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Development and Evaluation of an Assessment Method for Decentralized Stormwater Treatment Systems for Runoff from Traffic Areas

Maximilian Josef Huber

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Abstract

The runoff from traffic areas is polluted by several substances (e.g., heavy metals, hydrocarbons, compounds of de-icing salts, and solids) and must often be treated prior to discharge into receiving waters. In the last few years, many decentralized stormwater treatment systems have been developed and can be tested in Germany and worldwide by standardized methods. However, no detailed method is currently available for the determination of the service lives and the influences of de-icing salts on the remobilization of pollutants that were previously fixed onto filter materials. Decentralized stormwater treatment systems, which do not pose any risk for groundwater and surface water by discharging dissolved pollutants, can only be developed and operated with the knowledge of a realistic service life and a possible remobilization of previously retained heavy metals by de-icing salts. Therefore, the focus of this thesis was set on the development and evaluation of an assessment method for decentralized stormwater treatment systems for runoff from traffic areas.

In a first step, a literature study was performed to collect data from international monitoring programs (294 data sets) to determine the heavy metal concentrations of the most relevant heavy metals cadmium, chromium, copper, lead, nickel, and zinc in runoff from traffic areas. For these metals, the total and dissolved concentrations (median, average, and distribution) were determined separately for eight traffic area categories. In addition, the measured runoff concentrations of the metals antimony, cobalt, manganese, palladium, platinum, rhodium, titanium, and tungsten were compiled. Moreover, the influences of site-specific and method-specific factors on the results of monitoring programs were evaluated. As one result, the runoff concentrations of zinc were the most variable compared with the other heavy metals. Furthermore, the method of calculating average values has a huge impact on the results and the pH value of runoff waters does not correlate with the dissolved part of heavy metals in the field. In the 80s and 90s, the lead concentrations in runoff from traffic areas significantly decreased, whereas no trends were detected for copper and zinc.

The determined distributions of the dissolved metal concentrations were used to perform appropriate experiments for the evaluation of the heavy metal removal by filter materials in lab-, pilot-, and full-scale experiments. Although the removal mechanisms of filter materials consist of filtration (e.g., after precipitation), sorption, ion exchange, or biochemical transformation, the term “filter material” was deliberately chosen because it is commonly used by manufacturers and operators. At the beginning, a comparison of batch and column experiments was performed. Capacities and kinetics were determined that describe the removal of copper, nickel, and zinc by six filter materials. For batch experiments, the influences of two different types of shakers, a variation of the initial pH value (5 and 7), and the presence of a buffer was tested. For most experiments, the use of a rotary shaker, a pH buffer simulating ionic strength, and an initial pH value of 7 instead of 5

increased the capacities. In contrast to batch experiments, the column experiments were performed with more realistic boundary conditions. For the results of the column experiments, the differences between the filter materials were more significant compared with the batch test results. The capacities determined by column experiments decreased with increasing influent concentrations and shorter contact times. As a further result, lab-scale column experiments can be used as an indicator to determine the service lives of treatment systems. Batch experiments with heavy metals can only be used under identical and well-defined conditions to select appropriate filter materials and to prove the comparability of different production batches. In contrast, the cation exchange capacities, which were determined in accordance with the Standard Methods DIN EN ISO 11260:2011-09 and DIN EN 16070:2014-06, were not suitable to determine heavy metal removal capacities and to characterize the performance of filter materials. For the determination of realistic service lives and removal efficiencies of decentralized stormwater treatment systems for traffic area runoff, pilot-scale experiments are necessary. Therefore, a novel and standardized method was developed that consists of two parts. In Part 1, the filter material is rapidly preloaded in individual models with the heavy metals copper and zinc. In Part 2, the long-term performance of the system is assessed by simulating the last year of the filter material's service life with three different rain events. All tested treatment systems had different removal efficiencies, in particular in their long-term performances. The average removal efficiencies varied between 48.2% and 99.3% for copper and between 59.7% and 99.4% for zinc. The smallest values were determined during the rain event with the highest rain intensity. The applicability of the method was confirmed by a comparison of the novel test method results with field measurements, which were performed at four sites for two filter channel systems.

Moreover, the remobilization risk of previously retained heavy metals was determined for cadmium, copper, lead, nickel, and zinc under application of de-icing salts by lab-scale column experiments. Hereby, tests were performed with eight columns for each of the six filter materials to determine the heavy metal removal. Afterwards, three different experiments were performed with sodium chloride, a mixture of sodium chloride and calcium chloride, and a mixture of sodium chloride and magnesium chloride in duplicates. The mixture of sodium chloride and calcium chloride remobilized the most heavy metals. The remobilization increased with an increasing preloading of the filter and it also depends on the filter material and the heavy metal. The two preloaded columns without subsequent salt tests were used for further studies to describe the removal mechanisms.

A monograph was also published that provides an introduction into the topic decentralized stormwater treatment of runoff from traffic areas and metal roofs. It includes an overview of the system types, their removal mechanisms, the catchment areas, the discharge criteria, the receiving compartments, the legal requirements, the approval procedures, and the test methods.

Zusammenfassung

Verkehrsflächenabflüsse sind mit einer Vielzahl von Stoffen (z. B. Schwermetalle, Kohlenwasserstoffe, Auftausalze und Feststoffe) verunreinigt und bedürfen oftmals vor Einleitung in ein Gewässer einer Behandlung. In den letzten Jahren wurden zahlreiche dezentrale Behandlungsanlagen entwickelt, welche sowohl in Deutschland als auch weltweit anhand differenzierter Prüfverfahren getestet werden können. Jedoch ist derzeit keine detaillierte Methodik zur Bestimmung der Standzeit dieser Anlagen und zur Ermittlung des Einflusses von Auftausalzen auf die Remobilisierung von auf Filtermaterialien zurückgehaltenen Schadstoffen vorhanden. Nur bei Kenntnis einer realistischen Standzeit und einer möglichen Remobilisierung bereits zurückgehaltener Schwermetalle durch Auftausalze können dezentrale Behandlungsanlagen entwickelt und betrieben werden, die kein Risiko für den Eintrag gelöster Schadstoffe in Grund- und Oberflächengewässer bergen. Diese Arbeit behandelt daher die Entwicklung und Anwendung eines standardisierten Bewertungsverfahrens für dezentrale Anlagen zur Niederschlagswasserbehandlung von Verkehrsflächenabflüssen.

Als Basis für die Arbeit wurden in einem ersten Schritt 294 Datensätze internationaler Literatur ausgewertet, um die Schwermetallkonzentrationen der aufkommensrelevanten Stoffe Blei, Cadmium, Chrom, Kupfer, Nickel und Zink in Verkehrsflächenabflüssen zu bestimmen. Für diese Schwermetalle wurden die gesamten und gelösten Konzentrationen (Median, Mittelwert und Verteilung) getrennt für acht Verkehrsflächenkategorien bestimmt. Zusätzlich wurden gemessene Abflusskonzentrationen der Metalle Antimon, Kobalt, Mangan, Palladium, Platin, Rhodium, Titan und Wolfram zusammengestellt. Außerdem wurden orts- und methodenspezifische Einflüsse auf die Messwerte von Monitoring-Programmen evaluiert, wobei das Aufkommen von Zink besonders variabel ist. Anhand dieser Auswertung konnte u. a. gezeigt werden, dass die Art der Mittelung der Einzelergebnisse eines Standorts große Einflüsse auf die Charakterisierung der Verkehrsfläche hat und der pH-Wert des Abflusses nicht mit den gelösten Anteilen der Schwermetalle im Feld korrelierte. Während die Bleikonzentrationen der Verkehrsflächenabflüsse in den 80er und 90er Jahren deutlich zurückgingen, wurden keine Entwicklungen bei den Kupfer- und Zinkkonzentrationen festgestellt.

Anhand der ermittelten Konzentrationsverteilungen der gelösten Schadstoffe konnten geeignete Versuche an Filtermaterialien zur Bestimmung des Schwermetallrückhalts im Labor-, Pilot- und technischen Maßstab durchgeführt werden. Der Begriff „Filtermaterialien“ wurde bewusst gewählt, da er unter den Herstellern und Anwendern ein eingeführter Begriff ist. Filtermaterialien beinhalten jedoch u. a. die Wirkmechanismen Filtration (z. B. nach Fällung), Sorption oder Ionenaustausch. Zuerst wurden in vergleichenden Schüttel- und Säulenversuchen die Kapazitäten und Kinetiken von sechs Filtermaterialien bzgl. des Rückhalts von Kupfer, Nickel und Zink bestimmt. Dabei wurde der Einfluss der Schüttlerart, des pH-Wertes (5 oder 7) und der Verwendung von Puffern auf die

Ergebnisse der Schüttelversuche bestimmt. Bei den meisten Versuchen erfolgte eine Zunahme der Kapazitäten durch die Verwendung eines Vertikalschüttlers, eines pH-Wertes von 7 statt 5 und von Puffern. Anhand der Säulenversuche konnten die Unterschiede zwischen einzelnen Materialien im Vergleich zu den Schüttelversuchen differenzierter und unter realistischeren Bedingungen untersucht werden. Dabei reduzierten sich die anhand der Säulenversuche ermittelten Kapazitäten mit steigenden Zulaufkonzentrationen und kürzeren Kontaktzeiten. Die nach DIN EN ISO 11260:2011-09 und DIN EN 16070:2014-06 bestimmten Kationenaustauschkapazitäten ergaben, dass diese Methoden nicht zur Charakterisierung des Schwermetallrückhalts geeignet sind. Dahingegen sind Schüttelversuche mit Schwermetallen als definierte Methode zur Identifizierung geeigneter Materialien und zur Überprüfung von Produktionschargen sowie Säulenversuche im Labormaßstab zur Abschätzung von Standzeiten geeignet. Zur Bestimmung der tatsächlichen Standzeit und Behandlungsleistung dezentraler Anlagen zur Verkehrsflächenabflussbehandlung sind aber Versuche im Pilot-Maßstab notwendig. Die dafür entwickelte und standardisierte Methode besteht aus zwei Teilen: eine Vorbelastung der Filtermaterialien in geeigneten Modellen mit den Schwermetallen Kupfer und Zink sowie anschließend die Durchführung dreier Regenspenden zur Bestimmung der Rückhalteleistung im letzten Betriebsjahr vor dem Materialaustausch. Die untersuchten Anlagen zeigten besonders im Langzeitverhalten unterschiedliche Leistungen. Der mittlere Rückhalt variierte zwischen 48,2 % und 99,3 % für Kupfer und zwischen 59,7 % und 99,4 % für Zink. Die niedrigsten Werte wurden beim Regenereignis mit der höchsten Regenspende gemessen. Die Anwendbarkeit der Methode wurde durch die Beprobung zweier Rinnensysteme an vier Standorten nachgewiesen.

Anhand von Laborsäulenversuchen wurde anschließend das Remobilisierungsrisiko bereits gebundener Schwermetalle unter Auftausalzverwendung für Blei, Cadmium, Kupfer, Nickel und Zink bestimmt. Zuerst wurden für sechs verschiedene Filtermaterialien an je acht Säulen Versuche zum Schwermetallrückhalt durchgeführt. Je zwei Säulen wurden anschließend mit Natriumchlorid, einer Mischung aus Natrium- und Calciumchlorid sowie einer Mischung aus Natrium- und Magnesiumchlorid beschickt, wobei die Mischung aus Natrium- und Calciumchlorid die meisten Schwermetalle remobilisierte. Die Höhe der Remobilisierung schwankte zudem zwischen den einzelnen Filtermaterialien und den Schwermetallen und stieg mit zunehmender Beladung der Materialien an. Die beiden beladenen Säulen ohne anschließende Auftausalzversuche wurden für weitere Untersuchungen verwendet, um die grundlegenden Wirkmechanismen zu beschreiben.

Ebenfalls wurde eine Einführung in die Thematik der dezentralen Niederschlagswasserbehandlung für Verkehrsflächen- und Metalldachabflüsse erstellt. Diese Buchpublikation gibt einen Überblick zu den Anlagentypen und deren Wirkmechanismen, den Herkunftsflächen, den Zielkompartimenten, den rechtlichen Anforderungen sowie zu den Zulassungs- und Prüfverfahren.

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Abbreviations

AADT	Annual Average Daily Traffic
As	Arsenic
ASTRA	Bundesamt für Strassen, Schweiz (Federal Roads Office, Switzerland)
AUS	Australia
AUT	Austria
B	Boron
Ba	Barium
BBodSchV	Bundes-Bodenschutz- und Altlastenverordnung (German Soil Protection Ordinance)
BWK	Bund der Ingenieure für Wasserwirtschaft, Abfallwirtschaft und Kulturbau e. V. (German Association of Engineers for Water Management, Waste Management, and Land Management)
Ca	Calcium
Cd	Cadmium
CHE	Switzerland
Cl	Chloride
CN	Cyanide
Co	Cobalt
Cr	Chromium
CrO_4^{2-}	Chromate
CS	Channel System
Cu	Copper
DCMU	3-(3,4-dichlorophenyl)-1,1-dimethylurea (trade name: Diuron)
DEHP	di(2-ethylhexyl)-phthalate
DEU	Germany
DIBt	Deutsches Institut für Bautechnik (German Centre of Competence for Construction)
DWA	Deutsche Vereinigung für Wasserwirtschaft, Abwasser und Abfall e. V. (German Association for Water, Wastewater and Waste)

ETBE	Ethyl-tert-butyl ether
F	Fluoride
FCS	Filter Channel System
FSS	Filter Shaft System
GFS	Geringfügigkeitsschwellenwerte (de minimis threshold values)
GrwV	Grundwasserverordnung (Groundwater Ordinance)
Hg	Mercury
LANUV	Landesamt für Natur, Umwelt und Verbraucherschutz Nordrhein-Westfalen (North Rhine-Westphalian Agency for Nature, Environment, and Consumer Protection)
LAWA	Bund/Länder-Arbeitsgemeinschaft Wasser (German Working Group on Water Issues)
MCPP	Methylchlorophenoxypropionic acid (Mecoprop)
Mg	Magnesium
Mn	Manganese
Mo	Molybdenum
MTBE	Methyl-tert-butyl ether
MUSIC	Model for Urban Stormwater Improvement Conceptualization
Na	Sodium
NaCl	Sodium chloride
NH_4^+	Ammonium
Ni	Nickel
NJ	New Jersey
NJDEP	New Jersey Department of Environmental Protection
NLD	The Netherlands
NO_3^-	Nitrate
NRW	Nordrhein-Westfalen (State of North Rhine-Westphalia, Germany)
P	Phosphor
PAH	Polycyclic Aromatic Hydrocarbons

Pb	Lead
Pd	Palladium
PGE	Platinum-Group Elements
PP	Permeable Pavements
Pt	Platinum
Rh	Rhodium
Sb	Antimony
Se	Selenium
Sn	Tin
SO_4^{2-}	Sulfate
SS	Shaft System
SWMM	Stormwater Management Manual for Western Washington
TAPE	Technology Assessment Protocol – Ecology
TARP	Technology Acceptance Reciprocity Partnership
Tl	Thallium
TN	Total Nitrogen
TP	Total Phosphor
TPH	Total Petroleum Hydrocarbons
TrinkwV	Trinkwasserverordnung (Drinking Water Ordinance)
TSS	Total Suspended Solids
UBA	Umweltbundesamt (Federal Environment Agency)
USA	United States of America
V	Vanadium
VA	Virginia
VTAP	Virginia Technology Assessment Protocol
WA	Washingtion (state)
WHG	Wasserhaushaltsgesetz (Federal Water Act)
Zn	Zinc

Chapter 1: Introduction

1.1 Substances in Traffic Area Runoff

The occurrence of substances in traffic area runoff depends on several processes. Dustfalls and dry deposition during periods without rain entrain contaminants and remove them from the atmosphere (Hamilton et al., 1987; Ball, 2002). The processes during rain events are wet deposition (removal of additional substances from the atmosphere) and wash-off of previously deposited pollutants (Welker & Dittmer, 2005). Concerning the wash-off, CH2MHILL (1998) concluded that splashing and washing of pollutants from vehicles is more important than the wash-off of pollutants accumulated on road surfaces. Both the wash-off and the substances in the atmosphere can be linked to traffic related sources. The main traffic related sources of pollutants are summarized in Table 1, which contains seven heavy metals, three noble metals, five de-icing salt compounds, and four organic substances. For most heavy metals, the wear of brakes and tires are relevant sources. Moreover, braking, acceleration, and steering activities lead to increased abrasion of tires, higher use of brake linings, and increased automotive exhaust gas emissions (Muschak, 1990; Langbein et al., 2006). The corrosion and subsequent dissolution of zinc (Zn) from the surface of galvanized elements during rain events is also a relevant source (Gan et al., 2008). Further traffic related sources are the use of catalytic converters in vehicles (Wei & Morrison, 1994; Geiger-Kaiser & Jäger, 2005), road maintenance with de-icing salts (Novotny et al., 1998; Dai et al., 2012), road wear, and drip losses. All of these sources lead to a pollution of traffic area runoff.

The concentrations of the substances in the runoff from traffic areas are based on climatic site-specific factors and fixed site-specific factors, which consist of the three categories surrounding land use characteristics, traffic area site data, and operational characteristics (Shelley et al., 1987). To specify the land use characteristics, several categories are used: non-urban, urban, ultra-urban, transportation, residential, open, agriculture, commercial, mixed, and industrial (Kayhanian et al., 2003; Flint and Davis, 2007). Traffic area site data contain information on vegetation, topography (cut, at grade, or elevated; Driscoll et al., 1990), and road design (e.g., cross-sections, pavement surfaces, and drainage areas) (Huber et al., 2016e). The operational characteristics describe influences concerning traffic volumes (Annual Average Daily Traffic, AADT), percentage of trucks, driving-style categories (Horstmeyer et al., 2016), types and speed of vehicles, maintenance (e.g., mowing of the roadside shoulders, street sweeping, and winter services; Helmreich et al., 2010), and institutional regulations (e.g., phase-out of leaded gasoline). Climatic factors are dry deposition rates, antecedent dry periods, rain characteristics (volume, intensity, and duration), seasonal effects, and wind turbulences. These factors, which can have contradicting effects on the concentrations, have an influence on the real contamination of runoff from traffic areas.

Table 1: Substances in traffic area runoff waters and their traffic related sources (Ward, 1990; Wei & Morrison, 1994; Young et al., 1996; Sansalone & Buchberger, 1997a+b; Ball et al., 1998; Dierkes & Geiger, 1998; Novotny et al., 1998; Hares & Ward, 1999; Legret & Pagotto, 1999; Pahlke et al., 2000; Root, 2000; Davis et al., 2001; Borden et al., 2002; Denkhaus & Salnikow, 2002; Van Bohemen & Van De Laak, 2003; Geiger-Kaiser & Jäger, 2005; Hillenbrand et al., 2005; Mangani et al., 2005; Preciado & Li, 2005; Bleiwas, 2006; Masanao et al., 2006; Wilson, 2006; McKenzie et al., 2009; Kocher et al., 2010a+b; Dai et al., 2012).

Substance	Main sources
Antimony (Sb)	Brake linings, tires
Cadmium (Cd)	Batteries, brake linings, de-icing salts, fuel, insecticides, tires
Chromium (Cr)	Brake linings, metal plating, moving engine parts, tires
Copper (Cu)	Brake linings, de-icing salts, electronic equipment, fungicides, insecticides, metal plating, moving engine parts, pesticides, tires
Lead (Pb)	Brake linings, de-icing salts, fuel additive, grease, lubricating oil, tires, paints, vehicle exhaust, weights for tire balance
Manganese (Mn)	Brake linings, gasoline additives, moving engine parts, tires, vehicle exhaust
Nickel (Ni)	Asphalt surfaces, batteries, brake linings, diesel fuel, electronic equipment, lubricating oil, metal plating, stainless steel, vehicle exhaust
Zinc (Zn)	Batteries, brake linings, bridges, de-icing salts, galvanized car parts, grease, guardrails, lamp-posts, motor oil, road signs, safety fences, sign-posts, tires
Platinum-Group Elements (PGE): palladium (Pd), platinum (Pt), and rhodium (Rh)	Catalytic converters
Calcium (Ca)	Concrete surfaces, de-icing salts
Chloride (Cl)	De-icing salts
Cyanide (CN)	De-icing salts
Magnesium (Mg)	De-icing salts
Sodium (Na)	De-icing salts
Ethyl-tert-butyl ether (ETBE)	Fuel additive
Methyl-tert-butyl ether (MTBE)	Fuel additive
Polycyclic Aromatic Hydrocarbons (PAH)	Asphalt surfaces, vehicle exhaust
Total Petroleum Hydrocarbons (TPH)	Anti-freeze and hydraulic fluids, asphalt surface, drip losses, motor lubricants

Parameters usually monitored in runoff from traffic areas include solids, oxygen-consuming substances, nutrients, heavy metals, compounds of de-icing salts, and organic substances (e.g., Folkeson et al., 2009; Kayhanian et al., 2012). Several of these substances are analyzed as sum parameters such as solids (e.g., Total Suspended Solids (TSS)), oxygen-consuming substances (e.g., Chemical Oxygen Demand), and hydrocarbons (e.g., Polycyclic Aromatic Hydrocarbons (PAH) and Total Petroleum Hydrocarbons (TPH)). In addition, individual trace organic substances, pathogens, and toxicities are sometimes measured in runoff waters (e.g., Welker, 2014). The concentrations of heavy metals and trace organic substances (e.g., PAH) are one to several magnitudes lower than most of the other substances (e.g., solids, nutrients, and chloride (Cl)). Thus, traffic area runoff is polluted by substances in varying concentration ranges. At sites with winter services, the highest concentrations are measured for the parameter Cl with maximum values of 60.3 g/L (Jenewein & Schinner, 1982).

Summaries of the pollution of runoff from traffic areas are often found in dissertations (e.g., Grotehusmann, 1995; Haritopoulou, 1996; Zessner, 1999; Kasting, 2003; Li, 2014), in habilitations (e.g., Welker, 2004; Helmreich, 2010), and in research reports (e.g., Boller, 2003; Herrera, 2007; Helmreich et al., 2014) but also in journal papers (e.g., Ellis & Revitt, 1982). However, most authors had a special focus and subsequently considered only runoff data of one country, one traffic area category, or selected substances to obtain the data for their individual objectives. Moreover, most previous literature studies did not distinguish between different traffic area categories (e.g., parking lots, feeder streets, bridges, or highways) and the influences of site-specific data and method-specific factors on the results had not been evaluated on the basis of a large data set.

Concerning the metal runoff concentrations from traffic areas, most of the previous literature studies only summarize the runoff concentrations of cadmium (Cd), copper (Cu), lead (Pb), and zinc (Zn) that were most frequently measured in monitoring programs of the last 40 years. However, several Pb sources were substituted in the last decades and subsequently Pb runoff concentrations sharply decreased at the end of the 20th century (Kayhanian, 2012). Although the results of older monitoring programs are not relevant for current stormwater challenges, these Pb data are still considered in most literature studies. Further substances that might be relevant for authorities because of regulations (e.g., chromium (Cr) and nickel (Ni); cf. Section 1.3) have often not been considered in previous literature studies due to a lack of monitoring data. Further metals that were sometimes measured in the runoff of traffic areas include aluminum, antimony, arsenic, calcium, cobalt, iron, manganese, mercury, palladium, platinum, rhodium, sodium, titanium, and tungsten. Rarely measured metals in the runoff of traffic areas are barium, beryllium, boron, gold, magnesium, molybdenum, potassium, selenium, silicon, silver, thallium, tin, and vanadium. Furthermore, most monitoring programs do not distinguish between dissolved and total concentrations and do not publish annual heavy metal loads normalized per hectare of impervious catchment area, which can be used to calculate mass balances. Therefore, reliable runoff concentration ranges were missing for dissolved substances in the past. Nevertheless, the partitioning of heavy metals is often of particular concern because the dissolved fractions are directly biologically available and are subsequently relevant for toxic effects (e.g., Paulson & Amy, 1993; Crabtree et al., 2008a).

To differentiate the contamination level of the runoff from all types of traffic areas, categories of pollution are often used in standards and regulations because different traffic areas are influenced by specific factors. For example, runoff from bridges can be highly affected by the corrosion of galvanized elements (Wilson, 2006), highway runoff by guardrails and safety fences (Dierkes and Geiger, 1998), non-urban road runoff by the use of pesticides for farming (Mangani et al., 2005), road runoff by the percentage of trucks (Gan et al., 2008), and runoff of parking lots by different frequencies of use (Göbel et al., 2007) and drip losses (Grotehusmann & Kasting, 2002). Thus, the pollution sources vary at different traffic area categories. Consequently, the runoff concentrations can be different for each category. The categorizations of roads are mostly based on the AADT, main land use categories (urban, non-urban, or industrial), and the percentage of trucks. Differentiations are also considered for parking lots and runways of airports. Four different standards and local regulations are summarized in Table 2.

Less polluted traffic areas are commonly bikeways, footpaths, and roads with an AADT of less than 300 vehicles per day (DWA-M 153E, 2007; SN 640 347, 2009). In NRW (State of North Rhine-Westphalia, Germany), areas with low and medium runoff concentrations are called “category II”. Feldhaus et al. (2009) proposed a differentiation of “category II” between IIa (< 300 vehicles per day; low runoff concentrations) and IIb (> 2000 vehicles per day; medium runoff concentrations). Because the pollution of traffic areas with 300–2000 vehicles per day could not be generally specified, local authorities must decide individually about the pollution levels of roads within this AADT range. For traffic areas with expected medium and high runoff concentrations, the differentiation of these standards and regulations is more heterogeneous. In particular, the influences of industrial zones, trucks, non-urban land use, and frequencies of the use of parking lots are assessed differently. Moreover, the categorization of SWMM (2005) does not clearly distinguish between medium und high runoff concentrations. Nevertheless, these categories are useful to estimate the pollution of traffic areas because quantitative measurements at each site are not possible.

In general, most of the classification of traffic areas by these three categories of pollution is only verified by some research projects (e.g., Grotehusmann & Kasting, 2002; Steiner et al., 2006; Nadler & Meißner, 2007). Individual site-specific factors such as corrosion of galvanized elements, noise barriers, and stop-and-go traffic are not considered by these standards and local regulations. A statistically robust analysis of the pollution at different traffic areas to support these categorizations is not possible because of the varying site-specific factors and their large quantity of – sometimes contradicting – influences. However, a review of literature data can improve the differentiation of less and highly polluted categories of traffic areas and can also consider further categories (e.g., bridges).

Table 2: Categories of pollution for different traffic areas.

Reference	Low runoff concentrations	Medium runoff concentrations	High runoff concentrations
German Standard DWA-M 153E (2007)	Roads with less than 300 vehicles per day; bikeways; footpaths; car parking lots without frequent change of vehicles in housing areas	Roads with 300–15,000 vehicles per day; backyards; car parking lots without frequented vehicle change in mixed areas and industrial estates	Roads with more than 15,000 vehicles per day; car parking lots with frequent vehicle change; traffic areas with heavy pollution (e.g., because of farming, markets); truck parking lots
Swiss Standard SN 640 347 (2009)	Roads*; bikeways; footpaths; less frequently used car parking lots in residential zones	Roads*; logistics centers; frequently used parking lots	Roads (in most cases with more than 15,000 vehicles per day)*; highways; frequently used parking lots
Regulation of the State of North Rhine-Westphalia (NRW), Germany MUNLV (2004)	Less frequently used traffic areas (specifications are proposed by Feldhaus et al. (2009))		Logistics centers; highly frequented traffic areas; traffic areas in industrial zones; highly polluted traffic areas (e.g., farming, runways, places with waste or wastewater disposal)
Stormwater Management Manual for Western Washington, United States of America SWMM (2005)	Not defined	Urban highways with more than 15,000 vehicles per day**; other urban roads with more than 7500 vehicles per day**; non-urban roads with more than 15,000 vehicles per day**	

* depending on AADT, percentage of trucks, urban/non-urban, grade, de-icing salt application, and road sweeping; ** if runoff is discharged to fish-bearing streams, lakes, or to waters or conveyance systems tributary to fish-bearing streams or lakes, enhanced treatment is necessary (cf. Chapter 2)

1.2 Metal Emissions from Traffic Areas in Germany

For the year 2013, the UBA (Umweltbundesamt; Federal Environment Agency) published total annual emissions of road transportation of 0.8 t Cd, 25.7 t Cr, 2109 t Cu, 5.0 t Ni, 88.7 t Pb, and 1906 t Zn in Germany (Umweltbundesamt, 2015). The percentages of the traffic related emissions based on the total emissions are 11%, 45%, 98%, 5%, 42%, and 95% for Cd, Cr, Cu, Ni, Pb, and Zn, respectively. Considering these data, road transportation is an important source of total heavy metal emissions in Germany.

A further aspect is the trend of the atmospheric heavy metal emissions since 1990 (Umweltbundesamt, 2015). The trend shows that Cu and Zn emissions of road transportation are steadily increasing, and the Pb emissions decreased drastically in the last decade of the 20th century (Figure 1). The total atmospheric emissions of Cd, Cr, and Ni were low compared with the other three heavy metals and were constantly emitted for the last 15 years. The trend of the Pb emissions also verifies that a differentiation of Pb runoff concentration measurements between the 20th century and the 21st century must be considered as it was done in the worldwide and European reviews (cf. Chapter 5).

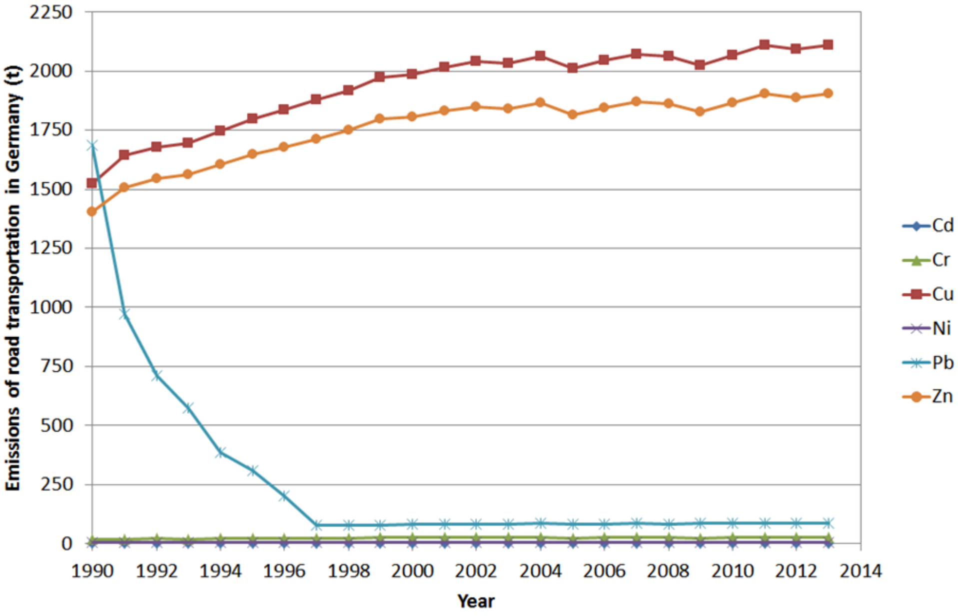

Figure 1: Trend of atmospheric heavy metal emissions of road transportation since 1990 (based on data from Umweltbundesamt (2015)).

Hillenbrand et al. (2005) calculated the sinks of traffic related emissions for three different compartments. Because approximately 73.5% of the traffic emissions originate from non-urban roads, most runoff water is infiltrated into the soil and groundwater system (Table 3). In urban catchment areas, most of the runoff is discharged by sewer systems. Most of this water is treated by a municipal wastewater treatment plant or pretreated by stormwater reservoirs and some water is discharged into receiving surface waters without treatment. Because of the dry deposition of heavy metals and the subsequent street sweeping, several heavy metals were disposed as waste in the past.

Table 3: Traffic related heavy metal sinks in Germany (adapted from Hillenbrand et al., 2005). Sources: Brake lining wear, tire wear, roadway abrasion, weights for tire balance, guardrails, and signs.

Pathway	Cu (t/a)	Pb (t/a)	Zn (t/a)
Soil	797	67.0	1935
Surface water	103	9.7	399
Waste disposal	32	3.4	117

The total heavy metal loads of stormwater runoff that are entering receiving waters by separate and combined sewer systems were calculated by Fuchs et al. (2002) as approximately 187 t/a Cu, 87.4 t/a Pb, and 1193 t/a Zn. These loads also include stormwater runoff from non-traffic related surfaces and are subsequently higher than the values in Table 3 for surface water. A reduction of these loads can be achieved by appropriate decentralized stormwater treatment systems at the runoff sites (cf. Chapter 2).

1.3 Regulations in Germany

In Germany, stormwater runoff that is collected and purposefully discharged is defined as wastewater in accordance with § 54 Wasserhaushaltsgesetz (WHG, 2009; Federal Water Act). Moreover, surface water and groundwater must be protected from substance emissions that are likely to cause adverse changes in water quality in a continuous or in a not inconsiderable extent (cf. § 9 WHG, 2009).

1.3.1 Infiltration into the Soil and Groundwater System

For infiltration into the soil and groundwater systems, the threshold values of the Bundes-Bodenschutz- und Altlastenverordnung (BBodSchV, 1999; German Soil Protection Ordinance) were previously used in Germany. With the implementation of the Grundwasserverordnung on 09 November 2010 (GrwV, 2010; Groundwater Ordinance), emissions of certain pollutants and pollutant groups, including Cu and Zn, have to be limited by suitable measures. For the protection of the groundwater against pollution and deterioration, the Geringfügigkeitsschwellenwerte (GFS; de minimis threshold values) of the Bund/Länder-Arbeitsgemeinschaft Wasser (LAWA, 2004; German Working Group on Water Issues) must be considered by the German authorities. The GFS values are defined as concentrations at which no significant toxicological effect can occur. Furthermore, the GFS take into account the requirements of the Trinkwasserverordnung (TrinkwV, 2001; Drinking Water Ordinance) for both health and aesthetic aspects (LAWA, 2004). The GFS of the year 2004 were updated in the years 2012/2013 (Moll & Quadflieg, 2014). The thresholds of several parameters were adjusted because of legal changes in the European Union and new technical findings. A new ordinance called Mantel-Verordnung (3rd draft, published on 23 July 2015) is intended to harmonize the GrwV and the BBodSchV and will further reinforce the binding character of the GFS. Table 4 compares the thresholds of all four regulations for selected inorganic and organic parameters. Thus, a tightening of the requirements for infiltration was implemented in recent years. In addition, new parameters such as Cl (250 mg/L), Ethyl-tert-butyl ether (ETBE) (2.5 µg/L), and Methyl-tert-butyl ether (MTBE) (5 µg/L) are also considered in the new ordinances.

In many cases, the adjustments of heavy metal thresholds, especially for Cu with 10–14 µg/L, mean that some thresholds are already exceeded by pure rain (Dauber et al., 1979; Dannecker et al., 1990;

Athanasiadis et al., 2007). Therefore, an infiltration of the precipitation into the soil and groundwater system would not be possible without a pretreatment. Thus, compliance issues regarding the implementation of the Mantel-Verordnung must be addressed soon.

Table 4: Selection of thresholds for infiltration into the soil and groundwater systems: thresholds of the BBodSchV (1999) for the pathway soil-groundwater, the GFS from 2004 (LAWA, 2004) and 2013 (Moll & Quadflieg, 2014), and the groundwater thresholds of the 3rd draft of the Mantel-Verordnung (2015). Strengthened requirements with respect to the previously valid BBodSchV are marked in bold.

Parameter	Unit	BBodSchV, 1999	GFS, 2004	GFS, 2013	3rd draft Mantel-Verordnung, 2015
Antimony (Sb)	µg/L	10	**5**	**5**	**5**
Arsenic (As)	µg/L	10	**10**	**2.6**	**10**
Barium (Ba)	µg/L	-	**340**	**186**	**340**
Boron (B)	µg/L	-	**740**	**180**	**740**
Cadmium (Cd)	µg/L	5	**0.5**	**0.25**	**0.25**
Chromium (Cr) (total)	µg/L	50	**7**	**3.4**	**7**
Chromate (CrO_4^{2-})	µg/L	8	-	-	-
Cobalt (Co)	µg/L	50	**8**	**5.7**	**8**
Copper (Cu)	µg/L	50	**14**	**10**	**14**
Lead (Pb)	µg/L	25	**7**	**7.2**	**7**
Mercury (Hg)	µg/L	1	**0.2**	**0.05**	**0.05**
Molybdenum (Mo)	µg/L	50	**35**	**35**	**35**
Nickel (Ni)	µg/L	50	**14**	**13**	**20**
Selenium (Se)	µg/L	10	**7**	**3**	**7**
Thallium (Tl)	µg/L	-	**0.8**	**0.2**	**0.8**
Tin (Sn)	µg/L	40	-	-	-
Vanadium (V)	µg/L	-	**4**	**4**	**4**
Zinc (Zn)	µg/L	500	**58**	**50**	**58**
Ammonium (NH_4^+)	mg/L	-	-	-	**0.5**
Chloride (Cl)	mg/L	-	**250**	**250**	**250**
Cyanide (CN) (total)	µg/L	50	50	50	50
Cyanide (CN) (easily accessible)	µg/L	10	**5**	10	**5**
Fluoride (F)	µg/L	750	750	750	750
Nitrate (NO_3^-)	mg/L	-	-	-	**50**
Sulfate (SO_4^{2-})	mg/L	-	**240**	**250**	**250**

1.3.2 Discharge into Surface Water

In Germany, no nationwide legally binding rules exist for the discharge of stormwater runoff into surface water. To guarantee a sufficient treatment of stormwater runoff, the generally accepted rules of technology must be considered in Germany for planning treatment systems (cf. § 60 WHG, 2009). As relevant rules and standards, DWA-M 153E (2007), BWK-M 3 (2007), and BWK-M 7 (2008) are applied. The DWA-M 153 (2007) has implemented an emission-based method and this standard will soon be replaced by the DWA-A 102. In contrast, the two BWK (Bund der Ingenieure für Wasserwirtschaft, Abfallwirtschaft und Kulturbau e. V.; German Association of Engineers for Water Management, Waste Management, and Land Management) rules represent immission-based approaches. The BWK-M 3 (2007) will be replaced in the future by a new worksheet (BWK-A 3).

Furthermore, the list of priority substances in the field of water policy was updated by the European Union in recent years to prevent a chemical pollution of surface water in the short- and long-term (DIRECTIVE 2008/105/EC, 2008; DIRECTIVE 2013/39/EC, 2013). Besides other parameters, the list includes mercury (Hg) and the heavy metals Cd, Ni, and Pb, which are also measured in traffic area runoff. The annual average values of these substances should not exceed 0.05 µg/L, 0.08–0.25 µg/L (depending on water hardness classes), 20 µg/L, and 7.2 µg/L in inland surface waters, respectively. Because all member states of the European Union have to achieve good chemical status of their surface waters, in which no pollutant may occur at a higher concentration than these environmental quality standards, it will not be allowed for authorities to discharge stormwater runoff from traffic areas, which exceed these values, into receiving waters on the long-term.

Chapter 2: Decentralized Treatment Systems for Traffic Area Runoff

2.1 Application and Functionality of Manufactured Decentralized Stormwater Treatment Systems

In recent years, various manufactured decentralized treatment systems have been developed to treat stormwater runoff at the source before it infiltrates into the subsurface or it is discharged to surface water (Dierkes et al., 2015a). They are particularly used when space is limited (Sample et al., 2012). Manufactured decentralized systems for treating stormwater runoff are suitable for several kinds of catchment areas. The drained catchment areas can be roofs, metal roofs, traffic areas, and combined surfaces, which sometimes also include building facades (Figure 2). The runoff of traffic areas can be low, medium, or highly polluted (cf. Table 2). In contrast, metal roof runoff (e.g., Zn or Cu roofs) always contains high concentrations of dissolved heavy metals (Helmreich, 2010) and it is not polluted with de-icing salts that are used by winter services on traffic areas. Most roofs (e.g., tiled roofs and shingle roofs) and combined surface areas (roofs and low polluted traffic areas such as sidewalks) are low or medium polluted. Appropriate decentralized systems must be used for the treatment of stormwater runoff to achieve a specific treatment efficiency. These individual systems take into account both the type and pollution of the connected catchment area and the receiving water that are based on legislative requirements and local regulations (Welker & Huber, 2014).

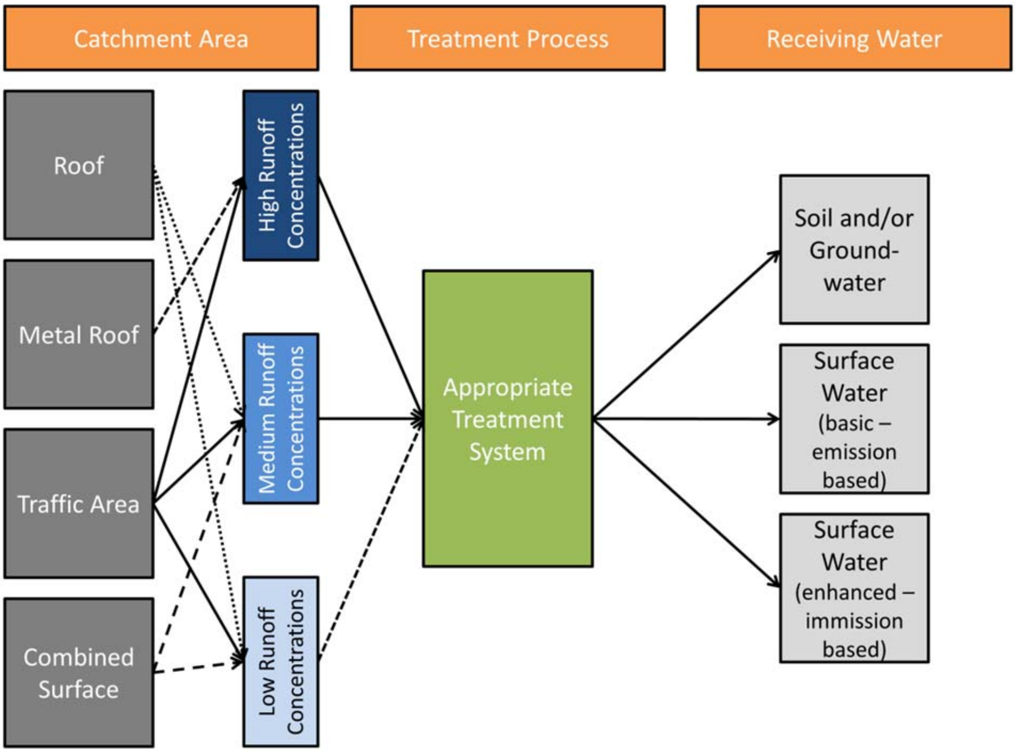

Figure 2: Application of decentralized stormwater treatment systems (adapted from Welker, 2015).

Most regulations differentiate between an infiltration of treated runoff into the subsurface or a discharge into surface water. Thus, different thresholds must be considered. For infiltration into the soil and groundwater system, distinct legislative requirements exist in Germany (cf. Section 1.3.1). However, the regulations are ambiguous for discharge into surface water (cf. Section 1.3.2).

The main removal mechanisms of manufactured decentralized stormwater systems are sedimentation, filtration, sorption, ion exchange, precipitation, and biochemical transformation (Huber et al., 2015d). In general, the systems consist of one, two, or three treatment steps. In a first step, sedimentation and/or filtration are the predominant retention mechanisms used to remove particle-bound substances. In several treatment systems, dissolved heavy metals are subsequently retained on filter materials by sorption, ion exchange, and precipitation (Liu et al., 2005). Thus, a second treatment step with a filter material is necessary to reduce the total concentrations of all metals (Hilliges et al., 2013; Maniquiz-Redillas & Kim, 2014). In the following chapters, these materials are consequently named “filter materials” because it is a commonly used term, although their removal mechanisms also include filtration after precipitation, sorption, ion exchange, or biochemical transformation. Nevertheless, the use of a filter material is often not required for basic treatment (e.g., discharge of runoff from low/medium polluted catchment areas into surface water – emission based). In addition, oil traps can be used to remove TPH, oil, and grease.

Manufactured decentralized stormwater treatment systems can be categorized into three types: permeable pavements (PP), filter channel systems (FCS), and filter shaft systems (FSS) (Dierkes et al., 2015a). These systems are commonly used as conforming products for enhanced stormwater treatment. For basic treatment (emission based discharge into surface water, cf. Figure 2), decentralized systems in gully pots, channel systems (CS), and shaft systems (SS) without a filter material are often used. An overview of all manufactured decentralized treatment systems for runoff from traffic areas and metal roofs, which are available in Germany and Austria, is presented in Huber et al. (2015d).

To guarantee a sufficient treatment, different test methods have been developed for manufactured decentralized systems treating the runoff from traffic areas (cf. Section 2.2). For the metal roofs, the preliminary test criteria of the Bavarian Environment Agency (Bayerisches Landesamt für Umwelt, 2011) must be applied to guarantee an appropriate runoff treatment in Bavaria, Germany. However, these criteria cannot be used for systems treating traffic area runoff because this runoff also includes both solids and de-icing salt compounds.

2.2 Assessment Procedures and Criteria

2.2.1 Worldwide Overview of Assessment Methods and Criteria Proposed by Authorities

Different assessment procedures were developed to test the performance of decentralized stormwater treatment systems for runoff from traffic areas. For approvals, further aspects such as environmental risk assessments, internal/external quality controls, and maintenance issues can also be mandatory. In addition, criteria for assessing field measurements are sometimes proposed. Table 5 presents a worldwide overview of test methods and criteria for manufactured stormwater treatment systems. All test methods are described in the following and the criteria that consider the removal of heavy metals or the remobilization of pollutants because of de-icing salt applications are reported in detail.

In Germany (DEU), the Deutsches Institut für Bautechnik (DIBt; German Centre of Competence for Construction) can approve decentralized systems for infiltration of runoff from traffic areas into the soil and groundwater system after passing several tests (e.g., removal of fine particles, TPH, and heavy metals; remobilization of heavy metals) (DIBt, 2012; DIBt, 2015). One procedure was developed for PP (DIBt, 2012) and one procedure for FCS and FSS (DIBt, 2015). One part of this laboratory test investigates the removal of dissolved heavy metals by three different rain events. The requested elimination is 80% for Cu and 70% for Zn (on average). Afterwards, a test simulating one rain event is performed under the application of sodium chloride (NaCl) to determine the risk of remobilization for Cu and Zn. In this part, the difference between the effluent concentrations and the influent concentrations must be below the threshold values of the BBodSchV (1999) (Cu 50 µg/L and Zn 500 µg/L). In NRW, a method was developed by the Landesamt für Natur, Umwelt und Verbraucherschutz Nordrhein-Westfalen (LANUV, 2012; North Rhine-Westphalian Agency for Nature, Environment, and Consumer Protection) for discharge into surface water as an emission-based approach. The test procedure consists of lab- and field experiments and it is mainly based on different TSS parameters (LANUV, 2012). The mandatory field measurements last at least twelve months.

In the Netherlands (NLD), a standard method was developed to approve FCS by laboratory tests (BRL 2036, 2008). Parts of the method are comparable with the DIBt procedures. However, it also considers the removal of four heavy metals (Cd, Cu, Pb, and Zn), which is determined by cation exchange capacities. Capacities of ≥40 cmol$^+$/kg are required for approval.

In Austria (AUT), the draft of the new Standard Method ÖNORM B 2506-3 Entwurf (2015) was published, which is based on a research report (Fürhacker et al., 2013). In lab-scale column tests, the filter materials of stormwater treatment systems for runoff from metal roofs (Cu and Zn) and traffic areas are evaluated by several tests (e.g., removal of fine particles, TPH, and heavy metals; remobilization of heavy metals). The requested elimination is 80% for Cu and 50% for Zn (on average). The Pb effluent concentrations must be below 9 µg/L. Afterwards, a remobilization test is performed under the application of sodium chloride to determine the risk of remobilization for Cu and Zn. In this part, the effluent concentration criteria are ≤50 µg/L Cu and ≤500 µg/L Zn.

Table 5: Overview of the test methods and criteria for stormwater treatment systems for traffic area runoff.

Reference	Country	Type of systems	Test procedure	Substances**	Receiving water
ASTRA, 2015a+b*	CHE	All	Field	Cu, TSS, Zn	Not specified
BRL 2036, 2008*	NLD	FCS	Lab	Cd, Cu, Pb, TPH, TSS, Zn	Groundwater
Department of Ecology, 2011	USA, WA	All	Lab & field	Cu, P, TPH, TSS, Zn	Groundwater and surface water
DIBt, 2012	DEU	PP	Lab	Cu, NaCl, TPH, TSS, Zn	Groundwater
DIBt, 2015	DEU	FCS, FSS	Lab	Cu, NaCl, TPH, TSS, Zn	Groundwater
NJDEP, 2009	USA, NJ	All	Field	TSS	Not specified
NJDEP, 2013	USA, NJ	All	Lab	TSS	Not specified
MUNLV, 2004; LANUV, 2012*	DEU	CS, FCS, FSS, gully pots, SS	Lab & field	TSS	Surface water
ÖNORM B 2506-3 Entwurf, 2015*	AUT	All	Lab	Cu, NaCl, Pb, TPH, TSS, Zn	Groundwater
Penrith, 2014	AUS	All	Modeling	TPH, TN, TP, TSS	Groundwater and surface water
Sample et al., 2012*	USA, VA	All	Lab or field	P, TSS	Not specified
Schmidt et al., 2015*	CHE	All	Lab & field	Cu, DCMU, MCPP, Zn	Not specified
Steiner et al., 2010	CHE	All	Field	Cu, DOC, PAH, TSS, Zn	Not specified
Victorian Stormwater Committee, 2006	AUS	All	Not specified	Dissolved substances, TN, TP, TSS	Not specified

* in development or reassessing; ** TSS, TPH, and P (phosphor) are individually defined by each method

In Switzerland (CHE), an assessment procedure will be proposed at the end of 2016 for decentralized stormwater treatment systems for runoff from traffic areas, roofs, and building facades. The novel method will consist of lab and field experiments. Among other experiences, it is based on previous lab-scale and field experiments with heavy metals (Cu and Zn) and trace organic substances (e.g., 3-(3,4-dichlorophenyl)-1,1-dimethylurea (DCMU) and Mecoprop (MCPP)) (Schmidt et al., 2015). The field experiments will be similar to the monitoring programs proposed by the Swiss Federal Roads Office (ASTRA) that include the measurement of Cu, TSS, and Zn (ASTRA 2015a; ASTRA 2015b). Moreover, assessment criteria for the performance of stormwater treatment systems for traffic area runoff exist that are used to evaluate field-monitoring results of at least twelve months. The criteria consist of absolute effluent concentrations discharging into receiving water, removal efficiencies, specific surface loadings, and hydraulic performances (Steiner et al., 2010). In addition to other parameters, the total heavy metal concentrations and removal efficiencies of Cu and Zn were considered as indicators for the performance of stormwater treatment systems. Furthermore, potential removal efficiencies of treatment systems have been categorized. For the Cu and Zn removal efficiencies, a range of < 60% to > 90% is postulated by classifying five different levels of treatment quality. For the total effluent concentrations, five ranges of < 5 µg/L to > 20 µg/L for Cu and of < 10 µg/L to > 40 µg/L for Zn are defined.

In the United States (USA), some methods have been developed in several federal states to test decentralized systems. In the State of Washington (WA), the Technology Assessment Protocol-Ecology (TAPE) of the Washington State Department of Ecology is used to evaluate and classify decentralized stormwater treatment systems by lab- and field-scale experiments (e.g., removal of TSS, Cu, Zn, phosphor (P), and TPH) (Department of Ecology, 2011). Besides requirements for the basic treatment, which include the removal of TSS, enhanced treatment facilities must exceed a removal of 30% dissolved Cu and 60% dissolved Zn in a field-monitoring. In New Jersey (NJ), a procedure for obtaining verifications of manufactured stormwater treatment systems was implemented by the New Jersey Department of Environmental Protection (NJDEP) to assess the removal of TSS and other parameters in the laboratory (NJDEP, 2013). As an amendment to the Technology Acceptance Reciprocity Partnership (TARP), which was proposed by the states California, Massachusetts, Maryland, New Jersey, Pennsylvania, and Virginia (VA), a field-testing for the removal of TSS can also be performed for final certification (NJDEP, 2009). Thus, TARP focuses primarily on sediments and compounds that can be removed with sediments, whereas TAPE addresses the removal of more substances that may also be dissolved (Sample et al., 2012). A new protocol will be the Virginia Technology Assessment Protocol (VTAP) that will be used to assess the removal of TSS and P by manufactured treatment systems (Sample et al., 2012). This new protocol is developed because TARP does not consider P, although P is regulated by Virginia’s stormwater management program.

In Australia (AUS), at least seven regulations of authorities exist, which are usually based on the removal in percentage of mean annual loads of solids, TPH, and nutrients (i.e., total nitrogen (TN) and total phosphor (TP)). Schmitt et al. (2011) summarized the criteria of six regulations. Only the criteria of one further and one updated program, which are similar to the ones reported by Schmitt et al. (2011), are described in the following: an approach that differentiates between different systems (primary, secondary, and tertiary treatment) is used in the state of Victoria (Victorian Stormwater Committee, 2006). For tertiary stormwater treatment systems (e.g., filter systems), a range of treatment performances has been defined for dissolved pollutants including Cu and Zn. For a moderate treatment performance, a removal of > 40% is postulated. In the development control plan of the city of Penrith, issues of the stormwater management are the flood control and the potential contaminants of stormwater runoff (Penrith, 2014). To safeguard receiving water by improving the quality of stormwater runoff, the following reductions of the post development mean annual loads are postulated: 90% of total gross pollutants (greater than 5 mm), 85% of TSS, 60% of TP, 45% of TN, and 90% of TPH. To calculate the annual loads and to prove these reductions, modeling with MUSIC (Model for Urban Stormwater Improvement Conceptualization) must be performed.

In summary, many different assessment methods and criteria are available, which sometimes also consider local requirements (e.g., nutrients). For the heavy metals, the effluent requirements of the different guidelines vary widely and the risk of remobilization of previously retained substances is only tested by three methods (DIBt, 2012; DIBt, 2015; ÖNORM B 2506-3 Entwurf, 2015) and exclusively for sodium chloride. Furthermore, there is currently no standardized procedure available by these assessment methods to determine quickly the filter material service lives and long-term performances of decentralized stormwater treatment systems.

The service life is defined as the point of time, when the filter material must be changed because of less efficient treatment. In general, two factors influence the determination of the service life of decentralized systems treating runoff from traffic areas: The service interval can be limited because of hydraulic failures of the plant (i.e., clogging of the filter material because of sediment accumulation and deposition) (Mercado et al., 2015). Otherwise, the service life can be limited because of the substance removal efficiency (e.g., a reduced retention of heavy metals). Clogging phenomena of the filter material can be simulated in the laboratory by using a full-scale system but realistic particle size distributions and site-specific factors of the system’s catchment area cannot be considered. Therefore, this factor needs to be determined by extensive and comparable field measurements that might take several years. In contrast, the terminal service life of the filter material (e.g., breakthrough of heavy metals) is rarely noticeable by the operator although this has a crucial effect on the receiving water. Although this effect can be simulated reproducibly at pilot-scale for decentralized systems, no assessment method is proposed by the authorities or manufacturers (cf. Section 2.2.2). The determination of the service life is also essential for a sustainable management of urban infrastructure because maintenance and the exchange of filter material have an effect on life-cycle costs of decentralized treatment systems (O'Sullivan et al., 2015).

2.2.2 Test Methods Used by Manufacturers

In addition to the procedures and methods proposed by the authorities, each manufacturer of decentralized stormwater treatment systems has developed different test methods to estimate maintenance intervals, service lives, and performances of their treatment systems. Moreover, internal and external quality control has to be conducted for each manufactured product (e.g., to prove the comparability of different production batches of the filter material). Most of these experiments are conducted in lab-scale.

Equilibrium batch experiments with different boundary conditions are used to determine sorption capacities of heavy metals (Dierkes, 2009) and to prove the comparability of different production batches (cf. Chapter 6). The service life of the filter material is often calculated by comparing these determined capacities (retention in g pollutant per kg filter material) and the total amount of filter material (kg) in the treatment system with the estimated pollutant loads of the connected catchment area. The determination of cation exchange capacities according to DIN EN ISO 11260:2011-09 (2011) is also used for the calculation of service lives and for quality control. However, none of these methods is suitable to determine realistic service lives and long-term performances (cf. Chapters 6 & 7). Several manufacturers also perform lab- and pilot-scale column experiments to determine removal efficiencies (Dierkes et al., 2013). Furthermore, some manufacturers have field-scale monitoring programs to prove the results of the lab-scale experiments and to determine maintenance intervals (e.g., Schriefer, 2013; Schriefer, 2014a; Schriefer, 2014b; Grontmij, 2015).

In summary, there are no detailed methods available for the determination of service lives, long-term performances, the effects of de-icing salts, and quality controls that also consider the effects of different experimental setups. Furthermore, the comparability of the results of different methods used by the manufacturers was not proven.

2.2.3 Determination of the Remobilization Risk under Application of De-icing Salts

Sodium chloride, calcium chloride, and magnesium chloride are widely used as de-icing salts for road maintenance (Stotz & Krauth, 1994; Marsalek, 2003; Bäckström et al., 2004; Nelson et al., 2009;

Bayerisches Landesamt für Umwelt, 2013). However, there is currently only one assessment method available that authorities have proposed to test the risk of remobilization for decentralized stormwater treatment systems and this method only evaluates the effect of sodium chloride on previously retained Cu and Zn (DIBt, 2012; DIBt, 2015; ÖNORM B 2506-3 Entwurf, 2015).

The monitoring results of systems treating the runoff from traffic areas highlight the importance to elucidate the heavy metal removal and remobilization risk under de-icing salt applications for all relevant de-icing salts and heavy metals. For example, during applications of sodium chloride, an increase in Cd, Cu, Ni, and Zn effluent concentrations was detected at a constructed wetland (Tromp et al., 2012). Thereby, the Cu effluent concentrations were higher than the maximum influent concentration. In addition, few studies have examined the remobilization behavior of heavy metals bound onto soils, which are used to treat runoff from traffic areas (Amrhein & Strong; 1990, Amrhein et al., 1992; Amrhein et al., 1993; Norrström & Jacks, 1998; Bäckström et al., 2004; Norrström, 2005). In most cases, a remobilization of heavy metals occurred during the application of different de-icing salts in cold seasons and the behavior of each metal differed.

Hence, heavy metal remobilization because of de-icing salt applications must be considered by assessment procedures for manufactured decentralized systems treating the runoff from traffic areas. A novel assessment method to evaluate the risk of remobilization of previously retained heavy metals must be developed to test these decentralized stormwater treatment systems. It is important to consider different heavy metals and de-icing salts because each combination behaves uniquely. For example, Nelson et al. (2009) tested two soils for the effects of sodium chloride and magnesium chloride on heavy metal mobility. Both soils showed different behaviors for each de-icing salt. The use of sodium chloride resulted in the largest release of Cu and Pb. Magnesium chloride released higher levels of Cd for both soils. Each soil showed different remobilization effects and all tested metals were released in different quantities.

2.3 Importance of this Research Topic

The rising importance of manufactured decentralized systems for runoff from traffic areas can be seen by the increasing number of approved systems in Germany (Figure 3). In each case, only the first approval is shown. For example, five of the systems were approved by the DIBt in the years 2006, 2009, and 2010. Four of the five systems received new approvals in the years 2011, 2014, and 2015 that are subsequently not shown for the years 2011, 2014, and 2015 in Figure 3. Because of the increasing number of systems that are available on the German market, the development and evaluation of an assessment method for decentralized stormwater treatment systems for traffic area runoff, which determines the service life and the effect of de-icing salt applications, is currently important to prevent negative effects on receiving waters.

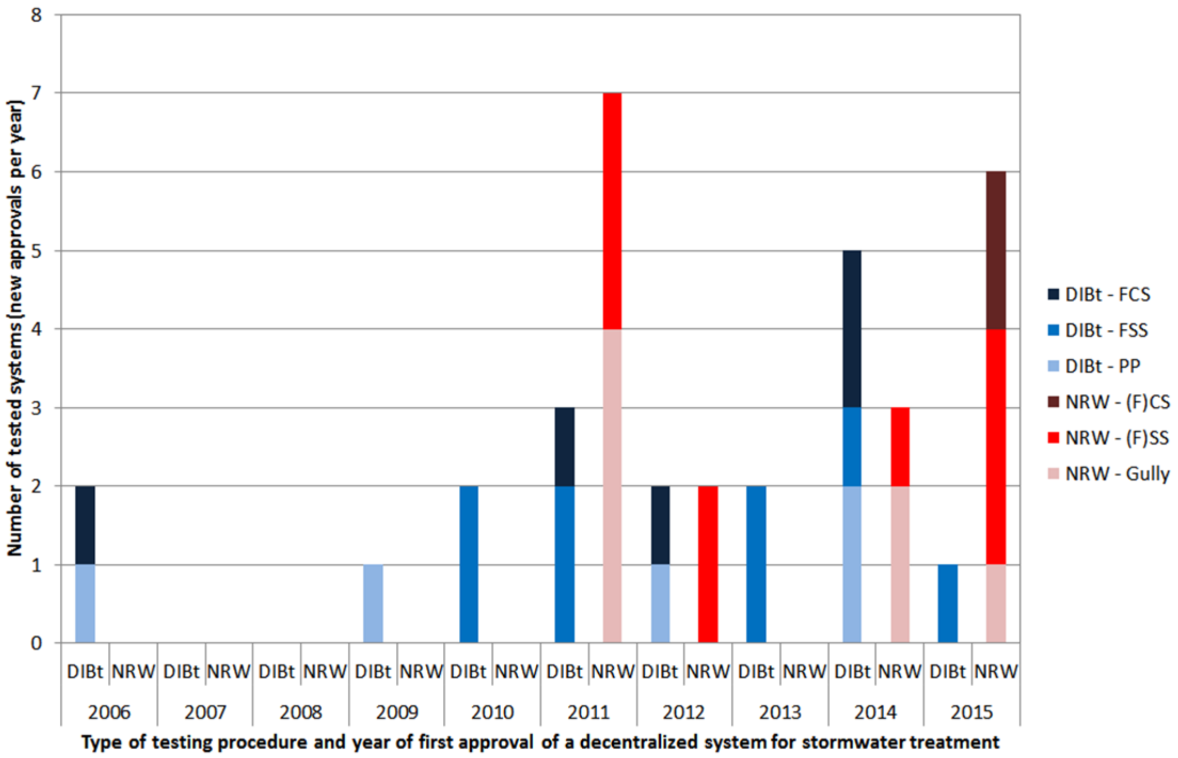

Figure 3: Approved decentralized stormwater treatment systems; data based on the websites DIBt (2016) and LANUV (2016) (01 March 2016). The systems are categorized as Filter Channel Systems (FCS), Filter Shaft Systems (FSS), Permeable Pavements (PP), Channel Systems without a filter (CS), Shaft Systems without a filter (SS), and systems installed in gully pots (Gully).

By using appropriate stormwater treatment systems, large amounts of the traffic and background emissions can be retained. The traffic emission loads of Hillenbrand et al. (2005) were used for a theoretical estimation of the reduction of heavy metals by stormwater treatment systems due to a lack of information concerning the area and pollution of all types of urban roads and parking lots. Hillenbrand et al. (2005) proposed that 26.5% of the traffic related emissions are produced on urban traffic areas. As manufactured stormwater treatment systems are mostly used for urban stormwater runoff, a theoretical heavy metal reduction load was calculated for urban traffic emissions (Figure 4). It was assumed that the particulate fractions of Cu, Pb, and Zn are 65%, 100%, and 55%, respectively. The reductions of these pollutants by the treatment systems were estimated to be 92% for all three particulate fractions, 80% for dissolved Cu, and 70% for dissolved Zn. The percentages of removal were adapted from the procedures proposed by DIBt (2015) and it is very ambitious to treat all urban stormwater runoff from traffic areas with these efficiencies. Thus, the loads presented in Figure 4 give only an indication of possible metal reductions and emphasize the use of stormwater treatment systems.

A reduction of the urban traffic emissions by approximately 10% might also be possible by street sweeping (Hillenbrand et al., 2005). Because more and more cars, signs, and guardrails exist, the metal loads of Cu and Zn are potentially increasing (cf. Figure 1) and the retained loads will also increase.

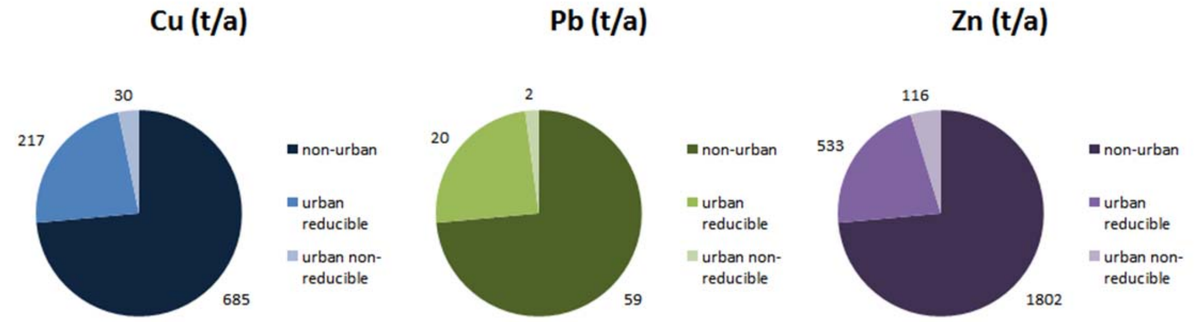

Figure 4: Estimation of heavy metal emissions in Germany and theoretical reduction of heavy metals by urban stormwater treatment systems (traffic related emission loads adapted from Hillenbrand et al. (2005)).

Chapter 3: Need for Action

Two aspects are important for the assessment of the relevance of treating individual substances in stormwater runoff from traffic areas. First, it must be determined, which parameters can have an eco-toxicological or human-toxicological effect. In general, these substances are implemented by the legislative requirements as thresholds to prevent negative impacts on receiving waters (cf. Section 1.3). Secondly, these parameters must occur in relevant concentrations in the runoff of traffic areas (cf. Section 1.1 and Chapter 5). Consequently, a treatment of traffic area runoff waters is necessary, if the threshold values of the corresponding ordinances are lower or comparable with the concentrations measured in monitoring programs.

The most relevant inorganic parameters of traffic areas are the four heavy metals Cd, Cu, Pb, and Zn, as well as the de-icing salt compound Cl. In addition, the metals Cr and Ni can occur in higher runoff concentrations compared with the threshold values for infiltration into the soil and groundwater system (Huber et al., 2015c; Welker, 2014). Thus, a treatment of the runoff from traffic areas is currently necessary in most cases because of contamination with heavy metals. An appropriate treatment of the runoff can be achieved with manufactured decentralized stormwater treatment systems (cf. Section 2.1). Because no procedure was available for the determination of service lives, long-term performances, and the remobilization behavior of previously retained substances, the main objective of this thesis was set on the development and evaluation of an assessment method for decentralized stormwater treatment systems for traffic area runoff. This method must determine the removal of relevant dissolved substances (i.e., heavy metals) and their risk to be remobilized by all relevant de-icing salts to avoid negative ecological and economic impacts on receiving water. The focus was set on the decentralized systems with filter materials because they are needed to retain dissolved metals.

For the development of the assessment method, the occurrence, influencing factors, and partitioning of all relevant substances must be identified. Thus, a literature study was performed to evaluate the partitioning and occurrence of heavy metals, alkali metals, and alkaline earth metals in traffic area runoff for the first time. It also included the site-specific (both fixed and climatic) and method-specific factors influencing runoff concentrations (cf. Chapters 5 & 8). The distributions of the heavy metals (both runoff concentrations and loads for different traffic area categories) are presented in three reviews. The two hypotheses of these papers are as follows:

The average metal runoff concentrations and their distributions can be determined by a literature study of monitoring programs whereupon the mean and median concentrations and the loads of each site are not comparable (Hypothesis 1).

Although the measured heavy metal pollution is influenced by site-specific and method-specific factors, the average total and dissolved metal concentrations vary significantly between different traffic area categories for a large collection of monitoring sites (Hypothesis 2).

Based on the determination of the dissolved heavy metal concentrations, an assessment method was developed to evaluate the service lives and long-term performances of decentralized systems treating traffic area runoff because such a method was not previously available (cf. Section 2.2). Subsequently, several tests were performed with filter materials for stormwater treatment in lab-, pilot-, and full-scale by novel standardized methods that can also be used for further applications such as quality control (cf. Chapters 6 & 7). The objectives of the three papers presenting the evaluation of the heavy metal removal performances were related to these hypotheses:

The results of batch experiments and lab-scale column experiments concerning simultaneous heavy metal removal differ widely because of varying boundary conditions (Hypothesis 3).

A novel test method for determining the filter material service lives and long-term performances can be developed and implemented to evaluate new and already existing decentralized systems (Hypothesis 4).

A validation of the determined service lives of channel systems by field measurements confirms the applicability of the developed method (Hypothesis 5).

Concerning the composition of de-icing salts, in most cases only Cl was monitored and the corresponding cations were often neglected. Because different de-icing salt compounds can have a different effect on the removal efficiencies of decentralized stormwater treatment systems for traffic area runoff, their use in road maintenance and their occurrence in the runoff from traffic areas must be subsequently evaluated (cf. Chapter 8). The hypothesis of this part of the thesis is:

Different types and amounts of de-icing salts are used in road maintenance by German authorities (Hypothesis 6).

After determining the relevance of three different de-icing salts in German traffic area runoff, a further aspect was implemented in the assessment method. Therefore, the heavy metal removal mechanisms and the risk of remobilization under de-icing salt applications were evaluated for five heavy metals to develop a complete assessment method for manufactured decentralized stormwater treatment systems for runoff from traffic areas (cf. Chapter 9). The corresponding hypothesis is:

The composition of the de-icing salt solution has a significant effect on the remobilization of heavy metals and the behavior of each heavy metal differs widely (Hypothesis 7).

Chapter 4: Methodology

The methods used in this thesis consist of literature studies, computer modeling, lab-scale, pilot-scale, and full-scale experiments (Figure 5). For each research topic, one or a combination of different methods was used. The research topics are also listed in Figure 5 with the corresponding numbers of the chapters of this thesis (left), the hypotheses (right), and the publications (peer reviewed journal papers, conference proceedings, and the monograph).

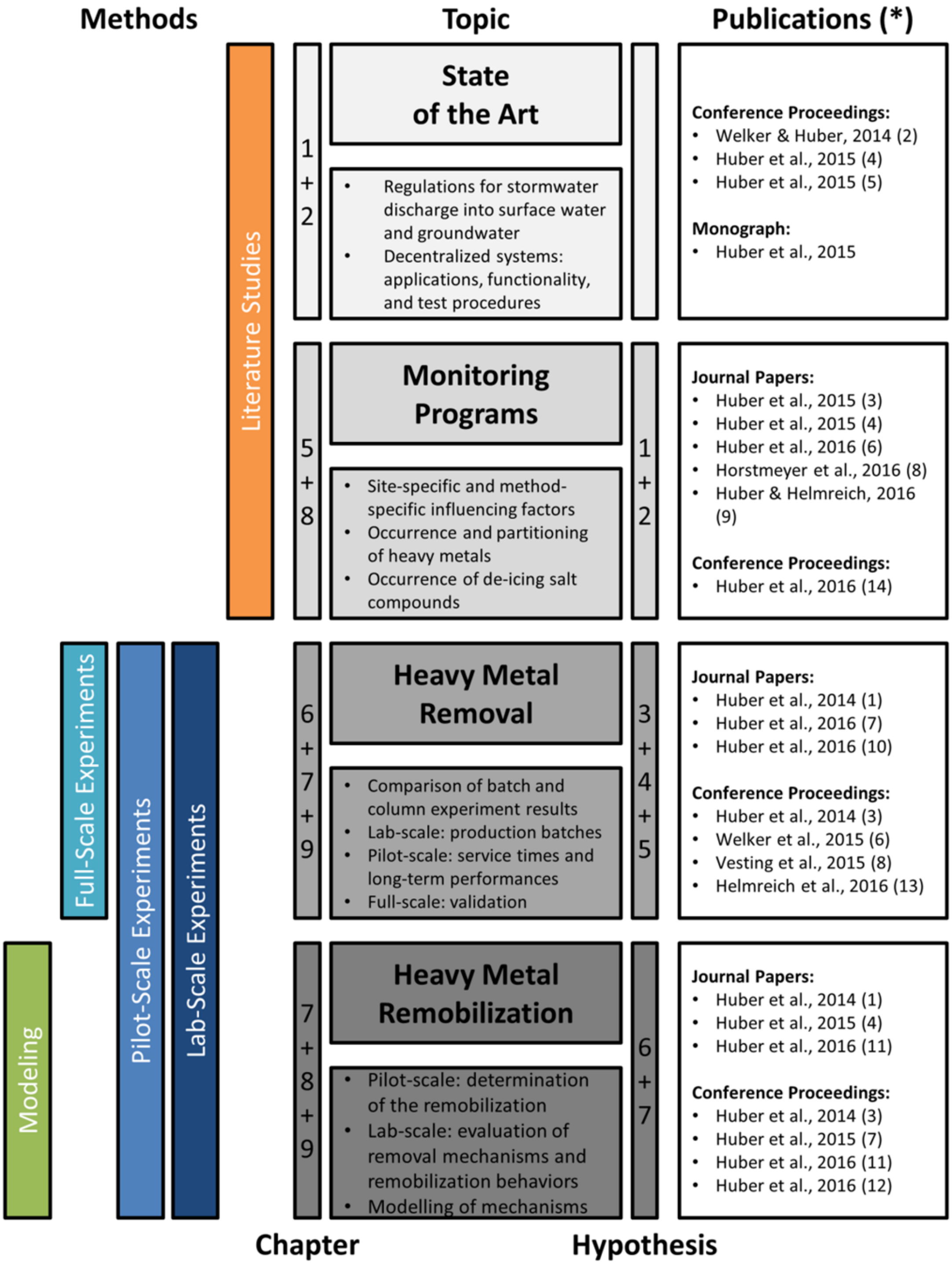

Figure 5: Methodology for the evaluation of decentralized treatment systems for traffic area runoff used for this thesis. (*) Number in brackets refers to the publication number of the publications list in the Appendix.

The literature studies were used to collect data from other monitoring and research programs that were published in peer-reviewed journal papers, reports, books, and non-reviewed articles (after validation). The data from the monitoring programs consist of substance concentrations in traffic area runoff and additional information concerning the monitoring program (e.g., fixed site-specific and climatic-specific influences; objectives, special conditions, and remarks; sample collection, preparation, analysis, and calculation methods). In addition, the annual heavy metal loads normalized per hectare of impervious catchment area were summarized and used for mass balances. In another literature study, information on the application, functionality, removal mechanisms, types of construction, test procedures, and assessment criteria were collected concerning stormwater treatment systems for runoff from traffic areas.

The lab-scale experiments consisted of batch and column experiments. Batch experiments with heavy metals were performed under identical and well-defined conditions to prove the comparability of different production batches and to select filter materials for subsequent column experiments. Batch equilibrium experiments, batch kinetic experiments, and experiments to determine cation exchange capacities were also conducted to determine heavy metal removals by filter materials for stormwater treatment and to evaluate factors influencing lab-scale studies. Column experiments were used as an indicator to determine the efficiencies and service lives of decentralized stormwater treatment systems. Lab-scale column experiments were also performed to study the remobilization of heavy metals during the application of three different de-icing salts.

To determine service lives and characterize long-term performances, pilot-scale experiments were performed on nine different filter systems and the retention of dissolved heavy metals was evaluated. The novel assessment method, which was developed for these pilot-scale experiments, consists of several steps including preloading the filter material in individual models with Cu and Zn by a load of n-1 years of the estimated service life (n). Subsequently, three representative rain events were simulated to evaluate the long-term performance by dissolved Cu and Zn during the last year of application. In addition, solid samples were taken at different full-scale treatment systems. Furthermore, two full-scale treatment plants were operated in NRW for monitoring their removal efficiencies and the pollution of road runoff.

To support the findings of the lab-scale column experiments concerning the removal and remobilization of heavy metals, computer modeling was used to simulate the speciation of heavy metals during removal and under application of different de-icing salts.

Detailed descriptions of the materials and methods that were used in this thesis are given in the “Material & Methods” Chapters of the following eight papers.

Chapter 5: Heavy Metal Pollution of Traffic Area Runoff

The results of the analysis of monitoring programs were published in three reviews. For the runoff concentrations, one review presents the results of the complete data set (international review, cf. Section 5.1) and one review presents the European data (cf. Section 5.2). The metal loads and the traffic related heavy metal emissions are summarized in the third review (cf. Section 5.3). The first two reviews deal with the following two hypotheses and the third review only with Hypothesis 1:

The average metal runoff concentrations and their distributions can be determined by a literature study of monitoring programs whereupon the mean and median concentrations and the loads of each site are not comparable (Hypothesis 1).

Although the measured heavy metal pollution is influenced by site-specific and method-specific factors, the average total and dissolved metal concentrations vary significantly between different traffic area categories for a large collection of monitoring sites (Hypothesis 2).

The reviews consider different aspects of the occurrence, influences, and partitioning of heavy metals. Because of less European data, the second review distinguishes between only five traffic area categories (instead of eight for the international review) for the metal distributions of Cd, Cr, Cu, Ni, Pb, and Zn. The mass balances published in the third review consider annual metal runoff loads and traffic related emission loads.

5.1 Review Paper: "Critical Review of Heavy Metal Pollution of Traffic Area Runoff: Occurrence, Influencing Factors, and Partitioning"

The first journal paper deals with the concentrations of relevant heavy metals in the runoff of eight different types of traffic areas (i.e., parking lots, bridges, roads with an AADT of < 5000, roads with 5000 < AADT < 15,000, roads with an AADT of > 15,000, urban highways with an AADT of > 30,000, non-urban highways with an AADT of > 30,000, and highways with an AADT of < 30,000). The data collected for the first review are based on the international literature study.

This review focuses on the objectives as follows: site-specific factors (both fixed and climatic) that influence the occurrence and partitioning of the heavy metals in traffic area runoff from different sites and that should be documented in monitoring programs were described in detail. The conditions of the monitoring methods, including sample collection, sample preparation, sample analysis, and calculation methods, were compiled and evaluated. The concentrations and fractionation of the heavy metals were summarized to produce a comprehensive data set of international monitoring programs, to characterize eight types of traffic areas, and to identify relevant heavy metals.

The following heavy metals from 294 sites were considered for the distribution analysis: Cd, Cr, Cu, Ni, Pb, and Zn. For Pb and Zn, the historical concentration trends were evaluated and only a decrease in the Pb concentrations was detected. Thus, only Pb data of the 21st century were used for the characterization of the different traffic areas. It was possible to determine the distributions of the total and dissolved concentrations of all six heavy metals. Their occurrence differed widely between the eight traffic area categories. In addition, the mean and median concentrations that were both available for each site were not comparable. Furthermore, the runoff concentrations of antimony, cobalt, manganese, platinum-group elements (PGE), titanium, and tungsten were considered. Tendencies such as seasonal effects, phase-out measures, and land use influences, which should be considered for planning stormwater treatment plants, were identified. In addition, the variability of Zn in stormwater runoff from traffic areas and continental trends were discussed. No link between lower pH values and a higher proportion of dissolved metals in the runoff was detected.

The results were published in Science of the Total Environment 541, 895–919. The basis for the database was set by the habilitation of Welker (2004). Huber collected most of the data, evaluated all documents, and wrote the review. Both co-authors contributed to the discussions and conclusions of the first review.

5.2 Review Paper: "Heavy Metal Pollution of Traffic Area Runoff – a European Review"

The second journal paper summarizes the concentrations of relevant heavy metals in the runoff of five different types of traffic areas (i.e., parking lots, highways, roads with an AADT of < 5000, roads with 5000 < AADT < 15,000, and roads with an AADT of > 15,000). The data of this review are based on the database of the international literature study (cf. Section 5.1) and they only include European runoff concentrations.

This paper focuses on the objectives as follows: site-specific factors (both fixed and climatic) that influence the occurrence and partitioning of the heavy metals in traffic area runoff from different sites were summarized. In addition, the influences of method-specific factors were compiled. The concentrations and fractionation of the heavy metals were summarized to produce a comprehensive data set, to characterize five types of traffic areas, and to identify relevant heavy metals that must be evaluated in assessment methods for decentralized stormwater treatment systems.

The concentrations and fractionation of Cd, Cr, Cu, Ni, Pb, and Zn were presented in detail. For Cu, Pb, and Zn, the historical concentration trends were evaluated and a trend was only detected for Pb. Thus, only Pb data of the 21st century were used for the characterization of the different traffic areas. The distributions of all six heavy metals differed widely between the five traffic area categories. The average total and dissolved metal concentrations varied significantly because of site-specific and method-specific factors and the concentrations of each site were not comparable. Furthermore, the maximum runoff concentrations of antimony, barium, cobalt, manganese, palladium, platinum, titanium, and tungsten were summarized. Continental trends for total Cu and Zn concentrations were also analyzed. In addition, the highway runoff concentrations were evaluated and discussed for each European country with available monitoring data. In conclusion, the categories of pollution for different traffic areas proposed by DWA-M 153E (2007) (cf. Table 2) were roughly verified and recommendations for further monitoring programs were summarized.

The second review was published in gwf Wasser/Abwasser 156, 896–909. The basis for the database was set by the review paper of Section 5.1. Huber collected most of the data, evaluated all documents, and wrote the paper. Both co-authors contributed to the discussions and conclusions.

5.3 Review Paper: "Stormwater Management: Calculation of Traffic Area Runoff Loads and Traffic Related Emissions"

In this third review, the annual metal runoff loads for different traffic areas (45 sites) were summarized on the basis of catchment areas (g/ha) for the metals Cd, Cr, Cu, Ni, Sb, Pb, and Zn. Further objectives were: to identify relevant trends and factors, to update and expand existing mass balances (heavy metal runoff loads and emissions) for Germany, and to characterize the mass fluxes of traffic related metals. Of these parameters, the annual traffic area runoff loads per hectare of catchment area were mostly published by monitoring programs for the three heavy metals Cu, Pb, and Zn. A historical trend for Pb was detected and the variations of the Zn loads were the highest compared with the other heavy metals because of site-specific factors. In addition, the annual loads were calculated at three monitoring sites for several years and the variations of these loads were evaluated for each year to determine the correlations of climatic site-specific factors with the runoff loads. For the heavy metal runoff loads, a calculation of average values was possible for highways and for all traffic areas whereupon the average loads of each site and traffic area category differed widely. The calculated average annual loads were used to perform mass balances for Germany to identify the total pollution loads of runoff from highways and all traffic areas. In addition, the traffic related emissions were calculated for Cd, Cu, Pb, and Zn for seven different sources (tire wear, brake lining wear, roadway abrasion, weights for tire balance, guardrails, lampposts/signs, and de-icing salts). However, an evaluation of the mass fluxes by a statistical analysis was not possible for the metals due to a lack of monitoring data.

This review was published in Water 8 (7), 294:1–21. Maximilian Huber collected the data, evaluated all documents, and wrote the paper. Brigitte Helmreich contributed to the results and discussions.

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Review

Critical review of heavy metal pollution of traffic area runoff: Occurrence, influencing factors, and partitioning

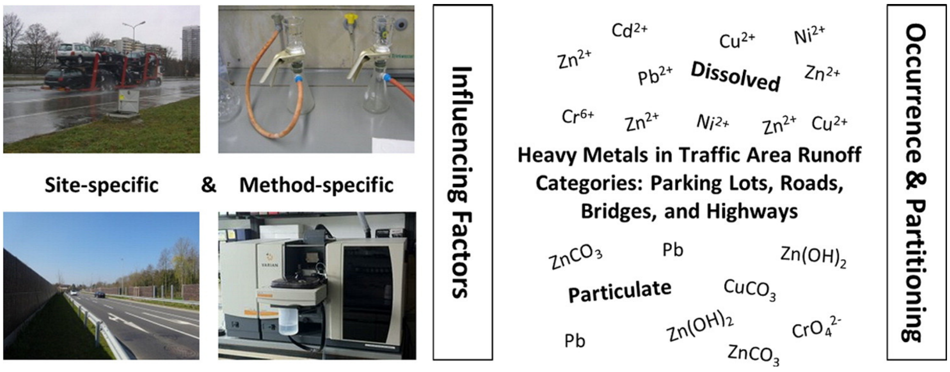
Maximilian Huber [a], Antje Welker [b], Brigitte Helmreich [a,*]

[a] Chair of Urban Water Systems Engineering, Technical University of Munich, Am Coulombwall 8, 85748 Garching, Germany
[b] Fachgebiet Siedlungswasserwirtschaft und Hydromechanik, Frankfurt University of Applied Sciences, Nibelungenplatz 1, 60318 Frankfurt am Main, Germany

HIGHLIGHTS

- Heavy metal runoff concentrations in parking lots, bridges, and roads
- Large dataset of dissolved and total metal concentrations in traffic area runoff
- Description of site-specific and monitoring method-specific factors
- Summary of traffic-related and anthropogenic heavy metals in road runoff
- Reduction in Pb concentrations over time is one of the robust chemical results.

GRAPHICAL ABSTRACT



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ABSTRACT

A dataset of 294 monitored sites from six continents (Africa, Asia, Australia, Europe, North and South America) was compiled and evaluated to characterize the occurrence and fate of heavy metals in eight traffic area categories (parking lots, bridges, and three types each of both roads and highways). In addition, site-specific (fixed and climatic) and method-specific (related to sample collection, preparation, and analysis) factors that influence the results of the studies are summarized. These factors should be considered in site descriptions, conducting monitoring programs, and implementing a database for further research. Historical trends for Pb show a sharp decrease during recent decades, and the median total Pb concentrations of the 21st century for North America and Europe are approximately 15 μg/L. No historical trend is detected for Zn. Zn concentrations are very variable in traffic area runoff compared with other heavy metals because of its presence in galvanized structures and crumbs of car tire rubber. Heavy metal runoff concentrations of parking lots differ widely according to their use (e.g., employee, supermarket, rest areas for trucks). Bridge deck runoff can contain high Zn concentrations from safety fences and galvanizing elements. Roads with more than 5000 vehicles per day are often more polluted than highways because of other site-specific factors such as traffic signals. Four relevant heavy metals (Zn, Cu,

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Abbreviations: AADT, average annual daily traffic; BR, bridge; d, dissolved; HL, highway with low AADT (<30,000); HN, highway with high AADT (>30,000) and non-urban land use; HU, highway with high AADT (>30,000) and urban land use; HWY, highway; p, particulate; Pb_21, dataset including only Pb concentrations measured in the 21st century; PL, parking lot; RL, road with low AADT (<5000); RM, road with medium AADT (5000 < AADT < 15,000); RU, road with high AADT (>15,000); t, total.

* Corresponding author.
E-mail address: b.helmreich@tum.de (B. Helmreich).

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Ni, and Cd) can occur in the dissolved phase. Knowledge of metal partitioning is important to optimize stormwater treatment strategies and prevent toxic effects to organisms in receiving waters.

Contents

1. Introduction

Traffic area runoff summarize precipitation- and snowmelt-related discharges of mostly impervious surfaces (sidewalks, parking lots (PL), feeder streets, major roads, and highways (HWY)). The main contamination sources of traffic area runoff are related to traffic, surrounding land use, atmospheric contamination, and other meteorological and environmental conditions (Muschack, 1990; Ball, 2002; Crabtree et al., 2009; Valtanen et al., 2014). It is difficult to determine the dominant sources of pollutants because most substances have more than one origin and the water quality data of runoff from different sites are extremely heterogeneous because of differing background levels, types of uses (Göbel et al., 2007), and method-specific factors.

The substance spectrum analyzed in traffic area runoff waters includes organic parameters such as polycyclic aromatic hydrocarbons, total petroleum hydrocarbons, chemical oxygen demand, biological oxygen demand, and total organic carbon; heavy metals such as Pb, Zn, and Cu; and materials from de-icing salts such as chloride (Smullen et al., 1999; Moy et al., 2003; Eriksson et al., 2007; Kayhanian et al., 2012).

As HWY account for a small percentage of urban land use, they contribute only a small portion of pollutant loads compared with other road surfaces (Shelley et al., 1987). In urban catchments, all road surfaces represent approximately 10%–15% of the total area (Bannerman et al., 1993; Ball, 2002), and in commercial and industrial areas, PL can constitute up to 46% of the total area (Bannerman et al., 1993). Therefore, it is essential to consider all types of traffic area runoff.

In most cases, runoff waters from PL and road surfaces contain higher levels of the heavy metals than other types of runoff in drainage systems such as conventional roof runoff (Schueler, 2000; Ball, 2002). Metals in roof and road runoff contribute up to 80% of the total mass flow in combined sewer systems (Ellis et al., 1987; Boller, 1997). The present review focuses on all heavy metals in traffic area runoff that have either traffic or anthropogenic sources. These metals are transported by stormwater runoff either attached to solids or in dissolved form depending on the prevailing redox and pH conditions (Ball, 2002). However, in most cases, only total metal concentrations are analyzed from runoff waters during measurement campaigns.

Some of these heavy metals can have acute or chronic impacts as a result of their accumulation in receiving waters in terms of aquatic habitats, drinking water resources, and recreational uses (Ellis and Revitt, 1982; Yousef et al., 1984). For potential toxic effects, the partitioning between the total and dissolved heavy metals is essential because the dissolved fractions are directly biologically available (Paulson and Amy, 1993; Crabtree et al., 2008). The toxicity of traffic area runoff has been investigated by various researchers (Gjessing et al., 1984; Pitt et al., 1995; McQueen et al., 2010). A particular link to the heavy metals was made by Tiefenthaler et al. (2001), who identified trace metals (particularly Zn) as important contributors to toxicity in PL runoff, and by Kayhanian et al. (2008), who identified dissolved Cu and Zn as the primary causes of toxicity in HWY runoff. In general, the toxicity of heavy metals is a function of several factors such as metal speciation and physical characteristics of receiving waters (Revitt and Morrison, 1987). If conditions change, particulate metals transported into receiving waters have the potential to repartition into the dissolved phase (Sansalone, 2002; Westerlund and Viklander, 2006). Metal partitioning is also important for designing appropriate stormwater treatment strategies (Hilliges et al., 2013; Maniquiz-Redillas and Kim, 2014).

The present review focuses on the objectives as follows:

- To describe site-specific factors (both fixed and climatic conditions) that influence the occurrence and partitioning of the heavy metals in traffic area runoff from different sites and that should be documented in investigation programs.
- To compile and evaluate the conditions of the monitoring methods, including sample collection, sample preparation, sample analysis, and calculation methods.
- To summarize the concentrations and fractionation of the heavy metals to produce a comprehensive dataset, to characterize different types of traffic areas, and to identify relevant heavy metals.

- To identify tendencies such as seasonal effects, phase-out measures, and land use influences that should be considered for planning stormwater treatment plants.

2. Material and methods

A dataset based on nearly 300 studies (n = 294) was analyzed. In addition to peer-reviewed journal papers, reports, books, and non-reviewed journal articles were considered after validation that presented heavy metal concentrations in traffic area runoff, including additional data concerning the monitoring program.

Several researchers evaluated street sweeping, the performance of grass swales, wetlands, detention ponds, biofiltration systems, porous asphalt surfaces, and artificial treatment plants. Only some characterized the quantity and quality of traffic area runoff containing heavy metal partitioning and influencing factors as their main objective. Therefore, the details of each monitoring program presented in publications differed and information regarding influencing factors such as those described in Section 3.1 are often missing.

To analyze historical trends and obtain a wide range of data, it is important to consider publications of the last decades. In North America and Europe, a large number of runoff data have been collected since the 1970s. Since the 1990s, data from Australia and New Zealand have been available, and since the 2000s, the number of publications from Asia, Africa, and South America has increased.

The date of the monitoring program was used instead of the publication date. If the date of the monitoring program was not available, the date of paper submission was used. All data are subdivided into eight categories: PL; bridges (BR); roads with an average annual daily traffic (AADT) of <5000 (RL), 5000 < AADT < 15,000 (RM), and >15,000 (RU); urban HWY with an AADT of >30,000 (HU); non-urban HWY with an AADT of >30,000 (HN); and HWY with an AADT of <30,000 (HL). If the AADT was not mentioned, the classification was performed from the description of the monitoring site, e.g., a feeder street was classified as category RL. Each site was linked to one category. The identification (ID) of each site consists of two capital letters according to the category and two consecutive numbers, e.g., PL01. These IDs are used in Tables 1, 2, and 3 and in Sections 4.3 and 4.4.

One important step was to ensure that each dataset from one site was only considered once, although it might have been published twice or more in different forms. All data of one site was recalculated to obtain one dataset per site, except where the investigation period was not continuous (e.g., a new monitoring program after several years) or site-specific factors varied because of construction work (e.g., new road surface). In these cases, one site provides two or three datasets (highlighted in Tables 1, 2, and 3 by adding a letter a, b, or c at the end of an ID).

At data collection, total (t), dissolved (d), and particulate (p) concentrations were distinguished, as were mean and median concentrations. If authors presented both mean and median values, both values were used in this review. For sites including all single-event concentrations, both mean and median concentrations were calculated. Some extreme measured concentrations were deleted for plotting the figures but all these concentrations are mentioned in Section 4.

For statistical analysis and plotting histograms and box plots, the software package SPSS 22 (IBM) was used. For box and whisker plots, the bottom and top of each box are the first and third quartiles and the band inside the box is the median. The whiskers represent 1.5 times the interquartile range (IQR). Outliers (>1.5 times IQR) are marked as small circles and extreme values (>3.0 times IQR) as stars.

3. Occurrence and influencing factors

Literature datasets have a wide variation due to site-specific and monitoring-method-specific factors (Fig. 1). Site-specific factors can be divided into fixed site-specific factors (spatial variance) and climatic site-specific factors (temporal variance); both affect the real pollution of surface runoff. Method-specific influences on the results, which should represent the real situation, are caused by the boundary conditions of a measurement program and occur during sample collection, sample preparation, sample analysis, and calculation.

3.1. Site-specific factors

3.1.1. Fixed site

Fixed site-specific factors can be divided into three categories: surrounding land use characteristics, traffic area site data, and operational characteristics (Shelley et al., 1987).

For the first factor, i.e., *surrounding land use characteristics*, several categories are used: non-urban, urban, ultra-urban, transportation, residential, open, agriculture, commercial, mixed, and industrial categories (Kayhanian et al., 2003; Flint and Davis, 2007). Driscoll et al. (1990) specified urban (undefined, commercial/residential, residential, and suburban) and non-urban (undefined rural, forest, undeveloped, agricultural, and desert) land use categories. Because the term residential land use could include a mixture of streets, driveways, rooftops, and lawns (Schueler, 2000), its use for analysis is restrictive and only comparisons of land use categories analyzed in one research program can provide an indication of their influence. E.g., the results of Dannecker et al. (1990) showed similar runoff pollution levels of an industrial road and a main street of an inner urban city.

The second factor, i.e., the *traffic area site data*, contains information about vegetation, topography, and road design. Only some data are available for surrounding *vegetation*, street surface covered with trees, and *topography*, which describes the slope and the configuration of the traffic area (cut, at grade, or elevated (Driscoll et al., 1990)). Jenewein and Schinner (1982) did not find a correlation of the slope with runoff concentrations. Most fixed site-specific data are available about *road design*, including information about cross-sections, pavement surfaces, and drainage areas. The cross-section describes the number of lanes per direction and the existence of curbs, median strips, hard shoulders, drainage systems, crash/noise barriers, and safety fences. The absence of curbs has a positive effect on reducing runoff concentrations (Driscoll et al., 1990), and further influences are described by Irish et al. (1998) and Li et al. (2008). Hard shoulders have an influence on water quality because they act as a sink for suspended solids and particle-associated metals (Berbee et al., 1999). Detailed information about the types of surfaces drained and sampled in monitoring programs is available in literature (62.9% of the datasets). Most surfaces are conventional asphalt surfaces (n = 122), followed by concrete (n = 33), asphalt and concrete (n = 15; mostly HWY with new lanes), porous asphalt (n = 9), asphalt and paver (n = 3; PL), paver (n = 2; PL), and crushed stone (n = 1; feeder street). Drapper et al. (2000) concluded that a concrete surface does not have a significant impact on pollutant runoff concentrations compared with asphalt surfaces. The surface condition has an effect on concentrations by either increasing runoff loads from decomposition products or reducing them during filtration into cracks and pot-holes (Driscoll et al., 1990). The size of the drainage area in the dataset of this study varies widely (3–429,000 m^2) and has an effect on the first flush (Lau et al., 2009) and metal concentration, when the ratio of the drainage area to traffic volume is considered (CH2MHILL, 1998). The same applies to the imperviousness of the surface. Noise barriers lead to an increase in runoff concentrations (Dierkes, 1999; Kocher et al., 2010a,b), and concrete barriers can retain more solids on road surfaces (Barrett et al., 1998). Speed limits and traffic signals have an influence on runoff concentrations because braking and acceleration activities lead to increased abrasion of tires, higher use of brake linings, and increased automotive exhaust gas emissions (Muschak, 1990; Langbein et al., 2006). Drapper et al. (2000) measured higher concentrations of Cu and Zn at locations with exit lanes due to sharp braking by exiting vehicles and road signs release metals as a result of rain and splashing water (Van Bohemen and Van de Laak, 2003).

Table 1
Register of studies investigating parking lot runoff.

ID	Literature	Location	Sampling period	Special conditions[a]	Discharge area[m^2]	Type and characteristics of parking lots	Number of events sampled	Type of sample collection	Sampling strategy	Pore diameter for filtration	Partition[b]	Metals detected[c]
PL01	Grotehusmann and Kasting (2002)	DEU	2001-01–2001-12	D, M, S	10,000	Tank farm and rest area for 41 trucks and four busses	40	Active-automatic	Flow-prop.	0.45	d, t	Cd, Cr, Cu, Hg, Pb, Pd, Pt, Zn
PL02	Grotehusmann and Kasting (2002)	DEU	2001-01–2001-12	D, M, S	5000	Tank farm and rest area for 122 cars	43	Active-automatic	Flow-prop.	0.45	d, t	Cd, Cr, Cu, Hg, Pb, Pd, Pt, Zn
PL03	Grotehusmann and Kasting (2002)	DEU	2001-01–2001-12	D, M, S	17,700	Tank farm and rest area for 18 trucks, 16 busses and 78 cars	42	Active-automatic	Flow-prop.	0.45	d, t	Cd, Cr, Cu, Hg, Pb, Pd, Pt, Zn
PL04	Pick et al. (2002)	DEU	1999-03–2000-11	–	300	Campus parking lot	–	Active-automatic	Mixed	–	t	*Cd (<1.0)*, Cr, Cu, Ni, Pb, Zn
PL05	Engelhard et al. (2012)	AUT	2010-04-02–2011-02-21	D	–	Supermarket	–	Passive-automatic	Mixed	–	t	Cd, Cr, Cu, Ni, Pb, Zn
PL06	Gromaire-Mertz et al. (1999)	FRA	1996-07–1997-05	–	–	Courtyard, old residential district with small businesses	7	Active-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Pb, Zn
PL07	Gnecco et al. (2005)	ITA	2002-01-23–2003-09-28	–	1000	Campus parking lot, typical urban residential area	9	Active-automatic	Time-prop. (equally timed)	–	d	Cu, Pb, Zn
PL08	Gnecco et al. (2008)	ITA	2005-11-15–2006-07-12	–	14,000	Airport parking area with 100 vehicles per hour	4	Active-automatic	Time-prop. (equally timed)	0.45	d, p	Al, Cu, Fe, Mn, Pb, Zn
PL09	Nowakowska-Blaszczyk and Zakrzewski (1996)	POL	–	S	–	–	–	–	–	–	t	Pb
PL10	Hogland et al. (1987)	SWE	1985	–	470	Research center	2	Active-automatic	Time-prop. (equally timed)	–	t	Al, Cd, Cr, Cu, Pb, Zn
PL11	Wei and Morrison (1994)	SWE	1992-05-26	–	–	Park and ride	1	–	–	0.45	d, t	Pt
PL12	Pitt et al. (1995)	USA	–	–	–	Park and ride	1	Manual	Random	0.45	d, t	Al, Cd, Cr, Cu, Ni, Pb, Zn
PL13	Caltrans (2003)	USA	2002-11-07–2003-04-24	–	–	Park and ride, commercial site within the foothills	8	Active-automatic	Flow-prop.	–	d, t	As, *Cd (<0.2)*, Cr, Cu, Ni, Pb, Zn
PL14	Caltrans (2003)	USA	2002-12-09–2003-02-12	–	–	Park and ride	8	Active-automatic	Flow-prop.	–	d, t	As, Cd, Cr, Cu, Hg, Ni, Pb, Zn
PL15	Caltrans (2003)	USA	2002-11-07–2003-02-15	–	–	Park and ride	8	Active-automatic	Flow-prop.	–	d, t	As, Cd, Cr, Cu, Hg, Ni, Pb, Zn
PL16a	Caltrans (2003)	USA	2002-11-29–2003-05-02	–	–	Park and ride, campus parking lot	8	Active-automatic	Flow-prop.	–	d, t	As, Cd, Cr, Cu, Ni, Pb, Zn
PL16b	Tiefenthaler et al. (2001)	USA	2000-07-15–2000-10-07	S	608	Park and ride, campus parking lot	5	Active-automatic	Time-prop. (equally timed)	0.45	d, t	Al, Cd, Cr, Cu, Fe, Ni, Pb, Zn
PL17	Caltrans (2003)	USA	2002-11-07–2003-04-14	–	–	Park and ride	8	Active-automatic	Flow-prop.	–	d, t	As, Cd, Cr, Cu, Ni, Pb, Zn
PL18	Caltrans (2003)	USA	2002-12-20–2003-04-14	–	–	Park and ride, rural area near foothills	4	Active-automatic	Flow-prop.	–	d, t	Cd, Cr, Cu, Ni, Pb, Zn
PL19	Caltrans (2003)	USA	2002-11-07–2003-04-12	–	–	Park and ride, flat business/retail area	8	Active-automatic	Flow-prop.	–	d, t	As, Cr, Cu, Ni, Pb, Zn

PL20	Caltrans (2003)	USA	2002-11-08–2003-04-14	–	–	Park and ride	8	Active-automatic	Flow-prop.	–	d, t	As, Cd, Cr, Cu, Ni, Pb, Zn
PL21	Caltrans (2003)	USA	2002-12-12–2003-03-14	–	–	Rest area with planter boxes and restrooms	8	Active-automatic	Flow-prop.	–	d, t	As, Cd, Cr, Cu, Ni, Pb, Zn
PL22	Caltrans (2003)	USA	2003-02-12–2003-04-12	–	–	Rest area, rolling grassy hills	3	Active-automatic	Flow-prop.	–	d, t	As, Cd, Cr, Cu, Ni, Pb, Zn
PL23	Caltrans (2003)	USA	2002-12-16–2003-04-17	–	–	Rest area, flat agricultural land	3	Active-automatic	Flow-prop.	–	d, t	As, Cd, Cr, Cu, Ni, Pb, Zn
PL24	Rushton (2001)	USA	1998-11–1999-11	–	1050	Florida Aquarium	30	Active-automatic	Flow-prop.	–	t	Cu, Fe, Mn, Pb, Zn
PL25	Rushton (2001)	USA	1998-11–1999-11	–	1050	Florida Aquarium	30	Active-automatic	Flow-prop.	–	t	Cu, Fe, Mn, Pb, Zn
PL26	Steuer et al. (1997)	USA	1993-10, 1994-05–1994-08	–	54,000	Commercial	12	Passive-automatic	Flow-prop.	0.45	d, t	Cu, Cd, Pb, Zn
PL27	McQueen et al., 2010	USA	2006-10-16–2007-07-17	nD, M	1254	Campus parking lot for 23 of 231 cars, residential/commuter	19	Active-automatic	First flush (2.54 mm)	–	t	Al, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn
PL28	McQueen et al., 2010	USA	2007-01-05–2007-07-30	nD, M	6398	Campus parking lot for 189 of 1089 cars, residential/commuter	12	Active-automatic	First flush (2.54 mm)	–	t	Al, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn
PL29	Line et al. (1997)	USA	1993-11-05	–	16,000	Vehicle maintenance	1	Active-automatic	First flush (5 min)	–	t	*As (<10)*, *Cd (<2)*, *Cr (<5)*, Cu, *Hg (<0.2)*, *Ni (<10)*, *Pb (<5)*, *Sb (<50)*, Zn
PL30	Line et al. (1997)	USA	1993-11-27	–	24,000	Vehicle maintenance	1	Active-automatic	First flush (5 min)	–	t	*As (<10)*, *Cd (<2)*, Cr, Cu, *Hg (<0.2)*, *Ni (<10)*, Pb, *Sb (<50)*, Zn
PL31a	Booth and Leavitt (1999)	USA	1996-11–1996-12-25	–	18	Employee parking lot with once-in, once-out daily usage	3	Passive-automatic	Flow-prop.	–	d, t	Ba, Cu, Fe, Mn, Pb, Zn
PL31b	Brattebo and Booth (2003)	USA	2001-11–2002-03	–	18	Employee parking lot with once-in, once-out daily usage	9	Passive-automatic	Flow-prop.	–	d, t	Cu, *Pb (<1)*, Zn
PL32	Bannerman et al. (1993)	USA	1991-05-05–1991-07-07	–	–	Commercial	5	Passive-automatic	Flow-prop.	0.7; 0.45	d, t	Cd, Cr, Cu, Pb, Zn
PL33	Bannerman et al. (1993)	USA	1991-05-05–1991-07-07	–	–	Industrial	9	Passive-automatic	Flow-prop.	0.7; 0.45	d, t	Cd, Cr, Cu, Pb, Zn
PL34	Hatt et al. (2009)	AUS	2007-01–2007-11	–	4500	Campus parking lot, top level of a multi-level carpark	17	Active-automatic	Flow-prop.	–	t	Cu, Mn, Pb, Zn
PL35	Maniquiz et al. (2010)	KOR	2006-06–2008-10	–	10,700	Vehicle registration office, commercial and light industrial	45	Manual	Time-prop. (first flush-enhanced)	–	t	Pb, Zn

[a] D = De-icing salt application, nD = no de-icing salt application, M = Maintenance, S = Sweeping.

[b] d = dissolved, p = particulate, t = total.

[c] Metals measured below study-specific detection limits are highlighted in this table with italics and the detection limits are presented in brackets behind these elements.

Table 2
Register of studies investigating road runoff.

ID	Literature	Location	Sampling period	Special conditions[a]	Discharge area[m^2]	AADT [Vehicles per day]	Number of events sampled	Type of sample collection	Sampling strategy	Pore diameter for filtration	Partition[b]	Metals detected[c]
RL01	Xanthopoulos and Hahn (1995)	DEU	–	–	–	3200	87	–	–	–	t	Cd, Cu, Ni, Pb, Zn
RL02	Dannecker et al. (1990)	DEU	1987	–	210	500	17	Active-automatic	Mixed	250, 0.45	t	Al, As, Ba, *Be (<0.2)*, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, Zn
RL03a	Dannecker et al. (1990)	DEU	1986	–	290	2300	16	Active-automatic	Mixed	250, 0.45	p, t	Al, As, *Be (<0.2)*, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, Zn
RL03b	Dannecker et al. (1990)	DEU	1987	–	290	2300	16	Active-automatic	Mixed	250, 0.45	p, t	Al, As, Ba, *Be (<0.2)*, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, Zn
RL04	Daub and Striebel (1995)	DEU	1994	–	–	<1000	3	Active-automatic	–	0.45, 0.1	d, p	Cu, Pb
RL05	Legret et al. (1996)	FRA	1991–1994	M	2800	2000	39	Passive-automatic	First flush	0.45	d, t	Cd, Cu, Pb, Zn
RL06	Van Dam et al. (1986)	NDL	1982-07–1985-12	M, S	176	3200	11	–	–	–	t	Cd, Cr, Cu, Pb, Zn
RL07	MacKay et al. (2011)	USA	2006-08-29	M	549	3000	1	Active-automatic	Time-prop. (equally timed)	0.45	d, t	Cu, Pb, Zn
RL08	MacKay et al. (2011)	USA	2006-08-29	M	1161	3000	1	Active-automatic	Time-prop. (equally timed)	0.45	d, t	Cu, Pb, Zn
RL09	MacKay et al. (2011)	USA	2008-08-15	M	1302	4000	1	Active-automatic	Time-prop. (equally timed)	0.45	d, t	Cu, *Pb (<5)*, Zn
RL10	MacKay et al. (2011)	USA	2008-09-06	M	279	2000	1	Active-automatic	Time-prop. (equally timed)	0.45	d, t	Cu, Pb, Zn
RL11	Gilbert and Clausen (2006)	USA	2002-06–2003-05	–	199	–	12	Passive-automatic	Mixed	–	t	Cu, Pb, Zn
RL12	Gilbert and Clausen (2006)	USA	2002-06–2003-05	–	405	–	12	Passive-automatic	Mixed	–	t	Cu, Pb, Zn
RL13	Gilbert and Clausen (2006)	USA	2002-06–2003-05	–	225	–	12	Passive-automatic	Mixed	–	t	Cu, Pb, Zn
RL14	Steuer et al. (1997)	USA	1993-10, 1994-05–1994-08	S	104,000	<3100	12	Passive-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Pb, Zn
RL15	Driscoll et al. (1990)	USA	1970s and early 1980s	–	5059	2000	–	–	–	–	t	Cu, Pb, Zn
RL16	Driscoll et al. (1990)	USA	1970s and early 1980s	–	1012	2500	–	–	–	–	t	Cu, Pb, Zn
RL17	Bannerman et al. (1993)	USA	1991-05-05–1991-07-07	S	–	100–400	10	Passive-automatic	Flow-prop.	0.7, 0.45	d, t	Cd, Cr, Cu, Pb, Zn
RL18	Bannerman et al. (1993)	USA	1991-05-05–1991-07-07	S	–	500–2150	9	Passive-automatic	Flow-prop.	0.7, 0.45	d, t	Cd, Cr, Cu, Pb, Zn
RL19	Waschbusch (1996)	USA	1994–1995	–	–	378	–	–	–	–	d, t	Cd, Cu, Pb, Zn
RL20	Davis and Birch (2010)	AUS	2007-08-19–2008-04-06	–	860	2000	4	Active-automatic	Time-prop. (first flush-enhanced)	250, 0.45	d, p	Cu, Pb, Zn
RL21	Maniquiz-Redillas and Kim (2014)	KOR	2010-05–2012-11	–	520	–	24	Manual	Time-prop. (first flush-enhanced)	0.45	d, t	Cd, Cr, Cu, Fe, Ni, Pb, Zn
RM01	Robien et al. (1997)	DEU	1990–1991	D, S	600	6000	4	Active-automatic	–	0.45, 0.1	d, p	Cd, Cu, Fe, Ni, Pb, Zn
RM02	Nadler and Meißner (2007)	DEU	1996-10-01–2005-09-08	D	17.3	6800	36	Passive-automatic	Mixed	–	t	Cd, Cr, Cu, Fe, Mo, Ni, Pb, Pt, Sb, V, Zn
RM02x	Nadler and Meißner (1999)	DEU	1997-10-01–1997-12-31	D	17.3	6100	1	Passive-automatic	Mixed	0.45	d, t	Cd, Cu, Pb, Zn
RM03	Muschack (1990)	DEU	1984-09–1985-08	–	–	14,200	31	–	–	–	t	Cd, Cr, Cu, Fe, Hg, Ni, Pb, Zn
RM04	Holthuis et al. (2012)	DEU	2008-09–2010-07	D	41,200	15,000	8	–	–	–	t	Cu, Fe, Zn
RM05	SEH (2010)	DEU	2008-08-20–2010-02-08	D	250	5500	18	Passive-automatic	Flow-prop.	–	t	*Cd (<50)*, *Cu (<100)*, *Pb (<100)*, Zn
RM06	SEH (2010)	DEU	2008-08-20–2010-02-08	D	250	5500	17	Passive-automatic	Flow-prop.	–	t	*Cd (<50)*, *Cu (<100)*, *Pb (<100)*, Zn
RM07	SEH (2010)	DEU	2008-08-20–2010-02-08	D	400	12,800	17	Passive-automatic	Flow-prop.	–	t	*Cd (<50)*, *Cu (<100)*, *Pb (<100)*, Zn
RM08	Krauth and Stotz (1993)	DEU	1991-12-17–1993-04-30	D	15,300	9870	111	–	–	–	t	Cd, Cr, Cu, Fe, Ni, Pb, Zn
RM09	Jenewein and Schinner (1982)	AUT	–	–	–	–	–	–	First flush	–	t	Cu, Pb, Zn
RM10	Rutz (2009)	CHE	2008-01-16, 2008-04-14, 2008-07-03	–	23,000	12,000	3	Manual	Random	–	d, t	Cd, Cr, Cu, Pb, Zn

RM11	von Ballmoos (2007)	CHE	2006-04-11, 2006-08-09	–	–	–	2	Manual	Random	–	t	*Cd (<1)*, *Cr (<5)*, Cu, *Pb (<5)*, Zn
RM12	Gromaire-Mertz et al. (1999)	FRA	1996-07–1997-05	M, S	602	–	7	Active-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Pb, Zn
RM13	Barbosa and Hvitved-Jacobsen (1999)	PRT	–	–	5970	6000	10	Active-automatic	Time-prop. (first flush-enhanced)	0.45	t	*Cd (<1)*, *Cr (<1)*, Cu, Pb, Zn
RM14a	Westerlund et al. (2003)	SWE	2000-03-25–2000-04-25	–	660	7400	4	Active-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Ni, Pb, Zn
RM14b	Westerlund et al. (2003)	SWE	2000-05-25–2000-06-26	–	660	7400	3	Active-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Ni, Pb, Zn
RM15	Pitt et al. (1995)	USA	–	–	–	–	1	Manual	Random	0.45	d, t	Al, Cd, Cr, Cu, Ni, Pb, Zn
RM16	MacKay et al. (2011)	USA	2007-08-09, 2008-04-28, 2008-05-02	M, S	1079	8000	3	Active-automatic	Time-prop. (equally timed)	0.45	d, t	Cu, Pb, Zn
RM17	MacKay et al. (2011)	USA	2006-09-03	M, S	1898	–	1	Active-automatic	Time-prop. (equally timed)	0.45	d, t	Cu, Pb, Zn
RM18	MacKay et al. (2011)	USA	2006-09-03	M, S	866	–	1	Active-automatic	Time-prop. (equally timed)	0.45	d, t	Cu, *Pb (<5)*, Zn
RM19	Fulkerson et al. (2007)	USA	2005-03-03–2005-07-30	nS	8100	5000	14	Manual	Time-prop. (equally timed)	0.7	d, p	MeHg, THg
RM20	Steuer et al. (1997)	USA	1993-10, 1994-05–1994-08	S	16,000	10,600	12	Passive-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Pb, Zn
RM21	Steuer et al. (1997)	USA	1993-10, 1994-05–1994-08	S	21,000	3100–5100	12	Passive-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Pb, Zn
RM22	Steuer et al. (1997)	USA	1993-10, 1994-05–1994-08	–	49,000	–	12	Passive-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Pb, Zn
RM23	Driscoll et al. (1990)	USA	1970s and early 1980s	–	1133	7300	–	–	–	–	t	Cu, Pb, Zn
RM24	Bannerman et al. (1993)	USA	1991-05-05–1991-07-07	S	–	2850–7300	10	Passive-automatic	Flow-prop.	0.7; 0.45	d, t	Cd, Cr, Cu, Pb, Zn
RM25	Bannerman et al. (1993)	USA	1991-05-05–1991-07-07	–	–	–	10	Passive-automatic	Flow-prop.	0.7; 0.45	d, t	Cd, Cr, Cu, Pb, Zn
RM26	Novotny et al. (1998)	USA	–	D	–	–	–	–	–	–	d, t	Cu, Pb, Zn
RM27	Novotny et al. (1998)	USA	–	D	–	–	–	–	–	–	d, t	Cu, Pb, Zn
RM28	Waschbusch, 1996	USA	1994–1995	D	–	6157	–	–	–	–	d, t	Cd, Cu, Pb, Zn
RM29	Novotny et al. (1998)	CAN	–	D	–	–	–	–	–	–	d, t	Cu, Pb, Zn
RM30	Novotny et al. (1998)	CAN	–	D	–	–	–	–	–	–	d, t	Cu, Pb, Zn
RM31	Brockbank et al. (1999)	AUS	–	–	–	–	–	–	–	–	d, p	Cu, Fe, Mn, Pb, Zn
RM32	Prestes et al. (2006)	BRA	2002-09–2004-03	S	1300	9000	21	Manual	Flow-prop.	0.45	d, t	Cd, Cu, Pb
RM33	Zhang et al. (2013)	CHN	2010-06-06–2011-08-04	nD	540	10,000	9	–	Time-prop. (first flush-enhanced)	–	d	Cu, Pb, Zn
RU01	Wüst et al. (1994)	DEU	1990-03-28, 1990-05-08, 1990-06-02	D, S	133	25,000	3	Manual	–	0.45, 0.1	d, p	Cd, Cu, Pb, Zn
RU02	Herrmann et al. (1992)	DEU	1991-07-14, 1991-08-15, 1991-08-17	nD, S	30	16,000	3	Active-automatic	–	0.45, 0.1	d, p	Cd, Cu, Fe, Ni, Pb, Zn
RU02x	Laschka et al. (1996)	DEU	1991	D, S	30	16,000	6	Active-automatic	–	–	t	Pt
RU03a	Helmreich et al. (2010)	DEU	2003-11–2005-11	D, S	400	57,000	63	Passive-automatic	First flush (1 mm rain)	0.45	d, t	*Cd (<0.5)*, Cu, Ni, Pb, Zn
RU03b	Hilliges et al. (2013)	DEU	2006-07-01–2007-06-30	D, S	100	57,000	24	Active-automatic	Flow-prop.	–	t	Cu, Pb, Zn
RU04	Dannecker et al. (1990)	DEU	1987	–	250	16,200	12	Active-automatic	Mixed	250, 0.45	t	Al, As, Ba, *Be (<0.2)*, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sb, Se, V, Zn
RU05	Dierkes (2010)	DEU	2009-01-12–2009-11-27	D	2300	18,000	11	Manual	Random	–	t	Cd, Cu, Pb, Zn
RU06	Gäth et al. (1990)	DEU	1988-12-01–1989-01-13	–	162.2	18,129	8	Active-automatic	Time-prop. (equally timed)	–	t	Cd, Cu, Pb, Zn
RU07a	Kasting et al. (2003)	DEU	2000-10–2001-09	D	15,000	45,000	–	Manual	Random	–	d, t	Cu, Pb, Zn
RU07b	Kasting and Grotehusmann (2007)	DEU	–	D	15,000	45,000	–	Manual	Random	–	d, t	Cu, Pb, Pt, Zn
RU07c	Grotehusmann and Kasting (2009)	DEU	2007-02-09–2008-01-07	D	15,000	45,000	10	Manual	Random	–	d, t	*As (<5)*, Cd, Co, Cr, Cu, Fe, *Hg (<0.2)*, *Mo (<5)*, Ni, Pb, Sb, V, Zn
RU08	Dierkes (1999)	DEU	1997-06–1999-02	D	–	26,000	28	Passive-automatic	Mixed	0.45	d, t	Cd, Cu, Pb, Zn
RU09	Daub and Striebel (1995)	DEU	1994	–	100	22,000	3	Active-automatic	–	0.45, 0.1	d, p	Cu, Pb
RU10	Schütte (1997)	DEU	1996-09–1996-12	–	–	15,000–20,000	10	–	–	–	t	Cd, Cu, Fe, *Hg (<0.5)*, Ni, Pb, Zn
RU11	Steiner et al. (2010)	AUT	–	–	–	–	–	–	–	–	t	Cr, Cu, Ni, Pb, Zn
RU12	Windhofer et al. (2012)	AUT	2011-10-21–2011-12-10	D	3300	26,000	4	–	Flow-prop.	–	t	Cu, Rh, Pt, Zn
RU13	Langbein et al. (2006)	CHE	2002-10-23–2004-07-08	–	1500	17,000	7	Active-automatic	Time-prop. (first flush-enhanced)	0.45	d, t	Cd, Cr, Cu, Fe, Ni, Pb, Zn

(*continued on next page*)

Table 2 (*continued*)

ID	Literature	Location	Sampling period	Special conditions[a]	Discharge area[m^2]	AADT [Vehicles per day]	Number of events sampled	Type of sample collection	Sampling strategy	Pore diameter for filtration	Partition[b]	Metals detected[c]
RU14	Steiner et al. (2006)	CHE	2002–2004	–	14.1	17,000	–	Passive-automatic	Mixed	–	t	Cd, Cr, Cu, Fe, Pb, Sn, Zn
RU15	Klimaszewska et al. (2007)	POL	2003-01–2003-05	S	–	59,000	13	Passive-automatic	Flow-prop.	–	t	Cd, Pb, Zn
RU16	Klimaszewska et al. (2007)	POL	2003-01–2003-05	S	–	39,400	8	Passive-automatic	Flow-prop.	–	t	Cd, Pb, Zn
RU17	Nowakowska-Blaszczyk and Zakrzewski (1996)	POL	–	S	–	–	–	–	–	–	t	Pb
RU18	Wei and Morrison (1994)	SWE	1992-05-26	–	–	–	1	–	–	0.45	d, t	Pt
RU19a	Bannerman et al. (1993)	USA	1991-05-05–1991-07-07	S	–	20,000	10	Passive-automatic	Flow-prop.	0.7, 0.45	d, t	Cd, Cr, Cu, Pb, Zn
RU19b	Waschbusch (1996)	USA	1994–1995	–	–	18,600	–	–	–	–	d, t	Cd, Cu, Pb, Zn
RU20	Bannerman et al. (1993)	USA	1991-05-05–1991-07-07	S	–	19,800	9	Passive-automatic	Flow-prop.	0.7, 0.45	d, t	Cd, Cr, Cu, Pb, Zn
RU21	Davis and Birch (2010)	AUS	2007-08-19–2008-04-06	–	1095	84,500	6	Active-automatic	Time-prop. (first flush-enhanced)	250, 0.45	d, p	Cu, Pb, Zn
RU22	Lloyd and Wong (1999)	AUS	1999-01-22, 1999-01-29	nS	1500	32,000	2	Manual	Time-prop. (equally timed)	–	t	Cd, Cu, Pb, Zn
RU23	Timperley et al. (2005)	NZL	2002-08-24–2002-09-18	–	–	17,000	4	Active-automatic	Flow-prop.	–	d, p	Cu, Pb, Zn
RU24	Chang et al. (2009)	CHN	2005-06-28, 2005-09-21	–	–	–	2	–	Time-prop. (equally timed)	0.45	d, t	Cd, Cr, Cu, Ni, Pb, Zn
RU25	Huang et al. (2007)	CHN	2005-06–2006-09	–	3875	30,000	5	Active-automatic	Time-prop. (first flush-enhanced)	–	t	Cu, Pb, Zn
RU26	Maniquiz et al. (2010)	KOR	2006-06–2008-10	–	5000	–	45	Manual	Time-prop. (first flush-enhanced)	–	t	Pb, Zn

[a] D = De-icing salt application, nD = no de-icing salt application, M = Maintenance, S = Sweeping, nS = no sweeping.

[b] d = dissolved, p = particulate, t = total.

[c] Metals measured below study-specific detection limits are highlighted in this table with italics and the detection limits are presented in brackets behind these elements.

Table 3
Register of studies investigating bridge runoff.

ID	Literature	Location	Sampling period	Special conditions[a]	Discharge area [m^2]	AADT [vehicles per day]	Number of events sampled	Type of sample collection	Sampling strategy	Pore diameter for filtration	Partition[b]	Metals detected[c]
BR01a	Legret and Pagotto (1999)	FRA	1995-03-28–1996-02-26	D, S	3200	12,000	49	Active-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Pb, Zn
BR01b	Pagotto et al. (2000)	FRA	1997-06-09–1997-11-09	D	3200	12,000	25	Active-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Pb, Zn
BR02a	Hallberg et al. (2007)	SWE	2005-05-03–2005-08-26	D	13,700	108,300	4	Active-automatic	Flow-prop.	0.45	d, t	Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn
BR02b	Hallberg et al. (2007)	SWE	2005-12-14–2006-02-14	D	13,700	108,300	5	Active-automatic	Flow-prop.	0.45	d, t	Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn
BR03	Driscoll et al. (1990)	USA	1970s and early 1980s	–	6070	42,000	–	–	–	–	t	Cu, Fe, Pb, Zn
BR04	Yousef et al. (1984)	USA	–	–	6825	55,000	11	–	–	–	d, t	Cd, Cr, Cu, Fe, Ni, Pb, Zn
BR05	Yousef et al. (1984)	USA	1982-08–1983-05	–	19,800	11,500	15	–	–	–	d, t	Cd, Cr, Cu, Fe, Ni, Pb, Zn
BR06	Driscoll et al. (1990)	USA	1970s and early 1980s	–	5787	70,000	–	–	–	–	t	Cd, Cr, Cu, Pb, Zn
BR07	Dean et al. (2005)	USA	2002-04-11–2002-06-16	–	544	–	4	Manual	Time-prop. (equally timed)	0.45	d, p	Cd, Cu, Pb, Zn
BR08	Wu et al. (1998)	USA	1995-09-22–1996-07-01	D, M	1497	25,000	10	Active-automatic	Time-prop. (pre-set time)	–	t	*Cd* (<0.5), Cr, Cu, Ni, Pb
BR09	Barrett et al. (1998)	USA	1994-04-29–1995-05-18	–	526	8780	26	Active-automatic	Flow-prop.	–	t	Cu, Fe, Pb, Zn
BR10	Barrett et al. (1998)	USA	1994-10-17–1995-05-08	–	1060	47,000	8	Active-automatic	Flow-prop.	–	t	Cu, Fe, Pb, Zn
BR11	Bourcier and Hindin (1979); Bourcier et al. (1980)	USA	1976-10-14–1977-03-23	M, S	2323	–	3	–	–	–[d]	d, t	Cr, Fe, Pb, Ti, W, Zn
BR12	Driscoll et al. (1990)	USA	1970s and early 1980s	–	401	42,000	–	–	–	–	t	Cu, Pb, Zn
BR13	Driscoll et al. (1990)	USA	1970s and early 1980s	–	890	17,000	–	–	–	–	t	Cu, Pb, Zn
BR14	Wilson (2006)	USA	2003-04-08, 2003-10-06, 2004-01-14	–	250	100,000–180,000	3	Active-automatic	Flow-prop.	–	d, t	As, Ba, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, Sb, V, Zn
BR15	Driscoll et al. (1990)	USA	late 1970s and early 1980	–	8498	53,000	35	–	–	–	t	Cd, Cr, Cu, Fe, Hg, Pb, Zn
BR16a	Frazer, 1990	CAN	–	–	–	–	29	–	–	–	t	Cd, Cu, Ni, Pb, Zn
BR16b	Marsalek et al. (1997)	CAN	16 months	–	–	83,700–102,100	53	Passive-automatic	Flow-prop.	0.45	d, t	Cd, Cu, Ni, Pb, Zn
BR17	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	5200	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR18	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	3548	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR19	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	15,524	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
RB20	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	23,672	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR21	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	15,766	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR22	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	27,000	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR23	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	12,156	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR24	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	13,302	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR25	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	14,312	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR26	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	33,000	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR27	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	33,000	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR28	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	35,248	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR29	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	33,191	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR30	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	26,950	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR31	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	26,826	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR32	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	50,304	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR33	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	46,235	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR34	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	32,000	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR35	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	32,000	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR36	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	45,000	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR37	Drapper et al. (2000)	AUS	1997-07–1999-01	–	–	5900	12	Passive-automatic	First flush (20 l)	–	t	Cu, Pb, Zn
BR38	Gan et al. (2008)	CHN	2006-04-06–2006-06-12	M	110	31,000	7	Active-automatic	Time-prop. (first flush-enhanced)	–	t	Cd, Cr, Cu, Ni, Pb, Zn
BR39	Yu and Zhao (2012)	CHN	2007-04-22–2007-09-01	–	–	–	3	–	Time-prop. (equally timed)	–	t	Cd, Cr, Ni, Pb, Zn

[a] D = De-icing salt application, M = Maintenance, S = Sweeping.
[b] d = dissolved, p = particulate, t = total.
[c] Metals measured below study-specific detection limits are highlighted in this table with italics and the detection limits are presented in brackets behind these elements.
[d] Centrifuging 50 ml of sample for 5 min at 681 g.

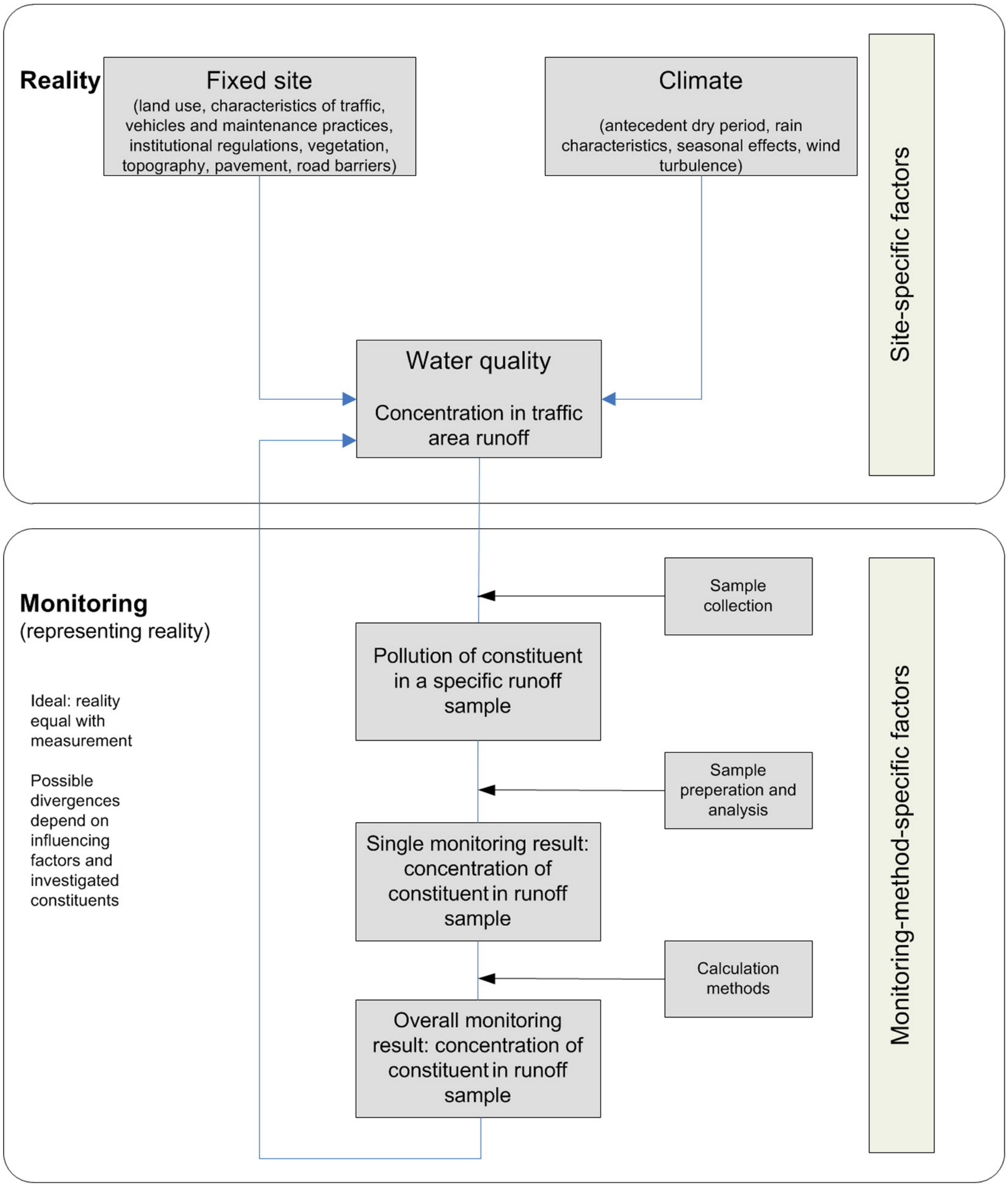

Fig. 1. Factors influencing monitoring programs and general flow schema for analysis of runoff characteristics and concentrations.

The final part of site-specific factors describes *operational characteristics* concerning traffic, types of vehicles, maintenance, and institutional regulations. Traffic volume is mostly characterized by *AADT* and varies in this review between 100 and 328,000 vehicles per day. Although some authors have concluded that AADT has the greatest influence on runoff concentrations of metals (Mangani et al., 2005; Crabtree et al., 2008), it might not be the best indicator for traffic area runoff concentrations and should not be used as a sole predictor because it only explains approximately 30% of the variation in these concentrations (Driscoll et al., 1990; Drapper et al., 2000; Kayhanian et al., 2003). Special cases of AADT are the traffic flow volume during sampling and between two sampling periods (Shinya et al., 2003; Hallberg et al., 2007; Li and Barret, 2008). CH2MHILL (1998) concluded that splashing and washing of pollutants from vehicles is more important than the wash-off of pollutants accumulated on road surface during dry periods and splashing corresponds with average vehicle speed during storms (Hallberg et al., 2007). It has not been specified whether the stated AADT is only valid for the monitored lanes or for all lanes in both directions; this makes it difficult to compare different studies, e.g., the site described by Aryal and Lee (2009), and to evaluate the impact of AADT on runoff concentrations. Another traffic-related aspect is that the *percentage of trucks* can be very high in industrial zones (approximately 37%) (Dannecker et al., 1990) and can be as much as 60% of the AADT (Gan et al., 2008). Further factors influencing runoff quality are construction works, car accidents, firefighting foams, and leakages (Hogland et al., 1987; Barrett et al., 1998; Holthuis et al., 2012). *Maintenance* factors, which differ widely among countries (Gan et al., 2008), are mowing of the roadside shoulders (Barrett, 2008), street sweeping (Gromaire-Mertz et al., 1999; Waschbusch, 2003; Horwatich and Bannerman, 2009; Helmreich et al., 2010), and winter services (Sansalone and Glenn, 2002; Bäckström et al., 2003; Crabtree et al., 2006; Hallberg et al., 2007; Klimaszewska et al., 2007; Tromp et al., 2012). The most important *institutional regulation* was the phase-out of Pb (cf. Section 4.1).

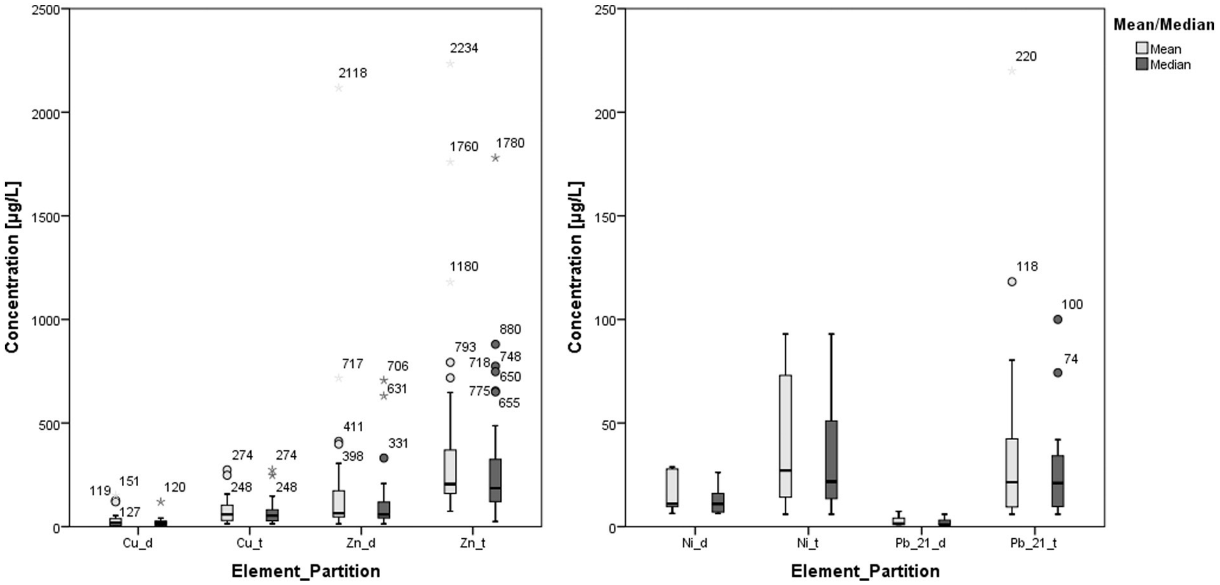

Fig. 2. Differences between mean and median values (d and t) for Cu, Zn (both left), Ni and Pb (both right) for highway runoff data (n = 41). Data were obtained from monitoring programs with more than two observed rain events, and both mean and median data were calculated for each site.

3.1.2. Climate

Climatic site-specific factors are dry deposition rates, antecedent dry periods (ADP), rain characteristics (volume, intensity, and duration), seasonal effects, and wind turbulence. Significant non-traffic-related sources are *dustfalls and dry deposition* during periods without rain, which entrains contaminants and removes them from the atmosphere (Hamilton et al., 1987; Ball, 2002). *ADP* were found to be a statistically significant factor for runoff concentrations due to the accumulation of substances on the traffic area surface (Harrison and Wilson, 1985; Hewitt and Rashed, 1992; Pitt et al., 1995; Irish et al., 1998; Drapper et al., 2000; Shinya et al., 2003; Westerlund et al., 2003; Mangani et al., 2005; Han et al., 2006a; Prestes et al., 2006; Wilson, 2006; Alo et al., 2007; Gan et al., 2008; Li and Barrett, 2008; Moores et al., 2010; Lee et al., 2011). However, Moy and Crabtree (2003), Shinya et al. (2003), Langbein et al. (2006), and Nason et al. (2012) found no relation between ADP and metal concentrations. This is most probably because of street sweeping, wind turbulence, and air turbulence caused by traffic (Bourcier et al., 1980; Helmreich et al., 2010). However, not all storm events completely clean the road surface and this effect prolongs the actual ADP (Cristina and Sansalone, 2003). A special case in (semi)arid regions is the first storm of each season characterized by higher runoff concentrations (Stenstrom and Kayhanian, 2005; Terzakis et al., 2008).

For *rain characteristics*, mostly poor correlations were obtained (Moy et al., 2003; Han et al., 2006a; Desta et al., 2007; MacKay et al., 2011; Nason et al., 2012; Zhang et al., 2013) and only rain history appears to have an effect on the washed-off substances (Steiner and Goosse, 2007). In contrast, some researchers found a correlation between runoff concentrations and storm intensities (Pitt et al., 1995; Tiefenthaler et al., 2001; Crabtree et al., 2006).

Moy et al. (2003) have observed an influence of *seasonal effects* on runoff concentrations; however, the conclusions are heterogeneous. Several studies have indicated that metal concentrations are higher in snowmelt runoff than in rainfall runoff because of the snow accumulation period, and the following melting period appears to be a critical

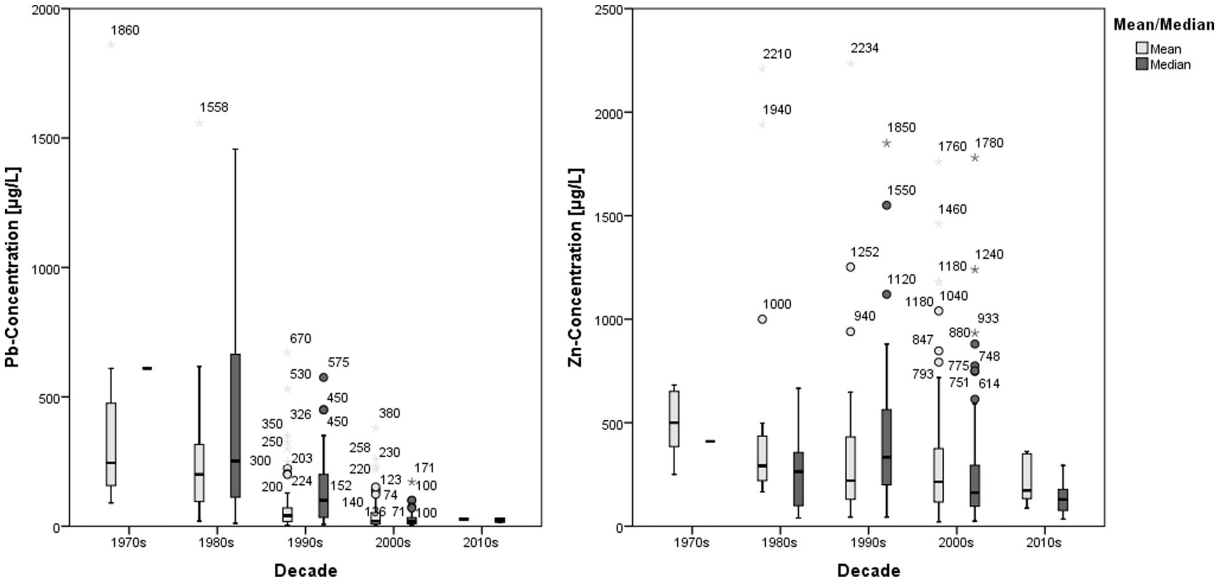

Fig. 3. Historical trends of Pb (n = 194 for mean; n = 146 for median) and Zn (n = 206 for mean; n = 148 for median) in traffic area runoff from the 1970s to date.

Table 4
Statistics of Fig. 3 — historical trends of lead (n = 194 for mean; n = 146 for median) and zinc (n = 206 for mean; n = 148 for median) in traffic area runoff from the 1970s to date.

Element	Decade	Calculation for all mean values from the literature										Calculation for all median values from the literature									
		Min	Median	Geometric mean	Mean	75th	90th	95th	Max	SD	95%[a]	Min	Median	Geometric mean	Mean	75th	90th	95th	Max	SD	95%[a]
Pb	1970s	90,0	293	346	558	923	[b]	[b]	1860	662	694	610	610	610	610	[b]	[b]	[b]	610	[b]	[b]
Pb	1980s	20,0	200	162	277	328	617	[b]	1558	346	167	11,0	252	230	393	685	995	1281	1457	377	146
Pb	1990s	4,0	41,0	40,7	79,6	71,5	227	339	670	119	28,8	7,0	100	73,3	133	200	350	450	575	133	38,2
Pb	2000s	1,4	19,2	20,7	42,2	56,0	118	152	380	59,9	12,0	2,3	18,0	17,2	26,1	34,2	59,0	92,3	171	28,2	7,0
Pb	2010s	24,0	27,5	27,3	27,5	[b]	[b]	[b]	31,0	4,9	44,5	15,0	31,0	37,3	52,7	[b]	[b]	[b]	112	52,0	129
Zn	1970s	250	455	776	3540	5240	[b]	[b]	19,100	7624	8000	410	410	410	410	[b]	[b]	[b]	410	[b]	[b]
Zn	1980s	166	292	366	535	467	1994	[b]	2210	614	316	40,0	264	217	366	364	655	2336	2892	565	238
Zn	1990s	43,8	220	230	329	450	620	940	2234	342	89,1	44,0	333	332	445	564	846	1486	1850	372	116
Zn	2000s	21,0	215	201	295	375	652	857	1760	287	52,4	25,0	162	170	264	304	667	891	1780	296	68,0
Zn	2010s	88,0	173	188	213	353	[b]	[b]	361	115	121	35,0	154	189	545	861	[b]	[b]	2560	991	1040

[a] 95% confidence errors on the means.
[b] No values because of small universe.

one for metal runoff concentrations (Lygren et al., 1984; Sansalone and Buchberger, 1996; Westerlund et al., 2003). A general increase of metals in traffic area runoff in winter was detected due to higher wear and tear as well as to an increased corrosion enhanced by de-icing salts and lower rain intensities (Legret and Pagotto, 1999; Hallberg et al., 2007; Helmreich et al., 2010). In contrast, Mungur et al. (1995) and Brezonik and Stadelmann (2002) detected the highest runoff loads in spring and summer, and Langbein et al. (2006) did not observe any seasonal dependency.

The final climatic factor, i.e., *wind and car-induced turbulence*, leads to a decrease in atmospheric deposition and reduces metal runoff concentrations (Stotz, 1987; Ball et al., 1998; Barbosa and Hvitved-Jacobsen, 1999; Preciado and Li, 2005). Wind turbulence increases with increasing vehicle speeds and decreases with the existence of hard shoulders and road barriers (Scheiwiller, 2008). In special cases such as in Auckland, New Zealand, frequent rain does not allow contaminants to be blown off the road surface (Timperley et al., 2005).

Further description of several site-specific factors for HWY can be found in Opher and Friedler (2010).

3.2. Monitoring-method-specific factors

In most studies, mean or median metal concentrations from runoff of a particular surface type are analyzed; other studies used pollutographs of single events. Further differences occur in sample preparation and analysis. The largest possible deviations arise during sampling and sample preparation, while the actual sample analysis causes less variation, if appropriate analytical methods are used.

3.2.1. Sample collection

The *type of sampling* (automatic or manual) depends on the research objective. For short-term sampling, *manual* (*grab*) *samples* are regularly used because they are a sampling requirement for stormwater permit applications (Line et al., 1997) and are easily manageable for an evaluation of the effectiveness of decentralized stormwater treatment plants (van Ballmoos, 2007; Dierkes, 2010; Rutz, 2012). *Automatic composite samplers* are used for a convenient collection of representative samples with up to several hundred subsamples of entire storm events (Ma et al., 2009; Steiner and Goosse, 2011b). Within the literature study, 13% of the datasets containing information about the sampling type were based on manual sampling, whereas 38% conducted passive-automatic sampling and 49% conducted active-automatic sampling. Active-automatic sampling can lead to the loss of particulate metals (Pagotto et al., 2000). This depends on the runoff composition, the mixture of the flowing water, the sampler intake orientation, and the tube and suction pump used. To minimize the fractionation effect during sampling, highest flow rates and shortest sampling tubes possible should be used; the loss of metals by adsorption can be avoided using polytetrafluoroethylene sampling tubes (Hewitt and Rashed, 1992). For the determination of pH and temperature, automatic samplers should not be used and manual sampling is recommended (Minervini, 2008). This recommendation is applicable to all rapidly degradable compounds and to substances that can adhere on container surfaces. To avoid cross-contamination of runoff samples, bottles must be changed for each rain event to prevent the mixing of samples from two events. Therefore, criteria for detecting a runoff are essential, as well as the event definition, which is directly linked to the sampling strategy and considers both the beginning and end of each event, to prevent a sampling of overlapping events. An appropriate cleaning of tubes, bottles, and the parts of the sampling device in contact with the runoff is necessary. For manual sampling, only bottle cleaning must be considered. Detailed examples of passive-automatic sampling devices have been given by Bannerman et al. (1993) and Li et al. (2008); an active-automatic sampling device has been described by Striebel et al. (1994). A comparison of grab and composite sampling techniques by Ma et al. (2009) concluded that a large number of grab samples (>30)

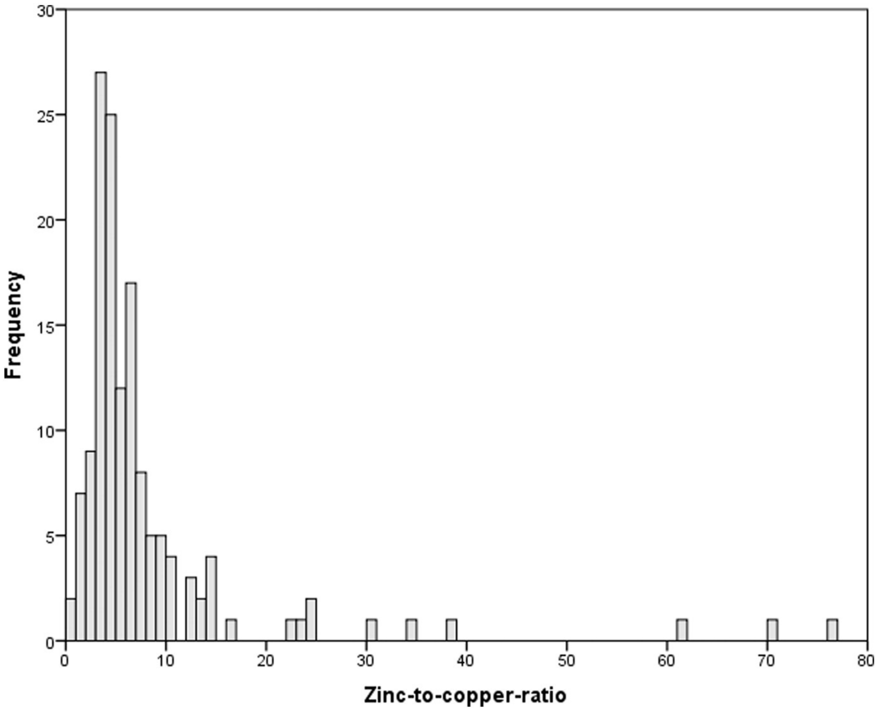

Fig. 4. Frequencies of Zn-to-Cu-ratios for all datasets containing median values (n = 141; mean = 8.1, SD = 10.9).

is required to approach the precision and accuracy of flow-proportional automatic sampling and for detecting a first flush effect, even more grab samples must be obtained. For manual sampling, the best strategy is to collect grab samples flow-proportionally (Stenstrom and Kayhanian, 2005). A storing of samples at 3 °C–6 °C is preferred (Hallberg et al., 2007; Hilliges et al., 2013; Scheiwiller, 2014).

The *sampling strategy* differs between flow-proportional (38% of datasets with information about the sampling strategy), time-proportional (24%), first flush (21%), mixed (12%), and random (5%) sampling.

In *flow-proportional sampling*, combined samples are mostly collected but discrete sampling is used for producing pollutographs (Legret and Pagotto, 1999) and a second device is required to trigger the sampling device. The second device can be, e.g., a magnetic inductive flow meter (Hilliges et al., 2013), a V-shaped overflow with an immersed ultrasonic flow meter (Legret and Pagotto, 1999; Hatt et al., 2009), a weir with a pressure probe (Hallberg et al., 2007), or a bubbler flow meter (Barrett et al., 1998; Nason et al., 2012). Criteria for sampling are very site-specific and can be 20 mL of sample for every 100 L of a runoff (Tromp et al., 2012). This strategy is limited by the detection limit of the second device sensor (Lau et al., 2009).

Time-proportional sampling can be subdivided into first flush-enhanced and equally timed sample collection. Han et al. (2006b) performed first flush-enhanced sampling, in which five grab samples were collected every 15 min in the first hour after the start of a detectable runoff. Subsequently, one sample was obtained every hour for the following 7 h and one or two additional grab samples were collected for events lasting longer than 8 h. Equally timed samples were obtained at specific time intervals by considering the sampling type and site-specific factors such as rain intensity, the slope of the catchment area and the season (Stotz, 1987). In most cases, the time interval is 5 min but it varies between a few minutes (2 min (Tiefenthaler et al., 2001) or 3 min (Chang et al., 2009)) up to 45 min (Golwer and Schneider, 1983) and 60 min (Gäth et al., 1990). Discrete time-proportional samples can be handled similar to mixed/combined flow-proportional samples by considering the data for simultaneous and continuous flow measurements.

For *first flush sampling*, which is a special case of time-proportional sampling, three types of triggering signals are used: time, volume, or rain depth. Line et al. (1997) sampled water of the first 5 min per rain event, Mangani et al. (2005) collected the first 10 L, Drapper et al. (2000) collected the first 20 L, Helmreich et al. (2010) collected the first 400 L (equaling a rain depth of 1 mm for this monitored site), and McQueen (2008) collected the first 2.5 mm runoff of a storm event. For an evaluation of the first flush effect, which might occur in the first 30 to 60 min, a sampling of the entire rain event is necessary (Han et al., 2006a). Definitions of the first flush effect are given by Wanielista and Yousef (1993), Saget et al. (1996), and Bertrand-Krajewski et al. (1998).

Mixed sampling by collecting the complete runoff volume of one event is an ideal method to obtain representative concentrations of a storm event. However, this strategy is only practical for small catchment areas (<20 m^2) and a settling of particles in a storage tank must be avoided (Kasting et al., 2003). Some mixed samples are taken over a time period containing several rain events (Nadler and Meißner, 2007; Engelhard et al., 2012).

All *randomly collected samples* were taken manually during storm events to evaluate the effectiveness of best management practices.

The type and strategy of sampling have an influence on metal concentration, as does the *location*, from which runoff samples are taken. E.g., sediments can be retained by the overflow of a measurement channel and are not sampled by the suction pump of an automatic sampler (Legret and Pagotto, 1999). Further aspects are related to the characteristics of the channel. A concrete channel has an influence on pH and long distances between the runoff edge, and the sampling device might allow the settling of solids in the channel during rain events with low intensities. Coatings and tube materials (e.g., stainless steel) can increase Ni concentrations in the samples (Nadler and Meißner, 1999).

Overall, as a result of rain events without runoff, equipment failure, and operational reasons such as holidays, not all rain events can be sampled: Grotehusmann and Kasting (2009) sampled and analyzed about half of all events during a one-year investigation period, Lee et al. (2011) monitored 50 out of more than 100 storm events during a

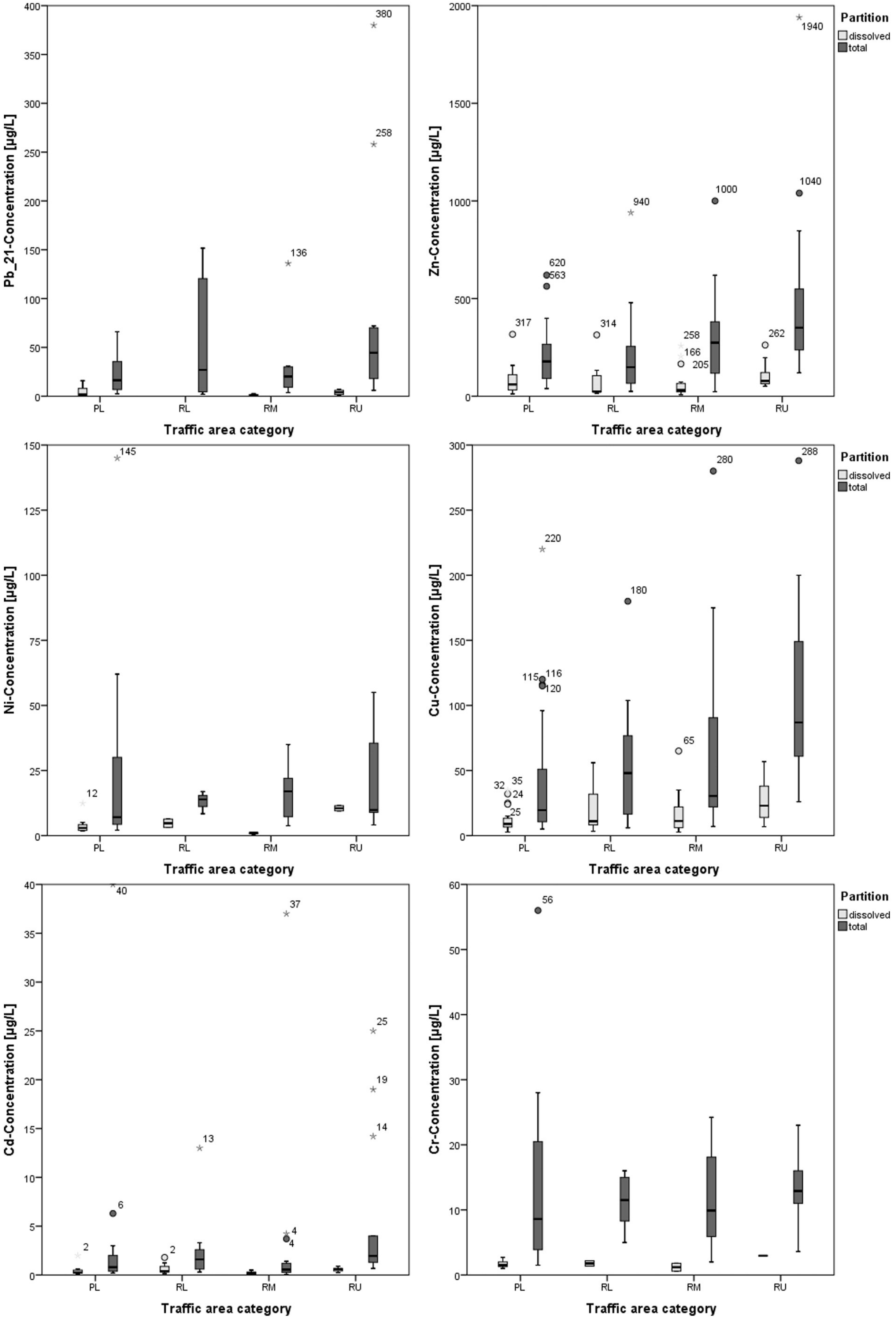

Fig. 5. Mean metal concentrations (d and t) in parking lot (PL) and road (RL, RM, and RU) runoff.

three-year period, and Terzakis et al. (2008) sampled 41 out of 58 rain events during a one-year period.

In conclusion, sampling programs had different objectives and constraints, and the sampling types and strategies used varied widely. Further influencing factors arise from equipment and operators.

3.2.2. Sample preparation and analysis

Prior to analysis, samples must be prepared according to the objectives. Only 36% of the datasets contain information about *preparation methods* (e.g., homogenization, subsampling, and filtration) and only 55 sets describe the holding time. For all metal analyses except those for total concentrations, the *holding time* is essential by having a critical effect on pH and on the fractionation of metals. Runoff samples were held for less than 1 h (Dean et al., 2005; Fulkerson et al., 2007), for 2–3 h (Barbosa and Hvitved-Jacobsen, 1999), for 7 h (Lau et al., 2009) or for a maximum of 24 h (Stotz, 1987; Stotz and Krauth, 1994; Booth and Leavitt, 1999; Brattebo and Booth, 2003; Flint and Davis, 2007; Moores et al., 2010; Lee et al., 2011; Nason et al., 2012; Borne et al., 2013) before delivery to a laboratory and beginning of preparation. Other studies stated the holding time as "immediately" or did not specify it properly. As Minervini (2008) discussed, the description of the sample collection and analysis often does not state how the pH analysis was performed, especially considering the maximum recommended holding time of 15 min (APHA, 2005). Only pH measurements in the field can comply with this requirement (Sansalone et al., 2005). As onsite filtration is risky owing to possible contamination of aqueous samples, samples should be fractionized as soon as practically possible within hours after collection in a laboratory to preserve the partitioning at the time of interest (Gnecco et al., 2008). For the *fractionation process*, a 0.45-μm-pore-diameter membrane filter is used (APHA, 2005) and the metals passing through the filter are the *dissolved part*. Filters with smaller pores (0.1 μm (Herrmann et al., 1992), 0.2 μm (Kluge et al., 2014), and 0.22 μm (Mangani et al., 2005)) and larger pores of up to 0.8 μm (Golwer and Schneider, 1983; Fulkerson et al., 2007) have also been applied. In addition, centrifuging (Bourcier and Hindin, 1979) and decanting before filtration (Bäckström et al., 2003) have been conducted. After filtration, the samples are mostly conserved with nitric acid to a pH of less than 2. The *soluble part* is determined as the dissolved part but from an acid preserved solution. Further factors associated with the filtration process are the diameter of the filter, the manufacturer, the volume of sample processed, and the amount of suspended solids in the sample, from which an inclusion/exclusion of colloidally-associated trace metals can occur (Horowitz et al., 1996). Measurements by Langbein et al. (2006) detected that the filtrate might contain fine colloids and particulate material in the nanometer range. An example of contamination during preparation and analysis of aqueous samples that was detected using blanks is described by Bannerman et al. (1993), in which the samples were contaminated with Zn during filtering procedure.

Preciado and Li (2005) described a procedure of using a chelex resin in a glass column to obtain the *resin-exchangeable part* of heavy metals, which is highly bioavailable.

To determine the potentially bioavailable part of a sample that can be *mobile* in different environmental conditions (i.e., for most metals at a lower pH), one part of a sample can be decanted and acidified followed by filtration after 24 h (Bäckström et al., 2003).

The *acid-exchangeable part* of metals is also called the total recoverable or pseudo-total concentration and is determined by adding a strong acid to the sample. Depending on the oxidizing potential of the acid and other equipment used (i.e., autoclave, microwave), a different part of all metals (i.e., all dissolved/soluble and a varying part of the particulate bound metals) is analyzed.

For the determination of *total concentration* of metals, aqua regia digestion (Nadler and Meißner, 1999; Hilliges et al., 2013), aqua regia digestion with a microwave (Shinya et al., 2000; Shinya et al., 2003), or lithium metaborate digestion must be used to destruct any particulate matter. The difference between the results of total and acid-exchangeable concentrations depends on the element, fixed site-specific factors, and climatic factors.

The particulate content is measured by separating and weighing the particulates gravimetrically. The metal concentration in *particulates*, i.e., the particulate bound metals, is usually taken as the difference of total or acid-exchangeable and dissolved or soluble concentrations but can also be determined in the filter residue by acid digestion, autoclaves, microwave digestion, or a combination of these digestion methods.

After sample preparation, the concentrations were determined in most cases using *atomic absorption spectroscopy* (AAS, 62 times), *inductively coupled plasma optical emission spectrometry/inductively coupled plasma mass spectrometry* (ICP-OES/ICP-MS, 108 times), or both techniques consecutively to reduce the detection limits of several elements (25 times), and to improve the differentiation of solid and liquid phases (Sansalone and Buchberger, 1997b).

To obtain precise and accurate data, it is essential to conduct several *quality assurance/quality control* (QA/QC) procedures. First, field replicate samples should be collected and analyzed. Second, field and method blanks, certified reference materials, matrix spikes, and duplicates or triplicates must be considered and analyzed to prevent failures during sample preparation/analysis (Line et al., 1997; Wilson, 2006). Line et al. (1997) used standard and spiked samples for a minimum of 10% of their field samples. A further QA/QC procedure is the use of materials from two manufacturers for every elemental standard used in the analysis (Westerlund and Viklander, 2006).

3.2.3. Calculation methods

Thomson et al. (1997) and Drapper et al. (2000) concluded that a minimum of 17 runoff events provides a statistically representative sample database. In the dataset of this review, the number of events sampled varied between 1 and 211 (Moxness, 1986), with a mean of 18.

The most influential factor for the calculation of mean or median values concerns the analyzed concentrations of samples below the *detection limits*. There are several methods of dealing with this type of result: they could be discarded and only observed concentrations could be used for the calculation (Pitt et al., 1995), or they could be set as the detection values (Barrett et al., 1998; Dierkes, 1999; Lange et al., 2003; Borne et al., 2013), or as numerical values of one-half the detection level (Herrmann et al., 1992; Bannerman et al., 1993; Laschka et al., 1996; CH2MHILL, 1998; Brattebo and Booth, 2003; Flint and Davis, 2007; Grotehusmann and Kasting, 2009; Lau et al., 2009). None of the studies included in the dataset specified, when a mean or median value is stated as "not detected" or when it is calculated using either the full or one-half the detection values and it is often not specified, how many of the analyses were below the detection limit. This has a crucial effect on the concentrations of elements near the detection limit such as Cd or dissolved Cr concentrations and might lead to an overestimation of the dissolved fraction. A comparison of the mean and median concentrations of a HWY runoff shows that the median Cd concentration was as high as the detection limit (1.0 μg/L) and only the mean concentration was slightly higher (1.6 μg/L) (Lange et al., 2003).

Some researchers selected the storm events used for analysis. The *main criterion for the selection* is associated with ADP to distinguish between different events or to discard several rain events. In the first case, different lengths of time between 30 min (Steiner et al., 2008) and 10 h (Barett et al., 1998) of dry weather are used to separate two events and the concentration of each event is used for the calculation of mean/median values. In the second case, an ADP between 24 h (Moy et al., 2003; Wilson, 2006) and up to five days was maintained before stormwater sampling was deployed (Hares and Ward, 1999). This approach, together with the influence of the ADP on runoff concentrations (longer ADP leads to higher concentrations, cf. Section 3.1), might lead to higher metal concentrations in a runoff in comparison with measurements obtained without a selection. A minimum rain depth, which varied between 1.5 mm (Steiner et al., 2008), 3.8 mm

(Nason et al., 2012), 5.1 mm (Horwatich and Bannerman, 2009), 6.4 mm (Wilson, 2006), and 13 mm (Brattebo and Booth, 2003), is considered to ensure that sufficient and representative sample volumes could be collected. Nason et al. (2012) considered a minimum rain duration of 1 h.

Further influencing factors involve data processing and plausibility tests for all relevant data such as flow measurements, clock time adjustments, and communication among sampling devices.

The most important criterion for representative datasets must be a sampling of a full range of seasonal conditions for a complete range of rainfall intensities, depths, and durations (Thomson et al., 1997; Moy et al., 2003; Stagge et al., 2012; Tromp et al., 2012) and all sources of critical pollutants should be considered (Sansalone and Buchberger, 1997b). However, most studies have been performed within a single year or a part of a year and their conclusions often do not reflect the seasonality and environmental variability of the site (Uhl and Kasting, 2002; Steiner and Goosse, 2011a).

Overall mean and median values presented in previous studies, which were calculated using different methods and should describe the real water quality (Fig. 1), are not assumed to be comparable. To prove the correctness of this hypothesis, mean and median concentrations that are both available for each HWY site and contain a minimum of three events are presented in Fig. 2. For the mean datasets, more extreme values and outliers are computed (for all four elements and for the dissolved and total fractions) in comparison with the median values. Therefore, for this review, mean and median datasets are not combined and are used separately to characterize the central tendency and variability of runoff concentrations among sites or site categories.

4. Occurrence and partitioning

4.1. Historical trends

Historical trends of Pb and Zn in traffic area runoff published in literature were analyzed from the 1970s to date. As described by Kayhanian (2012), for HWY runoff, Pb concentrations have been decreasing continuously in the last few decades. The decrease of Pb use as an antiknocking agent started in the USA in the mid-1980s and was completed in 1996 (FHWA, 1999). In Europe, the situation was not as homogenous as in the USA. In Germany, the beginning of the phase-out of leaded gasoline began in 1976, when the permitted concentration of Pb in gasoline decreased sharply to 150 mg/L (Golwer and Schneider, 1983). The phase-out in other countries of Western Europe started at the latest in 1986 (Berbee et al., 1999) and ended with the elimination of leaded gasoline in Italy in January 2002 (Mangani et al., 2005). At the same time, leaded gasoline was banned in China (Gan et al., 2008). Further reductions in Pb runoff concentrations are related to the substitution of Pb in other sources such as tires and brake linings (Sansalone et al., 1996; Legret and Pagotto, 1999; Davis et al., 2001), lubricating oil and grease (Ball et al., 1998), weights added to vehicles for tire balance (Root, 2000; Bleiwas, 2006), and Pb-based paints on roads and HWY (Mosley and Peake, 2001; Preciado and Li, 2005; Masanao et al., 2006). However, tires still contain Pb (Shinya et al., 2006; McKenzie et al., 2009).

A decline in Pb concentrations can be observed in the dataset of this review. Because of different numbers of measurements per year, the collected data were aggregated into decades to ensure that there were sufficient data for statistical analysis. Less data is available for the 1970s and 2010s compared to the other three decades (Fig. 3). The decline of Pb starting in the 1980s correlates with the statements mentioned above and is comparable to the one presented by Kayhanian (2012) for HWY runoff. Overall median concentrations of Pb in the 1980s are 252 μg/L (for all median values) and 200 μg/L (for all mean values). In the 2000s, overall median concentrations are one order of magnitude lower at 18.0 μg/L (for all median values) and 19.2 μg/L (for all mean values). All data is summarized in Table 4. The highest concentrations in the 21st century illustrated in Fig. 3 were measured in Poland by Klimaszewska et al. (2007), South Korea by Maniquiz-Redillas and Kim (2014), and China by Zhao et al. (2001). To increase the benefit of all other data analysis, only Pb data for the 21st century were used. These Pb concentration datasets shown in Figs. 5 and 6 are highlighted by the use of the index “21”. However, in this selected dataset, artifacts caused by the wash-off of Pb deposited on the upper part of road soils cannot be excluded completely.

For Zn, the trend is different. Neither a significant decline nor an increase in measured concentrations can be identified for the last few decades (Fig. 3). For the medians, overall concentrations of Zn in the 1980s are 264 μg/L (for all median values) and 292 μg/L (for all mean values). In the 2000s, overall median concentrations are 162 μg/L (for all median values) and 215 μg/L (for all mean values). All data is summarized in Table 4. Kayhanian et al. (2007) compared different Zn datasets obtained in California and concluded that there was a decrease in measured concentrations. In contrast, an increase in runoff concentrations is proposed as a result of an increasing use of Zn for safety fences (Dierkes and Geiger, 1998) and of galvanized car parts containing a Zn plating that should prevent corrosion (Sansalone and Buchberger, 1997a). In addition to these increasingly common applications, Zn has been used for decades in several structural elements, batteries (Davis et al., 2001), tires (Van Bohemen and Van de Laak, 2003; McKenzie et al., 2009), brake linings (Legret and Pagotto, 1999), motor oils and grease (Ball et al., 1998). Consequently, Legret and Pagotto (1999) indicated that Zn concentrations remain highly variable depending on the use of safety barriers made from galvanized steel and site-specific factors that have an influence on tire and brake wear. This is confirmed by the occurrence of several outliers and extreme values shown in Fig. 3. The highest values in the 21st century are affected by the corrosion of galvanized elements (Wilson, 2006; Gan et al., 2008) or by highly trafficked and congested roads (Helmreich et al., 2010; Moores et al., 2010) and HWY (Lau et al., 2009; Kluge et al., 2014). Preciado and Li (2005) identified Zn as a metal of future potential concern because it was second in concentration to Fe in most dust, air, and water samples.

4.2. Variability of Zn

Zn concentrations are very variable in traffic area runoff compared with other heavy metals as shown in Section 4.1. In recent years, most monitoring programs measured Zn and Cu, and frequencies of Zn-to-Cu-ratios are calculated for all datasets containing median values ($n = 141$, Fig. 4) to show the variability of Zn compared to Cu.

Calculation of Zn-to-Cu-ratios was first performed by Moores et al. (2010) for total median concentrations of HWY runoff in New Zealand. The calculated ratios in untreated runoff waters were within the range of 4–5 at all four sites (6–8 rain events per site; active-automatic and flow-proportional sampling) except at one site with porous asphalt that had a lower median concentration ratio of 2.4.

The most frequent ratios of Fig. 4 are between 3 and 5 with a larger deviation ($\text{mean} = 8.1$, $\text{SD} = 10.9$). For the highest ratios, either sample contamination or special site conditions are responsible. The highest value of 76.5 was calculated for a PL runoff that received water from a slate roof with Zn gutters (Gnecco et al., 2005). The ratio of 70.5 could be explained by the presence of a nearby Zn smelter (Driscoll et al., 1990), the ratio of 30.8 by vehicle maintenance (Line et al., 1997), and the ratio of 24.5 by the surrounding old residential district (Gromaire-Mertz et al., 1999). The ratio of 23.4 is correlated with a failure of galvanizing material of a BR (Wilson, 2006). Mangani et al. (2005) described the presence of safety fences at a construction site with a nearby agricultural land use, where metal-containing pesticides are used, that might have led to the high Zn-to-Cu-ratio of 22.7. Further ratios might be influenced by the presence of guardrails (ratios 14.0 and 16.5) (Gäth et al., 1990; Nadler and Meißner, 1999), and the erosion of galvanized utilities and noise barrier walls (ratio 14.8) (Gan et al., 2008). The Zn-to-Cu-ratios of 24.9, 34.2, 38.0, and 61.0 are caused by a temporal contamination of samples by a Zn-containing anti-corrosive paint have (Dierkes, 1999).

Table 5

Statistics of Figs. 5 and 6 — mean metal concentrations (dissolved and total) in parking lot (PL), road (RL, RM, and RU), bridge (BR), and highway (HL, HN, and HU) runoff.

Element	Traffic area category	Dissolved concentrations										Total concentrations									
		Min	Median	Geometric Mean	Mean	75th	90th	95th	Max	SD	95%[a]	Min	Median	Geometric Mean	Mean	75th	90th	95th	Max	SD	95%[a]
Pb_21	PL	1,0	1,7	2,5	4,3	8,0	12,0	[b]	15,9	4,6	2,6	2,5	16,3	15,5	23,1	36,3	59,8	65,8	66,0	19,5	9,1
Pb_21	RL	–	–	–	–	–	–	–	–	–	–	2,0	27,0	21,5	61,5	123	[b]	[b]	152	66,2	61,3
Pb_21	RM	0,13	0,4	0,4	0,9	2,3	[b]	[b]	2,8	1,3	2,0	3,7	20,3	18,3	32,3	30,5	[b]	[b]	136	43,1	36,0
Pb_21	RU	0,5	4,0	2,7	3,9	6,5	[b]	[b]	7,0	2,8	4,4	6,0	44,5	38,8	79,3	71,0	331	[b]	380	112	67,4
Pb_21	BR	0,13	0,14	0,4	1,4	[b]	[b]	[b]	3,8	2,1	5,3	24,0	41,7	42,5	49,9	83,7	[b]	[b]	92,3	32,3	51,4
Pb_21	HL	0,01	1,0	0,3	1,3	2,6	[b]	[b]	3,1	1,3	1,3	2,5	25,0	32,1	64,4	118	212	[b]	230	73,1	49,1
Pb_21	HN	12,8	12,8	12,8	12,8	[b]	[b]	[b]	12,8	[b]	[b]	4,4	16,0	18,7	31,7	64,5	[b]	[b]	90,0	34,1	35,8
Pb_21	HU	0,8	2,1	2,3	3,0	4,9	[b]	[b]	7,4	2,4	2,6	1,4	12,6	16,8	33,1	39,7	81,7	155	220	44,9	16,5
Zn	PL	12,0	60,4	56,1	77,2	113	157	293	317	69,1	30,6	39,0	178	155	201	295	397	586	620	147	53,7
Zn	RL	15,0	24,0	42,6	76,2	119	[b]	[b]	314	98,8	76,0	25,0	149	141	212	272	525	[b]	940	219	109
Zn	RM	7,9	31,0	41,4	68,0	73,0	226	[b]	258	77,2	42,8	23,0	274	203	285	384	533	886	1000	217	89,6
Zn	RU	51,0	78,3	97,7	113	159	[b]	[b]	262	71,6	55,1	120	351	379	474	556	982	1805	1940	397	176
Zn	BR	50,0	148	191	354	646	[b]	[b]	1226	432	332	77,0	498	611	2231	700	15,572	[b]	19,100	5607	3766
Zn	HL	5,0	66,5	54,3	76,9	108	177	[b]	191	53,0	30,6	32,3	217	207	306	371	642	1540	1760	359	155
Zn	HN	8,6	66,8	90	204	453	[b]	[b]	577	231	177	52,5	216	248	385	388	1202	1875	2210	477	193
Zn	HU	11,0	50,0	77,1	217	166	595	1838	2118	449	194	21,0	213	237	338	414	682	986	2234	358	103
Ni	PL	1,8	3,0	3,2	3,8	4,4	10,2	[b]	12,4	3,0	1,9	2,1	7,1	10,5	23,7	35,0	86,9	[b]	145	37,0	19,7
Ni	RL	3,1	4,7	4,5	4,7	[b]	[b]	[b]	6,3	2,2	20,2	8,4	13,9	12,5	13,1	[b]	[b]	[b]	16,9	4,3	10,7
Ni	RM	0,5	1,0	0,9	0,9	[b]	[b]	[b]	1,3	0,4	1,0	3,8	17,0	12,7	16,3	23,0	[b]	[b]	35,0	11,2	11,3
Ni	RU	9,5	10,5	10,5	10,5	[b]	[b]	[b]	11,5	1,4	12,7	4,1	9,8	13,8	20,5	40,4	[b]	[b]	55,0	20,2	21,2
Ni	BR	3,0	4,8	8,3	15,9	35,5	[b]	[b]	49,0	19,5	20,5	6,0	26,7	28,0	82,0	65,0	[b]	[b]	458	154	129
Ni	HL	–	–	–	–	–	–	–	–	–	–	6,0	12,0	15,6	22,6	35,2	[b]	[b]	73,0	25,3	26,5
Ni	HN	4,0	11,0	10,6	14,0	[b]	[b]	[b]	27,0	11,8	29,3	4,0	20,0	16,1	29,3	57,0	[b]	[b]	83,0	30,8	23,7
Ni	HU	1,9	9,6	9,9	14,9	28,3	[b]	[b]	28,8	12,5	15,6	2,0	8,7	9,5	19,2	31,5	68,8	[b]	93,0	25,1	12,1
Cu	PL	2,7	9,0	9,4	11,8	13,5	28,5	34,3	35,0	8,7	3,7	5,0	19,6	23,4	40,7	56,0	116	165	220	49,2	15,4
Cu	RL	3,3	11,0	14,5	20,3	37,8	53,2	[b]	56,0	17,9	11,4	6,0	48,0	37,5	53,7	77,4	104	[b]	180	43,9	21,2
Cu	RM	2,7	11,2	11,2	16,0	23,0	35,0	[b]	65,0	14,9	7,2	7,0	30,5	38,6	64,6	100	175	264	280	71,3	31,6
Cu	RU	6,8	23,0	22,2	26,4	39,0	55,0	[b]	56,8	15,9	9,6	26,0	86,9	86,9	105	149	197	275	288	66,6	29,5
Cu	BR	16,0	23,5	24,7	26,1	31,1	[b]	[b]	47,0	9,9	8,3	20,0	54,0	54,9	63,9	93,2	133	[b]	136	36,1	25,8
Cu	HL	5,7	18,0	18,9	23,3	31,1	56,9	[b]	64,0	16,4	9,9	13,3	48,0	49,4	60,7	91,7	124	138	140	38,8	17,2
Cu	HN	4,0	28,7	25,8	34,6	51,5	88,0	[b]	100	26,9	17,1	23,0	53,0	61,8	84,4	97,0	205	394	430	90,4	39,1
Cu	HU	4,1	15,4	18,8	35,5	49,1	122	144	151	42,0	17,3	13,0	56,4	51,4	63,5	74,7	118	145	274	45,5	13,2
Cd	PL	0,05	0,3	0,3	0,4	0,6	1,6	[b]	2,0	0,5	0,3	0,2	0,8	0,9	3,0	2,0	5,6	36,6	40,0	8,6	3,9
Cd	RL	0,14	0,4	0,5	0,7	1,2	[b]	[b]	1,8	0,6	0,6	0,3	1,6	1,5	2,7	2,8	12,0	[b]	13,0	3,8	2,7
Cd	RM	0,01	0,2	0,1	0,2	0,3	0,4	[b]	0,5	0,1	0,1	0,06	0,6	0,7	3,2	1,3	14,1	[b]	37,0	9,1	4,8
Cd	RU	0,25	0,6	0,5	0,6	0,7	[b]	[b]	0,9	0,2	0,2	0,67	2,0	2,7	5,6	6,5	22,0	[b]	25,0	7,8	4,5
Cd	BR	0,04	0,5	0,3	0,5	1,0	1,0	1,0	1,0	0,4	0,4	0,28	3,5	3,9	10,3	15,0	43,8	[b]	47,0	14,4	10,3
Cd	HL	0,02	0,1	0,1	0,5	0,8	[b]	[b]	2,6	1,0	1,1	0,05	0,9	0,7	1,8	1,9	7,6	[b]	9,0	2,6	1,6
Cd	HN	0,50	1,1	1,2	1,4	1,8	[b]	[b]	3,0	0,8	0,8	0,14	1,3	1,2	2,6	4,6	6,2	11,6	11,9	3,0	1,4
Cd	HU	0,10	0,9	0,8	1,3	1,7	[b]	[b]	3,8	1,2	1,1	0,06	1,7	1,6	4,1	2,4	12,5	31,3	35,0	7,9	2,9
Cr	PL	1,0	1,5	1,6	1,7	2,2	2,7	[b]	2,7	0,6	0,4	1,5	8,6	8,6	13,1	21,0	26,4	50,4	56,0	12,5	5,4
Cr	RL	1,4	1,8	1,7	1,8	[b]	[b]	[b]	2,2	0,6	5,3	5,0	11,5	10,4	11,2	15,3	[b]	[b]	16,0	4,3	4,5
Cr	RM	0,6	1,2	1,0	1,2	[b]	[b]	[b]	1,8	0,8	7,6	2,0	9,9	9,0	12,0	24,2	[b]	[b]	24,2	8,9	8,3
Cr	RU	3,0	3,0	3,0	3,0	[b]	[b]	[b]	3,0	[b]	[b]	3,6	12,9	11,6	13,2	17,8	[b]	[b]	23,0	6,4	6,7
Cr	BR	1,3	3,2	3,3	4,0	7,1	[b]	[b]	7,5	2,6	2,7	7,5	11,0	26,7	83,5	140	[b]	[b]	381	140	129
Cr	HL	–	–	–	–	–	–	–	–	–	–	4,9	12,5	14,4	18,6	38,0	[b]	[b]	40,4	14,4	13,3
Cr	HN	0,5	11,3	3,3	11,3	[b]	[b]	[b]	22,1	15,3	137	2,7	9,3	12,7	25,0	36,8	88,7	[b]	89,0	33,4	23,9
Cr	HU	1,5	3,2	3,3	4,5	8,0	[b]	[b]	11,9	4,3	5,3	1,0	7,3	8,6	17,9	13,8	69,3	103	105	26,8	12,5

[a] 95% confidence errors on the means.

[b] No values because of small universe.

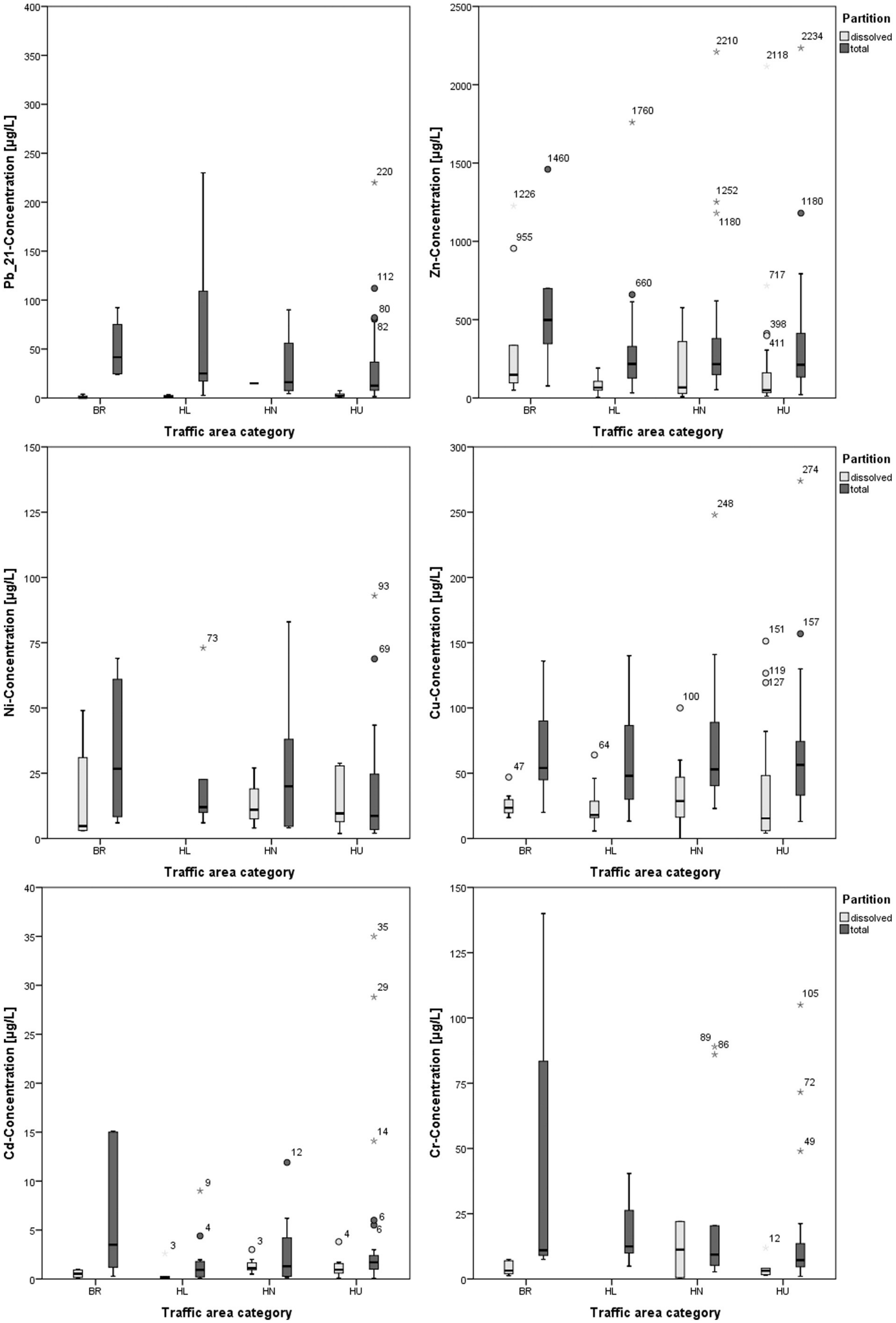

Fig. 6. Mean metal concentrations (d and t) in bridge (BR) and highway (HL, HN, and HU) runoff.

Table 6
Elements that are detected in sampling programs and that are related to traffic or might be used as a tracer for anthropogenic sources.

Element	Literature (partition)
Co	Dannecker et al. (1990) (p, t); Hares and Ward (1999)(t); Bäckström et al. (2003) (d, t); Wilson (2006) (d, t); Hallberg et al. (2007) (d); Herrera (2007) ([a]; d, t); Steiner and Goosse (2007) (t); von Ballmoos (2007) (t); Grotehusmann and Kasting (2009) (t)
Mn	Ellis and Revitt (1982) ([a]; d, t); Golwer and Schneider (1983) (t); Harrison and Wilson (1985) (d, t); Dannecker et al. (1990) (p, t); Sansalone and Buchberger (1997a) (d, t); Booth and Leavitt (1999) (d); Brockbank et al. (1999) (d, t); Hares and Ward (1999) (t); Shinya et al. (2000) (t); Rushton (2001) (t); Bäckström et al. (2003) (p); Shinya et al. (2003) (t); Preciado and Li (2005) (d, t); Hallberg et al. (2007) (d); Gnecco et al. (2008) (t); Hatt et al. (2009) (t); McQueen (2010) (t)
PGE	Wei and Morrison (1994) (Pt; d, t); Laschka et al. (1996) (Pt; t); Grotehusmann and Kasting (2002) (Pd; d, t; Pt; d, t); Moy et al. (2003) (Pd; t); Moy and Crabtree (2003) (Pt; t); Hilliges (2007) (Rh; t); Kasting and Grotehusmann (2007) (Pt; d, t); Nadler and Meißner (2007) (Pt; t); Windhofer et al. (2012) (Pt, Rh; t)
Sb	Dannecker et al. (1990) (p, t); CH2MHILL (1998) (t); Hares and Ward (1999) (t); Wilson (2006) (d, t); Herrera (2007) ([a]; t, d); Nadler and Meißner (2007) (t); Stachel et al. (2007) ([a]; t); Scheiwiller (2008) (t); Grotehusmann and Kasting (2009) (t); Steiner and Goosse (2009) (t)
Ti	Bourcier et al. (1980) (t); Golwer and Schneider (1983) (d, t); Fürhacker et al. (2013) (t)
W	Bourcier et al. (1980) (t); Bäckström et al. (2003) (d, t)

[a] Literature study.

The explanation for low Zn-to-Cu-ratios related to porous asphalt could only be confirmed by one Zn-to-Cu-ratio with a value of 1.2 (Berbee et al., 1999). For comparison, mean ratios of porous asphalt vary between 1.0 and 3.9 with one exception of a ratio of 8.5 (Stotz and Krauth, 1994), which strengthens the conclusion of Moores et al. (2010). Examples of low median ratios resulting from higher Cu concentrations could not be identified.

In conclusion, the calculation of median Zn-to-Cu-ratios can be used to evaluate datasets to detect possible sample contamination and to emphasize the importance of detailed site descriptions.

4.3. Characteristics of traffic area categories

The data for all traffic areas are subdivided into eight categories, as described in Section 2. The data for five categories, including PL, different roads without HWY, and BR, which are presented for the first time, are shown in detail in Tables 1, 2, and 3.

In Table 1, 35 PL are included, of which two were investigated in two monitoring programs (PL16a + b and PL31a + b). Most PL samples were collected in the USA; some samples were obtained in Europe (Germany, Austria, France, Italy, Poland, and Sweden). Only one PL runoff was collected in each of Australia and South Korea.

Generally, PL have low traffic densities. Thus, even in high-use parking areas such as at supermarkets, vehicle changes only occur up to four times per hour and might not exceed 50 vehicles per day per parking slot (Göbel et al., 2007). However, drip losses, tire and brake wear due to braking and steering activities, and exhaust gas emissions because of acceleration are higher compared with non-congested roads. Therefore, most of the Cu loads in runoff waters from industrial land uses originate from PL (Bannerman et al., 1993; Steuer et al., 1997) and PL runoff are a dominant contributor of total Zn and Cd loads in urban runoff (Steuer et al., 1997). Another factor is the contamination of parked vehicles (Pick et al., 2002). PL maintenance includes regular sweeping and cleaning of drains about three times a year (Grotehusmann and Kasting, 2002).

All types of road categories with an AADT of <5000 (RL, 21 sites), 5000 < AADT < 15,000 (RM, 33 sites), and >15,000 (RU, 26 sites) are presented in Table 2.

For RL, most published data are for US sites, with four sites in Germany and one site each in France, the Netherlands, Australia, and South Korea. The land uses are mostly urban and residential but also rural (RL07–RL10), industrial (RL03 + RL18), non-urban desert (RL15), and non-urban agricultural (RL16). RL mostly includes residential, feeder, and smaller collector streets, often with sidewalks and sometimes with side parking zones and curbs. Urban sites have single-family houses and two- to six-story buildings nearby. For RM, 14 datasets have been published for US sites, eight for German sites, and a total of twelve for sites in eight other countries. The land uses are mostly urban and residential but also include rural (RM11, RM13, RM16–18) and non-urban agricultural (RM23) uses. RM include collector roads, residential driveways, and other medium-traffic major roads with mostly two lanes, curbs, and limited on-street parking. RL and RM can be maintained by daily sweeping of street gutters and pressurized washing of sidewalks and gutters several times a week (Gromaire-Mertz et al., 1999). For porous asphalts pressurized water spraying and suction to prevent clogging is used (Legret et al., 1996; Dierkes et al., 1999). For RU, ten sites are located in Germany, three in Poland, three in the USA, and a total of ten sites in six other countries. All sites are highly trafficked arterial roads with almost no hard shoulders. The number of lanes varies between two and five for urban sites, which sometimes have medians, and only RU08 is a two-lane rural road with hard shoulders.

All BR are summarized in Table 3. 21 of the 39 sites are from a huge Australian study by Drapper et al. (2000), and 13 sites are located in the USA. One site in France was monitored with original asphalt surface and after renovation with porous asphalt (BR01a + b), one site in Sweden was selected to evaluate seasonal variations (summer/winter; BR02a + b), one site in Canada was monitored in two different decades (BR16a + b), and two sites are located in China. BR are monitored because contaminant sources are assumed to be exclusively traffic-related (Wilson, 2006), minimal infrastructure modifications are required for sampling (Drapper et al., 2000), and sites are better protected against vandalism (Marsalek et al., 1997). BR decks are intensively maintained (Wu et al., 1998).

All HWY sites are classified depending on the AADT according to Strecker et al. (1990). For HWY with an AADT of >30,000, all sites are distinguished between urban (HU, 64 sites) and non-urban (HN, 33 sites) land uses, which have been proposed to be the only factor having a statistically significant influence on pollutant concentrations for US datasets (Strecker et al., 1990; Kayhanian et al., 2007; Nason et al., 2012). The dataset also includes 35 HL sites. Data are available for five of six continents (no data for South America); 47 of the 132 sites are located in the USA, 24 in Germany, and eleven in each of Switzerland and the United Kingdom. The main difference between RU and HWY in European countries is that the latter mostly have hard shoulders and roads may have traffic signals in contrast to HWY.

4.4. Runoff concentrations of traffic area categories

All published mean metal concentrations (d and t) in PL, RL, RM, and RU runoff for Pb (n = 74), Zn (n = 154), Ni (n = 55), Cu (n = 164), Cd (n = 103), and Cr (n = 64) are plotted in Fig. 5 (Table 5). As datasets are not plotted because of fewer available data (n = 24 with 18 for PL). In most cases, the overall median concentrations for PL are lower than those for roads. Differences in pollution are related to the type of PL use: Employee PL (mostly once-in and once-out per day usage, PL31) are less polluted than highly frequented car PL (supermarket, PL05) and PL for trucks are higher loaded for almost all parameters in comparison to car PL (Grotehusmann and Kasting, 2002). RU is the road category with the highest runoff concentrations for all metals except Ni. Zn has the highest total runoff concentration, followed by Cu, Pb, Ni, Cr, and Cd. The highest dissolved concentrations have Zn and Cu, followed by Ni, Pb, Cr, and Cd. Thus, Pb and Cr are mostly in the particulate phase, whereas Ni and Cd are almost dissolved. For PL, median values for As (2.1 μg/L t; 1.1 μg/L d) are higher than those for Cd (0.8 μg/L t; 0.3 μg/L d). The total concentrations of PL08 (only one event analyzed in contrast to the dissolved values) and the very high values of RL21 are not plotted in Fig. 5.

In Fig. 6, all published mean metal concentrations (d and t) in BR, HL, HN, and HU runoff are plotted separately (Table 5). For BR, the number of datasets varies between seven for Pb and 20 for Zn. For HWY runoff, much more data are available for Pb (n = 61), Zn (n = 144), Ni (n = 42), Cu (n = 142), Cd (n = 83), and Cr (n = 44). For BR, the order of total and dissolved concentrations is equal to the ones presented in Fig. 5 but the Zn concentrations of BR runoff are higher compared with all other categories (cf. Sections 3.1, 4.2, and 4.3). Median values of total Cr concentrations (n = 13) are very high compared with all other datasets. This might be related to the generally very high total concentrations of BR11 (19,100 μg/L Zn, 1860 μg/L Pb, 140 μg/L Cr) and to specific conditions in China (BR38, BR39; 26.8–381 μg/L Cr). From a comparison of two monitoring programs of a French BR with asphalt and after renovation using porous asphalt (BR01a + b), it can be concluded that heavy metal runoff concentrations of porous asphalt are markedly lower and the type of BR deck surface does not affect heavy metal partitioning. A comparison of the effects of winter and summer on the runoff concentrations of a BR in Sweden (BR02a + b) showed higher mean concentrations during winter, and a comparison of two Canadian BR runoff (BR16a + b) indicated a sharp historical decrease in Zn and Pb concentrations and an increase in Cu and Ni concentrations during the 1990s.

For HWY, no significant difference among the three categories HL, HN, and HU could be detected. This contrasts with the conclusions obtained using US datasets that HL and HN have lower runoff concentrations compared with HU (Strecker et al., 1990). For comparison with this dataset, note that in this evaluation, all HN of US sites are in category HL. Thus, in HN, only European and two New Zealand sites are included. According to Strecker et al. (1990), the concentrations of HL should be lower than those of HN and HU. However, by considering worldwide data, the correlation among HWY runoff concentrations, AADT, and land use could not be verified. Thus, other site-specific aspects such as hard shoulders, median strips, noise barriers, and grade of congestion might be better indicators to characterize HWY runoff concentrations. This is confirmed by a comparison of RU data with HWY datasets. For RU, nearly all overall heavy metal median concentrations are higher in comparison to each of the three HWY categories. For special site conditions such as for RM03, the runoff concentrations of an urban road can be higher than most measurements of HWY because of braking and acceleration activities at traffic signals. The results of an evaluation of runoff concentrations of a HWY construction site not yet in use with two other sites on the same HWY showed that Zn runoff concentrations of the construction site are sometimes higher compared with the other two sites and that Cu and Pb concentrations of the construction site are lower (Mangani et al., 2005). This demonstrates the importance of site-specific factors for HWY runoff.

An analysis was performed to examine the hypothesis that a lower pH of the runoff corresponds to a higher proportion of dissolved metals (Mikkelsen et al., 1994; Sansalone and Buchberger, 1997b; Genç-Fuhrman et al., 2007). The dissolved concentrations in runoff samples are influenced by the capacity for rainwater and pH-modified runoff (cf. Section 4.6) to dissolve metals from a surface, which is influenced by kinetic factors, and by the rate of dissolution, which increases as pH decreases. An acidic pH of one entire event confirms the ionic form for Cu as most relevant (Gnecco et al., 2008). However, an evaluation of datasets for the percentages of Cu and Zn dissolved at the prevailing runoff pH could not confirm the speciation proposed by Genç-Fuhrman et al. (2007). The dissolved fraction of Cu in runoff from different sites was analyzed as 52% at a pH of 6.2 (Caltrans, 2003), 81% at a pH of 6.7 (Lau et al., 2009), and 72% at a pH of 8.0 (Mangani et al., 2005). The dissolved Cu fraction varies between 1% at a pH of 7.6 (Pitt et al., 1995) and 93% at a pH of 7.2 (Golwer and Schneider, 1983). For Zn, the dissolved fraction varies between 2% at a pH of 8.1 (Mangani et al., 2005), 11% at a pH of 7.1 (Lange et al., 2003), and 91% at a pH of 6.7 (Lau et al., 2009). Thus, nearly no decrease of the dissolved fraction of Zn and Cu can be found in the pH range between 6.2 and 8.1, and the hypothesis could not be confirmed. Potential reasons include the likelihood that dissolution is kinetically limited, the relative independence of dissolved and particulate sources, or differences in the sample preparation and analysis.

The results from all datasets show that Cu and Zn mostly exhibited intermediate behavior (Harrison and Wilson, 1985; Dean et al., 2005; Herrera, 2007; Kayhanian et al., 2007) but sometimes, they were more dissolved (Timperley et al., 2005; Wilson, 2006; Hilliges et al., 2013). The concentrations of Pb and Cr are mostly in the particulate phase; this was confirmed in several individual monitoring programs (Harrison and Wilson, 1985; Shinya et al., 2000; Timperley et al., 2005; Herrera, 2007; Kayhanian et al., 2007; Kayhanian, 2012; Hilliges et al., 2013). Ni (Shinya et al., 2000) and Cd (Wilson, 2006) are mainly in the dissolved form, although they are sometimes equally partitioned between the two phases (Harrison and Wilson, 1985; Dean et al., 2005; Kayhanian et al., 2007). The particulate fraction of heavy metals might be higher for snowmelt events than rain events (Westerlund et al., 2003). Bäckström et al. (2003) confirmed this for Pb and Zn, although other metals remained at a constant fraction during winter, and Hallberg et al. (2007) determined higher dissolved fractions of Cd, Cr, and Ni in winter compared to summer, whereas no significant differences occurred for Cu, Pb, and Zn. Helmreich et al. (2010) concluded that the fractionation of the heavy metals was not affected by seasonal variations but remarkable fluctuations were observed between different rain events with dissolved fractions greater than 90%. Similar results were confirmed by Dauber et al. (1979). However, Dean et al. (2005) stated that the partitioning of Pb was unaffected by flow rate.

4.5. Other elements

Other elements that are detected in sampling programs and that are related to traffic or might be used as a tracer for anthropogenic sources are listed in Table 6. As tracers, Co and W are potentially suitable for roads and traffic because of their low natural concentrations in environmental waters (Bäckström et al., 2003). Nadler and Meißner (1999) and Huang et al. (1994) suggest that Sb should be used as a marker, too.

Co is mainly used in steel and paints (Makepeace et al., 1995) and in diesel (Roeva et al., 1996). Steiner et al. (2006) concluded that traffic is not the main source for Co in soils, whereas Ward (1990) observed increased levels of Co in soils because of traffic activities. Bäckström et al. (2003) clearly demonstrated that roads act as a source for Co. Total mean concentration of Co varies between 1.5 μg/L (Grotehusmann and Kasting, 2009) and 13.3 μg/L (Bäckström et al., 2003) and dissolved mean concentrations are between 0.1 μg/L (Dannecker et al., 1990) and 15.1 μg/L (Hallberg et al., 2007); dissolved fractions vary between 4.1% (Dannecker et al., 1990) and 67% (Bäckström et al., 2003). The highest mass of Co (around 94%) in runoff was transported during winter at one site in Sweden (Bäckström et al., 2003).

Bourcier et al. (1980) measured W, which is used in tire studs, in concentrations between non-detectable (2.5 mg/L) and 25 mg/L in road runoff (completely in the particulate phase) and did not detect it in precipitation and dust samples. Bäckström et al. (2003) measured W at two sites in summer and winter and found that the particulate fraction is low and the concentrations increased by a factor of ten during winter (5.8–9.2 μg/L).

Sb is a very toxic element, and the main traffic-related sources are brake pads (Von Uexküll et al., 2005). Concentrations of Sb have been presented in ten publications (cf. Table 6). Total mean Sb concentrations are between 0.8 μg/L (Dannecker et al., 1990) and 10.7 μg/L (CH2MHILL, 1998). Dissolved concentrations were analyzed at three sites and vary between 1.2 μg/L (Dannecker et al., 1990) and 5.05 μg/L (Wilson, 2006).

Anthropogenic sources of Mn are moving engine parts (Ball et al., 1998; Drapper et al., 2000), gasoline additives such as methylcyclopentadienyl manganese tricarbonyl (MMT) (Young et al., 1996; Preciado and Li, 2005), brake wear (Ward, 1990; Sansalone et al., 1996), and tire wear (Hares and Ward, 1999). Values of Mn concentrations in runoff from different sites have been presented in 17 publications (cf. Table 6). Shinya et al. (2000) reported that Mn is mainly particle-bound, whereas Sansalone and Buchberger (1997a) and Harrison and Wilson (1985) reported that it is mainly dissolved. Total mean concentrations of Mn vary between 11.1 μg/L (Rushton, 2001) and 659 μg/L (Gnecco et al., 2008) (both PL), and dissolved mean concentrations are between 9 μg/L (Booth and Leavitt, 1999) and 255 μg/L (Sansalone and Buchberger, 1997a). Ward (1990) measured elevated levels of Mn in soils with increased traffic density.

Of the PGE only Rh, Pd, and Pt are related to traffic sources. Since the end of the 1980s, these three elements have been primarily used as catalytic converters and the number of automobiles equipped with these converters has increased sharply (Wei and Morrison, 1994). A three-way catalytic converter contains a total of 2–3 g of Rh, Pd, and Pt (Geiger-Kaiser and Jäger, 2005). Fritsche and Meisel (2004) determined the distribution of PGE in soils along major Austrian HWY and concluded that Rh, Pd, and Pt significantly exceeded natural background values. In road runoff, Rh was only detected by Hilliges (2007) at a concentration of 1.14 μg/L and by Windhofer et al. (2012) at a concentration of 0.08 μg/L. Pt concentrations vary between 0.0065 μg/L (Wei and Morrison, 1994) and 24 μg/L (Moy and Crabtree, 2003), and Pd concentrations are between 0.34 μg/L (Moy et al., 2003) and 8 μg/L (Grotehusmann and Kasting, 2002). The dissolved phases of Pd and Pt are high (65%–81%, Grotehusmann and Kasting, 2002). According to Laschka et al. (1996), Pt runoff concentrations depend on ADP, rain intensity, and rain duration.

Ti was only measured in three studies, and its main source is the wear of white road marking paint containing titanium dioxide as pigment (Makepeace et al., 1995), which is often renewed once a year (Golwer and Schneider, 1983). Another source is tire studs (Bourcier et al., 1980). Ti was not measured in precipitation samples by Bourcier et al. (1980) but was measured in road runoff with a mean concentration of 12.7 mg/L. Golwer and Schneider (1983) detected that Ti was mostly particulate and only 26 μg/L was dissolved. Fürhacker et al. (2013) measured Ti in one HU runoff sample with a total concentration of 630 μg/L.

4.6. Correlation between pH and type of surface

The partitioning of metals in runoff is strongly influenced by the pH of the rainfall, the solids present, the solubility of all substances, the pavement residence time, and the type of surface (Prestes et al., 2006). For asphalt, no significant level of alkalinity is provided to neutralize the pH of rainfall (Sansalone and Buchberger, 1997a), whereas a concrete pavement provides alkalinity to raise the runoff pH more than that of an asphalt pavement (Dierkes, 1999). Therefore, concrete surfaces should have a stronger influence on metal partitioning to the particulate phase than asphalt surfaces.

The evaluation of pH for 63 asphalt, 13 asphalt and concrete, 21 concrete, and four porous asphalt surfaces cannot provide significant differences. The median pH of asphalt surfaces was the lowest (6.8) and varied between 5.4 (Driscoll et al., 1990) and 8.2 (Zhang et al., 2013). A combination of asphalt and concrete surfaces had a median pH of 6.9 and varied between 6.4 (Lau et al., 2009) and 7.5 (Grotehusmann and Kasting, 2009). For concrete surfaces, a mean value of 7.2 was calculated, which is 0.4 units higher than the value for asphalt. The values varied between 5.6 (Driscoll et al., 1990) and 7.9 (Geiger-Kaiser and Jäger, 2005). For porous asphalts, the median value is 7.5.

The non-significant evaluation results of all datasets can be explained by two factors. First, the pavement residence times vary between each monitored site and samples were sometimes collected from a concrete channel that would have had an influence on the pH (Dierkes, 1999). Second, the pH of the rain itself can be different. Two-thirds of the measurements with concrete surfaces were conducted in the 1980s, when rain was more acidic. Conversely, the pH of the runoff of a concrete BR deck increased slightly with increasing flow rates (Dean et al., 2005). Drapper et al. (2000) compared asphalt and concrete BR decks and did not observe any variance in the pH or runoff concentrations. Aspects that might have an effect on porous asphalts have been described by Muschack (1990), and a literature review was provided by Barrett et al. (2006).

4.7. Continental trends

The runoff concentrations of Pb, Zn, and Cu that occur in the highest concentrations compared with other heavy metals were evaluated separately for each continent for which more than five datasets were available (i.e., Asia, Australia, Europe, and North America). For Pb, the recent concentrations are very low and the older, higher concentrations correlate well with the phase-out of leaded gasoline for all continents (cf. Section 4.1). The overall median Pb values for Europe and North America are approximately 30 μg/L and 15 μg/L for the 21st century. For Asia, both Pb values are approximately 50 μg/L, which correlates with the later Pb-phase out in Asia compared with Europe and North America (cf. Section 4.1). The Australian overall median Pb concentration is approximately five times higher than those of Europe and North America for the complete dataset and approximately three times for the dataset of the 21st century. The same results were obtained by Kayhanian et al. (2012) for HWY runoff, whereas the Asian HWY values consist out of one extreme value (Maniquiz et al., 2010). For Cu, the overall median concentrations are approximately 30 μg/L for North America and approximately twice as much for Asia, Australia, and Europe. For Zn, the overall median concentrations are approximately 170 μg/L for North America, 250 μg/L for Europe, 300 μg/L for Australia, and 620 μg/L for Asia. For Asia, it must be considered that fewer data are available compared with the other three continents and the data are mostly for heavily loaded traffic areas. In conclusion, the median values in North America are the lowest for Pb, Zn, and Cu, and the variance of Pb and Zn runoff concentrations among the continents is higher than the variance of Cu.

4.8. Planning stormwater treatment plants

Several factors concerning the occurrence and partitioning of heavy metals must be considered when planning stormwater treatment plants because the differentiation between the dissolved and particulate phases is of great concern for a successful treatment of stormwater to prevent negative effects on groundwater and surface water. As not all metals are present predominantly in the particulate phase (Cd, Ni, Zn, and Cu; cf. Section 4.4), an additional priority should be set for the reduction of dissolved metal concentrations in runoff treatment programs (Tiefenthaler et al., 2001). Because dissolved metals cannot be removed mechanically with solids (e.g., by sedimentation and filtration), a second treatment step (e.g., filter media) is necessary to reduce higher metal runoff concentrations (Hilliges et al., 2007; Steiner and Goosse, 2008; Minervini, 2010; Kayhanian et al., 2012). Maniquiz-Redillas and Kim (2014) investigated the partitioning of heavy metals in runoff and in the effluent of a stormwater treatment plant (manufactured eco-biofilter) and concluded that the fractionation of heavy metals plays an important role on the performance of the system and a reduction of dissolved metal concentrations is necessary. It is recommended that not only the initial runoff or the first 25% of the total runoff should be treated but the entire water volume (Flint and Davis, 2007; Maniquiz-Redillas and Kim, 2014). In addition, the individual site and climatic influencing factors (cf. Section 3.1) are relevant for the construction and operation of treatment plants to prevent negative toxic effects on ecosystems (Schueler, 2000; Nason et al., 2012; Dierkes et al., 2015). As discussed in Section 4.3, the treatment of runoff from large PL should be considered for an effective control of heavy metal loads. Furthermore, a proper control of BR runoff, runoff of congested streets, and arterial roads with traffic signals becomes a concern. A better knowledge of the historical trends (cf. Section 4.1) and seasonal variations of heavy metal runoff concentrations (cf. Section 4.4) can be used for optimization of treatment processes (Hallberg et al., 2007).

5. Conclusions

The conclusions of this review are summarized as follows:

- Trace element pollutant loads determined at a given site depend on the unique subset of site-specific (surrounding land use characteristics, traffic area site data, operational characteristics, and climatic factors) and method-specific factors (sample collection, preparation, analysis, and calculation). In literature, inconsistent conclusions have been obtained regarding how specific variables affect traffic area runoff. Thus, a combination of several interacting factors results in heavy metal runoff pollution, and these factors should be described in detail for each monitoring site. These factors should also be implemented in databases to allow further specifications in future.
- The calculation of median Zn-to-Cu-ratios can be used to detect possible sample contamination and relevant site-specific factors.
- Historical trends for Pb show a sharp decrease during the last decades, consistent with the phase-out of leaded gasoline; no historical trends were detected for Zn.
- The heavy metal runoff concentrations of PL differ widely according to their use (e.g., employee, supermarket, rest areas for trucks). BR deck runoff can contain high Zn concentrations from safety fences and galvanizing elements. Roads with more than 5000 vehicles per day are often more polluted than HWY because of other site-specific factors such as braking and acceleration at traffic signals. Worldwide HWY runoff concentrations are not significantly influenced by AADT and urban/non-urban land uses.
- Pb and Cr are mostly particle-bound, while Zn, Cu, Ni, and Cd occur at a higher fraction in the dissolved phase. Therefore, the treatment of runoff waters containing dissolved heavy metal pollutants by stormwater treatment strategies is important.
- Further traffic-related elements are Mn, Rh, Pd, Pt, and Ti. As tracers, Co, W, and Sb, which have anthropogenic sources, are potentially suitable for traffic areas.

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Belastung von Verkehrsflächenabflüssen mit Schwermetallen – ein europäischer Vergleich

Maximilian Huber, Antje Welker und Brigitte Helmreich

Regenwasserbewirtschaftung, Autobahnen, Einflussfaktoren, gelöste Schwermetalle, Parkplätze, Straßen

Verkehrsflächenabflüsse können mit einer Reihe von organischen und anorganischen Stoffen belastet sein. Zu den anorganischen Stoffen zählen insbesondere die Schwermetalle Zink, Kupfer, Blei, Nickel, Chrom und Cadmium. Die Höhe der Belastung hängt von ortsspezifischen Faktoren, die sich aus den kontinuierlichen Eigenschaften des Einzugsgebiets und den klimatischen Bedingungen zusammensetzen, und methodikspezifischen Einflussfaktoren ab. Diese bestimmen auch die ermittelte Verteilung der Schwermetalle zwischen der gelösten und der partikulären Phase. Diese Unterscheidung ist besonders für die Beurteilung der Toxizität des Niederschlagsabflusses sowie für dessen Behandlung in weitergehenden Behandlungsanlagen entscheidend. Im vorliegenden Beitrag werden historische Trends sowie das Aufkommen und die Verteilung von Schwermetallen in Verkehrsflächenabflüssen der Kategorien Parkplätze, Straßen und Autobahnen in Europa zusammengestellt.

Heavy Metal Pollution of Traffic Area Runoff – a European Review

Traffic area runoff waters are polluted by several organic and inorganic substances. The most concerning inorganic substances are the heavy metals zinc, copper, lead, nickel, chromium and cadmium. The discharge concentration in traffic area runoff waters depends on site-specific factors, which are composed of fixed-site conditions of the drainage area and climatic conditions, and method-specific influencing factors. These factors also determine the partitioning of heavy metals between the dissolved and the particulate phase. This distinction is relevant for the evaluation of the runoff toxicity and for its management in two-stage treatment plants. Historical trends and the impact of the traffic area categories parking lots, roads and highways on European runoff concentrations are described and evaluated.

1. Einleitung

Unter dem Begriff Verkehrsflächenabflüsse werden niederschlagsbedingte Abflüsse von Flächen zusammengefasst, die höchst unterschiedliche Nutzungen aufweisen. Diese reichen vom Gehweg, über Parkplätze und Anliegerstraßen bis hin zu Autobahnen [1]. Die meisten Messprogramme befassen sich mit den Abflüssen von Autobahnen, wobei auch die Abflüsse anderer Verkehrsflächenkategorien aufgrund ihrer deutlich höheren Flächenanteile in urbanen Gebieten für die Niederschlagswasserbewirtschaftung relevant sind. So können Parkplätze in Gewerbe- sowie Industriegebieten einen Anteil von bis zu 46 % an den urbanen Einzugsgebieten haben und der Anteil der Straßen kann 10–15 % der gesamten Stadtfläche betragen [2].

Eine Primärbelastung der Verkehrsflächenabflüsse sind die Inhaltsstoffe der Niederschläge, die aus der Atmosphäre ausgewaschen werden. Beim Auftreffen auf Oberflächen (befestigt und unbefestigt) und während des Abfließens von diesen Oberflächen nimmt der Niederschlag eine Vielzahl weiterer Stoffe in unterschiedlicher Konzentration auf. Diese Verschmutzung wird als Sekundärbelastung bezeichnet. Entsprechend der räumlichen und zeitlichen Variabilität ist das Spektrum der möglichen stofflichen Verunreinigungen der Verkehrsflächenabflüsse sowohl hinsichtlich der Art der auftretenden Stoffe als auch bezogen auf das Ausmaß der Belastung breit. Da viele der aufkommensrelevanten Stoffe mehr als eine Quelle aufweisen, sind die Hauptquellen der Verunreinigungen nur schwer bestimmbar. Dennoch ist eine Beschreibung der kontinuierlichen Eigenschaften des Einzugsgebietes (z. B. Querschnitt und Unterhalt der Straße) und der klimatischen Bedingungen (z. B. Dauer und Höhe des Regens) zur Quantifizierung der ortsspezifischen Einflüsse wichtig [3].

Neben den ortsspezifischen Einflüssen unterliegen die Ergebnisse auch methodikspezifischen Faktoren. Diese umfassen die Probennahme, die Probenaufbereitung bzw. Analytik sowie die Ergebnisauswertung, welche je nach Randbedingung des Messprogramms die Abbildung der realen Verhältnisse ermöglichen [4].

Die wichtigsten Stoffe in Verkehrsflächenabflüssen sind neben den Schwermetallen (z. B. Blei, Kupfer und Zink) die Bestandteile aus Auftausalzen (z. B. Chlorid) sowie organische Verbindungen (z. B. Mineralölkohlenwasserstoffe, polyzyklische aromatische Kohlenwasserstoffe und Methyl-tert-butylether) [5, 6]. Anhand von Messprogrammen kann die Aufkommensrelevanz dieser Stoffe in Verkehrsflächenabflüssen belegt werden.

Aus der Vielzahl an Stoffen werden im Folgenden aufgrund ihrer Aufkommens- und Wirkungsrelevanz nur die Schwermetalle behandelt. Neben der Höhe der Gesamtschwermetallkonzentrationen ist auch eine Betrachtung der Fraktionierung der Schwermetalle wichtig, da diese sowohl für die Bestimmung der Toxizität des Abflusses [7, 8, 9] als auch für dessen Behandelbarkeit in technischen Anlagen, welche sowohl die gelösten als auch partikulären Schwermetalle entfernen sollten [10], entscheidend ist.

Die Hypothese dieses Beitrags ist, dass mithilfe der Auswertung der Ergebnisse einer Vielzahl von Messprogrammen anhand von Verkehrsflächenkategorien Rückschlüsse auf die Zusammensetzung der Abflüsse trotz verschiedener Randbedingungen (orts- und methodikspezifische Faktoren) gezogen werden können. Ziel dieses Beitrags ist es daher, die orts- und methodikspezifischen Faktoren zu benennen, welche einen Einfluss auf die Höhe und Art der Belastung der Verkehrsflächenabflüsse haben, sowie das Auftreten und die Fraktionierung der Schwermetalle für verschiedene Verkehrsflächenkategorien zu beschreiben. Ferner werden historische Trends für die wichtigsten Schwermetalle aufgezeigt und es wird eine Einordnung der Verschmutzung europäischer Verkehrsflächenabflüsse im Vergleich zu anderen Kontinenten vorgenommen.

2. Methodik

Für diesen Beitrag wurde der Fokus auf die Erfassung und Auswertung europäischer Daten zu Verkehrsflächenabflusskonzentrationen gelegt. Diese entstammen einer von den Autoren dieses Beitrags erstellten Datenbank mit nationalen und internationalen Studien, die 294 einzelne Datensätze umfasst. Diese enthält neben den Schwermetallkonzentrationen weitere Stoffe sowie Informationen über die Probennahmeorte, die Analysen und Auswertungen. Dabei wird jeder Probennahmeort, auch wenn er mehrmals publiziert wurde, als ein Datensatz betrachtet. Ausnahmen sind die Orte, an denen zwischen den Messprogrammen viele Jahre (meist verbunden mit einer Änderung der durchschnittlichen täglichen Verkehrsstärke (DTV), des Straßenunterhalts, etc.), liegen bzw. konstruktive Änderungen am Straßenquerschnitt vorgenommen wurden, welche einen Einfluss auf die Ergebnisse haben (z. B. Erneuerung der konventionellen Deckschicht mit einem offenporigen Asphalt). Insgesamt konnten so 131 Studien ausgewertet werden. Bei der Datensammlung wurde immer zwischen gesamten, partikulären und gelösten Konzentrationen unterschieden. Ferner wurde zwischen der Angabe des arithmetischen Mittelwerts (im Folgenden als Mittelwert bezeichnet) und des Medians differenziert. Bei fehlenden Mittelungen wurden diese nach Möglichkeit aus allen Einzelergebnissen berechnet, sodass für viele Datensätze sowohl die Mittelwerte als auch Mediane erzeugt wurden.

Die Datenbank umfasst Messprogramme seit den 1970er-Jahren. Um aus diesen Daten auch historische Trends aufzeigen zu können, wurden die Konzentrationen zusammen mit den entsprechenden Untersuchungszeiträumen der Messprogramme abgespeichert und verwendet. Falls der Untersuchungszeitraum in einer Studie nicht angegeben war, wurde das Jahr, an dem der Zeitschriftenbeitrag eingereicht wurde, in der Datenbank hinterlegt.

Aus der Vielzahl der Stoffe, die in der Datenbank enthalten sind, werden in diesem Beitrag folgende Elemente ausgewählt, welche in den **Tabellen 1** und **2** mit ihrem Symbol abgekürzt werden: Aluminium (Al), Arsen (As), Barium (Ba), Cadmium (Cd), Kobalt (Co), Chrom (Cr), Kupfer (Cu), Eisen (Fe), Quecksilber (Hg), Mangan (Mn), Molybdän (Mo), Nickel (Ni), Blei (Pb), Platinum-Gruppen-Elemente (PGE; bestehen aus Palladium (Pd), Platin (Pt) und Rhodium (Rh)), Antimon (Sb), Selen (Se), Silicium (Si), Zinn (Sn), Titan (Ti), Vanadium (V), Wolfram (W) und Zink (Zn).

Da verschiedene Kategorien von Verkehrsflächenabflüssen untersucht wurden, wurden sämtliche Daten für die Auswertung in fünf Kategorien eingeteilt: Parkplätze, Straßen der Kategorie 1 (DTV ≤ 5000), Straßen der Kategorie 2 (5000 < DTV ≤ 15 000), Straßen der Kategorie 3 (DTV > 15 000) sowie Autobahnen. Wenn keine Angabe der DTV verfügbar war, wurde die Kategorisierung anhand der Beschreibung des Probennahmeortes vorgenommen, z. B. wurden Anliegerstraßen in die Kategorie 1 eingruppiert.

Für die statistische Auswertung und das Erstellen der Bilder wurde das Programmpaket SPSS 22 von IBM verwendet. Als Darstellung wurden Box-Whisker-Plots gewählt, bei denen das obere bzw. untere Ende der Box dem oberen bzw. unteren Quartil entspricht und der Strich in der Box

Tabelle 1: Übersicht der europäischen Messprogramme zur Charakterisierung von Autobahnabflüssen mit einer DTV ≤ 30 000; Fraktionen: t = gesamt, d = gelöst, p = partikulär; Elemente in kursiv unterhalb der Bestimmungsgrenze – diese ist, sofern veröffentlicht, in Klammern in µg/L angegeben

Land	Ort	Untersuchungszeitraum	DTV	Ereignisanzahl	Fraktion	Elemente	Literatur
CHE	Uri, Attinghausen, N2	08.03.2007–06.12.2008	21 000	17	t	Cd, Co, Cr, Cu, Fe, Mo, Ni, Pb, Sb, Zn	[65, 66, 67]
DEU	Saarland, Illingen, A1	–	25 470	–	t	Al, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Zn	[68]
DEU	Saarland, Nonnweiler, A1	–	15 360	–	t	Al, Cd, Cr, Cu, Fe, Hg, Ni, Pb, Zn	[68]
FRA	Loire-Atlantique, Nantes, A11, Brücke	28.03.1995–26.02.1996; 09.06.1997–09.11.1997	12 000	74	d, t	Cd, Cu, Pb, Zn	[43, 69]
GBR	Cowley, A417	06.1998–07.1999	23 650	10	d, t	Al, Cd, Cr, Cu, Ni, Pb, Pd, *Pt (< 0,15)*, Zn	[5]
GBR	Cumbria, Burton-in-Kendal, M6	01.12.1980–28.02.1982	30 000	10	d, p	Cd, Cu, Fe, Mn, Pb	[70]
IRL	Dublin, M4, Maynooth	12.2003–08.2005	29 140	42	d, t	*Cd, Cu (< 30), Pb (< 100),* Zn	[71]
IRL	Dublin, M4, Maynooth	12.2003–08.2005	29 140	42	d, t	*Cd*, Cu, *Pb (< 100)*, Zn	[71]
IRL	Dublin, M7, Kildare	12.2003–08.2005	25 760	42	d, t	*Cd*, Cu, Pb, Zn	[71]
IRL	Dublin, M7, Monasterevin	12.2003–08.2005	18 430	42	d, t	*Cd*, Cu, *Pb (< 100)*, Zn	[71]
ITA	SS73/bis, km 78	23.08.2002+24.10.2002	18 440	2	d, p	Al, Cu, Fe, Pb, Zn	[26]
ITA	SS73/bis, km 107	23.08.2002+24.10.2002	18 440	2	d, p	Al, Cu, Fe, Pb, Zn	[26]
NOR	Jessheim, E6	17.09.1980–13.05.1982	8000	26	t	Cd, Cr, Cu, Fe, Hg, Ni, Pb, Zn	[72]
PRT	Vila Real, IP4	–	6000	10	t	*Cd (< 1), Cr (< 1)*, Cu, Pb, Zn	[35]
SWE	Göteborg, E20	26.05.1992	–	1	d, t	Pt	[73]
SWE	Norsholm, E4	06.1998–06.1999	15 600	12	d, t	Al, Cd, Co, Cu, Fe, Mn, Pb, Si, W, Zn	[61]
SWE	Svaneberg, E20	06.1998–06.1999	7400	14	d, t	Al, Cd, Co, Cu, Fe, Mn, Pb, Si, W, Zn	[61]

Tabelle 2: Übersicht der europäischen Messprogramme zur Charakterisierung von Autobahnabflüssen mit einer DTV > 30 000; Fraktionen: t = gesamt, d = gelöst, p = partikulär; Elemente in kursiv unterhalb der Bestimmungsgrenze – diese ist, sofern veröffentlicht, in Klammern in µg/L angegeben

Land	Ort	Untersuchungszeitraum	DTV	Ereignisanzahl	Fraktion	Elemente	Literatur
AUT	Niederösterreich, Hinterbrühl, A21	04.11.2011–08.12.2011	42 000	4	t	Cd, Cr, Cu, Fe, Mn, Mo, Ni, Pb, *Pd*, Pt, Rh, Sb, V, Zn	[74]
AUT	Salzburger Land, A1, Schwarzenbergkaserne	12.2001–04.2003	> 60 000	29	t	Al, *Cd (< 1), Co (< 2),* Cr, Cu, Fe, Ni, Pb, Zn	[58]
AUT	Salzburger Land, A1, Baulos West	12.2001–04.2003	> 60 000	29	t	Al, Cd, *Co (< 2)*, Cr, Cu, Fe, Ni, Pb, Zn	[58]
AUT	Salzburger Land, A1, Messe	12.2001–04.2003	> 60 000	29	t	Al, *Cd (< 1), Co (< 2)*, Cr, Cu, Fe, Ni, Pb, Zn	[58]
AUT	Tirol, A13	–	–	–	t	Cu, Pb, Zn	[52]
AUT	Tirol, A13, Mautstelle	–	–	–	t	Cu, Pb, Zn	[52]
AUT	Wien, Kaisermühlen, A23	12.2012	> 240 000	1	t	Ba, Cd, Cr, Cu, Ni, Pb, Ti, V, Zn	[51]
AUT	–	–	–	45	t	*Cd (< 5), Cr (< 10)*, Cu, *Ni (< 1), Pb (< 10)*, Zn	[75]
CHE	Aargau, Wettingen, N1, Chlosterschür	24.08.2010–10.04.2012	109 000–120 000	7	t	Cu, Zn	[76, 77]
CHE	Aargau, Würenlos, N1, Neuwiesen	15.06.2011–02.06.2012	106 000–120 000	3	t	Cu, Zn	[77]
CHE	Basel-Landschaft, Birsfelden, N2, Hagnau	01.02.2008–31.12.2010	> 120 000	43	t	Cu, Zn	[78, 79]
CHE	Bern, Allmendingen, N6	2006–2007	59 000	–	d, t	*Cd (< 0,5)*, Cr, Cu, Pb, Zn	[16]
CHE	Bern, Bümpliz, N12, Pfaffensteig	06.03.2012–17.04.2013	38 985	–	t	Cu, Zn	[80]
CHE	Bern, Mattstetten, N1	2006–2009	74 000	–	d, t	*Cd (< 0,5)*, Cr, Cu, Pb, Sb, Zn	[16]
CHE	Zürich, Rossberg, N1	01.05.1976–30.04.1977	72 000	–	t	Cd, Cu, Pb, Zn	[81]
CHE	Zürich, Winterthur, N1, Chrebsbachknie	12.08.1996–14.11.2000	60 000	> 10	d, t	Cd, Cr, Cu, Ni, Pb, Zn	[82, 83, 84, 85]
DEU	Baden-Württemberg, Heilbronn-Obereisesheim, A6	02.1979–07.1979	47 000	48	t	Cd, Cr, Cu, Fe, Pb, Zn	[36]

▶▶

Land	Ort	Untersuchungszeitraum	DTV	Ereignisanzahl	Fraktion	Elemente	Literatur
DEU	Baden-Württemberg, Pleidesheim, A81	02.1978–09.1978	41 000	53	t	Cd, Cr, Cu, Fe, Pb, Zn	[36]
DEU	Baden-Württemberg, Ulm/West, A8/B10	05.1980–03.1981	52 100	44	t	Cd, Cr, Cu, Fe, Pb, Zn	[36]
DEU	Baden-Württemberg, Weinsberg, A6	04.1990–04.1991	34 675	-	t	Cd, Cr, Cu, Fe, Pb, Zn	[37]
DEU	Berlin, A113	05.11.2006–16.08.2007	140 000	20	d, t	As, Cd, Co, Cr, Cu, Fe, *Hg (< 0,1)*, Mo, Ni, Pb, Sb, V, Zn	[86]
DEU	Berlin, A115	02.2008–03.2008	91 000	3	d, p	Cd, Cr, Cu, Ni, Pb, Zn	[53]
DEU	Hessen, Frankfurt/Main, A3	09.1975–10.1977; 08.1978–08.1980	62 785-90 000	> 6	d, t	As, Ba, Cd, *Co (< 10)*, Cr, Cu, Fe, *Hg (< 0,1)*, Mn, *Mo (< 0,1)*, Ni, Pb, *Sb (< 0,2)*, *Se (< 2)*, Sn, Ti, V, Zn	[38]
DEU	Nordrhein-Westfalen, Bensberg, A4	2005–2007	71 220	82	t	Cd, *Co (< 10)*, *Cr (< 5)*, Cu, Fe, *Mo (< 25)*, *Ni (< 10)*, *Pb (< 25)*, *V (< 5)*, Zn	[18, 87]
DEU	Nordrhein-Westfalen, Gelsenkirchen, A2	06.1997–02.1999	61 061	20	d, t	Cd, Cu, Pb, Zn	[17]
DEU	Nordrhein-Westfalen, Gladbeck, A31	06.1997–02.1999	39 021	25	d, t	Cd, Cu, Pb, Zn	[17]
DEU	Nordrhein-Westfalen, Haltern, A43	06.1997–02.1999	–	–	d, t	Cd, Cu, Pb, Zn	[17]
DEU	Nordrhein-Westfalen, Köln-Ost, A3	11.08.2006–24.08.2007	156 000	20	d, t	*As (< 10)*, Cd, *Co (< 20)*, Cr, Cu, Fe, *Hg (< 2)*, *Mo (< 20)*, Ni, Pb, *Sb (< 10)*, V, Zn	[86]
DEU	Nordrhein-Westfalen, Köln-Poll, A4	12.07.1998–22.01.2000	> 50 000	33	d, t	Cd, *Cr (< 5)*, Cu, Hg, *Ni (< 10)*, Pb, Zn	[88]
DEU	Nordrhein-Westfalen, Köln-Porz, A59	12.07.1998–05.07.2000	> 50 000	29	d, t	Cd, Cr, Cu, Hg, *Ni (< 10)*, Pb, Zn	[88]
DEU	Nordrhein-Westfalen, Meckenheim, A61	2005–2007	73 310	88	t	Cd, *Co (< 10)*, *Cr (< 5)*, Cu, Fe, *Mo (< 25)*, *Ni (< 10)*, *Pb (< 25)*, *V (< 5)*, Zn	[18, 87]
DEU	Nordrhein-Westfalen, Mülheim, A3	06.1997–02.1999	89 498	13	d, t	Cd, Cu, Pb, Zn	[17]
DEU	Nordrhein-Westfalen, Oberhausen, A42	06.1997–02.1999	74 991	19	d, t	Cd, Cu, Pb, Zn	[17]
DEU	Nordrhein-Westfalen, Widdig, A555	2005–2007	69 368	88	t	Cd, *Co (< 10)*, *Cr (< 5)*, Cu, Fe, *Mo (< 25)*, *Ni (< 10)*, *Pb (< 25)*, *V (< 5)*, Zn	[18, 87]
GBR	Cumbria, Burton-in-Kendal, M6	12.04.1986–11.04.1987	37 600	7	d, p	Cd, Cu, Pb	[89]
GBR	London, A406	03.1993–02.1994	–	–	t	Cd, Cu, Pb, Zn	[90]
GBR	London, M25, junctions 5 and 6	01.1997–02.1998	120 000	10	t	Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sb, V, Zn	[57]
GBR	London, M25, junction 9	01.1997–02.1998	140 000	10	t	Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sb, V, Zn	[57]
GBR	Newbury, A34	05.2001–06.2002	37 192	10	d, t	Al, Cd, Cr, Cu, Ni, Pb, Pd, *Pt (< 0,15)*, Zn	[5]
GBR	Souldern Brook, M40	08.1999–10.2000	83 579	10	d, t	Al, Cd, Cr, Cu, Ni, Pb, Pd, *Pt (< 0,15)*, Zn	[5]
GBR	Weston on the Green, A34	09.2000–03.2002	64 953	10	d, t	Al, Cd, Cr, Cu, Ni, Pb, Pd, *Pt (< 0,15)*, Zn	[5]
GBR	Wiltshire, Brinkworth Brook, M4	17.12.1997–17.12.1998	70 830	10	d, t	Al, *Cd (< 4)*, *Cr (< 10)*, Cu, *Ni (< 10)*, *Pb (< 50)*, *Pd (< 0,5)*, Pt, Zn	[63]
GBR	Wiltshire, Swindon, M4	12.1998–03.2000	36 107	10	d, t	Al, Cd, Cr, Cu, Ni, Pb, Pd, *Pt (< 0,15)*, Zn	[5]
GRC	Crete, Hersonissos, E75	09.2006–08.2007	–	41	t	Cu, Ni, Pb, Zn	[91]
NLD	Amsterdam, A7	07.1994–09.1995	53 000	6	d, t	Cd, Cr, Cu, Ni, Pb, Zn	[39]
NLD	Amsterdam, A9	07.1994–09.1995	83 000	6	d, t	Cd, Cr, Cu, Ni, Pb, Zn	[39]
NLD	Laren, A1	27.01.2003–05.09.2004	-	80	d, t	Cd, Cr, Cu, Ni, Pb, Zn	[92]
SWE	Stockholm, E4, Brücke	03.05.2005–26.08.2005; 14.12.2005–14.02.2006	108 300	9	d, t	Al, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn	[93]

dem Median. Die Länge der Whisker gibt das 1,5-Fache des Interquartilsabstands (IQR) wieder. Ausreißer (>1,5-Fache des IQR) werden mit kleinen Kreisen und Extremwerte (>3,0-Fache des IQR) mit Sternen gekennzeichnet.

3. Einflussfaktoren

Die Ergebnisse von Messprogrammen zur Qualität von Verkehrsflächenabflüssen hängen von einer Vielzahl von Einflussfaktoren ab, weshalb die in der Literatur dokumentierten Qualitätsdaten aus verschiedenen Messprogrammen eine extrem weite Streuung aufweisen [4]. Grundsätzlich lassen sich ortsspezifische und methodikspezifische Faktoren unterscheiden. Die ortsspezifischen Faktoren variieren nicht nur örtlich, sondern auch über die Zeit und beeinflussen somit die reale Verschmutzung des Oberflächenabflusses. Folglich lassen sich diese Faktoren in Eigenschaften des jeweiligen Einzugsgebietes (örtliche Varianz) sowie Witterungs- und Klimaeinflüsse (zeitliche Varianz) unterscheiden. Dabei können die konstanten Eigenschaften des Einzugsgebietes in die drei Unterkategorien umgebende Landnutzung, Verkehrsflächeneigenschaften sowie Verkehrsgeschehen unterteilt werden.

Die erste Unterkategorie, Landnutzung, kann die Abflusskonzentrationen beeinflussen. Dies wurde beispielsweise für die beiden Kategorien städtisch und ländlich für amerikanische Autobahnabflüsse gezeigt [11, 12]. Eine weitere Kategorisierung (z. B. gewerblich, industriell, städtisch, ultra-städtisch und gemischt) ist nicht eindeutig, da deren Auslegung zwischen den einzelnen Messprogrammen variiert, und somit kann ein eindeutiger Zusammenhang zwischen diesen Kategorien und den Abflusskonzentrationen nicht gefunden werden.

Zu den Verkehrsflächeneigenschaften gehören neben Informationen zur Vegetation und Topographie Angaben zum Straßenquerschnitt und den eingesetzten Materialien. Derzeit sind wenige Informationen zur Topographie (Einschnitt, Dammlage etc.), zur straßennahen Vegetation, zur Steigung/Neigung der Straße sowie dem Vorhandensein von Bordsteinkanten, Lärmschutzwänden und Leitplanken in den Studien wiedergegeben, obwohl diese einen Einfluss auf die Konzentrationen im Abfluss haben [13, 14, 15, 16]. Beispielsweise führen Lärmschutzwände zu einem Anstieg der Konzentrationen [17, 18] und auch durch Betonbarrieren können mehr Feststoffe während der Trockenperioden zurückgehalten werden, welche während eines anschließenden Regens mobilisiert werden können [19]. Ferner haben Geschwindigkeitsbeschränkungen, Ampelanlagen sowie Ein- und Ausfahrten einen Einfluss auf die Abflusskonzentrationen, da durch das Bremsen und Beschleunigen ein höherer Reifen- und Bremsabrieb sowie ein erhöhter Ausstoß an Abgasen induziert wird [20, 21, 22]. Dahingegen beschreiben die meisten Studien die Anzahl der Fahrstreifen, die Materialoberfläche der Verkehrsfläche und die Größe des Einzugsgebietes. Die Anzahl der Fahrstreifen in Verbindung mit der DTV hat Auswirkungen auf die Staugefahr, welche einen Einfluss auf die Abflusskonzentrationen hat [23, 24]. Als Material für die Deckschicht wurde in Europa meist Asphalt (n=44), aber auch Asphalt und Beton (n=9), Beton (n=7) und offenporiger Asphalt (n=6) untersucht. Dabei führt nur die Betondecke zu einer signifikanten Erhöhung des pH-Werts in Verkehrsflächenabflüssen [25].

Das Verkehrsgeschehen umfasst die DTV, den Fahrzeugtyp, den Unterhalt der Straße sowie institutionelle Vorgaben. Obwohl einige Autoren die DTV als wichtigsten Einflussfaktor auf die Abflusskonzentrationen identifiziert haben [26, 27], ist die DTV oftmals nicht als bester Indikator für die Verschmutzung von Abflüssen geeignet, da sie als einzelne Variable nur teilweise die Abweichungen in den Konzentrationen zwischen verschiedenen Messorten beschreiben

Bild 1: Beispiele zweier Standorte in Deutschland zur Bestimmung der Verkehrsflächenabflusskonzentrationen urbaner Straßen (links: Wuppertal, rechts: München)

kann [28]. Ferner ist bei der Angabe der DTV darauf zu achten, dass sich die Werte oft auf beide Fahrtrichtungen beziehen, obwohl nur eine beprobt wurde und eine Beeinflussung durch die andere Richtung ausgeschlossen werden kann. Zu den Fahrzeugtypen zählen zum einen die Anteile des Schwerlastverkehrs an der DTV und zum anderen das Alter und der Unterhalt der Fahrzeuge, welche zwischen einzelnen Ländern stark variieren können [14]. Der Unterhalt der Straßen umfasst unter anderem den Winterdienst und die Straßenreinigung, welche beide einen erheblichen Einfluss auf das Stoffspektrum haben können [23, 29]. Als institutionelle Vorgabe kann beispielsweise das Verbot bleihaltigen Benzins angesehen werden. Diese wird in Abschnitt 4.1 genauer beschrieben.

Zu den klimatischen Einflussfaktoren zählen die Trockendeposition, die vorhergehende Trockenperiode, die Regencharakteristik (Volumen, Intensität und Dauer), saisonale Effekte sowie Windturbulenzen. Der wichtigste dieser Faktoren, der die Abflusskonzentrationen beeinflusst, ist die vorhergehende Trockenperiode [13], während der Feststoffe auf der Straße und im Straßenrandbereich akkumulieren können. Reduziert wird die Akkumulation unter anderem durch die Straßenreinigung sowie durch Windverfrachtungen (z. B. Verwehungen aufgrund vorbeifahrender Fahrzeuge) [23]. Weiterhin ist ein allgemeiner Anstieg der Schwermetallkonzentrationen in den Wintermonaten aufgrund der durch Auftausalze bedingten Korrosion in Verbindung mit geringeren Regenintensitäten festzustellen [23].

Beispiele für die Unterschiede zwischen zwei urbanen Einzugsgebieten, deren Abflüsse regelmäßig beprobt wurden, sind in **Bild 1** dargestellt. Während in Wuppertal die Straße anhand ihrer Steigung, der Vegetation, der Leitplanken aus Zink und der Nähe zu einem Kreuzungsbereich mit Blitzer gekennzeichnet ist, ist die Stelle in München deutlich stärker befahren (57 000 Kfz/d), hat eine erhöhte Staugefahr und mehr Fahrstreifen mit einer zusätzlichen Standspur. Gemeinsamkeiten der beiden Standorte stellen die Straßenlaternen sowie die teilerneuerten alten Fahrbahndecken aus Asphalt dar. Ferner ist Wuppertal mit die regenreichste Großstadt Deutschlands (jährliche Niederschlagshöhe: 1183 mm/a), wodurch sich die Niederschlagsmengen von denen in München (967 mm/a) unterscheiden. Folglich führen viele überlagernde Quellen und Einflussfaktoren, die die Belastung teils verstärken und teils reduzieren, zu einem unterschiedlichen Stoffaufkommen.

Als methodikspezifische Einflüsse gelten jene Randbedingungen des Messprogramms, die die Abbildung der realen Verhältnisse betreffen. Wichtige Teilschritte bei der Untersuchungsmethodik sind die Probennahme, die Probenaufbereitung bzw. Analytik sowie die Ergebnisauswertung. Bei der Probennahme gibt es zwei Arten (automatisch oder manuell), welche je nach Untersuchungsziel eingesetzt werden. Manuelle Probennahmen mit der Hand werden besonders bei kurzen Beprobungen genutzt, wie sie zum Nachweis der Funktionsfähigkeit dezentraler Anlagen [30] oder im Rahmen der Voraussetzungen zur behandlungsfreien Einleitung verwendet werden [31], da sie einfach umsetzbar sind. Dahingegen eignen sich automatische Probennehmer (sowohl aktive als auch passive) besonders zur Erzeugung repräsentativer Mischproben, wobei diese aus mehreren Hundert Teilproben bestehen können [32]. Entscheidend dabei ist auch die Probennahmestrategie, wobei zwischen volumenproportional (n=36 – alle Daten zur Anzahl der Probennahmestrategie für Europa), zeitproportional (n=22), First Flush (n=12), Mischprobe (n=25) und Zufallsprobe (n=9) unterschieden wird. Dabei werden zeitproportionale Probennahmen sowohl zu festen Zeitintervallen als auch zu variablen Zeiten genommen. Bei letzterer erfolgen zu Beginn des Regenereignisses mehr Probennahmen in kürzeren Intervallen als im weiteren Verlauf des Ereignisses. Neben der Art und Strategie haben weitere Aspekte wie der Ort der Probennahme (Straßenrand vs. Ablaufrinne, mögliche Vorsedimentation), der Probennehmerschlauch (Klassierungseffekte, Verunreinigungen etc.), die Kühlung der Proben sowie die Schulung des Personals Einflüsse auf die Messergebnisse. Auch hat die Anzahl der beprobten Regenereignisse einen Einfluss auf die Ergebnisse. So werden bei der Beprobung ausgewählter Regenereignisse meist nur größere Ereignisse mit höheren Schadstoffbelastungen untersucht, sodass dies im Jahresvergleich zu einem überhöhten Mittelwert/Median führen kann.

Bei der Probenvorbereitung für die Bestimmung der Schwermetallkonzentrationen ist immer die Art des Probenaufschlusses entscheidend (z. B. Säureaufschluss, Königswasseraufschluss), da nicht jeder Aufschluss sämtliche Schwermetalle für die anschließende Analytik detektierbar macht. Ferner ist bei der Bestimmung der gelösten Anteile die Dauer zwischen Probennahme und Filtration entscheidend. Als Zeitspanne zwischen Probennahme und Filtration wird in der Literatur 1 h [33, 34], 2–3 h [35] sowie maximal 24 h genannt [24, 36, 37]. Ähnlich verhält es sich auch mit der Messung des pH-Werts, welcher am besten binnen 15 Min nach Probennahme vor Ort gemessen werden sollte, da er sich aufgrund einer Gleichgewichtseinstellung der Stoffe in der Probe zügig ändern kann und damit auch den Anteil an gelösten und ungelösten Stoffen ändert.

Die größten möglichen Abweichungen zwischen Realität und Messwert treten somit bei der Probennahme und Probenvorbereitung auf, während die eigentliche Probenanalytik (z. B. Messung der Schwermetalle mittels Atomabsorptionspektrometrie) in der Regel geringere Schwankungen verursacht.

Bei der Ergebnisauswertung ist vor allem der Umgang mit Proben unterhalb der Bestimmungsgrenze ausschlaggebend. Diese werden je nach Autor gar nicht, mit der Hälfte der Bestimmungsgrenze sowie mit der ganzen Bestimmungsgrenze angesetzt. Auch unterscheidet sich das Vorgehen dahingehend, ab welchem Anteil an nichtbestimmbaren Konzentrationen eine mittlere Konzentration angegeben wird. Aufgrund der verschiedenen Herangehensweisen können beispielsweise Fehler bei der Bestimmung des gelösten Anteils auftreten, wenn die gelösten Anteile sehr gering sind und nahe an der Bestimmungsgrenze liegen. Dies kann auch einen großen Einfluss auf berechnete Mediane und Mittelwerte haben. Außerdem werden häufig die Ergebnisse der Messprogramme nicht frachtgemittelt angegeben, für deren Berechnung eine verlässliche Messung des Abflussvolumens notwendig ist. Dabei ist zu beachten, dass frachtgewogene Mittelwerte nicht direkt mit arithmetischen Mittelwerten zu vergleichen sind.

4. Ergebnisse und Diskussion

4.1 Historische Entwicklungen

Die historischen Entwicklungen für die drei aufkommensrelevanten Schwermetalle Zink, Kupfer und Blei wurden seit den 1970er-Jahren bis heute anhand der Literaturdaten ausgewertet und sind in **Bild 2** dargestellt. Dabei ist nur für Blei ein deutlicher Rückgang der Konzentrationen erkennbar, welcher mit institutionellen Vorgaben zusammenhängt.

Die Reduzierung der Blei-Konzentrationen im Benzin begann beispielsweise in Deutschland im Jahr 1976, als die zulässige Konzentration im Benzin von 850 mg/L auf 150 mg/L reduziert wurde [38]. Nachdem in den 80er-Jahren aufgrund des Einsatzes von Katalysatoren vermehrt zusätzlich bleifreies Benzin an den Tankstellen angeboten wurde, begann auch in allen anderen Staaten Westeuropas bis spätestens 1986 die Reduzierung der zulässigen Blei-Konzentrationen [39], sodass eine erhebliche Minimierung der Bleiquellen bis in die 90er-Jahre erzielt wurde. In fast allen Ländern der Europäischen Union sowie in der Schweiz darf seit Anfang 2000 kein verbleites Motorenbenzin mehr verkauft werden. Als letztes dieser Länder verbot Italien im Januar 2002 den Verkauf bleihaltigen Benzins [26]. Weitere Quellen für Blei, wie Auswuchtgewichte in Felgen [40], wurden ebenfalls substituiert, sodass einhergehend mit dem Verbot zum Verkauf bleihaltigen Benzins dessen Konzentrationen in Verkehrsflächenabflüssen weiterhin abgenommen haben. So nahm der Median der mittleren Blei-Konzentrationen in Europa von 163 µg/L in den 1980er-Jahren über 51 µg/L in den 1990er-Jahren auf 17 µg/L in den 2000er-Jahren ab. Für die Zeit nach 2009 sind bisher nur zwei Datensätze verfügbar, sodass ein hoher Einzelwert von 112 µg/L die bestehende Reduzierung der Bleiabflusskonzentrationen überprägt. Der Trend für Blei in Europa ist vergleichbar mit den Ergebnissen einer Untersuchung aus den USA [41].

Für Zink ist kein einheitlicher Trend feststellbar, obwohl einige Autoren annehmen, dass erhöhte Zink-Konzentrationen in Verkehrsflächenabflüssen aufgrund des zunehmenden Gebrauchs von Zink durch die steigende Anzahl an Leitplanken [42] und die Verwendung als Korrosionsschutz im Automobilbau [25] wahrscheinlich sind. Da Zink zusätzlich seit Jahrzehnten in weiteren Autoteilen, wie z. B. Reifen und Bremsen [43], verwendet wird und deren Abrieb je nach örtlichen Bedingungen schwankt, kann kein Trend für Zink gefunden werden. Ferner ist Zink aufgrund der Verwendung in Leitplanken, Straßenschildern und Brückenkonstruktionen an spezifischen Orten besonders aufkommensrelevant, weshalb stark erhöhte Zink-Konzentrationen oftmals mit diesen ortsspezifischen Einflüssen korrelieren. Ebenfalls ist für Kupfer kein Trend ermittelbar. Kupfer wird wie Zink in Autoteilen, z. B. Bremsen [43], verwendet und kommt daher fortwährend in den Verkehrsflächenabflüssen, insbesondere an Stellen mit starken Bremsvorgängen [21], vor.

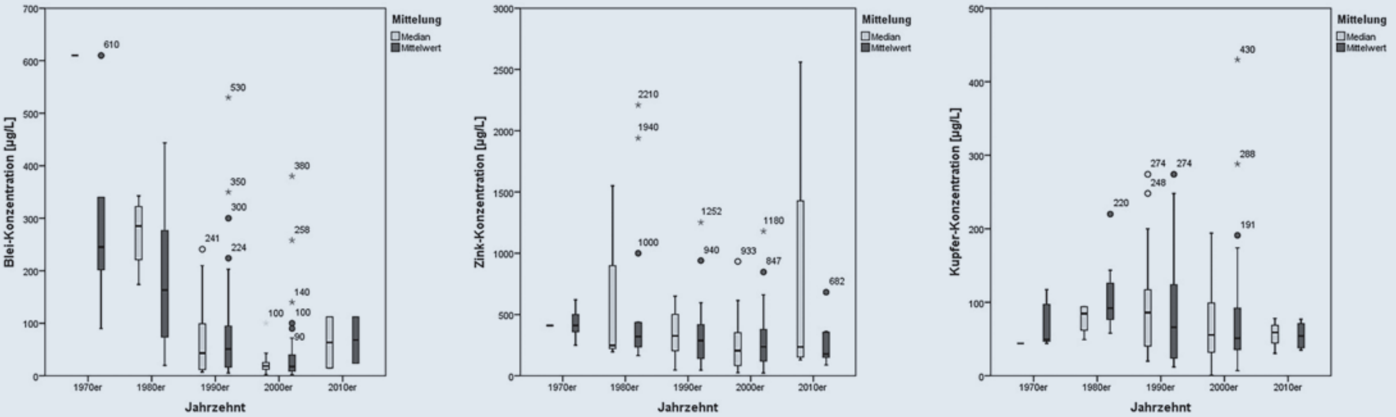

Bild 2: Historischer Trend der Blei-, Zink- und Kupfer-Konzentrationen (gesamt) in europäischen Verkehrsflächenabflüssen (Box-Whisker-Plots: oberes bzw. unteres Ende der Box entspricht dem oberen bzw. unteren Quartil und der Strich in der Box dem Median; kleine Kreise stellen Ausreißer und Sterne Extremwerte dar)

4.2 Parkplätze und Straßen

Die Datensätze zu den europäischen Verkehrsflächenabflüssen wurden – wie in Kapitel 2 beschrieben – in fünf Kategorien eingeteilt. Die mittleren Konzentrationen für die Parkplätze und die drei Straßenkategorien sind in **Bild 3** vergleichend für die sechs Schwermetalle Zink, Kupfer, Blei, Nickel, Cadmium und Chrom, sowohl für die gelösten als auch die gesamten Konzentrationen, dargestellt. Dabei gibt es in der Datenbank für die Straßen der Kategorie 3 die meisten Datensätze (n=20), für die Kategorie 2 (n=16) und die Parkplätze (n=11) weniger Daten und für die Straßen der Kategorie 1 in Europa sind den Autoren dieser Studie nur sieben Datensätze bekannt. Da in jedem Messprogramm nicht immer alle sechs dargestellten Schwermetalle gemessen wurden, kann die Anzahl für jeden Balken geringer sein. Dies trifft insbesondere auf die gelösten Konzentrationen zu und dabei vor allem auf Nickel, Cadmium und Chrom.

Die Ergebnisse dieser Auswertung für die europäischen Parkplatzabflüsse ergaben, dass die Konzentrationen für die Schwermetalle Kupfer, Zink, Blei und Cadmium vergleichbar mit den Konzentrationen für die Straßen sind. Bei den Blei-Konzentrationen ist zu beachten, dass die beiden höchsten Konzentrationen (350 µg/L und 107 µg/L) 1996 bzw. 1995 gemessen wurden. Auffällig sind die im Vergleich zu den Straßen hohen Chrom-Konzentrationen zwischen 8 µg/L und 28 µg/L (n=6). Für Nickel liegen nur zwei Gesamtkonzentrationen sowie eine gelöste Konzentration vor. Die gelösten Anteile der Schwermetalle Blei und Chrom sind sehr gering, wohingegen Kupfer, Zink und Cadmium deutlich stärker gelöst vorliegen.

In Deutschland werden Parkplätze nach DWA-M 153 je nach Nutzung in verschiedene Kategorien eingeteilt und die zugehörigen Flächenverschmutzungen werden entsprechend als gering, mittel oder stark eingestuft [44]. Ein Vergleich der Abflüsse dreier Tank- und Rastanlagen an deutschen Autobahnen ergab, dass die Lkw-Stellplätze, welche nach DWA-M 153 als stark verschmutzt eingruppiert sind, bei fast allen Parametern deutlich höher belastet waren als die Abflüsse der Pkw-Stellplätze [45]. Ferner sind laut der Eingruppierung nach DWA-M 153 stark frequentierte Pkw-Parkplätze (z. B. von Supermärkten) im

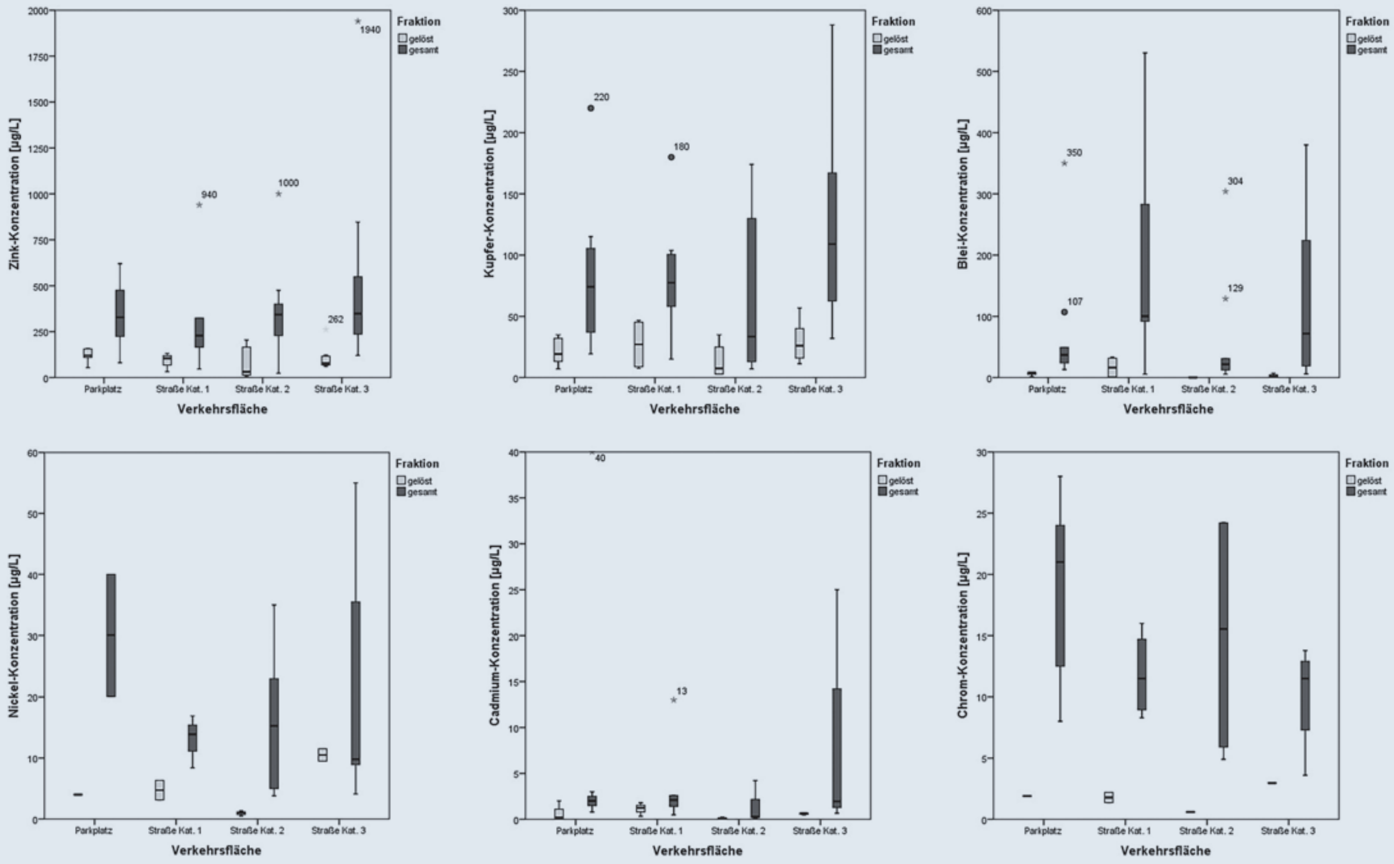

Bild 3: Schwermetallkonzentrationen an Zink, Kupfer, Blei, Nickel, Cadmium und Chrom (gelöst und gesamt) in Abflüssen von Parkplätzen und drei Straßenkategorien (Kategorie 1: DTV ≤ 5000; Kategorie 2: 5000 < DTV ≤ 15 000; Kategorie 3: DTV > 15 000) in Europa (Box-Whisker-Plots: oberes bzw. unteres Ende der Box entspricht dem oberen bzw. unteren Quartil und der Strich in der Box dem Median; kleine Kreise stellen Ausreißer und Sterne Extremwerte dar)

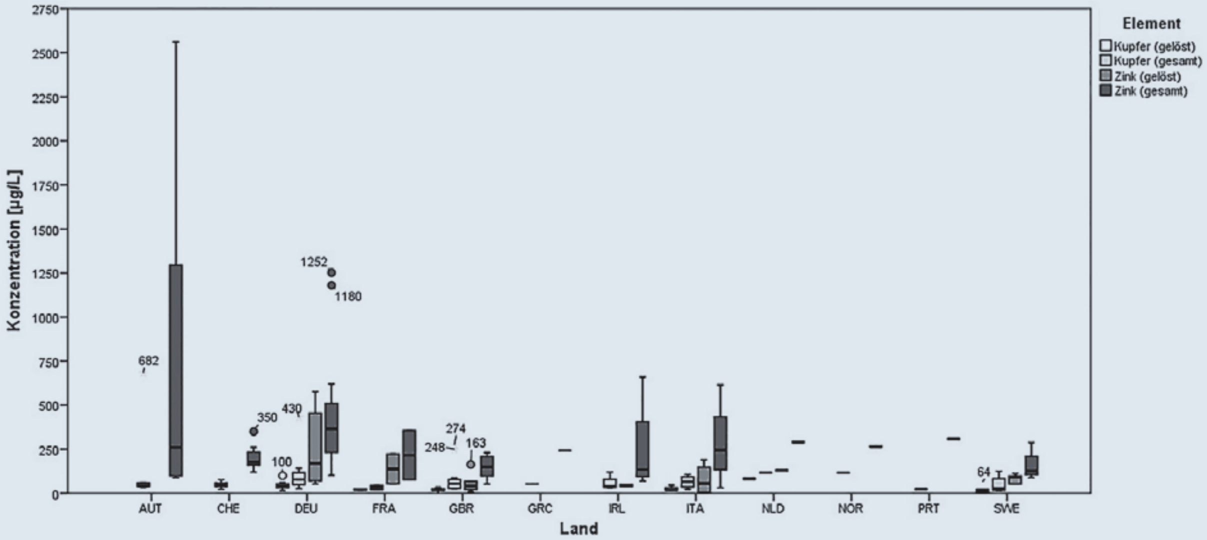

Bild 4: Belastung von Autobahnabflüssen europäischer Länder mit den Schwermetallen Zink und Kupfer (gelöst und gesamt) (Box-Whisker-Plots: oberes bzw. unteres Ende der Box entspricht dem oberen bzw. unteren Quartil und der Strich in der Box dem Median; kleine Kreise stellen Ausreißer und Sterne Extremwerte dar)

Gegensatz zu Hof- und Parkflächen mit geringer Frequentierung stark belastet. Dies konnte jedoch aus den wenigen in der Datenbank verfügbaren Konzentrationen für diese Verkehrsflächenkategorie nur bedingt abgeleitet werden. So weisen beispielsweise die Parkplatzabflüsse dreier Universitäten [46, 47, 48], bei denen von einer geringen täglichen Belegung ausgegangen werden kann, die geringsten Zink-, Kupfer- und Blei-Konzentrationen bezogen auf alle Parkplatzkategorien auf. Für den einzigen verfügbaren Supermarktplatz liegen die mittleren Abflusskonzentrationen bei 361 µg/L Zink, 77 µg/L Kupfer, 24 µg/L Blei und 40 µg/L Nickel (alles Gesamtkonzentrationen) [49] und somit oberhalb der Universitäts-Parkplatzabflusskonzentrationen und im Bereich der Pkw-Stellplätze der Autobahn-Rastanlagen. Etwas höhere Abflusskonzentrationen sind aus der Beprobung eines Flughafen-Parkplatzes bekannt [50]. Folglich kann davon ausgegangen werden, dass Parkplätze je nach Frequentierung, Fahrzeugkategorie und Fahrzeugverschmutzung stark belastet sind. Dies ist damit zu erklären, dass die Tropfverluste, die Abgasemissionen (auch kalter Motor) sowie die Brems- und Reifenabriebe auf dieser Verkehrsflächenkategorie besonders hoch sind, auch wenn die Belegung stark frequentierter Parkplätze maximal 50 Kfz/d pro Stellplatz zulässt [1].

Da in Deutschland die Höhe der stofflichen Belastung im Abfluss einer Straße im DWA-M 153 anhand der DTV festgelegt ist [44], wurden die drei Straßenkategorien entsprechend des Merkblattes gewählt. Die Kategorie 1 umfasst somit Wohn-, Anlieger-, Erschließungs- und Kreisstraßen und die Kategorie 2 Hauptverkehrsstraßen. Nach diesem Merkblatt werden die Hauptverkehrsstraßen mit überregionaler Bedeutung (Kategorie 3) mit über 15 000 Kfz/d als stark belastet bzw. nicht tolerierbar bezeichnet. Die Auswertung der Datensätze ergibt nur für die Zink-Konzentrationen mit steigender Straßen-Kategorie durchgehend eine Zunahme sowohl der Median-Werte (von 229 µg/L über 342 µg/L auf 349 µg/L) als auch der Maxima (von 940 µg/L auf 1940 µg/L). Bei den Kupfer-Konzentrationen sind lediglich die Konzentrationen der Kategorie 3 am höchsten. Bei den anderen Elementen als auch bei den weltweiten Daten sind die Tendenzen ähnlich. Besonders für Cadmium und Chrom ergeben sich Abflusskonzentrationen, die unabhängig von der DTV sind. Bei den Blei-Konzentrationen ist zu beachten, dass für die Kategorie 1 nur Werte vorliegen, die zwischen 1982 und 1994 gemessen wurden. Bei den Kategorien 2 und 3 wurden die Konzentrationen zwischen 1990 und 2009 gemessen, weshalb diese geringer als bei der ersten Kategorie sind, wobei die beiden Ausreißer der Kategorie 2 zu den ältesten Datensätzen zählen. Folglich sind die Straßenabflüsse unterschiedlich stark mit Schwermetallen belastet und die Höhe der Konzentrationen ist besonders durch örtliche Randbedingungen gekennzeichnet. Beispielsweise waren die Konzentrationen an einer Straße der Kategorie 2 (14 200 Kfz/d) in Hildesheim, Deutschland, aufgrund der Brems- und Beschleunigungsvorgänge an einer Ampel stark erhöht (Zink 436 µg/L und Kupfer 136 µg/L, Gesamtkonzentrationen) und lagen oberhalb der Konzentrationen, die üblicherweise an Autobahnen gemessen werden [20].

Insgesamt gesehen sind die Konzentrationen an Zink heutzutage am aufkommensrelevantesten, gefolgt von Kupfer und Blei. Je nach Ort folgen Nickel, Chrom und Cadmium. Die gelösten Anteile von Zink und Kupfer sind ebenfalls am höchsten, wobei Nickel aufgrund des höheren gelösten Anteils in höheren gelösten Konzentrationen als Blei und Chrom vorkommt. Die gelösten Konzentrationen an Blei und Chrom sind vergleichbar mit den gelösten Konzentrationen an Cadmium.

4.3 Autobahnen

Die meisten Messprogramme zu Verkehrsflächenabflusskonzentrationen in Europa haben sich mit der Charakteristik von Autobahnabflüssen beschäftigt (n=73), wobei die am häufigsten untersuchten Orte in Deutschland (DEU, n=24 an 20 Orten), in Großbritannien (GBR, n=11 an zehn Orten), in der Schweiz (CHE, n=11 an neun Orten), in Österreich (AUT, n=8) und in Schweden (SWE, n=6 an vier Orten) lagen. Weitere Länder mit mehr als einem Datensatz sind Irland (IRL, n=4), Italien (ITA, n=4 an zwei Orten), die Niederlande (NLD, n=3) und Frankreich (FRA, n=2 an einem Ort). Für Norwegen (NOR) und Portugal (PRT) liegt jeweils ein Datensatz vor. Auffällig bei den Ländern mit zahlreichen Messprogrammen ist, dass für Österreich keine gelösten Anteile vorliegen und in der Schweiz nur bei wenigen Messungen gelöste Konzentrationen bestimmt wurden. Die DTV der in **Bild 4** dargestellten Messergebnisse (Mittelwerte) variiert sehr stark und liegt zwischen 6000 Kfz/d in Portugal [35] und 240 000 Kfz/d in Österreich [51]. Eine Abhängigkeit der Belastung von der Landnutzung konnte nicht ermittelt werden, weshalb neben der Aufgliederung der Messergebnisse nach Ländern auf weitere Darstellungen verzichtet wurde. Alle Messprogramme zu Autobahnabflüssen sind in den **Tabellen 1** und **2** aufgeführt.

Die höchsten gesamten Zink-Konzentrationen in Autobahnabflüssen wurden in absteigender Reihenfolge an der A23 in Wien, Österreich [51], an der A13 an einer Mautstelle in Tirol, Österreich [52], an der A43 in Haltern, Deutschland [17], und an der A115 in Berlin, Deutschland [53], gemessen. Die höchsten gesamten Kupfer-Konzentrationen in Autobahnabflüssen wurden an der A23 in Wien, Österreich [51], und an der A115 in Berlin, Deutschland [53], gemessen. Ein Vergleich der Median-Konzentrationen ergibt, dass die höchsten Werte sowohl für Zink (gesamt 366 µg/L, gelöst 170 µg/L) als auch für Kupfer (gesamt 78 µg/L, gelöst 38 µg/L) in Deutschland gemessen wurden. Somit liegen die gelösten Anteile dieser beiden Schwermetalle bei jeweils knapp 50 %.

Insgesamt liegen die Zink-Konzentrationen an Autobahnabflüssen über alle Länder gemittelt bei 230 µg/L (gesamt) und 76 µg/L (gelöst) und die Kupfer-Konzentrationen bei 52 µg/L (gesamt) und 22 µg/L (gelöst), wobei die gelösten Anteile für Europa geringer sind als die in Deutschland ermittelten Anteile. Außerdem sind die Konzentrationen der beiden Schwermetalle in Autobahnabflüssen mit denen der Straßen aus **Bild 3** vergleichbar, wobei die Straßen der Kategorie 3 im Mittel höher als die Autobahnen belastet sind. Zum einen sind diese Hauptverkehrsstraßen mit überregionaler Bedeutung oftmals aufgrund spezifischer Randbedingungen (z. B. dichte Bebauung und Ampeln) höher belastet und zum anderen verursachen die hohen Fahrgeschwindigkeiten und der Wind auf den großen Straßenflächen der Autobahnen mit Standspuren mehr Verwehungen, als dies auf den Hauptverkehrsstraßen der Kategorie 3 der Fall ist, welche zu einer Reduzierung der Ablaufkonzentrationen beitragen. Im Gegensatz können Schallschutzwände diesen Effekt vermindern. Ferner können die Schwermetallkonzentrationen in Abflüssen von Autobahnen im Vergleich zu Außerortsstraßen geringer sein, da bei Autobahnen eine Standspur vorhanden ist, die zu einer Verdünnung der gemessenen Konzentrationen führen kann.

4.4 Kontinentale Trends

Die Verkehrsflächenabflusskonzentrationen für die beiden derzeit aufkommensrelevantesten Schwermetalle Zink und Kupfer wurden ebenfalls getrennt nach Kontinenten ausgewertet und sind in **Bild 5** als Gesamtkonzentrationen dargestellt. Bei der Auswertung wurden nur die vier Kontinente Asien, Europa, Nordamerika und Ozeanien berücksichtigt, da aus Südamerika und Afrika jeweils weniger als fünf Datensätze zur Verfügung standen. Dabei liegen für Europa mit 131 Datensätzen etwas mehr Daten als für Nordamerika (n=117) vor.

Als Ergebnis der Auswertung geht hervor, dass die gesamte gemittelte Median-Konzentration für Kupfer rund 30 µg/L für Nordamerika und ungefähr das Doppelte für Asien, Europa und Ozeanien beträgt. Die maximale Kupfer-Konzentration in Europa ist mit 430 µg/L ebenfalls höher als die in Nordamerika gemessene Konzentration von

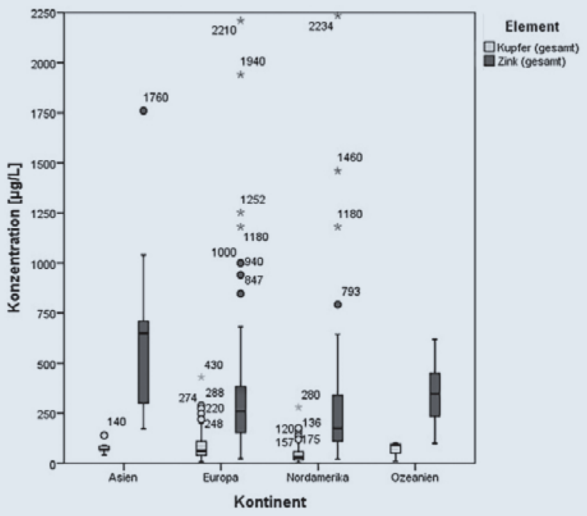

Bild 5: Vergleich der Zink- und Kupfer-Konzentrationen (gesamt) in Verkehrsflächenabflüssen verschiedener Kontinente (Box-Whisker-Plots: oberes bzw. unteres Ende der Box entspricht dem oberen bzw. unteren Quartil und der Strich in der Box dem Median; kleine Kreise stellen Ausreißer und Sterne Extremwerte dar)

280 µg/L. Die gesamte gemittelte Median-Konzentration für Zink liegt bei ungefähr 170 µg/L für Nordamerika, 250 µg/L für Europa, 300 µg/L für Ozeanien und 620 µg/L für Asien. Die höchsten Konzentrationen an Zink sind mit 1940 µg/L und 2210 µg/L für Europa und 2234 µg/L und 19 100 µg/L für Nordamerika nicht vergleichbar. Bei den Werten für Asien muss berücksichtigt werden, dass für Asien im Vergleich zu den anderen Kontinenten weniger Datensätze (n=13) verfügbar sind und die Probennahmen vornehmlich an hochbelasteten Verkehrsflächenkategorien erfolgten.

Zusammengefasst kann gesagt werden, dass die Median-Konzentrationen in Nordamerika für die beiden Schwermetalle am geringsten sind und die Unterschiede für Zink zwischen allen Kontinenten deutlich größer ausgeprägt sind als für Kupfer. Vergleichbare Ergebnisse wurden bereits in einer Übersicht von Autobahnabflüssen dargestellt [54].

4.5 Weitere Metalle

Zu den weiteren Metallen, die in Messprogrammen bestimmt werden und verkehrsbedingt erhöhte Konzentrationen aufweisen, gehören Kobalt (Diesel [55]), Mangan (Motoren [21], Bremsen [56], Reifen [57]), die Platin-Gruppen-Elemente (Palladium, Platin und Rhodium in Katalysatoren [58]), Antimon (Bremsen [59]), Titan (Fahrbahnmarkierungen [38]) und Wolfram (Spikes [60]).

Die höchsten in Einzeluntersuchungen gemittelten Konzentrationen in Europa reichen von unter 8 µg/L für Palladium [45] über bis zu 9,2 µg/L für Wolfram [61] bis zu 13,3 µg/L für Kobalt [61], bis zu 24 µg/L für Platin [62], bis zu 35 µg/L für Antimon [16], bis zu 86,6 µg/L für Barium [63], bis zu 329 µg/L für Mangan [57] und bis zu 630 µg/L für Titan [51]. Aus diesen Stoffen eignen sich Antimon, Kobalt und Wolfram aufgrund ihres geringen Vorkommens in der Umwelt als Tracer-Elemente zur Bestimmung des Einflusses des Verkehrs auf Umweltkompartimente [61, 64].

5. Zusammenfassung und Fazit

Zusammenfassend lässt sich sagen, dass die Einzelergebnisse eines Messprogramms für den untersuchten Ort charakteristisch sind und von dessen kontinuierlichen (umgebende Landnutzung, Verkehrsflächeneigenschaften sowie Verkehrsgeschehen) und klimatischen (Regen und Trockenperioden) Randbedingungen beeinflusst sind, wobei die Repräsentativität der über den gewählten Untersuchungszeitraum beprobten Ereignisse bezüglich der klimatischen Bedingungen (z. B. Verteilung der Regenintensitäten) nachzuweisen ist. Zusätzlich haben methodikspezifische Faktoren einen Einfluss auf die Abbildung der realen Verhältnisse. Dabei ist eine statistische Auswertung des Einflusses für jeden aufgeführten Faktor auf die jeweiligen Verkehrsflächenabflusskonzentrationen wegen der geringen Grundgesamtheit der entsprechenden Daten nicht möglich gewesen.

Aufgrund der breiten Datenbasis konnten für Europa viele Messprogramme mit verschiedenen Randbedingungen nach Verkehrsflächenkategorien (Parkplätze, drei Straßenkategorien und Autobahnen) ausgewertet werden. Als Ergebnis lässt sich für jede Kategorie eine Aussage zur Zusammensetzung und Belastung mit Schwermetallen treffen. Ein wichtiges Ergebnis ist, dass die Abflüsse an Hauptverkehrsstraßen mit überregionaler Bedeutung aufgrund der Randbedingungen meist stärker belastet sind als die Abflüsse von Autobahnen. Für eine fundierte Beschreibung der Belastung verschiedener Parkplatztypen (z. B. Mitarbeiterparkplätze, Supermarktparkplätze und Lkw-Stellplätze) fehlt derzeit eine breite Datenbasis, jedoch ist die Einteilung nach DWA-M 153 nachvollziehbar.

Neben den fehlenden Messprogrammen für Parkplätze sollten zusätzlich hochbelastete Innerortstraßen, vor allem mit besonderer Fahrweise wie Stop-and-go, untersucht werden, damit die Einflüsse des Fahrverhaltens auf die Abflusskonzentrationen besser bekannt sind, diese zukünftig statistisch ausgewertet und die Abflüsse entsprechend behandelt werden können.

Bei allen Messprogrammen sollten vermehrt die ortsspezifischen Randbedingungen, die Anzahl der beprobten Regenereignisse sowie die Art und Strategie der Probennahme vermerkt werden und auch der pH-Wert sowie die gelösten Konzentrationen der Schwermetalle bestimmt werden. Anhand dieser zusätzlichen Daten wird es möglich sein, sowohl an Parkplätzen, an denen derzeit viele dezentrale Behandlungsanlagen gebaut werden, als auch an Hauptverkehrsstraßen mit besonderen Gegebenheiten eine zukunftsorientierte Niederschlagswasserbewirtschaftung zu ermöglichen. Dabei ist für die weitergehende Behandlung von Verkehrsflächenabflüssen mit Filtermaterialien die Kenntnis der gelösten Anteile für eine ausreichende Reduzierung der Schwermetallkonzentrationen entscheidend, weshalb weitere Daten dazu benötigt werden.

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Autoren

Maximilian Huber, M.Sc.
Lehrstuhl für Siedlungswasserwirtschaft |
Technische Universität München |

Prof. Dr. rer. nat. habil. **Brigitte Helmreich**
b.helmreich@tum.de |
Lehrstuhl für Siedlungswasserwirtschaft |
Technische Universität München |
Am Coulombwall 8 |
D-85748 Garching

Prof. Dr.-Ing. habil. **Antje Welker**
Fachgebiet Siedlungswasserwirtschaft und Hydromechanik |
Frankfurt University of Applied Sciences |
Nibelungenplatz 1 |
D-60318 Frankfurt am Main

water

MDPI

Review

Stormwater Management: Calculation of Traffic Area Runoff Loads and Traffic Related Emissions

Maximilian Huber and Brigitte Helmreich *

Chair of Urban Water Systems Engineering, Technical University of Munich, Am Coulombwall 3, 85748 Garching, Germany; maximilian.huber@tum.de
* Correspondence: b.helmreich@tum.de; Tel.: +49-892-891-3719

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Abstract: Metals such as antimony, cadmium, chromium, copper, lead, nickel, and zinc can be highly relevant pollutants in stormwater runoff from traffic areas because of their occurrence, toxicity, and non-degradability. Long-term measurements of their concentrations, the corresponding water volumes, the catchment areas, and the traffic volumes can be used to calculate specific emission loads and annual runoff loads that are necessary for mass balances. In the literature, the annual runoff loads are often specified by a distinct catchment area (e.g., g/ha). These loads were summarized and discussed in this paper for all seven metals and three types of traffic areas (highways, parking lots, and roads; 45 sites). For example, the calculated median annual runoff loads of all sites are 355 g/ha for copper, 110 g/ha for lead (only data of the 21st century), and 1960 g/ha for zinc. In addition, historical trends, annual variations, and site-specific factors were evaluated for the runoff loads. For Germany, mass balances of traffic related emissions and annual heavy metal runoff loads from highways and total traffic areas were calculated. The influences on the mass fluxes of the heavy metal emissions and the runoff pollution were discussed. However, a statistical analysis of the annual traffic related metal fluxes, in particular for different traffic area categories and land uses, is currently not possible because of a lack of monitoring data.

Keywords: de-icing salt; Germany; heavy metal source; highway; mass balance; parking lot; pollution; road; site-specific factors; urban

1. Introduction

In most cases, four types of traffic related loads are used for calculating mass balances and fluxes [1]. Specific substance emission loads are presented in the literature for each vehicle per kilometer traveled (mg/(vehicle· km)) or for all vehicle kilometers traveled in a distinct catchment area during a specific period (mg/(ha· km)). Annual loads can be specified by the road length (g/km) or by the traffic area (g/ha). For the description of the pollution of a traffic area, a differentiation is necessary between total traffic related emission loads, atmospheric deposition loads (wet and/or dry), and runoff loads from traffic areas. For the calculation of runoff loads, long-term measurements of runoff concentrations and water volumes of a monitoring site with a distinct catchment area must be recorded, analyzed, and published. Although several researchers have evaluated the runoff concentrations of different traffic areas [2,3], only a part of these monitoring programs included the calculation of annual loads. This is based on the fact that during most monitoring programs, only a limited number of samples is collected and most objectives do not require the calculation of loads (e.g., manual (grab) samples for stormwater permit applications). Nevertheless, annual heavy metal loads normalized per hectare of impervious catchment area are necessary for mass balances and their determination is subsequently important for several purposes (e.g., modeling of stormwater quality and transportation into receiving water, calculation of metal masses removed by treatment systems, determination of pollution loads for laboratory test methods, calculation of regional or national mass balances, or the assessment of the environmental impact and benefit of different techniques to remove toxic substances in relation to their costs).

For stormwater management, determining traffic area runoff loads for different traffic area categories is necessary to design effective stormwater management practices [4]. Therefore, detailed information on differentiated traffic related sources (emissions because of leaded gasoline, tire wear, brake lining wear, roadway abrasion, weights for tire balance, guardrails, lampposts/signs, and de-icing salts) and areas (traffic area categories) is needed to determine their contributions to the total pollution of environmental compartments and to develop cost efficient mitigation strategies. Previously, urban area pollutant categories were often confined to broader land use categories such as commercial, industrial, or residential [4]. By this approach, the different pollution loads of traffic areas cannot be considered although all road surfaces represent approximately 10%–15% of the total urban area [5,6] and in commercial and industrial areas, parking lots can constitute up to 46% of the total area [5]. Thus, it is essential to consider the runoff loads more differentiated because of the large percentages of different traffic areas to the total urban area. However, most values for specific traffic area categories were available and summarized for highways and less were published for parking lots and other roads [7–9]. Because most values and ranges of pollutant loads published in books, reports, regulations, and standards are only based on a few studies, no reliable values are currently available for the main traffic area categories and a relation of the loads to climatic factors was also not evaluated previously. In addition, most published data summaries of runoff loads are based on the last century and they do subsequently not represent the latest trends.

In traffic area runoff, the occurrence of substances depends on several processes. Dustfalls and dry deposition during periods without rain entrain contaminants and remove them from the atmosphere [6,10]. These mechanisms are especially relevant for urban areas [11]. The processes during rain events are wet deposition (removal of additional substances from the atmosphere) and wash-off of previously deposited pollutants. Concerning the wash-off, Racin et al. [12] concluded that splashing and washing of pollutants from vehicles is more important than the wash-off of pollutants accumulated on road surfaces. Both the wash-off and the substances in the atmosphere can be linked to traffic related sources. For most heavy metals, the wear of brakes and tires are relevant sources. Moreover, braking, acceleration, and steering activities lead to increased abrasion of tires, higher use of brake linings, and increased automotive exhaust gas emissions [13,14]. The corrosion and subsequent dissolution of zinc (Zn) from the surface of galvanized elements during rain events is also a relevant source [15]. Further traffic related sources are the use of catalytic converters in vehicles [16], road maintenance with de-icing salts [17], road wear, and drip losses. All of these traffic related sources emit heavy metals, hydrocarbons, and further substances and subsequently lead to a pollution of runoff from traffic areas. Thus, the substances analyzed in traffic area runoff waters include solids, organic parameters, heavy metals, and compounds of de-icing salts [18,19]. Metals such as cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), lead (Pb), antimony (Sb), and Zn are crucial pollutants because of their toxicity, non-degradability, and their increase in all environmental compartments in consequence of their widespread industrial use [20,21]. Among these parameters, the three heavy metals Cu, Pb, and Zn were mostly measured in the highest concentrations in runoff from traffic areas [2]. Because of the relevant heavy metal concentrations, a treatment of the runoff water is often necessary to prevent negative effects on surface water, groundwater, and aquatic biota [22–24]. These stormwater treatment systems can be designed as a part of existing urban infrastructure for stormwater management [25].

The focus of this paper was set on the metals in traffic area runoff and their traffic related emissions. The hypothesis of this study was that a distinct calculation of mass balances for traffic related metals (emissions and runoff loads) of all types of traffic areas (i.e., highways, roads, and parking lots) is possible as a result of a literature study. The objectives of this paper were as follows: to summarize the distributions of annual metal loads of runoff from different types of traffic areas, to identify relevant trends and factors, to update and expand existing mass balances (heavy metal runoff loads and emissions) for Germany, and to determine the mass fluxes of traffic related metals.

2. Materials and Methods

The annual loads summarized in this paper were selected from a database of German and international studies with descriptions of the runoff from traffic areas. The database includes information on the monitored traffic area (categories: bridges, highways, parking lots, and roads), the substance concentrations and loads, and all influencing factors reported by the authors (e.g., land use characteristics, vegetation, topography, road design, operational characteristics, climatic factors, type of sampling, sampling strategy, sample preparation, sample analysis, and calculation methods). In addition to peer-reviewed journal papers, reports, books, and non-reviewed journal articles that presented data of metals in traffic area runoff were considered after validating the published results (i.e., to fulfill requirements such as a detailed description of the applied methods, the performance of quality control measures, and the reliability of the monitoring setup). The metal runoff concentrations and site-specific and method-specific (i.e., sample collection, preparation, and analysis) factors influencing the results were presented in another review [2]. From this database with more than 300 monitoring sites, this review includes all studies that presented traffic area runoff loads for the metals Cd, Cr, Cu, Ni, Pb, Sb, and Zn. The values were always reported as total (recoverable) metals that were measured after an appropriate digestion (i.e., strong acid or aqua regia digestion; sometimes combined with an autoclave or microwave). The sample preparation and analysis conducted by the selected studies had a low influence on the results because of quality assurance/quality control procedures [2]. All studies used automatic composite samplers to obtain a large number of samples for each rain event (active sampling) or a mixed sample of a part of the complete runoff (passive sampling). The highest uncertainties regarding all selected long-term monitoring programs are related to the handling of the raw data (e.g., time interval of the data logging and the link between flow-/rainfall-measurements and measured runoff concentrations) and the calculation methods for the annual loads (e.g., values below the detection limit or missing values because of a malfunction of a device). All monitored traffic areas were categorized in one of the three categories highway (H), parking lot (P), and road (R). Sites with reported runoff loads that are not completely linked to impervious traffic areas were categorized as special (S). Each monitoring site got an identification number (ID) that consists of one capital letter according to the category (H, P, R, or S) and two consecutive numbers, e.g., H01. These IDs were used in the Tables and Section 3.

The characteristics of the sites used for this review are summarized in Table 1. Table 1 includes data about the location (country), the period of sampling, the land use, special fixed site-specific factors, the road maintenance (i.e., winter services and sweeping), the average annual daily traffic (AADT), the discharge area of the monitoring site (i.e., the catchment area connected to the sampler), and the annual rainfall depth. The runoff loads of the metals are presented in Table 2. This novel evaluation of different types of traffic areas (H, R, and P) only used annual metal loads per hectare of catchment area (g/ha) that were calculated by the respective authors of the selected studies from runoff concentrations of impervious surfaces without a subsequent (pre-)treatment (i.e., some sites presented in [26–30] are summarized in the category S). Therefore, a conversion of units was sometimes performed (lb/acre, mg/m^2, or kg/ha in g/ha). In addition, if all required information was available, annual metal loads presented as g/km were recalculated to g/ha. A calculation of loads from runoff concentrations and the corresponding water volumes was not performed because of climatic site-specific factors (i.e., variable at each site) and several uncertainties (e.g., missing data about the percentage of analyzed runoff volumes to total volumes, the percentage of collected rain events to total rain events, the representativeness of the analyzed rain events in terms of intensities and durations, and the ambiguity between rainfall and runoff volumes).

The software package SPSS 22 (IBM) was used for statistical analysis and plotting. For box and whisker plots, the bottom and top of each box are the first and third quartiles and the band inside the box is the median. The whiskers represent 1.5 times the interquartile range (IQR). Outliers (>1.5 times IQR) are marked as small circles and extreme values (>3.0 times IQR) as stars. To analyze historical trends, the period of the monitoring program was used instead of the publication date. Because of the non-normal distribution of most parameters, the non-parametric Spearmen rank-order correlation was subsequently used for the correlation analysis following the example of Mosley and Peake [31]. For the correlation analysis, the sites H27, H28, and R02 were split up into two, four, and nine independent data sets, respectively, to consider the heavy metal loads and annual rainfall depths for each year. For H05, H06, and H08–H13, no values for the annual rainfall depths were used for the statistical analysis because of missing mean values.

Table 1. Characteristics of traffic area sites with heavy metal loads reported in the literature.

ID	Literature	Location *	Period of Sampling	Study Site Characteristics **	AADT (veh./d)	Discharge Area (m^2)	Rainfall (mm/yr)
H01	[32]	GBR	1973–1974	urban, B, D, S	57,600	36,000	533
H02	[1]	CHE	1976–1977	non-urban, D	72,000	54,300	1132
H03	[33]	FRA	1978–1979	–	13,600	–	–
H04	[26,27]	DEU	1978	non-urban, D, nS	41,000	13,000	792
H05	[34]	USA	1978–1981	urban	53,000	4937	810–1140
H06	[34]	USA	1978–1981	urban	42,000	401	810–1140
H07	[26,27]	DEU	1979	non-urban, D, nS	47,000	25,200	737
H08	[34]	USA	1979–1981	non-urban	8600	1133	1020–2290
H09	[34]	USA	1979–1981	non-urban	7700	728	1520–2540
H10	[34]	USA	1979–1981	non-urban	7300	1133	1780–2540
H11	[34]	USA	1979–1981	non-urban	2000	5059	178–381
H12	[34]	USA	1979–1981	urban, B, Zn smelter	17,300	890	432–711
H13	[34]	USA	1979–1981	non-urban	2500	1012	254–483
H14	[35]	FRA	1980–1982	non-urban	5500	1470	801
H15	[35]	FRA	1980–1982	non-urban	7000	–	620
H16	[36]	FRA	1993–1994	non-urban	30,000	13,000	–
H17	[30]	USA	1993–1995	urban, B	58,150	5341	1160
H18	[30]	USA	1994–1995	urban, B	8780	526	1670
H19	[37]	USA	1995–1996	urban	41,000	1670	945
H20	[37]	USA	1995–1996	urban	51,000	4940	945
H21	[28]	USA	1995–1996	urban, B, D	25,000	1497	–
H22	[38]	FRA	1995–1996	urban, B, D, S	12,000	3200	656
H23	[37]	USA	1995–1997	urban	120,000	1880	945
H24	[39]	DEU	1998–2000	urban, D	>50,000	75,800	774
H25	[39]	DEU	1998–2000	urban, D	>50,000	51,300	774
H26	[40]	CHE	1999–2000	non-urban	60,000	75,000	1050
H27	[41]	CHE	2006–2007	urban, D, S	59,000	25,000	876–1184
H28	[41]	CHE	2006–2009	non-urban, D, S	74,000	20,000	477–726
H29	[42]	DEU	2011–2012	non-urban, D	85,600	27.5	538
H30	[42]	DEU	2011–2012	urban, D	66,200	28.0	471
H31	[42]	DEU	2011–2012	non-urban, D	45,000	27.3	454
H32	[43]	CHE	2012–2013	urban, D, S	39,000	42,000	1147
R01	[44]	DEU	1988–1989	urban	18,129	162.2	609
R02	[45]	DEU	1996–2005	urban, D	6800	17.3	334–863
R03	[14]	CHE	2002–2004	non-urban	17,000	1500	1000
R04	[14]	CHE	2002–2004	urban	17,000	14.1	1000
R05	[46]	BRA	2002–2004	urban	9000	1300	–
R06	[47]	DEU	2006–2007	urban, D, S	57,000	100	739
R07	[48]	DEU	2014–2015	non-urban, D	20,600	9870	1200
R08	[48]	DEU	2015	urban, nD	9500	1600	800
P01	[29]	USA	1998–1999	urban	–	1050	1000
P02	[29]	USA	1998–1999	urban	–	1050	1000
P03	[49]	DEU	2001	non-urban, D, S	–	10,000	635
P04	[49]	DEU	2001	non-urban, D, S	–	5000	635
P05	[49]	DEU	2001	non-urban, D, S	–	17,700	769

Notes: * BRA (Brazil), CHE (Switzerland), DEU (Germany), FRA (France), GBR (Great Britain), and USA (United States of America); ** B = Bridge, D = De-icing salt application, nD = no De-icing salt application, nS = no Sweeping, S = Sweeping reported in the literature.

Table 2. Annual total heavy metal loads in runoff from traffic areas. The characteristics of all traffic area sites are described in Table 1.

ID	Literature	Cd Load (g/ha)	Cr Load (g/ha)	Cu Load (g/ha)	Ni Load (g/ha)	Pb Load (g/ha)	Zn Load (g/ha)
H01	[32]	–	972	3780	1000	12,900	19,000
H02	[1]	25	–	350	–	2500	1900
H03	[33]	–	–	–	–	1200	2300
H04	[26,27]	37	62	621	–	1330	2330
H05	[34]	–	–	223	–	4420	2320
H06	[34]	–	–	732	–	15,920	4210
H07	[26,27]	29	100	544	–	1160	2890
H08	[34]	–	–	34	–	130	220
H09	[34]	–	–	361	–	1670	2010
H10	[34]	–	–	565	–	5480	2560
H11	[34]	–	–	30	–	80	470
H12	[34]	–	–	118	–	690	10,400
H13	[34]	–	–	65	–	320	390
H14	[35]	–	–	–	–	410	1170
H15	[35]	–	–	–	–	820	1410
H16	[36]	–	–	–	–	140	1390
H17	[30]	–	–	230	–	470	1300
H18	[30]	–	–	80	–	200	450
H19	[37]	5.6	50.4	235	–	112	1300
H20	[37]	4.5	40.4	269	–	112	874
H21	[28]	30	90	220	90	200	–
H22	[38]	8.1	–	249	–	567	2080
H23	[37]	19.1	381	628	–	425	3230
H24	[39]	–	–	440	–	217	1960
H25	[39]	–	–	480	–	250	2120
H26	[40]	30	–	1070	–	560	1990
H27	[41]	–	–	440	–	80	1190
H28	[41]	–	–	359	–	55	1270
H29	[42]	1.4	–	805	–	88	2440
H30	[42]	0.6	–	241	–	44	878
H31	[42]	0.8	–	350	–	72	834
H32	[43]	–	–	750	–	–	3930
R01	[44]	–	–	500	–	1800	10,600
R02	[45]	1.7	41	225	41	83	2510
R03	[14]	4.0	119	471	–	179	3230
R04	[14]	3.2	75.4	398	–	170	2100
R05	[46]	8.0	–	310	–	500	–
R06	[47]	–	–	970	–	210	4820
R07	[48]	–	–	600	–	–	1300
R08	[48]	–	–	184	–	–	1060
P01	[29]	–	–	42	–	18	174
P02	[29]	–	–	33	–	17	147
P03	[49]	–	–	400	–	150	2400
P04	[49]	–	–	250	–	110	1000
P05	[49]	–	–	500	–	200	2000

For the calculations of median values, mean values, and standard deviations (SD) in Section 3.3, some data were excluded because of special site-specific factors (cf. Section 3.2.1). To avoid a bias because of the historical trends of the Pb data (cf. Section 3.2.2), the values of the 20th century were excluded for the discussion of recent loads. All of these exceptions are directly specified at the presentation and discussion of the particular results.

3. Results and Discussion

3.1. Annual Metal Runoff Loads for Different Traffic Areas

A summary of annual total metal loads of 45 sites is presented in Table 2 for all three traffic area categories (H, R, and P) and the six most relevant heavy metals (Cd, Cr, Cu, Ni, Pb, and Zn). Most of the values were published for highway sites (H, *n* = 32) with fewer values for other roads (R, *n* = 8) and parking lots (P, *n* = 5). The traffic area runoff loads of Cu, Pb, and Zn were mostly measured and published by other researchers. Less data are available for Cd, Cr, Ni, and Sb. For Sb, the annual average runoff load was only published for one urban road (16.8 g/ha at R02; [45]) and three highway sites (11.2 g/ha at H19, 5.6 g/ha at H20, and 17.9 g/ha at H23; [37]).

The most concentrated metal was Zn, followed by Pb, Cu, Ni, Cr, Cd, and Sb. The SDs between all monitoring results are quite high for all heavy metals summarized in Table 2: 12.9 g/ha Cd, 291 g/ha Cr, 587 g/ha Cu, 540 g/ha Ni, 3180 g/ha Pb, and 3330 g/ha Zn. In addition, the runoff loads of one to four sites are calculated as extreme values for most metals (Figure 1). Compared with the other heavy metals, the variations of the Pb and Zn data (e.g., cf. the SDs above and the IQRs presented in Figure 1) are the highest. For Pb, it is linked to the historical trends (cf. Section 3.2.2). In contrast, the contributions of different Zn sources to the runoff loads are highly variable between each site (e.g., brake linings, galvanized bridge parts, galvanized car parts, guardrails, lamp-posts, motor oil, tires, safety fences, and signs; [38,50,51]). Thus, these high loads are based on different fixed site-specific factors (cf. Section 3.2.1), operational characteristics (cf. Section 3.2.2) but can also be linked to climatic (cf. Section 3.2.3) and method-specific influences [2].

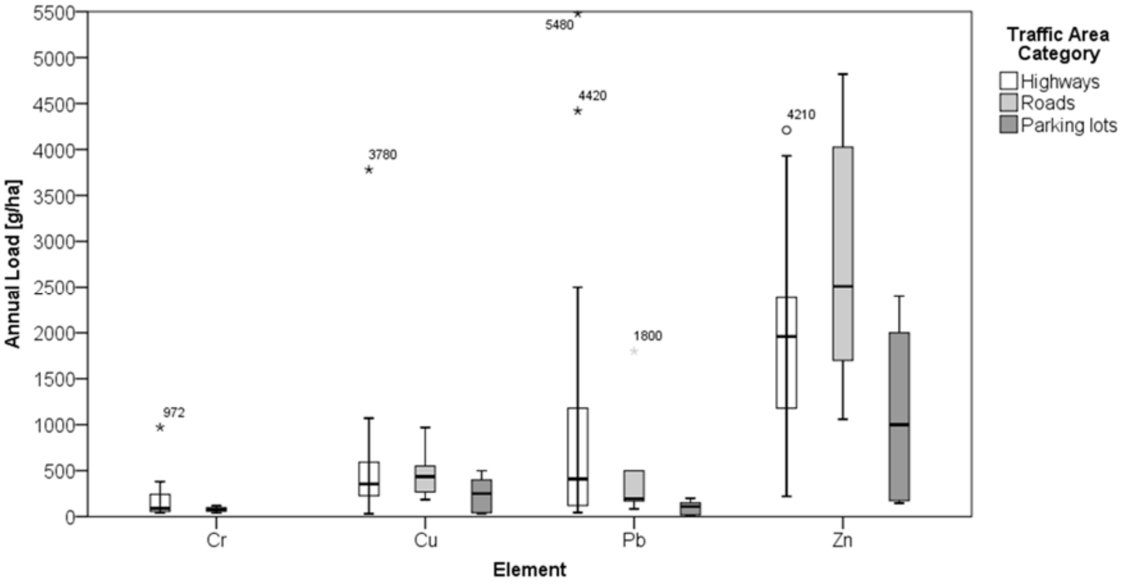

Figure 1. Annual total heavy metal loads of Cr (*n* = 10), Cu (*n* = 41), Pb (*n* = 42), and Zn (*n* = 43) in runoff from traffic areas for each of the three categories highways (H), roads (R), and parking lots (P).

For most values, the loads published for runoff from parking lots were lower than the loads of highway and road runoff (Table 2 and Figure 1). One further trend between the traffic area categories was found for the runoff loads from parking lots. The two parking lots P01 and P02 at the Florida Aquarium (USA) were less polluted compared with the three frequently used German highway parking lots for trucks (P03), cars (P04), and all types of vehicles (P05). Thus, the frequently used highway parking lots with high percentages of trucks have increased runoff loads and these values are comparable with the annual metal loads of the runoff from roads.

Strong and statistical significant correlations were determined for Cr with Pb, Cu, and Zn (Table 3) that represent the particulate fraction of these metals because Cr and Pb are mostly particulate in the runoff from traffic areas and Cu and Zn are also particulate but more dissolved [2]. Some significant correlations between the catchment area and the metal runoff loads were also calculated (Table 3). However, only an effect of the area on the first flush was previously reported [52].

Table 3. Spearman correlation coefficients of average annual daily traffic (AADT), catchment area, annual rainfall depths, and heavy metal runoff loads: correlation coefficient ⩽0.39 = weak correlation, 0.40–0.59 = medium correlation, and ⩾0.60 = strong correlation. Strong correlations are in bold type and no values are presented for Ni because of the small n (5 or 6 for all correlations).

Variable	Cd	Cr	Cu	Pb	Sb	Zn	AADT	Area	Rainfall
Cd	–	– –	– –	– –	– –	– –	– –	– –	– –
Cr	**0.76** ** *n* = 17	–	– –	– –	– –	– –	– –	– –	– –
Cu	0.49 * *n* = 24	**0.77** ** *n* = 18	–	– –	– –	– –	– –	– –	– –
Pb	**0.94** ** *n* = 24	**0.90** ** *n* = 18	0.43 ** *n* = 50	–	– –	– –	– –	– –	– –
Sb	−0.22 *n* = 12	0.08 *n* = 12	0.41 *n* = 12	−0.10 *n* = 12	–	– –	– –	– –	– –
Zn	0.21 *n* = 22	**0.69** ** *n* = 17	0.56 ** *n* = 51	0.50 ** *n* = 52	0.63 * *n* = 12	–	– –	– –	– –
AADT	0.44 * *n* = 24	**0.66** ** *n* = 18	0.47 ** *n* = 48	−0.01 *n* = 49	−0.32 *n* = 12	−0.03 *n* = 50	–	– –	– –
Area	**0.74** ** *n* = 24	0.54 * *n* = 18	0.41 ** *n* = 53	0.25 *n* = 52	−0.39 *n* = 12	−0.14 *n* = 53	0.43 ** *n* = 50	–	– –
Rainfall	**0.64** ** *n* = 22	0.29 *n* = 17	0.04 *n* = 43	0.22 *n* = 42	0.07 *n* = 12	−0.11 *n* = 45	0.02 *n* = 40	0.22 *n* = 44	-

Notes: * $p < 0.05$, statistical significance of the correlations; ** $p < 0.01$, statistical significance of the correlations.

3.2. Influences on the Metal Runoff Loads

3.2.1. Fixed Site-Specific Influences

One often-discussed factor is the impact of the AADT on the runoff pollution of traffic areas. However, AADT can only explain approximately 30% of the variations between different sites [53]. In this review, the AADT varies between 2000 vehicles per day and 120,000 vehicles per day for the highway and road sites (Table 1). Significant correlations between AADT and the heavy metals were found for Cd, Cr, and Cu (Table 3). Cr was the only metal with a strong correlation ($\delta = 0.66$). Thus, further site-specific factors have an influence on the annual loads. A general description of these site-specific factors that influence the metal runoff concentrations and loads from traffic areas is given in [2]. In the following, only the fixed site-specific factors that were responsible for the extreme values calculated for all 45 sites were discussed with the exception of Pb (cf. Section 3.2.2).

Most of the largest annual loads were measured for the urban site H01, which has a high AADT of 57600 vehicles per day. Further factors that might have led to the high runoff concentrations were the extreme use of de-icing salts, which had increased levels of metal contents (cf. Section 3.4.2), by the winter services (ca. 2100 $g/(m^2 \cdot yr)$), the grade of congestion during rush hours, and the road design (e.g., a section of the monitoring site was a bridge with concrete pillars) [32]. In addition to these high concentrations, uncertainties regarding the discharge area because of the steep slope at the ends of the drainage area causing additional water flowing into the monitored area might have led to the increased calculated annual runoff loads [32]. At H12, the presence of a nearby Zn smelter was responsible for the high Zn runoff loads [34]. The road design of R01 resulted in increased runoff loads because of the complete drainage of the catchment area by curbs and gutters (runoff volumes were calculated from the rainfall depths and an assumed runoff coefficient of 0.9), the presence of guardrails (at the median and at both sites of the road), and the stop-and-go traffic.

For the special sites (S, *n* = 10), grass swales were often used instead of conventional curbs and gutters to drain the catchment areas and the samples were often taken at the drainage pipes.

The category S consists of the highway sites S01–S04 and the parking lot sites S05–S10. Most metal runoff loads of category S (Table 4) are much lower than the ones reported for non-pre-treated runoff loads for comparable sites (Table 2). At S01, the surface was only sealed by 39.6% with asphalt and the rest of the drainage area were adjacent areas with surfaces vegetated by grass [26]. At S02, the catchment area consisted of 61% asphalt, which was drained to a catch basin, and the grassy shoulder that were both monitored [28]. A similar situation was present at S03 with 45% impervious asphalt and a grassy shoulder [28]. At S04, 37.6% of the drainage area was paved with asphalt and the highway runoff drained into a large grassy median that were both simultaneously monitored in mixed samples [30]. For the parking lots, several loads for pre-treated runoff were reported [29]: asphalt surfaces with subsequent vegetated swales (S05,S06), cement surfaces with subsequent vegetated swales (S07,S08), and pervious surfaces with subsequent vegetated swales (S09,S10). A comparison of the sites S05–S10 with the sites P01 and P02 shows that small alterations to parking lot designs can dramatically decrease runoff metal loads. The reduction of pollutants by grass swales was also measured for the adjacent sites H21/S02 and the data presented for S04 in [30]. These results concerning the runoff load reductions by pervious surfaces and vegetated swales can be used for designing stormwater management practices. For the design of vegetated swales, further site-specific factors such as the road design (e.g., crossings and roundabouts) and the grade of congestion (i.e., sites with frequent stop-and-go traffic) must be considered [54].

Table 4. Annual total heavy metal loads in pre-treated runoff from traffic areas.

Site	Literature	Cd Load (g/ha)	Cr Load (g/ha)	Cu Load (g/ha)	Ni Load (g/ha)	Pb Load (g/ha)	Zn Load (g/ha)
S01	[26,27]	7.2	12	130	–	360	715
S02	[28]	10	20	70	20	70	–
S03	[28]	50	50	100	50	130	–
S04	[30]	–	–	15	–	10	60
S05	[29]	–	–	8	–	2	37
S06	[29]	–	–	25	–	7	79
S07	[29]	–	–	8	–	3	42
S08	[29]	–	–	9	–	4	56
S09	[29]	–	–	3	–	1	20
S10	[29]	–	–	6	–	3	36

3.2.2. Historical Trends

The historical trends of traffic area runoff loads published in literature were analyzed for Cu, Pb, and Zn from the 1970s to date. The loads were aggregated into five decades and plotted as box and whisker plots in Figure 2.

For the Pb data, a significant decrease of the loads was detected as a function of time because of the phase-out and substitution of leaded gasoline. The decrease of Pb use as an anti-knocking agent started in the USA in the mid-1980s and was completed in 1996 [55]. In Europe, the situation was not as homogenous as in the USA. For example, Pb was almost completely phased-out in gasoline in Germany and several other countries of Western Europe since 1986 [56,57]. The phase-out in the European Union ended in January 2002, with the elimination of leaded gasoline in Italy [58]. At the same time, leaded gasoline was banned in China [15]. Further reductions of Pb usage are related to the substitution of Pb in tires and brake linings [38,59,60], lubricating oil and grease [50], and weights added to vehicles for tire balance [61,62]. These developments are responsible for the high Pb loads reported in older publications (cf. Table 2). However, tires still contain Pb [63,64] (cf. Section 3.4.2) and Pb is added to the gasoline used in classic cars. As a consequence, Pb is still found in runoff samples of the 21st century in low concentrations [2] and in younger vegetated infiltration swales in low contents [54]. Thus, only Pb data of the 21st century should be used for current mass balances (cf. Sections 3.3 and 3.4). For Cu and Zn, no historical trends were detected. The median values of the decades varied because of different characteristics of the sites monitored per decade. The evaluation of the Cu and Zn loads confirms the higher variability of Zn (e.g., cf. the IQRs presented in Figure 2) compared with Cu that is based on the different sources (e.g., presence of Zn in galvanized structures and crumbs of car tire rubber; cf. Section 3.4.2). In addition, Cu and Zn are simultaneously present in some sources such as brake linings [38,65] and, therefore, their runoff loads have a medium correlation (Table 3).

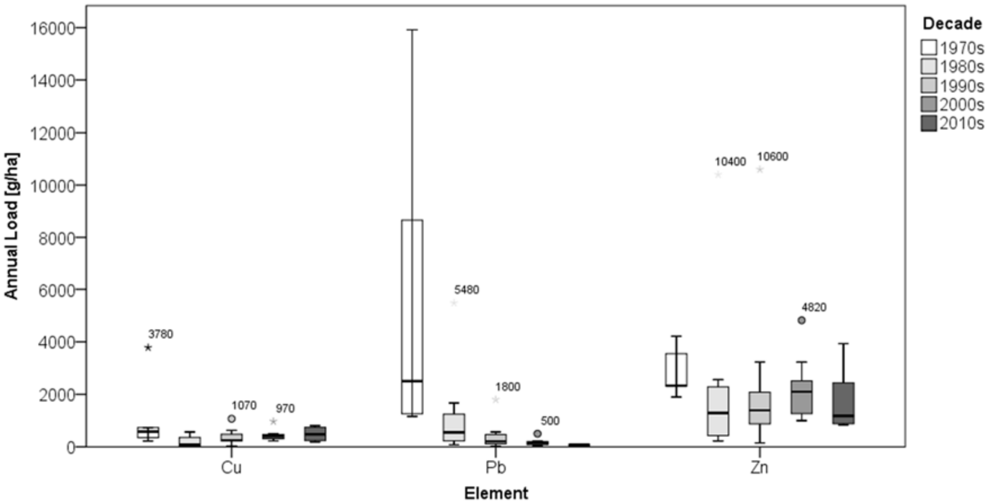

Figure 2. Historical trends of Cu (n = 41), Pb (n = 42), and Zn (n = 43) in traffic area runoff loads from the 1970s to date.

3.2.3. Annual Variations and Climatic Site-Specific Influences

Since most of the other sampling periods were only 1 to 2 years long (cf. Table 1), values of the annual runoff loads and annual rainfall depths are only available for each monitored year for the three traffic area sites R02, H27, and H28. Variations of the annual runoff loads (n = 9) were calculated for a nine-year monitoring program (R02) from the data presented by Nadler and Meißner [45]. The minima and maxima of the annual runoff loads calculated for each twelve months are 0.5–3.1 g/ha Cd, 17–76 g/ha Cr, 116–324 g/ha Cu, 20–60 g/ha Ni, 28–143 g/ha Pb, 10–23 g/ha Sb, and 1530–3550 g/ha Zn. The coefficients of variation (n = 9) for the annual runoff loads of R02 are 56% for Cd, 51% for Cr, 33% for Cu, 43% for Ni, 44% for Pb, 27% for Sb, and 30% for Zn. The catchment area, the sampling procedure, and most fixed site-specific factors (e.g., presence of a guardrail) were the same for the complete monitoring campaign. Only a small increase of the AADT was measured (6100 vehicles per day at the beginning and approximately 6800 vehicles per day in the second half of the monitoring program) and a different agricultural land use occurred in the east of the monitoring site. Since most annual metal loads of the second half of the monitoring program are lower than the loads of the first half, the influence of the AADT on the loads is not significant. Therefore, the variation of the heavy metals might be correlated with climatic factors such as the annual rainfall depth that varied largely between 334 mm and 863 mm (mean 667 mm; coefficient of variation 26%). However, a strong correlation ($\delta \geqslant 0.60$) between the seven metals and the annual rainfall depth was only calculated for Sb ($\delta = 0.68$, $p < 0.05$). Thus, the annual rainfall depths also had no significant influence on the variability of the annual heavy metal runoff loads for this site although most of the lowest loads were calculated for the year with low rainfall depths and long dry weather periods in the spring and summer.

Smaller annual variations of the heavy metal runoff loads were published for H28 (n = 4) for each of the four years that were monitored [41]: 340–381 g/ha for Cu, 47–63 g/ha for Pb, and 1200–1370 g/ha for Zn. The coefficients of variation (n = 4) are 5% for Cu, 13% for Pb, and 6% for Zn. The annual rainfall depths varied between 477 mm and 726 mm (mean 591 mm; coefficient of variation 22%) at this highway site. The small variations can be partially explained by varying street sweeping intervals of the hard shoulders [41]. Because of the small n, no correlation analyses were performed.

The variations of the annual rainfall depths at both sites were comparable (coefficients of variation are 26% and 22%). However, the variations between the metal runoff loads were much higher for R02 than for H28. These differences between the monitored years and sites can depend on a multitude of climatic boundary conditions (not only the rainfall depths), which can generally induce different deposition and wash-off processes, and on fixed site-specific factors (i.e., different median pollution loads of both sites). These influences on the median loads can be related to the different AADT and speed limits (50 km/h at R02 and 120 km/h at H28) and the presence of noise barriers, a forest, and hard shoulders at H28, which can also reduce the influences of climatic factors (e.g., wind turbulences) on the runoff loads [41]. Thus, the fixed site-specific factors also affected the influences of the climatic factors on the annual variations of the runoff loads.

At H27, with a speed limit of 120 km/h, the published annual heavy metal runoff loads were 470/410 g/ha for Cu, 71/88 g/ha for Pb, and 1010/1370 g/ha for Zn for the years 2006/2007 [41]. The annual rainfall depths were 876 mm and 1184 mm for the years 2006 and 2007, respectively. Thus, the rainfall depth was higher for 2007 and the runoff loads increased for Pb and Zn but not for Cu. Consequently, median runoff concentrations from the literature multiplied by different annual rainfall depths of a specific site cannot be used to describe the variance of the annual runoff loads because of climatic variations.

For all monitoring sites, the annual rainfall depth varied between 254 mm and 2540 mm (median 792 mm; 90th percentile 1200 mm). A strong correlation between the annual rainfall depths and the heavy metals was only calculated for Cd ($\delta = 0.64$, $p < 0.01$) for all monitoring sites (Table 3). The influence of the seasons on the pollution of traffic area runoff was evaluated by Helmreich et al. [66] with a considerable seasonal increase of pollutants in the runoff during the cold season. A correlation of the seasons could not be performed in this study because all published annual loads did not distinguish between different seasons. Therefore, only the results of single events can be used for this purpose. For example, the first storm of each season in (semi)arid regions is a special case for the climatic influences that is characterized by higher runoff loads [67,68].

In addition, the atmospheric deposition (wet and dry) must be considered as a climatic factor that affects the runoff loads. The deposition fluxes on traffic areas are strongly influenced by site-specific factors [69]. For example, the absence of curbs, hard shoulders, and noise barriers are site-specific influences that have a positive effect on reducing runoff concentrations by decreasing the deposition rates and the subsequent wash-off [41,70]. The deposition is also influenced by the traffic volume and the surrounding land use. Less data are available for urban traffic area sites (in particular roads and parking lots) at which higher fractions of metal emissions can be deposited and washed-off compared with non-urban traffic areas [11]. These rates are also influenced by the antecedent dry period [71]. However, Racin et al. [12] concluded that splashing and washing of pollutants from vehicles is more important than the wash-off of pollutants accumulated on road surfaces by precipitation (wet deposition). An increased importance of precipitation for runoff pollution of traffic areas in industrial zones is proposed by Dannecker et al. [72] and it is highly variable for each metal. Dry deposition at industrial sites also has an important effect, whereas the influences of traffic related emissions become more important at non-industrial urban sites [72]. For the non-urban sites H04, H07, and S01, the influence of the wet and dry deposition rates on the corresponding annual runoff loads was determined. The ratios of deposition loads to runoff loads for the sites H05/H07/S01 were 35%/31%/42% for Cd, 65%/2%/125% for Cr, 36%/23%/46% for Cu, 24%/27%/50% for Pb, and 54%/31%/80% for Zn [27]. Thus, the deposition affects the runoff loads at these non-urban highway sites and the influences depend on both the site and the metal.

In summary, the deposition is highly variable for each site because of site-specific factors and a robust data set is currently not available to determine the deposition loads for all types of traffic areas and the most relevant land use categories. For a rough estimation, the average deposition loads summarized by Zessner [73] can be used, which consist of traffic related and non-traffic related metals. He determined annual deposition loads of 1.4 g/ha Cd, 50 g/ha Cu, and 300 g/ha Zn for rural areas and annual deposition loads of 6.0 g/ha Cd, 250 g/ha Cu, and 1000 g/ha Zn for urban areas. Ilyin et al. [74] modeled annual deposition loads of 0.25–0.65 g/ha Cd and 8–20 g/ha Pb for Germany on a large scale. However, these data might underestimate the real values of metals deposited on traffic areas because of the large-scale model. For comparison, Kocher et al. [75] determined annual deposition loads for three different German highways and the rates varied strongly between each year, season, metal, and monitoring point (at ground level or 1.5 m height and with distance from the highways). Thus, method-specific factors also have an influence on the results. At 1.5 m height, the deposition rates determined by Kocher et al. [75] are in the range of the ones summarized by Zessner [73]. However, the values at ground level can be higher by a factor of up to approximately five for the measurements directly at the highway site. Thus, traffic related emissions highly affect the deposition rates on traffic areas.

A general description of the influences of climatic factors such as deposition rates, the antecedent dry periods, rain characteristics (volume, intensity, and duration), seasonal effects, and wind turbulence on the runoff from traffic areas is given in [2]. For rain characteristics, mostly poor correlations were obtained for the metal runoff concentrations. This is in accordance with the findings of this review (low correlation between annual rainfall depths and runoff loads). Thus, the differences of the runoff loads cannot be based on the uncertainties of the measured values and the high variability of the metal runoff loads between different years must be based on the variability of climatic factors and some fixed-site specific factors such as road maintenance (e.g., the variability of the use of de-icing salts by winter services; cf. Section 3.4.2). However, further data are needed to calculate the correlations between runoff loads and rainfall intensities or antecedent dry periods.

3.3. Average Runoff Loads for Different Traffic Area Categories

The values of the runoff loads presented in Table 2 were used to calculate median values, mean values, and SDs that could be used for mass balances (no values from H01 and Zn from H12). The calculated median annual runoff loads for all traffic areas are presented in Table 5.

Table 5. Median annual runoff loads (g/ha) for all traffic areas. For Pb, only data measured in the 21st century was used.

Parameter	*n*	Median	Mean	SD
Cd	16	6.8	13.0	12.9
Cr	9	75	107	106
Cu	40	355	384	254
Ni	2	66	66	35
Pb	13	110	149	119
Sb	4	14.0	12.9	5.7
Zn	41	1960	2018	1751

For the highway data, the calculated median runoff loads are presented in Table 6.

Table 6. Median annual runoff loads (g/ha) for all highway sites. For Pb, only data measured in the 21st century was used.

Parameter	*n*	Median	Mean	SD
Cd	12	14	16	14
Cr	6	76	121	130
Cu	27	355	388	260
Ni	1	90	90	–
Pb	5	72	68	18
Sb	3	11	12	6
Zn	29	1900	1773	1004

In a literature study, Hullmann and Kraft [76] presented annual average Cu and Zn loads for runoff from traffic areas that were summarized from several studies as approximately 800 g/ha for Cu and 2700 g/ha for Zn. These values are higher than the ones calculated in this study. For comparison, Driscoll et al. [7] reported the following ranges of annual heavy metal runoff loads measured in the USA ($n = 24$): 7.2–37 g/ha for Cd, 12–120 g/ha for Cr, 30–4670 g/ha for Cu, 70 g/ha for Ni, 80–21,200 g/ha for Pb, and 220–10,400 g/ha for Zn. The surrounding land use characteristics comprised nineteen urban sites (commercial, residential, and suburban) and five non-urban sites (forest, agricultural, and desert). Burton and Pitt [8] also summarized the runoff loads for parking lots, highways, and different land use categories that were published in five reports of the 1970s and 1980s. For parking lots, the reported values of the annual runoff loads are 10 g/ha Cd, 60 g/ha Cu, 800 g/ha Pb, and 800 g/ha Zn [8]. The published annual runoff loads are higher for highway sites: 20 g/ha Cd, 90 g/ha Cr, 370 g/ha Cu, 4500 g/ha Pb, and 2100 g/ha Zn [8]. With the exception of Pb, these annual highway runoff loads are only slightly higher than the median values calculated for highways in this review. In contrast, these summarized parking lot runoff loads are highly different compared with the parking lot values of Table 2 because the use of parking lots differ widely. Subsequently, the runoff loads of parking lots should not be summed up but considered in accordance to their type of use (cf. Section 3.1). Another relevant factor for parking lots is the contamination of parked vehicles with dirt, rust, and further substances [77] that must be considered for planning stormwater management strategies.

3.4. Heavy Metal Load Balances for Germany

3.4.1. Mass Balances of Heavy Metal Traffic Area Runoff Loads

In Germany, there are approximately 281.3 km^2 of highways and 339.5 km^2 of state roads [78]. The total amount of all types of traffic areas, which also include the areas for railroads, airports, and waterways, is 18,070 km^2 in Germany and the fraction of roads, places (e.g., parking lots and marketplaces), and sidewalks is 15,750 km^2 [79].

Since median values are preferred for the analyses of such field data [80], the median annual loads for highway runoff and the area of highways in Germany (281.3 km^2) were used to calculate the total amount of metal loads in runoff from German highways.. These total loads are presented as t/yr (1 t/yr = 1000 kg/yr) and were calculated as approximately 0.38 t/yr Cd, 2.1 t/yr Cr, 9.8 t/yr Cu, 2.5 t/yr Ni, 2.0 t/yr Pb, 0.31 t/yr Sb, and 53.4 t/yr Zn.

The calculated median annual heavy metal loads for all traffic areas, which are similar to the ones for the highway sites, and the total amount of all transportation areas (15,750 km^2) were used to estimate the total traffic area runoff loads for Germany. Thus, approximately 10.7 t/yr Cd, 119 t/yr Cr, 558 t/yr Cu, 103 t/yr Ni, 173 t/yr Pb, 22.1 t/yr Sb, and 3087 t/yr Zn are transported by runoff from traffic areas in Germany. Since less polluted traffic areas are also summarized in the total amount of transportation areas and no data are available for these categories regarding runoff loads, the calculated values tend to exaggerate the total runoff loads for Germany.

3.4.2. Mass Balances of Traffic Related Heavy Metal Emissions

Hillenbrand et al. [81] published heavy metal emission loads of different sources in a literature study for Germany. For the traffic related heavy metal emissions, they calculated the average loads for Cu, Pb, and Zn for the following sources: brake lining wear, tire wear, roadway abrasion, and Pb weights for tire balance. These data were adapted, updated, and extended by Cd and further sources (Zn weights for tire balance guardrails, lampposts/signs, and de-icing salts) for this review. The emission of Pb by leaded gasoline used by classic cars was not considered (only 0.6% of the cars in Germany are classic cars and their kilometric performances are low). A summary of the following results for the four metals Cd, Cu, Pb, and Zn is presented in Table 7. Both the metal contents of different sources (mg/kg) and the emission rates (mg/(vehicle· km)) are highly variable and only average values for each source are published to indicate their importance in relation to other emission sources.

Table 7. Traffic related heavy metal emissions in Germany (summary of mean values of the calculations presented in Section 3.4.2 that are based on different literature data).

Source	Cd (t/yr)	Cu (t/yr)	Pb (t/yr)	Zn (t/yr)
Tire wear	0.19	0.79	2.0	1255
Brake lining wear	0.03	928	61.5	309
Roadway abrasion	–	4.3	7.1	149
Weights for tire balance	–	–	2.2	6.6
Guardrails	–	–	–	313
Lampposts and signs	–	–	–	60
De-icing salts	0.70	1.75	11.6	1.75
Sum of all seven sources	0.92	935	84.4	2094

The first four sources are directly related to vehicles and the loads are determined by multiplying the metal contents of the material by their total abrasion masses. The tire wear was determined by Kocher et al. [82] based on 62 different tire samples for 13 metals and further substances. Kocher et al. [82] measured the metal contents of each tire (n = 62) and the coefficients of variation for the metal contents are 70% for Cd, 38% for Cr, 56% for Cu, 74% for Ni, 95% for Pb, 330% for Sb, and 35% for Zn. This is based on the different ages, manufacturers, and types of tires. The complete mass of tire wear is based on the emission factors reported in seven studies, which determined the tire wear as (mg/(vehicle· km)) for four vehicle categories (summarized in [81]), and the kilometric performances of the vehicles in Germany. By using the mean metal contents published by Kocher et al. [82] and an annual mass of tire wear of 111,420 t (coefficient of variation 29%) for Germany, the mean total emission loads for tire wear in Germany were calculated as 0.19 t/yr Cd, 0.51 t/yr Cr, 0.79 t/yr Cu, 0.97 t/yr Ni, 2.03 t/yr Pb, 0.42 t/yr Sb, and 1255 t/yr Zn. In contrast, Hillenbrand et al. [81] proposed annual emission loads of 2.7 t/yr Pb and 1620 t/yr Zn for tire wear. Thus, the calculated Pb and Zn loads of tire wear differ by approximately 20%–25% between these two studies. A summary of measured metal contents in tires and brakes was also presented by McKenzie et al. [64]. The contents also have a high variation for each metal (mostly one order of magnitude) and are in the order of the values summarized and measured by the German researchers [81,82]. The Cd emission load caused by brake wear was calculated with the metal content of 2.7 mg/kg presented by Legret and Pagotto [38], which is the median value of the contents summarized by McKenzie et al. [64] (coefficient of variation 120%), and the total brake lining abrasion mass of 12,500 t/yr published by Hillenbrand et al. [81]. The loads of brake lining wear for Cu, Pb, and Zn are taken from [81], which are based on the heavy metal contents of brake linings and their emissions published in eight studies separately for cars and trucks. In Germany, the brake linings of cars were responsible for 83%, 94%, and 80% of the total brake lining emission loads for Cu, Pb, and Zn, respectively. The emissions by weights for tire balances (Pb emissions reduced by 75% because of its substitution, mainly by Zn) and roadway abrasion were adapted from [81]. The roadway abrasion loads are based on the mean values of 18 road samples. The metal contents of the road surfaces varied between 0.0 and 31.5 mg/kg Cu, 0.9–29.0 mg/kg Pb, and 4–614 mg/kg Zn. Since only few data were available about the wear emission rates, in particular for different types of vehicles (e.g., cars, trucks, and buses), the uncertainties of the emission loads are high. Consequently, more data are needed to calculate reliable tire and break wear loads.

Guardrails, lampposts, and signs are parts of the road design and their heavy metal emissions must also be linked to road transportation [81]. The Zn emissions of lampposts and signs can only be a rough estimation because the number and surface area of traffic signs and further posts at traffic areas cannot be determined exactly. Most values are based on the assumptions proposed by Hillenbrand et al. [81].

The traffic related heavy metal emissions by de-icing salts for road maintenance were not previously evaluated for Germany. The novel calculated mean de-icing salt emissions (0.70 t/yr Cd, 1.75 t/yr Cu, 11.6 t/yr Pb, and 1.75 t/yr Zn) are based on an annual consumption of 3.5 million tons in Germany [78] and heavy metal contents of 0.2 mg/kg for Cd, 0.5 mg/kg for Cu and Zn, and 3.3 mg/kg for Pb in the de-icing salts [38]. However, these annual de-icing salt emissions are highly variable because of the different metal contents of the salts and the different annual consumptions that strongly depend on climatic factors. According to German regulation, the maximum permitted values for the soluble fraction of de-icing salts in Germany (sodium chloride, calcium chloride, and magnesium chloride) are 2 mg/kg Cd, 5 mg/kg Cu, 5 mg/kg Cr, 5 mg/kg Ni, 5 mg/kg Pb, and 20 mg/kg Zn [83]. In Great Britain, higher values were measured for a sodium chloride de-icing salt with 4.7 mg/kg Cr, 12 mg/kg Ni, 8.7 mg/kg Pb, and 6.0 mg/kg Zn [32]. Values of 1.2 mg/kg Cu and 1.6 mg/kg Pb were reported in [84] for sodium chloride (Cd and Zn not detected). For another sodium chloride de-icing salt, metal contents of 0.94 mg/kg Cd, 1.02 mg/kg Cr, 3.15 mg/kg Cu, 6.29 mg/kg Ni, 6.29 mg/kg Pb, and 1.57 mg/kg Zn were reported by Kobriger and Geinopolos [85]. Variations of the metal contents for highway de-icing salts were also published by Gupta et al. [86]: 0.003–0.01 mg/kg for Cr, 0–0.0004 mg/kg for Cu, 0.002–0.003 mg/kg for Ni, and 0.08–0.09 mg/kg for Pb. A comparison of the metal contents in de-icing salts with the ones measured in cooking salts [87] confirms that these small impurities can occur in all salts used in large quantities. Nevertheless, the total emission loads can be high because of the large amounts of salts used in Germany per year.

The total emissions are influenced by the highly variable annual consumption in Germany (0.46–5.26 million tons per year for the years 1990–2013). The metal contents of Cd, Cu, Pb, and Zn published in five references were multiplied by the annual consumption values in Germany for the years 1990–2013 (Table 8 and Figure 3). Each box and whisker represents the annual variation of the heavy metal emissions by de-icing salts for one element and the metal content of the corresponding reference. Thus, the mean de-icing salt emissions presented in Table 7 can only be a very rough estimation. In contrast to the other traffic related emission sources, the variability of these emissions mainly depends on climatic factors.

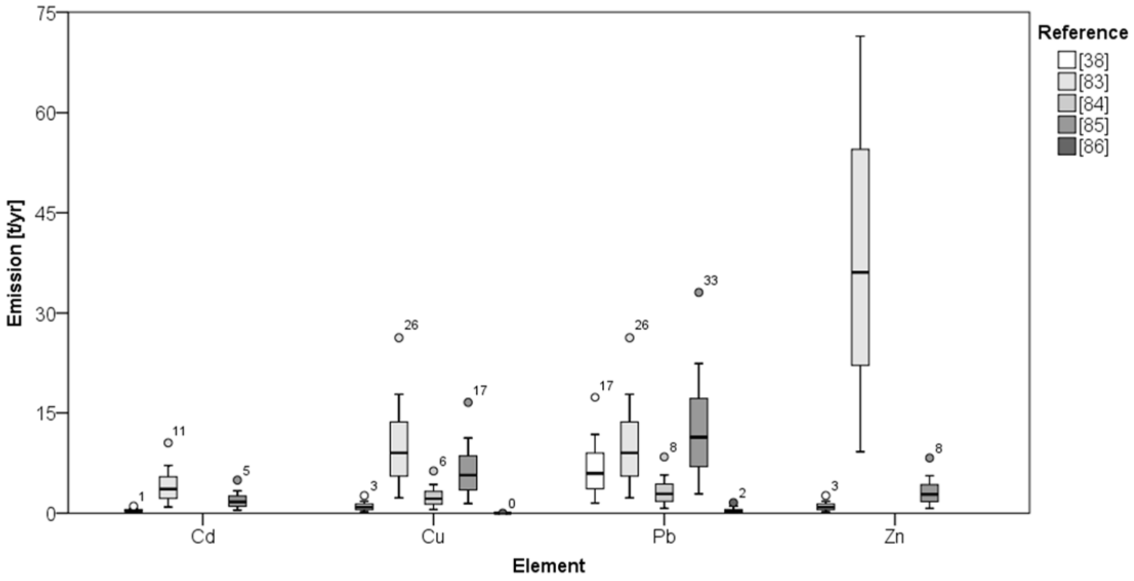

Figure 3. Variations of the traffic related heavy metal emissions by de-icing salts in Germany (1990–2013). Heavy metal contents of the de-icing salts differ largely within each reference.

Table 8. Variations of the traffic related heavy metal emissions (t/yr) by de-icing salts with different contents (cf. [38,83–86]) in Germany (1990–2013).

Parameter	**Min**					**Median**					**Mean**					**Max**				
	[38]	**[83]**	**[84]**	**[85]**	**[86]**	**[38]**	**[83]**	**[84]**	**[85]**	**[86]**	**[38]**	**[83]**	**[84]**	**[85]**	**[86]**	**[38]**	**[83]**	**[84]**	**[85]**	**[86]**
Cd	0.09	0.92	–	0.43	–	0.36	3.61	–	1.70	–	0.41	4.07	–	1.91	–	1.05	10.5	–	4.95	–
Cu	0.23	2.30	0.55	1.45	0.00	0.90	9.02	2.17	5.68	0.00	1.02	10.2	2.44	6.41	0.00	2.63	26.3	6.31	16.6	0.00
Pb	1.52	2.30	0.74	2.89	0.04	5.95	9.02	2.89	11.4	0.16	6.71	10.2	3.25	12.8	0.18	17.4	26.3	8.42	33.1	0.47
Zn	0.23	9.20	–	0.72	–	0.90	36.1	–	2.83	–	1.02	40.7	–	3.19	–	2.63	105	–	8.26	–

For the year 2013 (year 2000 in brackets), the German Federal Environment Agency published national total annual emissions for road transportation as 0.78 (0.82) t Cd, 25.7 (25.1) t Cr, 2109 (1986) t Cu, 5.0 (4.7) t Ni, 88.7 (80.8) t Pb, and 1906 (1805) t Zn [88]. There is a slight increase of the emissions for five of the six heavy metals in the 21st century. In 2013, the percentages of the traffic related emissions based on the total emissions are 11% for Cd, 45% for Cr, 98% for Cu, 5% for Ni, 42% for Pb, and 95% for Zn. Considering these data, road transportation is an important source of total heavy metal emissions in Germany. With the exception of the data for Cu, these values are comparable with the ones presented in Table 7. Denier van der Gon et al. [89] presented annual road transportation emissions for Germany for the year 2000 of 0.88 t Cd, 4.0 t Cr, 102.9 t Cu, 5.0 t Ni, 0.40 t Pb, and 209.8 t Zn. Most of these emission values are lower than the ones proposed by Hillenbrand et al. [81], calculated in this study, and published by Umweltbundesamt [88]. These differences are quite high and this is not only related to the variability of the data but also to the fact that each study considers different traffic related sources, which can also vary because of substitutions.

3.4.3. Transportation of Traffic Related Heavy Metals

A sustainable stormwater management combines the stormwater source control and the treatment of traffic area runoff loads to reduce the impact of heavy metals on receiving waters. Therefore, the influence of the traffic related heavy metal emissions on the runoff loads because of leaded gasoline, tire wear, brake lining wear, roadway abrasion, weights for tire balance, guardrails, lampposts/signs, and de-icing salts must also be considered to identify cost efficient mitigation strategies. This process was successfully implemented by the phase-out of Pb in recent decades (cf. Section 3.2.2). Nevertheless, not all traffic related emissions are found in the runoff because of wind turbulences, vehicles (traffic volume and vehicle speed), antecedent dry periods, and other site-specific factors (e.g., particle size distribution, land use, road surface roughness, or elevated configuration of the traffic area; [69]). For example, Hallberg et al. [90] found out that splashing corresponds with the average vehicle speed during storms. In Great Britain, Harrison et al. [91] measured that 90% of Pb emissions from fast-moving vehicles are deposited away from the immediate vicinity of a non-urban highway (up to several hundred meters). Consequently, most of the emitted Pb was not measured in the runoff. In contrast to Pb, Harrison and Johnston [92] showed that the Cd and Cu deposition fluxes were elevated close to the investigated highway section and decreased to background levels within 20–40 m. Langbein et al. [14] and Steiner et al. [93] measured the different fluxes of Cd, Cr, Cu, Pb, and Zn emissions at a non-urban road with approximately 17,000 vehicles per day in Switzerland (average vehicle speed between 60 km/h and 80 km/h). They evaluated that 17%–25% of the heavy metals were found in the runoff, 21%–38% in the splash water, 19%–45% were blown away and deposited in a distance of up to 25 m away from the road, and 17%–37% were deposited far away (>25 m). Thus, only some heavy metals were found in the runoff of a non-urban road and the fluxes of the emitted heavy metals varied between each element. Legret and Pagotto [38] calculated emission fluxes and estimated the percentages of Cd, Cu, Pb, and Zn that were removed by runoff waters. The following were considered as emission sources: tire and brake wear, gasoline, safety fences, de-icing salts, and atmospheric deposition (wet and dry). At the investigated rural highway bridge, 313% of the emitted Cd, 2% of Cu, 5% of Pb, and 37% of Zn were found in the runoff water and the rest was dispersed into the atmosphere as dust. For Cd, the authors assumed that some sources might be unrealized or underestimated. Hewitt and Rashed [94] also measured that the largest part of Pb (approximately 86%) was dispersed in the atmosphere. Most of these mass fluxes are proposed for non-urban sites and bridges. In contrast, a higher percentage of the traffic related emissions (e.g., 50%) might be found in urban runoff from traffic areas [14]. Thus, the heavy metal emissions in the urban areas influence the runoff concentrations and loads more directly [11]. Moreover, all traffic related emissions that were not found in the runoff (i.e., approximately 50%–98% according to the above data) would be deposited on non-traffic areas and consequently have an effect on the environment.

The influence of traffic related emissions on the runoff loads can be determined for de-icing salt applications on highways. Granato [95] calculated mean annual runoff loads as 4100 g/ha Cl, 1.8 g/ha Cr, 6.6 g/ha Cu, 2.0 g/ha Ni, and 3.6 g/ha Pb for the application of sodium chloride on highway runoff. Compared with the median runoff loads for highways calculated in this review (76 g/ha Cr, 355 g/ha Cu, 90 g/ha Ni, and 72 g/ha Pb), the de-icing salt emissions contribute to about 2.4%, 1.9%, 2.2%, and 5.0% of the total annual runoff loads for Cr, Cu, Ni, and Pb, respectively. Higher ratios were calculated for H01 with 38% for Cr, 56% for Ni, 2.2% for Pb, and 1.5% for Zn [32]. Thus, the use of de-icing salts with lower metal impurities can directly reduce the runoff loads by some percentages although de-icing salts are only used for several months per year.

In summary, the identification and subsequent reduction or substitution of relevant traffic related heavy metal sources (cf. Section 3.4.2) can help to fulfill the requirements of a sustainable stormwater management. Since the runoff loads are also influenced by special site-specific factors (e.g., nearby industries like Zn smelters) and non-traffic related emissions, a runoff treatment of distinct traffic area categories (e.g., highways, roads, and frequently used parking lots) is also necessary. This can be done by permeable pavements, grass swales, and technical stormwater treatment systems [96,97].

4. Conclusions

For stormwater management, an improved knowledge about the runoff loads of different traffic area categories is necessary. These loads were summarized for highways, roads, and parking lots. The highest loads were determined for highways, roads, and frequently used parking lots (in particular by trucks). In contrast to highways and roads, parking lots must be considered individually because of site-specific factors. The traffic related emissions in Germany were estimated for seven different sources (tire wear, brake lining wear, roadway abrasion, weights for tire balance, guardrails, lampposts/signs, and de-icing salts). Zn is mostly emitted by galvanized elements and tires, Cu and Pb by brakes, and Cd by de-icing salts. The calculated loads are comparable with the ones presented in other studies for most metals. However, a statistical analysis of traffic related metal mass fluxes (e.g., the contribution of metal emissions on the runoff loads, in particular for different traffic area categories and land uses) is not currently possible because of a lack of monitoring data (e.g., metal contents of different sources, emission factors, deposition rates for different traffic area categories and land use characteristics, and size characterizations of distinct catchment areas for different traffic area categories).

Nevertheless, the estimation of the runoff loads and the emission loads for Germany stated that the vehicles, the road design, and the winter services emit heavy metals in large quantities (0.93 t/yr Cd, 935 t/yr Cu, 84.4 t/yr Pb, and 2094 t/yr Zn) and the runoff also contains high amounts of metal loads per year. Currently, the most relevant metals are Cu and Zn because the annual Pb loads have decreased significantly in the last few decades and traffic related Cd and Ni contribute only 5% and 11% of the total emissions in Germany, respectively. However, the loads of the other metals can also be detrimental for receiving waters and aquatic biota. In particular, for the highly toxic metals Cd and Sb. Thus, efforts should be intensified to reduce traffic related loads (both emissions and runoff) and subsequently to minimize their input into receiving waters and soils.

For the future, more long-term data, which include a large variety of substances and a detailed description of climatic factors, must be measured and published to improve the determination of metal loads, mass balances, and fluxes and to reduce the large uncertainties of several values used in this review for the calculations.

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Chapter 6: Research Paper: "Evaluation of Factors Influencing Lab-Scale Studies to Determine Heavy Metal Removal by Six Sorbents for Stormwater Treatment"

Based on the results of Chapter 5, the heavy metals Cu, Ni, and Zn are relevant pollutants in the runoff from traffic areas. Thus, these metals must be retained by decentralized stormwater treatment systems. To remove dissolved and particulate heavy metals, the use of filter materials is necessary. For testing filter materials for urban stormwater treatment systems, it is important to assure that reproducible and reliable results are obtained. Therefore, the research objectives of this study were to identify relevant experimental boundary conditions concerning the simultaneous removal of Cu, Ni, and Zn and to evaluate important influencing factors by testing six filter materials with different removal mechanisms. Thus, this journal paper deals with the evaluation of different materials by batch and lab-scale column experiments. The hypothesis of the research article was:

The results of batch experiments and lab-scale column experiments concerning simultaneous heavy metal removal differ widely because of varying boundary conditions (Hypothesis 3).

The results of batch and small-scale column experiments concerning simultaneous heavy metal removal differed widely for each filter material. Hence, the following boundary conditions and factors should be considered to determine heavy metal removal capacities: batch experiments with the dissolved heavy metals of interest using synthetic water at pH 5 are suggested and the influence of the type and speed of the shaker must be considered. Adjustments of ionic strength and pH > 5 are more sensitive and should be avoided because they mostly lead to an overestimation of the removal capacities of filter materials. The filtration step at the end of the batch experiments has an effect on the results and does not simulate real situations. Batch experiments with other cations (e.g., magnesium and ammonium) should not be used as surrogates. Thus, the following applications are recommended: batch experiments with heavy metals can only be used under identical and well-defined conditions to prove the comparability of different production batches and to select filter materials for subsequent column experiments. In contrast, small-scale column experiments can be used as an indicator to determine roughly the efficiencies and service lives of treatment systems.

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Evaluation of Factors Influencing Lab-Scale Studies to Determine Heavy Metal Removal by Six Sorbents for Stormwater Treatment

Maximilian Huber, Sophia C. Badenberg, Moritz Wulff, Jörg E. Drewes and Brigitte Helmreich *

Chair of Urban Water Systems Engineering, Technical University of Munich, Am Coulombwall 8, 85748 Garching, Germany; maximilian.huber@tum.de (M.H.); sophia.badenberg@tum.de (S.C.B.); moritz.wulff@tum.de (M.W.); jdrewes@tum.de (J.E.D.)

* Correspondence: b.helmreich@tum.de; Tel.: +49-892-891-3719

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Abstract: For the development of decentralized treatment systems for road runoff, the determination of pollutant removal capacities is essential. The aim of this study was to evaluate the impact of boundary conditions on the simultaneous removal of copper, nickel, and zinc by six sorbents used for urban stormwater treatment (*i.e.*, granular activated alumina, anthracite, granular reactivated carbon, granular ferric hydroxide, calcium carbonate, and granular activated lignite). For batch experiments, capacities were determined at initial concentrations within the range of 2.5–180 mg/L with a rotary shaker. Further influences were investigated: the use of a horizontal shaker for concentrations of up to 1080 mg/L, a variation of the initial pH value (5 and 7), and the presence of a buffer. Furthermore, the influences of the filtration process on the capacities were studied. Kinetic experiments were conducted for contact times between 5 min and 120 min. Lab-scale column experiments with inflow concentrations of 2.5 mg/L (copper and nickel) and 5.0 mg/L (zinc) at an initial pH of 5 and a contact time of 11 min were performed for comparison. Selected experiments were subsequently carried out with changes in initial concentrations and contact time. One result is that it is essential to conduct batch experiments with the metals of interest. The capacities determined by column experiments deviated from batch experiments. Batch experiments under well-defined conditions can be used to evaluate different production batches. Column experiments give a more faithful capacity by considering realistic boundary conditions and should be preferred to determine efficiencies and service lives.

Keywords: batch experiments; column experiments; filter material; filtration; ion exchange; ionic strength; kinetics; pH value; sorption; urban stormwater management

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1. Introduction

Metals such as arsenic (As), cadmium (Cd), chromium (Cr), cobalt (Co), copper (Cu), lead (Pb), nickel (Ni), and zinc (Zn) are crucial pollutants because of their toxicity, lack of degradability, and their ubiquitous presence in the environment as a consequence of their industrial use [1–3]. Among these metals, the last four elements occur in highest concentrations in highway runoff [4,5]. Pb concentrations have decreased significantly in all traffic area runoff waters in the last few decades because of the phase-out of leaded gasoline and the substitution of Pb in other traffic-related sources. Thus, Cu, Ni, and Zn are, nowadays, measured in the highest runoff concentrations (maxima of mean concentrations per monitored site for the 21st century: 682 µg/L Cu, 458 µg/L Ni, and 2560 µg/L Zn—calculated from the worldwide data presented in [5]). These three heavy metals tend to be less particle-bound than other heavy metals such as Pb [6,7]. Sources of Cu in runoff water are brake linings [3,8], metal platings, moving engine parts [9], and pesticides applied to road medians [10]. Ni is present in diesel fuel [1], Ni-Cd batteries, stainless steel, and other electronic equipment [11]. The following are products containing Zn: batteries [12], brake linings [13], galvanized car parts [14], motor oils and grease [9], safety fences [15], and tires [16,17]. Furthermore, Cu and Zn show high accumulation potentials, biochemical activities, lifetimes, and solubility; Ni also has a high biochemical activity and lifetime [1].

To retain these heavy metals from traffic area runoff, decentralized stormwater treatment systems are used, which can be designed as an integral part of existing urban infrastructure [18]. Sedimentation and filtration are the retention mechanisms used to remove particle-bound metals. Dissolved heavy metals were retained on filter materials by sorption, ion exchange, and precipitation. To reduce particulate loadings for the filter materials, pretreatment ahead of the filter is required [19]. Both mechanical and chemical steps are necessary to sufficiently reduce dissolved and total metal concentrations [20,21]. Laboratory studies of the filter materials are necessary to develop new treatment systems, to control and optimize existing ones, and to determine application areas and service lives.

In the past, several types of laboratory studies have been conducted to evaluate different filter materials for stormwater treatment. Concerning the determination of cation exchange capacities, metals such as barium and magnesium are sometimes used as surrogates for the removal of heavy metals in time-efficient one-step methods [22]. In equilibrium batch experiments (48 h), the simultaneous removal of As, Cd, Cr, Cu, Ni, and Zn from stormwater was investigated by Genç-Fuhrman *et al.* [23] for eleven different sorbents with eight different initial metal concentrations. An equilibrium batch setup was also used by Wu and Zhou [24] to determine the simultaneous removal of the same six metals with eleven different initial concentrations on granular ferric hydroxide and its mixtures with zeolite and crystal gravel. Liu *et al.* [25] used manganese oxide coated polymeric media to evaluate the removal of Cd, Cu, Pb, and Zn in a recirculating flow-through reactor. In contrast to the static nature of batch experiments and their tendency to reach equilibrium, column experiments can help to assess the behavior of large-scale treatment systems with continuous influent concentrations, specific empty bed contact time (EBCT), and individual influent flow-rates [26]. Therefore, lab-scale column tests, which are less time-consuming and need smaller volumes of stock solution compared with pilot-scale experiments, can be used as an approach to evaluate the performance of sorbents at a small scale [27]. To evaluate filter materials for the removal of heavy metals, Li and Helmreich [28] conducted lab-scale column experiments with granular activated carbon and lignite to determine the removal of Zn. Liu *et al.* [19] used conventional and engineered media to evaluate the removal of Cd, Cu, Pb, and Zn (*i.e.*, silica sand, granular activated carbon, cementitious media, manganese oxide coated media, and other coated media).

However, a standardized protocol to compare filter materials for stormwater treatment systems for both batch and column experiments is missing. Therefore, the hypothesis of this study is that the results of batch- and lab-scale column experiments concerning simultaneous heavy metal removal differ widely because of varying boundary conditions.

The research objectives of this study were to identify relevant boundary conditions concerning the simultaneous removal of Cu, Ni, and Zn and the evaluation of important influencing factors by testing six sorbents with different removal mechanisms. By adjusting and standardizing these boundary conditions, an optimal use of the lab experiments can be achieved for their practical application in the field of stormwater treatment (e.g., type and amount of sorbent, removal efficiencies, ratio of drainage area to filter area, or service life). All of the materials used for this study were relatively coarse (main particle size 1.0–2.0 mm) to secure high permeability during service life and to prevent clogging of the filter. Furthermore, different production batches were evaluated to determine the use of batch experiments with well-defined methods for internal and external quality control.

2. Materials and Methods

2.1. Sorbents and Chemicals

The sorbents used were: granular activated alumina (alumina, Albemarle Corporation, Baton Rouge, LA, USA), anthracite (anthracite, EVERS GmbH & Co. KG, Hopsten, Germany), granular reactivated carbon (GAC, Chemviron Carbon GmbH, Beverungen, Germany), granular ferric hydroxide (GFH, HeGo Biotec GmbH, Berlin, Germany), calcium carbonate (calcium carbonate, Adolf Steinbach Steinindustrie-Schotterwerke GmbH & Co. KG, Bad Neustadt an der Saale, Germany), and granular activated lignite (lignite, Rheinbraun Brennstoff GmbH, Frechen, Germany). The physical characteristics of the sorbents, which were all commercially supplied, are provided in Table 1. Filter materials that retain heavy metals only by ion exchange and that can be improved or regenerated by sodium chloride (e.g., zeolites) [29,30] were not used in this study because they do not fulfill the requirements of a reliable heavy metal removal from traffic area runoff waters containing de-icing salts. All sorbents were sieved to a similar size fraction of 0.5–2.0 mm to avoid the influence of different size fractions of each sorbent on the retention of heavy metals [25,31]. The chosen size fraction (main particle size 1.0–2.0 mm) also fulfills the requirements of technical decentralized treatment systems (*i.e.*, permeability and filtration of particles). The sieved materials were subsequently dried at 105 ± 2 °C to achieve a constant weight, and finally stored in desiccators without additional treatment until usage.

Table 1. Physical characteristics of sorbents (based on dry mass).

Filter Material	Surface Area m^2/g	TC %	TS %	Al mg/kg	Ca mg/kg	Fe mg/kg	Mn mg/kg	Cu mg/kg	Ni mg/kg	Zn mg/kg
Alumina	263	-	-	462000	87.6	1270	7.43	18.6	1.98	9.11
Anthracite	1.4	88.2	0.91	931	1010	2260	20.5	16.0	2.57	1.56
Calcium carbonate	3.1	-	-	3520	318500	4370	78.7	3.50	2.91	5.04
GAC	896	91.6	0.83	6300	3050	3260	61.8	458	28.4	7.92
GFH	224	-	-	753	56300	431000	9090	2.00	41.3	87.6
Lignite	262	89.2	0.46	1090	18800	2330	83.1	0.17	1.69	0.37

The heavy metals used were: copper(II) nitrate trihydrate pure (AppliChem GmbH, Darmstadt, Germany), nickel(II) nitrate hexahydrate p.a. (Merck KGaA, Darmstadt, Germany), and zinc nitrate hexahydrate pure (AppliChem GmbH, Darmstadt, Germany). To determine the cation exchange capacities, barium chloride dehydrate p.a. (Merck KGaA, Darmstadt, Germany), magnesium sulfate heptahydrate p.a. (Merck KGaA, Darmstadt, Germany), sodium chloride 99.9% (AppliChem GmbH, Darmstadt, Germany), and ammonium chloride pure (Merck KGaA, Darmstadt, Germany) were used. pH adjustments and conservation of samples were performed with nitric acid 65% p.a. (Merck KGaA, Darmstadt, Germany). A combination of potassium dihydrogen phosphate p.a. (Th. Geyer GmbH & Co. KG, Renningen, Germany) and disodium hydrogen phosphate dihydrate p.a. (Merck KGaA, Darmstadt, Germany) was used as a buffer.

2.2. Experimental Setup

For all of the experiments with heavy metals, the concentration of Zn was always twice as high as the concentration of Cu and Ni, respectively. This represents the higher concentrations of Zn in traffic area runoff waters [5]. Equal concentrations of Cu and Ni made it easier to detect a possible competition between the removals of each metal.

2.2.1. Batch Experiments with Heavy Metals

For the equilibrium batch tests, sorption experiments were carried out in 250 mL low-density polyethylene (LDPE) bottles, at pH 5, and with initial metal concentrations from 2.5 mg/L to 90 mg/L for Cu and Ni, and from 5.0 mg/L to 180 mg/L for Zn. Deionized water (electric conductivity of <1 µS/cm) was used for preparing all stock solutions and the initial pH value was adjusted with nitric acid. At pH 5, all three heavy metals are present in dissolved form [23]. This initial pH value was chosen because filter materials in stormwater treatment systems are mainly designed to retain dissolved substances. The filter material dosage was always 8.0 g of filter material in 200 mL stock solution. No delay occurred between the addition of the solution to the sorbent in the bottles and the beginning of the shaking. Shaking was performed on a rotary shaker at a speed of 16 rpm for 24 ± 0.5 h. Subsequently, samples were filtrated with a cellulose nitrate membrane filter (0.45 µm pore size, Sartorius, Göttingen, Germany) to separate the dissolved and solid phases. All experiments were carried out in duplicate at room temperature (21 ± 2 °C).

Control experiments were conducted with all six sorbents without adding heavy metals. They followed the same experimental procedure as described previously to observe a possible release of heavy metals from the sorbents. In addition, further blank and spike samples were included into the experimental setup to detect possible sample contaminations during the filtration process and subsequent sample preparations and analyses. Samples with the same heavy metal concentrations, but without sorbents, were used to evaluate any possible heavy metal losses because of adsorption to the flasks and bottles. No significant losses of heavy metals were detected for any materials.

Differences between the rotary shaker (16 rpm) and a horizontal shaker (125 rpm) were examined for concentrations of up to 540 mg/L (Cu and Ni) and 1080 mg/L (Zn). In addition, the presence of a buffer was tested at pH 5 and pH 7 with the rotary shaker. A pH value of 7 represents commonly observed values for traffic area runoff [5]. Since the runoff water from traffic areas consists of several compounds that constitute a buffer capacity, other studies used sodium hydrogen carbonate buffers in the stock solution [23,24]. Since carbonate leads to a precipitation of heavy metals similar to the sorbent calcium carbonate and its addition influences the removal mechanisms, a phosphate buffer was used to minimize pH changes in this evaluation. Furthermore, the influence of the filtration process on the measured concentrations was studied by using further samples without sorbents, and by following the same experimental procedure.

Batch kinetic experiments were conducted with two concentration ranges (10 mg/L and 90 mg/L for Cu and Ni, respectively; 20 mg/L and 180 mg/L for Zn). The contact times were 5 min, 10 min, 20 min, 30 min, 60 min, and 120 min, respectively.

For the evaluation of different production batches, only Cu with an initial concentration of 63.5 mg/L was used. In contrast to the other experiments, the dosages were 0.4 ± 0.05 g material and 0.3 ± 0.05 g material in 250 mL solution.

2.2.2. Column Experiments

Lab-scale column experiments were conducted with inflow concentrations of 2.5 mg/L (Cu and Ni) and 5.0 mg/L (Zn) at an initial pH of 5 and at ambient temperature (21 ± 2 °C). Deionized water (electric conductivity of <1 µS/cm) was used for influent preparation. pH adjustments were performed with nitric acid. The experimental apparatus was constructed of glass and all pipes were of polyvinyl chloride. The glass column had an inner diameter of 2.0 cm and a length of 40 cm, providing a sorbent bed height of 23 cm. A glass screen was placed at the bottom of the column to support the filter materials. The feed water percolated through the glass columns in down-flow mode (from top to bottom) and the flow-rate was adjusted to about 6.5 mL/min by a peristaltic pump (ECOLINE VC-MS/CA8-6, Ismatec SA, Glattbrugg, Switzerland). As a result, the EBCT was maintained at 11 min. The experiments were performed in duplicate from Monday to Friday. During weekends, the filter materials were kept saturated without flow. Operation of all columns was terminated when the removal efficiencies of all metals were less than 20%. However, for the two tests representing the best performing removal efficiencies, this operational criterion was not achieved during a four-week testing period.

One influent sample per stock solution and several effluent samples at designated times were collected in 250 mL LDPE bottles. Filtration of samples was not conducted. Selected experiments were done with changes in initial concentrations and contact time.

2.2.3. Batch Experiments According to Standard Methods

Further batch experiments were performed according to two standard protocols [32,33]. Both procedures were often used in the field of stormwater treatment to determine sorption capacities of heavy metals although they do not use heavy metals as adsorbates, but other cations as substitutes.

The cation exchange capacity of the first method (CEC 1) is determined by shaking 2.50 g of filter material with 30 mL of barium chloride solution three times each for 60 min to extract sodium, potassium, calcium, and magnesium [32]. After each step, the solution is separated from the filter material by centrifugation and subsequently rejected. The filter material is then shaken again with a fresh barium chloride solution to preload the material with barium for 17 h. After centrifugation, 30 mL of magnesium sulfate solution is added to the filter material to exchange the sorbed barium ions with magnesium under the precipitation of barium sulfate. After 24 h of shaking, the decrease of dissolved magnesium is determined in filtrated samples of the last step. The result of the magnesium sorption is given in cmol^+/kg and this value is often proposed to be valid for other metal cations. This standard protocol was developed for soils but is also used for technical filter materials. All experiments were done in quadruplicate.

The cation exchange capacity of the second method (CEC 2) is determined by preparing a sample of the filter material (10 g) with sodium chloride and then adding the sorbent to a 300 mL solution of ammonium chloride [33]. This cation exchange capacity is calculated by the decrease of ammonium in a filtrated sample after 2 h of shaking. It is assumed that the exchange capacity related to ammonium is comparable to the total cation exchange capacity. This standard was developed for natural zeolites. All experiments were carried out in triplicate.

2.3. Analytical Procedures

The same analytical procedures and equipment were used during all experiments to minimize the influences of sample preparation and analysis on the results.

The concentrations of heavy metals were measured by flame atomic absorption spectrometry (AAS, Varian Spectrometer AA-240FS, Palo Alto, CA, USA) according to Standard Method 3111 [34] with detection limits of 50 μg/L (Cu), 50 μg/L (Ni), 20 μg/L (Zn), 2500 μg/L (aluminum), 100 μg/L (calcium), 100 μg/L (iron), 50 μg/L (manganese), and 50 μg/L (magnesium). Some samples were also measured with a graphite furnace AAS (Varian Spectrometer AA-240Z with GTA 120, Palo Alto, CA, USA) according to Standard Method 3113 [34] with detection limits of 5 μg/L (Cu) and 5 μg/L (Ni). Total metal concentrations were measured with AAS in an *aqua regia* digestion of the samples to quantify their contents in the filter materials. Quality control was maintained by using control and blank samples, and taking standard solution quantity checks of every five samples.

Ammonium was measured using cuvette tests (Hach Lange GmbH, Düsseldorf, Germany). Total carbon (TC) and total sulfur (TS) were determined with a multi EA 4000 element analyzer (Analytik Jena AG, Jena, Germany). The solid samples were directly decomposed and oxidized at 1350 °C and the resulting oxides were quantified simultaneously with a non-dispersive infrared (NDIR) detector. Specific surface areas of the sorbents were determined according to ISO standard 9277 [35] using a SA 3100 plus (Beckman Coulter, Brea, CA, USA) with nitrogen as adsorption gas (BET method).

For all samples, the pH value and electric conductivity were measured immediately after sample collection. Measurements of the pH value were performed with a glass electrode (WTW Sentix 60, Weilheim, Germany) according to Standard Method 4500-H^+ and electric conductivity (detection limit: 0.1 μS/cm) was determined by using a WTW ProfiLine Cond 1970i (WTW, Weilheim, Germany) according to Standard Method 2520 B [34]. Subsequently, all samples were conserved with nitric acid at pH < 2. Sorbent samples were weighed using an analytical balance (Sartorius MC1 Research RC 210D, Göttingen, Germany) with an accuracy of ±0.1 mg.

2.4. Data Evaluation

Heavy metal capacities, representing their contents on the sorbent, were calculated for each experiment and each sorbent. This was done by using the analyses of the corresponding metal concentrations before and after the experiments with Equation (1):

$$q_e = (c_0 - c)/X, \tag{1}$$

where q_e (mg/g) is the sorbed amount of adsorbate on the filter material, c_0 (mg/L) is the initial metal concentration, c (mg/L) is the equilibrium or averaged effluent metal concentration of adsorbate in solution, and X is the filter material dosage (g/L).

The Langmuir isotherm model (a non-linear isotherm with two-fitting parameters) was used for the calculation of q_m (2):

$$q_e = q_m K_l c/(1 + K_l c), \tag{2}$$

where q_m (mg/g) is the saturated amount of adsorbate on the filter material, and K_l (L/mg) is the Langmuir parameter representing the consumed energy in the sorption process.

For the column experiments, breakthroughs in this study were defined at $c/c_0 = 0.1$ and $c/c_0 = 0.5$. The corresponding capacities were $q_{10\%}$ and $q_{50\%}$, respectively.

3. Results and Discussion

3.1. Batch Experiments

Comparing the removal of heavy metals by all investigated materials, anthracite had the lowest capacities because it was not pretreated and it had the smallest specific surface area (1.4 m^2/g). Anthracite was the only sorbent with metal leaching in the control batches without any heavy metals being present in the stock solution. The leached concentrations observed after shaking were 13.5 μg/L Cu, 41.4 μg/L Ni, and 30.3 μg/L Zn.

For all other equilibrium experiments, Cu showed the highest affinity for all sorbents. For the highest concentrations, smaller quantities of Ni and Zn were retained. Almost no removal of Ni was observed at initial concentrations of 540 mg/L Ni (pH 5, no buffer) for alumina or calcium carbonate. Zn was not removed by GAC or calcium carbonate at an initial concentration of 1080 mg/L Zn under competitive conditions (*i.e.*, in the presence of other metals). At the same concentrations and pH values, the removal mechanisms including sorption, ion exchange, and precipitation preferentially removed Cu from the solutions. With increasing metal concentrations, the equilibrium pH value decreased for all sorbents. This trend was also reported by Wu and Zhou [24] for GFH. During the shaking, the shift of the pH value of the solution is linked to the presence of the filter material, the removal of metals from the solution, and the exposure to limited amount of atmosphere in the bottle. In the case of poor removal (e.g., by anthracite in this study), the difference between the concentration measurements of the stock solution and after shaking is very small, which might lead to small calculated capacities because of the analytical accuracy (Figure 1).

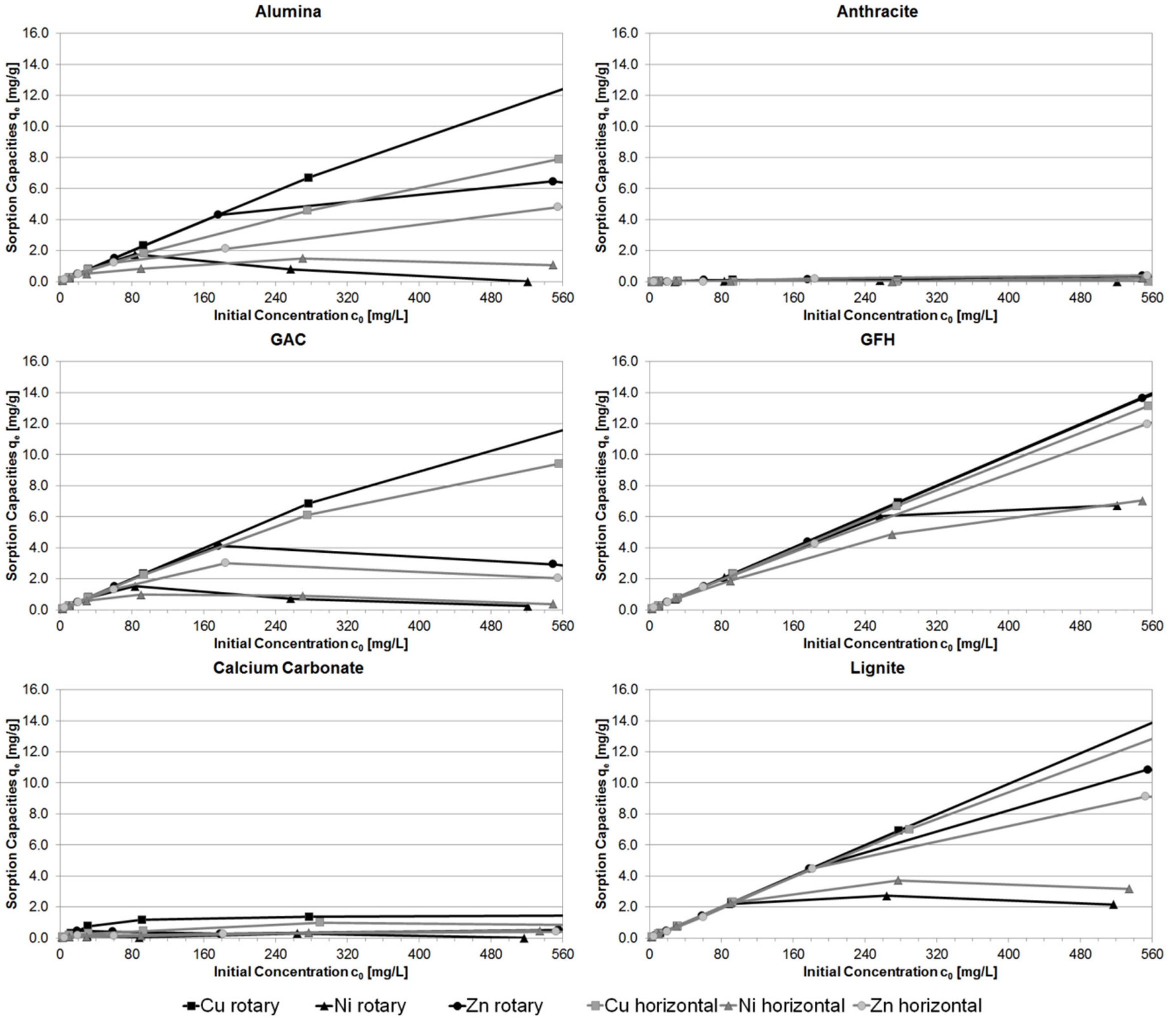

Figure 1. Influences of a rotary shaker (16 rpm) and a horizontal shaker (125 rpm) on sorption capacities at pH 5 without a buffer.

The effect of different types of shakers on the removal capacities was determined for the equilibrium experiments. The speed of each shaker was adapted in order to ensure that the sorbents were in suspension. The same substrate dosage and stock solutions (*i.e.*, initial heavy metal concentrations, pH 5, and no buffer) were used. The capacities determined in the experiments with the rotary shaker were often higher (Figure 1). These differences are likely because of material abrasion and dispersion during shaking [36], which subsequently led to larger surface areas. This effect depends on the type of material and, as a consequence, different behaviors between the sorbents occurred. The absolute differences were low for anthracite and high for alumina. For Cu, the capacities of all of the substrates, determined using the rotary shaker, were between 98% and 268% of the ones determined using the horizontal shaker. At the lowest concentrations, when nearly all of the Cu ions were removed, the difference was very small for all sorbents (factor 1.05) but increased for the highest concentration (factor 1.37). The higher capacities of Ni determined using the horizontal shaker at increasing concentrations can be explained by minor retention of Cu and Zn compared with the rotary shaker on the material surface. Therefore, the competition was lower and Ni ions had more opportunities to sorb onto the material.

Quevauviller *et al.* [37] investigated the influence of shaking types and speed on the determination of extractable trace metals in sediments. They reported that lower amounts of metals were extracted by experiments with horizontal shakers at a speed of less than 40 rpm compared with speeds of up to 150 rpm. In experiments with rotary shakers operated at 30 rpm, 20% more Cu was extracted, compared with experiments using a horizontal shaker at 130 rpm. This effect was less pronounced for Ni than for Cu [37].

Due to the small amount of materials used in batch experiments (8.0 g) an extensive disintegration of the material was possible during rotary shaking intervals of 24 h. This was observed during the experiments with calcium carbonate. Abrasion led to significant experimental difficulty and representative analyses were hardly possible, which was also observed by Liu *et al.* [25] for manganese oxide-coated polymeric media. Therefore, it is essential to record the type of shaker and its speed to assure reproducible and reliable results.

The initial pH value has an influence on the sorption capacities. For example, Smith [31] investigated the Cd removal by a recycled iron sorbent and determined a capacity increase as the pH increased within the range of 4–7. Liu *et al.* [25] calculated the equilibrium capacities for Pb removal that was observed at pH 5, 6, and 7. Between pH 5 and 6 the capacity increased slightly, whereas it was several times higher for pH 7 compared with pH 6. Reed *et al.* [38] also reported higher Pb removal by GAC for elevated pH values. Athanasiadis *et al.* [30] measured higher Zn uptakes of a zeolite with an increasing initial pH value. For example, at higher pH values more metals are precipitated [24], the pH value is at or above the point of zero charge that leads to negatively charged sorbent surfaces and higher retention of cations [23], and sorption also increases because metal ions exchange hydrogen ions of the sorbent surfaces more likely [39]. Thus, higher capacities for all six sorbents were expected for an initial pH value of 7 instead of 5.

The results of the experiments with the variation of the initial pH value are presented in Figure 2. A buffer was used for all curves and the results without a buffer are shown as single points for comparison. For the best materials (alumina and GFH), nearly no differences in the removal of heavy metals occurred with and without buffer. Anthracite, which showed nearly no removal for experiments without a buffer, had removal capacities for all heavy metals for the experiments with a pH buffer at pH 5 and all capacities increased for the experiments at pH 7. The behavior of calcium carbonate was similar. For GAC, differences occurred only for the removal of Ni that was better removed at pH 7 instead of pH 5. However, the use of a buffer had a negative effect on the Ni capacities compared with experiments without buffer addition. Lignite that has larger amounts of primarily oxygen-containing functional groups (e.g., carboxylic, phenolic, and carbonyl groups) than GAC [40–42] also showed differences in the Ni capacities. At pH 5 without a buffer, Ni was completely removed. For the experiments with a buffer, Ni was not completely removed and the removal increased with increasing pH value. This might be related to the functional groups and to a preferred removal of the cations present in the buffer (*i.e.*, potassium and sodium) compared with Ni. In consequence, an increase of the initial pH value led to higher removal capacities whereas the addition of a buffer showed different results.

The difference of the removal capacities for the used buffer and the pH values correlated with the filtration step at the end of the equilibrium experiments. Some heavy metals can be associated with colloidal sorbent compounds and can pass through the 0.45 µm-filter and are subsequently analyzed [31]. In contrast, larger colloids and particulate heavy metals are retained by the filter. The greatest difference between total and filtrated heavy metal concentrations was observed for a stock solution with a buffer at an initial pH of 7. This difference in initial and equilibrium concentrations could be related to a capacity that is equal for all experiments at pH 7 (Figure 3). A comparison of this capacity (equilibrium experiment with no sorbent) with the total capacities of alumina and anthracite (equilibrium experiments with sorbents) illustrated that all heavy metals were partially removed by the filtration process. Thus, the capacities of anthracite at pH 7 and in the presence of a pH buffer (Figure 2) were not related to the sorbent but mainly to the experimental setup. Similar results were obtained by Liu *et al.* [25]. They investigated the removal of Pb during equilibrium batch experiments for pH values between 5 and 11. At pH values > 6, Pb was not only adsorbed onto the sorbent surface but also precipitated. Thus, 89% of Pb was removed at pH 7 by precipitation in the presence of the sorbent, and 62% of Pb was precipitated in the solution without sorbent (same experimental setup) and, therefore, retained during the filtration process. Results consistent with this study were also reported by Genç-Fuhrman *et al.* [23] and Wu and Zhou [24] at an initial pH of 6.5.

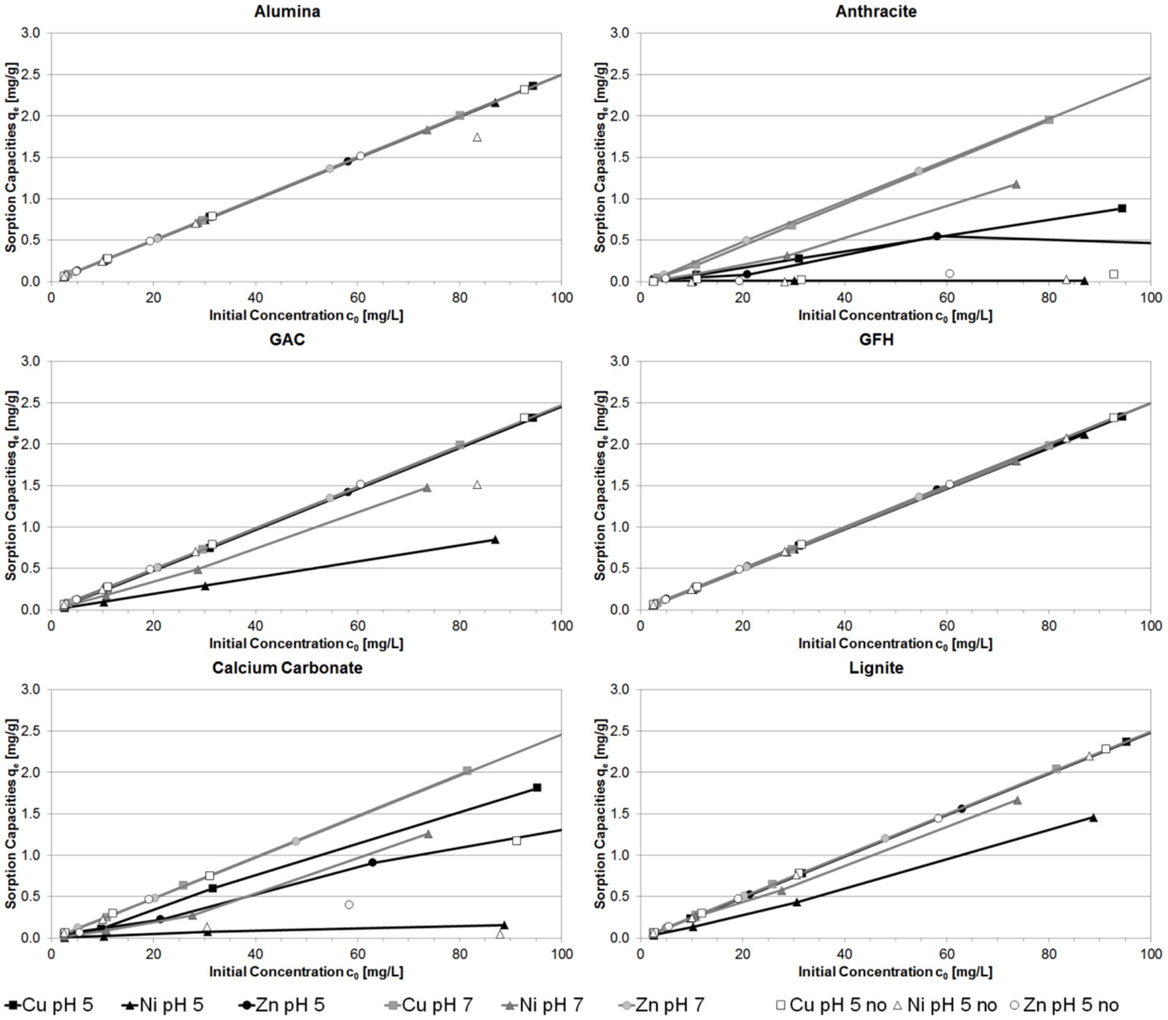

Figure 2. Influences of a buffer at pH 5 and pH 7 for the rotary shaker. For comparison, pH 5 without a buffer (metal pH 5 no) is also presented.

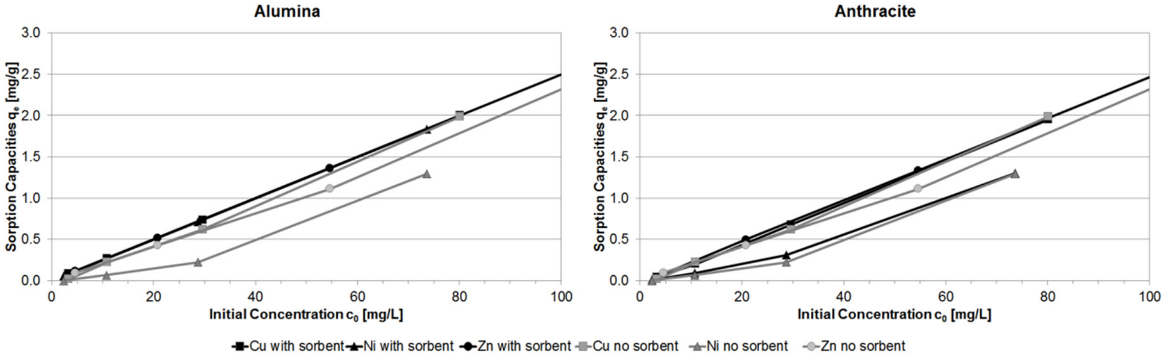

Figure 3. Influence of the filtration process at pH 7 for experiments using a buffer and a rotary shaker: heavy metal removal for different initial concentrations for two sorbents and for the same initial concentrations without any sorbent (metal no sorbent).

The addition of a buffer and other chemicals to adjust the ionic strength, led to an oversaturation of the heavy metal minerals and salts in the synthetic solutions. Thus, higher metal concentrations tended to precipitate with compounds of the added chemicals and not with the constituents of the sorbents. They were also retained during the filtration process and contributed to the overall removal. Consequently, the removal of dissolved heavy metals by sorbents for stormwater treatment systems should be determined at pH 5 without the addition of a buffer.

Batch kinetic experiments can be used to evaluate capacities for the design of treatment systems. In stormwater treatment systems, the contact time between the influent and the filter material is in the order of several minutes and varies widely because rain intensities can be very heterogeneous and the retention volumes are very small in decentralized systems. These short EBCTs can lead to an incomplete reaction of the heavy metals with the sorbents. Therefore, the kinetics of the heavy metal removal is very important to characterize the suitability of a material for this application. Since the removal mechanisms in porous media are controlled by physical-chemical processes or by chemical-dominant surface reactions (*i.e.*, adsorption, desorption, surface complexation, ion exchange, and precipitation), the capacities after several minutes or hours depend on the filter material and the substances of interest as some processes are rate limited (e.g., intraparticle transport and dissolution of carbonates) [31,43,44].

For all kinetic experiments, a higher initial concentration correlated with less heavy metal removal efficiencies at the same time, and the pH values increased with time for all sorbents except anthracite because of the sorbent specific removal mechanisms (e.g., dissolution of the sorbent minerals or release of phenols) [23]. Following an initial fast reaction stage, a slower stage continued for the four sorbents having the best removal capacities (Figure 4).

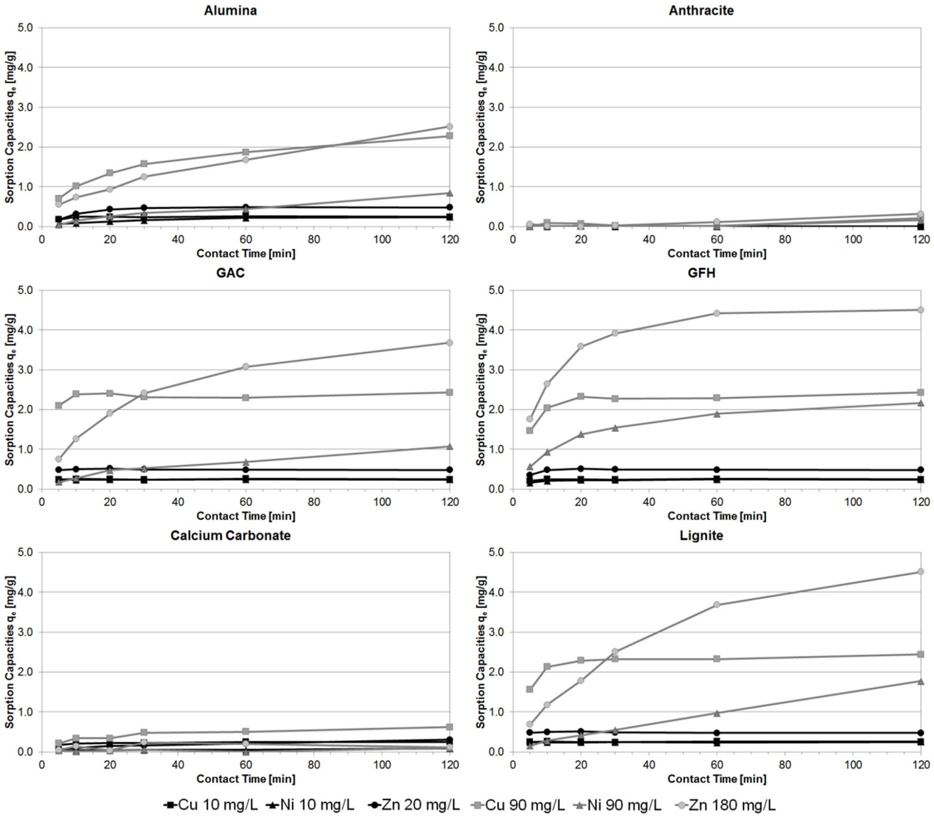

Figure 4. Results of the kinetic experiments with two initial concentrations (rotary shaker, pH 5, and no buffer).

Sorption rates were fast for GAC with nearly 100% of the equilibrium capacities within 5 min for all metals at lower concentrations and Cu at higher concentrations, where the removal of Zn was slower and the removal capacity of Ni was only 71% after 2 h compared with the result of the equilibrium experiment (Zn 89%, respectively). Lignite had a similar behavior but had slower efficiencies compared with GAC for the higher concentrations until 20–30 min. The initial fast rates can be explained by the large surface areas (*cf.* Table 1) and the following reactions were rate-limited by diffusion through the micropores of the sorbent because easily available surface areas were occupied by previously-sorbed metals [39]. Similar results were calculated for GFH with slower rates in the first 5 min for the lower concentrations and a subsequent faster increase in removal efficiencies. For higher concentrations, GFH had the highest removal rates compared with all other sorbents. For alumina, the capacities increased slowly, especially for Ni. Calcium carbonate exhibited slow increases in the removal capacities over time (rate-limited dissolution) with varying results likely because of the abrasion of the material. Therefore, in the range of 5–20 min, which is relevant for stormwater treatment systems, differences occurred between all sorbents for all heavy metals. However, batch kinetic experiments are not suitable to evaluate contact times shorter than 5 min. Thus, these conditions representative of high rain intensities should be simulated using column experiments.

The effect of different temperatures was not evaluated as lab-experiments were always performed at ambient temperatures. Changes in the behavior of heavy metals because of temperature effects in labs were not considered significant according to Quevauviller *et al.* [37] and Pehlivan and Arslan [39].

3.2. Column Experiments

Column studies were carried out with appropriate solid-to-solution-ratios [36] and filtration was not conducted when preparing the samples. The differing behavior of the sorbents during the batch kinetic experiments were reflected in the performance of the filter materials in the lab-scale column tests. One of the aims of these column studies was to achieve maximum capacities of a sorbent under specific boundary conditions. These capacities are needed to design full-scale systems for stormwater treatment (e.g., amount of sorbent and service life) [45]. The design of a full-scale system requires the prediction of the concentration-time profile or the breakthrough curve using lab-scale experiments. The breakthrough curves had a very high coincidence with the pH drift curve for all sorbents (Figure 5). In general, the pH drop correlated with decreasing heavy metal removal for the experiments with an EBCT of 11 min. This pH effect can be used in monitoring the removal process.

The best material (GFH) had a breakthrough of 10% at *ca.* > 2200, 1300, and 1470 bed volumes for Cu, Ni, and Zn, respectively. The capacities of the column experiments were also comparable to the ones of the batch experiments (Table 2). No differences occurred between alumina and GFH in the equilibrium batch experiments for concentrations <100 mg/L and there were only small differences in the kinetic studies for lower concentrations. For the column experiments, the differences were more evident. A 10%(50%)-breakthrough of Ni, Zn, and Cu was observed after treatment of approximately 70(240), 190(310), and 380(950) bed volumes by alumina, respectively. The high efficiency of GFH compared with alumina cannot be linked exclusively to the specific surface area [24] and the medium pore diameter because they are both similar. However, GFH has a 4.5 times larger pore volume (0.717 cm^3/g) that supports heavy metal pore diffusion, surface interaction, surface complexation with hydroxide groups, and precipitation with calcium carbonate (*cf.* Table 1). Furthermore, a regeneration of the filter material during weekends was observed (Figure 5).

Although GAC had the largest specific surface area (896 m^2/g), it had very low capacities in the column experiments and the steepest gradient from breakthrough to exhaustion. The simultaneous steep decrease of the pH value can be linked to the formation of hydroxide complexes of Zn and Ni after the breakthrough point, when the surface area is limited for sorption of metal ions. These low capacities of GAC limits its applicability for stormwater treatment systems as it was also reported by Liu *et al.* [19] and Li and Helmreich [28]. Both GAC and lignite had Zn and Ni values above 1.0 for c/c_0 as a result of competition because Cu is preferentially retained. As sorption is one of the key removal mechanisms, a partial displacement of previously sorbed Ni and Zn occurred as the remaining capacity decreased and led to effluent concentrations that were higher than the influent concentrations. A higher selectivity of lignite for Cu removal (followed by Zn and Ni) was also measured by Jochová *et al.* [44].

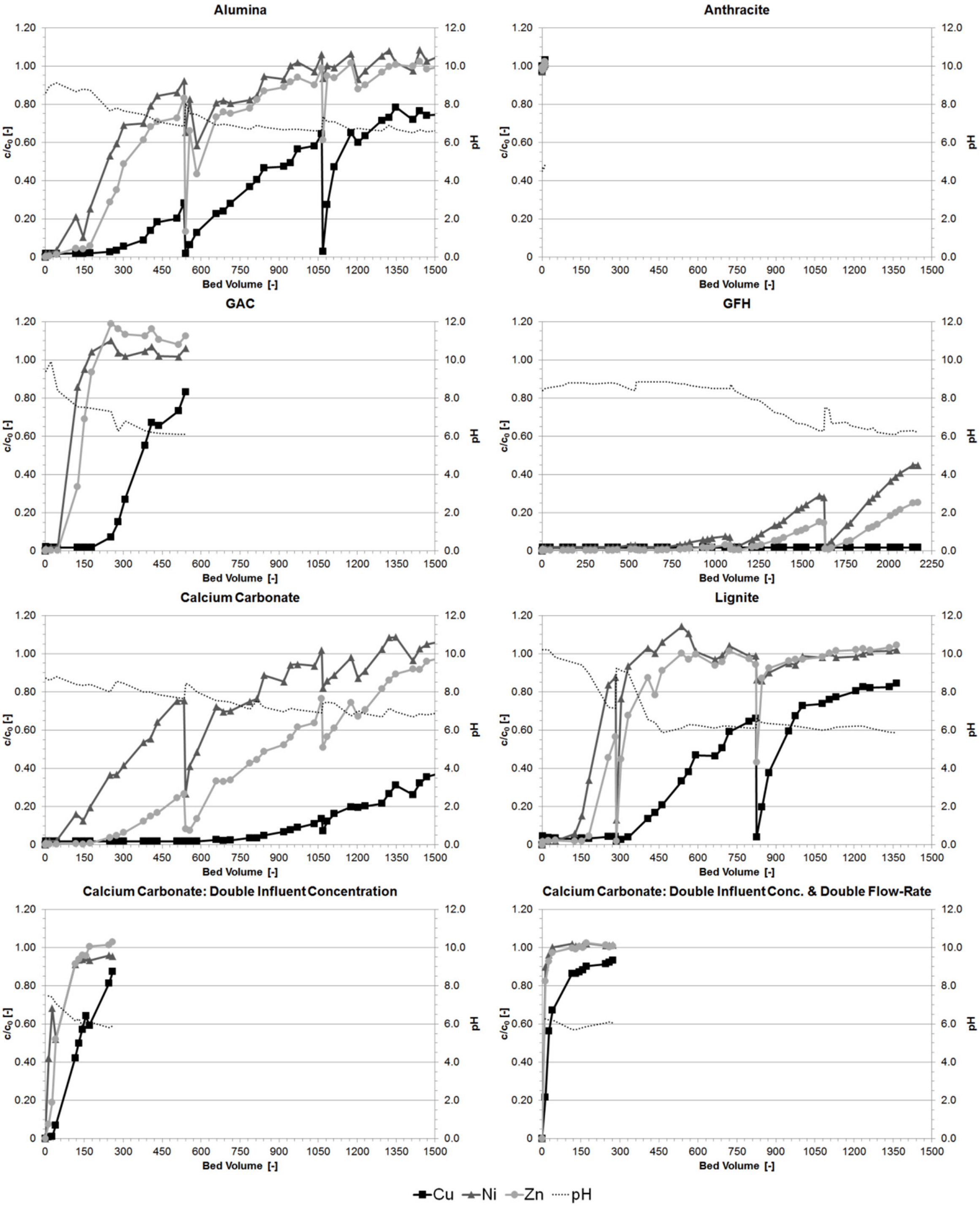

Figure 5. Heavy metal removal and pH drift curves of all column experiments conducted with influent concentrations of 2.5 mg/L Cu, 2.5 mg/L Ni, and 5.0 mg/L Zn at a flow-rate of 6.5 mL/min (exceptions: two calcium carbonate experiments with variations; GFH has a different scale).

Anthracite had an exhaustion of the capacity before the first sampling (Figure 5).

In contrast to the batch experiments, calcium carbonate had higher capacities in column experiments (Table 2). Therefore, the batch experiment setup had a negative effect while determining sorption capacities because of the abrasion and disintegration of the material. Despite its medium Ni and Zn removal efficiencies, calcium carbonate showed a high Cu removal because Cu is predominantly precipitated as copper carbonate at the prevailing pH values (*cf.* Figure 5) [23].

Table 2. Comparison of different sorption capacities; batch experiments: highest values measured (q_e) for two initial concentrations (mg/L) at pH 5 with the rotary shaker without a buffer and calculated using the Langmuir isotherm model (q_m); column experiments: at 10% breakthrough point ($q_{10\%}$) and at 50% breakthrough point ($q_{50\%}$)—all values in mg/g.

Filter Material	**Cu**					**Ni**					**Zn**				
	$q_{e,2.5}$	$q_{e,540}$	q_m	$q_{10\%}$	$q_{50\%}$	$q_{e,2.5}$	$q_{e,540}$	q_m	$q_{10\%}$	$q_{50\%}$	$q_{e,2.5}$	$q_{e,540}$	q_m	$q_{10\%}$	$q_{50\%}$
Alumina	0.06	12.7	13.5	1.46	3.07	0.06	1.75	1.77	0.25	0.71	0.12	6.45	6.47	1.35	2.01
Anthracite	0	0.09	-	-	-	0	0.06	-	-	-	0	0.36	-	-	-
Calcium carbonate	0.06	1.45	1.45	1.60	>2.27	0.06	0.30	0.31	0.13	0.42	0.12	0.53	0.53	1.06	2.25
GAC	0.06	11.9	12.0	1.40	1.81	0.06	1.51	1.52	0.28	0.41	0.12	4.11	4.44	0.71	1.22
GFH	0.06	14.4	21.8	>8.92	>8.92	0.06	6.73	6.82	4.90	>7.57	0.12	24.0	23.5	11.1	>11.3
Lignite	0.06	14.6	16.6	1.27	2.00	0.06	2.73	2.73	0.44	0.60	0.12	10.9	11.2	1.26	1.62

Selected experiments were performed with calcium carbonate because of its rate-limiting removal mechanism to evaluate the influence of changes in initial concentrations and contact time. These are the main factors varying between different rain events treated in decentralized systems. For decreasing EBCT, capacities and efficiently treated bed volumes decreased (Figure 5). The $q_{50\%}$-capacity decreased for Zn from 2.25 mg/g to 0.42 mg/g (double influent concentration) and to 0.14 mg/g (double influent concentration and double flow-rate). These results indicated that the impact of the EBCT for calcium carbonate was comparable to the ones for GAC and manganese oxide-coated media [19,38]. Inglezakis *et al.* [43] also determined for a zeolite that the breakthrough point (set at $c/c_0 = 10\%$) could be observed after a smaller amount of treated bed volumes as the flow-rate increased and the EBCT decreased. This effect also occurred in the experiments of this study (Figure 5) and it influenced the capacities negatively because the steepness of the breakthrough curve also increased. In contrast to the manganese oxide-coated media tested by Liu *et al.* [19], the breakthrough capacities of calcium carbonate decreased with increasing concentrations for the same EBCT. For the same removal efficiencies, a longer EBCT must be maintained for higher influent concentrations [38]. Therefore, for the same EBCT and higher influent concentrations, the removal efficiencies decreased and the steepness of the breakthrough curve increased (Figure 5). All breakthrough curves can be used to evaluate service lives for specific removal efficiencies under defined boundary conditions of the column experiments. These individual breakthroughs can be linked to meeting regulatory requirements.

3.3. Comparison of Batch and Column Experiment Capacities

A comparison of the capacities determined for the three heavy metals is summarized in Table 2. These removal capacities are important because they are needed to design full-scale systems for stormwater treatment (e.g., type and amount of sorbent, ratio of drainage area to filter area, or service life) [45]. For realistic stormwater runoff concentrations (2.5 mg/L and 5.0 mg/L) in batch experiments, all filter materials exhibited approximately 100% removal (with the exception of anthracite) and, therefore, achieved the same capacities (0.06 mg/g for Cu and Ni, 0.12 mg/g for Zn). For the highest initial concentrations of the batch experiments (540 mg/L and 1080 mg/L) the sorbents can be differentiated by the capacities. However, aspects such as a decrease in the equilibrium pH value because of the high metal concentrations make it difficult to evaluate the results as some metals were not removed. These capacities are comparable with the ones calculated using the Langmuir isotherm model.

In column experiments, the sorbents were evaluated by realistic concentrations under an EBCT of 11 min, which represents lower rain intensities. The capacities were much lower than the maximal sorption capacities calculated from the batch experiments, with the exception of GFH and calcium carbonate. GFH was the best material in the lab-scale column tests and calcium carbonate was disintegrated during the batch experiments. These differences were observed because of the reaction kinetics and the experimental setup. For a calcareous soil, a comparison of the retention of heavy metals in batch and column experiments showed similar results [46]. A comparison of the batch ($q_{e,2.5}$ or $q_{e,5.0}$) and column capacities ($q_{10\%}$ and $q_{50\%}$) using the same stock solution concentrations showed that larger amounts are sorbed on the column materials because of the higher solid-to-solution-ratios [36].

Lab-scale column experiments are more representative than batch experiments because of a constant flow, higher solid-to-solution-ratios, and the missing of the filtration step, which makes it possible to compare the efficiencies of filter materials and the effluent water quality [47]. A variation of the flow-rate to evaluate varying rain intensities can be achieved without high efforts during one experimental run by considering the site-specific hydrology and the flow-rate that is relevant for the filter system. Column experiments are applicable to simulate the behavior of stormwater filter materials for high rain intensities (contact times < 5 min) and, therefore, kinetic effects are better considered compared with batch kinetic experiments. In addition, there is no abrasion of filter materials that could influence the results. Thus, column experiments give a more realistic capacity by considering real boundary conditions and should be preferred to determine efficiencies and service lives. The determined service life is only valid for the dissolved substances and further field experiments are needed to verify the performance in the presence of particulate matter that can cause clogging. Batch experiments can be used to exclude sorbents that are not appropriate for the removal of a specific metal from subsequent and more expensive column experiments and to evaluate different production batches (*cf.* Section 3.6). To date, a lack of standardized methods does not allow a comparison of various results of one sorbent used in different research programs [37].

3.4. Comparison of Different Batch Experiment Capacities

In addition to the batch experiments with heavy metals, further batch experiments were conducted with surrogates according to standard methods to evaluate their ability to determine the capacities of different sorbents. Both methods use other cations (magnesium and ammonium) that replace previously exchanged cations (barium and sodium, respectively). Due to the exchange of cations being a selective process, the use of surrogates assumes that their affinity for being removed is comparable with the cations of interest. This is especially critical for the method CEC 2 because it was developed for natural zeolites and not for filter materials in general. It must be noted that the method CEC 1 is not suitable for materials containing significant amounts of calcite or gypsum [32]. This applies to the sorbent calcium carbonate.

The results of both methods that are described in Section 2.2.3 are shown in comparison with sorption capacities of Cu, Ni, and Zn in Table 3. All capacities are presented in the unit cmol^+/kg to enable a comparison as it is suggested by the two standard methods. By comparing the absolute numbers of the sorption capacities with the results of CEC 1, differences between each sorbent are apparent. GFH and lignite have comparable results regarding the sorption capacities for all three heavy metals, although they have different CEC 1 values. Therefore, different removal mechanisms are responsible for the retention of heavy metals. For alumina, CEC 1 values and the sum of the heavy metal sorption capacities are comparable. However, the individual heavy metal capacities indicate that Cu is completely retained, Zn less, and Ni only up to 67%. Thus, the method CEC 1 does not consider the diverse behavior of the heavy metals and overestimates the capacity for more mobile metals such as Ni. By using the method CEC 2, a capacity was calculated for anthracite that does not retain any heavy metals (*cf.* Figure 1). Furthermore, a ranking of the capacities according to CEC 2 is not comparable with the results of the batch experiments with heavy metals. The use of the method CEC 2 for natural zeolites is also limited because sodium chloride is used to enhance the sorption behavior of natural zeolites by pretreatment [30]. The use of sodium chloride during application of CEC 2 would, therefore, falsify the capacity results for pretreated and untreated zeolites.

Table 3. Comparison of different capacities determined by batch experiments at pH 5 with the rotary shaker and without a buffer, CEC 1, and CEC 2—all values in cmol$^+$/kg.

Filter Material	**Four Sorption Capacities**				**CEC 1**	**CEC 2**
	120 mg/L Cu	**120 mg/L Ni**	**240 mg/L Zn**	**480 mg/L $\sum$(Cu+Ni+Zn)**	**480 mg/L Mg**	**180 mg/L NH_4^+**
Alumina	8.42	5.93	15.0	29.4	28.5	7.33
Anthracite	0	0	0	0	0	5.37
Calcium carbonate	3.00	1.34	0	4.34	7.94	3.45
GAC	8.38	3.39	12.4	24.2	3.99	3.63
GFH	8.44	8.96	17.0	34.4	40.2	10.7
Lignite	8.44	8.93	17.0	34.4	9.39	2.98

3.5. Implementation of Ionic Strength and the pH value of Stormwater Runoff

In lab-scale experiments with sorbents for stormwater treatment systems, chemicals such as NaCl [23,24], $NaClO_4$ [31], and $NaNO_3$ [19,25,39] are used to adjust ionic strength, which shall represent the one measured in traffic area runoff. These chemicals contain further cations and chloride that might have an effect on sorption efficiencies (e.g., the presence of other cations can diminish the available number of sorption sites and chloride can form complexes that enhance the mobility of heavy metals [48]).

The parameter of field measurements that is often quantified and mostly related to ionic strength is the electric conductivity because hardness is only sometimes measured. However, the values vary between 11.3 µS/cm and 52000 µS/cm in the dataset presented in [5] with a median value of 160 µS/cm. Thus, the ionic strength relates to site-specific factors that are mainly influenced by the road surface and road maintenance. It should at least be distinguished between warm and cold climates and between asphalt and concrete surface runoff waters. Hence, a median ionic strength is not representative for several sites or seasons. Winter maintenance practices, such as de-icing salt applications, generate chlorides and associated cations, in particular sodium and calcium, that can have an effect on the removal of heavy metals [49,50]. Thus, the ionic strength previously used in lab experiments does not reflect the amount and composition of the cations and anions of real stormwater runoff. Since each element has a unique effect on the removal, additional compounds to simulate ionic strength should be used carefully.

In summary, the consideration of ionic strength in lab-scale experiments is ambiguous as it varies in reality because of de-icing salt maintenance and other site-specific factors that lead to different compositions of the stormwater runoff, which cannot be reproduced in the lab. Therefore, the use of no buffer or ionic strength might be a worst case scenario for the determination of dissolved heavy metal removal to prevent oversaturated systems. However, for treatment systems in cold climates, de-icing salts must be considered. Additional experiments might be a better solution to observe their mobility and remobilization behavior.

The initial pH value has an effect on the measured capacities (*cf.* Figure 2). The pH values of 6.0, 6.5, and 7.0 [23–25,31] are often used in lab experiments that represent the median value of 6.9 for traffic area runoff waters [5]. A higher initial pH value results in more metals being precipitated because of the experimental setup [25]. This leads to an overestimation of the capacities of sorbents (especially for higher metal concentrations), which are predominantly increased by the filtration step at the end of all batch experiments by which larger colloids and particulate heavy metals are retained (*cf.* Figure 3). This effect does not occur in real stormwater treatment systems because the wire mesh or geotextile that is used to restrain the sorbents has larger pore diameters and these fine heavy metals can pass through them. Therefore, the enhanced precipitation has a great influence on the results, and the comparability between the real situation and the lab experiments is limited. Using realistic influent pH values is not suitable to test the performance of filters for stormwater treatment systems that have to remove dissolved heavy metals. Therefore, to determine the sorbent performance and to investigate the removal mechanisms of decentralized stormwater treatment systems, it is preferred to perform batch experiments with the dissolved heavy metals of interest using synthetic water at pH 5.

3.6. Evaluation of Different Production Batches

A well-defined method for batch experiments can also be used to evaluate different production batches in a fast and inexpensive way. This method must contain information on the initial pH value, the concentrations of the metals of interest, the substrate dosage, the type and speed of the shaker, the shaking time, the temperature, and the sample preparation. The addition of a buffer or ionic strength should be avoided. All experiments should be performed in triplicate and blank samples are recommended.

The evaluation of different production batches of GFH for the removal of Cu is plotted in Figure 6. All samples were filtrated (0.45 μm) after 24 h of shaking and before analysis. The first two batch experiments marked with * contained 0.4 ± 0.05 g material instead of 0.3 ± 0.05 g material in 250 mL solution. A lower filter material dosage had an effect on the results that is comparable to the one of increasing concentrations for the same dosages. A decreased dosage led to fewer variations of the results (Figure 6). Comparing the four batches with the same experimental setup, it can be seen that the results are reproducible for each production batch and there is only a slight variation between different production batches.

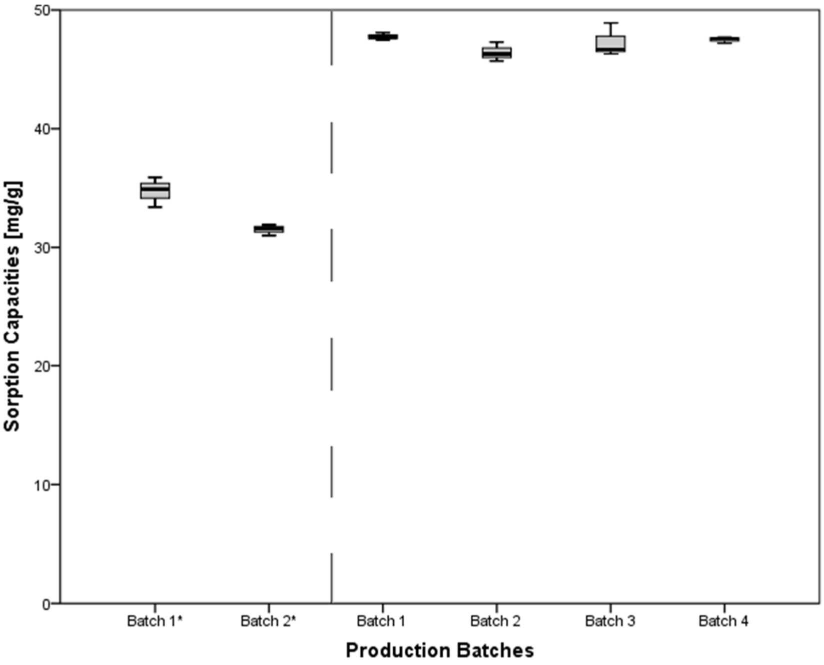

Figure 6. Evaluation of different production batches of GFH by sorption capacities for Cu: batch experiments marked with * contained 0.4 ± 0.05 g sorbent instead of 0.3 ± 0.05 g sorbent in 250 mL solution.

This method can be used to evaluate the performance of the sorbent in comparison with different production batches under identical experimental conditions. Such an evaluation is needed for internal and external quality control, which is conducted several times per year by the manufactures of technical decentralized stormwater treatment systems. This quality control proves that the filter materials of one treatment system always have the same characteristics. Therefore, the performance of different plants should be comparable. However, this method is not suitable to design full-scale treatment systems or to predict the service life because it overestimates the sorbent capacity.

4. Conclusions

For testing sorbents for urban stormwater treatment systems, it is important to assure that reproducible and reliable results were obtained. Hence, the following influencing factors must be considered for the determination of capacities:

- Experiments with the dissolved heavy metals of interest using synthetic water at pH 5 is suggested and the influence of the type and speed of the shaker must be considered.
- Adjustments of ionic strength and pH > 5 are more sensitive and should be avoided because they mostly lead to an overestimation of the removal capacities of sorbents.
- The filtration step at the end of the batch experiments has an effect on the results and does not simulate real situations.
- Batch experiments with other cations (e.g., magnesium and ammonium) should not be used as surrogates.

The results of batch and lab-scale column experiments concerning simultaneous heavy metal removal differed widely for each sorbent. Thus, the following applications are recommended:

- Batch experiments with heavy metals can only be used under identical and well-defined conditions to prove the comparability of different production batches and to select sorbents for subsequent column experiments.
- Column experiments can be used as an indicator to determine the efficiencies and service lives of treatment systems.

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Chapter 7: Assessment Method for the Heavy Metal Removal

7.1 Research Paper: "Development of a Novel Test Method to Determine Service Lives"

Because batch and small-scale column experiments are not suitable to determine service lives and long-term performances of filter materials (cf. Chapter 6), a novel test method was developed during this thesis, which can be used to assess manufactured decentralized stormwater treatment systems for runoff from traffic areas in the laboratory. The corresponding hypothesis was: *A novel test method for determining the filter material service lives and long-term performances can be developed and implemented to evaluate new and already existing decentralized systems* (Hypothesis 4).

This novel test method was developed to determine – within a few days – the service lives and long-term removal efficiencies for dissolved substances in stormwater treatment systems. It consists of two parts: in Part 1, the filter system is rapidly preloaded with heavy metal loads of n-1 years. The variable n represents the estimated service life of the filter material in years that has been determined in previous batch or small-scale column experiments (cf. Chapter 6). In Part 2, the long-term performance of the system is assessed by simulating the last year of the filter material service life. For all experiments, Cu and Zn are used and the filter systems are represented in individual pilot-scale models. To calculate the amount of Cu and Zn for each test, the connected catchment area and a filter model factor are considered. The experimental results of one FCS are presented. In addition, the heavy metal contents of Cu and Zn as a function of depth (0–10 cm) were measured for this system. These contents were compared with the field measurements of a full-scale treatment plant, which were available for the metals Cd, Cr, Cu, Ni, Pb, and Zn after 2 years, 5 years, and 9 years of operation (service life: 10 years). However, further pilot-scale experiments in the laboratory and field measurements were required to show the applicability of the novel method (cf. Section 7.2). In addition, the influence of sodium chloride and calcium chloride on the remobilization of Cu and Zn was evaluated at the end of the service life for the FCS. More results concerning the heavy metal remobilization were presented in Huber et al. (2014a) for five filter systems. In some cases, both heavy metals were released and in other cases, only Zn showed higher mobility. The pH of the effluent was often reduced by the de-icing salts. For a few materials, Cu and Zn showed nearly no increased mobility during all de-icing salt experiments. Thus, a more standardized method must be developed to assess the remobilization of five heavy metals by three de-icing salts (cf. Chapter 9).

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Ein neues Laborverfahren zur Ermittlung von Standzeiten dezentraler Anlagen zur Behandlung von Verkehrsflächenabflüssen

Abwasserbehandlung, Verkehrsflächenabfluss, Niederschlagswasserbehandlungsanlage, dezentral, Standzeit, Schwermetalle, Substrat

Maximilian Huber, Antje Welker, Martina Dierschke, Jörg E. Drewes und Brigitte Helmreich

Im Rahmen eines vom Bayerischen Landesamt für Umwelt (LfU) beauftragten Forschungsvorhabens wurde ein auf Grundlage der Prüfverfahren des Deutschen Instituts für Bautechnik (DIBt) für dezentrale Niederschlagswasserbehandlungsanlagen basierendes Laborverfahren entwickelt, um realistische Standzeiten und Reinigungsleistungen sowie deren zeitliche Veränderungen in Bezug auf gelöste Schwermetalle zu ermitteln. Die Evaluierung dieses neuen Verfahrens erfolgte anhand von Laboruntersuchungen an kommerziellen Behandlungssystemen für Verkehrsflächenabflüsse, die bereits durch das DIBt zugelassen wurden, und das Ergebnis wird anhand eines Rinnensystems erläutert. Anhand dieses Verfahrens können realistische Standzeiten mit einer einheitlichen Methodik ermittelt werden, wodurch Behörden, Planern und Betreibern ein objektiverer Vergleich der Anlagen ermöglicht wird.

A Novel Laboratory-Scale Test Method to Determine the Service Time of Decentralized Plants Treating Street Surface Runoff

In a project initiated by the Bavarian Environment Agency (Bayerisches Landesamt für Umwelt, LfU) a laboratory-scale test method was developed to determine the service time and efficiencies to remove dissolved heavy metals in decentralized water treatment systems based on standard procedures proposed by the Deutsches Institut für Bautechnik (DIBt). This method was evaluated in the laboratory using commercial treatment systems, which have already national approvals by the DIBt for treating street surface runoff, and the results are presented for one channel system. The novel method developed during this study enables a standardised and more accurate estimation of service intervals of decentralized treatment systems and therefore provides regulatory authorities, designers and operators with a more objective basis for performance assessment.

1. Einleitung

In den letzten Jahren wurden zahlreiche dezentrale Behandlungsanlagen für Verkehrsflächenabflüsse entwickelt, welche eine allgemeine bauaufsichtliche Zulassung nach einer Prüfung durch das Deutsche Institut für Bautechnik (DIBt) erhalten können. Das Kernstück der DIBt-Zulassungsgrundsätze für Filteranlagen ist die Wirksamkeitsprüfung [1]. Dabei werden die Rückhaltevermögen für Abfiltrierbare Stoffe (AFS) sowie für Mineralölkohlenwasserstoffe (MKW) an der jeweiligen Originalanlage geprüft. Der Rückhalt von Schwermetallen und die mögliche Remobilisierung der Schwermetalle durch das Auftausalz Natriumchlorid (NaCl) werden an einem verkleinerten, repräsentativen Filtermodell durchgeführt, um so wenig wie möglich schwermetallhaltiges Abwasser zu produzieren. In den Zulassungsgrundsätzen werden die Prüfanordnung, die Zusammensetzung des Beschickungswassers, die Höhe und Dauer der Regenspenden sowie die Art der Probenahme, der Analytik und der Ergebnisauswertung ausführlich beschrieben. Jedoch ist bei diesem Prüfverfahren keine detaillierte Methodik zur Ermittlung einer realistischen Standzeit der Anlagen vorgesehen. Derzeit werden diese von den Herstellern mittels unterschiedlicher Verfahren (z. B. Bestimmung der Kationenaustauschkapazität oder der Adsorptionskapazität bzw. Säulenversuche) ermittelten und im Zulassungsverfahren vorgeschlagenen Standzeiten der Substrate vom DIBt, ggf. nach einer Korrektur, übernommen.

Daher hat das Bayerische Landesamt für Umwelt (LfU) ein Projekt in Auftrag gegeben, welches von der Technischen Universität München in Kooperation mit der FH Frankfurt am Main bearbeitet wurde. Das Ziel dieses Vorhabens war, die Reinigungsleistungen und realistische Standzeiten sowie deren zeitliche Veränderungen für dezentrale Niederschlagswasserbehandlungsanlagen für Verkehrsflächenabflüsse durch eine reproduzierbare Testmethode zu ermitteln.

Grundsätzlich können bei der Bestimmung der Standzeit zwei Effekte zum Tragen kommen, welche bei der Entwicklung einer Methodik für die Standzeitermittlung bei Verkehrsflächenabflüssen untersucht werden müssen:

a) Die Standzeit ist limitiert durch das hydraulische Betriebsversagen der Anlagen. Häufig wird hierbei eine **Kolmation** beobachtet, welche meist durch erhöhte Feststoffbelastung in Kombination mit kolmationsfördernden Randbedingungen der Örtlichkeit (z. B. mineralische Streustoffe, Baustaub oder organische Belastung aus Pollenflug, Blüten sowie Laub) ausgelöst wird.
b) Die Standzeit ist limitiert durch das stoffliche Betriebsversagen der Anlagen bzw. eine signifikante **Reduktion der stofflichen Rückhalteleistung**. Hierbei ist im Regelfall die Rückhaltekapazität des Filtersubstrats hinsichtlich der **Schwermetalle** erreicht.

Da eine Kolmation (Fall a) wesentlich und nicht vorhersehbar durch die Randbedingungen am Einbauort der Anlagen bestimmt wird (z. B. kurzzeitige und massive Fahrbahnverschmutzungen durch Baustellenverkehr), kann die Kolmation nur in umfassenden und vergleichenden Feldversuchen an verschiedenen Standorten festgestellt werden. Sollte es in der Realität zu einer Kolmation an einem vom DIBt zugelassenen System kommen, wird dem Betreiber der Handlungsbedarf sichtbar aufgezeigt. Da diese Anlagen keinen Notüberlauf besitzen dürfen, erfolgt ein Rückstau des Niederschlagswassers im Falle einer Kolmation auf die Straße, wodurch Handlungszwang besteht.

Beim Überschreiten der Standzeit des Filtersubstrats (Fall b) hingegen (z. B. Filterdurchbruch oder reduzierte Fällwirkung) können unzulässig hohe Schadstoffkonzentrationen, z. B. Schwermetalle, unbemerkt in den Ablauf der Anlage und somit in das Boden-Grundwasser-System gelangen. Dieser Effekt lässt sich unter Laborbedingungen nachstellen und wurde im Rahmen dieses Vorhabens untersucht.

Die am häufigsten bei diesen Behandlungsanlagen auftretenden Wirkungsweisen der Filtersubstrate in Bezug auf den Schadstoffrückhalt werden nachfolgend beschrieben, wobei die ersten beiden Mechanismen abhängig vom pH-Wert sind:

1. Sorption: Der wichtigste Prozess zur Entfernung von gelösten Stoffen ist die Adsorption an festen Oberflächen (Bsp. Aktivkohle). Dabei unterscheidet man zwischen Physisorption und Chemisorption. Bei der Physisorption werden die Stoffe aufgrund von schwachen Van-der-Waals-Kräften an der Materialoberfläche zurückgehalten. Dahingegen resultiert die Chemisorption aus elektrostatischen und kovalenten Wechselwirkungen. Das Prinzip dieser Bindung besteht in der Ausbildung sehr stabiler Einheiten der oberflächennahen, funktionellen Gruppen (z. B. Carboxyl-, Hydroxy- bzw. Phenolgruppen) des Filtersubstrats mit zahlreichen Stoffen im Niederschlagswasser. Ein Beispiel ist der Ionenaustauschprozess, für den auf den Ionenaustauschermaterialoberflächen (Bsp. Zeolith) geladene Ionen wie Natriumionen (Na^+) oder Calciumionen (Ca^{2+}) durch elektrostatische Kräfte an entgegengesetzt geladenen Oberflächenladungen gebunden sein müssen. Die so gebundenen Ionen sind leicht durch andere Ionen gleicher Ladung austauschbar. Beispielsweise werden die auf den Oberflächen befindlichen Na^+ und Ca^{2+} während des Anlagenbetriebs durch Schwermetallkationen wie Zn^{2+} (Zinkionen) oder Cu^{2+} (Kupferionen) unter Berücksichtigung äquivalenter Ladungen unspezifisch ausgetauscht.
2. Fällung: Durch eine Erhöhung des pH-Wertes kann es bei gelösten Schwermetallionen zu einer Ausfällung als entsprechende Hydroxid- oder Carbonatverbindung kommen; die Schwermetallionen werden somit in eine wasserunlösliche Form überführt (Bsp. Carbonatgestein).
3. Filtration: Dabei handelt es sich um ein mechanisch-physikalisches Verfahren, bei dem partikuläre Schadstoffe zurückgehalten werden, indem sie entweder größer als der Porenraum des Filtersubstrats oder kleiner als dieser sind und durch Partikelträgheit, Adhäsion oder Sperreffekte zurückgehalten werden.

Im Folgenden wird das in diesem Vorhaben entwickelte Verfahren zur Bestimmung der Standzeiten und Reinigungsleistungen in Bezug auf gelöste Schwermetalle dargestellt und anhand der Untersuchung eines Substrats aus einer bereits durch das DIBt zugelassenen Anlage erläutert. Die Ergebnisse dieser standardisierten Testmethode können zukünftig Behörden, Planern und Betreibern einen objektiven Vergleich der auf dem Markt verfügbaren Anlagen ermöglichen, da derzeit z. B. der amtliche Sachverständige bei der wasserwirtschaftlichen Beurteilung dieser technischen Behandlungsanlagen im Hinblick auf Reinigungsleistungen und Standzeiten zumeist auf Herstellerangaben angewiesen ist.

2. Auswahl der Untersuchungsparameter

Als Vorgaben zur Bewertung der Leistungen von dezentralen Behandlungsanlagen werden derzeit die Prüfwerte der Bundes-Bodenschutz- und Altlastenverordnung (BBodSchV) vom 12.07.1999 für den Wirkungspfad Boden-Grundwasser zugrunde gelegt [2]. Demnach sind im Ablauf der Behandlungsanlage bspw. Konzentrationen von 50 µg/L Kupfer (Cu) und 500 µg/L Zink (Zn) zu erreichen bzw. zu unterschreiten. Mit der Mantelverordnung

Tabelle 1. Prüfwerte der BBodSchV für den Wirkungspfad Boden-Grundwasser und die Prüfwerte Grundwasser des Entwurfs der Mantel-V vom 31.10.2012 für ausgewählte anorganische Parameter [2, 3].

	Einheit	BBodSchV	Mantel-V
Pb	µg/L	25	7
Cd	µg/L	5	0,25
Cr	µg/L	50	7
Cu	µg/L	50	14
Ni	µg/L	50	20
Zn	µg/L	500	58
Cl	mg/L	-	250

Tabelle 2. Schwerpunktwerte sowie oberer und unterer Wertebereich für ausgewählte Stoffe in Verkehrsflächenabflusskonzentrationen.

	Einheit	Schwerpunktwert	Wertebereich
Pb	µg/L	160	10–300
Cd	µg/L	3,0	0,2–6
Cr	µg/L	10	1–25
Cu	µg/L	85	30–280
Ni	µg/L	20	2–60
Zn	µg/L	450	200–620
Cl	mg/L	7 500*	

* Spitzenwert

(Mantel-V, derzeit Entwurf vom 31.10.2012) [3] sollen unter anderem die Verordnung zum Schutz des Grundwassers vom 09.11.2010 und die derzeit gültige BBodSchV geändert werden. **Tabelle 1** stellt ausgewählte Prüfwerte der BBodSchV und des Entwurfs der Mantel-V gegenüber. Eine Verschärfung der Prüfwerte durch die Einführung der Mantel-V ist derzeit möglich. Dies würde sich direkt auf zukünftige Anforderungen an alle Behandlungstechniken für die anschließende Versickerung auswirken.

Bei der Auswahl der Stoffe für eine Standzeituntersuchung von Behandlungsanlagen von Straßenabflüssen ist neben der Aufkommensrelevanz die praktische Untersuchbarkeit der einzelnen Stoffe wichtig. Zunächst wurde anhand einer Literaturstudie geklärt, welche Stoffparameter in Verkehrsflächenabflüssen aufkommensrelevant und somit in signifikanten Mengen im Straßenabfluss vorhanden sind. Anschließend haben versuchspraktische Erwägungen (z. B. toxische Effekte einzelner Stoffe) die Untersuchung der aufkommens- und wirkungsrelevanten Stoffparameter eingeschränkt.

Die wesentlichen Ergebnisse der Literaturstudie zur Festlegung des Beschickungswassers für die Untersuchungen zum Standzeitverhalten sind im Folgenden dargestellt.

Ausgewählte Verkehrsflächenabflusskonzentrationsbereiche sind in **Tabelle 2** dargestellt.

2.1 Schwermetalle

Die in vielen Behandlungsanlagen eingesetzten Filtersubstrate dienen vornehmlich dem Rückhalt gelöster Stoffe. Für diese Untersuchung sind daher nur in gelöster Form vorliegende Schwermetalle relevant, da davon ausgegangen wird, dass partikuläre Anteile in der jeweiligen mechanischen Behandlungsstufe der Anlagen (z. B. Hydrozyklon oder Sedimentationseinheit) aus dem Niederschlagswasser entfernt werden und somit die Entfernung der ungelösten Anteile beispielsweise mit einer Prüfung auf den Feststoffrückhalt nachgewiesen werden kann. In diesem Zusammenhang wurden häufig vorkommende Schwermetalle auf ihre Relevanz (Aufkommen, Schadwirkung und gelöster Anteil) überprüft.

Chrom (Cr) gilt neben Blei (Pb) als eines der immobilsten Schwermetalle und liegt in der Regel überwiegend partikulär gebunden vor [4, 5]. Außerdem kommt Chrom in Verkehrsflächenabflüssen nur in niedrigen Konzentrationen vor. So wurden beispielsweise in einer LfU-Studie in Augsburg im Mittel 6 µg/L Cr [6] und in einer Schweizer Untersuchung in Burgdorf ca. 10 µg/L Cr [7] gemessen.

Für **Nickel** (Ni) wurden ebenfalls nur geringe Konzentrationen in Verkehrsflächenabflüssen nachgewiesen. Beispielsweise wurden in einer LfU-Studie im Mittel 5 µg/L Ni [6], bei Untersuchungen in Bayreuth ca. 9,9 µg/L Ni und etwa 36 µg/L Ni [8] sowie von der TU München in München im Mittel 35 µg/L Ni [9] gemessen.

Die Aufkommensrelevanz von Chrom und Nickel ist somit gering.

Blei liegt in Verkehrsflächenabflüssen fast vollständig partikulär vor und dessen Konzentrationen nehmen seit Jahren kontinuierlich in den betrachteten Verkehrsabflüssen ab [6, 10, 11, 12]. Aufgrund der Umstellung auf bleifreies Benzin und des Ersatzes der Bleigewichte an Autofelgen wird Blei immer weniger aufkommensrelevant. Blei gilt zudem im Boden als immobilstes Schwermetall [4, 13].

Die Untersuchung des Rückhalts von **Cadmium** (Cd) ist aufgrund der guten Stofflöslichkeit, der Wirkungsrelevanz und der möglichen Wechselwirkung mit anderen Kationen erstrebenswert, kann aber in Laborversuchen aufgrund seiner Toxizität und der Entsorgungsproblematik nicht mit vertretbarem Aufwand abgebildet werden. Zudem liegen die Cadmiumkonzentrationen in Verkehrsflächenabflüssen in neueren Studien bei unter 1 µg/L [6, 11, 14].

Als Bestandteile des Beschickungswassers für diese Versuche zur Standzeituntersuchung werden somit die beiden aufkommensrelevanten Schwermetalle **Kupfer** und **Zink** verwendet. Die in der durchgeführten Literaturstudie bestimmten Schwerpunktkonzentrationen (Gesamtgehalt, **Tabelle 2**), welche die partikulären und gelösten Stoffanteile beinhalten, betragen ca. 85 µg/L Cu und etwa 450 µg/L Zn. Die gelösten Anteile liegen jeweils deutlich darunter. Für diese beiden Stoffe ist zudem im Entwurf der Mantel-V im Vergleich zur BBodSchV eine deutliche Verschärfung der Prüf- bzw. Grenzwerte angesetzt (siehe **Tabelle 1**), die möglicherweise bei zukünftigen Versuchen berücksichtigt werden muss.

2.2 Eisen, Natrium und Calcium

Weitere aufkommensrelevante Kationen im Straßenabfluss sind Eisen, Natrium (Na) und Calcium (Ca). Dabei liegt frisch korrodiertes **Eisen** partikulär vor und kann somit durch Sedimentation entfernt werden [6, 15]. Da einige der vom DIBt zugelassenen und auf dem Markt erhältlichen Substrate eisenhaltig sind und das Eisen bei diesen Anlagen für die stoffliche Rückhalteleistung entscheidend ist, würde die Berücksichtigung von Eisen bei diesen Untersuchungen zu ungleichen Versuchsbedingungen führen, weshalb aus versuchspraktischen Erwägungen darauf verzichtet wurde.

Die Einflüsse von **Natrium** und **Calcium** auf das Standzeitverhalten werden durch die Berücksichtigung von chloridischen Auftausalzen (Straßenwinterdienst) untersucht, wodurch auch der in **Tabelle 2** aufgeführte Spitzenwert von Chlorid (Cl) berücksichtigt wird. NaCl ist das in Deutschland am häufigsten verwendete Auftausalz [16]. Außerdem wird in diesem Vorhaben zusätzlich das zweiwertige Calcium neben dem einwertigen Natrium abgebildet. Die Berücksichtigung von Calcium bildet nicht nur einen möglichen Anteil im auf den Straßen ausgebrachten Feuchtsalz (Calciumchlorid ($CaCl_2$)) ab, sondern auch den Abrieb der Fahrbahnoberfläche, welcher zusammen mit den örtlichen Gegebenheiten den Calciumeintrag in situ darstellt.

3. Darstellung des neu entwickelten Laborverfahrens

3.1 Versuchsaufbau

Bei den Standzeitversuchen werden die zu untersuchenden Rinnensysteme mit Originalbreite und einer Baulänge von 0,50 m bzw. 1,00 m im Versuchsmodell verwendet. Die Einfüllhöhen, die Substratmengen sowie die Einbringverfahren entsprechen den Angaben der Einbauanleitungen der Hersteller. Die Rinnen selbst werden mithilfe einer Beregnungsanlage über 625 Edelstahlkanülen (Durchmesser 0,90 mm) beschickt, die an einer quadratischen, geschlossenen Acrylwanne mit einer Kantenlänge von 1 m oberhalb der Herstellersysteme befestigt sind. In der Wanne befindet sich ein unter Druck stehendes Wasserreservoir, welches mittels einer Pumpe über den Vorlagebehälter gespeist wird. Die Beregnungsintensität wird über den Durchfluss der Pumpe sowie den Wasserdruck im Wasserreservoir gesteuert, über das Schwebekörper-Durchflussmessgerät bestimmt und über die Waage kontrolliert **(Bild 1)**. Bei diesem Versuchsaufbau werden die Rinnen sowohl direkt von oben als auch über Edelstahlbleche seitlich mit dem Beschickungswasser beaufschlagt **(Bild 2)**.

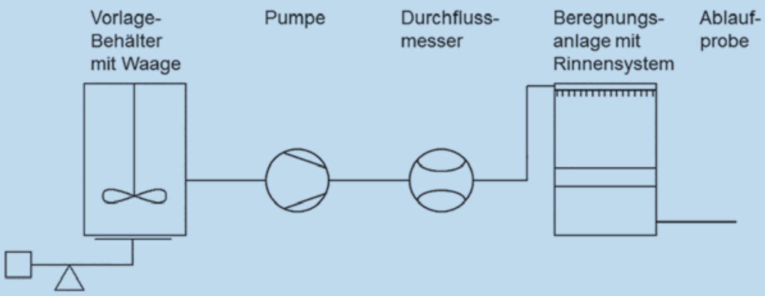
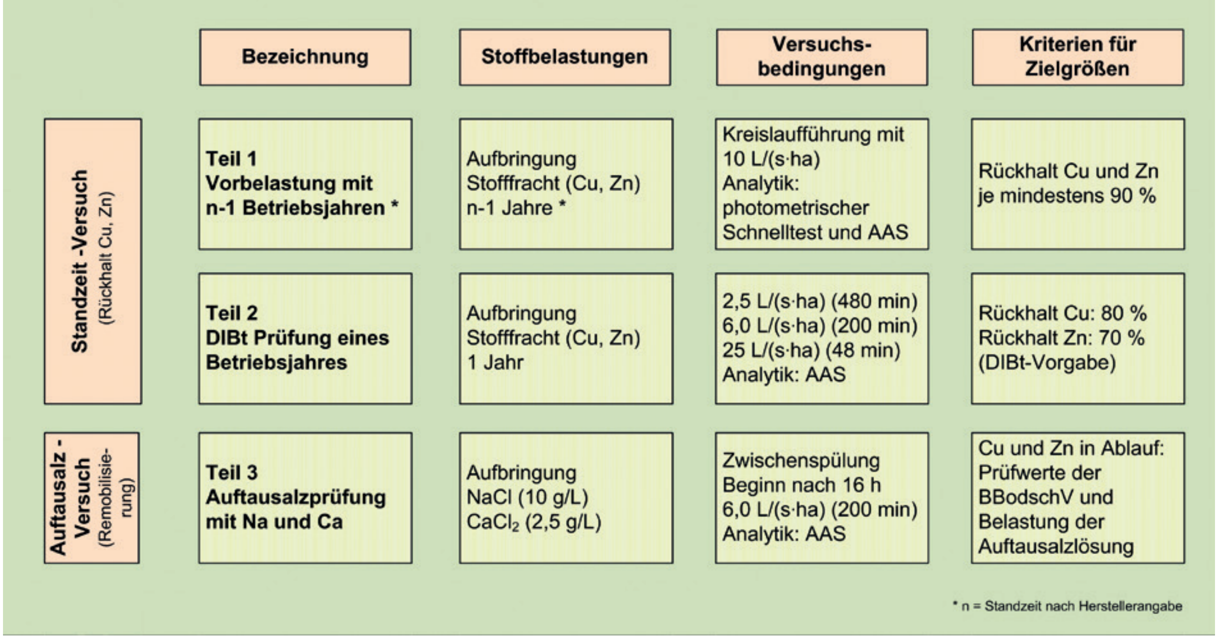


***Bild 1**. Versuchsaufbau für Rinnensysteme – Fließbild.*

***Bild 2**. Versuchsaufbau für Rinnensysteme – Ansicht der Beregnungsanlage, der beiden Edelstahlbleche sowie in der Mitte unten die beiden hintereinander angeordneten Rinnenkörper (Doppelbestimmung).*

Das bei diesen Versuchen verwendete Beschickungswasser besteht aus deionisiertem Wasser, welches künstlich mit den aufkommensrelevanten Schwermetallen Zink und Kupfer angereichert wird. Die beiden Schwermetalle werden in Kombination als gut lösliche Nitrat-Salze zugegeben, damit die Wechselwirkungen zwischen beiden Schwermetallen und den jeweiligen Substraten zeitgleich untersucht werden können, wobei die in den Chemikalien enthaltenen Anionen keinen Einfluss auf die Funktionsweise der Substrate haben. Die elektrische Leitfähigkeit (LF) des deionisierten Wassers ohne Zusätze beträgt ca. 1 µS/cm. Der pH-Wert der Lösung wird mit Salpetersäure auf pH 4,8 bis pH 5,1 eingestellt, damit die beiden Schwermetalle vollständig gelöst vorliegen. Es wird kein Puffer hinzugegeben, damit sich der pH-Wert des Wassers in der Anlage entsprechend den darin vorherrschenden Bedingungen zügig verändern kann. Somit können die substratspezifischen Wirkungsmechanismen zum Rückhalt von Kupfer und Zink, zu denen die Adsorption, der Ionenaustausch sowie die Fällung mit Filtration gehören, realitätsnah abgebildet werden.

Die auf den Anschluss befestigter Verkehrsflächen bezogenen, gelösten Jahresfrachten von 135 mg/m^2 Zn und 15,5 mg/m^2 Cu werden entsprechend der Modell-



	Bezeichnung	Stoffbelastungen	Versuchsbedingungen	Kriterien für Zielgrößen
Standzeit -Versuch (Rückhalt Cu, Zn)	**Teil 1** **Vorbelastung mit n-1 Betriebsjahren ***	Aufbringung Stofffracht (Cu, Zn) n-1 Jahre *	Kreislaufführung mit 10 L/(s·ha) Analytik: photometrischer Schnelltest und AAS	Rückhalt Cu und Zn je mindestens 90 %
	Teil 2 **DIBt Prüfung eines Betriebsjahres**	Aufbringung Stofffracht (Cu, Zn) 1 Jahr	2,5 L/(s·ha) (480 min) 6,0 L/(s·ha) (200 min) 25 L/(s·ha) (48 min) Analytik: AAS	Rückhalt Cu: 80 % Rückhalt Zn: 70 % (DIBt-Vorgabe)
Auftausalz - Versuch (Remobilisierung)	**Teil 3** **Auftausalzprüfung mit Na und Ca**	Aufbringung NaCl (10 g/L) $CaCl_2$ (2,5 g/L)	Zwischenspülung Beginn nach 16 h 6,0 L/(s·ha) (200 min) Analytik: AAS	Cu und Zn in Ablauf: Prüfwerte der BBodschV und Belastung der Auftausalzlösung

* n = Standzeit nach Herstellerangabe

Bild 3. Versuchsdurchführung – Darstellung der einzelnen Teile.

dimensionierung als gelöste Gesamtfrachten auf die zu untersuchenden Anlagen aufgebracht und beruhen auf folgenden Annahmen: Für die Berechnung der gelösten Frachten für Zink und Kupfer wurden die im DIBt-Verfahren verwendeten Schwerpunktkonzentrationen herangezogen, welche aus einer umfangreichen Literaturstudie festgelegt wurden und mit den eigenen Literaturrecherchen (siehe **Tabelle 2**) sehr gut übereinstimmen [18]. Die für die Versuche verwendeten gelösten Anteile der Schwerpunktkonzentrationen liegen bei ca. 200 µg/L Zn und bei etwa 23 µg/L Cu. Diese spiegeln somit die Konzentrationen typischer Verkehrsflächenabflüsse wider, lassen jedoch keine Unterscheidung nach durchschnittlicher täglicher Verkehrsstärke (DTV), Nutzung der Fläche sowie weiteren Faktoren zu. Die Abflussannahme beruht auf Auswertungen der Niederschlagsstation Mühldorf am Inn in Bayern durch das LfU [19]. An dieser Station betrug die langjährige mittlere Niederschlagshöhe 888 mm und der Jahresabflussbeiwert lag bei 0,76. Das Ergebnis wird in Zulassungsverfahren für Niederschlagswasserbehandlungsanlagen als Worst-Case-Szenario fachlich anerkannt.

Für die Berechnung der gelösten Gesamtfrachten werden die maximale Anschlussfläche und die vom Hersteller angegebene Standzeit der Anlage verwendet. Außerdem erfolgt die Skalierung der Gesamtfrachten auf das im Modell anteilig verwendete Substratvolumen linear.

3.2 Versuchsdurchführung

Das entwickelte Verfahren gliedert sich in drei Teile, welche in **Bild 3** dargestellt sind und in diesem Kapitel erläutert werden.

Im **Teil 1** der Versuchsdurchführung wird eine Vorbelastung mit einer Fracht von (n-1) Jahren aufgebracht, wobei n für die vom Hersteller angegebene Standzeit der Anlage in Jahren steht. Abhängig von den aufzubringenden Zink- und Kupferfrachten werden für diesen Versuchsteil Wassermengen zwischen 35 L und 120 L verwendet, wodurch sich für jedes Versuchsmodell spezifische Schwermetallkonzentrationen im Vorlagebehälter ergeben. Bei den auf diese Weise gewählten Wassermengen liegen die Konzentrationen bei allen Vorbelastungsversuchen in vergleichbarer Größenordnung vor. Aus jeder Vorlage wird eine Probe entnommen und analysiert, um die Soll-Konzentrationen zu überprüfen. Die Durchführung von Teil 1 erfolgt für alle Systeme gleich. Dabei werden (n-1) Jahre als Vorbelastung mit einer Regenspende von 10 L/(s·ha) aufgebracht. Der Ablauf wird bei allen Versuchen komplett in einem entsprechend großen Behälter aufgefangen. Nach einem Durchgang der Vorlage werden die darin enthaltenen Ablaufmischkonzentrationen an Zink und Kupfer mit einem photometrischen Küvetten-Schnelltest bestimmt, dessen Genauigkeit und Bestimmungsgrenzen in diesem Fall ausreichend sind. Gegebenenfalls wird der Ablauf weitere Male unter gleichen Bedingungen durch das Versuchsmodell gepumpt, bis mindestens 90 % von jeder der beiden Schwermetallfrachten in der Anlage zurückgehalten wurden. Abschließend werden zwei Ablaufmischproben entnommen und mittels AAS analysiert.

Beim **Teil 2** werden die Schwermetallfrachten des n-ten Jahres aufgebracht, welche somit die Frachten eines Jahres darstellen. Die Durchführung erfolgt analog zu den Wirksamkeitsprüfungen im DIBt-Verfahren unter Verwendung des mit (n-1) Jahren vorbelasteten Modells. Die berechnete n-te Jahresfracht wird in drei gleich große Frachtportionen aufgeteilt, die in jeweils einer Teilregenspende auf das Versuchsmodell aufgebracht werden. Die drei Teilregenspenden betragen 2,5 L/(s·ha), 6,0 L/(s·ha) und 25 L/(s·ha) mit den entsprechenden Dauern von 480 min, 200 min und 48 min. Somit ergibt sich bei jeder Teilregenspende eines Modells im Zulauf das gleiche Wasservolumen. Die im Zulaufwasser gleichzeitig vorhandenen Schwermetallkonzentrationen sind gegenüber der Realität erhöht und betragen unabhängig von der zu untersuchenden Anlage immer 6250 µg/L Zn und 718 µg/L Cu [1]. Daher wird in Anlehnung an das DIBt-Prüfverfahren ein definierter Stoffrückhalt, welcher 70 % für Zink und 80 % für Kupfer beträgt [1], gefordert, der in Abhängigkeit von den derzeit gültigen gesetzlichen Anforderungen berechnet wurde. Vor jedem Versuchsbeginn wird eine Zulaufprobe entnommen und analysiert, um die Soll-Konzentrationen zu überprüfen. Im Ablauf erfolgen pro Teilregenspende vier Probenahmen mit je zwei Probeflaschen nach jeweils einem Viertel der Versuchsdauer, in denen jeweils die beiden Schwermetalle Zink und Kupfer bestimmt werden.

Nach dem Aufbringen der drei Teilregenspenden wird der Versuchsaufbau mit Trinkwasser gespült, um das mit Schwermetallen belastete Beschickungswasser aus dem Versuchsmodell vollständig zu entfernen. Die Menge an verwendetem Trinkwasser entspricht der einer Teilregenspende aus Teil 2. Es werden ebenfalls eine Zulaufprobe zur Analyse der Trinkwasserzusammensetzung sowie nach jeweils der Hälfte der Spüldauer je zwei Proben im Ablauf zur Bestimmung der Schwermetallkonzentrationen entnommen.

Frühestens 16 Stunden nach der Spülung beginnt **Teil 3** der Untersuchung, welcher sich mit dem Rückhalt der Schwermetalle unter Salzeinfluss befasst. Es wird das gleiche, belastete Modell verwendet, welches auch bei den Versuchsteilen 1 und 2 verwendet wurde. Bei diesem Versuch wird eine Auftausalzmischung mit Zulaufkonzentrationen von 10 g NaCl/L [20] und 2,5 g $CaCl_2$/L verwendet. Das Wasservolumen entspricht immer der in Teil 2 pro Teilregenspende verwendeten Wassermenge und die Chloridzulaufkonzentration liegt bei 7660 mg/L. Als Regenspende wird in Teil 3 abweichend vom DIBt 6,0 L/(s·ha) bei einer Dauer von 200 Minuten verwendet, da in der Realität im Winter niedrigere Regenspenden deutlich häufiger auftreten als im Sommer. Eine Messung der Chlorid-, Zink- und Kupferkonzentrationen in der Auftausalzlösung wird durchgeführt, um zum einen die richtige Dosierung der Auftausalze und zum anderen eventuelle Verunreinigungen der verwendeten Salze mit Schwermetallen feststellen zu können. Im Ablauf werden vier Proben mit je drei Probeflaschen nach jeweils einem Viertel der Versuchsdauer genommen. Jeweils eine Probe wird auf Chlorid untersucht und die anderen Proben werden auf Zink und Kupfer analysiert. Analog zum DIBt-Verfahren beträgt bei diesem Versuchsteil die zulässige Zink-Ablaufkonzentration (500 + Messwert Auftausalzlösung) µg/L und die zulässige Kupfer-Ablaufkonzentration (50 + Messwert Auftausalzlösung) µg/L. Anhand dieses Versuchs ist eine Aussage über eine mögliche Schwermetallremobilisierung im Filtersubstrat möglich. Diese potenziell mögliche Rücklösung von zuvor zurückgehaltenen Schwermetallionen durch Auftausalze in der Behandlungsanlage wird durch erhöhte Ablaufkonzentrationen im vorgeschlagenen Verfahren detektiert. Somit wird durch den Versuchsteil 3 sichergestellt, dass die geforderten Ablaufkonzentrationen auch am Ende der untersuchten Standzeit während der simulierten Winterdienstsaison eingehalten werden. Die einzuhaltenden Schwermetallablaufkonzentrationen orientieren sich an den Werten der BBodSchV und müssen eventuell bei einer Umsetzung der Mantel-V angepasst werden. Gleiches gilt für einen möglichen Chlorid-Prüfwert in der Mantel-V.

3.3 Materialien und Analysenmethoden

Die für die Schwermetalle verwendeten Chemikalien waren Zinknitrat-Hexahydrat reinst (AppliChem GmbH) und Kupfer(II)-nitrat-Trihydrat reinst (AppliChem GmbH). Für die pH-Wert-Einstellung wurde Salpetersäure 65 % zur Analyse (Merck KGaA) verwendet.

Bei den Untersuchungen zum Einfluss von Auftausalz auf eine mögliche Rücklösung der Schwermetalle wurde Trinkwasser vom Forschungsgelände der TU München in Garching als Beschickungswasser mit einer LF von ca. 580 µS/cm verwendet, dem gleichzeitig die Salze NaCl und $CaCl_2$ zugegeben wurden. Als NaCl wurde ein handelsübliches Auftausalz (esco Steinsalz, 25-kg-PE-Sack) nach TL-Streu verwendet [21], welches laut Hersteller eine Löslichkeit von fast 100 % sowie einen hohen NaCl-Gehalt von rund 99 % aufweist. Als $CaCl_2$ reinst wurde eine getrocknete und gepulverte Chemikalie (AppliChem GmbH) verwendet.

Zur Probenahme wurden 250 mL PE-LD-Flaschen (VWR Collection) verwendet. Bei jeder entnommenen Wasserprobe wurden neben den Zink- und Kupfer-Konzentrationen mittels Atomabsorptionsspektrometrie (AAS) nach DIN 38406-E8 bzw. DIN 38406-E7 auch der pH-Wert nach EN ISO 10523:2012, die LF nach DIN EN 27888-C8 sowie die Temperatur nach DIN 38404-C4 bestimmt [22]. Bei den Ablaufmischproben aus Teil 1 wurden zur Bestimmung photometrische Küvetten-Schnelltests der Firma HACH LANGE GmbH verwendet (LCK 329 für Cu und LCK 360 für Zn). Bei den Versuchen mit Auftausalz wurde Cl mittels Ionenchromatographie nach EN ISO 10304-1:2009-D19 analysiert [22]. Alle Proben wurden mit Salpetersäure 65 % zur Analyse (Merck KGaA) konserviert. Bei zusätzlichen Analysen wurden Na, Ca und Magnesium mittels AAS nach DIN 38406-E14 (Na) bzw. DIN 38406-E3-1 (Ca und Magnesium) bestimmt. Der Königswasseraufschluss bei der schichtweisen Beprobung zur Ermittlung der Zn- und Cu-Konzentrationen erfolgte nach DIN 38414-S7.

4. Ergebnisse und Diskussion

4.1 Laborverfahren

Die Durchführbarkeit des in diesem Vorhaben entwickelten Verfahrens wird im Folgenden beispielhaft anhand der Untersuchung eines kommerziellen Rinnensystems, welches bereits durch das DIBt zur Behandlung von Verkehrsflächenabflüssen zugelassen ist und eine Standzeit von zehn Jahren hat, aufgezeigt.

Die Schwermetallvorbelastung, welche im Teil 1 neun Jahren entspricht und auf zwei Rinnenelemente desselben Herstellers mit einer Baulänge von je 0,50 m aufgebracht wurde, wurde in je 60 L deionisiertem Wasser gelöst. Somit ergaben sich Zulaufkonzentrationen von 114 000 mg/L Zn und 12 900 mg/L Cu. Die Vorbelastung konnte während der Untersuchung auf das Rinnensubstrat beim ersten Durchgang durch das System mit einer Vorbelastungsregenspende von 10 L/(s·ha) aufgebracht werden. Aus den gemessenen Ablaufmischkonzentrationen (849 mg/L Zn und 80 mg/L Cu) ergaben sich im Mittel ein Zinkrückhalt von 99,3 % und ein Kupferrückhalt von 99,4 %, welche beide deutlich über den jeweils geforderten 90 % an Zink- und Kupferrückhaltewerten lagen.

Beim Teil 2 des Verfahrens wurden pro Teilregenspende 86,4 L Wasser benötigt. Die maximal gemessene Zinkkonzentration im Ablauf betrug 75,1 µg/L und die maximal gemessene Kupferkonzentration lag bei 7,5 µg/L. Somit lagen alle Ablaufkonzentrationen unterhalb der Prüfwerte der BBodSchV für den Wirkungspfad Boden-Grundwasser und die geforderten Rückhalte von mindestens 70 % Zink und 80 % Kupfer wurden erreicht. Bei allen drei Regenspenden ergaben sich Kupferrückhaltewerte von jeweils 99,2 %. Bei Zink betrug der Rückhalt 99,5 % bei der mittleren Regenspende mit 6,0 L/(s·ha) und 99,4 % bei den anderen beiden Regenspenden.

Beim Teil 3 lagen die Kupferkonzentration in der Auftausalzlösung bei 16,9 µg/L und die Zinkkonzentration bei 702 µg/L. Alle gemessenen Ablaufkonzentrationen waren bei beiden Rinnen unterhalb der Prüfwerte der BBodSchV für den Wirkungspfad Boden-Grundwasser, wobei die Kupferwerte sogar durchwegs unterhalb der Bestimmungsgrenze von 5 µg/L lagen. Die höchste gemessene Zinkablaufkonzentration betrug 36,2 µg/L. Die nach diesem Laborverfahren im Ablauf zulässigen Konzentrationen, getrennt nach BBodSchV und Mantel-V, sind in **Tabelle 3** aufgeführt. Daraus ergibt sich, dass die an die Anlage gestellten Anforderungskriterien selbst bei einer möglichen Verschärfung durch die Mantel-V (zulässige Kupferkonzentration 30,9 µg/L und zulässige Zinkkonzentration 760 µg/L) eingehalten wurden. Die während dieses Versuchsteils gemessenen Chloridablaufkonzentrationen betrugen 6840 mg/L im Mittel und lagen damit nur geringfügig unterhalb der Zulaufkonzentration von 8290 mg/L, welche etwa den in der Realität gemessenen Spitzenwerten in Verkehrsflächenabflüssen entspricht.

Tabelle 3. Zulässige Ablaufkonzentrationen für das dargestellte Rinnensystem auf Grundlage der Prüfwerte zur Beurteilung des Wirkungspfads Boden-Grundwasser gemäß BBodSchV (zul. AK_B) und Mantel-V (zul. AK_M) [2, 3].

	Einheit	Auftausalzlösung	Prüfwert BBodSchV	zul. AK_B	Prüfwert Mantel-V	zul. AK_M
Cu	µg/L	16,9	50	66,9	14	30,9
Zn	µg/L	702	500	1 200	58	760

Aus diesen Ergebnissen folgt, dass eine Einführung eines Prüf- oder Grenzwertes für Chlorid zu Problemen führen würde. So sieht der Entwurf der Mantel-V [3] einen Prüfwert von 250 mg/L Cl vor (siehe **Tabelle 1**). Alle derzeit vom DIBt zugelassenen Behandlungsanlagen sowie Oberboden können Chlorid aus chemisch-physikalischen Gründen nicht zurückhalten und würden somit den nach Mantel-V geforderten Wert während der Streusalzphase im Winter nicht einhalten können. Dieses gilt in gleichem Maße für Flächenversickerungsanlagen und Mulden- bzw. Mulden-Rigolen-Systeme nach DWA-A 138 [23]. Dies würde auch bereits eingebaute Systeme für die anschließende Versickerung betreffen.

Hier ergibt sich weiterer Forschungsbedarf. So muss z. B. geklärt werden, wie sich die zeitlich begrenzte Chloridspitzenkonzentration auf den Boden-Grundwasserkörper auswirkt und ob die Betrachtung einer mittleren Jahreskonzentration in diesem speziellen Fall legitim ist. Eine Einschränkung der Auftausalzausbringung hat in jedem Fall weitreichende Folgen für die Verkehrssicherheit, insbesondere auf Autobahnen. Somit kann diese Thematik an dieser Stelle nicht abschließend beurteilt werden.

Mit dem in diesem Vorhaben entwickelten Verfahren war es möglich, im Labor Aussagen zur Standzeit und zu Veränderungen im Rückhalt der beiden Leitparameter Zink und Kupfer zu treffen. So konnte an dem als Beispiel dargestellten Rinnensystem die Herstellerangabe zur Standzeit bestätigt werden. Im zweiten Versuchsteil konnte keine Verschlechterung der Ablaufwerte im Verlauf der drei Regenspenden festgestellt werden, sodass von einem Puffer bei der Standzeit zum Schwermetallrückhalt für diese Anlage ausgegangen werden kann. Außerdem wurden während der Untersuchung bei diesem Rinnensystem keine Schwermetalle durch Na^+ und Ca^{2+} remobilisiert. Somit kann durch dieses Vorgehen eine weitreichendere Aussage zu einer möglichen Schwermetallremobilisierung getroffen werden, als dies mit der derzeitigen Salzprüfung der DIBt-Zulassungsgrundsätze möglich ist.

4.2 Vergleich der Substratschwermetallgehalte im Labor mit Feldmessungen

Das in der dargestellten Untersuchung verwendete Substrat hatte vor Versuchsbeginn einen Zinkgehalt von

51 mg/kgTR und einen Kupfergehalt von 23 mg/kgTR (TR: Trockenrückstand). Nach der Versuchsdurchführung im Labor wurden die obersten 10 cm des verwendeten Filtersubstrats beprobt und bezüglich des Zink- und Kupfergehaltes analysiert. Die bestimmten Schwermetallgehalte lagen nach dem Aufbringen von zehn Jahresfrachten im Mittel bei 468 mg/kgTR für Zink und bei 70 mg/kgTR für Kupfer. Ein Vergleich der im Labor ermittelten Ergebnisse mit regelmäßigen Feldbeprobungen einer seit neun Jahren im Betrieb befindlichen Rinne desselben Herstellers zeigt, dass die beiden Stoffe Zink und Kupfer auch am untersuchten Standort aufgrund ihrer Aufkommensrelevanz im Vergleich zu anderen Schwermetallen den größten Anteil der zurückgehaltenen Mengen ausmachen (siehe **Bild 4**). Dadurch wird die Auswahl, nur die beiden aufkommensrelevanten Stoffe Zink und Kupfer im Labor zu betrachten, nochmals bestätigt.

Bei der Feldmessung lagen die Ausgangsgehalte bei 23 mg/kgTR Zn und 11 mg/kgTR Cu, welche geogen als Bestandteile der Mineralien im Substrat enthalten waren und nicht eluiert werden konnten. Sie lagen damit unterhalb der Werte der Laborrinne vor Versuchsbeginn. Ein Vergleich der Anreicherungen aus neun Jahren in situ gemessener Schwermetallgehalte (186 mg/kgTR Zn und 39 mg/kgTR Cu) mit denen im Labor ergibt in der Tendenz eine gute Übereinstimmung. Es zeigte sich, dass die getroffenen Annahmen für die Schwermetallkonzentrationen zur Versuchsdurchführung im Vergleich zu den tatsächlichen Belastungen einer Rinne in situ höher waren. Damit wird sichergestellt, dass an anderen Einbauorten mit höheren Belastungen die sich daraus ergebenden höheren Frachten in der gleichen Zeit zurückgehalten werden können. Somit erscheint diese Methode für die Standzeitbestimmung anhand der durchgeführten Evaluierung des Laborverfahrens an einer Feldbeprobung geeignet zu sein.

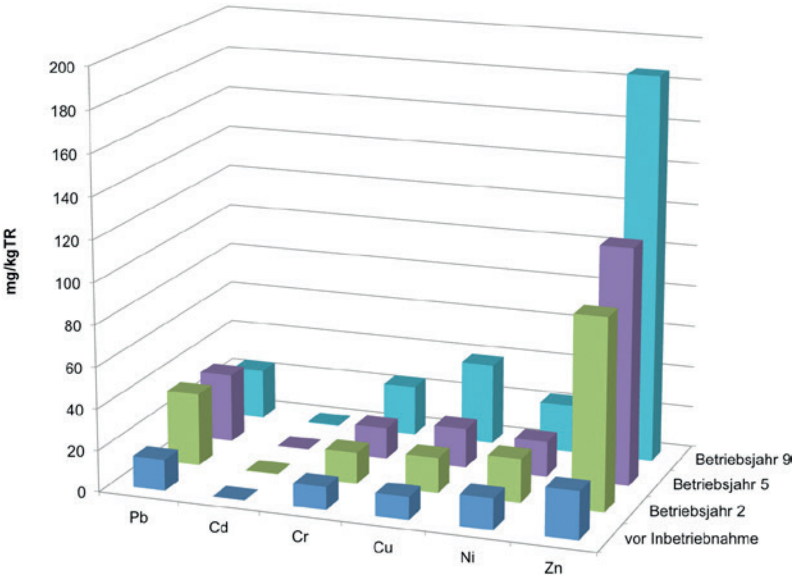


***Bild 4**. Zeitlicher Verlauf der Schwermetallgehalte eines in situ beprobten Rinnensubstrats [5].*

5. Fazit

Die Ermittlung von Standzeiten dezentraler Behandlungsanlagen ist nach wie vor ein schwieriges und teilweise offenes Thema. Dies liegt vor allem daran, dass sehr viele Einflussfaktoren das Auftreten eines Versagensfalls an sich und dessen Ausmaß stark beeinflussen. In vielen Fällen wird die Kolmation als limitierender Prozess für die Standzeit identifiziert. Gerade für diesen Prozess ist keine Labormethode verfügbar und auch kaum zu entwickeln.

Zur Ableitung einer Methodik zur standardisierten Standzeitermittlung bei Niederschlagswasserbehandlungsanlagen für Verkehrsflächenabflüsse ist es unabdingbar, verfügbare Betriebsdaten auszuwerten. Weiterhin sind Betriebsbegehungen inklusive Messungen der hydraulischen Kennwerte wünschenswert. Hieraus könnte eine Verknüpfung der hydraulischen Leistungsfähigkeit mit der Charakteristik der Anlagen und der jeweiligen Örtlichkeit vollzogen werden. Damit ließen sich Belastungsklassen definieren und die Vorhersage von Kolmationseffekten verbessern.

Insgesamt ist festzuhalten, dass realistische Standzeiten mit Bezug auf den gelösten Schwermetallrückhalt für Rinnensysteme nach dem beschriebenen Verfahren, welches in drei Teile gegliedert ist und die die Standzeit limitierenden Schwermetalle Zink und Kupfer berücksichtigt, erfolgreich bestimmt werden können.

Auch für Schachtsysteme konnten mit dem beschriebenen Verfahren wertvolle Erkenntnisse zum Standzeitverhalten gewonnen werden. Da sich bei den Standzeitversuchen der Schachtsysteme durch den spezifischen Modellaufbau Unterschiede ergeben, erscheinen hierzu weitere Untersuchungen wünschenswert. Dazu gehören ein Vergleich verschiedener Modellfaktoren bei der Abbildung der Schachtfiltereinheiten als Säulenmodelle sowie eine Beurteilung der Bedeutung von Ruhephasen sowohl während als auch zwischen den einzelnen Versuchsteilen, da innerhalb der Untersuchung Niederschläge ohne die in der Realität vorhandenen Trockenzeiten aufgebracht werden. Diese haben bei den Schachtsystemen aufgrund der Funktionsweise der Substrate und des größeren Verhältnisses aus befestigter Verkehrsanschlussfläche zu Filteroberfläche eine höhere Bedeutung als bei den Rinnensystemen.

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Autoren

Maximilian Huber, M.Sc.
Wissenschaftlicher Mitarbeiter
Prof. Dr.-Ing. **Jörg E. Drewes**
PD Dr. rer. nat. habil. **Brigitte Helmreich**
(Korrespondenzautorin) |
E-Mail: b.helmreich@tum.de |
Technische Universität München |
Lehrstuhl für Siedlungswasserwirtschaft |
Am Coulombwall 8 |
D-85748 Garching

Prof. Dr.-Ing. habil. **Antje Welker**
Dipl.-Ing. **Martina Dierschke**
FH Frankfurt |
Fachgebiet Siedlungswasserwirtschaft und Hydromechanik |
Nibelungenplatz 1 |
D-60318 Frankfurt am Main

7.2 Research Paper: "A Novel Test Method to Determine the Filter Material Service Life of Decentralized Systems Treating Runoff from Traffic Areas"

The selection of the two authoritative parameters (Cu and Zn), the boundary conditions, and the original procedure of the novel method were presented in Section 7.1. After completing the assessment of eight more systems, the final experimental setup was described in this journal article that does not contain an application of de-icing salts (cf. Chapter 9). The assessment method can be used to determine service lives and long-term performances of all types of decentralized stormwater treatment systems with filter materials in pilot-scale models. Furthermore, additional field samples were collected to show the applicability of the novel procedure. The corresponding hypothesis was:

A validation of the determined service lives of channel systems by field measurements confirms the applicability of the developed method (Hypothesis 5).

The validation of the results of two channel systems at four field-monitoring sites confirmed the applicability of the novel test method despite different site-specific characteristics. Moreover, the long-term performances of the nine decentralized filter systems varied significantly for the two authoritative parameters Cu and Zn between the different rain events, which have an impact on management strategies to prevent the transport of pollutants into the environment. Furthermore, the type and amount of the filter material influenced the retention of Cu and Zn significantly for all systems. The results of the novel test method provide regulatory authorities, designers, and operators with a more objective basis for performance assessment. In addition, further substances such as nutrients can have a crucial impact on the receiving water (Sample et al., 2012). An application of these substances in the novel test method is possible after a calculation of the annual loads. Thus, this method was successfully expanded to evaluate the simultaneous removal of Cu, Zn, and P. Both the service life (Part 1) and the long-term performance (Part 2) were determined for all three substances for one FSS. The relevance of P was also proposed by Welker et al. (2015). In lab-scale experiments, the removal of Cu, Zn, P, and trace organic substances (i.e., ETBE and MTBE) was tested. The results showed that ETBE and MTBE were more easily removed than P. Furthermore, a relevance of ETBE and MTBE in traffic area runoff could not be confirmed by current monitoring programs presented by Grotehusmann et al. (2014) and Vesting et al. (2015). Moreover, MTBE can be easily removed by activated carbon (Li, 2012; Wichern et al., 2012).

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Research article

A novel test method to determine the filter material service life of decentralized systems treating runoff from traffic areas

Maximilian Huber [a], Antje Welker [b], Martina Dierschke [b], Jörg E. Drewes [a], Brigitte Helmreich [a, *]

[a] Chair of Urban Water Systems Engineering, Technical University of Munich, Am Coulombwall 3, 85748 Garching, Germany
[b] Fachgebiet Siedlungswasserwirtschaft und Hydromechanik, Frankfurt University of Applied Sciences, Nibelungenplatz 1, 60318 Frankfurt am Main, Germany

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ABSTRACT

In recent years, there has been a significant increase in the development and application of technical decentralized filter systems for the treatment of runoff from traffic areas. However, there are still many uncertainties regarding the service life and the performance of filter materials that are employed in decentralized treatment systems. These filter media are designed to prevent the transport of pollutants into the environment. A novel pilot-scale test method was developed to determine – within a few days – the service lives and long-term removal efficiencies for dissolved heavy metals in stormwater treatment systems. The proposed method consists of several steps including preloading the filter media in a pilot-scale model with copper and zinc by a load of n-1 years of the estimated service life (n). Subsequently, three representative rain events are simulated to evaluate the long-term performance by dissolved copper and zinc during the last year of application. The presented results, which verified the applicability of this method, were obtained for three filter channel systems and six filter shaft systems. The performance of the evaluated systems varied largely for both tested heavy metals and during all three simulated rain events. A validation of the pilot-scale assessment method with field measurements was also performed for two systems. Findings of this study suggest that this novel method does provide a standardized and accurate estimation of service intervals of decentralized treatment systems employing various filter materials. The method also provides regulatory authorities, designers, and operators with an objective basis for performance assessment and supports stormwater managers to make decisions for the installation of such decentralized treatment systems.

1. Introduction

Parameters commonly monitored in runoff from traffic areas include organic substances, heavy metals, and compounds of de-icing salts (Eriksson et al., 2007; Folkeson et al., 2009; Fraga et al., 2016). Heavy metals such as cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni), and zinc (Zn) are highly relevant pollutants because of their toxicity, non-degradability, and their increasing presence in the environment as a consequence of their widespread industrial use (Roeva et al., 1996; Tiefenthaler et al., 2001). Among these heavy metals, Cu, Pb, and Zn commonly occur in highest concentrations in runoff from highways and other traffic areas (Kayhanian et al., 2012; Revitt et al., 2014; Huber et al., 2016a). However, Pb concentrations have decreased significantly in traffic area runoff in the last few decades because of the phase-out of leaded gasoline and the substitution of Pb in other traffic-related sources (Kayhanian, 2012; Huber et al., 2016a). In addition, Pb occurs mostly particle-bound, while Cu and Zn are usually more present in the dissolved phase (Timperley et al., 2005; Wilson, 2006; Kayhanian et al., 2007). Helmreich et al. (2010) concluded that the fractionation of these heavy metals is not affected by seasonal variations but remarkable fluctuations can occur between different rain events with dissolved fractions higher than 90%. Therefore, Cu and Zn are relevant dissolved substances in runoff from traffic areas during all seasons because of their total concentrations, fractionation behavior, and toxicity.

To retain these heavy metals from traffic area runoff, decentralized stormwater treatment systems are used, which can be

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* Corresponding author.
E-mail address: b.helmreich@tum.de (B. Helmreich).

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designed as an integral part of existing urban infrastructure for stormwater management and rainwater harvesting (Scholes et al., 2008; Inamdar et al., 2013; Scholz, 2015). In a first step of these treatment systems, sedimentation and filtration are the predominant retention mechanisms used to remove particle-bound substances. Subsequently, dissolved heavy metals are retained on filter materials by sorption, ion exchange, or precipitation (Liu et al., 2005). Thus, a second treatment step with a filter material is necessary to reduce the total concentrations of all metals (Hilliges et al., 2013; Maniquiz-Redillas and Kim, 2014). In recent years, various technical decentralized stormwater treatment systems have been developed to treat the runoff of traffic areas when space is limited (Sample et al., 2012; Dierkes et al., 2015a). Three categories of these urban drainage systems are: permeable pavement, filter channel system (FCS), and filter shaft system (FSS). These systems are commonly used as conforming products in order to treat stormwater runoff at the source before it infiltrates into the subsurface or is discharged to surface water. Permeable pavements are similar to those of traditional concrete block pavements. They are specifically designed for infiltration of stormwater through the pores of the pavers and/or the joints between the pavers, which are filled with filter media to retain pollutants, into the various pavement layers and subsequently into the soil (Dierkes et al., 2015a). A FCS consists of a typical drainage channel (concrete or polypropylene) that is filled by filter materials (mixture, layers, or only one material). Each FCS has a specific retention volume between the grate and the filter media and the stormwater runoff percolates (down-flow mode) through the filter media and the treated water is subsequently drained by holes in the bottom of the channel or by a pipe. The pollutants are retained by sedimentation on top of the filter and within the filter media by filtration and (bio-)chemical processes. Some FCS have a sedimentation box/trap (polypropylene or steel) ahead of the filter. FSS usually consist of a sedimentation unit and a subsequent filter, which is integrated either into the system or in another shaft (concrete or polypropylene). The stormwater can be treated by the filter vertically in both up-flow and down-flow mode, or in a radial direction. Some FSS are permanently submerged and others run dry after each rainfall event (Dierkes et al., 2015a). In Germany, the Deutsches Institut für Bautechnik (DIBt, German Center of Competence for Construction) can approve these decentralized systems for the infiltration of runoff from traffic areas after performing several tests (e.g., removal of fine particles, total petroleum hydrocarbons, and heavy metals; remobilization of Cu and Zn under application of sodium chloride) (DIBt, 2012; DIBt, 2015). In the United States, several methods have been developed to test decentralized systems. In the State of Washington, the Technology Assessment Protocol-Ecology (TAPE) of the Washington State Department of Ecology is used to evaluate and classify decentralized stormwater treatment systems by lab- and field-scale experiments (e.g., removal of total suspended solids, Cu, Zn, phosphor, and petroleum hydrocarbons) (Department of Ecology (2011)). In New Jersey, a procedure for obtaining verifications of stormwater treatment systems was implemented by the New Jersey Department of Environmental Protection to assess the removal of total suspended solids in the laboratory (NJDEP, 2013). As an amendment to the TARP (Technology Acceptance Reciprocity Partnership) Protocol, a field testing for the removal of total suspended solids can also be performed for final certification (NJDEP, 2009). However, there is no standardized method available to quickly determine the filter material service life of decentralized stormwater treatment systems.

In general, two factors influence the determination of the service life of decentralized systems treating runoff from traffic areas:

- The service interval is limited because of a hydraulic failure of the plant (i.e., clogging of the filter material because of sediment accumulation and deposition) (Mercado et al., 2015). This is often related to an increased solids load in combination with site-specific factors that enhance clogging (e.g., mineral substances such as grit and construction site dust, or organic loads of pollen and leaf litter) (CH2MHILL, 1998).
- The service life is limited because of the substance removal efficiency (i.e., a reduced retention of substances). In most cases, the capacity of the filter material for heavy metal removal is exhausted.

Clogging phenomena of the filter material can be simulated in the laboratory by using a full-scale system but realistic particle size distributions and site-specific factors of the system's catchment area cannot be considered. Therefore, this factor needs to be determined by extensive and comparable field measurements that might take several years. The terminal service life of the filter material (e.g., breakthrough of heavy metals) is rarely noticeable by the operator although this has a crucial effect on the receiving water. However, this effect can be simulated reproducibly at pilot-scale for decentralized systems. Nevertheless, a standardized test method for the determination of the service life and long-term performance of different types of filter systems is currently not published by other researchers. This lack regards the complete procedure (e.g., input parameters such as estimated service lives and metal loads, rainfall events, breaks, and effluent requirements) and the performing of an experimental study.

Since the treatment of traffic area runoff is a serious issue, the evaluation of decentralized filter systems for the removal of heavy metals is very important. An appropriate experimental setup is necessary to evaluate the removal of dissolved substances as the use of filter systems for stormwater treatment is increasing worldwide and keeping track of the filter usability and performance is currently a difficult task because of the multitude of manufacturers of filter systems. Such a method is also needed because, depending on the type of the system, field experience of 5–20 years is often missing for existing systems. Because of the missing experience and information concerning their long-term performance and service lives, it is currently difficult to guarantee a sustainable and cost efficient stormwater management by decentralized systems. The determination of the service life is also essential for a sustainable management of urban infrastructure because maintenance and the exchange of filter material have an effect on life-cycle costs of decentralized treatment systems (O'Sullivan et al., 2015). In addition, the benefit of using a novel method, which must consist of several well defined steps that represent the real conditions realistically, for testing new product developments and existing decentralized systems must also be based on a rapid performance assessment within a few days for toxic dissolved substances.

The hypothesis of this study is that a novel test method can be developed and implemented to evaluate the long-term performance of new and already operational decentralized systems. This novel assessment method can provide a simulation of the service lives of the filter materials and their long-term performances within a few days prior to field applications. The objectives of this study are the development of a pilot-scale test method, the evaluation of nine channel and shaft systems to determine their service lives and efficiencies to remove dissolved heavy metals, validation of results, and the delivery of stormwater treatment guidelines of authorities for the assessment of heavy metal retention performances by standardized procedures for decentralized stormwater treatment systems.

2. Material and methods

2.1. Method

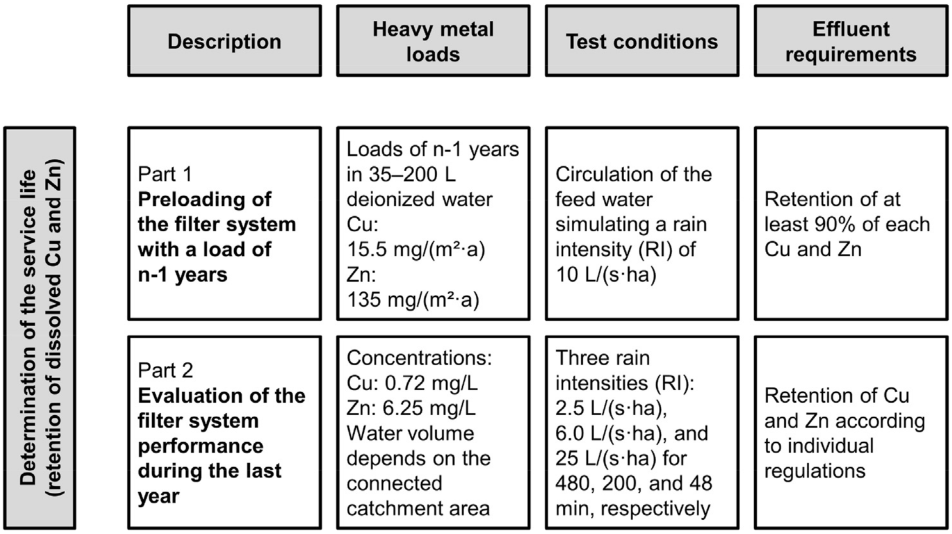
2.1.1. Scheme and requirements

The novel assessment method consists of two parts (Fig. 1). In Part 1, the filter system is rapidly preloaded with heavy metal loads of n-1 years. The variable n represents the estimated service life of the filter material in years that has been determined in previous batch or small-scale column experiments. By these lab-experiments, the capacities of the filter media, which is used in the full-scale system in the same composition, are determined for all substances of interest. Appropriate procedures for the determination of capacities are proposed by Huber et al. (2016b) with information on the initial pH value, the concentrations of the metals of interest, the substrate dosage, the type and speed of the shaker, the contact time, the influence of the ionic strength (e.g., buffer), and the sample preparation. The determined capacities (mg/kg) are multiplied for each substance (e.g., Cu and Zn), which has to be retained, by the total amount of filter material (kg) that is used in the full-scale system. This maximum amount of retained substances (mg) in the filter system without an exchange of the filter media (i.e., the service life) is subsequently divided by the corresponding typical annual loads (mg/a) (described in the following; values must be multiplied by the connected catchment area of the full-scale system) to roughly determine service lives for each substance. The estimated service life of the filter system is equal the rounded down value of the smallest value calculated for all substances. For the estimated service lives calculated from batch experiment results, a reduction of the measured capacities by a factor between four and eight is recommended.

For the calculation of the annual loads, which are specified per m^2 catchment area, total runoff concentrations of 443 μg/L Zn and 76.3 μg/L Cu are used in accordance with concentrations measured in field studies for road runoff (Welker, 2005). Assuming an average dissolved fraction of 45% for the total Zn concentrations and of 30% for the total Cu concentrations (Huber et al., 2016a), mean dissolved concentrations of 199 μg/L Zn and 22.9 μg/L Cu are calculated. Particulate heavy metals are not considered in this method because their removal mechanisms are not sorption, ion exchange, or precipitation but filtration. Therefore, metals that are bound onto particles in the influent of a system do not limit the service life of the filter material concerning the substance removal efficiency. Particulate fractions of pollutants are considered by other tests that are designed to evaluate the removal of fine particles (<200 μm) because most particulate heavy metals in highway runoff were occurring in this size fraction (Sansalone and Buchberger, 1997; Roger et al., 1998). Runoff is calculated as 677 mm (annual precipitation height 888 mm, mean annual runoff coefficient 0.76; Mühldorf am Inn, Germany) (Bayerisches Landesamt für Umwelt, 2008). Thus, the annual loads for traffic areas are 135 mg/m^2 Zn and 15.5 mg/m^2 Cu (DIBt, 2015). Values for other heavy metals can be calculated in the same manner after determination of the concentrations and the dissolved fractions (cf. Huber et al., 2016a). In Part 2, the long-term performance of the system is assessed by simulating the last year of the filter material service life. For all experiments, Cu and Zn are used and the filter systems are represented in individual pilot-scale models (cf. Section 2.2). To calculate the amount of Cu and Zn for each test, the connected catchment area of the full-scale system and a filter model factor to scale down the catchment area are considered.

2.1.2. Experimental setup

In Part 1, the preloading of the filter system with a load of n-1 years is performed. The total loads of Part 1 are calculated by multiplying the annual loads (135 mg/m^2 Zn and 15.5 mg/m^2 Cu) by n-1 years and the catchment area of the filter model. The heavy metals are dissolved in 35–200 L of water to obtain heavy metal concentrations in one order of magnitude for all systems (the amount of water also depends on the model of the filter system). Deionized water without a buffer is used as feed water and the pH value is adjusted to 4.9 ± 0.3. Thus, all heavy metals are present in the dissolved form (Genç-Fuhrman et al., 2007). The feed water circulates through the filter model with a rain intensity (RI) of 10 L/(s·ha), representing a precipitation of 3.6 mm/h, until 90% of each Cu and Zn are retained. One influent sample and several effluent samples (one sample after each complete circulation of the feed water) are taken. Afterwards, the filter materials are either submerged in the feed water or dried at room temperature (20 ± 2 °C)



	Description	Heavy metal loads	Test conditions	Effluent requirements
Determination of the service life (retention of dissolved Cu and Zn)	Part 1 **Preloading of the filter system with a load of n-1 years**	Loads of n-1 years in 35–200 L deionized water Cu: 15.5 mg/(m²·a) Zn: 135 mg/(m²·a)	Circulation of the feed water simulating a rain intensity (RI) of 10 L/(s·ha)	Retention of at least 90% of each Cu and Zn
	Part 2 **Evaluation of the filter system performance during the last year**	Concentrations: Cu: 0.72 mg/L Zn: 6.25 mg/L Water volume depends on the connected catchment area	Three rain intensities (RI): 2.5 L/(s·ha), 6.0 L/(s·ha), and 25 L/(s·ha) for 480, 200, and 48 min, respectively	Retention of Cu and Zn according to individual regulations

Fig. 1. Scheme of both parts of the filter material service life experiments (n equals the estimated service life in years that has been determined in previous batch or small-scale column experiments).

for several hours in accordance with the full-scale systems.

In Part 2, the filter system's performance during the last year is evaluated. The annual loads of Cu and Zn are the same as in Part 1. Part 2 is performed using three different RIs with the same amount of water to simulate the long-term performance for defined rain events: 2.5 L/(s·ha) for 480 min, 6.0 L/(s·ha) for 200 min, and 25 L/(s·ha) for 48 min (DIBt, 2015). They equal precipitations of 0.9 mm/h, 2.2 mm/h, and 9.0 mm/h, respectively. These rainfall intensities are selected after completing a comprehensive statistical analysis of typical rainfall intensities across Germany (longtime average) (Dierkes et al., 2015a). The water volume of each experiment depends on the connected catchment area of the filter model and is calculated from the RI and its duration (i.e., 7.2 L/m^2 per RI). The heavy metal loads of the last year are divided equally within the three RIs (45 mg/m^2 Zn and 5.17 mg/m^2 Cu per RI). Thus, the concentrations of each RI are always 0.72 mg/L Cu and 6.25 mg/L Zn, which are higher than most values of real runoff waters (Kayhanian et al., 2012; Huber et al., 2016a). Deionized water without a buffer is used as feed water and the pH value is adjusted to 4.9 ± 0.3. A break of 15 h is maintained between RI 2.5 L/(s·ha) and RI 6.0 L/(s·ha). This represents only a small antecedent dry period and is linked to the experimental setup (RI 2.5 L/(s·ha) takes one day and the other two RI one further day). Afterwards, RI 6.0 L/(s·ha) and RI 25 L/(s·ha) are performed in one run with changing flow rates. The retention of Cu and Zn is determined by measuring the influent concentrations and eight effluent concentrations per RI. Two effluent samples are taken per RI at the end of each quarter (e.g., after 12 min, 24 min, 36 min, and 48 min for RI 25 L/(s·ha)). The removal efficiencies are calculated as the ratio of the difference of the influent concentration and the arithmetic average of all eight effluent concentrations and the corresponding influent concentration. The efficiencies can directly be compared with effluent requirements or local regulations (cf. Section 4.2).

All experiments are performed in duplicate. The influent concentrations are adjusted with an accuracy of ±10% and the flow rate with an accuracy of ±5%. The experimental setup consists of a feed water tank with a scale, a pump, a flow meter and the model of the filter system (Fig. 2). The scale is used for preparing the feed water and to control the flow meter. The pump is adjusted by the flow meter to simulate different RIs. The model of the filter system depends on the type of the system and each system has an individual model. The different models are based on the fact that the functionality of each system varies because of specific design features (e.g., up/down/radial-flow, submerged/run dry after rainfall, and with/without pre-treatment), filter materials (e.g., type, grainsize, amount, filter bed height, and retention mechanisms), catchment areas, and estimated service lives (1–10 years). All filter models must consider the real flow conditions, the flow paths, and the contact times of the full-scale systems (DIBt, 2015). To simulate the service lives of FSS, adapted pilot-scale models are used to reduce the amount of water for each experiment (Dierkes et al., 2015b). However, at least 35 L of feed water should be used in Part 2 per RI (DIBt, 2015). Segments of the full-scale systems are used to test all FCS and some FSS. The model must be in accordance with the information of the manufacturer's instructions for full-scale systems for the filter materials, the heights, the amounts, and the procedures of filling.

2.2. Filter systems

An evaluation of three different FCS and six FSS, which are commonly used in urban stormwater management, was performed. The systems were selected to represent a wide range of commercial decentralized treatment systems, which were developed in Germany and are distributed worldwide, with filter materials designed to treat runoff from traffic areas (Huber et al., 2015a). The functionalities and maintenance of several systems selected for the following experiments are described by Dierkes et al. (2015a). The values n and catchment area were set in accordance with the specifications of the manufacturers and batch experiment results. A linear downscaling of the total loads of each filter system on the filter model was performed. The modelling factor, which was used to scale down the catchment areas, was 1:1 for all FCS and varied between 1:4 and 1:8000 for the FSS (Table 1).

All full-scale FSS are available in different sizes (i.e., different shaft diameter or different amounts of segments with filter material) and can subsequently be adopted for several catchment area sizes. Since the systems and the filter media are completely scaled up for different catchment areas (e.g., same ratio of catchment area to filter media surface area and same flow paths), the filter model factors and, therefore, the removal efficiencies are the same. Because of the different shaft designs, individual filter models that consisted of a representative segment or column were used for all FSS. For example, a column (inner diameter 8.0 cm) representing a section of the filter segments (e.g., two full segments for 1500 m^2 catchment area and four segments for 3000 m^2) was used to test the performance of FSS3 with a filter model factor of 1:82 (Fig. 3, left). For FSS1, a quarter of the filter unit was used for the pilot-scale experiments. Columns representing cross-sections of the filters are used for the other FSS. For the FCS, a section of each channel was used with full width and 0.5 m length (Fig. 3, right) or 1.0 m length. The rain-simulating-device for the FCS consisted of 625 hollow stainless steel needles (inner diameter 0.90 mm) that were fixed onto a square plate (1 m^2). The feed water was delivered with different RIs from the pressurized water reservoir installed on top of the FCS. All experiments were performed in duplicate.

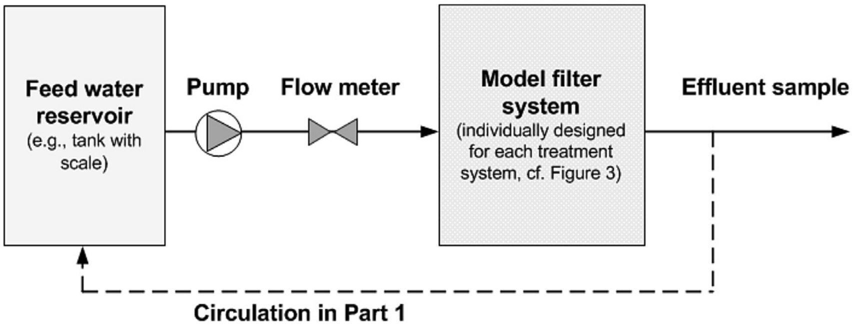


Fig. 2. Scheme of the experimental setup.

Table 1
Results of the evaluation of all treatment systems for Cu and Zn in Part 2 (means of duplicates).

Description	Type of system	Channel system (FCS)			Shaft system (FSS)					
	Connected catchment area	12–20 m^2/rm[a]			500–3000 m^2				10,000 m^2	
	Estimated service life	10 years			3–4 years				1 year	3 years
	System ID	FCS1	FCS2	FCS3	FSS1	FSS2	FSS3	FSS4	FSS5	FSS6
	Filter model factor	1:1	1:1	1:1	1:4	1:100	1:82	1:82	1:8000	1:430
	Main filter media component	Natural minerals	Calcium carbonate	Activated carbon	Zeolite	Zeolite	Granular ferric hydroxide	Granular ferric hydroxide	Activated carbon	Calcium silicate
Cu-Retention	RI 2.5 L/(s·ha)	99.2%	99.3%	72.7%	57.6%	55.4%	99.3%	98.8%	87.6%	95.8%
	RI 6.0 L/(s·ha)	99.2%	99.3%	66.6%	47.1%	54.4%	99.1%	97.9%	97.5%	89.0%
	RI 25 L/(s·ha)	99.2%	99.3%	56.0%	39.9%	51.2%	85.3%	79.5%	83.3%	79.7%
	Average	99.2%	99.3%	65.1%	48.2%	53.7%	94.6%	92.1%	89.5%	88.2%
Zn-Retention	RI 2.5 L/(s·ha)	99.4%	98.5%	68.3%	73.2%	65.3%	99.7%	98.6%	89.7%	98.4%
	RI 6.0 L/(s·ha)	99.5%	99.5%	66.9%	66.5%	58.9%	98.2%	94.4%	94.5%	89.5%
	RI 25 L/(s·ha)	99.4%	99.7%	55.3%	60.5%	55.0%	70.4%	49.7%	24.8%	74.6%
	Average	99.4%	99.2%	63.5%	66.7%	59.7%	89.4%	80.9%	69.6%	87.5%

[a] rm: running meter.

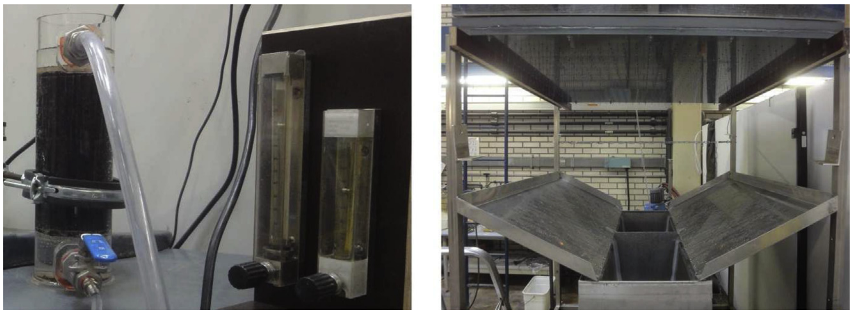


Fig. 3. Experimental setups: FSS model as a column with flow meters (left) and FCS model as two channels (full width) with rain-simulating-device (right).

2.3. Chemicals, and materials

For the heavy metals, copper(II) nitrate trihydrate pure (AppliChem GmbH, Germany) and zinc nitrate hexahydrate pure (AppliChem GmbH, Germany) were used. Deionized water (electric conductivity of <1 µS/cm) was used for preparing the feed water. Feed water pH adjustments and preservation of all samples were performed with nitric acid 65% p.a. (Merck KGaA, Germany). All samples were collected in 250 mL low-density polyethylene bottles (VWR Collection, Germany).

2.4. Analytical procedures

The pH value and electric conductivity were measured immediately after each sample collection using a glass electrode (WTW Sentix 60) according to Standard Method 4500-H^+ and a WTW ProfiLine Cond 1970i (detection limit: 0.1 µS/cm) according to Standard Method 2520 B (APHA, 2005), respectively. Subsequently, all samples were preserved with nitric acid at $pH < 2$.

For Part 1, Cu and Zn concentrations were first measured using cuvette tests (Hach Lange GmbH, Germany) to reduce the duration of the test method. Subsequently, the total concentrations of both heavy metals were measured in all samples by flame atomic absorption spectrometry (AAS, Varian Spectrometer AA-240FS) according to Standard Method 3111 (APHA, 2005) with detection limits of 50 µg/L (Cu) and 20 µg/L (Zn). Cu samples with concentrations below 50 µg/L were repeatedly measured with a graphite furnace AAS (Varian Spectrometer AA-240Z with GTA 120) according to Standard Method 3113 (APHA, 2005) with a detection limit of 5 µg/L. Total metal concentrations were measured with AAS in an aqua regia digestion of selected samples to determine the differences of metal concentrations between acidified ($pH < 2$) and digested effluent samples, and to quantify the metal contents in dried filter material samples in an aqua regia digestion. No differences were determined by both methods for the effluent samples. Quality control was maintained by using control and blank samples, and taking standard solution quantity checks every five samples.

3. Results

3.1. Part 1: preloading

The preloading of each filter system was achieved within 24 h. For FSS5, no preloading was conducted because the estimated service life was 1 year. For most systems, the retention of Cu was higher than the Zn retention. For two FCS, a circulation of the feed water (14.0 mg/L Cu and 121.5 mg/L Zn in 120 L) was not necessary because the mixed effluent concentrations after the first percolating of the feed water (120 L) through the filter media were approximately 0.1–0.6% for Cu and 0.7–3.7% for Zn of the influent concentrations for all experiments (retention >90%, cf. the terminating criteria in Section 2.1.2).

3.2. Part 2: removal efficiencies and long-term performance

The results of Part 2 for the FCS and the FSS, which describe the long-term performances of the filter systems and their heavy metal retention efficiencies in the last year, were more heterogeneous (Table 1). Differences occurred between both heavy metals, the RIs, the system types, and each filter system. The two FCS with only one preloading run had the highest removal efficiencies for both metals and all RI (99.2–99.4%). All other filter elements showed larger differences for all RI between each other and each metal. In general, most of the FSS and FCS showed high performances for removing the dissolved heavy metals. The lower retention of FSS1 and FSS2 might be related to the higher ratio of the connected catchment area to the surface area of the filter but it can also be linked to their unique design (e.g., type of filter media).

Besides the determination of the retention capacities and the removal efficiencies of decentralized systems, removal processes must also be considered. For most systems, the retention of Cu was higher than the retention of Zn. This observation is consistent with their speciation because Cu is less dissolved in the percolating water at pH values higher than 6.5 and Zn at pH values higher than 8.5 (Genç-Fuhrman et al., 2007). The pH of the effluent samples varied between minimum values of 7.1 for FSS3 and 8.2 for FCS2, and maximum values of 11.1 for FCS1 and of 11.4 for FSS2. In general, a high removal efficiency (cf. Table 1) can be achieved at lower pH values with granular ferric hydroxide (adsorption and precipitation) and calcium carbonate (precipitation) and at higher pH values with natural minerals (adsorption, ion exchange, and precipitation). Less removal efficiencies were measured for filters with activated carbons and zeolites.

An increase of the RI led to a decrease of the contact time between the feed water and the filter material. For most of the systems evaluated, this decrease of contact time correlated with a decrease in performance. In addition, the absolute contact times varied between the different systems and for some systems, very short contact times for RI 25 L/(s·ha) (<1 min) led to a significant decrease of their heavy metal removal (e.g., FSS5). Consequently, the performance of the systems correlated with the hydraulic loading rate.

The differences between the performances were also linked to more exhausted capacities of the filter materials because of the preloading and the subsequent loading. These trends of the removal efficiencies during Part 2 led to an increase of the ratio c/c_0 (effluent concentration/influent concentration) for most filter systems (Fig. 4, which does not represent typical breakthrough curves because of the preloading (Part 1) and the varying RIs of Part 2). However, this trend was not detected for the two FCS (Fig. 4a and b). Both heavy metals were continuously retained by these FCS and no significant variations of the pH values were measured. Both FCS treated low amounts of bed volumes (Fig. 4a and b), i.e., the ratio of filter material to water volume treated was the highest. Therefore, the capacities of these filter materials (cf. Section 4.1) were lower than the ones of all other filter materials. FCS2 treated 27% more bed volumes than FCS1, although the removal efficiencies and the amount of treated water were comparable (the amount of water per RI was the same because of the same connected catchment areas of these systems although the volume of the filter media is different in the pilot-scale and the corresponding full-scale FCS because of the design features). Concerning Part 2, all other systems had significantly higher amounts of bed volumes treated. For FSS3 (Fig. 4c) and FSS6 (Fig. 4d), the trend of increasing effluent concentrations was observed for RI 6.0 L/(s·ha) and RI 25 L/(s·ha) because of decreasing contact times and increasing loadings of the filter materials within one RI. The removal efficiencies decreased for FSS3 continuously during RI 25 L/(s·ha), whereas in the experiments with FSS6, the efficiencies and the pH value decreased stepwise for all three RIs. This is also linked to the different capacities and removal mechanisms of the filter materials of FSS3 (sorption and precipitation) and FSS6 (sorption and ion exchange). In addition, the performances of the filter materials were correlated with the pH value of the effluent samples. The pH value decreased with increasing effluent concentrations for all filter systems (Fig. 4c and d). Consequently, the type of the filter material and the amount of the treated bed volumes influenced the retention of Cu and Zn significantly for all systems (heavy metal loading rates).

4. Discussion

4.1. Validation of the test method

The results of the novel test method were compared with field measurements to confirm the applicability of the pilot-scale experiments. The only field measurements available were the heavy metal contents of the filter materials in the full-scale systems FCS1 and FCS2 as a function of depth. The total heavy metal contents of the loaded pilot-scale filter material samples were sometimes lower than the contents of the field samples. This is because of the fact that the systems FCS1 and FCS2 do not have a separate treatment step to retain particulate matter. Thus, the total runoff concentrations were removed by the filter material and were subsequently measured in the field samples. However, the test method only considers dissolved metal concentrations. Consequently, the field measurements have much higher metal contents in the top layer of the filter material because herein most of the solids were retained.

A long-term monitoring was available for the system FCS1 at three different locations after several years of operation (Schriefer, 2013, 2014a, 2014b). One system was built in 2004 in a parking lot of a buyback center in Berlin, Germany (four sample collections after 1 year, 2 years, 5 years, and 9 years). The system was also installed and monitored at two frequently used parking lots of building supplies stores in Bristol, Great Britain (built in 2006; one sample collection after 8 years), and Würzburg, Germany (built in 2005; three sample collections after 2 years, 4 years, and 9 years). The main results of all pilot-scale and field samples are summarized in Table 2. The heavy metal contents of the blank samples (i.e., new and unloaded material) were much lower than the loaded contents of Zn and slightly lower for Cu. Concerning the blank samples, both metals could not be eluted by water within 24 h (<5 μg/L Cu and <10 μg/L Zn) (Schriefer, 2014b). For all samples, the contents of Zn were higher compared with the contents of Cu, and all contents decreased from top to bottom of the three filter layers (i.e., in the flow direction). The contents at the Bristol location were higher than the contents at all other sites because of different site-specific factors that had a significant influence on the total contents (Horstmeyer et al., 2016). At the Bristol site, the contents of the field-samples were recalculated for comparison with the pilot-scale samples (45% for Zn and 30% for Cu, cf. Section 2.1). The fictive average "dissolved" contents, extrapolated for 10 years, are approximately 317 mg/kg Zn and 54 mg/kg Cu (also considering the different blank contents). Therefore, the results of the test method samples (320 mg/kg Zn and 52 mg/kg Cu) were comparable with the field samples at a highly polluted catchment area (Huber et al., 2016a) for the same service life. The contents of both heavy metals at the other two field sampling sites were lower and the comparison of the contents in Würzburg after 4 years and 9 years proved the increase of the heavy metals in the top layer after several years.

The field samples of FCS2 were collected at a monitoring site in Augsburg, Germany, where the system was installed and operated for four years. The ratio of the filter surface area (A_F) to the connected catchment area (A_U) of each test channel varied (Table 3). The catchment area consisted of a road with medium average annual daily traffic of approximately 7000 vehicles per day. The site was characterized by a high annual load of solids (up to 4500 kg/ha)

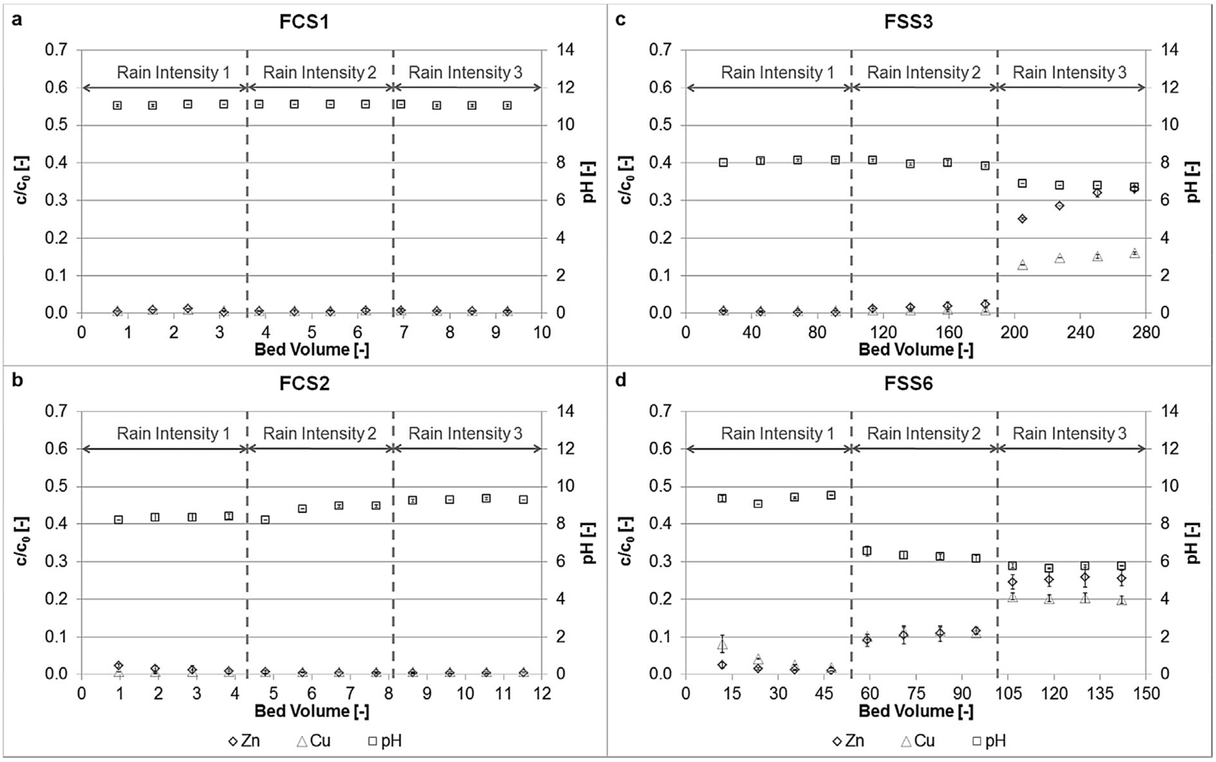


Fig. 4. Effluent concentration trends of both heavy metals (averages and standard deviations of four samples) and the pH value curves of two channel systems (left) and two shaft systems (right) of Part 2.

Table 2
Comparison of the test results of FCS1 (own pilot-scale samples) with field samples from Schriefer (2013; 2014a; 2014b). Heavy metal contents (mg/kg) based on dry matter as a function of depth (three layers).

Type of sample	Pilot-scale samples				Field samples									
	Blank		Loaded		Blank		Berlin		Bristol		Würzburg		Würzburg	
Service life	0 years		10 years		0 years		9 years		8 years		4 years		9 years	
Element	Cu	Zn	Cu	Zn	Cu[a]	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn
0–10 cm	23	51	70	468	5–11	23	39	186	206	1130	19	434	165	610
10–20 cm	23	51	51	332	5–11	23	13	52	44	230	8	51	–	–
20–30 cm	23	51	36	160	5–11	23	12	44	29	129	6	55	–	–
Average	23	51	52	320	5–11	23	21	94	93	496	11	180	–	–

[a] Range of contents.

Table 3
Comparison of the test results of FCS2 with field samples. Heavy metal contents (mg/kg) based on dry matter as a function of depth (six layers).

Type of sample	Pilot-scale samples				Field samples					
	Blank		Loaded		Blank		Augsburg		Augsburg	
Service life	0 years		10 years		0 years		4 years		4 years	
A_F: A_U	–		2.1%		–		4.8%		2.4%	
Element	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn	Cu	Zn
0–3 cm	3.5	5.0	–	–	3.1	6.4	351	1080	379	1270
3–6 cm	3.5	5.0	–	–	3.1	6.4	204	533	453	1060
6–9 cm	3.5	5.0	–	–	3.1	6.4	24.6	149	370	849
9–12 cm	3.5	5.0	–	–	3.1	6.4	26.7	109	229	442
12–15 cm	3.5	5.0	–	–	3.1	6.4	9.8	36.6	41.4	94.1
13–18 cm	3.5	5.0	–	–	3.1	6.4	11.3	52.9	13.9	55.5
Average	3.5	5.0	57	458	3.1	6.4	105	327	248	628

and the presence of a guardrail. All samples had a similar behavior to the samples of FCS1: the contents of Zn were higher than the contents of Cu, the contents decreased in flow direction, and the metal contents of the field samples were higher compared with the pilot-scale samples for comparable ratios of A_F:A_U. This is also linked to the missing mechanical pre-treatment step. Nevertheless, a comparison of the field and pilot-scale results proves the applicability of the test method for this highly polluted site.

In summary, the field sample contents of Cu and Zn varied widely at different locations because of individual site-specific factors. Furthermore, the time of operation and the ratio of A_F to A_U (i.e., the same channel with different catchment areas) differed between several samples. However, as a result of these comparisons, the applicability of the novel test method was verified because the heavy metal contents of the pilot-scale samples were in a realistic range and showed mostly conservative results. Therefore, the test results can be used by stakeholders to manage stormwater treatment challenges.

4.2. Criteria for assessing performance data of treatment systems

The main objective of this paper was the development of a pilot-scale method to determine the service life and the long-term performance of decentralized treatment systems. This method has been tested for nine systems (cf. Table 1). The assessment of the different system performances under these pilot-scale conditions depends on specific boundary conditions of the operation site and is varying because of different requirements in several countries. As a guideline to assess the removal efficiencies determined in Part 2, different criteria concerning heavy metal removal are summarized in the following.

In Germany, the DIBt can approve decentralized systems for groundwater infiltration of runoff from traffic areas after passing several tests. One part of this laboratory test investigates the removal of dissolved heavy metals. The requested elimination is 80% for Cu and 70% for Zn (on average) (DIBt, 2015).

In the State of Washington (USA), the performance goals of stormwater treatment systems have been summarized in the Technical Guidance Manual for Evaluating Emerging Stormwater Treatment Technologies of TAPE (Department of Ecology (2011); Sample et al., 2012). Besides requirements for the “basic treatment”, which include the removal of total suspended solids, “enhanced treatment” facilities must exceed a removal of 30% dissolved Cu and 60% dissolved Zn in a field monitoring.

In Switzerland, requirements are evaluated on the basis of absolute effluent concentrations discharging into receiving water, removal efficiencies, specific surface loadings, and hydraulic performances (Steiner et al., 2010). In addition to other parameters, the total heavy metal concentrations and removal efficiencies of Cu and Zn were considered as indicators for the performance of stormwater treatment systems. Furthermore, potential removal efficiencies of technical treatment systems have been categorized. For the Cu and Zn removal efficiencies, a range of <60% to >90% is postulated by classifying five different levels of treatment quality (Steiner et al., 2010). These categories are used to compare different types of systems after a field monitoring of at least twelve months.

A similar approach is used in Australia (Victorian Stormwater Committee, 2006). For tertiary treatment systems (e.g., filter systems), a range of treatment performances has been defined for dissolved pollutants including Cu and Zn. For a moderate treatment performance, a removal of >40% is postulated.

In summary, the effluent requirements of the different guidelines vary widely and can be directly linked to the results of Part 2 of this pilot-scale test method (cf. Table 1). The requirements represent local regulations, which depend on the receiving water (groundwater or surface water), site-specific factors, and types of connected catchment areas/systems. The guidelines can be used by stormwater managers to classify different decentralized systems.

4.3. Impact of the novel method

Although stormwater runoff waters consist of many different pollutants, the use of the most common dissolved heavy metals ensured that the testing procedure remained relatively straight forward and can be easily performed (Dierkes et al., 2015a). Because field measurements are expensive and take several years, a pilot-scale method can be used to evaluate filter systems within days and should be used before their application.

The results of the novel test method provide regulatory authorities, designers, and operators with a more objective basis for performance assessment. With this method, the estimated service lives of the filter materials of decentralized stormwater treatment systems can be verified or discarded. This is in contrast to theoretical approaches that can only be used to rank different systems without determining removal efficiencies (Scholes et al., 2008). The novel standardized protocol also verifies the long-term performances of the systems, although the runoff water quality and the treatment technologies of decentralized systems are variable (Sample et al., 2012). The efficiencies of the treatment plants, which are measured in Part 2 (cf. Table 1), can be linked to different guidelines/regulations (cf. Section 4.2) and the measured effluent concentrations are also relevant to allow infiltration or discharge to surface water. The results enable stakeholders to identify realistic service intervals of the decentralized stormwater treatment systems independently from the manufacturer's statements. Thus, the costs for maintenance can be better calculated and a reliable comparison of different systems is achieved by these tests. A realistic service life also enables a sustainable use of the filter materials.

In contrast to the novel method, which is based on pilot-scale models of the decentralized stormwater treatment systems, lab-scale models are only suitable to roughly estimate the service lives of these systems. For example, the maximum contents of Cu and Zn for FSS3 determined by batch experiments were 14,400 mg/kg and 24,000 mg/kg, respectively. These contents were several times higher than the ones measured in this study (1100 mg/kg for Cu and 9560 mg/kg for Zn). The results concerning the granular ferric hydroxide of FSS3 are comparable with other filter materials (Huber et al., 2016b). In addition, batch experiments are not suitable to assess the removal efficiencies under flow conditions with appropriate solid-to-solution-ratios (Bürgisser et al., 1993; Poddar et al., 2013). As a benefit, the pilot-scale experiments of this study describe the reality better than lab-scale column experiments (e.g., inner diameter of approximately 20 mm) because the real flow paths and contact times of the full-scale systems can be implemented in the pilot-scale models by using the original grain sizes of the filter media. Furthermore, individual models for each system can also be implemented by pilot-scale models (cf. FCS).

In other studies, permeable pavements were tested with the same rain-simulating-device that was used for the FCS in this study (Fach et al., 2001; DIBt, 2012). Thus, it is theoretically possible to perform tests with this method to determine the service lives of FCS, FSS, and permeable pavements that are used to retain pollutants such as dissolved heavy metals. In addition, further substances such as nutrients can have a crucial impact on the receiving water (Sample et al., 2012). An application of nutrients and further metals is possible after a calculation of the annual loads. For example, this method was successfully expanded to evaluate the simultaneous removal of Cu, Zn, and phosphor by the FSS6 (annual load of phosphor: 101 mg/m^2). Both the service life and the long-term performance of the system were determined for all three substances.

4.4. Restrictions of the novel method

This novel method does not consider all boundary conditions that have an influence on the service life. Service lives that are limited because of a hydraulic failure of the plant (cf. Section 1) cannot be determined by this assessment method. To determine this factor, decentralized systems must be monitored at several sites over periods of some years. In addition, the presence of other dissolved contaminants in stormwater runoff (both further metals and other non-metals), which are currently not implemented into the novel assessment method, may influence the performance and the service life of the filter.

In this study, the effect of de-icing salts on the remobilization of heavy metals was not considered. De-icing salts such as sodium chloride, calcium chloride, and magnesium chloride are mostly used on traffic areas (Bäckström et al., 2004; Nelson et al., 2009; Huber et al., 2015b). For example, sodium chloride can lead to an increase of heavy metal concentrations in the effluent of technical decentralized systems such as constructed wetlands (Tromp et al., 2012). The effect of all three de-icing salts on filter systems will be published elsewhere.

5. Conclusions

Decentralized filter systems are an integral part for the management of urban stormwater runoff. By implementing the assessment method developed in this study, it was possible to characterize the performance of nine different filter systems. Thus, the developed method can be used to determine the filter material service lives and the changes in the long-term performance of the removal efficiencies by the two authoritative parameters Cu and Zn. The estimated service lives were confirmed for all systems. However, the retention efficiencies varied significantly between the nine filter systems because of different filter materials and filter system designs. The removal efficiencies decreased with an increase of the flow rate, the exhaustion of the capacities, the ratio of drainage area to filter material surface area, and the dissolved part of the heavy metals (i.e., a decrease of the pH value). To assess the efficiencies, different guidelines of authorities were summarized to correlate local regulations with the test criteria of this study. Lower performances can be suitable for distinct receiving waters whereas the same efficiencies might not be appropriate for other treatment goals because of local requirements. The validation of the results for two channel systems confirmed the applicability of the novel test method despite different site-specific characteristics. In summary, the method provides regulatory authorities, designers, and operators with a more standardized basis for performance assessment.

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Chapter 8: Research Paper: “De-icing Salts in Roads Maintenance – Occurrence and Impact on Decentralized Systems Treating Traffic Area Runoff for Infiltration”

The use of de-icing salts in road maintenance and their occurrence in runoff from traffic areas must be determined for the implementation of a more standardized method to test the influence of de-icing salts on the remobilization of previously retained heavy metals (cf. Chapter 9). A detailed survey was conducted to collect data about the application of de-icing salts from German authorities that use de-icing salts for winter services on roads, parking lots, and bikeways. Thereby, the used loads of de-icing salts were evaluated for selected German roads and highways. In addition, the total consumptions were summarized for three federal states, also for different categories of traffic areas. In addition, the database of the reviews (cf. Chapter 5) was used to summarize runoff data from Germany, Austria, and Switzerland. The presented runoff concentrations included the de-icing salt compounds Ca, Cl, CN, K, Mg, and Na. The corresponding hypothesis was:

Different types and amounts of de-icing salts are used in road maintenance by German authorities (Hypothesis 6).

The evaluation of the de-icing salt use showed that the amount of salts used per year has increased in the last decades although new and more efficient techniques were implemented. Furthermore, the annual loads of salt per m² highway surface were higher compared with the values for state roads. Moreover, each authority and municipality uses different types of de-icing salts. Sodium chloride is the most commonly used de-icing salt. Mixtures of sodium chloride with calcium chloride or magnesium chloride are also used. In some cases, pure brines of calcium chloride or magnesium chloride are used in Germany. In addition, the runoff concentrations from traffic areas confirmed that the de-icing salts occur in high concentrations (several g/L). Thus, de-icing salts are relevant constituents of runoff from traffic areas in moderate and cold climates with winter services. They can have an effect on decentralized stormwater treatment systems because of their high concentrations, e.g., 50.3 g/L Na (Jenewein & Schinner, 1982), and their behavior (Nelson et al., 2009).

In addition, three different methods (cf. Section 2.2.1, Section 7.1, and Chapter 9) were compared that can be used to evaluate the impact of de-icing salts on decentralized treatment systems for infiltration into the soil and groundwater system.

The following paper was published in gwf Wasser/Abwasser 156, 1138–1152. Most of the runoff data were based on the database, which was created for the reviews (cf. Chapter 5). Huber collected further research data, performed a survey to get more data from German authorities, evaluated all documents, and wrote the manuscript. All authors contributed to the discussions and conclusions.

Auftausalze im Straßenwinterdienst – Aufkommen und Bedeutung für dezentrale Behandlungsanlagen von Verkehrsflächenabflüssen zur Versickerung

Maximilian Huber, Antje Welker, Jörg E. Drewes und Brigitte Helmreich

Regenwasserbewirtschaftung, Auftausalzverbrauch, Labormethoden, Niederschlagswasserbehandlungsanlagen, Verkehrsflächenabflusskonzentrationen

In Deutschland werden Natrium-, Calcium- und Magnesiumchlorid sowie deren Mischungen als Auftausalze im Straßenwinterdienst verwendet. Diese Auftausalze werden mit Ausnahme kleinerer Kommunen als Feuchtsalz ausgebracht und werden meist in den Monaten November bis April in den Verkehrsflächenabflüssen wiedergefunden. Dabei werden vereinzelt mehr als 10 g/L Salz in Verkehrsflächenabflüssen gemessen. Diese im Vergleich zu anderen Schadstoffen deutlich höheren Konzentrationen können die Behandlungsprinzipien dezentraler Anlagen zur Verkehrsflächenabflussbehandlung negativ beeinflussen. Eine Remobilisierung bereits zurückgehaltener Schadstoffe durch die verwendeten Auftausalze ist möglich. Zur Quantifizierung des Remobilisierungsrisikos wurden zwei Labormethoden entwickelt. Anhand dieser Methoden kann die Einhaltung der rechtlichen Anforderungen auch während des Einsatzes von Auftausalzen untersucht werden.

De-icing Salts in Roads Maintenance – Occurrence and Impact on Decentralized Systems Treating Traffic Area Runoff for Infiltration

In Germany, sodium, calcium and magnesium chloride and their mixtures are used as de-icing salts in winter roads maintenance. With the exception of small municipalities, pre-wetting technologies are used for spreading the salt on the roadway and de-icing salts are usually found in the months of November to April in traffic area runoff waters. Salt concentrations higher than 10 g/L are sometimes measured in traffic area runoff. These concentrations are significantly higher compared with other pollutants and may negatively affect the treatment steps of decentralized stormwater treatment systems for traffic area runoff. A remobilization of pollutants by the use of each de-icing salt is possible. To quantify the risk of remobilization, two laboratory-scale test methods were developed. By these methods, the compliance with legal requirements can be examined during the use of de-icing salts.

1. Einleitung

Verkehrsflächenabflüsse können mit einer Vielzahl von Stoffen, wie Schwermetallen, Mineralölkohlenwasserstoffen (MKW), Polycyclischen Aromatischen Kohlenwasserstoffen (PAK), Ethyl-tert-butylether (ETBE), Methyl-tert-butylether (MTBE) sowie Auftausalzen, verunreinigt sein und bedürfen oftmals vor der Einleitung in ein Gewässer einer Behandlung. Unter diesen Verunreinigungen weisen besonders die Bestandteile der Auftausalze starke saisonale Schwankungen auf. Diese sind aufgrund der in den Wintermonaten

(Ende Oktober bis April) deutlich erhöhten Abflusskonzentrationen aufkommensrelevant. Der Verbrauch an Auftausalzen unterliegt dabei örtlichen und jährlichen Schwankungen.

Zur Behandlung von Verkehrsflächenabflüssen wurden in den letzten Jahren zahlreiche dezentrale Anlagen als Alternativen zu zentralen Anlagen, wie Regenklärbecken bzw. Retentionsbodenfiltern, entwickelt, um die Abflüsse von versiegelten Flächen vor Ort zu bewirtschaften. Dabei müssen die dezentralen Anlagen für die anschließende Versickerung weitergehende Anforderungen an den Schadstoffrückhalt berücksichtigen, welche meist durch eine Kombination aus einer mechanisch-physikalischen Behandlung und einem oder mehreren (bio-)chemischen Verfahren erfüllt werden. Die Verfahrenskombinationen der dezentralen Anlagen zur Versickerung müssen dabei die rechtlichen Rahmenbedingungen beachten, welche sich derzeit aus den Prüfwerten der Bundes-Bodenschutz- und Altlastenverordnung (BBodSchV) für den Wirkungspfad Boden-Grundwasser [1] und der Einführung der Verordnung zum Schutz des Grundwassers (GrwV) [2] ergeben. Diese Prüfwerte sind so definiert, dass bei deren Einhaltung trotz einer möglichen Erhöhung der Konzentration im Grundwasser keine relevanten toxikologischen Wirkungen auftreten können. Beispielsweise sind als Parameter nicht nur Schwermetalle aufgeführt, sondern auch Chlorid und Cyanide, welche Bestandteile von Auftausalzen und der darin verwendeten Antibackmittel sind [3].

Dezentrale Anlagen zur Behandlung von Verkehrsflächenabflüssen für die anschließende Versickerung können bundesweit durch das Deutsche Institut für Bautechnik (DIBt) geprüft und zugelassen werden [4]. Wichtigster Punkt der Zulassungsgrundsätze ist die Wirksamkeitsprüfung der Behandlungsanlage unter Laborbedingungen, anhand derer der Rückhalt an Abfiltrierbaren Stoffen (AFS), MKW und Schwermetallen ermittelt wird. Anschließend wird in der Teilprüfung zum Salzeinfluss auf den Zink- und Kupferrückhalt die Beständigkeit der eingesetzten Filtermaterialien gegenüber Natriumchlorid (NaCl) als repräsentativem Auftausalz untersucht. Jedoch ist bei diesem Prüfverfahren derzeit keine detaillierte Methodik zur Ermittlung des Einflusses weiterer Auftausalze auf die Remobilisierung von auf Filtermaterialien zurückgehaltenen Schadstoffen vorhanden. Folglich gibt es aktuell keine dezentrale Behandlungsanlage für Verkehrsflächenabflüsse in Deutschland, bei der die Filterstabilität aller aufkommensrelevanten Auftausalze, welche seit Jahren im differenzierten Winterdienst (Unterscheidung der Wahl des Streustoffes und der ausgebrachten Menge nach Straßenkategorien, deren Trassierung und dem Einsatzfall) verwendet werden [5], nachgewiesen wurde. Daher ist die Entwicklung einer Methodik zur Durchführung weitergehender Untersuchungen für die Bestimmung einer möglichen Remobilisierung von bereits auf Filtermaterialien zurückgehaltenen Schwermetallen durch die auf deutschen Straßen verwendeten Auftausalze notwendig.

In diesem Beitrag wird ausgehend von der Darstellung des Verbrauchs der Auftausalze im Straßenwinterdienst und der in Messprogrammen bestimmten Abflusskonzentrationen der fünf wichtigsten Auftausalzbestandteile (Calcium (Ca), Chlorid (Cl), Kalium (K), Magnesium (Mg) und Natrium (Na)) erläutert, welche Einflüsse Auftausalze auf die Behandlung von Verkehrsflächenabflüssen in dezentralen Anlagen zur anschließenden Versickerung haben. Abschließend werden zwei Methoden zur Bestimmung der Salzstabilität dezentraler Anlagen unter aufkommensrelevanten Auftausalzen im Labor vorgestellt, anhand derer die Wirksamkeit der Filtermaterialien – auch während des Einsatzes von Auftausalzen – nachgewiesen werden kann. Die Verknüpfung dieser Aspekte ermöglicht es, die Einflüsse der Auftausalzverwendung auf die dezentrale Verkehrsflächenabflussbehandlung unter Berücksichtigung rechtlicher Vorgaben zu erfassen.

2. Auftausalze im Straßenwinterdienst

Das in Deutschland auf Straßen am häufigsten verwendete Auftausalz ist NaCl, wobei Auftausalze wie Calciumchlorid ($CaCl_2$) und Magnesiumchlorid ($MgCl_2$) sowie deren Mischungen im Feuchtsalz (FS) seit Jahren zusätzlich im Straßenwinterdienst verwendet werden. Dabei wird zwischen Feuchtsalz, das 30 % Sole (Salzlösung) und 70 % Trockensalz enthält (FS30) und solches, das 100 % Sole (FS100, Flüssigstreuung) enthält, unterschieden [6]. Als Ausgangskonzentrationen der Lösungen werden meist 21–23 Massen (M.)-% NaCl, 20–21 M.-% $CaCl_2$ bzw. 17,5 M.-% $MgCl_2$ verwendet. Diese drei Stoffe werden auch in der aktuell geltenden TL-Streu (Technische Lieferbedingungen für Streustoffe des Straßenwinterdienstes) als Einzige neben den abstumpfenden Streustoffen (Sand, Splitt) aufgeführt [3]. Bei der Wahl der verwendeten Auftausalze spielt sowohl der Preis, die regionale Verfügbarkeit, die Tauwirksamkeit, als auch die Taugeschwindigkeit eine wesentliche Rolle [7]. Dabei werden die $CaCl_2$- und $MgCl_2$-Solen besonders bei Meistereien eingesetzt, die einen hohen Anteil an Steigungsstrecken und gegebenenfalls weitere lokale Besonderheiten, wie Engstellen und Brücken, unterhalten [8]. Weitere Stoffe, wie Formiate, Acetate, Harnstoffe, Alkohole und Glykole, werden derzeit für den Einsatz auf Straßen in Deutschland nicht empfohlen [6, 9], werden aber beispielsweise auf Flughäfen zur Enteisung verwendet.

2.1 Auftausalzverbrauch in Deutschland

In den letzten Jahrzehnten ist trotz der flächendeckenden Einführung des differenzierten Winterdienstes [8] und der zunehmenden Umstellung auf Feuchtsalztechnik FS30 sowie neuerdings auf Flüssigstreuung FS100 [10] der gesamte Auftausalzverbrauch in Deutschland gestiegen. Als repräsen-

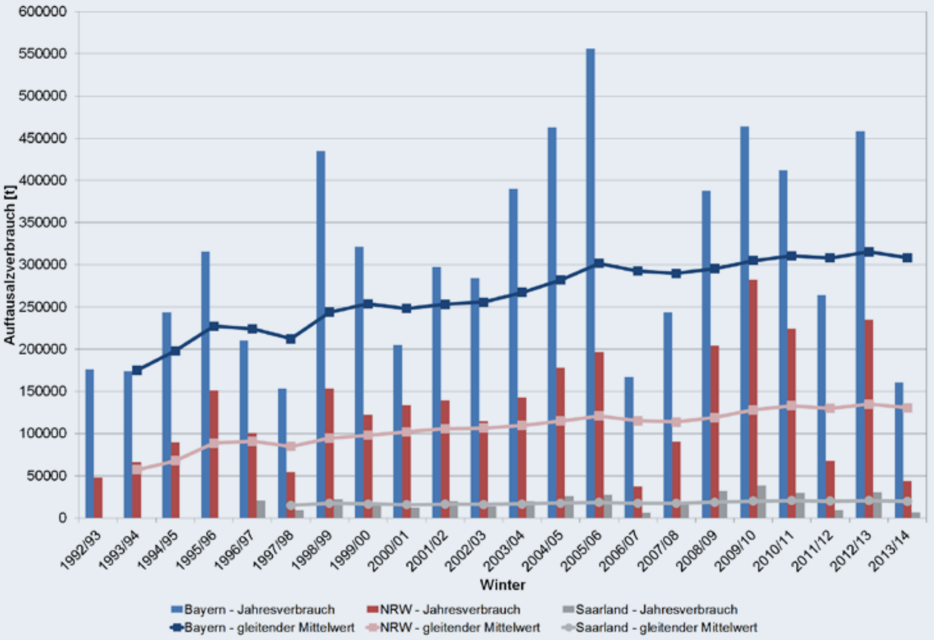

Bild 1: Historischer Verlauf des gesamten Auftausalzverbrauchs in Bayern, NRW und Saarland [11–15]

tative Beispiele ist dies für die drei Bundesländer Bayern, Nordrhein-Westfalen (NRW) und Saarland in **Bild 1** dargestellt, bei denen der Verbrauch tendenziell im Jahresmittel steigt. Zu erkennen sind außerdem die pro Winter stark unterschiedlichen Verbräuche, die beispielsweise in den Wintern 2009/10 aufgrund einer lang anhaltenden Kälte- und Schneefallperiode (Dezember bis April) sowie 2010/11 aufgrund eines strengen Winters mit andauernden Schneefällen (November bis Januar) besonders hoch waren [10].

Betrachtet man den historischen Verlauf des durchschnittlichen Auftausalzverbrauchs pro Kilometer für einzelne Straßenkategorien in Bayern und NRW, so ergibt sich ein differenziertes Bild (**Bild 2**). Zum einen folgt der Verbrauch aller sieben Kategorien in **Bild 2** dem Verlauf der Gesamtverbräuche aus **Bild 1** proportional, sodass die Verbräuche pro Kategorie zwischen den einzelnen Jahren deutlich schwanken. Für Bundesautobahnen liegen die Werte pro Winter zwischen 5,7 t/km und 79,3 t/km, bei Kreisstraßen zwischen 1,3 t/km und 10,8 t/km. Zum anderen sind die durchschnittlichen Verbräuche der Bundesautobahnen deutlich höher als die der anderen Straßenkategorien. Zum Vergleich sind in Bayern die Verbräuche pro Kilometer auf den Autobahnen 3,7-fach höher als auf den Bundesstraßen. In NRW sind die Verbräuche pro Kilometer auf den Autobahnen 4,5-fach höher als auf den Bundes- und Landesstraßen, welche in NRW nicht getrennt erfasst werden. Unter Berücksichtigung der durchschnittlichen Fahrbahnbreite [16] dürften die Verbräuche der Bundesautobahnen aber nur ca. 2,5-fach höher sein, wenn pro m^2 und Jahr die gleiche Menge auf diesen Straßenkategorien verwendet würde. Die erhöhten Verbräuche der Bundesautobahnen stehen auch in Verbindung mit dem Anforderungsniveau an den Winterdienst für den Zeitraum der Befahrbarkeit, welcher auf den Bundesautobahnen 24 Stunden täglich beträgt. Dahingegen erstreckt sich der Zeitraum für wichtige Straßen des überörtlichen Verkehrs nur von 6:00 Uhr bis 22:00 Uhr. Des Weiteren ist der Verbrauch pro Kilometer in Bayern im Vergleich zu NRW aufgrund anderer klimatischer Bedingungen für jede Straßenkategorie höher.

Die Auswertung des Auftausalzverbrauches verschiedener Autobahnmeistereien (AM), kommunaler Meistereien (KM) und Straßenmeistereien (SM) für die Winter 2010/11 bis 2012/13 ist in **Bild 3** dargestellt. Dabei ist ebenfalls die Schwankung des Verbrauchs zwischen den Wintern aufgrund der Darstellung als Box-Whisker-Plots ersichtlich, bei denen das obere bzw. untere Ende der Box dem oberen bzw. unteren Quartil entspricht und der Strich in der Box dem Median-Wert. Bei Meistereien mit wenigen Datensätzen sind die mittleren Verbräuche nur als waagerechter Strich dargestellt. Ebenfalls ist bei den AM der durchschnittliche Verbrauch deutlich höher als bei den anderen beiden Kategorien, wobei bei AM02 der Verbrauch an Auftausalzen aufgrund langer Steigungsstrecken im Mittelgebirge besonders hoch ist. Ferner weist sowohl eine KM als auch eine SM jeweils höhere Durchschnittswerte im Vergleich zu den anderen Meistereien der jeweiligen Kategorie auf. Bei KM08 handelt es sich um eine Großstadt im Süden Deutschlands und bei SM05 um eine Meisterei, die ebenfalls im deutschen Mittelgebirge viele Steigungsstrecken unterhält.

Ein Vergleich der verwendeten Auftausalze in vier deutschen Großstädten ergab, dass sich die verwendete Feuchtsalztechnik von Stadt zu Stadt unterscheidet und es innerhalb der Städte aufgrund des differenzierten Winterdienstes Unterschiede gibt. In Berlin wird FS30 mit NaCl und

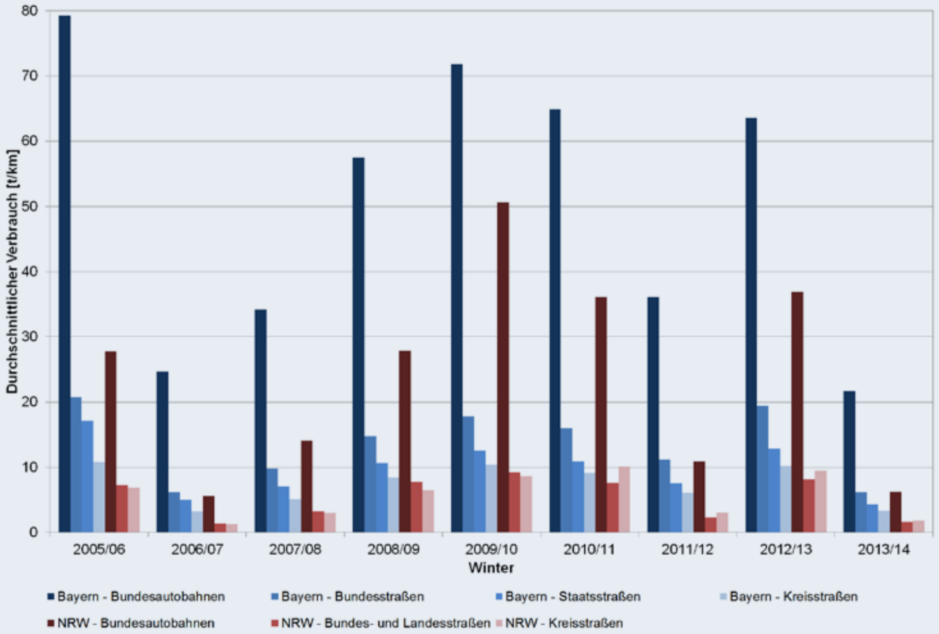

Bild 2: Historischer Verlauf des Auftausalzverbrauchs in Bayern und NRW getrennt nach Straßenkategorien – die Staatsstraßen in Bayern entsprechen den Landesstraßen in NRW [12–14]

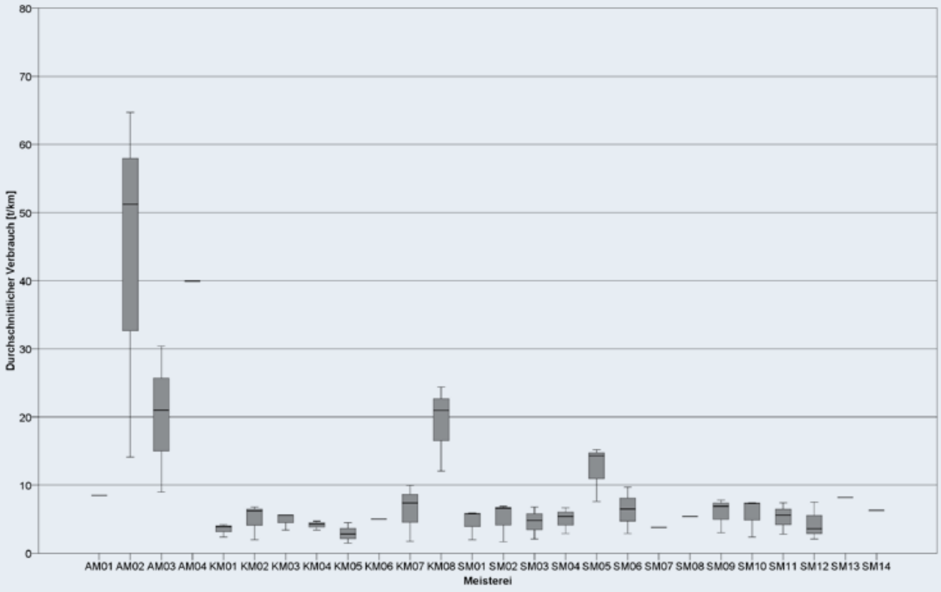

Bild 3: Auftausalzverbrauch in Deutschland von verschiedenen Autobahnmeistereien (AM), kommunalen Meistereien (KM) und Straßenmeistereien (SM) für die Winter 2010/11 bis 2012/13 (Box-Whisker-Plots: oberes bzw. unteres Ende der Box entspricht dem oberen bzw. unteren Quartil und der Strich in der Box dem Median)

$CaCl_2$ als Sole verwendet [17]. Dabei werden Auftausalze im Regelfall nur in der Einsatzstufe E1 (Straßen mit besonderer Verkehrsbedeutung oder öffentlichen Personennahverkehr) sowie auf den Bundesfernstraßen verwendet [18]. In Frankfurt/Main wird vorwiegend NaCl als Feuchtsalz auf den Hauptverkehrs- und Durchgangsstraßen (Kategorie A) sowie auf Zubringer- und Erschließungsstraßen (Kategorie B) eingesetzt, wobei zusätzlich wenige Tonnen pro Jahr $CaCl_2$ auf sensiblen Belägen/Fußgängerbrücken sowie mehrere Hundert Tonnen pro Jahr Splitt auf den restlichen Wohnstraßen der Kategorie C verwendet werden [19, 20]. In Hannover wird auf den Straßen (ca. 420 km) sowie den Rad- und Gehwegen der Stufe 1 FS30 mit NaCl und $MgCl_2$ als Sole verwendet, auf Radwegen testweise auch FS100 mit $MgCl_2$ sowie auf Straßen der Kategorie 2 Splitt [21, 22]. Auf den ca. 2300 Straßenkilometern in München wird auf den ca. 805 km Salzrouten FS30 mit NaCl sowie eine Splitt-Streuung in Sonderfällen im Nebenstraßennetz auf einer Länge von ca. 560 km angewandt [23]. Folglich werden aktuell alle drei Auftausalze in deutschen Großstädten in unterschiedlichen Kombinationen verwendet.

2.2 Konzentrationen an auftausalzbedingten Stoffen in Verkehrsflächenabflüssen

Aufgrund der Unterschiede zwischen den Wintern, den Straßenkategorien sowie den Meistereien sind die Ergebnisse der Messprogramme aus Deutschland, Österreich und der Schweiz sehr unterschiedlich (vgl. **Tabelle 1**). Dabei

Tabelle 1: Übersicht der Messprogramme in Deutschland, Österreich und der Schweiz zur Charakterisierung von Auftausalzbestandteilen in Verkehrsflächenabflüssen; die gesamten Konzentrationen sind in [mg/L] angegeben und in Klammern sind die Mittelwerte (MW), sofern verfügbar, aufgeführt

Land	Ort	Untersuchungszeitraum	DTV	Ereignisanzahl	Winterdienst	Elemente und Konzentrationen	Literatur
AUT	Salzburger Land, A1, Schwarzenbergkaserne	12.2001–04.2003	> 60 000	29	Ja	5,4–160 Ca (37,0 MW) 1,4–5200 Cl (646 MW) 0,7–15 K (3,7 MW) < 0,1–8,3 Mg (2,4 MW) 2,4–3400 Na (440 MW)	[44]
AUT	Salzburger Land, A1, Baulos West	12.2001–04.2003	> 60 000	29	Ja	15–240 Ca (71,5 MW) 1,7–2600 Cl (582 MW) 1,6–13 K (5,6 MW) < 0,1–14 Mg (4,6 MW) 5,9–1500 Na (410 MW)	[44]
AUT	Salzburger Land, A1, Messe	12.2001–04.2003	> 60 000	29	Ja	13–350 Ca (55,0 MW) 1,3–7900 Cl (1255 MW) 0,1–29 K (8,1 MW) < 0,1–10 Mg (2,5 MW) 5,4–5700 Na (984 MW)	[44]
AUT	Tirol, A13, Steigungsabschnitt	–	–	–	–	1–60300 Cl (7537 MW) 0,3–193 K (26,6 MW) 1,1–50250 Na (5467 MW)	[45]
AUT	Tirol, A13, Mautstelle	–	–	–	–	1–7816 Cl (1378 MW) 0,9–18,0 K (6,3 MW) 2,4–6064 Na (1016 MW)	[45]
AUT	Tirol, Stadtstraße	–	–	–	–	1–485 Cl (86,0 MW) 0,8–50,5 K (7,5 MW) 1,5–348 Na (82,1 MW)	[45]
CHE	Bern, Burgdorf, Kirchbergstrasse	23.10.2002–08.07.2004	17 000	7	–	Ca (34,9 MW) Ca (21,5 MW – gelöst)	[46]
CHE	Graubünden, Klosters-Serneus, N28a, Klosters	11.04.2006–09.08.2006	–	2	–	15,0–746 Cl	[47]
CHE	Zürich, Birmensdorf, Waldeggstrasse, Ristet	16.01.2008–03.07.2008	12 000	3	–	15,3–2350 Cl (815 MW)	[48]
CHE	Zürich, Rossberg, N1	01.05.1976–30.04.1977	72 000	–	Ja	Cl (159 MW)	[42]
DEU	Baden-Württemberg, Heilbronn-Obereisesheim, A6	02.1979–07.1979	47 000	48	Ja	Cl (152 MW)	[49]
DEU	Baden-Württemberg, Pleidesheim, A81	02.1978–09.1978	41 000	53	Ja	Cl (108 MW)	[49]
DEU	Baden-Württemberg, Ulm/West, A8/B10	05.1980–03.1981	52 100	44	Ja	Cl (357 MW)	[49]
DEU	Baden-Württemberg, Weinsberg, A6	04.1990–04.1991	34 680	–	Ja	Ca (115 MW) Ca Sommer (17 MW) Ca Winter (205 MW) Cl (2061 MW) Cl Sommer (36 MW) Cl Winter (3921 MW) Mg (3,5 MW) Mg Sommer (2,1 MW) Mg Winter (4,8 MW)	[50]
DEU	Bayern, Augsburg, Derchinger Straße	01.10.1996–08.09.2005	6800	36	Ja	14–123 Ca (44 MW) 1,0–4100 Cl (584 MW) 1,8–2840 Na (380 MW)	[51]
DEU	Bayern, Bayreuth, B22, Königsallee	14.07.1991–17.08.1991	16 000	3	–	1–56 Cl	[52]
DEU	Bayern, Bayreuth, Meranierring	13.06.1990–06.08.1990	6000	3	–	0,2–4,5 Cl	[52]
DEU	Bayern, München, Landshuter Allee	11.2003–11.2005	57 000	63	Ja	17,6–10 400 Na	[53]
DEU	Bayern, München, Landshuter Allee	01.07.2006–30.06.2007	57 000	24	Ja	< 25–2480 Na	[54]

▶▶

Land	Ort	Untersuchungszeitraum	DTV	Ereignis-anzahl	Winter-dienst	Elemente und Konzentrationen	Literatur
DEU	Berlin, A113	05.11.2006–16.08.2007	140 000	20	–	14–175 Ca (34 MW) 3–1110 Cl (117 MW) 5–568 Na (62 MW)	[55]
DEU	Hamburg, Hamburg-Billbrock, Pinkertweg	1986	2300	16	–	Ca (15,1 MW) Cl (10,7 MW) K (1,7 MW) Mg (1,4 MW) Na (5,0 MW)	[43]
DEU	Hamburg, Hamburg-Billbrock, Pinkertweg	1987	2300	16	–	Ca (13,7 MW) Cl (4,1 MW) K (1,7 MW) Mg (1,0 MW) Na (21,3 MW)	[43]
DEU	Hamburg, Jungiusstraße	1987	16 200	12	–	Ca (14,5 MW) Cl (3,9 MW) K (1,9 MW) Mg (1,0 MW) Na (3,0 MW)	[43]
DEU	Hamburg, Redder	1987	500	17	–	Ca (11,7 MW) Cl (4,5 MW) K (2,6 MW) Mg (1,3 MW) Na (2,6 MW)	[43]
DEU	Hamburg, Halenreie	09.2008–07.2010	15 000	8	Ja	9,7–1270 Cl (25,2 MW)	[56]
DEU	Hessen, Frankfurt/Main, A3	09.1975–10.1977; 08.1978–08.1980	62 790–90 000	> 6	Ja	Ca (40 MW) Cl (74 MW) K (4,6 MW) Mg (1,8 MW) Na (64 MW)	[57]
DEU	Hessen, Gießen, A485	26.08.2014–27.08.2014	ca. 40 000	1	Nein	5,8–8,7 Ca 1,1–2,6 K 1,4–2,1 Mg 11,5–42,2 Na	*
DEU	Niedersachsen, Allertal West, A7	01.2001–12.2001	Parkplatz	42	Ja	1–2471 Cl (120 MW)	[58]
DEU	Niedersachsen, Brake, L875	27.04.2013–31.08.2013	6000–7000	2	–	23,0–35,4 Ca 26,7–55,8 Na	*
DEU	Niedersachsen, Garbsen Nord Z1, A2	01.2001–12.2001	Parkplatz	40	Ja	2–2449 Cl (157 MW)	[58]
DEU	Niedersachsen, Garbsen Nord Z2, A2	01.2001–12.2001	Parkplatz	43	Ja	2–7580 Cl (604 MW)	[58]
DEU	Niedersachsen, Hannover, B3	10.2000–09.2001	45 000	–	Ja	Cl (168 MW)	[59]
DEU	Niedersachsen, Hannover, B3	09.02.2007–07.01.2008	45 000	10	–	31–88 Ca 10–1320 Cl (186 MW) 12–850 Na	[55]
DEU	Niedersachsen, Hildesheim, Marienburger Str., P5	09.1984–08.1985	14 200	31	-	Cl (80,2 MW)	[60]
DEU	Nordrhein-Westfalen, Bensberg, A4	2005–2006	71 220	76	Ja	1,6–185 Ca (45,9 MW) 0,5–4760 Na (258 MW)	[61]
DEU	Nordrhein-Westfalen, Bensberg, A4	2006–2007	71 220	6	Ja	15,5–113 Ca (67,4 MW) 0,5–365 Na (154 MW)	[62]
DEU	Nordrhein-Westfalen, Köln-Ost, A3	11.08.2006–24.08.2007	156 000	20	–	8–46 Ca (20 MW) 3–829 Cl (93 MW) 7–456 Na (59 MW)	[55]
DEU	Nordrhein-Westfalen, Köln-Poll, A4	12.07.1998–22.01.2000	> 50 000	33	–	1–6600 Cl (501 MW) 1–170 Cl Sommer (22,0 MW) 13–6600 Cl Winter (1372 MW)	[63]

Land	Ort	Untersuchungszeitraum	DTV	Ereignisanzahl	Winterdienst	Elemente und Konzentrationen	Literatur
DEU	Nordrhein-Westfalen, Köln-Porz, A59	12.07.1998–05.07.2000	> 50 000	29	Ja	1,0–9300 Cl (402 MW) 1,0–5,0 Cl Sommer (2,5 MW) 3,0–9300 Cl Winter (748 MW)	[63]
DEU	Nordrhein-Westfalen, Meckenheim, A61	2005–2006	73 310	63	Ja	1,8–603 Ca (79,4 MW) 0,7–5574 Na (812 MW)	[61]
DEU	Nordrhein-Westfalen, Meckenheim, A61	2006–2007	73 310	25	Ja	4,7–53,8 Ca (12,9 MW) 1,0–66 Na (13 MW)	[62]
DEU	Nordrhein-Westfalen, Widdig, A555	2005–2006	69 370	65	Ja	4,3–83,4 Ca (25,1 MW) 1,5–5160 Na (691 MW)	[61]
DEU	Nordrhein-Westfalen, Widdig, A555	2006–2007	69 370	23	Ja	3,4–31,0 Ca (12,5 MW) 0,5–189 Na (58 MW)	[62]
DEU	Nordrhein-Westfalen, Wuppertal, L417	14.10.2014–07.11.2014	20 670	4	Nein	16,3–33,3 Ca 1,3–2,1 K 4,0–6,3 Mg 3,6–12,0 Na	[26]
DEU	Nordrhein-Westfalen, Wuppertal, L417	10.12.2014–11.12.2014	20 670	2	Ja (NaCl)	18,6–34,2 Ca 0,9–1,6 K 3,2–4,5 Mg 63,0–387 Na	[26]

* eigene unveröffentlichte Untersuchungen

decken einige Messergebnisse nur Teile der Jahreszeiten ab und die Konzentrationen werden je nach Studie als Bereiche, Mittelwerte oder Mediane angegeben. Wegen fehlender Angaben, ob Straßenwinterdienst mit Auftausalzen erfolgte bzw. welche Auftausalzkombination verwendet wurde, sowie der oftmals fehlenden Unterscheidung zwischen Sommer- und Wintermonaten ist eine statistische Auswertung der Daten nicht möglich. Die in den drei Ländern maximal gemessenen Konzentrationen betragen 603 mg/L Ca, 60 300 mg/L Cl, 193 mg/L K, 14 mg/L Mg und 50 250 mg/L Na. Somit wurden Salzkonzentrationen größer 10 000 mg/L in den Abflüssen gemessen. Es ist zu beachten, dass für Kalium (n = 14) und Magnesium (n = 12) den Autoren nur wenige Datensätze zur Verfügung stehen.

Für die Messungen, die Werte für den Sommerzeitraum angegeben haben bzw. die außerhalb des Einsatzes von Auftausalzen durchgeführt wurden, wurden maximale Konzentrationen von 33,3 mg/L Ca, 170 mg/L Cl, 2,6 mg/L K, 6,3 mg/L Mg und 42,2 mg/L Na ermittelt.

Eine beispielhafte Berechnung der Salzabflusskonzentrationen aus den aufgebrachten Salzmengen ist in [24] angegeben. Ausgehend von einer Autobahn mit einer befestigten Fahrbahnbreite von 20 m und der Ausbringung von insgesamt 2000 kg/km NaCl innerhalb eines abflussunwirksamen Zeitraums (z. B. vier Streueinsätze mit je 25 g/m^2) und einer anschließenden Schmelzwasserabflusshöhe von 10 mm sind im Mittel ca. 10 g/L NaCl, also ca. 4000 mg/L Na und ca. 6000 mg/L Cl, im Verkehrsflächenabfluss rechnerisch zu finden. Durch Variation der Ausgangsdaten in Kombination mit den Daten der **Bilder 2 und 3** können weitere Konzentrationen für Strecken berechnet werden, für die keine Abflussmessungen vorliegen. Mit diesem Ansatz können auch reale Abflussereignisse nachgerechnet werden. Als grobe Näherungen können für die Straßenbreiten die Regelquerschnitte RQ 29,5 (Autobahnen, Breite der befestigten Fläche beträgt 2 x 11,5 m) bzw. RQ 10,5 (Bundesstraßen, Breite der befestigten Fläche beträgt 7,5 m) zugrunde gelegt werden. Für eine Bundesstraße mit einem Auftausalzverbrauch von 113 kg/km NaCl (z. B. ein Streueinsatz mit 15 g/m^2 oder ca. 1/65 der im Durchschnitt bei den ausgewerteten SM im Jahr ausgebrachten Mengen) und einer anschließenden Schmelzwasserabflusshöhe von 30 mm, sind im Mittel ca. 0,5 g/L NaCl, also ca. 200 mg/L Na und ca. 300 mg/L Cl, im Verkehrsflächenabfluss zu finden. Eine Berücksichtigung von Verwehungen und Spritzwasserverlusten (sowohl des Feuchtsalzes als auch des Niederschlags/Schmelzwassers, vgl. auch [16]) kann im Einzelfall in Betracht gezogen werden.

3. Prinzipien dezentraler Anlagen zur Behandlung von Verkehrsflächenabflüssen

Derzeit ist eine Vielzahl dezentraler Behandlungsanlagen für Verkehrsflächenabflüsse auf dem deutschen Markt verfügbar, die für unterschiedliche Einsatzorte und angeschlossene Flächen konzipiert sind sowie variierende Reinigungsziele verfolgen. Eine Einteilung der Systeme sowie deren technischer Ausführungen und Behandlungsprinzipien ist schwierig, da sich die Produkte in vielen Aspekten erheblich unterscheiden. Jedoch können die Anlagen grob nach der Einbauweise (oberflächennah, z. B. Rinnen, versus oberflächenfern, z. B. Schächte) und dem Reinigungsprinzip

(z. B. Siebung, Leichtstoffabscheidung, Sedimentation und Ionenaustausch) unterschieden bzw. klassifiziert werden [25].

In den dezentralen Anlagen werden die Behandlungsprinzipien der Dichtetrennung, der Filtration, der Sorption, der (bio-)chemischen Umsetzung, der Fällung und des Ionenaustausches entweder alleine oder in Kombination verwendet, wobei jedes Prinzip nicht zum Rückhalt aller Schadstoffe geeignet ist. Bei den einstufigen Anlagen werden die Schadstoffe meist rein mechanisch-physikalisch (z. B. durch Sedimentation) zurückgehalten, wohingegen bei den Anlagen, die aus einer Kombination mehrerer Behandlungsprinzipien bestehen, vorwiegend nach der mechanisch-physikalischen Behandlung eine oder mehrere (bio-)chemische Verfahren nachgeschaltet sind (z. B. Ionenaustausch und Fällung im Filter).

Beim Einsatz von Auftausalzen verändert sich die Zusammensetzung und Dichte des zu behandelnden Niederschlagswassers von Verkehrsflächen. Die Erhöhung der Dichte durch das Auftausalz hat zur Folge, dass u. a. die Sedimentation der (Fein-)Partikel erschwert wird [26] und dadurch der mechanisch-physikalische Rückhalt in dezentralen Anlagen reduziert werden kann. Dabei wird nicht nur der Rückhalt an AFS, sondern damit verbunden auch der von partikulär gebundenen Schwermetallen verringert. Zusätzlich haben in diesen Fällen die Kationen der Auftausalze (Natrium, Calcium und Magnesium), welche in deutlich höheren Konzentrationen als die Schwermetalle vorkommen (Faktor ca. 1:1000), einen Einfluss auf die chemischen Verfahren zum Schadstoffrückhalt. Zum einen können bei Filtermaterialien aus Ionenaustauschern, bei denen durch elektrostatische Kräfte Kationen an negativ geladenen Oberflächen zurückgehalten werden, bereits aus vorhergehenden Regenereignissen zurückgehaltene Schwermetalle durch die Auftausalzbestandteile ausgetauscht und somit wieder in Lösung gebracht werden [27]. Bei der Remobilisierung ist eine Rücklösung von Schwermetallen, die bereits seit Monaten oder Jahren gebunden wurden, möglich, wodurch die Ablaufkonzentrationen der dezentralen Anlagen für einzelne Schwermetalle höher als die Zulaufkonzentrationen sein können [28]. Zum anderen können auch aufgrund schwacher Van-der-Waals-Kräfte (Physisorption) an der Materialoberfläche zurückgehaltene Stoffe remobilisiert werden. Ferner können freie Plätze durch Auftausalzkationen belegt werden, welche somit nicht mehr bzw. erschwert dem Schadstoffrückhalt zur Verfügung stehen. Durch Reaktionen der Auftausalzbestandteile mit den Filtermaterialien und anderen Schadstoffen können sowohl während des Regenereignisses als auch während der Trockenperioden bei dauereingestauten Systemen weitere Rücklösungen erfolgen [29]. Durch Ausbildung eines chemischen Gleichgewichts kann sich der pH-Wert des zu behandelnden Wassers verringern [30], sodass einzelne, bereits gefällte Schwermetalle wieder in Lösung gehen und somit aus dem System ausgetragen werden können. Folglich ist es unerlässlich, dass die eingesetzten dezentralen Anlagen ihre Leistungsfähigkeit auch während der Wintermonate mit Auftausalzeinsatz sicherstellen und dabei die Anforderungen, u. a. zum Rückhalt von Schwermetallen, einhalten.

Derzeit ist noch nicht geklärt, wie sich die unterschiedlich hohen Konzentrationen an Calcium, Kalium, Magnesium und Natrium an verschiedenen Orten – auch ohne den Einsatz von Auftausalzen – auf den Schadstoffrückhalt eines Behandlungssystems auswirken.

Ein Rückhalt von Chlorid ist bei keiner dezentralen Anlage vorgesehen, sodass die in den Verkehrsflächenabflüssen gemessenen Chlorid-Konzentrationen von bis zu 60 300 mg/L ebenfalls im Ablauf der Behandlungsanlagen zu finden sind.

4. Rechtliche Rahmenbedingungen für dezentrale Anlagen zur Versickerung

Als Vorgaben zur Bewertung der Leistungsfähigkeit dezentraler Behandlungsanlagen wurden bislang die Prüfwerte der BBodSchV vom 12.07.1999 für den Wirkungspfad Boden-Grundwasser zugrunde gelegt [1]. Mit Einführung der GrwV vom 09.11.2010 [2] sind Einträge bestimmter Schadstoffe und Schadstoffgruppen, darunter Kupfer und Zink, durch geeignete Maßnahmen(-programme) zu begrenzen. Bei der Festlegung der Maßnahmen(-programme) sind, so die Aussage vom Umweltbundesamt und der Länderarbeitsgemeinschaft Wasser (LAWA), die Länder zum Schutz des Grundwassers vor Verschmutzung und Verschlechterung an die Geringfügigkeitsschwellenwerte der LAWA von 2004 gebunden. Geringfügigkeitsschwellenwerte sind als Konzentrationen definiert, bei denen trotz einer Erhöhung der Konzentration im Grundwasser keine relevanten toxikologischen Wirkungen auftreten können. Zusätzlich werden die Anforderungen der Trinkwasserverordnung, wie z. B. für die Parameter Chlorid und Sulfat, berücksichtigt, welche sowohl gesundheitliche als auch ästhetische Aspekte berücksichtigen [31]. Eine Aktualisierung der Ableitung von Geringfügigkeitsschwellenwerten für das Grundwasser von 2004 erfolgte in den Jahren 2012/2013 aufgrund rechtlicher Änderungen in der Europäischen Union und neuen fachlichen Erkenntnissen [32].

Die Mantel-Verordnung („Verordnung zur Festlegung von Anforderungen für das Einbringen oder das Einleiten von Stoffen in das Grundwasser, an den Einbau von Ersatzstoffen und für die Verwendung von Boden und bodenähnlichem Material“, derzeit Entwurf vom 31.10.2012) [33] soll die GrwV und die BBodSchV harmonisieren und die Verbindlichkeit der Geringfügigkeitsschwellenwerte weiter untermauern. **Tabelle 2** stellt die Anforderungen für ausgewählte Parameter gegenüber. Eine Verschärfung der

Tabelle 2: Auswahl der Prüfwerte der BBodSchV für den Wirkungspfad Boden-Grundwasser, der Prüfwerte Grundwasser des Entwurfs der Mantel-Verordnung vom 31.10.2012 und der Geringfügigkeitsschwellenwerte der LAWA aus den Jahren 2004 und 2013. Verschärfungen der Anforderungen gegenüber der bisher gültigen BBodSchV sind fett markiert

Parameter	Einheit	BBodSchV, 1999	Geringfügigkeitsschwellenwerte, 2004	Entwurf Mantel-Verordnung, 2012	Geringfügigkeitsschwellenwerte, 2013
Antimon	µg/L	10	**5**	**5**	–
Arsen	µg/L	10	10	10	**2,6**
Barium	µg/L	–	**340**	**340**	**186**
Blei	µg/L	25	**7**	**7**	**7,2**
Bor	µg/L	–	**740**	**740**	**180**
Cadmium	µg/L	5	**0,5**	**0,25**	**0,25**
Chrom, gesamt	µg/L	50	**7**	**7**	**3,4**
Chromat	µg/L	8	–	–	–
Kobalt	µg/L	50	**8**	**8**	**5,7**
Kupfer	µg/L	50	**14**	**14**	**10**
Molybdän	µg/L	50	**35**	**35**	–
Nickel	µg/L	50	**14**	**20**	**13**
Quecksilber	µg/L	1	**0,2**	**0,05**	**0,05**
Selen	µg/L	10	**7**	**7**	**3**
Thallium	µg/L	–	**0,8**	**0,8**	**0,2**
Vanadium	µg/L	–	**4**	**4**	–
Zink	µg/L	500	**58**	**58**	**50**
Zinn	µg/L	40	–	–	–
Ammonium	mg/L	–	–	**0,5**	–
Chlorid	mg/L	–	**250**	**250**	–
Cyanid, gesamt	µg/L	50	–	–	–
Cyanid, leicht freisetzbar	µg/L	10	**5***	**5***	10*
Fluorid	µg/L	750	750	750	–
Nitrat	mg/L	–	–	**50**	–
Sulfat	mg/L	–	**240**	**250**	–
ΣPAK	µg/L	0,2	0,2	0,2	–
ETBE	µg/L	–	–	–	**5**
MTBE	µg/L	–	**15**	**15**	**2,5**

* 50, wenn kein leicht freisetzbares Cyanid vorliegt

Anforderungen für sämtliche Behandlungstechniken von Verkehrsflächenabflüssen zur anschließenden Versickerung ist somit bereits formaljuristisch gegeben (betroffene Parameter sind in **Tabelle 2** fett markiert).

Weiterhin ist für die Anforderungen an dezentrale Behandlungstechniken zu diskutieren, inwiefern das Hinzukommen neuer Parameter (z. B. Chlorid, ETBE und MTBE) aufgrund der Relevanz in Verkehrsflächenabflüssen berücksichtigt werden muss.

Zu den für Verkehrsflächenabflüsse aufkommensrelevanten Parametern aus **Tabelle 2** gehören die drei Schwermetalle Blei, Kupfer und Zink sowie die Auftausalzbestandteile Chlorid und Cyanid [34, 35]. Ferner können in Verkehrsflächenabflüssen die anorganischen Parameter Cadmium, Chrom, Nickel und Quecksilber in Konzentrationen oberhalb des Entwurfs der Mantel-Verordnung vorkommen [35].

Bei der aktuellen Verschärfung der Anforderungen sind besonders die Werte für Cadmium, Kupfer, Zink und Chlorid für die bestehenden Behandlungstechniken relevant. Dabei ist Cadmium ein sehr mobiles Element, welches sich in den Behandlungsanlagen durch die Filtermaterialien verlagern kann. Ein Durchsickern aus den Verkehrsflächenabflüssen in das Grundwasser ist somit möglich. Außerdem ist zu beachten, dass viele der derzeitigen Messprogramme eine Bestimmungsgrenze von 0,5 µg/L für Cadmium haben [36] und damit eine gesicherte Aussage zur ausreichenden Behandlungsfähigkeit von Cadmium in dezentralen Behandlungsanlagen, aber auch in zentralen Anlagen und

Oberböden, fehlt. Ferner führen die Verschärfungen der Prüfwerte für Kupfer und Zink dazu, dass an hochbelasteten Stellen (z. B. Stop+Go-Bereiche) ein zu hoher Austrag der Schwermetalle in das Grundwasser erfolgen kann. Die Aufnahme von Chlorid mit einer Konzentration von 250 mg/L führt dazu, dass nun neben Cyanid, welches in Verkehrsflächenabflüssen in Gesamt-Konzentrationen von bis zu 190 µg/L vorkommt [37], der Hauptbestandteil der aufkommensrelevanten Auftausalze bei der Versickerung direkt beachtet werden muss. Gleichzeitig müssen die Kationen der Auftausalze in Verbindung mit den strengeren Anforderungen an den Schwermetallrückhalt sowie den möglichen Schwermetallremobilisierungen aufgrund der verwendeten Behandlungsprinzipien stärker bei der Filtermaterialwahl berücksichtigt werden.

5. Methoden zur Bestimmung der Salzstabilität dezentraler Anlagen

Zur Sicherstellung der Einhaltung der Anforderungen (siehe Kapitel 4) durch dezentrale Anlagen sind aufgrund der Behandlungsprinzipen und den damit verbundenen Risiken einer Schwermetallremobilisierung (siehe Kapitel 3) Methoden zur Bestimmung der Salzstabilität der verwendeten Filtermaterialien unerlässlich.

Die ersten Prüfverfahren zur Salzstabilität in Deutschland wurden mit NaCl konzipiert. Dies fand auch Eingang in die Zulassungsgrundsätze des DIBt (siehe Kapitel 5.1). Durch den Einsatz von alternativen Verbindungen wurden zwei Methoden entwickelt, um auch die Auswirkungen von weiteren Auftausalzen ($CaCl_2$ und $MgCl_2$) zu untersuchen (siehe Kapitel 5.2 und 5.3). Die Versuche werden i. d. R. an verkleinerten Filterelementen (Modelle) im Labormaßstab durchgeführt.

Als Grundlage für die in den Versuchen verwendeten Konzentrationen an Auftausalzen dienten sowohl die Empfehlungen und Merkblätter zum Winterdienst [3, 6, 9, 10], welche durch eine Umfrage bei zahlreichen Meistereien eruiert wurden, als auch die gemessenen Verkehrsflächenabflusskonzentrationen sowie die aus den Auftausalzverbräuchen rechnerisch ermittelten Konzentrationen. Davon ausgehend wurde der Fokus auf das in Deutschland auf Straßen am häufigsten verwendete Auftausalz NaCl sowie auf die beiden FS30-Kombinationen aus NaCl mit $MgCl_2$ bzw. NaCl mit $CaCl_2$, welche z. B. in Hannover und Berlin verwendet werden (siehe Kapitel 2.1), gelegt. Die Konzentration an NaCl wurde aus Auftausalzverbräuchen rechnerisch zu 10 g/L ermittelt (siehe Kapitel 2.2) und anhand gemessener Konzentrationen plausibilisiert (siehe **Tabelle 1**). Da für Magnesium wenige Messergebnisse vorliegen und kein Monitoringstandort bekannt ist, an dem $MgCl_2$ als Auftausalz ausgebracht wurde, konnten dessen Konzentrationen nur theoretisch berechnet werden. Unter der Annahme, dass die Verhältnisse aus NaCl und $MgCl_2$ im FS30 gleich dem im Verkehrsflächenabfluss sind, werden ausgehend von den 10 g/L an NaCl (70 %) bei einer $MgCl_2$-Sole (30 %), die mit 26,3 M.-% $MgCl_2$ einen um 50 % höher als optimal empfohlenen Salzanteil enthält (Worst-Case-Betrachtung, z. B. generell höherer Anteil (21 % statt 17,5 %) und Fehler bei der Herstellung der Sole), ca. 1,1 g/L $MgCl_2$ abflusswirksam. Für Calcium ist die Abschätzung einer effektiven Verkehrsflächenabfluss- und somit Anlagenzulaufkonzentration schwieriger, da Calcium, welches für die Remobilisierungsbetrachtung im Gegensatz zum Chlorid relevant ist (siehe Kapitel 3), sowohl aus dem Auftausalz als auch aus erhöhtem Fahrbahnabrieb stammen kann. Da die Umstellung von Trockensalz auf FS30 schrittweise erfolgte und in kleineren Meistereien immer noch nicht abgeschlossen ist sowie aufgrund fehlender Beschreibungen der Randbedingungen in Messprogrammen, können auch für Calcium die gemessenen Abflusskonzentrationen nicht direkt mit verschiedenen Feuchtsalztechniken und dem Straßenabrieb korreliert werden. Daher wurde in einem Worst-Case-Szenario die höchste im Mittel eines Regenereignisses gemessene Konzentration von 603 mg/L Ca herangezogen und ebenfalls um 50 % erhöht (905 mg/L Ca bzw. 2,5 g/L $CaCl_2$). Damit soll sichergestellt werden, dass sowohl der Anteil des Calciums aus dem Auftausalz (analog zur Herangehensweise beim $MgCl_2$ wären dies ca. 1,4 g/L $CaCl_2$) als auch der erhöhte Fahrbahnabrieb berücksichtigt werden.

5.1 Salzprüfung beim DIBt

Die bundesweite Zulassung von dezentralen Niederschlagwasserbehandlungsanlagen deckt den Anwendungsfall „Einleitung Verkehrsflächenabflüsse in das Boden-Grundwasser-System“ ab [4]. Durch die Anforderungen an den Rückhalt gelöster Schadstoffe (z. B. Kupfer und Zink) werden beim DIBt vorwiegend Anlagen mit einer Filterstufe zugelassen. Das Ausmaß einer möglichen Rücklösung bereits zurückgehaltener Schwermetalle wird unter den Randbedingungen der Salzprüfung getestet. Ein weiterer Effekt, der im Winterbetrieb auftreten kann, ist die mögliche Veränderung der Filterstabilität. Unter hohem Salzeinfluss können in Filterstufen Kolmationseffekte auftreten [38]. Auch dieser Einfluss wird beim Prüfverfahren des DIBt bei der Vorlage von kolmationsfördernden Randbedingungen (z. B. Ton/Schluff-Anteil > 5 %) berücksichtigt.

In der Salzprüfung des DIBt wird NaCl verwendet. Die Prüfung wird an einem verkleinerten Filterelement (z. B. Säule) oder an der Originalanlage durchgeführt. Die Randbedingungen des Maßstabs der Prüfanlage orientieren sich dabei an der Vorgabe, möglichst geringe Mengen an belastetem Prüfwasser zu erzeugen. Dazu legt der Hersteller einen zusammen mit der benannten Prüfstelle konzipierten Versuchsplan vor, der vom DIBt geprüft wird. Die Salzprüfung erfolgt im Anschluss an die Prüfung auf den Rückhalt an gelösten Kupfer- und Zinkverbindungen (Schwermetallprüfung). In der Schwermetallprüfung wird eine Stoffjahresfracht auf das Filterelement in drei charakteristischen Prüfregenspenden aufgebracht. Kupfer und Zink werden in gelöster Form in entionisiertem Wasser (pH 4,5–5,2) appliziert. Die Prüfung gilt als bestanden, wenn mindestens 80 % (Kupfer) bzw. 70 % (Zink) der Schwermetallfracht von der Anlage zurückgehalten wurde. Frühestens 16 h und spätestens 72 h nach Beendigung der Prüfung des Zink- und Kupferrückhalts wird dasselbe Filterelement auf eine mögliche Rücklösung der Schwermetalle durch Auftausalze geprüft. Hierzu wird eine mit Auftausalzen (NaCl gemäß TL-Streu 10 g/L, siehe Kapitel 2.2) versetzte Lösung aus entionisiertem Wasser verwendet. Das Filterelement wird mit dieser Lösung 200 Min. lang mit einer Intensität von 6 L/(s·ha) beaufschlagt. Vor Beginn der Versuche werden die Säulen zunächst mit 6 L/(s·ha) entionisiertem Wasser gespült, bis die Leitfähigkeit im Ablauf stabil ist. Die Probennahme im Ablauf erfolgt nach einem genau festgelegten Programm [4].

Die Salzprüfung nach DIBt ist bestanden, wenn die gemittelte Gesamtkonzentration aller Einzelproben die zulässigen Ablaufkonzentrationen gemäß **Tabelle 3** nicht überschreitet. Diese Werte entsprechen den Prüfwerten der BBodSchV für Kupfer (50 µg/L) und Zink (500 µg/L). In der Berechnung werden die möglichen Kupfer- und Zinkbelastungen der Tausalzlösung berücksichtigt. Des Weiteren wird verlangt, dass auch die Konzentration in den Einzelproben bestimmte Gehalte nicht übersteigt.

5.2 Methode 1: Untersuchungen zur Salzstabilität im Rahmen eines Projekts zur Standzeitermittlung

In dem Forschungs- und Entwicklungs-Projekt „Untersuchung von Anlagen zur Behandlung des Niederschlagswassers von Verkehrsflächen“, welches vom Bayerischen Landesamt für Umwelt (LfU) beauftragt wurde, wurde ein Laborverfahren zur Standzeitermittlung dezentraler Behandlungsanlagen entwickelt [39], da während der Prüfung beim DIBt keine detaillierte Methodik zur Ermittlung einer realistischen Standzeit der Anlagen vorgesehen ist. Dieses Verfahren besteht aus drei Teilen. Während des ersten Teils wird eine Vorbelastung mit Zink und Kupfer in Abhängigkeit der Anlagenstandzeit (bis zu zehn Jahre) auf das Filtermaterial im Labormodell aufgebracht (**Bild 4**, links). Im zweiten Teil wird analog zu den Prüfgrundsätzen des DIBt eine Stoffjahresfracht an Zink und Kupfer auf das vorbeladene Filterelement in drei charakteristischen Regenspenden aufgebracht (siehe Kapitel 5.1). Die Randbedingungen (mit Zink und Kupfer beladenes Filterelement, Dauer der Wartezeit, Spülung vor Auftausalzuntersuchung, Regenspende, Regendauer und Probennahme) im dritten Teil sind analog zu den aktuellen DIBt-Prüfgrundsätzen, welche aufgrund der Erkenntnisse aus diesem Projekt teilweise modifiziert wurden. Im Unterschied zum DIBt-Verfahren wird bei der neu entwickelten Untersuchungsmethode eine Auftausalzmischung aus Trinkwasser mit Zulaufkonzentrationen von 10 g NaCl/L und 2,5 g $CaCl_2$/L verwendet. Somit liegt die Chlorid-Zulaufkonzentration bei 7660 mg/L und ist mit den in **Tabelle 1** angegebenen Konzentrationen realer Verkehrsflächenabflüsse vergleichbar.

Tabelle 3: Festgesetzte Prüfwerte für Zink und Kupfer im Ablauf (zulässige Ablaufkonzentration nach [3])

Schwermetall	Zulaufkonzentration Schwermetall in der Auftausalzlösung [µg/L]	Zulässige Ablaufkonzentration [µg/L]
Zink	Messwert Auftausalzlösung	≤ 500 + Messwert Auftausalzlösung
Kupfer	Messwert Auftausalzlösung	≤ 50 + Messwert Auftausalzlösung

Anhand dieser Laboruntersuchungen wurde festgestellt, dass bei einigen Filtermaterialien, die mit den beiden Schwermetallen Kupfer und Zink beladen sind, beim Einsatz eines Feuchtsalzgemisches aus NaCl und $CaCl_2$ eine Rücklösung von Kupfer und Zink stattfinden kann, obwohl bei reinem NaCl keine Remobilisierungen auftraten [40]. Außerdem kam es bei wenigen der beladenen Filtermaterialien während der Feuchtsalzuntersuchung zur selektiven Remobilisierung von Zink. Es wurde bei einem Teil der Filtermaterialien festgestellt, dass die Feuchtsalzaufbringung zu einer Veränderung des pH-Wertes im Anlagenablauf führte. Besonders kritisch ist zu sehen, dass weder die untersuchten dezentralen Behandlungsanlagen noch der zu Vergleichszwecken verwendete Oberboden, der in Versickerungsmulden und Banketten eingesetzt wird, Chlorid zurückhalten können und sie somit nach GrwV in Verbindung mit den Geringfügigkeitsschwellenwerten schon heute nicht mehr zur Behandlung von Verkehrsflächenabflüssen bei anschließender Versickerung geeignet wären.

5.3 Methode 2: Weitergehende Untersuchungen im Rahmen des Projekts „Schadstoffremobilisierung durch Auftausalze“

Die grundlegende wissenschaftliche Beschreibung und Erklärung des Remobilisierungsverhaltens von verschiedenen, gleichzeitig auf Filtermaterialien zurückgehaltenen Schwermetallen bei der dezentralen Behandlung von Verkehrsflächenabflüssen unter Verwendung unterschiedlicher Auftausalze ist das Ziel des Forschungsvorhabens „Schadstoffremobilisierung durch Auftausalze“, welches von der Bayerischen Forschungsstiftung und der Firma FRÄNKISCHE ROHRWERKE gefördert wird. Als Schwermetalle werden dabei Zink, Kupfer, Nickel, Blei und Cadmium verwendet, um weitreichendere Aussagen zum Rückhalt unterschiedlich mobiler Schwermetalle treffen zu können. Daneben werden die Wirkmechanismen zum Schwermetallrückhalt der einzelnen Filtermaterialien beschrieben. Abschließend

Bild 4: Beispiele für Versuchsaufbauten: Methode 1 (links) und Methode 2 (rechts)

wird eine mögliche Schwermetallremobilisierung anhand des Einsatzes dreier verschiedener Auftausalze in getrennten Versuchen untersucht.

Bei den Säulenversuchen (**Bild 4**, rechts) erfolgt zuerst eine Beladung der Materialien mit Zink, Kupfer, Nickel, Blei und Cadmium (Zink: 5000 µg/L, Kupfer, Nickel und Blei: je 2500 µg/L und Cadmium: 625 µg/L) bis zum Schwermetalldurchbruch. Der Durchbruch wurde als Zeitpunkt, an dem die Ablaufkonzentration an Kupfer oder Zink als Leitparameter mindestens 50 % der Zulaufkonzentration beträgt, definiert. Sollte in den nachfolgenden Auftausalz-Untersuchungen eine Schwermetallremobilisierung auftreten, werden weitere Säulen mit demselben Filtermaterial befüllt und mit einer mittleren Belastung von ca. einer Jahresfracht beladen. Dabei wird jedes Material in acht Säulen vergleichbar beladen, sodass in den folgenden vier Untersuchungen (drei verschiedene Auftausalzkombinationen sowie weitergehende Analysen) jeweils zwei Säulen (Doppelbestimmung) zur Verfügung stehen, um differenzierte Aussagen zum Remobilisierungsrisiko treffen zu können.

Nach der Beladung werden unterschiedliche Feuchtsalzversuche mit reinem NaCl (10 g/L), mit einer definierten Mischung aus NaCl und $CaCl_2$ (10 g/L bzw. 2,5 g/L) und mit einer definierten Mischung aus NaCl und $MgCl_2$-Hexahydrat (10 g/L bzw. 2,5 g/L) mit einer Regenspende von 6 L/(s·ha) für je 200 Minuten unter Verwendung von entionisiertem Wasser an getrennten Säulen durchgeführt.

Weitergehende Analysen, wie Korngrößenverteilung, Porenvolumen, Porengrößenverteilung, Schwermetallgehalte, spezifische Oberfläche sowie Beschaffenheit der Oberfläche, dienen dazu, unterschiedliche Wirkmechanismen zu identifizieren, um die in Kapitel 3 erläuterten Wechselwirkungen zu untersuchen und das Verhalten der Filtermaterialien bei Auftausalzeinwirkung unter realen Bedingungen zu dokumentieren [41].

Aufgrund der differenzierten Herangehensweise bei den Säulenversuchen ist es für fünf Schwermetalle möglich, Aussagen zum Remobilisierungsrisiko anhand zweier unterschiedlicher Beladungszustände der Filtermaterialien zu treffen. Als Ergebnis kann festgehalten werden, dass die sechs untersuchten Materialien die Schwermetalle unterschiedlich gut zurückhalten. Dabei unterscheidet sich sowohl der Zeitpunkt, an dem der Durchbruch zu 50 % erfolgt, als auch bei der Beladung mit nur einer Jahresfracht der prozentuale Schwermetallrückhalt deutlich. Ferner werden Kupfer und Blei meist besser als die anderen Schwermetalle gebunden, sodass die Durchbrüche zu 50 % anhand des Parameters Zink bestimmt wurden. Im Anschluss an die Beladung wurde das mögliche Auftreten einer Schwermetallremobilisierung durch drei verschiedene Auftausalzmischungen getrennt erfasst. Die ersten Ergebnisse zur Remobilisierung bestätigen die Erkenntnisse der ersten Methode (Kapitel 5.2), jedoch sollen in Kombination mit den noch auszuführenden, weitergehenden Analysen detaillierte Aussagen ermöglicht werden. Diese Ergebnisse werden Gegenstand einer weiteren Publikation sein.

5.4 Vergleich und Anwendungsbereich der Methoden 1 und 2

Die erste Methode hat im Gegensatz zur zweiten Methode den Vorteil, dass innerhalb einer Versuchswoche im Labor Aussagen zur Standzeit als auch zur Schwermetallremobilisierung getroffen werden können. So wird die zu untersuchende Anlage innerhalb eines Tages gealtert, an zwei Tagen auf den Schwermetallrückhalt im letzten Betriebsjahr getestet sowie in den beiden darauffolgenden Tagen anhand von Salzversuchen auf die Rücklösung von Schwermetallen untersucht. Dabei werden für eine Doppelbestimmung nur zwei Labormodelle benötigt. Aufgrund der größeren Modelle im Vergleich zu Methode 2 (siehe **Bild 4**) können lediglich die beiden aufkommensrelevantesten Schwermetalle Kupfer und Zink untersucht werden (Toxizität sowie Entsorgungsproblematik). Sobald bei einer Salzuntersuchung eine Remobilisierung ermittelt wurde, kann die Höhe einer möglichen Remobilisierung bei den nachfolgenden Salzuntersuchungen am gleichen Material aufgrund der bereits zurückgelösten Schwermetalle nicht mehr bestimmt werden.

Im Gegensatz dazu ist der Aufwand für die zweite Methode deutlich höher. Es werden anstelle zweier Modelle acht parallel zu beschickende Säulen pro Material benötigt. Dabei werden diese je nach Rückhaltekapazität des Materials zwischen drei und dreißig Tage lang mit den fünf relevantesten Schwermetallen beaufschlagt, wobei während der Beladung die Simulation von Trockenperioden möglich ist. Zwei Säulen dienen den weitergehenden Analysen und je zwei der anderen sechs Säulen der getrennten Untersuchung der drei relevanten Auftausalz(-kombinationen). Diese Untersuchungen eignen sich zur quantitativen Bestimmung des Einflusses jedes Auftausalzes auf an Filtermaterialien zurückgehaltenen Schwermetallen sowie zur Beschreibung der Rückhaltemechanismen. Aufgrund der Verwendung von fünf Schwermetallen kann zusätzlich nachgewiesen werden, welche davon besonders gut zurückgehalten werden und ob bzw. bei welchen Auftausalzen diese remobilisiert werden können. Jedoch ist bei dieser Methode eine genaue Erfassung der Standzeit aufgrund der kleinen Säulendurchmesser nicht möglich (Randeffekte).

6. Fazit und Ausblick

Zu den in Deutschland aufkommensrelevanten Auftausalzen zählen neben dem am weitesten verbreiteten NaCl, ebenfalls $CaCl_2$, $MgCl_2$ und deren Mischungen. Bei der Verwendung des Auftausalzes spielt sowohl der Preis, die regionale Verfügbarkeit, die Tauwirksamkeit als auch die Taugeschwindigkeit eine Rolle. Somit können $CaCl_2$ und $MgCl_2$ regional begrenzt in einer Kommune, nur an bestimmten Orten, wie sensible Beläge und Fußgängerbrücken, innerhalb einer Kommune oder temporär bei tiefen Temperaturen eingesetzt werden. Die eingesetzten Mengen orientieren sich an den Empfehlungen der FGSV und die über ein Jahr pro m² Straßenfläche eingesetzten Mengen sind auf Autobahnen im Vergleich zu anderen Straßenkategorien am höchsten. Dabei hat sich die Feuchtsalztechnik im staatlichen Straßenwinterdienst überall mit Ausnahme kleinerer Kommunen durchgesetzt. Ausgehend von den aufgebrachten Mengen an Auftausalzen, können die Chlorid-Abflusskonzentrationen abgeschätzt werden, auch wenn keine Abflusskonzentrationen gemessen wurden. Diese rechnerisch ermittelten Chlorid-Konzentrationen liegen dabei meist über dem Geringfügigkeitsschwellenwert von 250 mg/L, was durch zahlreiche im Winter gemessene Konzentrationen bestätigt wird.

Die Verwendung von Auftausalzen hat für dezentrale Behandlungsanlagen für Verkehrsflächenabflüsse zur Folge, dass ein Teil der Prozesse zum Schadstoffrückhalt an Filtermaterialien durch die erhöhten Salzkonzentrationen negativ beeinflusst werden kann. Somit müssen Filtermaterialien vor dem Einsatz in der Realität an Verkehrsflächen mit Auftausalzeinsatz bei Untersuchungen im Labor zeigen, dass deren Rückhaltemechanismen durch den saisonalen Einsatz von Auftausalzen nicht bzw. nur geringfügig beeinträchtigt sind und somit filterstabil sind. Anhand der vorgestellten Methodik mit weitergehenden Untersuchungen (siehe Kapitel 5.3) ist es erstmalig möglich, die Einflüsse der aufkommensrelevanten Auftausalze im Labor getrennt für fünf Schwermetalle zu erfassen und die Wirkmechanismen der Filtermaterialien zu beschreiben. Sollte sich bei solchen Untersuchungen zeigen, dass eine Anlage nicht für alle Auftausalze einen sicheren Schadstoffrückhalt bietet, dürfen diese dezentralen Anlagen nur an Standorten eingesetzt werden, an denen entsprechende Auftausalze verwendet werden, welche zu keiner Schadstoffremobilisierung führen. Ziel muss es immer sein, durch die Einhaltung der rechtlichen Anforderungen die in Verkehrsflächen aufkommensrelevanten Stoffe in dezentralen Anlagen ausreichend zurückzuhalten und somit negative Auswirkungen auf das Grundwasser zu verhindern.

In Zukunft kann die Umsetzung der Verschärfung der Geringfügigkeitsschwellenwerte aus den Jahren 2012/2013 zu erhöhten Anforderungen an die dezentralen Anlagen führen, sodass auch geringe Remobilisierungen durch Auftausalze nicht mehr erlaubt sein werden. Denn eine weitere Verschärfung der Schwermetallkonzentrationen, besonders für Cadmium, Kupfer und Zink, würde bedeuten, dass deren Zielwerte in vielen Fällen bereits durch den reinen Niederschlag überschritten werden [42, 43]. Eine Versickerung des Straßenabflusses wäre somit oft nicht möglich, da der Zielwert für Kupfer von 10 µg/L im Anlagenablauf durch derzeitige Behandlungstechniken nur schwer eingehalten wird.

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Autoren

Maximilian Huber, M.Sc.
Lehrstuhl für Siedlungswasserwirtschaft |
Technische Universität München |

Prof. Dr.-Ing. **Jörg E. Drewes**
Lehrstuhl für Siedlungswasserwirtschaft |
Technische Universität München |

Prof. Dr. rer. nat. habil. **Brigitte Helmreich**
b.helmreich@tum.de |
Lehrstuhl für Siedlungswasserwirtschaft |
Technische Universität München |
Am Coulombwall 8 |
D-85748 Garching

Prof. Dr.-Ing. habil. **Antje Welker**
Fachgebiet Siedlungswasserwirtschaft und Hydromechanik |
Frankfurt University of Applied Sciences |
Nibelungenplatz 1 |
D-60318 Frankfurt am Main

Chapter 9: Research Paper: "Heavy Metal Removal Mechanisms of Sorptive Filter Materials for Road Runoff Treatment and Remobilization under De-icing Salt Applications"

First results showed that Cu and Zn were not (or less) released with pure sodium chloride but with a mixture of sodium chloride and calcium chloride (cf. Section 7.1). However, only the effect of sodium chloride on previously retained Cu and Zn is currently considered in test procedures of authorities (DIBt, 2012; DIBt, 2015; ÖNORM B 2506-3 Entwurf, 2015). Thus, the objective of the following paper was to elucidate the heavy metal removal and remobilization under de-icing salt applications for five heavy metals and three de-icing salts (sodium chloride, a mixture of sodium chloride with calcium chloride, and a mixture of sodium chloride with magnesium chloride) using lab-scale column experiments. Because more and more manufactured decentralized treatment systems are used (cf. Section 2.3), the focus was set on six commercially supplied filter materials. The hypothesis was:

The composition of the de-icing salt solution has a significant effect on the remobilization of heavy metals and the behavior of each heavy metal differs widely (Hypothesis 7).

The risk of remobilization was determined after preloading the filter material with Cd, Cu, Ni, Pb, and Zn. The removal efficiencies of each heavy metal varied for all filter materials. In most cases, Cu and Pb were best retained. The best material removed about six times more Zn compared with all other materials. Concerning the subsequent de-icing salt experiments, the results were more heterogeneous. The behavior of each filter material, each heavy metal, and each de-icing salt was different. In general, Cu was released the least from most of the selected filter materials by the three de-icing salts. In most cases, the mixture of sodium chloride and calcium chloride resulted in the strongest effect on remobilization compared with the other two de-icing salts. Pure sodium chloride had a minor effect on the remobilization for most heavy metals and filter materials. The results can be helpful for the development and improvement of filter materials used in decentralized systems. Advanced analyses were used to specify the removal mechanisms. For example, the metal distribution in a particle was evaluated by energy-dispersive X-ray spectroscopy and laser ablation tests. Computer modeling was used to support the findings of the lab-scale column experiments by simulating the speciation of heavy metals, also under the application of different de-icing salts. Data and figures of the modeling are presented in the conference proceeding Huber et al. (2016b).

The following research paper was published in Water Research 102, 453–463. Huber, Hilbig, Badenberg, Fassnacht, and Helmreich conceived the experiments. Huber, Hilbig, Badenberg, and Fassnacht designed, performed, and evaluated the experiments. Fassnacht and Huber did the computer modeling. Huber also wrote the manuscript. All authors contributed to the discussions.

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Heavy metal removal mechanisms of sorptive filter materials for road runoff treatment and remobilization under de-icing salt applications

Maximilian Huber [a], Harald Hilbig [b], Sophia C. Badenberg [a], Julius Fassnacht [a], Jörg E. Drewes [a], Brigitte Helmreich [a, *]

[a] Chair of Urban Water Systems Engineering, Technical University of Munich, Am Coulombwall 3, 85748 Garching, Germany
[b] cbm—Centre for Building Materials, Technical University of Munich, Baumbachstraße 7, 81245 Munich, Germany

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ABSTRACT

The objective of this research study was to elucidate the removal and remobilization behaviors of five heavy metals (i.e., Cd, Cu, Ni, Pb, and Zn) that had been fixed onto sorptive filter materials used in decentralized stormwater treatment systems receiving traffic area runoff. Six filter materials (i.e., granular activated carbon, a mixture of granular activated alumina and porous concrete, granular activated lignite, half-burnt dolomite, and two granular ferric hydroxides) were evaluated in column experiments. First, a simultaneous preloading with the heavy metals was performed for each filter material. Subsequently, the remobilization effect was tested by three de-icing salt experiments in duplicate using pure NaCl, a mixture of NaCl and $CaCl_2$, and a mixture of NaCl and $MgCl_2$. Three layers of each column were separated to specify the attenuation of heavy metals as a function of depth. Cu and Pb were retained best by most of the selected filter materials, and Cu was often released the least of all metals by the three de-icing salts. The mixture of NaCl and $CaCl_2$ resulted in a stronger effect upon remobilization than the other two de-icing salts. For the material with the highest retention, the effect of the preloading level upon remobilization was measured. The removal mechanisms of all filter materials were determined by advanced laboratory methods. For example, the different intrusions of heavy metals into the particles were determined. Findings of this study can result in improved filter materials used in decentralized stormwater treatment systems.

1. Introduction

Parameters commonly monitored in runoff from traffic areas include solids, organic substances, heavy metals, and compounds of de-icing salts (Folkeson et al., 2009). Heavy metals such as Cd, Cu, Ni, Pb, and Zn are often measured because of their environmental relevance and can occur in notable runoff concentrations (Huber et al., 2016c). Chloride is also determined in high runoff concentrations from traffic areas with winter services because NaCl, $CaCl_2$, and $MgCl_2$ are widely used as de-icing salts (Bauske and Goetz, 1993; Nelson et al., 2009).

In recent years, dozens of manufactured decentralized stormwater treatment systems have been developed to treat traffic area runoff prior to discharge into surface water or local groundwater (Dierkes et al., 2015). To retain dissolved and particulate substances, commonly a mechanical step (e.g., sedimentation or filtration) followed by a subsequent treatment step with a filter material is necessary (Hilliges et al., 2013). The term "filter material" was deliberately chosen because it is commonly used by manufacturers and operators, although the removal mechanisms of filter materials consist not just of filtration (e.g., after precipitation), but also of sorption or ion exchange. These decentralized treatment systems for traffic area runoff can be approved for use after passing several tests (e.g., remobilization of Cu and Zn by NaCl; DIBt, 2015). However, no standardized testing method is provided among these approval processes to determine the impact of all relevant de-icing salts upon the remobilization of heavy metals that were previously retained by filter materials. Consequently, there is currently no manufactured decentralized treatment system for road runoff available in Germany with proven filter stability against NaCl, $CaCl_2$, and $MgCl_2$, although Cl^- and the corresponding cations can have an effect upon retained heavy metals. For example, during applications of NaCl in road maintenance, increases in Cd, Cu, Ni, and Zn

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* Corresponding author.
E-mail address: b.helmreich@tum.de (B. Helmreich).

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concentrations were detected in the effluent of a constructed wetland (Tromp et al., 2012). Thereby, the Cu effluent concentrations were higher than the maximum influent concentration and the system itself became a source of contamination during the cold season. Hence, the consideration of heavy metal releases triggered by de-icing salts is relevant for systems treating traffic area runoff in moderate and cold climates.

A few studies have examined the remobilization behavior of heavy metals bound to roadside soils (e.g., Amrhein et al., 1992; Bauske and Goetz, 1993; Bäckström et al., 2004). Nelson et al. (2009) tested two soils for the effects of NaCl and $MgCl_2$ on heavy metal mobility (Cd, Cu, and Pb). Both soils exhibited different behaviors for each de-icing salt and heavy metal, in particular for Cu. The use of NaCl resulted in the largest remobilization of Cu and Pb. $MgCl_2$ released higher levels of Cd for both soils. Huber et al. (2014) published results concerning the remobilization of Cu and Zn by NaCl and $CaCl_2$ for two soils and five filter materials used for stormwater treatment. In some cases, both heavy metals were released and in other cases, only Zn showed higher mobility. These effects depended upon the material and the de-icing salt, e.g., some heavy metals were not released with NaCl alone but by a mixture of NaCl and $CaCl_2$. In addition, the pH of the effluent was often reduced by the use of de-icing salts in this study. However, neither of these studies experimented with all of the relevant five heavy metals and three de-icing salts. Thus, the focus of further research should be on the identification of the impact of relevant de-icing salts upon previously retained metals. Moreover, as more and more manufactured decentralized treatment systems are used, commercially supplied filter materials must be tested as these materials are much higher loaded than roadside soils because of their high ratio of drainage area to filter material surface area (Huber et al., 2014).

The objectives of this study were to elucidate the removal and remobilization behaviors of five heavy metals under the application of three de-icing salts using lab-scale column experiments. In addition, the removal mechanisms were specified for all six commercially supplied filter materials, and the influence of two preloading levels upon the remobilization was determined for the material with the highest removal capacity.

2. Material and methods

2.1. Filter materials and chemicals

The following filter materials were used in this study: granular activated carbon (GAC, Donau Carbon GmbH, Germany), a mixture (called alumina) of granular activated alumina (55%; Albemarle Corporation, USA) and porous concrete (45%; Xella Deutschland GmbH, Germany), granular activated lignite (lignite, Rheinbraun Brennstoff GmbH, Germany), half-burnt dolomite (dolomite, Rheinkalk Akdolit GmbH & Co. KG, Germany), and two granular ferric hydroxides with different calcium contents (FerroSorp, HeGo Biotec GmbH, Germany; GFH, Wasserchemie GmbH & Co. KG, Germany). Prior to conducting any experiments, lignite and dolomite were sieved to the size fraction of the other materials (0.5–2.0 mm) to minimize the influence of different size fractions of each filter material upon the heavy metal retention (Smith, 1998). Subsequently, all materials were dried at 105 ± 2 °C to achieve a constant weight, and finally stored in desiccators without additional treatment until use. The chemical and physical characteristics of all blank materials are presented in Tables 1 and 2.

The heavy metal salts used were $CdSO_4 \cdot {}^8/_3H_2O$ (AppliChem GmbH, Germany), $Cu(NO_3)_2 \cdot 3H_2O$ (AppliChem GmbH, Germany), $Ni(NO_3)_2 \cdot 6H_2O$ (Merck KGaA, Germany), $Pb(NO_3)_2$ (AppliChem GmbH, Germany), and $Zn(NO_3)_2 \cdot 6H_2O$ (AppliChem GmbH, Germany). Adjustments of pH values and preservation of all liquid samples were performed with nitric acid 65% p.a. (Merck KGaA, Germany). As de-icing salts, NaCl (AppliChem GmbH, Germany), dried, powdered $CaCl_2$ (AppliChem GmbH, Germany), and $MgCl_2 \cdot 6H_2O$ (AppliChem GmbH, Germany) were used.

2.2. Experimental setup

The experimental apparatus was constructed of glass and all pipes were made of polyvinyl chloride. The glass column had an inner diameter of 2.0 cm and a length of 40 cm, providing a filter material bed height of 14.4 cm. Thus, the ratio of the inner diameter to the largest particle was at least 10:1 to obtain appropriate lab-scale design parameters (Fanfan et al., 2005). A glass screen was placed at the bottom of the column to support the filter materials. The influent was directed through the glass columns in up-flow mode (from bottom to top) at ambient temperature (21 ± 2 °C). All sorbent samples were weighed using an analytical balance (MC1 Research RC 210D, Sartorius AG, Germany) with an accuracy of ±0.1 mg.

For heavy metal removal, lab-scale column experiments were conducted with influent concentrations of 0.625 mg/L (Cd), 2.5 mg/L (Cu, Ni, and Pb), and 5.0 mg/L (Zn). These concentrations are higher than most values measured in runoff from traffic areas (to shorten the duration of the experiments), and the different values were chosen to simulate realistic proportions (Huber et al., 2016c). The initial pH was 4.9 ± 0.3 to obtain dissolved metals. Deionized water (electrical conductivity <1 μS/cm) was used for influent preparation and no buffer was used to simulate a worst-case scenario for the determination of dissolved heavy metal removal (e.g., to avoid oversaturated conditions) (Huber et al., 2016a). The flow rate was adjusted to about 6.5 mL/min by a peristaltic pump (ECOLINE VC-MS/CA8-6, Ismatec SA, Switzerland). Thereby, the empty bed contact time was maintained at about 7 min. The experiments were performed in eight columns per material from Monday to Friday. During weekends, the filter materials were kept saturated without flow (representing dry-weather periods). The preloading part was terminated when the breakthrough of Zn was at least 50% in the effluent. This removal efficiency corresponds to the elimination criteria often requested by international regulations for stormwater treatment systems (Huber et al., 2016b). Consequently, a breakthrough of 50% correlates with the end of the service life (i.e., the maximum preloading of a real filter). In addition, one shorter preloading run was performed with the filter material that had the highest removal capacity to test the effects of different preloading levels on remobilization. At the end of the preloading, each column was flushed with deionized water for 60 min to remove the dissolved heavy metals, which were not fixed onto the filter material. The flushing process was controlled by measuring the heavy metal concentrations in effluent samples for all tests. Thus, the effluent concentrations measured in the subsequent de-icing salt experiments were only related to remobilization and no remobilization occurred during the previous deionized water flushes.

Six of the eight columns containing filter materials with preloaded heavy metals were used for the de-icing salt experiments. Two columns were operated with pure NaCl solution (10 g/L), two with a mixture of NaCl (10 g/L) and $CaCl_2$ (2.5 g/L), and two with a mixture of NaCl (10 g/L) and $MgCl_2$ hexahydrate (2.5 g/L). All de-icing salt concentrations were taken from the literature (Huber et al., 2015) and the experiments simulated a typical runoff event (6.0 L/(s·ha), which corresponds to 2.2 mm/h, for 200 min). The ratio of the filter surface area to the drainage area of the decentralized treatment system that was selected for the de-icing salt tests was 1:1820. Therefore, the corresponding flow rate was

Table 1
Chemical characteristics of all blank filter materials based on dry matter. For alumina, the characteristics of the two components are specified.

Parameter	Main elements										Heavy metals					
	Al	TC	Ca	Fe	K	Mg	Mn	Na	TS	Si	Cd	Cu	Cr	Ni	Pb	Zn
Unit	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg
Dolomite	<0.02	8.74	28.7	0.04	<0.02	17.1	<0.02	0.02	<0.02	0.09	<0.3	3.0	52.2	1.4	<5	4.9
FerroSorp	0.12	2.18	6.16	41.5	<0.02	0.46	0.92	0.02	0.06	5.56	0.3	<1	50.3	50.7	14.9	90.0
GAC	0.07	96.3	0.21	0.07	0.96	0.09	<0.02	0.07	0.06	0.39	<0.3	4.5	53.9	5.5	<5	13.2
GFH	0.05	0.09	0.28	60.0	0.02	0.03	0.14	<0.02	0.53	0.18	<0.3	<1	31.3	44.1	22.6	11.9
Granular activated alumina	46.2	0.08	<0.02	0.13	<0.02	<0.02	<0.02	0.19	0.04	0.05	<0.3	18.6	161	2.0	<5	9.1
Lignite	0.10	89.2	2.27	0.70	0.06	0.88	<0.02	0.25	0.46	0.04	<0.3	<1	21.6	2.0	<5	<1
Porous Concrete	0.56	1.82	15.9	0.46	0.13	0.20	<0.02	0.03	0.62	30.6	0.5	16.7	165	9.1	5.4	57.8

20.5 mL/min. The influent pH value was not adjusted for the de-icing salt tests. After the de-icing salt experiments, each column was flushed with deionized water for 15 min to remove most of the salts that were not fixed onto the filter material and to obtain comparable solid samples.

For all preloading experiments, one influent sample per stock solution and several effluent samples at designated times were collected in 50 mL polypropylene bottles. For each de-icing salt test, one influent sample and four effluent samples (each representing a 50 min period) were collected. This sampling strategy was in accordance with the DIBt procedure (DIBt, 2015). Selected samples of all tests were additionally filtrated to determine the dissolved concentrations of the heavy metals (0.45 μm pore size, VWR International GmbH, Germany).

Finally, three layers of each column were separated to specify the attenuation of heavy metals as a function of depth. All samples were subsequently dried at 40 ± 2 °C in a vacuum for 24 h and stored separately in glass vessels pending analysis.

2.3. *Analyses*

For all water samples, measurements of the pH value were immediately performed after sample collection with a glass electrode Sentix 60 (WTW GmbH, Germany) according to DIN EN ISO 10523:2012-04. Subsequently, all samples were preserved with nitric acid at $\mathrm{pH} < 2$. The concentrations of heavy metals were measured by flame atomic absorption spectrometry (AAS, Varian Spectrometer AA-240FS, Palo Alto, USA) according to the APHA Standard Method 3111 with limits of quantification (LOQ) of 1.0 mg/L (Ca), 20 μg/L (Cd), 50 μg/L (Cu), 0.1 mg/L (Fe), 0.5 mg/L (Mg), 1.0 mg/L (Na), 50 μg/L (Ni), 200 μg/L (Pb), and 20 μg/L (Zn). Graphite furnace AAS (Varian Spectrometer AA-240Z with GTA 120, Palo Alto, USA) was used to reduce the LOQ for Cu, Ni, and Pb (5 μg/L) in accordance with the APHA Standard Method 3113. Quality control was maintained by using control and blank samples and by performing standard solution quantity checks every five samples.

The loaded solid and blank samples of all filter materials were fused at 1000 °C for 15 min with three times their mass of $LiBO_2$ (or at 500 °C for 10 min with ten times their mass of NaOH). Subsequently, the melt pellets were digested and quantitative chemical analyses of the solutions were performed using inductively coupled plasma optical emission spectroscopy (ICP-OES, Ultima 2 ICP-OES, HORIBA Jobin Yvon, France) for Al, Ca, Cr, Fe, K, Mg, Mn, Na, S, and Si (LOQ for all elements was 0.02 wt%). Heavy metals (Cd, Cu, Cr, Ni, Pb, and Zn) were determined with ICP-OES after an inverse aqua regia digestion in a pressure vessel (300 mg sample in 6 mL HNO_3 and 2 mL HCl for 20 min at 180 °C and ca. 20 bar; LOQ: Cd: 0.3 mg/kg, Pb: 5 mg/kg, all others: 1 mg/kg).

For all blank and selected loaded samples, the physical characteristics dry residue (105 °C), loss on ignition (550 °C and 1000 °C), and bulk density were determined according to DIN EN 12880:2001-02, DIN 18128:2002-12, and DIN EN 1097-3:1998-06, respectively. The characteristics of the pore system (pore volumes and porosity) were determined by Hg intrusion in a pressure range from 0.004 MPa to 400 MPa (AutoPore III, Micromeritics GmbH, Germany) to detect the pore diameter in the range from 345 μm to 3.2 nm. Specific surface areas of the filter materials were determined according to Standard Method ISO 9277:2010-09 using a SA 3100 plus (Beckman Coulter, Brea, USA) with N_2 as an adsorption gas (BET method). In addition, the total carbon (TC) and total sulfur (TS) were determined with a multi EA 4000 element analyzer at 1350 °C (Analytik Jena AG, Germany).

To characterize selected blank and loaded samples, the following advanced methods were used: X-ray diffraction data were collected using an XRD 3003 TT (GE Sensing & Inspection Technologies GmbH, Germany) with θ-θ configuration and Cu Kα radiation ($\lambda = 1.54$ Å). Intensities were recorded over scattering angles between 5° and 70° at steps of 0.02° and ZnO was used as an internal standard. The electroacoustic principle was used for

Table 2
Physical characteristics of all blank and dried filter materials. For alumina, the characteristics of the two components are specified.

Parameter	Dry residue (105 °C)	Loss on ignition (550 °C)	Loss on ignition (1000 °C)	Bulk density	Pore volume (5–345,000 nm)	Pore volume (5–50 nm)	Porosity	Surface area (BET)
Unit	%	%	%	g/L	cm^3/g	cm^3/g	%	m^2/g
Dolomite	99.9	0.3	31.1	1130	0.132	0.031	28.2	5.4
FerroSorp	90.0	10.9	16.5	590	0.717	0.636	72.2	224
GAC	97.0	12.4	96.3	430	0.354	0.026	27.5	986
GFH	55.0	8.8	11.9	1120	0.154	0.000	8.9	237
Granular activated alumina	99.8	7.6	8.7	970	0.159	0.103	27.4	263
Lignite	99.2	8.8	89.2	470	0.450	0.014	38.2	262
Porous Concrete	99.4	5.2	11.6	350	1.239	0.397	85.4	29.8

the zeta potential (as a surrogate for surface charge) measurements (Electroacoustic Spectrometer DT-1200, Quantachrome GmbH & Co. KG, Germany); for these experiments only, the samples were grinded with a ball mill. For infrared (IR) spectroscopy, a Spectrum 100 (Perkin Elmer, USA) was used with Golden Gate ATR technique. Sample surface characteristics were determined with a Scanning Electron Microscope (SEM) using a JEOL JSM 5900 LV (JEOL Ltd, Japan) and an energy-dispersive X-ray spectroscopy (EDX) system (RÖNTEC AG, Germany); each sample was sputtered with a 5 nm gold layer prior to analysis. For lignite, a Euro EA-CHNSO Elemental Analyzer (HEKAtech GmbH, Germany) was used to determine the contents of H, N, and O at 1500 °C. The laser ablation inductively coupled plasma mass spectrometer (LA-ICP-MS) consisted of a laser ablation system NWR 213 (New Wave Research, USA) and a quadrupole-ICP-MS NexION 300D (Perkin Elmer, USA). The material was removed from the sample surface with a high-performance neodymium-doped yttrium aluminum garnet laser (213 nm). 100 μm laser spots with different ablation depths of about 10 μm and 50 μm were applied for each measurement. The ablated material was transported via helium as carrier gas into the ICP-MS.

Speciation calculations for half-burnt dolomite, which had a well-defined composition, were carried out using PHREEQC 3.3.2 at a wide pH range to model the behavior of all five heavy metals. The input data consisted of the inflow concentrations of Ca, Cd, Cl, Cu, Mg, N, Na, Ni, Pb, S, and Zn according to the chemicals used for each simulated test. Different TC concentrations, which were measured in selected effluent samples, were also considered.

3. Results and discussion

3.1. Heavy metal removal

The simultaneous heavy metal removal was determined during eight experiments. For FerroSorp, which had the highest removal capacity, a second test was performed to evaluate the effect of different preloading levels (low and high; cf. Section 3.4) on the remobilization. Because GFH has a high moisture content (45.0%), one test was performed with dried GFH (comparable to the other experiments) and one with the original moisture.

At the beginning of the column experiments, with the exception of GFH, the effluent pH increased from 4.9 ± 0.3 (influent solution) to approximately 8.7 (FerroSorp), 9.3 (alumina), 10.2 (GAC), 10.7 (dolomite), and 10.9 (lignite). The pH value decreased during the experiments for all materials with increasing exhaustion of the capacities, which was also reported for heavy metal removal by Liu et al. (2005) and Huber et al. (2016a). GFH was the exception, with a constant effluent pH value of 4.0–4.1 (cf. Fig. 1).

With the exception of half-burnt dolomite, the heavy metals Cu and Pb were better retained by the filter materials than any of the other three heavy metals. Furthermore, the removal efficiencies decreased in the order of Zn > Ni > Cd for alumina, GAC, and lignite. For dolomite, the metals were retained in the order Ni > Zn = Cu > Pb = Cd; at the end of the preloading, Cd and Pb were mainly dissolved (ca. 54–83%) in the effluent, whereas Cu, Ni, and Zn were more undissolved (ca. 41–98%). The breakthrough curves of all heavy metals and the corresponding pH-drift curves are plotted in Fig. 1 for FerroSorp and GFH. For the high-preloaded FerroSorp, all heavy metals were removed to a large extent during the first week (until a bed volume of 986; bed volume = bed height * column crosssectional area). Then, the removal efficiency decreased steadily during continuous flow and increased stepwise after the weekends without flow. The sequence of decreasing removal efficiencies was continuously given by Pb > Cu > Cd > Zn > Ni, which agrees with the adsorption tendencies of divalent transition elements by the hydrous oxides proposed by Liu et al. (2005), which follows the Irving-Williams series. The pH decreased to approximately 6.6. The low preloading of FerroSorp was terminated after 569 bed volumes. The removal efficiencies were comparable to those of the high-preloaded FerroSorp at the same bed volume range. In contrast to FerroSorp, no removal was measured for GFH with the exception of a low Pb retention (Figs. 1 and 2). A similar result was obtained for GFH with its high natural moisture content for this experimental setup. Consequently, no de-icing salt experiments were performed with GFH.

The measured heavy metal contents of the three layers are shown as the means of two columns in Fig. 2. The contents are not shown for dolomite because a breakthrough of 50% for Zn was not achieved (as the permeability was reduced because of precipitated metal salts) and a loss of heavy metals was calculated for the sample preparation (e.g., metal salts adhered to the glass wall).

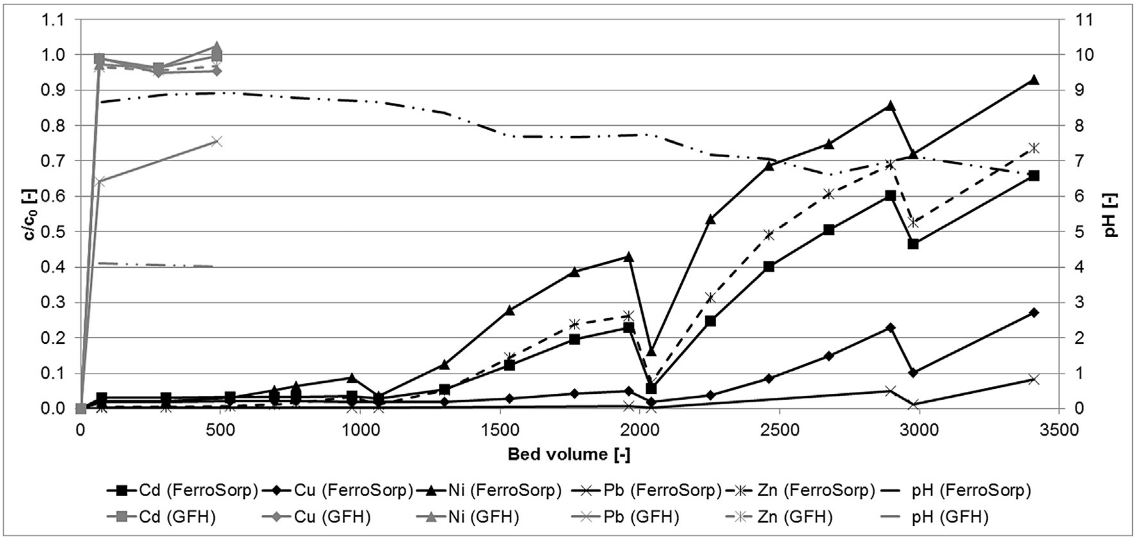

Fig. 1. Breakthrough curves of FerroSorp (high preloading, black) and GFH (dried, gray) for all five heavy metals and the corresponding pH-drift curves.

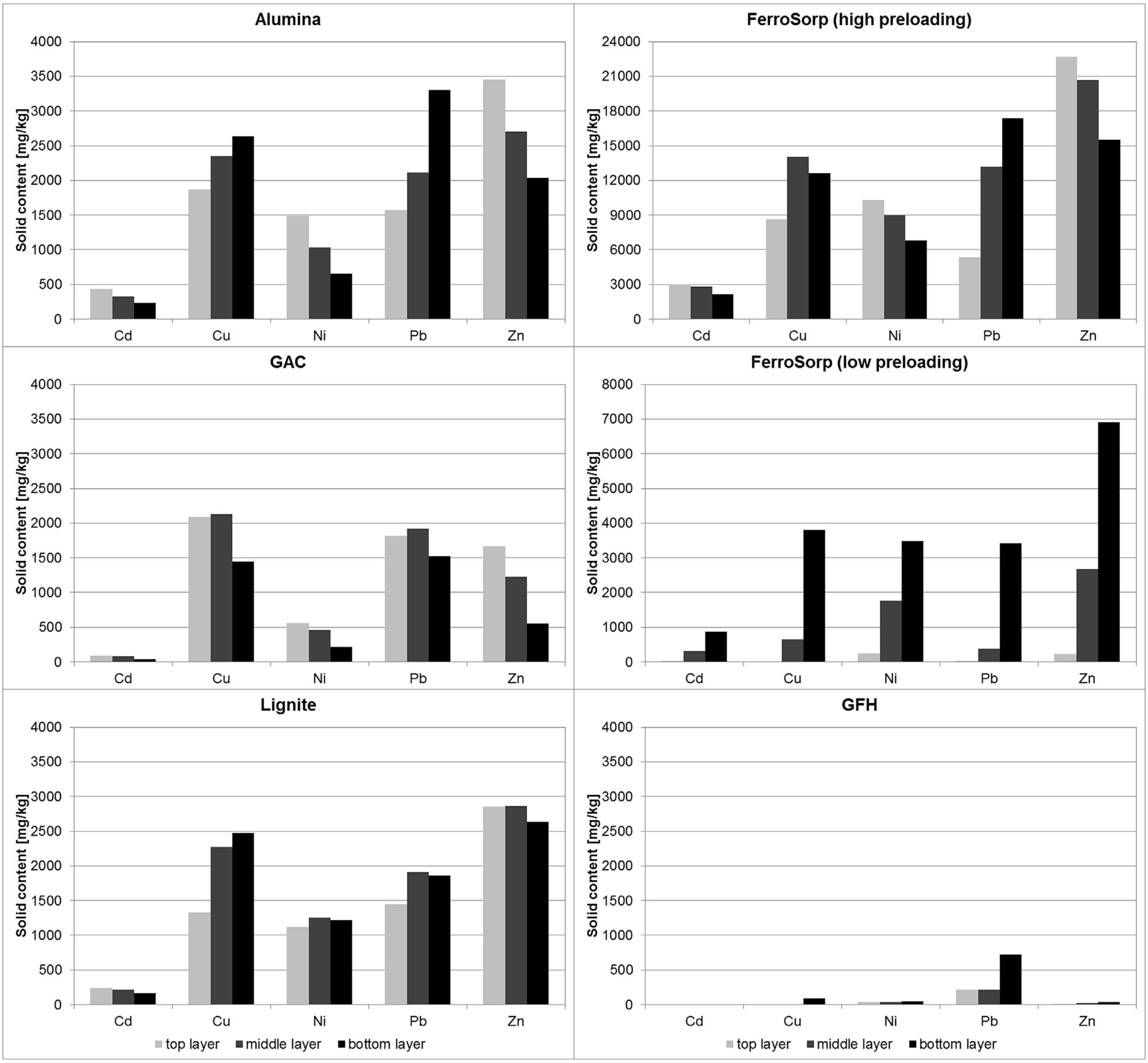

Fig. 2. Heavy metal contents of five filter materials as functions of depth after the preloading in the up-flow mode (values are the means of duplicates). For FerroSorp, both preloading levels are presented.

Therefore, the average metal contents of dolomite were calculated from the differences of influent and effluent concentrations to be approximately 379 mg/kg Cd, 1680 mg/kg Cu, 1720 mg/kg Ni, 1350 mg/kg Pb, and 3420 mg/kg Zn. For all filter materials, the total amount of each heavy metal retained was always much higher than the corresponding solid contents of the blank materials (cf. Table 1). The highest capacities were measured for FerroSorp (high preloading), followed by comparable results for alumina, FerroSorp (low preloading), and lignite. GAC had lower sorption capacities, and GFH showed only a slight Pb removal in these experiments. More mobile elements (i.e., Cd, Ni, and Zn) had an earlier breakthrough and the highest contents in the top layer, whereas Cu and Pb mostly had the highest contents in the bottom layer. For GAC, all heavy metals had a fast breakthrough of at least 50% and a competition between the metals was observed (Cu and Pb replaced other metals). Consequently, the heavy metal attenuation of GAC was different from that of the other filter materials.

The removal of metals by filter materials for stormwater treatment has also been determined by other researchers. For the same GFH, Genç-Fuhrman et al. (2007) measured the third-best removal in a batch experiment comparison of eleven materials. It retained Cr most effectively, but also Cd, Cu, Ni, and Zn. The arsenic removal was moderate in contrast to the results of Sperlich et al. (2005). However, Genç-Fuhrman et al. (2007) used an initial pH value of 6.5 and NaCl/$NaHCO_3$ to simulate the ionic strength that led to the precipitation of some metals. Sperlich et al. (2005) performed column experiments at a pH of 7 with ionic strength simulated by NaCl. These differences in the experimental setup can explain the large variances in the removal efficiencies for GFH (Huber et al., 2016a).

3.2. Removal mechanisms

3.2.1. Alumina

The mixture consisted of a granular activated alumina (87.3% Al_2O_3; 0.19% Na) and a porous concrete with low impurities (Table 1). The pH value of the effluent decreased during the heavy metal removal test from 9.3 to 6.6. The removal mechanisms are based on the high surface area and the interactions of metals with the surface, but also on the high porosity of the porous concrete (Table 2). The zeta potential decreased from −41 mV (unloaded sample at original pH value) to −10 mV at the end of the preloading, confirming that alumina had a high removal capacity for metal cations because of outer-sphere complexation. In addition, the negatively charged surface enables sorption by ligand exchange. They could also precipitate at the prevailing pH values, mainly as hydroxides, although the metal concentrations in solution might be less than the solubility limits. Considering literature data, some cations might be removed by sorption after pore diffusion or ion exchange (Genç-Fuhrman et al., 2007), the latter being proposed by the measurements of a decreasing Na content in the filter material during the experiment.

3.2.2. Dolomite

Half-burnt dolomite is obtained from the thermic dissociation of dolomite at 650–750 °C. The main components were determined to be $CaCO_3$ (approximately 71.6%), $MgCO_3$ (0.9%), and MgO (26.8%). The pH increased by the reaction of H_2O with MgO to form $Mg(OH)_2$ and the dissolution of carbonates and their subsequent reactions. High effluent concentrations of Ca (3.1–13.2 mg/L) and Mg (5.9–7.9 mg/L) and a low reduction of the materials' C content were measured. The heavy metals were mainly retained at pH 10 by precipitation as hydroxides and carbonates as identified by the PHREEQC calculations at a pH of 9.7–10.7 and a TC of 3.5 mmol/L. The main metal species were: free cations (Cd), hydroxides (Cd, Cu, and Zn), and carbonates (Cd, Ni, Pb, and Zn). In addition, simulations showed that Ni had the highest affinity for carbonate precipitation. This effect increased with decreasing TC. Thus, Cd, Pb, and Zn were precipitated less as carbonates because of the presence of Ni. This speciation can explain the low removal of Cd and Pb and the high removal of Ni. The calculations were confirmed by the analyses of preloaded dolomite by SEM/EDX (Fig. 3). By evaluating the backscattered electrons (lighter places indicate heavier elements), clusters of retained heavy metals were found on the surface (site B). At these sites, C and O were also measured, indicating the precipitation of the metals. Acosta et al. (2011) also determined the removal of Pb and Zn by carbonate precipitation on the surface of dolomite.

3.2.3. FerroSorp

This porous ferric hydroxide has a large surface area and pore volume. It mainly consists of Fe, Si, Ca, TC, and Mn compounds. During the removal experiment, almost no Fe (<0.1 mg/L), some Mg (<0.5–3.3 mg/L), and large amounts of Ca (3.0–13.4 mg/L) were measured in the effluent. The TC content of the filter material, which was mostly present as $CaCO_3$, decreased to 1.22% (Ca to 1.73%). Thus, the Ca content decreased by approximately 72% and the TC content by approximately 44%. Consequently, heavy metals must be precipitated as carbonates and might also be retained by ion exchange. Further pH-dependent removal mechanisms, such as sorption (cf. Section 3.1) and surface complexation (hydroxide groups), were proposed by Wu and Zhou (2009) and supported by zeta potential measurements. Moreover, the porous structure of FerroSorp enhances the diffusion of heavy metals into the particle.

On the surface, only high amounts of Pb were measured by SEM/EDX (Fig. 4). To determine the intrusion of all metals into the particle, LA-ICP-MS tests were performed with particles of the bottom layer; at and near the surface (0–60 μm), a high retention of Pb was measured. Cu was also found near the surface and mainly within a particle depth of up to 100 μm. The more mobile elements, Cd, Ni, and Zn, were mostly found in the depth range of 40–300 μm. In particular, Ni diffused into the whole particle (40–1000 μm) in large amounts compared with the other heavy metals. The distribution of the heavy metals was displayed for a high resolution of the first 130 μm in Fig. 4. Therefore, most heavy metals diffused into the pores, where they were retained at internal sites. The higher decrease of Ca and Mg at and near the surface indicates surficial and some internal precipitation and ion exchange. The evaluation method for the LA-ICP-MS tests, as well as the complete results, will be published elsewhere.

During the preloading experiment, adsorption and precipitation likely took place first in the outer porous system because of the low diffusion coefficients (Sperlich et al., 2005). With more and more heavy metals retained, these metals diffused into the particle. A temporary desorption of a heavy metal and its intra-particle transport to an adjacent site was also possible. Since intra-particle pore and surface diffusion are rate-limited (Badruzzaman et al., 2004), the removal of FerroSorp decreased although the inner porous system was not saturated. The rate-limitation of some processes and the differences between the metals were also detected by the increased removal efficiencies after the weekends without flow (Fig. 1). The generally high removal efficiencies might be related to the large macropores and mesopores of the material, which are beneficial for high adsorption kinetics (Badruzzaman et al., 2004).

3.2.4. GAC

This material is based on coconut shells and is activated by steam and an alkaline reaction to obtain a large surface area (986 m^2/g). It mainly consists of TC, K, Si, and Ca. This used GAC has large contents of micropores (0.483 cm^3/g) and mesopores (0.071 cm^3/g) (Kaya et al., 2008) and these physical characteristics support the adsorption of substances, which primarily takes place in these small pores. Nevertheless, its chemical structure affects its interaction with inorganic compounds. During the heavy metal removal experiment, the Ca content decreased to 0.04%, which can be linked to ion exchange and/or precipitation. Largitte et al. (2014) tested a similar GAC for the removal of Pb, and the precipitation as carbonates, oxalates, oxides, and/or hydroxides was determined. In addition, metals can be adsorbed to GAC by weak intermolecular (van der Waals forces) and electrostatic interactions (ionic metals and surface oxide groups). No heavy metals were detected by SEM/EDX on the surface, indicating removal of the metals in the pores by a film diffusion-controlled adsorption mechanism as proposed by Kaya et al. (2008) for this GAC. However, a distinct determination of the removal mechanisms is not possible because of the heterogeneity and small amounts of functional groups in combination with other cations on the surface.

3.2.5. GFH

For this poorly crystallized β-FeOOH, the point of zero charge (pH_{pzc}) was determined to be 7.5–8.0 (Sperlich et al., 2005). Thus, at pH values of 4.0–4.1, the surface of the GFH was positively charged, which does not promote metal cation removal by electrostatic interactions (all metals were measured to be free ions in the influent at pH 5). In addition, no significant precipitation could occur at the measured pH values and only a slight removal of Pb was determined, probably by sorption. The low pH might be linked

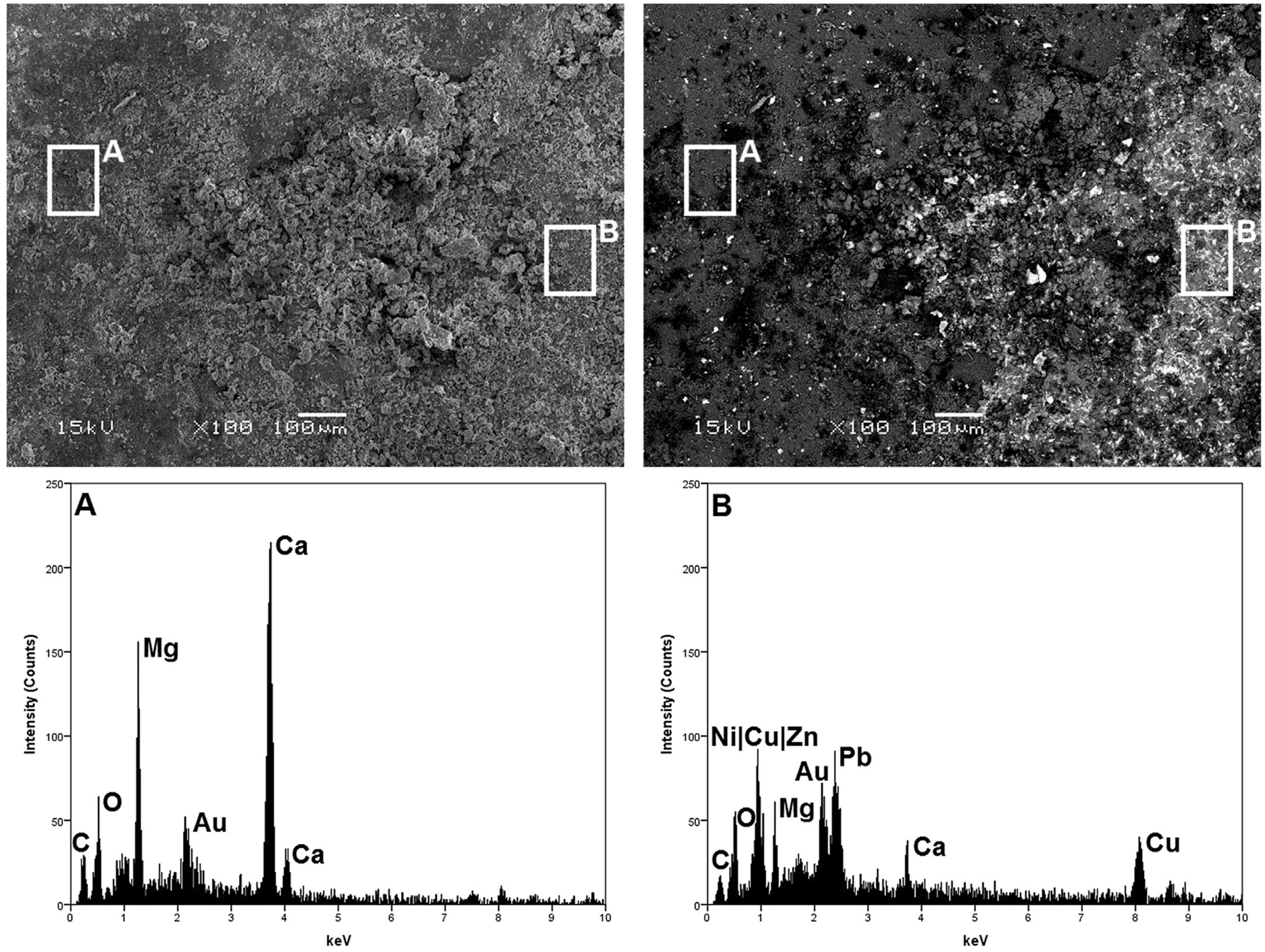

Fig. 3. Analyses of preloaded dolomite (SEM/EDX): detection of secondary electrons (top left), of backscattered electrons (top right), and elements (below).

to the absence of $CaCO_3$, which is often added to GFH. However, this GFH was selected to determine the influence of $CaCO_3$ in ferric hydroxides on the removal and subsequent remobilization of heavy metals.

3.2.6. Lignite

This lignite was activated by a rotary-hearth furnace process that increased the carbon content, surface area, and mesopore volume and decreased the number of functional groups (Murakami et al., 1995). The used lignite mainly consisted of 89.2% TC, 5.15% O, 2.27% Ca, 0.88% Mg, 0.46% TS, 0.26% N, 0.25% Na, and 0.22% H. By XRD analysis, the presence of $CaCO_3$ was determined and Ca was detected on the surface in agglomerates using SEM/EDX. No functional groups were detected via IR spectroscopy. Qi et al. (2011) also described IR spectroscopy curves showing no specific absorption bands for a steam-activated coconut shell with an O content of 3.8%. Only a large variation between the blank and the preloaded samples was measured for the Ca and Na contents (preloaded sample: 1.66% Ca and 0.06% Na). The effluent concentrations were approximately 41 mg/L Ca and 9.7 mg/L Na at the beginning of the preloading test and <1.0 mg/L Ca and <1.0 mg/L Na at the end. Thus, the exhaustion of the heavy metal removal correlated with the decrease of Ca and Na. Therefore, the retention mechanisms were mainly precipitation (e.g., as carbonates or sulfur compounds) and ion exchange. The physical characteristics (cf. Table 2) also enable the removal of heavy metals by adsorption.

3.3. Remobilization

Remobilization of a heavy metal is defined as a direct or indirect metal release during de-icing salt application. The main processes during these experiments were the complexation of heavy metals with Cl^-, ion exchange, competition of de-icing salt cations with positively charged metal species for sorption sites on the filter material, and change of metal species due to decrease of the pH value (Bäckström et al., 2004; Norrström, 2005; Acosta et al., 2011). Consequently, the important factors are the pH value and the retention sites (Nelson et al., 2009).

Each remobilization test represented one realistic traffic area runoff event on a short-term basis (91 bed volumes). The objective was to differentiate between heavy metals, de-icing salts, and filter materials, rather than to determine the total amount of metals that could be remobilized. The results for the amounts of heavy metals measured in the effluent per test are presented in Fig. 5. The presented amounts are based on the individual filter material masses of each column.

Alumina had low remobilized amounts with no significant differences between the heavy metals. The highest measured effluent

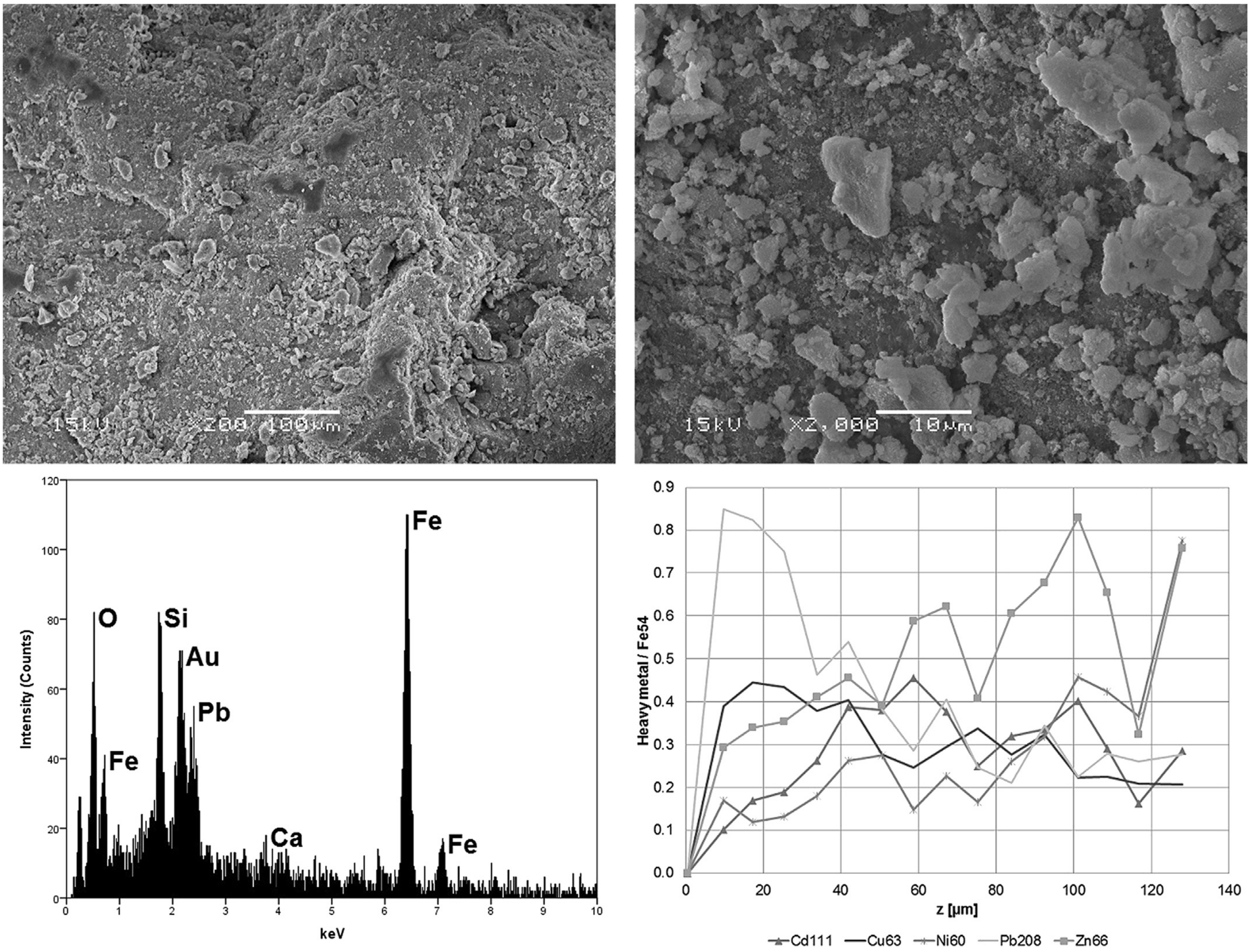

Fig. 4. Analyses of high-preloaded FerroSorp (SEM, EDX, and LA-ICP-MS): detection of secondary electrons (top), detection of elements on the surface (bottom left), and the results of the LA-ICP-MS for the qualitative heavy metal retention as a function of intra-particle attenuation (bottom right; normalized by Fe, which was evenly distributed; verified by Si).

concentrations (the average of each sampling for the two columns) were 119 µg/L Cd (NaCl + $CaCl_2$), 87 µg/L Cu (NaCl + $CaCl_2$), 71 µg/L Ni (NaCl + $MgCl_2$), 96 µg/L Pb (NaCl + $CaCl_2$), and 220 µg/L Zn (NaCl + $CaCl_2$). All values originated from the first sampling. The average effluent pH values were 9.0 (NaCl), 8.5 (NaCl + $MgCl_2$), and 8.1 (NaCl + $CaCl_2$). The lowering of the pH could be a response to ion exchange (Bäckström et al., 2004). For all tests, Ni, Cd, and Pb were mainly dissolved (>95%). By contrast, Cu and Zn were more particulate (0–44%).

Dolomite had moderate remobilized amounts with the exception of a high Pb release. The highest measured effluent concentrations (averages of each sampling) were 372 µg/L Cd (NaCl + $MgCl_2$), 292 µg/L Cu (NaCl + $CaCl_2$), 141 µg/L Ni (NaCl + $CaCl_2$), 791 µg/L Pb (NaCl + $CaCl_2$), and 375 µg/L Zn (NaCl + $CaCl_2$). All values originated from the first two samplings. The average effluent pH values were 10.3 (NaCl), 9.9 (NaCl + $MgCl_2$), and 10.3 (NaCl + $CaCl_2$). For most samples, Cd and Pb were mainly dissolved (>90%) and all other metals were more particle-bound with high variations, in particular Cu with a particulate fraction of up to 99%. Modeling with PHREEQC determined that Cd was mostly complexed with Cl^-, with less than 10% existing as free ion. Acosta et al. (2011) also found that Cd and Pb were very mobile in the presence of $CaCl_2$ because they were more efficiently replaced by Ca and more readily complexed with Cl^- than Cu or Zn. These effects were similarly measured for NaCl with lower released metal concentrations. A generally higher remobilization of Pb, which was previously precipitated on the surface, was also determined for soils with increasing carbonate contents (Acosta et al., 2011).

FerroSorp (high preloading) showed a high remobilization, in particular for Ni. The highest measured effluent concentrations (average of each sampling) were 1720 µg/L Cd, 157 µg/L Cu, 4010 µg/L Ni, 91 µg/L Pb, and 1990 µg/L Zn. The values originated from the first and last samplings for the NaCl + $CaCl_2$ test, which also indicated different mechanisms or rate-limitation processes (e.g., diffusion). Nevertheless, Ni was mostly remobilized at the beginning, although it was mainly bound within the particle (cf. Fig. 4). The difference between the Cd remobilization by NaCl + $CaCl_2$ and the other de-icing salts was highest for FerroSorp, indicating that a cation exchange by Ca (both having similar ionic radii) was likely (Amrhein et al., 1992). The average effluent pH values were 7.4 (NaCl), 7.2 (NaCl + $MgCl_2$), and 6.9 (NaCl + $CaCl_2$). For all tests, all five heavy metals were mainly dissolved (>95%). For the tests with NaCl + $MgCl_2$ and NaCl + $CaCl_2$, higher particulate fractions of Cu (7–23%), Pb (19–47%), and Zn (5–8%) were measured at the last sampling (200 min) with no differences in the effluent pH values

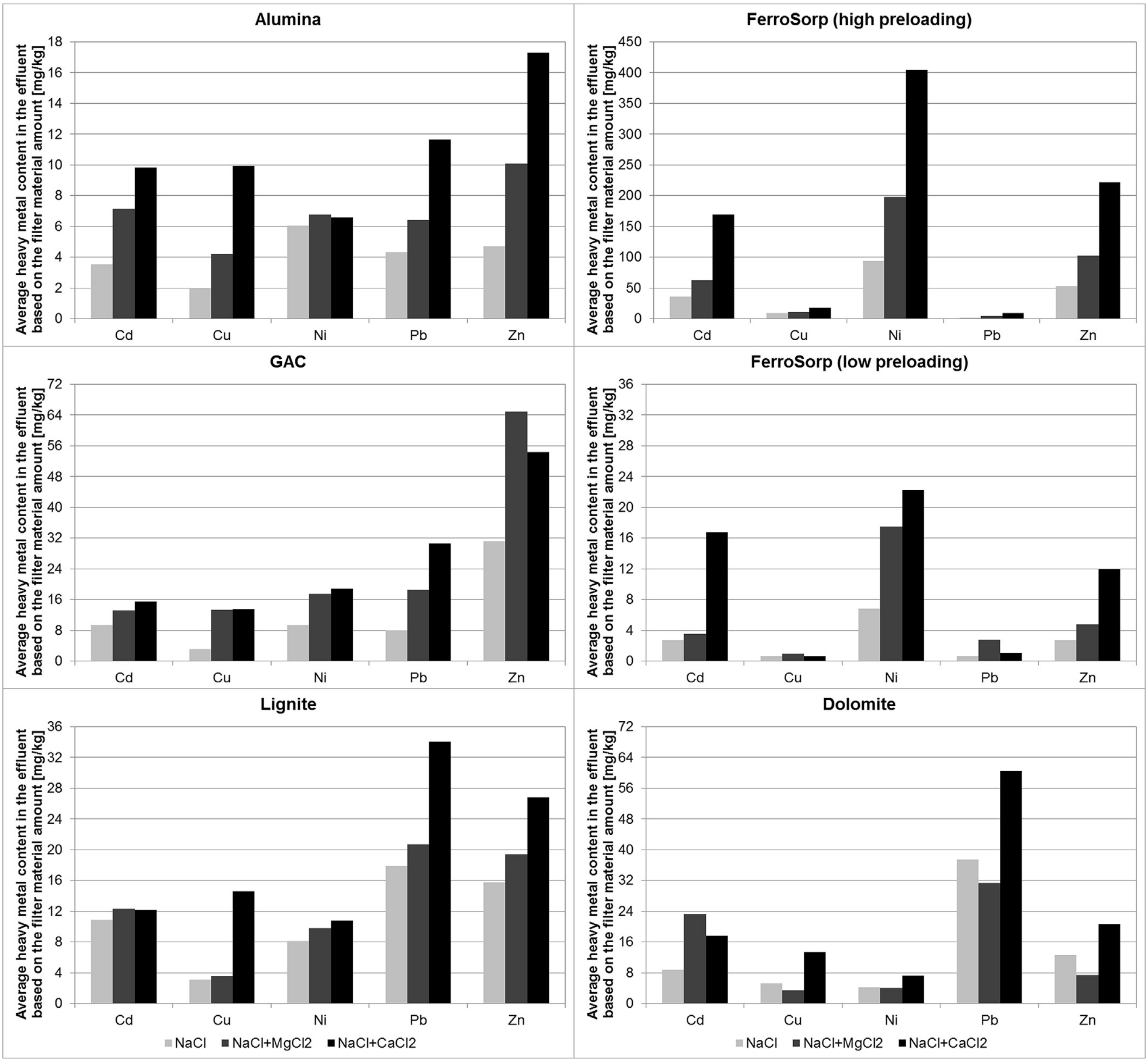

Fig. 5. De-icing salt experiments: amounts of heavy metals measured in the effluent, calculated as the average of both columns for de-icing salt tests and based on the filter material mass of each column (i.e., eight water samples per salt test that are represented for each metal in one bar). For FerroSorp, both preloading levels were evaluated.

compared with earlier samplings.

GAC, which had previously retained low amounts of heavy metals (Fig. 2), showed mostly moderate remobilized amounts. The highest measured effluent concentrations (average of each sampling) were 143 μg/L Cd, 116 μg/L Cu, 150 μg/L Ni, 163 μg/L Pb, and 418 μg/L Zn. All values originated from the first sampling for the NaCl + $CaCl_2$ test. The average effluent pH values were 7.6 (NaCl), 7.5 (NaCl + $MgCl_2$), and 7.6 (NaCl + $CaCl_2$). For all tests, Cd was mainly dissolved (>93%), followed by Zn, Ni, Pb, and Cu (52–91%).

Lignite also had moderate remobilized amounts. The highest measured effluent concentrations (averages of each sampling) were 107 μg/L Cd, 197 μg/L Cu, 97 μg/L Ni, 210 μg/L Pb, and 229 μg/L Zn. All values originated from the first sampling for the NaCl + $CaCl_2$ test. The average effluent pH values were 9.5 (NaCl), 9.1 (NaCl + $MgCl_2$), and 9.2 (NaCl + $CaCl_2$). For all tests, Cd was mainly dissolved (>95%) and all other metals were more particle-bound with high variations.

In summary, each filter material behaved differently during the de-icing salt experiments because of their differing chemical compositions. Concerning the heavy metals, Cu was mostly released the least by the three de-icing salts, followed by Pb. The most mobile metal was Cd, which had the highest dissolved fraction and total remobilized amounts of previously retained metals of 3.0% (alumina), 4.7% (dolomite), 6.3% (FerroSorp, high preloading), 21.7% (GAC), and 5.8% (lignite) during the NaCl + $CaCl_2$ tests. For dolomite, this percentage was also very high for Pb (4.5%) during the NaCl + $CaCl_2$ experiment. The Cd values were higher for the NaCl + $MgCl_2$ experiments for dolomite (6.1%) and lignite (5.9%). The high affinity of Cd for forming highly soluble complexes with Cl^- has been reported in previous studies (e.g., Norrström, 2005;

Nelson et al., 2009), and this resulted in the measured Cd remobilization. Because of the relatively small amounts of released metals, the differences of metal attenuation as functions of depth before and after the de-icing salt tests could not be experimentally determined. The pH values were always the highest for the NaCl tests and often the lowest during the NaCl + $CaCl_2$ experiments, indicating a correlation between remobilization and pH decrease. However, the absolute pH values were different between the materials. For a comparison between the filter materials, no correlation of the pH values and the remobilized amounts was determined for each de-icing salt. A correlation between remobilization and the removal mechanism was also not found because of the large diversity of mechanisms and their different combinations for each filter material. In most cases, NaCl + $CaCl_2$ application resulted in the highest remobilization and pure NaCl application resulted in the lowest. The difference between the remobilizations by Ca and Mg was likely because of their differences in size and atomic mass; Ca is larger and heavier than Mg and probably replaces heavy metals faster and more easily (Acosta et al., 2011). In addition, Ca and Mg can compete more effectively for cation exchange sites than the monovalent Na (Amrhein et al., 1992). Consequently, all de-icing salts had an impact on the high-preloaded filter materials and the effluent concentrations could be very high.

3.4. *Effect of the preloading level on the remobilization*

The effect of the preloading level was investigated for FerroSorp because it had the highest removal capacity. The heavy metal capacities of the low-preloaded FerroSorp were lower by a factor of approximately 5 than those of the high-preloaded filter material (Fig. 2). For all remobilization tests, the effluent concentrations of the high-preloaded material were much higher than those of the low-preloaded filter material. For high/low-preloading experiments, the maxima of the average concentrations per test were 1240/123 µg/L for Cd, 128/6.8 µg/L for Cu, 2960/163 µg/L for Ni, 63.0/20.1 µg/L for Pb, and 1620/87.5 µg/L for Zn. All maxima were obtained for the tests with NaCl + $CaCl_2$, with the exceptions of Cu and Pb for the low-preloading experiment (i.e., the remobilization by NaCl + $MgCl_2$). Thus, high-preloaded filter materials released higher heavy metal concentrations under the same simulated de-icing salt runoff event than low-preloaded materials, and these concentrations can be relevant for the evaluation of toxicity effects. The average pH values of the effluent samples for the high/low-preloading experiments were 7.4/9.1, 7.2/8.7, and 6.9/8.1 for the tests with NaCl, NaCl + $MgCl_2$, and NaCl + $CaCl_2$ (with comparable influent pH values of 7.0, 6.9, and 8.2), respectively. The highest ratios of remobilized heavy metals (Fig. 5) to previously retained metals (Fig. 2) were 6.3%/4.2% Cd, 0.1%/0.1% Cu, 4.6%/1.2% Ni, 0.1%/0.2% Pb, and 1.1%/0.4% Zn for the high/low-preloading experiments. Therefore, significantly higher fractions of previously retained metals were remobilized from the higher preloaded material for Cd, Ni, and Zn.

The heavy metal contents of these manufactured filter materials were much higher than the contents of roadside soils that have been evaluated by other researchers for heavy metal remobilization. For example, Acosta et al. (2011) tested nine soils with heavy metal contents of 1.07–14.1 mg/kg Cd, 15–89 mg/kg Cu, 31–2764 mg/kg Pb, and 34–401 mg/kg Zn. For $CaCl_2$, a total release of Cd of up to 4.1% was measured. It decreased for $MgCl_2$ (3.0%) and NaCl (1.8%) for the high ionic strength of each de-icing salt (0.3 M). The releases of Pb and Cu were smaller and very low for Zn (0.0–0.2%). Thus, Cd was previously retained less and had the highest remobilization ratios, whereas Cu was highly retained and subsequently less remobilized (Acosta et al., 2011). Bauske and Goetz (1993) reported similar results with a release of 15.7% Cd and 1.9% Zn by NaCl + $CaCl_2$ from soil containing 2.99 mg/kg Cd and 658 mg/kg Zn. Norrström (2005) evaluated the remobilization of Cd, Cu, Pb, and Zn from two soils from infiltration trenches for highway runoff (1.5–2.7 mg/kg Cd, 155–194 mg/kg Cu, 171–324 mg/kg Pb, and 607–781 mg/kg Zn). In total, 37–45% of Cd, 0.1–0.2% of Pb, and 4.7–5.0% of Zn were leached by NaCl. Thus, higher fractions of the more mobile elements were remobilized. Despite the influences of the test conditions (e.g., ionic strength, durations, batch or column experiments), similar trends were detected.

To exclude the effects of different preloading levels, a comparison of FerroSorp (low preloading) with alumina and lignite, which were similarly preloaded (Fig. 2), was performed. The lowest/highest releases of heavy metals from these three materials were measured as 0.7%/6.3% of Cd, 0.0%/1.2% of Cu, 0.4%/1.3% of Ni, 0.1%/2.4% of Pb, and 0.1%/1.3% of Zn for all de-icing salt tests. The lowest percentages were all determined for the experiment with FerroSorp/NaCl and the highest rates for lignite/NaCl + $CaCl_2$. However, for some combinations of heavy metal and de-icing salt, the least remobilization was determined for alumina.

4. Conclusions

- The differentiated approach of this study made it possible to elucidate the removal and remobilization of five heavy metals and six filter materials in lab-scale column experiments, both against three different de-icing salts.
- The removal efficiencies of each heavy metal and their retention mechanisms varied for all filter materials. Cu and Pb were retained best except in the case of half-burnt dolomite.
- Concerning the de-icing salt experiments, the results were more heterogeneous. The behavior of each filter material, each heavy metal, and each de-icing salt was different. In general, Cu was released the least by the three de-icing salts. The NaCl + $CaCl_2$ mixture resulted in a stronger effect on remobilization than the other two de-icing salts. Pure NaCl had a minor effect on the remobilization for most heavy metals and filter materials. A correlation between the heavy metal solid content and the remobilization level was found.
- The use of a mixture of different filter materials might be beneficial for reducing remobilization under all three different de-icing salts.
- Findings of this research can be utilized in the development and improvement of decentralized stormwater treatment systems using filter materials. By using these improved materials, a more environmentally friendly operation of these systems for traffic area runoff can be ensured.

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Chapter 10: Conclusions, Impact, and Novel Research Topics

10.1 Highlights of the Research Results

For the first time, the pollution of different traffic area runoff categories (i.e., parking lots, bridges, and three types of both highways and roads) was summarized in an extensive literature study for total and dissolved heavy metal runoff concentrations. Therefore, a large data set ($n = 294$) of validated dissolved and total metal concentrations in traffic area runoff was created and analyzed. The calculated fractionation of heavy metals helped to obtain dissolved parts of the most relevant metals Cd, Cr, Cu, Ni, Pb, and Zn, which can be used to evaluate toxic effects and the performance of stormwater treatment systems. In addition, runoff concentrations of further traffic related metals, anthropogenic metals, and de-icing salt compounds were summarized. A description of site-specific and method-specific factors was presented to evaluate their influences on the monitoring results. Analyses of historical and continental trends were performed and the reduction in Pb concentrations over time was one of the robust chemical results. Moreover, the annual heavy metal loads normalized per hectare of impervious catchment area were summarized and used to calculate mass balances.

The second research topic addressed the removal of heavy metals from traffic area runoff. The heavy metal retention of different filter materials was determined by lab-scale, pilot-scale, and full-scale experiments. At the beginning, method-specific differences of heavy metal capacities were determined by an analysis of the influences of various experimental setups. After the comparison of the results of four different lab-scale methods for metal removal (Cu, Ni, and Zn), the suitability of different methods was evaluated by testing six filter materials for stormwater treatment with diverse properties. As a result, the following recommendations were given: a guideline for manufacturers was proposed with a well-defined description of batch experiments for internal and external quality control that could be used for testing different production batches but also for selecting appropriate filter materials for subsequent column experiments. Further recommendations included testing the removal efficiencies and service life by using small-scale column experiments. In addition, a pilot-scale performance assessment method was developed to determine the service life of various filter materials for dissolved substances. The novel method is applicable to a wide range of filter systems (i.e., channel systems, shaft systems, and permeable pavements) and allows a rapid evaluation (within a few days) of long-term performances and service lives of all of these manufactured decentralized systems. The determined removal efficiencies can be compared with the presented assessment criteria based on stormwater treatment guidelines. Furthermore, the novel method was validated by field measurements of two different filter channel systems at four sites and it can be extended for further dissolved substances (e.g., nutrients).

The third topic of the development of an assessment method for decentralized stormwater treatment systems for runoff from traffic areas included the evaluation of the heavy metal remobilization by various de-icing salts. At the beginning, the composition of the de-icing salts used by winter services was evaluated in order to identify realistic compositions for further tests. In Germany, the de-icing salts consist of different types of compounds (e.g., sodium chloride, a mixture of sodium chloride with calcium chloride, and a mixture of sodium chloride with magnesium chloride). An assessment method was developed that considered all three de-icing salts and five heavy metals (Cd, Cu, Ni, Pb, and Zn). As a result, the behavior was different for each of the six filter materials tested, each heavy metal, and each de-icing salt. In general, Cu was released the least from most of the selected filter materials by the three de-icing salts. Compared with the other two de-icing salts, the mixture of sodium chloride and calcium chloride resulted in the strongest effect on remobilization. Pure sodium chloride had a minor effect on the remobilization for most heavy metals and filter materials. In addition, higher preloaded filter materials released more heavy metals than less preloaded filter materials. After completing the lab-scale column experiments, advanced analyses were conducted with the unloaded and loaded filter materials to specify the attenuation of heavy metals as a function of depth and to describe the removal mechanisms. In addition, computer modeling was used to support the findings of the lab-scale column experiments by simulating the speciation of heavy metals.

The results of this research can be implemented into the development and improvement of manufactured stormwater treatment systems using filter materials. Both novel assessment methods can be used to evaluate new and already existing decentralized systems for stormwater treatment. By these findings, a more environmentally friendly operation of these systems for traffic areas can be ensured and a longer service life is possible.

10.2 Improvement of the State of the Art

The pollution classification of traffic areas by three categories was only verified by some research projects (e.g., Grotehusmann & Kasting, 2002; Steiner et al., 2006; Nadler & Meißner, 2007). As novel contributions, the reviews differentiated the pollution of various traffic area categories for both European and worldwide data. The results can be used to update existing rules and standards and to emphasize the monitoring of runoff from highly polluted sites (e.g., sites with stop-and-go traffic, crossings, and entries/exits of parking lots at supermarkets). In addition, the better estimation of the pollution of different traffic areas supports the use of appropriate treatment systems that are neither oversized nor have an insufficient treatment. Thus, the initial costs and the maintenance are reduced.

The critical review can also be used to identify all relevant site-specific and method-specific factors of monitoring programs concerning stormwater runoff. Because these factors have an influence on the results, it is important for research reports to record and present site-specific and method-specific data in an appendix. As a result of the analysis of different monitoring programs, it is also helpful to present the complete runoff data sets together with the rain characteristics because they are required for data evaluation as the authors often have a different objective compared with other researchers. In addition, the method-specific factors that were summarized and discussed in the reviews can be used to develop novel monitoring schemes.

Previously, the experimental results of manufacturers were not comparable because the influences of different lab-scale boundary conditions on the heavy metal removal were not summarized or evaluated (cf. Section 2.2.2). Based on the new results regarding stormwater treatment systems with filter materials, standardized methods for quality control, selection of filter materials, and performance evaluation are available.

For the determination of service lives and long-term performances, no assessment method or criteria had been proposed by authorities (cf. Section 2.2). By developing the novel test method, a standardized and more accurate estimation of service intervals of decentralized treatment systems was implemented. The method also provides regulatory authorities, designers, and operators with a more objective basis for the performance assessment and supports stormwater managers to make decisions for the installation of manufactured decentralized treatment systems.

In recent years, the composition of de-icing salts used in experiments was mainly based on information provided by German state authorities, which use sodium chloride in most cases. Consequently, only the effect of sodium chloride on previously retained Cu and Zn is currently considered in authorities' test procedures (DIBt, 2012; DIBt, 2015; ÖNORM B 2506-3 Entwurf, 2015). However, as a result of the survey, municipalities use different types of de-icing salts on urban traffic areas. This is important for manufactured decentralized stormwater treatment systems because they are mainly installed at these sites. The differentiated approach of the second assessment method enabled an evaluation of the effect of different de-icing salts on all relevant heavy metals for six commercially used filter materials. The results established an understanding of the different mechanisms and will lead to the development of filter materials that are less affected by a remobilization of pollutants due to the three tested de-icing salts.

10.3 Impact of the Results on other Projects

Several results of the research project presented in Section 7.1 were implemented in an update of the DIBt procedure (DIBt, 2011; DIBt, 2015). For example, the rain duration and its intensity for the test with de-icing salts was changed to more realistic values (200 min with 6.0 L/(s·ha) instead of 48 min with 25 L/(s·ha)). In addition, the current procedure of the DIBt (2015) only allows the use of deionized water for all experiments instead of drinking water to avoid the possibility of different results from various institutes. The novel method for the determination of service lives and long-term performances was successfully applied by manufacturers of decentralized stormwater treatment systems and is currently under consideration for implementation into the approval process of the DIBt.

The method developed in this thesis for the evaluation of the service lives and long-term performances of decentralized systems was also applied in another research project to test the removal of dissolved substances (i.e., Cu, TP, and Zn) by a semi-decentralized treatment plant. The results will be used to compare the pilot-scale performances determined in the laboratory with field measurements. Such a testing procedure, which also includes TP and/or TN, might be beneficial to support the export of manufactured decentralized treatment systems into other countries (e.g., Australia and USA).

The evaluation of the use of different de-icing salts by German authorities affected the experimental setup of other research projects. For example, Barjenbruch et al. (2016) determined the influence of a mixture of sodium chloride and calcium chloride on previously retained Cu and Zn for two different decentralized treatment systems in the laboratory according to the new findings presented in Huber et al. (2015b).

The summary of the influences of method-specific factors on the measurements (cf. Chapter 5) was used to improve the setup of a monitoring campaign at two sites in NRW, Germany, which consisted of the measurement of the road runoff and the effluent water quality of a decentralized stormwater treatment system (parameters: Cu, ETBE, MTBE, PAH, TPH, TSS, and Zn). The summary of site-specific factors was used to describe the influences of both catchment areas on the pollution. The results of this monitoring are presented in Wichern et al. (2015).

10.4 Suggestions for further Research

The composition of the feed water has a strong effect on the results of all types of experiments (e.g., batch experiments, lab-scale column experiments, and pilot-scale experiments) as the results of Huber et al. (2014a), Welker et al. (2014), and Wichern et al. (2015) indicate. An effect of ionic strength on the heavy metal removal was mainly measured for decentralized systems with filter materials using ion exchange as the main removal mechanism and for filter materials with limited capacities. For example, the effect of using drinking water (approximately 300–500 µS/cm) instead of deionized water (< 1 µS/cm) was tested by two different experimental setups. For the determination of the breakthrough curves of Cu and Zn, drinking water had a strong effect on the removal capacities (Wichern et al., 2015). Thus, the capacities decreased significantly for the experiments with drinking water compared with deionized water. Moreover, a higher remobilization of Cu and Zn was measured for several de-icing salt tests with drinking water instead of deionized water (Huber et al., 2014a). To exclude this influence, deionized water was used in the experiments presented in this thesis. However, the effect of ionic strength should be considered in a further study because institutes for internal and external quality control often have “deionized” water with an electric conductivity of > 1 µS/cm that can have an effect on the results. In addition, not only the electric conductivity has an influence on the results but also the composition of the feed water. For example, two feed waters with an electric conductivity of approximately 200 µS/cm were compared for the removal of Cu and Zn by the same filter material (Welker et al., 2014). One of the feed waters consisted of deionized water with a defined amount of calcium and the other of deionized water with a defined amount of magnesium. The filter material evaluated with the calcium containing feed water showed a decreased retention of Cu and Zn. This effect is also important for the novel assessment method (Huber et al., 2016d) if more dissolved substances are tested simultaneously (e.g., heavy metals with TN, TP, or trace organic substances). For example, in the presence of P, the retention of Cu and Zn increased and the remobilization of both metals decreased. In addition, the influences of pretreatment before the first experimental run (e.g., flushing of the filter model) and of the breaks between different experimental runs should be addressed in a further research project.

The relevant dissolved heavy metals (Cu and Zn) are considered in the novel method to determine the service lives and long-term performances (cf. Chapter 7). Moreover, the experimental setup of the assessment method allows for testing of further dissolved substances, also simultaneously, because different requirements are proposed by the authorities (cf. Table 5). In the future, further parameters such as trace organic substances (e.g., MTBE and ETBE) might be added because of new legislative requirements (cf. Section 1.3). Thus, an implementation of further substances into the novel methods should be carried out in the future in accordance with legal requirements and new monitoring results.

The novel method for the determination of service lives should also be applied to permeable pavements to assess their performances. This evaluation can be used to compare the long-term performances of all three types of decentralized treatment systems with filter materials. In addition, modified permeable pavements might also be beneficial in terms of water balances (Dierkes & Lucke, 2015).

As mentioned in Section 2.2.1, this novel method cannot consider all boundary conditions that have an influence on the service life, such as a limitation due to a hydraulic failure of the plant. To determine this factor, decentralized systems must be monitored at several sites over periods of at least two years. Thus, a full-scale monitoring of tested treatment systems should be performed with a special focus on the service life of permanently submerged systems, the maintenance, and the measurement of further substances. It should monitor additional substances such as de-icing salt compounds, fine particles, and new priority pollutants, which might be higher than the values of the updated GFS of 2013 (groundwater) (Moll & Quadflieg, 2014) or of the environmental quality standards (surface water) (DIRECTIVE 2013/39/EC, 2013). These substances should be monitored because the necessity for treating these trace organic substances in runoff from traffic areas cannot be evaluated currently due to a lack of monitoring data. These parameters are antimony, benzo(a)pyrene, benzo(g,h,i)-perylene, cyanide, cobalt, di(2-ethylhexyl)-phthalate (DEHP), diuron, fluoranthene, mercury, and vanadium. In urban runoff, brominated diphenylethers, nonylphenols, and octylphenols might also occur in relevant concentrations (Stachel et al., 2007; Grotehusmann et al., 2014). By using a well-known location (e.g., "Landshuter Allee" in Munich), a long-term monitoring of this site can be used to characterize the less reported annual pollution loads (kg/ha) for heavy metals (cf. Section 5.3). In addition, the monitoring-site can also be used to verify the influences of different site-specific factors (e.g., road maintenance and climatic factors) because this site was used in three earlier research programs (Hilliges et al., 2007; Helmreich & Horn, 2009; Helmreich et al., 2010; Hilliges et al., 2013). Furthermore, the results of Chapter 5 might be beneficial for developing the monitoring setup.

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Appendix

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1. Huber, M., Welker, A., Dierschke, M., Drewes, J.E., Helmreich, B., 2014. Ein neues Laborverfahren zur Ermittlung von Standzeiten dezentraler Anlagen zur Behandlung von Verkehrsflächenabflüssen. gwf Wasser/Abwasser 155 (5), 630–638.
2. Hinken, L., Huber, M., Weichgrebe, D., Rosenwinkel, K.-H., 2014. Modified ADM1 for modelling an UASB reactor laboratory plant treating starch wastewater and synthetic substrate load tests. Water Research 64, 82–93.
3. Huber, M., Welker, A., Helmreich, B., 2015. Belastung von Verkehrsflächenabflüssen mit Schwermetallen – ein europäischer Vergleich. gwf Wasser/Abwasser 156 (9), 896–909.
4. Huber, M., Welker, A., Drewes, J.E., Helmreich, B., 2015. Auftausalze im Straßenwinterdienst – Aufkommen und Bedeutung für dezentrale Behandlungsanlagen von Verkehrsflächenabflüssen zur Versickerung. gwf Wasser/Abwasser 156 (11), 1138–1152.
5. Helmreich, B., Huber, M., Drewes, J.E., 2015. Schwermetalle in Metalldachabflüssen und Möglichkeiten zur dezentralen Behandlung. gwf Wasser/Abwasser 156 (12), 1238–1245.
6. Huber, M., Welker, A., Helmreich, B., 2016. Critical review of heavy metal pollution of traffic area runoff: Occurrence, influencing factors, and partitioning. Science of the Total Environment 541, 895–919.
7. Huber, M., Badenberg, S.C., Wulff, M., Drewes, J.E., Helmreich, B., 2016. Evaluation of Factors Influencing Lab-Scale Studies to Determine Heavy Metal Removal by Six Sorbents for Stormwater Treatment. Water 8 (2), 62:1–19.
8. Horstmeyer, N., Huber, M., Drewes, J.E., Helmreich, B., 2016. Evaluation of site-specific factors influencing heavy metal contents in the topsoil of vegetated infiltration swales. Science of the Total Environment 560–561, 19–28.
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10. Huber, M., Welker, A., Dierschke, M., Drewes, J.E., Helmreich, B., 2016. A novel test method to determine the filter material service life of decentralized systems treating runoff from traffic areas. Journal of Environmental Management 179, 66–75.
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Monograph

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Conference Talks

1. Huber, M., 2014. Durchführung von Laborversuchen zur anaeroben Industrieabwasserbehandlung und Modellierung der Ergebnisse mit dem ADM1. Forum „Young Scientists“. 47. Essener Tagung, Essen, 19–21 March 2014.
2. Welker, A., Huber, M., 2014. Dezentrale Behandlungsanlagen für Verkehrsflächenabflüsse – Stand der Technik. 42. Abwassertechnisches Seminar: Zukunftsfähige Bewirtschaftungskonzepte für Niederschlagswasser, Garching, 10 July 2014. Berichte aus der Siedlungswasserwirtschaft, Technische Universität München, Vol. 211, ISSN 0942-914X, pp. 89–106.
3. Huber, M., Dierschke, M., Welker, A., Helmreich, B., 2014. Laborverfahren zur Ermittlung von Standzeiten von Anlagen zur Behandlung des Niederschlagswassers von Verkehrsflächen. Aqua Urbanica 2014, Innsbruck (Austria), 23–24 October 2014. ISBN 13: 978-3-902978-28-8.
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8. Vesting, A., Huber, M., Giga, A., Helmreich, B., Wichern, M., 2015. Erfahrungen aus Praxisuntersuchungen eines dezentralen Behandlungssystems zur Reduktion von Kohlenwasserstoffen und organischen Spurenstoffen aus Verkehrsflächenabflüssen. Aqua Urbanica 2015, Stuttgart, 07–08 October 2015. Stuttgarter Berichte zur Siedlungswasserwirtschaft, Vol. 225. ISBN 13: 978-3-8356-7292-5, pp. 131–141.
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10. Huber, M., 2015. Dezentrale Anlagen zur Behandlung von Verkehrsflächenabflüssen. 1. Erfahrungsaustausch SABA ASTRA, Ittigen (Switzerland), 05 November 2015.
11. Huber, M., Fassnacht, J., Hilbig, H., Drewes, J.E., Helmreich, B., 2016. Auftausalzeinfluss auf die Remobilisierung von auf Filtermaterialien zur Behandlung von Verkehrsflächenabflüssen zurückgehaltenen Schwermetallen. Wasser 2016, Jahrestagung der Wasserchemischen Gesellschaft, Bamberg, 02–04 May 2016. ISBN 13: 978-3-936028-94-2, pp. 421–426.
12. Huber, M., Hilbig, H., Drewes, J.E., Helmreich, B., 2016. Heavy Metal Removal and Remobilization under De-icing Salt Applications using Lab-scale Column Experiments. NOVATECH-Conference, Lyon (France), 28 June – 01 July 2016, pp. 1–7.
13. Helmreich, B., Huber, M., Vesting, A., Giga, A., Wichern, M., 2016. Erfahrungen mit einem dezentralen Behandlungssystem für Verkehrsflächenabflüsse – Vom Labormaßstab in die technische Umsetzung. 15. Regenwassertage der DWA, Darmstadt, 05–06 July 2016.
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