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RESEARCH ARTICLE

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# Cost versus environment?

## Combined life cycle, techno-economic, and circularity assessment of silicon- and perovskite-based photovoltaic systems

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## Abstract

Photovoltaics will play a key role in future energy systems, but their full potential may not be realized until their life cycles are optimized for circularity and overall sustainability. Methods that quantify flows of compound and minor element mixtures, rather than non-mixed elemental flows, are needed to prospectively analyze and predict inventory and performance for complex technology life cycles. This study utilizes process simulation to resolve the mass and energy balances needed to rigorously analyze these complexities in circular systems. Using physics-based prospective inventory data, we simultaneously assess the environmental and techno-economic performance of three photovoltaic life cycles and predict the effects of circularity on resource efficiency, carbon footprint, and levelized cost of electricity. One inventory dataset is generated per life cycle to ensure alignment between assessments and to identify trade-offs between environmental and techno-economic performance with respect to circularity, so linking circularity and sustainability. The linked material and energy resource and techno-economic models allow for the impacts of carbon taxation and the moderating effects of circularity to be explored. In addition to the clear environmental benefits of increased circularity, we find that it could dampen the cost impact of taxation. While confirming that perovskite-based modules, single junction or in tandem with silicon, clearly outperform the silicon market standard both techno-economically and environmentally, we show that maximum circularity does not automatically deliver the most sustainable outcome. The approach enables assessment of the combined impacts of specific technological, commercial, and policy choices made by different actors along the photovoltaic value chain. This article met the requirements for a gold–gold *JIE* data openness badge described at http://jie.click/badges.

KEYWORDS
circularity, industrial ecology, photovoltaics, process simulation, sustainability, techno-economics

# 1 | INTRODUCTION

## 1.1 | Current and emerging photovoltaic technologies

Manufacturers of photovoltaic (PV) technology have been successfully improving techno-economic performance with materials and cell architectures that increase power conversion efficiency (PCE), while reducing material consumption and production costs. This has resulted in levelized costs of electricity (LCOE) lower than that of fossil-based power generation (Fraunhofer-ISE, 2021). Wafer-based crystalline silicon (Si) PV, in particular the "passivated emitter rear cell" (PERC) architecture, is expected to remain the market leader for at least the next decade (VDMA, 2021). A promising emerging technology is based on mixed lead (Pb) halide compounds that crystallize in the "perovskite" structure. Perovskite absorbers have seen the steepest rise in PCE with the record single-junction cell efficiency currently 25.7%, up from 14% less than 10 years ago (NREL, 2022). Their high PCE and low cost, among others, allow perovskite cells to compete with existing commercial technologies (Liu et al., 2021). Challenges that still prevent commercialization are the long-term stabilities of some component materials and interfaces. Including a cesium cation ($Cs^+$) in certain perovskite structures has been shown to improve thermal and moisture stability (Saliba et al., 2016). A detailed comparison of PV types, advantages, and disadvantages is provided by Muteri et al. (2020).

The rapid progress in perovskite research has also driven fast development of perovskite-based tandem devices (Werner et al., 2018). Perovskite/perovskite and perovskite/Si tandem configurations achieve higher efficiencies by taking advantage of perovskites' tunable bandgap to better exploit short-wavelength photon energy (Leijtens et al., 2018). In four-terminal tandem configurations, two independently manufactured sub-cells are stacked on top of each other. They can operate independently these to maximize performance (Leccisi & Fthenakis, 2020). With a theoretical four-terminal efficiency limit of approximately 46% (Eperon et al., 2017), these devices exceed the theoretical single-junction limit of 33% (Shockley & Queisser, 1961) by far. Liu et al. (2022) recently reported a $Cs^+$-doped perovskite module efficiency of 21.08%, and Si and perovskite/Si tandem efficiencies are expected to reach 22.2% and 28% by 2031, respectively (VDMA, 2021).

Figure 1 depicts the structures of the (a) perovskite, (b) silicon, and (c) perovskite/silicon tandem modules described above and discussed in this paper.

## 1.2 | End-of-life PV and circular flows

Solar modules deployed in the 1990s, primarily Si based, are reaching the end of their useful lives, with rapid increases in PV waste volumes expected by 2030. It has been estimated that the recyclable materials in end-of-life (EoL) devices accumulated by 2050 could be used to produce about 630 GW of new capacity (IRENA & IEA-PVPS, 2016). That is, if all of it could be recovered at purities high enough for re-use in PV systems. Despite the high energetic and economic value of contained Si, most recycling facilities presently only recover bulk materials like glass cullet, cabling, and aluminum frames (Isherwood, 2022). Integrated processes aimed at recovering Si and other elements are complex, and while some have been demonstrated at laboratory or pilot scale, commercial examples barely exist (Deng et al., 2022). Promising recycling options for perovskites have only been investigated at laboratory scale (Liu et al., 2021). The further development of these processes, complemented by innovative design-for-recycling to simplify dismantling and separation processes, will maximize the quantities and purities of materials brought back into the economy (Norgren et al., 2020). However, the recovery of materials from EoL devices is subject to limits imposed by the laws of physics, including solution thermodynamics. Complete "unmixing" and recovery of individual elements is impossible due to the irreversibility of processes, as described by the second law of thermodynamics. The effects of these limitations on material and energy flows must be analyzed using fit-for-purpose tools to assess the contribution of circular strategies such as recycling to overall sustainability in detail, as circular flows do not necessarily guarantee sustainable outcomes (Geissdoerfer et al., 2017; Korhonen et al., 2018).

Besides the physical limits, circular flows are unlikely to occur unless driven by economic or regulatory incentives. In the Si PV case, for instance, still-low waste volumes and low demand for high-quality integrated recycling have limited investment in innovation and, as a consequence, recycling costs remain high (Cui et al., 2022). With the expected increase in waste volumes, programs like the European Green Deal (European Commission, 2020a) that promote circular economy (CE) and circular business models are important to stimulate investment and accelerate development despite the limited present demand. Also important are regulations that stipulate recovery targets for specific materials and penalize pollutant emissions. The effects of such measures also need to be quantified to assess whether they do, in fact, enhance overall sustainability.

Lindgreen and colleagues highlight the lack of assessment approaches that quantify the links between circularity and sustainability, that is, the environmental, economic, and social impacts of circular strategies. Such approaches are needed to ensure systemic change for sustainable development rather than mere incremental improvements driven by *"promises of economic gains through resource efficiency"* (Lindgreen et al., 2020).

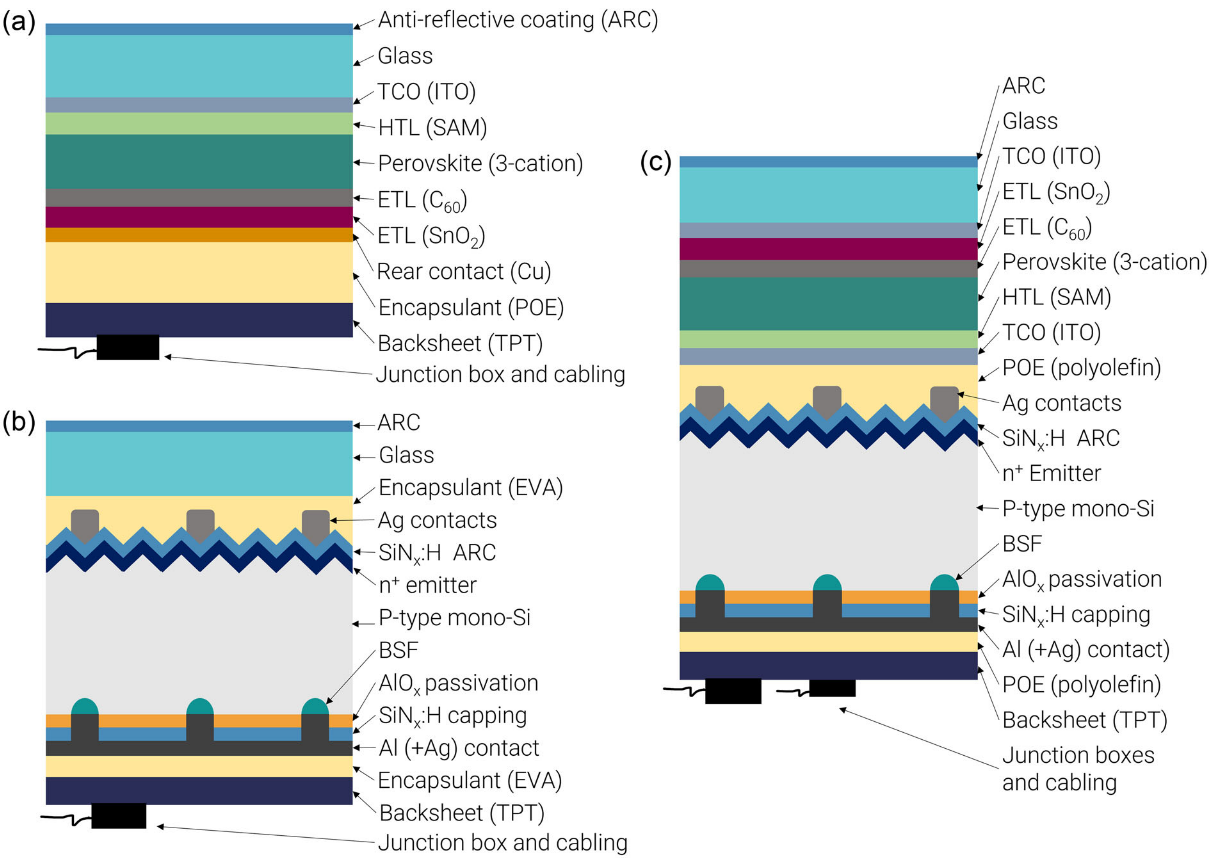

**FIGURE 1** (a) Perovskite (single-junction), (b) Silicon (passivated emitter rear cell architecture), and (c) perovskite/silicon tandem (four-terminal) module configurations. We assume reference power conversion efficiencies of 18%, 21.7%, and 27.3% for perovskite, silicon, and tandem modules, respectively, based on previous publications for similar modules (Sofia et al., 2020). Explanations of the acronyms used in Figure 1 are given in Supporting Information S2.

### 1.3 | Assessing resource, environmental and economic performance

Environmental and economic performance are usually estimated using standardized life cycle assessment (LCA) (ISO, 2006) and techno-economic analysis (TEA), respectively, which are well-known approaches. After goal and scope definition, the foundation of LCA and TEA is the "inventory" of the system at hand, which refers to the mass and energy flows into and out of a system. Material flow analysis (MFA) is most often used and touted as the ideal tool to map life cycle inventory (Graedel, 2019). At its core, MFA refers to doing a mass balance, a fundamental concept that ensures adherence to the law of mass conservation. While suitable for single-element bulk material flows, methodological limitations hamper its use in prospective assessments when not integrated with high-detail approaches like process simulation (Baars et al., 2022). MFA does not suffice when the system includes complex material and minor element combinations such as those found in PV and other technologies, as it does not consider solution chemistry and cannot predict the distribution of minor elements between process outputs (Reuter et al., 2019). Process simulation implicitly performs MFA, but considers the enthalpy, entropy, and thus, free energy of all compounds and solutions, which is necessary to resolve complex mass and energy balances. This allows it to generate physics-based inventory data and gives it predictive capabilities, which is particularly relevant in the context of prospective assessments in which mass and energy flows are not based on historical data but need to be predicted. For these reasons, process simulation is the foundation of the work presented in this paper. More detail is provided in Section 2.2.1.

The integration of LCA and TEA is done in various ways and to various degrees and can enhance decision making in technology development (Wunderlich et al., 2021). Methodological challenges remain, often associated with inconsistent functional units and system boundaries, and discrepancies in assumptions when combining standalone LCAs and TEAs; there is a research gap with respect to tools that simultaneously perform LCA and TEA, while allowing for the influence of changes in process parameters to be investigated (Mahmud et al., 2021). Examples of integrated resource, environmental, and economic performance assessments of PV systems are scarce. Zhang and colleagues compare the environmental impacts and costs of three perovskite technologies to identify material and manufacturing method combinations that could deliver the best environmental and economic performance. The authors identify trade-offs *within* sustainability dimensions and highlight that additional methods are needed to quantify trade-offs *between* them (Zhang et al., 2022).

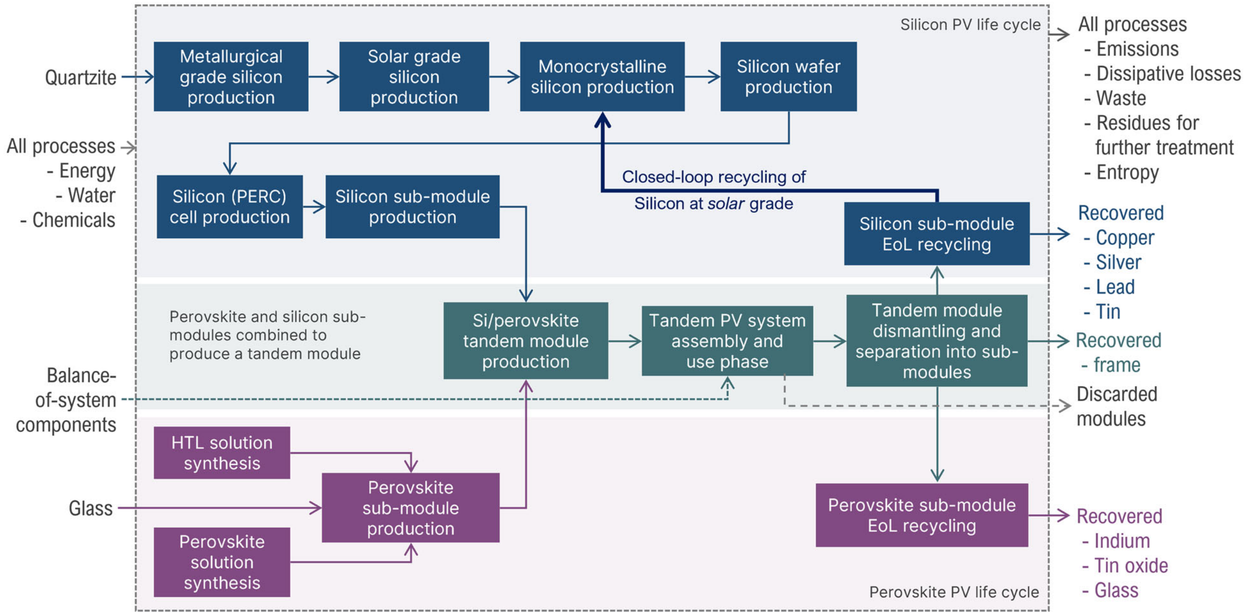

**FIGURE 2** Four-terminal silicon/perovskite tandem life cycle and system boundary with the silicon and perovskite subsystems shown in the top and bottom sections, respectively. All arrows represent mass flows between processes. The silicon and perovskite systems can be visualized by connecting the respective sub-module production step with its end-of-life recycling process. The system can be considered cradle-to-cradle with respect to Si, and cradle-to-gate for other recovered products.

### 1.4 | Purpose of this paper

The cost of solar PV energy is already well below that of traditional power sources. While it is generally accepted that PV systems have negligible environmental impact during use (Battisti & Corrado, 2005; Lunardi et al., 2018; Muteri et al., 2020), their production and recycling processes introduce additional costs and environmental impacts. These need to be analyzed rigorously to avoid burden shifting between life cycle stages and to identify trade-offs between them. Whether increased circularity increases overall sustainability needs to be confirmed, as it is not guaranteed. By establishing its influence on life cycle inventory, circularity can be linked to environmental and economic performance via LCA and TEA to determine if and how it contributes to sustainability.

In this paper, process simulation is used to generate physics-based inventory datasets for each of the described PV systems to assess resource, environmental, and techno-economic performance consistently. We then analyze system responses to changes in the closed-loop recycling of solar-grade Si—our measure of circularity—to link sustainability and circularity via the inventory. To analyze and compare the circular PV systems, material recoveries and the energy return on investment ($\mathrm{EROI}_{\text{PE-eq}}$) are used as indicators of resource efficiency. Carbon footprint, that is, $CO_2$-equivalent ($CO_2$e) emissions, is used as the environmental impact indicator, and LCOE as techno-economic performance indicator. The approach is further applied to evaluate the potential impacts of carbon taxation on cost, and how circularity might function as a moderator of its effects. To the best of our knowledge, this is the first study to employ thermodynamic process simulation to analyze the effects of circularity on resource, environmental, and techno-economic performance in a single assessment to compare the three contemporary PV systems, and to identify trade-offs between sustainability dimensions.

## 2 | METHODS

### 2.1 | Defining the life cycle systems

The Si system is expected to remain the market leader for at least the next decade and is the reference to which the other systems are compared. The overall tandem system and main production and recycling steps included are shown in Figure 2.

The top section in Figure 2 represents the tandem's silicon subsystem, including the shown production steps and a recycling process that recovers solar-grade Si, silver, copper, aluminum, lead, and tin. The closed-loop recycling of Si connects EoL Si recycling and monocrystalline silicon production (the thicker arrow in the top section of Figure 2). This loop is the focus of this paper and represents any reference to *circularity*. The bottom section depicts the perovskite subsystem, where indium, tin, and glass are recovered but not returned to the life cycle, as the focus of this paper is on Si circularity. Combining the sub-modules into the tandem is shown in the middle section. EoL tandem modules are disassembled into the two sub-modules and recycled in dedicated processes to maximize the quantities and qualities of recovered materials, also preventing cross-contamination with Pb compounds (Kadro & Hagfeldt, 2017). The top, middle, and bottom sections combined represents the tandem life cycle.

## 2.2 | Resource flows

### 2.2.1 | Process simulation-based life cycle inventory

Process simulation is indispensable in any process design activity and the value it adds to CE life cycle assessments has been recognized (Reuter, 1998; Reuter et al., 2019). To develop the simulation models, all unit operations that make up the aggregated blocks in Figure 2 are modeled separately and linked by material flows to create a model for that process. Process blocks are connected to create a deterministic simulation model of the whole life cycle, in the tandem system case comprising 122 unit processes, 653 material and energy flows, and 226 compounds, ions, and elements. The model automatically enforces the laws of mass and energy conservation. Because stream compositions and enthalpies are available, solution chemistry can be accounted for—the true losses from the system can be quantified via "excess" enthalpy and entropy, quantities that represent additional, usually unaccounted-for losses that occur when materials are joined in complex solutions rather than simply blended. This reveals the true non-circularity of systems. Furthermore, thermodynamic equilibrium relationships can be used to predict element distributions where process data are not available.

Closed-loop recycling and its system-wide effects are modeled in the foreground system, which avoids having to select EoL calculation approaches that are often unnecessarily complicated or counter-intuitive (Guinée & Heijungs, 2021). We do not intentionally apply the EoL ("avoided burden") or cutoff ("recycled content") recycling approach, as they merge when modeling closed loops in the foreground (Nordelöf et al., 2019). This simplifies direct assessment of the effects of CE strategies on sustainability performance. As the recycling processes do not exist commercially, simulation models are based on combinations of processes described in the literature, the authors' industry experience and own calculations, and thermodynamic equilibrium predictions to fill data gaps. Simulation models are created using HSC Chemistry (Mogroup, 2021). Detailed process descriptions are provided in Supporting Information S1 (Section S1).

We create neural network (NN)-based surrogate functions as proxies for simulation results using MATLAB (MathWorks, 2021). NNs enable generalized nonlinear process modeling of complex systems without having to define regression equations beforehand (Reuter et al., 1992). Computational efficiency is thereby enhanced to analyze inventory over parameter ranges. Simulations are run with random combinations of the independent variables of interest and the corresponding updated mass and energy flows recorded in datasets, which are then imported into MATLAB to create NNs that reliably reproduce simulation results in a fraction of the time it would take the simulation itself. This allows quantification of the selected sustainability indicators over ranges of, for example, closed-loop recycling rate, PCE, and PV system lifetime. The procedure is described in more detail in a previous publication (Bartie et al., 2021a).

### 2.2.2 | Resource efficiency

$\text{EROI}_{\text{PE-eq}}$, the ratio of energy delivered by, and that harvested to produce a PV system (Raugei et al., 2016) is used as an indicator of energetic resource efficiency assuming an average irradiation of 1700 kWh/(m$^2$ year), performance ratio (PR) of 0.75, a 30-year lifetime, and grid efficiency ($\eta_{\text{grid}}$) of 0.30. PR refers to the ratio of a PV system's rated power and that which it delivers, and $\eta_{\text{grid}}$ to the efficiency with which a particular grid converts all energy harvested from the environment into an energy carrier, in this case electricity (Ibid.).

We distinguish between collection, recycling, and recovery rates. The *recoveries* of Si and other materials are determined by the efficiency of the recycling processes. The *recycling* rate is an independent variable used to specify the quantity of recovered Si returned to PV production and the quantities of other recovered materials sold. For clarity, we assume a *collection* rate of 100% so that the quantity of Si recycled also represents the quantity of originally consumed Si returned for re-use. At a recycling rate of 0%, however, nothing is returned or sold despite the hypothetical 100% collection rate, because all recoverable materials remain locked in unliberated "urban minerals," that is, spent PV modules. Until they pass through the recycling process, they only have potential value—we do not pre-emptively assign cost or environmental impact credits unless recycling actually occurs.

## 2.3 | Estimating carbon footprint

Distinctions are made between Scope 1, 2, and 3 $CO_2$e emissions (GHG Protocol, 2011) to assess carbon footprints. Scope 1 refers to direct $CO_2$e emissions from manufacturing and recycling, as calculated in the simulation models. Scope 2 refers to indirect emissions associated with the consumption of purchased energy. We convert power consumption quantities into Scope 2 emissions using the carbon intensity of the German electricity market mix (0.55 $kgCO_2e$/kWh; Wernet et al., 2016). Scope 3 refers to the embodied emissions of materials and components not modeled in the foreground. We include these for glass, aluminum frames, mounting systems, and cabling using published emission factors (de Wild-Scholten, 2013; Frischknecht et al., 2016; Stolz et al., 2017, 2020). In line with the majority of PV system assessments, two functional units—per $m^2$ of modules produced, and per kWh energy generated over the lifetime—are used to express carbon footprint. The former is useful for comparisons of production and recycling emissions. The latter considers PCE, lifetime, and solar irradiation, thereby accounting for PV system performance during its use phase.

It is acknowledged that considering only one impact category carries the risk of shifting burdens to from one environmental issue to others (Rosenbaum et al., 2018). The Product Environmental Footprint Category Rules (PEFCR) for PV electricity consider climate change, particulate matter formation, and resource use the most relevant impact categories (European Commission, 2020b). These and human toxicity impacts have been shown to tend in the same direction for PV (Laurent et al., 2018). Therefore, the risk of burden shifting is considered low.

## 2.4 | Cost calculations

### 2.4.1 | Minimum sustainable price

Discounted cash flow and net present value (NPV) analyses are used to estimate the minimum sustainable module price (MSP, expressed as $/Watt), which is needed to calculate LCOE. A description of the NPV calculation is given in Supporting Information S1 (Section S2). The module price at which NPV is zero is the minimum price that sustains the manufacturer while providing investors with their expected return. We assume a 6% weighted average cost of capital (WACC) for MSP calculations, in line with recently published analyses (KPMG, 2020; Roth et al., 2021; Steffen, 2020). To estimate potential revenue from recycled products, it is assumed that silver, copper, indium, and tin dioxide are recovered at saleable purities in accordance with the recycling process developers' claims of recovering pure indium metal sponge (Li et al., 2011) and other metals at purities greater than 99%, all of which can be sold to the PV industry (Huang et al., 2017). Module frames are recovered as aluminum scrap and glass as cullet. The link between Si circularity and cost is established by adjusting the total Si cost for PV production based on the share of recycled Si content. Taking an integrated life cycle perspective, the hypothetical revenue from recovered Si breaks even with the cost of recycled Si, which is assumed to be two thirds of the global Si MSP of $15/kg suggested by Woodhouse et al. (2020). For materials other than Si, potential revenue is estimated using calculated recoveries and current prices.

### 2.4.2 | Levelized cost of electricity

LCOE is a useful indicator of the economic performance of power supply technologies and a decision support tool when comparing them. It is the ratio of total economic investment in, and total energy generated by a PV system over its lifetime and is represented by Equation (1) (Sofia et al., 2018).

$$\mathrm{LCOE} = \frac{I_{\mathrm{system}} + \sum_1^n \frac{\mathrm{OM}}{(1+r)^n}}{\sum_1^n \frac{E(1-d)^n}{(1+r)^n}} \tag{1}$$

$I_{\mathrm{system}}$ is the initial PV system investment, OM is the annual operation and maintenance cost, $n$ is the system lifetime, $E$ is the energy yield in the first year, $r$ is the nominal discount rate, and $d$ is the annual degradation rate. The initial investment and *OM* were estimated using recently published breakdowns of area- and power-related costs (Zafoschnig et al., 2020). To calculate $E$, we assumed an average insolation of 1700 kWh/($m^2$ year), a 0.5%/year degradation rate, and a performance ratio of 0.75. Cost estimation methods and assumptions are summarized in Supporting Information S1 (Section S2 and Table S2).

## 3 | RESULTS

The detailed mass and energy balance data for all 122 unit processes derived from, among others, reaction equations, distribution coefficients, Gibbs free energy minimization, and published information are available in a data repository (see Bartie et al., 2022). Also provided are separate inventory datasets for the perovskite, silicon, and tandem systems, each with EoL recycling rates of 0%, 50%, and 100%, as examples. These data form the basis of all results presented in this paper.

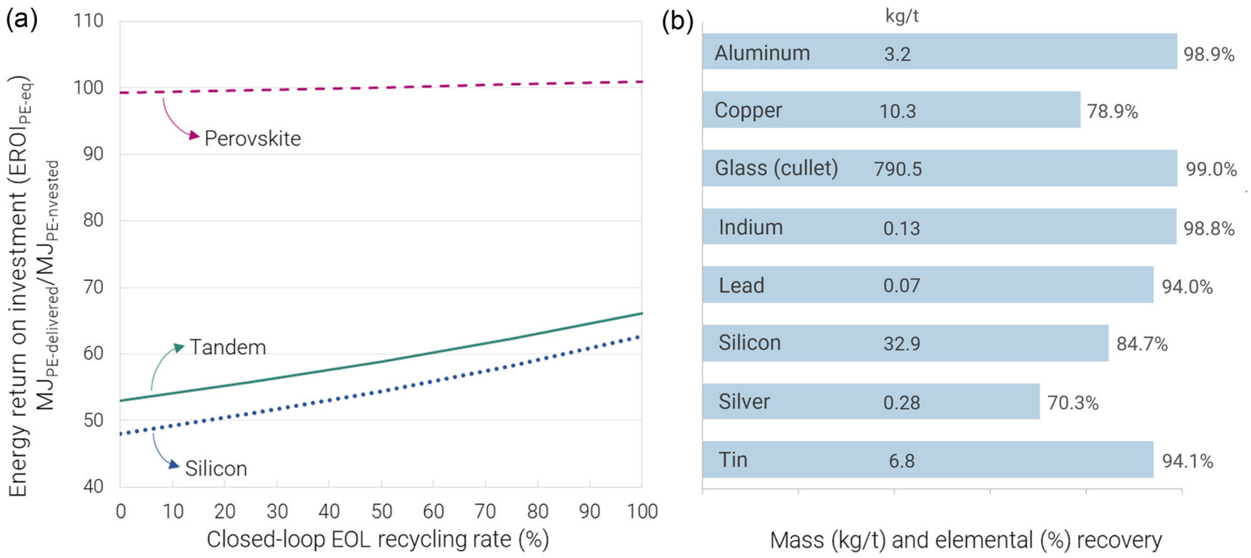

**FIGURE 3** (a) Energy return on energy investment ($EROI_{PE\text{-}eq}$) and (b) end-of-life (EoL) recoveries as a percentage of the element entering the recycling process and as the mass recovered per tonne of EoL modules recycled. Note that the recoveries are independent of the recycling rate. Tin and lead are recovered as oxides. $EROI_{PE\text{-}eq}$ values have been normalized to an average irradiation of 1700 kWh/(m$^2$ year), PR of 0.75, lifetime of 30 years, and a grid efficiency ($\eta_{grid}$) of 0.30. Underlying data can be found in Supporting Information S2.

Direct comparisons of PV systems are notoriously challenging because of the number of cell material combinations and configurations, deposition methods, electricity inventories, different system boundaries, and methodological assumptions, among others. Nonetheless, we have normalized results as described in each case to enable valid comparisons. All comparisons are based on our results at a closed-loop recycling rate of zero to ensure alignment with other studies.

## 3.1 | Resource efficiency

### 3.1.1 | Energy generated for energy invested

Figure 3a depicts $EROI_{PE\text{-}eq}$ as a function of circularity. We calculate 48, 53, and 99 for the silicon, tandem, and perovskite systems without recycling, respectively. Fthenakis and Leccisi (2021) recently reported a monocrystalline-Si $EROI_{PE\text{-}eq}$ of 38 (34 normalized), and Jia et al. (2021) 32 (39 normalized). Our more optimistic 48 is expected because of a higher PCE (21.7% vs. 20.5% and 20.2%) and potentially lower estimate of energy consumption in the background system. Variations of perovskite cell configurations are many (Li et al., 2021), which adds to the difficulty of like-for-like comparisons. We exclude BoS components to align with other studies and calculate an $EROI_{PE\text{-}eq}$ of 187, which lies between the normalized 223 and 155 calculated from Ibn-Mohammed et al. (2017) and Tian et al. (2021) who modeled fairly similar modules. We have not found published $EROI_{PE\text{-}eq}$ values for four-terminal tandems.

Between not recycling at all and recycling all recovered Si, $EROI_{PE\text{-}eq}$ increases by 30% and 25% for the silicon and tandem systems, respectively. The amount of Si returned to the life cycle strongly influences power consumption in the silicon and tandem life cycles. When returned at solar grade, both the metallurgical- and solar-grade Si production processes are bypassed (see Figure 2). In bypassing these processes, their high energy consumptions are avoided. This is not relevant in the perovskite system, as it does not make use of Si. However, a 1.6% increase in $EROI_{PE\text{-}eq}$ is still observed. This is attributed to the generation of electricity from heat recovered during recycling, which reduces net power consumption. Regardless of recycling rate, the perovskite system's return is considerably higher than that of both the tandem and silicon systems, because its production energy investment is at least 60% lower based on our calculations.

### 3.1.2 | Recovery of valuable and hazardous materials

Figure 3b shows the recoveries of key elements, that is, the maximum amount of each element that can be returned to the same or similar life cycle in a closed loop, or sold for use in a different application. Note that recovery rates are independent of recycling rate as they are expressed per tonne

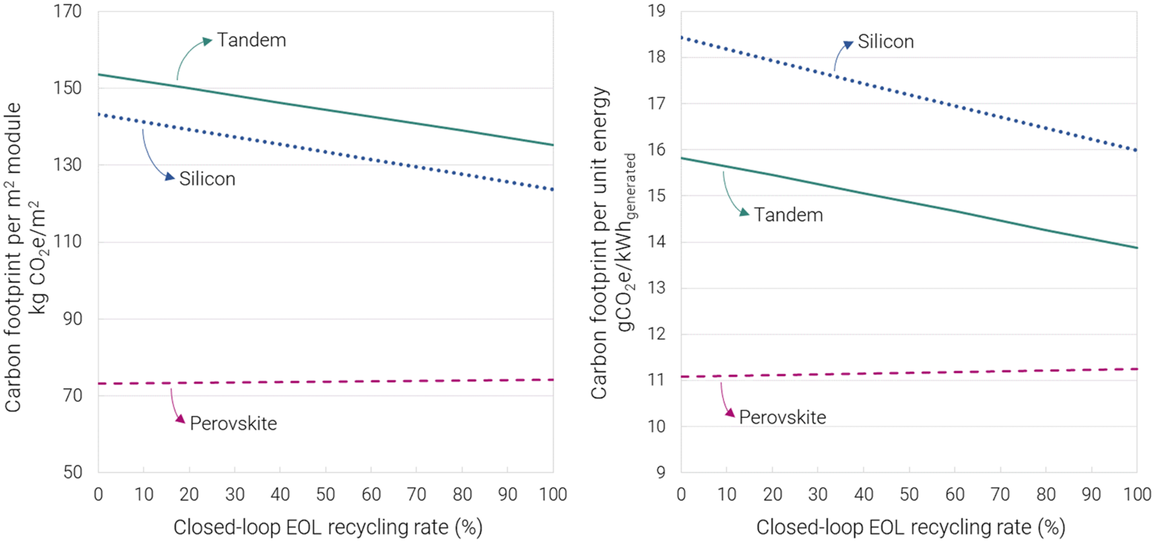

**FIGURE 4** Carbon footprints (a) per $m^2$ module produced, and (b) per lifetime energy generated, normalized to an average irradiation of 1700 kWh/($m^2$ year), PR of 0.75, and a lifetime of 30 years. Breakdowns of Scope 1, 2, and 3 emissions can be found in Supporting Information S1 (Section S3, Figure S2) and all underlying data in Supporting Information S2.

of modules recycled. Even at a closed-loop Si recycling rate of 100%, 84.7% of the Si entering the recycling process is returned, while the remainder is lost. Thus, even with total circularity, the Si material loop cannot be completely "closed."

## 3.2 | Carbon footprint

Life cycle carbon footprints are depicted in Figure 4 for the two functional units described in Section 2.3.

We estimate 143, 73, and 153 kg$CO_2$e/$m^2$, and 18.4, 11.4, and 15.8 g$CO_2$e/kWh for the silicon, perovskite, and tandem systems without closed-loop recycling, respectively. Our 18.4 g$CO_2$e/kWh for Si is in line with the normalized 21.3 and 16.7 g$CO_2$e/kWh calculated from Jia et al. (2021) and Lunardi et al. (2018), respectively. Fthenakis and Leccisi (2021) reported a higher 23 g$CO_2$e/kWh (normalized to 26) due to their larger system boundary. We also find reasonable agreement with previous perovskite studies. Again excluding BoS components, we calculate a footprint of 3.1 g$CO_2$e/kWh, compared with 2.0 (Ibn-Mohammad et al., 2017), 4.9 (Gong et al., 2015), and 4.9 reported by Tian et al. (2021). We have not found published footprints for four-terminal tandems.

Because the four-terminal tandem is a straight-forward combination of perovskite and silicon modules, its higher manufacturing footprint (Figure 4a) is expected. Including lifetime performance (Figure 4b), the tandem's higher PCE compensates for the increased manufacturing emissions to lower its footprint to below that of the silicon reference. Compared to silicon, the same amount of energy will be delivered by a smaller tandem system in a given time period, reducing resource consumption and impacts. The silicon and tandem system footprints decrease with increasing circularity, again because of avoiding two Si production steps and the associated Scope 2 emissions. Contrary to Tian et al. (2021), we find the perovskite system's footprint to worsen with increased circularity, which comes down to the choice of recycling process. The incineration of encapsulation and backsheet materials in our process causes a net increase in emissions—while power generated from recovered heat reduces net Scope 2 emissions, it is not enough to compensate for the increase in direct $CO_2$ emissions from incineration. Tian et al. (2021) assumed selective layer dissolution to recover substrates and other components but did not specify the recycling treatment applied for de-encapsulation, as it was likely not the focus of their study.

Figure 5 depicts the sensitivity of carbon footprint to system lifetime and PCE for the three systems. The labeled datapoints represent the footprints at the respective reference PCE and lifetime. At constant PCE, the perovskite and tandem footprints remain lower than the silicon system's 18.4 g$CO_2$e/kWh at lifetimes down to 18.1 and 25.5 years, respectively. With a 30-year lifetime, the tandem system footprint is less than that of the silicon reference at PCEs down to 23.4%. The minimum perovskite PCE would be less than the 16% lower limit shown.

Considering the constant $CO_2$e emission contours in Figure 5, the area above any given contour is greatest in the perovskite system, followed by the silicon and tandem systems. Qualitatively, this could be interpreted to mean that it would be least challenging to achieve lower footprints in the perovskite system and most challenging in the tandem system if PCE and lifetime are the only factors considered. Important, however, is that the full ranges of lifetime and PCE are not available in all three systems. For instance, if the recent 21.1% perovskite PCE (Liu et al., 2022), and the 22.2% and 28% expected for monocrystalline Si and tandems, respectively (VDMA, 2021), are taken as upper limits (the dashed vertical lines), it is

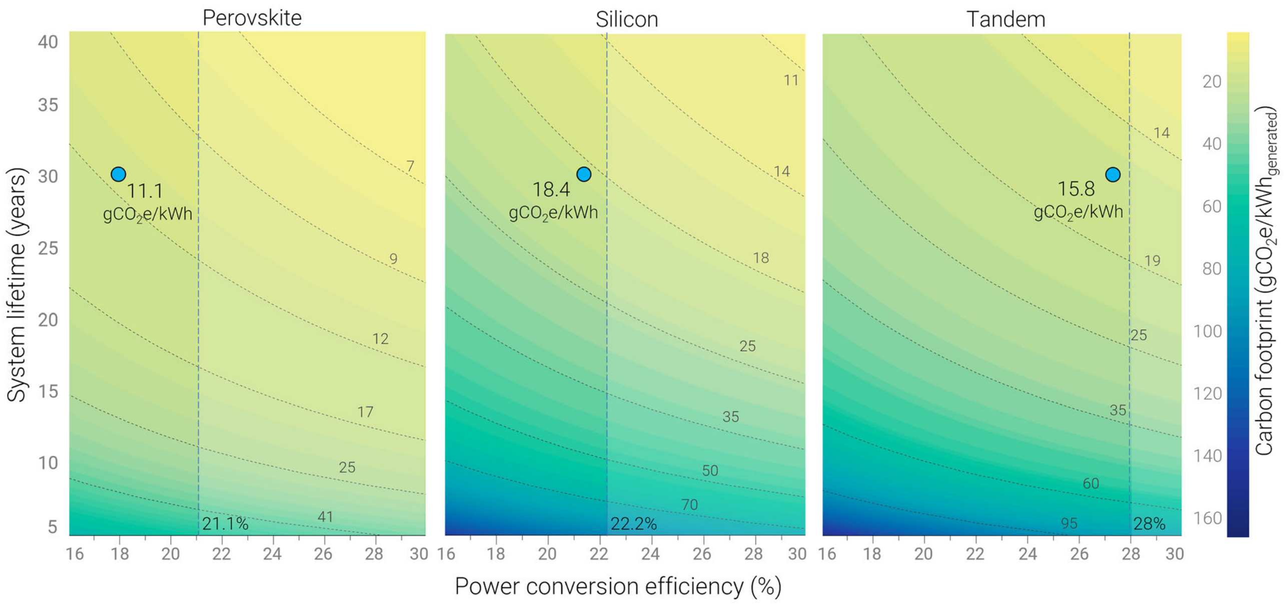

**FIGURE 5** The sensitivity of carbon footprint to system lifetime and power conversion efficiency (PCE) for the case with no recycling. The labeled datapoints show the footprint for each system at its reference PCE (18%, 21.7%, and 27.3% for perovskite, silicon, and tandem systems, respectively) with a 30-year lifetime, annual irradiation of 1700 kWh/(m$^2$ year), performance ratio of 0.75, and an annual relative degradation rate of 0.5%. The dashed vertical lines indicate current perovskite (21.1%) (Liu et al., 2022) and projected Si (22.2%) and tandem (28%) efficiencies (VDMA, 2021). Underlying data can be found in Supporting Information S2.

clear that footprints lower than the reference can be achieved in both the perovskite and tandem systems. Although lifetime limits may currently exist, we assume that continued development would result in perovskite lifetimes similar to that of current commercial technologies.

## 3.3 | Techno-economic assessment and interaction with carbon footprint

Figure 6 depicts the sensitivity of LCOE to lifetime and PCE.

Our reference LCOEs are 3.92, 4.82, and 4.48 c/kWh for the perovskite, silicon, and tandem systems, respectively. The associated MSPs are 0.20, 0.31, and 0.38 \$/Watt, respectively. As expected, our values agree with the 0.21, 0.32, and 0.36 reported by Liu et al. (2020) and Sofia et al. (2020) as our assumptions are closely aligned with those studies. Perovskite LCOE remains below that of the silicon reference if its lifetime exceeds 17.6 years compared to 18.1 years for a lower carbon footprint. A perovskite lifetime greater than 18.1 years would, therefore, give it both environmental and economic advantage over the silicon system. The same applies in the tandem system at the reference PCEs—a lifetime greater than 25.5 years is needed, and carbon footprint is the deciding factor (cf. 23.7 years for a lower LCOE). Alternatively, if 30-year lifetimes can be guaranteed, tandem LCOE will remain below that of the silicon system down to PCEs of 24.8% (compared to 23.4% for carbon footprint). Here, LCOE is the deciding factor—at PCEs above 24.8%, the tandem outperforms the silicon system in both the economic and environmental dimensions. As before, the minimum perovskite PCE would be less than the 16% lower limit shown.

As mentioned, the full ranges of lifetime and PCE are not technically achievable in all systems. There may be additional thresholds beyond which a system would not be considered a sound investment or environmentally acceptable. The sustainability of a particular system can only be maximized where the feasible operating windows bounded by these thresholds overlap, that is, within the bounds of all technical, environmental, and techno-economic limits.

Figures 7a and 7b, respectively, show the variation of LCOE and carbon footprint with circularity.

The increase in recycled content brought about by closed-loop recycling lowers silicon system LCOE (Bartie et al., 2021b). In the perovskite and tandem systems, on the other hand, increased circularity increases LCOE. As explained, the perovskite system does not benefit from the direct displacement of an energy-intensive and expensive raw material that would contribute significantly to lowering power consumption and cost, such as high-grade Si. In essence, the four-terminal tandem system represents the net effect. Viewed in isolation from a profit-only perspective, increased circularity seems unfavorable in the perovskite and tandem systems. From a sustainability perspective, however, Figures 7a and 7b together show that, in the tandem system, a trade-off exists between LCOE and carbon footprint with respect to circularity—increased circularity brings about lower CO$_2$e emissions, while the delivered energy becomes more expensive. Therefore, all other things being equal, an optimum level of Si circularity exists that minimizes both cost and environmental impact.

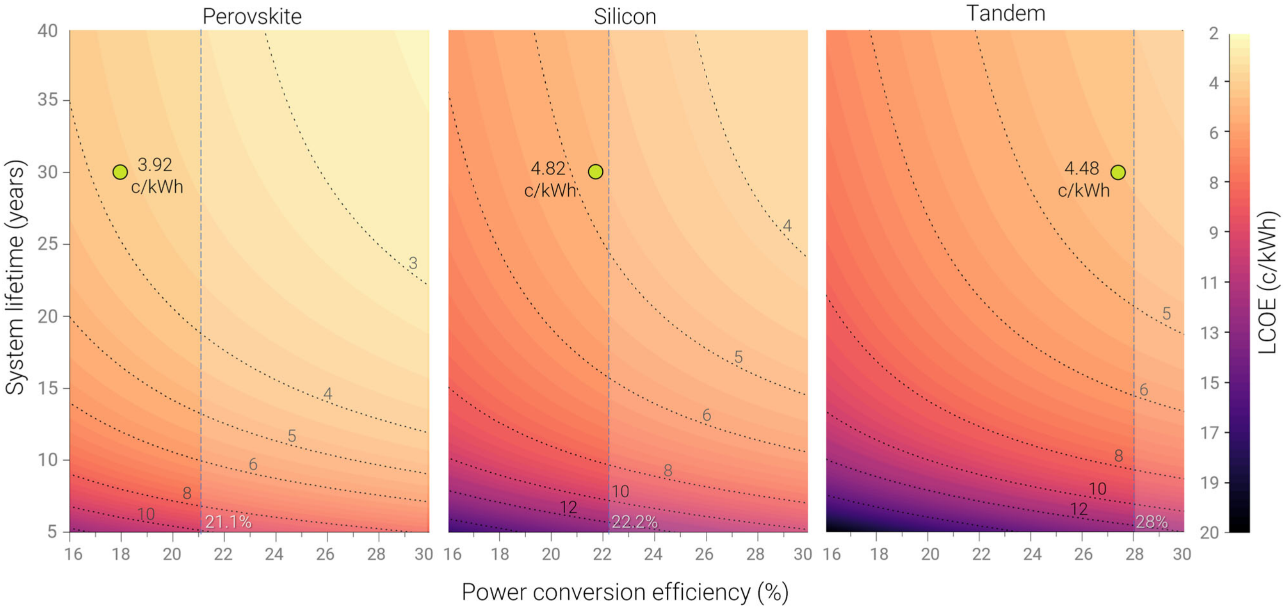

**FIGURE 6** The sensitivity of levelized costs of electricity (LCOE) to system lifetime and power conversion efficiency (PCE) with no-recycling case. The labeled datapoints indicate the LCOE for each system at its reference PCE (18%, 21.7%, and 27.3% for the perovskite, silicon, and tandem systems, respectively) with a 30-year lifetime, annual irradiation of 1700 kWh/(m$^2$ year), performance ratio of 0.75, and an annual relative degradation rate of 0.5%. The dashed vertical lines show current perovskite (21%) (Liu et al., 2022) and projected Si (22.2%) and tandem (28%) efficiencies (VDMA, 2021). An assessment of the sensitivity of LCOE to overall recycling cost and discount rate can be found in Supporting Information S1 (Section S2 and Figure S1) and all underlying data in S2.

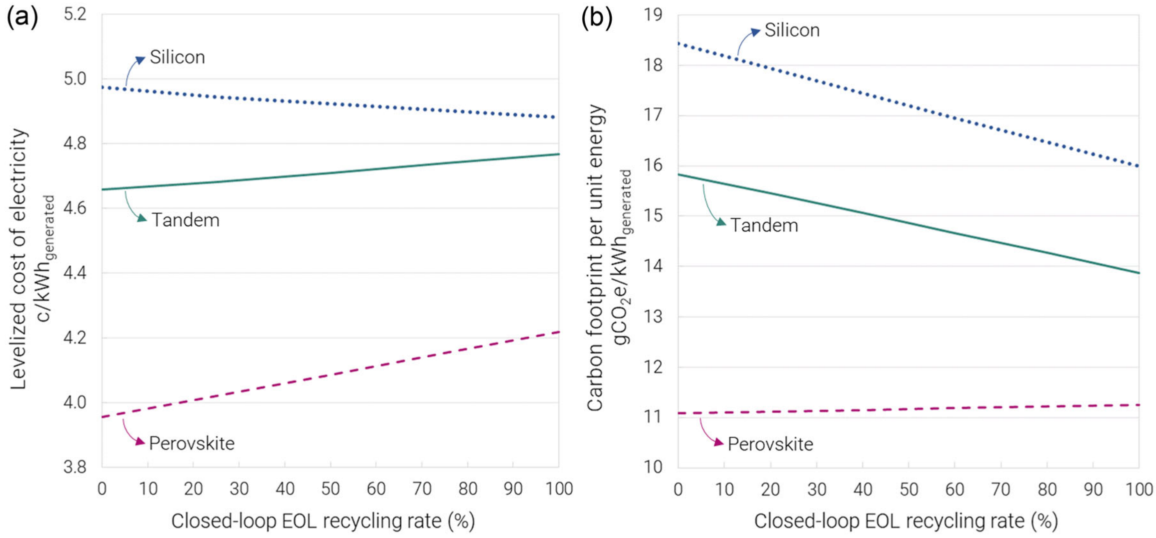

**FIGURE 7** The variation of (a) levelized costs of electricity and (b) $CO_2$e emissions with end-of-life recycling rate. Figure 4b is repeated here as 7b for convenience. Underlying data can be found in Supporting Information S2.

### 3.3.1 | Carbon tax

Carbon taxation is generally considered an effective policy measure for stimulating emission reductions. Our approach facilitates estimation of the effects such a tax might have if imposed on the energy sector. Figure 8a shows the effects of hypothetical taxes (between 25 and 400 \$/t$CO_2$e emitted) and circularity on tandem LCOE. The public revenue stream generated by the tax increases LCOE, as expected. Increased circularity at the low tax has little effect—the \$25 line is almost parallel to the zero-tax line. As the tax increases, line slopes become less positive, and eventually negative, above the breakeven tax rate. Below the breakeven rate, increased circularity always increases cost, but as the tax rate approaches the breakeven rate, the cost increase becomes less pronounced, that is, increased circularity softens the tax's cost-increasing effect. Above the breakeven rate,

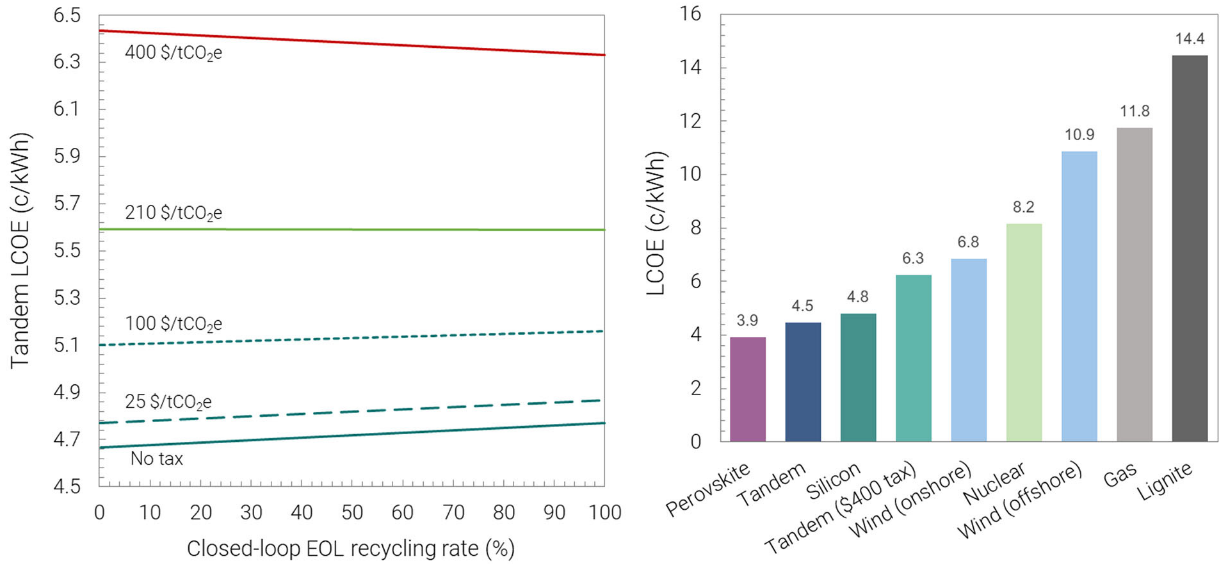

**FIGURE 8** (a) The combined effects of recycling and carbon tax on levelized costs of electricity (LCOE) in the tandem system, and (b) a comparison of photovoltaic LCOE with that of other electricity sources (Fraunhofer-ISE, 2021; EIA, 2022 for nuclear). Underlying data can be found in Supporting Information S2.

increased circularity always reduces cost, and this effect becomes stronger the higher the tax. Based on our assumptions, the breakeven tax is \$210/tCO$_2$e. With this value significantly above any current, for example, Sweden's \$134/tCO$_2$e (Sweden Ministry of Finance, 2021) or predicted tax rates (Jaumotte et al., 2021), it is highly unlikely that closing materials loops alone would be enough to reverse tax-induced cost increases. Despite these findings, Figure 8b shows that, even at \$400/tCO$_2$e, PV LCOEs remain below that of other electricity sources with no taxes applied.

### 3.3.2 | The contribution of transport to carbon footprint and MSP

With PV supply chains being globally distributed, it is worth investigating the impacts and costs of material and product movement across the globe. We estimated the contribution of transport to carbon footprint and MSP for various combinations of manufacturing and recycling location for the tandem system. Results are presented in Supporting Information S1 (Section S4 and Figure S3).

## 4 | DISCUSSION

An important aspect of this work is the method by which the inventory data were generated and the amount of detail included. Contrary to methods that account for material streams as if they are flowing through a system as pure elements, process simulation accounts for the flows of the compounds and solutions actually present, also including their thermochemical properties. The rigorous mass and energy balances calculated as a result allow each flow in the system to be quantified in terms of mass and energy (for enthalpy and entropy) units per unit of time, so that all flows and thermodynamic losses from the system can be quantified in the same units as the energy delivered by the energy carrier, in this case PV electricity.

Our results have shown that the perovskite system is the best performer in terms of our indicators for resource efficiency (EROI$_{\text{PE-eq}}$), environmental impact (CO$_2$e emissions), and techno-economic performance (LCOE), with the tandem in second place, and the silicon system in third, regardless of the degree of circularity. The caveat is that the perovskite and tandem systems achieve the same long lifetimes current commercial technologies do. The perovskite and tandem lifetimes must exceed 18.1 and 25.5 years, respectively, to outperform the silicon system both environmentally and techno-economically.

Compared to the Si system, the tandem will be more effective at enhancing the sustainability of whichever life cycle system consumes the energy it delivers, because both the embodied carbon footprint and the levelized cost of that energy will be lower, and a system 20% smaller in physical size would deliver a given amount of energy within a fixed period. While the perovskite system's footprint and LCOE are considerably lower, a 21% larger system would be needed.

Although recycling increases *direct* (Scope 1) emissions in all systems, the increased consumption of high-quality recycled Si considerably reduces electricity consumption in the silicon and tandem systems. The reduction in associated *energy-related* (Scope 2) emissions compensates for the direct emissions added through recycling several times over. Importantly, this is conditional upon the further development and commercialization of recycling processes that recover Si at solar grade. There is no similar benefit in the perovskite system because of the absence of large quantities of input materials as energy intensive as Si. While the recovery and recycling of intact glass substrates appear to be a promising option for perovskite recycling (instead of downcycling glass into cullet), a recent analysis of 13 potential approaches revealed that all but one are environmentally *more* detrimental than using virgin coated glass, mainly because of the solvents needed for delamination (Rodriguez-Garcia et al., 2021). However, the perovskite system's footprint is already 30%–40% lower than that of the silicon, and 19%–30% lower than that of the tandem system.

Increased circularity in the silicon system is beneficial in terms of both carbon footprint and LCOE, while the opposite is true for the perovskite system. In the tandem system, the trade-off that emerges indicates that an optimum level of circularity exists at which both cost and environmental impact will be minimized, but with compromises in both dimensions. The implication is that total circularity does *not* deliver the most sustainable outcome in this case, highlighting the importance of the assertion that circularity does not automatically come with an overall sustainability guarantee (Korhonen et al., 2018). The advantage of our approach is the use of fully aligned inventory data to calculate the environmental and techno-economic indicators as a function of circularity, so avoiding the introduction of additional uncertainty resulting from potential data and system boundary inconsistencies.

However, to gain a more complete picture of interactions within the system, future work should investigate further options and constraints that may influence cost and footprint. For instance, while we found the perovskite footprint to increase with circularity, Tian et al. (2021) reported the opposite. In this example, recycling process design and its associated costs and impacts play key roles in creating, modifying, or undoing any sustainability trade-offs. Results from studies such as that by Rodriguez-Garcia et al. (2021) mentioned earlier should be incorporated in future life cycle simulations to analyze the system-wide effects of different production and recycling approaches. Another example is the potential revenue from recycled products. All processes have to operate within economic, societal, and environmental impact constraints to be viable. Also, depending on the supply and demand for recycling and the products from recycling, this may involve increased focus on the recovery of certain elements at the expense of others.

The linked resource and economic models also allowed us to quantify the potential cost effects of carbon taxation, while investigating the role of closed-loop recycling in modifying these effects at the same time. We found that, besides clear environmental benefits in the tandem system, recycling dampens the tax's cost impact. Therefore, increasing the recycled content in PV modules alone would not be expected to fully compensate for the cost impact of taxation. Additional measures upstream in the supply chain and higher up in the CE material hierarchy (e.g., by reducing consumption, re-using, and refurbishing) are needed before recycling becomes inevitable. If this occurs with overall sustainability, rather than merely cost reduction in mind, the tax will have the intended effect. It could, of course, also be counteracted with measures that reduce cost but not emissions, such as cross-border carbon leakage for which other mechanisms like border tax adjustments need to be implemented. From a business perspective, however, the lower perovskite and tandem LCOEs provide more room to move in terms of margins and investment in emission reduction and/or energy storage technologies relative to the silicon reference.

There are limitations associated with the obtained results. Power consumption has a significant impact on carbon footprint as Scope 2 emissions and is therefore sensitive to location-specific grid compositions. Although PV supply chains are typically globally distributed, all results presented in this paper are based on the German electricity mix for the sake of simplicity. Results are based on static simulations—we have not considered potential evolution of the electricity mix or innovations in manufacturing technologies and efficiencies over time. Although standard methods have been used for recycling cost estimates, they should be seen as preliminary, as none of the recycling processes exists commercially. While we only quantified carbon footprint and considered the risk of burden shifting to be low, future assessments should include other impact categories to examine whether other trade-offs exist. We considered two of the sustainability dimensions in this study. To gain a comprehensive understanding of overall sustainability, this approach will be expanded to include the effects of societal impacts.

## 5 | CONCLUSION AND OUTLOOK

PV has a key role to play in the decarbonization of future energy systems, but its full potential will only be realized once PV life cycles achieve sustainable circularity. To confirm whether this is occurring, reliable methods are needed to assess sustainability and how it is influenced by circularity. We presented a novel approach that uses process simulation to generate physics-based inventory data that comply with the laws of mass conservation, and the first and second laws of thermodynamics, as opposed to elemental flows based on linear input–output transformations. By linking simulations with bottom-up cost models, we evaluated and compared the resource efficiencies, carbon footprints, and LCOEs of three contemporary PV technologies. Direct evaluation of the simultaneous, system-wide effects of circularity, PCE, system lifetime, and carbon taxation on the selected sustainability indicators are a further novelty, which are facilitated by NN-based surrogate functions that serve as proxies for simulation results. Assessments of resource efficiency, and environmental and techno-economic performance, as well as the effects of policy measures can, therefore, all be conducted within the same framework from a consistent foundation of physics-based inventory data. As a result, potential trade-offs among the sustainability dimensions and in relation to CE strategies can be identified and quantified with a view to maximizing overall sustainability.

With rapid technological development in the PV and other industries that make use of complex material combinations to achieve required functionalities and efficiencies, agile approaches that ensure data consistency and adherence to the laws of conservation and thermodynamics—such as that presented in this paper—are needed. This rigorous quantification of life cycle mass and energy flows (including thermodynamic losses) provides the true starting points and performance assessments along development paths aimed at increasing overall sustainability.

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## CONFLICT OF INTEREST STATEMENT

The authors declare no conflict of interest.

## DATA AVAILABILITY STATEMENT

The data that support the findings of this study are openly available in Zenodo at https://doi.org/10.5281/zenodo.7102103, reference number 7102103, and in the supporting information (S1 and S2) of this article.

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## SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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