

A comparative life cycle toxicity assessment of perovskite/silicon tandem photovoltaics

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Abstract

Perovskite/silicon tandem solar cells (PSTs) have emerged as a promising photovoltaic (PV) technology that can exceed the theoretical power conversion efficiency limit of single-junction solar cells. To determine the future potential benefits of PSTs, it is crucial to accurately assess their environmental impacts and recyclability. Here, we present the first complete life cycle toxicity assessment of PST panels. For this, we evaluated the toxicity of material procurement, manufacturing, and use stages, and compared them with the toxicity of crystalline silicon PV and CdTe PV, as well as other electricity sources. For the end-of-life (EoL) stage, we developed three variants of panel recycling processes and compared their toxicity impacts with the case of procuring the materials required to manufacture a new panel. We found that the life cycle toxicity of PV in general is mainly driven by metal emissions. PSTs in particular emit more metals—and these are more toxic—than other PVs, but less than conventional sources of energy. A lower silver content—or more sustainable silver procurement—would be the first step towards making PSTs more environmentally sustainable. Concerning the EoL analysis, all proposed variants are less impactful than procuring materials from the market. Their largest benefits can be found in the recovery of the bottom glass and crystalline silicon sub-cell, the copper cables, and the top glass.

Introduction

Heavy reliance on renewable electricity production is a key pathway to mitigate climate change.¹ Thanks to their worldwide development in recent years, photovoltaics (PVs) are today one of the cheapest sources of electricity.² Perovskite solar cells are among the most promising emerging PV technologies.³ Perovskite solar cells' power conversion efficiencies (PCE) have increased from 14.1% to 34.6% from 2013 to 2024.⁴ Moreover, when stacked on top of a silicon PV cell, perovskite/silicon tandem (PST) can surpass the PCE of single junction cells, and increase up to 33.7%.⁴ As a result, PSTs have attracted tremendous attention in the last few years as highly promising to bring very high-performance PV to the market, which is critical to further lower the levelized cost of electricity from solar energy.⁵ Practically, having achieved efficiencies well beyond those of established technologies, current research efforts concentrate on expanding the operation stability of the cells, and on scaling up the production process for their manufacturing.⁶

Since their inception, the environmental performance of perovskite PV has been the subject of several studies.^{7–13} The majority of these studies employed life cycle assessment (LCA), a tool for the evaluation of potential impacts of products, processes, and human activities from their cradle to grave.¹⁴ Both in and out of the LCA community, the use of lead as part of the perovskite absorber layer has received close attention due to its acute toxic effects.^{15,16} To some, lead in perovskite PVs represents a low level of contamination.¹⁷ In terms of freshwater ecotoxicity, despite its low solubility, it is harmful to zebrafish (*Danio rerio*) at concentrations as low as 10 μM .¹⁸ To human health, the inhalation of toxic fumes is one of the largest lead-related burdens, and a 100 m² roof half-covered with perovskite modules could release up to 20 g of this substance.¹⁹ Perovskite PVs' tendency to degrade and potentially leach lead compounds in case of failure has encouraged LCA

practitioners to evaluate scenarios beyond normal operation.¹⁹ Celik et al.²⁰ showed that all the PbI_2 from the active layer—and 11.1% of the CuSCN hole transport layer would be released in less than a year if the panel was to suffer a crack, i.e., 3 cm^2 of crack area in 1 m^2 of solar module surface. The toxicity impacts of these lead and copper emissions would be more significant than the upstream toxicity generated by the production of the metals themselves. Furthermore, a fire safety assessment of PST was conducted with a focus on lead-related aspects. The study demonstrated that, in the absence of glass-glass encapsulation, most of the PbI_2 products evaporated into the atmosphere.²¹ Despite the importance of a total release of lead, Billen et al.²² indicated that electricity from perovskite PVs resulted in four times less lead than other sources of electricity in the US, and their emissions are about 20 times less harmful. In a recent study from Chen et al., it was demonstrated that the use of self-healing encapsulation materials can reduce lead leakage from perovskite PVs by up to 95%.²³

In this study, we aim to conduct a life cycle toxicity assessment (LCTA) of PST focusing on their metal components and inorganic emissions, while also comparing their impacts with those other electricity generating technologies. We modeled a complete recycling treatment process, and for the first time, we evaluated the importance of panel recycling and metal recovery in terms of LCTA.

Methods

Goal and scope

Our objective is to assess the life cycle toxicity of PST PVs. The purpose of this analysis is twofold: First, to elucidate if the toxicity potential of PSTs and their metal emissions differ significantly from other electricity sources; second, to evaluate the potential environmental

benefits of recovering their metal content and other materials at the EoL of the PST (Figure 1). Our intention is to help PST manufacturers, solar scientists, and recyclers select less harmful materials and processes. Because of that, we aspire for our results to be used in comparative assertions disclosed to the public.

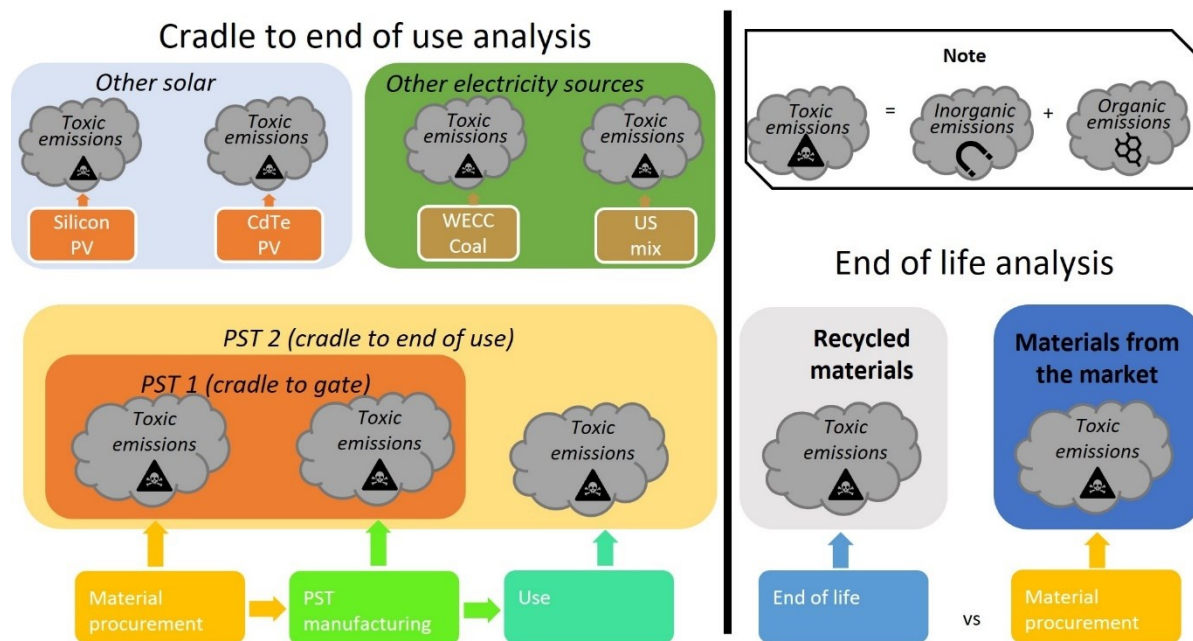


Figure 1 Systems under study in the cradle to end of use and EoL analyses

The system boundaries of our first analysis are cradle to end-of-use (Figure 1). Scenario PST 1 contains material procurement—which in turns includes extraction of raw materials, as well as their processing, and transport—and the manufacturing of the complete device. As an alternative scenario, PST 2 also includes the use phase, modeled as an accidental release of lead at the end of its useful life. For the EoL analysis, we compared the recycling of the PST with the primary material procurement of its components to assess if currently available recycling approaches have lower toxicity impacts.

Each analysis has its own reference point for comparison. For the cradle-to-end-of-use analysis, which aims to compare PSTs with other energy sources, we used 1 kWh of low-voltage AC delivered as the reference point. This analysis required calculating the energy generated over the entire lifetime of the solar panels after installation. In contrast, for the end-of-life (EoL) analysis, we used a 1 m² panel as the reference point. This analysis focused on the material composition of the manufactured solar panel.”For this LCTA, we evaluated the freshwater ecotoxicity and human toxicity impact categories according to the USEtox methodology²⁴ using GaBi.²⁵ We also built our inventories using this program according to information from literature from 2020-2022 for commercial or soon-to-be-commercialized PV technologies, and from the Ecoinvent database 3.6.^{26,27}

Modeling approaches and life cycle inventories

Cradle to end of use analysis

The inventory for the reference PST was first described by Frischknecht et al.²⁸, which we adapted for a potential production in the US in our previous work²⁹— see Table 1. The initial scenario—PST 1 considered the complete reference PST. In PST 2 we added a complete release of the lead present in the PST to the worst possible compartment, which is fresh water and agricultural soil for ecotoxicity and human toxicity, respectively.

Table 1 Scenarios for cradle to end-of-use analysis

	Scenario	Process	Source
PST scenarios	PST 1	PST panel production	Ecoinvent 3.6 ²⁷ , and Frischknecht et al ²⁸ (adapted in Rodriguez-Garcia et al. ²⁹)
	PST 2	PST 1 + lead release	
Other solar	Si PV	Photovoltaic panel, single-Si, at plant	Frischknecht et al. ²⁸ (adapted in Rodriguez-Garcia et al. ²⁹)
	CdTe PV	Photovoltaic laminate, CdTe, First Solar Series 6, at plant	

Other sources	Coal	WECC, US only: electricity production, hard coal	Ecoinvent 3.6 ²⁷
	US mix	US: market group for electricity, low voltage	Ecoinvent 3.6 ²⁷

WECC is the Western Electric Coordinating Council, the reliability council that covers the westernmost part of the US (<https://www.wecc.org/>)

The inventories from Frischknecht et al.²⁸ and Rodriguez-Garcia et al.²⁹ were built on an area basis. To calculate the energy produced by the PSTs scenarios we followed Roffeis et al.¹⁵, and considered a PCE of 25%, an average performance rate of 76.3%, and a lifetime of 25 years. Also following Frischknecht et al.²⁸ we developed two reference scenarios for existing PV modules: single junction monocrystalline silicon (single-Si) PV and CdTe PV, with PCEs of 22% and 19.5%, respectively.^{15,30} In addition, we selected three Ecoinvent activities as reference scenarios for existing electricity-generating technologies in the United States. They are electricity from coal production in the Western Electric Coordinating Council (WECC) region, and the low voltage mix for the whole US. These three inventories are based on 2019 data.²⁶

The different metals that make the PST, taken from Ecoinvent as global market activities are detailed in Table S1. They include the different production processes that make up a substantial share of the market for a given product. For example, the market for Al frame includes the production of this material in Europe and the rest of the world (RoW). In addition, market activities also include average transportation from the production sites to an undefined customer. By modeling the market activities in detail, we evaluated where the emissions and impacts of the different metals come from with a higher level of detail.

End-of-life analysis

To quantify the relative importance of EoL in toxicity terms, we developed a treatment process for the separation and recovery of metals from the reference PST (Figure 1), including three alternatives for delamination (Figure S1). The treatment train consists of two sub-systems: out-module, and in-module recycling. The out-module subsystem includes two steps, panel disassembly and cable and frame treatment. Both are based on the updated Full Recovery End-of-Life Photovoltaic (FREL P) process, first described by Latunussa et al.³¹, and updated by Ardente et al.³² We also modeled the recycling of aluminum and copper recovered from frames and cables according to these two sources. We modeled tin recovery according to Iannicelli-zubiani et al.³³, and lead recovery according to Ecoinvent.²⁷ In the in-module recycling, we also used Latunussa et al.³¹ and Ardente et al.³² for cell separation and bottom cell recovery. We assumed the high-frequency knife button used to separate PV glass from the rest of the device could also separate the top and bottom cells of the tandem. For the remaining in-module steps, we followed three different techniques for the recovery of top glass and PbI_2 , so both can be used again in the manufacturing of perovskite cells. One is based on Binek et al.³⁴, another on Li et al.³⁵ and Zhang et al.³⁶, and the last one on Feng et al.³⁷ These techniques were initially developed for treating perovskite cells only, and cells with a different architecture than the one we address here. However, similar processes have been used for the treatment of cells closely related to those whose recycling we model.³⁸ Therefore, we assume these techniques could be easily adapted for the delamination of PSTs. The filtration and drying required for the recovery of PbI_2 are modeled based on Peters et al.³⁹ We allocated by mass the impact caused by the recovery of the different materials.

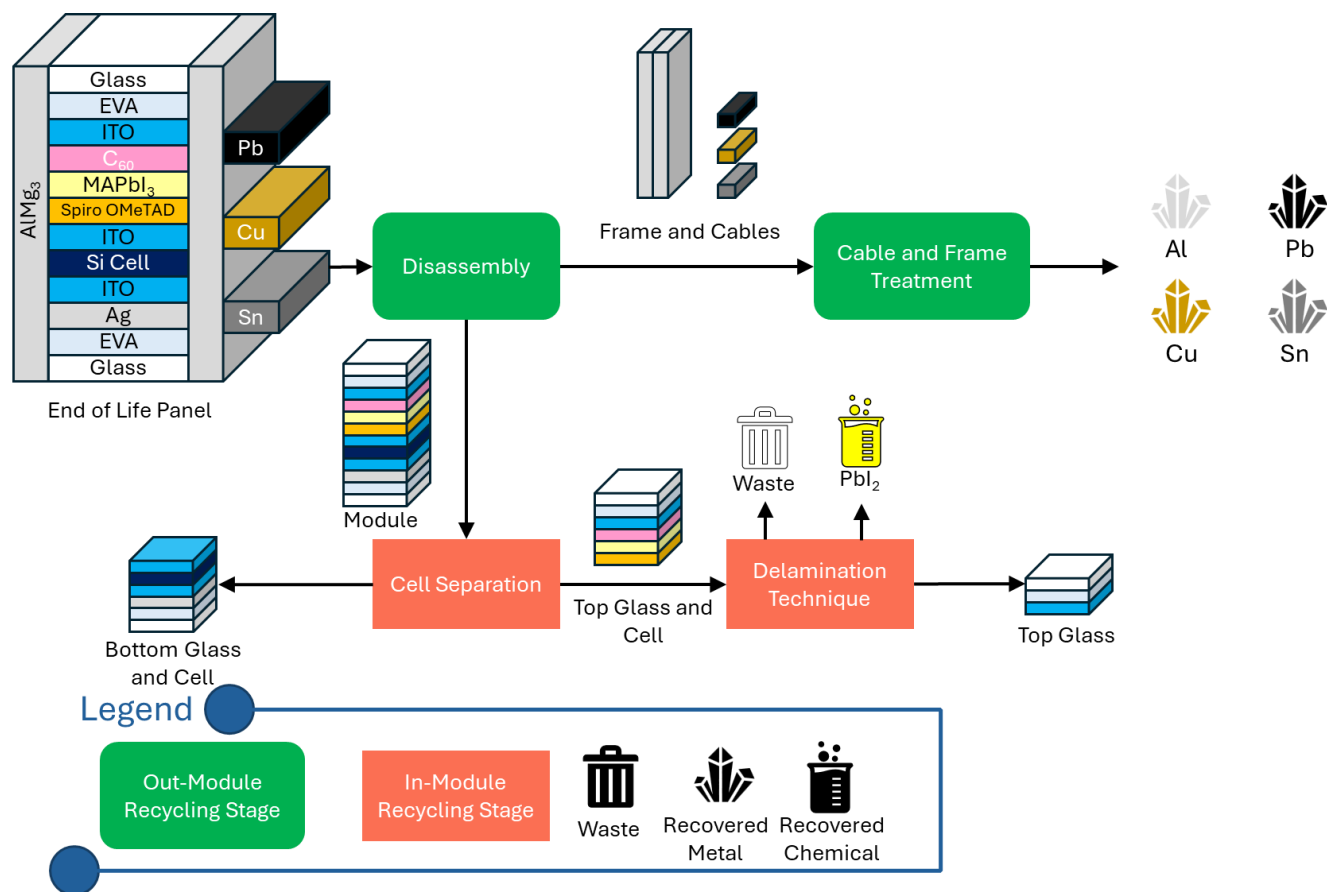


Figure 2 Proposed recycling process for PSK/Si tandem panels.
The three delamination alternative techniques are presented in Figure S1

We presented the relevant layers to recycle in Table S2. The pretreatment and flat glass separation inventories are taken from Ardente et al.³² on the basis that 1,000 kg of PV waste are equivalent to 73 m².³¹ As modeled, the delamination is based on Binek et al.³⁴, Zhang et al.³⁶, and Feng et al.³⁷ techniques.³⁸ Inventories for the common recycling steps, and the three delamination techniques can be found in Tables S3-6.

Results and discussion

Cradle to end-of-use toxicity

While toxicity comes both from inorganic and organics emissions, we found that the effect of the latter is almost negligible for electricity produced by PSTs or the US mix, and relatively minor

when it comes to electricity produced with other PVs—Figure 3. Therefore, the following comparison focuses on inorganic emissions and the toxicity they generate.

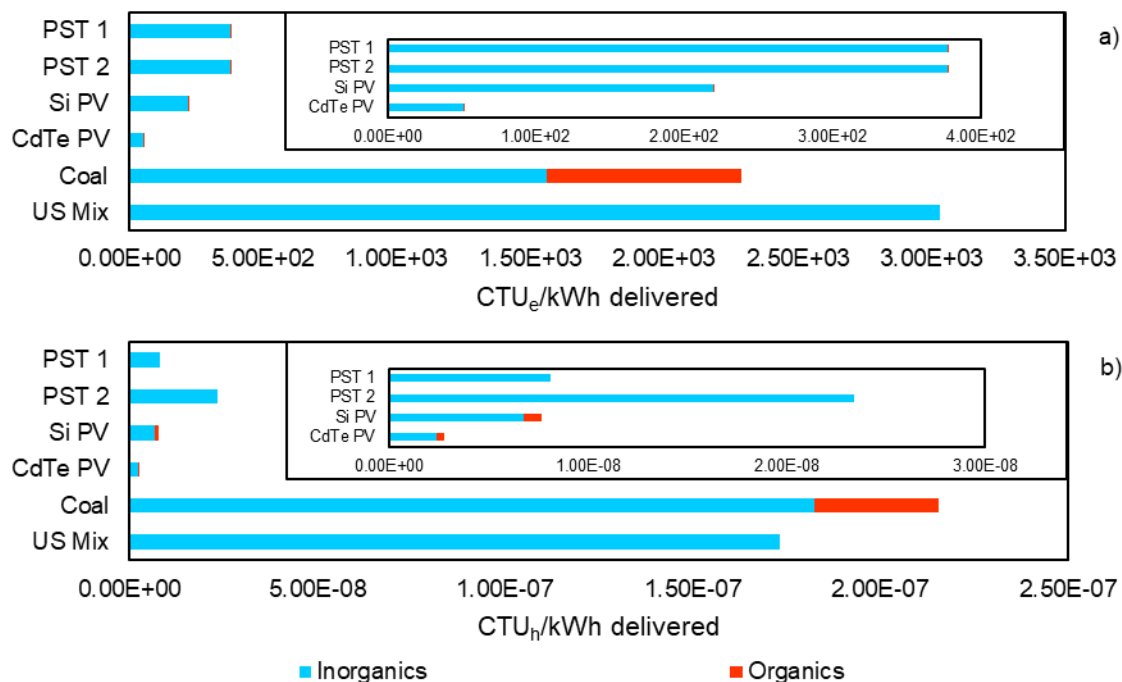


Figure 3 Impacts caused by the delivery of 1 kWh of AC low voltage electricity
a) freshwater ecotoxicity, and b) human toxicity.

We found that electricity from PSTs is more environmentally friendly in terms of both freshwater ecotoxic, and human toxic than electricity from coal or the current US electricity mix—between 6 and 26 times less harmful. This is regardless of the impact of a total loss of lead from PSTs. However, the electricity provided by PSTs is 1.06-1.7 times more impactful than other PVs. A comparison between single-Si PV and PST shows that the manufacturing of PSTs requires 65% more electricity, and 10% more materials than manufacturing single-Si PVs, ~3.5 times more silver.²⁸ This increases PSTs impacts despite this technology having higher PCE. CdTe PV has lower emissions and impacts than both PST and single-Si PV. Compared with single-Si PV, CdTe PV requires 20% less aluminum in its frame, 97% less copper, and no silver, besides having a

lighter active layer.²⁸ In addition, CdTe's active layer is far less toxic. On a per-area basis, it generates less than 0.5% of the freshwater ecotoxicity impact, and less than 5% of human toxicity than the single-Si active layer—Figure S2.

Figure 3 also shows that it is only in human toxicity terms—as opposed to emission in mass or to freshwater ecotoxicity—that an accidental release of lead will be relevant. If this lead reaches agricultural soil at the end of the useful life of the PST, for instance when deployed in agrivoltaics applications, it will triple its human toxicity impact. A release to other compartments excluding air would make the effect of these accidental emissions negligible. For example, the characterization factor for freshwater is more than 1,000 times lower than that of agricultural soil. Despite this accidental release, electricity from PSTs still has lower human toxicity impacts than that from coal combustion, or the US electricity mix. Only if the accident occurs within the first 15 months of operation would electricity from PSTs be more harmful than that of conventional sources.

Figure 4 shows the relative impacts of the different inorganic emissions coming from upstream processes e.g., mining of metals to be used in panel production, etc. For PSTs, aluminum and iron are the two largest emissions—45% each, Figure 4.a.—and cause the practical totality of the ecotoxicity impact of electricity supplied from PST modules—90% and 10% respectively, Figure 4.b. However, we found their impacts were negligible in terms of human toxicity. Most of the impact in this last category is caused by silver, arsenic, chromium, mercury, and zinc emissions.

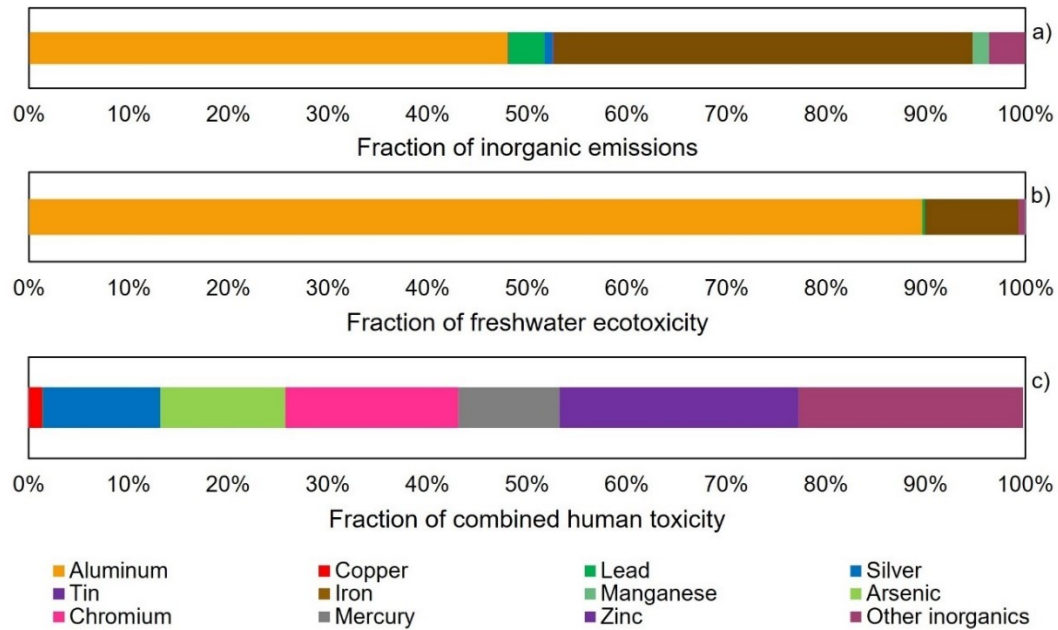


Figure 4 Inorganic emissions as a fraction of the a) mass, b) freshwater ecotoxicity impact, and c) combined human toxicity impact. Unless already included in the figure, other inorganics include Sb, As, Ba, Be, Cd, Cs, Cr, Co, Fe, Mn, Hg, Mo, Ni, Pa, Rh, Sc, Se, Sr, Te, Tl, Th Ti, W, V, Zn, and Zi. There is neither release of In in any of the processes evaluated, nor a characterization factor for this metal in USEtox 2.2

Figure 5 shows the sources of inorganic emissions in terms of mass (a), freshwater ecotoxicity (b), and human toxicity (c). Metal procurement is responsible for 71% of all emissions and their related ecotoxicity, and for 67% of the human toxicity impacts. The remaining emissions and impacts come mostly from the fabrication of the single-Si sub-cell—24%—while 2% comes from the electricity used to manufacture the tandem structure, and the remaining 4% from various other sources—e.g., transportation, auxiliary chemicals, and other elements of the panel. 43% of all inorganics released during metal procurement, 45% of the ecotoxicity and 23% of the human toxicity come as a result of silver procurement—Figure 5. This is particularly relevant when we consider that the silver—required for the back contact metallization in PST modules—is used in smaller quantities than other metals (<0.5% of metal weight). Copper cables—~5% of metal weight—are the second main source of inorganic emissions during metal procurement. Finally, the procurement of Al—~94% of metal weight— releases only 5% of all inorganics. A detailed

description of the fraction of different inorganic emissions, broken down by their metal procurement source can be found in tables S7-9.

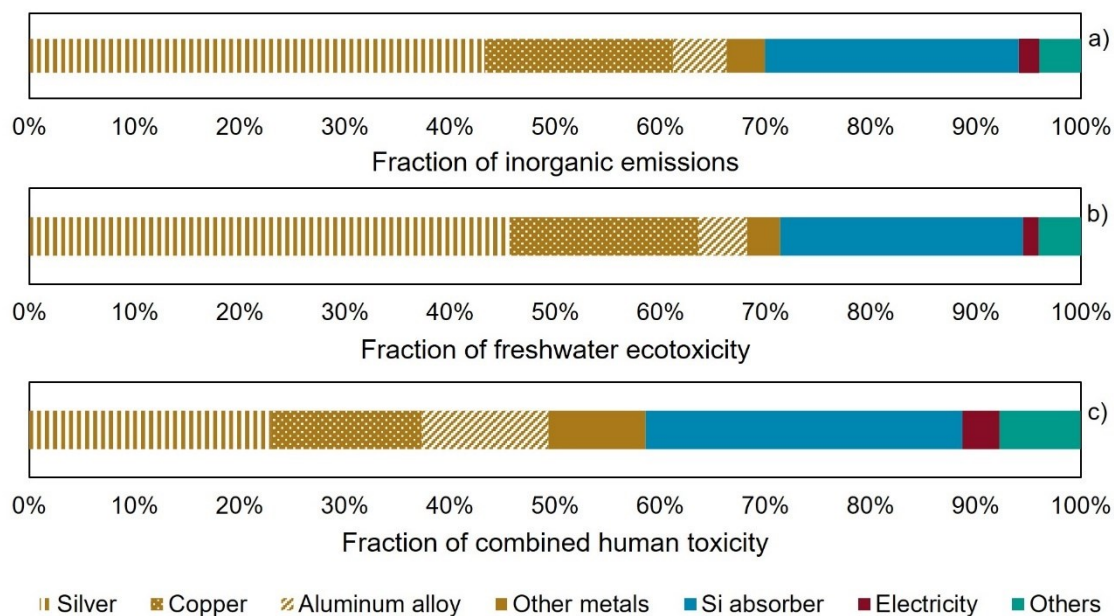


Figure 5 Sources of inorganic emissions as a fraction of the a) mass, b) freshwater ecotoxicity impact, and c) combined human toxicity impact (See Table S.10 for the raw data of the figure)

Due to the impacts of silver, copper, and aluminum alloy procurement, we evaluated which of their production processes contributed the most to inorganic emissions—and thus to the toxicity—of PSTs. For all inorganics assessed, emissions from Al frame procurement are roughly divided between production in Europe and the rest of the world according to their market shares—33% and 67% respectively—Figure S3. Emission percentages from copper and silver procurement are also very similar for all inorganics with the exception of arsenic. However, neither of them follow market shares. For example, electrowinning represents 17.5% of the copper market, but it emits more than a third of most inorganics, indicating it is a particularly harmful copper source. Similarly, primary silver production in the rest of the world—which in this case is the entire world minus Chile, Quebec, and Sweden—has 76.3% of the market share. However, it is responsible for

more than 90% of all inorganic emissions excluding arsenic. For that reason, we further evaluated this production process and found out most of its emissions came from the impoundment of sulfidic tailings—see Figure 6. Since silver procurement is responsible for 62% of all the inorganic emissions released during metal procurement, any improvement in the treatment of these tailings would translate into a significant reduction of the life cycle toxicity of PSTs.

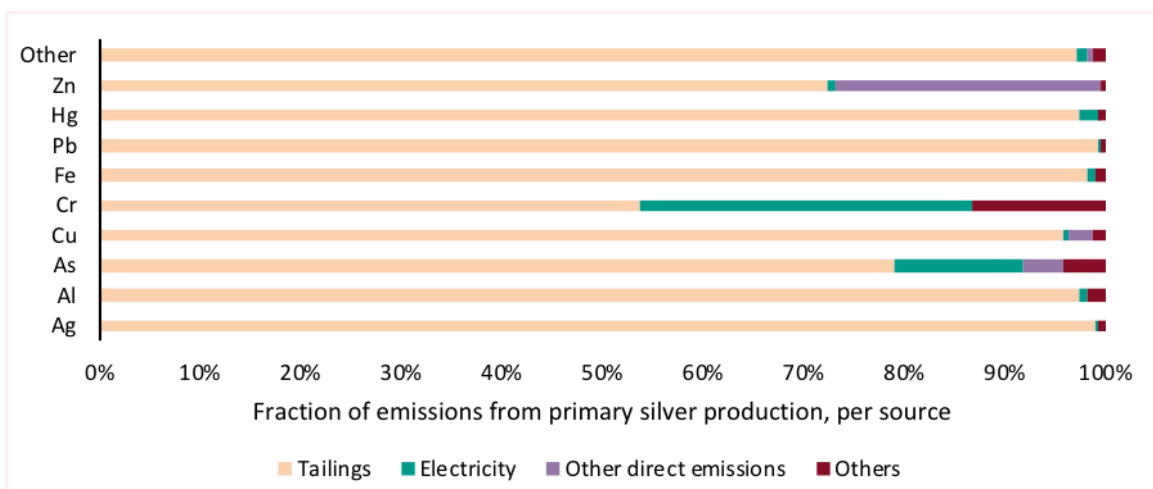


Figure 6 Fraction of inorganic emissions from silver primary production in the rest of the world (RoW: silver-gold mining operation, with refinery), detailed by source

Assessment of end-of-life

In Figure 7.1, we presented the freshwater ecotoxicity and combined human toxicity impacts of recovering valuable components and toxic metals (lead) from PST, allocated by mass. It also included as a benchmark the impacts of procuring those materials from the market, which would represent the environmental cost of building a completely new device. Overall, recovery of components from PST is between 7 and 112 times less impactful than procuring those same materials from the market. The rank between the different techniques from highest impact to lowest impact is Binek et al. > Feng et al. > Zhang et al.^{34,36,37} The recovery of the top glass drives this classification. This is because the steps for out-module metal recovery and bottom glass and cell recovery in-module are all based on FRELP, not on the delamination techniques. The latter

are unique in how they recover the in-module lead from the absorber and the top glass. Since the top glass has the largest mass, the cleanest technique would be the one recovering it with the lowest environmental impact: the Zhang technique³⁶. We can draw a similar conclusion from Figure 7.2—both a) and b). There we showed the fraction of the total impacts caused by the recovery or manufacturing of each layer. The fraction of impacts allocated to the recovery of the top glass in this technique is much smaller than in the other two. The reason for that is the lack of a drying process after delamination, which is consistent with the idea postulated in Rodriguez-Garcia et al.³⁸ that solvent reuse and recovery might be insufficient to make certain recycling techniques sustainable. Contrarily, the Zhang technique³⁶ presents the highest impact on the recovery of lead. Like the Feng technique³⁷, it requires two steps after delamination to recover lead, and although it uses fewer toxic chemicals, the amount of water needed makes it a less competitive technique. That said, while the benefits of recovering in-module lead and top glass are limited, the recovery of the bottom glass and cell is more likely to be beneficial— as it is 200-750 times less impactful than the initial material. As shown in Figure 7—again, both a) and b)—most of the toxicity potential of the initial device is in this layer. It is thus, in environmental terms, the key driving force for recycling PSTs.

The results of the cradle to end-of-use section agree with those presented here. Freshwater and human toxicity concentrates in the metal components and the single-Si sub-cell, and within the former, in silver, copper, and aluminum production. This is also shown in Figure 7 a) and b). Since both the single-Si sub-cell and silver are included in the “bottom glass and cell,” the recovery of this component is of the utmost importance. We can do that at a low environmental cost during the common—FREL—part of the recycling process, and clearly benefits the overall result. To a lesser extent, it is the same situation for copper recovery, but not for aluminum. The recycling of

this last metal is only slightly less toxic than procuring Al used in frame from the market. Finally, EoL results also de-emphasized the deleterious role of lead might have. Although we can recover lead—both in-module and out-module—at a lower impact than if purchased from the market, environmental benefits are not as significant as the recovery of the silicon cell, silver, or copper.

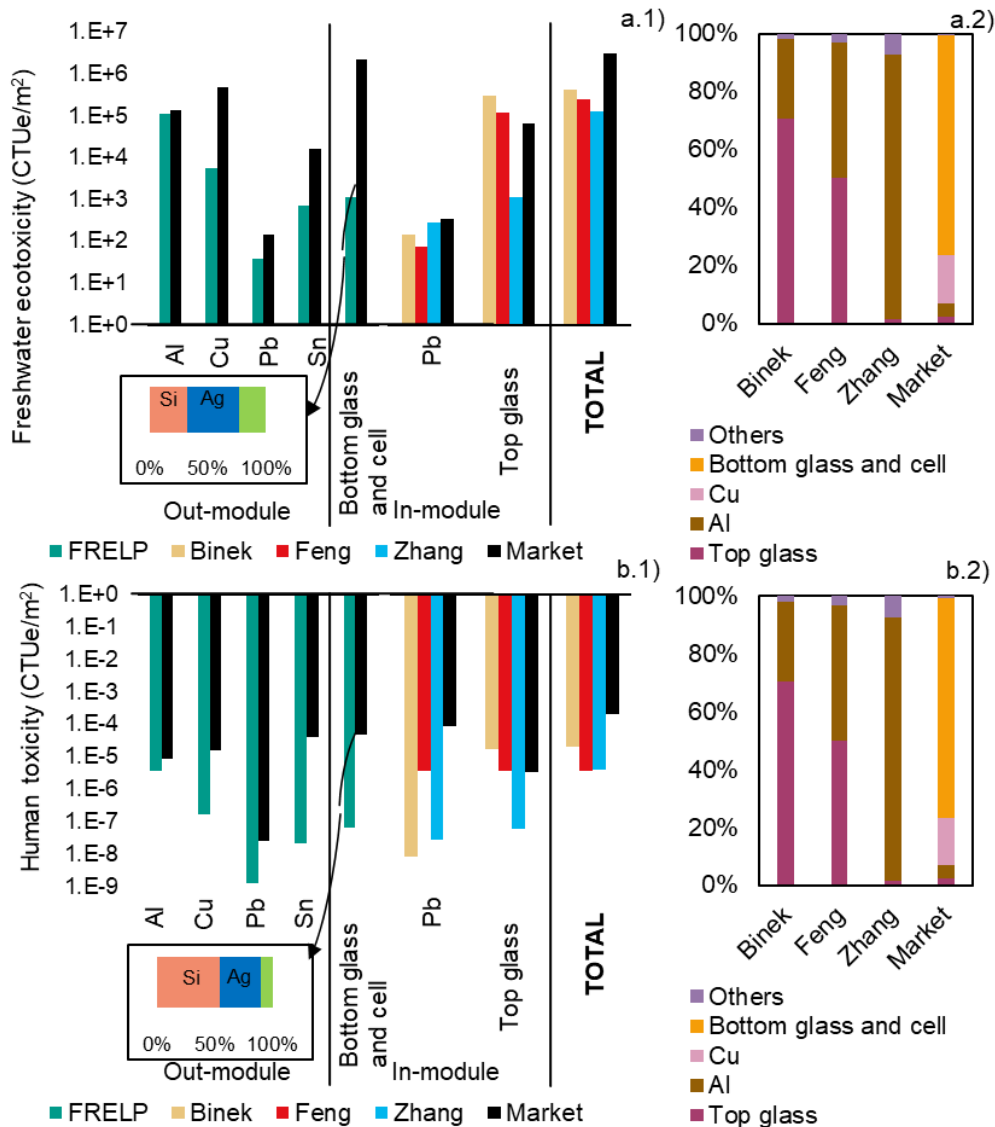


Figure 7 a) Freshwater ecotoxicity and b) human toxicity during recycling for the recovery of metals, top and bottom glass, and caused by procuring the same products using materials straight from the market a.1) per metal/layer, and b.1) as a share of the total impacts. The market scenario includes release of lead

Conclusions

In this study, we have evaluated the life cycle toxicity of PSTs using USEtox 2.2. as impact assessment method. We then compared their cradle to end-of-use impacts with those of other electricity sources. We contrasted the impacts of its EoL—three variants of a proposed recycling process—with those of procuring the materials needed to manufacture a new PST from the market. We found that the life cycle toxicity of PV in general is mainly driven by metal emissions. PSTs in particular emit more metals—and these are more toxic—than other PVs, but less than conventional sources of energy. A lower silver content—or more sustainable silver procurement—would be the first step towards making PSTs more environmentally sustainable. Concerning the EoL analysis, all proposed variants are less impactful than procuring materials from the market. Their largest benefits can be found in the recovery of the bottom glass and crystalline silicon sub-cell, the copper cables, and the top glass.

CRedit authorship contribution statement

Conceptualization: I.C., Methodology: G.R.G., Formal Analysis: G.R.G., Investigation: G.R.G., Resources: I.C., Data curation: G.R.G., Writing – Original Draft: G.R.G., I.C., Writing – Review & Editing: G.R.G., Z.Z., J.K., E.A., S.D.W., I.C., Visualization: G.R.G., Supervision: I.C., Project Administration I.C., Funding Acquisition: I.C.

Declaration of competing interests

The authors declare no competing interests.

Supporting Information

See supporting information regarding details of the presented data in figures and inventories of the solar PV manufacturing and recycling process

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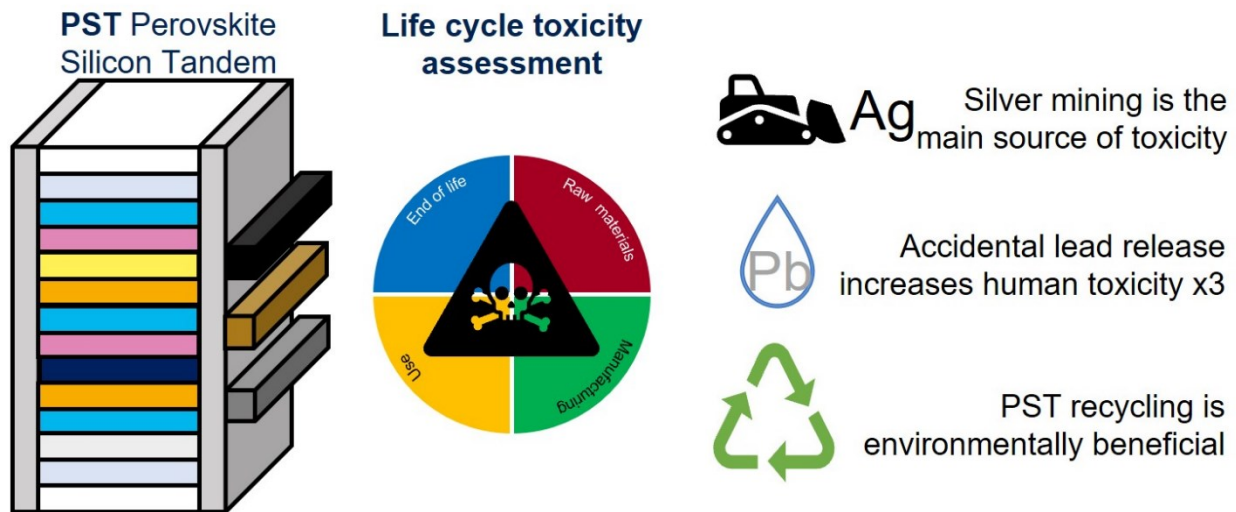
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More sustainable solar energy can be generated from PST by reducing silver content or by sourcing silver more sustainably.