

Comparison of Perovskite Solar Cells with other Photovoltaics Technologies from the Point of View of Life Cycle Assessment

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A review of the life cycle sustainability of perovskite solar cells (PSCs) is presented, distinguishing results between simulated laboratory-based and simulated industrial-based PSCs, comparing this technology with the commercial photovoltaic (PV) technologies. Laboratory-based perovskites are seriously affected by the unrealistic energy consumption of the deposition routes. Moreover, other hot spots such as lead and solvent toxicity are much less relevant. A cluster analysis of single-junction PSCs allows us to differentiate two clusters, in one of them, the most numerous, environmental impacts are in a similar range to other thin film PVs. Despite more progress at the industrial scale being needed, the current studies point out the promising results in terms of energy payback time and the environmental impacts at the industrial-scale, that are the lowest compared with commercial PVs, if 1 kWp is used as the functional unit. In addition, PSCs present less supply risk than other photovoltaic technologies. Only cesium, which is dispensable, can present a specific risk. Current production costs could be reduced if barrier foils and transparent conductive oxide coated plastics can be found. However, this cost could be competitive selling into niche markets. In addition, their lightness, flexibility and ease of integration give them very advantageous qualities for social acceptance.

Converting all this incident energy would suppose nearly 158 000 EJ per year, which greatly exceeds the 585 EJ of primary energy (PE) consumed in 2017.^[3] This fact makes solar power technologies converting directly incident sunlight into usable electrical energy, hence, PVs, powerful candidates to ease the environmental issues derived from the present system of energy production. However, the potential of PV to provide electricity to our societies in a post-carbon energy system is limited by a Shockley–Queisser limit of about 33%,^[4] together with land and sources availability.

There are several PV technologies, each with a different degree of maturity. Crystalline Si PVs represent the most deployed type with 95% of the market share, 75% in monocrystalline silicon (Mono-Si), with a growing share, and 20% for multi-crystalline silicon (Multi-Si), with a continuously declining share.^[5] Mono-Si and Multi-Si present moderately high operating efficiencies between 20% and 22%, a deep


industrial implementation constructed in parallel with the development of the electronic industry and low toxicity.^[6] Another key advantage of this technology in a large production scenario is that it can be entirely produced with relatively abundant materials.^[7] The only argument against crystalline Si as the ideal PV material is the chemistries required for purification, reduction, and crystallization of pure silicon from sand, which are highly energy

1. Introduction

Earth receives from the sun ≈ 432 EJ in 1 h, out of which 18 EJ per hour are reflected off from the surface and lost into space.^[1] Despite the fact that this amount of energy is available to be converted to usable energy by photovoltaics (PVs), nowadays, this power technology is just converting about 4 EJ per year.^[2]

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demanding and polluting.^[8] Furthermore, it is a highly optimized technology, which leaves little margin for enhancing the efficiency.^[9]

Thin-film PV types possess the remaining 5% of the market share in 2017.^[10] This type of PV technology needs less energy in the manufacture process. Among the absorbing materials used for this technology, large-scale production of CdTe and Cu(In,Ga)Se₂ (CIGS) cells is constrained by the scarcity of Te and In.^[11] The highest efficiency for a single-junction device belongs to GaAs absorbing material,^[12] despite its Ga content also supposes a bottleneck for large-scale deployment.^[13]

Dye-sensitized solar cells (DSSCs),^[14–16] full organic PV (OPV) solar cells,^[17,18] perovskite solar cells (PSCs),^[19–22] and quantum dot solar cells (QDSCs)^[23,24] technologies are considered as emerging PV technologies.^[25] In general, emerging technologies may not have reached the market yet or have only been introduced into minor niche markets. Among these technologies, DSSC has reached the highest level of maturity, suitable for different niches of the market, i.e., building-integrated PV (BIPV) systems and indoor energy-harvesting applications.^[14]

Among all the types of currently existing PV technologies, those based on halide PSCs raise much interest among the scientific community, as they potentially offer high efficiencies at a low cost.^[26] In roughly a decade, as hybrid organic inorganic methylammonium (MA) lead halides in a perovskite crystal arrangement were discovered as a promising absorbing material for DSSCs,^[27] their record efficiency for a little area cell is positioned at 25.5%.^[28] This efficiency with non-concentrated light is comparable to that of already commercialized PV technologies with the highest efficiencies, such as crystalline Si (26.1%), CIGS (23.4%), CdTe (22.1%), and GaAs (27.8%).^[29] A second pillar standing for their potentiality is that they can be manufactured through solution processing, which is a simple and cost-effective process of production.^[30] These advantages are still not enough for successful commercialization of PSCs, because some challenges remain to be solved. The main one is to achieve long lifetimes with good stability at module level. Although many signs of progress have been made, it is still challenging for PSCs to pass the most popular international standards of IEC61215:2016 for mature PV technologies. Second, the toxicity problems must be addressed, especially those arising from the use of lead^[31] and solvents.^[32]

Comparison of different PV technologies using a single parameter as efficiency is unfair, as it does not consider all the impacts originated by the fabrication and utilization process. In this context, a very powerful tool to analyze the environmental impacts of a product or a service is the life cycle assessment (LCA). The LCA methodology has been used to analyze different aspects of PSC, as it has been discussed in previous reviews.^[19,21,25,33–38] Here, we present a review of the LCA of PSC, treating distinct issues than in previous reviews, focusing in the comparison of PSCs with currently commercial PV technologies. These distinguished aspects treated herein comprise the difference between laboratory-based and industrial-based simulated PSCs, harmonized comparison of PSCs with silicon-based, CdTe and CIGS PV technologies using a functional unit independent of the lifetime; consequently, stability issues are not considered, as it is field experiencing a continuous progress, and currently, there is no clear benchmark. The review includes

the discussion, based on the LCA methodology, of hot topics of the PSCs such as the end-of-life (EOL), lead toxicity, solvents toxicity, and encapsulation. In addition, in the way to progress from LCA to life cycle sustainability assessment (LCSA)^[39,40] for PSCs, we present some briefs about costs, criticality, and social impacts.

2. Methodology

Solar cells are conceived to generate electricity from the sunlight energy. Their main advantage lays in the fact that they can generate electricity for a wide variety of purposes from a renewable source, without generating neither emissions nor residues in the process. However, are not solely the whole of the product impacts generated during its use stage, but during the entire life cycle. In the case of PSCs, although devices based on perovskite are produced via a relatively simple process, it becomes necessary to verify that impacts generated during their whole life cycle are acceptable. For this purpose, LCA emerges as the most appropriate tool.

2.1. LCA Methodology

LCA is a methodology to evaluate the environmental burdens associated with a product, process, or activity by identifying and quantifying energy and materials used and wastes released to the environment, to assess the impact of those energy and material uses and releases to the environment, and to identify and evaluate opportunities to affect environmental improvements. The assessment from cradle-to-grave includes the entire life cycle of the product, process, or activity, which encompasses: extracting and processing raw materials; manufacturing, transportation, and distribution; use, re-use, and maintenance; recycling and final disposal. Sometimes, the assessment comprises a partial product life cycle. This is the case of the cradle-to-gate, which comprises from resource extraction to the factory gate.

According to ISO 14040 and 14044, LCA consists of four phases: goal and scope definition, inventory analysis, impact assessment, and interpretation. The inventory analysis involves collecting data to create a life cycle inventory (LCI) of the inputs (energy and materials) and outputs (environmental releases and waste) associated with each stage of the life cycle. The impact assessment translates the LCI data into potential environmental impacts. To this end, the impact categories under study must be defined, the inventory data must be assigned to specific impact categories, and the level of impact must be evaluated according to predefined assessment methods.

2.2. Functional Unit

The International Energy Agency Photovoltaic Power Systems Programme (IEA PVPS) Task 12 has compiled PV-specific LCA guidelines,^[41] e.g., functional unit, life expectancy, impact categories, etc., as well as LCI for major commercial PV technologies.^[42,43] In this context, the functional unit allows consistent comparisons to be made of various PV systems and of other electricity-generating systems that can provide the same

function. kWh of electricity generated is the recommended functional unit,^[41] with the inconvenience that it is necessary to assume a lifetime for the solar cell to perform the calculation.^[19] Life expectancy for mature module technologies is 30 years. Despite the continuous progress in the increments of PSC stability, PSCs currently cannot ensure this life expectancy. Comparisons with so much difference in lifetime, that, on the other hand, is continuously evolving, would bring even more uncertainty. Consequently, we have used another functional unit, the nominal power, kW_p , more convenient for emerging PV with different efficiencies,^[25,44,45] and noting that the differences in the amount of kWh fed to the grid may differ between the systems analyzed. This functional unit considers the PV module area, A , and the maximum efficiency [power conversion efficiency (PCE)], η , under standard test conditions, which are a light intensity, E , of 1 kW m^{-2} and a cell temperature of $25\text{ }^\circ\text{C}$, according to the standard IEC 61215 (Equation (1)).^[46]

$$kW_p = A \cdot \eta \cdot E \quad (1)$$

2.3. System Boundaries

This review is focused on the environmental impacts of the PV panel, materials, manufacture of the panel, and EOL. Excluding mounting systems, cabling, inverters, and all further components needed to produce electricity and supply it to the grid and the construction stage. Except for the estimation of the Energy payback time (EPBT), the use stage is not considered.

The most common PSC devices include four architecture types that are mesoporous, planar, inverted planar, and inverted mesoporous. The materials considered in the life cycle inventories for a mesoporous n-i-p architecture are the substrates, the transparent conducting oxide (TCO) such as fluorine-doped tin oxide (FTO) and indium-doped tin oxide (ITO), that is deposited on a substrate and serves as a front electrode; an electron transport layer (ETL) scaffold with nanoscale pores; a perovskite absorber layer that covers the scaffold, forming a compact layer, penetrating into the scaffold, and leading to an intermixed layer; a hole transport layer (HTL), that is a hole conductor deposited on the perovskite layer; and a metal back electrode. The planar n-i-p architecture contains a compact ETL layer instead of a mesoporous ETL-perovskite intermixed layer. When the deposition order is changed and the HTL layer is deposited first, the device is fabricated in the p-i-n structure, as either inverted planar or inverter mesoporous.

These data of the PSCs devices reviewed are extracted from the literature: Architecture, substrate material and front contact, ETL layer, composition of active layer, deposition method, HTL layer, back contact, encapsulant, PCE, and environmental impacts for 1 m^2 .

2.4. Environmental Impact Assessment

In this review, two of the most common impact categories used to compare PV technologies are selected, the cumulative energy demand (CED) and the global warming potential (GWP).^[25,33,44] The CED, in MJ, quantifies the PE inputs of the included life

cycle stages, and the GWP quantifies the greenhouse gas emissions in kg of carbon dioxide equivalents ($\text{kg CO}_2\text{-eq}$). The environmental impacts of these categories are harmonized for kW_p with Equation (1).

2.5. Environmental Impacts Interpretation

One common metric for LCA interpretation is the EPBT. EPBT is defined as the period required for a renewable energy system to generate the same amount of energy (in terms of PE equivalent) that was used to produce the system itself, Equation (2).

$$EPBT = \frac{CED}{\frac{E_{\text{agen}}}{\eta_G} - E_{O\&M}} = \frac{CED}{\frac{I \cdot PR \cdot 3.6}{\eta_G} - E_{O\&M}} \quad (2)$$

where E_{agen} is the mean annual electricity generation, η_G is the PE to electricity conversion efficiency, and $E_{O\&M}$ is the annual PE demand for operation and maintenance. E_{agen} can be expressed as a function of the irradiation in $\text{kWh m}^{-2}\text{ year}^{-1}$, I , and the performance ratio, PR , which quantifies the overall system losses due to temperature effects, soiling, shading, and inefficiency of its components. In this case, CED and $E_{O\&M}$ are in MJ m^{-2} . To harmonize the results, the values for I , η_G , PR , and $E_{O\&M}$ are $1700\text{ kWh m}^{-2}\text{ year}^{-1}$, 0.35, 0.75, and 0 MJ m^{-2} , respectively.

3. Review of LCAs of PSCs

The literature review is divided in LCA of single-junction PSCs based on laboratory data, tandem of PSCs with other PV technologies, and industrial simulation-based perovskites. Table 1 and 2 show a summary of LCA studies with materials and perovskite deposition processes for single-junction and tandem PSCs, respectively. Furthermore, the next sections are focused on hot topics as EOL, lead toxicity, solvent toxicity, and encapsulation.

3.1. Prospective LCA at Laboratory Scale

The first LCAs of PSCs were published in the year 2015; see Table 1. One of the first LCA study was focused in comparing two laboratory-scale perovskite layer deposition processes, which are solution- and vapor-based processes.^[47] The research of this team was continued comparing tin-based and lead-based PSCs discerning several disadvantages on the first one.^[48] In other LCA, the perovskite was deposited in a sequential method, the PbI_2 reagent is spin-coated and the methylammonium iodide (MAI) is dipped.^[49] The main goal of this study was to compare two devices using TiO_2 and ZnO as ETL. A subsequent LCA,^[50] also published in 2015, evaluated a device where perovskite is deposited through the previous same two-step deposition method, although with titanium dioxide nanotubes as ETL and a liquid HTL, inherited from DSSCs, but it was soon identified as an instability inducer, and so replaced it with the solid-state HTL.^[26]

Over the following five years, several topics were subject of LCAs. One of them was to compare the three more used recipes for perovskite deposition in laboratory using spin-coating for both planar and mesoporous architectures: spin-coating of stoichiometric precursor solution, spin-coating of precursor solution

Table 1. LCA studies on single-junction PSCs with materials for each layer, perovskite deposition layer, efficiency, carbon footprint (GWP), and CED per kW_p. First column indicates the observation number and the reference. Source of the energy consumption for perovskite layer deposition is indicated. Symbol (*) means data obtained by the authors of the study.

N	Front	ETL	Meso	Active layer	Deposition	HTL	Back	PCE [%]	GWP Kg CO _{2eq}	CED MJ
1 ^[64]	Glass: FTO	SnO ₂	–	MAPbI ₃	Spin-coating ^[52,74]	Free	Carbon	14.5	308	5131
2 ^[64]	Glass: FTO	TiO ₂	TiO ₂	MAPbI ₃	Spin-coating ^[52,74]	Free	Carbon	11.5	387	6443
3 ^[53]	Glass: FTO	TiO ₂	TiO ₂ , ZrO ₂	MAPbI ₃	Screen printing*		Carbon	11.0	304	6580
4 ^[53]	Glass: FTO	TiO ₂	TiO ₂	MAPbI ₃	Spin-coating 1:1* ^[51]	Spiro	Au	19.0	391	8773
5 ^[51]	Glass: FTO	TiO ₂	–	MAPbI ₃	Spin-coating 1:3*	Spiro	Au	11.4	504	9253
6 ^[51]	Glass: FTO	TiO ₂	–	MAPbI ₃	Spin-coating 1:1*	Spiro	Au	10.4	554	10 190
7 ^[51]	Glass: FTO	TiO ₂	–	MAPbI ₃	Spin-coating + dipping*	Spiro	Au	15.0	522	9929
8 ^[51]	Glass: FTO	TiO ₂	TiO ₂	MAPbI ₃	Spin-coating 1:1*	Spiro	Au	12.3	606	10 956
9 ^[65]	Glass: FTO	TiO ₂	TiO ₂	MAPbI ₃	Vapor deposition ^[47]	Spiro	Au, PET	15.1	346	4773
10 ^[65]	Glass: FTO	TiO ₂	TiO ₂	Cs _(x) FA _(1-x) PbI _(3-y) Br _y	Spin-coating ^[74]	CuSCN	Cu, PET	21.1	212	2374
11 ^[72]	Glass: FTO	TiO ₂	TiO ₂	MASnI _{3-x} Br _x	Spin-coating*	Spiro	Au	5.7	3099	157 068
12 ^[72]	Glass: FTO	TiO ₂	TiO ₂	MAPbI ₃	Spin-coating*	Spiro	Au	20.0	1209	60 300
13 ^[72]	Glass: FTO	–	TiO ₂	FAPbI ₃	Spin-coating*	Spiro	Au	15.6	1458	75 578
14 ^[72]	Glass: FTO	TiO ₂	TiO ₂	CsPbBr ₃	Spin-coating*	Spiro	Au	4.9	3074	157 377
15 ^[72]	Glass: FTO	TiO ₂	TiO ₂	MAPbI ₂ Cl	Spin-coating*	Spiro	Au	10.9	1635	82 018
16 ^[54]	Glass: ITO	TiO ₂		MAPbI ₃	Thermal evaporation and slot die ^[57]	NiO	Ag	13.8	740	8590
17 ^[52]	Glass: FTO	SnO ₂		MAPbI ₃	Spray ^[74]	CuSCN	MoO _x Al	15.0	1047	17 000
18 ^[52]	Glass: FTO	SnO ₂		MAPbI ₃	Vacuum deposition ^[74]	CuSCN	MoO _x Al	15.0	1253	20 267
19 ^[52]	Glass: FTO	SnO ₂		MAPbI ₃	Spray ^[74]	Free	Carbon	15.0	847	13 800
20 ^[49]	Glass: FTO	TiO ₂	TiO ₂	MAPbI ₃	Spin-coating + dipping* ^[74]	Spiro	Au, PET	9.1	242	4945
21 ^[49]	Glass: FTO	ZnO		MAPbI ₃	Spin-coating + dipping ^[74]	Spiro	Ag, PET	11.0	173	3636
22 ^[48]	Glass: FTO	TiO ₂	TiO ₂	CH ₃ NH ₃ SnI ₃	Spin-coating ^[47]	Spiro	Au	6.4	14552	140 760
23 ^[47]	Glass: FTO	TiO ₂		PbCl ₂ /CH ₂ NH ₃ I	Vapor deposition*	Spiro	Ag	15.4	7452	70 307
24 ^[47]	Glass: ITO	[6,6]-phenyl-C ₆₁ -butyric acid methyl ester (PCBM)		PbCl ₂ /CH ₂ NH ₃ I	Spin-coating*	PEDOT:PSS	Al	11.5	7126	79 558
25 ^[50]	Glass: FTO	TiO ₂	TiO ₂	MAPbI ₃	Spin-coating + dipping*	Liquid electrolyte	Glass: Pt	6.5	4400	155 077
	PSC, industrial simulation	–	–	–	–	–	–	–	–	–
26 ^[77]	Glass: ITO	ZnO	–	MAPbI ₃	Screen printing*	NiO	Al, EVA	16.0	–	3613
	Partial LCA	–	–	–	–	–	–	–	–	–
27 ^[58]	Glass: FTO	TiO ₂	–	MAFACsPbI ₃ Br	Spin-coating, FIRA*	Spiro	Au	17.3	–	–
28 ^[58]	Glass: FTO	TiO ₂	–	MAFACsPbI ₃ Br	Spin-coating ^[51]	Spiro	Au	17.3	–	–
29 ^[73]	–	–	–	Cs _x [MA _{0.17} FA _{0.83}] _(1-x) Pb[_{0.83} Br _{0.17}] ₃	Spin-coating*	16–22%	–	–	–	–

using lead chloride precursor, and the two-step deposition method.^[51] Spin-coating is the most used deposition method on a laboratory scale, and this is also reflected in the LCAs; see Table 1. However, it is not appropriate on an industrial scale. Other deposition methods present a greater potential to be used in a large-scale arrangement^[21] and for which there are still few LCAs: spray,^[52] screen printing,^[53] and slot die.^[54] This last study combines laboratory data,^[49,55] ecoinvent data,^[56] IEA PVPS LCI,^[42] and data from suppliers for slot die.^[57]

The required time for the annealing of the perovskite layer is other handicap for the commercialization of the PSCs. Use of fast IR annealing (FIRA), notable improvement in the annealing step of the perovskite is evaluated through LCA.^[58] FIRA uses infrared rays to anneal the perovskite layer of a planar device in just 1.2 s, instead of the antisolvent method followed by relatively long annealing (tens of minutes) in hotplate performed after perovskite deposition, to produce the final perovskite crystallization. Compared with conventional annealing, FIRA

Table 2. LCA studies on tandem PSCs with materials for each layer, perovskite deposition layer, efficiency, carbon footprint (GWP), and CED per kW_p. First column indicates the observation number and the reference. The column PV indicates the PV technology in tandem with PSC. Source of the energy consumption for perovskite layer deposition is indicated. All tandems are encapsulated with EVA^[54,66] or PET with a pressure sensitive adhesive.^[67]

N	PV	Front	ETL	Active layer	Deposition	HTL	PCE [%]	GWP Kg CO _{2eq}	CED MJ
30 ^[54]	SHJ	Ag, ITO	TiO ₂	MAPbI ₃	Thermal evap. & slot die ^[57]	NiO	23.8	1698	20 004
31 ^[66]	Si	MoO ₃ , ITO, Ag	TiO ₂	MAPbI ₃	Spin-coating ^[47]	Spiro	27.0	7650	19 671
32 ^[66]	Si	MoO ₃ , ITO, Au	TiO ₂	MAPbI ₃	Spin-coating ^[48]	Spiro	27.0	7650	19 671
33 ^[66]	Si	ZnO, ITO, Al	PCBM	MAPbI ₃	Spin-coating ^[47]	PEDOT:PSS	24.0	4800	17 049
34 ^[67]	Si	MoO ₃ , ITO, encapsulant	PCBM	MAPbI ₃	Spin-coating ^[52,74]	Spiro	21.0	802	14 286
35 ^[67]	CIGS	FTO, encapsulant	TiO ₂	MAPbI ₃	Spin-coating ^[52,74]	Spiro	19.5	397	8128
36 ^[67]	CZTS	Al, encapsulant	PCBM	MAPbI ₃	Spin-coating ^[52,74]	PEDOT:PSS	6.0	546	9150
37 ^[67]	PSC_Sn	Glass: ITO	PCBM	CH ₃ NH ₃ (Sn,Pb)I ₃	Spin-coating ^[48,52,74]	NiO	21.0	284	1995

method presents environmental impacts one order of magnitude lower respect a conventional process using an antisolvent. Therefore, it is undoubtedly more adequate for large scale production, not only for its lower environmental impact but also for its lower cost. The fact that the annealing can be performed outside a glove box through this method is another advantage for large-scale production.^[59]

The cathode of precious metals such as gold or silver is responsible for a high environmental impact and a high economic cost. The carbon stack configuration allows replace them by carbon, and it is suitable to be produced at large scale.^[60] Another important advantage of this configuration is the elimination of the HTL layer, usually composed of Spiro-OMeTAD, which is also expensive and decreases the stability of the whole device.^[61] The most comprehensive LCA of a carbon stack configuration considers mesoporous titania and zirconia layers via screen printing in a pre-industrial process,^[53] whereas the perovskite can be uniformly infiltrated in the stack with a robotic dispenser.^[62] Large-area carbon-stack modules are manufactured through this process.^[63] Other LCAs for this configuration include spray deposition^[52] and spin-coating.^[64] Alternatives to the Spiro-OMeTAD such as CuSCN,^[52,65] poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS),^[47,66,67] or NiO^[54,67] are also included in the LCAs.

Several LCA studies were focused on analyzing new perovskite compositions to mainly improve their performance properties stability, reproducibility, and efficiency. The principal difference in their composition supposed the introduction of formamidine (FA) and cesium cations. For instance, introducing FA cation boosted its PCE above 20%.^[68] Through the so-called compositional engineering, researchers sought improving the PCE and stabilizing the perovskite phase.^[69] Both properties were also improved while tuning the perovskite's bandgap, by combining Cs with FA in the cationic mixture.^[70] Recently, the stability was further optimized by introducing Rb to the perovskite composition.^[71] In this line, the effect of MA, FA, and Cs and different halides was assessed.^[65,72,73]

Main conclusion from this review is that the environmental impacts are seriously affected by the unrealistic energy consumption of the deposition routes used at laboratory scale.^[21,47,49–51] In addition, it has to be highlighted that several studies^[49,52,64,65,67] have considered energy data from the same source, originally for

OPV at laboratory scale^[74] and applied different use factors. Use factors are also applied to correct the excessive losses of the materials and solvents during processing.

Other environmental hotspot is the use of gold as cathode. Substituting the gold cathode by, for instance, copper is considered as a necessary step toward commercialization of PSCs.^[51,65,75] Other high impacts are related to the use of glass and TCO.^[73]

Environmental impacts of single-junction PSC are represented in **Figure 1** for GWP and **Figure 2** for CED, and further discussion is provided in Section 4. These box plots represent the median, the lower, and the upper quartiles for each PV technology. The lines extending from the boxes indicate the variability outside the upper and lower quartiles, and outliers are plotted as individual points. This variability is very high for CED, and in the case of GWP, PSCs analyzed by Krebs, Urbina, and co-workers^[47,48] are plotted as outliers in **Figure 1**.

The values in Table 1 should be considered with great caution when making comparisons. Comparisons are valid when they are expressly made by the same researcher and with the same assumptions. However, energy and material consumptions are

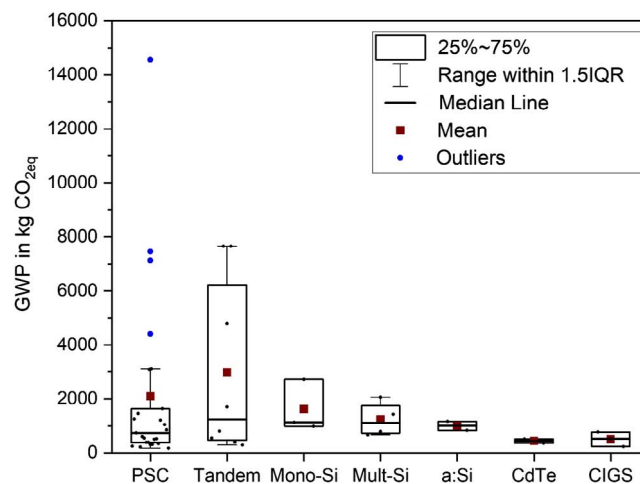


Figure 1. GWP compared for 1 kW_p. Data are in Table 1 for PSC and PSC-ind, in Table 2 for tandem PSC, and in Table 3 for the rest of PV technologies.

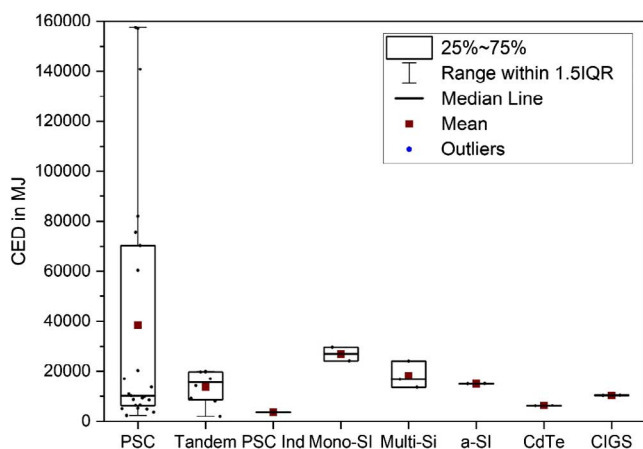


Figure 2. CED compared for 1 kW_p. Data are in Table 1 for PSC and PSC-ind, in Table 2 for tandem PSC, and in Table 3 for the rest of PV technologies.

still so unrealistic and with different estimation criteria that are very specific comparisons, such as a change of a material in a layer or a change in the deposition process or a change in architecture, if between different researchers may not be clarifying. Therefore, our interest is focused on statistically analyzing the GWP and CED of the PSCs compared with other PV technologies.

3.2. Perovskite Tandem Solar Cells

Halide perovskite can be combined with the first and second generation solar cells in either monolithic 2-terminal or stacked 4-terminal tandem solar cells with high efficiencies. Several LCA studies are published, addressing this matter covering Si-PSC tandems,^[66,67] silicon heterojunction (SHJ)-PSC tandem,^[54] and further PV technologies as CIGS-PSC, Cu₂ZnSnS₄ (CZTS)-PSC, and PSC of Sn-PSC;^[67] see Table 2. Authors underline that the ability of perovskite to transmit both electricity and light after failure is essential to improve their environmental results, as the silicon layer continues functioning.^[66] All results confirm that the tandem perovskite-Si is beneficial to the environment, and these results are expected to improve as tandems technology matures.^[54,66,67] Options of interest include tandems constructed with a wide bandgap lead-based perovskite top cell and a low bandgap bottom cell of Si, CIGS, CZTS, and Sn-PSC. With a higher PCE and a lifetime comparable to state-of-the-art devices, the all-perovskite tandem was found to be the most promising PV technology for lowering the environmental impacts of solar PV.^[67]

Environmental impacts of single-junction PSC and tandem PSC are in the same range, considering their high uncertainty; see Figure 1 for GWP and Figure 2 for CED.

3.3. Advancing toward Large-Scale Production

Applying LCA to support upscaling of a laboratory scale generates results with high uncertainty;^[76] see Figure 1 and 2. An alternative is industrial-based simulated PSCs.

One industrial-based simulated PSC is a techno-economic analysis that scrutinized a module configuration with high

potential to be industrialized because of including inexpensive materials and high-throughput deposition techniques.^[77] The module is manufactured by sputtering the Al and ITO electrodes, alongside screen printing the charge transport layers (NiO and ZnO) and the perovskite. CED for materials and electricity consumption was included in the study. This study is compared with other PV technologies in Section 4. This result highlights the differences in electricity demand between spin-coating and other deposition techniques such as sputtering and screen printing. Laboratory spin-coating is ranked as having the highest energy demand with the lowest throughput.^[57]

3.4. End-of-Life

For the sustainability of any PV technology, an adequate EOL of each module is mandatory. An increase in the recyclability of traditional PV technologies,^[78] as long as clear and efficient recycling methods for the emerging PV technologies, is essential for the expected deployment.

For PSCs, some important environmental, regulatory, and practical aspects that may arise from their dismantling are discussed.^[79] Authors of this study warn that if highly efficient and simple reuse and recycling processes are not established, we can eventually have large PVs waste volumes with a time span of 20 or 30 years, representing a 10% (60 million tons) of all e-waste produced globally.

Several methods of producing PSCs from disposed materials have already been proposed, for instance, a clean process to recover the lead from disposed car batteries.^[80] Lead iodide obtained by this method has similar material characteristics to that stemming from virgin lead. Also addressed to optimize the usage of lead, another study reports a method to recycle until 99.8% of the lead in PSCs, using deep eutectic solvents.^[81] Other studies present clean solutions to extend the lifetime of the perovskite absorbing layer in PSCs.^[82,83] The lifetime of PSCs can also be extended by dismantling them and recovering the principal components.^[84,85]

EOL environmental impacts are included in some LCA.^[48,51,66,67,75] One study included two EOL scenarios for PSCs^[51] to evaluate a recycling process where the principal layers are separated and reused. Both EOL scenarios are designed from recycling processes reported in the bibliography.^[86,87] The recycling process proposed significantly diminishes the toxicity impact. Therefore, proper recycling process for PSCs would notably improve their sustainability when produced at large scale.^[21]

3.5. Lead and Substitutes

Commercial deployment of PSCs poses a critical concern on lead's toxicity. Agents of diverse nature can liberate these toxic species into the environment, ranging from a simple rain falling on a damaged module to natural disasters or fire; probably, this is the most pernicious, because it can liberate lead into the atmosphere.^[88] In principle, although with some discrepancies,^[89] a proper encapsulation of PSCs is vital to protect the environment from the toxic substances contained in the cell.^[90-92]

Scientists from diverse research fields performed relevant studies to contribute with empirical results to shed light on

the toxicity of lead. A relevant number of these studies are gathered in a recent review, which covers PSCs apart from other PV technologies.^[93] The review finds relatively few studies addressing the toxicity generated by the potential leachates of PSCs, which considers insufficient. More data about the environmental consequences of leachates are then necessary to perform more accurate studies on the impact to the environment and health. In line with a conclusion of a previous study,^[90] authors propose considering the worst-case scenario in the event of spillage, when designing PV devices and regulatory policies.

Lead content in electronics is restricted to mass concentrations lower than 0.1% in homogeneous materials through the “Restriction of Hazardous Substances (RoHS) Directive.”^[94] As “homogeneous material,” this Directive considers “one material of uniform composition throughout or a material, consisting of a combination of materials, that cannot be disjointed or separated into different materials by mechanical actions, such as unscrewing, cutting, crushing, grinding, and abrasive processes.”

PV panels are currently exempt from the RoHS Directive according with the article 2.4 (i) “ This Directive does not apply to...(i) PV panels intended to be used in a system that is designed, assembled, and installed by professionals for permanent use at a defined location to produce energy from solar light for public, commercial, industrial, and residential applications.” However, this consideration does not specifically extend to solar cells embedded in consumer electronics or portable systems. In PSCs, this criterion could not be met, with the current perovskite compositions, whether perovskite is considered homogeneous material. The definition of what comprises the definition of “homogenous material” will be crucial for incorporation into consumer electronics sold in the European Union (EU).^[79] Due to the ambiguity of the directive, it is necessary to initiate a critical debate on the exploitation of lead halide perovskites, clearly stating “if” and “how” they can enter the consumer market.^[95]

Several researchers try to replace the lead in PSCs with alternative metallic cations. Among the candidate elements to substitute lead, we can find bismuth,^[96] germanium,^[97] cuprum,^[98] antimony,^[99] or tin,^[100] that is the most extensively studied. Sn-based PSCs are susceptible to degradation in ambient air, moisture, water, high temperature, and UV lights, which affect the overall performance of the cell. The major reason for poor stability and performance of tin-based perovskites includes the rapid oxidation of Sn²⁺ into its most stable Sn⁴⁺ state, uncontrolled crystallization of Sn perovskite, and mismatch of band alignment of Sn perovskite with either ETL or HTL.^[101] The Sn-based PSC performance and efficiency can be improved by controlling crystallization and mitigating the oxidation of Sn²⁺.^[101,102] In this line, the promising all-perovskite tandem solar cells retained 95% of their initial efficiency after 1000 h in a damp heat test (85 °C with 85% relative humidity) with encapsulation.^[102]

A summary of the progress of low-lead halide perovskites is exposed along with the necessary criteria to replace lead in perovskites.^[103] However, lead-free perovskites are a minority, as they still present low efficiencies,^[104] and some cases also have stability lower than Pb-based halide perovskites. Pb²⁺ presents excellent structural and electronic properties that make difficult waiving its usage.^[105] Therefore, a promising alternative to

reduce the toxicity of PSCs is partially replacing lead with these elements.^[105]

There are few LCA studies applied to PSCs focused explicitly on evaluating the toxicity of lead in perovskites.^[48,72,106,107] Nevertheless, an important number of the LCA studies about PSCs present a low relevance of lead as a conclusion.^[47,49,51,52,75,108] Two of the LCA studies focusing on the toxicity of lead in PSCs compare impacts of lead in PV devices based on perovskite with those of the electricity from the grid.^[106,107] In both studies, the production of electricity from perovskite PV devices results beneficial. The other two LCA studies of this kind compare a lead-based with a tin-based perovskite device. The first of both studies compares the environmental performance of a representative tin-based PSC^[48] with two lead-based PSCs.^[47,48] The second one includes a tin-based PSC to contrast its environmental impacts with that of four lead-based PSCs.^[72] GWP and CED are higher for tin-based PSC, see Table 1, largely due to low efficiency. Meanwhile, neither lead nor tin represents a major environmental impact because of the little amount of them used.

3.6. Solvents

Many industrial entities aim to commercialize PSCs because of the high performance, but also because this type of semiconductor can be processed from solution, a key feature that enables low cost and fast production. For mass production of halide perovskites, appropriate handling of solvents must be considered during the initial development phase to mitigate environmental impacts while optimizing performance.^[109]

Some polar aprotic solvents used for halide perovskite film fabrication may cause concern due to their toxicity.^[32,110–112] The most commonly used solvent for perovskite layer deposition, *N,N*-dimethylformamide (DMF), is included in the Candidate List of Substances of Very High Concern (SVHC), as part of Regulation by the European Chemical Agency (ECHA; Registration, Evaluation, Authorization and Restriction of Chemicals, REACH). The authorization process “*aims to ensure that SVHCs are progressively replaced by less dangerous substances or technologies where technically and economically feasible alternatives are available.*”^[113] Moreover, DMF, *N,N*-dimethylacetamide (DMAC), and *N*-methyl-2-pyrrolidone (NMP) are recognized as toxic to the human reproductive systems and other favorable solvents such as gamma-butyrolactone (GBL), although it is classified as a drug in some countries; 1,3-dimethyl-3,4,5,6-tetrahydroimidin-2(1H)-one (DMPU), 1,3-dimethylimidazolidin-2-one (DMI), and tetrahydrofuran (THF) are labeled “dangerous.” Dimethyl sulfoxide (DMSO) is not classified.

Recently, toxicity and other environmental impacts included for human health of eight polar aprotic solvents are used for perovskite deposition: DMF, DMSO, DMAC, NMP, DMI, GBL, THF, and DMPU were assessed with the LCA methodology.^[109] The impacts of solvent production, removal, and emissions were considered. Post-processing solvent removal was modeled for different scenarios: direct emission of solvent vapor to urban air, solvent condensation with subsequent incineration, and solvent recovery (with and without further purification).

Fate and exposure models were applied to determine missing human health characterization factors for several solvents and

to update existing values with available toxicity data published from the registration dossiers submitted to the ECHA under the REACH regulation.^[113] USEtox,^[114] the best available model for characterizing human health toxicity of chemicals, was modified with newly available toxicity data for detrimental impacts on human health beyond the binary “carcinogenic” classification.

DMF presents the highest impact due to air emission during solvent removed from the thin film. However, when the entire life cycle is considered, others including NMP have much greater impact due to high energy consumption during production. Energy consumption is directly related to the increased impact of GWP. Adopting an EOL treatment reduces the environmental impact, except for THF and incineration for DMSO. Direct solvent recovery using a condenser is an ideal option but must be tested to determine if the purity of the recovered solvent is sufficient. Some solvents are much more prone to deprotonation and production of side complexes that would require a more complex purification process.^[109]

Of the solvents assessed, DMSO has the lowest total impact being the solvent, which is both the most environmentally friendly and least deleterious to human health.^[109] Although some issues are acknowledged,^[115–121] DMSO can decompose exothermically near its atmospheric boiling point, and the presence of other chemicals such as acids, bases, several halides, sodium hydride, or perchloric acid can reduce the onset temperature; there is an odor issue with heavy use in water treatment plants, and DMSO can also carry contaminants/toxins on the skin or dissolved materials with it across the membranes.

Unique DMSO^[122] or combinations,^[123] such as DMSO/2-butoxyethanol^[124] or DMSO/PbS quantum dots,^[125] are achieving decent efficiencies on the small scale, and these can be motivation for upscaling. Beyond DMSO, other green solvents in the roll-to-roll process are worth mentioning, such as acetonitrile/methylamine.^[126]

3.7. Encapsulation

Encapsulation of PSCs can play an effective role in improving the stability. The encapsulation layer can act as a barrier layer by restricting the diffusion of oxygen and moisture, preventing the penetration of UV light, reducing the sensitivity to sharp thermal fluctuations and also inhibiting the irreversible escape of volatile decomposition products possibly forming from the perovskite materials, resulting in the protection of the cathode interface and the active layer from deterioration.^[127–129]

One of the most widespread approaches is based on glass–glass encapsulation, where the PSC device is sandwiched between two glass sheets with a polymer processed by thermoforming (e.g., ethylene vinyl acetate (EVA), Surlyn, polyolefins, and polyisobutylene [PIB]) or with UV-curable sealants. Also, edge sealants (mainly butyl rubber and PIB) or UV epoxy adhesives should be applied for preventing, or at least delay, moisture and oxygen ingress from the lateral perimeter, thus extending the lifetime of PSCs. Glass–glass with EVA encapsulation is commonly used for Si, CdTe, and CIGS PV modules.^[42,130] Some perovskite tandems^[54,66] as well as the industrial-based simulated PSC^[77] also use this encapsulation.

Some thermoforming solutions for encapsulating PSCs have passed, at least partially, the IEC standards; e.g., a solution with polyolefin passed the IEC 61646 standard for thin films with 200 thermal cycling (between -40 and 85 °C) and 1000 damp heat tests (85 °C–85% relative humidity);^[131] additionally, similar packaging passed the standard for a tin-lead iodide perovskite because of the use of an ITO–perovskite heterojunction as the hole contact instead of PEDOT:PSS.^[102] PIB used as blanket and edge seal passed partially the IEC 61215 standard for crystalline silicon PV modules with 200 thermal cycling (between -40 and 85 °C), but only 540 h in the damp heat tests (85 °C–85% relative humidity).^[132]

The relatively high temperature (>100 °C) required for the thermoforming is only suitable for highly temperature resistant perovskite materials, such as FAPbI₃ and triple cation perovskites. Hence, with the aim of reducing the impact of heat sealing on the performance of PSCs, in many research works, thermoforming sealant has been replaced by UV-curable adhesives despite the higher costs.^[127,132]

The toxicity of lead might require special and additional encapsulation to reduce Pb leakage.^[79,90] One satisfactory solution is the use of polymers that are able to self-heal when heated to temperatures higher than their glass transition temperatures and the use of a top glass cover.^[92]

Glass–glass encapsulation techniques are very affordable, relatively straightforward, and extremely efficient, because glass has the best water and oxygen blocking properties as transparent material. However, it is incompatible with flexible applications. Hence, alternative methods in which rigid glass coverslips are replaced by flexible films have recently been developed as flexible glass and transparent polymers.^[127,128]

Flexible glass was shown to exhibit ultralow moisture diffusion values. One option is a laser-assisted hermetic glass frit encapsulation to seal HTL-free PSCs without the use of sealants.^[133] However, it requires high processing temperature (120 °C). Therefore, more in-depth studies are currently underway with the aim of developing a similar hermetic encapsulation procedure with processing temperature lower than 85 °C to enable the use of highly efficient HTM-based devices (for devices with superior PCE) and reducing the high fabrication costs.^[127,133]

Various commercial transparent polymer-based barriers have been investigated as device encapsulants for PSCs, among these are polyethylene terephthalate (PET), polytetrafluoroethylene (PTFE), poly(methyl methacrylate) (PMMA), and polycarbonate (PC). One of the main limitations is again the temperature of the thermoforming. Consequently, encapsulation approaches relying on low-temperature lamination have been designed, e.g., two-step encapsulation process, thermoplastic polymeric films with integrated adhesives, or in situ-polymerized aliphatic polyurethane-based resins.^[127,134] Laboratory PSCs usually are not encapsulated, with some exceptions. PET^[49,65,67] is used as a barrier foil pre-laminated with a pressure sensitive acrylic adhesive.

Commercial polymeric films present low restriction to diffusion of oxygen and moisture. To improve these properties, polymer composite films (with graphene oxide or SiO₂ dispersed in a polymeric matrix) or solution-processed hydrophobic barrier coatings have recently gained noticeable attention.

Currently, the most technologically promising, but also challenging, option is thin-film encapsulation (TFE). It consists in the direct deposition of a single protective flexible ultrathin layer (e.g., Al₂O₃, SiO_x, SiN, etc.) or a multilayer stack, composed of multiple pairs of organic and inorganic layers, on top of perovskite devices using vacuum deposition techniques such as chemical or physical vapor deposition (CVD and PVD), plasma-enhanced CVD (PECVD), atomic layer deposition (ALD), and other vacuum coating techniques. Among these, ALD has been proved to be particularly suitable for PSCs.^[128,135,136] However, despite its effectiveness, ALD is an expensive approach, because it requires high-cost vacuum-based equipment and processes, as well as a detailed understanding of the interaction among the deposition process, the barrier layer material, and the device structure.

From the recycling perspective, polymeric materials can be classified as thermosets and thermoplastics. Thermoplastics (EVA, Surllyn, PIB, PET, PMMA, PTFE, PC, etc.) are made up of linear molecular chains, and this polymer softens on heating and hardens when cooled. Thermoset polymers (epoxy, polyester, thermosetting polyurethane, etc.) differ from thermoplastics, as they do not require heating to form the plastic material but utilize a curing process where a chemical reaction takes place, forming cross links. All polymers can be incinerated with energy recovery. However, only thermoplastics polymers can be mechanically recycled, because they can be re-melted and reprocessed into end products. The mechanical recycling does not involve the alteration of the polymer during the process. Mechanical recycling is impaired if incompatible polymers are mixed. This mismatch can happen, e.g., with a two-step encapsulation process or thermoplastic polymeric films with integrated adhesives. Chemical recycling is other option, in which polymers are chemically converted to monomers or partially depolymerized to oligomers through a chemical reaction, although it is not fully developed.^[137]

While the encapsulant material requires environmental stability to protect the cell from years of environmental influence, it should ideally be easily removable or thermally decomposable without the formation of toxic products to properly remove the active layer. If solvents are required to remove the encapsulant, the solubility of the lead halide absorber, ETL and HTM, and the resulting solvent contamination has to be considered.^[79,129] Experiences from recycling crystalline Si, CIGS, and CdTe PV modules conclude that eliminating the encapsulant is the most difficult process. Therefore, some recycling strategies for crystalline silicon PV modules are focused on recovering only the back-contact metals and the substrate glass, after the pre-disassembly process.^[138,139] However, this is not a valid recycling strategy for PSCs, because lead has to be removed due to its toxicity.

4. Environmental Comparison with Current PV Technologies

Environmental impacts of laboratory-based PSCs, included in Table 1 and 2, are compared in the literature with crystalline-Si,^[49,50,52,64,65,72] CdTe,^[49,50,52,64,65,72] CIGS,^[52,64] DSSC,^[37,50,64,72] OPV,^[37,49,64] amorphous-Si,^[50,52,64,72] GaAs,^[64] and quantum dot PV.^[37]

In addition, PSCs were compared with the rest of PV technologies in some reviews.^[25,36,38] A couple of works were focused on comparing the toxicity risk of lead from PSC with toxicity of the electricity grid, mostly based on fossil fuels.^[106,107] Toxicity potential of electricity from lead PSC over a 20 year operational lifetime would be ≈20 times lower than that of grid electricity.^[106] In addition to toxicity, the environmental benefits from avoided grid electricity during a lifetime of 25 years exceed environmental burdens from using perovskite-based cells for climate change.^[107]

Some studies on EPBT underline the potential that PSC is an environmentally sustainable electricity generation technology, because their EPBT is shorter compared with the rest of the existing PV technologies.^[49,52,64,65] Although for other studies, PSCs have EPBT higher than 10 years.^[47,50]

The environmental impacts of PSCs, for 1 kWh as functional unit, are higher than the commercial PV technologies. The reason is the shorter lifetime assumed for the PSCs, 1–5 years against 25–30 years for the rest of technologies, making them environmentally uncompetitive if the current lifetimes are not improved.^[25,36,38,49,50,52,64,65,72]

In this article, the functional unit kW_p is selected to avoid the consideration of the lifetime. Results for the GWP and CED of the PSCs reviewed are in Table 1 and 2 for single-junction and tandem PSCs, respectively. These impacts are compared with those of selected mono-Si,^[130,140] multi-Si,^[130,140,141] amorphous-Si,^[130] CdTe,^[130] and CIGS,^[130,142] whose values are in Table 3 and Figure 1 and 2, respectively. The assumptions for selecting these solar cells are that the studies were relatively recent (after 2011), and that the impacts of the inverter, frame, mounting, cable, and connectors were not included. The impacts of these components are the same for rigid PV panels, independently of the technology. Avoiding the common elements is a typical strategy of the LCA methodology to be focused on the differences.

EPBT is obtained with Equation (2) using the CED of Table 1–3, and it is plotted in Figure 3. Note that not all PSCs have enough

Table 3. GWP and CED for 1 kW_p of the selected PV technologies.

Ref.	Technology	Electricity mix	PCE [%]	GWP [Kg CO _{2eq} kW _p ⁻¹]	CED [M] kW _p ⁻¹
[140]	Mono-Si	Germany	15.8	986	–
[130]	Mono-Si	Union for the Coordination of the Transmission of Electricity (UCTE)	14.8	1133	24 170
[130]	Mono-Si	China	14.8	2730	29 700
[140]	Multi-Si	Germany	15.2	787	–
[141]	Multi-Si	China	12.7	2060	24 100
[130]	Multi-Si	UCTE	14.1	667	13 710
[130]	Multi-Si	China	14.1	1431	16 790
[130]	a-Si	UCTE	7.0	827	15 100
[130]	a-Si	China	7.0	1170	15 000
[130]	CdTe	UCTE	11.9	358	6320
[130]	CdTe	China	11.9	518	6260
[130]	CIGS	UCTE	12	523	10 400
[130]	CIGS	China	12	766	10 300
[142]	CIGS	Netherlands	15	230	–

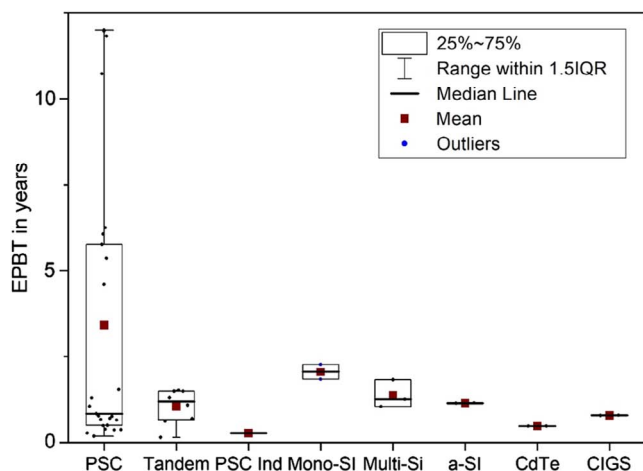


Figure 3. EPBT. Data are in Table 1 for PSC and PSC-ind, in Table 2 for tandem PSC, and in Table 3 for the rest of PV technologies.

stability to reach the estimated EPBT. Improving the long-term stability of PSCs is critical to the deployment of this technology. Oxygen and moisture chemical degradation remains a more challenging issue, and strategies to keep devices dry and oxygen free are being developed. A first indirect approach is the exploration of different charge transport layers, which have been intensely researched with the focus on improvement of photocurrent and, therefore, PCE. Stability has also benefited from this approach, because many interfacial layers have proved to be critical to avoid the chemical degradation of devices.^[19] A second approach is the development of encapsulation materials and techniques, which avoid the ingress of oxygen, moisture, and other contaminants into the device, as described in Section 3.7. Currently, the lifetimes of PSCs are difficult to compare due to the different lifetime measurements and analysis procedures. Stability protocols, such as ISOS, should be used to report stability in PSC research, and this will allow fair comparison between materials and device architectures.^[143,144] Despite this, it is worth noting that some PSCs have passed the IEC 61646 or IEC 61215 standards to accelerate different degradation pathways, specifically 1000 h damp-heat test (85 °C–85% relative humidity).^[71,102,131] So far, the longest lifetime reported for PSCs is over 9000–10 000 h, under certain stress conditions.^[145,146]

Results for the median PCE, EPBT, GWP, and CED are in Table 4. Median values EPBT, GWP, and CED for the single-junction and tandem PSCs at laboratory scale are lower than for silicon PV and higher than CdTe and CIGS. Noticeable is the highest PCE of tandem PSC. Due to the large dispersion observed in Figure 1–3, a hierarchical analysis cluster with the furthest neighbor method is performed with the single junction at laboratory scale considering two clusters; see Figure 4. In this figure, the cluster labeled high includes the PSCs analyzed by Krebs, Urbina, and co-workers^[47,48] and Zhang et al.^[50,72] The rest of the PSCs are included in the cluster labeled low. The criteria for these two cluster in Figure 4 are that the median EPBT, GWP, and CED for the cluster labeled high are one order of magnitude higher than for the cluster labeled low and the highest values of all PV technologies analyzed. The low PSCs with data

Table 4. Median PCE, EPBT, GWP, and CED of the PV technologies.

	PCE [%]	EPBT [years]	GWP [kg CO _{2eq} kW _p ⁻¹]	CED [MJ kW _p ⁻¹]
PSC lab low	14.2	0.66	448	8681
PSC lab high ^[45,46,51,53]	10.9	6.25	3099	82 018
Tandem	22.4	1.19	1250	15 667
PSC Ind	16.0	0.28	n/a	3613
Mono-Si	14.8	2.06	1133	26 935
Multi-Si	14.1	1.26	1109	16 790
a-Si	7.0	1.15	999	15 050
CdTe	11.9	0.48	438	6290
CIGS	11.7	0.79	523	10 350

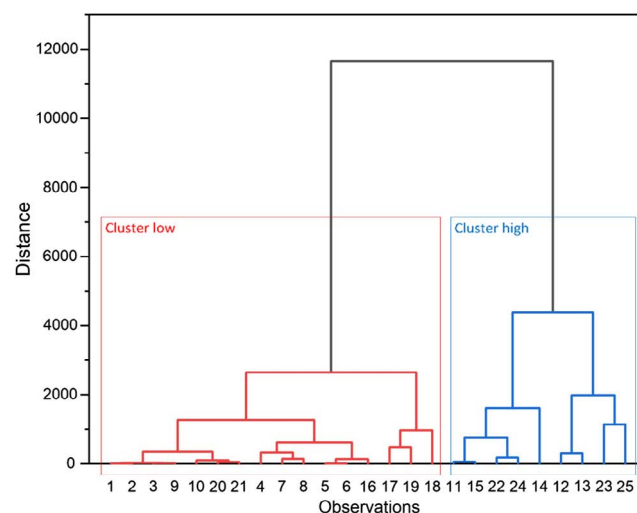


Figure 4. Dendrogram with Euclidian distances of the hierarchical cluster analysis using the furthest neighbor method with variables GWP, CED, and PCE of the observations included in Table 1.

at laboratory scale are in a similar range to CdTe and CIGS for EPBT (0.66 years), GWP (448 kg CO_{2eq} kW_p⁻¹), and CED (8681 MJ kW_p⁻¹). Specifically, the GWP for PSC-low is almost the same as for the CdTe and lower than CIGS. The CED and EPBT for the PSC-low are between the values of CdTe and CIGS. Note that the estimated EPBT for the PSC-low is lower than the longest lifetime reported for PSCs.^[145,146]

Results for the only PSC simulated at industrial scale are the lowest of the PV technologies, see Table 4, at least seven times lower than for commercial silicon PV. The EPBT is 0.28 years, and the CED is 3613 MJ kW_p⁻¹. These favorable results are achieved in one study at industrial scale described in Section 3.3, in which energy data are obtained from technology suppliers and material usage efficiency for perovskite deposition is 80%,^[77] whereas several authors have considered that materials and energy efficiency at laboratory scale are about 5–20%.^[47,52,54,67,72] An advance on this rating is the ideality coefficient (IC).^[53] The IC quantifies how close a given fabrication procedure from the ideal process is. Its value is the result of dividing the environmental impact of the ideal process by the impact of the process to

compare, and it is expressed in percentage. Note that a technology requires an IC for each impact category as high as possible to reduce as much as possible the environmental impacts. This coefficient was applied to a pre-industrial module and the laboratory-based PSCs.^[53] The anode and substrate layers of the pre-industrial process reach ICs in the range of 56–90%; meantime, these layers at laboratory scale do not surpass 3%. Deposition of perovskite is the least optimized process for the both PSCs with ICs lower than 0.1%.

To clarify the differences in the environmental impacts, electricity consumption, and materials of mono-Si, CdTe, and CIGS, obtained from IEA PVPS LCI,^[42] they are compared with the inputs obtained from PSC-ind work.^[77] Electricity consumption of mono-Si is very high 298 kWh m⁻², whereas the electricity consumption of PSC-ind is the lowest, 24.8 kWh m⁻², also lower than the electric consumption for CIGS (44.7 kWh m⁻²) and quite similar to CdTe (27.9 kWh m⁻²). Electricity consumption is responsible for 93% of CED for mono-Si and 41% for CdTe, CIGS, and PSC. In addition to the lower electricity consumption, the environmental impacts of the materials are also lower. However, the PSC-ind LCI does not include emissions, waste treatments, and some auxiliaries, which could slightly increase CED and EPBT.

These results are very promising for the implantation of halide PSCs as a promising commercial PV technology, especially for applications with short lifetimes, or in the case of PV panels, if the PSC cell is replaced seven times with a life span of about 4 years. Specifically, the PSC will be environmentally competitive (for CED) with mono-Si PV (30 years of lifetime) if PSC stability is higher than 6841 h of illumination (1700 h of illumination year⁻¹). However, more LCA studies of PSCs on pre-industrial scale and industrial simulations are needed as just an industrial simulation and with only PE data is not enough to draw conclusions from PSCs with respect to other PV technologies.

5. From Environmental LCA to Sustainable LCA

Sustainability integrate three concepts: environment, economy, and society. For the environmental part, there is already an internationally standardized tool, the LCA, which review for PSCs is presented in Section 3. Life cycle costing (LCC) is the logical counterpart of LCA for the economic assessment. LCC surpasses the purely economic cost calculation by considering the use and EOL phases and hidden costs. Some studies about production costs for PSCs are reviewed in Section 5.1, because the remaining costs to complete the LCC have not been found. Geopolitical supply risk or the economic resource scarcity potential of resources for PSCs production, as integrated within the LCSA framework,^[147] is described briefly in Section 5.2. Finally, some aspects about social and aesthetic impacts are detailed.

5.1. Cost Analyses

Several technoeconomic analyses are performed on simulated PSCs manufacturing processes,^[77,148–152] see **Table 5**. Some authors have pointed out the lower cost of Cai et al.,^[148] 0.22–0.25 \$ W_p⁻¹, is due to the fact of this analysis neglected some important material costs in the module fabrication.^[77,150]

Table 5. Production costs of PSCs obtained in different studies in comparison with the price index of standard crystalline Si.

Ref.	Cost [\$ m ⁻²]	Factory [MW]	Substrate	PCE	Cost [\$ W _p ⁻¹]
[77]	32	100	Glass	16	0.41
[148]	40	100	Glass	12	0.25
[148]	51	100	Glass	19	0.22
[149]	96	100	Glass		0.57
[150]	35	100	Flexible	10	1.10
[152]	231	<3.6	Flexible	12	1.93
[152]	86	3.6–100	Flexible	12	0.72
[151]	–	3.6	Flexible	18	3.30
[151]	–	1000	Flexible	18	0.53
[151]	63.3	100	Tandem	26	0.85
Crystalline Si standard, price index ^[153]					0.25

Considering annual productions higher than 100 MW, production costs are under 1 \$ W_p⁻¹ with the efficiencies of 12% or higher. This cost is higher than standard crystalline Si with an average price of 0.25 \$ W_p⁻¹.^[153] Production costs can be reduced if lower cost materials, specifically barrier foils and TCO-coated plastics, can be found. However, this cost could be competitive selling into niche markets for \$1 W_p⁻¹ or greater, representative of Internet of Things, building-integrated, and vehicle-integrated markets.^[151]

In addition to the previous results, the levelized cost of energy (LCOE) is estimated for modules with 16% PCE and a 30 year lifetime in €6.3 kWh^[77] and €9 kWh.^[149] Considering these futuristic PCE and lifetime, the cost of manufacture must be reduced to be competitive with the commercial solar PV. In this sense, the global weighted average LCOE for solar PV can be expected to continue its downward trend to reach by €4 kWh by 2030.^[154]

5.2. Supply Risk

PSCs will be feasible if the supply is guaranteed with new mineral resources and a lack of supply disruptions. Disruptions to supply may occur due to governmental interventions, market imbalances, or physical impediments within the supply chain.^[155] Non-fuel minerals, that are at greatest risk of a disruption in supply, and whose supply disruption would have the most consequences, may be considered critical. Criticality assessments are performed for silicon-based CdTe and CIGS^[156–158] but not yet for PSCs. PV materials, in relative order of most to least critical in Europe, are: Pt, Ge, Te, In, As, Si, Sn, Se, Mo, Ag, Cd, Zn, Ga, Au, Al, Fe, and Cu.^[156]

According with the list of critical materials of the EU,^[159] lead has a supply risk value of 0.1, lower than Cu, the last one of the PV materials analyzed, with a value of 0.2. To be used as a reference, silicon metal has a value of 1, and Pt has a value of 2.1.

Despite the fact that there are no specific studies of the criticality of perovskites, it is possible to indicate these possible supply risks: precious metals for the cathode and TCO^[160] for the

potential use of cesium.^[161] The two first ones are common to thin films, and there are alternatives with less supply risk.

Cesium is one of the materials with the least coverage in the criticality assessments. In 2018, Cs was, for the first time ever, included in the draft list as a critical mineral for economic and national security interests by the US government,^[162] and recently, data about the supply risk are provided.^[161]

Supply risk determined with the EU methodology^[163] has a value of 1.5 and can be considered potentially critical. If Cs resources are not increased, either by including as yet unaccounted resources of several countries or by finding new deposits, and assuming as constant the current extraction rate, these resources would be completely depleted in the year 2056, whereas the reserves (the economically mineable part of a measured mineral resource) will be depleted in the year 2025.^[161]

5.3. Social and Aesthetic Impacts

The main object of this research is the assessment of the environmental impact of PSC technology to compare with results from other technologies; however, other impacts exist that should not be underestimated. For instance, industry involved in sustainable development strives to differentiate itself by showing environmental responsibility, but increasingly, they also pay attention on social and economic responsibility. Indeed, progressively, we find companies that, because they show total indifference to social impacts, are under scrutiny and facing greater social pressure to display and disseminate this information.^[164]

In this sense, the raw material supply chains analysis becomes crucial to assess the social and economic impact when a product is manufactured. Social life cycle analysis (S-LCA) has emerged in the literature as a suitable method for evaluating social risks in commodities.^[164–166] To develop this method, raw material country sectors are identified, and specific social indicators are selected. As a result, direct and indirect impacts are detected, and important risks originating in the supply chain are identified.^[165] It should be highlighted that knowledge of the place of origin of the raw materials as well as the place where production takes place are of vital importance to carry out the S-LCA.^[167,168] Although there is a lack of social data and the analysis can be difficult to carry out, the results obtained with this method make it possible to establish priorities for political actions in the most affected areas.

To complete the comparative study of the impact caused by different PV technology systems, it would be advisable to carry out an S-LCA, which would consider the origin of the raw materials used as well as the place where they are produced or can be produced. The results obtained with this method would make it possible not only to establish priorities at political level, but also to identify those technologies most favorable to socially sustainable development.

On the other hand, the threat of climate change has led to a clear commitment to the use of renewable energies in recent years, and an example of this is the energy policy pursued in the EU over the last decade.^[169–172] The necessary energy transition to be carried out implies the integration of renewable energies in the city, and in this process, solar technologies have a very important role. Integrating solar technologies into the city means

altering the urban landscape, and this should also be considered as a social impact. Precisely, the aesthetic impact caused by the integration of renewable energies in the city is a current topic of interest in the scientific literature that has increased its relevance in recent years.

Aesthetic perception depends on several objective factors. Indeed, according to a study carried out in 2018, the objective factors most influencing the aesthetic perception of solar systems are visibility and the degree of integration.^[173] Therefore, certain characteristics of the different solar technologies, such as the possibility of placing them in different parts of a building or their ease of aesthetic integration into building elements of the envelope, could give them advantages over other technologies in the long term.

Several studies have shown favorable results assessing the aesthetic perception of integrated PV systems.^[174–176] However, it must be borne in mind that BIPV systems, those that have been integrated into the construction elements of buildings, are more widely accepted by society than building-applied PV systems, those that are applied with a substructure on the envelope.^[177] Hence, it is foreseeable that technologies, which have a greater capacity to be part of the building envelope construction system, or to be integrate harmoniously with the aesthetics of the building of which they are part, will, in the future, have a greater acceptance by society. For example, technologies such as those based on thin films or halide perovskites can be bonded to other materials as a second skin, thus allowing better integration options than other more rigid and heavy panel-shaped technologies such as crystalline silicon. Also, technologies that can be manufactured in different colors offer a wider range of possibilities for technical designers to harmonize with the architectural whole.

In recent years, we find, in the literature, a growing concern for aesthetic aspects, such as color or transparency, in new emerging thin film technologies while preserving a significant level of efficiency.^[178–180] Controlling the inherent properties of the photoactive layer has been an active area of research as part of the effort to directly tune the colors of solar cells. However, when using this method, the efficiency of a solar cell is greatly influenced by the color of its photoactive layer.^[179] Fortunately, new methods based on placing colored filters with a narrow bandwidth on the light-illuminated side of a solar cell next to it seem to be a good alternative to avoid this loss of efficiency without the need for the cells to be transparent.^[179]

Colored and semitransparent OPV cells have proved their applicability in buildings with the maximum transmission efficiencies exceeding 25% and constant performance.^[181] Regarding DSSCs, we found studies advocating their high aesthetic potential, especially in transparent and colored applications. Furthermore, this technology has a promising future in indoor applications since, unlike most inorganic solar cells, DSSCs can still provide a useful amount of energy in low light conditions.^[182]

In the specific case of PSC, we know that they cannot yet be considered technically or industrially mature; however, very interesting literature on its applicability to BIPV systems was found.^[183,184] These studies highlighted a wide variety of possibilities, such as electrochromic and thermochromic cells, colorful solar cells, flexible solar cells, or semitransparent solar cells. In fact, the properties of this type of solar cell have shown great

promise for specific innovative applications in windows, which would make it possible to control factors such as the absorption capacity of the glass, or the degree of transparency.^[185,186] We also found interesting articles that confer outstanding capabilities to PSCs, with great versatility in design and color, while maintaining an energy conversion efficiency of 18–18.9%.^[180,181]

Overall, although some of these new emerging thin-film technologies are still in their early stages, their lightness, flexibility, and ease of integration give them very advantageous qualities for aesthetic impact assessment in comparison with other PV technologies.

6. Conclusion

A revision of LCAs of laboratory-based PSCs, tandems with perovskite, and industrial-based simulated PSCs is presented in this article, and put in context with respect to other PV technologies. It is highlighted that the main drawback arising from the results of the environmental impacts of PSCs at an early stage of development stems from the unrealistic energy consumption of the laboratory-scale production processes.

Furthermore, controversial topics as PSCs' EOL, lead content, and perovskite deposition solvents toxicity are herein discussed. A proper recycling process is considered fundamental for sustainable PSCs deployment, as it is found to diminish the toxicity impacts. From the LCA performed thus far, it is concluded that they do not represent a serious toxic risk. Among all solvent used to deposit PSCs, DMSO presents the lowest environmental impact, despite presenting some drawbacks when handling it.

A harmonized comparison for 1 kW_p of PSCs and other commercialized PV technologies is included. Despite the excessive energy consumption and the great uncertainty of the laboratory-based single-junction and tandem PSCs, their environmental impacts are lower than for silicon PV. A cluster analysis of single-junction PSCs allows us to differentiate two clusters; in one of them, the most numerous, GWP, CED, and EPBT are in a similar range to CdTe and CIGS PV technologies.

The comparison is much more advantageous for the industrial-based simulated PSC, in which energy data are obtained from technology suppliers and high material usage efficiency for perovskite deposition. Industrial-based presents lower CED and EPBT than silicon-based PV, CdTe, and CIGS, e.g., seven times lower environmental footprint respect commercial Si PV.

A final synopsis about cost of PSCs, criticality of raw materials, and social impacts of PSCs is provided to facilitate future LCSA. Technoeconomic analyses of PSCs reveal that current costs make PSCs competitive for some niche markets, such as the Internet of Things, BIPV, and vehicle-integrated. The overall costs of PSCs need to be reduced, however, to be competitive with crystalline Si. Cost reduction should arrive from a decrease mainly of barrier foils and transparent conductive oxide-coated plastics, better than glass. PSCs coincide with the supply risk for cathode and TCO with other thin film technologies and, however, present less supply risk than other PV technologies. Only Cs can present a specific risk for perovskites with the current information that we have about Cs reserves, although it is not an indispensable element. In addition, their lightness, flexibility, and ease of integration give them very advantageous qualities for social

acceptance. This work highlights the potential of PSCs to reduce environmental impacts and reduce EPBT, despite more analysis for an industrial environment is required. Further increase in long term stability of this technology, in the line of the last results reported in the literature, will contribute to make this technology even more appealing for commercialization.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

R.V. and I.M.S. designed the study. R.V. and J.A.A.B. wrote the first version of manuscript. N.S.P. wrote Section 5.3. I.M.S. and N.S.P. assisted in the work analysis. All authors contributed to the final version of the manuscript.

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