Life Cycle Assessment (LCA) of Perovskite PV Cells Projected from Lab to Fab

Ilke Celik^a (Ilke.Celik@rockets.utoledo.edu) , Zhaoning Song^b
(Zhaoning.Song@rockets.utoledo.edu), Alexander J. Cimaroli^b
(Alexander.Cimaroli@rockets.utoledo.edu), Yanfa Yan^b (Yanfa.Yan@utoledo.edu), Michael J.
Heben^b (Michael.Heben@utoledo.edu) and Defne Apul^{*a} (defne.apul@utoledo.edu)

^a 2801 W. Bancroft, Department of Civil Engineering, University of Toledo, Toledo, OH, 43606

^b 2801 W. Bancroft, Dept. of Physics and Astronomy, University of Toledo, Toledo, OH, 43606

*Corresponding author tel: (419)-530-8132; email: defne.apul@utoledo.edu

Abstract

Perovskite photovoltaic cells (PVs) have attracted significant worldwide attention in the past few years. Although the stability of the power conversion is a concern, there is great potential for perovskites to enter the global PV market. To determine the future potential of perovskites, we performed a cradle-to-gate environmental life cycle (LCA) for two different perovskite device structures suitable for low cost manufacturing. Rather than examining current laboratory deposition processes like dipping and spinning, we considered spray and co-evaporation methods that are more amenable to manufacturing. A structure with an inorganic hole transport layer (HTL) was developed for both solution and vacuum based processes, and an HTL-free structure with printed with back contact was modeled for solution based deposition. The environmental impact of conventional Si PV technology was used as a reference point. The environmental impacts from manufacturing of perovskite solar cells were lower than that of mono-Si. However, environmental impacts from unit electricity generated were higher than all commercial PV technology mainly because of the shorter lifetime of perovskite solar cell. The HTL-free perovskite generally had the lowest environmental impacts among the three structures studied. Solution based methods used in perovskite deposition were observed to decrease the overall electricity consumption. Organic materials used for preparing the precursors for perovskite deposition were found to cause a high marine eutrophication impact. Surprisingly, the toxicity impacts of the lead used in the formation of the absorber layer were found to be negligible. Energy payback times were estimated as 1.0 to 1.5 years.

Key Words: Perovskite LCA, ecotoxicity, scalable manufacturing

1. Introduction

Increasing interest in atmospheric carbon reduction and a fast growing demand for renewable energy promise a great future for large-scale use of solar cells. While photovoltaics (PV) currently contribute ~1 % of the global electricity generation, this percentage is expected to grow remarkably reaching 18 % in the US and 16 % globally by 2050 [1, 2]. The past decade has already witnessed the rapid growth of the global PV market, and total global capacity was close to 180 gigawatts (GW) in 2014 mainly due to the substantial reduction in the manufacturing costs of market-dominant crystalline Si solar cells. Between 2010 and 2014 the total installed cost of utility-scale PV systems fell by 29 - 65% [3]. While the levelized cost of electricity (LCOE) can vary greatly as a function of discount rate, conversion efficiency, system degradation, solar insolation, and operation and maintenance conditions [4], most of the utility scale projects in 2014 delivered electricity at a reasonable LCOE of \$ 8 ¢/kWh in 2014 [3]. Compared to first generation wafer based (crystalline mono and poly Si) solar cells, second generation thin-film solar cells (e.g. CdTe and Cu(In,Ga)(Se,S)₂) have lower conversion efficiency but their lower manufacturing and material costs have made them quite competitive in recent years, reaching a current global market share of ~10% in 2015, which was double their share in 2007 [5]. The manufacturing cost of solar PV energy will be further reduced if promising alternatives being developed in laboratories can be commercialized.

One emerging promising PV technology is based on methylammonium lead halide perovskites. These organic-inorganic hybrid materials have progressed rapidly over the past few years and become one of the most attractive areas for PV research [6]. A large number of studies emerged with a variety of different material preparation methods and diverse device architectures. To date, the best performing perovskite device has achieved 21% efficiency [7], which is competitive with the established PV technologies [8]. This fast learning curve for perovskites is extraordinary considering more than three decades were required to achieve similar efficiency advances with either Si, CdTe, or Cu(In,Ga)(Se,S)₂.

Perovskites, named after the Russian mineralogist Lev Perovski, are materials that have the same crystal structure as calcium titanium oxide (CaTiO₃). In the field of PV research, the term perovskites mainly refers to a class of organic-inorganic hybrid methylammonium lead halides (CH₃NH₃PbX₃, where X= I, Br, or Cl). These perovskites have a tunable direct bandgap ranging from 1.5 to 2.2 eV [9] that covers the whole visible solar spectrum and high optical absorption. High-efficiency devices can be fabricated with very thin layers, which promises reduced materials costs. For example, ~300 nm is needed to form a high efficiency perovskite cell, while Si and thin film (e.g. CdTe or Cu(In,Ga)(Se,S)₂) absorber layers are typically ~300 µm and ~2 µm thick, respectively [10]. These characteristics along with other important properties such as ultrafast charge generation [11], high carrier mobilities for both electrons and holes [12], and relatively long carrier lifetimes [13, 14] lead to excellent performance in PV devices.

Furthermore, perovskites exhibit a large degree of tolerance in synthesis [15] and may be easier to manufacture into high-efficiency devices than current silicon and thin film cells. Despite this promise, there remain significant technical challenges that must be overcome before perovskites can be fabricated in modules at a large scale. For example, perovskite solar cells display hysteresis in their current-voltage characteristics [16], and more importantly their stability issues have not been resolved. Perovskite solar cells degrade due to oxygen, light, moisture, and high temperatures [16, 17].

Most of the prior work on perovskite solar cells have focused on their technical properties and performance. Yet, if perovskite cells were to enter the commercial market and ultimately play an important role in supplying electricity globally, it will be important to understand their environmental impacts as well. To date, four ex-ante life cycle assessment (LCA) studies have been conducted on perovskite PV cells to shed light on their potential environmental impacts [18-21]. One of these studies focused on liquid perovskite solar cells [19]. The other three focused on solid perovskite solar cells but modeled materials (e.g. gold, silver, spiro-MeOTAD) and methods (e.g. spin coating) [18-21] that may be difficult to scale up from lab to commercial fabrication. In this paper, we evaluate the environmental impacts of perovskite PV technology at scale by modeling device architectures that we believe represent the expected low-cost production conditions. As described in the methods section, we went through a rigorous elimination process and determined and modeled the materials (FTO-coated glass, SnO₂, perovskite, CuSCN, and MoO_x/Al) and manufacturing methods (spray deposition and co-evaporation under vacuum) that we believe are more likely to be used during commercialization. In addition, seeing as the hole transport layer (HTL) was found to have significant environmental impacts (*vide infra*), a new promising HTL-free device was modeled. The global PV market share is currently dominated by mono-Si (36 %) and poly-Si (56 %) [22]. Mono-Si has higher environmental impact than other commercial PVs (poly-Si, a-Si, CdTe and CIGS) [19]. To more clearly interpret the results, we present the results after normalizing the impacts to mono-Si. We discuss the relative importance of different PV components and electricity use in manufacturing the cells. A sensitivity and uncertainty analysis was also conducted.

2. Methodology

2.1. Goal and Scope

The goal of this study was to evaluate the life cycle environmental impacts from perovskite solar cell designs that may be commercially fabricated. Life cycle assessment is a technique for assessing the environmental impacts of a product or service throughout its life cycle from raw material acquisition, to production, through use and disposal/recycling phases [23]. Since perovskite cells are not currently commercially fabricated, we conducted an 'ex-ante' LCA

study. We anticipated and modeled the most scalable methods and materials for a commercial PV market. Evaluation of ex-ante LCA results can identify potential opportunities for improvement, inform research directions and guide the sustainable design of perovskite solar cells.

The system boundary of the study was cradle to gate. Since the perovskite technology is still far from scalable manufacturing, the operation and end of life phases include much greater uncertainty than the production phase and were not included in the LCA model for this reason.

LCA guidelines suggest that the environmental profiles be interpreted based on the function of the product[23]. The function of PV modules is to generate electricity and our selected functional unit was 1 kWh of electricity generated for the entire life time of the PV. All results were presented for this functional unit. In addition, following the approach of recent PV LCA studies we present the impacts from manufacturing 1m² of module [19, 21, 24].

2.2. Selection of the Perovskite PV Cell Device Architecture

The conventional structure of perovskite PV cells typically consists of a fluorine-doped tin oxide (FTO) coated glass substrate, a TiO₂ electron transport layer (ETL), a perovskite absorber, a Spiro-MeOTAD hole transport layer (HTL), and a gold back contact. Each layer of the device can be made from these or other alternative materials (Table 1). The performance of perovskite solar cells is mainly determined by the quality of the perovskite absorber. Changing other layers in the perovskite solar cell into alternative materials with similar functionality does not significantly affect the physics of the device, and consequently, results in similar device performance [6]. In this study, we selected the materials that are most likely to be used in low-cost commercial scale fabrication.

The first layer, also known as the top contact layer, is commercially available TCO coated glass; coated either with fluorine doped tin oxide (FTO) or indium tin oxide (ITO). We modeled FTO because it is lower in cost and is already used more frequently in the solar industry for this reason[25]. Moreover, realizing the challenges with the global availability of indium, U.S. Department of Energy's Solar Energy Technologies Program aims to reduce the amount of indium used in thin film technology; indium has been designated a 'critical metal' with high supply risk and high importance for the clean energy industry [26].

138 *Table 1 Alternatives for device layers. The materials selected and modeled in this study are in bold. In the structure*
 139 *without the HTL, CuSCN was not used and MoO_x/Al was replaced with C-Paste.*

Layer	Function	Alternative Chemicals
Top Contact Layer	Collects electrons	FTO ITO
Electron Transport Layer	Conducts electrons	TiO ₂ ZnO Al ₂ O ₃ SnO₂
Absorber Layer	Harvests light	CH ₃ NH ₃ PbI _x Br _{3-x} CH ₃ NH ₃ PbI _x Cl _{3-x} CH₃NH₃PbI₃
Hole Transfer Layer	Conducts holes	Spiro-MeOTAD P3HT PTAA CuSCN CuI NiO
Back Contact Layer	Collects holes	Au MoO_x/Al Ag C-Paste

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 141 For the electron transport layer, of the four available materials, SnO₂ was selected as offering
 142 the most promise for commercial production due to the low-temperature process used for
 143 deposition and energy band match with the perovskite. TiO₂ has been commonly used in
 144 perovskite solar cells, however, it typically requires temperatures as high as 450 °C to 500 °C
 145 which may be incompatible with low-cost solar technology [27]. Similarly, Al₂O₃ and ZnO were
 146 found to be not suitable for low cost production due to the highly energy intensive methods
 147 that are used for their manufacturing [28]. Among all the alternative materials, SnO₂ provides a
 148 low-temperature and solution-processable approach for high efficiency (>18%) perovskite
 149 device, which is compatible with roll-to-roll manufacturing of low-cost perovskite solar cells [29,
 150 30].

151 For the absorber layer, we considered that compositional engineering of CH₃NH₃PbI₃ is made
 152 possible by incorporating/substituting other species such as chlorine, bromine, and
 153 formamidinium (FA) to achieve improved device performance and stability [9, 31-33]. However,
 154 since the amounts of additive chemicals can vary from lab to lab and the necessity of such
 155 compositional modification is yet to be proven, we chose pure CH₃NH₃PbI₃ for our study. Note
 156 that devices based on pure CH₃NH₃PbI₃ alone have shown efficiencies of 19.7%, which is very

close to the current record value. To provide scalable manufacturing by an easy processing method, pure $\text{CH}_3\text{NH}_3\text{PbI}_3$ was selected in our study.

In the HTL, dozens of organic small molecules, conducting polymers, organometallics, and inorganic compounds have been used in perovskite solar cells. Among them, the most commonly used organic HTL materials include 2,2',7,7'-tetrakis (*N,N*-di-*p*-methoxyphenylamine) 9,9'-spirobifluorene (Spiro-MeOTAD) [34, 35], poly(3-hexylthiophene-2,5-diyl) (P3HT), [36, 37] and poly(bis(4-phenyl) (2,4,6-trimethylphenyl)amine) (PTAA), [32, 38]. However, these materials are very expensive [39] and their instability may represent a potential hurdle to the commercialization of this type of solar cell [40]. To solve this problem, alternative inorganic HTLs, including CuI, CuSCN [41, 42], and NiO [43, 44] have been developed. Use of inorganic HTL material can lead to high performance perovskite devices with high hole mobility, better stability, ease of synthesis, and thus low production cost [45, 46]. For our study, we selected CuSCN, a copper-based semiconductor, which shows the highest energy conversion efficiency (15.6%) so far for an inorganic HTL [42].

To complete the PV device, a patterned metal contact is deposited. Gold, silver, and aluminum are among the common materials reported in the literature [28, 47, 48]. Among them, gold and silver anode contacts provide better device performance due to the higher work function and better energy band alignment. However, both of these metals are relatively expensive and are not suitable for mass production. Aluminum is commonly used as a cathode electrode due to a low work function, but it can be used as an anode electrode with a MoO_x interface layer to achieve a 11.4% efficiency [49]. Thus, we propose to use less expensive MoO_x/Al as the back contact for the solution- and vacuum-based perovskite cells in this study.

Besides the standard device architecture, an HTL-free structure that was developed in a recent study has also attracted much attention. The device without a HTL shows a potential to reduce material costs and process complexity by using low-cost solution-processed carbon-paste back contact. The HTL-free perovskite cells have recently demonstrated a reliable stability during a 3-month outdoor test in Saudi Arabia, showing a potential to realize perovskite PV cells [50]. To compare it with standard devices, we also modeled the environmental impacts for the HTL-free device with the carbon electrode.

2.3. Selection of the Manufacturing Processes for the Perovskite Cell

We sought to optimize not only the materials but also the manufacturing processes. The ultimate device architecture and deposition methods that we expect to see in industrial scale production are shown in Figure 1. For the solution based method we followed steps 1, 2, 3a, 4

and 5b shown in Figure 1. For the vacuum based method we followed steps 1, 2, 3b, 4, 5b (for the device with HTL) and steps 1, 2, 3a, 5a (for the HTL-free structure).

Figure 1 Device architecture (a) and deposition techniques (b) of Perovskite devices expected to be commercialized.

Steps 1 and 2 are common for all three of the devices we modeled. In step 1, the FTO coated glass substrates (Pilkington, TEC 8 or TEC 15) are sequentially cleaned with acetone and isopropanol in an ultrasonic bath for 10 min each [51]. Then, in step 2, a 60 nm compact layer of tin dioxide (SnO_2) is deposited on FTO by spraying a 0.1 M $\text{SnCl}_2 \cdot \text{H}_2\text{O}$ in ethanol and annealing it at 180°C for 1 h.

In Step 3, the $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite can be deposited as the absorber layer using several possible methods that can be classified as either solution or vapor based (Figure 2) [46]. Solution-based processes for depositing $\text{CH}_3\text{NH}_3\text{PbI}_3$ include spin-coating, spraying, dip coating,

and printing [52]. Among these deposition techniques, spin coating is the most widely used for fabricating uniform thin films with nanoscale thickness for small-sized PV devices. However, most of the solution is spun off the substrate during the processing, leading to a material loss around 90 % [18]. Thus, spin coating may not be a feasible option for large-scale commercial fabrication of thin film PV devices. Sequential deposition method consisting of a PbI_2 thin film deposition followed by dipping the film into a $\text{CH}_3\text{NH}_3\text{I}$ solution can be used to achieve high-performance perovskite devices in the laboratory. Although this dipping technique is partially employed in the fabrication of other thin film solar modules [53], the extra step to deposit PbI_2 thin films would add complexity and lower fabrication efficiency. Thus, the dip coating technique is not considered optimal for commercialization. Consequently, we concluded that spray and inkjet coating would be the most feasible deposition methods for the commercialization of perovskite solar cells. These two techniques are known to promise low-cost production of solar PV by reducing material usage and energy input in the thin film deposition process, and are similar in that, both employ piezoelectric nozzles to generate micron-sized liquid droplets to deposit precursor solutions. Here, spray deposition was chosen to represent the solution based deposition approach because it is used for device fabrication in University of Toledo's PV labs.

Vapor based deposition can be achieved by vacuum [54] and non-vacuum [28] methods. As for the perovskite deposition, co-evaporation of $\text{CH}_3\text{NH}_3\text{PbI}_3$ and PbI_2 offers better control of the thickness, uniformity, and the grain structure of the perovskite layer [46] than non-vacuum, and therefore, has potential to be commercialized in the future [55]. Also, in contrast to non-vacuum methods that combines vacuum and solution based methods, it can be applied one step. Therefore, for practical consideration for mass production, one step vacuum method was chosen.

We assumed that the commercial scale perovskite cell fabrication would include a 300 nm perovskite absorber prepared by either the solution based spray deposition or the vapor based vacuum deposition method. In the solution-based deposition shown as Step 3a in Figure 1, the precursor solution consisting of 0.1 M PbI_2 (Sigma-Aldrich, 99 %) and 0.3 M $\text{CH}_3\text{NH}_3\text{I}$ (synthesized in house) are dissolved in N, Ndimethylformamide (Sigma-Aldrich, 99.8 %) (DMF) and spray-deposited at 70 °C for 30 min followed by drying at 100 °C for 1h. In the vacuum-based deposition shown as Step 3b in Figure 1, PbI_2 and $\text{CH}_3\text{NH}_3\text{I}$ are co-evaporated in a vacuum chamber ($< 10^{-6}$ Torr base pressure, $< 10^{-4}$ Torr deposition pressure) for around 30 min. During the deposition, the source temperatures for PbI_2 and $\text{CH}_3\text{NH}_3\text{I}$ are fixed at 270 and 100 °C, respectively.

In step 4, a 700 nm HTL is deposited by printing the precursor solution of 0.1 M CuSCN (Sigma-Aldrich, 99.5%) in chlorobenzene (Sigma-Aldrich, 99.8%) [41]. For the HTL-free device, no process is needed for this step. Finally, in step 5a, a 100 nm metal back contact (MoO_x/Al) is

deposited by evaporating at around 700 °C. The back contact for the HTL-free structure was deposited from a solution of commercial graphite paste in isopropanol by spray or printing (step 5b in Figure 1).

Figure 2 Possible routes for deposition of perovskites. The techniques used in this study are shown in bold.

2.4. Life Cycle Inventory

At the University of Toledo, the Photovoltaic Innovation and Commercialization (PVIC) center produces perovskite and other thin film cells in research labs. Life cycle inventory data were collected from the PVIC lab for manufacturing of 2" X 2" lab scale cells. We assumed 65 % of the area to be active, which is the average of previously reported values (Gong et al. used 70 % and Zhang et al used 60 %) [19, 21]). We also collected inventory data from the literature and from the Ecoinvent database. All modeling was done using GaBi 6.0 software.

Electricity consumption values were taken from the literature, and necessary assumptions were made in order to scale-up lab data to an industrial fabrication [51, 56]. Intermediate products that are not included in the Ecoinvent database were synthesized within the model assuming 100 % reaction efficiency (i.e. all inputs would react completely to form the desired product) (Table S.3). Electricity consumption for these chemical syntheses were not done on a reaction specific basis; average values were taken from the study of García et al. which incorporates a

methodology from Geisler [51, 57]. We note that the eventual contribution of electricity consumption during chemical syntheses was less than 0.1% to the total electricity consumption in this study.

Vacuum apparatus (e.g. control systems and vacuum pumps) used for the perovskite absorber and the metal back contact would likely run nonstop in a manufacturing facility. In absence of other data, we assumed these apparatus to run only during the deposition. While some of the material is wasted during deposition, deposition efficiencies are not well documented in the literature. For vacuum efficiencies a range of 30 to 50 % have been recommended [18, 55]. We used the upper value (50 %) of this range in our analysis assuming the manufacturers will prefer more efficient processes that reduce total waste and cost. For solution deposition a higher efficiency of 80 % [58] was used.

Emissions and waste streams associated with deposition were modeled as being released to different mediums during the production stage. Volatile organic compounds and waste from thermal annealing and vacuum deposition were assumed to be emitted to air. Lead emissions used for the perovskite layer were assumed to be emitted to fresh water. Other solid wastes in the solution-based methods were assumed to be released to industrial soil. All the energy input was modeled by waste heat output.

To compare the environmental impacts from perovskite cells to those of commercial technologies, we also modeled the first (mono-Si, Poly-Si) and second (amorphous-Si(a-Si), CdTe and CIS) generation solar cells. There is some variation in published energy and greenhouse gas emission metrics for these commercially fabricated solar cells [59-61] and part of this variation stems from the variability in the location the solar cells are manufactured [62]. To be consistent, we extracted all data for the commercial technologies from the Ecoinvent database. To further help with interpretation, we show the effect of the electricity mix in the supporting information (See Supporting Information, Figures S2 and S3).

2.5. Life cycle impact assessment

Nine midpoint environmental impact categories were modeled using TRACI impact assessment model: acidification (kg SO₂ equiv.), ecotoxicity (CTU,e), eutrophication (kg N equiv.), global warming potential (GWP) (kg CO₂ equiv.), human toxicity (CTU,h), cancer and non-cancer, and primary energy demand (PED) (MJ). TRACI is the most commonly used impact assessment method in the U.S. while some other impact assessment methods like ReCiPe, EcoIndicator and ILCD are more commonly used in Europe [63-65]. Our preliminary analysis using different impact assessment methods suggested some interesting findings with the marine eutrophication impact. Therefore, we also include results for this impact category, modeled

using the ReCiPe method. Finally, using the approach in Bhandari et al. we also present the energy payback time (EPBT)[61].

One important goal of perovskite deployment should be to reduce the total environmental impact from electricity generation. Yet, the meaning of the ‘total’ impact has been interpreted in different ways in PV LCA studies. Since the different types of environmental impacts are all calculated in different units, they are not directly additive or comparable to each other. Often, researchers use normalization methods embedded in existing life cycle impact assessment methods such as Ecoindicator 99, CML, and ReCiPe to compare across different impact categories or to add the different impact categories towards estimating a single ‘total’ impact score. In the U.S. the normalization is done using the US emissions and population data presented by Ryberg [66]. The fundamental concept in all of these normalization approaches is to divide the impact from PV LCA (for each impact category) with the total impact for a region and with the population in that region (e.g. for Europe, U.S. or global). The challenge with this approach is that the normalization factors are dependent on the region and different results and interpretations can be obtained depending on the normalization method. In addition, the absolute values resulting from normalization are not easy to interpret in this way. To overcome these challenges, in this paper, we use crystalline mono-Si as the reference point [67]. Since crystalline mono-Si is the second most dominant PV technology in the market and has the largest environmental impacts compared to commercial PVs, it is important to understand the impacts of new technologies in reference to the technology it may displace. To facilitate this interpretation, we normalized each impact from the different technologies to the corresponding impact from mono-Si.

3. Results and Discussion

3.1 Impacts from Manufacturing of the Cells

The impacts from manufacturing of the cells are shown in Figure 3. Based on the Ecoinvent database, and as was also reported by Gong[20], mono-Si has the highest and a-Si has the lowest total manufacturing environmental impact among commercial technologies (Figure 3). For each environmental impact category, the impacts from manufacturing mono-Si are higher than those of other commercial technologies. Total impacts from the three perovskite solar devices were about 10 to 30 % lower than that of mono-Si but still higher than those of second generation technologies. The acidification impacts from perovskite cells were considerably higher than in mono-Si. Checking our life cycle inventory, we note that the reason for high acidification in our model is due to the upstream emissions of sulfur compounds (sulphate, sulfur and sulphide) to freshwater during electricity production and tin extraction.

Figure 3 Comparison of perovskite devices with commercial PV technologies when normalized to mono-Si for selected impact categories. Note that GWP and PED stand for global warming potential and primary energy demand. Within each impact category, the impact (per m² of module manufactured) from each technology was divided by the impact from mono-Si. For mono-Si the bar height is unity for each impact category. For nine impact categories, the total impact for mono-Si is given as nine units. The data for mono-Si, poly-Si, a-Si, CdTe and CIS are directly extracted from Ecoinvent database.

3.2 Impacts from Generating 1 kWh of Electricity

While the impacts per unit area given in Section 3.1 are helpful for direct comparison of manufacturing of the cells, the ultimate goal for solar PV sustainability is the reduction of the total impact per unit electricity generated. To convert the impact results from manufacturing of the cells to impact results from unit electricity generated (i.e. the functional unit of the study), several other parameters are needed as shown in Equation 1;

$$Impact_{kWh} = \frac{Impact_{m^2}}{I \times \eta \times PR \times LT} \quad \text{Equation 1}$$

where,

I = insolation constant (kWh/m²-yr);

η = module efficiency (%);

PR = performance ratio of the module (%);

LT =lifetime of the PV technology (yr).

Impact_{m²}=Impact per 1m² module area

Impact_{kwh}=Impact per 1 kWh of electricity generated

Some of these parameters can be kept constant when comparing different technologies. For example, performance ratio and insolation are often kept constant at 75 % and 1700 kw/m²-yr, respectively; and module lifetime is often estimated as 30 years[61]. For perovskite solar cells, module lifetime does not currently exist but values of 1, 2, 5, 15 and 30 years have been used in the literature [18, 19, 21]. For efficiencies, once again, there is some ambiguity and different perspectives in what value should be used. In evaluating perovskite LCA results, Gong et al. [21] made the comparison with Laleman's inventory, which was based on 2011 efficiency data. However, since 2011 the module efficiencies have increased from 14.0 to 22.9 % for mono-Si, 13.2 to 18.5 % for poly-Si, 6.5 to 10.9 % for a-Si, and 7.1 to 17.5 for CdTe and 10.7 % to 17.5% for CIGS [22]. The latest best cell efficiencies reported by NREL are 25.0 % for mono-Si, 20.8 % for poly-Si, 13.6 % for a-Si, 21.5 % for CdTe and 21.7 % for CIGS [7]. Darling and You reported that efficiencies may decrease 19.6 % to 49.6 % from cell to module [24]. With the intent of comparing perovskite cells to other technologies in a future scenario, we used 15 % efficiency for our modeled perovskite cells which is 25 % lower than the champion perovskite cell efficiency (20.1%) [9]. This efficiency value is in the higher part of the efficiencies (6.4 %, 6.5 %, 9.1 %, 11 %, 11.5 %, 15.4 %) used in prior perovskite PV LCA studies [18-21]. We assumed a 30 year lifetime for all technologies except perovskite solar cells where an average value of 5 years was assumed. When perovskite lifetime is assumed to be much shorter than others, this has a large effect causing perovskite PV devices to have 5-8 times higher impacts than mono-Si (Table 2). The difference between perovskite and commercial technologies other than mono-Si is even

more drastic suggesting perovskite is not environmentally competitive with a 5 year lifetime. While both the efficiency and lifetime affect the impacts per kWh, the short lifetime assumption is the primary reason for the very high environmental impacts from perovskite cells.

Table 2 Environmental impacts from generation of 1kWh of electricity.

	mono-Si	poly-Si	a-Si	CdTe	CIS	Solution	Vacuum	HTL-free
Acidification [kg SO ₂ -equiv.]	9.3E-05	8.0E-05	7.6E-05	9.5E-05	5.3E-05	1.4E-03	1.5E-03	1.2E-03
Ecotoxicity [CTUe]	1.8E-01	1.6E-01	9.3E-02	2.1E-01	1.6E-01	1.5E+00	1.6E+00	5.0E-01
Eutrophication [kg N-equiv.]	1.2E-04	8.7E-05	4.8E-05	6.7E-05	1.3E-04	1.0E-03	9.2E-04	8.4E-04
GWP [kg CO ₂ -equiv.]	2.3E-02	2.0E-02	1.2E-02	1.2E-02	1.7E-02	1.5E-01	1.8E-01	1.2E-01
Human toxicity, [CTUh]	1.7E-09	1.4E-09	1.1E-09	1.0E-09	1.3E-09	9.4E-09	1.1E-08	7.5E-09
PED [MJ]	1.1E-08	1.1E-08	4.9E-09	1.1E-08	8.0E-09	3.1E-08	3.7E-08	2.4E-08
Marine Eutrop. [kg N-equiv.]	4.6E-01	4.1E-01	2.1E-01	1.9E-01	2.9E-01	2.5E+00	3.0E+00	2.0E+00
Total fresh- water use [kg]	1.1E-05	8.1E-06	1.2E-06	2.8E-06	4.4E-06	9.3E-05	2.4E-05	8.9E-05

3.3 Uncertainty and Sensitivity Analysis

As shown in Section 3.2, deploying perovskite cells in the market at expected efficiency and relatively short lifetime would not reduce the environmental impacts of PV technologies. Therefore, we asked a relevant question for these two parameters. Figure 4a shows how the improvement in lifetime of perovskite PV cells can bring a fast reduction in their total environmental impacts. The environmental performance of HTL-free perovskite cells may

supersede that of mono-Si cells within 26 years lifetime. For HTL-free perovskite device to supersede many of other commercial technologies, its lifetime has to be 40 years. Solution and vacuum based cells need even longer lifetimes for them to perform better than commercial technologies. With 15 % efficiency, solution and vacuum based perovskite cells need 33-34 year lifetimes to perform better than mono-Si. In contrast, if the lifetimes of perovskite cells are fixed to five years, perovskite solar cells would not have lower environmental impacts than commercial technologies even if their efficiency could increase up to 30 % (Figure 4b). Therefore, a longer lifetime should be targeted for further improvement of perovskite technology.

Figure 4 Sensitivity of total environmental impacts to lifetime (Figure 4a) and efficiency (Figure 4b) assumptions. Baseline perovskite cells efficiencies and lifetime assumed were 15 % and 5 years. This graphs test whether it is possible for perovskite cells to reduce overall impacts with efficiency or lifetime. Figure 4a shows the variation of perovskite impacts when perovskite cell lifetime is varied. Figure 4b shows the variation of perovskite impacts when its efficiency is varied. The parameters for commercial technologies were kept constant as in Section 3.2. All results are presented as total impacts normalized to mono-Si impacts as in Figure 3. Unlike figure 3, the mono-Si equivalent impact is calculated for 1kWh electricity generated not for manufacturing of 1m² of module.

From Figure 4, we can conclude that improvements in both the efficiency and the lifetime of perovskite solar cells are needed for perovskite cells to have lower environmental impacts than commercial technologies. Given the challenges with prediction to a future date, the relevant question to ask is about the probability of perovskite solar cells having impacts less than commercial technologies. We analyze this question in Figure 5. Based on the resulting cumulative density functions, we would expect the total environmental impact of HTL-free structure to be lower than that of mono-Si 55 % of the time. The HTL-free structure intersects with the other technologies at cumulative probabilities ranging from about 25 to 40 %. For solution and vacuum based perovskite cells, the chances of them having lower impacts are even lower.

The uncertainty analysis clearly depends on the input distribution (uniform assumed) used in Monte Carlo analysis which results in a cumulative probability distribution that is steep at first but then levels off very slowly suggesting that the total impacts from perovskite solar cells could be many times higher than those of mono-Si. (The x-axis scale was cut off at 35 but extends 266.) On the other hand, there is also a probability of all three perovskite cells having lower environmental impacts from all commercial technologies but this chance is only 10 %. In this analysis we did not incorporate the uncertainty resulting from the LCA model itself which is difficult to characterize but we estimate that the current modeling of perovskite cells reveal higher energy requirements (and corresponding higher impacts) than commercial technologies due to lack of commercial scale data for their production (see section 3.4). Once perovskite solar cells are being manufactured commercially, it is quite possible that their environmental impact data obtained not from lab estimation but from actual facilities will be lower. As we obtain facility level data for perovskite manufacturing and as we gain more confidence in perovskite cells having higher lifetimes and efficiencies, the shape of the cumulative density function will shift towards being steeper early on suggesting that the chances of perovskite cells having lower total environmental impacts than existing technologies will be higher than what we currently estimate in Figure 5.

* x shows median of total mono-Si equiv. of the perovskite cells.

Figure 5 Probability of perovskite cells having total impact less than that of commercial technologies. Monte Carlo simulations were run using @Risk software to create the data in Figure 5. Uncertainties in three parameters were propagated using uniform distributions. Insolation constant was varied from 1266 to 2187 kWh/m²/yr to represent the variation across the USA [61]. Efficiency was varied from 10 to 30 % with the higher value representing the thermodynamic limit for photovoltaic conversion, known as the Shockley-Queisser limit.[24, 68]. Lifetime was varied from 1 to 30 years following Zhang et al.'s[21] approach. Median values are indicated with a red x. Median/mean values were 7.90/14.3, 9.97/18.18 and 10.25/18.56 for HTL-free, solution based and vacuum based cells, respectively.

3.4 Energy Requirements of Perovskite Cells

We had anticipated the perovskite PV cell energy requirements to be lower than those of the thin film PV cells due to their simpler manufacturing methods and simpler structures. Contrary to our expectations, the PED of perovskite solar cells were lower than that of mono-Si but higher than the thin film cells (see the heights of the purple bars in Figure 3). PED incorporates the direct energy requirements of processing the materials and the manufacturing of the cells. If we leave out the energy requirements of the materials and analyze only the electricity demand of the perovskite solar cell manufacturing facility, the electricity requirement for vacuum, solution, and HTL-free devices are 821, 665, and 504 MJ/m², respectively. These values

are among other perovskite studies [18, 20, 21] (See Section 3.5 for the comparisons). In the EcoInvent database the electricity requirements for first and second generation solar cells range from (59 to 484 MJ/m²). Contrary to our expectations, the manufacturing electricity requirements of perovskite solar cells are higher than those of all current commercial technologies. We believe the reason for this is the incomplete specification of an industrial scale perovskite manufacturing process and the modeling assumptions that reflected this situation.

3.5 EPBT and GWP impacts: Comparison to Prior Perovskite LCA studies

Recent LCA studies on perovskite PV cells have shown large variation in results. Here we compare only the EPBT and GWP metrics since they were the only ones we could directly extract and compare from prior studies (Figure 6). In GWP impacts, the reported values vary by almost 50 fold among the three published cradle to gate studies. For solution and vapor based deposition methods, Espinoza et al. [18] found GWP emissions to be around 1100 g CO₂ eq. per kWh. However, Gong et al. found 24 and 32 g CO₂ eq. per kWh for the perovskite cells with ZnO and TiO₂ ETL, respectively. Zhang reported GWP as 414 g CO₂ eq per kWh for liquid titania perovskite cells[21] and Serrano-Lujan calculated this value as 1880 g CO₂ eq. per kWh The GWP in our study was in the lower end of the reported values (99 to 147 g CO₂ equiv. per kWh).

There is a large variation in the literature in perovskite solar cells' estimated EPBTs as well. Our estimates of the EPBT of the three perovskite structures varied between 1.05-1.54 years whereas values reported by both Gong et al. (0.19-0.27 years) and Espinoza et al. (1.1 years) were lower. (Espinoza et al. also reported EPBT of around 17 and 1 years when they assumed the lifetime of the perovskite to be 1 and 15 years, respectively, but we assume this was an error. We omitted these values from the comparison since EPBT should not depend on system lifetime.) In contrast, estimates from Zhang et al [21] are much greater (5.3-55.4 years).

Ultimately, it is helpful to compare the EPBTs not only among perovskite solar cells but also with existing commercial technologies. Bhandari et al [59] recently compiled and harmonized the EPBTs for Si and thin film based solar cells. These data also display a wide variation in EPBT for a given technology even when the data are harmonized to the same insolation and performance ratios [59]. Yet, if we consider the mean of the harmonized EPBTs given in Bhandari et al (4.1 yr for mono-Si, 3.1 yr for poly-Si, 2.3 yr for a-Si, 1.0 yr for CdTe and 1.7 yr for CIGS [61]) the perovskite solar cells that we modeled appear quite competitive with all of the commercial technologies.

Another way to interpret the perovskite EPBTs is to compare them to standard organic PV which has shown great advances in the last decade [69] and seem to promise much lower environmental impacts than commercial technology due to the use of inexpensive materials

and solution processing methods. For example, Espinosa et al. [70] report that polymer PVs with 3 % efficiency can have lower environmental impacts than crystalline silicon technology and deliver EPBT as low as 1.35 yr. In another study with a 10 % efficiency assumption, an EPBT of as short as 12 days was estimated for organic PV [24]. Roes estimates that a polymer module on a flexible substrate can have lower CO₂ equiv. than poly-Si even at a 2.6 yr lifetime [67]. More LCA studies directly comparing organic PV to perovskite PV would be helpful in the future to improve our understanding of the environmental impacts of perovskites in relation to organic PV.

The variations in results among the published perovskite LCA studies stem from the different materials, deposition methods, electricity inventories, and assumptions used in the models (See Table 3). For example, performance ratios, power conversion efficiencies, and active area assumptions are slightly different among the published data. If we analyze the choice of electricity mix in the models, we note that in our study, we extracted the data from Ecoinvent database and used the average US electricity mix in the models. Yet, Espinoza et al. and Serrano et al. used Denmark's low-voltage electricity. Similar to our study, Gong et al. and Zhang et al. used the average U.S. electricity mix, which is higher GWP emissions per unit of MJ electricity produced (Figure S2).

While there could be many reasons for variations, we estimate that the large CO₂ equiv. difference between Espinosa and Gong can be contributed to the direct process energy to deposit the layers. Gong estimated the direct process energy needed to fabricate 1 m² module as 7.78 kWh (TiO₂) and 4.56 kWh (ZnO) [19] while Espinosa found the same value as 1080 kWh (Solution) and 1460 kWh (Vacuum)[18]. We can see this difference more clearly in the deposition of Spiro-MeOTAD (HTL). For the deposition of that layer Gong only considered the spin coating process which consumes 0.244 kWh (per m²) while Espinosa modeled annealing and nitrogen glovebox energy requirement in addition to spin coating and reported 276 kWh (per m²). In contrast, a much higher direct process energy value is reported by Serrano-Lujan et al. [20]; this study is based on the model given in Espinoza [18] and reports a value of 31,700 kWh (per m²) for direct process energy. Since Zhang modeled dye synthesized solar cell, the materials (layers) and processes are quite different in that study and cannot be directly compared.

As another example of the effect of the materials and modeling assumptions, we note that both our study and Gong et al. used FTO as the top contact layer but the percentage contribution of the PED of this layer is much lower in this study (15 %) than in Gong's study (27 %). The PV companies have been using commercially available TCO glasses for this layer (e.g., Pilkington, TEC 15). In our study, we selected the low-iron solar glass available in the Ecoinvent database to model this glass. However, Gong separately modeled the glass and the FTO deposition. Similarly, in contrast to our study where we modeled aluminum as the metal electrode, Gong et

al. modeled gold, silver paste, and found the embedded energy to be primarily dominated (70 to 85 % of total) by the energy demand for these materials. This is an expected result since gold and silver are considered precious materials and embedded energy of gold and silver are much higher than aluminum (see Figure S.2).

Figure 6 Comparison of GWP and EPBT data from this study with prior studies. Data shown are for cradle to gate and 5 year lifetime assumption for all studies. Since Zhang et al [21] did not provide an average EPBT value, we plotted the lowest point of the uncertainty range they reported. Also, EPBT value of tin based perovskite cell was not available provided.

Table 3 Comparison of the modeling assumptions in published Perovskite PV LCA models

	Espinosa et al. 2015 [18]	Gong et al. 2015 [19]	Zhang et al. 2015 [21]	Serrano-Lujan et al. 2015 [20]	This study
System boundary	Cradle-to-gate	Cradle-to-grave	Cradle-to-gate	Cradle-to-gate	Cradle-to-gate
Lifetime	1 - 15 yr	2 yr	1 to 30 yr	1 yr	5 yr
Performance Ratio	80 %	80 %	75 %	80 %	75 %
Active Area	Not provided	70 %	60 %	Not provided	65 %
Power conversion efficiency	11.5 % (solution) 15.4 % (vacuum)	9.1 % (TiO ₂) 11.0 % (ZnO)	6.5 %	6.4 %	15 %
Material Structure	FTO/TiO ₂ / Lead-P/S-MeOTAD/Ag (for solution) ITO/REDOT:PSS/ Lead-p/PCBM/Al (for Vacuum)	FTO/TiO ₂ / Lead-p/S-OMeTAD/Au ITO/ZnO/ Lead-p /S-OMeTAD/Ag	FTO/TiO ₂ / Lead-p /electrolyte/ Pt.	FTO/TiO ₂ /Tin-P+TiO ₂ /S-MeOTAD/Au	FTO/SnO ₂ /Lead-P/CuSCN/ MoO _x -Al (for solution and vacuum) FTO/SnO ₂ /Lead-P/C-paste (for HTL-free)
Fabrication methods	SD: Oxygen plasma treatment, Spin coating, Drying, Sintering, evaporation, annealing VD: Evaporation, Spin coating, Vacuum	TiO ₂ : Screen printing, Sintering, Spray pyrolysis, spin coating ZnO: Ultrasonic cleaning, spin coating,	Spin coating, dipping, annealing	Spin coating/ annealing/ evaporation [71]	Ultrasonic cleaning, spray pyrolysis, Screen printing, co-evaporation vacuum, annealing,
Electricity inventory	Denmark Electricity, low voltage	US Electricity mix	US Electricity mix	Denmark Electricity, low voltage	US Electricity mix

553 3.6 Hot spot analysis of Perovskite Solar Cells: Comparison of Electricity versus Material 554 Impacts

555 In sections 3.6-3.8 we present hot spot results that can help guide researchers and
556 manufacturers on how to reduce the impacts from perovskite cells. A possible question to ask is
557 whether the impacts come mainly from electricity use versus materials (section 3.6). If the
558 impacts are from electricity, then this means the focus should be in reducing electricity use
559 during manufacturing. In that case, the relevant question is which layer one should focus on to
560 reduce the impact (section 3.7). Finally, the hot spot analysis can also be done for the specific

561 materials to evaluate if some materials should be substituted with others to decrease impact
562 (section 3.8).

563 For all of the impact categories studied, except ecotoxicity and marine eutrophication, the
564 major impacts were from electricity consumption during manufacturing, which contributed
565 from 50 to 90 % of the different impacts (Figure 7). In particular, impacts from electricity are
566 around 80 % for GWP, human toxicity (cancer and non-cancer), PED and freshwater use. These
567 results emphasize the importance of using electricity efficient manufacturing methods and
568 clean electricity sources to reduce environmental impacts.

569 From our data, it is possible to roughly estimate how the impacts may change if more energy
570 efficient processes are used by a manufacturer. For instance, in scaling up the vacuum based
571 model to commercial production, if we consider that our approximation using lab data were
572 low and that the industry may reduce the processing energy by 20 % (in comparison to our
573 current model), we can say that the electricity dominates 40 to 90 % of the impacts (see Figure
574 7). A 20 % reduction in processing energy would therefore results in 8 to 18 % reduction in final
575 impacts. Corresponding LCIA can be calculated by using values given in Table 2.

576 Interestingly, prior perovskite LCA studies reported contradictory results for electricity versus
577 materials impacts. In Espinoza's study, electricity contributed to more than 90 % of the impacts
578 likely due to energy intensive deposition choices used in that study [18]. Yet, in Gong's study
579 the material choices such as gold or silver, resulted in a dominant contribution of materials to
580 the impacts categories [20]. Our study lies somewhere in between Espinoza's and Gong's
581 results. These differences are a good example of how sensitive PV LCA results are to the specific
582 device architecture and deposition methods modeled. More research in comparing specific
583 materials and deposition methods would be helpful in guiding future research and better
584 interpreting the variation of results in the literature.

585 Figure 7 is also helpful in comparing the three devices with respect to the individual impact
586 categories. From this comparison, we see that for all except the marine eutrophication impact,
587 the highest, medium, and lowest impacts are observed for vacuum, solution, and HTL-free
588 devices, respectively. The differences are fairly small with solution and HTL-free devices being
589 about 10 to 20 % lower than the vacuum based device. The primary reason for the lower
590 impacts in these two devices is that the solution based deposition of the perovskite layer in
591 solution and HTL-free devices result in the lower energy consumption which then reduces the
592 environmental impacts for these devices compared to the vacuum deposition. (In Figure 2, the
593 solution based device had higher impact than the vacuum based device primarily due to the
594 marine eutrophication impact.)

595 Marine eutrophication shows an exception to the general order of impacts with vacuum having
596 the highest and HTL-free having the lowest impacts. Marine eutrophication is also the primary

reason the solution based device had higher impact than the vacuum based device in Figure 2. Marine eutrophication impact is largely affected by the use of DMF in solution based deposition of perovskite which caused a much greater marine eutrophication impact in solution and HTL-free devices compared to the vacuum based device.

Another interesting observation is with the use of copper thiocyanate. HTL free device does not use this compound and results in about a 60 % lower ecotoxicity and 30 % human toxicity compared to vacuum and solution based devices.

Figure 7 Comparison of environmental impacts among solution, vacuum and HTL-free devices. The results are plotted as normalized to the highest value within each impact category.

3.7 Hot Spot analysis of Perovskite Cells. Which steps in cell preparation use the most electricity?

Process breakdown of the electricity consumption during manufacturing of perovskite solar cells is shown in Figure 8. Pre-treatment consists of the processes that have to be done before depositions, including cleaning of solar glass or stirring and heating the chemicals necessary for solution based depositions (steps 1, 2, 3a and 4). Depositions include the processes such as screen-printing, spray, or vacuum (steps 2, 3a, 3b, 4 and 5). This analysis shows that the highest electricity requirement for processing the cells come from the absorber layer (perovskite deposition) for all three devices. In the case of solution based deposition of perovskites, pretreatment process also has high electricity requirements, larger than the deposition process itself. As was expected, vacuum-based deposition of perovskites was found to be much more energy intensive (about twice as much) than solution based perovskites. This caused the

perovskite deposition in the vacuum based device to require about three quarters of the total energy consumption during cell manufacturing.

From Figure 8 we also see that post processing contributes to about less than seven percent in all three devices suggesting that the focus in reducing electricity consumption should be in pre-processing and deposition steps. With respect to deposition the preference should be in solution based deposition. In the back contact layer, the HTL free device was deposited using vacuum whereas the other two devices were deposited using solution processing. As was in the perovskite deposition, the high energy requirements of vacuum processing are also evident in this layer. These results show that to reduce electricity usage in thin film manufacturing, vacuum deposition should be avoided and instead solution deposition should be used.

Among the three modeled device architectures, the substrate and ETL were the same and therefore contributed equal amounts of electricity to cell production. Cleaning of the solar glass using sonication was responsible for 9 to 14 % of the electricity consumption. Using detergent based chemicals instead of sonication may be a way to reduce energy consumption for the substrate layer.

Figure 8 Breakdown of direct process energy requirements in each layer

3.7 Hot spot analysis of perovskite cells: Which materials in cell preparation contribute more to the impacts?

Material impacts from the vacuum based method were lowest for most of the impact categories (Figure 7). Part of the reason for this is that vacuum based method uses less organic compounds (e.g. DMF and chlorobenzene) that contribute a lot to the impacts from materials (Figure 9). Marine eutrophication impact of the solution and the HTL-free structures were especially dominated by organics (around 95 %) and were 16 to 20 times higher than that of the vacuum based method. The primary reason for this was the use of DMF in the solution-based deposition of the perovskites.

Other than the organic compounds, tin and the waste stream contributed considerably to the impacts from materials in all three perovskite cell devices. Raw tin usage dominated acidification, GWP, human-toxicity, cancer and non-cancer, PED and fresh water use categories. High toxicity findings of this study is consistent with Serrano-Lujan[20]. Waste streams (that were dominated by copper and tin including waste to industrial soil) dominated the ecotoxicity and eutrophication impacts. This result is quite higher than what we expected; however similar to ours, Ryberg et al. also found questionably high impacts from waste released to industrial soils [66].

Figure 9 Comparison of material impacts among solution, vacuum and HTL-free devices. The results are plotted as normalized to the highest material impact value within each impact category (For raw data see Table S 8).

We note that the contribution from lead is insignificant in all impact categories in all three perovskite devices (Figure 9). Cradle-to-gate environmental impacts of lead used in the PV solar cell come from mining of lead and the release of lead back into the environment when some lead waste is created during deposition of the absorber. These impacts contributed less than 0.2% of total impacts in all of the impact categories. Thus, our results confirm the conclusions from previous studies [18-21]. However, to our knowledge, Espinosa and Gong did not include any lead emissions during manufacturing [18, 19]. Although Zhang et al. discussed the release of metals to the environment (iron, magnesium, and aluminum), lead emissions may not have been considered in that study either [21]. Unlike prior studies, we modeled a direct freshwater release of the lead which was assumed as a material loss during perovskite layer deposition. The negligible impact from lead may be due to the limited mass of lead used in the cell. Another likely reason is the limited accuracy in modeling the toxicity of metals in LCA studies. For example, the TRACI model used in this study is based on the USETOX model which has ‘interim’ characterization factors for metals [72]. Also, ecotoxicity characterization in USETOX is only valid for fresh water. USETOX model is embedded in the other LCA studies as well. For example, Espinosa et al., Serrano-Lujan et al., and Zhang et al. used the International Reference Life Cycle Data System (ILCD) in their models which is also based on the USETOX model [18,20,21]. In contrast, Gong et al. used Ecoindicator and CML, both of which are based on USES-LCA [19]. USES-LCA has differentiated characterization factors for lead related to aquatic, sediment and terrestrial ecotoxicity potentials [73]. Based on this different toxicity model, Gong et al. did not find lead to pose a major concern either [19]. The only study that showed significant toxicity from lead is Serrano-Lujan’s cradle-to-grave lead perovskite model. In this study, lead emissions at the end of life were modeled in a landfill scenario which included leaching of 70% of total lead content of the module into soil [20]. This model resulted in >10 times higher fresh water toxicity compared to the cradle-to-gate system boundary. Much uncertainty still remains at the end-of-life phase of perovskite cells. More work in this area is needed to better understand the environmental impact of lead in perovskite cells.

4. Conclusions

The goal of this study was to evaluate the environmental impacts of vacuum, solution, and HTL-free perovskite solar cell devices using fabrication approaches that are amenable to large scale manufacturing. A comparison of environmental impacts was made with mono-Si as a reference point. We found that manufacturing of perovskite solar modules causes 10 to 30 % lower impacts than manufacturing mono-Si PV. However, if perovskite cells were to enter the market, their environmental impacts would be higher than those of all commercial PV technologies mainly because of their shorter lifetimes. Monte Carlo analysis varying lifetime and efficiencies of perovskite cells showed that HTL-free structure could perform equal or better than mono-Si cells with about 55 % cumulative probability.

To our surprise, the energy requirements of perovskite solar cells were not lower than those of commercial technologies. This result is likely due to incomplete specification of the commercial scale fabrication of perovskite solar cells. The EPBT and GWP varied from 1.0 to 1.5 years and 100-150 g CO₂ equivalence (per kWh). These values are within previously reported data for other perovskite solar cells. Solution-based methods with spray used in perovskite deposition were observed to decrease the overall electricity consumption.

Among the three structures modeled, the HTL-free structure had the lowest environmental impacts in all impact categories, except marine eutrophication. Marine eutrophication impact of the solution and HTL-free structure were dominated by organics (around 95 %). Material impacts attributed to the vacuum based method were lower than other two methods, except for ecotoxicity. The ecotoxicity value of HTL free structure was around 30 % of the vacuum and solution based methods because it is missing the HTL made from copper thiocyanate. The impact of lead used in the absorber layer was negligible, which is consistent with the literature.

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