PerTPV – Perovskite thin film photovoltaics

Grant agreement 763977

Deliverable 4.1

Initial assessment of environmental impacts from cradle to cradle of existing prototypes

WP4

Lead beneficiary: FHNW

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Delivery date: 30/09/2019

Confidentiality level: Public

The PerTPV project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 763977.

Revision History

Author Name, Partner short name	Description	Date
Vinzenz Müller, FHNW Dirk Hengevoss, FHNW Felix Schmidt, FHNW Markus Lenz, FHNW	Draft deliverable	18/09/2019
Henry Snaith, UOXF	Revision 1	27/09/2019
Henry Snaith, UOXF	Final version (rejected)	30/09/2019
Vinzenz Müller, FHNW Dirk Hengevoss, FHNW Felix Schmidt, FHNW Markus Lenz, FHNW	Revision 2	21/11/2019
Vinzenz Müller, FHNW Dirk Hengevoss, FHNW Felix Schmidt, FHNW Markus Lenz, FHNW	Revision 3	17/04/2020
Henry Snaith, UOXF	Final version 2	17/04/2020
Henry Snaith, UOXF	Final Version	05/10/20

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Abbreviations

ACN	Acetonitrile
ACS	American chemical society
Ag	Silver
AR	Anti-reflective
Au	Gold
BOS	Balance of system
C_{60}	Fullerene
CdTe	Cadmium telluride
CED	Cumulative energy demand
CSEM	Centre Suisse d'Electronique et de Microtechnique
D4.1	Deliverable 4.1
DMDP	P,P′-di(2-ethylhexyl)methanediphosphonic acid
DMF	Dimethylformamide
DMSO	Dimethyl sulfoxide
€	Euro
E	Ecosystems
EA	Ethyl acetate
EC-JRC	European Commission, Joint Research Centre
EDTMP	N,N,N′,N′-ethyl enediaminetetrakis (methylenephosphonic acid)
EPFL	École Polytechnique Fédérale de Lausanne
ETL	Electron transport layer
EVA	Ethylene-vinyl acetate
FHNW	University of Applied Sciences and Arts Northwestern Switzerland
FTO	Fluorine-doped tin oxide
GCI-PR	Green Chemistry Institute Pharmaceutical Roundtable
GFF	Geometrical fill factor
GHS	Global harmonized system
GWP	Global warming potential
HH	Human health
HTL	Hole transport layer
ILCD	International Reference Life Cycle Data
IPCC	Intergovernmental Panel on Climate Change
ISE	Institute for Solar Energy Systems

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ISO	International Organization for Standardization
ITO	Indium tin oxide
(k)g CO_2 eq	(Kilo-)gram carbon dioxide equivalents
kWh	kilowatt hour
LCA	Life cycle assessment
LCI	Life cycle inventory
LCIA	Life cycle impact assessment
LCOE	Levelized cost of electricity
m^2	Square meter
MJ	Mega joule
Mo	Molybdenum
mPt	MilliPoints
NREPBT	Non-renewable energy payback time
NiO	Nickel(II) oxide
OXFORD PV	Oxford Photovoltaics Limited
PCE	Power conversion efficiency
PEDOT:PSS	Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate
PerTPV	Perovskite thin-film photovoltaic
PET	Polyethylene terephthalate
PO	Polyolefin
ppm	Parts per million
PSA	Pressure-sensitive adhesive
PSC	Perovskite solar cell
PV	Photovoltaic
SnO_2	Tin(IV) oxide
Spiro-OMeTAD	N2,N2,N2′,N2′,N7,N7,N7′,N7′-octakis(4-methoxyphenyl)-9,9′-spirobi[9H-fluorene]-2,2′,7,7′-tetramine
(c-/n-) TiO_2	(Compact-/nanoparticulate-) titanium dioxide
UV	Ultraviolet
VTT	Technical Research Centre of Finland Ltd
W/m^2	Watt per square meter
WP4	Work package 4
yr	year
ZnO	Zinc oxide

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Summary

As part of work package 4 (WP4) of the perovskite thin-film photovoltaic (PerTPV) project, this deliverable (D4.1) shares the results of the life cycle analysis (LCA) of the production process of perovskite solar cells (PSCs) from raw material extraction up to the production of the PSCs including the recycling of lead iodide (PbI_2) from the perovskite layer (from cradle to cradle, see Figure 1) of four chosen prototypes. Data on these prototypes (three single-junction PSCs and one all-perovskite tandem PSC) were obtained from the project partners. The goal of this LCA was to compare the PSC prototypes to benchmark photovoltaic (PV) technologies and assess which material use should be avoided or minimised from an environmental point of view. The environmental impact was measured through International Reference Life Cycle Data (ILCD) indicators, global warming potential (GWP), cumulative energy demand (CED) as well as non-renewable energy payback time (NREPBT). The assessment of GWP, CED and NREPBT showed that the prototypes of the PerTPV project were able to achieve lower carbon and energy footprints compared to silicon PV technologies and compete with CdTe solar cells, another thin-film PV technology. The impacts assessed through ILCD indicators stemmed mainly from the bottom (sputtering of indium tin oxide (ITO)) and top (gold) contact layers. Comparing literature data on deposition techniques suggested a benefit of solution deposition (e.g. printing) over vacuum deposition methods (e.g. sputtering and thermal evaporation) due to lower material waste and energy consumption. Within the scope of this LCA (from cradle to cradle), the extraction of lead (Pb) and its use in the production of the perovskite layer did not significantly increase the analysed impact indicators. We estimated that the Pb content of 1 m^2 of PSC panels would equal only between $^1/_9$ and $^1/_{15}$ of the “guide value” for Pb in topsoil (see Chapter 2 for details). Additionally, in the literature ([1], [2]) effective ways to reduce the risk of Pb leaching after breaking of PSC panels have been presented.

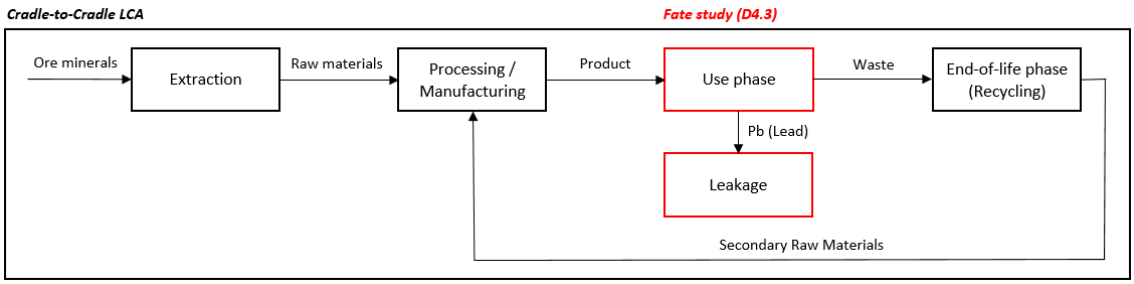

Figure 1: System boundaries of the LCA from cradle to cradle (i.e. from raw material extraction to recycling). The environmental fate study (red) analysing the leakage of Pb during the use phase and potential measures to reduce it will be addressed in deliverable 4.3.

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1. Introduction

The PerTPV project aims to advance perovskite thin-film photovoltaic (PerTPV) technology by increasing the efficiency, stability, scalability and module fabrication methodologies that are compatible with high-volume manufacturing. The project targets increased power conversion efficiencies (PCE) of >25% and >30% for single-junction and tandem perovskite solar cell (PSC) devices, respectively. Furthermore, it aims at increasing service lifetimes beyond 25 years whilst decreasing production costs (levelized cost of electricity [LCOE] < 0.03 €/kWh). To achieve this, the project develops different organic and inorganic materials and production processes whilst accounting also for the associated environmental impacts.

Work package 4 (WP4) of the PerTPV project is dedicated to the following:

- Assessing the environmental, health and safety impacts of PSC manufacturing, deployment and recycling;
- identifying potential environmental constraints; and
- developing a scalable protocol for end-of-life recycling.

While the PerTPV project partners are developing new materials/designs for their PSC modules, the project also analyses the first PSC prototypes from an environmental perspective in the form of a life cycle analysis (LCA).

This deliverable (D4.1) presents the results of the LCA, which focuses on three commonly used environmental impact indicators: global warming potential (GWP, [kg CO_2 eq]), cumulative energy demand (CED, [MJ]) and indicators of the International Reference Life Cycle Data (ILCD, [mPt]) system. These parameters are presented normalized to 1 m^2 PSC as well as to kWh electricity produced. Additionally, the non-renewable energy payback time (NREPBT, [months]) shows how long it will take until the PSC produces net energy. A special focus is given to the use of lead (Pb) within the active layer and solvents used for the deposition methods. Prior to the LCA, the partners were provided with recommendations on solvents regarding their adverse environmental effects (i.e. following a 'safe-by-design' approach). The goal of D4.1 is to provide a LCA that establishes a base line assessment for existing PSC prototypes of the partners and allows identifying components and processes causing most impacts to foster environmental improvements during an early stage of development in the project.

2. The use of lead (Pb)

To address one of the major social acceptance concerns of perovskites - the use of lead in the photoactive layer - current literature is used as base to estimate environmental concentrations and potentially adverse effects.

Data on soil heavy metal concentrations in the EU based on more than 23'000 topsoil samples (upper 0.2 m), can be used as the reference value for the Pb background concentration [3]. The mean Pb concentration (mg Pb/kg topsoil) was 15.3 ppm (min. 1.63, max. 151.12, std. deviation 8.33). The prototypes analysed here included between 600 and 1'000 mg Pb per m^2 PSC. For each m^2 PSC that would 100% leach into the

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ground, 3–5 ppm Pb would be added to the m^2 topsoil below the module (assuming a soil density of 1.3 t/m^3, 0.2 m topsoil)[1]. The ordinance on impacts on soil contamination in Switzerland states 50 ppm as the so called “guide value” for Pb. If the guide value is not exceeded there is no risk, even in the long term, on any negative impact including soil fertility [4]. One can argue on the ultimate design of an outdoor installation and the resulting ratio of Pb leached and soil contaminated. Nevertheless, to increase Pb concentration from a reference soil (15 ppm Pb) to the guide value (50 ppm Pb) in a depth of 20 cm and in an area of 1 m^2, Pb from a PSC area of 9.1–15.2 m^2 can be leached entirely.

Hailegnaw et al. [5] calculated in 2015 that, even if one in 300 PSC panels would break and release all the Pb within 20 years, the amount of Pb emitted would still be lower compared to the state-of-the-art coal power plant per kWh of produced electricity. Similarly, Fabini [6] assumed that with a lead intensity (total lead content per unit electricity produced) of 38 µg/kWh [2] and a panel break and release rate of 1%, 1.6 tonnes of Pb would be released each year (if all US electricity was produced by PSC). This would be insignificant (<1%) compared to the estimated 5’900 – 93’000 t/year lead in US coal wastes from electricity production.

While these estimates show a relative reduction of lead emissions compared to coal power plants, lead emitted through damaged PSC modules still has to be kept to a minimum since representing point sources. Jiang et al. [1] showed that encapsulating the modules with self-healing epoxy resins and a glass cover could reduce Pb leakage from damaged modules drastically (minus 94% versus non-encapsulated cells). The encapsulation offered protection upon simulated hail impact followed by acid rain, as well as elevated temperatures expected during a sunny day (45 °C for 4 h).

Li et al. [2] included a Pb absorbing coating to the front and back of the cell achieving an on-device sequestration efficiency of 96%. The materials used are a transparent Pb-absorbing P,P′-di(2-ethylhexyl)methanediphosphonic acid (DMDP) film on the bottom and a mixture of Pb-chelating agents such as N,N,N′,N′-ethyl enediaminetetrakis (methylenephosphonic acid) (EDTMP) blended in a host polymer matrix.

Despite being efficient in limiting Pb emissions, both approaches (self-healing epoxy resin; Pb absorbing coating) still need to prove their efficiency from an economic and environmental point of view.

While some local increases in Pb soil concentrations are possible due to PSC leaching, proper operation with immediate replacement of damaged modules, as well as disposal at the end of the life will limit the potentially adverse effects of Pb. On device sequestration and appropriate encapsulation approaches may further reduce possible risks during the use phase of the life cycle.

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[1] ~0.6 g Pb (from 1 m^2 PSC of prototypes A, B and C) per 1 m^2 of topsoil to a depth of 0.2 m; density 1.3 t/m^3
= plus 2.3 ppm Pb/m^2 topsoil;
~1 g Pb (from 1 m^2 PSC of prototype D) per 1 m^2 of topsoil to a depth of 0.2 m; density 1.3 t/m^3
= plus 3.8 ppm Pb/m^2 topsoil;
= 5 ppm Pb/m^2 topsoil

[2] For prototype D (1’000 mg Pb/m^2) the lead intensity would be 99 µg/kWh.

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3. Selection of solvents for a 'safe-by-design' approach

A number of solvent selection guides [7] by major pharmaceutical companies are available, including from GlaxoSmithKline [8] [9] [10], Pfizer [11] and Astra Zeneca [12]. The American Chemical Society (ACS) Green Chemistry Institute Pharmaceutical Roundtable (GCI-PR) has also produced an industry-wide solvent selection guide [13]. Prat et al. (2014) compiled the data given in these guides and developed a methodology based on a set of safety, health and environment criteria that aligned with the Globally Harmonised System (GHS) as well as European regulations [14]. The solvents were classified into four categories: 'recommended (green)', 'problematic (yellow)', 'hazardous (red)' and 'highly hazardous (dark red)' (the other guides may use similar categories).

Combining the available data, overall, 160 different solvents were assessed for this PerTPV. For instance, acetonitrile (ACN) is a popular solvent in perovskite research. Prat et al. (2016) first classified ACN as green and then changed the classification to yellow because of the low occupational threshold value of ACN [15]. Alder et al. (2016) also classified acetonitrile as yellow [7]. Following a conservative 'safe-by-design' approach, the use of ACN was highlighted as potentially problematic and should therefore be substituted if possible to avoid potentially negative impacts in the life cycle.

4. Prototypes

For this LCA study, three single-junction and one tandem (all perovskite) PSC prototypes of four PerTPV partners (CSEM, EPFL, OXFORD PV, VTT) with respective materials and deposition methods were analysed (Table 1, Table 2). These prototypes do not represent the final architectures developed by the partners but an intermediary stage helpful to establish which materials and deposition methods should be favoured.

All four perovskite layers (referring to the layer between electron transport layer (ETL) and hole transport layer (HTL), not the entire solar cell) that were analysed contained lead (Pb) and the impact of Pb was accounted for in the LCA, but the exact compositions of the perovskite layers are not shared here due to confidentiality.

The amount of materials used for the assessment of the environmental impacts was calculated by multiplying the respective layer thickness with its density for an area of 1 m^2. The functional unit of 1 m^2 was chosen because it is commonly used in LCA studies and therefore eases comparison. However, such sized prototype cells do not yet exist, and such large modules and the scale-up from the small-size prototype of around 1–5 cm^2 to 1 m^2 may have led to inaccuracies in the estimates.

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Table 1: Composition and deposition methods of prototype D (estimated module efficiency = 23%), the tandem PSC. Exact composition and thickness not disclosed due to confidentiality.

	Prototype D (tandem-cell)		
Layer	**Thickness [nm]**	**Materials**	**Deposition method**
Encapsulation	1.00E+06	EVA foil	Thermal laminator
Substrate	2.00E+06	Glass	
Bottom contact 1	150-250	FTO	Sputtering
Bottom contact 2	15-25	ITO	Sputtering
Electron transport	15-25	PEDOT:PSS Solvents: IPA	Solution deposition
Narrow gap perovskite	350-450	Perovskite material (confidential) Solvents: DMF, DMSO, EA	Solution deposition
Recombination junction 1	15-25	C_{60}	Thermal evaporation
Recombination junction 2	15-25	Diamine derivate	Thermal evaporation
Wide gap perovskite	450-550	Perovskite material (confidential) Solvents: DMF, DMSO, EA	Thermal evaporation
Hole transport	40-60	ITO	Sputtering
Top contact	150-250	Ag	Screen print
Front glass	3.20E+06	AR-coated Glass	Thermal laminator

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Table 2: Composition and deposition methods of the single-junction PSCs, prototypes A (estimated module efficiency = 5%), B (estimated module efficiency = 15%) and C (estimated module efficiency = 20%). Exact composition and thickness not disclosed due to confidentiality.

	Prototype A (single-junction)			Prototype B (single-junction)			Prototype C (single-junction)		
Layer	**Thickness [nm]**	**Materials**	**Deposition method**	**Thickness [nm]**	**Materials**	**Deposition method**	**Thickness [nm]**	**Materials**	**Deposition method**
Encapsulation (2 layers: top & bottom)	8.00E+04	PET with UV Barrier	Lamination	3.20E+06	PO foil	Lamination			
	2.50E+04	Epoxy resins repr. PSA		4.00E+06	Glass				
Substrate	1.25E+05	PET		7.00E+05	Glass		4.00E+06	Glass	
Bottom contact	50-150	ITO	Sputtering	250-350	ITO	Sputtering	450-550	FTO	
Electron transport	20-30	Nanoparticles (SnO_2) Solvents: H_2O, IPA	Printing	15-25	C_{60}	Thermal evaporation	25-35	c-TiO_2 Solvents: EtOH	Spray pyrolysis
				5-15	SnO_2	Atomic layer deposition	200-300	n-TiO_2 Solvents: EtOH	Spin-coating
Perovskite	350-450	Perovskite material (confidential) Solvents: DMF, DMSO, EA	Printing	350-450	Perovskite material (confidential) Solvents: DMF, DMSO, EA	Spin-coating	450-550	Perovskite material (confidential) Solvents: DMF, DMSO, EA	Spin-coating
Hole transport	150-250	Spiro-OMeTAD Solvents: CB, tBP, Li-TFSI	Printing	15-25	NiO	Sputtering	250-350	Spiro-OMeTAD Solvents: CB, tBP, Li-TFSI	Spin-coating
Top contact	50-150	Ag	Thermal evaporation	5-15	Ag	Thermal evaporation	50-100	Au	Thermal sublimation

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5. Methodology

The LCA methodology is standardised in ISO 14040:2006 [16] and consists of the following steps:

1. **Goal and scope:** Definition of system boundaries and functional units.
2. **Life cycle inventory (LCI):** Elaboration of a mass balance for a process including all inputs and outputs.
3. **Life cycle impact assessment (LCIA):** Assessment of the environmental consequences of the LCI, such as climate change, natural resource depletion, ozone depletion, ecotoxicity, etc. with specific indicators.
4. **Interpretation:** Identification of processes and flows that confer the main environmental impacts and recommendation of improvement measures.

This LCA is executed according to the ILCD Handbook [17].

5.1. Goal and scope

The goal of the LCA was to establish a baseline for the existing PSC prototype of the PerTPV partners, identifying which components and processes have the highest potential impact so they can be addressed during the project. The LCA considered the life cycle phases from cradle to cradle (i.e. from resource extraction to recycling, see Figure 1), but for the final phase the focus was laid on the recycling of the Pb containing perovskite layer (lead iodide, PbI_2) only, excluding the impact of recycling the remaining PSC. The proper handling of solar cell modules is mandatory [18], but concrete recycling schemes for PSC still have to be developed. D4.1 focus on the perovskite layer does only manifest a primary analysis and further impacts and gains from recycling the entire PSC have to be added in the future, once concrete recycling schemes have been developed. The data used to estimate the impact of recycling PbI_2 were derived from experiments performed at FHNW. As done in other studies (e.g. [19]), comparing the deposition methods focused on the electricity consumption needed for the process. The environmental impacts of PSCs were benchmarked with competing photovoltaic (PV) technologies. The single-crystalline silicon (scSi) PV module was chosen due to its high market share and a cadmium telluride (CdTe) PV module was used as a representative of thin-film PV technology. The impact of the deposition process for the benchmark technologies was calculated by subtracting the electricity and heat process inputs in the SimaPro/ecoinvent processes[3]. European electricity mixes were considered for the production of the benchmark technologies (see Chapter 5.2).

The functional unit was 1 m^2 of a PSC prototype for reasons discussed in Chapter 4. To account for the electricity production, the results were converted to the environmental impact related to the generation of 1 kWh of electricity. This considered the “average European irradiation weighted by the installed PV capacity per country in 2012” of (1’331 $kWh/m^2/yr_{[2012]}$) [20, p. 19] and module lifetimes of 1–30 years. The impact refers only to

[3] “Photovoltaic laminate, CdTe {DE}| market for | Cut-off, U” and “Photovoltaic laminate, single-Si wafer {GLO}| market for | Cut-off, U”

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the PSCs without the balance of system (BOS). The environmental impact from the transportation of materials was only included for the encapsulation, substrate and front glass (all with a material weight of more than 0.1 kg of that material per m^2).

5.2. Life Cycle Inventory

The material and energy flows related to processes and products, wastes and emissions were modelled using the data provided by the project partners (Table 1 and Table 2) and/or were derived from the literature (Table 3) and FHNW's own data collection. The basic data (e.g. chemicals, electricity, etc.) and the LCI of the benchmark technologies were taken directly from the ecoinvent 3.5 database [21]. The inventory for the recycling process (Table 4) was developed within the FHNW but does only present an initial protocol.

Table 3: Some of the material compositions of the layers were adapted from literature. The composition of the materials from Table 1 and Table 2 that are not listed here were either directly taken from ecoinvent or stemmed from FHNW's own datasets.

Literature reference	**Material**
Alberola-Borràs et al. 2018a [22]	TiO_2
Alberola-Borràs et al. 2018b [23]	Perovskite (CSEM, EPFL, OXFORD PV)
García-Valverde et al. 2010 [24]	ITO sputtering
Gong et al. 2015 [25]	Perovskite (VTT), Spiro-OMeTAD, Silver paste (Ag)
Itten and Stucki 2017 [26]	NiOx (Nickel oxide) sputtering
Tsang et al. 2015 [19]	FTO sputtering
	Deposition method
Alberola-Borràs et al. 2018c [27]	Spray coating or thermal evaporation
Espinosa et al. 2011 [28]	Lamination
García-Valverde et al. 2010 [24]	ITO sputtering
Itten and Stucki 2017 [26]	NiOx (Nickel oxide) sputtering
Tsang et al. 2015 [19]	FTO sputtering

Table 4: Processes and materials used for the calculation of the impact of recycling 1 m^2 of PSC assuming 100% PbI_2 recovery. Assumptions made by FHNW.

Step	**Material/ Process**	**Amount/Unit**
Shredding	Power; time	0.75 kW; 60 s (0.0125 kWh)
Leaching	HNO_3 (70%); H_2O	0.077 kg; 5.57 kg
Filtration	Ultra filtration	0.56 kW; 10 s (0.0016 kWh)
Concentration	Ion exchange resin	0.001 kg

5.3. Life Cycle Impact Assessment

For the LCIA GWP, CED and the ILCD indicators (Table 5) were chosen. The total value of the summed ILCD single score illustrates the performance of the PSCs. The ILCD indicators with a small impact were summed up as 'further indicators' - in total 16 were analysed. The ILCD indicators (unit milliPoints [mPt]) use EC-JRC Global normalization and equal weighting of all categories (EC-JRC Global, equal weighting). This was chosen in order to be able to compare the results to previous studies [29]. The purpose of the points is to facilitate the comparison of the relative differences.

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The normalization and weighting setting was chosen in order to enable the comparison of the results to previous studies [29]. This decision however led to a strong influence of the category 'Mineral, fossil & renewable resource depletion'. The ILCD results presented here therefore have to be interpreted carefully considering that no specific weighting imposes the assumption that all impact categories (e.g. climate change as well as eutrophication) are of equal importance.

The NREPBT was calculated according to Frischknecht et al. (2016) [30]. The project partners reported the PCE, whereas for the case of the two established PV technologies, the PCE were derived from the Fraunhofer Institute for Solar Energy Systems (ISE) Photovoltaics Report 2019 [31].

Table 5: Indicators used in this assessment and their purpose.

Indicator	Purpose
Cumulative Energy Demand (CED)	The CED [MJ] indicates the total amount of primary energy involved in the life cycle of a product.
Non-Renewable Energy Payback Time (NREPBT)	The EPBT is the time [months] required for the primary renewable and non-renewable CED/kWh to reach the same value as the reference electricity. The reference electricity refers to the efficiency of the European (RER) medium voltage electricity grid at the consumer end (10.9 MJ/kWh acc. to ecoinvent 3.5). The NREPBT approach uses only non-renewable energy for the calculation of the EPBT. Therefore, it avoids comparing the PV modules against a higher EPBT if in the future a higher portion of the energy grid run on renewable energy which has a lower EPBT.
Global Warming Potential (GWP)	The GWP [kg CO_2 equivalents] contains the Intergovernmental Panel on Climate Change (IPCC) climate change factors for a time span of 100 years.
International Reference Life Cycle Data (ILCD 2011)	ILCD [mPt] includes all categories of environmental impact which are equal weighted (normalization: EC-JRC Global): human toxicity (with and without cancer effects), freshwater ecotoxicity, ionising radiation human health (HH) and mineral, fossil and renewable resource depletion, climate change; ozone depletion; particulate matter; ionising radiation ecosystems (E; interim); photochemical ozone formation; acidification; terrestrial, marine and freshwater eutrophication; land use; water resource depletion. The categories with a low ILCD impact were summarized as 'further indicators' (i.e. climate change; ozone depletion; particulate matter; ionising radiation ecosystems (E; interim); photochemical ozone formation; acidification; terrestrial, marine and freshwater eutrophication; land use; water resource depletion).

ILCD, CED and GWP impacts were not only calculated per m^2 but also per kWh electricity generated according the following equation taken from Celik et al. [32]. That allows to compare PSCs not only to other PV technologies but also to other electricity generating technologies. However, the additional impact from the equipment needed for the transition from DC to AC electrical energy output (e.g. inverters) needs to be added separately.

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$$\text{Impact}_{kWh} = \frac{\text{Impact}_{m^2}}{I \times \eta \times PR \times LT} \quad [32]$$

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

$Impact_{m^2}$ = Impact per 1 m^2 module area

I = Irradiation constant [kWh/m^2/yr]; 1'331 kWh/m^2/yr$_{[2012]}$ [20]

η = module efficiency [%]; PCE of prototype A = 5%, B = 15%, C = 20%, D = 23% (PCE not yet confirmed) (see caption of Table 1 and Table 2; the current efficiencies as estimated by the project partners), for CdTe 16% and for scSi 17% [31]

PR = performance ratio of the module [%]; 75% [33]

LT = lifetime of the PV technology [yr]; 30 yr [33]

The NREPBT was calculated as following (adapted from [34]):

$$\text{EPBT [yr]} = \frac{CED\ production\ kWp}{Avoided\ RER\ electricity}$$

$$\text{CED production kWp} \left[\frac{\text{MJ}}{\text{kWp}}\right] = \frac{Module\ area}{power} \times CED\ production\ m^2$$

$$\frac{\text{Module area}}{\text{power}} \left[\frac{\text{m}^2}{\text{kWp}}\right] = \frac{1000}{Module\ power/_{area}} = \frac{1000}{Module\ efficiency\ [\%] \times 1000};$$

$$\text{CED production } m^2 \left[\frac{MJ}{m^2}\right] = \text{CED value per } m^2\text{, see Chapter Results}$$

$$\text{Avoided RER electricity} \left[\frac{MJ}{m^2}\right] = Annual\ electricity\ generation \times RER\ electricity\ efficency$$

$$\text{Annual electricity generation} \left[\frac{kWh}{kWp}/yr\right] = 1'331$$

$$\text{RER electricity efficiency} \left[\frac{CED\ tot\ MJ}{kWh\ RER}\right] = \text{CED of 1 kWh electricity RER medium voltage } = 10.87$$

6. Results and discussion

Initially the impacts from the ILCD, CED and GWP analysis were calculated per m^2 for all four prototype cells and compared with two established PV cells (see Chapter 6.1). The exact numbers of the results can be found in the tables (Table 7 to Table 15) at the end of the Annex. Next, ILCD, CED and GWP were calculated per kWh and the GWP per kWh as function of the lifetime of the PCE was shown (see Chapter 6.2). Then, the NREPBTs were calculated (see Chapter 6.2). Finally, the ILCD, CED and GWP impacts of recycling were shown (Chapter 6.3). The impacts of the deposition processes (see Table 1, Table 2 and Table 3) were included into each layer.

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6.1. Production impacts of the prototypes

Figure 2 shows for a prototype with a high PCE (i.e. porotype C with 20% and prototype D with 23%) that in order to minimize the environmental impact measured with the ILCD indicators the tandem PSC (prototype D, 28.6 mPt/m^2) should be favoured over the single cell (prototype C, 145.9 mPt/m^2). Prototype D's main impact stem from the indicator category 'mineral, fossil & renewable resource depletion' with 18.3 mPt/m^2 (64% of the impact) compared to 55.4 mPt/m^2 (38% of the impact) coming from 'human toxicity non-cancer effects' for prototype C. The prototypes with lower PCE (i.e. prototype A with 5% and prototype B with 15%) do have 57% (prototype A) and 35% (prototype B) lower ILCD impacts. Dividing the mPt/m^2 through the PCE shows that both prototypes D (1.243) and B (1.247) show a favoured ILCD-PCE ratio compared to prototypes A (2.440), C (7.295) and benchmarks CdTe (1.719) and scSi (4.024).

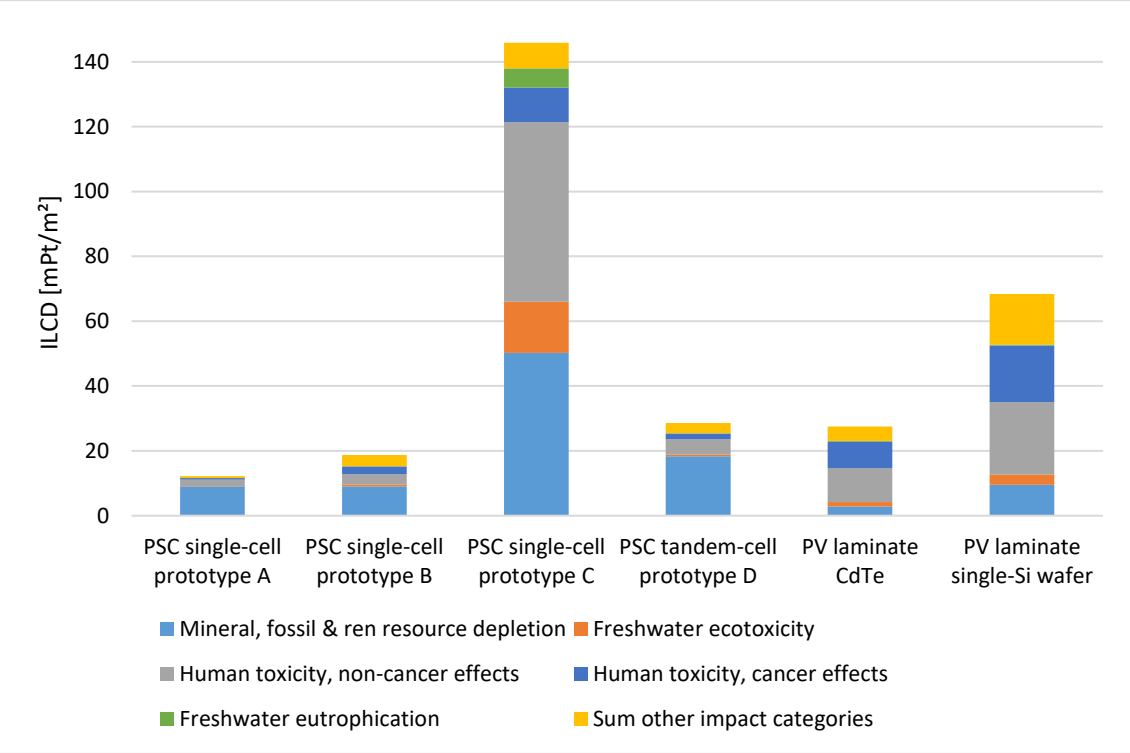

Figure 2: Comparison of the five ILCD indicators with the biggest impact (see legend; in total 16 were analysed) of the four perovskite solar cell prototypes and two benchmark PV technologies. Given are the total milliPoints per square meter from the ILCD single score method (the points purpose being the facilitation of seeing the relative differences).

Two network diagrams in the Annex (A1 and A3) show that the main contributing materials to the impacts of prototype C are the use of Au as top contact and to the impacts of prototype D ITO as bottom contact and hole transporter. In an additional stacked columns chart in the Annex (A5) the impact is divided between what comes from the production of the materials and the deposition methods. Almost all of the environmental impact measured by the ILCD categories comes from the associated emissions of the production of the materials used.

Figure 3 shows the CED and GWP per m^2 of produced PSC. Per m^2 only prototype C (1'640 MJ/m^2) has a higher CED value than the benchmark CdTe PV module (1'125

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MJ/m^2), prototype B's being slightly lower (1'087 MJ/ m^2). The main energy demand of single-crystalline silicon (scSi) PV modules stems from the Czochralski process to produce single crystal silicon (Annex A2). The materials' contribution tends to have a slightly higher impact than the deposition method. The GWP shows a similar pattern as the CED.

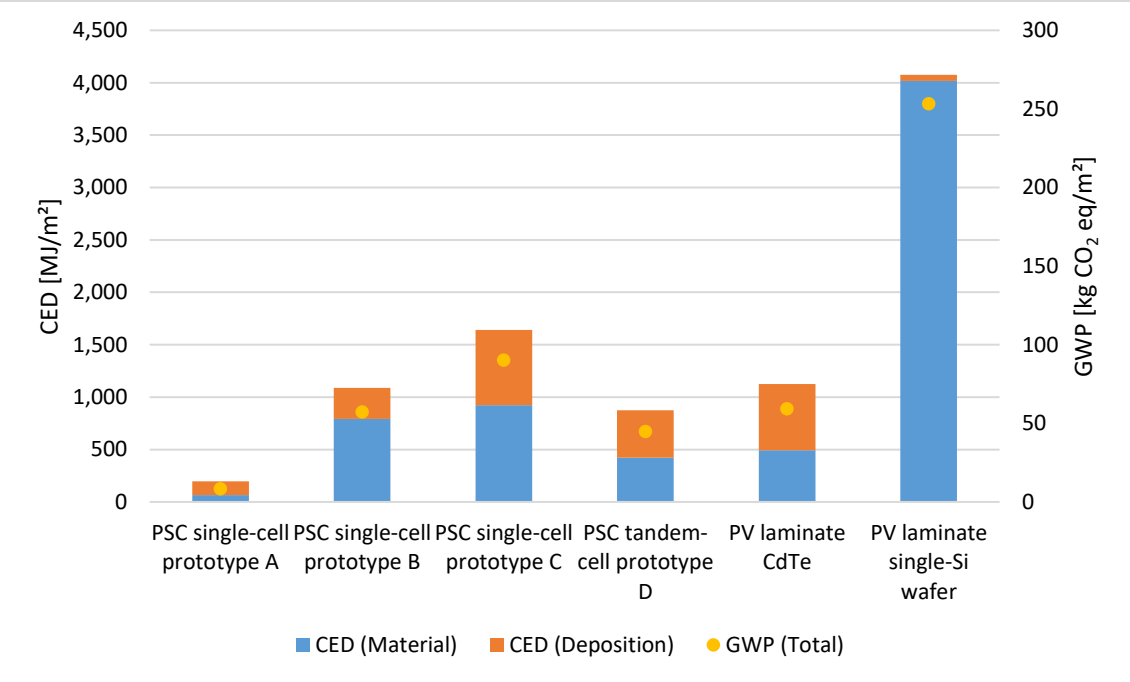

Figure 3: Comparison of the CED and GWP (yellow dot) for each of the four prototypes and two benchmark solar cells used on the market per m^2 module produced. The CED is divided into the contributions from the materials used (blue) and the deposition methods (orange).

Comparison to Maranghi et al. [29] (Supporting Information, Table 1, multiply by 0.0001 to convert MJ/cm^2 to MJ/m^2) shows that the results presented lie within the range of previous studies.

In one of these previous studies, Gong et al. (2015) arrived at similar results as those for the single-junction PSCs from this project: for the TiO_2 perovskite module (see Table 6), they found slightly more than 20 kg CO_2 eq/m^2 and 450 MJ/m^2, and for the zinc oxide (ZnO) perovskite module, they found slightly less than 20 kg CO_2 eq/m^2 and 400 MJ/m^2 [25]. The main impacts were derived from the ITO glass and silver paste. Celik et al. (2016) compared vacuum- and solution-based devices with HTL-free devices; they calculated 821, 665 and 504 MJ/m^2, respectively [32].

Table 6: Material compositions of previously studied PSCs and their literature reference.

Previously studied PSC	**Materials used in those studies**
TiO_2 module [25]	FTO glass, TiO_2, Au
ZnO module [25]	ITO glass, ZnO, Ag
SnO_2 module [32]	FTO, SnO_2, $MAPbI_3$, CuSCN, MoO_x/Al, C-Paste
SnO_2 module [26]	ITO, SnO_2, $MAPbI_3$, NiO, Ag

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6.2. Electricity generation

The impact of the electricity per m^2 PSC is measured with ILCD, CED, GWP and NREPBT at a solar irradiation of 1'331 kWh/m^2/$yr_{[2012]}$, assumed lifetimes of 30 years, current PCEs as mentioned in chapter 5.3 and an assumed performance ratio of 75%. As mentioned in chapter 5.1, the impact from the BOS is not considered.

Figure 4 compares the ILCD impact for the production of 1 kWh electricity (note that the ILCD Pt are given in µPt compared to mPt in Chapter 6.1). The main difference to Figure 2 can be seen for prototype A which has a low assumed efficiency and prototype D that has a high assumed efficiency. Both prototypes B and D show a lower ILCD impact than CdTe PV modules (both prototypes 4.2 µPt/kWh compared to 5.7 µPt/kWh).

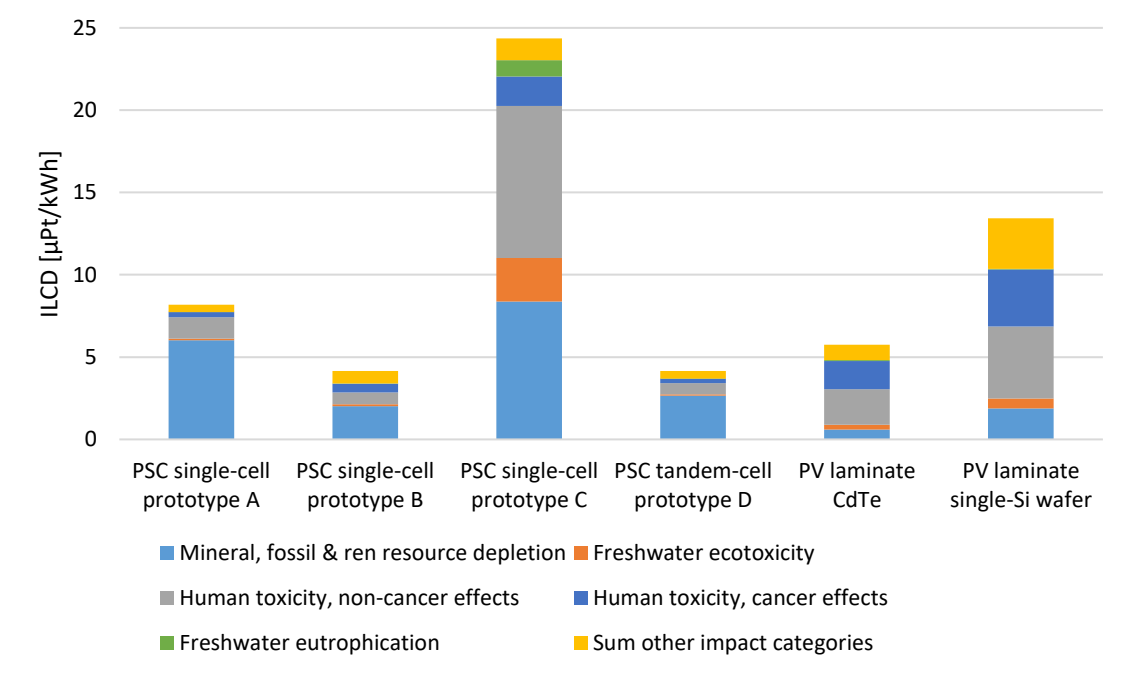

Figure 4: Comparison of the five ILCD indicators per kilowatt-hours. Solar irradiation: 1'331 kWh/m^2/$yr_{[2012]}$; lifetime: 30 years.

Figure 5 shows the CED and GWP per kWh electricity produced. The energy demand for the generation of 1 kWh is higher for the prototypes B and C (0.24 MJ/kWh for B and 0.27 MJ/kWh for C compared to 0.23 MJ/kWh for CdTe) while prototypes A and D are produced with less energy intensive materials and deposition methods.

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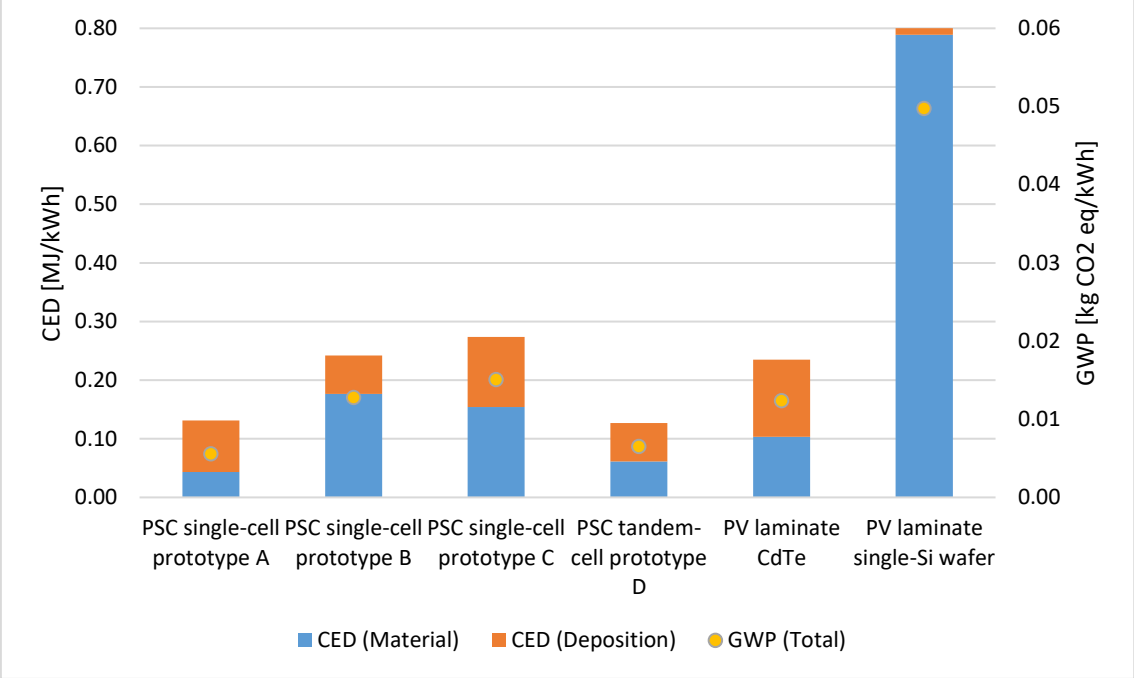

Figure 5: Comparison of the CED and GWP (yellow dot) per kWh electricity generated. The CED is divided into the contributions from the materials used (blue) and the deposition methods (orange). Solar irradiation: 1'331 kWh/m^2/yr$_{[2012]}$; lifetime: 30 years.

The impact of the electricity generation is also analysed in terms of GWP [kg CO_2 eq / kWh] over the lifetime. Apart from the current PCEs (between 5–23%) the project-targeted (25–30%) PCEs are also considered (Figure 6, dotted lines). In grey and black are the benchmark technologies.

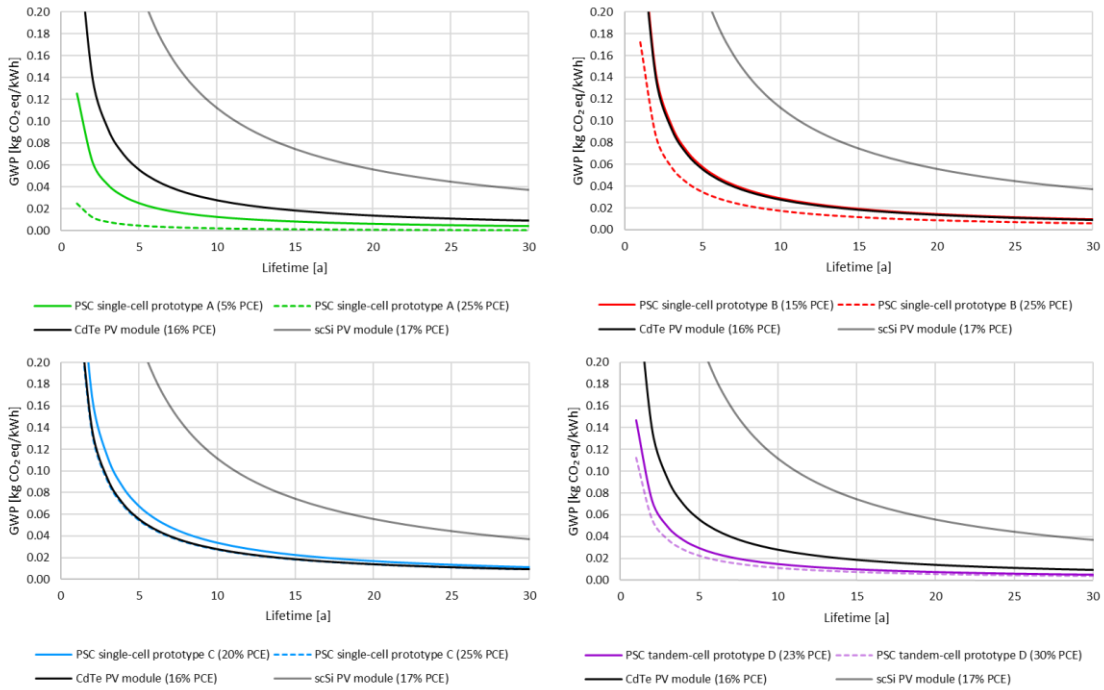

Figure 6: GWP of the four prototypes (prototype A in green, prototype B in red, prototype C in blue and prototype D in violet) for current (continuous line) and targeted (dotted) power conversion efficiencies and

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the two benchmark technologies (cadmium telluride PV in black and single-crystalline silicon PV in grey) in [kg CO_2 eq/kWh] as function of the lifetime [a]. Solar irradiation: 1'331 kWh/m^2/yr$_{[2012]}$.

One should note that this initial study only considers encapsulated cells but does not include the BOS. The impact of these compounds is excluded for the prototypes and benchmark technologies, but they can be expected to be similar for all PV technologies.

The study demonstrates that the longer the lifetime of a solar cell, the lower the specific environmental impact of the produced electricity (Figure 6), with the main reduction of the GWP impact achieved in the initial years of electricity production. For example, after a lifetime of 7 years, the prototype A (PCE 5%) has its GWP impact reduced to below 0.02 kg CO_2 eq/kWh[4]. Prototypes D and C with PCEs of (23% and 20%) and a similar GWP impact of the production require 8 and 9 years to achieve a similar value. Prototype B would reach this target at 15 years. Using similar amounts of materials but achieving the target PCE of 25% would lead to a drastic drop for prototype B to under 9 years to reach that mark.

The NREPBT (Figure 7) is also assessed. This refers to the efficiency of the European medium voltage electricity grid (10.87 MJ/kWh, according to ecoinvent 3.5). The NREPBT from current estimated PCE to targeted PCE would drop the most for prototype A from 3.3 to just 0.7 months (~20 days), then prototype B from 6 to 3.6 months and prototype C from 6.8 to 5.4 months. Prototype D, the tandem PSC would improve the NREPBT from 3.2 to 2.4 months. As a comparison, the NREPBT of CdTe PV modules is 3.3 months while that of single-crystalline silicon modules is 17.6 months.

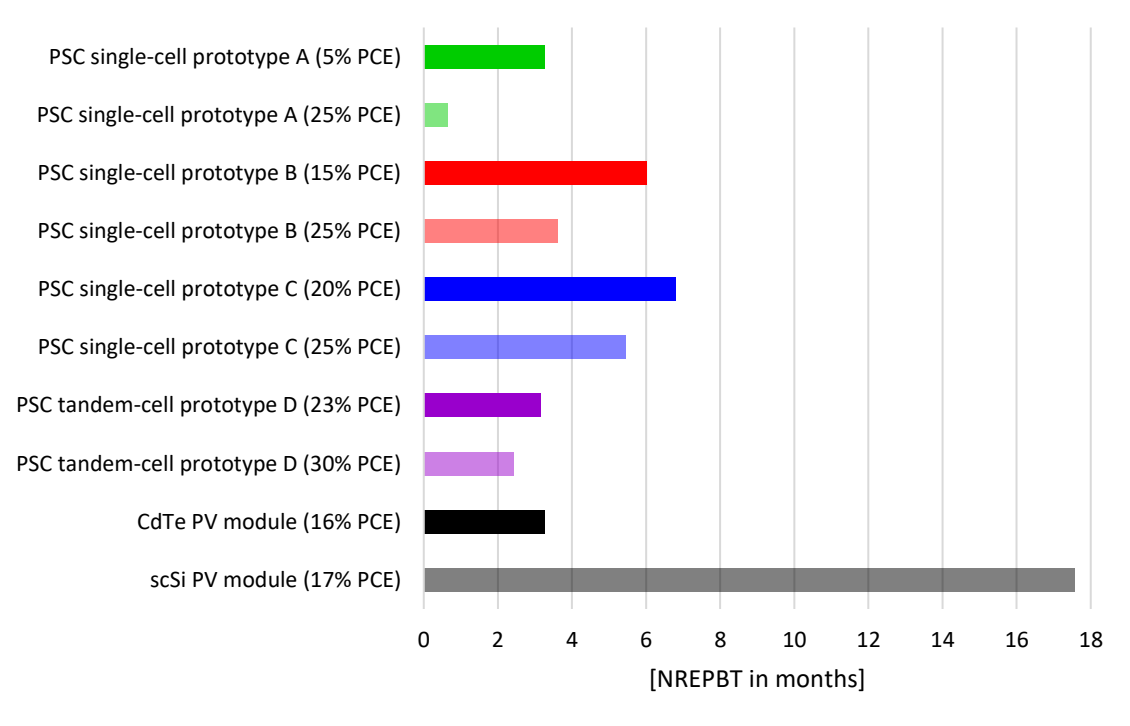

Figure 7: Non-renewable energy payback time in months of the four perovskite solar cell prototypes for current and targeted power conversion efficiencies and the two benchmark technologies.

[4] 20 g CO_2 eq/kWh is an arbitrary point of reference in order to facilitate comparison between prototypes

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Gong et al. (2015) calculated 2.6 months for their ZnO perovskite module, whereas their benchmark single-crystalline silicon module reached approximately 28 months [25]. However, Itten and Stucki (2017, see Table 6) calculated 18 months as the NREPBT for their perovskite solar cell module [26].

6.3. Recycling of PbI_2

PSCs will and must be recycled, as they contain valuable (e.g. Ag, Au, In) and harmful (e.g. Pb) materials. In this LCA, the focus was set on recycling the Pb in the form of PbI_2 showcasing one possible recycling scenario but thus omitting additional benefits from the recycling of valuable materials. A comparison was made between the impact caused by the recycling process itself (shredding, leaching, filtration and concentration) recovering 1 g of PbI_2 ("Impact Recycling PbI_2") and the impact caused by the primary raw material production of 1 g of PbI_2 used per 1 m^2 which can be substituted through the perovskite layer recycling ("Avoided Impact"). The ILCD impact of the investigated early stage PbI_2 recovery from the perovskite layer recycling for PSCs seems based on this analysis slightly higher (1.3x) compared with the impacts caused by the current primary raw material production of PbI_2 (Figure 8). The remaining overall impact caused by the analysed recycling scheme was visualized with a black arrow. The CED (7%) and GWP (4%) impacts cannot be offset significantly (Figure 9). Almost all impact from recycling of PbI_2 stems from the use of nitric acid during the leaching process (see Annex A4).

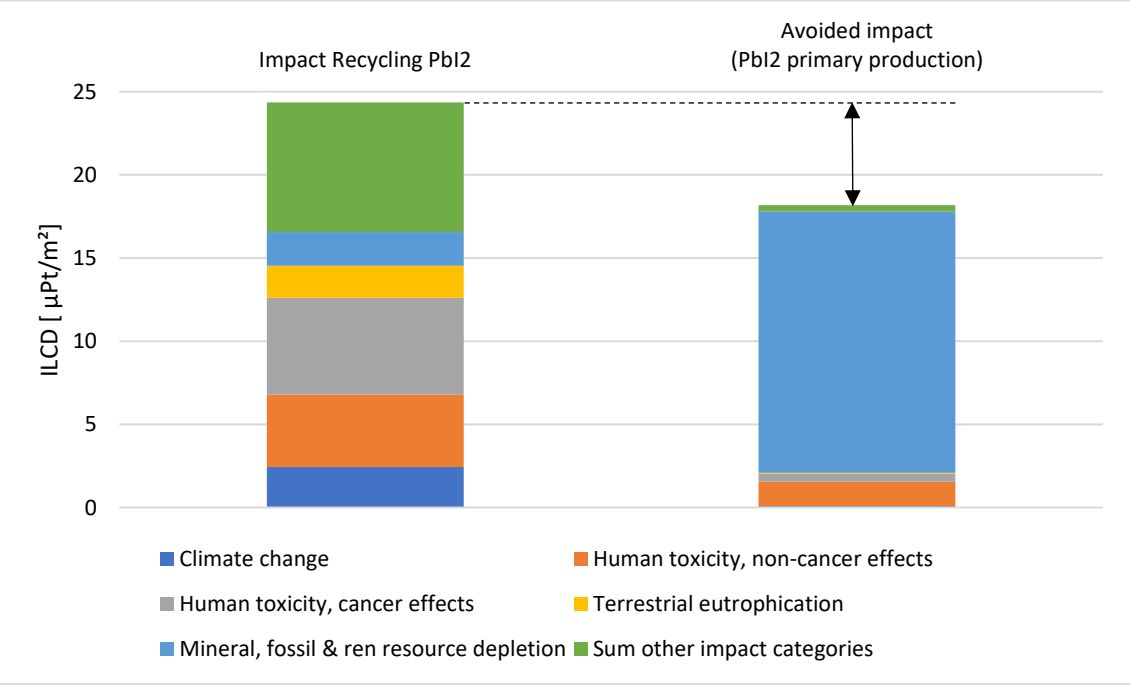

Figure 8: Comparison of the of the five ILCD indicators with the biggest impact ([µPt] see legend; in total 16 were analysed) caused through the recycling process itself compared to the avoided impact due to eliminating the need to produce the raw material PbI_2 used in the perovskite layer. The black arrow shows the remaining impact caused by the recycling of PbI_2.

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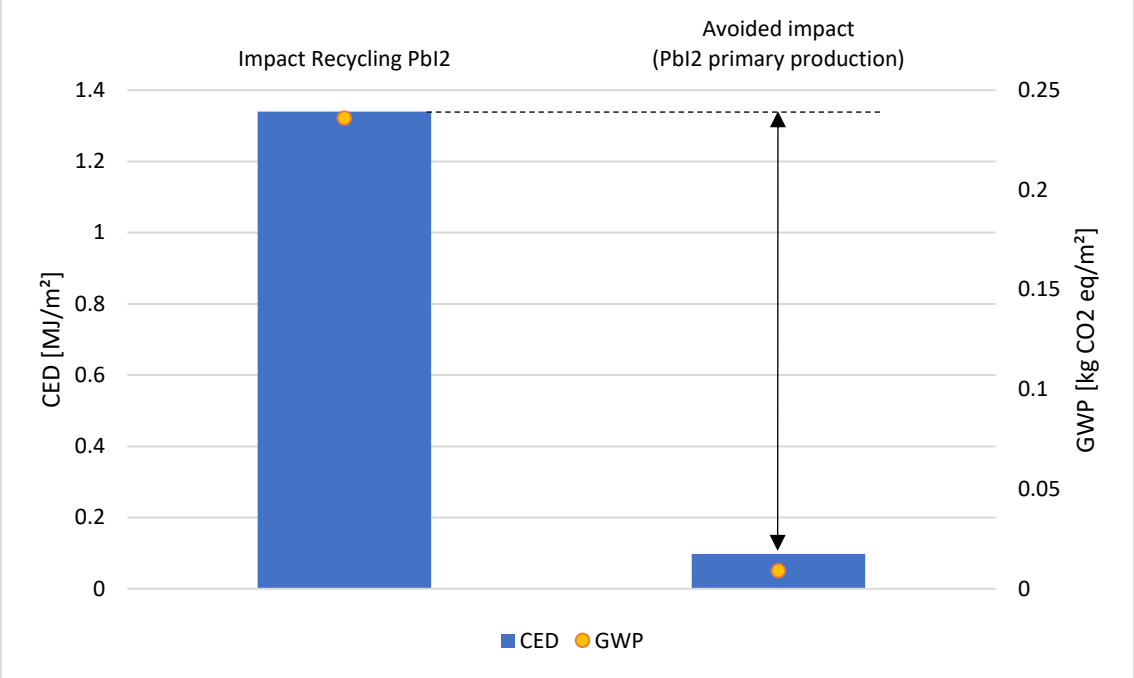

Figure 9: Comparison of the Cumulative Energy Demand (blue column) and Global Warming Potential (yellow dot) of the impact caused through the recycling process itself compared to the avoided impact (CED and GWP) due to eliminating the need to produce the raw material PbI_2 used in the perovskite layer. The black arrow shows the remaining impact caused by the recycling of PbI_2.

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7. Conclusion

This LCA revealed for the three areas of focus that:

a) in production:

- the main difference in the environmental impact from cradle to cradle between the PSCs stems from the different bottom and top contact materials;
- metal use in general and Au and indium (in the form of ITO) use in particular should be limited as much as possible due to the main impacts associated with these metals; developers should consider if it can be substituted by Ag, Al or carbon paste and FTO;
- deposition techniques should also be closely analysed, as some methods (e.g. spin-coating and anti-solvent deposition) may lead to large waste of materials [35] [36] and/or are not easily scalable;
- in general, the energy consumption of printing processes is lower than for sputter coating or thermal evaporation;

b) in electricity generation:

- the GWP impact per kWh decreases with lifespan and should therefore reach at least 10 years to be able to compete with the established PV-technologies;
- either the choice of materials and deposition methods (prototype A) or the PCE (prototype D) must be optimized to outperform the CdTe benchmark technology;

c) in recycling of PbI_2:

- the impact of the recycling process itself is partially mitigated through the avoidance of primary raw material production and avoided deposition.

Regarding concerns about Pb that is contained in the perovskite layer, calculations showed that all Pb from about 9.1–15.2 m^2 PSC modules would need to leach into 1 m^2 topsoil with an average reference concentration of ~15 ppm before even reaching a concentration of possible effects (e.g. guide values for Pb of 50 ppm). Furthermore, there are already two recent mitigation technologies available that could reduce that the leaching risk from broken panel significantly. According to Alberola-Borràs et al. (2018), other applications – such as lead-acid batteries, crystalline solar cell panels (during its production), and weather-proofing lead sheets on roofs – are estimated to create more significant Pb emissions during their life cycle than PSC [22]. However, the solubility and mobility (and potentially bioavailability) is much higher for PSC-borne Pb than metallic Pb used e.g. in soldering [37]. Therefore, safe design and implementing of a reverse logistic and recycling system for end of life PSC should be considered before commercialising PSC and will be addressed in a later stage of this WP. This is not only important from the point of environmental impacts, but even more so from the point of social acceptance. Finally, and most importantly, increased device lifetime and PCE leading to a higher total amount of generated electricity over the lifetime decrease environmental impacts and the costs of the generated electricity, i.e. per kWh.

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9. Annex

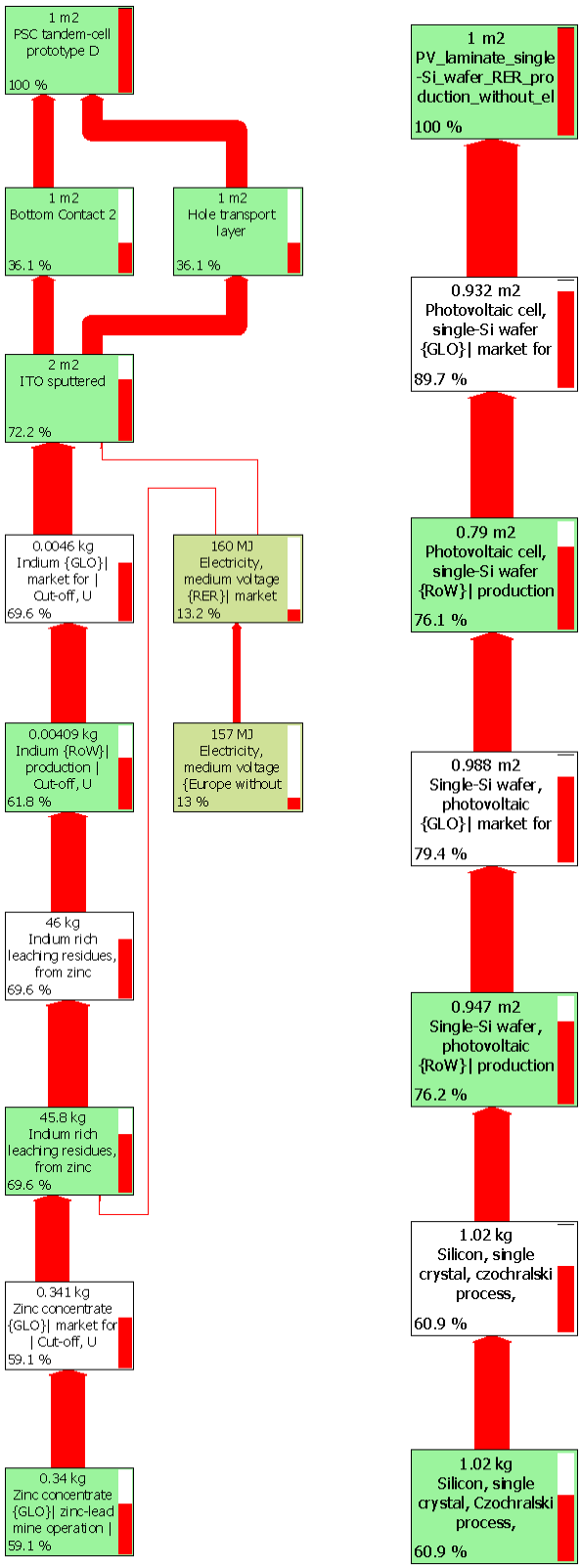

Figure A 1 (Left): Network illustration of ILCD results of prototype D. Main impact derives from the use of ITO as bottom contact and hole transporter material. (Right): Network illustration of CED results of single-crystalline silicon (scSi) PV module.

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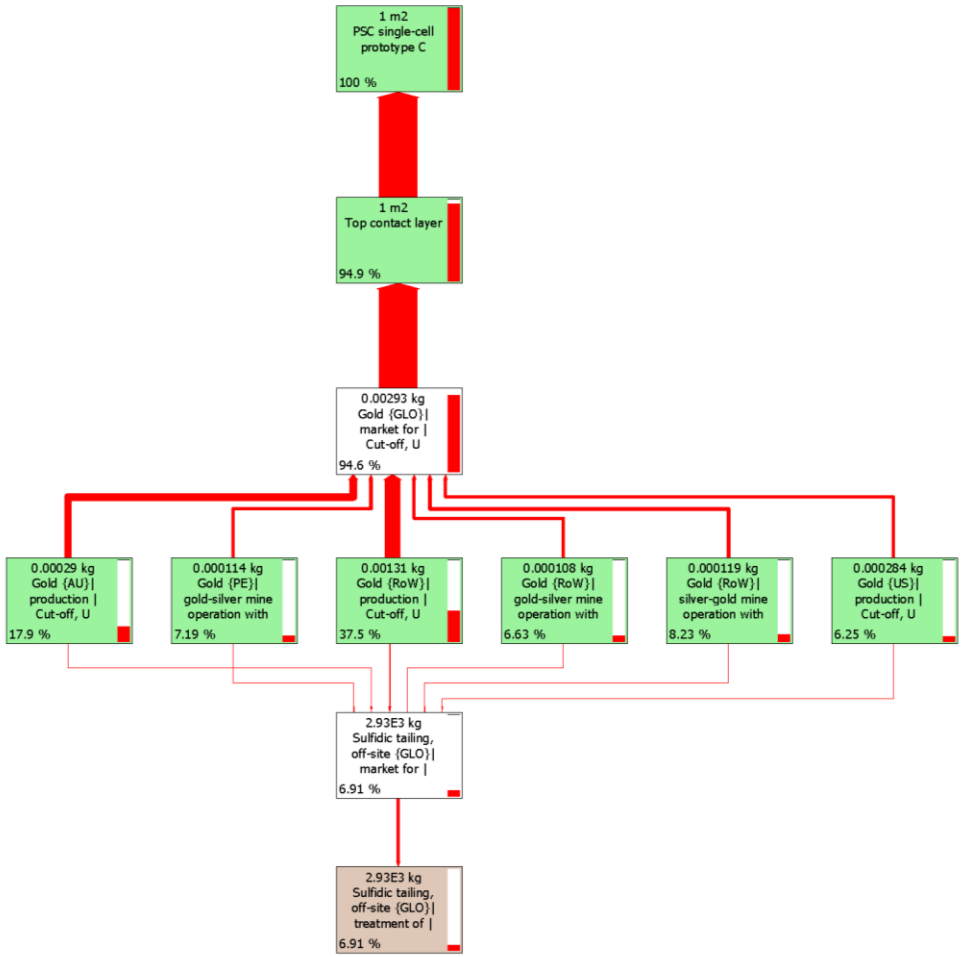

Figure A 2: Network illustration of ILCD results of prototype C. Main impact derives from the use of gold as top contact material.

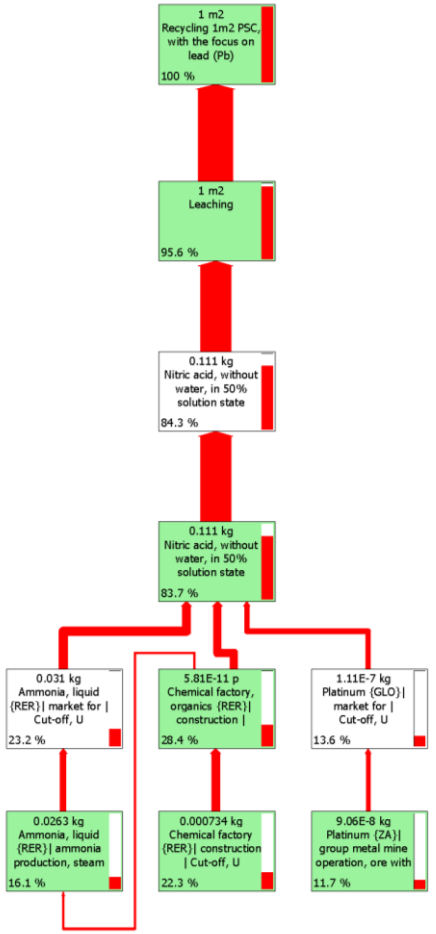

Figure A 3: Network illustration of ILCD results of recycling. Main impact derives from the use of nitric acid during the leaching process.

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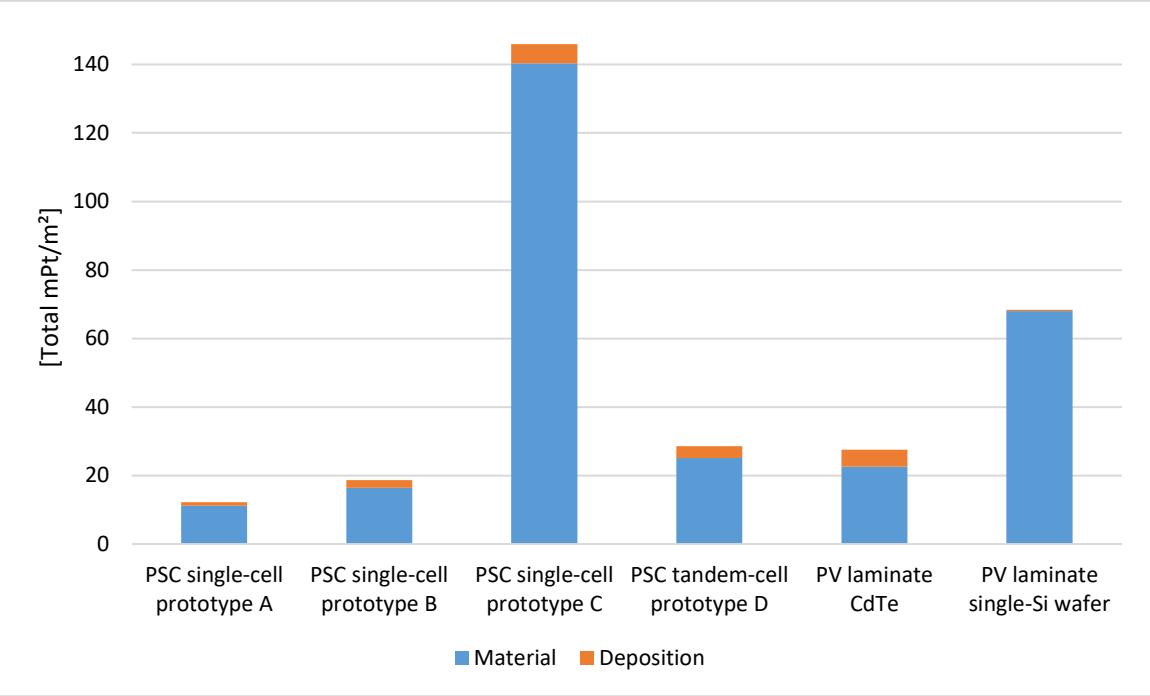

Figure A 4: This figure shows that for the ILCD impact categories the materials have a much higher impact than the deposition methods. This has to do with the weighting of the normalized results (SimaPro normalization and equal weighting of all categories was used).

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ILCD results[5]:

Table 7: ILCD result of prototypes and benchmark PV technologies in mPt per m².

[Total mPt/m²]	PSC single-cell prototype A	PSC single-cell prototype B	PSC single-cell prototype C	PSC tandem-cell prototype D	PV laminate CdTe	PV laminate single-Si wafer
Climate change	0.079	0.533	0.850	0.423	0.556	2.358
Ozone depletion	0.004	0.029	0.051	0.025	0.030	0.146
Human toxicity, non-cancer effects	1.937	3.243	55.368	4.716	10.405	22.284
Human toxicity, cancer effects	0.460	2.360	10.656	1.785	8.207	17.548
Particulate matter	0.062	0.473	0.965	0.359	0.628	2.911
Ionizing radiation HH	0.304	1.003	2.323	1.192	1.552	3.240
Ionizing radiation E (interim)	0.000	0.000	0.000	0.000	0.000	0.000
Photochemical ozone formation	0.037	0.327	0.996	0.243	0.341	1.300
Acidification	0.074	0.551	1.174	0.447	0.817	1.697
Terrestrial eutrophication	0.058	0.390	1.334	0.325	0.461	1.184
Freshwater eutrophication	0.016	0.033	5.926	0.046	0.216	0.276
Marine eutrophication	0.018	0.156	0.528	0.119	0.171	0.556
Freshwater ecotoxicity	0.148	0.565	15.839	0.516	1.386	3.113
Land use	0.000	0.001	0.003	0.001	0.002	0.003
Water resource depletion	0.025	-0.014	-0.256	0.053	-0.093	2.204
Mineral, fossil & ren resource depletion	9.024	9.034	50.170	18.348	2.847	9.568
Total	**12.246**	**18.683**	**145.927**	**28.596**	**27.526**	**68.389**

[5] All values given with three decimal places.

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Table 8: ILCD result of prototypes and benchmark PV technologies in µPt per kWh.

[Total µPt/kWh]	PSC single-cell prototype A	PSC single-cell prototype B	PSC single-cell prototype C	PSC tandem-cell prototype D	PV laminate CdTe	PV laminate single-Si wafer
Climate change	0.053	0.119	0.142	0.061	0.116	0.463
Ozone depletion	0.003	0.006	0.009	0.004	0.006	0.029
Human toxicity, non-cancer effects	1.293	0.722	9.244	0.685	2.171	4.377
Human toxicity, cancer effects	0.307	0.525	1.779	0.259	1.713	3.447
Particulate matter	0.041	0.105	0.161	0.052	0.131	0.572
Ionizing radiation HH	0.203	0.223	0.388	0.173	0.324	0.636
Ionizing radiation E (interim)	0.000	0.000	0.000	0.000	0.000	0.000
Photochemical ozone formation	0.025	0.073	0.166	0.035	0.071	0.255
Acidification	0.049	0.123	0.196	0.065	0.170	0.333
Terrestrial eutrophication	0.038	0.087	0.223	0.047	0.096	0.233
Freshwater eutrophication	0.011	0.007	0.989	0.007	0.045	0.054
Marine eutrophication	0.012	0.035	0.088	0.017	0.036	0.109
Freshwater ecotoxicity	0.099	0.126	2.644	0.075	0.289	0.612
Land use	0.000	0.000	0.001	0.000	0.000	0.001
Water resource depletion	0.017	-0.003	-0.043	0.008	-0.019	0.433
Mineral, fossil & ren resource depletion	6.027	2.011	8.376	2.664	0.594	1.879
Total	**8.178**	**4.159**	**24.364**	**4.152**	**5.745**	**13.433**

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Table 9: ILCD result of recycling and avoided primary production of PbI_2 in mPt per m^2.

[Total mPt/m²]	**Impact Recycling PbI2**	**Avoided impact (PbI2 primary production)**
Climate change	2.435	0.087
Ozone depletion	0.063	0.004
Human toxicity, non-cancer effects	4.358	1.459
Human toxicity, cancer effects	5.818	0.492
Particulate matter	1.043	0.089
Ionizing radiation HH	1.149	0.056
Ionizing radiation E (interim)	0.000	0.000
Photochemical ozone formation	0.758	0.033
Acidification	1.652	0.074
Terrestrial eutrophication	1.919	0.045
Freshwater eutrophication	0.074	0.006
Marine eutrophication	0.438	0.016
Freshwater ecotoxicity	1.846	0.131
Land use	0.001	0.000
Water resource depletion	0.778	-0.013
Mineral, fossil & ren resource depletion	2.020	15.701
Total	**24.353**	**18.181**

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CED results:

Table 10: CED result of prototypes and benchmark PV technologies in MJ per m^2.

[MJ/m²]	PSC single-cell prototype A	PSC single-cell prototype B	PSC single-cell prototype C	PSC tandem-cell prototype D	PV laminate CdTe	PV laminate single-Si wafer
Materials	65.140	791.782	922.426	422.011	493.994	4'017.913
Deposition methods	131.147	295.203	717.416	453.302	631.704	56.139
Total	196.287	1'086.985	1'639.842	875.313	1'125.698	4'074.052

Table 11: CED result of prototypes and benchmark PV technologies in MJ per kWh.

[MJ/kWh]	PSC single-cell prototype A	PSC single-cell prototype B	PSC single-cell prototype C	PSC tandem-cell prototype D	PV laminate CdTe	PV laminate single-Si wafer
Materials	0.044	0.176	0.154	0.061	0.103	0.789
Deposition methods	0.088	0.066	0.120	0.066	0.132	0.011
Total	0.131	0.242	0.274	0.127	0.235	0.800

Table 12: CED result of recycling and avoided primary production of PbI_2 in MJ per m^2.

[MJ/m²]	Impact Recycling PbI2	Avoided impact (PbI2 primary production)
Total	1.340	0.098

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GWP results:

Table 13: GWP result of prototypes and benchmark PV technologies in kg CO_2 eq per m^2.

[kg CO2 eq/m²]	PSC single-cell prototype A	PSC single-cell prototype B	PSC single-cell prototype C	PSC tandem-cell prototype D	PV laminate CdTe	PV laminate single-Si wafer
Total	8.367	57.379	90.354	44.959	59.291	253.376

Table 14: GWP result of prototypes and benchmark PV technologies in kg CO_2 eq per kWh.

[kg CO2 eq/kWh]	PSC single-cell prototype A	PSC single-cell prototype B	PSC single-cell prototype C	PSC tandem-cell prototype D	PV laminate CdTe	PV laminate single-Si wafer
Total	0.006	0.013	0.015	0.007	0.012	0.050

Table 15: GWP result of recycling and avoided primary production of PbI_2 in kg CO_2 eq per m^2.

[kg CO2 eq/m²]	Impact Recycling PbI2	Avoided impact (PbI2 primary production)
Total	0.236	0.009

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