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Citation

Costa, D., Martulli, A., Oever, A. van den, Müller, A., Degens, R., Rajgolapan, N., ... Vermang, B. (2025). Life cycle assessment of novel two-terminal (2T) and four-terminal (4T) perovskite/CIGS solar cells. *Progress In Photovoltaics*, 33(11), 1271-1289.
doi:10.1002/pip.70008

Version: Publisher's Version

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RESEARCH ARTICLE

Life Cycle Assessment of Novel Two-Terminal (2T) and Four-Terminal (4T) Perovskite/CIGS Solar Cells

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Received: 28 September 2024 | **Revised:** 23 March 2025 | **Accepted:** 12 June 2025

Funding: This study received funding from the European Union's Horizon 2020 research and innovation program under grant no. 850937 (PERCISTAND).

Keywords: climate change | copper indium gallium selenide (CIGS) | cumulative energy demand (CED) | energy payback time (EPBT) | perovskite | prospective life cycle assessment (pLCA) | tandem solar cells

ABSTRACT

Perovskite solar cells are an emerging photovoltaic technology promising higher efficiency than monocrystalline silicon solar cells. This study assesses the environmental impacts of novel two-terminal (2T) and four-terminal (4T) perovskite/copper indium gallium selenide (CIGS) tandem solar cells at technology readiness levels (TRL) 3 (2T configuration) and 4 (4T configuration). Based on the life cycle assessment (LCA) methodology, impacts are assessed from cradle to gate, focusing on climate change impacts per 1 m² of solar cell, 1 kW_p of module capacity, and the environmental 1 kWh of produced energy. Other relevant environmental impact categories, cumulative energy demand (CED) and energy payback time (EPBT), are also considered. The impacts of critical parameters are a sensitivity analysis. The LCA results indicate global warming impacts (GWI) of 49 and 50 kg CO₂eq/m², 170 and 174 kg CO₂eq/kWp for the 2T and 4T configurations, respectively. For both configurations, the GWI ranges from 3.2 to 5.6 gCO₂eq/kWh. Prospective investigated in climate change impact assessments suggest that manufacturing impacts may increase by 4% by 2050 without climate policies because of changes in the European energy mix. Conversely, stringent climate policies could reduce climate change impacts by 77%. The sensitivity analyses identify electricity use as the most critical factor for climate change impacts. CED values are 1219 and 1266 MJ/m² for the 2T and 4T configurations, respectively. For impacts per energy output, the CED is 4231 and 4380 MJ/kWp for the 2T and 4T configurations, respectively. For both configurations, the CED ranges from 80 to 140 kJ/kWh. The average EPBT values are 0.74 and 0.76 years for the 2T and 4T configurations, respectively.

1 | Introduction

Photovoltaic (PV) technologies are essential to achieving the energy transition and mitigating climate change, since they convert sunlight into electricity, reducing dependence on fossil fuels and lowering greenhouse gas emissions. The PV market is currently

dominated by single-junction devices, particularly single-junction silicon (Si) solar cells, which have matured to efficiencies approaching their theoretical maximum limit of around 29% [1]. Perovskite solar cells, part of the third generation of PV cell technologies, incorporate innovative, nonsilicon-based materials and novel device concepts [2]. Interest in perovskite

materials has grown since 2009, when they were shown to efficiently sensitize titanium dioxide (TiO_2) for visible-light conversion in photoelectrochemical cells [3, 4]. The term “perovskite” refers to any material with the crystal structure of calcium titanate (CaTiO_3), namely, ABX_3 [5, 6]. Perovskites are particularly appealing because of their cost-effectiveness in deposition processes and their efficiency compared with monocrystalline Si cells [7].

The use of perovskite on copper indium gallium selenide (CIGS) tandem cells is a promising thin-film PV technology, offering high potential for power conversion efficiency (PCE) at low cost, although it remains underexplored in the scientific literature [8, 9]. The PERCISTAND (Development of all thin-film PERovskite on CIS TANdem photovoltaics) project has developed novel two-terminal (2T) and four-terminal (4T) planar and inverted planar tandem perovskite/CIGS solar cells, achieving technology readiness level (TRL) 3 for the 2T configuration and TRL 4 for the 4T configuration, as a proof-of-concept to compete with conventional Si PV cells in terms of efficiency. Tandem cells consist of two solar cells that are mechanically coupled to maximize photon absorption [8]. By stacking two or more compatible solar cells in a tandem configuration, the solar spectrum can be utilized more efficiently, yielding PCEs that exceed the limits of single-junction devices [10].

The architecture of the developed tandem cells can be either monolithic (2T tandem), where the high bandgap cell is fabricated over the lower bandgap cell, or mechanically stacked (4T tandem), where the two subcells are optically coupled but electrically isolated. In the 2T setup, both layers are connected in series, sharing the same current, which reduces the number of electrical contacts but requires precise current matching to avoid limiting the performance of the entire stack. In contrast, the 4T configuration allows each subcell to operate independently at optimal efficiency, but it requires more electrical contacts [11]. In both configurations, light first interacts with the high-bandgap absorber, capturing higher energy photons, whereas the low-bandgap absorber captures lower energy photons.

Life Cycle Assessment (LCA) is a standardized methodology to assess the environmental impacts of a product or service from a life cycle perspective [12, 13]. LCA is recommended as the best framework to assess environmental impacts [14], and it has been widely used to analyze solar technologies [2, 15, 16]. Even though several studies have assessed the environmental impacts of perovskite solar cells, few have investigated the impact of tandem structures [15, 16]. For emerging technologies, such as perovskite-CIGS cells, prospective LCA can support the evaluation of the environmental impacts before the technology is fully implemented or developed. Prospecting the future impacts of technologies is important to provide informed decision-making guidance for the development of technologies. To date, only [17] have presented the prospective environmental impacts of perovskite solar tandem cells. However, [17] considered perovskite cells mounted on passivated emitter and rear contact (PERC) cells, primarily relying on secondary data from the scientific literature.

This study comprehensively assesses the environmental impacts of perovskite/CIGS modules using primary data through

the LCA methodology. It assesses the monolithic (2T) and the stacked (4T) perovskite on CIGS PV cells developed in the PERCISTAND project, using primary data obtained from PV manufacturers in the project consortium. Next to a conventional LCA, this study conducts a prospective assessment of the impacts on climate change until 2050 under different socioeconomic scenarios, to inform future environmental impacts of these emerging technologies. In addition to prospective LCA results, the cumulative energy demand (CED) and energy payback time (EPBT) of the two devices are also assessed.

2 | Methodology

2.1 | Goal and Scope Definition and System Description

The system boundaries (Figure 1) consider the solar cells from cradle to gate. The functional units are defined the assessment of as 1 m^2 for PV module cell manufacturing, 1 kWp of module cell capacity, and 1 kWh of delivered energy by the PV cell. A lifespan of 25 years of operation is considered representative of PV systems currently in use [18]. The calculation of the energy yield considers three distinct locations, as described in Section 2.3. The cells have a PCE of 28.8% for the 2T perovskite/CIGS configuration and 28.9% for the 4T perovskite/CIGS configuration.

Figure 2 shows the layout of the two devices and the materials used. Both devices are produced on glass substrates encapsulated with ethylene vinyl acetate (EVA) and glass. The detailed layouts of the devices, including functionality, main materials per layer, thickness, and deposition methods, are shown in Tables S1 and S2. The CIGS cell has zinc oxide (ZnO) and cadmium-sulfide (CdS) as a window layer and buffer layer, with the CIGS absorber deposited on the molybdenum (Mo) rear contact through coevaporation. The CIGS and perovskite ($\text{Cs}_{0.18}\text{FA}_{0.82}\text{PbI}_3$) cells are connected through a recombination junction layer of sputtered aluminum-doped ZnO (Al:ZnOx , AZO). The top cell uses sputtered nickel (II) oxide (NiOx) and a thin layer of 2PACz ((2-(9H-carbazol-9-yl)ethyl)phosphonic acid) as the hole transport layer, deposited with slot-die coating on indium tin oxide (ITO). The electron transport layer (ETL) is composed of tin (IV) oxide (SnO_2) deposited by atomic layer deposition (ALD) and C60 (buckminsterfullerene) deposited by physical vapor deposition (PVD). The top transparent conductive oxide (TCO) layer is comprised of ITO via sputtering.

In the 2T configuration, the two subcells are electrically connected in series through a recombination layer or tunnel junction. The 2T configuration requires only one transparent electrode, lowering the manufacturing costs due to reduced material usage and fewer deposition steps. The 4T configuration employs the same materials as the 2T (with different thicknesses), yet it includes a recombination junction and has an EVA spacer layer between the top and bottom cells. As the cells in the 4T configuration are independent, it requires two electrodes. In both configurations, the top cell is partially transparent to increase the quantity of energy photons captured by the adjacent absorber layer. This optimized utilization of both high- and low-bandgap light increases the overall efficiency of the module.

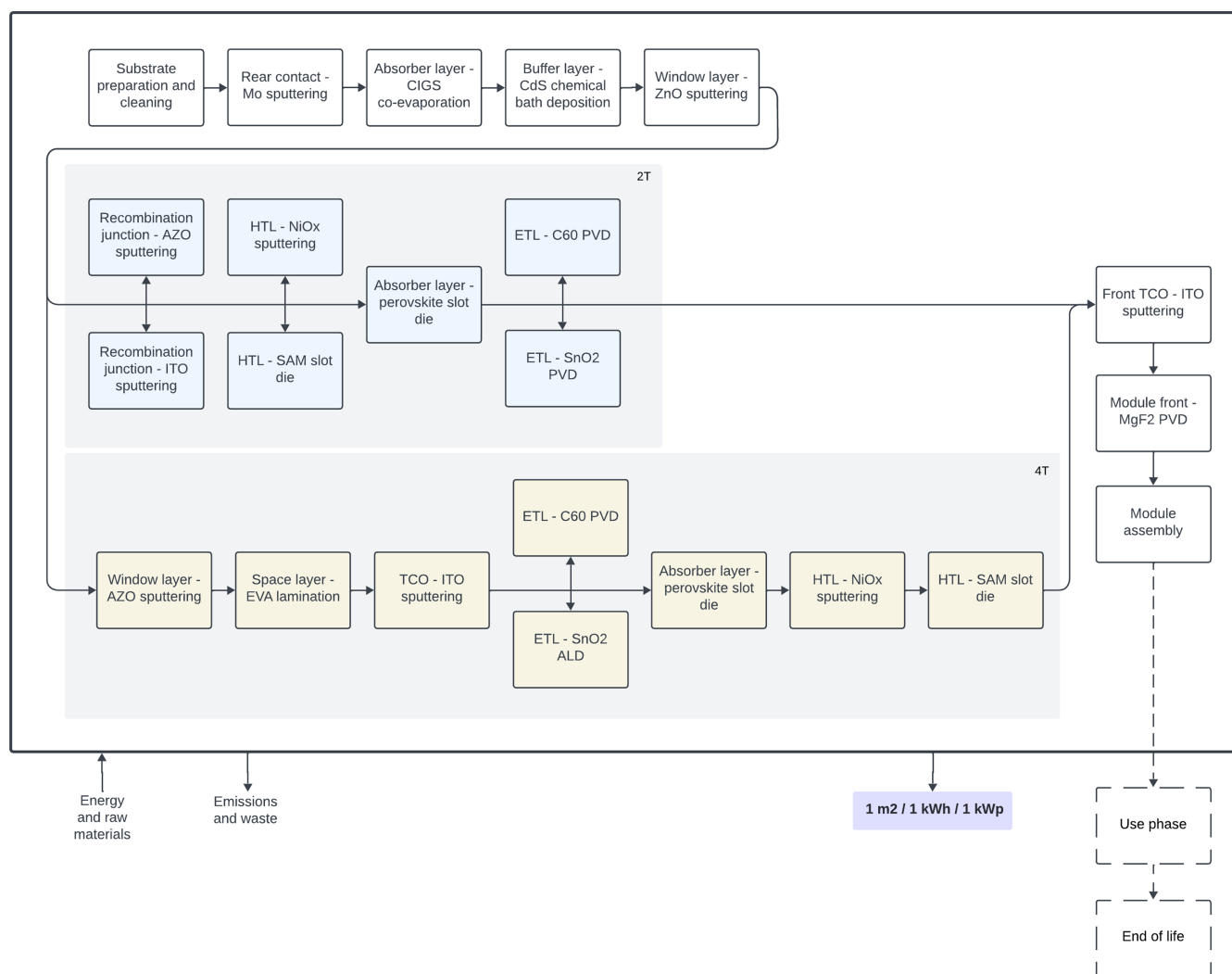


FIGURE 1 | System boundaries for the perovskite/CIGS tandem systems. Blue boxes represent the 2T configuration, and yellow boxes the 4T configuration. The purple box highlights the system output. Dashed lines represent life cycle stages that are not considered in this study. ALD—atomic layer deposition, AZO—aluminum-doped zinc oxide, C60—buckminsterfullerene, CdS—cadmium-sulfide, CIGS—copper indium gallium selenide, ETL—electron transport layer, EVA—ethylene vinyl acetate, HTL—hole transport layer, ITO—indium tin oxide, MgF₂—magnesium fluoride, Mo—molybdenum, NiOx—nickel oxide, PVD—physical vapor deposition, SAM—self-assembled monolayer, SnO₂—tin (IV) oxide, TCO—transparent conductive oxide, ZnO—zinc oxide.

2.2 | Life Cycle Inventory (LCI)

Foreground primary data to compile the LCI were obtained from the PV manufacturers and developers of CIGS and perovskite PV technologies in the PERCISTAND project. These data considered the layer properties of the cells and the electricity used for the different deposition methods. The source of background data is the ecoinvent database version 3.8 cutoff [19]. The detailed LCI data on the material used in each layer of the cell stack for both configurations are presented in Tables S3 and S4.

Additional modeling assumptions and data sources for the chemicals are given in Tables S5–S14. Waste from metals is assumed to be incinerated as hazardous waste, whereas glass is assumed to be treated and landfilled. For all layers, the market group “electricity, medium voltage” for Europe was considered representative of the European electricity mix, since the exact location for the manufacture of the solar cells in Europe

is unknown. Electricity consumption in different layers was estimated based on the nominal power of equipment and their operational time, obtained from PV cell manufacturers or the scientific literature [20–22].

2.3 | Energy Yield

Since perovskite top cells still lack the sufficient stability necessary to conduct real-world, long-term measurements, energy yield modeling is adopted to evaluate the performance of the devices under study in realistic outdoor conditions. Knowledge of the realistic energy yield of the solar cells under study is critical to assess the industrial relevance of perovskite/CIGS tandem PV and support their comparison with existing studies. To achieve that, an energy yield model was developed in PERCISTAND, resulting in an open-source software package called “EYCalc”¹ [23, 24]. Energy yield simulations in

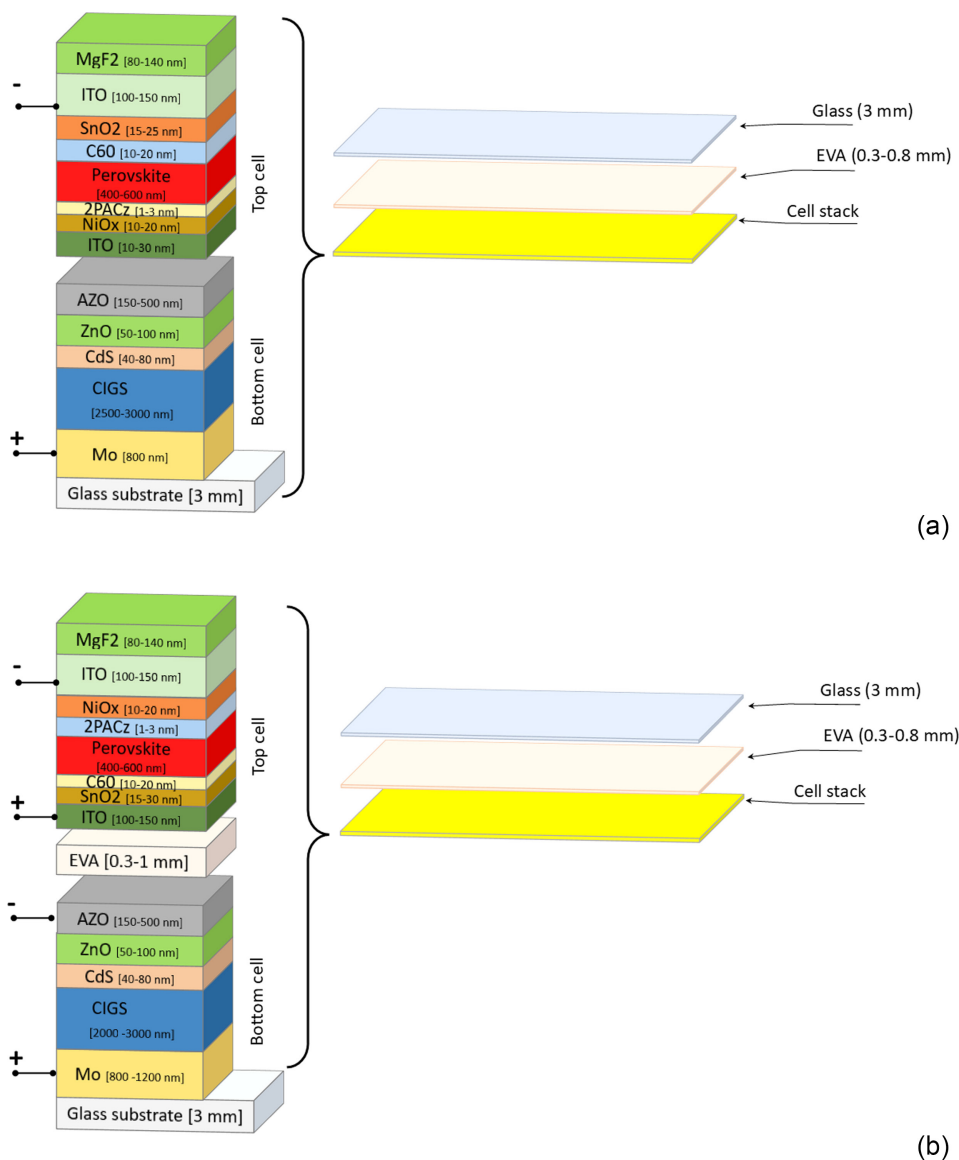


FIGURE 2 | Layout of the 2T (a) and 4T (b) configurations. 2PACz—(2-(9H-carbazol-9-yl)ethyl)phosphonic acid, AZO—aluminum-doped zinc oxide, C60—buckminsterfullerene, CIGS—copper indium gallium selenide, CdS—cadmium sulfide, EVA—ethylene vinyl acetate, ITO—indium tin oxide, MgF2—magnesium fluoride, Mo—molybdenum, NiOx—nickel oxide, SnO₂—tin (IV)oxide, ZnO—zinc oxide.

TABLE 1 | Energy yield for different climate conditions.

Device	PCE (%)	Energy yield (kWh/m ² year)		
		Oceanic	Tropical	Desert
2T perovskite/CIGS	28.8	353	487	610
4T perovskite/CIGS	28.9	361	492	624

Note: Degradation is not considered.
Abbreviation: PCE—power conversion efficiency.

realistic irradiation conditions were performed for the 2T and 4T configurations for three locations with different climatic conditions: (i) oceanic climate, (ii) tropical climate, and (iii) desert climate. The calculated energy yields of both devices are shown in Table 1.

The energy yield modeling platform “EYCalc” includes four individual modules: (i) irradiance module, (ii) optics module, (iii) energy yield core module, and (iv) electrics module [23, 24]. The irradiance module computes the irradiance of selected locations with a time resolution of 1 h. The irradiance is angularly and spectrally resolved, considering the meteorological conditions and the cloud cover of the selected locations. The optics module calculates the angularly and spectrally resolved absorptance of each layer of the solar cell stack. The irradiance and the absorptance are inputs to the energy yield core module, which computes the photogenerated current density of the absorbent materials (perovskite and CIGS) with a time resolution of 1 h. Based on the hourly resolved photocurrent density of each subcell and the parameterization of the equivalent circuit model representing the tandem solar cell, the electrics module calculates the maximum power point for both 2T and 4T configurations with an hourly resolution. The hourly resolved generated power feeds back

into the energy yield module that calculates the annual energy yield and further details. It should be noted that degradation was disregarded since comparative experimental data for the two configurations under consideration are yet missing.

2.4 | Life Cycle Impact Assessment (LCIA) and Prospective Assessment

LCIA was conducted based on the LCI presented in the [Supporting Information](#). The LCIA was conducted using the open-source software package Brightway [25] and its graphic user interface Activity Browser [26]. These tools support the use of graphic user interface, which comprise all scenario LCI databases in a single structure and facilitate the calculation across scenarios [27].

For the calculation of the LCIA, the [environmental footprint](#) method (EF 3.1) was used [28]. The EF method was chosen based on Recommendation 2021/2279 [29]. This study investigates the most relevant environmental impact categories of PV systems at the midpoint level [30, 31] in the context of the Product Environmental Footprint (PEF): acidification potential, abiotic depletion potential (mineral, fossil, and renewable resource depletion), climate change, freshwater ecotoxicity, human toxicity potential (cancer and noncancer effects), and particulate matter formation potential. However, the focus of the assessment is on climate change impacts.

For the prospective LCA assessment, the Python tool premise version 2.1 [32] was used to modify the ecoinvent 3.8 cutoff database for the years 2030, 2040, and 2050, to reflect projected energy policy trajectories, including emerging technologies, and to modify market shares and the efficiency of technologies based on scenarios from the Integrated Model to Assess the Global Environment (IMAGE) [33, 34]. For the prospective assessment, only impacts on climate change are discussed, since IMAGE explores pathways to reach overall climate targets, neglecting many mechanisms that are important to other impact categories. Consequently, the reliability of the premise-generated databases for other impact categories is low [35]. The IMAGE scenarios are defined based on representative concentration pathways (RCPs) and shared socioeconomic pathways (SSPs). RCPs describe different future climate scenarios with their greenhouse gas concentrations and respective radiative forcing values by the year 2100 [36, 37], whereas the SSPs provide narratives for future socioeconomic developments [36, 37].

The selected scenarios are SSP2-base (no climate policy), SSP2-RCP1.9 (which limits global temperature to 1.5°C above preindustrial levels by the end of the century, as outlined in the Paris Agreement), and SSP2-RCP2.6 (2°C target). The SSP2 scenarios are considered “middle-of-the-road” scenarios, which extrapolate the past and current global development into the future [38]. In the SSP2-base scenario, technological progress will be made in the base storyline, but no technological breakthroughs are foreseen. Since there are no restrictions on the use of fossil fuels, the increase in global energy demand after 2030 will lead to an increase in fossil

energy consumption [39]. In the SSP2-RCP2.6 storyline, ambitious climate change policies are implemented to reach the Paris Agreement goal of limiting global warming to below 2°C (1.6°C–1.8°C) [40]. Finally, in the SSP2-RCP1.9 scenario, the policy ambitions are even more stringent by limiting global warming to well below 2°C (1.2°C–1.4°C) [40].

2.5 | CED and EPBT

The CED method was used to evaluate all energy consumption associated with manufacturing [41]. The CED indicator quantifies the energy over the life cycle of the system, including direct energy consumption (e.g., electricity) and indirect energy contributions (i.e., embodied energy of materials). The CED was used to calculate the EPBT, which is defined as the time required for a renewable energy system to generate the same amount of energy that was used to produce the system. The EPBT was calculated according to Equation (1) [42], considering only the modules.

$$EPBT = \frac{CED}{E_{out}} \quad (1)$$

where

- EPBT = energy payback time (years)
- CED = cumulative energy demand (MJ)
- E_{out} = yearly energy output (MJ/year)

2.6 | Sensitivity Analysis

Sensitivity analysis was conducted to assess the robustness of the results and their sensitivity to the data and assumptions. First, a perturbation analysis was performed following Equation (2). The perturbation analysis aims to identify which material waste rates most affect all environmental categories in the different layers. The waste rates per layer are provided in the [Supporting Information](#). This analysis foresees changing the different waste rates by +10% and deriving the results' sensitivity by calculating a sensitivity factor (SF) and the relative changes in results. It supports assessing each change that leads to minor or major deviations of results.

$$SF = \frac{\frac{\Delta \text{result}}{\text{initial result}}}{\frac{\Delta \text{parameter}}{\text{initial parameter}}} \quad (2)$$

Because of the intensive use of electricity during cell manufacturing, a sensitivity analysis based on scenarios was conducted, changing the aggregate energy mix market to that of different European countries. The scenarios consider manufacturing in countries with current production of PV cells [43] and modules and the countries within the project consortium involved in the development of solar cells, namely, Belgium, France, Germany, and Switzerland. In addition, Estonia and Sweden, respectively, were considered representative cases of the most and the least carbon-intensive energy mixed in Europe [44].

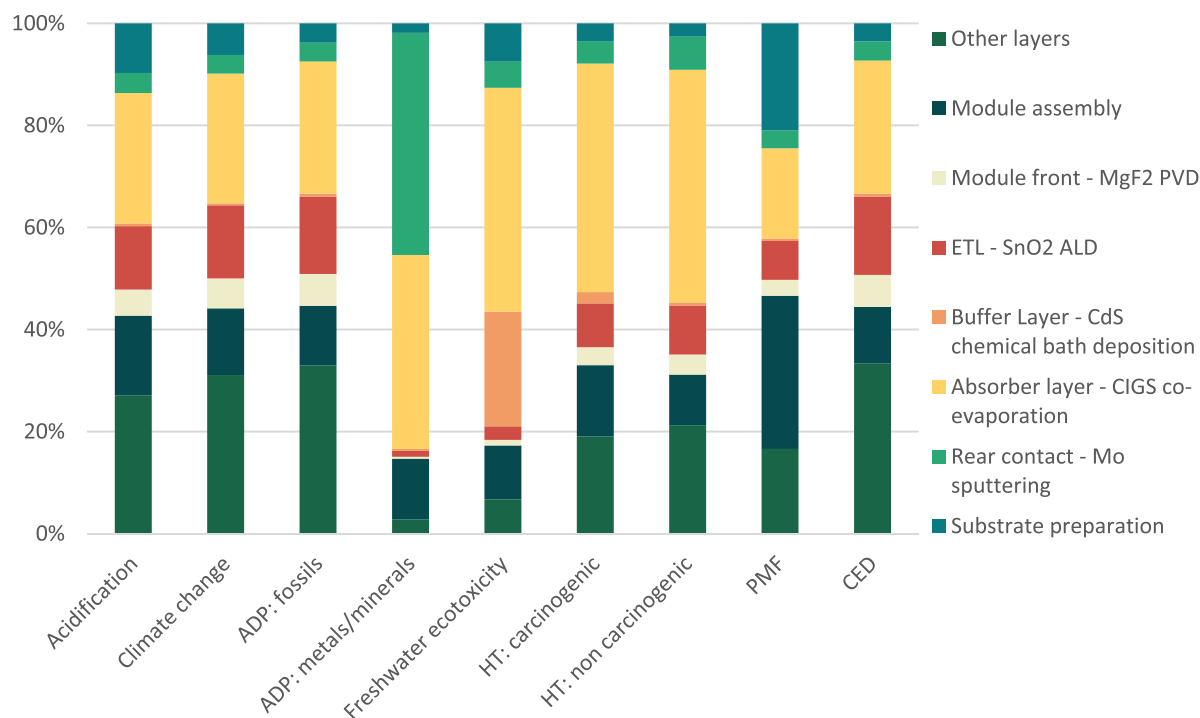


FIGURE 3 | Life cycle impact assessment results for the 2T configuration. ADP—abiotic depletion potential, ALD—atomic layer deposition, CdS—Cadmium Sulfide, CED—cumulative energy demand, CIGS—Copper indium gallium selenide, ETL—electron transport layer, HT—human toxicity, MgF₂—magnesium fluoride, Mo—Molybdenum, PMF—particulate matter formation, PVD—physical vapor deposition, SnO₂—tin (IV) oxide.

3 | Results and Discussion

This section presents the results of the LCA (Section 3.1), considering all environmental impact categories, as well as the CED and the EPBT (Section 3.2). Section 3.3 details the current and prospective impacts on climate change. These sections are followed by the sensitivity analysis (Section 3.4), a comparison with similar studies, and a discussion of the limitations of the study (Section 3.5).

3.1 | Environmental Impacts

Figures 3 and 4 show the share of environmental impacts of the most relevant manufacturing steps for the 4T per 1 m², respectively. The total absolute values for both configurations are shown in Table 2. The absolute values for all layers of the systems under assessment are presented in the Supporting Information (Tables S15 and S16). Figures 3 and 4 also present the results of the CED, which will be discussed in Section 3.2. The discussion of the impacts on climate change is presented in Section 3.3.

The differences in the results for the two configurations are marginal as the main differences between them are the inclusion of the spacer layer (EVA lamination) in the 4T configuration and differences in the material used for the front TCO (ITO sputtering). In both assessments, the impact of the laser scribing steps across the different environmental impact categories is aggregated to support a better analysis of the contribution of the different manufacturing steps.

For both cell configurations, the most critical steps are substrate preparation, manufacturing of rear contact (Mo sputtering), absorber layer (CIGS coevaporation), buffer layer (CdS chemical bath deposition), ETL (SnO₂ ALD), and module assembly. The manufacturing steps contribute more than 5% to at least one environmental impact category. The other manufacturing steps are grouped under “other layers”. For both configurations, the laser scribing contributes less than 1% to all environmental impact categories.

In both configurations, the absorber layer (CIGS coevaporation) is the manufacturing step contributing the most to all impact categories, except particulate matter formation and abiotic depletion of metals/minerals. Considering both configurations, the impacts of the absorber layer range from 17% in particulate matter formation to 45% in carcinogenic human toxicity. Module assembly is the largest contributor to particulate matter formation (30% in the 2T configuration and 29% in the 4T configuration). The rear contact (Mo sputtering) is the largest contributor to the abiotic depletion of metals and minerals (about 44% in both configurations).

For acidification, the total impact is 2% higher in the 4T configuration compared with the 2T. The main contributions to this impact category are the production of the absorber layer (25% and 26% contributions for the 2T and 4T configurations, respectively), followed by module assembly and the ETL (SnO₂ ALD) (15% and 12% contribution in both configurations). For both configurations, the impacts can be attributed to sulfur dioxide and nitrogen oxide emissions (about 71% and 25%, respectively). In both cases, for the process contributions, most of the impacts

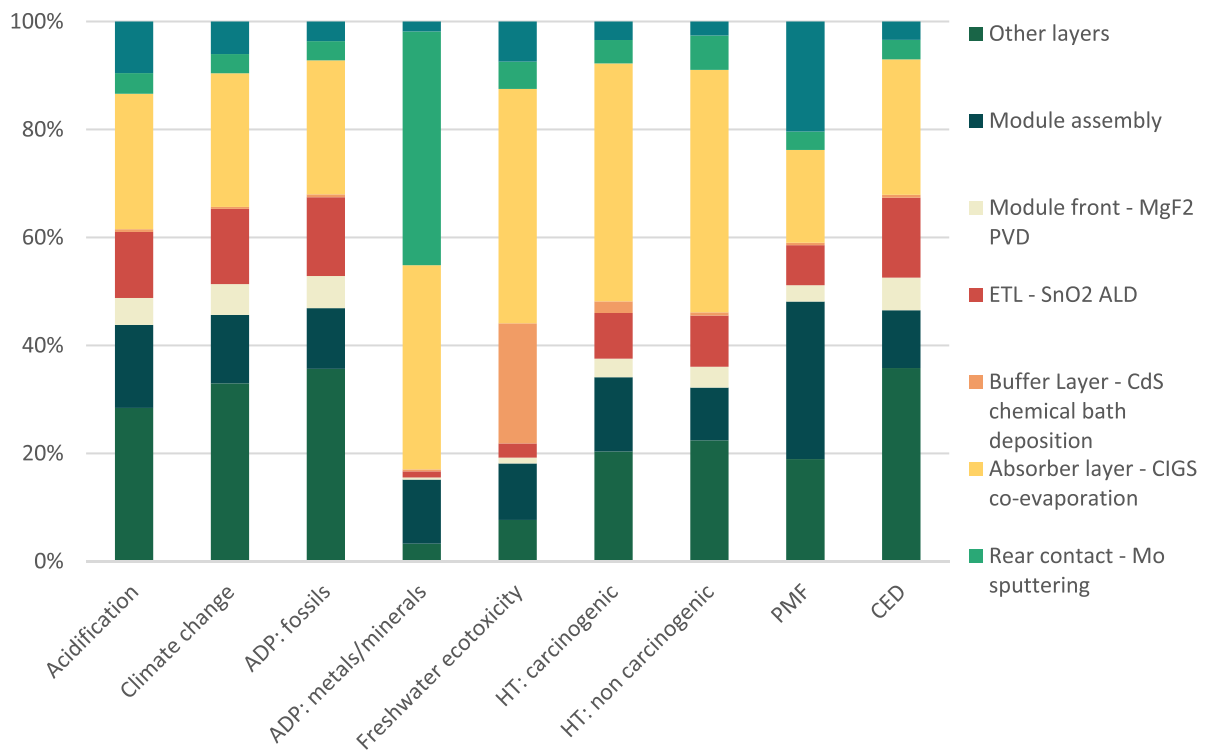


FIGURE 4 | Life cycle impact assessment results for the 4T-stack configuration. ADP—abiotic depletion potential, ALD—atomic layer deposition, CdS—cadmium sulfide, CED—cumulative energy demand, CIGS—copper indium gallium selenide, ETL—electron transport layer, HT—human toxicity, MgF2—magnesium fluoride, Mo—molybdenum, PMF—particulate matter formation, PVD—physical vapor deposition, SnO₂—tin (IV) oxide.

TABLE 2 | Absolute values per configuration and relative changes reported for 1 m² of PV module.

Impact category	2T configuration	4T configuration	Changes from 2T to 4T
Acidification (mol H + eq)	3.06E-01	3.11E-01	+2%
Climate change (kgCO ₂ eq)	4.90E+01	5.04E+01	+3%
ADP: fossils (MJ)	1.01E+03	1.05E+03	+4%
ADP: metals/minerals (kg Sbeq)	1.72E-03	1.73E-03	+1%
Freshwater ecotoxicity (CTUe)	3.53E+02	3.57E+02	+1%
Human toxicity: carcinogenic (CTUh)	2.54E-08	2.58E-08	+2%
Human toxicity: noncarcinogenic (CTUh)	7.36E-07	7.47E-07	+2%
Particulate matter formation (disease incidence)	1.48E-06	1.52E-06	+3%
Cumulative energy demand (MJ)	1.22E+03	1.27E+03	+4%

for this impact category derive from the consumption of electricity of fossil origin (about 40%), followed by heat and power cogeneration (16%) and flat glass production (around 13%).

The impact on abiotic depletion of fossil fuels is 4% larger for the 4T contribution than the 2T stack. For both, the manufacturing steps contributing the most are the absorber layer (CIGS co-evaporation) (26% and 25% for the 2T and the 4T configurations, respectively), followed by the ETL (SnO₂ ALD) (about 15% for both configurations) and module assembly (about 12% for both configurations). The mining and consumption of uranium contribute the most to effects in this impact category, mostly linked

to electricity generation. Nuclear plants generated around 22.8% of the total electricity produced in the European Union in 2023, explaining the contribution of uranium to this impact category [45]. Natural gas consumption is the second largest contributor to abiotic fossil fuel depletion, at about 22%.

The absolute contribution to the abiotic depletion of elements is nearly the same for the 2T and 4T configurations. The impacts are mostly explained by the manufacturing of the rear contact (Mo sputtering) and the absorber layer (CIGS coevaporation). In both cases, these steps account for 88% of the total contribution to this impact category. Similarly, for both configurations, the

impacts are mostly due to copper mining. This can be explained by the fact that almost all Mo production in the world originates from molybdenite, and approximately 50% of the world's Mo production comes from Cu (copper)–Mo ores as byproducts [46].

The impacts on freshwater ecotoxicity and human toxicity (carcinogenic and noncarcinogenic) are 1% higher in the 4T configuration. For freshwater toxicity, the impacts are mostly linked to the absorber layer (CIGS coevaporation) and the buffer layer (CdS chemical bath deposition), which contribute 44% and 23% of this impact category, respectively. The impacts on freshwater toxicity are mostly due to thiourea production (about 23% of total impacts) and the production of zinc (about 30%). Indium is a critical element recovered as a byproduct of zinc ores, and today, about 90% of indium is isolated as a byproduct of zinc mining [47, 48].

The 4T configuration has a 2% higher impact for human toxicity due to carcinogenic substances and a 1% higher impact for noncarcinogenic substances. The absorber layer (CIGS coevaporation) is the largest contributor to the potential impacts of both human toxicity impact categories. For carcinogenic human toxicity, this manufacturing step contributes 45% in the 2T configuration and 44% in the 4T configuration. For noncarcinogenic human toxicity, this manufacturing step contributes 46% in the 2T configuration and 45% in the 4T configuration. For carcinogenic human toxicity, the impacts are mostly explained by the use of indium, copper, and gallium, which are linked to elementary flow emissions of chromium VI, benzo(a)pyrene, arsenic, and formaldehyde. The impacts of noncarcinogenic human toxicity are mostly explained by the use of indium and copper, which are linked to elementary flow emissions of arsenic, mercury, and lead.

The particulate matter impacts are 3% higher in the 4T configuration. For both configurations, particulates < 2.5 μm and sulfur dioxide emissions are the largest contributors (53% and 30%, respectively). The manufacturing steps contributing the most to this environmental impact category are the same for

both configurations: absorber layer (26% and 25% of the total contribution for the 2T and 4T configurations, respectively) and module assembly (16% and 25% of the total contribution for the 2T and 4T configurations). For both configurations, the processes contributing to this environmental impact category are electricity generation from fossil fuels followed by flat glass production.

3.2 | CED and EPBT

The CED and the EPBT values are shown in Table 3. For the 2T and the 4T devices, the CED is mostly explained by the absorber layer (CIGS coevaporation), with 25% and 26% contributions, followed by the ETL (SnO_2 ALD), with 15% contribution in both configurations, and module assembly, with 11% contribution in both configurations (Figures 3 and 4). Concerning energy carriers, the CED is mostly explained by uranium consumption (about 33% in both configurations), followed by natural gas (about 18% in both configurations) and hard coal (about 12% in both configurations). About 33% of the total CED originates in Europe (including Switzerland) in the case of the 2T configuration and 34% in the 4T configuration. The average EPBT (for all irradiation conditions) is 0.74 years for the 2T configuration and 0.76 years for the 4T configuration. The EPBT is a measure of the performance of the technology/system, which quantifies how long it takes the system to recover all the energy that went into its manufacture. It can be used as a measure of the commercial viability of PV cell technologies [49].

3.3 | Current and Prospective Impacts on Climate Change

For the reference year 2021, using the original ecoinvent 3.8 database, the total contribution of the 2T to climate change is 49 kg $\text{CO}_2\text{eq}/\text{m}^2$, slightly lower than the 4T configuration, which has total emissions of 50 kg $\text{CO}_2\text{eq}/\text{m}^2$. Table 4

TABLE 3 | Cumulative energy demand (CED) and energy payback time (EPBT) of 2T and 4T perovskite solar tandem cells. CIGS—copper indium gallium selenide.

Device	CED (kJ/kWh)			EPBT (years)			CED (MJ/m ²)	CED (MJ/kWp)
	Oceanic	Tropical	Desert	Oceanic	Tropical	Desert		
2T Perovskite/CIGS	138	100	80	0.97	0.70	0.56	1219	4231
4T Perovskite/CIGS	140	102	81	0.98	0.72	0.57	1266	4380

TABLE 4 | Global warming impact (GWI) of 2T and 4T perovskite solar tandem cells per kWh in different solar insulation scenarios. CIGS—copper indium gallium selenide.

Device	GWI (kgCO ₂ eq/m ²)	GWI (gCO ₂ eq/kWh)			GWI (kgCO ₂ eq/kWp)
		Oceanic	Tropical	Desert	
2T Perovskite/CIGS	49	5.55	4.02	3.21	170
4T Perovskite/CIGS	50	5.58	4.09	3.23	174

shows the impacts on climate change considering the different solar insulations under assessment. Similar to the other environmental impact categories, the process contribution to impacts on climate change is mostly driven by electricity generation based on fossil fuels (hard coal and lignite), which contributes 47% of the impacts in the 2T configuration and 46% in the 4T configuration. This is followed by heat and power cogeneration, contributing to about 21% of the impacts in the 2T configuration and 19% in the 4T

configuration. For both configurations, the absorber layer is also the largest contributor (25%), ETL–SnO₂ sputtering (14%), module assembly (13%).

Concerning the prospective scenarios, the climate change impacts in 2050 in the SSP2-Base scenario increased by 4% compared with the original ecoinvent 3.8 database in the year 2021 for both 2T (Figure 5) and 4T (Figure 6). For the SSP2-RCP1.9 and SSP2-RCP2.6 scenarios, the reductions in climate

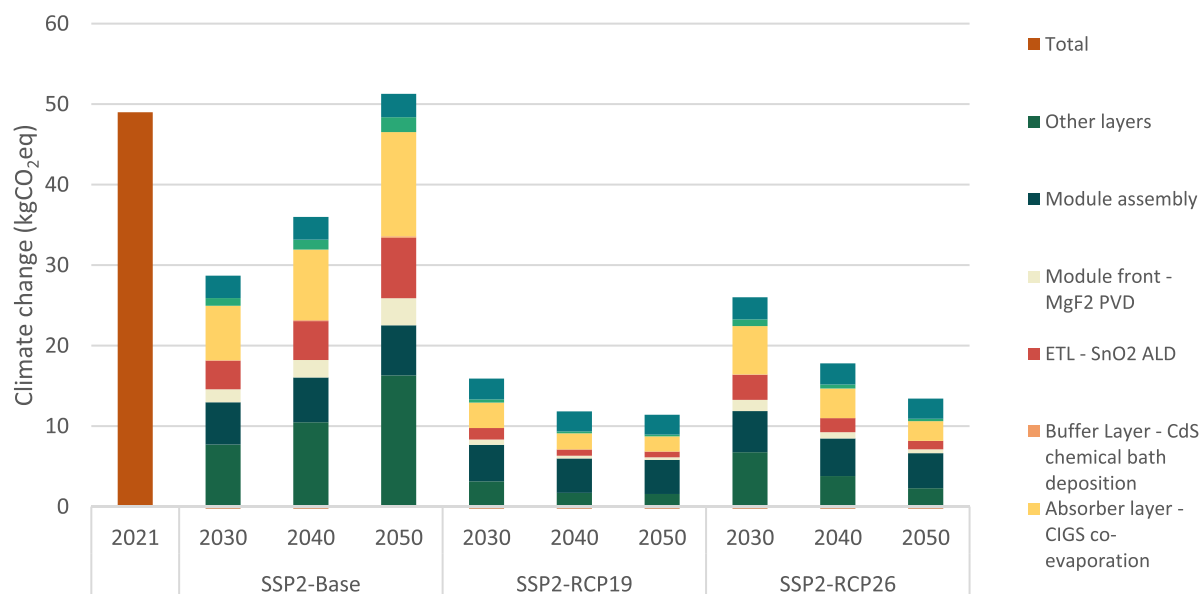


FIGURE 5 | Prospective impacts on climate change for the 2T configuration. ALD—atomic layer deposition, CdS—cadmium sulfide, CIGS—copper indium gallium selenide, ETL—electron transport layer, MgF₂—magnesium fluoride, PV—physical vapor deposition, RCP—representative concentration pathways, SnO₂—tin (IV) oxide, SSP—shared socioeconomic pathway RCP—representative concentration pathways, SSP2—shared socioeconomic pathway. Values for 2021 correspond to results from ecoinvent 3.8.

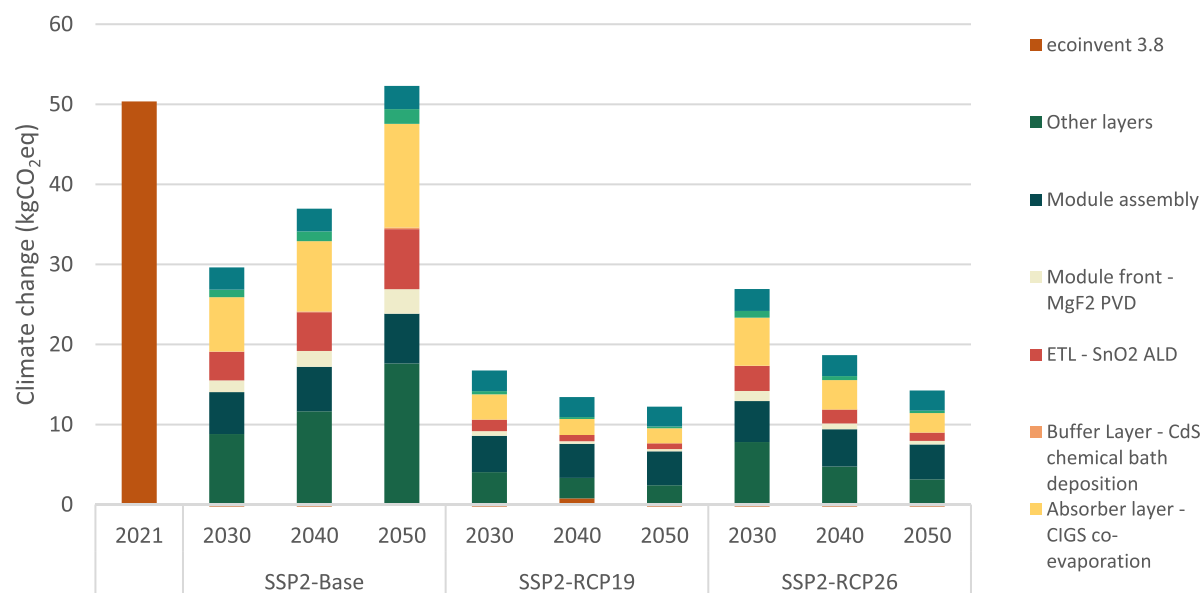


FIGURE 6 | Prospective impacts on climate change for the 4T configuration. ALD—atomic layer deposition, CdS—cadmium sulfide, CIGS—copper indium gallium selenide, ETL—electron transport layer, MgF₂—magnesium fluoride, PVD—physical vapor deposition, RCP—representative concentration pathways, SnO₂—tin (IV) oxide, SSP—shared socioeconomic pathway. Values for 2021 correspond to results from ecoinvent 3.8.

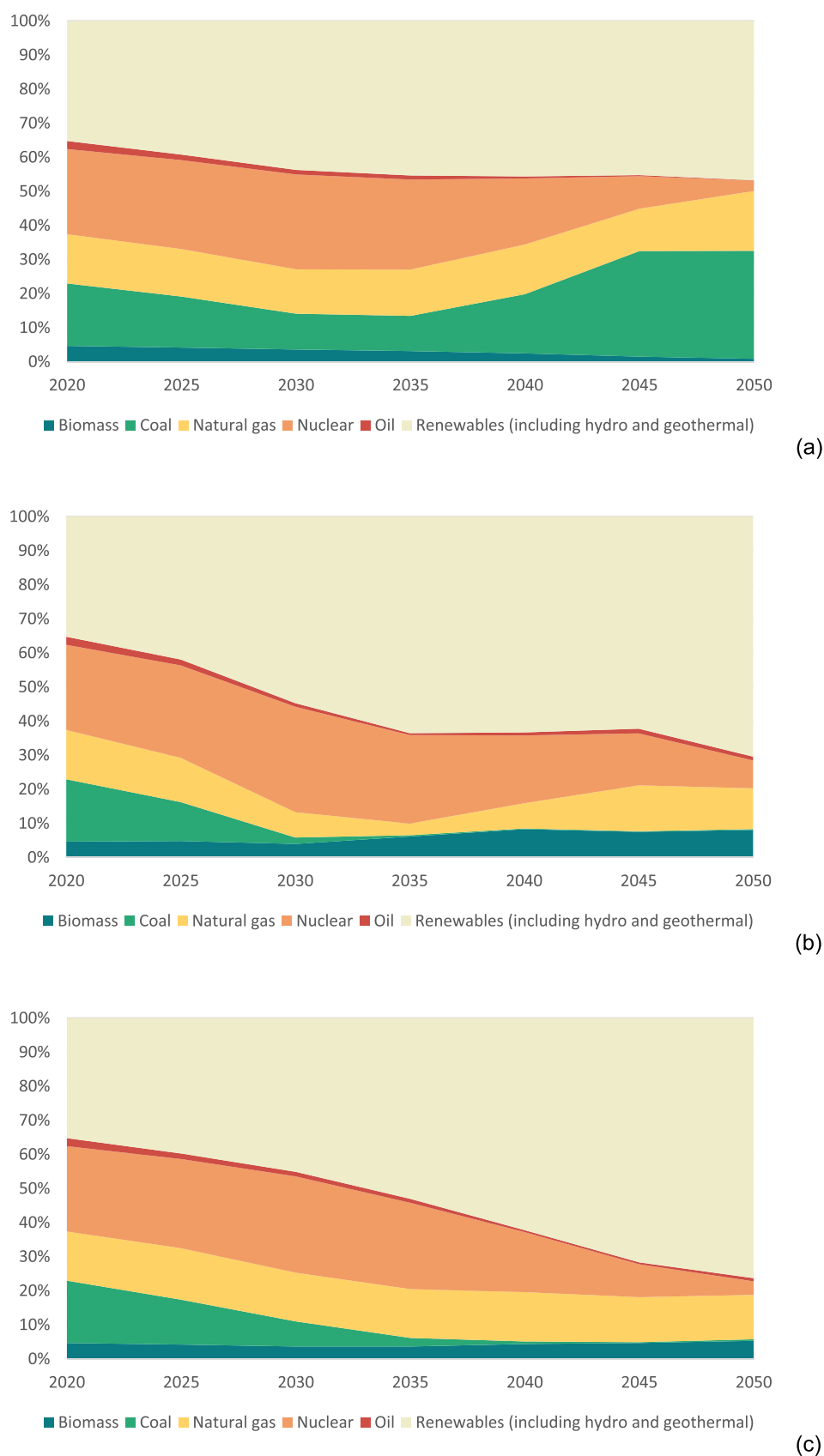


FIGURE 7 | Energy generation mix of Western Europe as projected by IMAGE for the scenarios SSP2-Base (a), SSP2-RCP19 (b), and SSP2-RCP26 (c). Figures generated with the Python library premise. IMAGE—Integrated Model to Assess the Global Environment, RCP—representative concentration pathways, SSP—shared socioeconomic pathway.

change impacts by 2050 are 77% and 73% compared with current impacts in the 2T and 75% and 72% in 4T, respectively. In all scenarios, the changes in climate change impacts can be mostly explained by the shift in energy carriers in the energy mix (Figure 7). The large reductions in climate change impacts for the SSP2-RCP1.9 and SSP2-RCP2.6 scenarios in future years align with the high shares of renewable energies in the energy mix in these scenarios. In the SSP2-Base scenario, the climate change impacts for the year 2030 are lower than those of the base year 2021, which can be explained by the reduction in the share of coal to 11% until 2030 (compared with 18% in 2020) and the increase in renewable energy until 2030 by 44% (compared with 35% in 2020). However, in this scenario, climate change impacts increase again between 2030 and 2050 because of the increased emissions per kWh of electricity in the Western European electricity mix because of an increased share of coal (32% of total electricity generation) and reduced progression of renewable electricity generation (47% of the total generation). Finally, an important difference between the ecoinvent 3.8 database and the prospective databases is their different geographical resolution. In the ecoinvent database, the electricity mix is available for Europe or individual countries, whereas in the prospective databases, the electricity mix is available for parts of Europe but not for Europe as a whole. Consequently, the European electricity mix used for the 2021 scenario is not fully comparable with the one used for the prospective scenarios, which are based on Western Europe only, excluding high-emitting countries such as Poland. This database difference exacerbates the big gap between the 2021 base case and the prospective scenarios in 2030.

3.4 | Sensitivity Analysis

For the perturbation analysis, all waste rates for the different manufacturing stages are changed one at a time, considering increases of 10%. The results for the 2T and 4T configurations are shown in Figure 8. Layers with SFs smaller than 0.05 in all impact categories, that is, changes smaller than 5% when the waste rate is changed by 10%, were excluded from the visualization. The results show little influence in the environmental impact categories of most waste rates over the different layers. For both configurations, the environmental impact categories most affected by the sensitivity to the waste rate are the abiotic depletion of mineral resources and freshwater ecotoxicity. For both configurations, the changes in waste rates in the absorber layer and the rear contact layer are the most critical.

The sensitivity analysis of the impacts on climate change considered different European energy mixes and is reported in Table 5 for both solar cell configurations under study. The results for other impact categories are presented in the [Supporting Information](#). The results show Sweden to have the energy mix with the greatest reductions in climate change impacts of up to 73%. These results show that the location of manufacturing at an industrial scale matters for the total impacts of these novel technologies. However, the choice of location is also constrained by socioeconomic aspects, such as labor and investment costs in different regions. In addition, it is also relevant to assess how the

energy mixes of the countries under consideration will change in the future and how this can affect production in each of the assessed countries.

3.5 | Comparison With Similar Studies and Limitations

Table 6 presents a nonexhaustive comparison of the obtained results to those obtained in similar studies in the scientific literature. The results are also compared with the literature reviews from [16] and [15], where the minimum and maximum values of different studies are given, as well as average values for CIGS technologies. Based on this comparison, the results obtained in this study are within the minimum and maximum range of existing studies, even though a high variability of results exists.

However, a direct comparison with existing studies is not straightforward because of the experimental setup of the cells under study and the different configurations identified in the literature. Various researchers have reported the results of LCA studies of perovskite solar cells, demonstrating a variety of configurations and fabrication processes that have been proposed in recent years [15, 16, 55]. This extensive range of cell configurations results in a wide variability of material flows as well as energy used throughout the different processes. For example, several routes are available for the deposition of perovskites [51]. In addition, the low TRL of the systems reported in studies used for the comparison of results also entails uncertainties concerning the use of material and scalability of manufacturing processes. Similarly to PCE, the number of operational years also presents great variability in the scientific literature, ranging from 1 to 30 years [11, 15, 16, 55].

Further complicating this issue, available LCA studies on solar cells often make methodological choices and assumptions regarding functional units, system boundaries, and LCIA methods. This calls for the harmonization of methodological assumptions when assessing the environmental performance of solar cells. Studies should also consider the assessment of social and economic impacts using the life cycle sustainability framework (LCSA). Concerning the environmental impacts, several reviews of the environmental performance of solar cells have been published in recent years, demonstrating that the global warming impact (GWI) and CED are the most frequent impact assessment categories used [56]. Therefore, the comparison with previous studies focuses on the results retrieved in the scientific literature for these impact categories, but it also includes the EPBT.

The results of the prospective assessment shed light on how the environmental impacts of the analyzed solar cells can change in future years under different scenario assumptions. The reduction of impacts on climate change until 2050 is aligned with the results reported by [17], which corresponded to average reductions of about 75% and 72% for tandem devices in the SSP2-RCP19 and SSP2-RCP26 scenarios, respectively. For the SSP2-Base scenario, [17] identified reductions between 46% and 48% of impacts on climate change, which contrasts with the results obtained in this study. In their assessment, [17] considered

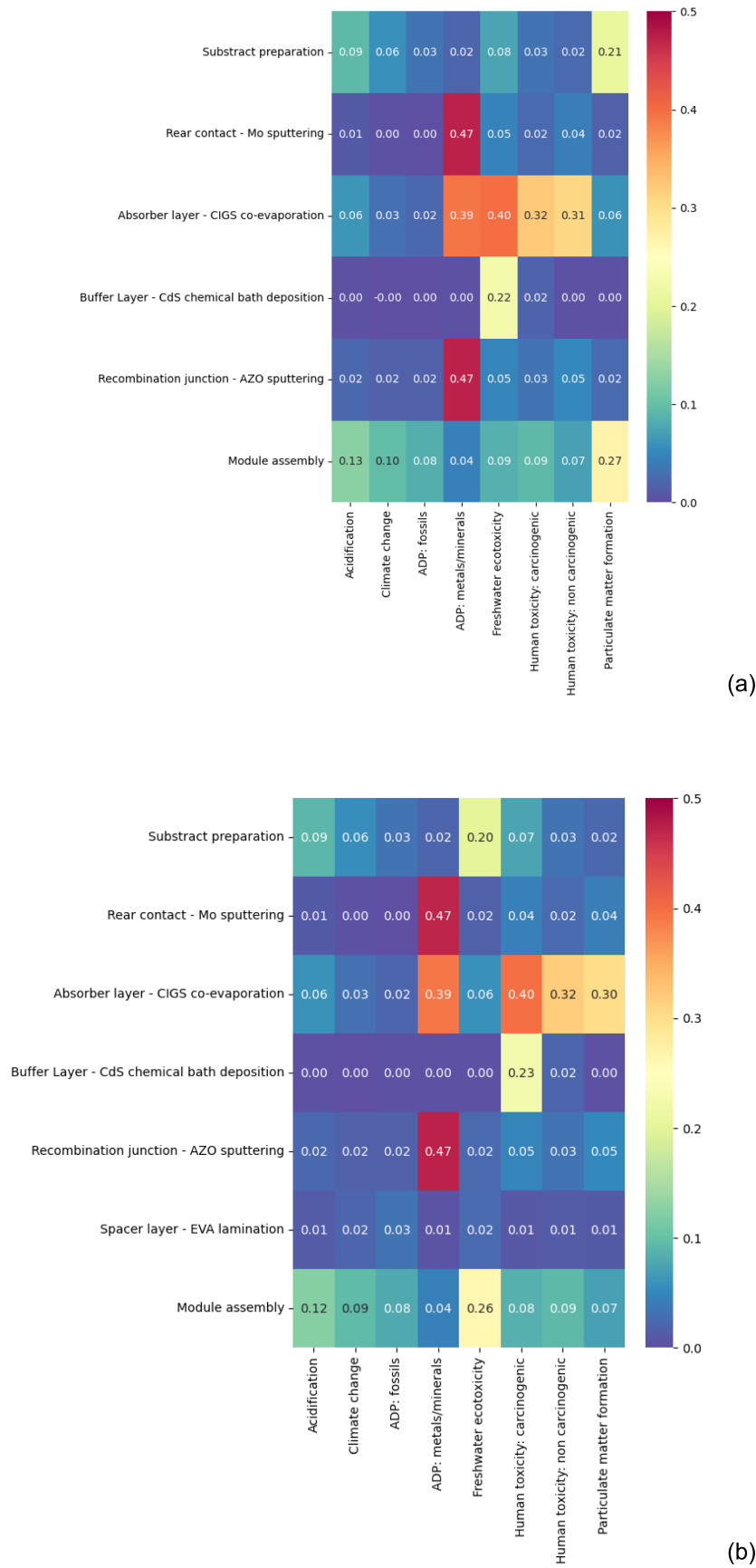


FIGURE 8 | Perturbation analysis of material waste rates for the 2T (a) and 4T (b) configurations. ADP—abiotic depletion potential, AZO—aluminium-doped zinc oxide, CdS—cadmium sulfate, CIGS—copper indium gallium selenide, EVA—ethylene vinyl acetate, Mo—molybdenum.

TABLE 5 | Sensitivity of the impacts on climate change considering different European energy mixes. Energy mixes are obtained from [19] using the EF 3.1 method.

Energy mix	Relative changes of results		Energy mix (gCO ₂ eq/kWh)–ecoinvent
	2T	4T	
EU-mix (base case)	—	—	390
Belgium	−27%	−24%	258
Estonia	92%	71%	866
France	−63%	−57%	79
Germany	41%	31%	556
Hungary	10%	9%	439
Sweden	−73%	−64%	44
Switzerland	−71%	−65%	39

changes in manufacturing processes toward the commercialization of the cells over the years based on experts' assumptions and secondary data sources, which are not considered in this study, that only consider the future changes of the wider economy. That aspect can be the reason for the divergence in the SSP2-Base scenario. Generally speaking, future impacts can be reduced because of learning effects during the upscaling of the emerging technology at higher TRLs in the foreground system and by the progress and technological change of the wider economy in the background system [57]. Upscaling lab-sized cells to large-area modules is a challenge since it must account for the efficiency and stability of the solar cells, which is further complicated because of the variability in the composition of perovskite materials. This study focuses on including prospective elements of the latter kind, leaving upscaling effects to higher TRLs for future research.

The results of the assessment of human toxicity impact categories should be interpreted with care. Even though Cd did not significantly impact the results because of its small amounts, there is pressure to eliminate cadmium from PV cells because of its toxicity. In addition, Regulation (EC) 1907/2006 (Registration, Evaluation, Authorisation and Restriction of Chemicals—REACH) [58], for example, has restricted the use of cadmium in many industrial applications. Previously, the replacement of the CdS buffer layer with zinc oxysulphide (Zn(O,S)) via ALD was shown to present lower impacts than the existing chemical bath deposition of CdS [59]. Moreover, even though lead is present in the absorber layer, its use did not significantly affect the environmental impact categories for human toxicity, which can be explained by the low mass that is used. Although the amount of lead in each cell is small, large-scale deployment could lead to cumulative environmental contamination if cells are not properly disposed of or recycled.

The results of CED and EPBT are within the ranges reported by existing studies. However, several ambiguous definitions and methods for CED exist, as well as methods for the calculation

of EPBT [49], causing divergent results in the scientific literature. Differences in the CEDs reported by different studies can be attributed to differences in CED definition [60]. The assessment of the EPBT only considered the PV cells. It does not consider the inverters and their efficiency, the mounting system, and auxiliary materials (e.g., cables), and the energy grid efficiency, which could influence the results and conversion of output PV electric energy to the generation side, resulting in longer EPBTs [49].

Similar to other studies, electricity consumption was shown to be the most critical aspect influencing the environmental impacts of perovskite solar cell manufacture. Therefore, it is critical to ensure the use of energy mixes with low carbon intensity. For that, strategies like green energy tariffs can be investigated since they could reduce the environmental impacts linked to solar cell manufacturing. Under a green tariff program, the supplied electricity is obtained from renewable energy sources, which could reduce the environmental impacts of solar cell manufacture.

Moreover, perovskite materials are prone to degradation when exposed to environmental factors (e.g., moisture, oxygen, heat, and UV light). This study assumes constant energy yield over time without cell degradation. However, future research should consider the impact of degradation effects on the output of electricity, which are particularly critical for the lifespan of the perovskite solar cells and their commercialization [61].

A concern regarding the environmental impacts of PV systems that is not assessed in this study is the evaluation of their end of life (EoL). The EoL of PV is particularly critical because of the potential treatment and disposal of hazardous metals that could be released into the environment. The EoL of PV systems is not often included in LCA studies because of the lack of data, even though they can have significant contributions to the total impacts [56]. Waste from PV systems in the European Union has been regulated by the Waste Electric and Electronic Equipment (WEEE) Directive since 2012 [62]. The WEEE Directive determines that PV modules shall be treated and recycled, therefore not incinerated or landfilled at their EoL. Currently, private companies have performed the reuse, repair, and refurbishment of PV components, and limited regulations and/or standards exist for the testing, certification, and labeling of refurbished PV modules [63].

In this context, the lack of established recycling processes for perovskite solar cells is an important concern for its commercialization [54]. Developing effective and economically viable recycling methods is essential to mitigate the environmental impact. Particularly in the case of perovskite cells, to date, no cells have reached the EoL, and only results on a lab scale have been presented in the scientific literature. Currently, some processes have been considered for the dismantling and recovery of materials contained in perovskite solar cells [64]. Poll et al. [65], for instance, provided a proof-of-concept for the electrochemical recovery of lead, achieving a recovery efficiency of 99.8%. Binek et al. [66] demonstrated a recycling process for perovskite based on methylammonium lead iodide (MAPbI₃) that proves that the conductive glass, as well as the

TABLE 6 | Comparison of results with existing studies.

Authors	System	System boundaries	Perovskite deposition	FU	PCE (%)	Lifetime (years)	GWI (kgCO ₂ eq/m ²)	GWI (gCO ₂ eq/kWh)	GWI (kgCO ₂ eq/kWp)	CED (MJ/m ²)	CED (kJ/kWh)	CED (MJ/kWp)	EPBT (years)
This study—2T configuration	CIGS/Perovskite	Cradle-to-gate	Slot die coating	m ² /kWh/kWp	28.8	25	49	3.21–5.55	170	1219	80–138	4231	0.56–0.97
This study—4T configuration	CIGS/Perovskite	Cradle-to-gate	Slot die coating	m ² /kWh/kWp	28.9	25	50	3.23–5.58	174	1266	81–140	4380	0.57–0.98
[50]	2T CIGS/tin-lead based perovskite	Cradle-to-gate	Spin coating	m ²	19.5	5	77	58 ^a	397	1819 ^b	1371 ^a	9325	0.58–0.67
[51]	Single junction carbon-based perovskite	Cradle-to-gate	Solution-based (spray)	m ² /kWh	15.0	30	157	150	1253	2550 ^b	2666	17,000	1.3
[51]	Single junction carbon-based perovskite	Cradle-to-gate	Vapor-based (vacuum)	m ² /kWh	15.0	5	188	180	1047	3040 ^b	3179	20,267	1.6
[52]	2T Si-perovskite tandem	Cradle-to-gate	Spray coating	m ² /kWh/kWp	28.0	10–30	38–163	4.5–19 ^c	580	700–2900	82–338	2500–10,400	0.30–0.73
[52]	CIGS	Cradle-to-gate	—	m ² /kWh/kWp	23.4	15–17.5	94	18–20 ^c	535–620	1500	284–331	8600–10,000	0.73–0.86
[53]	Perovskite/Si tandem with silver, gold, or aluminum as top electrodes	Cradle-to-gate	Spin coating	kWh	27.0	1	~103–122	~250–300	490–558	5036–6019	2571–3238	20,983–22,284	1.6–1.7
[16]	CIGS	Cradle-to-gate	—	m ²	10.5–15.0	25–30	35–92	9–23 ^d	271–557 ^d	1236–1936	304–476 ^d	9694–15184 ^d	1.35–3.32

(Continues)

TABLE 6 | (Continued)

Authors	System	System boundaries	Perovskite deposition	FU	PCE (%)	Lifetime (years)	GWI (kgCO ₂ eq/m ²)	GWI (gCO ₂ eq/kWh)	GWI (kgCO ₂ eq/kWh)	CED (MJ/m ²)	CED (kJ/kWh)	CED (MJ/kWh)	EPBT (years)
[16]	Perovskite tandem	Cradle-to-gate/Cradle-to-grave	—	m ²	6.0–27.0	25–30	33–2066	157–25245 ^d	200–12521 ^d	419–5311	157–25245 ^d	2539–32188 ^d	0.34–19.28
[54]	Carbon-based perovskite	Cradle-to-grave	Ink-jet printing	kWh	14.40	25	28	92–152	195	736	2405–4000	5111	0.90–1.40
[17]	4T PERC Si/perovskite tandem monofacial	Cradle-to-grave	Slot die printing and thermal cure	kWh	26.5	30–40	321 ^e	35	1212	NA	NA	NA	NA
[17]	4T PERC Si/perovskite tandem bifacial	Cradle-to-grave	Slot die printing and thermal cure	kWh	30.5	30–40	317 ^e	30	1039	NA	NA	NA	NA

Abbreviations: CED, cumulative energy demand; CIGS, copper indium gallium selenide; EPBT, energy payback time; FU, functional unit; GWI, global warming impact; NA, not available; PCE, power conversion efficiency; PERC—passivated emitter and rear contact; Si, silicon.

^aValues converted considering the reported performance ratio (PR) of 80%.

^bBased on the primary energy demand. Calculation based on the highest energy payback time value reported.

^cBased on values reported by the study: solar insulation of 1800 kWh/m²/year and a lifetime of 20 years (representative of the central values reported in the study). The PR is 0.85.

^dCalculated considering central PCE values.

^eImpacts per m² were calculated assuming parameters indicated in the manuscript during 30 years of operation and no degradation over the operational years.

toxic lead precursor, can be reused. However, the toxicity and flammability of the solvents used also require further investigation concerning their safety, environmental, and health impacts.

4 | Conclusion

Emerging PV technologies, such as perovskite, need to meet technical requirements besides economic and environmental performances to achieve a successful large-scale commercialization. This study presents the cradle-to-gate environmental impacts of perovskite solar cells with two configurations, 2T and 4T, at TRL 3 and 4 using LCA and the indicators CED and EBPT. The LCA, CED, and EBPT results show marginal differences between the two analyzed configurations. The impacts on climate change and abiotic depletion of fossil fuels are marginally higher for the 4T cells, about 3% and 5% higher than the 2T cells. For the impact categories particulate matter emissions and abiotic depletion, the impacts are nearly the same. The CED of the 4T configuration is 4% higher than that of the 2T configuration, and the EPBT for the 4T configuration is about 2% higher than that of the 2T configuration.

The results show that the source of electricity is the most critical contributor to the environmental impacts. The impacts for acidification potential, abiotic depletion of fossil fuels, and particulate matter are linked to the use of electricity to manufacture solar cells. Improvements in these impact categories imply the replacement of energy sources since the results showed a high impact because of the use of fossil energy sources in the European energy mix. The sensitivity analysis confirms that the results are highly impacted by the carbon intensity of the energy mix used in manufacturing. Changing the energy mix used for manufacturing can reduce impacts on climate change by up to 73%. The prospective assessment shows that the total greenhouse gas emissions could be reduced by up to 77% by 2050 in both systems if stringent climate scenarios were adopted.

For other impact categories, a reduction in the impacts of abiotic depletion of elements is linked to the reduction of Mo use. The impacts on freshwater ecotoxicity can be reduced by minimizing the consumption of indium. For toxicity, the reduction of the use of indium and copper should be foreseen. Finally, the perturbation analysis identified the waste rate on the absorber layer and rear contact as critical aspects of impacts on the abiotic depletion of mineral resources for both cells. For both configurations, the waste rate on the recombination junction was also critical to this impact category. The findings align with similar studies in the scientific literature. However, comparisons with these other studies are complicated by the employment of different perovskite technologies and methodologies to assess the environmental impacts.

Assessing different EoL strategies and the effects of degradation rates are aspects not covered in this study that can be explored in future research. Future studies should also consider the effects of upscaling in the foreground system to understand the impact of the increase of the TRL through technological learning effects on environmental impacts. This is

particularly relevant since improvements such as higher cell efficiency, reduction in energy consumption during module production, and EoL strategies might change at higher TRLs. Finally, exploring the economic and social aspects of the commercial production of perovskite solar cells is a critical aspect that must be further analyzed using frameworks such as LCSA.

Author Contributions

Daniele Costa: conceptualization, data curation, formal analysis, writing – original draft preparation. **Alessandro Martulli:** data curation. **Anne van den Oever:** formal analysis. **Amelie Müller:** writing – review and editing, formal analysis. **Roel Degens:** writing – review and editing. **Neethi Rajagolapan:** writing – review and editing. **Ulrich W. Paetzold:** data curation. **Sebastien Lizin:** writing – review and editing. **Bart Vermang:** funding acquisition.

Acknowledgments

This study received funding from the European Union's Horizon 2020 research and innovation program under grant no. 850937 (PERCISTAND). The authors thank Romain Carron for his support during the execution of this work.

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

Endnotes

¹ <https://github.com/PerovskitePV/EYcalc>.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section. **Table S1.** Layout of 2T cell block (from top to bottom). Legend: 2PACz –2-(9H-carbazol-9-yl)ethyl)phosphonic acid, AZO – aluminum-doped zinc oxide, C60 – buckminsterfullerene, CdS – cadmium-sulfide, CIGS – copper indium gallium selenide, EVA – ethylene vinyl acetate, ITO – indium tin oxide, MgF₂ – magnesium fluoride, Mo – molybdenum, NiOx – nickel (II) oxide, SAM – self-assembled monolayer,

SnO₂ – tin (IV) oxide, ZnO – zinc oxide. **Table S2.** Layout of 4T cell block (from top to bottom). Legend: 2PACz – 2-(9H-carbazol-9-yl)ethyl) phosphonic acid, AZO – aluminum-doped zinc oxide, C60 – buckminsterfullerene, CdS – cadmium sulfide, CIGS – copper indium gallium selenide, EVA – ethylene vinyl acetate, ITO – indium tin oxide, MgF₂ – magnesium fluoride, Mo – molybdenum, NiOx – nickel (II) oxide, SAM – self-assembled monolayer, SnO₂ – tin (IV) oxide, ZnO – zinc oxide. **Table S3.** Life cycle inventory for manufacturing 2T solar cells. Values are reported for 1 m² of solar cell. *Source:* personal communication with manufacturers and developers of CIGS and perovskite photovoltaic technologies. Legend: AZO – aluminum-doped zinc oxide (Al:ZnOx), C60 – buckminsterfullerene, CdS – cadmium sulfide, CIGS – copper indium gallium selenide, CsI – cesium iodide, DMF – N,N-dimethylformamide, DMSO – dimethyl sulfoxide, ETL – electron transport layer, EVA – ethylene vinyl acetate, FAI – formamidine iodide, ITO – indium tin oxide, MgF₂ – magnesium fluoride, Mo – molybdenum, NiOx – nickel (II) oxide, PbI₂ – lead (II) iodide, PVD – physical vapor deposition, SAM – self-assembled monolayer, SnO₂ – tin (IV) oxide, ZnO – zinc oxide. **Table S4.** Life cycle inventory for manufacturing 4T solar cells. Values are reported to 1 m² of solar cell. *Source:* personal communication with manufacturers and developers of CIGS and perovskite photovoltaic technologies. Legend: AZO – aluminum-doped zinc oxide (Al:ZnOx), C60 – buckminsterfullerene, CdS – cadmium sulfide, CIGS – copper indium gallium selenide, CsI – cesium iodide, DMF – N,N-dimethylformamide, DMSO – dimethyl sulfoxide, ETL – electron transport layer, EVA – ethylene vinyl acetate, FAI – formamidine iodide, ITO – indium tin oxide, MgF₂ – magnesium fluoride, Mo – molybdenum, NiOx – nickel (II) oxide, PbI₂ – lead (II) iodide, PVD – physical vapor deposition, SAM – self-assembled monolayer, SnO₂ – tin (IV) oxide, ZnO – zinc oxide. **Table S5.** Life cycle inventory for the production of 1 kg of aluminum-doped zinc oxide (AZO) life cycle inventory. *Source:* adapted from Celik et al. (2016). **Table S6.** Life cycle inventory for the production of 1 kg of buckminsterfullerene (C60). *Source:* Anctil et al. (2011). **Table S7.** Life cycle inventory for the production of 1 kg of cesium iodine (CsI). *Source:* Alberola-Borrás et al. (2018). **Table S8.** Life cycle inventory for the production of 1 kg of formamidine iodide – FAI (HC(NH₂)₂I). *Source:* Zhang et al. (2017). **Table S9.** Life cycle inventory for the production of 1 kg of formamidine acetate – FAAC (C₃H₈N₂O₂). *Source:* Zhang et al. (2017). **Table S10.** Life cycle inventory for the production of 1 kg of hydriodic acid (HI). *Source:* Zhang et al. (2015). **Table S11.** Life cycle inventory for the production of 1 kg of nickel (II) oxide (NiOx). *Source:* Leccisi and Fthenakis (2021). **Table S12.** Life cycle inventory for lead iodide (PbI₂) production. *Source:* Leccisi and Fthenakis (2021). **Table S13.** Life cycle inventory for thiourea production. *Source:* van der Hulst et al. (2020). **Table S14.** Life cycle inventory for calcium cyanamide production (CaCN₂). *Source:* van der Hulst et al. (2020). **Table S15.** Life cycle impact assessment for the 2T configuration per 1 m². Legend: AD – acidification, ADP – abiotic depletion potential, ALD – atomic layer deposition, AZO – aluminum-doped zinc oxide, C60 – buckminsterfullerene, CdS – cadmium-sulfide, CED – cumulative energy demand, CIGS – copper indium gallium selenide, ETL – electron transport layer, FE – freshwater ecotoxicity, HT – human toxicity, HTL – hole transport layer, ITO – indium tin oxide, MgF₂ – magnesium fluoride, Mo – molybdenum, NiOx – nickel (II) oxide, PMF – particulate matter formation, PVD – physical vapor deposition, SAM – self-assembled monolayer, SnO₂ – tin (IV) oxide, TCO – transparent conductive oxide, ZnO – zinc oxide. **Table S16.** Life cycle impact assessment for the 4T configuration per 1 m². Legend: AD – acidification, ADP – abiotic depletion potential, ALD – atomic layer deposition, AZO – aluminum-doped zinc oxide, C60 – buckminsterfullerene, CdS – cadmium-sulfide, CED – cumulative energy demand, CIGS – copper indium gallium selenide, ETL – electron transport layer, EVA – ethylene vinyl acetate, FE – freshwater ecotoxicity, HT – human toxicity, HTL – hole transport layer, ITO – indium tin oxide, MgF₂ – magnesium fluoride, Mo – molybdenum, NiOx – nickel (II) oxide, PMF – particulate matter formation, PVD – physical vapor deposition, SAM – self-assembled monolayer, SnO₂ – tin (IV) oxide, TCO – transparent conductive oxide, ZnO – zinc oxide. **Table S17.** Sensitivity analysis based on energy mix scenarios for the 2T configuration. Legend: AP – acidification potential, CC – climate change, ADP – abiotic depletion potential, FE – freshwater ecotoxicity, HT – human toxicity, PMF – particulate matter formation.