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Assessing the sustainability of emerging technologies: A probabilistic LCA method applied to advanced photovoltaics

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A B S T R A C T

A key source of uncertainty in the environmental assessment of emerging technologies is the unpredictable manufacturing, use, and end-of-life pathways a technology can take as it progresses from lab to industrial scale. This uncertainty has sometimes been addressed in life cycle assessment (LCA) by performing scenario analysis. However, the scenario-based approach can be misleading if the probabilities of occurrence of each scenario are not incorporated. It also brings about a practical problem; considering all possible pathways, the number of scenarios can quickly become unmanageable. We present a modelling approach in which all possible pathways are modelled as a single product system with uncertain processes. These processes may or may not be selected once the technology reaches industrial scale according to given probabilities. An uncertainty analysis of such a system provides a single probability distribution for each impact score. This distribution accounts for uncertainty about the product system's final configuration along with other sources of uncertainty. Furthermore, a global sensitivity analysis can identify whether the future selection of certain pathways over others will be of importance for the variance of the impact score. We illustrate the method with a case study of an emerging technology for front-side metallization of photovoltaic cells.

1. Introduction

Whenever a new technology is proposed, the main concern from an environmental perspective is whether it will satisfy certain societal needs at the expense of introducing unwanted environmental burdens. This has happened often in the past, sometimes resulting in global-scale environmental issues that were not foreseen. Life cycle assessment (LCA) is until now the only environmental assessment method that can systematically reveal undesired environmental trade-offs that may result when an existing technology is replaced by a new one (Guinee, 2002). Because of this, the application of LCA in early research and development (R&D) stages has gained considerable traction in recent years (Cucurachi et al., 2018) and is even recognized by the European Union as an essential component of the R&D projects it is funding (European Commission Joint Research Centre, 2019).

The LCA method was originally developed to study systems for which sufficient information about material and energy inputs and outputs, as well as the cause-effect relationships throughout the entire supply-chain of a technology is obtainable. This is already challenging for well-established technologies, let alone for technologies that are in development and have not yet been commercialized. In both cases, many uncertainties arise from missing or inaccurate data, spatial and temporal variability of process parameters, spatial and temporal variability of characterization models, and inaccuracy of characterization models, amongst other sources (Huijbregts et al., 2003; Igos et al., 2018; Lloyd and Ries, 2008). The standard approach for dealing with these uncertainties in LCA is to represent them using stochastic parameters with probability distributions (e.g., uniform, normal or lognormal) instead of fixed values, and then propagate them by random sampling and calculation of the resulting impacts in numerous Monte Carlo simulations. Rather than a single impact score, this approach produces a probability distribution for the impact score which can

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also be described by its mean, mode, variance, confidence intervals, and/or other statistical descriptors (Groen et al., 2014).

For emerging technologies, the challenge of dealing with uncertainty is even greater because these technologies have not been tested in a real operating environment and many design aspects have not been settled yet (Arvidsson et al., 2017; Bergerson et al., 2019; Hetherington et al., 2014; Villares et al., 2017). At any given point in time during the R&D process, there are many unknowns as to how the numerous technical and economic roadblocks to a successful marketable product will be eventually overcome, if they are overcome at all. In addition to this, the technology must be evaluated in the future economic and environmental context in which it will be deployed. An LCA model that attempts to forecast the impacts of such an unproven and immature technology therefore has potentially larger and more diverse sources of uncertainty (Table 1).

Following the typology of Huijbregts et al. (2003), some of these uncertainties can be represented as "parameter" uncertainties, e.g. when the quantities of material and energy inputs and outputs required in each manufacturing step may decrease as a result of future process optimizations. If reasonable estimates for the expected changes in these quantities is within reach, then this type of variation can be incorporated via the aforementioned Monte Carlo methods using most LCA software. Other perhaps more consequential types of uncertainty are related to which specific manufacturing steps will ultimately enable the early design or concept to become technically and economically feasible. Numerous and widely diverse engineering solutions are proposed and tested during early R&D stages, and these may or may not be a part of a technology's future product system configuration once it reaches maturity. We refer to these different possible configurations as "technological pathways", each of which is further pursued and investigated in subsequent R&D stages in order to find the one that ensures technical and economic feasibility. This type of uncertainty can be classified as "scenario uncertainty" and has often been addressed in LCA by modelling each technological pathway as a separate scenario (Arvidsson et al., 2017; Cucurachi et al., 2018; Valsasina et al., 2017).

Assessing and comparing different scenarios is useful when a design choice can be made on sustainable grounds (Höjer et al., 2008). However, the usefulness of this approach is more limited when there is no choice, rather a technological pathway that will eventually emerge as the −often only - economically and technically viable option. If the LCA results are meant to guide funding decisions that must be made with the *current* state of information, a comparative assessment of two or more scenarios can be misleading, even more so if the probability of one occurring is higher than the other. Another limitation is of a more practical nature; considering all the different possible technological pathways, the number of scenarios will most likely become unmanageable and their interpretation confusing if not impracticable.

To address these limitations, in this paper we propose a probabilistic approach in which all technological pathways being pursued by the developer are combined in a single product system. The competing pathways are activated or deactivated in each Monte Carlo run according to their probabilities of success by stochastic triggers or switches that are built into the LCA model. This type of model setup builds upon those proposed by other authors for combining different scenarios and/or modelling choices in single product systems (Azari Jafari et al., 2018; Gregory et al., 2016; Huijbregts et al., 2003; Mendoza Beltran et al., 2016). It has been shown that these models allow the joint propagation of parameter, scenario and model uncertainties, producing a single probability distribution for the studied system's impact score.

The framing and methods we propose extend and refine the previous work of these authors in various ways. First, in applying this approach to emerging technologies we propose a clear separation between (i) uncertainty about the potential success of competing technological pathways, and (ii) uncertainty introduced by subjective modelling choices or preferences related to allocation, system boundaries, and future external scenarios. The former constitutes an inherent uncertainty about the product system and its effect is appropriately reflected by a single output impact score distribution. The latter, on the other hand, is best investigated as separate scenarios, in order to distinguish the effects of subjective choices and make them more transparent.

To further differentiate between (i) and (ii), we note that the stochastic triggers we use in (i) to activate technological pathways are objective parameters with a *true* value: each pathway either can or cannot overcome the technical and economic barriers the technology concept faces, but this is unknown at present by the developer. This true value −the uncertainty of which is adequately characterized by a Bernoulli distribution - will only be found by future R&D and testing. On the other hand, subjective value choices as in (ii) do not have an empirical "true" value and their joint propagation risks masking the effect of such subjective choices,

Table 1
Additional uncertainty sources specific to LCA of emerging technologies.

LCA phase	Uncertainty source	Uncertainty type	Context in LCA of emerging technologies
Goal and scope	Functional unit	Scenario	The technology may ultimately be used in ways different than the one projected initially, or it may be used for multiple/different purposes.
	System boundary: end-of-life (EOL)	Scenario	The possibilities for reuse/recycling often develop after the technology has been deployed, and/or when it is economically feasible. It is not known if and how this will happen. Regulations may change with respect to EOL requirements.
Inventory	Unit process	Scenario	The manufacturing methods will most likely change as the technology moves from the lab to industrial scale.
	Flow quantities	Parameter	Cost and process optimizations will likely lead to reduced or substituted material and energy input/output flows.
	Allocation	Parameter	The parameters used to establish the criteria for allocation of multifunctional processes might change in time. E.g., forecasted market values in the case of economic allocation.
Impact assessment	Characterization model	Model	Novel materials may have unknown or insufficiently studied impact mechanisms or pathways.
	Characterization model: fate	Parameter	Landscape parameters that affect transport and fate of substances may change in time, e.g. global temperature.
	Characterization model: exposure	Parameter	Parameters that affect exposure e.g. population densities or diets may change in time.
	Characterization model: effect	Model	Marginal changes may result in exponentially larger effects as the baseline condition deteriorates. E.g. impact of increased radiative forcing on ecosystems.

reducing model transparency (Gregory et al., 2016).

Second, our method investigates the effects of uncertainty about the probabilities (chances of success) of each pathway/scenario, which most likely exists in early R&D. This uncertainty about the input probabilities is often called second-order uncertainty (Borsotto et al., 2006; Sankararaman and Mahadevan, 2013). We characterize these uncertainties using different types of probability distributions for these parameters other than uniform, allowing for a more refined and realistic representation of the expectations of technology developers.

Finally, we demonstrate the application of a global sensitivity analysis (GSA) method that is suitable for such a model and highlights which uncertainties - including those from competing technological pathways as well as second-order uncertainties - are most relevant from an environmental perspective. Our aim with this is to identify incentives to more actively pursue research towards resolving the most sensitive ones. If they cannot be resolved, the information can and should be used to select the more relevant pathways that merit further investigation via e.g. local sensitivity analysis. In this case, the definition of scenarios for further investigation as a subsequent step becomes more objective and systematic, as the modeller will have quantitative criteria to select those that are most relevant.

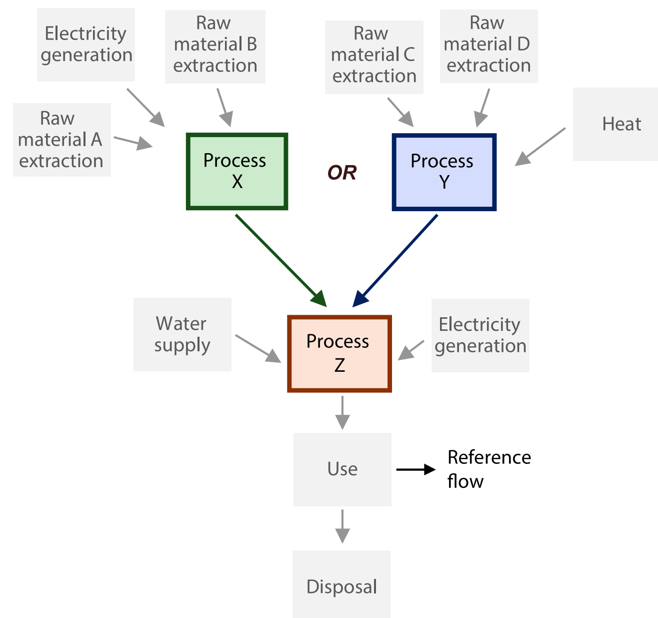

Fig. 1. Product system with a process (Z) that requires an input from two competing, mutually exclusive process (X or Y).

2. Methods

2.1. Configuring the parametrized product system

To perform LCA calculations on a single system that combines different technological pathways, we use random parameters that activate or deactivate the inputs from the competing processes according to their underlying probabilities of occurrence (i.e. chances of success). To each competing process, we attach a random trigger that takes on a value of 0 or 1, so that it activates or deactivates the process flow according to a defined Bernoulli distribution function. The Bernoulli distribution is a discrete distribution that has two possible outcomes: success (=1) occurs with probability π, and failure (=0) occurs with probability 1- π, where $0 < p < 1$ (Forbes et al., 2011).

Step 1: Identify the relevant technological pathways. The first step is to screen for the possible technological pathways that are being pursued, and the corresponding unit processes that are to be included in the single product system. This can be aided by a quick-scan lab-scale LCA and by eliciting expert knowledge and expectations of technology developers. The result of this step is a tree of possibilities that includes a number of pathways to fulfil the intended function(s) of the technology. This step would screen for alternative competing unit processes in all life-cycle stages, including manufacturing but also use and end-of-life options.

Step 2: Set up the product system. The competing unit processes (Process X and Process Y) are connected as providing simultaneous inputs to Process Z as shown in Fig. 1.

Step 3: Determine the required flows. Each competing process may contribute in a different way. For example, process Z may use either 1 kg of the product made by process X or 2 kg of the product made by process Y. Both quantities are added to the process Z as if they occur simultaneously, so the inputs of process Z are 1 kg of product from process X and 2 kg of product from process Y.

Step 4: Determine the probabilities of occurrence of each flow. The probability of occurrence of X or Y will most likely be determined based on expert knowledge or expectations from the technology developers about technical and/or economic feasibility. For example, they may be estimated by looking at trends in related technologies, or by using economic forecasts for each alternative as a proxy. The criteria should be tightly linked to the functional unit of the technology, and the chances each option has of contributing to this function in an optimal (technical and economic) way. We define π as the probability of process X being selected, where π is a value between 0 and 1. Then the probability of process Y being selected is 1- π.

Step 5: Define parameter T. We will use a random number T to switch each flow on or off, by taking 1 for 'on' and 0 for 'off'. We generate T from a Bernoulli distribution, which is equivalent to a binomial distribution with 1 single trial (n = 1) and probability π.

$$T \sim \text{bin}(n = 1, \pi)$$

If there are more than two competing unit processes for the same element of the technology's product system, the generalized version of the Bernoulli distribution can be used, namely the categorical distribution. In this case we would define the probability of process X as π_x, the probability of process Y as π_y, and the probability of process Z would be $\pi_z = 1 - (\pi_x + \pi_y)$. A similar result can be achieved by nesting the alternatives so that their combined probabilities result in the desired individual probabilities (see Supplementary Information for implementation notes).

Step 6: Apply the triggers to each flow. Because they are competing processes, only one flow can be activated at a time. This is achieved by multiplying process Z's input from Process X by *[T]* and the input from Process Y by *[1-T]*.

Step 7: If applicable and known, add uncertainty to the probability of occurrence (success) of each flow. The probabilities of each flow occurring may be given as a range, rather than fixed. For example, "the chance of using process X instead of process Y may be between 30% and 50%". In this case, a uniform distribution with minimum 0.3 and maximum 0.5 can be used. The uncertainty about the probabilities can be characterized in even more detail by using non-uniform distributions. Such is the case when a range of probabilities is expected, but there is more confidence around a certain value. For example, the chance of using process X instead of process Y is between 30% and 50%, but most likely 40%. This can be characterized by a triangular distribution with min 0.3, max 0.5 and mode 0.4. To implement this, the uncertainty distribution is directly applied to parameter π in the equations above. Wide ranges can be used in this step when there is limited knowledge about the probabilities. The relevance of this second-order uncertainty will be investigated afterwards in the global sensitivity analysis, indicating whether further efforts are necessary to make the predictions more accurate.

Step 8: Run the Monte Carlo simulation. The Monte Carlo simulation is run for the single product system. In each run, uncertain flows and characterization factors will take on random values according to their underlying probability distributions, and the effects propagated towards the calculation of the impact score. In the same way, the random triggers will randomly activate or deactivate the alternative technological pathways, according to their chances of success. The sampling in each run is done in a dependent way as recommended by Henriksson et al. (2015) and Mendoza Beltran et al. (2018), in order to ensure that shared unit processes across both systems take the same random values in each run. The inventory or impact assessment output will represent a future system that has a probability π of using Process X and a probability π *-1* of using Process Y.

Step 9: Global sensitivity analysis. Several sensitivity indices and the corresponding algorithms to calculate or estimate them have been proposed for GSA (Borgonovo and Plischke, 2016). These methods can calculate or estimate how much each uncertain input contributes to the model's output variance, for all or a subset of uncertain input parameters. For our model we propose the delta moment-independent sensitivity measures (Borgonovo, 2007) which had previously been implemented in LCA by Cucurachi et al. (2016). Various methods have been proposed to estimate the delta measures (Derennes et al., 2019; Plischke et al., 2013); we used the *betaKS3* MatLab subroutine developed and provided by E. Plischke and E. Borgonovo upon request (Borgonovo and Iooss, 2017).

The sensitivity measure and corresponding estimation algorithm we propose present several important advantages for our model: (i) it accounts for possible correlations between uncertain input parameters; (ii) it has a significantly faster computation time and less memory usage, which is essential for models with tens or hundreds of thousands of uncertain parameters as in the case of large LCA databases like ecoinvent (Frischknecht et al., 2005); (iii) it is independent of the model and only requires the values taken by the uncertain input parameters and the outputs (impact scores) in each Monte Carlo run, making them easy to apply in LCA; (iv) it is moment-independent, i.e. reflects expected changes in the actual output distribution rather than an approximated curve fit (typically a lognormal distribution with an estimated mean and variance). This is especially important in our framing given that, as we will show, the superposition of different technological pathways may produce output impact score distributions with more than one peak (multimodal or heteroscedastic). In such cases, variance based sensitivity measures would not provide accurate estimates of importance. Finally, (v) it can take uncertain input parameters with discrete distributions, such as the binomially distributed triggers we used.

2.2. Case study of emerging photovoltaic technologies

We applied the method to a real-life case study in order to determine whether it was computationally feasible, if the results are in line with expectations and to further explore what types of conclusions can be drawn from the analysis. For this, we chose an emerging technology for metallization of the front electric contacts of photovoltaic (PV) cells that uses silver or copper metallic nanoinks. The special properties of the nanoparticles in the ink enhance the cell's performance by reducing the shadow, i.e. the area of cell that is covered by the metallic patterns and does not receive sunlight. It can also reduce the amount of silver required vs. traditional screen-printing methods. The case study is an ideal situation to investigate whether secondary materialization is occurring, while many possible configurations of the manufacturing and mainstream use of the technology are yet to be resolved. The concept of secondary materialization, introduced by Williams et al. (2002), suggests that *"technological progress tends to increase energy and material use associated with products and is thus a counterforce to efficiency improvements attributed to dematerialization"*.

Preparation of the metallic nanoinks starts with the manufacturing of metallic nanoparticles via one of two possible routes; physical (or "top-down") methods apply energy to fracture larger particles to nanoscale sizes, and chemical (or "bottom-up") methods create the nanoscale particles from even smaller molecules using chemical reactions (Kamyshny and Magdassi, 2017). We based our calculations for these processes on the life-cycle inventories reported by Pourzahedi and Eckelman (2015) and Slotte and Zevenhoven (2017). The nanoinks consist of a solution of metallic nanoparticles in alcohol/hydrocarbon (for silver) or polymer (for copper) and are deposited in patterns on the front side of the cell by inkjet printing to form an initial "seed layer". The printed patterns then have to be sintered, using either a thermal (laser) or a chemical process that consolidates the metallic particles in the pattern (Renn et al., 2017). Sintering of silver nanoparticles can be done in open air, while copper nanoink requires an oxygen-free atmosphere to avoid formation of undesired oxides on the contacts (Hermerschmidt et al., 2018). Once sintered, the fingers are grown to a final thickness of 12.5 μm by electroplating. Three busbars are placed on the cell using the conventional screen-printing methods that are used for the fingers in most commercially available silicon PV cells. Fig. 2 and Table 2 show the different competing alternatives and the parameter values used in the model.

In addition to the five stochastic triggers *T1-T5* and their uncertain probabilities of success π*1*- π*5*, we also included three input parameters subject to the more conventional form of uncertainty commonly addressed in LCA. First, we varied the amount of sintering gas mixture consumed per PV cell, dividing it by a random, triangularly distributed value (*P6*) with min:1, mode:5 and max:10. Second, we considered uncertainty in the amount of electrolyte solution consumed in electroplating per PV cell, i.e. how many cells can be treated per batch. We represented this by a parameter *P7* that divided the amount of solution required by a random, triangularly distributed value with min:10, mode:50 and max:100. Finally, we considered a potential increase in cell conversion efficiency of between 0.5 and 2%. We represented this by a parameter *P8* that multiplied the PV cell area required to produce 1 kWh by a uniformly distributed value between 0.98 and 0.995.

We then ran a (dependent) Monte Carlo simulation of $n = 1000$ runs to calculate and compare the impact scores of the nanoink printed PV cell with a conventional screen-printed PV cell. For this comparison we defined the functional unit as the generation of 1 kWh of electricity. For the conventional cell, we used the inventory data for single-Si photovoltaics from the LCA database ecoinvent v2 (Frischknecht et al., 2005), and incorporated uncertainty in the background input/output flows provided by ecoinvent. We focused on four impact categories: climate change, ozone depletion, human toxicity and freshwater aquatic ecotoxicity, all based on the ReCiPe impact assessment method (Goedkoop et al., 2009).

We then used the modified null hypothesis significance test proposed by Heijungs et al. (2016) to determine whether the differences in impact scores between the types of systems were statistically significant. The choice of the modified version of the test responds to the fact that it is well suited for early stages in technology development, where we the size (or relevance) of the difference is important. In other words differences that are not

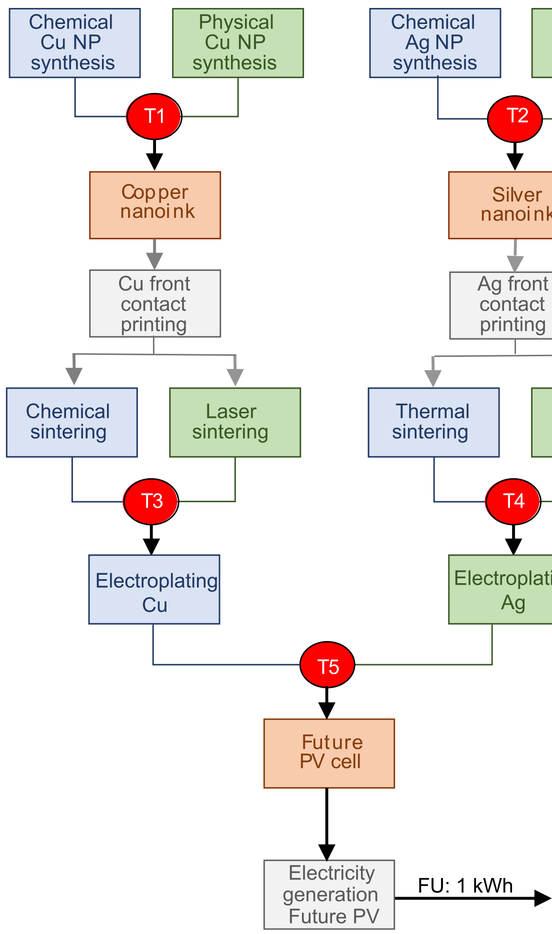

Fig. 2. Product system for the generation of electricity using a solar cell with nanoink-printed front contacts, considering different alternative manufacturing pathways. T variables identify the triggers that select one or the other of a competing pair of unit processes for each pathway.

relevant enough should not provide a basis to deter continued research and development while the potential benefits of the technology are still uncertain. To implement the modified null hypothesis significance test we used the excel based tools developed by Mendoza Beltran et al. (2018).

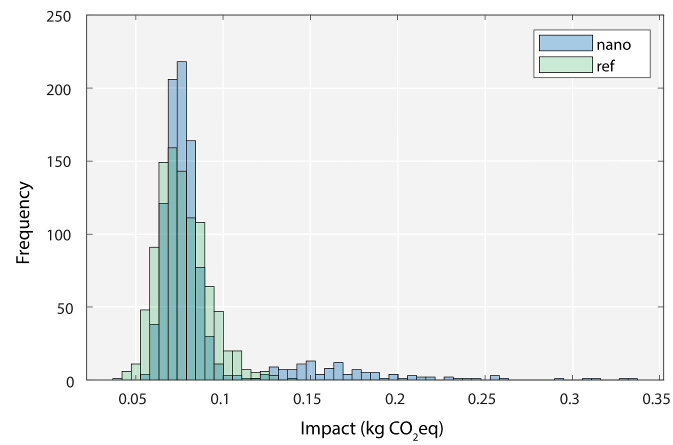

Fig. 3. Comparison of climate change impacts of a PV system with nanoink-printed cells (nano) and a conventional screen-printed cells (ref).

3. Results and discussion

3.1. Comparative impact assessment of PV systems

The distribution of the climate change impact scores for both types of PV systems (nanoink-printed and conventional screen-printed cells) are shown in Fig. 3. The impact score distributions of both systems mostly overlap around 0.08 kg CO_2eq, except for an additional peak around 0.15 kg CO_2eq for the nanoink-printed cells. This is in line with our expectation to find multimodal output distribution curves, and further strengthens the case for the use of moment-independent global sensitivity measures (this is further discussed in Section 3.2). By looking at the impact contributions of the individual foreground processes, we were able to determine that the additional peak around 0.15 kg CO_2eq corresponded to the chemical sintering pathway for the copper nanoink option which had a low probability of success (hence the lower frequencies), but was the only pathway that could result in impacts in this higher range.

Having a single probability distribution for the impact scores, we can draw general conclusions about the expected impacts of the nanoink-printed PV technology. For climate change, for example, the impacts will range between 0.05 and 0.2 kg CO_2 eq, and the

Table 2
Parameter definitions for possible manufacturing pathways of nanoink printed front contacts in photovoltaic cells. T variables identify the triggers (Fig. 2) and π values the probability for the least likely unit process in the competing pair.

T	Description	π	Expected chance of success	Uncertainty about chance of success π: type	Uncertainty about chance of success π: parameters	Justification
T1	Synthesis route for Cu nanoparticles. Success = chemical route, failure = physical route.	π1	0.7	Triangular	Min: 0.5 Mode: 0.7 Max: 0.8	Chemical methods provide more control over particle size and shape, which may ultimately be more important for the nanoink.
T2	Synthesis route for Ag nanoparticles. Success = chemical route, failure = physical route.	π2	0.7	Triangular	Min: 0.5 Mode: 0.7 Max: 0.8	Chemical methods provide more control over particle size and shape, which may ultimately be more important for the nanoink.
T3	Sintering method for Cu nanoink. Success = chemical sintering, failure = laser sintering.	π3	0.2	Triangular	Min: 0.1 Mode: 0.2 Max: 0.3	Based on initial trials, the chemical sintering method had not performed as well as the laser methods. In addition to this, it may be easier to upscale the laser process.
T4	Sintering method for Ag nanoink. Success = thermal sintering, failure = laser sintering.	π4	0.5	Uniform	Min: 0 Max: 1	At the time of assessment, there was no particular indication of the performance of each method.
T5	Metallic nanoink used for seed printing of front contacts. Success = Cu nanoink, failure = Ag nanoink.	π5	0.8	Triangular	Min: 0.5 Mode: 0.5 Max: 0.8	Based on preliminary tests for technical feasibility, copper-based nanoink seemed “more promising”, while silver-based nanoink was not discarded.

impact will remain below 0.167 kg CO_2 eq with 95% confidence. These and other statistics are summarized in Table 3.

The boxplot in Fig. 4 shows the mean and confidence intervals for the *differences* in impact scores, relative to the reference system and for the four impact categories investigated. A positive percentage value (above the dotted red line) means a higher impact score for the nanoink printed cells. The medians (central black lines) of all values are higher, suggesting a slightly worse performance for the nanoink-printed cells. However, the difference in performance does not appear to be strongly conclusive, given that an important part of the boxes (25th and 75th percentiles) in all cases remains below 0%.

In order to discern whether these differences were statistically significant or not, we used the modified null hypothesis significance test (Heijungs et al., 2016) with an alpha-value of 0.05 and a d-value of 0.2. The test concluded that only the climate change and freshwater ecotoxicity impact scores of the reference screen-printed cell was lower. For the other impact categories, the differences were not statistically significant.

3.2. Global sensitivity analysis (GSA)

The Borgonovo delta sensitivity measures (Borgonovo, 2007) are listed for the stochastic triggers and other uncertain foreground parameters in Table 4. The most important contribution to variance in the climate change impact score comes from trigger T3, which selects between the chemical and laser sintering for the copper nanoink pathway. This is followed in order of importance by trigger T5, which selects between the copper and silver nanoink front contacts for the cell. The third most important parameter was not a trigger, but the amount of gas mixture that could be used to treat each cell in the chemical sintering procedure. The three most sensitive parameters are therefore in the copper nanoink with chemical sintering route. These can all be traced to the potentially very large impact contribution that can result from formic acid consumption in the chemical sintering route for copper.

Table 3
Statistical descriptors for the impact score distributions of the nanoink-printed PV system (nano) and the conventional screen-printed system (ref).

Statistical parameter	Nanoink printed system	Ref system
Climate change (kg CO2 eq)		
Arithmetic mean	0,088	0,077
Geometric mean	0,083	0,076
Median	0,077	0,075
5% confidence interval	0,064	0,057
95% confidence interval	0,167	0,103
Ozone depletion (kg CFC-11 eq)		
Arithmetic mean	1.73E-08	1.54E-08
Geometric mean	1.62E-08	1.50E-08
Median	1.50E-08	1.49E-08
5% confidence interval	1.17E-08	1.03E-08
95% confidence interval	3.25E-08	2.25E-08
Human toxicity (kg 1,4 DCB eq)		
Arithmetic mean	0.229	0.212
Geometric mean	0.185	0.173
Median	0.170	0.159
5% confidence interval	0.085	0.081
95% confidence interval	0.534	0.502
Freshwater ecotoxicity (kg 1,4 DCB eq)		
Arithmetic mean	0.0026	0.0024
Geometric mean	0.0024	0.0022
Median	0.0023	0.0021
5% confidence interval	0.0013	0.0013
95% confidence interval	0.0049	0.0043

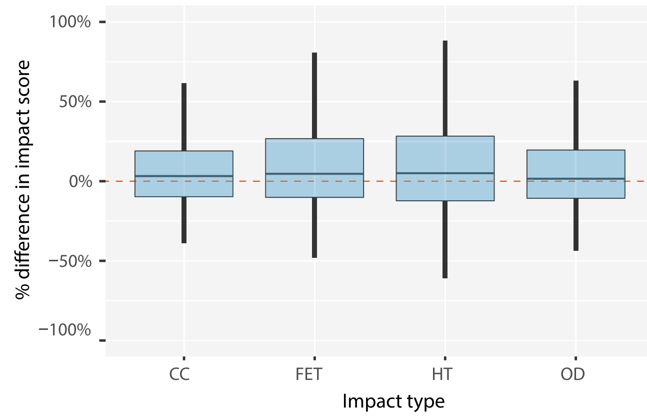

Fig. 4. Distribution of difference in impact scores of nanoink-printed cell, relative to the impact score of the screen-printed cell (ref). CC: Climate Change; OD: Ozone Depletion; HT: Human Toxicity; FET: Freshwater Ecotoxicity.

Table 4
Delta sensitivity measure estimates for the climate change impacts of the PV system with nanoink printed front contacts.

Uncertain input parameter	δ est.	Rank
π1: Chance of success of T1	0.01	10
π2: Chance of success of T2	0.00	6
π3: Chance of success of T3	0.02	5
π4: Chance of success of T4	0.02	4
π5: Chance of success of T5	0.02	9
T1: Chem. vs. phys. synthesis of Cu nanoparticles	0.00	12
T2: Chem. vs. phys. synthesis of Ag nanoparticles	0.01	11
T3: Chem. Vs. laser sintering: Cu ink	0.20	1
T4: Thermal vs. laser sintering: Ag ink	0.01	13
T5: Cu vs. Ag printed front contacts	0.10	2
Qty. of gas mix required for Cu nanoink sintering	0.04	3
Qty. of solution required for electroplating	0.01	7
Cell conversion efficiency increase	0.01	8

3.3. Factor fixing

With the sensitivity ranking obtained from the GSA, we proceeded to factor fixing (Saltelli et al., 2008) in order to investigate further how the environmental profile of the technology would change if the most sensitive parameters were fixed. In this case, we tested trigger T3, which by the final stages of this study was looking less likely to favour a chemical sintering route for copper nanoink due to various technical challenges. Therefore, we updated T3 to a constant value of 0 so that the laser sintering route was always chosen for copper-based nanoink. We then ran a similar Monte Carlo simulation for the updated system and produced the results shown in Fig. 5.

With all other triggers left to vary freely, the impact profile of this updated technology improved considerably. The peak around 0.15 kg CO_2eq disappeared, and the spread of the impact score distribution diminished noticeably. The geometric mean of the climate change impact score for the updated system decreased by 10% (75 g CO2 eq) and the 95% confidence interval by 46% (90 g CO2 eq). The geometric means for ozone depletion, human toxicity and freshwater ecotoxicity decreased by 15%, 3% and 8% respectively.

We performed a similar significance test on the updated results in order to confirm if − under these new constraint −statistically significant differences could be observed. The results indicate that discarding the chemical sintering of copper nanoink as an optional pathway results in a statistically significantly lower climate change impact score for the nanoink-printed cells vs. the conventional screen-printed cells. For other impact categories, there are no statistically significant differences. The calculations and results of all

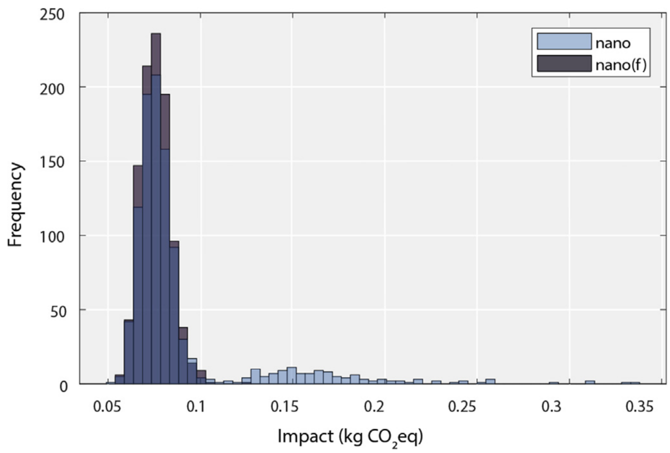

Fig. 5. Comparison of climate change impacts of a PV system with nanoink-printed cells with both laser and chemical sintering alternatives for copper nanoink (nano) and with only laser sintering alternative for copper nanoink (nano(f)).

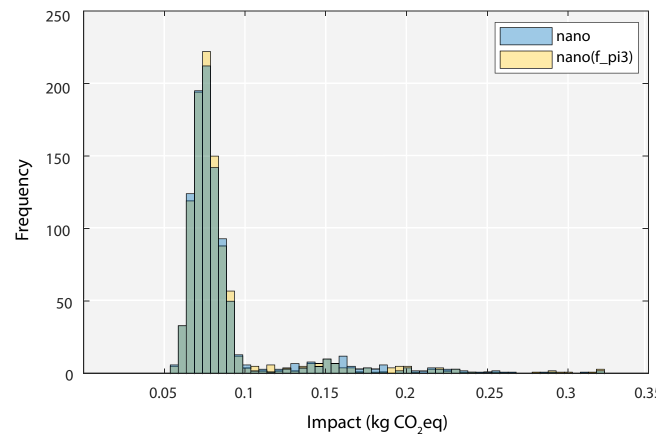

Fig. 6. Comparison of climate change impacts of a PV system with nanoink-printed cells with uncertain chance of success for chemical sintering alternatives for Cu nanoink (nano) and with certain probability of success (nano(f_pi3)).

significance tests are provided in the Electronic Supplementary Information.

3.4. Insights from the application of the method

An important aspect addressed in our method is the fact that the chances of success π are uncertain, and must be determined using subjective criteria to a certain degree. The implementation of Step 7 allowed us to factor this in and investigate the relevance of these uncertainties by including the uncertain parameters π in the global sensitivity analysis. The results of our case study suggested that these second-order uncertainties about the probabilities of success π of each trigger did not have important effects on the model's output variance.

There are theoretical reasons to believe that uncertainty about the probability π has no influence on the overall result in a Monte Carlo type of sampling. After all, when we sample from a binomial distribution with probability π and sample size n (say, 1000), the expected number of times we have chosen a certain technological pathway is $n\times\pi$. When we modify the setup and use a binomial distribution with probability equal to $\pi+\varepsilon$, where ε is, for instance normally distributed with mean 0 and standard deviation σ, the expected number of times we have chosen this technological pathway is $n \times \pi + 0 = n \times \pi$, because the expected value of this normal distribution is 0.

To further verify this, we fixed parameter $\pi 3$ in order to give a certain chance of success for *T3* of 20% and repeated the Monte Carlo simulation. The results are shown in Fig. 6, showing only a very small shift in the distribution curves as expected. Further exploration of this perhaps unexpected finding is out of scope for this study, but we believe worthy of investigation in future work. Nevertheless, addressing uncertain probabilities in the method makes an important step in moving from probability theory to possibility theory (Dubois and Prade, 1988), without yet making the full turn.

4. Conclusions

The application of the probabilistic method to the case study proved that calculation of such a model is feasible and the results fall within expectations as verified by the shapes of Figs. 3–6. Additionally, we demonstrated the important analytical possibilities offered by the method, and successfully addressed the conceptual and practical limitations of the scenario approach for the specific case of uncertain technological pathways. This probabilistic approach better represents the fundamental reality of the technological system under scrutiny when these pathways will only be resolved in a future stage. In early R&D stages, and with the existing state of knowledge of the system, these possible branches of the technology are better represented as a single system with a single range of potential impacts and specific probabilities attached to each value. This interpretation is fundamentally different from making numerous *if/then* conclusions about the system's environmental performance in different scenarios. It can especially provide a more robust basis and —if desired-a more conservative basis (e.g. based on confidence intervals) for considering future environmental impacts in current decisions.

The proposed framing also demonstrated to be better suited for a global sensitivity analysis that allowed us to identify the most sensitive parameters from a wider spectrum of uncertainty sources, including whether the future selection of one unit process instead of another is relevant for the variance in the system's impact score. We were further able to demonstrate -both analytically and experimentally-that uncertainties about the chances of success of each pathway do not influence the results. This is an important takeaway because it affords robustness to the proposed approach while forgoing the need to characterize and incorporate such uncertainties.

The combination of the probabilistic LCA model with GSA can now be used to answer two fundamental questions about the sustainability of an emerging technology in a more robust and realistic way. The first question being whether an emerging technology with unresolved pathways is likely to outperform the incumbent technology, and to what degree of confidence. The second question being to what extent the assessment depends on the chances of success of the technological pathways being pursued.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Carlos F. Blanco: Conceptualization, Methodology, Formal analysis, Software, Investigation, Writing - original draft, Visualization. **Stefano Cucurachi:** Conceptualization, Methodology, Formal analysis, Writing - review & editing, Supervision. **Jeroen B. Guinée:** Methodology, Validation, Writing - review & editing. **Martina G. Vijver:** Writing - review & editing, Supervision. **Willie J.G.M. Peijnenburg:** Writing - review & editing, Supervision. **Roman Trattnig:** Investigation, Data curation, Validation. **Reinout Heijungs:** Methodology, Formal analysis, Writing - review & editing, Validation.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2020.120968.

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