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Uncertainty quantification in the techno-economic analysis of emission reduction technologies: a tutorial case study on CO$_2$ mineralization

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The pathways toward net-zero greenhouse gas emissions by 2050 should be designed based on solid scientific evidence. *Ex ante* system analysis tools, such as techno-economic assessments (TEAs), are key instruments to guide decision-makers. As *ex ante* TEAs of CO$_2$ mitigation technologies embody a high level of uncertainty, the informed use of uncertainty analysis becomes crucial for meaningful interpretation and communication of TEA outputs. To foster enhanced appreciation and the use of uncertainty analysis, we compare multiple uncertainty analysis methods for *ex ante* TEAs, using a case study on CO$_2$ mineralization in the cement industry. We show that local sensitivity analysis tools such as one-way analysis, which are most often used by TEA practitioners, may not suffice for deriving reliable conclusions and provide guidance on how to apply global sensitivity analysis methods, such as variance-based indicators for TEAs in this field.

1 Introduction

Data-driven decision-making on the research and development (R&D) and investment in CO$_2$ mitigation technologies, such as carbon capture and utilization (CCU) technologies, is key to achieving the goal of reaching net-zero greenhouse gas emissions by 2050. However, i) many technologies and systems are still at a low level of maturity, ii) underlying physio-chemical mechanisms have often not yet been fully investigated, iii) the level of process or system design is still preliminary, and iv) future environmental conditions (financial, policy, technology development, societal, *etc.*) cannot yet be fully anticipated. All the approaches and technologies needed to meet our climate goals do not exist at the scale and/or maturity needed. *Ex ante* system analysis tools that embody a high level of uncertainty (Van der Spek et al., 2021; Mendoza et al., 2022), among which techno-economic assessments (TEAs) for the evaluation of economic performance and life-cycle assessments (LCAs) for the evaluation of environmental impacts are needed to guide decision-makers in this process (Cremonese et al., 2020; Strunge et al., 2022a; Langhorst et al., 2022).

The rigorous use of uncertainty analysis methods has been advocated to increase the transparency of techno-economic studies and improve their utility (Van der Spek et al.,

2017a; Van der Spek et al., 2017b; Van der Spek et al., 2020; Rubin et al., 2021; Van der Spek et al., 2021). TEA studies take different forms thorough the development process of the technology, from simplified studies using mass and energy balances to very detailed ones based on high-fidelity technology modeling (and/or measured plant data) and bottom-up costing methods (i.e., by starting with the design and costing of each major equipment) (Van der Spek et al., 2020). In principle, the uncertainty analysis methods used must fit the complexity of the TEA model and its purpose. For instance, an effective design for tax relief programs for CO_2 storage requires TEA models of incumbent technologies, which can appropriately incorporate potential tax reliefs (Fan et al., 2018). Additionally, some uncertainty analysis methods come with high computational costs and data requirements, whereas others are much more straightforward to undertake, both considerations requiring TEA modelers to rationally weigh which method(s) to select for a given case. This is not trivial, and most frequently, the simplest uncertainty analysis methods are selected (i.e., local sensitivity analysis methods). For example, a non-exhaustive review of 21 studies presenting TEAs of CO_2 mineralization processes showed that 11 publications (50%) used simple local sensitivity analysis methods (either one-at-a-time sensitivity analysis (Pedraza et al., 2021) or one-way sensitivity analysis (Huijgen, 2007; Huijgen et al., 2007; Hitch and Dipple, 2012; Pasquier et al., 2016; Naraharisetti et al., 2019; McQueen et al., 2020), and ten publications (45%) did not include any uncertainty analysis (Kakizawa et al., 2001; Iizuka et al., 2004; Katsuyama et al., 2005; O'Connor, 2005; Gerdemann et al., 2007; Eloneva, 2010; Sanna et al., 2012; Pérez-Fortes et al., 2014; Sanna et al., 2014; Mehleri et al., 2015). Only one publication (5%) applied a global sensitivity analysis method (Strunge et al., 2022b). We must acknowledge that more recent studies appeared to be more likely to incorporate some form of uncertainty analysis, highlighting the evolvements of this research field in recent years. Seemingly, when uncertainty analysis is incorporated, methods other than local sensitivity analysis are usually neglected and/or methods are selected without a clear rationale, possibly leading to errors in their use and especially the interpretation of model outputs. A result may be that conclusions are drawn on, for instance, economic viability that is not supported by the performed local sensitivity analysis of the uncertain input data.

Here, we present a tutorial case study where we discuss and show the use of a range of quantitative uncertainty analysis methods to inform TEA practitioners on the different options available, their use and utility, and good and, perhaps, poor practices. Overall, we aim to advance the appreciation and use of uncertainty analysis in the *ex ante* TEA literature to strengthen the quality of the TEAs that are undertaken, leading to a better-informed policy.

As a case study for techno-economic modeling of CCU technologies, we used an integrated TEA model of a CO_2 mineralization process that produces a supplementary cementitious material (SCM) as cement replacement, as reported earlier in Strunge et al. (2022b). We discuss in detail seven common approaches to uncertainty analysis that may be relevant to the TEA of CO_2 mitigation technologies (i.e., one-at-a-time sensitivity analysis, one- and multiple-way sensitivity analysis, scatterplot analysis, rank correlation, variance-based methods, and density-based methods) while acknowledging that many other methods (for specific other applications) have been developed (e.g., classification

tree analysis if an analysis of smaller subsets of the input and output space is necessary or entropy mutual information analysis for non-monotonic relationships (Mishra et al., 2009)).

2 Case study and modeling

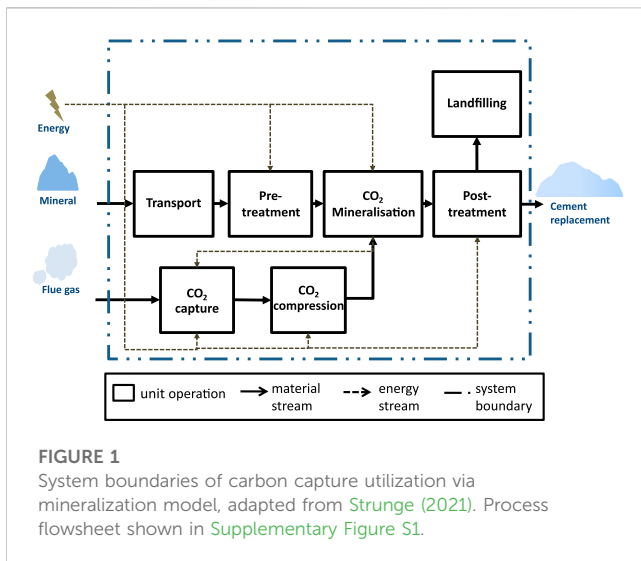## 2.1 CO_2 mineralization for SCM production

Being a major emitter of anthropogenic CO_2 (Favier et al., 2018) with one of the highest carbon intensities per unit of revenue (Czigler et al., 2020), the cement industry needs economically viable solutions to reduce emissions and reach net zero (European Cement Association, 2014; Bellmann and Zimmermann, 2019; Czigler et al., 2020). For this sector, among other strategies, CO_2 mineralization has been proposed as a means of CO_2 utilization, where CO_2 is reacted with activated minerals (e.g., magnesium- or calcium-rich minerals such as forsterite (Mg_2SiO_4) present in olivine-bearing rocks or lizardite ($Mg_3Si_2O_5(OH)_4$) present in serpentine-bearing rocks). As an exemplification, the mineralization reaction of CO_2 with forsterite is shown as follows:

$$Mg_2SiO_4 + 2CO_2 \rightarrow 2MgCO_3 + SiO_2 + \text{heat} \uparrow \qquad (1)$$
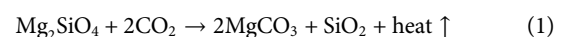
The product [mixture of carbonate (i.e., in the case of forsterite ($MgCO_3$) and silica (SiO_2))] can be used as an SCM in the cement industry. SCMs are materials that can be added to cement blends to archive certain properties or, more commonly, lower the amount of clinker (the cement's main reactive component) needed in cement blends to reduce emissions (Favier et al., 2018). The by-product of CO_2 mineralization silica makes the product mixture a valuable SCM for cement blends. Amorphous silica is a widely accepted pozzolanic additive in cement production. While the main product, carbonate, is inert when added to cement, amorphous silica and calcium hydroxide (CaOH) present in cement react to produce additional binding products (e.g., calcium silicate hydrates), leading to a comparable or increased strength to using cement alone (Wong and Abdul Razak, 2005). Hence, mineralization products not only permanently store CO_2 as carbonates but also reduce emissions by partially replacing conventional cement/clinker production when used as SCMs (Sanna et al., 2012; Sanna et al., 2013; Benhelal et al., 2018; Woodall et al., 2019; Ostovari et al., 2020; Ostovari et al., 2021).

In Strunge et al. (2022b), we showed via integrated techno-economic modeling that the application of CO_2 mineralization for the production of SCM could generate a net profit of up to €${}_{2021}$32 per tonne of cement under certain conditions (i.e., the resulting products must be used as SCMs in cement blends and the storage of CO_2 in minerals must be eligible for emission certificates or similar).

The CO_2 mineralization process considered here is a direct aqueous carbonation approach based on Eikeland et al. (2015) and Gerdemann et al. (2007) (Figure 1; Supplementary Figure S1), in which ground minerals are reacted with captured CO_2 in a pressurized stirred tank using an aqueous slurry with additives. We advanced this process by designing a post-processing train that i) partially separates unreacted minerals via gravity separation and ii) separates magnesium carbonate ($MgCO_3$) from the reaction

FIGURE 1
System boundaries of carbon capture utilization via mineralization model, adapted from Strunge (2021). Process flowsheet shown in Supplementary Figure S1.

products, able to produce SCMs with different properties [i.e., different silica (SiO_2) contents] (Strunge et al., 2022b; Kremer et al., 2022). We selected the conditions with the lowest costs as the nominal case (i.e., olivine-bearing rocks were used as feed minerals, the reaction pressure was set at 100 bar, and the reaction temperature was set at 190°C) (Strunge et al., 2022b). For this case study, the mineralization plant was assumed to be located at the cement plant's site (located in the north of Germany) to reduce the costly transport of flue gas or CO_2. As feed minerals (i.e., olivine-bearing rocks) are currently mined in Norway, Italy, Greece, or Spain (Kremer et al., 2019), they are transported to the mineralization plant, where they are first mechanically activated via crushing and grinding (pre-treatment), followed by the mineralization in continuously stirred reactors under elevated pressure and temperature, in an aqueous slurry with carbonation additives (i.e., NaCl and $NaHCO_3$). CO_2 is introduced in gaseous form into the mineralization slurry after being separated from the flue gas via monoethanolamine (MEA) post-combustion capture. Following the reaction, the slurry and unreacted minerals are recycled, and the products are purified (post-treatment) to produce an SCM for the cement industry (Figure 1). This purification step is needed as the carbonation reaction produces magnesium carbonate and silica. Because the former is inert when blended with cement, thus reducing its compressive strength, and the latter reacts with cement (i.e., increasing its strength), the silica content has to be increased through purification to use the carbonation products as SCM (Bremen et al., 2022; Strunge et al., 2022b; Kremer et al., 2022). Consequently, some of the inert products must be landfilled (e.g., in the limestone quarry) (Figure 1).

The integrated TEA model, as is commonly the case, combines multiple approaches. We calculated mass and energy balances from first principles (e.g., energy transfer for heat exchangers) in combination with literature values (e.g., energy demand for grinding). The reaction conditions (e.g., pressure, temperature, and concentration) and resulting yield were based on literature values. A post-processing train did not exist yet and was therefore designed and subsequently simulated on Aspen Plus (Strunge et al., 2022b).

2.2 TEA model implementation

We described the methodology of the used TEA model in depth earlier in Strunge et al. (2022b). Hence, the following gives only a short overview of the approach followed. The model was developed following recent guidelines for TEA in CCU (IEAGHG, 2021; Rubin et al., 2021; Langhorst et al., 2022). The performance indicator chosen for this assessment was the levelized cost of product (LCOP) in €$_{2021}$ per tonne SCM$_{CCU}$ produced. This indicator combines the total capital requirements (TCR) and operational expenditures (OpEx). We discounted the capital costs using the interest rate i and the lifetime of the plant L to evaluate the real cost of capital for the proposed plants as follows (Smith, 2005):

$$LCOP = \alpha \cdot TCR + OpEx, \tag{2}$$

$$\alpha = \left(\frac{i}{1 - (1 + i)^{-L}} \right). \tag{3}$$

We calculated the TCR building up from the total direct cost (TDC) and total overnight cost (TOC) (Eqs 4, 5, 6):

$$TOC = \sum TDC \cdot \left(1 + f_{indirect}\right) \cdot \left(1 + f_{process}\right) \cdot \left(1 + f_{project}\right)$$
$$\cdot \left(1 + f_{owner}\right). \tag{4}$$

Here, $f_{indirect}$, $f_{process}$, $f_{project}$, f_{owner} represent indirect costs, process contingencies, project contingencies, and owners' costs. To calculate the TCR for an nth-of-a-kind plant, we used the following equations (Rubin et al., 2013; Rubin et al., 2021):

$$TCR = \left(\frac{TOC}{\dot{m}_{SCM}} \right) \cdot N^{-E} \cdot \dot{m}_{SCM} \cdot (1 + i)^{t_{construction}} \tag{5}$$

$$E = \frac{\ln(1 - LR)}{\ln(2)}. \tag{6}$$

N characterizes the number of plants built, LR the learning rate, E the experience factor, i the interest during construction, and $t_{construction}$ the estimated time for construction.

We estimated OpEx using mass and energy balances as a basis to calculate the costs of utilities and feedstocks and the costs of material transport:

$$OpEx = \sum w_i \cdot \pi_i + \dot{m}_{mineral,in} \cdot \sum \pi_j \cdot d_j + OpEx_{fixed}, \tag{7}$$

where the amount of feedstock or utility needed is represented by w_i, π_i is the price of feedstock or utility, π_j is the price of transportation means (i.e., truck, train, or ship), and d_j is the distance for material transported. OpEx$_{fixed}$ consists of insurance and local taxes, maintenance costs, and labor, which we derived on a factorial basis from TPC and the plant's capacity (Peters et al., 1991; Anantharaman et al., 2018).

The model was specified in MATLAB 2019b, which allowed for combining the technical and economic performance estimation into one model and running local and global sensitivity analysis methods on the integrated TEA model. For all global uncertainty analysis calculations, we used UQLab v1.4.0 (Marelli and Sudret, 2014), which is fully MATLAB-based, to easily link it to the TEA model.

TABLE 1 Used input variables for the uncertainty analyses. *Only used in local sensitivity analysis because these variables are dependent on each other (e.g., yield of the reaction increases with pressure not modeled in the TEA model).

Category	Description	Abbreviation/ symbol
Process assumptions	Recycling of the reaction solution (additives + water): the recycling rate determines how often the solution of water and additives can be recycled in the process. High recycling rates lead to lower operational costs	$Add. Rec$
	Reaction rate: the reaction rate determines the time needed to obtain a desired yield and thus with smaller rates the size of the reactors rise, leading to higher capital and operational expenditures	$k_{reaction}$
	Pressure*: elevated pressure of the reactor is necessary to increase the reaction rate via the dissolution of CO_2 in the slurry, where it can react on the mineral particle's surface. Higher pressures lead to higher capital and operational expenditures for compression	$P_{reaction}$
	Temperature*: increased temperature is needed to initiate the reaction. Higher temperature needs lead to higher capital costs for heat exchangers and higher operational costs for natural gas	$T_{reaction}$
	Yield of the reaction*: a higher yield will lead to longer residence time in the reactors and hence increase capital and operational expenditures for the reactors but will require fewer separation steps after the reaction to purify the product, which decreases the capital and operational expenditures for the separation step	$Yield$
	Solid–liquid ratio in slurry: the solid–liquid ratio determines the amount of solid in the slurry. Higher values reduce the amount of water and additives needed and require smaller reactors, leading to a decrease in capital and operational expenditures for these accounts, as the density of the slurry increases operational expenditures for stirring rise	$X_{S/L}$
	Unreacted mineral recovery rate: after the reaction, a share of the unreacted mineral is recovered and fed back to the reactor. Higher values increase electricity demand and capital costs but reduce costs for feed minerals	$X_{unreacted\ mineral}$
	Silica content in the SCM_{CCU}: the SCM produced by this process is a mixture of SiO_2 and inert material (i.e., unreacted mineral and magnesium carbonate). Higher contents of SiO_2 in the SCM require an increased purification effort because the process produces more carbonates than silica. Higher contents lead to increased capital and operational expenditures	X_{SiO_2}
	Purity of feed mineral: the feed minerals are of natural origin; they can contain impurities (e.g., Fe_2SiO_4), which may act inert when reacted with CO_2. Higher purities will decrease both capital and operational expenditures	$p_{mineral}$
Capital expenditures assumptions	Lifetime of the plant: the lifetime determines how many years the plant operates. Long lifetimes will reduce annualized capital expenditures	L_{plant}
	Number of plants to reach NOAK: the model estimates the costs of the mature plant, with costs decreasing through learning effects with each iteration of building a plant. This value determines how many plants will have to be built to reach the nth-of-a-kind (NOAK). Higher values decrease capital expenditures	$No\ of\ plants$
	Learning rate: the learning rate factors in cost reductions archived though iterative learning. Higher values decrease capital expenditures	$Learning\ rate$
	Combined process and project contingencies: the contingencies used in the model combine both project and process contingencies. Higher values of contingencies increase capital expenditures	$Contingencies$
	Overall interest rate on capital: the interest rate combines interest on loans and equity needed to obtain the capital for constructing a plant. Interest will be paid over the entire lifetime of the plant. Higher interest rates lead to higher annualized capital expenditures	i
	Operating time of the plant per year: the operation time determines how much the plant is running on full load. Higher operation times will require a smaller plant for the same output and hence will reduce capital expenditures	$t_{operating}$
Prices of utilities, feedstock, and product	Price of electricity	$\pi_{electricity}$
	Price of natural gas	$\pi_{natural\ gas}$
	Price of feedstock mineral	$\pi_{mineral}$
	Price of sodium bicarbonate	π_{NaHCO_3}
	Price of sodium chloride	π_{NaCl}
	Price of MEA	π_{MEA}
General	Transport distance: distance from the mining site of feed mineral to the cement plant. Higher values increase operational expenditures	$Transport\ distance$

2.3 Quantity of interest and selected input variables for the uncertainty analyses

For the case study, we used the output variable LCOP as the so-called quantity of interest (a term often used in uncertainty quantification for the output parameter of which the sensitivity is tested) to compare the different uncertainty analysis methods. We chose to vary the following input variables (Table 1).

3 Uncertainty analysis methods in TEA

This section discusses the uncertainty analysis method we investigated here. It first gives a general introduction to uncertain TEA problems before discussing local sensitivity analysis in more detail. It then introduces global sensitivity analysis methods and approaches to characterize uncertainty and variability in model inputs.

A general formulation of an uncertain TEA problem can be specified as a function $g(\mathbf{x})$, where $\mathbf{x} = (x_1, x_2, \ldots, x_n)$ represents the input space (e.g., process variables and feedstock prices) and $\mathbf{y} = (y_1, y_2, \ldots, y_m)$ the model's output space (e.g., LCOP and profit). The initial or base values of the input space are denoted as $\mathbf{x}^0 = (x_1^0, \ldots, x_n^0)$ and the base value output space as $\mathbf{y}^0 = (y_1^0, \ldots, y_n^0)$:

$$\mathbf{y} = g(\mathbf{x}) \, with \, \mathbf{y}^0 = g(\mathbf{x}^0). \tag{8}$$

As many parameters or variables of the input space x^0 are not fully known (e.g., the actual yield of the process when in operation), TEAs require assumptions to be made for some of the uncertain inputs.

A helpful categorization of uncertainty was suggested by Rubin (2012), who distinguishes between "uncertainty," "variability," and "bias." Although true uncertainty means the precise value of a parameter is not yet known (e.g., reaction yield of the process at scale), variability simply means a variable can take on different values (e.g., over a time period or at different locations) and the modeler chooses one (e.g., the temperature in a certain location). The principal difference between uncertainty and variability is that the precise value of uncertain parameters is not known, nor is the probability of the parameter taking on a certain value, whereas variable parameters are known or at least knowable, allowing quantification of a probability density function. This does not mean an uncertain parameter should not be quantified, they can and need to be, but their quantification is a guess or good estimate at best, rather than a (series of) measured value(s) *per se*. The uncertainty analysis methods discussed in this study can be used to assess uncertainty and variability, and hereafter, we refer to both simply as uncertainty. Bias refers to assumptions that (intended or unintended) change the results (e.g., choosing the highest or lowest reported reaction yield for the assessment of a chemical process) (Rubin, 2012; Van der Spek et al., 2020). Bias analysis is challenging as only third-party reviews of a study's assumptions, and their reasoning might be able to detect these (Rubin, 2012).

The goals of uncertainty analyses can be manifold, including testing the robustness of a model and providing insight into changes in outputs due to changes in inputs and their probabilities to determine key drivers of uncertainty and gain insights into the

strength of the model or its input data (Saltelli et al., 2008; Van der Spek et al., 2020). In this study, we focused on the commonly used goal to determine key drivers of uncertainty on the output of the model (i.e., creating a ranking of the most influential parameters on the model's uncertainty) and investigated their input–output relationships, frequently called sensitivity analysis (Mishra and Datta-Gupta, 2018). In the following, we give an overview of seven suggested sensitivity analysis methods (Table 2).

3.1 Local sensitivity analyses

Local sensitivity analyses (LSA) are the most commonly used methods in *ex ante* TEA, in which one or multiple input variables are varied around a base value. The simplest form of local analysis is the one-at-a-time (OAT) local sensitivity analysis, in which input variables of interest x_i are varied using two realizations (e.g., ± 10%, ± 50% or a defined minimum and maximum) around the base value x_i^0 (Sagrado and Herranz, 2013; Van der Spek et al., 2020). Overall, given the local nature of the OAT analysis, selecting a small range for the variation (e.g., ± 10% or ± 15%) is recommended to investigate the local sensitivities of the model around the base values. The resulting output values with varied inputs allow the modeler to test the local robustness of the model and determine the strength of a local input–output relationship (e.g., a strong relationship is present if varying x_i by ± 10% leads to the high increase/decrease of y_j compared to the base value y_j^0) and the direction of the relationship (e.g., when an increase in x_i leads y_j to increase from y_j^0, we call it a positive relationship). The nature of the relationship (e.g., linear or nonlinear) cannot be fully investigated with OAT. A main advantage of OAT lies in its computational costs, where the model for n relevant input variables needs to be run $C = (2 \cdot n + 1)$ times, which translates for 10 variables into 21 runs to be computed. We utilized the following definition for OAT indicators (Ikonen, 2016):

$$OAT\left[y_{i,j}\right] = \frac{\left|y_{i,j}^+ - y_{i,j}^-\right|}{y_j^0}. \tag{9}$$

We can then plot the output responses for each varied input $\frac{y_{i,j}^\pm}{y_j^0}$ in, for instance, a tornado graph while using the OAT indicators as a ranking order (i.e., the variable with the highest value of its OAT indicator is shown at the top, followed by the second highest value on the second spot from the top).

Another commonly used approach is one-way local sensitivity analysis, which is an extension of the OAT approach. Instead of using only two realizations of each input variable around the base value, a modeler varies the input variables using predefined intervals (e.g., ten steps in the predefined interval of ± 10% of x_i^0). This approach has higher computational costs and needs $C = (2 + k) \cdot n + 1$ runs, with the number of added steps k (excluding the extremes) and the number of relevant input variables n. For 10 variables with eight added steps (excluding extremes), this translates to 101 model runs. Spider plots are a common way to present the results of the one-way sensitivity analysis, in which the strength, direction, and nature of the local input–output relationship can be determined through a comparison of the slopes (Mishra and Datta-Gupta, 2018).

TABLE 2 Uncertainty analysis methods considered in this publication.

	Local sensitivity analysis			Global sensitivity analysis			
	One-at-a-time	One-way	Multiple-way	Scatterplot-analysis	Spearman rank correlation	Sobol (variance-based)	Borgonovo (density-based)
Using a Monte Carlo simulation	✗	✗	✗	✓	✓	✓	✓
Short description	Changing one input (x_i) at-a-time using two extremes (Borgonovo and Plischke, 2016)	Changing one input (x_i) at-a-time over an interval. (Borgonovo and Plischke, 2016)	Changing multiple inputs $x_i, \ldots, x_j$ at-a-time over an interval (Van der Spek et al., 2020)	Visual analysis of input–output scatterplots (Mishra and Datta-Gupta, 2018)	Regression over ranked input–output pairs (Marelli et al., 2021)	Measuring how much an output variance could be reduced if inputs $x_i, \ldots, x_j$ were fixed (Saltelli et al., 2008)	Measuring the distance between the output density when no inputs are fixed and the density where x_i is fixed (Borgonovo and Plischke, 2016)
Advantages	Computationally cheap	Nonlinearities can be detected	Simple interaction effects can be detected	Visual interpretation possible	Comparably simple to compute	Interaction and total effects can be analyzed in depths	Moment independent
Disadvantages	Only one or a few inputs are varied at-a-time			Computationally costly			

A drawback of these two methods is that no interaction effects can be investigated, as in each run, only one variable is changed at a time (Borgonovo and Plischke, 2016; Van der Spek et al., 2020; Van der Spek et al., 2021). However, most TEA models are likely to contain interaction effects because they often contain related parts. For example, process variables that impact capital expenditures might have a different influence on the LCOP depending on the interest on capital. To tackle this in a simple way, multiple-way sensitivity analysis can be used (Borgonovo and Plischke, 2016). Like the one-way local sensitivity analysis, input variables vary along a predefined interval, but instead of only varying one input variable, multiple variables are changed at a time. For example, variable pairs are varied in a two-way approach, and variable triplets are varied in a three-way approach. This allows modelers to identify combinations of variables with a high impact on the output that might not have been discovered using one-way analysis. The computational costs are significantly higher compared to the other local sensitivity analysis measures. This analysis requires $C = (2 + k)^q \cdot \left(\frac{n!}{(n-q)! \cdot q!}\right) + 1$ runs, with q being the number of ways of the analysis. Hence, two-way analysis ($q = 2$) for 10 variables and eight added steps requires 4,501 model runs. To analyze the results graphically, for a two-way sensitivity analysis, the same graphical approach can be used as described for the one-way or OAT local sensitivity analysis, albeit in three dimensions. For higher dimensions, the values themselves must be evaluated (e.g., setting a maximum/minimum value for y_i and collecting all combinations of x_i that lead to this realization).

3.2 Global sensitivity analyses

Although local sensitivity analyses require comparably low computational costs, a major drawback is their limited ability to consider probabilities; hence, some considered realizations (e.g., ± 15%) might be arbitrary (Saltelli et al., 2008). Therefore, global or probabilistic sensitivity analyses (GSA) are frequently suggested (Borgonovo and Plischke, 2016; Van der Spek et al., 2020; Van der

Spek et al., 2021). In contrast to local sensitivity analysis methods, global sensitivity analysis methods study how variations in probabilistic output parameters can be attributed to different probabilistic input parameters (Mishra et al., 2009). Although some of these input–output relationships could be solved analytically in theory, due to the complexity and heterogeneity of TEA models (i.e., they often comprise multiple different connected models), global sensitivity analysis methods are commonly conducted using statistical techniques coupled with a random or quasi-random sampling of the model (Hastie et al., 2009). Here, probabilities for the variables being a certain realization (i.e., probability density functions, PDFs) need to be assigned, for example, by estimation (called uncertainty characterization). Then, using Monte Carlo (or quasi-Monte Carlo) sampling (MCS), samples of the input variables are drawn simultaneously, and the resulting outputs of the models are calculated (called uncertainty propagation). The simplest method of uncertainty propagation is random sampling. More sophisticated approaches to sampling (e.g., Latin hypercube sampling or low discrepancy series sampling using Halton or Sobol sequence) have been developed to avoid that simple random sampling leads to clustering in some areas of input space, thus underrepresenting other areas (Saltelli et al., 2008). Following uncertainty propagation and utilizing the obtained input and output space, input–output relationships can be determined, and variables with high importance are identified (called uncertainty importance evaluation) (Mishra and Datta-Gupta, 2018).

3.3 Uncertainty characterization

The uncertainty characterization of the input parameters is arguably the most important step in probabilistic uncertainty/sensitivity analysis and requires experience and careful balancing of real knowledge of the uncertainty *versus* the ambitions of the modeler (e.g., a modeler might be drawn to choosing overly optimistic or pessimistic values to fit their goal). Often, we are

inclined to fit probability density functions to collected data without accounting for its quality (e.g., completeness), leading to the propagation of incorrect or incomplete uncertainty, directly impacting the PDF of the parameter of interest. Care should be taken, for instance, to maintain PDFs within a range that is physically possible. For example, many quantities have natural limits (e.g., you cannot have a negative number of people). Multiple methods have been suggested to assist the modeler in selecting their uncertainty characterization (Harr, 1984; Hawer et al., 2018; Mishra and Datta-Gupta, 2018; Van der Spek et al., 2020; Van der Spek et al., 2021). We here exemplify three approaches that can help TEA practitioners define reasonable PDFs: i) a decision tree by Hawer et al. (2018), ii) the maximum entropy principle (Harr, 1984), and iii) simply choosing a uniform distribution for all inputs.

  i) Hawer et al. (2018) (in the following referred to as Hawer's method) provided a useful decision tree that gives suggestions on the probability density functions that should ideally be assigned, mainly depending on the data quality and nature (e.g., discrete or continuous) of the input parameter (Supplementary Figure S2). The decision tree guides the modeler to assign PDFs either subjectively by relying on assumptions on a potential distribution (e.g., through assessing the likelihood to have outliers in the dataset) or when more data are available, more objectively (e.g., through estimating a PDF using a maximum likelihood method on a dataset). This uncertainty characterization method can result in many different PDFs being assigned to the input variables (e.g., uniform, triangular, normal, logistic, and lognormal). Although we find this a comprehensive method for assigning PDFs, many options require detailed knowledge of the data, which may not always be publicly available for new or commercially sensitive technologies.
  ii) An approach that requires slightly less knowledge is the maximum entropy principle, where five different types of PDF are assigned (uniform, triangular, normal, beta, and Poisson) (Harr, 1984), subject to known constraints in the available data (e.g., the bounds and mean, Supplementary Table S1). The general idea of this approach is to use all available information but not to add assumptions to estimate the PDFs (Mishra and Datta-Gupta, 2018). In comparison to Hawer's method, the maximum entropy principle uses fewer options for describing the input data.
  iii) The simplest method of assigning probability densities is assigning a uniform distribution in which all realizations have the same probability. This approach requires the least knowledge of the input parameters and can be performed by only knowing or defining a range for each input variable.

Although all approaches to assigning a PDF aim to harmonize under which conditions a certain PDF is assigned, modelers' choices and interpretation of the underlying data quality inherently introduce a bias, which needs careful consideration. A method to reduce this bias can be the definition of several subjective probability distributions by multiple experts (Mishra and Datta-Gupta, 2018). Here, the modeler relies on multiple experts to, for example, estimate quantiles of distribution (e.g., minimum value relates to the 0th quantile and maximum value

relates to the 100th quantile). Although this method can reduce the modeler's bias, many TEA practitioners might not have access to experts for performing these estimations. Therefore, this approach is not discussed further in this article. In any case, TEA practitioners and the users of TEA results should always consider that even well-quantified PDFs only represent reality but may not capture it completely.

3.4 Uncertainty importance evaluation

Several approaches have been developed to describe the uncertainty importance of the output space $\boldsymbol{y}$ (Borgonovo, 2007; Borgonovo and Plischke, 2016; Mishra and Datta-Gupta, 2018). In this study, we focused on a number of common approaches, namely, scatterplot analysis, rank correlation, variance-based methods, and density-based methods (Borgonovo and Plischke, 2016). The main difference between these approaches is their underlying assumptions. Hence, they are better suited for different applications. A major distinction can be made whether a method is a parametric (i.e., assumptions for the distribution of the inputs are made when calculating the input ranking) or non-parametric (i.e., no assumptions for the distribution of the inputs are made when calculating the input ranking) (Hoskin, 2012). In general, parametric measures will have higher accuracy but more stringent requirements on when they can be applied. Of the approaches we discuss here, rank correlation and the density-based measure are non-parametric, the variance-based measure is parametric, and the scatterplot analysis is qualitative.

3.4.1 Scatterplot analysis

Scatterplots are suited to illustrate bivariate relationships and allow a visual determination of input–output relationships. Therefore, we plot the output sample of output j ($\boldsymbol{y_j}$), which was generated by MCS against the realizations of each of the input variables ($\boldsymbol{x_i}$). The strength, direction, and nature of the relationships can be observed visually, where a strong relationship will lead to a smaller variance in the sample (Mishra and Datta-Gupta, 2018).

The computational costs for scatterplot analyses are significantly higher than those for local methods (Mishra and Datta-Gupta, 2018). Although the number of runs will depend on the nature of the model itself, most models will require at least C = 100–1,000 runs to reach convergence (Saltelli et al., 2008; Mishra and Datta-Gupta, 2018).

3.4.2 Spearman rank correlation

The Spearman rank correlation coefficient (SRCC) assesses how well the relationship between the input sample x_i and output sample y_j can be described using a monotonic function (Helton et al., 1991). Here, the nature of the relationship (e.g., linear) does not influence the SRCC. Therefore, it is widely applicable, including to TEA, where Several input–output relationships can be expected. The SRCC is calculated by ranking the inputs and outputs from the Monte Carlo sample. This is performed by assigning each input–output pair $x_{i,l}$ and $y_{i,l}$ the ranks $x_{i,l}^{trans} y_{j,l}^{trans}$, which for **n** variables consist of the ranks $1 \ldots n$ (e.g., if the smallest value of $x_{i,l}$ corresponds to the highest value of $y_{i,l}$, we assign the ranks $x_{i,l}^{trans} = 1$

and $y_{j,l}^{trans} = n$). We calculated the SRCC for each desired input and output pair $x_{i,l}, y_{j,l}$ (e.g., input interest rate and output LCOP) (Helton et al., 1991; Mishra and Datta-Gupta, 2018) as follows:

$$SRCC\left[\boldsymbol{x}_i, \boldsymbol{y}_j\right] = \frac{\sum_l \left(x_{i,l}^{trans} - \bar{x}_i\right)\left(y_{j,l}^{trans} - \bar{y}_j\right)}{\left[\sum_l \left(x_{i,l}^{trans} - \bar{x}_i\right)^2 \sum_l \left(y_{j,l}^{trans} - \bar{y}_j\right)^2\right]^{\frac{1}{2}}}. \quad (10)$$

The underlying assumption for using this measure is that the input–output relationship is characterized by a monotonic function (i.e., no inflection must be present in the relationship) (Helton et al., 1991), which first needs to be established, for instance, by visual inspection of the input–output relation. As the SRCC can be seen as the linear regression between the ranks, a non-monotonic function will not lead to sufficient answers (as the ranks are not linear) (Marelli et al., 2021), or it might not be possible to assign ranks at all if a value appears twice.

The computational costs for this method again depend on the nature of the model and require at least C = 100–1,000 runs (Saltelli et al., 2008; Mishra and Datta-Gupta, 2018).

3.4.3 Sobol indices

Variance-based methods assess how the expected variance of the output model changes when knowing an input realization with certainty. In the variance-based method, the indices by Sobol (1993) are commonly used (Borgonovo and Plischke, 2016). These variance based indices allow modelers not only to provide a quantitative measurement of the strength, direction, and nature of the global input–output relationship as the first sensitivity (i.e., the effect of one input variable alone on the output space), but also to investigate higher orders of input–output relationships (i.e., the effect of multiple variables collectively on the output space). Usually, the first-order effect and the total order effect are calculated and compared, which can uncover the interaction effects of input variables (which cannot be identified using the other suggested methods) (Borgonovo and Plischke, 2016). As Sobol's variance-based measure depends on a particular moment of the output distribution (its variance), it may lead to misleading results when input variables influence the entire output distribution without significantly influencing the variance (Borgonovo and Tarantola, 2008).

The general idea for calculating Sobol indices lies in the decomposition of the model function (Eq. 11), in which g_0 equals the expected value of $g(x_1 \ldots x_n)$ added with summands of the partial functions of each variable input variable and all their combinations (Sobol, 1993; Marelli et al., 2021):

$$g(x_1 \ldots x_n) = g_0 + \sum_{i=1}^n g_{i(x_i)} + \sum_{1 \le i < q \le n} g_{i,q}\left(x_i, x_q\right) \cdots$$
$$+ g_{1,2,\ldots n}(x_1, \ldots x_n). \quad (11)$$

Following Sobol (1993), we define the total and partial variance for inputs i_1 till i_s as follows:

$$D = \int g^2(x)dx - g_0^2, \quad (12)$$

$$D_{i_1 \ldots i_s} = \int \ldots \int g_{i_1 \ldots i_s}^2 \left(x_{i_1}, \ldots, x_{i_s}\right)dx_{i1} \ldots dx_{i_s}. \quad (13)$$

The first-order and higher-order indices $S_{i_1 \ldots i_s}$ are then defined as the partial variance divided by the total variance (Eq. 14).

Consequently, the total Sobol index S_i^T for a variable i can be calculated as the sum of all indices in which i is present. As this definition is not practical to compute (i.e., all indices must be computed separately), the total index can additionally be derived using the sensitivity index of all variables excluding i, $S_{\sim i}$ (Eq. 15) (Marelli et al., 2021). It is important to note that for this measure, the input variables must be independent (Sobol, 1993; Borgonovo, 2007):

$$S_{i_1 \ldots i_s} = \frac{D_{i_1 \ldots i_s}}{D}, \quad (14)$$

$$S_i^T = \sum_{(i_1 \ldots i_s) \supset i} S_{i_1 \ldots i_s} = 1 - S_{\sim i}. \quad (15)$$

Because the calculation of each partial variance can be cumbersome and quickly makes thousands or millions of calculations be computed, multiple shortcut methods have been proposed (Saltelli et al., 2008; Marelli et al., 2021). For this study, we used the Janon estimator (Janon et al., 2014) (Eq. 16), allowing quick calculation of first-order and total-order effects. We considered two independent Monte Carlo samples $\boldsymbol{x} = (x_1, \ldots, x_n)$ and $\boldsymbol{x} = (x_1', \ldots, x_n')$ resulting in $y = g(\boldsymbol{x})$ and $y^v = g(x_1', \ldots, x_{v-1}', x_v, x_{v+1}', \ldots, x_n')$ and computed them as follows:

$$S_v = \frac{\frac{1}{N}\sum y_i y_i^v - \left(\frac{1}{N}\sum\left(\frac{y_i + y_i^v}{2}\right)\right)^2}{\frac{1}{N}\sum \frac{y_i^2 + y_i^v 2}{2} - \left(\frac{1}{N}\sum \frac{y_i + y_i^v}{2}\right)^2}. \quad (16)$$

The computational costs for Sobol indices are significantly higher than other global sensitivity methods. Using the Janon estimator to calculate first-order and total-order indices, the required model runs are $C = k \cdot (2 \cdot n + 2)$ with n being the number of variables and k being the number of runs for the measure to converge (i.e., at least 100–1,000 runs), translating to at least 2,200–22,000 runs for 10 variables.

3.4.4 Borgonovo indices

Because Sobol indices are moment dependent (i.e., the second moment: variance), Sobol indices cannot sufficiently analyze the sensitivities of inputs if they cannot be fully measured by the variance, which can, for example, be the case if selected PDFs for inputs have long tails (Borgonovo, 2007). Density-based approaches have been developed to counter this, which take the shape of the output distributions and compare it to the shape of the input distributions. Borgonovo (2007) developed the density-based method we used in this study.

We calculated the Borgonovo indices using the conditional and unconditional probability distribution function $f_{y_j | x_i}$ and f_{y_j} of the output $\boldsymbol{y}_j$ for each input variable of interest $\boldsymbol{x}_i$. The general approach for output j and input i can be described as follows (Eq. 17), with $E_{\boldsymbol{x}i}$ being the expected value of $\boldsymbol{x}_i$ (Borgonovo, 2007):

$$\partial_i = \frac{1}{2} E_{\boldsymbol{x}i}\left[\int\left|f_{y_j}\left(y_j\right) - f_{y_j | x_i}\left(y_j | x_i\right)\right|dy\right]. \quad (17)$$

To compute these, we used the histogram-based approach, which is used by default in UQLab (Marelli and Sudret, 2014; Marelli et al., 2021). To approximate the conditional distribution $\boldsymbol{y}_j | \boldsymbol{x}_i$, we drew samples from the input and binned them into classes

TABLE 3 Base case assumptions.

Category	Abbreviation/symbol	Base value
Process assumptions	$Add.Rec$	95%
	$k_{reaction}$	1.4717 s
	$P_{reaction}$	100 bar
	$T_{reaction}$	190°C
	$Yield$	60%
	$X_{S/L}$	12.5 wt%
	$X_{unreacted\ mineral}$	60 wt%
	X_{SiO_2}	40 wt%
	$p_{mineral}$	80 wt%
Capital expenditures assumptions	L_{plant}	30 years
	$No\ of\ plants$	20
	$Learning\ rate$	10.55%
	$Contingencies$	40%, 30%
	i	7.69%
	$t_{operating}$	8,000 h
Prices of utilities, feedstock, and product	$\pi_{electricity}$	62€ MWh^{-1}
	$\pi_{natural\ gas}$	32€ MWh^{-1}
	$\pi_{mineral}$	12 € t^{-1}
	π_{NaHCO_3}	209 € t^{-1}
	π_{NaCl}	61.9 € t^{-1}
	π_{MEA}	1,230€ t^{-1}
General	$Transport\ distance$	1,200 km

of x_i. We computed a distribution of y_j in each of the classes, providing us an approximation of the conditional distribution $f_{y_j|x_i}$. We calculated the unconditional distribution f_{y_j} directly from overall distribution of y_j (Marelli et al., 2021).

The computational costs of this measure are again dependent on the nature of the model but can be expected to be at least C = 100–1,000 runs.
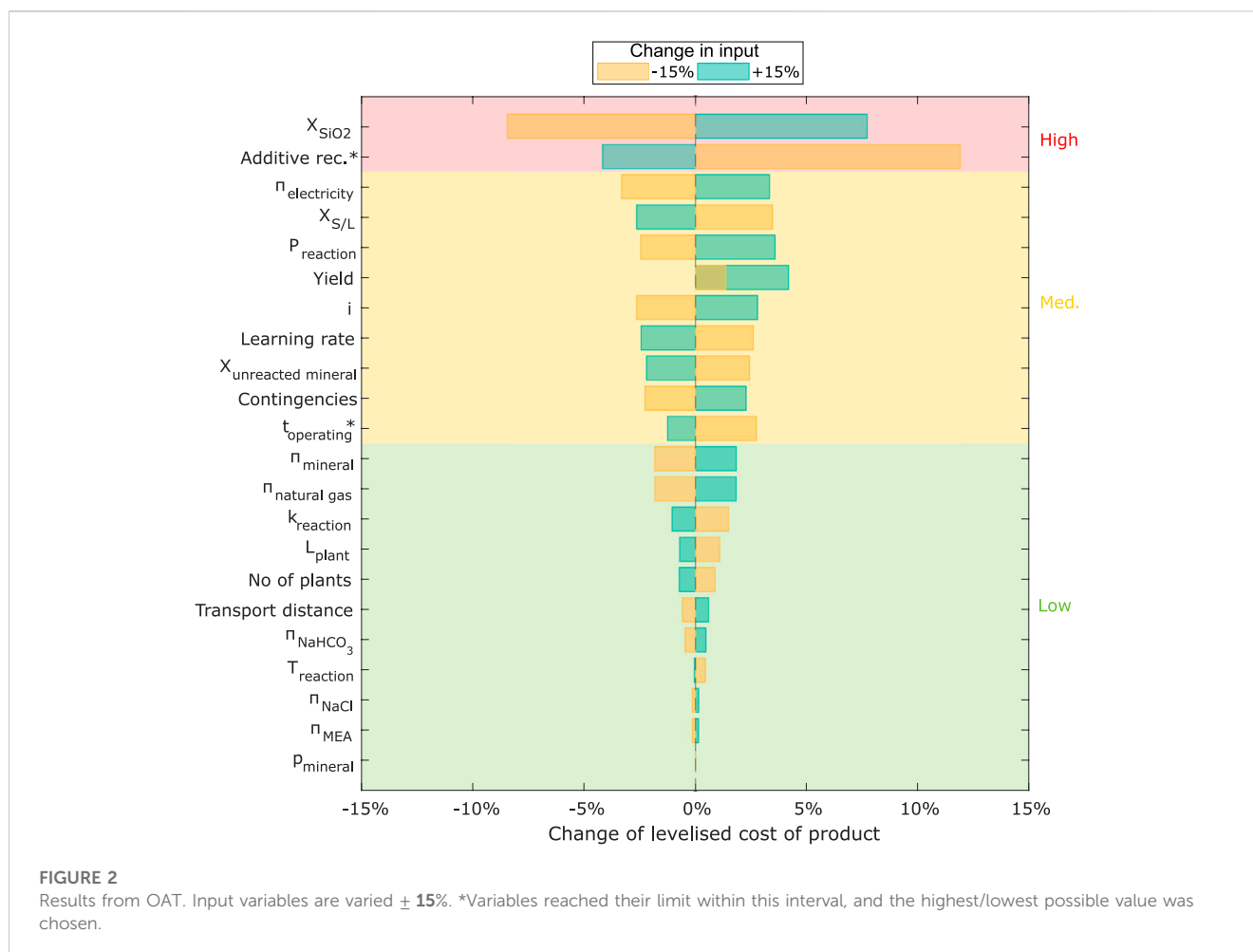
4 Results and illustration of uncertainty analysis methods

This section discusses the implementation of the deliberated uncertainty analysis methods in the mineralization case study. We first calculated the results using the base case assumptions (Table 3) for a mineralization plant with a capacity of 272 kt_{SCM} a^{-1} (this size was chosen to replace 20% of cement of a cement plant producing 1.36 Mt_{cement} a^{-1}), leading to a levelized cost of the product of €129 t_{SCM}^{-1} produced via CO_2 mineralization. As previously discussed by Strunge et al. (2022b), these costs can be offset by replacing cement production and reducing the costs for CO_2 emission certificates (e.g., from the European Emission Trading

System). In the following, we applied the in Section 3 discussed uncertainty analysis tools (i.e., one-at-a-time sensitivity analysis, one- and multiple-way sensitivity analysis, scatterplot analysis, rank correlation, Sobol analysis and Borgonovo analysis) to the case study.

4.1 Exemplification of local sensitivity analysis methods

For the OAT sensitivity analysis, we varied the input variables around the base values by ± 15% (Figure 2). The graph shows observably different local sensitivities of the different input variables. To ease the interpretation of the results, we clustered the local sensitivities following the induced change in the output (Δy_i) subjectively into three categories [high sensitivity ($\Delta y_i \geq 5\% \cap \Delta y_i \leq -5\%$), medium sensitivity ($5\% > \Delta y_i \geq 2.5\% \cap -5\% < \Delta y_i \leq -2.5\%$), and low-to-no sensitivity]. The variables X_{SiO_2} and $Add.Rec$ produce the highest changes of the output variable LCOP (high sensitivity). To put the shown values into perspective, X_{SiO_2} induced the highest change on the LCOP with +12%, which means by increasing X_{SiO_2} by 15%, the

FIGURE 2
Results from OAT. Input variables are varied ± **15**%. *Variables reached their limit within this interval, and the highest/lowest possible value was chosen.

LCOP increases from €129 (base case) to €144 t_{SCM}^{-1}. Variables toward which LCOP shows medium sensitivity are $\pi_{electricity}$, $X_{S/L}$, $P_{reaction}$, Yield, i, Learning rate, and Contingencies. All other variables we can cluster as low-to-no impact on the LCOP. Additionally, the results suggest that the sensitivity toward Yield might not follow a linear relationship because both output bars have the same direction (Figure 2), which needed further investigation using one-way local sensitivity analysis (the following section).

For the one-way local sensitivity analysis, we varied the input values in 10 steps (including the extremes) within the interval of ± 15% around the base values (Figure 3). Visual analysis of the graph reveals that multiple relationships in addition to Yield are nonlinear (i.e., $t_{operating}$, $X_{S/L}$, $P_{reaction}$, and $T_{reaction}$). The detected nonlinearities are not surprising as all the input variables in question (i.e., $t_{operating}$, $X_{S/L}$, $P_{reaction}$, and $T_{reaction}$) influence either the design of the reactor (e.g., increasing the reaction pressure leads to a different wall thickness of the reactor) or other equipment (e.g., heat exchangers), which do not scale linearly in the model. Because a comparison of the slopes is infeasible for variables with different directions of the input–output relationship and the presence of nonlinearities, we used the categories high sensitivity, medium sensitivity, and low-to-no sensitivity with the same boundaries as for the OAT to determine a ranking of the input variables. We ranked the impact after their

highest value in each category [i.e., $\max\left(\left|\frac{y_{i,j}^+}{y_j^0}, \frac{y_{i,j}^-}{y_j^0}\right|\right)$]. This determination of the ranking came to a similar conclusion as the OAT analysis: the variables with the highest impact are $Add.\,Rec$ and X_{SiO_2} followed by the medium impact variables Yield, $P_{reaction}$, $X_{S/L}$, $\pi_{electricity}$, i, $t_{operating}$, and Learning rate. Figure 3 clearly shows that one-way sensitivity analysis is a simple method to identify nonlinearities in input–output relationships. However, a visual determination of the ranking order and deriving impact categories in spider plots can become challenging when dealing with a multitude of input variables of interest.

We performed the two-way sensitivity analysis on the six inputs with the highest impacts (Figure 4). This analysis aimed to investigate which combination of these inputs has a particularly high impact on the output and thus need to be investigated thoroughly. To interpret the results, we again clustered the local sensitivities following the induced change in the output (Δy_i). We used the three categories: high sensitivity ($\Delta y_i \geq 10\% \cap \Delta y_i \leq -10\%$), medium sensitivity ($10\% > \Delta y_i \geq 5\% \cap -10\% < \Delta y_i \leq -5\%$), and low-to-no sensitivity. Note that compared to the categories used in the OAT or one-way analysis, here we chose intervals with cut-off values twice as high (e.g., high sensitivity is defined as $\Delta y_i \geq 10\%$ instead of $\Delta y_i \geq 5\%$), as combinations of factors with high sensitivity will lead to bigger changes.

The results show that nine combinations, including the variables X_{SiO_2}, $Add.\,Rec$, $\pi_{electricity}$, $X_{S/L}$, $P_{reaction}$, and Yield lead to
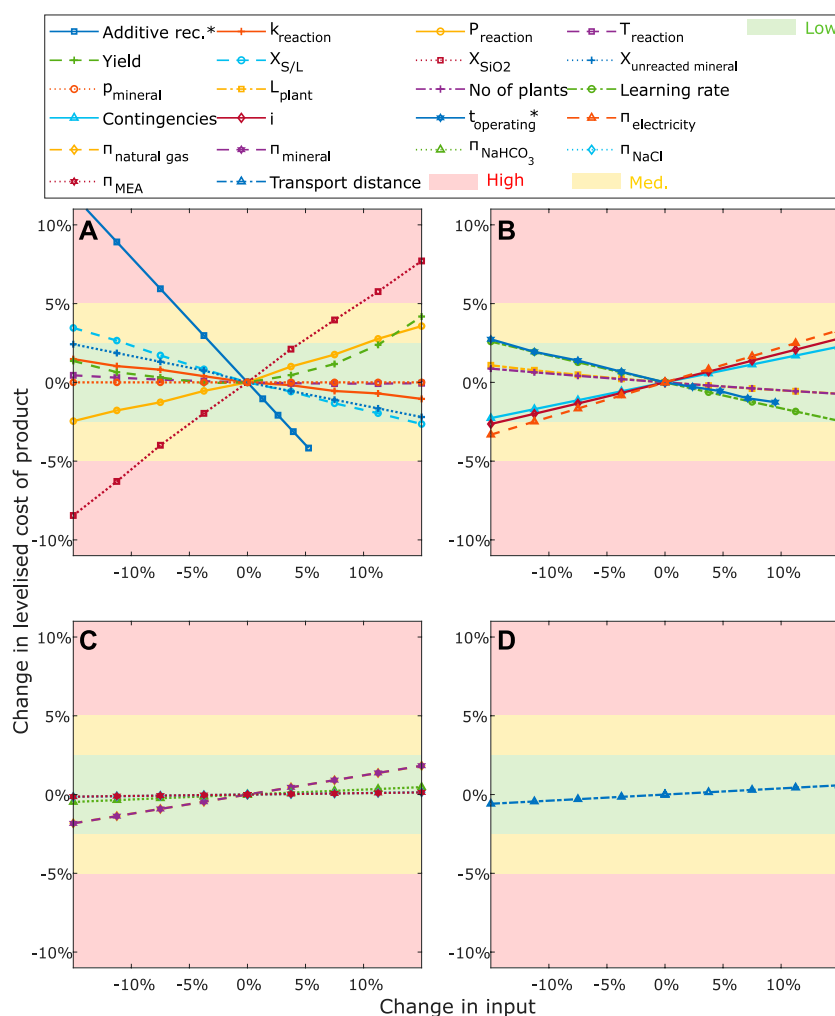
FIGURE 3
Results from one-way LSA. Input variables are varied $\pm$ **15**% for **(A)** process assumptions, **(B)** capital expenditures assumptions, **(C)** prices of utilities and feedstock, and **(D)** general assumptions. *Variables reached their limit within this interval, and the highest/lowest possible value in this interval was chosen.

combinations with high sensitivity (Figure 4). In Figure 4, the areas which are most desirable (i.e., decrease LCOP) or most undesirable (i.e., increase LCOP) are clearly marked. The most undesirable combinations are low X_{SiO_2} and low $Add.Rec$, as well as low $Add.Rec$ and low $X_{S/L}$ (−15% from base value), leading to an increase in LCOP from €129 (base case) to €156 t_{SCM}^{-1} (+21%), as well as €152 t_{SCM}^{-1} (+18%), respectively. For some variable pairs (e.g., $\pi_{electricity}$, $Yield$), we additionally see that much of the mapped space does not lead to large changes in the output (i.e., categorized as low-to-no sensitivity) and hence can be seen of lower priority during the assessment or further research.

4.2 Exemplification of uncertainty characterization methods

Following the exemplification of LSA methods, we applied the aforementioned global sensitivity analysis methods to the case study.

For the comparison of global uncertainty analysis methods, we removed dependent inputs (i.e., $P_{reaction}$, $T_{reaction}$, and $Yield$), which are here onward only represented by reaction rate, as they would have to be changed simultaneously, for which the model is not detailed enough (i.e., no reaction model present). Alternatively, if the model had been more detailed (i.e., including a reaction model), a dependence structure (i.e., copula) could have been used (Soepyan et al., 2018). As discussed in Section 3.2, we first started with the uncertainty characterization, followed by the uncertainty importance evaluation in Section 3.4.

This section illustrates how uncertainty characterization (i.e., the selection of PDFs) can influence the output of a Monte Carlo simulation by applying the three methods discussed in Section 3.3 (i.e., Hawer's method, maximum entropy principle, and assigning uniform distributions) to our case study. With different PDF choices resulting from the three methods, the uncertainty quantification moreover depends on the confidence of the modeler to determine certain moments of the distribution (e.g.,
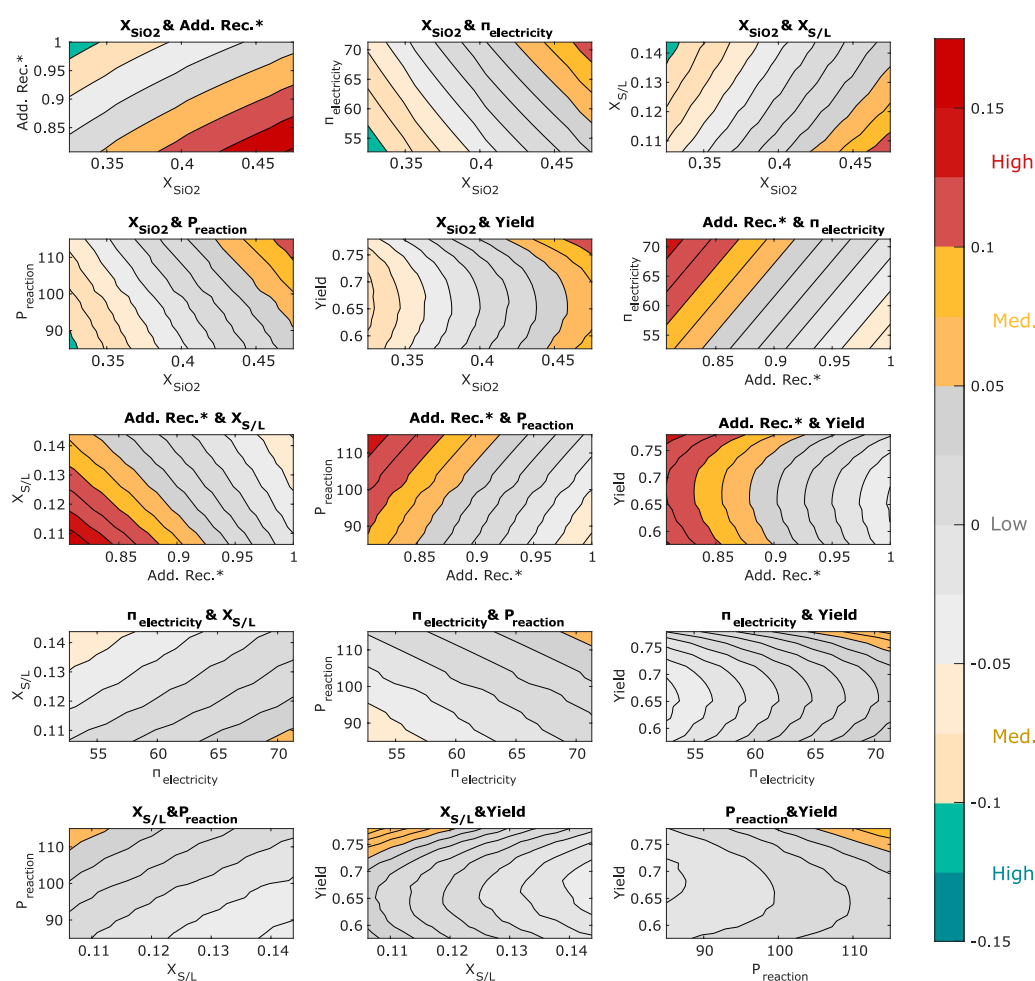
FIGURE 4
Results from two-way LSA. The six most influential input variables determined via OAT are varied in combination ±15%: silica content in the SCM$_{CCU}$ (X_{SiO_2}) (fraction), solid−liquid ratio of the slurry ($X_{S/L}$) [fraction], recycling of the reaction solution (additives + water) (**Add. Rec**) (fraction), pressure (**P$_{reaction}$**) (bar), yield (fraction), and price of electricity ($\pi_{electricity}$) (€ MWh^{-1}). The colors indicate an increase in the output variable LCOP (red) or a decrease (green). *Variables reached their limit within this interval, and the highest/lowest possible value was chosen.

mean and variance). In the approaches by Hawer et al. (2018) (i.e., Hawer's method) and the maximum entropy approach of Harr (1984), a modeler with low confidence (pessimistic) in the data quality will choose simple distributions (e.g., a triangular distribution), whereas modelers with higher confidence (optimistic) in the data quality will be inclined to assign more complex methods (e.g., selecting a normal distribution or beta distribution). To exemplify this effect, we applied the maximum entropy principle, assuming high and low confidence in the data. The derived input samples and the selected PDFs are shown in Figure 5 and Supplementary Table S2.

Figure 5 shows that for these inputs with the highest uncertainty (where a probability density is truly unknown, mostly process-related inputs in this case study, e.g., $k_{reaction}$, $Additive\ rec.$, and $X_{S/L}$), we assign uniform distributions, regardless of the uncertainty quantification method, whereas for variables with more available data (here, mostly pricing data, e.g., $\pi_{electricity}$ or $\pi_{mineral}$), we choose different distributions, such as

triangular, normal, lognormal, or beta distributions, depending on the uncertainty quantification method (Figure 5; Supplementary Table S2).

A comparison of the resulting output distributions when using the different uncertainty quantification methods reveals clear differences in the shape of the distribution and the mean and the width of the confidence interval for the quantity of interest (here, levelized cost of product) (Figure 6). First, note that the mean values, so the expected value from the MCSs (altering all variables at the same time), derived here (€145–€173 t$_{SCM}$$^{-1}$; see Figure 6), are in a similar range or exceed the maximum values for LCOP obtained using LSA methods (maximum value from OAT €144 t$_{SCM}$$^{-1}$, from two-way analysis €156 t$_{SCM}$$^{-1}$; see Section 4.1), which some might consider as extremes in the LSA. The use of uniform input distributions for all variables results in the highest mean (20% higher than using Hawer's method) and (naturally) leads to an increase of approximately 25% in the width of the 95% confidence
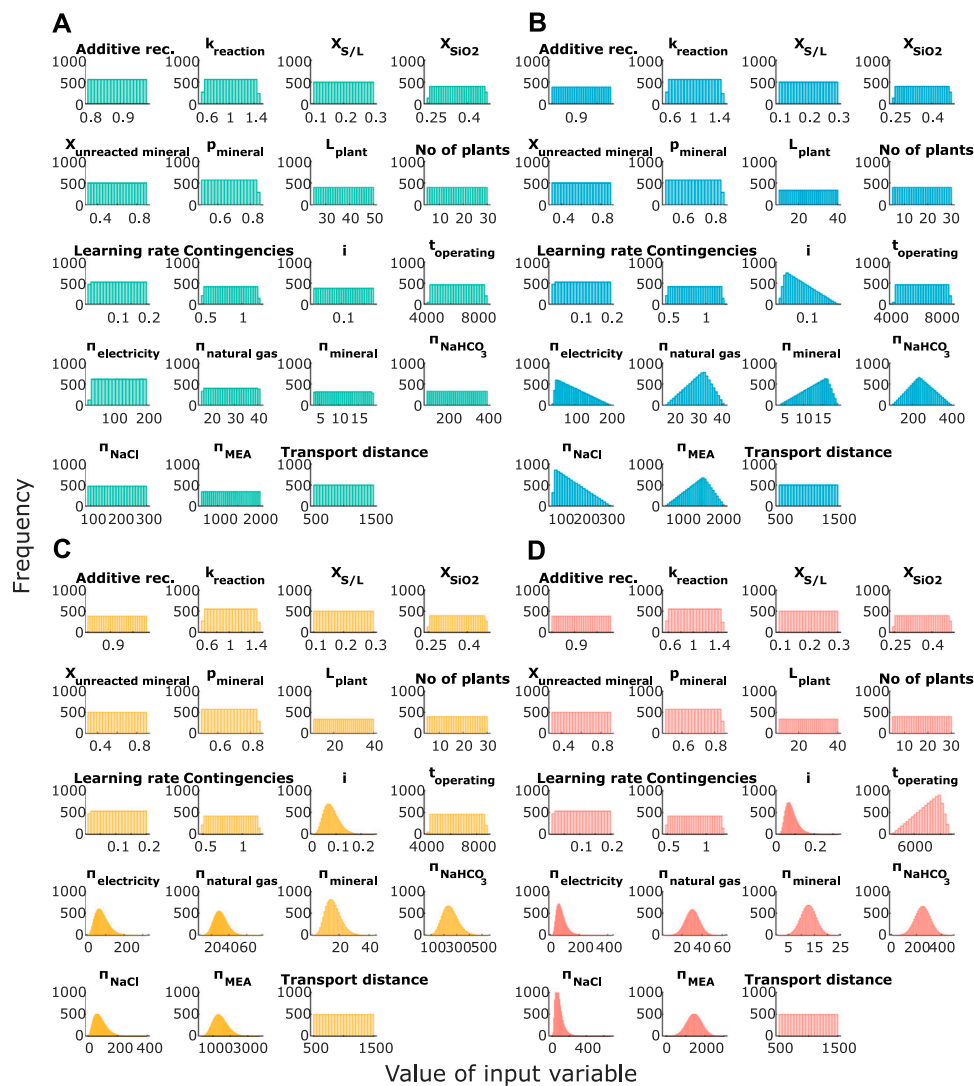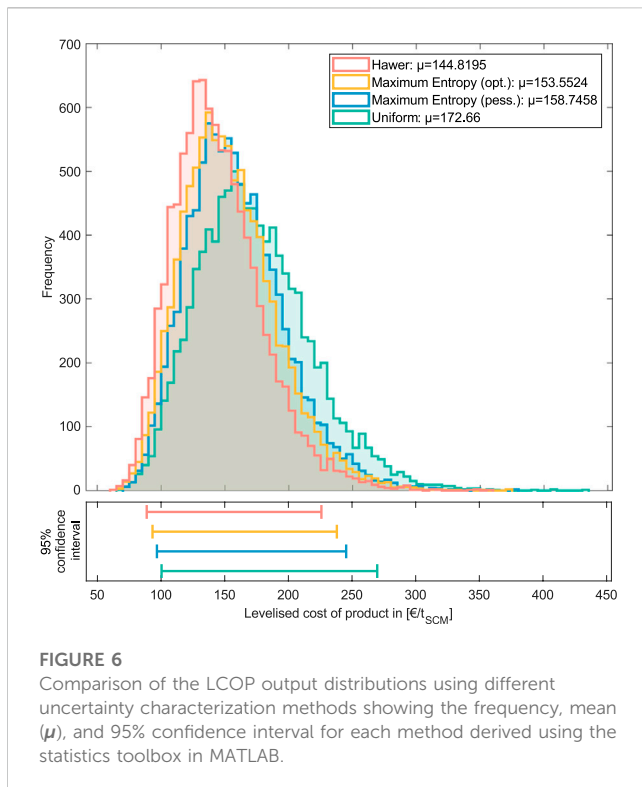
FIGURE 5
Input samples following different uncertainty characterization approaches: **(A)** uniform, **(B)** maximum entropy (pessimistic), **(C)** maximum entropy (optimistic), and **(D)** Hawer's decision tree. Altering the input variables, additive recovery (**Add. Rec**) (fraction), reaction rate constant ($k_{reaction}$) (s), solid−liquid ratio in reactor ($X_{S/L}$) [fraction], silica content in the SCM_{CCU} (X_{SiO_2}) (fraction), unreacted mineral recovery ($X_{unreacted mineral}$) (fraction), mineral purity ($P_{reaction}$) (fraction), lifetime of the plant (L_{plant}) (years), number of plants to reach maturity (**No of plants**) (natural number), learning rate on CAPEX (**Learning rate**) (fraction), combined process and project contingencies (**Contingencies**) (fraction), interest rate on capital (*i*) (fraction), operation hours per year ($t_{operating}$) (h), price of electricity ($\pi_{electricity}$) (€ MWh^{-1}), price of natural gas ($\pi_{natural gas}$) (€ MWh^{-1}), price of mineral ($\pi_{mineral}$) (€ t^{-1}), price of sodium bicarbonate (π_{NaHCO_3}) (€ t^{-1}), price of sodium chloride (π_{NaCl}) (€ t^{-1}), price of monoethanolamine (π_{MEA}) (€ t^{-1}), and transport distance of feed minerals (transport distance) (€ t^{-1}). Bounds shown in Supplementary Table S3.

interval compared to applying Hawer's method. A difference can additionally be seen between the maximum entropy principle (optimistic) and Hawer's method. The results suggest that higher values of the output are less likely to follow Hawer's method than when following the maximum entropy principle. This might be because we fit beta distributions to variables with high data availability following the maximum entropy principle assuming a confident modeler, whereas we fit lognormal and normal distributions when applying Hawer's method. Although this effect will not always be statistically significant, given the unknown nature of some of the input parameters, which method

has the highest accuracy cannot generally be concluded. However, we can conclude that different uncertainty quantification methods generate different output distributions. Therefore, particular care shall be taken when using MCS outputs for decision-making: someone cannot claim to provide a 95% confidence interval of an output if the exact nature of the input PDF is unknown, although this is very commonly done. Furthermore, clear communication of the assigned PDFs (and rationale) for MCSs must be a key element of *ex ante* system analyses to increase transparency and informed interpretation of results.

FIGURE 6
Comparison of the LCOP output distributions using different uncertainty characterization methods showing the frequency, mean (*μ*), and 95% confidence interval for each method derived using the statistics toolbox in MATLAB.

4.3 Exemplification of uncertainty importance evaluation methods

We applied the aforementioned methods for measuring uncertainty importance (Section 3.4) to the case study. We here used Hawer's method for uncertainty characterization. Note that for the comparison of the uncertainty importance evaluation methods, we again cluster the variables subjectively into three categories (high sensitivity, medium sensitivity, and low-to-no sensitivity). In contrast to the used categories for the LSA methods in Section 4.1, here, the values of the indices do not translate into a practical interpretation (e.g., an increase in LCOP by 10%).

In the scatterplot analysis, the visual determination of the most influential input variables concluded that $\pi_{electricity}$ and i show the most influence on the output, followed by $Learning\,rate$, X_{SiO_2}, and $X_{S/L}$ (Figure 7). In particular, i and $\pi_{electricity}$ stick out, as they seem to be the most influential, but they are not detected by the LSA as highly influential. Overall determination of the ranking order via scatterplot analysis can show high subjectivity to it. Hence, it has been suggested to couple scatterplot analyses with SRCC analysis (Mishra and Datta-Gupta, 2018). The SRCC analysis shows that X_{SiO_2} and $\pi_{electricity}$ show the most influence on LCOP followed by $Learning\,rate$, i, and $X_{S/L}$ (Figure 8). As the variance-based measure, we calculated the first-order and total-order Sobol indices for each variable. The results show that the most influential variables broadly match the ones concluded by the spearman rank correlation (X_{SiO_2}, $\pi_{electricity}$ followed by $Learning\,rate$, i, and $X_{S/L}$) only with a couple of switches in their ranks (Figure 9). Additionally, i, $Learning\,rate$, and $X_{S/L}$

cause a significantly higher sensitivity because of interaction effects ($S_i^T > S_i$). The interaction effects might arise because they all directly impact the capital costs. For example, i influences the annual costs of capital, $Learning\,rate$ influences the TCR, and $X_{S/L}$ influences the reactor size. Hence, the influence of either of these variables change, depending on the value of the other (e.g., for lower learning rates, the TCR will be higher and hence changes in i have a larger impact on the overall LCOP). Nevertheless, the impact of the observed interaction effects is small and does not significantly change the overall ranking of variables. The results of Borgonovo density-based indices show that the most important inputs are X_{SiO_2} and $\pi_{electricity}$, followed by $Learning\,rate$, $X_{S/L}$, and i, which are consistent with the previously shown GSA results only with a couple of order switches (Figure 10).

5 Discussion

The rankings of all input variables on the output described in Sections 4.1, 4.3 are summarized in Figure 11, where they are compared to the Sobol index (which, as the most comprehensive method, is often considered the gold standard among sensitivity indicators) (Roussanaly et al., 2021).

Figure 11 shows that the scatterplot method could only be used for a limited number of rankings because determining minor differences in the plots was challenging. All other global SA methods led to an almost unanimous ranking with only a few switches in positions (in particular for the first eight ranks), whereas there are noticeable differences with the rankings provided by the local SA methods (Figure 11). Using LSA methods, the uncertainty importance of some variables was highly overestimated (e.g., $Add.\,Rec$), although the importance of others was underestimated (e.g., $Learning\,rate$). To examine whether these differences between the LSA and GSA methods arose solely due to the difference in input intervals (i.e., ±15% for LSA and estimated minimum and maximum for GSA), we repeated the OAT analysis using the same boundaries as used for GSA methods (Figure 11). This expanded LSA approach led to identifying the same five variables with the highest impacts as determined by GSA methods. However, significant differences between the obtained rankings by LSA and GSA methods remained present, indicating that probabilistic inputs and interaction between variables are important when deriving these rankings.

To compare the derived rankings quantitively, we applied the approach suggested by Ikonen (2016) to first transform rankings into Savage scores, followed by a correlation analysis to analyze the consistency between the uncertainty analysis methods. Savage scores were developed by Savage (1956) and had the advantage that inputs with higher ranks (i.e., 1st or 2nd) receive a significantly higher score, whereas less influential parameters receive very similar scores (Supplementary Equation S1). Calculated Pearson correlation coefficients (PCC) (Supplementary Equation S2) between the Savage scores of the uncertainty analysis methods are shown in Table 4. The results confirm that, in this case study, the GSA methods (i.e., SRCC, Sobol indices, and Borgonovo indices)
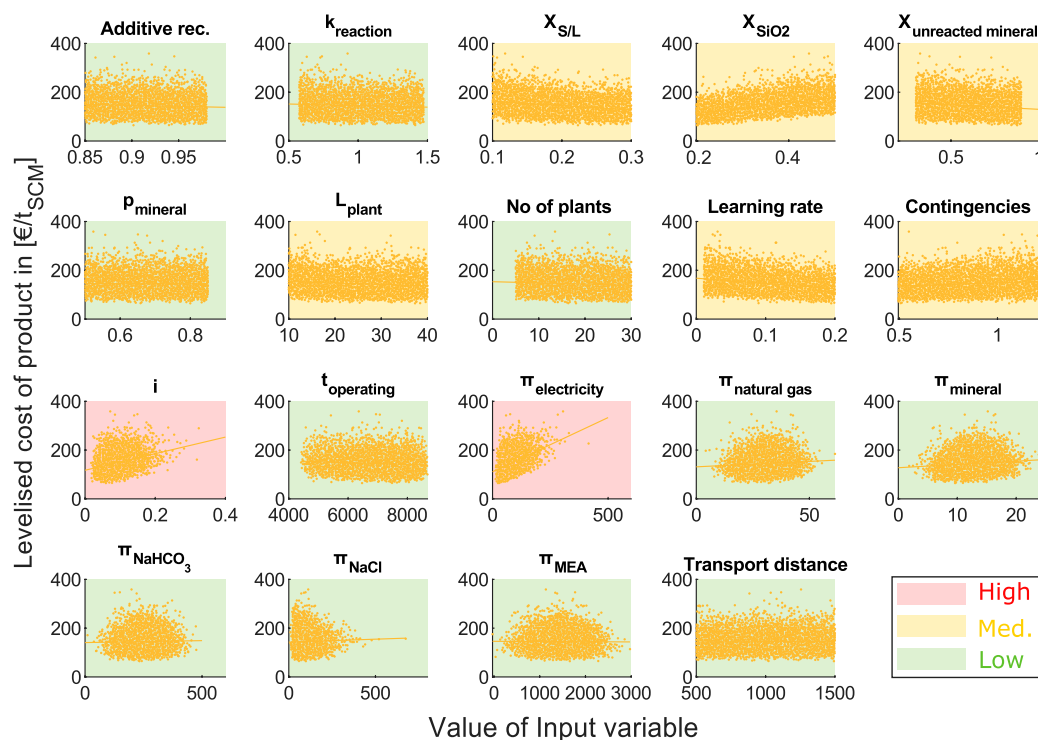
FIGURE 7
Results of scatterplot analysis. Variables with high sensitivity marked in red, medium sensitivity marked in yellow, and low sensitivity marked in green. Altering the input variables, additive recovery (**Add. Rec**) (fraction), reaction rate constant ($k_{reaction}$) (s), solid−liquid ratio in reactor ($X_{S/L}$) (fraction), silica content in the SCM$_{CCU}$ (X_{SiO_2}) (fraction), unreacted mineral recovery ($X_{unreacted\,mineral}$) (fraction), mineral purity ($P_{reaction}$) (fraction), lifetime of the plant (L_{plant}) (years), number of plants to reach maturity (**No of plants**) (natural number), learning rate on CAPEX (**Learning rate**) [fraction], combined process and project contingencies (**Contingencies**) (fraction), the interest rate on capital (**i**) (fraction), operation hours per year ($t_{operating}$) (h), price of electricity ($\pi_{electricity}$) (€ MWh^{-1}), price of natural gas ($\pi_{natural\,gas}$) (€ MWh^{-1}), price of mineral ($\pi_{mineral}$) (€ t^{-1}), price of sodium bicarbonate (π_{NaHCO_3}) (€ t^{-1}), price of sodium chloride (π_{NaCl}) (€ t^{-1}), price of monoethanolamine (π_{MEA}) (€ t^{-1}), and transport distance of feed minerals (transport distance) (€ t^{-1}).

showed high consistency in the results (a PCC close to the maximum value of 100% was reached, indicating an almost perfect correlation between the derived rankings). However, the correlation between LSA and GSA methods shows a much lower strength (PCC of 77%–87% for OAT and PCC of 54%–91% for one-way sensitivity analysis).

Although the correlation between the GSA approaches was high, small changes in ranks were expected as they are all calculated differently and have slightly different underlying assumptions. The results of the GSA methods show that position switches mainly occurred to variables with underlying interaction effects (i.e., in this case study: *i*, *Learning rate*, *Contingencies*, and $X_{S/L}$), which can only be fully investigated using Sobol indices (but at a higher computational cost) (Figure 11). Note that the nature of our case study was still fairly simple, and case studies with much larger nonlinearities and interaction effects exist, for which changes in rankings between the GSA approaches should become more distinctive. Although the rankings of all quantitative GSA methods were consistent, non-parametric measures (i.e., SRCC and Borgonovo indices) provided the impression that inputs that we categorize as being of low sensitivity might have a higher impact than those shown with

the other methods because the differences in values between high impact variables and low impact were smaller (Figures 8, 9, 10).

6 Summary and recommendations

The case study and analyses in this publication lead to a number of general recommendations on the use of uncertainty analysis in *ex ante* TEA studies. We showed that LSA methods can be insufficient in identifying all inputs of high importance and characterize inputs as important while they may not be. Therefore, we recommend the wider use of global SA methods to improve the utility of uncertainty analysis in *ex ante* TEAs and make such studies more valuable for policy- and decision-making.

This study also showed the effect of using different uncertainty characterization approaches for GSA, where the characterization method used and the confidence of the modeler can have a non-negligible influence on the computed confidence intervals, thus communicating the message. Therefore, policy- and decision-making should only rely on computed confidence intervals when there is high confidence in the inputted probability density functions (indicating they
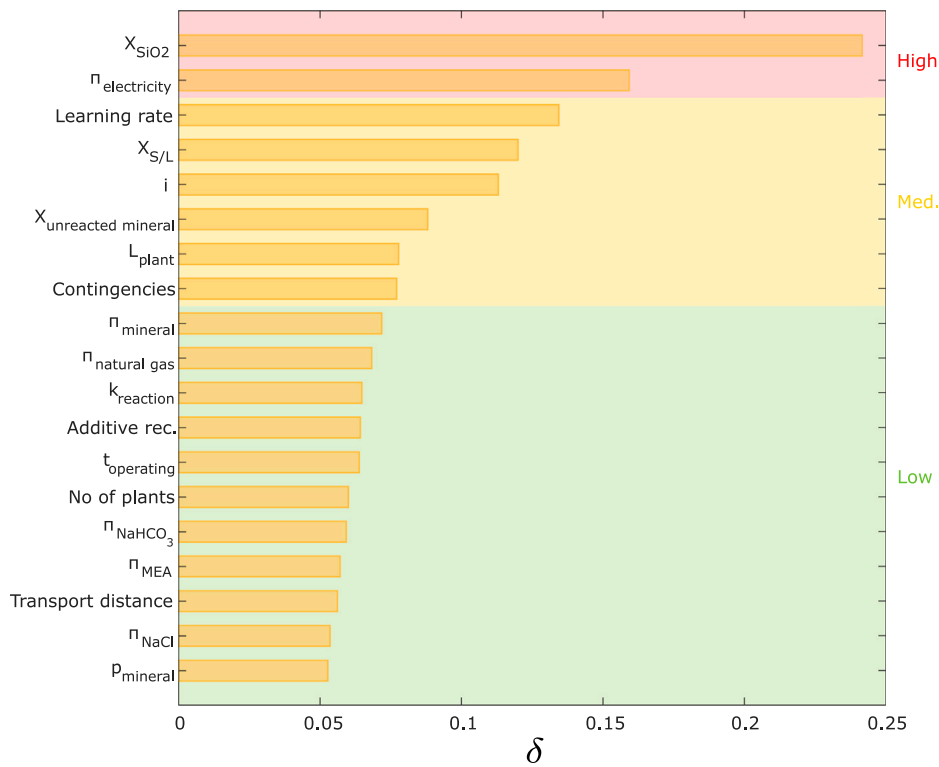
FIGURE 8
Results of SRCC analysis.

FIGURE 9
Results of Sobol analysis.
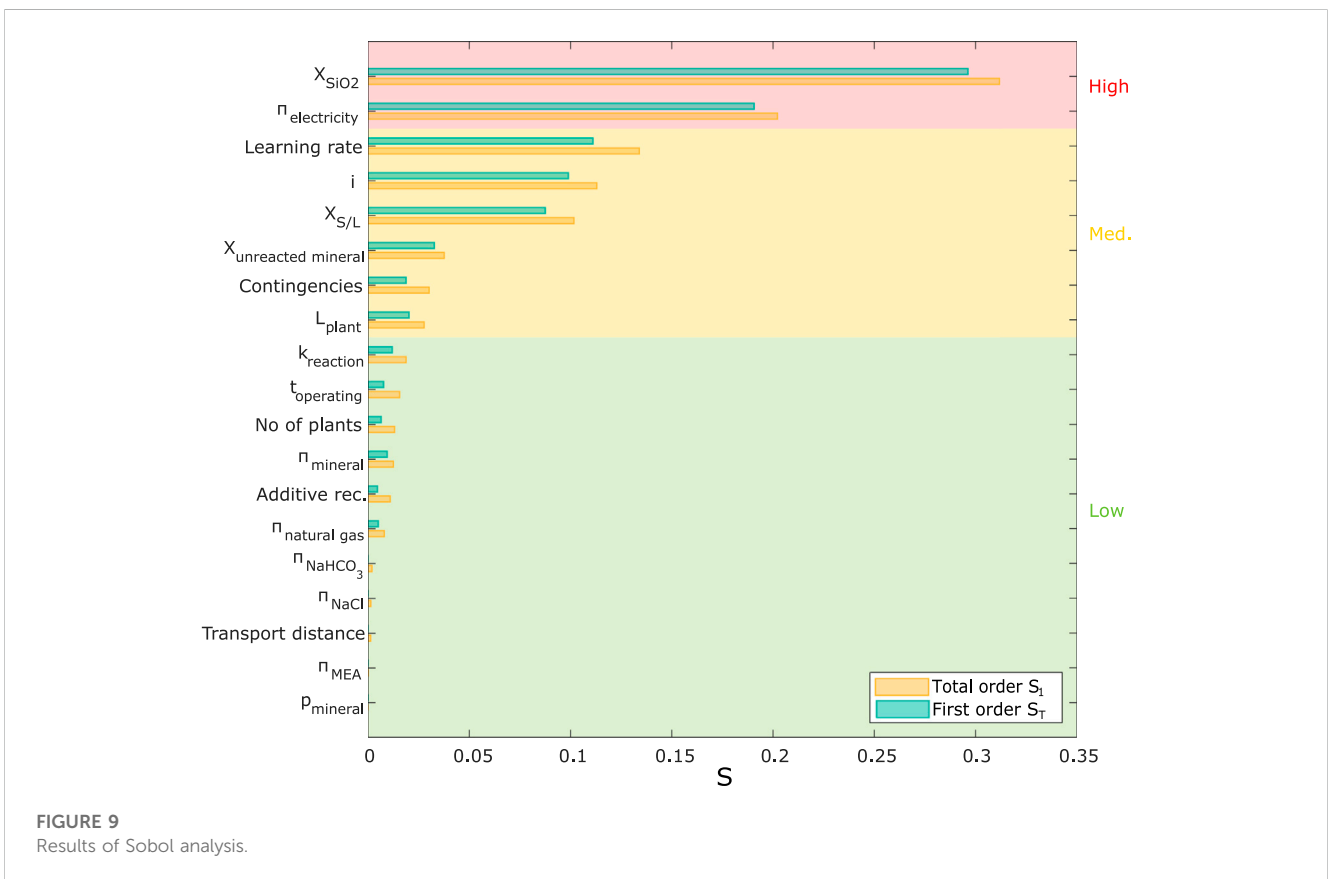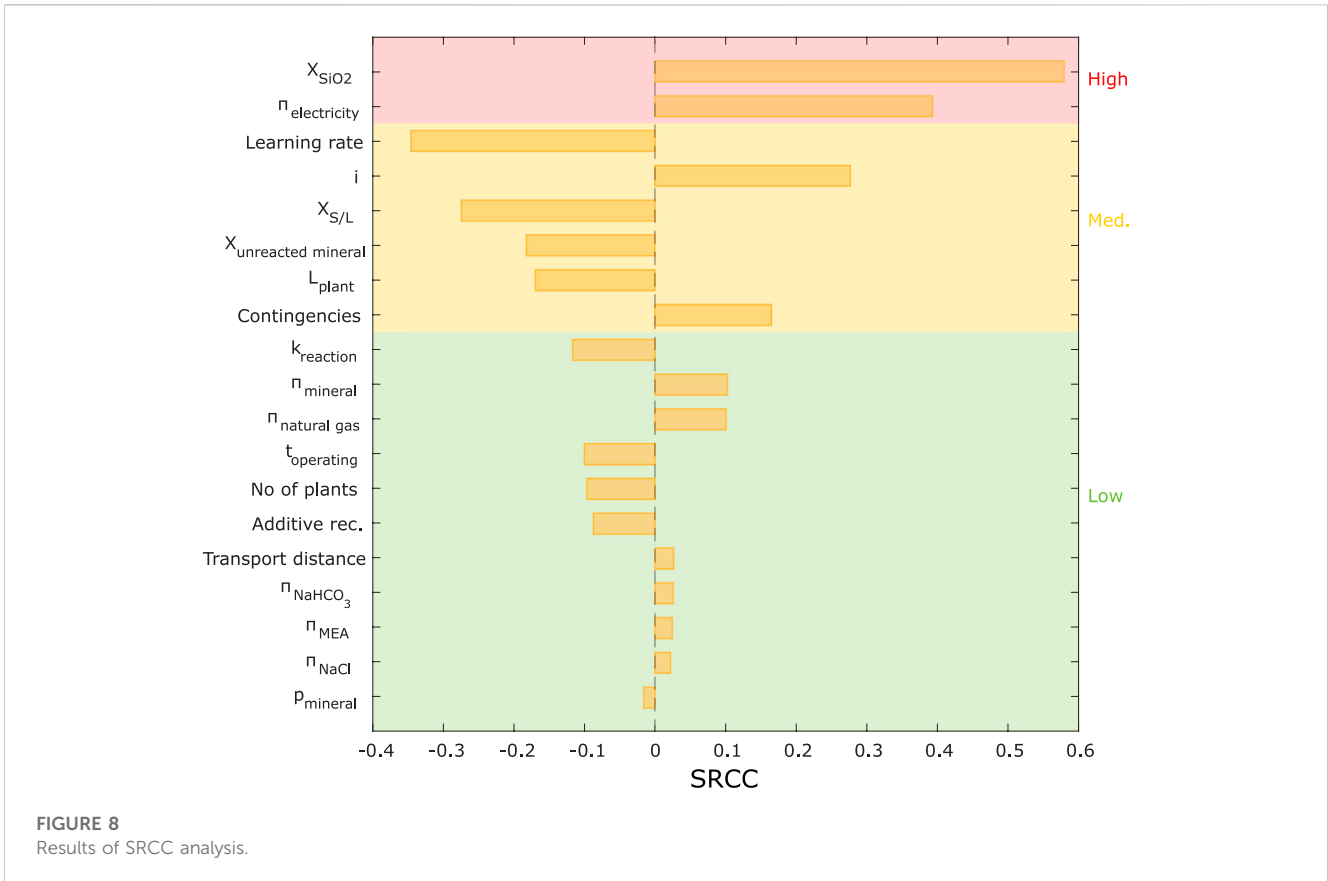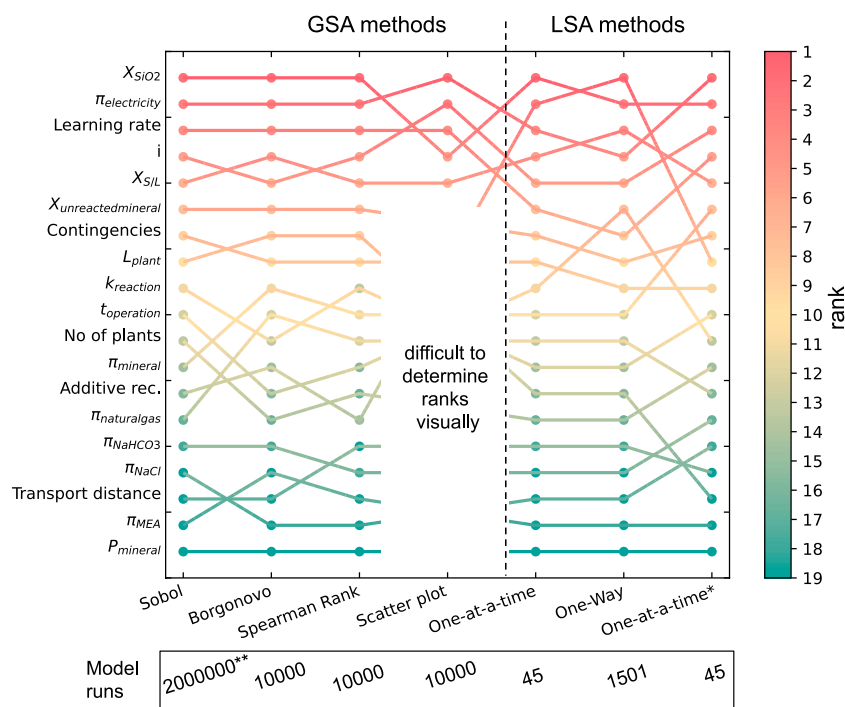
FIGURE 10
Results of Borgonovo analysis.

FIGURE 11
Comparison of ranks derived from each sensitivity analysis method. Variables only used in the LSA but not in the GSA are not shown. *For the second OAT, the same boundaries were used as for the GSA methods. **Model convergence for the Sobol indices was only found at k = 50,000 runs for the used 19 input variables.

TABLE 4 Pearson correlation coefficients between the calculated Savage scores for the rankings of each uncertainty analysis method. Scatterplot analysis was excluded as it did not yield a ranking for each variable (Figure 11). *For the second OAT, the same boundaries were used as for the GSA methods.

	GSA			LSA		
	Sobol	Borgonovo	Spearman rank	One-at-a-time	One-way	One-at-a-time*
Sobol	100%	98%	99%	81%	56%	88%
Borgonovo	—	100%	99%	82%	81%	87%
Spearman rank	—	—	100%	79%	54%	87%
One-at-a-time	—	—	—	100%	91%	77%
One-way	—	—	—	—	100%	61%
One-at-a-time*	—	—	—	—	—	100%

represent variability rather than true uncertainty) and when all known information has been used by the modeler. Given that this is seldomly true for *ex ante* studies, we argue against using GSA to answer strictly prognostic (what will) type of questions in the *ex ante* technology/system analysis domain but limit the use of GSA to identify sensitivities instead.

Regarding uncertainty importance evaluations, all quantitative GSA methods used in this case study (i.e., SRCC, Sobol, and Borgonovo) could compute consistent ranking orders (i.e., the results were largely the same). Because Sobol analysis entails much higher computational costs, it may suffice to use SRCC or Borgonovo indices instead. However, we first recommend investigating the presence and severity of interaction effects (e.g., through only using a smaller group of variables for the Sobol analysis or through multiple-ways LSA) because interaction effects are likely to cause changes in rankings between the used methods. SRCC or Borgonovo indices should then preferably be applied when small or no interaction effects are present. In particular, this may be of major importance with more complex and/or nonlinear models.

In conclusion, we recommend *ex ante* TEA modelers to i) use LSA methods only when computational power is truly limited, ii) refrain from using GSA of *ex ante* techno-economic models for answering prognostic questions, iii) investigate parameter interactions *a priori* and use Sobol indices when significant interaction effects are present or can be expected, and iv) otherwise use SRCC or Borgonovo (or other "cheap") indices to avoid high computational costs. The results from this study suggest that using these recommendations will not only harmonize the results from different TEA studies but also increase their utility for public and private policy- and decision-making.

Data availability statement

The datasets presented in this study can be found in online repositories. The names of the repository/repositories and accession number(s) can be found at: https://doi.org/10.5281/zenodo.7863667.

Author contributions

TS: conceptualization, methodology, formal analysis, and writing—original draft. PR: conceptualization and writing—review. MV: conceptualization, methodology, formal analysis, and writing—review. All authors contributed to the article and approved the submitted version.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Supplementary material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenrg.2023.1182969/full#supplementary-material

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