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Relative contribution and sensitivity analyses for decision-making in the environmental assessment of bio-based phase change materials

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ABSTRACT

This paper aims to investigate the environmental impact of the production of bio-based phase change materials to contribute to the advancement and development of energy systems with a low environmental impact. It uses relative contribution, local, and global sensitivity analysis to target critical inputs for decision-making. The materials selected are jojoba oil, coconut oil, lauric, myristic, and stearic acids, xylitol, and adipic acid, with melting temperatures varying from 14 °C to 152 °C. The methodology follows the ISO guidelines for the life cycle assessment of all materials, for which the boundary system has considered three unit processes: farming, transportation, and manufacturing. It was found that to produce 1 kg of adipic acid, the global warming potential is 13 kg $CO_{2\ eq}$, while for jojoba oil, it is only 0.96 kg $CO_{2\ eq}$. For phase change materials like jojoba oil, coconut oil, myristic acid, and stearic acid, farming dominates the global warming potential, while manufacturing takes over the major share of this environmental indicator for the remaining materials. The local sensitivity analysis results show that jojoba oil's global warming potential can increase by nearly 38 % if irrigation water reaches its upper bound. On the other hand, global sensitivity analysis shows the Sobol index by which inputs contribute to output uncertainty. For instance, the jojoba oil global warming uncertainty could be affected by 60 % by irrigation water. Results show that the higher melting temperature phase change materials have a higher footprint than the lower melting temperature ones, except for coconut oil, as a consequence of more process inputs and production process complexity. Overall, relative contribution analysis helps locate critical unit processes and inputs, while local sensitivity analysis allows for identifying how critical input variation affects the targeted indicator, and the global sensitivity analysis is useful to understand the critical inputs in terms of uncertainty.

1. Introduction

Numerous actions are taken worldwide to mitigate environmental harm as the global push for environmentally sustainable technologies intensifies. COP28's target of tripling the global installed capacity of renewable energy by 2023 is among the measures [1]. According to IEA, renewable energy technologies play a special role in keeping the global temperature rise below 1.5 °C [2]. A way to shift from fossil fuels or integrate even more renewables can be by including technologies that store renewable energy for use during off-peak periods. A potential technology in this respect refers to latent heat thermal energy storage (TES) systems. In these technologies, conventional PCM (phase change material) like paraffin has a wide range of melting temperatures, suitable for various applications [3]. On the other hand, bio-based PCMs have gained great attention, however, their environmental performance has not been discussed completely.

In a TES system, the PCM stores or releases heat during its melting or solidification process [4]. Solid-liquid PCMs are classified into inorganic and organic materials [3]. Within the organic category, bio-based PCMs' properties make them suitable for numerous applications: Bio-based PCMs are composed of organic components partially or fully derived from biomass, vegetable, and tropical oils, are generally non-toxic, with melting temperatures varying from −80 °C to 275 °C [5]. They are environmentally friendly compared to other materials and can also be a way of recycling side products of the food sector, such as expired palm oil [6]. Additionally, other advantages of bio-based PCS are the thermal stability over a wide range of temperature applications and limited flammability, while biodegradability, odor generation, leakage, and demand supply are some of their disadvantages [7]. However, they also offer other barriers. As they are derived from plants and trees, they are

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associated with deforestation and environmental and social challenges, which requires work on developing better practices on farming or extraction methods, bio-fertilizers and food waste [8]. The present paper aligns well with these barriers, providing insights on the environmental impacts during the production of such PCMs.

Bio-sourced PCMs have also received significant focus in various applications like in buildings, heating and cooling, photovoltaic panels cooling, and electronic devices cooling [9]. In the building sector, coconut oil has been applied within bamboo houses wall for temperature control, demonstrating effectively in regulating temperature in tropical highlands [10]. A vertical double-tube TES system study with coconut oil and bees wax (with and without nano particles) investigated the melting and solidification of the PCM. Among the results, it was found that 2 wt% addition of Gr-Cu nanoparticles showed a 67.59 % and 56.32 % higher melting and solidification rates compared to the bio-based PCM without enhancement [11]. Used materials can also be applied as PCMs, one example is waste cooking oil after pre-treating to remove large particles [6,12].

Life cycle assessment (LCA) has been used to evidence the environmental performance of conventional PCMs. For instance, the environmental behavior of paraffin RT35 HC has been used in the domestic hot water system, showing that the use of PCM could reduce by 24 % the kg CO_2 Eq. [13]. An environmental assessment using ReCiPe 2008 endpoint compared palm oil as bio-PCM, paraffin, and expired palm oil has been reported [6]. It revealed that raw palm oil has the highest emissions, mainly associated with soil use and water consumption during its cultivation. With IPCC 100y, the same study showed that raw palm oil, paraffin, and expired palm oil have a global warming potential (GWP) of 2.771 kg CO_2 eq, 0.207 kg CO_2 eq, and 0.053 kg CO_2 eq. This is a consequence of expired palm oil being treated as a residue with zero environmental load during its production. In some cases, the environmental assessment of PCM is conducted using proxy or generic LCA data due to the unavailability of LCI information. For instance, Motte at all [14] simulated myristic acid in a building-integrated solar collector, and the environmental load regarding the PCMs was generic fatty acid data from Ecoinvent. For the production of bio-based PCMs, one study shows that 70 % of CO_2 emissions in the production of lauric acid originate from the fresh fruit bunches. Nevertheless, some LCA studies have been carried out regarding materials used as PCMs. Although these works have not been aimed at thermal energy storage purposes, they are worth mentioning. Yani et al. (2021) investigated the life cycle of the coconut oil industry, considering the steps of the manufacturing process only, i.e., a gate-to-gate study. The main raw material is copra, and the products consist of coconut oil, coconut pulp pellets, and waste (solid, liquid, and gas). GHG emissions contribute to a global warming potential of 2.9 kg CO_2 eq per kilogram of coconut oil.

Decision-making in LCA results can be conducted by indicating the contribution of inputs to the final indicators and, additionally, by using Local Sensitivity Analysis (LSA) or Global Sensitivity Analysis (GSA). Briefly, in the LSA, inventory inputs are varied individually to investigate the influence on the outputs [15]. The main limitation is that it does not consider the correlation between parameters, and they must be analyzed one at a time [16]. On the other hand, GSA is defined as the study of how output uncertainty can be apportioned to different uncertainties in the model input [17]. One of the methods for GSA is calculating the Sobol Indices, which are based on the variance [18]. An appropriate tool for implementing GSA is the sensitivity analysis library written in Python, known as SALib, which offers various sampling and sensitivity analysis methods [19]. However, what has not been discussed is the assessment potential provided by integrating contribution, LSA, and GSA. This paper will show that despite the current common preference for one technique over another, both LSA and GSA can be used in decision-making due to their distinct mathematical nature, along with contribution analysis.

The objectives of this study are twofold. First, to present an environmental assessment using LCA on a cradle-to-gate level for seven bio-based PCMs with melting temperatures ranging from 14 °C to 152 °C. Second, to combine contribution and sensitivity analyses for robust decision-making in the life cycle assessment of bio-based phase change materials. The aim is that the results obtained will support studies that focus on the use phase of such materials, where the environmental impact, informed per kilogram of material, can be scaled down or up according to the mass of the system. The contribution and sensitivity results aim to support decisions that can reduce environmental impacts in the manufacturing and reflect such reductions at the time of designing bio-based PCM TES systems. The methodology has particular advantages since it includes the propagation of input uncertainty to outputs, i.e., to the LCA indicators, and is well-documented to ensure replicability. The results are useful for the cradle-to-grave environmental assessment of thermal energy storage systems. The limitations are treated carefully as they open doors for further improvements.

2. Methodology

This study followed ISO 14040 and 14,044 [20,21] guidelines for the main requirements of LCA works, incorporating the four phases: goal and scope definition, life cycle inventory, life cycle impact assessment method, and interpretation. The entire work was carried out with the methodological structure shown in Fig. 1, consisting mainly of the 6 steps that are explained next.

The methodology steps are conceptualized to provide a robust environmental assessment to identify the most critical unit process and input. Table 1 shows the inputs necessary for the whole production of each PCMs for the three unit processes considered.

Step 1. The materials selected for the assessment were based on two main factors. The first is the bio-based nature. Several works have reported conventional PCMs such as paraffins, but a discussion on the environmental impacts of bio-based materials for TES is still lacking in literature. Second, the working temperature range of these materials was thought to support studies on cooling and heating applications for commercial, residential, and industrial sectors. Therefore, the materials included in this work are jojoba oil (JO), coconut oil (CO), lauric acid (LA), myristic acid (MA), stearic acid (SA), xylitol (XY) and adipic acid (AA) for melting temperatures of 14.7 °C, 25 °C, 44 °C, 54 °C, 69 °C, 92.55 °C and 153.5 °C [22,23], respectively. Their manufacturing LCA were carried out considering some assumptions: jojoba oil and coconut oil production chain were based on electricity use, and no residues are used to produce heat; the fatty acids manufacturing process are based on the coconut oil fractionation; xylitol is manufactured from sugarcane bagasse and adipic acid is produced based on corn stove residues. Also, three unit processes were established for investigating the use of contribution and sensitivity analysis on their production: farming, transportation and manufacturing.

Step 2. In this step, the goal and scope were conceptualized such that within the system boundaries, all PCMs production had the same unit processes. The three unit processes are farming, transportation, and manufacturing. This way, an evaluation of the most contributing unit process is allowed. Within the unit processes, the specific material flow, energy flow, and reactions are specific to each PCM. As a material used for energy storage, the PCM's main feature is the amount of energy that can be stored for later use. So, one possible FU for this study could be a specific amount of stored thermal energy. However, the materials considered have different properties with subsequent different melting temperatures, and the amount of PCM is dependent on operational characteristics of the TES system and storage medium properties. This would also imply different amounts of each PCM. Thus, 1 kg of the material at the factory's gate is considered as FU. By doing so, the final environmental impacts will be presented per kg of each material, such as kg CO_{2eq} per kg of PCM, and it facilitates scaling up or down these values according to the mass of PCM obtained for a specific tank design.

Step 3. Life cycle inventory is a critical step in any life cycle study. In this step, the production routes of the PCMs described were studied, and

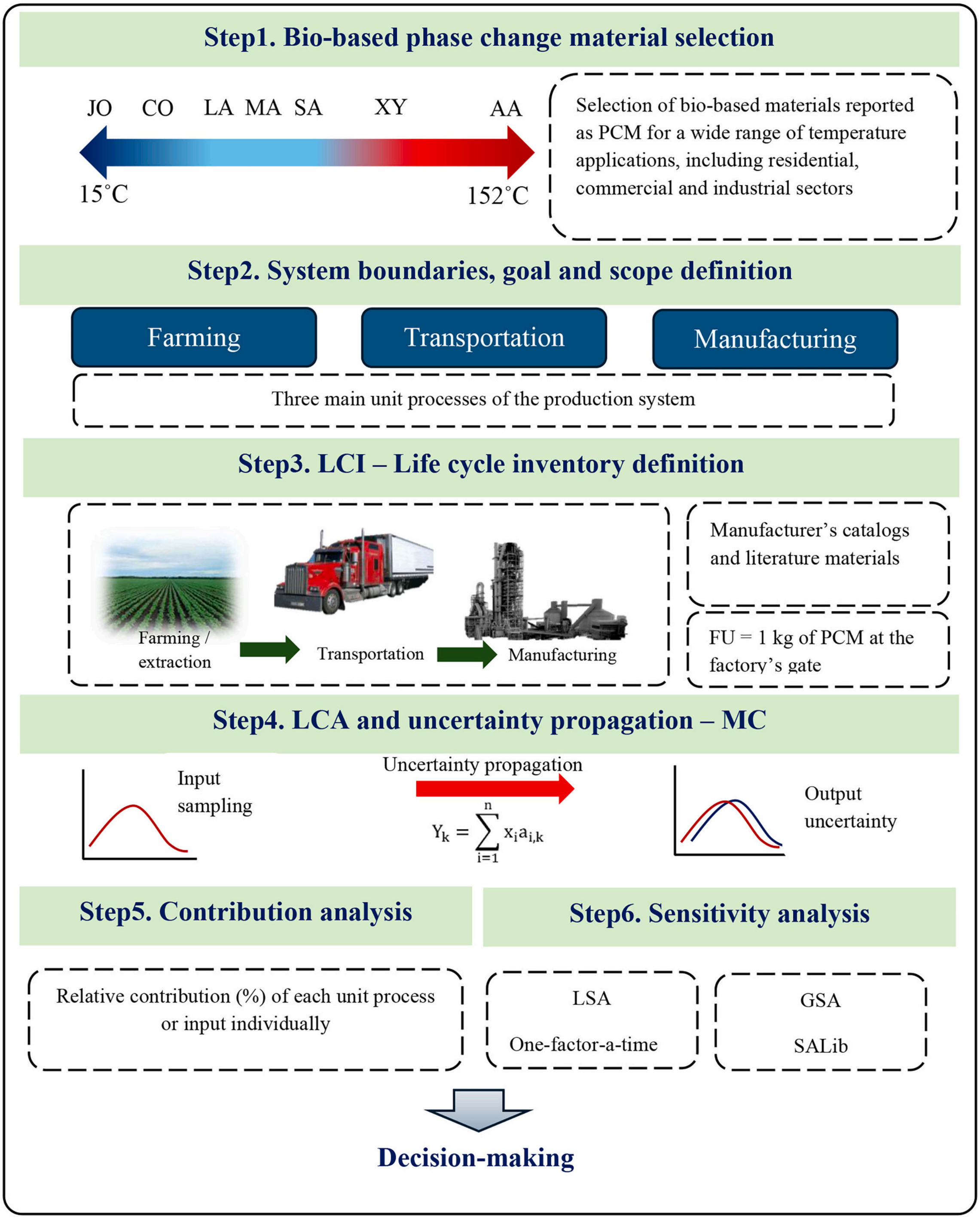

Fig. 1. Flowchart of the methodology structure adopted in this work. JO: Jojoba oil, CO: coconut oil, LA: lauric acid, MA: myristic acid, SA: stearic acid, XY: xylitol, AA: adipic acid.

the available information was compiled using manufacturer catalogs and published papers. A brief description of the inventory phase in the production of each PCM is given as follows. Regarding the transportation unit process, a standard distance of 100 km was considered for all materials.

Jojoba oil (JO): Major information regarding the production of jojoba oil was obtained from published literature for the farming and cultivation, harvesting, and oil-pressing stages [24]. The material flow used in the inventory is shown in Fig. 2 (a).

Coconut oil (CO): Information regarding CO production was gathered from various sources, including farming irrigation guidelines [25,26]. Plantation management information and plant fertilizing materials were also accounted for [27]. Energy and material consumption data were obtained from manufacturers' catalogs and direct assistance from the manufacturer, and the material flows were calculated directly based on the production machine's catalogs. This procedure was adopted and considered reliable based on published literature using machine specifications (Sandouqa and Al-Hamamre, 2019). The material flow of CO production is presented in Fig. 2 (b).

Fatty acids (LA, MA, SA). For the production data of LA, MA, and SA, various materials were inventoried. The main process is based on fractionation dividing wall column [28]. However, additional data is

Table 1
Definition of inputs used in the inventories of each PCM and used in Eq. 1 for propagating uncertainty, local sensitivity analysis, and global sensitivity analysis. Note that the amount of each material or energy flow is presented in the production flow charts of Figs. 2, 3, 4, and 5.

Input	Jojoba oil (JO)	Coconut oil (CO)	Lauric acid (LA)	Myristic acid (MA)	Stearic Acid (SA)	Xylitol (XY)	Adipic acid (AA)
x_1	Irrigation water	Nitrogen fertilizer	Nitrogen fertilizer	Nitrogen fertilizer	Nitrogen fertilizer	Irrigation water	Irrigation water
x_2	Nitrogen fertilizer	Potassium fertilizer	Potassium fertilizer	Potassium fertilizer	Potassium fertilizer	Nitrogen fertilizer	Phosphorus fertilizer
x_3	Phosphorous fertilizer	Irrigation water	Irrigation water	Irrigation water	Irrigation water	Phosphorus fertilizer	Nitrogen fertilizer
x_4	Potash	Transp. distance	Transp. distance	Transport. distance	Transp. distance	Potassium fertilizer	Maize grain
x_5	Diesel	Process water	Process water	Process water	Process water	Poultry manure	Pesticide
x_6	Pesticide	Electricity	Electricity	Electricity	Electricity	Electricity -farming	Diesel
x_7	Transp. distance		Steam	Steam	Steam	Diesel	Lubricating oil
x_8	Electricity		Cooling water	Cooling water	Cooling water	Transp. Distance	Electricity
x_9			Heating	Heating	Heating	Process water	Transp. Distance
x_{10}			Cooling	Cooling	Cooling	Sulfuric acid	Natural gas
x_{11}						Quicklime	Sulfuric acid
x_{12}						Hydrogen	Ammonia
x_{13}						Nickel	Potassium hydroxide
x_{14}						Electricity	Enzimes
x_{15}						Steam	Acetronitrile
x_{16}							Compressed air
x_{17}							Acetic acid
x_{18}							Water
x_{19}							Hydrogen
x_{20}							Cooling
x_{21}							Heating
x_{22}							Electricity

Farming
Transportation
Manufacturing

required for other processes, such as splitting, and the heating or cooling demands [29–31]. Material properties were obtained from the NIST Webbook [22]. Fig. 3 depicts the material flow for LA, MA, and SA.

Xylitol (XY). As shown in the literature, two production routes have been discussed for the production of XY, chemical and biotechnological production routes, both of which use biomass as raw materials. An environmental assessment comparing chemical and biotechnological routes for xylitol production has been reported [32]. Although it showed that the biotechnological route is associated with a lower GWP, this study opted for the chemical pathway because it is the conventional technology already in use. The results will differ slightly from theirs because they used a dataset representing the Indian scenario, while in this study, emphasis is given to datasets representing the global market. Thus, the study conducted by [33] was inventoried for the production of xylitol. The material flow is shown in Fig. 4.

Adipic acid (AA). Data was collected from different sources. Apart from farming and transportation, the manufacturing phase contained information on glucaric acid and adipic acid production. The feedstock considered was corn stover obtained during corn cultivation [36,37]. Then, additional work was inventoried for the glucaric acid production stage, material, and energy flow [38]. The final production stage inventory comprises the conversion of glucaric acid to adipic acid [39]. The materials flow is shown in Fig. 5.

Step 4. Uncertainty propagation was carried out to transfer uncertainties from inputs to outputs. Outputs in this study are the LCIA indicators, such as GWP, obtained after the use of an LCIA method, in this case, ReCipe 2016 midpoint (H) [40], collected using Ecoinvent as a database [41] and SimaPro as a tool to provide the Ecoinvent factors [42]. This LCIA methods allows the investigation of the following indicators: global warming potential (GWP), ozone depletion potential (ODP), ionizing radiation potential (IRP), particulate matter formation potential (PMFP), photochemical oxidant formation potential: ecosystems (EOFP), photochemical oxidant formation potential: humans (HOFP), terrestrial acidification potential (TAP), freshwater eutrophication potential (FEP), marine eutrophication potential (MEP), human toxicity potential: carcinogenic (HTPc), human toxicity potential: non carcinogenic (HTPnc), terrestrial ecotoxicity potential (TETP), freshwater ecotoxicity potential (FETP), marine ecotoxicity potential (METP), agricultural land occupation potential (LOP), water consumption potential (WCP), surplus ore potential (SOP), fossil fuel potential (FFP). In addition, the Ecoinvent datasets used to represent the materials and energy flows are found in Table S1 in the supplementary information. Each input was sampled with the Monte Carlo method with a triangular distribution, including 10,000 samplings, according to

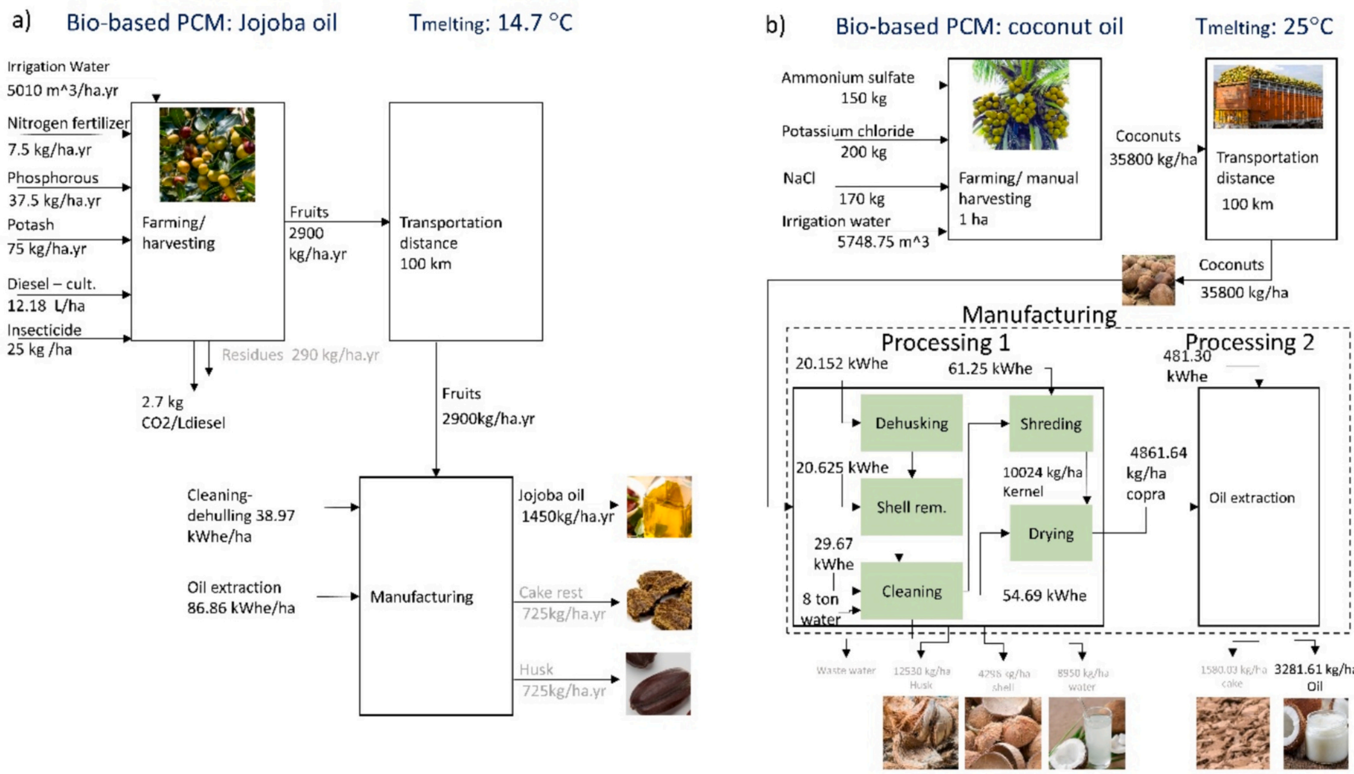

Fig. 2. Material flow assumptions to produce: (a) jojoba oil – JO and (b) coconut oil – CO. Note that data to produce JO come from (Sandouqa and Al-Hamamre 2019) while CO uses data from manufacturers catalogs.

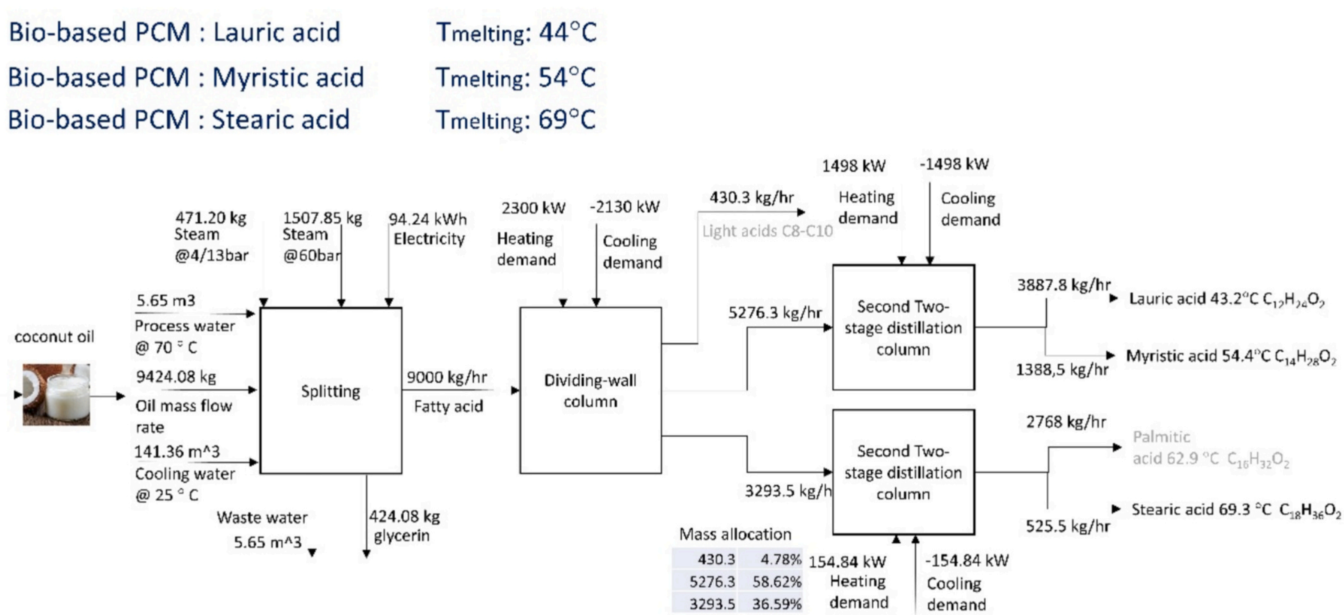

Fig. 3. Fatty acids production flow diagram with LCI information. Note that the 3 fatty acids under study are obtained in the same manufacturing processes by means of separation. Data was collected from different sources [28,29,31].

published literature [43]. Moreover, the triangular function fits well in this case because it uses the maximum and minimum values obtained from Monte Carlo sampling. Each data point obtained is applied in Eq. 1 to propagate the uncertainty to the output, allowing the indicators to be evaluated with a maximum and minimum or standard deviation value. In this step, one important assumption regards the range of variation of each input, it is set to a variation in the range of ±10 based on the approach used in the literature [44]. Eq. 1, x_i represents an input flow, Y_k represents the midpoint indicator, $a_{i,k}$ is the Ecoinvent coefficient for the corresponding input and indicator, and n represents the number of inputs. The coefficients ($a_{i,k}$) for all inputs are presented in Tables S2, S3, S4, S5, and S6 in the Supplementary Information of the Excel file.

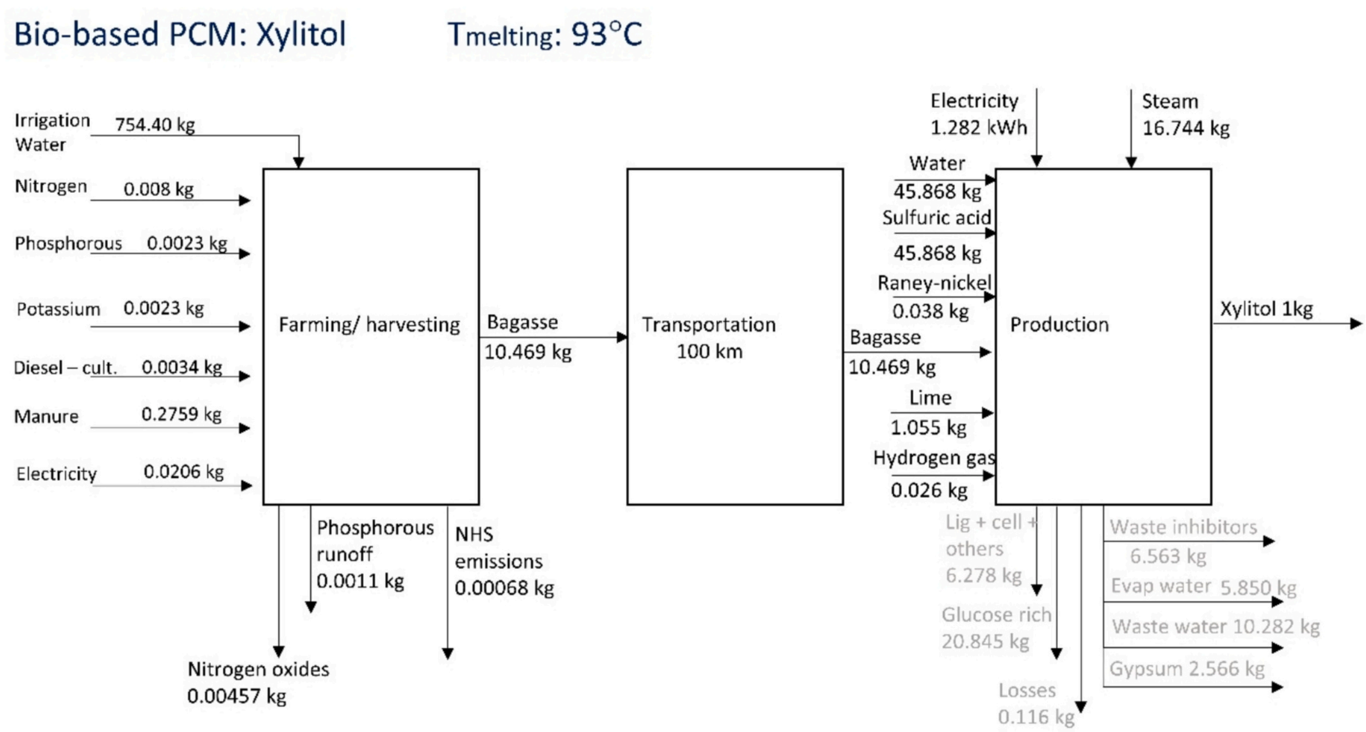

Fig. 4. Xylitol production flow diagram with LCI information. Data was collected from different sources [34,35].

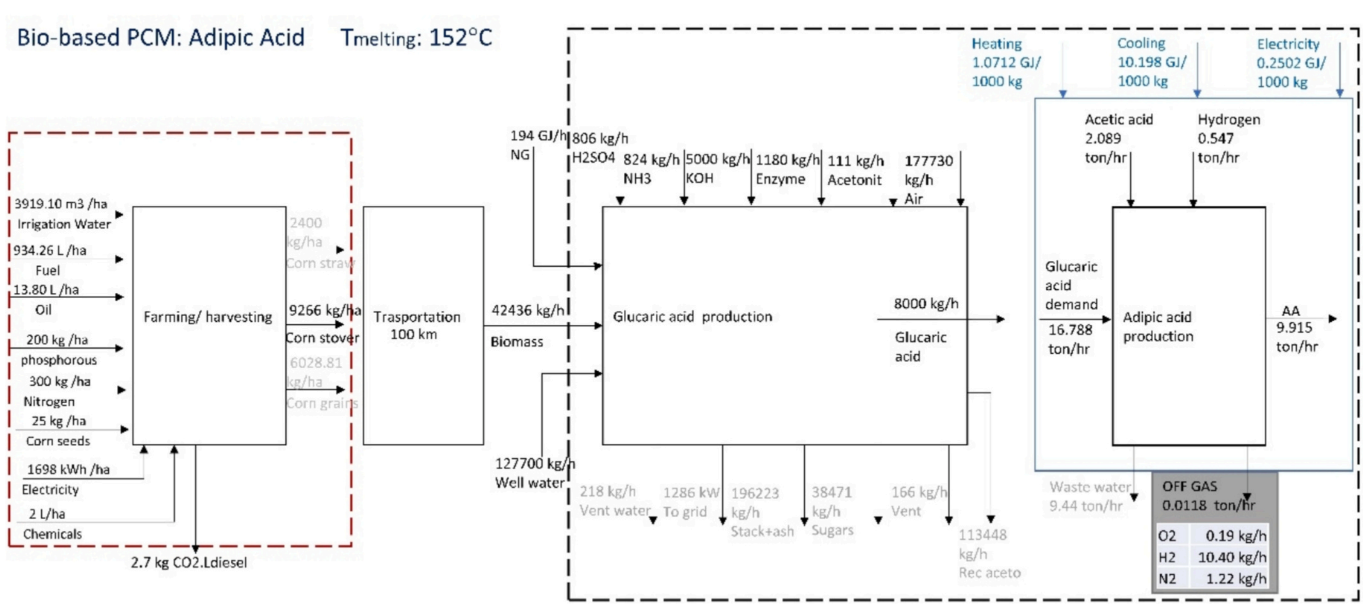

Fig. 5. Adipic acid production flow diagram with LCI information. Data was collected from different sources [36–39].

$$Y_k = \sum_{i=1}^{n} x_i a_{i,k} \tag{1}$$

For this study, to exemplify the use of Eq. (1), the mid-point indicator Particle Matter Formation Potential of the production of JO is demonstrated. It can be obtained by expanding Eq. 1 to Eq. 2, where $n = 8$. Table 1 can be used to link each x variable with its corresponding input name, then the numerical value of each one can be obtained from the Figures representing the production inventory (Figs. 2 to 5).

$$Y_{PMFP} = x_1 a_{1,PMFP} + x_2 a_{2,PMFP} + x_3 a_{3,PMFP} + x_4 a_{4,PMFP} + x_5 a_{5,PMFP} + x_6 a_{6,PMFP} + x_7 a_{7,PMFP} + x_8 a_{8,PMFP} \tag{2}$$

Step 5. The objective of the contribution analysis is to measure the relative contribution of each unit process or the relative contribution of each input individually. In this study, the relative contribution means the percentage to which a unit process or input is responsible for a specific indicator. Initially, the results are analyzed and discussed based on the three unit processes (farming, transportation, and manufacturing), identifying the one with the highest contribution to the indicator. Then, the discussion is extended to the individual inputs, helping to find the individual most critical ones.

Step 6. Local sensitivity analysis (LSA) and global sensitivity analysis (GSA) are carried out to investigate how the inputs can implicate in the outputs of the LCA study. In the LSA, inventory inputs are varied independently to investigate the influence on the LCA outputs, this means that while one input parameter receives a small perturbation the others remain constant, and the influence on the environmental impacts is visualized, such approach is known as one step at a time (Wei et al., 2015). The LSA is conducted using Eq. 3, where ΔXi represents the variation of input Xi and ΔYi the corresponding changes to output Yi .

$$\underline{S}\left(X_i, p_j\right) = \frac{\Delta X_i / X_i}{\Delta Y_i / Y_i} \tag{3}$$

On the other hand, the GSA is the study of how output uncertainty can be apportioned to different uncertainties in the model input [17]. In this work, GSA is calculated using the variance-based method, in which Sobol indices are used to represent the sensitivity of outputs to inputs, in other words it measures the impact of the variance of the input on the variance of the output. The first-order Sobol index, S_i, is represented by Eq. 4, where V denotes variance, E is the expectation, and X_i is the input variable being treated. Moreover, the Sobol indices were obtained using the sensitivity analysis library in Python (SAlib) [45] was chosen to perform the GSA of the input-to-output parameters.

$$S_i = \frac{V[E(Y|X_i)]}{V(Y)} \tag{4}$$

3. Results and discussion

Three main unit processes were maintained under the boundary system for the cradle-to-gate analysis of the PCMs: farming, transportation, and manufacturing. Then, LCA was carried out using global datasets from Ecoinvent and SimaPro as software. Subsequently, the LCIA method ReCiPe 2016 midpoint (H) was applied, obtaining eighteen impact category indicators to assess the PCM's environmental performance. To simplify, four indicators were selected for further discussion based on the consequences they have on endpoint areas. For example, an increase in respiratory disease can be caused by particulate matter, so particulate matter formation potential (PMFP) is included. Global warming contributes to increasing malnutrition, damage to freshwater and terrestrial species; thus, global warming potential (GWP) is considered. Additionally, as bio-based PCMs are directly related to soil management practices and agriculture, water use and terrestrial ecotoxicity are included for detailed discussion, therefore, water consumption potential (WCP) and land occupation potential (LOP) are also reported.

3.1. Uncertainty propagation

The total impacts obtained per kg of each material after uncertainty propagation are shown in Fig. 6, presented as the mean and standard deviation bars. Overall, the higher the average value of the indicator, the more dispersed the results, as shown by the standard deviation bars, which have been obtained with Monte Carlo sampling. The GWP is detailed in Fig. 2 (a). JO and CO showed the lowest kg of $CO_{2\ eq}$ among these PCMs, while AA holds the highest amount of kg $CO_{2\ eq}$ per kg of material, nearly fourteen times that of JO. Between CO and AA, the GWP of materials like LA, MA, SA, and XY varies between 1.68 and 9.20 kg $CO_{2\ eq}$. Figs. 2 (b), (c), and (d) contain the results of PMFP, LOP, and WCP, allowing the identification of the highest and lowest values among

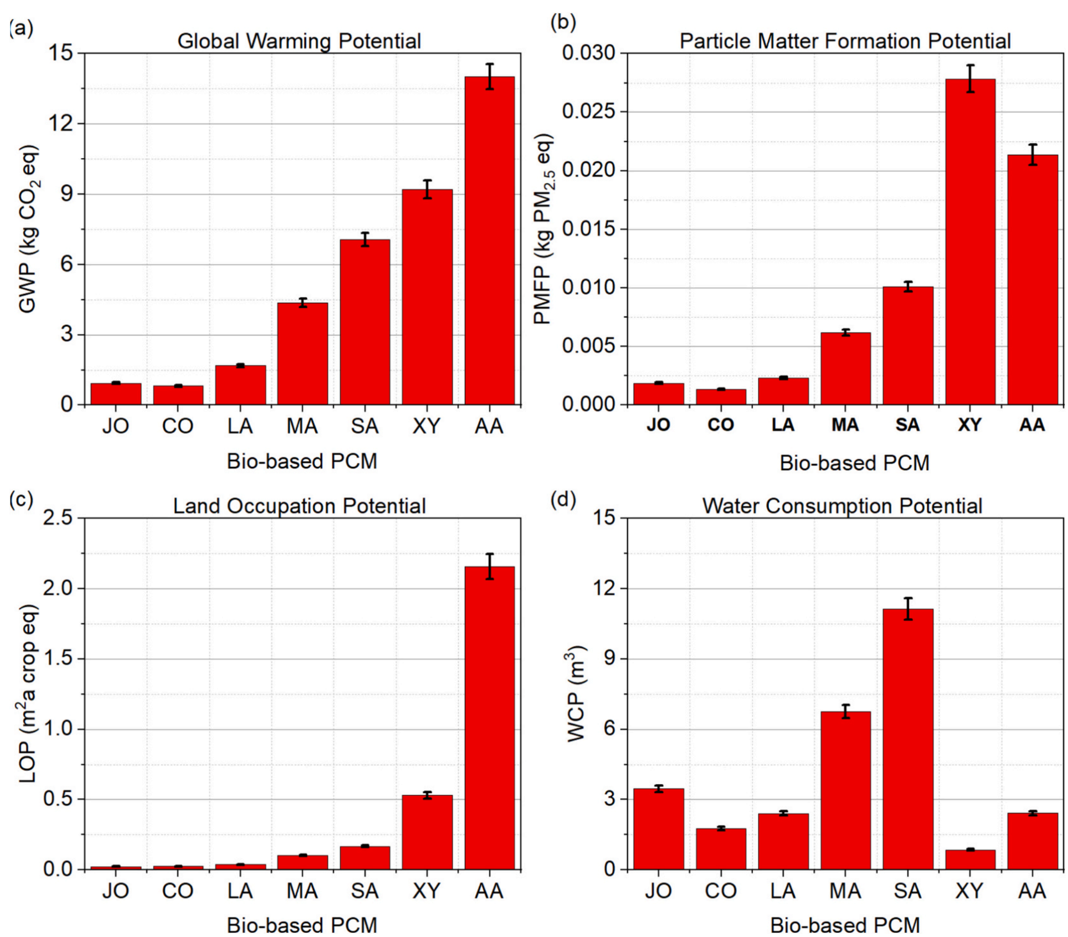

Fig. 6. Total environmental impact for the FU = 1 kg in the production of each PCM: (a) GWP; (b) PMFP; (c) LOP; (d) WCP.

the PCMs investigated. WCP, on the contrary to the other indicators, presented a higher variability among the materials studied, not following a tendency such as shown for GWP and LOP. The greater majority of this water consumption potential is related to the farming activity in the production of all PCMs. The standard deviation bars indicated in the figure are intended to highlight the fact that the environmental impact is within a sample, which was conducted using Monte Carlo sampling. This allows for the expectation that the environmental impact value falls within maximum and minimum limits. However, these variations can be expected due to various sources of error, such as time, geography, technology, and uncertainty in the data used in accordance with ISO 14040/14044 recommendations.

The overall results presented in Table 2 may vary according to the production method adopted for each material. For instance, in the case of coconut oil, the production system considered in this study assumes the use of electricity rather than fossil fuels, thereby promoting reliance on cleaner energy sources. Nevertheless, the environmental benefits remain highly dependent on the energy mix of the production site's geographical region. To address this variability, global Ecoinvent datasets were employed. Similarly, the environmental profile of adipic acid is strongly influenced by its production route. Conventional processes are based on petrochemical feedstocks, typically initiating with benzene, a compound of high toxicity (Skoog et al., 2018). The adoption of alternative pathways, particularly those derived from biomass, represents a promising strategy to mitigate the associated environmental impacts.

A comparison between the obtained results and those reported in the existing literature is presented, however, it is important to bear in mind that for some reported studies the authors have considered distinct production process, LCA boundaries, assumptions, and LCIA methods. Table 2 summarizes the total environmental impacts for all materials investigated, i.e., farming, transportation, and manufacturing. For JO, the GWP is 14 % higher than the value reported by Sandouqa [24], with a GWP of 0.802 kg $CO_{2\ eq}$ per kg of oil using the IPCC method. This difference may be attributed to the datasets in this study since the same materials and energy flows were used. Additionally, the authors used LCIA method IPCC, whose characterization factors for CO_2, CH_4, and N_2 are 1, 23, and 296, respectively, while in this work the LCIA method used, ReCiPe 2016 midpoint (H), whose value for the factors are 1, 34/36 (methane/fossil methane), and 298 for the same factors, which could increase the final results. For CO, a GWP of 2.9 kg CO_2 eq per kg of the material has been reported [46], approximately 3.5 times higher than the amount found in this study. In the reported study, the manufacturing fuel was charcoal, which could be one of the reasons for this difference. Also, LCA assumptions and the LCIA method affect the results. As shown, the CO inventory uses electricity as the main using electricity as the main source of energy, and depending on the country's electricity mix, the emissions could be lower or higher. In this case, global datasets were chosen to have a better representation of the world's emissions based on the electricity mix. Regarding the environmental impacts of LA, MA, and SA, reported research is normally attributed to generic fatty acids [47] from Econvent [41], which is currently 3.91 kg CO_2 eq per kg fatty acid. This value falls within the range of variations obtained in this study, from 1.68 kg CO_2 eq for LA to 7.06 for SA, however, this work considers the energy and materials flow for the fractionation of each fatty acid, being more representative for each acid. Concerning AA, Corona et al. [48] reported for different scenarios a GWP of 4.87 kg CO_2 eq per kg_{AA}. They used consequential LCA assumptions with system expansion, the LCIA method TRACI 2.1 midpoint, and considered AA obtained from high lignin fermentation by-product (HLFB), assigning to it zero load as it was treated as a by-product. However, as this manuscript does not deal with consequential LCA, the results should not be comparable with the results obtained by Corona et al. Paraffin wax has a GWP of 0.207 kg $CO_{2Eq.}$ [6], lower than bio-based PCMs. However, the LCA method used, IPCC 100y, and load assignment conditions must be taken into account, which also affect the results of the LCA . Therefore, comparisons with bio-based PCMs should be cautious. On the other hand, sodium acetate has a GWP of approximately 18.3 kg $CO_{2Eq.}$ [49], demonstrating that bio-PCMs are highly competitive in terms of global warming.

3.2. Contribution analysis

The unit process relative contribution is shown in Fig. 7. The percentage of contribution from farming, transportation, and manufacturing varies from 0 to 100 % and must be analyzed individually for each material. GWP's relative contribution is seen in Figure B (a). JO and CO are the PCMs in which farming contributes to more than 50 %. MA and SA production GWP is also dominated by farming, although it is slightly higher than manufacturing. In the case of JO and CO, the results point to an agricultural activity related to high carbon footprint emissions as compared to the other unit processes. XY and AA equivalent emissions are dominated by the manufacturing sector. Transportation contributed the least with the exception of CO, where transportation slightly contributed more than manufacturing. Coconut transportation from farming to manufacturing sites requires a large volume per amount of produced oil, demanding more from transportation and contributing more to the GWP compared to manufacturing and other PCMs. Indeed, according to the European Environment Agency, road transportation is one of the largest sources of greenhouse gas emissions in the EU [50]. On the other hand, manufacturing is the critical unit process for the rest of the PCMs.

PMFP, which combines the effects of NO_x, NH_3, SO_2, or $PM_{2.5}$

Table 2
Summary of the total environmental impact using the LCA methodology.

Indicator	Unit	JO	CO	LA	MA	SA	XY	AA
GWP	kg CO_{2eq}	9.40E-01	8.19E-01	1.68E+00	4.37E+00	7.06E+00	9.23E+00	1.30E+01
ODP	kg CFC_{11eq}	2.70E-06	1.66E-06	2.36E-06	6.54E-06	1.08E-05	3.66E-06	2.04E-05
IRP	kBq Co-60_{eq}	4.88E-02	3.74E-02	5.96E-02	1.62E-01	2.65E-01	1.82E-01	8.33E-01
HOFP	kg $NO_{x\ eq}$	2.41E-03	2.30E-03	3.91E-03	1.05E-02	1.71E-02	1.64E-02	3.18E-02
PMFP	kg $PM_{2.5eq}$	1.87E-03	1.33E-03	2.32E-03	6.20E-03	1.01E-02	2.79E-02	2.09E-02
EOFP	kg $NO_{x\ eq}$	2.51E-03	2.40E-03	4.10E-03	1.10E-02	1.79E-02	1.74E-02	3.36E-02
TAP	kg SO_{2eq}	4.09E-03	2.78E-03	5.03E-03	1.34E-02	2.17E-02	8.87E-02	5.48E-02
FEP	kg P_{eq}	7.22E-04	4.90E-04	8.30E-04	2.23E-03	3.63E-03	3.45E-03	8.52E-03
MEP	kg N_{eq}	8.09E-05	2.97E-05	4.75E-05	1.29E-04	2.11E-04	5.99E-04	4.25E-03
TETP	kg 1,4-DCB	2.37E-01	2.96E-02	5.01E-02	1.34E-01	2.18E-01	2.42E-01	3.12E+00
FETP	kg 1,4-DCB	2.52E-03	3.08E-04	4.82E-04	1.31E-03	2.14E-03	1.31E-02	7.74E-02
METP	kg 1,4-DCB	4.54E-03	3.87E-03	6.55E-03	1.75E-02	2.85E-02	2.13E-02	9.32E-02
HTPc	kg 1,4-DCB	1.20E-03	3.54E-04	6.23E-04	1.68E-03	2.74E-03	4.16E-03	2.68E-02
HTPnc	kg 1,4-DCB	1.34E-02	8.57E-03	1.33E-02	3.61E-02	5.91E-02	1.28E-01	1.48E-01
LOP	m^2a $crop_{eq}$	2.41E-02	2.45E-02	3.74E-02	1.02E-01	1.68E-01	5.36E-01	2.15E+00
SOP	kg Cu_{eq}	1.23E-02	6.01E-03	9.05E-03	2.47E-02	4.06E-02	2.34E-01	7.17E-02
FFP	kg oil_{eq}	9.03E-02	7.26E-02	1.30E-01	3.46E-01	5.64E-01	4.83E-01	1.16E+00
WCP	m^3	3.46E+00	1.76E+00	2.41E+00	6.75E+00	1.11E+01	9.49E-01	2.41E+00

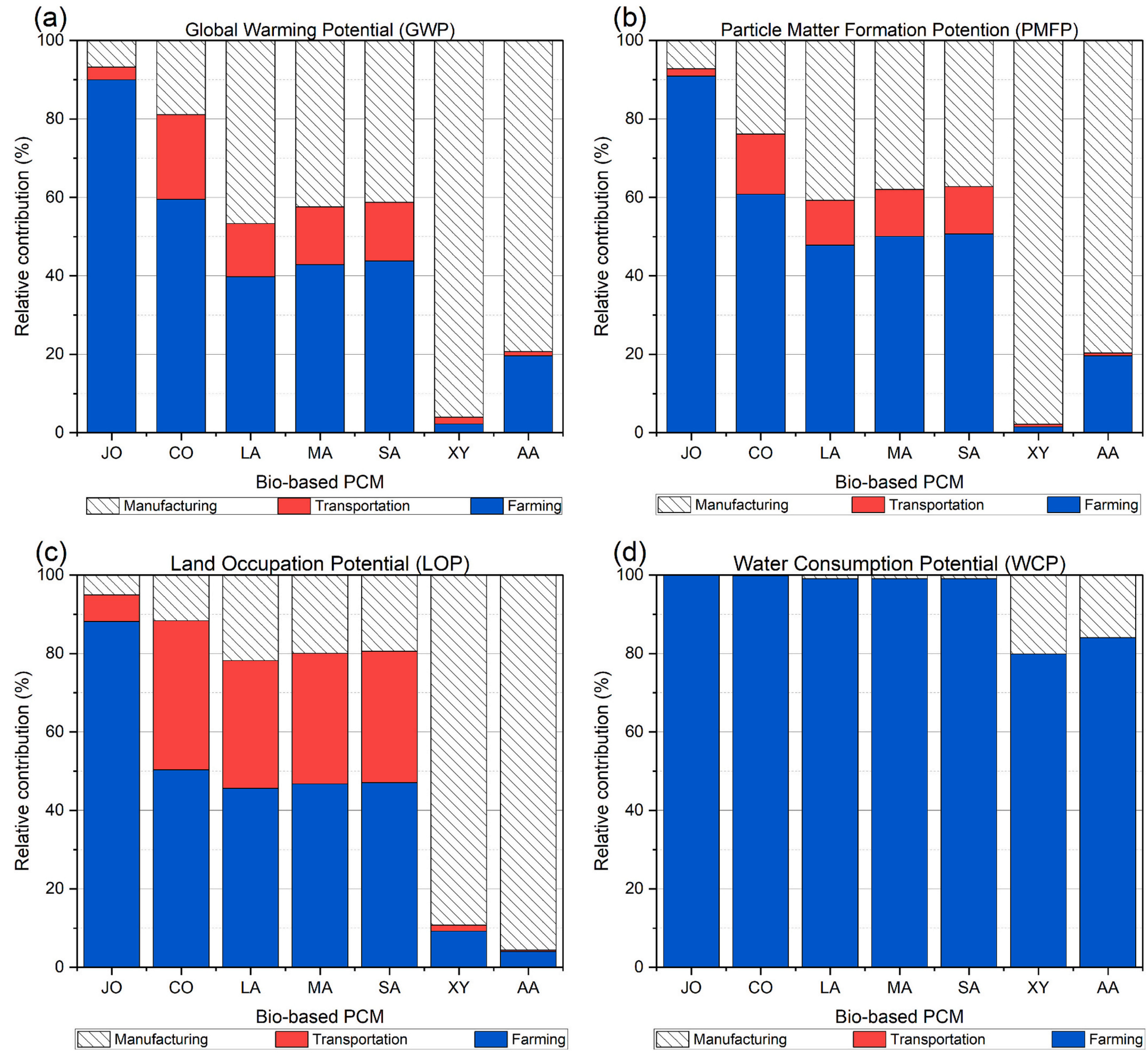

Fig. 7. Percentage contribution of farming, transportation, and manufacturing to the environmental indicator of individual PCM: (a) GWP; (b) PMFP; (c) LOP; (d) WCP.

emissions, atmospheric fate, and chemistry to measure the human intake of $PM_{2.5}$ and its final impacts on human health, is shown in Figure B(b). The value of PMFP for JO, CO, LA, MA, and SA production is dominated by the farming activities, within a range of 90 % to 47 %. XY and AA production equivalent emissions of particulate matter come mostly from manufacturing, with 97.8 % and 78.6 %, respectively. Transportation contributed the most to the PMFP of CO, reaching a maximum of nearly 15.3 %. The results show that for PCMs like XY and AA, aiming at manufacturing may be a priority if reducing the PMFP is desired, while for the rest of the materials, the attention shifts to farming activities.

Different from the GWP and PMFP, LOP shows a higher participation of transportation in the total impact of producing the PCMs. Once again, farming is the leading activity contributing to LOP for JO, CO, LA, MA, and SA. Meanwhile, the XY and AA impact of LOP is caused primarily by the manufacturing activity, reaching more than 85 %. Transportation, on the other hand, shifted to impressive values surpassing the effect of manufacturing on the LOP of JO, CO, LA, MA, and SA. A coherent interpretation of the relative contribution is given by considering that, although farming dominates in the production of JO, its impacts are much smaller than those of AA, as can be seen in Fig. 2.

The contribution based on the individual input is presented in Fig. 8. Similarly to the results obtained from Fig. 7, the same interpretation can be applied. The sum of all inputs for each PCM adds to 1. The objective is to support the previous results of identifying the most contributing unit process by now indicating how the inputs behave individually. For example, the farming process for JO is composed of inputs x_1 to x_6 (Irrigation water, nitrogen fertilizer, phosphorous fertilizer, diesel, pesticide). The GWP of this PCM is mostly affected by x_1 and x_6, followed by x_4 in the farming process, with overall contributions of 35.17 %, 21 %, and 20.53 %, respectively. It was identified that farming activity is the main factor responsible for the WCP of JO, and the results show that it is mainly a consequence of irrigation activity, attributed to input x_1.

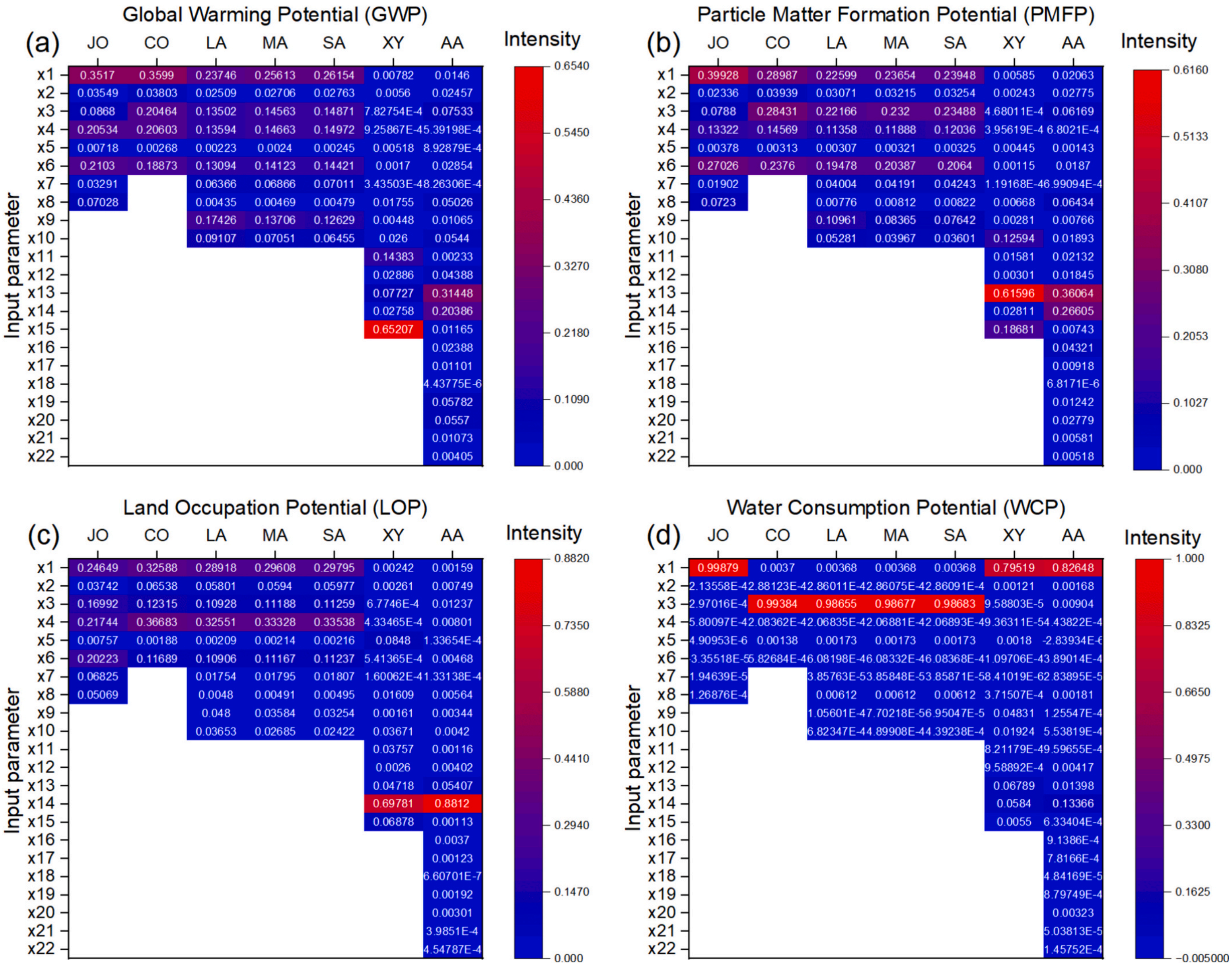

Fig. 8. Percentage contribution of inputs to each PCM: (a) GWP; (b) PMFP; (c) LOP; (d) WCP.

3.3. Local sensitivity analysis

The LSA was carried out based on the lower and upper bounds of each input after sampling with the Monte Carlo method. The triangular distribution function fitted well due to the range of variation for each input, i.e., a maximum and minimum value. Using eq. 3, the sensitivity was calculated for all PCMs. Unlike the contribution analysis, the LSA helps identify the maximum and minimum values of each environmental impact indicator when a specific input reaches its upper and lower bounds, respectively. Indeed, it is another method to help identify which input is most critical in an LCA, but it should not be confused with the contribution, which refers to the percentage that an input has contributed to the total value of a defined indicator. An LSA is conducted in the LCA to see the inputs behavior for GWP, PMFP, LOP, and WCP of JO, shown in Fig. 9. In Fig. 9 (a), GWP is highly sensitive to input x_1 (irrigation water), for which a variation between its lower and upper bounds can reduce the GWP by nearly 33 % and increase it by approximately 38 %, respectively. Inputs x_4 and x_6 (potash and pesticide) manage to hold a reduction of almost 19 % and an increase of nearly 22 % in the total amount of kg $CO_{2\ eq}$. All of these inputs are located in the unit process farming, the most critical unit process, in agreement with the results found in the relative contribution section. PMFP sensitivities to inputs are seen in Fig. 9 (b). Coincidentally, x_1 variation implicates in more than 40 % increase and a decrease of nearly 37 %, causing the most sensitivity to this indicator, followed by x_6. Once again, it shows that farming has parameters that considerably impact the formation of particulate matter. An analysis of the sensitivity index of the LOP, depicted in Fig. 9 (c), shows it is very sensitive to four parameters, being x_1 the most influential input with more than 20 % impact in the LOP, followed by x_4, x_6 and x_3 (phosphorous fertilizer), all of them belonging to the farming unit process. Unlikely, WCP, shown in Fig. 9 (d), is extremely sensitive to only x_1, with an index of nearly 100 %, due to the need for high water volumes necessary for the irrigation process. Having the WCP as a target for mitigating impacts, the changes in irrigation techniques and improvement in the process efficiencies can benefit the sensitivity to this input.

3.4. Local sensitivity analysis

Similarly, a sensitivity analysis of the indicators for AA production can be conducted based on Fig. 10. In contrast, AA contains 22 inputs in the production process, allowing for mitigation through the adjustment of various parameters. The GWP, shown in Fig. 10 (a), was found to be most sensitive to input x_{13} (potassium hydroxide), indicating that when this input reaches its upper bound, the amount of carbon dioxide equivalent peaks by about 30 %, while reducing the input to its lower bound can result in a reduction of approximately 28 % for this indicator. This sensitivity is followed by indicator x_{14} (enzymes), which can lead to

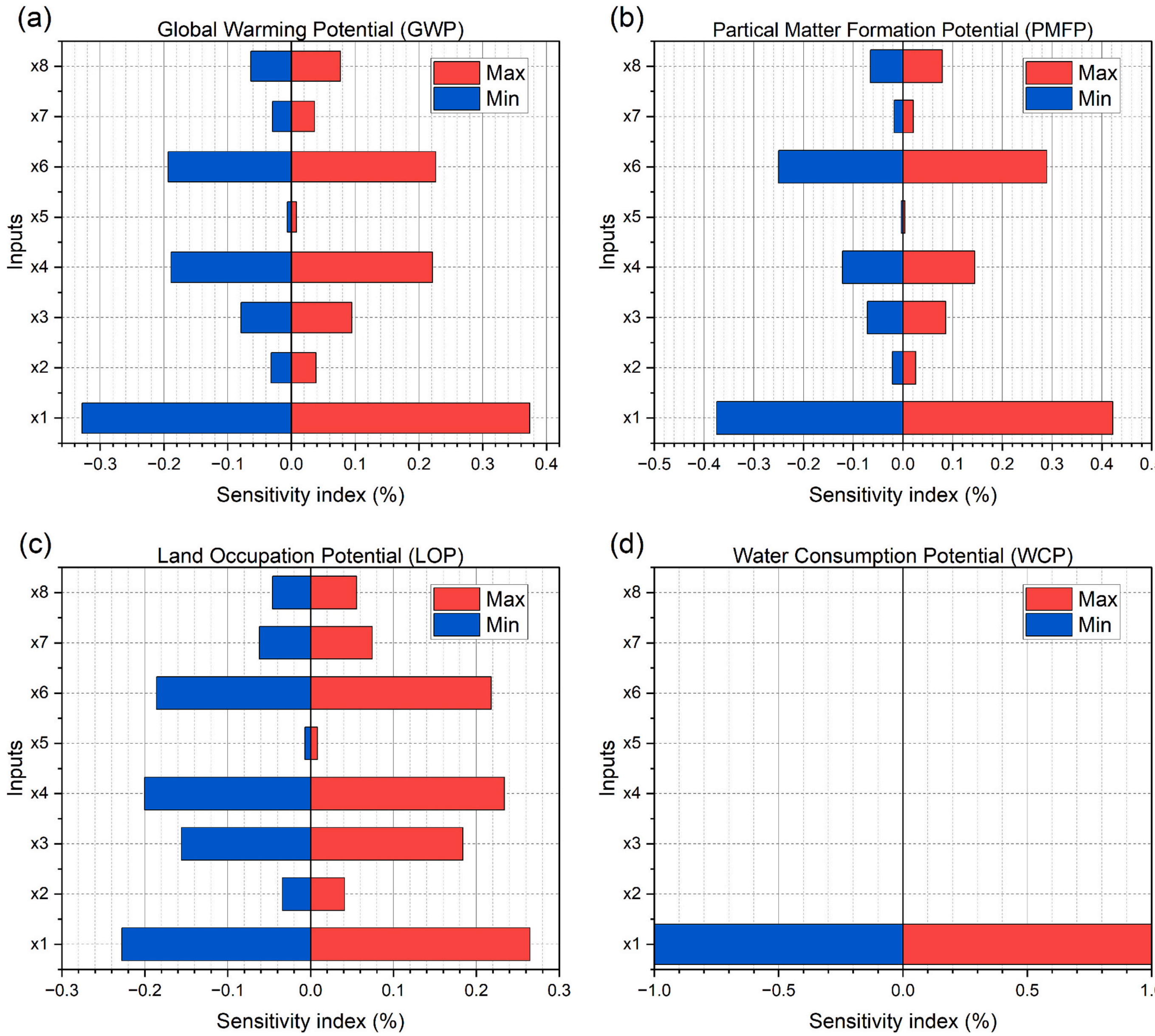

Fig. 9. Local sensitivity analysis using one-factor-at-a-time theory for JO: (a) GWP; (b) PMFP; (c) LOP; (d) WCP.

an increase of approximately 22 % and a decrease of 18 %. These inputs are located in the manufacturing unit process, which, in the case of AA, is the most contributing unit process. Other inputs also contribute to the mitigation of GWP values, but with sensitivity indices below 10 %, as can be observed. Regarding PMFP, similarly to GWP, inputs x_{13} and x_{14} exhibit the highest sensitivity. PMFP can reach a maximum of 38 % and a minimum of 34 % if x_{13} reaches its upper and lower bounds, respectively, while varying x_{14} results in PMFP values fluctuating between 20 % and 30 %. Interestingly, LOP and WCP are highly sensitive to only one input. The sensitivity index of LOP to x_{14} is nearly 90 %. According to the dataset used to represent x_{14}, this activity is strongly linked to the use of electricity and steam in the production of the enzymes. In the case of WCP, sensitivity is associated with activities related to agriculture, predominantly linked to irrigation, represented by input x_1 (irrigation water). The sensitivity index shows that when aiming for WCP, maximum and minimum values of approximately 99 % can be achieved.

3.5. Global sensitivity analysis

The Sobol indices for all PCMs and the four discussed indicators are presented in Fig. 11. The results of the Sobol indices for inputs with GWP are shown in Fig. 11 (a). The uncertainty of GWP is impacted mostly by input x_1 (irrigation water) for PCMs from JO to SA, with minor contributions from other inputs, such as x_3, x_4, x_6, and $x_{9,}$ responsible for more than 10 % of LA's uncertainty. XY variance, on the other hand, is dominated by one input only, x_{15} (steam), while AA uncertainty comes mainly from x_{13} (potassium oxide) and x_{14} (enzymes). These values are implicated by the effects not only of the upper and lower bounds but also the intensity of the coefficient a_i, in such a manner that even if a parameter varies in a short range, it can have a significant uncertainty contribution if the ai for the input is significantly higher. A similar interpretation can be carried out for the other indicators presented in Figs. 11 (b), (c), and (d). WCP uncertainty, as seen, is impacted mainly by the uncertainty of one input only, such as x_1 for JO, XY, and AA (nitrogen fertilizer), and x_3 for the rest of the PCMs.

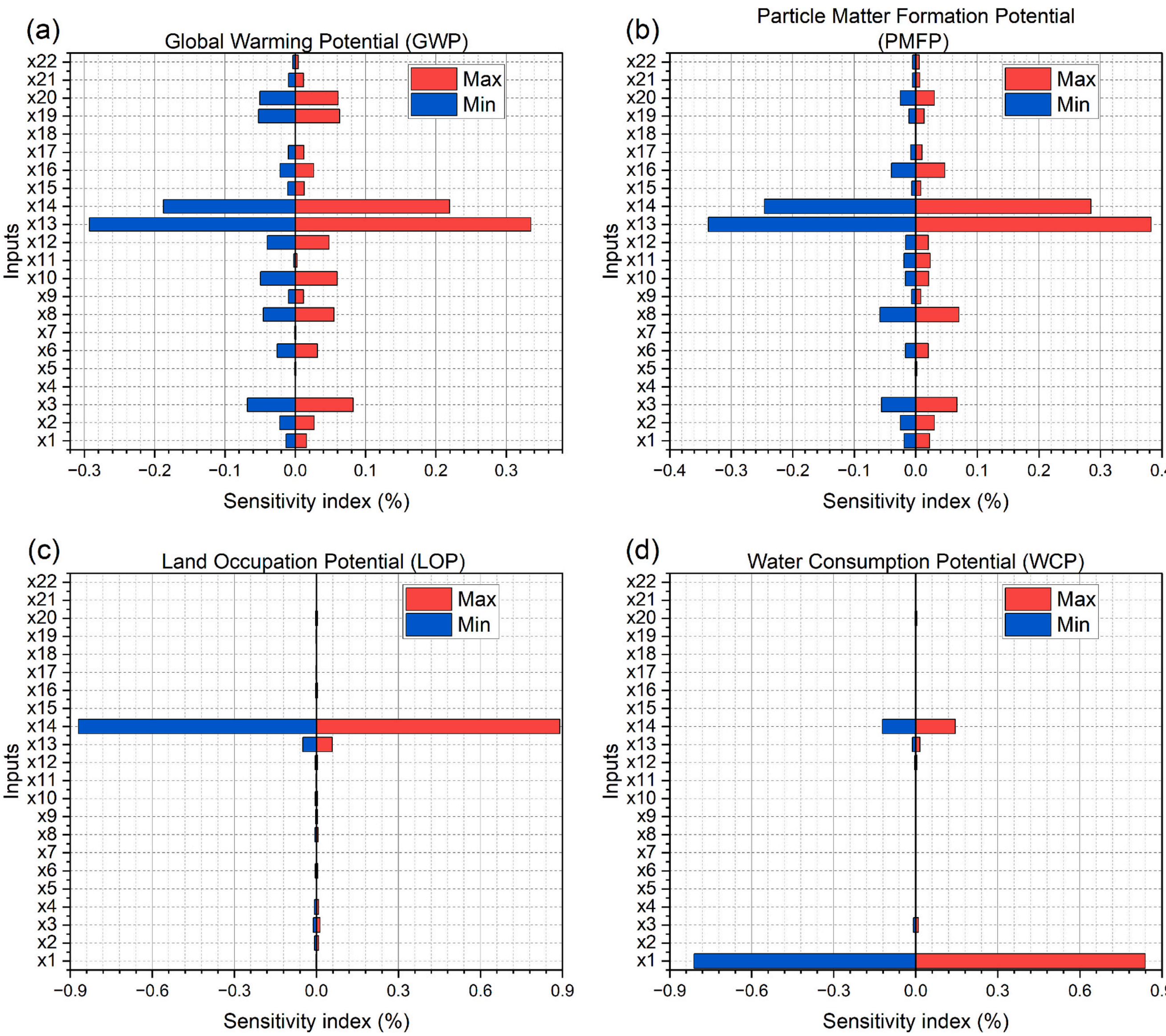

Fig. 10. Local sensitivity analysis using one-factor-at-a-time theory for AA: (a) GWP; (b) PMFP; (c) LOP; (d) WCP.

3.6. Decision-making based on contribution and sensitivity analysis

This section summarizes how the interpretations from the contribution and sensitivity analysis can lead to decisions regarding the environmental impacts of the addressed bio-based PCMs. Calculating the relative contribution helps locate the critical unit process or individual input variable, as shown in Figs. 3 and 4. It represents a stationary percentage share of unit processes or inputs to a specific environmental indicator. This discussion refers to bio-based AA to explain more clearly the interpretation of such indices and how it can support decision making in mitigating environmental impacts associated to the production of PCMs. One example is identifying the hot spots of PFMP in the production of AA, where manufacturing is responsible for nearly 80 % of this indicator, which in turn inputs x_{13} (potassium hydroxide) and x_{14} (enzymes), holding up to more than 60 % of these indicators. It can lead to solutions in replacing these inputs with lower environmental impact, although care must be taken to respect process efficiency requirements.

On the other hand, if the intention is to understand how the variation of these inputs and their uncertainties impact an indicator, LSA and GSA are used, respectively. For example, the LSA of x_{13} and x_{14} tells to what extent the output (in this case the LCA indicator) will increase or decrease when these inputs reach their upper and lower bounds, respectively. Considering the Monte Carlo sampling carried out, the lower and upper bounds of indicator x_{13} are 0.95 and 1.16 kg, respectively, which means that if x_{13} reached such values, the PMFP could experience a reduction of nearly 34 % and an increase of 38 %. Attaining the lower bound of an input would be the optimal solution, meaning that the environmental impact would reduce to its lowest value based on a single input, however, this is not always the case due to the interaction of multiple variables and the consequences on the operational requirements. In this way, LSA can be used as a tool for calculating the effects on the environmental impacts, caused by the variations attributed to each parameter, paying attention to the specificity of each process and respecting the ranges of variation of each variable in a real process. In this study, the upper and lower bounds obtained from Monte Carlo sampling were used, but in any case, the LSA allows investigating the variations of small changes such as 1 % or any other desired value. On the other hand, the GSA will show the uncertainty contributions to an indicator. For the same material (AA), it is seen that x_{13} has a Sobol Index of 0.6078 in the PMFP, meaning it is to more than 60 % of this indicator's uncertainty. It is then followed by input x_{14} with a Sobol Index of 0.33. More than 90 % of the uncertainty is a consequence of two

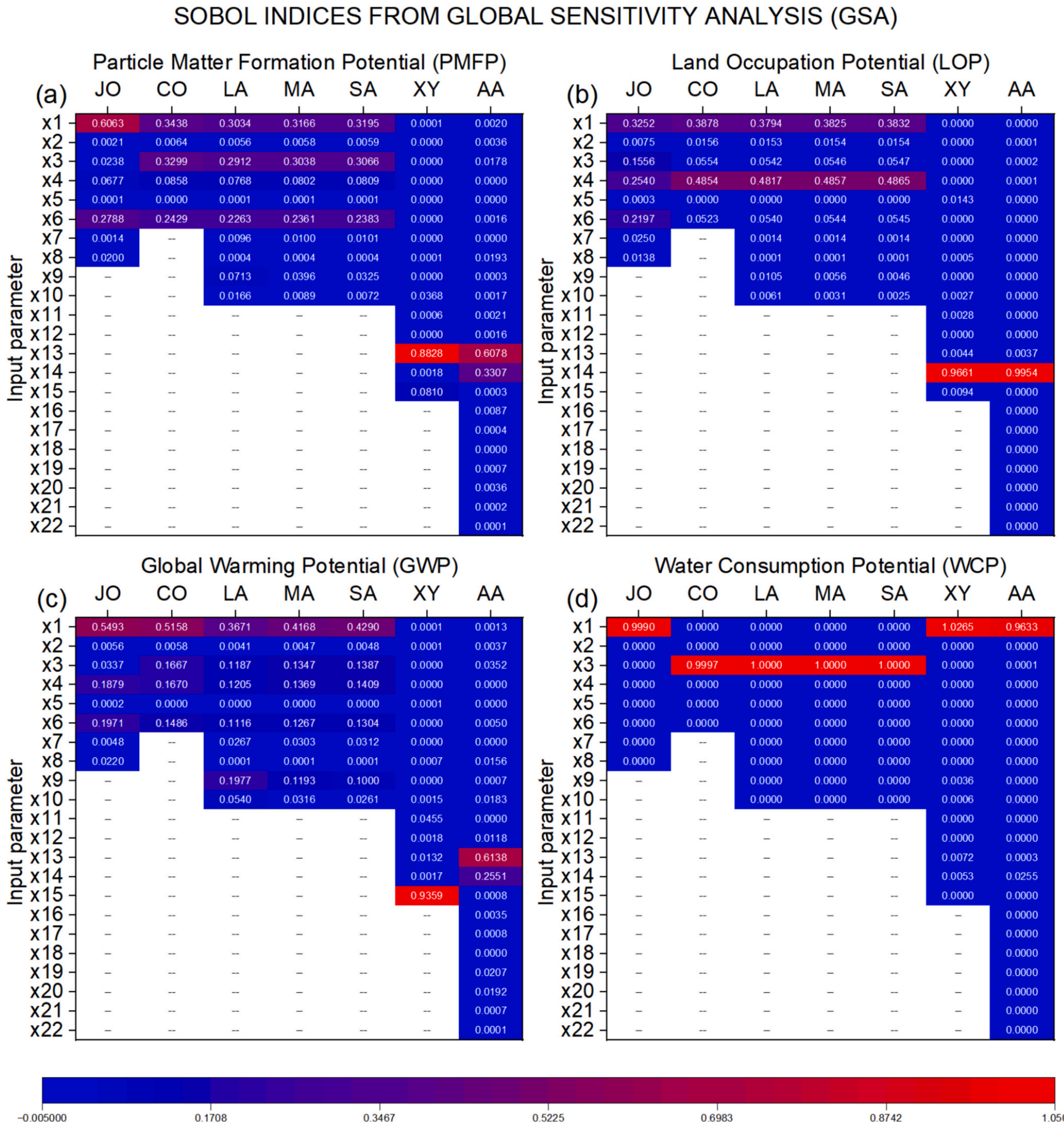

Fig. 11. Global sensitivity analysis with Sobol índices for the indicators (a) GWP; (b) PMFP; (c) LOP; (d) WCP.

indicators only. Then, GSA could be used to target actions against the uncertain outputs, which could be obtained by looking carefully at the variance or standard deviation of an input. It is important to bear in mind that results of relative contribution and sensitivity analyses might not always indicate the same input(s) as being the most important. In the case mentioned, the inputs with high contribution rates coincide with high sensitivity. However, i an input amount is low but is very uncertain, it might result in low relative contribution and high sensitivity. Additionally, the mathematical model used to calculate sensitivity may influence this behavior between relative contribution and sensitivity. In this study a linear model was used, such as Eq. 1, but if a different model with variables correlating among themselves was applied distinct dynamism should be expected. If the analysis led to decision to mitigate the environmental impacts of such processes, the use of cleaner or more efficient technologies in the production of phase change materials could reduce energy consumption and pollutant emissions, reducing environmental impact. This contributes to more sustainable processes, with less waste generation and greater use of natural resources.

Identifying the critical inputs with contribution and sensitivity analysis may facilitate finding strategies and develop policies to mitigate environmental impacts, such as the GWP in the production of bio-based PCMs. For instance, in the phase of farming, strategies can concentrate on water management. The use of newly developed technologies such as smart irrigation systems using artificial intelligence (AI), remote sensing, the Internet of Things (IoT), and machine learning (ML) enhances agricultural sustainability [51]. Also, the integration of renewable energy sources in the farming step could be a solution for those activities that rely on the use of diesel fuel, for instance [52].

One of the limitations of this work regards the study of specific waste generation in the production scale of the PCMs, including the farming, transportation and manufacturing phases. For instance, several types of waste can be produced in the manufacturing chain, and the effects that they have on the LCA as well as the resource consumption for their treatment could be investigated in more detail. One approach could be close partnership between manufacturers and LCA researchers, that could result in exchange of important and robust information for the improvement of LCA results. This could be positive and help reduce the number of assumptions that are made during waste treatment.

4. Conclusions

In summary, this study investigated the environmental impacts of bio-based PCMs by looking at overall indicator results and including a robust methodology to account for the effects of uncertainty propagation. The procedure included relative contribution, local sensitivity analysis, and global sensitivity analysis to support decision-making regarding the environmental impacts in the production of selected materials. The main conclusions drawn from this work are presented.

- Based on the results of the inventoried material, the overall GWP potential showed an increasing pattern. With the exception of CO, the higher the melting temperature of the PCM, the higher the GWP. For instance, the production of 1 kg of JO is associated with 0.96 kg $CO_{2\ eq}$, while the same amount of AA points to 13 kg $CO_{2\ eq}$. However, not all indicators follow this pattern.
- Relative contribution is useful in locating the most contributing unit process or input to an environmental indicator. The results represent punctual percentages but are helpful in deciding which unit process or input needs to be prioritized in the environmental assessment of a process.
- LSA was carried out based on the lower and upper bounds of each input after Monte Carlo sampling. LSA provides important information regarding changes to the environmental indicators in the production of the PCMs and can be useful to align strategies in reducing the environmental impacts.
- Additionally, this study also showed that GSA can be an asset to support decision-making by identifying the parameters that contribute the most to the indicator's uncertainty.

CRediT authorship contribution statement

Humberto Santos: Writing – review & editing, Writing – original draft, Software, Methodology, Conceptualization. **Silvia Guillen-Lambea:** Writing – review & editing, Supervision, Resources, Funding acquisition.

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Declaration of competing interest

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

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.est.2025.118961.

Data availability

We declare that all data supporting and generated in this study are available as stated within the text, as well as in the supplementary material.

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