



## Research article

# A life cycle assessment of CCU process to produce a nanocomposite from ethanol plant CO<sub>2</sub> emission

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## ABSTRACT

The use of rubber septa for controlled release of semiochemicals has raised important discussions about their efficiency and environmental impact since they are composed of fossil raw material. The life cycle assessment (LCA) of the synthesis of a nanocomposite type calcium nanocarbonate/Kraft lignin (NC-CN-KL) obtained from CO<sub>2</sub> capture aimed to quantify the environmental loads involved in the production process. The synthesis evaluated by the LCA was performed on a laboratory scale, since it is a synthetic route classified as technological readiness level 4 (TRL-4) and is in the study and development stage. The LCA was performed according to the principles of the ISO 14044/2006 series of standards, from 101.854 g of nanocomposite as a functional unit. The limitations of the study arose from its synthesis scale, absence of LCA data on the rubber septa and other nanocomposites. The results obtained in the LCA identified electricity and other energy generation processes as the largest contributors to environmental loads for all environmental impact categories studied and suggest that research should focus on these inputs when choosing the sources used in energy nanocomposite formulation processes. LCAs for this synthesis to obtain NC-CN-KL should be carried out on a pilot scale, and it is expected that this work will contribute to the formulation of the material and decision-making, especially regarding the choice of the energy matrix.

## 1. Introduction

Climate change and all its consequences have been discussed vigorously and exponentially by the scientific community in recent decades. Global warming, a product of the triggering of the greenhouse effect, is an alarming signal for the whole world, as it affects humanity through the exacerbated increase in temperature on Earth [1]. This is due to the massive emissions of greenhouse gases (GHG) (e.g., carbon dioxide, methane and nitrous oxide) into the atmosphere [2], especially from anthropogenic activities, which have affected humans and the environment in which they live.

Because of this, many efforts are being focused on achieving net zero GHG emissions, which corresponds to a balance between the gases emitted and the gases collected from the atmosphere [3,4], with the main goal of keeping the temperature increase below 2 °C in

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the coming years [5].

GHGs can be found from natural activities such as the respiration of living beings and the decomposition of matter, and from industrial activities, of anthropogenic origin [6]. Among the gases that influence the greenhouse effect, carbon dioxide (CO<sub>2</sub>) stands out, due to its excessive abundance compared to other gases [7].

Causing heat retention in the ozone layer, GHGs come from various anthropogenic activities such as energy generation [8], construction [9] and automobile use [10]. These are, therefore, essential activities for the daily life of human beings, but they require a thorough look at the control of disordered GHG emissions.

To this end, countries, entities and researchers are focused on discussing strategies to mitigate GHG emissions, mainly targeting the activities with the greatest impact and CO<sub>2</sub>, one of the GHGs that absorbs energy in the Earth's cover [11]. Global conventions such as the Paris Agreement and the United Nations Framework Convention on Climate Change have triggered the first steps to outline GHG mitigating actions.

Among the most prominent actions today are the technologies related to Carbon Capture and Storage (CCS) [1] and Carbon Capture and Utilization (CCU) [12], which deal with the capture and use of CO<sub>2</sub> as a raw material, respectively.

CCS is based on the principle of long-term storage of CO<sub>2</sub>, allowing for the integration of better strategies to use this gas in the future [13] and CCU prioritizes the use of CO<sub>2</sub> as a raw material to obtain other value-added materials [14]. There is also a significant prospect for the integration of CCS and CCU through Carbon Capture Storage and Utilization (CCSU), which aims to incorporate large GHG emitters such as power plants [15,16] and the mining sector [12].

The use of CO<sub>2</sub> as an input [17] is a promising alternative for GHG mitigation [18,19]. Many studies have been developed with the purpose of evaluating the environmental loads of these technologies and validating the use of CO<sub>2</sub> as a raw material [17,20,21].

Nanotechnology, for example, has been highly discussed and noted as a promising technology for obtaining value-added materials from captured CO<sub>2</sub> [22]. [6] presents a survey of the main nanomaterials obtained from captured CO<sub>2</sub>, among them are nanocomposites, functionalized nanomaterials, nanocatalysts and nanore coatings.

For agriculture - a sector that contributes significantly to GHG emissions [19] - the issue of nanomaterials can favor both CO<sub>2</sub> mitigation and obtaining nanocomposites for controlled release and nanocorrectives for the soil [23–25].

In this sector (agriculture), nanocomposites - such as the one obtained through the synthesis described in this work – can have the objective of controlled release of semiochemicals (natural compounds from animals and plants), which in turn have the mission of providing pest control in agricultural systems [26]. A suitable controlled release of semiochemicals depends on the release rates in the medium and has therefore been the reason for great efforts to optimize existing technologies, in addition to the sustainable appeal that comes from the change in the use of rubber septa, a fossil-based input.

To this end, validation is required for these technologies to be implemented in GHG emitting activities, especially since they are new technologies that are at an initial Technology Readiness Level (TRL) scale [27].

To quantify the environmental impacts of a given process, quantification models are proposed, selected, and applied according to the research needs [28]. used a quantification model that determined the fouling rate of a membrane bioreactor, which was necessary to evaluate the cleaning rate of the bioreactor. Another study showed that the use of a quantification model for the contribution of membrane cleaning can improve absorption, filtration, and performance mechanisms [29]. This is because the main obstacle to membrane technology for water and effluent treatment is fouling [30]. Therefore, the use of quantification models can assist in decision-making in processes in the most diverse areas, favoring both the process stages and the disposal of their inputs in the environment. Life Cycle Assessment (LCA) is a tool that measures the environmental impacts of a given process or product from the steps involved in the system [31], allowing the comparison of alternative technologies with conventional technologies [32]. In the case of GHG mitigation technologies, LCA's are performed focusing on CO<sub>2</sub> emissions [8] and the decarbonization potentials of technologies can be assessed.

Leonzio et al. (2023) used LCA to compare the use of CCSU, simulating the use of CO<sub>2</sub> at larger scales and were able to verify that the tool validates the use of CO<sub>2</sub> as an alternative for GHG emissions [33]. Through LCA Kim et al. (2019) can evaluate CO<sub>2</sub> sequestration in an energy plant [34]. The tool also allows analyzing the reduction of all impact categories [35] and emissions of other substances such as (sulfur dioxide) SO<sub>2</sub> and (nitrous oxide) NO<sub>x</sub> [36].

In view of this, this work aimed to measure the environmental impacts of a synthesis to obtain a nanocomposite for use in agriculture of the calcium nanocarbonate/Kraft lignin type, obtained from the precipitation of CO<sub>2</sub> from ethanol plants. For this, a gate-to-gate LCA was performed, analyzing the steps involved during the laboratory process. It was then possible to highlight the integration of CCU in GHG emitting activities and the use of this gas, which in addition to being made feasible by its abundance, has added value.

## 2. Materials and methods

### 2.1. Materials

The raw materials used to obtain the NC-CN-KL were of high purity grade, except when informed: calcium chloride dihydrate (CaCl<sub>2</sub>·2H<sub>2</sub>O) from Sigma Aldrich; distilled water; Kraft lignin provided by a paper industry (Suzano Paper and Pulp); ammonium hydroxide (NH<sub>4</sub>OH) from Sigma Aldrich; carbon dioxide (CO<sub>2</sub>) from White Martins (volumetric composition of 100 % of CO<sub>2</sub>, simulating the ethanol plant emission); acetone (C<sub>3</sub>H<sub>6</sub>O) from Sigma Aldrich; and electricity from the local power grid.

## 2.2. CCU process development for the nanocomposite production

The synthesis of the NC-CN-KL – a CCU process - was initiated by means the bubbling of CO<sub>2</sub> in calcium chloride dihydrate solution (CaCl<sub>2</sub>·2H<sub>2</sub>O), in order to obtain an inorganic-organic carrier for controlled release of agrochemicals (i.e., semiochemical and antibiotics) for pest control in agriculture and livestock. The applied methodology and the product characterization can be accessed in a previous published article [25].

The synthesis carried out in multiple steps, which were summarized and explained in the flowchart presented in Fig. 1, in order to understand, subsequently, the choice of system boundaries for performing the LCA.

## 2.3. The LCA description for the CCU process

The life cycle assessment of the synthesis followed the ISO 14044/2006 [31] standard series on Environmental Management, which addresses the requirements and guidance for performing LCA.

The LCA was performed in the SimaPro® software, version 9.2.0.1, with data from the Ecoinvent database version 3.7.1, allocation, cut-off by classification-unit.

## 2.4. Goal definition, functional unit, and system boundaries

The objective of this LCA was to evaluate the environmental loads of the synthesis to obtain the NC-CN-KL. Since it is a technology under development and classified as TRL-4, the function of this study was to explain the potential impacts of the synthesis performed in laboratory scale, aiming its application in pilot scale. The function of this system is to replace conventional petrochemical carrier as the rubber septa used for pest control in agriculture. The system boundaries (Fig. 2) were defined as gate-to-gate in order to understand the impacts of the process of obtaining the nanocomposite. The CO<sub>2</sub> capture and material application steps were not considered.

## 2.5. Inventory

The data of inputs and outputs the synthesis for obtaining the NC-CN-KL and their respective quantities and qualities of data collection are presented in Table 1. All inventory data was defined through laboratory tests and optimized (during syntheses) in order

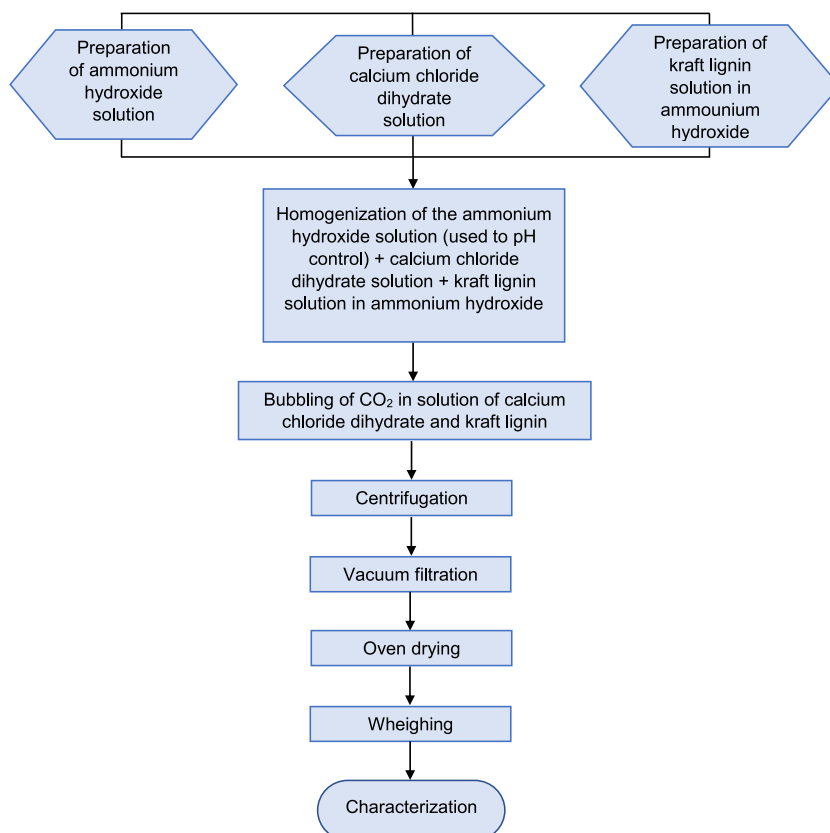


Fig. 1. Flowchart of obtaining the inorganic-organic NC-CN-KL.

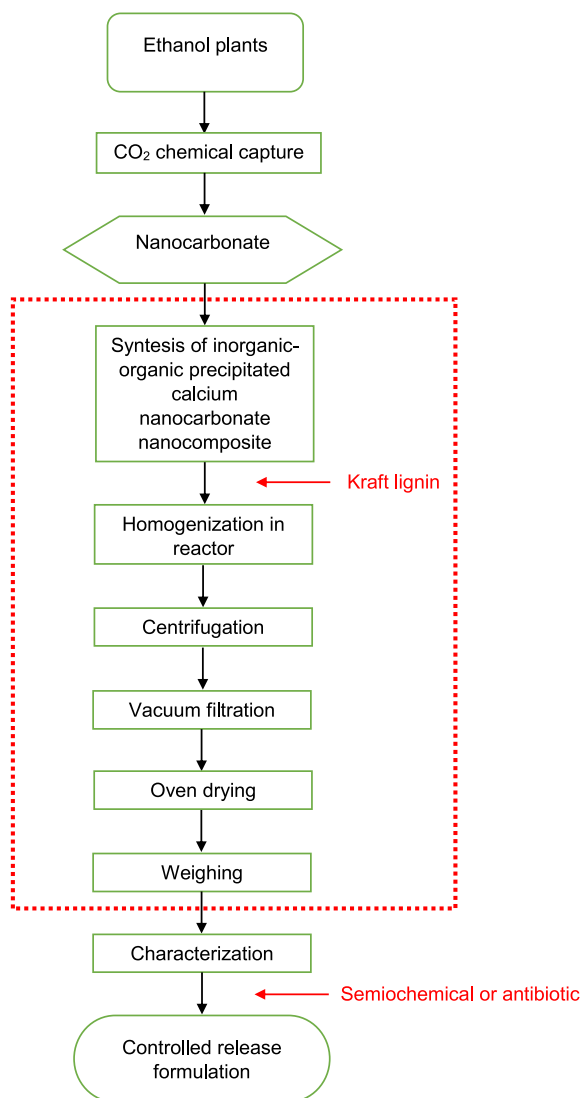


Fig. 2. System boundaries highlighted in red.

Table 1

Inputs and outputs of the synthesis process for the obtention of the nanocomposite.

Reagents and products	Quantity	Unit	Quality
<b>Inputs</b>			
Calcium chloride dihydrate (CaCl <sub>2</sub> ·2H <sub>2</sub> O)	14.709	g	Checked
Kraft lignin	36.0093	g	Checked
Ammonium hydroxide solution 29 % (NH <sub>4</sub> OH)	125	mL	Checked
Distilled water	1574.2817	mL	Checked
Acetone	50	mL	Esteemed
Carbon dioxide (CO <sub>2</sub> )	75	m <sup>3</sup>	Checked
Distilled water (nanocomposite recovery)	800	mL	Esteemed
Electricity	78.35	kWh	Esteemed
<b>Outputs</b>			
Composite	101.854	g	Verified
Supernatant (water + waste)	1350	mL	Esteemed
Nanocomposite recovery water	800	mL	Esteemed

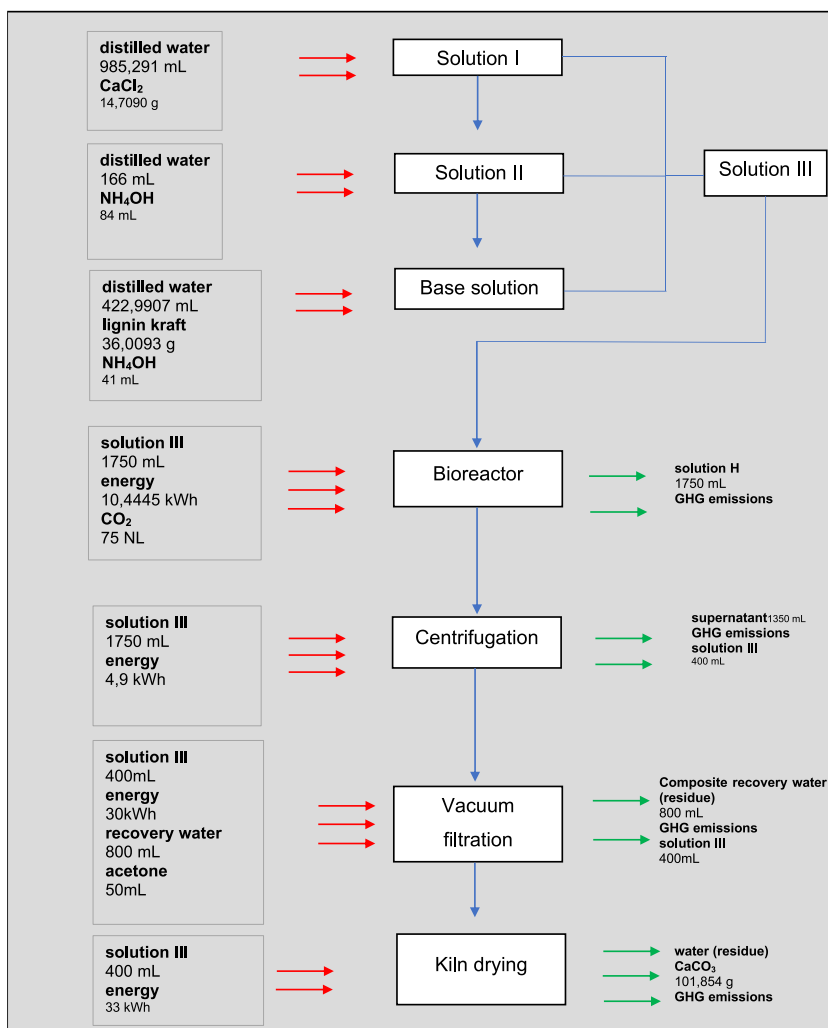


Fig. 3. Flowchart for obtaining the Kraft nanocarbonate/lignin composite.

to guarantee the yield for obtaining the nanocomposite.

Data validation was performed by mass balance (Fig. 3) and was correlated with the functional unit. It should be noted that emissions from the bioreactor, centrifugation, filtration, and drying stages were attributed to the use of electrical energy.

Limitations were found regarding the supply of the inventory data for the Kraft lignin procurement process. Both the supplier and the software databases had no inventory data. To insert the Kraft lignin production process in the LCA of the synthesis for obtaining the nanocomposite, a literature search was performed and the inventory developed by Bernier et al. was adopted [37].

## 2.6. Environmental impact evaluation

The impact categories were defined according to comprehensive literature on CO<sub>2</sub> capture technologies, especially because it is a nanocomposite synthesis integrated with the chemical capture of CO<sub>2</sub>, being the most relevant: climate change, terrestrial acidification, freshwater ecotoxicity, fossil depletion, freshwater eutrophication, ozone depletion and human toxicity [38–42].

The analysis model adopted in this LCA was the ReCiPe Midpoint (H) version 1.13/World Recipe (2010) H, as it associates emissions and their quantification with specific environmental impact indicators, based on impact categories, considering those mentioned above. As for the analysis of LCA impacts, the characterization stage (mandatory) was considered to identify the substances of greatest impact for each category and to quantify the overall environmental loads.

## 2.7. Interpretation

Following the considerations of ISO 14044/2006 significant issues were identified in this step, data from the previous steps were reviewed through sensitivity analysis, conclusions, limitations and recommendations were formulated from the results of the

environmental impact assessment.

The data review was performed through data revalidation and therefore consistency and sensitivity review. From this, all limitations and recommendations resulting from this LCA came from the nanocomposite synthesis.

### 3. Results and discussion

#### 3.1. Environmental impact evaluation

From the seven categories of environmental impacts selected as described in section 3.3, this section presents the results for each of them, highlighting the substances of greatest contribution to their environmental loads.

Fig. 4 presents the graph of contribution obtained at 1 % cutoff point for the environmental impact category of climate change. In this category, all contributors are GHG, with emphasis on carbon dioxide of fossil origin with 70 % contribution.

Fig. 5 presents the graph of contribution obtained at 1 % cut-off point for the ozone depletion environmental impact category, highlighting the methane gas substance as the main contributor.

Fig. 6 presents the contribution graph obtained at 1 % cut-off point for the environmental impact category terrestrial acidification. The main contributing substances of the inventory for this category were sulfur dioxide, nitrogen oxide and ammonia.

Fig. 7 presents the graph of contribution obtained at 1 % cut-off point for the environmental impact category freshwater eutrophication. The substances phosphate and phosphorus obtained the highest contribution in this category.

Fig. 8 presents the contribution graph obtained at 1 % cut-off point for the environmental impact category human toxicity. In this category, the substances manganese and barium were the largest contributors.

Fig. 9 presents the contribution graph obtained at 1 % cut-off point for the environmental impact category freshwater ecotoxicity. The prominent substance in this category is copper.

Fig. 10 presents the graph of contribution obtained at 1 % cut-off point for the environmental impact category fossil depletion. Natural gas, as feedstock, is the largest contributor to this category with 66 %.

Fig. 11 presents the graph of the results of environmental loads of the synthesis of obtaining the NC-CN-KL for each of the seven categories of environmental impact listed, according to characterization of the method ReCipe Midpoint (H) version 1.13/World Recipe (2010) H. In evidence, the areas in dark blue correspond to the input electrical energy.

When analyzing the processes involved in the life cycle of obtaining the nanocomposite, it becomes even more evident the electric energy and/or elementary processes of energy generation as the main contributors to the environmental loads of the synthesis. This result may be linked to the composition of the Brazilian energy matrix, since it was observed through the inventory analysis, influences of energy generated by hydroelectric, coal and oil, for example. The importance of a detailed look at this input is mainly because power generation is one of the sectors that stands out in terms of GHG emissions. However, and intending to perform an LCA of this work on a pilot scale, a discussion about the integration of CO<sub>2</sub> capture in thermoelectric power plants is essential.

Through LCA it was identified that in terms of global warming, two sub-critical and supercritical coal-fired power plants presented benefits when they had CO<sub>2</sub> capture plants [43]; however, it is necessary to observe other impact categories. Another alternative is

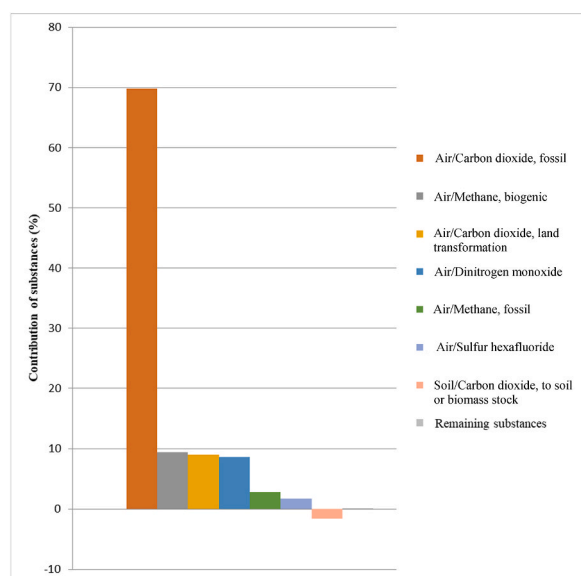
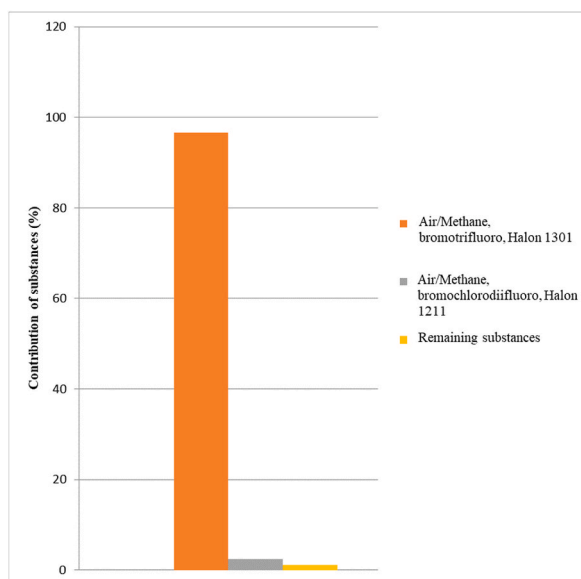
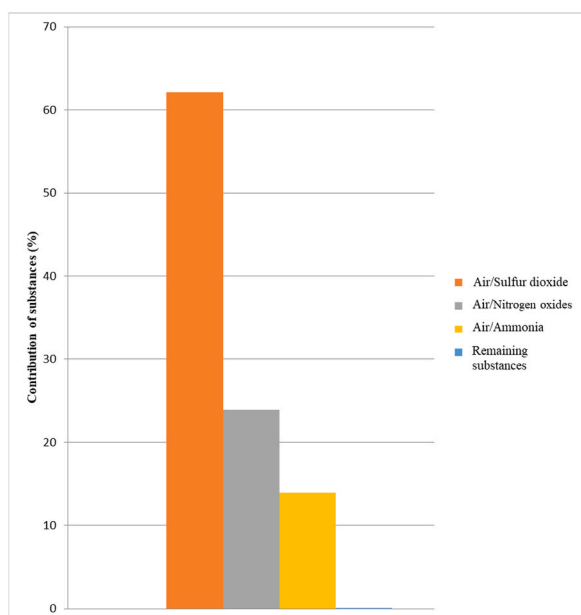


Fig. 4. Contribution of the main substances responsible for the environmental loads of the climate change category. Orange: air/fossil CO<sub>2</sub>; grey: air/biogenic methane; yellow: air/CO<sub>2</sub> from land transformation; blue: air/N<sub>2</sub>O; green: air/fossil methane; purple: air/fossil methane; pink: soil/CO<sub>2</sub> from soil or biomass stock.



**Fig. 5.** Contribution of the main substances responsible for the environmental loads of the ozone depletion category. Orange: air/methane, bromotrifluoro-, Halon 1301; grey: air/methane, bromochlorodifluoro-, Halon 1211; yellow: other substances.

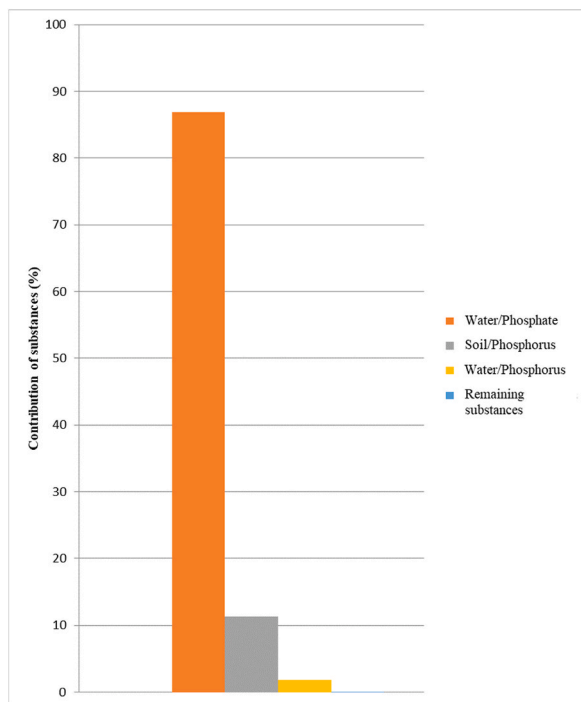


**Fig. 6.** Contribution of the main substances responsible for the environmental loads of the category terrestrial acidification. Orange: air/SO<sub>2</sub>; grey: air/N<sub>x</sub>O<sub>x</sub>; yellow: air/ammonia; blue: other substances.

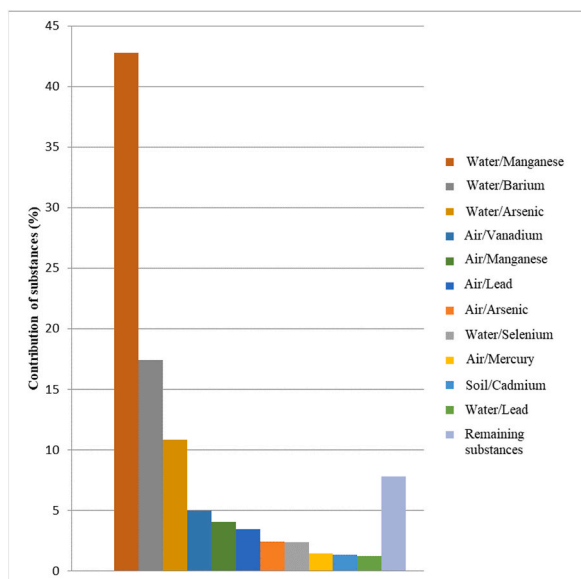
discussed by Briones-Hidrovo et al. (2022), who evaluated the use of biomass to generate electricity in a bioenergy system with carbon capture and storage (BECCS), where the results are satisfactory for categories related to climate change, but also highlight the importance of assessing impact categories related to land and water [44].

Table 2 shows the elementary processes with the highest environmental load for each of the environmental impact categories addressed.

In order to obtain a better understanding of the results regarding the categories with the greatest impact, Fig. 12 presents the graph of the results of the environmental loads of the synthesis of obtaining the NC-CN-KL for each of the seven environmental impact categories listed, according to the characterization of the ReCipe Midpoint (H) method version 1.13/World Recipe (2010) H in the normalized format configured in the software.



**Fig. 7.** Contribution of the main substances responsible for the environmental loads of the freshwater eutrophication category. Orange: water/phosphorus; grey: soil/phosphorus; yellow: water/phosphorus; blue: other substances.

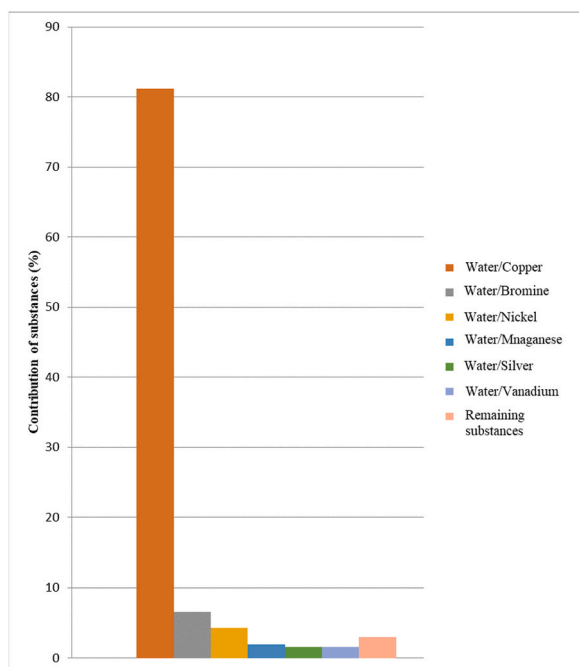


**Fig. 8.** Contribution of the main substances responsible for the environmental loads in the human toxicity category. Dark orange: water/manganese; dark grey: water/barium; yellow: water/arsenic; blue: air/vanadium; green: air/manganese; dark blue: air/lead; soft orange: air/arsenic; soft grey: water/selenium; soft yellow: air/mercury; soft blue: soil/cadmium; soft green: water/lead; magenta: other substances.

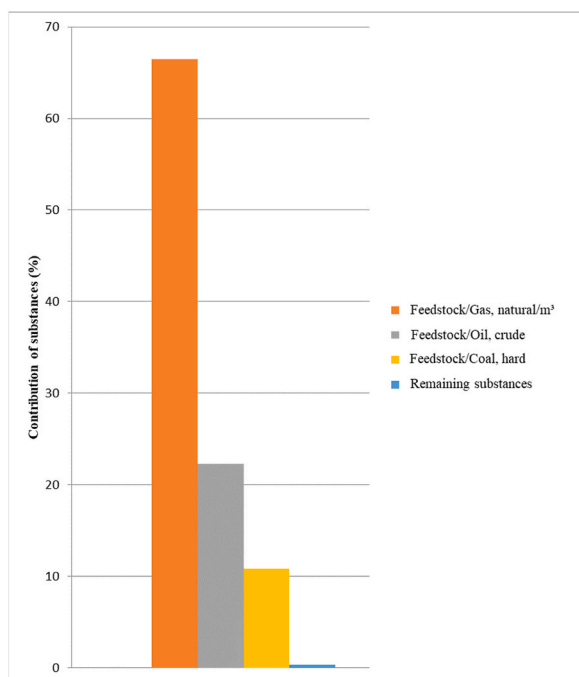
### 3.2. Interpretation

From the life cycle inventory and its interpretation, it was noted that the processes and substances with the greatest contribution to environmental loads are related to the inputs and outputs of electrical energy and its involved processes. All the impacts of energy generation are linked to the Brazilian energy matrix, the country where the nanocomposite was synthesized and the LCA was





**Fig. 9.** Contribution of the main substances responsible for the environmental loads in the freshwater ecotoxicity category. Orange: water/copper; grey: water/bromine; yellow: water/nickel; blue: water/manganese; green: water/silver; magenta: water/vanadium; pink: other substances.



**Fig. 10.** Contribution of the main substances responsible for the environmental loads of the fossil depletion category. Orange: raw material/natural gas; grey: raw material/crude oil; yellow: raw material: hard coal; blue: other substances.

performed.

In this case, because it is a laboratory scale experiment, the results of the input "electrical energy" highlight the importance of a new LCA at pilot scale, in order to seek environmentally adequate energy alternatives. Baker et al. (2022) through tools that integrate LCA with other inventory analysis tools, identified the relevance of cross-referencing laboratory scale data with pilot scale data, as this

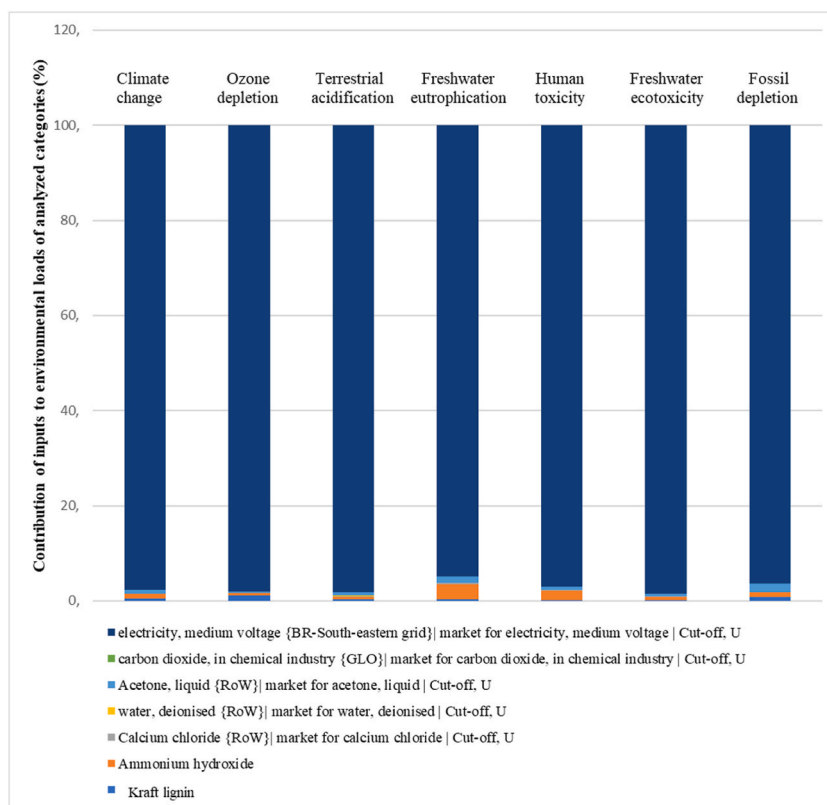


Fig. 11. Contribution of each input of the synthesis for each category of environmental impact studied.

Table 2

Relationship between the environmental impact categories with their respective processes of greatest contribution in terms of environmental load.

Environmental impact category	Process of greatest contribution for this category analyzed in SimaPro® software
CC	Electricity, high voltage {BR-South-eastern grid}  electricity production, natural gas, combined cycle Power plant  Cut-off, U
OD	Natural gas, high pressure {RoW} petroleum and gas production, on-shore   Cut-off, U
TA	Electricity, high voltage {BR-North-eastern gride}  electricity production, hard coal   Cut-off, U
FE	Spoil from Hard coal mining {GLO}   treatment of, in surface landfill   Cut-off, U
FEC	Scrap copper {Europe without Switzerland}   treatment of scrap copper, municipal incineration   Cut-off, U
FD	Natural gas, high pressure {RoW} natural gas production   Cut-off, U
HT	Spoil from Hard coal mining {GLO}   treatment of, in surface landfill   Cut-off, U

CC = climate change, OD = ozone depletion, TA = terrestrial acidification, FE = freshwater eutrophication, FEC = freshwater ecotoxicity, FD = fossil depletion, HT = human toxicity.

brings the study of environmental loads closer to the most real way possible [45]. However, this issue does not invalidate the goal of seeking a green product in this work, but emphasizes to consider the process as a whole, so that the correct study of speculations is possible. As an example, the storage and use of CO<sub>2</sub> from a combined natural gas cycle as an input for dimethyl ether was studied through LCA; the LCA showed that the storage can decrease the Global Warming Potential by up to 97 %, while the use of CO<sub>2</sub> reaches up to 68 %, but the latter is linked to obtaining a chemical product [46].

The recognition by the previous literature [47,48], that the energy sector contributes considerably to GHG emissions, justifies the search for integration of CO<sub>2</sub> capture processes [49] in power generation plants and tools such as LCA to validate the use of CO<sub>2</sub> as a benefit, especially when studying the Climate Change category [50] and when comparing storage *versus* CO<sub>2</sub> use [33].

All inventory data was reviewed and limitations regarding their quality were taken into consideration for the interpretation of the LCA. It is evident in this LCA limitations such as the quantity of inputs for inventory analysis, absence of scaling data and absence of data from the Kraft lignin obtaining process.

In order to optimize the data from this LCA, it is recommended to perform a new LCA with the scaled data (absent for the nanocarbonate synthesis) for the development of the CCU type process at TRL-6 or beyond. Another aspect to be considered is the inherent lack of data robustness – as, for instance, demonstrated by means a sensitivity analysis [51] - of the study derived from the absence of scale-up at pilot-plant. Once again, it can be reached by means the scaling to TRL-6 or beyond using batch production

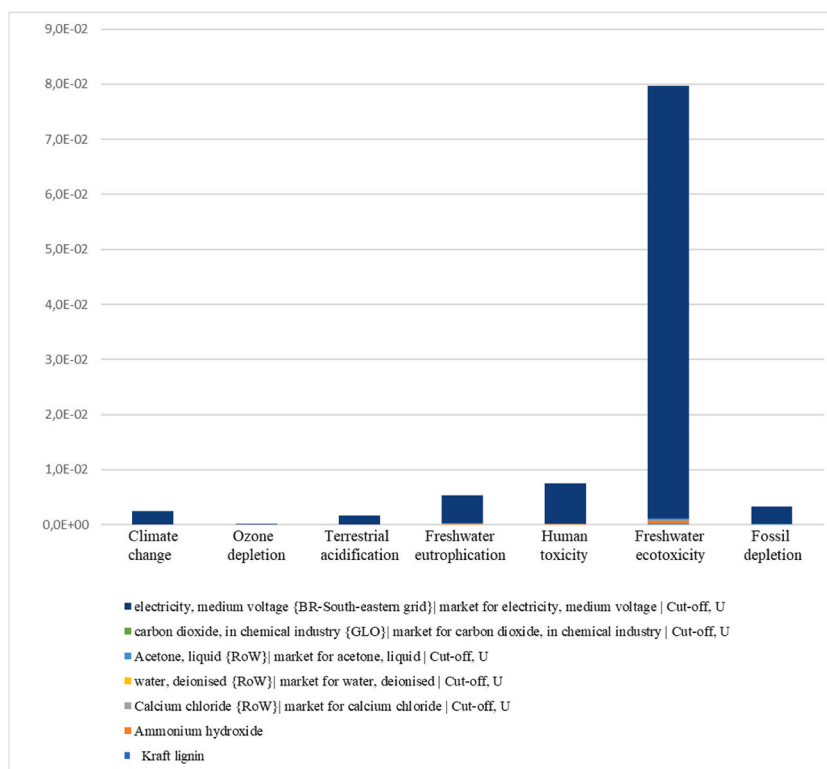


Fig. 12. Contribution of each input of the synthesis for each category of environmental impact studied in the normalized format.

strategy.

Limitations are present in all LCAs and should be highlighted especially in unpublished works, as is the case with the research described.

Nanotechnology has gained strength in obtaining agrochemical inputs in recent years, but it is worth highlighting the challenges (cost, acceptance, lack of data, etc.) that exist as a new topic [52,53].

#### 4. Conclusions

The main objective of the LCA was achieved and the main result refers to the high environmental loads of energy generation and other similar processes analyzed from the inventory analysis, such as burning of coal and other fossil sources, natural gas and water used in industrial processes for energy generation. This question suggests analyzing the energy mix in a complementary LCA aiming at mitigating the environmental impacts of energy generation.

The freshwater ecotoxicity category was identified through data normalization as the category with the greatest environmental impacts.

As for the limitations, we highlight the absence of data from the process of obtaining Kraft lignin during data collection and, therefore, the absence of data in Ecoinvent, the database used in this LCA. For an LCA, data quality is one of the most required items for the reliability of the results.

It is worth mentioning that this LCA was performed for a technology in TRL-4, that is, for a technology recently developed on a laboratory scale. It is expected, therefore, that many study limitations will be found at this TRL level. In this study, the estimates of the synthesis data stand out from the other limitations, as they directly interfere with the accuracy of the environmental loads. Another important limitation is the geographic area where the synthesis was developed, since it was developed in Brazil, making it difficult to search for information in the database that considers the country's characteristics. It is also worth mentioning the inventory of Kraft lignin, one of the main inputs of this LCA.

To formulate the inventory of Kraft lignin, it was necessary to use an inventory proposed in another work mentioned in this research. However, it was not possible to consider a reliable inventory of the specific process for the lignin incorporated in the synthesis in question.

It is therefore recommended that a new LCA be carried out for a higher level of industrial maturity considering the pilot scale for the green technology of obtaining NC-CN-KL, as well as repeating the synthesis, in order to collect the data with greater precision.

However, the material obtained presented good yield and its prospects for development and application are considered viable and favor the objective of the nanocomposite. The studies of their environmental loads carried out in this work tend to qualitatively and

quantitatively guide their development at higher scales.

### CRedit authorship contribution statement

**Ana Paula Rodrigues de Souza:** Writing – original draft, Methodology, Investigation. **Silvio Vaz:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization. **Érica Gonçalves Gravina:** Methodology, Investigation, Formal analysis. **Bruno Eduardo Lobo Baeta:** Supervision.

### Consent to participate

All authors consent to participate in the manuscript writing and publication.

### Consent for publication

All authors consent to publish this manuscript.

### Ethical approval

Not applicable.

### Data and code availability statement

LCA data have been deposited at Federal University of Ouro Preto public repository (<http://www.repositorio.ufop.br/jspui/handle/123456789/15096>) with accession number 123456789/15096.

### Declaration of competing interest

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

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