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*Original Research*

# Life Cycle Assessment of Different Sorbents at Early Stage

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## Abstract

Climate change caused by man-made carbon-dioxide emissions has become a global consensus. The studies of technologies for $CO_2$ capture have received continuous attention over the past few decades. In our lab, we have developed different biomass sorbents in previous studies, compared to other biomass sorbents, which have more desirable performance for $CO_2$ uptake. However, the application of these sorbents is still an emerging technology at the early stages of development. In order to optimize experimental design or develop large-scale production in the future, life cycle assessment has been employed to assess the environmental impacts of the sorbents prepared by different raw materials and conditions. According to the primary analysis on adsorption capacity and environmental benefits, five $CO_2$ uptake sorbents which synthesized by glucose or wheat flour as the biomass source of carbon were selected in this study. The result shows that, from the environmental perspective, the 700-PC is the best $CO_2$ uptake sorbent in five sorbents at ambient pressure, and the 800-N-PC is the best $CO_2$ uptake sorbent at high pressure. For the source of the environmental burdens, the electricity production is the largest environmental burden in all sorbents, but it has obvious laboratory features and is easy to decreased with the proceeding of industrialization. The environmental burden of glucose is bigger than that of wheat flour. The environmental burden of sodium bicarbonate is lower than that of potassium hydroxide. The environmental benefit of N-doping is different in different conditions, at the ambient pressure, the sorbent without N-doping is better than that with N-doping, but it is opposite at high press. Additive of cobalt has great environmental benefits for sorbents. Meanwhile, we also found that some factors that have great impacts on environment have not been studied before. Hence, in order to get the best environmental friendship sorbents, the further research can be conducted on these issues.

**Keywords**: LCA, $CO_2$ uptake sorbents, glucose, wheat flour, lab-scale

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## Introduction

In recent years, climate change has become a serious global environmental problem, which is greatly due to the increased release of $CO_2$ through human activities, such as burning coal and natural gas to produce electricity [1]. In order to capture and sequestrate $CO_2$, efficient $CO_2$ uptake sorbents for both pre- and post-combustion have received continuous attention over the past few decades [2-5].

Currently, chemical absorption is the most applicable technology for $CO_2$ capture in power plants, but has several drawbacks (i.e., corrosion rate and high energy regeneration requirements) [6]. Compared to the chemical absorption, porous solid sorbents are promising alternatives, because of their desirable characteristics, such as high adsorption capacity, insensitivity to moisture [7], property variability, low cost [8], good selectivity for $CO_2$, easy regeneration, and good reusability [9]. Among them, biomass materials are more desirable as carbon sources as they are readily available, renewable and cheap [5].

In our lab, wheat flour and glucose have been employed as a biomass carbon source to synthesize biomass sorbents for $CO_2$ uptake. According to the results of the previous studies, some of these sorbents show outstanding desirable characteristics and higher $CO_2$ capture capability, [5, 10, 11]. While due to the common raw materials, simple synthesis method, desirable characteristics and low costs, they are more likely to be industrialized than other sorbents. However, in the process of synthesizing biomass sorbents, some chemical reagents and energy have been used, sometimes in order to purse the best results, more chemical reagents and energy have been consumed. This is not a problem in the lab-scale, but if we want to study further industrial applications, the environment impacts of chemical reagents and energy should be pay much more attention.

Life cycle assessment (LCA) is a powerful holistic tool for the assessment of the environment impacts of new technologies to develop an understanding of their potential environmental burdens, which has already been successful applied in many fields [12, 13]. In this study, LCA is employed to assess the environment impacts of our biomass sorbents. In the previous studies, fourteen different biomass sorbents have been synthesized. According to the primary analysis on the preparation conditions and absorption effect of $CO_2$ uptake, five biomass sorbents are selected to develop LCA study. The result of this study can help researchers to optimize experimental design or to select appropriate adsorbents to develop large-scale production in the future.

## Materials and Methods

### Goal and Object

The goal of this study is (1) to evaluate the life cycle environmental impacts of different sorbents, (2) to discuss the environmental benefits of different raw materials and different preparation conditions and (3) to point to reduction opportunities for researchers to optimize experimental design or develop large-scale production in future.

The object of this study is five biomass sorbents synthesized by glucose or wheat flour as a biomass carbon source in our lab. For glucose sorbents, three different sorbents are selected according to primary analysis on adsorption capacity and environmental benefits. In order to relates well with our previous studies, the same names are adopted in this study, which are PC, NC-3 and Co-N-PC700, respectively. PC is synthesized by glucose and sodium bicarbonate in pyrolysis condition. The preparation of NC-3 is similarly to PC and urea is added as source of nitrogen doping to improve the intrinsic activity of the sorbent. Similarly to NC-3, cobalt acetate is added in Co-N-PC700. For wheat flour sorbents, similarly to glucose sorbents, two sorbents are selected, which are 700-PC and 800-N-PC. 700-PC is synthesized by wheat flour, sodium bicarbonate and potassium hydroxide in

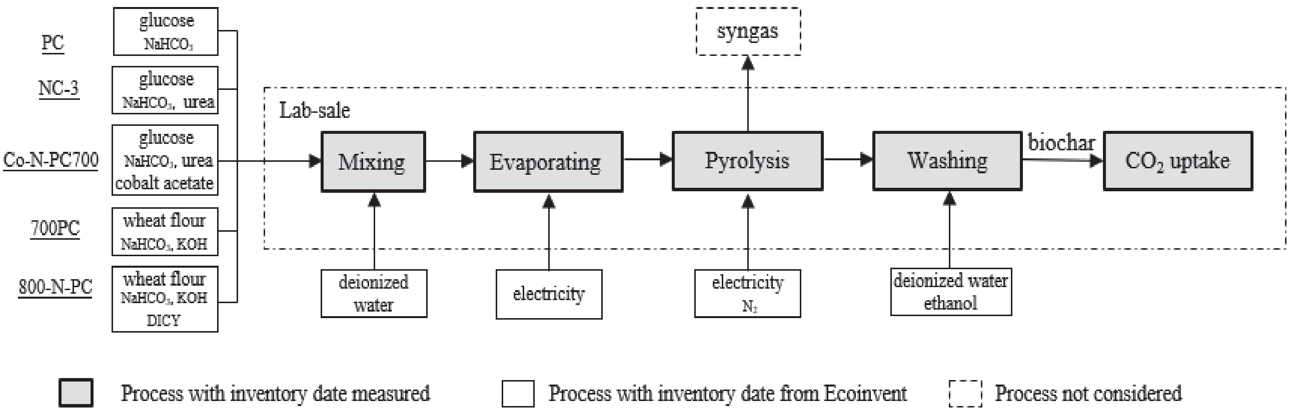

Fig. 1. Boundary describing the cradle-to-use life cycle assessment of five sorbents.

pyrolysis condition. Compared to 700-PC, 800-N-PC adds dicyandiamide as a source of nitrogen doping and rises the pyrolysis temperature. (see Section 1, 2 in SM)

## Boundary and Function

As show in Fig. 1, the boundary of this study is to assess cradle-to-use life cycle impacts of five biomass sorbents for application in $CO_2$ uptake, which include the activities from raw material extraction and processing to pyrolysis and $CO_2$ uptake. Since it is still in the lab-scale stage, no recycling or reuse of reaction media is considered, the reuse of syngas is also not considered and the transportation of materials (such as whale flour, glucose) is not included, too. Since the produced adsorbents have different adsorption capacities, the functional unit was determined to be 1 kg $CO_2$ at ambient pressure and 25ºC. Because of different adsorption mechanism at different pressure, the same adsorbents show different adsorption capacity. So for wheat flour sorbents, we also assess the scenario at 10 bar and 25ºC.

## Life Cycle Inventory

Life cycle inventory (LCI) analysis is the second phase in an LCA study [16]. The LCI is the data collection portion of all recognized inputs/outputs to or from the system boundary [17]. On this basis, our laboratory experiments provided the process data for assessing cradle-to-use life cycle impacts of the produced adsorbents. Inputs and outputs in each stage were converted to the scale of 1 kg $CO_2$ uptake. Table 1 present the life cycle inventory for generating final products. The Ecoinvent 3.5 Database was the source of inventory data for all input materials and energy in this study; however, there were no data for dicyandiamide (DICY) and cobalt acetate in this database or elsewhere. For DICY, we assumed that the emissions and resource use of producing DICY is the same as that of producing melamine. This analogy is considered reasonable since most DICY is processed in a similar manner to melamine. Lacking production data for cobalt acetate, its production was modeled in detail based on the stoichiometric formula (see Section 3 in SM).

## Impact Categories and Environmental Indicators

LCA models are developed with SimaPro v.9.0 software to quantify the impacts on a "cradle-to-use". The impact categories are those suggested by the ReCiPe 2016 Midpoint methodology, specifically: Global warming, Stratospheric ozone depletion, Ionizing radiation, Ozone formation (Human health), Fine particulate matter formation, Ozone formation (Terrestrial), Terrestrial acidification, Freshwater eutrophication, Marine eutrophication, Terrestrial ecotoxicity, Freshwater ecotoxicity, Marine ecotoxicity,

Table 1. The synthesis inventory date of 1kg $CO_2$ uptake for five sorbents.

| PC | | Unit | Amount used |
|---|---|---|---|
| Input | Glucose | kg | 54.69 |
| | Sodium bicarbonate | kg | 54.69 |
| | Deionized water | kg | 2959.41 |
| | $N_2$ | kg | 174.33 |
| | Ethanol | kg | 104.04 |
| | Electricity | MJ | 1903.45 |
| Output | $CO_2$ uptake | kg | 1.00 |
| Nc-3 | | | |
| Input | Glucose | kg | 50.76 |
| | Sodium bicarbonate | kg | 50.76 |
| | Urea | kg | 67.67 |
| | Deionized water | kg | 2746.45 |
| | $N_2$ | kg | 161.78 |
| | Ethanol | kg | 96.56 |
| | Electricity | MJ | 1578.17 |
| Output | $CO_2$ uptake | kg | 1.00 |
| Co-N-PC700 | | | |
| Input | Glucose | kg | 47.35 |
| | Sodium bicarbonate | kg | 47.35 |
| | Urea | kg | 47.35 |
| | Deionized water | kg | 2462.12 |
| | Cobalt acetate | kg | 0.95 |
| | $N_2$ | kg | 150.92 |
| | Ethanol | kg | 74.72 |
| | Electricity | MJ | 1375.61 |
| Output | $CO_2$ uptake | kg | 1.00 |
| 700-PC at ambient pressure | | | |
| Input | Wheat flour | kg | 17.22 |
| | Deionized water | kg | 1635.67 |
| | Sodium bicarbonate | kg | 17.22 |
| | Potassium hydroxide | kg | 10.33 |
| | $N_2$ | kg | 54.88 |
| | Ethanol | kg | 68.70 |
| | Electricity | MJ | 922.80 |
| Output | $CO_2$ uptake | kg | 1.00 |
| 700-PC at high pressure | | | |

Table 1. Continued.

| | | | |
|---|---|---|---|
| Input | Wheat flour | kg | 7.73 |
| | Deionized water | kg | 734.38 |
| | Sodium bicarbonate | kg | 7.73 |
| | Potassium hydroxide | kg | 4.64 |
| | $N_2$ | kg | 24.64 |
| | Ethanol | kg | 30.84 |
| | Electricity | MJ | 414.32 |
| Output | $CO_2$ uptake | kg | 1.00 |
| 800-N-PC at ambient pressure | | | |
| Input | Wheat flour | kg | 19.94 |
| | Deionized water | kg | 2192.98 |
| | Sodium bicarbonate | kg | 19.94 |
| | Potassium hydroxide | kg | 11.96 |
| | DICY | kg | 19.94 |
| | $N_2$ | kg | 68.53 |
| | Ethanol | kg | 79.55 |
| | Electricity | MJ | 1297.42 |
| Output | $CO_2$ uptake | kg | 1.00 |
| 800-N-PC at high pressure | | | |
| Input | Wheat flour | kg | 4.71 |
| | Deionized water | kg | 517.60 |
| | Sodium bicarbonate | kg | 4.71 |
| | Potassium hydroxide | kg | 2.82 |
| | DICY | kg | 4.71 |
| | $N_2$ | kg | 16.17 |
| | Ethanol | kg | 18.77 |
| | Electricity | MJ | 306.22 |
| Output | $CO_2$ uptake | kg | 1.00 |

Human carcinogenic toxicity, Human non-carcinogenic toxicity, Land use, Mineral resource scarcity, Fossil resource scarcity, Water consumption. For clarity of presentation, our results are normalized based on the report of Sleeswijk et al. [18], which has been input into the SimaPro v.9.0 software.

## Results and Discussion

### Assessment of Glucose Sorbents

Fig. 2 shows the normalized environmental impacts of PC, NC-3 and Co-NPC-700, which are 666.26, 679.89 and 586.41, respectively. Obviously, the Co-NPC-700 is a better environmental friendship sorbent than others by comprehensive consideration of the adsorption capacity and environmental benefits. As shown in Fig. 3, the environmental burden is from the use of raw materials and energy in the process of sorbents synthesis, such as glucose, sodium bicarbonate, cobalt acetate, deionized (DI) water, nitrogen, ethanol and electricity.

Electricity production is the main contributor of the environmental burden in PC, NC-3 and Co-NPC-700, which are 57.26%, 46.53% and 47.02%, respectively. According to the synthesis process of three sorbents, electricity is mainly used for water evaporation and pyrolysis, which has obvious laboratory features. Because, first, the water is used as solvent to mixed uniformly the reactants, such as glucose, sodium bicarbonate and cobalt acetate, so the evaporation in oven is a good way to separate out the dissolved matters from the water in the lab scale, but we do not pay more attention to the energy efficiency and the electricity consumption of the water evaporation. Second, the thermal efficiency of the research in the lab is relatively low and the by-products of the synthesis, such as biogas, is not considered to recycle. But if the process will become industrialized, a lot of measures of saving energy and reused will be applied, so the electricity consumption will be greatly decreased in industrial scale.

Urea production is the next contributor of the environmental burden, which is 14.61% and 11.85% in NC-3 and Co-NPC-700 synthesis, respectively. Since the urea is not used in PC synthesis, the environmental burden of urea production in PC synthesis is 0%. Introduction of nitrogenous groups onto carbon surface is known to increase $CO_2$ capture [1, 19]. The result of our experiments proved that the $CO_2$ uptake capacity of NC-3 is higher 40% than PC at ambient pressure and 25ºC [20]. This is partly because urea can lead to the formation of the pillared-layered pore channels and a higher micropore volume ($V_{mic}$) [20], which determined the low pressure $CO_2$ uptake due to a large caging effect [3, 21]. Besides, the presence of N atoms, especially pyrrolic-C, is closely linked to the $CO_2$ adsorption [21], as they can serve as Lewis bases to interacte with Lewis acidic $CO_2$ [10]. However, urea as the source of nitrogen can cause environmental burdens in its production process, especially for terrestrial ecotoxicity, mineral resource scarcity, water consumption and fossil resource scarcity. (see Table S1-3 in SM) As we know, the amount of N-doping is partly due to the nitrogen source material and partly due to the synthesis condition, for example, the amount of nitrogen in Co-NPC-700 is 4.89at%, which is higher than that synthesized at 800ºC and 900ºC [10]. Thus, there are two ways to decrease the environmental burden of the nitrogen source material for further experimental design or industrial application. The first is to try to find and use the more environment-friendly nitrogen source materials and the second is to try to find more mild synthesis conditions.

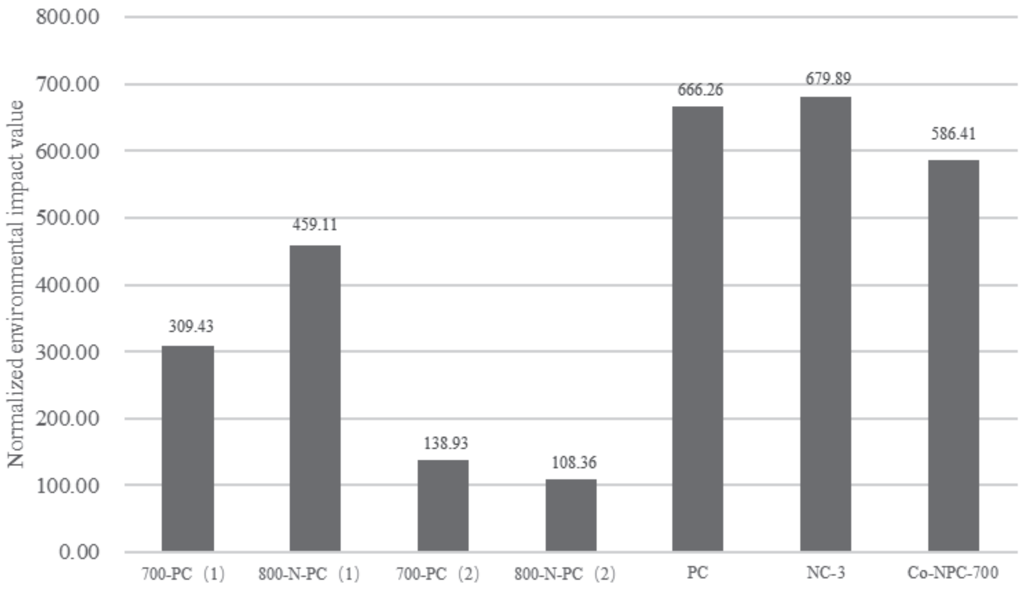

Fig. 2. Life cycle assessment results for five sorbents. (1) represents $CO_2$ uptake at ambient pressure. (2) represents $CO_2$ uptake at high pressure.

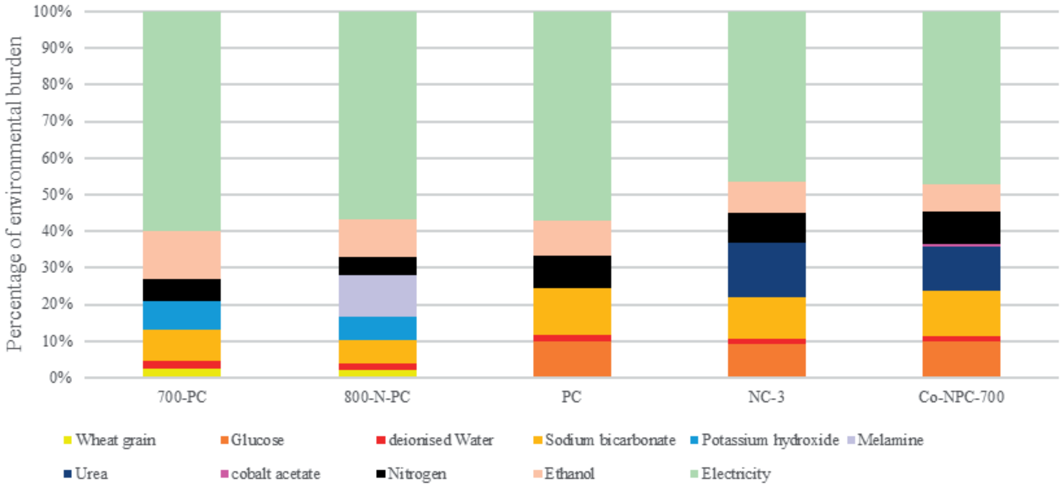

Fig. 3. Percentage of environmental burden of five sorbents for different raw materials and energy.

Sodium bicarbonate production is the third contributor of the environmental burden in PC, NC-3 and Co-NPC-700, which are 12.62%, 11.48% and 12.41%, respectively. According to our previous studies, we have confirmed that sodium bicarbonate as activator is very important for sorbents production process, because the sorbent from direct carbonization of glucose was nonporous [20]. Additionally, sodium bicarbonate as activator is considered to be green and scalable, because the decomposition product of sodium carbonate can be easily removed by water washing [20]. However, sodium carbonate is one of the chemicals which can cause environmental burden in its production process, especially for human non-carcinogenic toxicity, marine ecotoxicity, mineral resource scarcity and terrestrial ecotoxicity (see Table S1-3 in SM). In our previous studies, the amount of the sodium carbonate is the same with glucose in weight, but we do not pay more attention to the impacts of the $CO_2$ uptake capacity due to the change of amount of sodium carbonate. Hence, in order to decrease the environmental burdens from sodium bicarbonate production and achieve the best balance between environmental benefits and adsorption capacity, we suggest to develop the study of the impact of the change of amount of sodium carbonate to ascertain the best ratio in the further experimental design or industrial application model.

Glucose is the source of carbon for every sorbent, the environmental burden of glucose is 10.08%, 9.17% and 9.92% in PC, NC-3 and Co-NPC-700, respectively. Glucose is made of corn starch, the main environmental burdens are land use, marine eutrophication and stratospheric ozone depletion(see Table S1-3 in SM). Since the glucose is the basic material for the $CO_2$ uptake sorbent, the best way to decrease its environmental burdens is to increase the input-output ratio of biomass sorbents, the current input-output ratio in our previous experiments is about 12%, which is relatively low.

The environmental burdens of ethanol, deionized water and nitrogen are relatively small, especially for deionized water, the contributor is less than 1.5%, and the contributor of ethanol and nitrogen are both less than 10%. According to the process of sorbents synthesis, the ethanol, deionized water and nitrogen are used as assistant materials, the ethanol is used to wash the sorbents, the deionized (DI) water is used to dissolve the raw material and wash the sorbents, nitrogen is used as shielding gas to avoid the raw material from the oxygen during pyrolysis. At the lab-scale, all of them are not consider to reuse, but similarly to the electricity, the environmental burdens of them are easy to decreased with the reuse of them at large-scale production.

Cobalt acetate is only used to prepare Co-NPC-700 to enhance the $CO_2$ uptake capacity. The LCA result shows that the environmental burden of cobalt acetate production is are very little, which is only 0.71%. But introduction of cobalt groups onto carbon surface is greatly increase $CO_2$ capture [10], so cobalt acetate can be considered an environment friendly-ship additive for biomass sorbents. Similarly to the sodium bicarbonate, the best amount of cobalt acetate is not studied in our previously studies, so the best balance between environmental benefits and adsorption capacity is not uncertain for added cobalt acetate. Hence, we suggest developing the study on the effect of the amount of cobalt in the further experimental design or industrial application model.

### Assessment of Wheat Flour Sorbents

The environmental assessments of the 700-PC and 800-N-PC at ambient and high pressure are assessed in this section. At ambient pressure, the normalized environmental impacts of the 700-PC and 800-N-PC are 309.43 and 459.11, respectively. Obviously, the 700-PC is a better environmental friendship sorbent than the 800-N-PC. This is because, compared to the 700-PC, the 800-N-PC which synthesized with N-doping and at higher calcination temperature has created more BET surface and total pore volume, but the micro-porosity, which is generally believed that $CO_2$ uptake in ambient condition is determined by micro-porosity because of a large caging effect [3, 21], is decreased [5]. Thus, the 700-PC is a relatively desirable sorbent for post-combustion $CO_2$ capture.

At high pressure, the normalized environmental impacts of the 700-PC and 800-N-PC are 138.93 and 108.36, respectively. This is because $CO_2$ adsorption at high pressures also proceeds in the wider micropores and narrow mesopores [3, 22]. Although the environmental burden is increased for the 800-N-PC due to the addition of dicyandiamide and synthesis at higher temperature, the $CO_2$ capture capability was also greatly increased correspondingly due to the higher BET surface and total pore volume, thus the comprehensive benefits of the 800-N-PC is better than the 700-PC at high pressure.

Similarly to the glucose sorbents, the normalized environmental impacts of electricity is the highest in wheat flour sorbents, but it is also easy to decreased with industrialization. The ethanol, deionized (DI) water and nitrogen are used as assistant materials, their normalized environmental impacts are also little and easy to decreased with industrialization. It is noteworthy that besides sodium bicarbonate, potassium hydroxide is also used as activator to create a porous structure. According to the results of LCA, the normalized environmental impacts of sodium bicarbonate and potassium hydroxide production were close to each other (see Table S4-7 in SM), but the amount of potassium hydroxide addition is only 60% of sodium bicarbonate at the process of wheat flour sorbents synthesis, so it is clearly that the normalized environmental impacts of potassium hydroxide is larger than that of potassium hydroxide. Thus, for the environmental benefits, sodium bicarbonate is a more desirable activator for sorbents than potassium hydroxide.

### The Comparison of Glucose Sorbents and Wheat Flour Sorbents

According the results of LCA, at ambient pressure, the Co-NPC-700 is the best sorbents in glucose sorbents, and the 700-PC is the best sorbents in wheat flour sorbents. The normalized environmental impacts of two sorbents are 586.41 and 309.43, respectively, which is obvious that the 700-PC is a better environmental friendship sorbent than the Co-NPC-700. This is partly because the Co-NPC-700 yield from the glucose is only 12%, which is lower than that of 700-PC (30%) from wheat flour. Besides, the $CO_2$ capture capability of 700-PC is also better than that of the Co-NPC-700, which are 4.4 $mmmolg^{-1}$ [5] and 4 $mmmolg^{-1}$ [10], respectively. Therefore, according to the boundary of this study and comprehensive consideration of environmental and adsorption benefits, the 700-PC is a more desirable sorbent than others at ambient pressure.

## Conclusions

In this study, the environmental impacts of five $CO_2$ uptake sorbents synthesized by glucose or wheat flour have been assessed by LCA, the result shows that the 700-PC is the best $CO_2$ uptake sorbent in five sorbents at ambient pressure from the environmental perspective, and the 800-N-PC is the best $CO_2$ uptake sorbent at high pressure.

The environmental burden of sorbents is from the production of raw materials and energy. Electricity production is the largest environmental burden in all sorbents, but it has obvious laboratory features and is easy to decrease with the proceeding of industrialization. Similarly to electricity, the environmental burdens of ethanol, deionized (DI) water and nitrogen which used as assistant materials are also easily to decreased with the proceeding of industrialization. Glucose and wheat flour are the source of carbon for sorbents, the environmental burden of glucose is bigger than that of wheat flour, mainly because the sorbent yield from glucose is lower than that from wheat flour. Sodium bicarbonate or potassium hydroxide is important for all sorbents as activator to create a porous structure, and the environmental burden of sodium bicarbonate is lower than that of potassium hydroxide. The environmental benefit of N-doping is different in different conditions, at the ambient pressure, the sorbent without N-doping is better than that with N-doping according to comprehensive consideration of environmental benefits, but it is opposite at high press, because of different adsorption mechanism at different press. The additive of cobalt has great environmental benefits for sorbents, because the benefit of $CO_2$ capture from cobalt is larger than the environmental burdens from cobalt.

In summary, this study assess the sorbents which have prepared in our previous studies, and meanwhile, we also found that some factors, such as the effects on different mixing ways for raw material, different cobalt content, different activators and their different content, that have great impacts on environment have not been studied before. Thus, in order to get the best environmental friendship sorbents, the further research can be conducted on the above issues.

## Acknowledgments

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## Conflict of Interest

The authors declare no conflict of interest.

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# Supplementary Material

## Section 1. Synthesis Process of Glucose Sorbents

### Preparation of PC

Glucose (0.9 g) and sodium bicarbonate (0.9 g) were dissolved in pure water and then the water was evaporated at 105ºC; Heat treatment of the mixture was done at 700ºC for 2 h with a heating rate of 5ºC min$^{-1}$ under $N_2$ flow. Finally, the carbonized materials were grinded and washed with water and ethanol for several times, then dried in an oven [1].

### Preparation of NC-3

Glucose (0.9 g), sodium bicarbonate (0.9 g), and urea (1.2 g) were dissolved in pure water and then the water was evaporated at 105ºC; Heat treatment of the mixture was done at 700ºC for 2 h with a heating rate of 5ºC min$^{-1}$ under $N_2$ flow. Finally, the carbonized materials were grinded and washed with water and ethanol for several times, then dried in an oven [1].

### Preparation of Co-NPC-700

5 g of glucose, 5 g of sodium bicarbonate, and 5 g of urea were fully dissolved in 150 mL of deionized (DI) water, followed by mixing with 10 mL of cobalt acetate solution (0.1 g of cobalt acetate dissolved in DI water) under vigorous stirring. After evaporation at 100ºC, the mixture was pyrolyzed under a $N_2$ flow at a rate of 5ºC min$^{-1}$ at 700ºC and kept for 2 h. Finally, Co-NPC 700 was obtained after alternate washing with water and ethanol [2].

## Section 2. Synthesis Process of Wheat Flour Sorbents

### Preparation of 700-PC

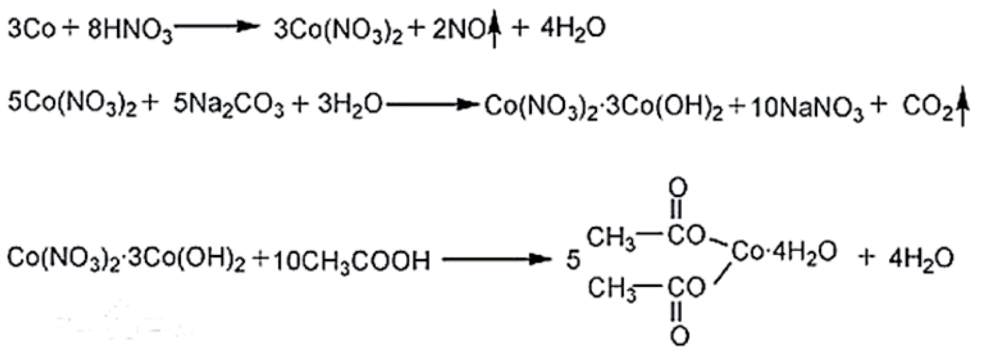
2 g of wheat flour was added into 40 mL deionized water under magnetic stirring. Then, 2 g sodium bicarbonate and 1.2 g potassium hydroxide were dissolved in 50 mL water. The mixture was then added to the above flour solution. After magnetic stirring and drying at 90ºC, the mixture was put in a tube furnace and pyrolyzed under a $N_2$ flow at 700ºC for 2 h. The heating ramp rate was 5ºC/min. The carbonized materials were pestled to powders, and washed with water and ethanol. Finally, the powders were dried in an oven [2].

### Preparation of 800-N-PC

2 g of wheat flour was added into 40 mL deionized water under magnetic stirring. Then, 2 g sodium bicarbonate and 1.2 g potassium hydroxide were dissolved in 50 mL water, which was then mixed with a 30 mL water solution containing 2.0 g dicyandiamide (DICY). The mixture was then added to the above flour solution. After magnetic stirring and drying at 90ºC, the mixture was put in a tube furnace and pyrolyzed under a N2 flow at 800ºC for 2 h. The heating ramp rate was 5ºC/min. The carbonized materials were pestled to powders, and washed with water and ethanol. Finally, the powders were dried in an oven [2].

## Section 3. Life Cycle Inventory of Cobalt Acetate

Production of cobalt acetate is not available in the Ecoinvent database, and there is no obvious analog. Therefore, cobalt acetate production had to be modeled in detail. The modeling followed the Cobalt Metal synthesis route. Briefly, metal cobalt was mixed with nitric acid to prepare cobalt nitrate solution, then soda was added to produce the cobalt carbonate precipitation. The precipitation was washed with water, and then acetic acid was added to prepared cobalt acetic. After acidification, the reaction solution was filtrated, adjusted to pH = 2 and concentrated to the relative density about 1.17-1.20. Then some acetic acid was added to the concentration. The solution was crystallized for 7-10 days under 20ºC, centrifuged and dried at 30-40ºC. Raw material consumption quota is cobalt metal (>99%) 265kg/t, acetic acid 1000 kg/t, nitric acid 1100 kg/t. The chemical reaction equation is as follows.



$$3Co + 8HNO_3 \longrightarrow 3Co(NO_3)_2 + 2NO\uparrow + 4H_2O$$

$$5Co(NO_3)_2 + 5Na_2CO_3 + 3H_2O \longrightarrow Co(NO_3)_2 \cdot 3Co(OH)_2 + 10NaNO_3 + CO_2\uparrow$$

$$Co(NO_3)_2 \cdot 3Co(OH)_2 + 10CH_3COOH \longrightarrow 5(CH_3COO)_2Co \cdot 4H_2O + 4H_2O$$

Table S 1. The normalized value of different environmental impacts and percentage of different raw materials and energy for PC.

| Environmental impact type | Normalized value | Percentage of different raw materials and energy | | | | | |
|---|---|---|---|---|---|---|---|
| | | Water | Sodium bicarbonate | Nitrogen | Ethanol | Glucose | Electricity |
| Global warming | 0.12 | 0.60% | 9.51% | 10.56% | 15.49% | 9.04% | 54.80% |
| Stratospheric ozone depletion | 0.02 | 0.37% | 2.72% | 3.99% | 1.38% | 69.90% | 21.64% |
| Ionizing radiation | 0.15 | 0.54% | 4.76% | 9.92% | 1.69% | 8.90% | 74.19% |
| Ozone formation, Human health | 0.09 | 0.62% | 10.88% | 10.05% | 16.23% | 11.65% | 50.58% |
| Fine particulate matter formation | 0.06 | 0.66% | 11.73% | 11.54% | 8.01% | 9.34% | 58.73% |
| Ozone formation, Terrestrial ecosystems | 0.10 | 0.61% | 10.67% | 9.80% | 18.14% | 11.47% | 49.32% |
| Terrestrial acidification | 0.08 | 0.53% | 19.51% | 8.34% | 10.84% | 14.59% | 46.19% |
| Freshwater eutrophication | 0.63 | 0.56% | 10.63% | 9.61% | 13.52% | 8.60% | 57.09% |
| Marine eutrophication | 0.04 | 0.09% | 1.30% | 1.31% | 0.58% | 88.82% | 7.91% |
| Terrestrial ecotoxicity | 1.55 | 0.59% | 28.12% | 4.17% | 12.89% | 22.33% | 31.89% |
| Freshwater ecotoxicity | 22.69 | 0.43% | 13.52% | 5.19% | 6.91% | 9.87% | 64.07% |
| Marine ecotoxicity | 21.86 | 0.57% | 22.43% | 6.26% | 11.35% | 15.07% | 44.31% |
| Human carcinogenic toxicity | 599.29 | 1.73% | 11.85% | 9.54% | 9.15% | 9.69% | 58.03% |
| Human non-carcinogenic toxicity | 19.22 | 0.56% | 23.24% | 6.17% | 11.74% | 15.66% | 42.63% |
| Land use | 0.02 | 0.07% | 4.95% | 0.80% | 1.37% | 87.55% | 5.26% |
| Mineral resource scarcity | 0.00 | 1.36% | 32.09% | 2.67% | 17.24% | 24.53% | 22.11% |
| Fossil resource scarcity | 0.27 | 0.45% | 5.99% | 7.77% | 38.29% | 7.28% | 40.22% |
| Water consumption | 0.05 | 21.95% | 18.47% | 13.67% | 7.76% | 17.31% | 20.84% |
| Total | 666.26 | 1.61% | 12.62% | 9.18% | 9.25% | 10.08% | 57.26% |

Table S 2. The normalized value of different environmental impacts and percentage of different raw materials and energy for NC-3.

| Environmental impact type | Normalized value | Percentage of different raw materials and energy | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | Water | Sodium bicarbonate | Nitrogen | Ethanol | Glucose | Urea | Electricity |
| Global warming | 0.14 | 0.47% | 7.50% | 8.33% | 12.22% | 7.13% | 25.72% | 38.62% |
| Stratospheric ozone depletion | 0.02 | 0.35% | 2.52% | 3.71% | 1.29% | 64.90% | 9.28% | 17.95% |
| Ionizing radiation | 0.15 | 0.51% | 4.43% | 9.24% | 1.58% | 8.30% | 14.16% | 61.78% |
| Ozone formation, Human health | 0.09 | 0.54% | 9.52% | 8.79% | 14.19% | 10.19% | 17.26% | 39.52% |

Table S 2. Continued.

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Fine particulate matter formation | 0.07 | 0.55% | 9.79% | 9.64% | 6.68% | 7.80% | 21.74% | 43.80% |
| Ozone formation, Terrestrial ecosystems | 0.11 | 0.53% | 9.32% | 8.56% | 15.85% | 10.02% | 17.22% | 38.50% |
| Terrestrial acidification | 0.10 | 0.40% | 14.80% | 6.33% | 8.22% | 11.07% | 27.88% | 31.30% |
| Freshwater eutrophication | 0.61 | 0.53% | 10.16% | 9.18% | 12.92% | 8.22% | 10.24% | 48.75% |
| Marine eutrophication | 0.04 | 0.09% | 1.25% | 1.26% | 0.56% | 85.63% | 4.40% | 6.81% |
| Terrestrial ecotoxicity | 2.58 | 0.33% | 15.74% | 2.34% | 7.22% | 12.50% | 45.93% | 15.95% |
| Freshwater ecotoxicity | 23.25 | 0.39% | 12.25% | 4.70% | 6.26% | 8.94% | 15.61% | 51.84% |
| Marine ecotoxicity | 25.15 | 0.46% | 18.10% | 5.05% | 9.16% | 12.16% | 23.13% | 31.94% |
| Human carcinogenic toxicity | 605.00 | 1.59% | 10.90% | 8.77% | 8.42% | 8.91% | 13.76% | 47.66% |
| Human non-carcinogenic toxicity | 22.14 | 0.45% | 18.72% | 4.97% | 9.46% | 12.61% | 23.09% | 30.69% |
| Land use | 0.01 | 0.07% | 4.85% | 0.79% | 1.35% | 85.89% | 2.44% | 4.61% |
| Mineral resource scarcity | 0.00 | 0.93% | 21.85% | 1.82% | 11.74% | 16.70% | 33.51% | 13.45% |
| Fossil resource scarcity | 0.33 | 0.34% | 4.52% | 5.86% | 28.86% | 5.49% | 27.85% | 27.08% |
| Water consumption | 0.09 | 11.27% | 9.49% | 7.02% | 3.99% | 8.89% | 49.78% | 9.56% |
| Total | 679.79 | 1.46% | 11.48% | 8.35% | 8.41% | 9.17% | 14.61% | 46.53% |

Table S 3. The normalized value of different environmental impacts and percentage of different raw materials and energy for Co-NPC-700.

| Environmental impact type | Normalized value | Percentage of different raw materials and energy | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | Water | Sodium bicarbonate | Nitrogen | Ethanol | Glucose | Urea | Cobalt acetate | Electricity |
| Global warming | 0.12 | 0.50% | 8.37% | 9.30% | 11.31% | 7.96% | 21.52% | 0.77% | 40.27% |
| Stratospheric ozone depletion | 0.02 | 0.30% | 2.27% | 3.34% | 0.96% | 58.43% | 6.27% | 13.34% | 15.10% |
| Ionizing radiation | 0.13 | 0.53% | 4.78% | 9.96% | 1.41% | 8.94% | 11.45% | 0.70% | 62.23% |
| Ozone formation, Human health | 0.08 | 0.56% | 10.28% | 9.49% | 12.71% | 11.00% | 13.98% | 2.11% | 39.88% |
| Fine particulate matter formation | 0.06 | 0.58% | 10.72% | 10.55% | 6.07% | 8.53% | 17.84% | 0.91% | 44.80% |
| Ozone formation, Terrestrial ecosystems | 0.10 | 0.55% | 10.09% | 9.27% | 14.23% | 10.84% | 13.98% | 2.09% | 38.94% |
| Terrestrial acidification | 0.08 | 0.43% | 16.33% | 6.98% | 7.53% | 12.22% | 23.08% | 1.15% | 32.28% |
| Freshwater eutrophication | 0.53 | 0.55% | 10.97% | 9.92% | 11.58% | 8.88% | 8.30% | 0.59% | 49.21% |
| Marine eutrophication | 0.04 | 0.08% | 1.27% | 1.28% | 0.47% | 86.99% | 3.36% | 0.08% | 6.46% |
| Terrestrial ecotoxicity | 2.09 | 0.36% | 18.08% | 2.68% | 6.87% | 14.35% | 39.55% | 1.00% | 17.11% |
| Freshwater ecotoxicity | 20.01 | 0.41% | 13.27% | 5.10% | 5.63% | 9.69% | 12.69% | 0.72% | 52.50% |
| Marine ecotoxicity | 21.46 | 0.48% | 19.79% | 5.53% | 8.31% | 13.29% | 18.97% | 1.01% | 32.63% |
| Human carcinogenic toxicity | 522.43 | 1.65% | 11.77% | 9.48% | 7.54% | 9.62% | 11.15% | 0.68% | 48.11% |
| Human non-carcinogenic toxicity | 18.90 | 0.48% | 20.46% | 5.43% | 8.57% | 13.78% | 18.92% | 1.03% | 31.33% |

Table S 3. Continued.

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Land use | 0.01 | 0.07% | 4.90% | 0.79% | 1.13% | 86.64% | 1.85% | 0.29% | 4.35% |
| Mineral resource scarcity | 0.00 | 0.43% | 10.46% | 0.87% | 4.66% | 8.00% | 12.03% | 57.53% | 6.02% |
| Fossil resource scarcity | 0.27 | 0.38% | 5.19% | 6.73% | 27.52% | 6.31% | 24.01% | 0.76% | 29.09% |
| Water consumption | 0.08 | 12.56% | 11.00% | 8.14% | 3.83% | 10.31% | 43.28% | 0.53% | 10.36% |
| Total | 586.41 | 1.52% | 12.41% | 9.03% | 7.55% | 9.92% | 11.85% | 0.71% | 47.02% |

Table S 4. The normalized value of different environmental impacts and percentage of different raw materials and energy for 700-PC at ambient pressure.

| Environmental impact type | Normalized value | Percentage of different raw materials and energy | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | Wheat | Water | Sodium bicarbonate | Potassium hydroxide | Nitrogen | Ethanol | Electricity |
| Global warming | 0.06 | 2.82% | 0.68% | 6.20% | 7.16% | 6.89% | 21.19% | 55.06% |
| Stratospheric ozone depletion | 0.01 | 66.16% | 0.46% | 1.91% | 3.14% | 2.81% | 2.05% | 23.47% |
| Ionizing radiation | 0.07 | 1.55% | 0.65% | 3.25% | 7.23% | 6.78% | 2.43% | 78.11% |
| Ozone formation, Human health | 0.04 | 5.25% | 0.71% | 7.12% | 7.16% | 6.57% | 22.26% | 50.94% |
| Fine particulate matter formation | 0.03 | 3.98% | 0.78% | 7.94% | 6.97% | 7.81% | 11.36% | 61.17% |
| Ozone formation, Terrestrial ecosystems | 0.05 | 5.13% | 0.69% | 6.92% | 6.96% | 6.36% | 24.68% | 49.27% |
| Terrestrial acidification | 0.04 | 8.70% | 0.64% | 13.45% | 6.75% | 5.75% | 15.67% | 49.04% |
| Freshwater eutrophication | 0.31 | 5.30% | 0.62% | 6.81% | 6.64% | 6.15% | 18.16% | 56.31% |
| Marine eutrophication | 0.01 | 76.06% | 0.21% | 1.77% | 1.91% | 1.79% | 1.65% | 16.61% |
| Terrestrial ecotoxicity | 0.65 | 4.94% | 0.78% | 21.27% | 12.25% | 3.16% | 20.45% | 37.15% |
| Freshwater ecotoxicity | 10.38 | 2.19% | 0.52% | 9.30% | 6.56% | 3.57% | 9.98% | 67.88% |
| Marine ecotoxicity | 9.66 | 3.00% | 0.72% | 15.98% | 10.25% | 4.46% | 16.97% | 48.62% |
| Human carcinogenic toxicity | 279.48 | 2.58% | 2.05% | 8.00% | 7.64% | 6.44% | 12.96% | 60.33% |
| Human non-carcinogenic toxicity | 8.46 | 3.12% | 0.71% | 16.63% | 10.54% | 4.41% | 17.62% | 46.98% |
| Land use | 0.01 | 90.20% | 0.06% | 2.51% | 1.25% | 0.41% | 1.46% | 4.11% |
| Mineral resource scarcity | 0.00 | 11.26% | 1.69% | 22.67% | 12.90% | 1.89% | 25.54% | 24.05% |
| Fossil resource scarcity | 0.14 | 1.76% | 0.47% | 3.56% | 5.15% | 4.61% | 47.68% | 36.77% |
| Water consumption | 0.04 | 41.84% | 18.02% | 8.64% | 2.48% | 6.39% | 7.61% | 15.01% |
| Total | 309.43 | 2.61% | 1.91% | 8.55% | 7.77% | 6.22% | 13.15% | 59.78% |

Table S 5. The normalized value of different environmental impacts and percentage of different raw materials and energy for 800-N-PC at ambient pressure.

| Environmental impact type | Normalized value | Percentage of different raw materials and energy | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | Wheat | Water | Sodium bicarbonate | Potassium hydroxide | DICY | Nitrogen | Ethanol | Electricity |
| Global warming | 0.09 | 2.06% | 0.58% | 4.52% | 5.22% | 18.01% | 5.42% | 15.45% | 48.74% |
| Stratospheric ozone depletion | 0.01 | 58.19% | 0.47% | 1.68% | 2.76% | 7.37% | 2.67% | 1.80% | 25.06% |
| Ionizing radiation | 0.11 | 1.19% | 0.58% | 2.48% | 5.52% | 10.34% | 5.59% | 1.85% | 72.45% |
| Ozone formation, Human health | 0.06 | 4.11% | 0.64% | 5.57% | 5.61% | 12.65% | 5.55% | 17.43% | 48.44% |
| Fine particulate matter formation | 0.05 | 2.94% | 0.66% | 5.85% | 5.14% | 16.03% | 6.21% | 8.38% | 54.79% |
| Ozone formation, Terrestrial ecosystems | 0.07 | 4.03% | 0.63% | 5.44% | 5.48% | 12.56% | 5.39% | 19.41% | 47.06% |
| Terrestrial acidification | 0.06 | 6.06% | 0.52% | 9.37% | 4.70% | 22.66% | 4.32% | 10.91% | 41.47% |
| Freshwater eutrophication | 0.44 | 4.34% | 0.59% | 5.57% | 5.43% | 7.88% | 5.43% | 14.85% | 55.91% |
| Marine eutrophication | 0.01 | 68.57% | 0.22% | 1.60% | 1.72% | 6.47% | 1.74% | 1.49% | 18.18% |
| Terrestrial ecotoxicity | 1.35 | 2.75% | 0.51% | 11.84% | 6.82% | 39.71% | 1.89% | 11.38% | 25.10% |
| Freshwater ecotoxicity | 15.66 | 1.68% | 0.46% | 7.14% | 5.03% | 11.77% | 2.96% | 7.66% | 63.29% |
| Marine ecotoxicity | 15.32 | 2.19% | 0.60% | 11.67% | 7.48% | 19.05% | 3.51% | 12.39% | 43.10% |
| Human carcinogenic toxicity | 412.19 | 2.02% | 1.86% | 6.28% | 6.00% | 10.69% | 5.46% | 10.18% | 57.51% |
| Human non-carcinogenic toxicity | 13.40 | 2.28% | 0.60% | 12.15% | 7.70% | 19.24% | 3.48% | 12.88% | 41.68% |
| Land use | 0.01 | 87.91% | 0.07% | 2.45% | 1.22% | 1.64% | 0.43% | 1.42% | 4.87% |
| Mineral resource scarcity | 0.00 | 7.58% | 1.32% | 15.27% | 8.69% | 28.91% | 1.37% | 17.20% | 19.66% |
| Fossil resource scarcity | 0.22 | 1.32% | 0.41% | 2.67% | 3.87% | 18.60% | 3.74% | 35.84% | 33.55% |
| Water consumption | 0.06 | 27.05% | 13.49% | 5.59% | 1.60% | 31.10% | 4.46% | 4.92% | 11.78% |
| Total | 459.11 | 2.04% | 1.73% | 6.67% | 6.06% | 11.35% | 5.24% | 10.26% | 56.64% |

Table S 6. The normalized value of different environmental impacts and percentage of different raw materials and energy for 700-PC at high pressure.

| Environmental impact type | Normalized value | Percentage of different raw materials and energy | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | Wheat | Water | Sodium bicarbonate | Potassium hydroxide | Nitrogen | Ethanol | Electricity |
| Global warming | 0.03 | 2.82% | 0.68% | 6.20% | 7.16% | 6.89% | 21.19% | 55.06% |
| Stratospheric ozone depletion | 0.00 | 66.16% | 0.46% | 1.91% | 3.14% | 2.81% | 2.05% | 23.47% |
| Ionizing radiation | 0.03 | 1.55% | 0.65% | 3.25% | 7.23% | 6.78% | 2.43% | 78.11% |
| Ozone formation, Human health | 0.02 | 5.25% | 0.71% | 7.12% | 7.16% | 6.57% | 22.26% | 50.94% |
| Fine particulate matter formation | 0.01 | 3.98% | 0.78% | 7.94% | 6.97% | 7.81% | 11.36% | 61.17% |
| Ozone formation, Terrestrial ecosystems | 0.02 | 5.13% | 0.69% | 6.92% | 6.96% | 6.36% | 24.68% | 49.27% |
| Terrestrial acidification | 0.02 | 8.70% | 0.64% | 13.45% | 6.75% | 5.75% | 15.67% | 49.04% |

Table S 6. Continued.

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| Freshwater eutrophication | 0.14 | 5.30% | 0.62% | 6.81% | 6.64% | 6.15% | 18.16% | 56.31% |
| Marine eutrophication | 0.00 | 76.06% | 0.21% | 1.77% | 1.91% | 1.79% | 1.65% | 16.61% |
| Terrestrial ecotoxicity | 0.29 | 4.94% | 0.78% | 21.27% | 12.25% | 3.16% | 20.45% | 37.15% |
| Freshwater ecotoxicity | 4.66 | 2.19% | 0.52% | 9.30% | 6.56% | 3.57% | 9.98% | 67.88% |
| Marine ecotoxicity | 4.34 | 3.00% | 0.72% | 15.98% | 10.25% | 4.46% | 16.97% | 48.62% |
| Human carcinogenic toxicity | 125.48 | 2.58% | 2.05% | 8.00% | 7.64% | 6.44% | 12.96% | 60.33% |
| Human non-carcinogenic toxicity | 3.80 | 3.12% | 0.71% | 16.63% | 10.54% | 4.41% | 17.62% | 46.98% |
| Land use | 0.00 | 90.20% | 0.06% | 2.51% | 1.25% | 0.41% | 1.46% | 4.11% |
| Mineral resource scarcity | 0.00 | 11.26% | 1.69% | 22.67% | 12.90% | 1.89% | 25.54% | 24.05% |
| Fossil resource scarcity | 0.06 | 1.76% | 0.47% | 3.56% | 5.15% | 4.61% | 47.68% | 36.77% |
| Water consumption | 0.02 | 41.84% | 18.02% | 8.64% | 2.48% | 6.39% | 7.61% | 15.01% |
| Total | 138.93 | 2.61% | 1.91% | 8.55% | 7.77% | 6.22% | 13.15% | 59.78% |

Table S 7. The normalized value of different environmental impacts and percentage of different raw materials and energy for 800-N-PC at high pressure.

| Environmental impact type | Normalized value | Percentage of different raw materials and energy | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | Wheat | Water | Sodium bicarbonate | Potassium hydroxide | DICY | Nitrogen | Ethanol | Electricity |
| Global warming | 0.02 | 2.06% | 0.58% | 4.52% | 5.22% | 18.01% | 5.42% | 15.45% | 48.74% |
| Stratospheric ozone depletion | 0.00 | 58.19% | 0.47% | 1.68% | 2.76% | 7.37% | 2.67% | 1.80% | 25.06% |
| Ionizing radiation | 0.03 | 1.19% | 0.58% | 2.48% | 5.52% | 10.34% | 5.59% | 1.85% | 72.45% |
| Ozone formation, Human health | 0.01 | 4.11% | 0.64% | 5.57% | 5.61% | 12.65% | 5.55% | 17.43% | 48.44% |
| Fine particulate matter formation | 0.01 | 2.94% | 0.66% | 5.85% | 5.14% | 16.03% | 6.21% | 8.38% | 54.79% |
| Ozone formation, Terrestrial ecosystems | 0.02 | 4.03% | 0.63% | 5.44% | 5.48% | 12.56% | 5.39% | 19.41% | 47.06% |
| Terrestrial acidification | 0.01 | 6.06% | 0.52% | 9.37% | 4.70% | 22.66% | 4.32% | 10.91% | 41.47% |
| Freshwater eutrophication | 0.10 | 4.34% | 0.59% | 5.57% | 5.43% | 7.88% | 5.43% | 14.85% | 55.91% |
| Marine eutrophication | 0.00 | 68.57% | 0.22% | 1.60% | 1.72% | 6.47% | 1.74% | 1.49% | 18.18% |
| Terrestrial ecotoxicity | 0.32 | 2.75% | 0.51% | 11.84% | 6.82% | 39.71% | 1.89% | 11.38% | 25.10% |
| Freshwater ecotoxicity | 3.70 | 1.68% | 0.46% | 7.14% | 5.03% | 11.77% | 2.96% | 7.66% | 63.29% |
| Marine ecotoxicity | 3.62 | 2.19% | 0.60% | 11.67% | 7.48% | 19.05% | 3.51% | 12.39% | 43.10% |
| Human carcinogenic toxicity | 97.29 | 2.02% | 1.86% | 6.28% | 6.00% | 10.69% | 5.46% | 10.18% | 57.51% |
| Human non-carcinogenic toxicity | 3.16 | 2.28% | 0.60% | 12.15% | 7.70% | 19.24% | 3.48% | 12.88% | 41.68% |
| Land use | 0.00 | 87.91% | 0.07% | 2.45% | 1.22% | 1.64% | 0.43% | 1.42% | 4.87% |
| Mineral resource scarcity | 0.00 | 7.58% | 1.32% | 15.27% | 8.69% | 28.91% | 1.37% | 17.20% | 19.66% |
| Fossil resource scarcity | 0.05 | 1.32% | 0.41% | 2.67% | 3.87% | 18.60% | 3.74% | 35.84% | 33.55% |
| Water consumption | 0.01 | 27.05% | 13.49% | 5.59% | 1.60% | 31.10% | 4.46% | 4.92% | 11.78% |
| Total | 108.36 | 2.04% | 1.73% | 6.67% | 6.06% | 11.35% | 5.24% | 10.26% | 56.64% |

## References

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