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Supporting Information

A bioplastic with high strength constructed from cellulose hydrogels by changing aggregated structure

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Assessment process of CP and PE based on life cycle assessment (LCA) and analytic hierarchy process (AHP).

(1) Goal definition and scoping

The first step of life cycle assessment is the goal and scope definition. The LCA scopes of the studied cases include as following:

(a) For production upstream, the pollutant emissions at the stages of biomass growth and petroleum extraction are taken into accounts for cellulose plastics and PE, respectively. The pollutant emissions from raw material transportation are also considered.

(b) For production stage, as a key stage, the pollutant emissions during the whole production process of wood pulps into the cellulose plastic, and petroleum refined and synthesized into polyethylene are taken into accounts. In addition, the indirect pollutant emissions from electricity, chemicals and industry water consumed for plastic production are also taken into accounts.

(c) For production downstream, the environmental impacts resulted from disposal of both plastic materials are taken into account, while the environmental impacts from the use of them are not considered at present. Here cellulose plastic is considered as 100% recyclable material, whereas polyethylene is as 80% recyclable based on the literatures[1], and the landfill is used to the disposal of PE.

(2) Inventory analysis

The main pollutants of atmospheric emissions include CO, NOx, PM_{10}, SO_2, CH_4, N_2O, CO_2, and etc. NH_3-N, COD and etc are considered as main water effluents emission. The solid wastes include disposal of PE, ash and slag. Data concerning the demand for chemicals, energy consumption of transport and pollutant emissions during the production upstream and downstream processes, are taken from references[2-4], including national statistical data, and cleaner production standard of China. For the production process, the pollutant emissions from cellulose plastics is calculated based on the lab experiments and similar process, and those from polyethylene material are based on the industry production data and the statistical data[2-5]. The principle of LCA for cellulose plastics and PE are based on the international standards (ISO 14040, 1997; ISO 14041, 1998; ISO 14042, 2000) and CML 2010[6].

(3) Environment impact assessment

Considering environmental impacts have different importance depending on the geographical scale that is observed, therefore, three sets of weights are developed based on local, regional and global perspectives, which take the geographical scale of the environmental impacts into account.

Based on the weights from AHP, the aggregated index of total environmental load (*SI(k)*) for each perspective can be obtained as following:

$$SI(k) = \sum_{i=1}^{n} W(i,k) \times NP(i) = \sum_{i=1}^{n} SE(i,k) \qquad (1)$$

where *W(i,k)* is weight of the *i* impact category for each perspective *k*, *NP(i)* is normalized environment load of the *i* category impact, *SE(i,k)* is the standardized environmental load of the *i* impact category for each perspective *k*. The detailed data collection and calculation process are below.

The various pollutants have the different environment impact and damage, thus the impact evaluation is divided into two steps: classification and aggregation to determine the relative weights of various impact categories and obtain a single index of impact assessment.

(a) Classification of environmental impacts: In the classification step, the pollutant emissions of the system are divided into different impact categories. In this study, the functional unit for the evaluation of life cycle is 1000 kg of plastic material. Additionally, six impact categories, including global warming (GW), acidification (AC), photochemical ozone formation (POF), eutrophication (EP), human-toxicity (HT), and solid waste (SW), are considered. The characterization factors of various pollutant emissions are taken from CML 2010[6].

The total environmental load (*ED(i)*) for each impact category *i* can be obtained as follow:

$$ED(i) = \sum_{x=1}^{m} [PE(x) \times C(x,i)] \quad (2)$$

where *ED(i)* is the total environment load of the *i* impact category, *PE*(*x*) is amount of the *x* pollutant emissions, *C(x,i)* is the characterization factor of the *i* impact category of the *x* pollutant.

Subsequently, the environmental load for each impact category is normalized by a corresponding normalization reference (*NR(i)*) which is taken from CML 2010[6,7], therefore the normalized environment load of each impact *NP(i)* is calculated as follow :

$$NP(i) = \frac{ED(i)}{NR(i) / Pop_{2000}} \quad (3)$$

where *NP(i)* is normalized environment load of the *i* category impact; *NR*(*i*) is normalization reference of the *i* category impact; Pop_{2000} is the world's population in 2000.

(b) Aggregation of environmental impacts: To establish weights to aggregate the LCA impact categories into an aggregated index, the AHP is used to address impact weights in this study.

The relative weights for each impact category are evaluated according to the AHP procedure[8-10]. According to the six impact categories considered in this study, 6×6 matrices are established with numbers between 1 and 5 expressing the important degree of one impact category relative to the other based on nine-point intensity scale[8,9].

The weight ($W(i, k)$) for the i impact category and the k perspective can be obtained as following:

$$W(i,k)=\frac{[\sum_{j=1}^{n}\left(A_{ij}/\sum_{i=1}^{n}A_{ij}\right)]_k}{n} \quad \text{(i, j=1-6, n=6, } k\text{=regional, gobal, local) (4)}$$

Where A_{ij} is the pair-wise comparison matrix, indicating the relative importance of impact category i compared to impact category j.

The pair-wise comparison matrices for the relative important values of impact categories from three perspectives as well as the resulting weights per impact category are shown in Tables S1- S3.

Table S1. Pair-wise comparison matrix for regional perspective.

	GW	AC	POF	EP	HT	SW	*W(i,R)*
GW	1	1/5	1/3	1/5	1/3	1	0.053
AC	5	1	3	1	3	5	0.317
POF	3	1/3	1	1/3	1	3	0.130
EP	5	1	3	1	3	5	0.317
HT	3	1/3	1	1/3	1	3	0.130
SW	1	1/5	1/3	1/5	1/3	1	0.053
Total	18	3.07	8.67	3.07	8.67	18	1.00

Table S2. Pair-wise comparison matrix for global perspective.

	GW	AC	POF	EP	HT	SW	*W(i,G)*
GW	1	2	3	3	4	5	0.364
AC	1/2	1	2	2	3	4	0.230
POF	1/3	1/2	1	1	2	3	0.136
EP	1/3	1/2	1	1	2	3	0.136
HT	1/4	1/3	1/2	1/2	1	2	0.081
SW	1/5	1/4	1/3	1/3	1/2	1	0.052
Total	2.62	4.58	7.83	7.83	12.50	18	1.00

Table S3. Pair-wise comparison matrix for local perspective.

	GW	AC	POF	EP	HT	SW	*W(i,L)*
GW	1	1/3	1/4	1/4	1/5	1/5	0.044
AC	3	1	1/2	1/2	1/3	1/3	0.094
POF	4	2	1	1	1/2	1/2	0.156
EP	4	2	1	1	1/2	1/2	0.156
HT	5	3	2	2	1	1	0.275
SW	5	3	2	2	1	1	0.275
Total	22	11.33	6.75	6.75	3.53	3.53	1.00

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Supplementary Figures

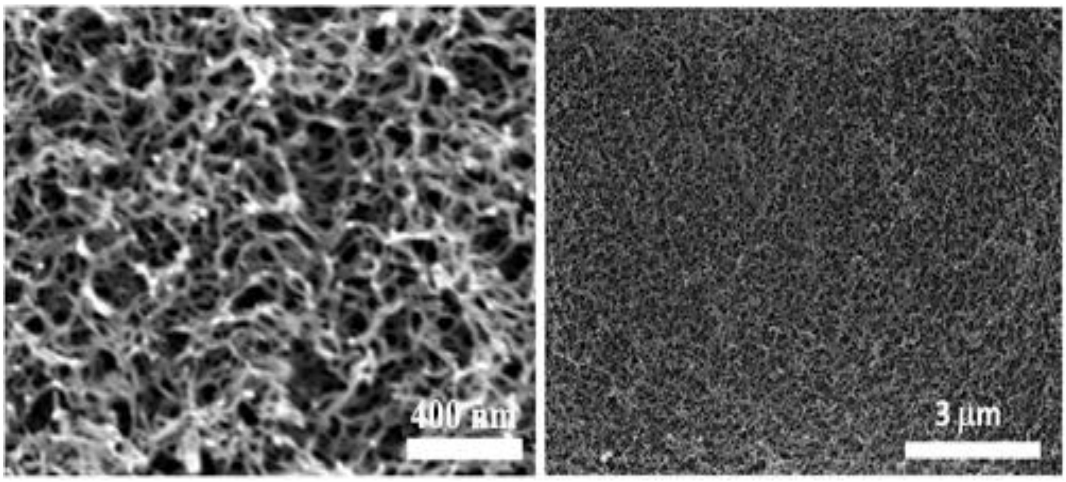


Fig. S1 SEM image of inner part (left) and surface part (right) of the cellulose hydrogel.

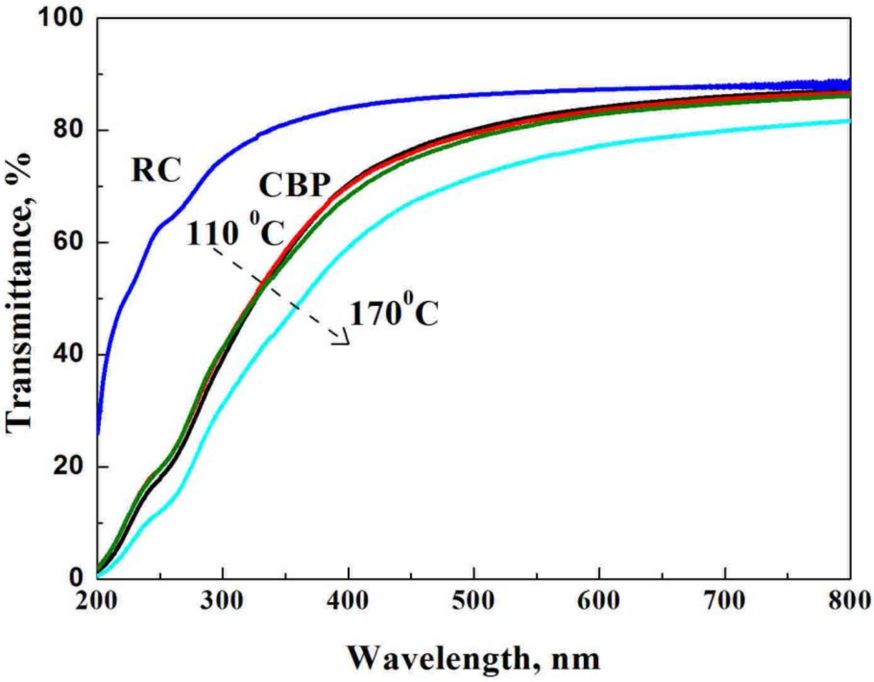


Fig. S2 UV-vis spectroscopy of the CBP sheets (0.3 mm thick) prepared from the cellulose hydrogels (3 mm thick) by hot-pressing at different temperatures (110, 130, 150, and 170 ^{o}C). The regular-dried cellulose film (RC) is approximately 0.1 mm thick for comparison.

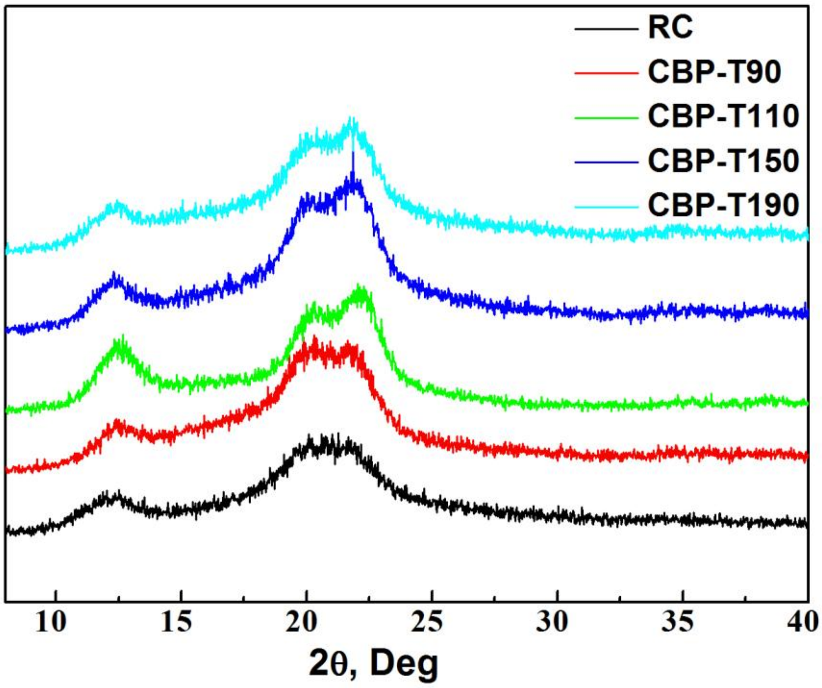
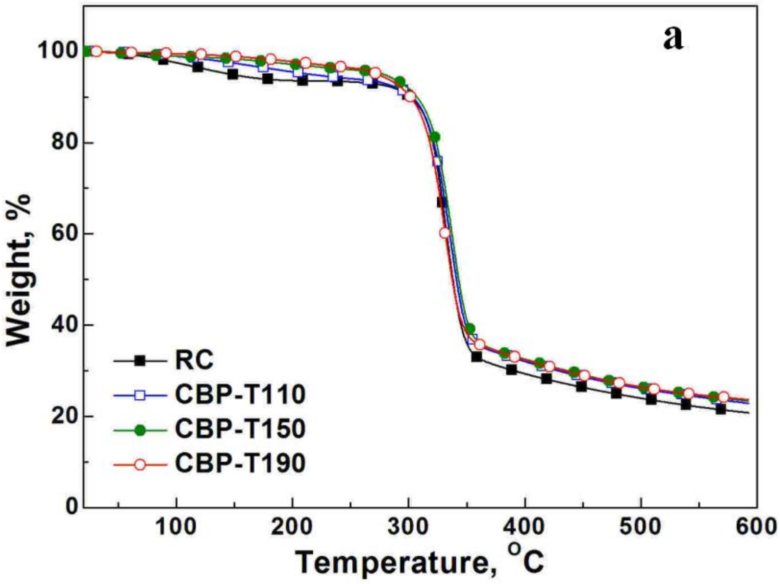


Fig. S3 X-ray diffraction profiles of the cellulose samples (RC, regular drying at ambient temperature; CBP-T90, 90 ^{o}C; CBP-T110, 110 ^{o}C; CBP-T150, 150 ^{o}C; CBP-T190, 190 ^{o}C), respectively.



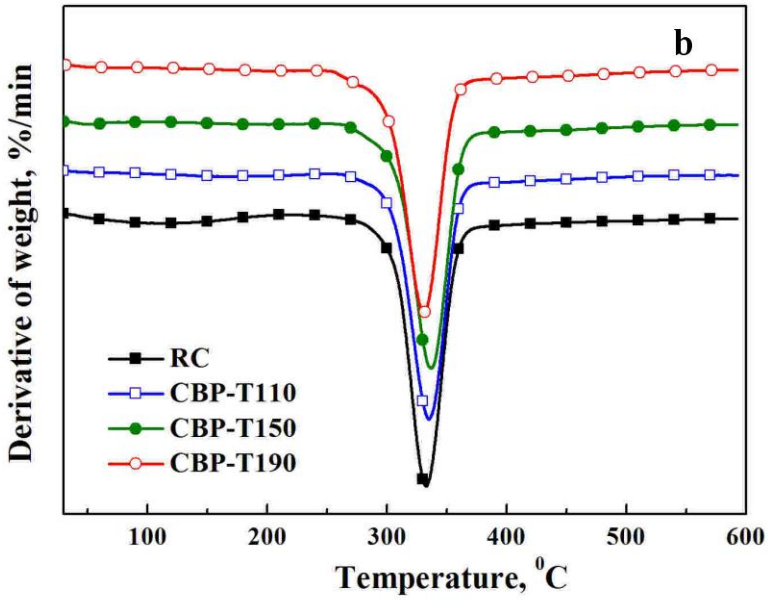


Fig. S4 Thermogravimetry (a) and derivative thermogravimetry (b) curves of cellulose bioplastics prepared by hot-pressing at different temperatures (CBP-T110, 110 °C; CBP-T150, 150 °C; CBP-T190, 190 °C) under nitrogen atmosphere, respectively. The regular dried cellulose film (RC) with 0.1 mm thickness was evaluated for comparison.

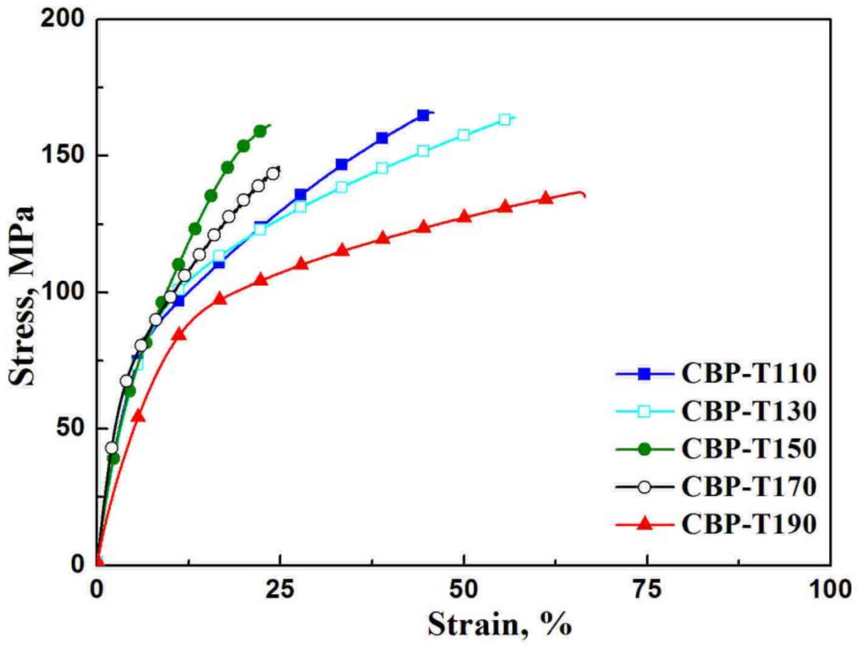
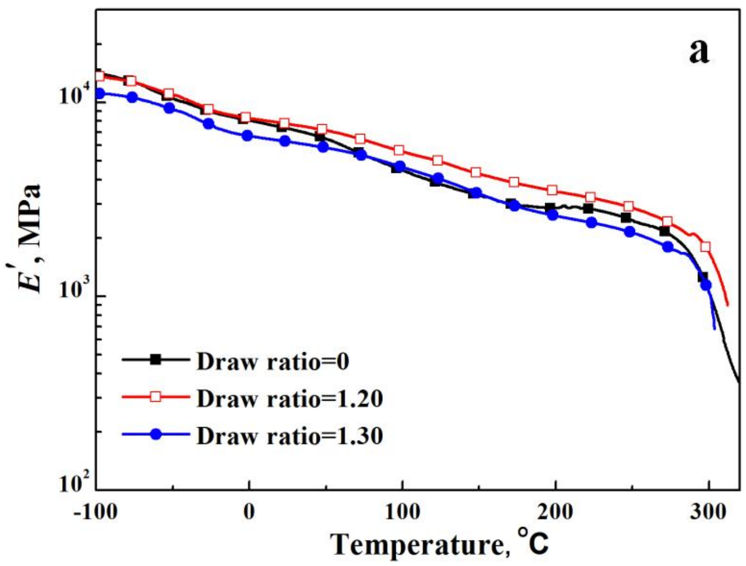


Fig. S5 Representative stress–strain curves of cellulose bioplastics prepared from 3-mm-thickness cellulose hydrogels by hot-pressing at different temperatures (CBP-T110, 110 ^{o}C; CBP-T130, 130 ^{o}C; CBP-T150, 150 ^{o}C; CBP-T170, 170 ^{o}C; CBP-T190, 190 ^{o}C), respectively.



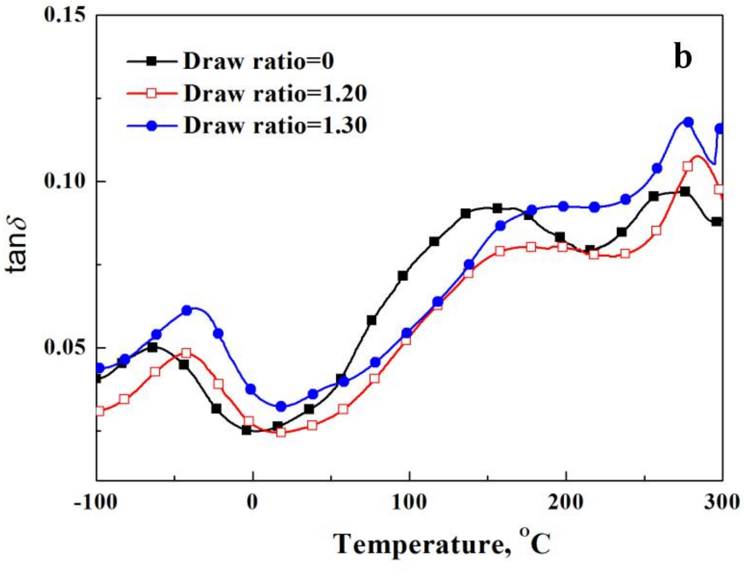


Fig. S6 Representative DMA temperature sweeps that show tensile storage moduli *E'* and loss tangents tanδ of CBPs prepared by hot-pressing at 110 °C, and then oriented with different draw ratio as a function of temperature, respectively.

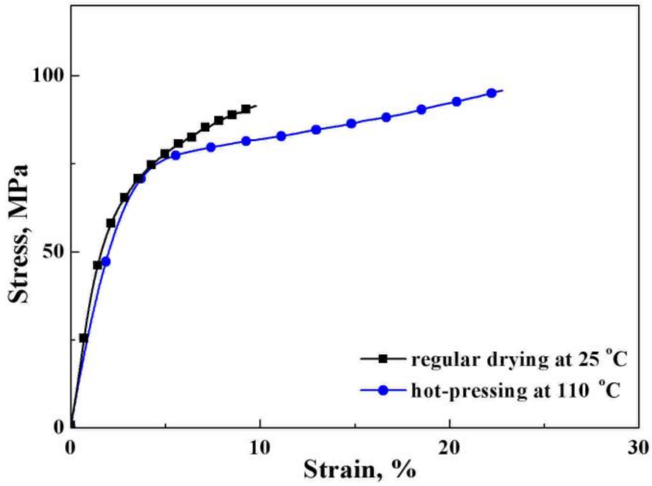


Fig. S7 Stress-strain curves of the cellulose films prepared with 1-mm-thickness cellulose hydrogels by regular drying at ambient temperature and hot-pressing at 110 °C, respectively. Both cellulose films were 0.1 mm in thickness after drying.

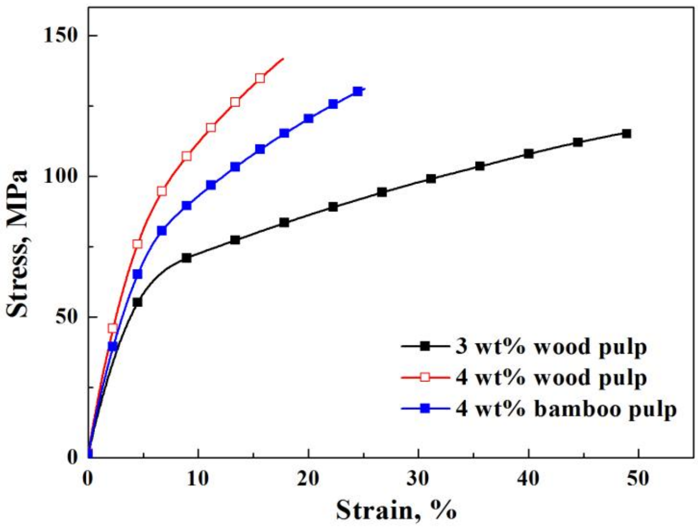


Fig. S8 Stress-strain curves of the CBP with 0.35 mm thickness, prepared from 3-mm-thickness cellulose hydrogels with various raw materials (wood pulp and bamboo pulp) by hot-pressing at 110 °C, respectively.

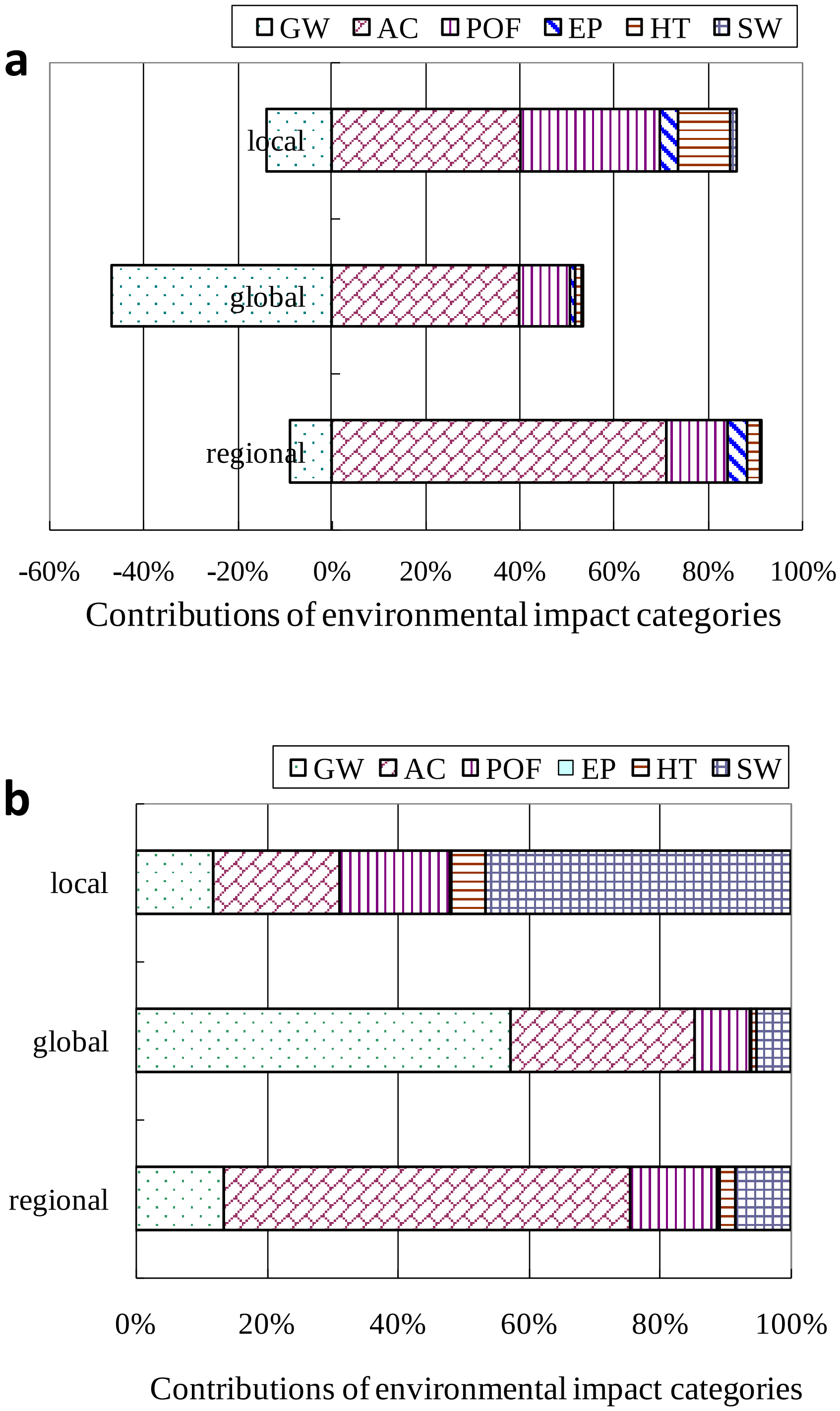


Fig. S9 Environmental impact of CBP and PE. (a) Contributions of environmental impact categories for CBP; (b) Contributions of environmental impact categories for PE. (GW, global warming; AC, acidification; POF, photochemical ozone formation; EP, eutrophication; HT, human-toxicity; SW, solid waste)