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Country-level potential of carbon sequestration and environmental benefits by utilizing crop residues for biochar implementation

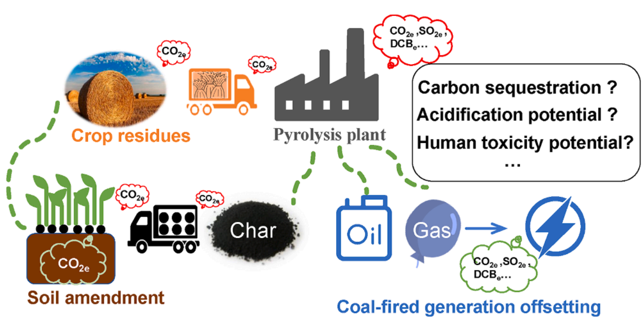
Qiushuang Yang [a], Ondřej Mašek [b], Ling Zhao [c,a,*], Hongyan Nan [c], Shitong Yu [c], Jianxiang Yin [a], Zhaopeng Li [c], Xinde Cao [c]

[a] *China-UK Low Carbon College, Shanghai Jiao Tong University, Shanghai 201306, China*
[b] *UK Biochar Research Centre, School of Geosciences, University of Edinburgh, Kings Buildings, Edinburgh EH93JN, UK*
[c] *School of Environmental Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, China*

HIGHLIGHTS

- Biochar soil storage and biofuel utility achieve over 900 $kgCO_{2e}\cdot t^{-1}$ sequestration.
- Biochar implementation could reduce around 4.5% of annual national carbon emission.
- Central south, east and northeast of China contribute 65% of the national GWP100.
- Biochar system could mitigate aquatic ecotoxicity and soil acidification, etc.
- Bio-energy offsetting coal-fired power generation dominates carbon negativity.

GRAPHICAL ABSTRACT



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ABSTRACT

Conversion of biomass into biofuel and biochar with a subsequent soil storage is assumed as a prospective strategy of reducing atmospheric CO_2 concentrations. However, substantial uncertainties exist in this field regarding the country-level potential of biochar carbon sequestration, indirect effects of biochar implementation on overall environment, and dominating factors. This study conducted a life cycle assessment of country-wide incorporation of biochar in agriculture, and associated potential benefits. Results showed that over 920 kg CO_{2e} (CO_2-equivalent) could be sequestrated via converting 1 t of crop residues into biochar. As an example, based on crop residues availability statistics for China in 2014, the estimated annual carbon sequestration potential could be as high as 0.50 Pg CO_{2e} (1 Pg = 1 × 10^9 t). The most significant potential for biochar carbon sequestration was identified in the central south, east and northeast of China, which contributed 65% of the national biochar carbon sequestration potential. The biochar system could also contribute to mitigation of the following environmental problems: marine aquatic biodiversity destruction, surface soil and water acidification, etc. Sensitivity analysis demonstrated that biochar yield, carbon content in biochar, electricity conversion efficiencies of bio-oil and pyrolysis gas were the critical parameters determining the biochar system's overall carbon sequestration potential and environmental effects. This study provides guidance on evaluating biochar's potential carbon sequestration capacity and comprehensive environmental impacts, as well as research and development needs.

* Corresponding author at: School of Environmental Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, China.
E-mail address: wszhaoling@sjtu.edu.cn (L. Zhao).

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1. Introduction

With the rapid increase in global population and the ever-increasing reliance on fossil fuel resources for energy and material, one of the most serious problems that human being faces is global warming and resulting climate change. This is due to the rising concentration of greenhouse gases (GHG) in the atmosphere, especially CO_2, which is predicted to double by the end of the 21st century [1]. Mitigating global warming therefore requires a significant reduction in atmospheric GHG concentrations. Globally, large production of waste biomass and lack of appropriate recycling technology contribute dramatically to GHG emissions [2]. For instance, in China, farmers burn straw in field to avoid hindering next round of planting. And open burning of crop residue, especially during the harvest seasons, has led to a series of serious environmental problems, such as emission of CO_2, CO, NO_x, photochemical oxidation and particulates, destroy of soil microbial community, etc. Xu et al. [3] reported that in 2016, emissions of carbon monoxide and $PM_{2.5}$ by crop residue burning were 3.63×10^6 and 6.96×10^5 tons in Northeast China, 9.77×10^5 and 1.36×10^5 tons in Southwest China, and 1.24×10^5 and 1.19×10^4 tons in Guangdong province alone. The sustainable management of large amounts of agricultural and forest residues is also a big challenge for countries around the world.

Biomass pyrolysis yields a range of solid, liquid and gaseous outcomes with applications as biofuels [4], source of chemicals [5], etc. Biochar, the solid product of biomass pyrolysis, is an important substance with many environmental [6] and agricultural benefits [7], which has gained a lot of attention [8]. A large volume of research has been conducted and published on many aspects of biochar applications in agriculture, environmental management [9], and carbon storage [10]. Although the recalcitrance of biochar-carbon stemming from its aromatic structure has been demonstrated [11], the extent of carbon negativity of biochar systems still requires further research and demonstration, due to the energy and material requirements of the system [12]. Several studies have applied life cycle analysis (LCA) methods to investigate the total carbon sequestration potential of the process, where the carbon footprints of a product, activity, or process from cradle to grave are evaluated by considering all inputs and outputs of energy and carbon [13], and they incorporated many influencing factors such as materials collection, pyrolysis, transport, conversion of bio-oil/gas to electricity, etc [14].

However, there are outstanding issues with analysis of biochar systems due to the complexity and diversity of applications and impacts on environment, resulting in different considerations and system boundaries being used by different researchers. For example, Roberts et al. [15] selected cornstover and yard waste to study the material and energy flows in a slow pyrolysis system. They reported that the net GHG emission reduction was around 870 kg $CO_{2e}\cdot t^{-1}$ dry feedstock, and 65% of the total reduction was ascribed to carbon remained in biochar. However, this study did not incorporate the promoting effect of biochar on crop yield. Kauffman et al. [16] performed a LCA of fast pyrolysis and emphasized the benefits of biochar on agricultural productivity, and impact of biochar on emissions from indirect land use change. Peters et al. [17] analyzed a slow pyrolysis system which generated heat and biochar from lignocellulosic crops, and compared the life-cycle environmental performances of different scenarios; they mainly compared the effects of different biochar utilities such as direct combustion and natural gas substitutes, in which all secondary data were specific for Spain.

Despite there have been wide researches in this field, a more comprehensive and systematic research is needed due to: 1) knowledge system related to energy and mass balance of biochar production, environmental impacts (marine aquatic ecotoxicity, acidification, etc.), and biochar-induced agricultural benefits (promoting plant growth, fertilizers saving, etc.) keeps growing [18]; 2) as the research developed, more accurate data could be obtained and models are also evolving; 3) it is necessary to evaluate the GHG mitigation potential of biochar systems at a country level based on the actual availability of biowastes. In this regard, Woolf et al. [19] assessed the biomass amount of the whole world and reported that biochar could reduce the annual global net emissions of GHGs (CO_2, CH_4, and N_2O) by a maximum of 1.80 Pg CO_{2e} per year, which was equivalent to 12% of the total annual anthropogenic GHG emissions.

Before the large-scale application, more insights into the potential carbon sequestration and the overall environmental impacts of biochar implementation are still needed, especially from the macro perspective [20]. Therefore, in this study, with the aim of answering a question "if all of a country's crop residues were used to produce biochar for soil storage and biofuel for energy recovery without endangering food security, what's the maximum amount of carbon that could be sequestrated?", a life cycle model was established based on the updated GaBi software, and a systematic assessment for biochar's potential in carbon sequestration and environmental amelioration was performed based on the amounts of crop residues generated at a country-level with China as an example. Besides, the potential of biochar system influencing the main environmental issues such as abiotic depletion, acidification, eutrophication, etc. were also quantified. Sensitivity and scenario analysis were conducted to identify the critical parameters in the biochar system, and to guide the future application of biochar technology and supporting policies.

2. Methods and data

2.1. Goal, functional unit and system boundary

The goal is to explore the carbon sequestration potential and environmental impacts of biochar implementation in China using LCA analysis. All abbreviations and the corresponding full names can be seen in Table 1. The system boundary is presented in Fig. 1, in which the functional unit (FU) was set as 1 t of crop residues. The crops selected in this study included grain, bean, tuber, oil crop, cotton, sugarcane and hemp, which are the most common crops in China. In this study we did not consider the planting process of crops and only crop residue management was assessed. Collection and spreading operations in the field were powered by electricity, and the transportation process was considered to consume diesel. Building of pyrolysis plant and construction, operation and maintenance of the pyrolysis system were included in the slow pyrolysis process. Energy produced as bio-oil and pyrolysis gas was utilized to offset coal-fired electricity generation. The biochar was used as a soil amendment in the field [21]. Environmental benefits of biochar including carbon sequestration [22], crop yield increase [23,24], fertilizer saving [25], N_2O emission reduction [26], and soil organic carbon (SOC) enhancement [27] were incorporated in the calculations. Considering the possible mineralization of biochar-carbon in soil, only the stable carbon fraction was involved in the calculation [28,29]. Although several studies have reported that biochar may reduce the methane emission from soil [30], the data available was not yet conclusive, therefore this potential benefit was not included in this study.

2.2. Data source, inventory analysis and calculation program

All material and energy flow data within the system boundary were identified based on 1-t crop residue and a time span of one year. The life cycle inventory (LCI) is shown in Table S1. Inventory data were collected from literature and national statistics, which were selected to be as relevant as possible to the Chinese context (Table S2). If Chinese data were lacking, then related data from research in other countries were cited. Due to the diversity of data sources, we filtered out the representing data or adopted the averaged values in order to reflect the Chinese context. The latest national GHG emission inventory data from 2014 were published by the Chinese government in 2019 [31],

Table 1
Abbreviations and the corresponding full names.

Abbreviation	Full name
GHG	atmospheric greenhouse gas
LCA	life cycle analysis
FU	functional unit
SOC	soil organic carbon
LCI	life cycle inventory
$E_{o/g}$	bio-oil/pyrolysis gas conversion electricity
$M_{o/g}$	bio-oil/pyrolysis gas mass
$CV_{o/g}$	bio-oil/pyrolysis gas calorific value
$CE_{o/g}$	bio-oil/pyrolysis gas electricity conversion efficiency
C_{cs}	carbon sequestration in soil
M_b	biochar mass
C_b	carbon content in biochar
C_{sc}	stable carbon content in biochar
C_{ci}	sequestrated carbon from increased crop yield
r_b	biochar spreading ratio
y_c	crop yield
y_i	crop yield increase
C_c	carbon content in crop
A_N	reduced N-fertilizer mass
$A_{f,N}$	N-fertilizer application mass in the field
S_c	crop planting area
R_N	N-fertilizer reduction
A_P	reduced P-fertilizer mass
$A_{f,P}$	P-fertilizer application mass in the field
R_P	P-fertilizer reduction
A_K	reduced K-fertilizer mass
$A_{f,K}$	K-fertilizer application mass in the field
R_K	K-fertilizer reduction
E_{N_2O}	reduced N_2O emission
E	N_2O-N emission
R_{N_2O}	N_2O emission reduction
R_{SOC}	sequestrated carbon from reduced SOC mineralization
R	average SOC reserve in field
r_{SOC}	SOC mineralization reduction
ADP	abiotic depletion potential
AP	acidification potential
EP	eutrophication potential
GWP100	global warming potential at 100-year scale
ODP	ozone layer depletion potential
POCP	photochemical ozone creation potential
HTP	human toxicity potential
ETP	ecological toxicity potential
TETP	terrestric ecotoxicity potential
FAETP	freshwater aquatic ecotoxicity potential
MAETP	marine aquatic ecotoxicity potential
EI(j)	environmental impact potential of the system on the j-th environmental impact category
$EI(j)_i$	contribution of the i-th substance to the j-th environmental impact category
M_i	emission of the i-th substance
$EI(j)_i$	characterization factor for the i-th substance on the j-th environmental impact category
LCC	life cycle cost
NMVOC	non-methane volatile organic compound

therefore, all evaluations carried out were based on the Chinese data in 2014. It is notable that the methodology and basic calculations could be applied to other inventory data easily if updated data become available in the future.

There were some formulas in our calculation program as follows [32]:

$$E_{o/g} = -\frac{M_{o/g} \times CV_{o/g} \times CE_{o/g}}{3.60} \tag{1}$$

where, $E_{o/g}$, bio-oil/pyrolysis gas conversion electricity, kWh; $M_{o/g}$, bio-oil/pyrolysis gas mass, kg; $CV_{o/g}$, bio-oil/pyrolysis gas calorific value, $MJ \cdot kg^{-1}$; $CE_{o/g}$, bio-oil/pyrolysis gas electricity conversion efficiency, %; 3.60, MJ-kWh conversion coefficient.

$$C_{cs} = -M_b \times C_b \times C_{sc} \times 3.67 \tag{2}$$

where, C_{cs}, carbon sequestration in soil, kg CO_{2e}; M_b, biochar mass, kg; C_b, carbon content in biochar, %; C_{sc}, stable carbon content in biochar, %; 3.67, C-CO_2 conversion coefficient.

$$C_{ci} = -\frac{M_b}{r_b} \times y_c \times y_i \times C_c \times 3.67 \tag{3}$$

where, C_{ci}, sequestrated carbon from increased crop yield, kg CO_{2e}; r_b, biochar spreading ratio, $t \cdot ha^{-1}$; y_c, crop yield, $t \cdot ha^{-1}$; y_i, crop yield increase, %; C_c, carbon content in crop, %.

$$A_N = -\frac{M_b}{r_b} \times \frac{A_{f,N}}{S_c} \times R_N \tag{4}$$

where, A_N, reduced N-fertilizer mass, kg; $A_{f,N}$, N-fertilizer application mass in the field, kg; S_c, crop planting area, ha; R_N, N-fertilizer reduction, %.

$$A_P = -\frac{M_b}{r_b} \times \frac{A_{f,P}}{S_c} \times R_P \tag{5}$$

where, A_P, reduced P-fertilizer mass, kg; $A_{f,P}$, P-fertilizer application mass in the field, kg; R_P, P-fertilizer reduction, %.

$$A_K = -\frac{M_b}{r_b} \times \frac{A_{f,K}}{S_c} \times R_K \tag{6}$$

where, A_K, reduced K-fertilizer mass, kg; $A_{f,K}$, K-fertilizer application mass in the field, kg; R_K, K-fertilizer reduction, %.

$$E_{N_2O} = -\frac{M_b}{r_b} \times E \times 3.14 \times R_{N_2O} \tag{7}$$

where, E_{N_2O}, reduced N_2O emission, kg; E, N_2O-N emission, kg; R_{N_2O}, N_2O emission reduction, %; 3.14, N-N_2O conversion coefficient.

$$R_{SOC} = -\frac{M_b}{r_b} \times R \times r_{SOC} \times 3.67 \tag{8}$$

where, R_{SOC}, sequestrated carbon from reduced SOC mineralization, kg CO_{2e}; R, average SOC reserve in field, $t \cdot ha^{-1}$; r_{SOC}, SOC mineralization reduction, %.

The different ways biochar can contribute to GHG emissions and carbon sequestration were quantified by formulas (2) to (8). The inputs and outputs of materials and energy for each approach in the biochar system are listed in Table 2.

2.3. Assessment of environmental impacts

LCA software, GaBi 8.70, developed by the Thinkstep group was employed for background data support and computational implementation of the impact categories. Detailed information on GaBi is available on the website of http://www.gabi-software.com/china/index/. GaBi offers various quantitative methods such as CML 2001, EDIP 2003, ReCiPE 1.08, and UBP 2013. Among them, CML 2001 is a calculating method with more transparency and less uncertainty, thus it was chosen to quantify the ten environmental impacts of applying pyrolysis technology [33]. More information about this method can be found on the website of the Institute of Environmental Science at Leiden University of the Netherlands (IESLUN, 2019) [34]. These impacts could be divided into three categories: 1) Resource consumption; 2) Environmental damage; 3) Toxicity potential. Resource consumption was evaluated using an index ‘abiotic depletion potential (ADP)’, which reflects the depletion of non-renewable resources. Environmental impact classification factor consists of acidification potential (AP), eutrophication potential (EP), global warming potential at 100-year scale (GWP100), ozone layer depletion potential (ODP) and photochemical ozone creation potential (POCP). The third impact focuses on. HTP refers to potential toxicity to human induced by substances released from processes, and the latter is described as terrestric ecotoxicity potential (TETP), freshwater aquatic ecotoxicity potential (FAETP) and marine aquatic ecotoxicity potential (MAETP). The theoretical basis of calculating these environmental impacts is expressed by the following formula:

$$EI(j) = \sum EI(j)_i = \sum [M_i \times EI(j)_i] \quad (9)$$

where, EI(j), environmental impact potential of the system on the j-th environmental impact category, $kgreferent_e(referent - equivalent, suchasSb_e, CO_{2e}, etc.) \cdot t^{-1} cropresidues$; $EI(j)_i$, contribution of the i-th substance to the j-th environmental impact category, $kgreferent_e \cdot t^{-1} cropresidues; M_i$, emission of the i-th substance, $kg \cdot t^{-1} cropresidues; EI(j)_i$, characterization factor for the i-th substance on the j-th environmental impact category. For a detailed introduction to the characterization factors for each impact category in the CML2001 method, refer to http://cml.leiden.edu/software/data-cmlia.html. More detailed information on these environmental impacts is provided in the SI (Page S14). The calculation of characterization, normalization and Monte-Carlo analysis was performed in GaBi software.

2.4. Uncertainty analysis and statistical calculation

The uncertainty of the calculation results was determined by Monte-Carlo analysis (normal distribution, n = 5000). In the calculation, the variation range of all parameters in the biochar system was set to ± 10%. There would be discrepancies which measured the uncertainty of this study between the Monte Carlo simulation results and the LCA results.

2.5. Sensitivity analysis and scenario calculation

Sensitivity analysis was conducted for a fluctuation of the parameters in the system by ± 10%. After identifying the key parameters in the system based on the sensitivity coefficients, different hypothetical values were set for these parameters to perform scenario analysis.

2.6. Economic feasibility analysis

Life cycle cost (LCC) analysis was performed to determine the economic feasibility of biochar implementation. In this section, the expenditures and incomes of the biochar system were summarized and calculated, and then the benefit of its application was obtained.

Table 2
Input-output inventory based on 1-t crop residues.

Process	Input	Output
Crop residue collection	Crop residues 1 t Electricity 68.98 kWh	Collected crop residues 1 t
Crop residue transportation	Collected crop residues 1 t Diesel 1.49 kg	Crop residues after transportation (1% mass loss) 990.00 kg
Slow pyrolysis	Crop residues 990.00 kg Brick 15.19 kg Concrete 0.0046 m^3Steel 0.69 kg Electricity 205.47 kWh Tap water 272040 kg	Biochar 348.28 kg Pyrolysis gas 344.12 kg Bio-oil 297.59 kg
Coal-fired generation offsetting	Pyrolysis gas 344.12 kg Bio-oil 297.59 kg	Electricity (offsetting coal-fired generation) 678.13 kWh
Biochar transportation	Biochar 348.28 kg Diesel 0.52 kg	Biochar after transportation (3% mass loss) 337.83 kg
Biocharspreading	Biochar 337.83 kg Electricity 53.11 kWh	Biochar 337.83 kg
Biochar field application	Biochar 337.83 kg	Carbon storage in soil 599.22 $kgCO_2e$ Fixed carbon from crop yield increase 18.50 kg CO_2eReduced N-fertilizer amount 0.10 kg Reduced P-fertilizer amount 0.02 kg Reduced K-fertilizer amount 0.01 kg Reduced N_2O emission 0.01 kg Fixed carbon from reduced SOC mineralization 104.15 $kgCO_2e$

SOC: soil organic carbon.

3. Results and discussion

3.1. Mass and energy flow of biochar system

Based on the collected data listed in Table S1 and Table 2, an average

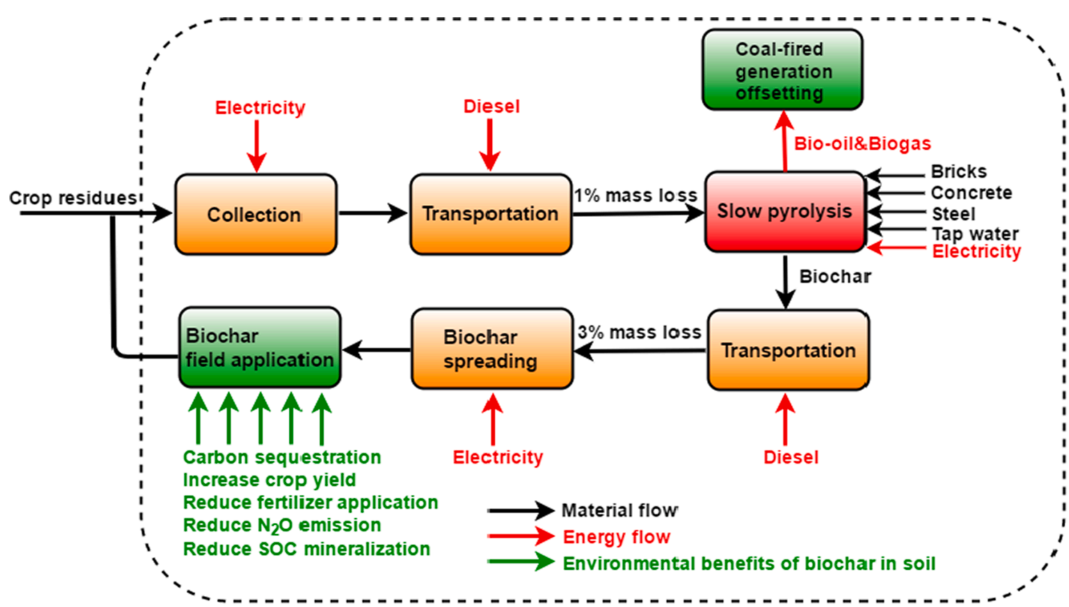

Fig. 1. System boundary of the biochar production and field application.

mass loss of 1.00% was assumed for transporting biomass feedstock to the pyrolysis plant, and the average yield of biochar was about 35.0% of the raw material, with an additional 3.00% mass loss of transporting biochar to the field. Therefore, based on 1-t crop residues evaluation, 337.83 kg (Table 2) of biochar was finally incorporated into soil for achieving carbon sequestration [35]. It is worth noting that during the biochar production process, there was inevitably a total consumption of 327.56 kWh electricity and 2.01 kg of fuel for 1 t feedstock, however, if the produced pyrolytic oil/gas were efficiently recovered as energy, it could offset the pyrolytic energy consumption. Existing studies have proven that the calorific values of pyrolytic oil/gas were as high as 17.5 and 6.00 $MJ·kg^{-1}$, respectively; The advanced coal-fired electricity generation technologies could achieve about 35% electricity conversion efficiency. Therefore, the recycled energy could act as 678.13 kWh of electricity generated from coal-fired (Table 2). In this regard, the whole LCA of biochar system exhibited a negative (i.e., favorable) effect on climate warming, and the GWP100 value reached −921.30 kg CO_{2e} (FU = 1 t crop residues). Detailed data involved in each process was presented in Table S3.

In this system, slow pyrolysis was the largest contributor to the system's carbon emissions (392.40 kg CO_{2e}), including the construction, operation and maintenance of the pyrolysis plant. In addition, the collection of crop residues in field and spreading biochar to soil also consumed considerable energy, which was equivalent to emissions of 75.66 and 78.74 kg CO_{2e}, respectively. Spreading biochar onto the field could be conducted together with other soil management practices, offering additional options for decreasing the energy consumption. In contrast, the transportation process consumed comparatively small amounts of energy (the emission was only 0.88 kg CO_{2e}). Despite these unavoidable emissions, the carbon negativity of biochar was still prominent, largely due to the carbon sequestration potential of biochar when applied to soil (-743.83 kg CO_{2e}), and offsetting carbon emission by fossil fuel for power generation (-725.15 kg CO_{2e}). Taking the original calorific value and conversion efficiency of 33–35% (Table S1), the pyrolytic oil/gas produced by slow pyrolysis could be utilized to offset 678.13 kWh of coal-based electricity (FU = 1 t of crop residues) according to formula (1) (Table 2). Upgrading and high-efficiency utilization of pyrolytic oil/gas are still being researched [36,37], and future developments are expected to increase its conversion efficiency [38,39].

The biochar produced by pyrolysis of 1-t crop residues could be incorporated into the soil to achieve stable carbon storage of 599.22 kg CO_{2e}. Due to the role of biochar in promoting plant growth, more atmospheric carbon was assimilated into the crops, which accounted for the additional carbon captured by biochar system of 18.50 kg CO_{2e}. More importantly, biochar offers the prospects of reducing the excessive use of nitrogen and phosphorus fertilizers in China, thus alleviating associated environmental issues such as soil acidification, water eutrophication and loss of nutrient resources. According to our calculations, application of 337.83 kg biochar (from 1 t biomass) could reduce 0.10 kg of N-fertilizer, 0.02 kg of P-fertilizer and 0.01 kg of K-fertilizer demand per year. Besides, it was estimated that biochar application would also reduce soil N_2O emission by 0.01-kg. Furthermore, an additional fixing of 104.15 kg of CO_{2e} could be achieved via the inhibition of SOC mineralization by biochar amendment (Table 2). As a prospective soil amendment, biochar also finds its place in remediating soil pollution, i.e., it can stabilize heavy metals or adsorb organic contaminants, while this was not incorporated into the evaluation of carbon sequestration potential in this study because it is worthy further study.

3.2. Carbon sequestration potential of biochar application in China

To assess the maximum annual carbon sequestration potential of biochar application at a country level, China was taken as an example, and results could be used as a reference to analyze the situations of any other country. The data of available crop residue resources in China 2014 were listed in Table S2. Since the special administrative regions of China including Hong Kong, Macao and Taiwan are administered separately and the data were not available, we only used data from the main land of China. The crop residues potentially available as feedstock for biochar production amounted to 5.41×10^8 t without compromising agricultural production sustainability and competing with other established uses. Therefore, the annual carbon sequestration potential of biochar production was estimated at approximate 4.98×10^8 t CO_{2e} (Fig. 2a). Majority of this contribution was achieved by three staple food crops in China, i.e., rice, wheat and corn, with total annual production of 5.41×10^8 t. Among these, biochar from corn residues alone contributed over 45% of the total carbon sequestration potential, making it the single most important feedstock. According to the governmental report, the Second Biennial Update Report on Climate Change of PRC, the total net GHG emissions in China was 11.19 billion t CO_{2e} in 2014 [31], thus applying pyrolysis technology to convert crop residues into biochar could mitigate approximately 4.50% of China's total GHG emissions. In contrast, Jin et al. [40] reported that the annual average CO_2 emissions from open burning of main crop straws (rice, wheat, corn, rape and soybean straw) in mainland China was$4.22 \times 10^8 t$ during 2000–2014.

The main carbon emissions associated with biochar production were related to heating in the pyrolysis process (2.12×10^8 t CO_{2e}), biomass collection (4.09×10^7 t CO_{2e}) and biochar spreading (4.26×10^7 t CO_{2e}). On the other hand, emissions related to transportation were much smaller, amounting to 4.76×10^5 t CO_{2e} in total. In contrast, reuse of pyrolytic oil/gas could offset fossil fuel consumption for generating 3.67×10^{11} kWh of electricity, thus avoiding emissions of 4.02×10^8 t CO_{2e}. Furthermore, biochar for soil amendment could achieve sequestration of additional 3.92×10^8 t CO_{2e} (Fig. 2a, Table S4).

Besides the total carbon sequestration potential of biochar in the country, we also investigated the regional distribution, based on the data on available crop residues [41] in different main regions of China [42,32], as shown in Table S5. Fig. 3 shows the carbon sequestration potential of biochar implementation in various provinces in China. Because of the abundant crop residue resources, the potential of the central south and east of China accounted for 22.32% and 22.07% of carbon sequestration respectively. Northeast, north and southwest of China, separately covered 20.03%, 15.41% and 11.07%. The lowest carbon sequestration potential of 9.09% was obtained in northwest of China. The central south, east and northeast of China contributed almost 65% of the national biochar carbon sequestration potential. Among them, Henan, Heilongjiang, and Shandong were the provinces that deserve the most attention. Results interestingly revealed the prominent influences of geographical environment on crop production, availability of residues, and the resulting carbon sequestration potential. For instance, the climate of the east of China is relatively wet and warm, offering relatively high crop residue yields, and it could be a suitable pilot area for applying pyrolysis technology. In contrast, the cold North China and dry West China showed a relatively low level of agricultural productivity and a corresponding low carbon sequestration potential.

3.3. Potential environmental impacts

Environmental impacts of each process in the biochar life cycle are presented in Fig. 2b, and Table 3 lists the annual values of each impact calculated from the sum of subprocesses. The results show that the biochar system had positive environmental impacts, except in case of abiotic depletion potential (ADP) and ozone layer depletion potential (ODP). Compared to the open burning of crop residues, converting them into biochar brought many environmental advantages. Zhang et al. [43] reported that crop residue burnings in China was responsible for the substantial release of harmful atmospheric substances including CO_2, $PM_{2.5}$, NMVOC (non-methane volatile organic compound), CH_4, NO_x, SO_2 and NH_3, and releasing values of these substances in China's 2014 were 305.20, 1.77, 1.85, 1.77, 0.53, 0.16 and 0.12 Tg (1 Tg = 1×10^6 t), respectively. In fact, all these positive environmental effects were greatly related to the offsetting of coal-fired electricity generation by the

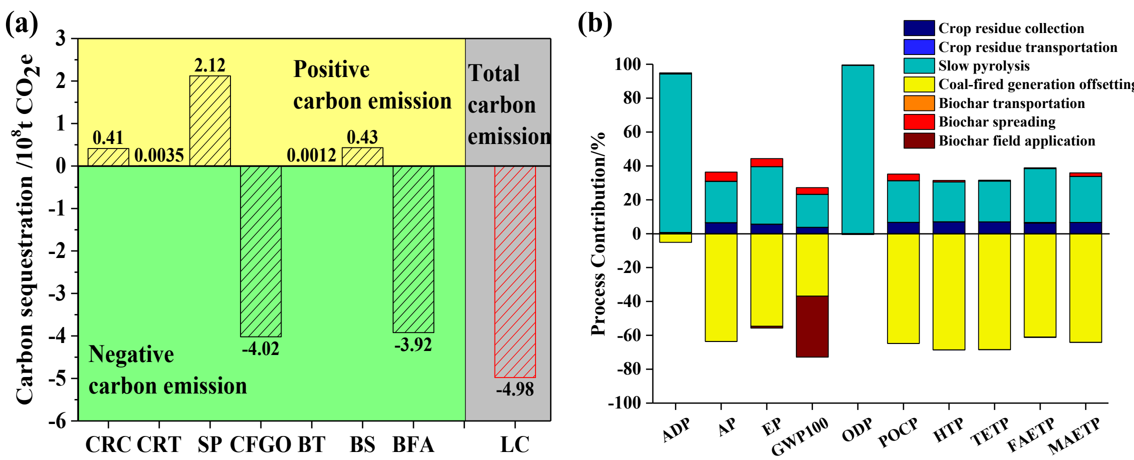

Fig. 2. (a) Annual carbon emission of each process in the biochar production-application system based on the latest data of total available crop residues in China (2014), and (b) The main environmental impacts of each process in biochar-production-application system. CRC: crop residue collection; CRT: crop residue transportation; SP: slow pyrolysis; CFGO: coal-fired generation offsetting; BT: biochar transportation; BS: biochar spreading; BFA: biochar field application; LC: the whole life cycle. ADP: abiotic depletion potential; AP: acidification potential; EP: eutrophication potential; GWP100: global warming potential at 100-year scale; ODP: ozone depletion potential; POCP: photochemical oxidation potential; HTP: human toxicity potential; TETP: terrestric ecotoxicity potential; FAETP: freshwater aquatic ecotoxicity potential; MAETP: marine aquatic ecotoxicity potential.

Table 3
Environmental impact values of the whole biochar life cycle.

Category	Annual value	Normalized value[1]
ADP	4.50×10^4/ $kgSb_e$	1.25×10^{-4}
AP	-7.25×10^8/ $kgSO_{2e}$	-3.03×10^{-3}
EP	-2.46×10^7/ $kgPO_4^{3-}{}_e$	-1.56×10^{-4}
GWP 100	-4.98×10^{11}/ $kgCO_{2e}$	-1.18×10^{-2}
ODP	2.82/ $kgR11_e$	1.24×10^{-8}
POCP	-7.33×10^7/ kgC_2H_{4e}	-1.99×10^{-3}
HTP	-7.90×10^{10}/ $kgDCB_e$	-3.06×10^{-2}
TETP	-1.86×10^9/ $kgDCB_e$	-1.70×10^{-3}
FAETP	-1.44×10^9/ $kgDCB_e$	-6.10×10^{-4}
MAETP	-3.18×10^{13}/ $kgDCB_e$	-1.63×10^{-1}

ADP: abiotic depletion potential; AP: acidification potential; EP: eutrophication potential; GWP100: global warming potential at 100-year scale; ODP: ozone depletion potential; POCP: photochemical oxidation potential; HTP: human toxicity potential; TETP: terrestric ecotoxicity potential; FAETP: freshwater aquatic ecotoxicity potential; MAETP: marine aquatic ecotoxicity potential.

[1] Normalized value $= \frac{\text{Characterization result}}{\text{normalized reference value}}$. It was used to compare different environmental impact categories of the biochar system. Normalization is usually based on the total amount of per capita emissions or per capita resource consumption in a specific range, such as the global, EU or a certain region. This paper used the CML2001-Jan.2016 method in GaBi, and the world's per capita emission in 2000 (CML2001 - Jan.2016, World, year 2000) was adopted as the normalized reference value.

energy utilization of pyrolytic oil/gas. In Fig. 2b, it can be clearly seen that offsetting the coal-fired generation had the greatest positive effects (negative values) on all environmental impacts. It indicated that highly efficient conversion of pyrolytic oil/gas into power played a vital role in mitigating environmental deterioration, while the biochar field application mainly contributed to reduced GWP100. Heating for pyrolysis no doubt caused the highest adverse impact on the environment (positive values in Fig. 2b), especially to the ADP and ODP, which accounted for about 95–100% of the total contributions among the different processes. Both crop residue collection and biochar spreading processes showed a minor adverse influence on these environmental issues. The impact of transportation was even lower. Therefore, reducing the energy consumption of pyrolysis process should be the predominant consideration, which determined the success or failure of biochar technology application. People have been recognized this point and novel thermo-chemical technologies are being investigated for biochar production such as solar pyrolysis [44], synergetic pyrolysis with cement kiln [45], and microwave pyrolysis [46,47], etc. Development of these technologies is promising to greatly reduce the energy consumption of pyrolysis in the future, and minimize their emissions and environmental impact.

In order to compare the different impacts of biochar on the environment, the characterization values were normalized using the CML2001-Jan.2016 method in GaBi, and the world's per capita emissions in 2000 (CML2001 - Jan.2016, World, year 2000) was adopted as the reference value [48]. The normalized data, listed in Table 3, were almost all negative for these issues except ADP and ODP, indicating that biochar application would benefit most environmental issues, while it might be disadvantageous for abiotic depletion potential and ozone depletion potential. Both ADP and ODP are mainly related to fossil fuel consumption by the slow pyrolysis process, biomass collection/transportation, and biochar spreading/transportation. Among these, slow pyrolysis accounted for almost 99% based on the values in Table S6. Table S6 also showed that the only positive mitigating effect on these two impact categories were ascribed to the produced bioenergy for offsetting coal-fired power generation, and biochar field application had no influence on them; while biochar amendment could bring obvious positive influences on all the other impact indexes except ADP and ODP. However, these adverse effects were relatively minor, indicated by the lower values (ADP: 1.25×10^{-4}; ODP: 1.24×10^{-8}), compared to the mitigating effects in the other impact categories, thus validating the feasibility of biochar implementation. The degree of benefit of the biochar system on different categories decreased in the following order: MAETP > HTP > GWP100 > AP > POCP > TETP > FAETP > EP > ADP and ODP. Han et al. [49] reached similar conclusion, where compared to open field burning, pyrolysis system showed higher values in some categories such as ADP, ODP, and HTP, while it had considerably lower GWP and EP impacting values.

It is worth noting that biochar implementation could remarkably decrease the human toxicity potential (HTP: -3.06×10^{-2}), marine aquatic ecotoxicity potential (MAETP: -1.63×10^{-1}), and acidification potential (AP: -3.03×10^{-3}) (Table 3). This could be ascribed both to the avoidance of open burning of biowastes, and the substitution of fossil fuels by pyrolytic oil/gas. These results highlighted again the significance of reducing fossil fuels burning (HTP: -2.69×10^2; MAETP: -1.33×10^5; AP: -3.13×10^1) (Table S6). In China, coal-fired power generation is currently the primary cause of many environmental issues, especially air pollution due to release of a large amount of soot, GHGs, sulfur oxides and nitrogen oxides into the atmosphere. Soot seriously affects the air quality and human health; GHGs aggravate the climate change, and sulfur/nitrogen release results in acid rain; all these emissions ultimately deteriorate the acidification and marine aquatic ecotoxicity. Therefore, replacing coal with renewable heat and power obtained as a by-product of biochar production will directly benefit the environment. In addition to renewable heat and power generation from the biochar system, for which other alternatives exist, biochar soil application benefits HTP, MAETP and AP with corresponding values of -6.24×10^{-4}, -7.27×10^{-1} and -3.53×10^{-4}, respectively (see Table S6). This inferred that as one of the agricultural benefits of biochar technology, reducing the amount of nitrogen and phosphate fertilizer applied in soil also contributed considerably to the reduced eutrophication, acidification, as well as human/marine ecotoxicity (Table 2) of biochar systems.

Monte-Carlo analysis (normal distribution, n = 5000) was performed to determine the sensitivity of the results to varying inputs. The variation range of all parameters were set in the biochar system to be ± 10%. As shown in Table S7, the standard deviations (σ) of all environmental impact categories are not over ± 10%, proving that the results are acceptable.

3.4. Sensitivity analysis and scenario calculation

To gain further insights into the relative influence of different parameters of the biochar system on its environmental impact, sensitivity analysis was conducted for the LCA results. The parameters are shown in Table S8, and their impacts on three important indexes, GWP100, ADP and HTP, were analyzed. The specific calculation process can be found in SI (Formula 1, Table S9), and Fig. 4 presents the obtained sensitivity coefficients resulting from variations of the parameters by ± 10%. Results showed that sensitivities of GWP100, ADP and HTP to all 15 parameters varied greatly, and significant differences could also be observed among these three environmental impacts.

Almost all key parameters of the biochar system had a direct influence on the GWP100, among them, biochar yield, carbon or stable carbon content in biochar, conversion efficiencies of pyrolytic oil/gas to power are the most influential (Fig. 4a). In contrast, GWP100 showed a very low sensitivity to transportation and fertilizer reduction. This indicated that developing novel slow pyrolysis technologies to achieve high biochar yield and improving bio-energy conversion efficiency are critical in further promoting the environmental performance of biochar technology. The parameters that had the greatest impact on ADP included power conversion efficiencies of pyrolytic oil/gas and energy consumption of pyrolysis plant. The sensitivity of ADP to power generation efficiency was twice higher compared to pyrolysis system energy demand (Fig. 4b). Electricity conversion efficiencies of pyrolytic oil/gas also had great impacts on HTP, with HTP being much more sensitive to these two processes than ADP, while following the same trend. The fluctuations of energy consumption by slow pyrolysis and crop residue collection also had notable effects on HTP, and the higher the energy consumption, the larger the HTP value was (Fig. 4c). The parameters related to the carbon storage of biochar in the soil (No. 9–15) all showed limited effects on both ADP and HTP.

To further conduct a quantitative analysis, a series of different hypothetic values for bioenergy converting efficiencies and biochar yield were set to simulate different application scenarios. The calculated values were presented in Table S10-S12, and results can be seen in Fig. S1. Rising of bioenergy converting efficiencies from 25% to 40% could induce an increase in GWP100 from −0.42 to −0.54 Pg CO_{2e} (bio-oil) and −0.45 to −0.50 Pg CO_{2e} (pyrolysis gas) (Fig. S1a); Biochar yield increasing from 25% to 40% would enable GWP100 to be improved from-0.32 to −0.58 Pg CO_{2e}. Our previous studies explored the biochar-engineering strategies for producing composite biochar via doping inorganic minerals [50,51] such as triple superphosphate, bone meal, magnesium chloride [52,53], etc. to increase the carbon retention by around 25% in biochar during pyrolysis [54]. Similarly, increase on efficiency of bio-oil based power generation efficiency from 25% to 40% led to ADP decrease from 4.40×10^4 to 4.32×10^4 kg Sb_e (Fig. S1b), and HTP changing from -5.25×10^{10} to -9.77×10^{10} kg DCB_e (Fig. S1c). The increase in efficiencies of pyrolysis gas conversion from 25% to 40% presented only a small alteration to ADP, while it decreased HTP remarkably from -6.47×10^{10} to -8.26×10^{10} kg DCB_e, indicating the importance of effective reclamation of exhaust pyrolytic gas.

3.5. Economic feasibility analysis

Economic feasibility analysis was performed from the prospective of biochar producer. The LCC inventory of the biochar system used in this study is shown in Table 4. In this section, we still used 1-t crop residues as the FU, and the evaluated time span was 1 year. All expenditure and income estimates were shown as U.S. Dollars. Land purchase or rental costs were not considered since the land cost for constructing a pyrolysis

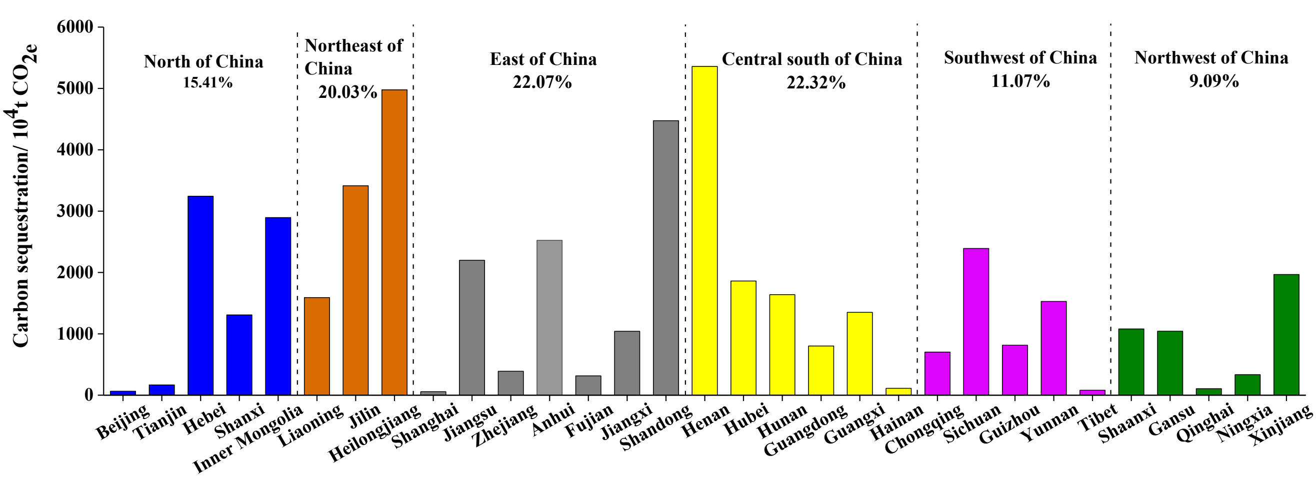

Fig. 4. Sensitivity analysis of GWP100 (a), ADP (b) and HTP (c). GWP100: global warming potential at 100-year scale; ADP: abiotic depletion potential; HTP: human toxicity potential. Parameter 1: electricity consumption in crop residue collection, kWh; Parameter 2: diesel for crop residue transportation, kg; Parameter 3: pyrolysis plant electricity demand, kWh; Parameter 4: biochar yield, %; Parameter 5: diesel for biochar transportation, kg; Parameter 6: electricity consumption in biochar spreading, kWh; Parameter 7: bio-oil electricity conversion efficiency, %; Parameter 8: pyrolysis gas electricity conversion efficiency, %; Parameter 9: carbon content in biochar, %; Parameter 10: stable carbon content in biochar, %; Parameter 11: biochar spreading ratio, t·ha-1; Parameter 12: crop yield increase, %; Parameter 13: fertilizer reduction (N/P/K), %; Parameter 14: N2O emission reduction, %; Parameter 15: SOC mineralization reduction, %.

plant would be highly varied and negligible compared to the other costs. The cost of crop residue itself was also very low and many farmers burn the crop residue in the field to avoid hindering next round of planting. The cost of equipment and materials was referred to the values provided by Yang et al. [55] in his research about running a pyrolysis plant. The annual treatment scale of the plant was assumed as 2.56×10^3 t, with 13 part-time employees and an operational life of 20 years. The employees' salary was on average 8.40 USD (U.S. Dollar) for one person per day [56]. Wages are not very high because such factories are usually located in underdeveloped areas or rural areas, and rely to a large extent on relatively low-skilled labor. In addition, due to the high level of automation, these employees only need to work part-time. The life cycle expenditure mainly included the costs of material, equipment and labor. The income came from the sale of biochar and pyrolytic oil/gas. As a result, the biochar implementation derived from 1 t of crop residues would be profitable at 46.92 USD (Table 4). Considering the crop residue yield in China, the annual profit of the biochar system could reach 25.38 billion USD with total deployment on 3.66×10^6 ha of land processing 5.41×10^8 t of agricultural residues. Therefore, the biochar implementation was economically feasible. As the carbon market grows and matures, the economic benefits from the carbon abatement will also be an important part in biochar system, and biochar production will be even more economical and competitive in the future.

Table 4
Life cycle cost based on 1 t of crop residues.

Item	Price	Amount	Benefit /USD[1]
Biomass collection, storage and transportation	37.02 USD [1]	/	−37.02
Equipment in the pyrolysis plant	2.20 USD [2]	/	−2.20
Labor	15.57 USD [3]	/	−15.57
Electricity (Coal-fired generation)	0.07 USD/kWh [4]	326.84 kWh	−22.88
Diesel	1.13 USD/kg [5]	2.00 kg	−2.26
Brick	0.10 USD/kg [6]	15.19 kg	−1.52
Concrete	39.90 USD/m^3 [7]	0.0046 m^3	−0.18
Steel	0.70 USD/kg [6]	0.69 kg	−0.48
Tap water	0.52 USD/t [8]	272040 kg	−141.46
Biochar	0.63 USD/kg [6]	343.53 kg	+216.42
Bio-oil	0.10 USD/kg [6]	293.93 kg	+29.39
Pyrolysis gas	0.07 USD/kg [6]	352.54 kg	+24.68
Total			**+46.92**

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[1] '−'indicates expenditure, and '+' indicates income.

4. Conclusions

Pyrolytic conversion of biomass resources into biochar and biofuels is a promising strategy for effective utility of crop residue to improve environment and reduce atmospheric greenhouse gas concentrations. In this study, a comprehensive life cycle analysis identified the following critical parameters of the biochar system for enhancing its carbon sequestration potential and environmental benefits: 1) energy consumption of pyrolysis process, 2) bio-oil/gas utilization efficiency for offsetting coal-fired power generation, and 3) biochar-induced agricultural amelioration. Reducing pyrolysis energy consumption, improving bio-oil/gas converting electricity efficiency and soil sequestration of biochar-carbon played vital roles in global warming mitigation. Optimizing the former two factors also can be related closely to ameliorating almost all environmental issues, especially to abating the release of human-toxic substances, protecting marine aquatic biodiversity, and relieving surface soil and water acidification. Based on the available latest data on crop residue in China, the annal carbon sequestration potential of a country-level biochar system was estimated at 0.50 Pg CO_{2e}, accounting for 4.50% of the total annual national greenhouse gas emissions. The central south and east of China were the focus districts for achieving carbon sequestration. The methodology, parameters, and results of this study could be utilized for analyzing the applicability of biochar system in any other country, and this study could guide large-scale implementation of biochar, as well as informing further development of relevant technologies and systems.

CRediT authorship contribution statement

Qiushuang Yang: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Software, Validation, Writing - original draft. **Ondřej Mašek:** Validation, Visualization, Writing - review & editing. **Ling Zhao:** Resources, Project administration, Funding acquisition, Methodology, Supervision, Validation, Writing - review & editing. **Hongyan Nan:** Investigation, Methodology, Software. **Shitong Yu:** Conceptualization, Methodology, Software. **Jianxiang Yin:** Conceptualization, Methodology. **Zhaopeng Li:** Investigation, Formal analysis. **Xinde Cao:** .

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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Appendix A. Supplementary data

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