Carbon-negative transition by utilizing overlooked carbon in waste landfills

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*Correspondence: cbzhou@rcees.ac.cn

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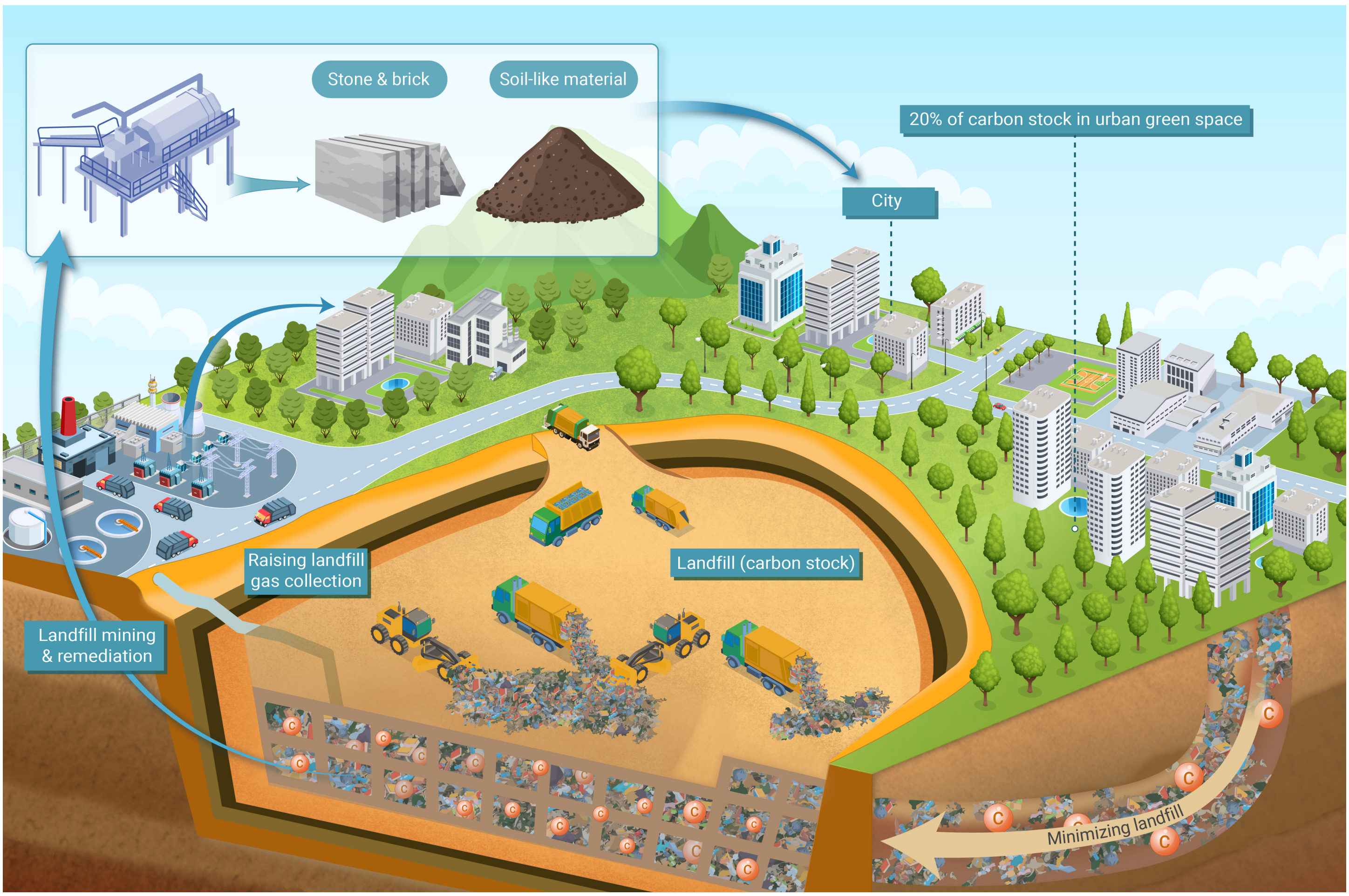
GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- A new model offers insights into landfill carbon stock accounting.
- The carbon stock from landfills at the prefecture-level in China is estimated from 2001 to 2020.
- The carbon stock in landfills holds substantial resource potential and environmental impact.
- Landfill mining and in-situ restoration could transform landfills into carbon-negative sectors.

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[1]Stake Key Laboratory of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China
[2]College of Resources and Environment, University of Chinese Academy of Sciences, Beijing 101408, China
[3]Santa Fe Institute, Santa Fe 87501, USA
[4]Department of Earth System Science, Stanford University, Stanford 94305, USA
[5]Department of Environmental Studies, New York University, New York 10003, USA
[6]The Bartlett School of Sustainable Construction, University College London, London WC1E 6BT, UK
[7]School of Economics, Beijing Institute of Technology, Beijing 100081, China
*Correspondence: cbzhou@rcees.ac.cn

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Landfills play a crucial role in urban climate solutions, as the decomposition of their "hidden" carbon stock contributes to 8.8% of global methane emissions. While controlling landfill gas emissions is the most commonly used intervention, a systematic approach to manage the carbon cycle in landfills remains elusive. In this study, we developed a quantitative solid-water-gas coupling model to estimate the carbon stock in landfills across 346 cities in China. Our findings reveal a standing landfill carbon stock 506.3 ± 4.2 Tg, which could potentially substitute for 20% of soil organic carbon in green spaces and 1 year of residential electricity consumption of cities. Our scenario analyses show that by implementing a life-cycle package of interventions (incl., input minimization, stock utilization, and leakage reduction), the total carbon stock in landfills could be reduced to 230.1 Tg with a negative annual carbon emission (-57.1 Tg CO_2 eq/year) reached by 2030. These interventions could cumulativly cut greenhouse gas (GHG) by 753.3 Tg, representing 62.2% of the landfill-related GHG emissions and 2.0% of China's carbon debt towards the 1.5°C warming targets. Landfill mining contributes 52.3% of these reductions, while in-situ aerobic restoration accounts for 14.4%, positioning landfills as a potential carbon-negative sector that can drive cities towards carbon-neutrality.

INTRODUCTION

It was estimated that nearly 70% of municipal solid waste (MSW) has been stored away in landfills, one of the fundamental infrastructures of cities.[1] As a result, a vast amount of post-used carbon has been stored in landfills for a long period, making it a vital carbon stock in the city. Previous studies indicate that landfills potentially account for about 11% ~ 12% of the urban carbon stock,[2-3] however, the restoration of these carbon stocks is often neglected because landfills are out of sight for urban citizens. The organic carbons in landfills are in two major forms: (i) organic carbon that can naturally degrade, such as textile, wood, and food waste (hereafter degradable organic matter, or DOC), (ii) organic carbon that would not naturally degrade at a meaningful speed, such as plastics (hereafter fossil organic carbon, or FOC).[4]

These organic carbon in landfills undergoes dynamic transformations between solid (stored waste), liquid (leachate), and gaseous forms (methane and carbon dioxide) through biochemical degradation processes, which is distinct from other comparative stable carbon stocks in cities, such as urban soil and buildings.[1,5-6] Most notably, the decomposition of such "hidden" carbon stocks in urban landfills was the third-largest source of anthropogenic methane emissions in the world, accounting for 8.8% of global methane emissions in 2021,[7-8] while remote sensing analysis demonstrated that landfills are the dominated source of methane emissions in cities.[9-10]COP27 highlighted tackling landfill-source methane emissions is the key to limiting global warming to 1.5°C.[11]

Additionally, fire accidents in landfills are occurring at a higher frequency in part because of global warming, and the consequent burning of plastic waste can generate extra greenhouse gas (GHG) emissions.[12] If the current waste management system remains unchanged, the amount of MSW buried into landfills and its methane emissions will double by 2050.[13] Furthermore, stored waste in landfills can result in a number of negative local impacts. Mismanaged leachate (generated mainly from food waste and rainwater) results in water body hypoxia and/or eutrophication, threatening the safety of both water and food supplies.[14] Evidences show that plastic waste could break down into microplastics in landfills and leak with leachate.[15]

On the other hand, carbon stocks in landfills also have great potential to be recycled and utilized as secondary materials or renewable energy. It has been demonstrated that humus soil and waste-derived fuel can be obtained from landfill mining.[16-17] Similar to other countries in the world, landfilling was also the most fundamental method for MSW disposal in China in recent decades; therefore, there is a large amount of stored waste in landfills. Many of these landfills, once located in unpopulated areas, are now incorporated into urban built-up area due to the rapid urbanization. These in-city landfills represent a significant resource pool to tap into, while at the same time posing a threat to public health if not managed well.[18-19]

Current research and policies mainly focus on phasing out landfilling, an undesirable sector in the city, for mitigating its climate-change impact.[20] For instance, the European Waste Framework Directive have been launched to reduce landfilled waste, while countries such as Austria, Germany, and the Netherlands have reached 'virtual elimination of landfilling.[21] China has also explored a zero-landfilling policy in the developed cities since 2020. Admittedly phasing out landfilling would benefit GHG reduction, a large number of existing landfills still continuously emit GHG over decades.[22] How and to what extent landfills (both existing and newly-built ones) can mitigate GHG emissions are also crucial for cities' strategies to fight climate-change. In addition to minimizing landfilling rate, interventions for reducing GHG emissions from landfills may also include increasing the rate of methane collection[23] and implementing landfill remediation.[24] Particularly, ex-situ restoration (e.g., landfill mining) has been implemented by many countries in the world, while it enables utilizing organic carbon stocks in landfills and then contributes to carbon mitigation.[25-26]

Understanding the characteristics, flows, and fates of the organic carbon stock in landfills provides a scientific basis for formulating policies to mitigate their GHG emissions.[23] Several studies have examined the carbon stocks and flows in specific landfills by field-sampling and laboratory analysis,[27-28] besides, limited research has presented national-scale information on the carbon flow in landfills (e.g., the US and China).[2-3] However, current methods fail to fully capture the relationship between solid, liquid, and gas phase transformations in landfilled waste,[29] limiting our ability to comprehend the dynamics of landfilled carbon stock. As a result, comprehensive datasets with higher spatial and temporal resolution (e.g., localized waste compositions and climatic parameters) are inadequate for formulating more targeted mitigation countermeasures for urban landfills.

Here, we present a bottom-up accounting approach to analyze the quantity and composition profiles of carbon stocks in landfills, with an quantitative solid-water-gas coupling transformation model, which is innovative for evaluating the climate-change impact of the landfilling sector. We analyzed the temporal trends and spatial patterns of organic carbon stock, flow, and shift

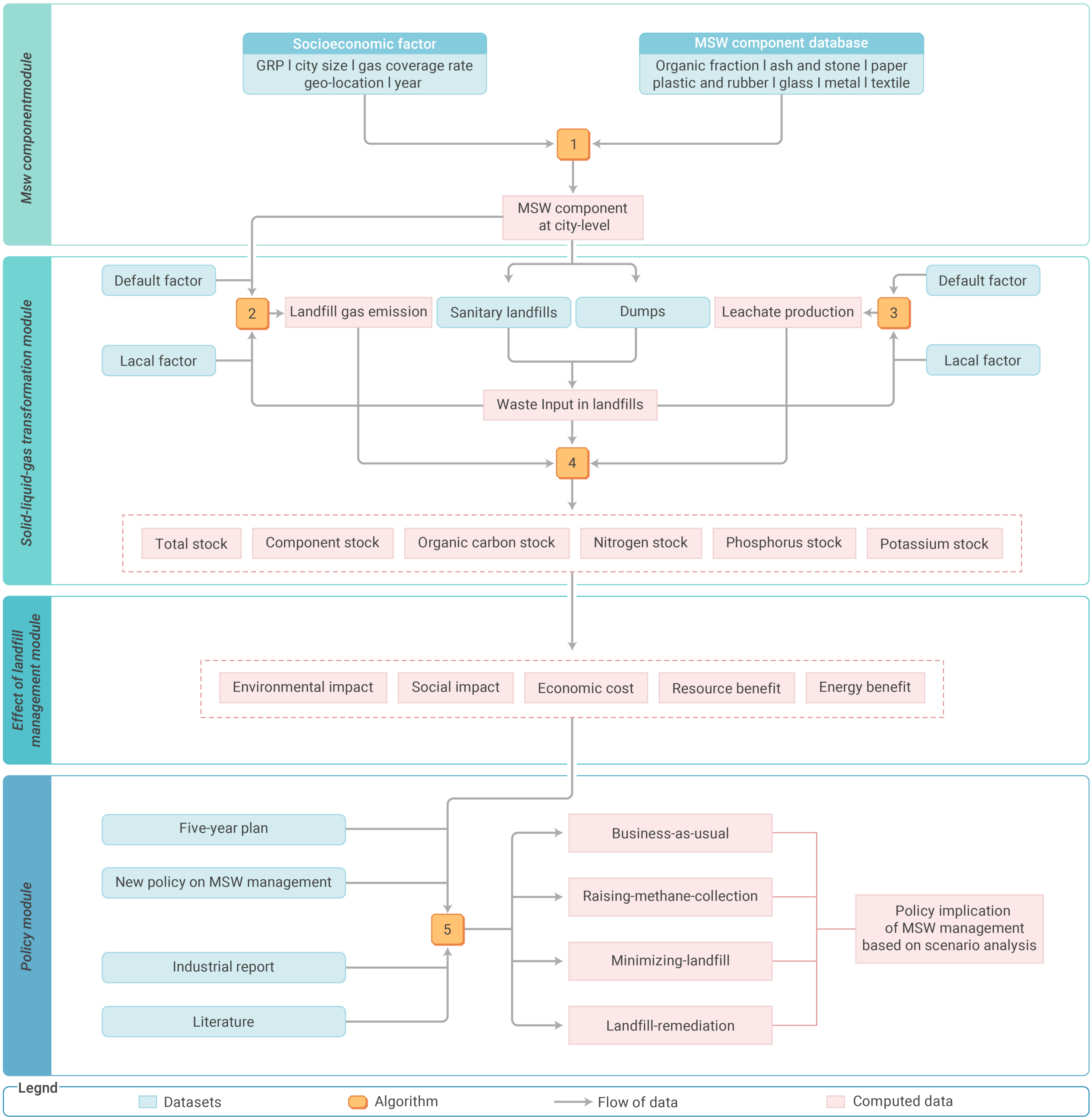

Figure 1. Framework of the landfill organic carbon stock estimation and management.

in urban landfills across 346 cities in China from 2001 to 2020. The significance of landfill organic carbon pool to urban carbon cycle was revealed from the perspectives of resource utilization, energy recovery, and GHG emissions. Moreover, future scenarios from 2020 to 2030 and a life-cycle package of interventions was put forward to mitigate GHG emissions. This work contributes new models and data for understanding the carbon cycle in urban systems, and provides a scientific basis for a low-carbon transition in the waste sector.

MATERIALS AND METHODS

Quantifying landfill organic carbon stock follows four module: (1) the MSW component module, (2) the solid-liquid-gas transformation module, (3) the effect of landfill management module, and (4) the policy module (Figure 1). The first two modules are applied for quantifying the organic carbon stocks, while the environmental impact and benefits of different landfill management method are analyzed by the third module. The last module is used for setting intervention scenarios and analyzing the implication of different policies.

MSW component module

To fill the knowledge gap of inadequate and incomplete MSW component datasets in cities, in our previous work, we collected 503 pieces of MSW component data covering 135 cities from literature.[30-31] A propagation neural network model was used to find the correlations between socioeconomic factors and MSW component. Here, this model was applied to forecast the

MSW component of China from 2001-2030.

Estimate landfill gas and leachate production. Methane emissions are calculated by using Equation (1) based on the first-order decomposition model recommended by IPCC.

$$E_{CH4,r,p} = \sum\nolimits_{T} (e^{-(T-1)k_{r,p}} - e^{-Tk_{r,p}}) \times MSW_{L,r,p} \times MCF \times DOC_{r,p} \times DOC_{f,r,p} \times F \times 16/12 \times (1-OX) \quad (1)$$

where, $E_{CH4,r,p}$ represents the amount of methane production for each city ($Tg{\cdot}a^{-1}$), T is the time when the input MSW is stored in landfills. $k_{r,p}$ is the constant of methane production rate for each city, calculated by Equation (S1), $MSW_{L,r,p}$ refers to the amount of MSW disposed of in sanitary landfills and dumps (Tg), which is derived from China Urban-Rural Construction Statistical Yearbook,[32] MCF is methane correction factor, which is 1.0 for sanitary landfills and 0.61 for dumps,[33] $DOC_{r,p}$ represents the biodegradable organic carbon content in MSW for each city, calculated by Equation (S2), $DOC_{f,r,p}$ is the fraction of DOC that can be oxidized for each city, calculated by Equation (S3), F represents the volume fraction of methane in landfill gas, set to 0.5,[34] 16/12 is the accounting factor that converts the mass of carbon to the mass of methane, label p and r represent the city and geographic region (Table S1), OX is the oxidation factor, shown in Table S2.

Equation (2) was used to calculate the amount of leachate produced from landfill for each city.

$$L_{r,p} = MSW_{L,r,p} \times F_r \quad (2)$$

where, $L_{r,p}$ is the amount of leachate produced by landfills, 10000 $m^3{\cdot}a^{-1}$, F_r is the amount of leachate generated per ton of MSW entering into the landfills, $m^3{\cdot}t^{-1}$, shown in Table S3, label p and r represent the city and geographic region.

Carbon storage in landfills. Disposed amounts of MSW in sanitary landfills and dumps are obtained from China Urban-Rural Construction Statistical Yearbook, 2001 to 2020.[32] Carbon leakage from landfills is mainly through leachate and landfill gas. Therefore, the carbon stock of landfill is calculated by the material flow analysis model, i.e., the amount of carbon input into landfills minus the amount of carbon output in the form of landfill gas and leachate, by using Equation (3). The amounts of carbon in landfill gas and leachate were calculated by Equation (4) and (5), while the input of carbon in landfills was obtained by using Equation (6).

$$C_{Fin,r,p} = C_{input,r,p} - C_{E,r,p} - C_{L,r,p} \quad (3)$$

where, $C_{Fin,r,p}$ is the annual carbon stock of landfills (dumps and sanitary landfills) for each city, $Tg{\cdot}a^{-1}$, $C_{input,r,p}$ refers to the carbon storage of MSW that enters into landfills every year for each city, $Tg{\cdot}a^{-1}$. $C_{E,r,p}$ and $C_{L,r,p}$ are the amount of carbon outflow in the form of landfill gas and leachate, $Tg{\cdot}a^{-1}$.

$$C_{E,r,p} = E_{CH4,r,p} \times 12/16 \times 2 \quad (4)$$

$$C_{L,r,p} = L_{r,p} \times \frac{COD + 0.0582}{3} / 100000000 \quad (5)$$

$$C_{input,r,p} = MSW_{L,r,p} \times \sum\nolimits_{i=1}^{n=5} C_{i,r,p} \times c_i \quad (6)$$

where, $C_{i,r,p}$ is the MSW component, including organic fraction, plastic and rubber, paper, wood, and textile for each city, which are derived from our previous work, Ma et al. (2020),[30] c_i is the carbon content for each MSW component. COD is the concentration of chemical oxygen demand in landfill leachate, m^3 ton^{-1}, detailed parameters can be found in Tables S4-S8.

Benefits and environmental impact module

We used three metrics, incl., utilization potential of soil-like material (UPSL), electricity generation potential (EGP), and contribution to methane emissions (CME), respectively, to assess the resource utilization potential, renewable energy potential, and environmental impact of organic carbon stock in landfills. These metrics were calculated for each city and then compiled into seven regions (Table S1).

Contribution to methane emissions. The solid waste sector is the third largest contributor to global non-CO_2 GHG emissions.[16,35] The main GHG emissions in landfills are methane and nitrous oxide. However, While the carbon storage of landfills is the focus in this study, there is limited discussion regarding nitrous oxide. Therefore, we used the proportion of methane emissions from landfills to total methane emissions in a specific city to measure the environmental impact of landfill organic carbon stock, as shown in Equation (7).

$$CME_{r,p} = CCH_{4,r,p} / CH_{4anthro,r,p} \quad (7)$$

where, $CME_{r,p}$ is the contribution to methane emissions of landfills for each city, $CCH_{4,r,p}$ is the cumulative methane emissions from landfills for each city from 2001 to 2018. $CH_{4anthro,r,p}$ is the total methane emissions for each city from 2001 to 2018, which is derived from Emissions Database for Global Atmospheric Research (EDGAR) Version 6.0 with 6-s resolution.[36]

Electricity generation potential. To characterize the waste incineration potential for each city, we used the ratio of the converted electricity through incineration of the organic carbon in combustible components in landfills to electricity consumption of urban residents in 2020 by using Equation (8).

$$EGP_{r,p} = (Pla_{r,p} \times HV_{pla} + W_{r,p} \times HV_W + T_{r,p} \times HV_T)/3600/Ele_{r,p} \times EGE_r \quad (8)$$

where, $EGP_{r,p}$ refers to electricity generation potential of the stocks of plastics, wood, and textile in landfills for each city, $Pla_{r,p}$, $W_{r,p}$, $T_{r,p}$ are the stocks of plastics, wood, and textile in landfills for each city, kg, HV_{pla}, HV_W, HV_T are calorific values of plastics, wood and textile, $kJ{\cdot}kg^{-1}$, which are derived from Nie (2013),[37] $Ele_{r,p}$ refers to electricity consumption of urban residents in 2020 for each city, kWh, EGE_r refers to electricity generation efficiency for incineration of region r, shown in Table S9.

Utilization potential of soil-like material. Recovery of organic materials from MSW is crucial for urban nutrient cycling and organic carbon resource conservation.[38] The proportion of organic carbon storage in soil-like material of landfills to that in 100-cm soil of urban green space was used as a metric to evaluate resource potential of material stocks in landfills, calculated by Equation (9)-(11).

$$UPSL_{r,p} = S_{TOC,r,p} / S_{GREEN,r,p} \quad (9)$$

$$S_{GREEN,r,p} = C_{TOC,r,p} * Area_{GREEN,r,p} * depth * SD_{r,p} / 1000000 \quad (10)$$

$$SD_{r,p} = \sum_{i=1}^{n=5} layer_i * SD_{i,r,p} / 100 \quad (11)$$

where, $UPSL_{r,p}$ is the proportion of the organic carbon stock in soil-like material to that in urban green space (100 cm) for each city, $S_{TOC,r,p}$ is the organic carbon stock in soil-like material of landfills for each city, ton, $S_{GREEN,r,p}$ is the organic carbon stock in 100 cm soil of the built-up area for each city, ton, $C_{TOC,r,p}$ is the average organic carbon contain in soil for each city, $g{\cdot}kg^{-1}$, derived from the soil organic matter content map in China with 1-kilometer grid from 2010-2018,[39] $Area_{GREEN,r,p}$ is the built-up area for each city, m^2, which is obtained from the China Urban-Rural Construction Statistical Yearbook,[32] depth is the soil depth, taken 100 cm here, $SD_{r,p}$ is the soil bulk density of 100cm for each city, $kg{\cdot}(m^3)^{-1}$, $SD_{i,r,p}$ is the soil bulk density at depth i in, incl., 0 - 5 cm, 5 - 15 cm, 15 - 30 cm, 30 - 60 cm and 60 -100 cm, derived from the global soil bulk density dataset with 1-kilometer grid.[40]

Policy module and scenario settings

We set up four scenarios based on the current policies of MSW management implemented and encouraged in China. The four scenarios are the business-as-usual, the raising-methane-collection, the minimizing-landfill, and the landfill-remediation scenarios. The base year for the scenario analysis is 2020, and the analysis spans from 2021 to 2030, the year for achieving the global methane reduction goals.[11] The differences between these four scenarios and detailed parameters of the four scenarios are shown in Tables S10-S14.

Business-as-usual scenario. It describes that MSW management in China will follow the same policies applied in the year 2020. This scenario provides a baseline scenario for how landfill organic carbon stocks and their

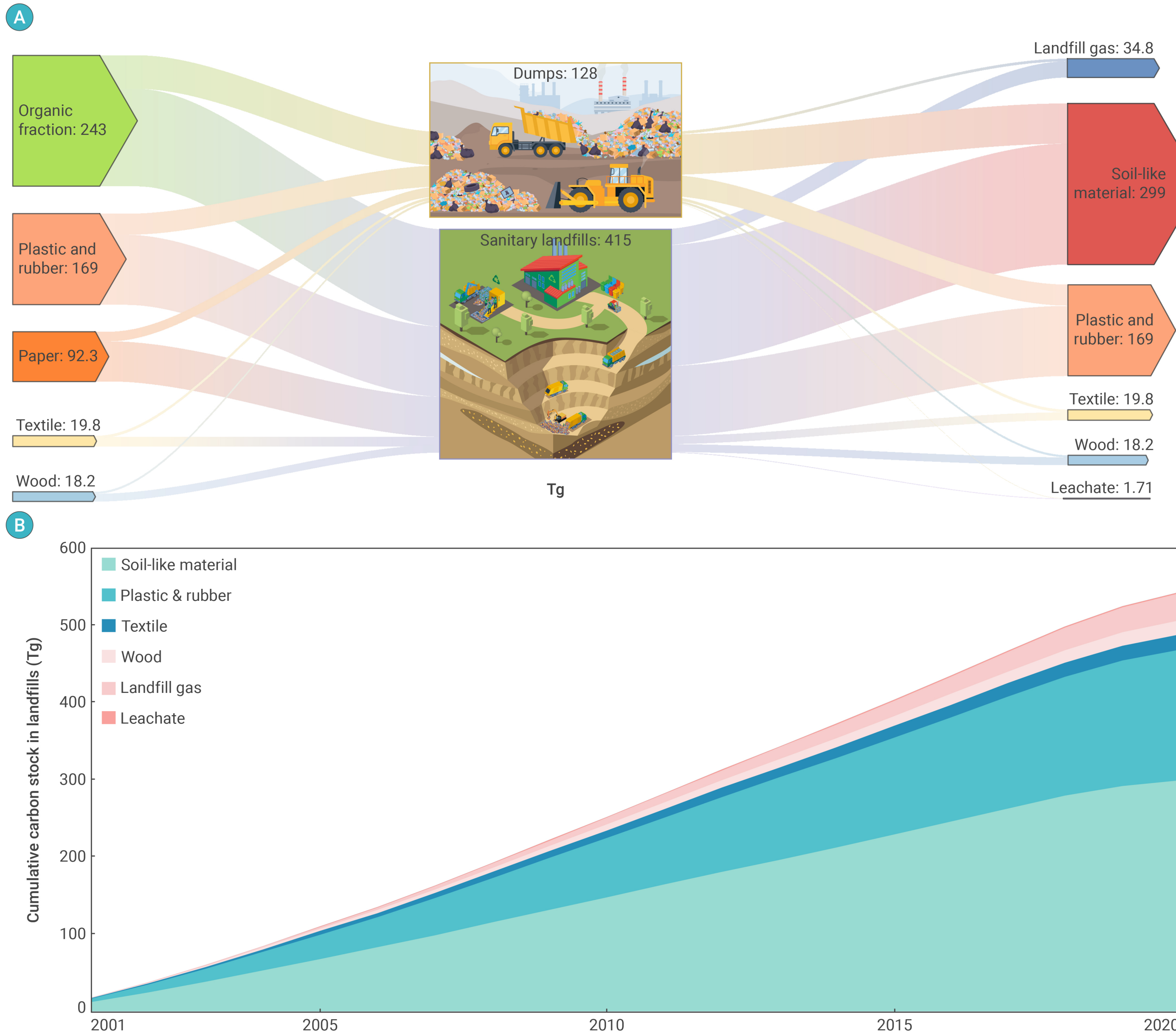

Figure 2. Cumulative organic carbon stocks in landfills in China (2001-2020) (A) The material flow of cumulative organic carbon stock from landfills, (B) temporal trends of cumulative organic carbon stocks in landfills including sanitary landfills and dumps. Note: soil-like material, plastic and rubber, textile and wood are the carrier of organic carbon stock in landfills, landfill gas and leachate refer to the forms of organic carbon transferred out of landfills.

associated GHG emissions change over the next decade. The forecast of the amount of MSW collected and transported was from Chhay et al. (2018),[41] while the changes of composition of MSW in China from 2021 to 2030 were obtained by Ma et al. (2020).[30] No additional intervention strategies will be imposed. Detailed parameters of the business-as-usual scenario are shown in Table S11.

Raising-methane-collectionscenario. It describes that the methane collection rate will be increased in MSW landfills in China significantly compared with business-as-usual to reduce methane emissions. The methane collection rate will increase from 24% in 2020 to 40% in 2030, according to Cai et al. (2018).[16] Detailed parameters of the raising-methane-collection scenario are shown in Table S12.

Minimizing-landfill scenario. It considers the new policies and recommendations leading to the changes of MSW treatment and disposal methods in China before 2020, including zero-waste cities, compulsory MSW source separation, and the 14th Five-Year Plan,[42] in addition to the increased methane collection rate as outlined in the raising-methane-collection scenario. These policies and plans include detailed targets for developing waste treatment facilities, for instance, the incineration rate for MSW reaches 65% by 2025 and aims for zero dumping. We used linear interpolation to extrapolate these data to 2030. The proportion of MSW dumping remains at 0%, the proportion of sanitary landfills reduces to 12%, and the proportion of incineration increases to 68% by 2030. Besides, a 20% diverting rate of food waste is used as the goal for all cities by 2030, which is the current rate in developed mega-cities (Beijing and Shanghai) in China. Besides, we assume that 76.1% of the separated food waste is anaerobically digested and the remaining food waste is composted.[43] The GHG mitigation in this scenario is the sum of the reduction in methane emissions from landfills and the rise in incineration and composting. Detailed parameters of the minimizing-landfill scenario are shown in Table S13.

Landfill-remediation scenario. It further considers implementing eco-remediation interventions gradually (incl., landfill mining and in-situ aerobic remediation) in China after 2020, in addition to all policies in the minimizing-landfill scenario. The total amount of material stock after landfill mining used for incineration depends on the remaining capacity for incineration in China, i.e., the operating capacity of incineration minus the MSW disposed of in

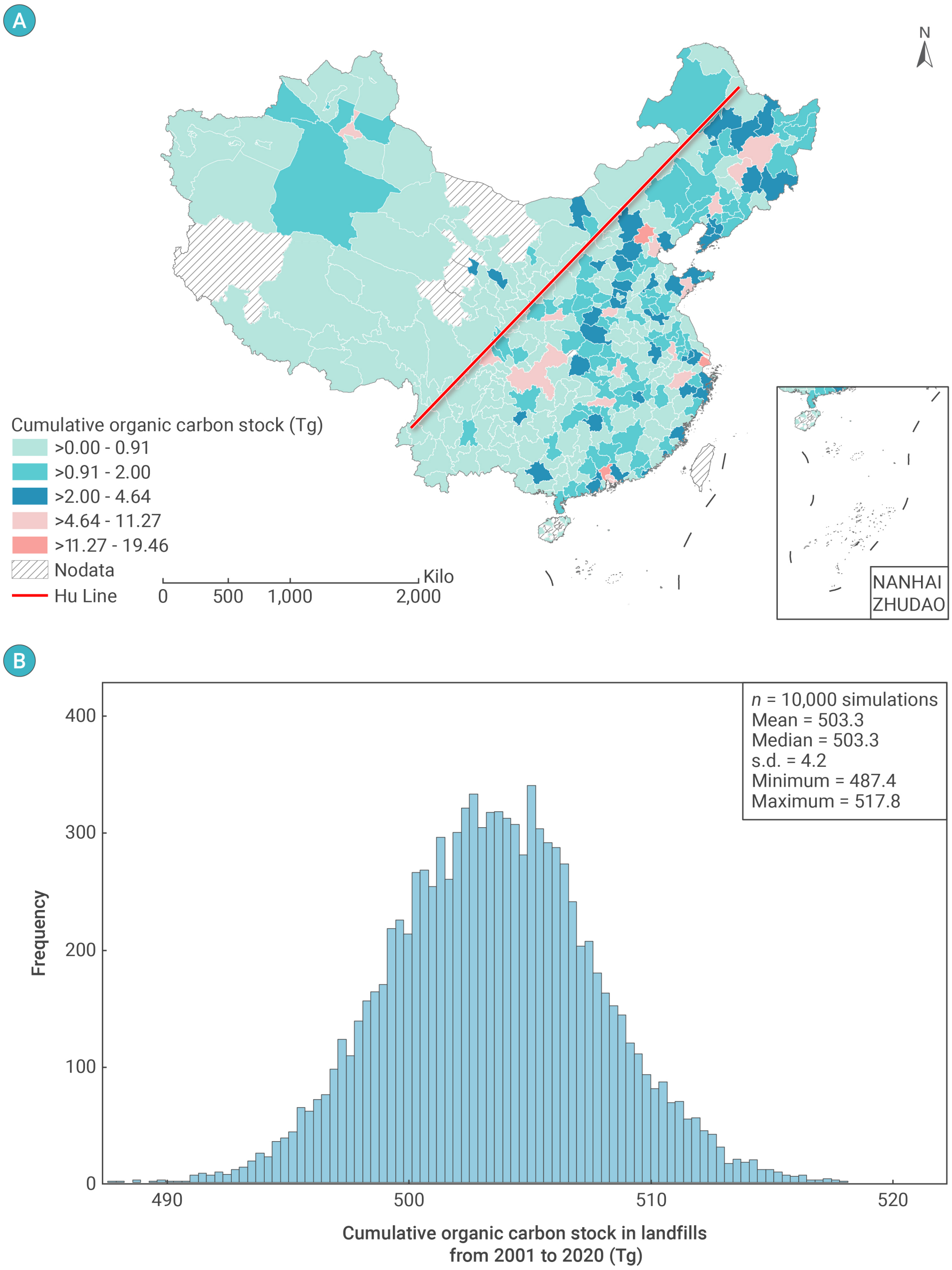

Figure 3. The spatial distribution of cumulative organic carbon stocks in landfills in China (A) cumulative organic carbon stocks in landfills at the city-level, (B) Monte Carlo analysis with 346 cities and 10,000 trials on possible variations of the national carbon stocks in landfills.

incineration plants, according to the 14^{th} Five-Year Plan of China. When landfill mining is adopted, we assume 50% of soil-like material and all plastics and rubber, textile and wood are incinerated to generate electricity, while the remaining 50% of the soil-like material is used for urban landscaping.[44] The organic carbon stocks in other post-closure landfills are assumed to be remediated by in-situ aerobic remediation. Detailed parameters of the landfill-remediation scenario are shown in Table S14. It should note that GHG emissions from the incineration of biodegradable organic carbon, such as organic fractions, paper, textile, and wood, are not included in our GHG inventory.[45] In addition to GHG emission reductions resulting from increased methane collection rates, the relative GHG emission reductions due to the substitution of urea fertilizer by organic matter or soil-like substances, and the replacement of coal-fired power generation with incineration power generation were also considered here. The economic feasibility of the landfill remediation scenarios was analyzed. The costs included in situ screening, aerobic remediation, closure of landfill, incineration; while the benefits included the output of electricity and nitrogen fertilizer. Detailed parameters used for the cost-benefit analysis are shown in Table S15.

Details of scenario settings and models used in this study are shown in supplementary information section 1.1 - 1.3. All the data and code used in this study are disclosed in supplementary data and supplementary code.

Uncertainty analysis. Monte Carlo simulation was used to quantify the uncertainty of carbon stock in landfills in China from 2001 to 2020.[30] It is assumed that the uncertainty of all variables in quantitative-based solid-water-gas coupling transformation model obeys a Gaussian distribution. If the uncertainty of a variable can be calculated or there is a reference value in the literature, the uncertainty was used. If a variable lacks uncertainty, 30% of the total range was used as a reference uncertainty. 10,000 Monte Carlo simulations were run for each variable. The results were brought into the MFA model to calculate the uncertainty of China's landfill carbon stock. Moreover, the 95% confidence interval of the results of 10,000 Monte Carlo simulations was considered as the uncertainty of the study. In addition, in order to analysis the uncertainty caused by the application of soil-like materials from landfill mining, we set that 30% or 70% of the soil-like materials from landfill mining are put into incineration plants, and the remaining are used for landscaping, as shown in supplementary information section 1.5.

RESULTS

Organic carbon stock in landfills over time and space

Over the past two decades, the cumulative input of organic carbon in China amounts to 542.8 Tg, of which 414.8 Tg (76%) went to sanitary landfills and 128.1 Tg (24%) to open dumps. Of these carbon inputs, 34.8 Tg (6.4%) were eventually released into the air in the form of landfill gas (methane and carbon dioxide), and 1.7 Tg (0.3%) was either transferred to wastewater treatment plants or lost to aquatic environments in the form of leachate (Figure 2A). The remaining standing stock (i.e., cumulative organic carbon stock in landfills) amounts to 506.3Tg (93.3% of input), mainly stored in four major forms: soil-like material (299.2 Tg, 55% of input), plastic and rubber (169.1Tg, 31% of input), textile (19.8 Tg, 3.6% of input), and wood (18.2 Tg, 3.4% of input) (Figures 2A & 3B).

For the organic carbon in landfill gas, annual emissions increased, from 0.2 Tg/year in 2001 to 3.1 Tg/year in 2020 (Figure S1a). However, this increasing trend is mostly driven by increasing gas emissions from sanitary landfills (0.13 Tg/year in 2001 to 2.84 Tg/year in 2020). In contrast, gas emissions from open dumps peaked around 2012 and decreased ever since, largely due to decreasing use of dumps over time.

For the organic carbon in leachate, total output peaked early in 2008 (0.13 Tg/year) and has been decreasing steadily since then. The overall decreasing trend is mostly driven by marked decrease of leachate from open dumps, again reflecting the decreasing use of dumps and the effectiveness of sanitary landfills in reducing leachate (Figure S1b).

For the remaining standing stock, the annual increment of carbon stock (open dumps and sanitary landfills combined) showed an almost linear increase, rising from 17.6 Tg/year in 2001 to 506.3 Tg/year in 2020, representing a 28.8 folds increase over 20 years (Figure 2B). However, when we

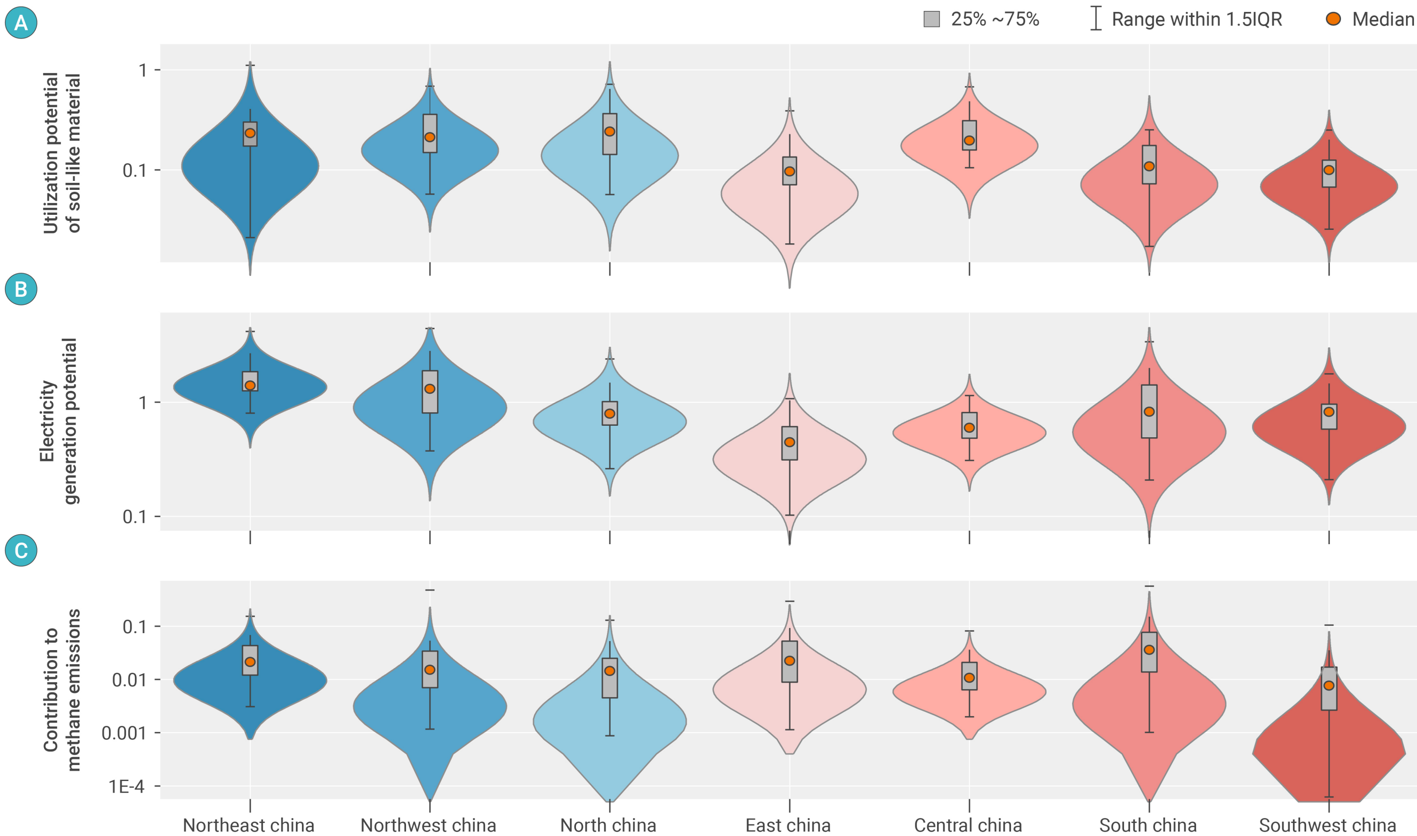

Figure 4. Benefits and environmental costs of organic carbon stock in landfills in each region of China (A) utilization potential of soil-like material (UPSL), (B) electricity generation potential (EGP), (C) contribution to methane emissions (CME).

teased apart the stocks in dumps and sanitary landfills (Figures S2a-b), we demonstrate a saturating rate of increase in open dumps. Correspondingly, we see a faster than linear increase in annual stock increment for sanitary landfills, driven by the increasing rate of urban waste production combined with the increasing popularity of sanitary landfills. Overall, the use of dumps to dispose of MSW has dramatically decreased over the past two decades in China, from 47.8% in 2001 to 0.3% in 2020.[32] Interestingly however, sanitary landfills and dumps do not differ significantly in their composition profiles: soil-like material (58.5% vs. 61.0%), plastic and rubber (33.9% vs. 31.8%), textile (3.8% vs. 4.1%), and wood (3.8% vs. 3.1%). This result indicates that, on average, the composition profile of the carbon stock is not so sensitive to the way of waste disposal.

The organic carbon stock in landfills reflects geographical distribution. In general, the spatial distribution of cumulative organic carbon stock in landfills was consistent with the law of Hu line, a watershed of high- and low-density population (and economy) of China (the red line shown in Figure Figure 3). That is, the total amount of the organic carbon stock of MSW landfills on the eastern side of the line (Northeast China, East China, South China, and Central China) was significantly higher than that on the western side (413.1 Tg vs. 93.2 Tg). Furthermore, the cumulative organic carbon stock of landfills in 31 provincial capital cities (9.0% of the prefecture-level cities in China) accounted for 36.2% of that in China over the last two decades (Figure 3A). Shanghai, Beijing, Guangzhou and Shenzhen, the four most developed and densely populated cities in China, accounted for 12.7% of the national total carbon stocks in landfills. The spatial distribution of cumulative organic carbon, as well as its main forms (i.e., soil-like material, plastic, textile, and wood), in sanitary landfills and dumps at city-level was shown in Figures S3-S5. The cumulative organic carbon in dumps increased gradually from south to north and from west to east of China. Cities in Northeast China have the highest proportion of organic carbon stock in dumps. Particularly, in northeastern cities such as Harbin, Changchun, and Jilin, the cumulative organic carbon stocks of dumps were 3.02 Tg, 2.13 Tg, and 1.36 Tg, accounting for 48.7%, 33.3%, and 65.2% of their total disposed organic carbon, respectively. A possible reason is that the sanitary landfilling rate in Northeast China was lower than that in other regions (see Figure S6), in addition, the biochemical degradation rate of organic carbon is much slower in frigid regions.

Utilization potential and environmental impacts of organic carbon stock in landfills

We devised three metrics to quantify the potential utility or harm of landfill-bound organic carbon: utilization potential for soil-like material, electricity generation potential, and contribution to methane emissions (Figure 4 & Figure S7).

First, soil-like material in landfills can be potentially used as organic soil to support urban green spaces. We thus derived the first metric, utilization potential for soil-like material (UPSL), by using the ratio of carbon stock in soil-like material to that in the topsoil (0 - 1 m) of urban green spaces. By evaluating this metric across all cities, we found that the average UPSL value of cities in China was 0.20 (the 25th - 75th percentiles: 0.09 - 0.24) (Figure S8a). This result suggests that MSW landfills are a vital organic carbon pool in cities. Among them, cities with larger UPSL were mainly located in Northeast China (0.39), North China (0.27), and Northwest China (0.26) (see Figures 4A & S9a). In North and Northeast China, the high UPSL values were mainly due to comparatively small urban green space coverage,[32] In Northwest China, it may due to the relatively low content of organic carbon in urban green space.[46]

Second, organic carbon in the form of combustible components (i.e., plastic, rubber, textile, and wood) has the potential to be converted into electricity to aid urban energy consumption in the future. We thus derived the second metric, electricity generation potential (EGP), by using the ratio of the converted electricity through incineration of the organic carbon in these combustible components to the annual electricity consumption of all urban residents (Figure 4B). The average EGP value of cities in China was 0.92 (the 25th-75th percentiles: 0.49 - 1.16). The EGP of three-quarters of cities in China was higher than 0.50, indicating that the stored waste in these cities can provide urban residents with electricity for at least half a year (Figure S8b). In

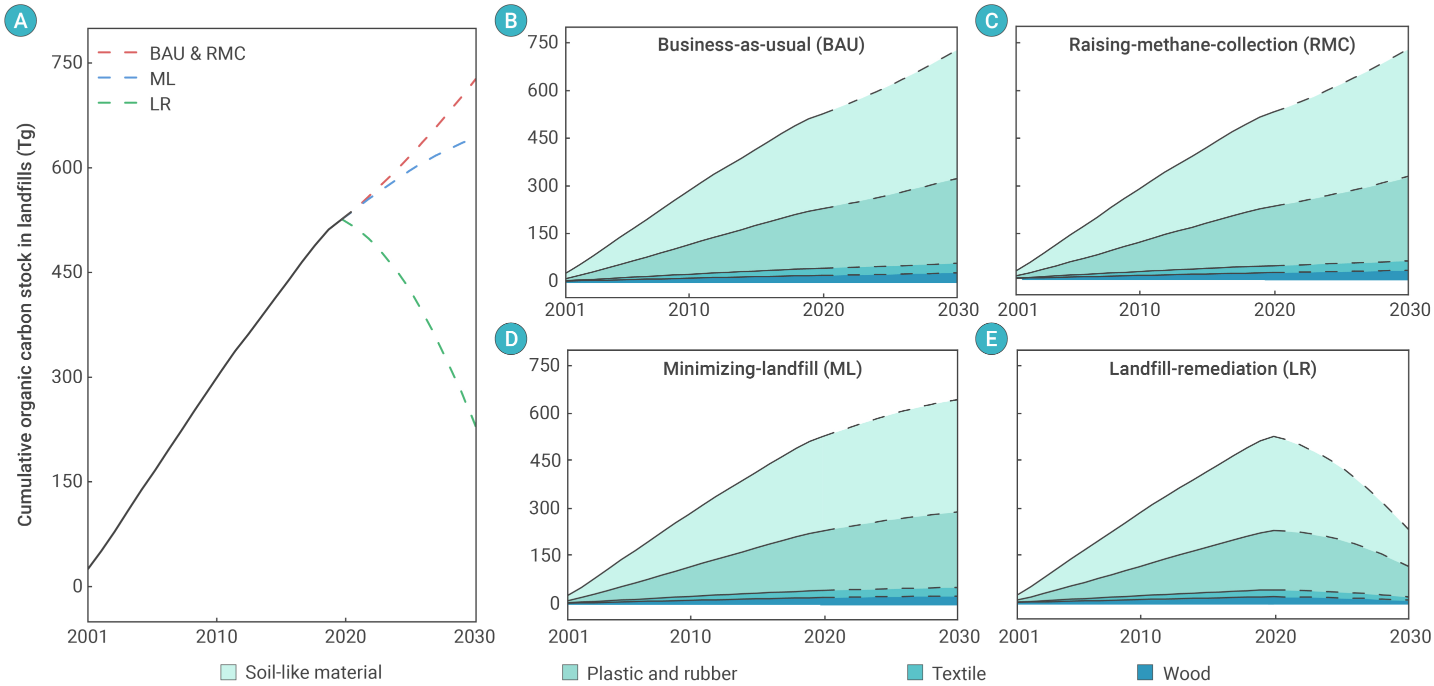

Figure 5. Scenario analysis of cumulative organic carbon stock from landfills in China from 2001 to 2030 (A) Overview of cumulative organic carbon stock, summarizing the results under four scenarios. (B)-(E) are cumulative organic carbon stock from landfills by carrier in the business-as-usual (BAU), raising-methane-collection (RMC), minimizing-landfill (ML), and landfill-remediation (LR) scenarios, respectively. Note: soil-like material, plastic and rubber, textile and wood are the carriers of organic carbon stock.

more than 81.53% of the cities, mainly distributed in Northeast China and Northwest China, the combustible organic carbon in landfills can even supply electricity for more than one year. For instance, the EGP values of Wuwei and Qitaihe were 4.44 and 4.17, respectively (see Figure S9b).

Third, methane emissions from landfills can negatively impact our environment. We derived a third metric, the contribution to methane emissions (CME), as the ratio of landfill-derived methane emissions to total anthropogenic methane emissions for each city (Figure 4C). The average CME value of cities in China was 0.039 (the 25th-75th percentiles: 0.007-0.044) (Figure S8c). Cities with high CME were mainly located in South China, East China, and Southwest China (Figure 4C). For instance, Shenzhen, Haikou, Guangzhou, and Zhuhai were the cities with the largest CME, which were 0.69, 0.57, 0.41, and 0.39, respectively (Figure S9c). These regions are located in the subtropic zone, and the temperature and precipitation are higher, therefore, the organic carbon biodegrades faster than that in the cold area. Furthermore, the consuming amounts of fruits and vegetables, which contain a high fraction of DOC, in southern cities were greater than those of northern cities.[30] Therefore, it leads to higher methane emissions in landfills of the cities in the subtropic zone.[36] Moreover, previous studies showed that methane, odor, and leachate are generated through the anaerobic digestion process of organic matter.[27] Therefore, methane emissions can also be used to reflect the local environmental impacts of carbon stocks in landfills.

Synergistic reduction of carbon stocks and GHG emissions of landfills

According to related MSW interventions such as landfill gas collection system upgrading, waste input decreasing, ecological restoration (aerobic restoration and stock mining), we designed four scenarios to analyze the shift in organic carbon stock and relative GHG emissions reduction in landfills by 2030 (Figures 5-6 & S10). For the business-as-usual scenario, the policy interventions of the MSW management system remain the same as in 2020. For the raising-methane-collection scenario, the methane collection process is improved, but other MSW management measures remain the same as in the business-as-usual scenario. For the minimizing-landfill scenario, new policies, such as zero-waste city programs and the source separation of MSW, will be continuously implemented in China and thus change the landfilling rates, as well as the compositions of landfilled waste. For the landfill-remediation scenario, in-situ remediation and landfill mining programs will be applied based on the settings of the minimizing-landfill scenario. The four scenarios are progressive, i.e., the interventions set by the latter are based on the former. Moreover, the methane emissions in this study are relative, that is, GHG emissions reduction through utilizing landfill organic carbon stock or MSW instead of widely used products are also included.

Results show that, except in the landfill-remediation scenario, the organic carbon stocks in the landfills of the other three scenarios keep increasing compared to those in 2020 (Figure 5). In the business-as-usual and raising-methane-collection scenarios, the organic carbon stock of landfills shows an exponential growth trend, reaching 615.4 Tg and 727.4 Tg by 2025 and 2030, increasing by 16.9% and 38.2% compared with that in 2020, respectively. In these two scenarios, the organic carbon stock in the form of soil-like material has the highest proportion of the total stock in landfills, fluctuating between 55.5% and 56.6%. In the minimizing-landfill scenario, interventions are based on the goals of China's 14th Five-Year Plan (2021 - 2025), i.e., the mismanaged dumps rate decreases to 0%, the incineration rate of MSW reaches 68%, the percentage of food waste diverting from mixed MSW stream reaches 20%, and the landfilling rate drops to 12% in 2030. These policies will result in a slowdown in the growth of the organic carbon stock in landfills with the increase of only 2.2% annually, which is 58.1% of that under the business-as-usual scenario. Interestingly, the landfill-remediation scenario is the only one in which the net amount of organic carbon stock in landfills decreases by 2030, reducing by 18.7% in 2025 and 56.3% in 2030 compared to that in 2020.

Promoting the methane collection rate, decreasing the landfilling rate, and implementing eco-remediation of landfills all produce positive effects on GHG emissions reduction from the MSW management sector. The business-as-usual scenario takes into account the increase of waste generation and the decrease of food waste fraction, and the total amount of methane generated by landfills in 2030 is 1577.8 Tg CO_2eq (Figure 6). In the raising-methane-collection scenario, the methane collection rate in sanitary landfills increases from 24% to 40%, which results in a 5.7% reduction in the final methane emissions from landfills (i.e., 1487.8 Tg CO_2eq) by 2030. In the minimizing-landfill scenario, the GHG emissions from landfills are 1408.6 Tg CO_2eq, and the GHG emissions reduction reaches 251.1 Tg CO_2eq. The decreasing share of landfilling, resulting in 64.2% of the total GHG emissions reduction. Furthermore, in this scenario, the increase of the incineration rate generates an additional 66.7 TWh of electricity, and the increase in the composting rate produces additional organic fertilizers (equivalent of 6.4 million tons of urea

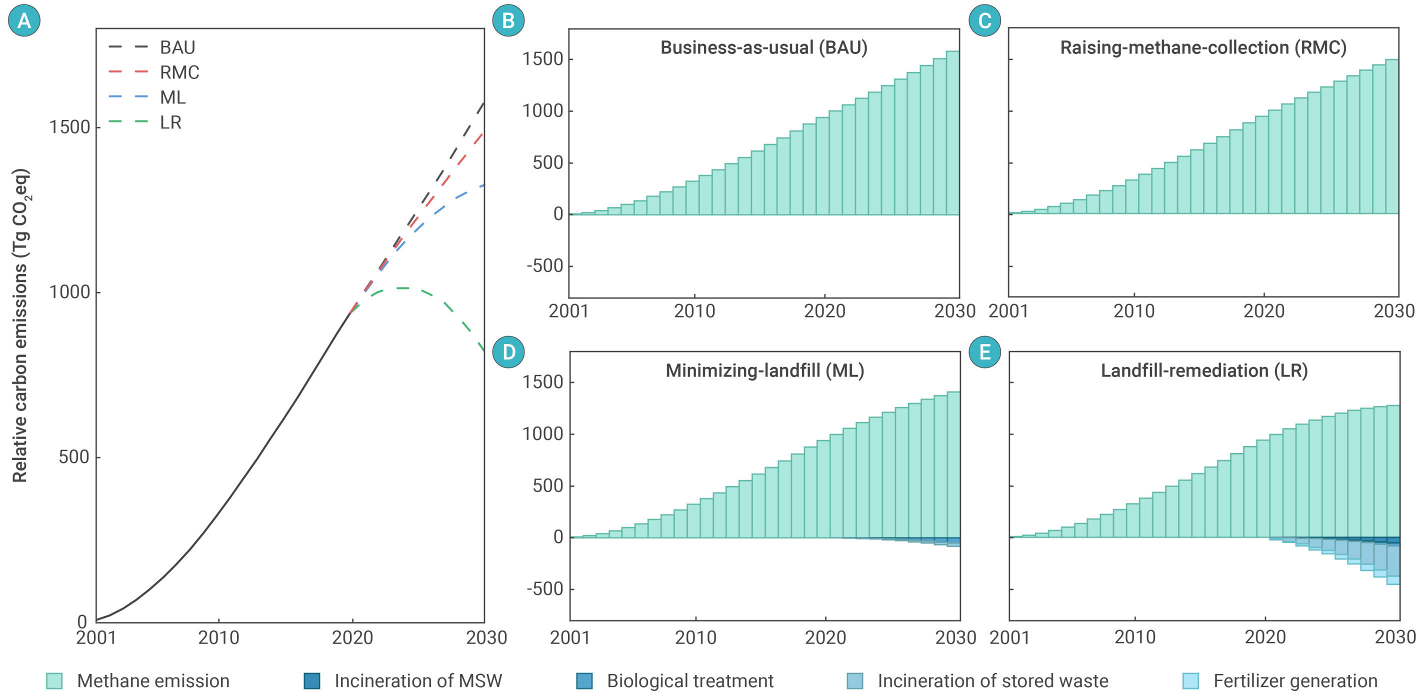

Figure 6. Scenario analysis of cumulative GHG reduction from landfills in China from 2001 to 2030 (A) Overview of cumulative GHG reduction, summarizing the results under four scenarios. (B), (C), (D), (E) are cumulative GHG emissions and emissions reduction from landfills by measure in the business-as-usual (BAU), raising-methane-collection (RMC), minimizing-landfill (ML), and landfill-remediation (LR) scenarios, respectively. Note: the legends of incineration of MSW, compost, incineration of stored waste, and fertilizer generation, represent cumulative GHG emission reduction from adopting relative interventions, the results are based on life cycle assessment methodology, and the GHG emission reductions from recovered energy and recycling nutrients are included.

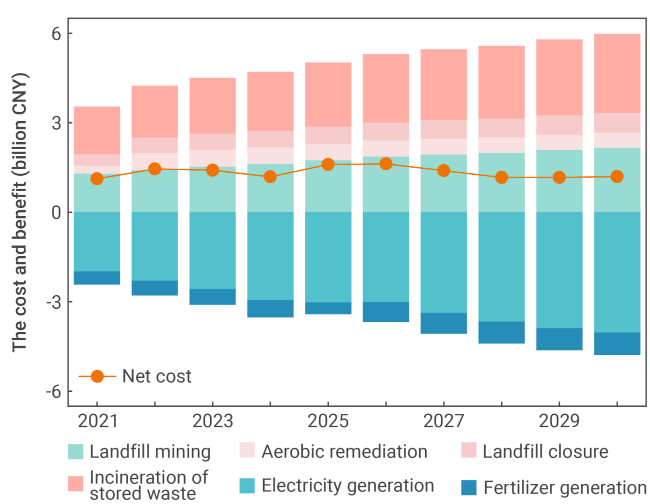

Figure 7. The cost-benefit analysis of the landfill-remediation scenario Note: positive values represent costs, negative values represent benefits.

fertilizer), which both contribute to GHG emissions reduction from the perspective of life cycle analysis.

The landfill-remediation scenario achieves the greatest GHG emissions reduction, the cumulative GHG emissions are only 824.4 Tg CO_2eq by 2030, accounting for 54.8% of the business-as-usual scenario. Energy recovery from stored plastic and rubber, textile and wood, and fertilizer recycling from stored soil-like material contribute to net GHG reductions of 290.7 and 79.2 Tg CO_2eq, accounting for 38.6% and 10.5% of the total GHG emissions reduction in the landfill-remediation scenario, respectively. Besides, by applying landfill mining, 695.7 TWh of power generation and 13.4 million tons of urea fertilizer substitutes are produced, 10.4 and 2.1 times higher than those in the minimizing-landfill scenario, respectively. Besides, in-situ aerobic remediation and landfill mining will also lead to a reduction of methane emissions from landfills by 108.6 and 23.8 Tg CO_2eq, respectively, accounting for 14.4% and 3.2% of the total emissions reduction. It indicates that more than 66.7% of the carbon reduction was due to the application of landfill restoration, mainly due to the recycling and substituting resource in stored waste. Interestingly, the landfill-remediation scenario is the only one that can transform the landfill into a carbon-negative sector (Figure S10). By 2030, the GHG emissions from urban landfills in China will be reduced to -57.1Tg CO_2eq/year.

The economic feasibility of ecological remediation of landfills in China under the landfill-remediation scenario is presented in Figure 7, with a detailed cost and benefit analysis available in Table S15. Generally, there is a net cost of 132.6 billion CNY for the eco-remediation of landfills, with a total cost of 501.3 billion CNY and a total benefit of 368.7 billion CNY; the annual net cost varies from 11.1 to 16.2 billion CNY from 2021 to 2030. With respect to specific costs and benefits, the costs of landfill mining, aerobic remediation, closure of landfill, incineration are 176.3, 50.8, 58.2, and 215.9 billion CNY; the benefits of electricity and fertilizer production are 308.3 and 60.4 billion CNY; respectively. The costs of landfill mining and the benefit of electricity are the most crucial economic issue for the eco-remediation scenario.

DISCUSSION

The underestimated amount of organic carbon in landfills

One of our innovations is the solid-liquid-gas transformation model of stored waste in landfills. The mass balance among different phases of organic carbon can be analyzed by applying this model, with city-specific coefficients input, such as waste composition, temperature, precipitation, etc. By increasing the spatial and temporal resolution of the quantification approach,[31] we found that the DOC stock of MSW landfills in China in 2014 was twice that of previous estimates (15.8 vs. 7.2 Tg/year).[3]

What's more, due to our unique methodology, we were able to fill a key knowledge gap in previous work. Stubbins et al. (2021) proposed that landfills are an important but unknown link in understanding the global plastic-carbon cycle.[4] However, plastic-carbon stock remains poorly studied and poorly known, especially across cities in China. Our study found that the FOC in landfills by 2020, in the form of plastic waste, was 169.1 Tg, equivalent to 2.2 times the total plastic use (including heavy-duty PVC etc.) in China in

2020,[47] and 11.3 times the plastic used for packaging.[48] This means that the organic carbon stock of landfills is ignored, and the overlooked landfill DOC and FOC may have huge resource value, but also might cause unpredictable environmental problems, including GHG emissions.

Significance of overlooked carbon stocks in landfills

Our results suggest that organic carbon stock in landfills will be a very potent source of methane emissions, but it has also great resource and energy potential if properly recycled and used. We demonstrate that the organic carbon reserve of soil-like material in landfills, on average, is equivalent to 20% of what is stored in urban green spaces, which are considered as the most vital organic carbon pool in urban areas.[2-3] This landfill organic carbon pool can be potentially utilized when taking appropriate engineering measures. Additionally, the organic carbon in the combustible fractions in landfills have great potential for renewable energy generation. In theory, the amount of electricity converted from the heat value stored in combustible waste is equivalent to nearly a year's electricity consumption for average urban residents. In fact, for cities with high MSW input but less energy consumption, this value becomes more impressive. For example, Wuwei can potentially power the entire city for 4.4 years by converting combustible trash into energy. Moreover, we calculated that methane emissions from landfills accounted for 3.9% of the total anthropogenic methane emissions (including agricultural source outside of cities), suggesting a non-trivial role for the global methane cycle. Further details on the significance of carbon stocks in landfills are discussed in supplementary information section 1.4.

Mitigating GHG emissions from landfills through life-cycle interventions

Organic carbon stock in landfills has significant impacts on both short- and long-term GHG emissions. According to the business-as-usual scenario, methane emissions from landfills in 2030 may increase by 638.6Tg CO_2eq compared with those in 2020, due to rapid urbanization and expanding consumption in China.[49] Collection and treatment of landfill gas were considered as the key measures to reduce GHG emissions from landfills. This study suggests that increasing the methane collection rate (from 24% to 40%) will only reduce GHG emissions by a total of 89.9 Tg CO_2eq over the next decade compared to the business-as-usual scenario. Thus, increasing methane collection alone is still insufficient for a more ambitious carbon reduction goal.

The minimizing-landfill scenario indicates that reducing the amount and share of landfilling, as the measures taken by the Chinese government currently, will lead to a considerable contribution to decreasing GHG emissions from landfills. In recent years, the implementation of relevant laws, regulations, and programs (such as the Anti-Food-Waste Law, the Opinions on Further Strengthening the Control of Plastic Pollution, Zero-waste Pilot Cities, and Compulsory Waste Source Separation Program) will reduce the amount of food waste and plastic waste in landfills in the future.[50-51] With further progresses in sustainable MSW management in China, the DOC input to landfills will decrease, and thus eventually reduce GHG emissions.[43] In the minimizing-landfill scenario, the cumulative organic carbon stock of landfills is only reduced by 11.6%, while the GHG emission reductions is as high as 48.5% compared with GHG emissions in 2020. It indicates that if those policies can take effect in the future, methane emissions from the MSW sector could easily achieve the COP27 target, reducing 30% of total emissions by 2030.[11] The cumulative GHG emissions reduction also accounts for 0.6% of the GHG emission gaps for China to achieve 1.5°C warming targets by 2030.[52-53]

Moreover, landfill mining and ecological remediation of post-closure landfill provide a feasible approach to optimizing the urban carbon cycle. The landfill-remediation scenario demonstrates that landfill mining can not only achieve resource/energy utilization of landfill organic carbon, but also significantly reduce methane and leachate production from landfills.[54-55] In recent decades, landfill remediation has been regarded as an important option in MSW management in China, and there were successful cases in many cities (e.g., Beijing and Wuhan). Although landfill mining can bring significant environmental benefits, technological innovation and infrastructure capacity are also urgently needed for the utilization of soil-like materials, plastics, and other stored recyclables. The capacity of incineration plants is crucial for implementing landfill mining programs, due to their potential for disposing residues and harvesting renewable energy. The construction of incineration plants in China is lagging but accelerating, with the number of plants increasing by 7.3 folds over the past ten years, which enables the country to carry out landfill mining in the future (see Figure S11).

Based on our analysis, the landfill-remediation scenario is the only negative carbon emission scenario, the GHG emissions will be -57.1 Tg CO_2eq in 2030 (Figure S10). According to the China Fourth National Communication on Climate Change, GHG emissions from the landfill sector was 1210.2 Tg CO_2eq in 2017, indicating a remarkable negative-carbon transition for the landfill sector. The cumulative GHG reduction by 2030 is 753.3Tg, accounting for 62.2% of the GHG emission from the landfill sector in China[56] and 2.0% of the GHG emission gaps for China to achieve the 1.5°C warming targets by 2030 (Figure S10). The cumulative GHG mitigation is largely due to the ecological remediation of landfills (e.g., aerobic remediation and landfill mining). How to utilize the organic carbon stocks of landfills will bring huge uncertainty to the GHG mitigation of landfills in the future. Compared with incineration of organic carbon stock, the increased resource utilization of organic carbon stock will bring more GHG emissions reduction from landfills (see details in supplementary information section 1.5). Therefore, stock waste to material may be a more potential way to mine and utilize landfill stored substances.

In addition, it indicates that ecological remediation of landfills for mitigating the GHG emission can be economically feasible. A total economic input of 501.3 billion CNY, with a net cost of 132.6 billion CNY by waiving the benefits of electricity and fertilizer production, could contribute a GHG reduction of 369.9 Tg. The benefit from land reclamation could be another vital factor to raise the economic feasibility of ecological remediation; however, due to differentiated land prices among cities, it was not discussed in this work. Besides, although it was not quantified in this research, recycling carbon-free materials from landfills, such as metal and glass, is also closely related with the urban carbon cycle, due to their high energy consumption for producing primary materials.[44,57] However, it is worth noting that environmental pollution monitoring and controlling should be highlighted during the landfill mining process. For instance, odor may emit if the stabilization degree of the stored waste is low, or the gas collecting system at working surface of excavation is ineffective. In addition, the soil-like materials may contain heavy metals, impeding recycling it into farmland.[58] Therefore, economic input for controlling the pollutant leakage could be very vital for implementing landfill mining programs.

Innovative technologies for landfill retrofitting, such as dry bio-landfilling, could throw positive impacts on carbon reduction of landfills in the future.[59] According to our preliminary analysis, if dry bio-landfills was implemented after 2020, cumulative GHG emissions from landfills in China are expected to further decrease by 46.8% by 2030 (see details in Supplementary Information, Section 1.6 and Figure S12). It indicates that innovative approaches to designing, constructing, and maintaining landfills could also contribute to the GHGs mitigation. It merits in-depth researches in the future, covering the whole life-cycle of landfill's low-carbon transition.

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AUTHOR CONTRIBUTIONS

Shijun Ma: Methodology, Formal analysis, Visualization, Writing (original draft). Mingzhen Lu: Visualization, Writing (review & editing). Guang Yang: Writing (review & editing). Yuehao Zhi: Writing (original draft), Methodology. Zutao Ouyang: Writing (review & editing). Jing Meng: Writing (review & editing). Heran Zheng: Writing (review & editing). Ningxin Huang: Data collection, Writing (review & editing). Zhiying Yang: Data collection. Chuanbin Zhou: Methodology, Writing (review & editing), Visualization, Funding acquisition. All authors contributed to and approved the manuscript.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

Data are available from the corresponding author upon reasonable request.

SUPPLEMENTAL INFORMATION

It can be found online at https://doi.org/10.59717/j.xinn-geo.2024.100109