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Original Research

Environmental sustainability opportunity and socio-economic cost analyses of phosphorus recovery from sewage sludge

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ABSTRACT

Although phosphorus (P) recovery and management from sewage sludge are practiced in North America and Europe, such practices are not yet to be implemented in China. Here, we evaluated the environmental sustainability opportunity and socio-economic costs of recovering P from sewage sludge by replacing the current-day treatments (CT; sludge treatment and landfill) and P chemical fertilizer application (CF) in China using life cycle assessment and life cycle costing methods. Three potential P recovery scenarios (PR_1–PR_3: struvite, vivianite, and treated sludge) and corresponding current-day scenarios (CT_1–CT_3 and CF) were considered. Results indicated that PR_1 and PR_2 have smaller environmental impacts than the current-day scenarios, whereas PR_3 has larger impacts in most categories. PR_3 has the lowest net costs (sum of internal costs and benefits, 39.1−54.7 CNY per kg P), whereas PR_2 has the lowest external costs (366.8 CNY per kg P). Societal costs for production and land use of 1 kg P by P recovery from sewage sludge (e.g., ~527 CNY for PR_1) are much higher than those of P chemical fertilizers (~20 CNY for CF). However, considering the costs in the current-day treatments (e.g., ~524 CNY for CT_1), societal costs of P recovery scenarios are close to or slightly lower than those of current-day scenarios. Among the three P recovery scenarios, we found that recovering struvite as P fertilizer has the highest societal feasibility. This study will provide valuable information for improved sewage sludge management and will help promote the sustainable supply of P in China.

1. Introduction

Phosphorus (P) is life's bottleneck on the planet [1−3]. It is vital for the development of all living organisms [4]. Global food production has largely depended on the extraction of rock phosphate for fertilizer even though it is a non-renewable resource and cannot be replaced by other elements [5−7]. To feed the projected 9.7 billion residents by 2050, the total global food demand is expected to increase by 35−56% from 2010 to 2050 [8]. However, sustained P supply for food production is still not assured [2,9,10]. Despite the likelihood of discovering new P deposits and improved technology, rock phosphate is expected to be depleted within 100−400 years [11,12]. Therefore, establishing a circular P flow, rather than the one-way flow, between agriculture and the urban food system will be essential to satisfy the future needs for P while protecting a critical natural resource [4,13]. In urban regions, P in human excreta is usually collected by sewer pipes, treated in wastewater treatment plants (WWTPs), and ends up in sewage sludge, which could account for up to 98% of the P ingested by human beings [14]. On the global scale, P remaining in sewage sludge represents around 25% of the P demand in agriculture, but only a small percentage has been recycled [15]. Notably, efforts to increase P recovery from human waste would also advance several Sustainable Development Goals declared by the United Nations, such as “Zero Hunger” (SDG 2), “Sustainable Cities and Communities” (SDG 11), and “Clean Water and Sanitation” (SDG 6) [16].

P recovery from human waste could provide important

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opportunities to close the biogeochemical P cycle at a regional or system level while reducing the environmental burdens associated with sewage sludge treatments [17,18]. A good example of advancing P recovery from sewage sludge has emerged in the European Union (EU) [19,20]. To encourage large-scale production of P fertilizer from domestic organic and secondary raw materials, the EU adopted new regulations on fertilizer application in 2019 [21]. Sludge contains mineral and organic P, which can be applied on land as fertilizer and soil amending agent after treatment to the relevant standards [22]. This practice has already been used in some European and African countries (e.g., Sweden, Finland, and Rwanda) [4,23]. In recent decades, some new technologies have been developed to recover P from sewage sludge to avoid concerns related to heavy metals, pathogens, and social acceptability [24,25]. One proposed route is to recover P from a digest solution in the form of struvite ($NH_4MgPO_4 \cdot 6H_2O$), which could then be applied as a slow-release fertilizer on cropland [26,27]. Recently, P recovery in the form of vivianite [$Fe_3(PO_4)_2 \cdot 8H_2O$] has been proposed as a promising pathway [28,29], along with phosphate compounds like calcium phosphates [30]. Local businesses have profited from the reuse of human waste in Kigali (Rwanda) and Accra (Ghana) [23]. Furthermore, 17 struvite recovery facilities have been set up by Ostara, which could serve 11.5 million people in North America and Europe [31].

However, a remaining unsolved question surrounds the overall environmental impacts of recovering P from sludge and how it compares with the current-day sludge treatments (e.g., landfill, incineration) and P chemical fertilizer application. Life cycle assessment (LCA) provides a useful tool to evaluate changes in terms of different environmental impacts by altering or replacing production processes [32–34]. Bradford-Hartke et al. [35] compared the environmental benefits and burdens of P recovery from six municipal wastewater systems. They found that P recovery did not necessarily present net environmental benefits and depended on the methods of P recovery. Linderholm et al. (Linderholm et al., 2012) evaluated the environmental impacts of four P recovery pathways to supply Swedish agriculture. They concluded that using sewage sludge directly on croplands was the most efficient option regarding energy use and greenhouse gas emissions. However, Pradel and Aissani [36] suggested that sludge-based P fertilizers appeared to be less environment-friendly than mineral fertilizers from the “product” perspective because of the contribution of the upstream burden in sludge production and P recovery. In recent years, life cycle costing (LCC), an approach extended by LCA, has also been widely used to evaluate the economic costs or benefits of the selected scenario [37,38]. Tonini et al. [39] estimated that the societal costs incurred for P products derived from sewage sludge, manure, and meat and bone meal were 81%, 50%, and 10%, respectively, lower than the rock-derived superphosphate. Rashid et al. [40] calculated that the life cycle costs of three upgraded processes with the additions of nutrient removal and P recovery technologies were on average 24% higher than the original processes. By combining the LCA and LCC into a common framework, the decision-making process around these technologies could be more effective and comprehensive [41].

China, with an urban population of 0.9 billion, has the largest wastewater treatment capacity across the world [42]. In 2020, around 65 billion cubic meters of municipal wastewater was produced, and more than 90% of it was treated by advanced wastewater treatment, mainly relying on the sludge process [43]. About 60 Mt of sewage sludge with 80% moisture content was produced in 2019 [44]. It is estimated that around 70% of the sludge was not properly managed, resulting in a waste of resources and pollution of water, air, and soil [45]. In recent years, rapid declines in the stock of phosphate rock and P losses in the environment have led to concerns over the sustainable P supply in China [46,47]. In 2020, phosphate rock was added to the list of strategic minerals in China [48]. These changes have urged the government to reconsider the opportunity for P recovery from sewage sludge that has been previously neglected [49].

This study aimed to evaluate the environmental sustainability opportunity and socio-economic costs of recovering P from sewage sludge by replacing current-day treatments (sludge treatment and landfill) and P chemical fertilizer application in China. Three potential P recovery scenarios from sewage sludge (i.e., struvite, vivianite, and treated sludge) and corresponding current-day scenarios were considered and evaluated. The environmental impacts and societal costs (i.e., internal and external costs, benefits) of each scenario were calculated based on LCA and LCC methods. This study is expected to provide valuable information for improved sewage sludge management, which would turn waste into resources and promote a sustainable P supply in China.

2. Methods and materials

2.1. Standard life cycle assessment

Our LCA analysis for each scenario strictly followed the four-step procedure as specified in the ISO standards 14040 and 14044 [50,51]. The first step consisted of defining the goals and scopes of the analysis, such as the system boundary, functional unit, and assumptions. The second step was the life cycle inventory (LCI) analysis, which listed and considered all inputs and outputs related to each process in the system [36]. The third step was life cycle impact assessment, which determined the consequences related to the inputs and outputs of the target system. Different categories of environmental impact (e.g., climate change, ecosystem quality, human health, and resources) could be emphasized based on the study's goal. Finally, these results were interpreted to describe the connection between inventory data and environmental impact categories.

2.2. Functional unit and system boundary

The primary objective of this study was to quantify the environmental impacts and socio-economic costs of P recovery from sewage sludge for three scenarios (PR_1–PR_3) by replacing the current-day scenarios, including current-day treatments (CT_1–CT_3; sludge treatment and landfill) and P chemical fertilizer application (CF) (Fig. 1). In this study, production and land use of 1 kg of bioavailable P was identified as the functional unit to provide the reference for the LCI. Bioavailable P is the combined quantity of P immediately available to plants and P that can be transformed into available forms by naturally occurring process [39]. In the current-day scenarios, the most common sludge treatment in China is by disposal as landfill [52]. For the P recovery scenarios, three pathways were considered, including recovery in the forms of struvite (PR_1), vivianite (PR_2), and treated sludge (PR_3). Recovered P products were applied on the cropland as replacements for P chemical fertilizers (Fig. 1).

Among the three P recovery scenarios, P recovery as struvite (PR_1) has been implemented in North America and Europe [17,53]. P recovery as vivianite (PR_2) has been developed recently, and pilot-scale tests have been reported [28]. Relative to the recovery of struvite, the reaction conditions used to produce vivianite are less demanding, and P recovery efficiency from digest solutions is higher [54,55]. Vivianite has a higher commercial value than struvite in addition to potential applications in agriculture [56,57]. Therefore, P recovery as vivianite is likely to be a promising pathway in the future. The application of treated sludge has also

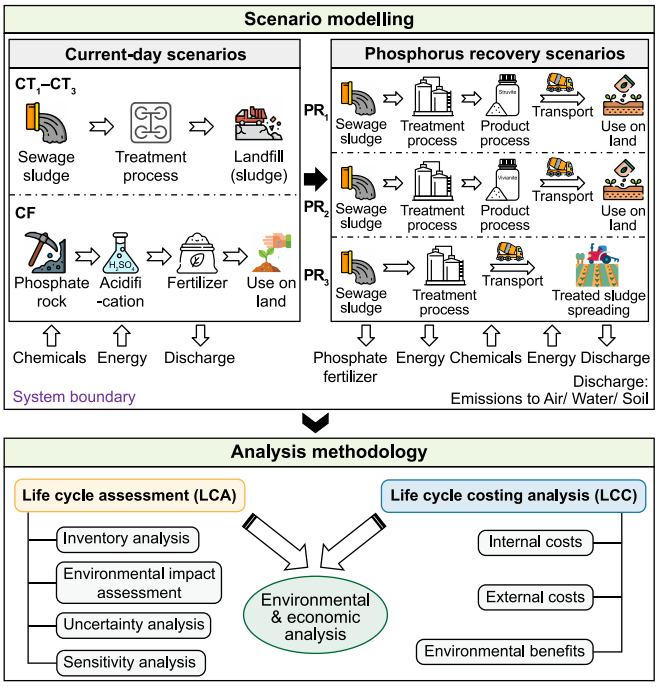

Fig. 1. System boundary and schematic diagram of the methodology applied in this study.

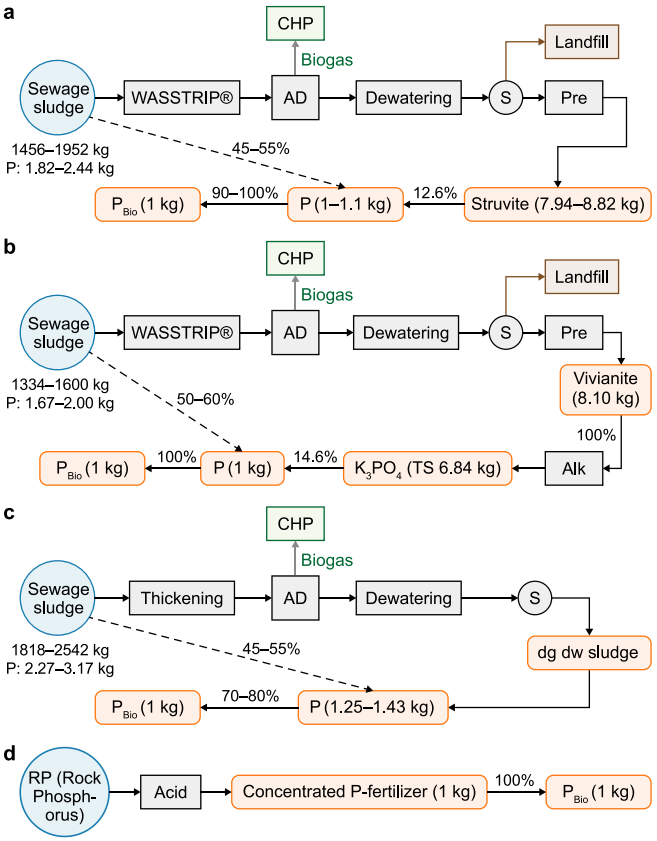

Fig. 2. Detailed processes and parameters in three P recovery scenarios and P chemical fertilizer application. **a**, Struvite scenario; **b**, Vivianite scenario; **c**, Treated sludge scenario; **d**, P chemical fertilizer application. WASSTRIP®: Waste Activated Sludge Stripping to Remove Internal Phosphorus; AD: Anaerobic digestion; CHP: Combined heat and power; S: Solid–liquid separation; Pre: Precipitation; Alk: Alkali dissolution; dg dw sludge: Digested and dewatered sludge.

been introduced as a potential P recovery route (PR_3). In recent years, European countries have started to apply sewage sludge as nutrient fertilizers and soil amending agents [58,59]. In China, the application of sewage sludge has been allowed in landscaping [44]. Detailed processes and parameters for different P recovery pathways are displayed in Fig. 2.

2.3. Life cycle inventories for different scenarios

Three P recovery pathways were assessed starting from sewage sludge, all of which included assessments of anaerobic digestion (AD), combined heat and power (CHP), dewatering, and solid–liquid separation (Fig. 2). Major technologies applied in sludge treatment and P recovery included WASSTRIP® (waste activated sludge stripping to remove internal phosphorus) and precipitation (for PR_1 and PR_2), thickening (for PR_3), alkali dissolution (for PR_2), spreading (for PR_3), landfill (for CT_1–CT_3), and land transport (for PR_1–PR_3). The WASSTRIP® process is designed to release P upstream of the digestion tanks, thereby releasing more P from sludge and improving the P recovery efficiency [39]. Separated P-containing supernatant is then sent together with the rejected water from sludge thickening after digestion to the precipitation reactor (Ostara Pearl®), where struvite or vivianite is produced [39]. It is noteworthy that because of the low solubility of vivianite, potassium hydroxide is added to produce potassium phosphate solution in fertilizer applications [60]. In this study, we assumed that the P recovery from sludge occurred in WWTPs with sludge production of 70000 tons yearly [39]. It was assumed that the P content accounted for 2.5% of the total solid [61,62]. The moisture content of sewage sludge was assumed to be 95%, and the moisture content of treated sludge used for landfill or crop spreading was assumed to be 60% [34,63].

Depending on P recovery pathways, chemicals required for the P recovery process can include ferric chloride, calcium oxide, lime (for PR_1–PR_3), magnesium hydroxide (for PR_1), and potassium hydroxide (for PR_2). Ferric chloride has two major functions: it controls hydrogen sulfide production in the anaerobic digesters and precipitation of struvite and vivianite in the digested-sludge thickening stage [27], and it coordinates with calcium oxide in the dewatering stage to improve dewatering efficiency. Energy is typically consumed in diesel, electricity, and thermal energy. Given that the thermal energy produced by biogas utilization in the CHP stage was sufficient to offset the thermal energy consumption in the whole stage [64], the thermal energy was not calculated separately. Furthermore, a large amount of electrical energy could also be recovered in the CHP stage, reaching up to 3.7–3.8 times the electrical energy consumed in the AD process [36,65,66]. Land transport requirements included the transportation of raw materials, P-containing fertilizer, and remaining sludge. The product was transported from P recovery facilities to the croplands, and the remaining sludge was transported to landfill sites by truck in loads of 16 tons. All transport distances were set to 30 km in the considered scenarios [67]. Discharge included the emissions to air, water, and soil in the form of gases, ions, and metal elements and was considered part of the output inventory. Major processes involved in CT were sludge treatments (including thickening, AD, and dewatering) and landfill (Figs. 1 and S1). In CF, we assumed that 7.6 kg of rock phosphate was needed to produce 1 kg of bioavailable P in China (P_2O_5 content: 30%) [68]. Sulfuric acid was used in the production of P chemical fertilizers. In this study, the product system was modeled using openLCA 1.10 [69], and the Ecoinvent database 3.7 [70,71] was used to model processes of the background system [72]. Technical data used to simulate the foreground

LCA system was mainly derived from literature and survey data (Table 1). Some data came from parameters of specific cases. A detailed input–output table and parameter values are presented in Tables 1 and S1.

2.4. Life cycle impact assessment and life cycle costing

In this study, environmental impacts for different scenarios were determined by the EF2.0 midpoint method [78–80]. Four major impact groups were considered: climate change, ecosystem quality, human health, and resources. Specific impacts include climate change (CC, kg CO_{2eq}), freshwater and terrestrial acidification (FTA, mol H^{+}_{eq}), freshwater eutrophication (FEU, kg P_{eq}), marine eutrophication (MEU, kg N_{eq}), carcinogenic effect [CE, comparative toxic units for humans (CTUh)], ozone layer depletion (OLD, kg CFC-11_{eq}), land use (LU, points), and dissipated water (DW, m^3 $water_{eq}$). Alongside impact assessment, LCC analysis was used to quantify the societal costs. The societal costs included internal costs, external costs, and benefits. Internal costs mainly included the fixed costs of the equipment and the costs of material and energy during the operation stage [37]. External costs refer to the economic costs caused by the environmental impacts of these scenarios [81]. Benefits were mainly from energy recovery and resource substitution. The compound-specific shadow price and monetization factors were used to estimate the external costs induced by the environmental impact [82,83]. Market prices for different inputs in the LCA are provided in Table S2. The fixed cost was converted into unit equivalents at a discount rate of 5% in Table S3. The factors to monetize different categories of environmental impact are provided in Table S4.

2.5. Uncertainty and sensitivity analysis

This study used Data Quality Indicator (DQI) to provide a semi-quantitative analysis of input variables in the LCA [84]. DQI applied a pedigree matrix approach to assessing the qualities of input variables, including “reliability”, “completeness”, “temporal correlation”, “geographic correlation”, and “technological correlation” [70,85]. Each indicator was further divided into five quality levels ranging from 1 to 5 (Table S5). Each level corresponded to an uncertainty factor. To address uncertainty in LCA, Monte Carlo simulation was applied to obtain a rigorous conclusion. In this study, we used 1000 iterations in uncertainty analysis. Sensitivity analysis was performed on the LCA results to identify the main parameters causing environmental impact [66]. This was achieved by changing the value of each inventory data by 10% of the original values and calculating the sensitivity [86].

3. Results

3.1. Environmental and health impacts

Environmental and health impacts on four typical categories (i.e., climate change, ecosystem quality, human health, and resources) for three P recovery scenarios (PR_1–PR_3) and the current-day scenarios (CT_1–CT_3 and CF) are presented in Fig. 3. The detailed

Table 1
Life cycle inventory for scenarios of three P recovery (PR_1-PR_3), current-day treatments (CT_1-CT_3), and P chemical fertilizer application (CF) (based on 1 kg bioavailable P).

Category	Input & Output	Unit	PR_1	PR_2	PR_3	CT_1	CT_2	CT_3	CF	Reference
Chemicals consumption	Magnesium hydroxide	kg	2.24	-	-	-	-	-	-	Estimated
	Ferric chloride	kg	1.96	9.82	2.51	1.96	1.69	2.51	-	[28,73]
	Calcium oxide	kg	3.27	2.82	4.19	3.27	2.82	4.19	-	[73]
	Potassium hydroxide	kg	-	5.2	-	-	-	-	-	Estimated
	Lime	kg	11.96	10.30	-	11.96	10.30	15.30	-	Estimated
	Sulfuric acid	kg	-	-	-	-	-	-	5.0	Estimated
	Liquid ammonia	kg	-	-	-	-	-	-	0.488	Estimated
Energy consumption	Diesel	L	0.67	0.58	0.85	0.50	0.43	0.63	-	[39,74]
	Electricity	KWh	35.0	33.0	44.2	28.9	24.9	39.7	-	[52,65,74-76]
Other consumption	Spreader	kg	0.078	0.067	3.23	-	-	-	0.034	[75]
	Tractor	kg	0.028	0.024	1.16	-	-	-	0.012	[75]
	Landfill	kg	136	117	-	136	117	174		Estimated
	Transport[a]	t • km	8.68	7.71	10.5	8.16	7.02	10.4	0.38	Estimated
Emissions to air	Carbon dioxide	kg	49.7	42.7	70.9	49.7	42.7	63.4	-	[36,74]
	Carbon monoxide	kg	0.036	0.031	0.046	0.036	0.031	0.046	-	[36]
	Methane	kg	1.25	1.08	4.34	1.25	1.08	1.60	-	[52]
	Nitrogen	kg	0.06	0.05	0.08	0.06	0.05	0.08	-	[36]
	Nitrogen oxides	kg	0.033	0.028	0.058	0.033	0.028	0.042	-	[36,52]
	Sulfur oxides	kg	0.013	0.011	0.061	0.013	0.011	0.016	-	[36,52]
	VOCs	kg	0.007	0.006	0.009	0.007	0.006	0.009	-	[36]
	Hydrogen sulfide	kg	0.73	0.63	0.93	0.73	0.63	0.93	-	[36]
	Ammonia	kg	1.61	1.39	2.06	1.61	1.39	2.06	-	[36]
	Dinitrogen monoxide	kg	-	-	0.128	-	-	-	-	[77]
Emissions to soil	Cadmium	g	-	-	0.102	-	-	-	-	[36]
	Chromium	g	-	-	3.48	-	-	-	-	[36]
	Copper	g	-	-	24.34	-	-	-	-	[36]
	Lead	g	-	-	3.48	-	-	-	-	[36]
	Mercury	g	-	-	0.063	-	-	-	-	[36]
	Nickel	g	-	-	2.09	-	-	-	-	[36]
	Zinc	g	-	-	48.7	-	-	-	-	[36]
Emissions to water	Nitrate	kg	-	-	10.33	-	-	-	-	[36]
	Phosphorus	kg	-	-	0.009	-	-	-	-	[36]
Avoided product	Electricity	KWh	50.16	43.32	64.22	50.16	43.32	64.22	-	[36,65,66]
	Inorganic nitrogen fertilizer, as N	kg	0.48	-	-	-	-	-	-	Estimated
	Inorganic phosphorus fertilizer, as P_2O_5	kg	2.29	2.29	2.29	-	-	-	-	Estimated
	Inorganic potassium fertilizer, as K_2O	kg	-	4.51	-	-	-	-	-	Estimated
	Tap water	kg	-	5.56	-	-	-	-	-	Estimated

[a] 30 km, 16 ton, truck.

environmental impacts of each scenario are provided in Table S6. As shown in Fig. 3, relative to the current-day scenarios, PR_1 and PR_2 usually presented smaller environmental impacts, while PR_3 showed different results depending on the category of environmental impact (e.g., higher impacts on climate change and eutrophication but smaller impacts on land use and dissipated water). Compared with CT_1+CF and CT_2+CF, impacts on climate change declined by 11.2% and 38.4%, respectively, for PR_1 and PR_2, and the impacts on land use decreased by 37.5% and 50.0%, respectively (Table S6). However, impacts on climate change and marine eutrophication for PR_3 increased by 132.7% and 1273.0%, respectively, relative to CT_3+CF (Table S6).

If only based on the "product" perspective (1 kg of bioavailable P), we found that the environmental impacts of using P chemical fertilizers (CF) would be much smaller than those caused by using P recovered from sewage sludge (PR_1–PR_3). For instance, the impacts on freshwater and terrestrial acidification were 4.8 mol H^+_{eq} per kg P for PR_1 and 4.1 mol H^+_{eq} per kg P for PR_2, whereas only 0.1 mol H^+_{eq} per kg P for CF (Table S6). The impacts on land use were 163.3, 114.0, and 108.7 points per kg P, respectively, for PR_1, PR_2, and PR_3, whereas only 22.7 points per kg P for CF (Table S6). However, when considering the impacts by replacing the current-day treatments, the P recovery scenarios (mainly PR_1 and PR_2) are superior. CT_1–CT_3 induce large environmental burdens, which would be even higher than PR_1–PR_3 in some impacts. For example, the impact on land use was 238.0 points per kg P for CT_1, whereas it was only 163.3 points per kg P for PR_1. For the impact on climate change, CT_2 had an environmental impact of 77.4 kg CO_{2eq} per kg P, whereas PR_2 only had a burden of 49.9 kg CO_{2eq} per kg P, illustrating the potential environmental benefits of replacing the current-day treatments. Overall, although replacing P chemical fertilizer with recovered P may not appear as an environment-friendly option, net environmental benefits would be obtained for most impact categories if considering the environmental burdens of current-day treatments (Fig. S2). As shown in Fig. S2, a positive value (when PR > CT + CF) represented a net environmental burden of the P recovery scenario relative to the current-day scenario, whereas a negative value (when PR < CT + CF) represented a net environmental benefit of the P recovery scenario relative to the current-day scenario.

When focusing on the three P recovery scenarios, we found that PR_3 usually had the largest environmental impacts in most categories. For example, the environmental impacts on climate change for PR_1 and PR_2 were not greatly different (i.e., 83.0 kg CO_{2eq} per kg P for PR_1 and 49.9 kg CO_{2eq} per kg P for PR_2) and were much lower than that for PR_3 (255.9 kg CO_{2eq} per kg P) (Fig. 3; Table S6). This was because more N_2O and CH_4 emissions are produced in sludge spreading, both of which have stronger greenhouse effects for each molecule than CO_2 [87,88]. Regarding freshwater eutrophication impact, PR_1 and PR_2 can achieve environmental benefits with values of -2.3×10^{-3} kg P_{eq} per kg P and -8.4×10^{-4} kg P_{eq} per kg P, respectively; whereas environmental burden would be imposed by PR_3 (1.1×10^{-2} kg P_{eq} per kg P) (Fig. 3; Table S6).

Monte Carlo simulation was used to obtain the mean, standard deviation, extreme value, and median of the environmental impacts (Table S7). For some impact categories with low uncertainty (e.g., climate change, freshwater, and terrestrial acidification), the uncertainty did not change the overall property of environmental burden or benefit. For example, results indicated that the impact on freshwater and terrestrial acidification for PR_1 is bounded by a minimum value of 3.6 mol H^+_{eq} per kg P and a maximum value of 6.8 mol H^+_{eq} per kg P, resulting in the range of −26.5% to 38.8% when compared with the mean value of 4.9 mol H^+_{eq} per kg P (Table S7). For some impact categories with high uncertainty (e.g., freshwater eutrophication, carcinogenic effects), the property of environmental burden or benefit for some scenarios (e.g., PR_2, CT_2) may be changed by the uncertainty. For instance, compared with the mean value of -6.9×10^{-4} kg P_{eq} per kg P for freshwater eutrophication in PR_2, the maximum value was 2.3×10^{-2} kg P_{eq} per kg P, which could lead to a shift from environmental benefit to burden (Fig. 3; Table S7). When using standard deviation as the judgment criterion for uncertainty, the uncertainty in PR_2 had the greatest impact on LCA results when compared with the other scenarios (Fig. 3).

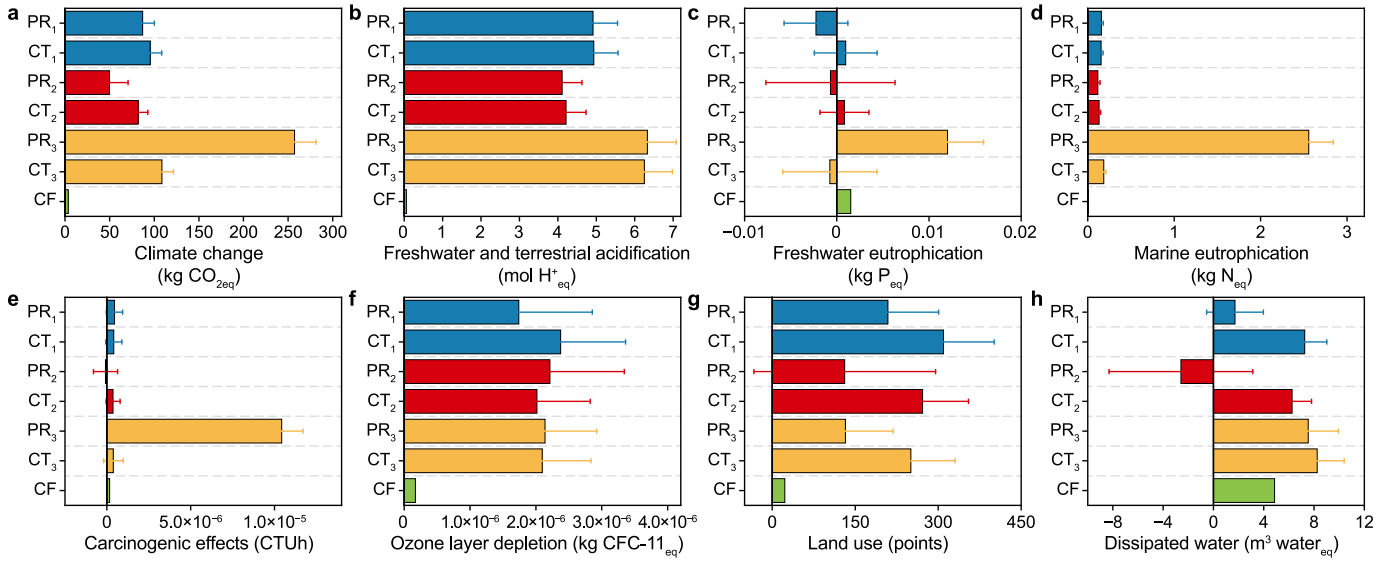

Fig. 3. Environmental and health impacts for different scenarios with uncertainty.
PR_1–PR_3: P recovery scenarios (struvite, vivianite, and treated sludge); CT_1–CT_3: Current-day treatments (sludge treatment and landfill, corresponding to the P recovery scenarios); CF: P chemical fertilizer application. **a**–**h**: Results for climate change (**a**), freshwater and terrestrial acidification (**b**), freshwater eutrophication (**c**), marine eutrophication (**d**), carcinogenic effects (**e**), ozone layer depletion (**f**), land use (**g**), and dissipated water (**h**). The bars represent the environmental impact values for each scenario, with negative values to the left of the zero axis (beneficial to the environment) and positive values to the right (harmful to the environment). Error bars represent the uncertainty. Eq: Equivalent; CTUh: Comparative toxic units for humans.

3.2. Impacts of key processes for three P recovery scenarios

Fig. 4 shows the relative importance of different processes in determining the environmental impact (detailed results are shown in Table S8). We divided these processes into AD (with CHP), dewatering, transport, sludge disposal, energy recovery, resource substitution, and other operations. Regarding climate change impacts (Fig. 4a), AD contributed 72.5% of the environmental burden of PR_1 and 63.2% of the burden of PR_2. For PR_3, the contributions from AD and sludge disposal were similar, respectively contributing 54.9% and 60.9% of the total impacts. For PR_1 and PR_2, due to electricity recovery, respective impacts of −53.2 and −45.9 kg CO_{2eq} per kg P were observed on climate change (Fig. 4a; Table S8).

For impacts on ecosystem quality, most negative impacts occurred in the disposal of the remaining sludge (Fig. 4b–d). The major impacts on freshwater and terrestrial acidification (Fig. 4b) were caused by sludge disposal, mostly because of the released ammonia. For freshwater and marine eutrophication (Fig. 4c and d), the largest impact was observed for PR_3, followed by PR_2 and PR_1. Land application of sludge can result in leaching and runoff of P and N, resulting in a larger impact on PR_3. For freshwater eutrophication, other operations (e.g., WASSTRIP®/thickening, precipitation, and land application) had large impacts, especially for PR_2, and accounted for 62.6% of the environmental burdens. The discharge of nitrate (NO_3^-) in sludge spreading was a major factor resulting in eutrophication. A large fraction of total nitrogen could be eventually transferred to the groundwater and surface water as nitrate (Hansen et al., 2006; [72]. Energy recovery and resource substitution significantly offset the environmental burdens of freshwater eutrophication in PR_1 and PR_2, which provided respective environmental benefits of -1.3×10^{-2} and -1.9×10^{-2} kg P_{eq} per kg bioavailable P (Fig. 4c; Table S8).

For carcinogenic effects (Fig. 4e), negative impacts were caused by releasing heavy metal pollution into the soil. Among the three P recovery scenarios, PR_3 had the highest carcinogenic effect (up to 1.1×10^{-5} CTUh per kg P) relative to PR_1 and PR_2, with most of the impacts incurred in sludge disposal. The impacts on ozone layer depletion (Fig. 4f) were related to methane emissions. We found that the impacts were closely related to the dewatering and sludge disposal stages for PR_1 and PR_3 and other operations for PR_2.

For impacts on land use and dissipated water (Fig. 4g and h), most negative impacts were related to dewatering and sludge disposal for PR_1 and PR_3 and other operations for PR_2. In PR_1, the dewatering had an impact of 144.5 points per kg P, and sludge disposal had an impact of 164.0 points per kg P. In PR_2, other operations had an impact of 247.0 points per kg P. However, through energy recovery and resource substitution, environmental benefits could be obtained on land use (−266.2 to −457.9 points per kg P for PR_1–PR_3) and dissipated water (−10.4 to −21.2 m^3 $water_{eq}$ per kg P) (Fig. 4g and h; Table S8).

3.3. Life cycle costing analysis

Internal costs for PR_1–PR_3 ranged from 87.2 to 159.9 CNY per kg P and were much higher than the internal costs for CF (8.2 CNY per kg P) (Table 2). However, when considering current-day treatments, the gaps between PR and CT + CF in the internal costs could be greatly reduced (Table 2). For PR_1, the internal cost was estimated to be 101.5–136.1 CNY per kg P, while the internal cost for CT_1+CF was 93.6–122.7 CNY per kg P. Similarly, for PR_2, the internal cost was 133.5–159.9 CNY per kg P, while that of CT_2+CF was 86.7–102.2 CNY per kg P. Conversely, the internal cost of PR_3 (87.2–121.9 CNY per kg P) was lower than that of CT_3+CF (115.7–158.6 CNY per kg P), mainly because the sludge landfill cost was not included in PR_3. Most internal costs were incurred in the operation process for the P recovery scenarios, whereas the transport cost was relatively low (3.5–6.1 CNY per kg P) (Table 2). Regardless of the P recovery scenario, energy consumption was usually responsible for most operation costs (34–48%). In addition, given that not all sludge could be reused, the disposal of the remaining sludge made up 28–40% of operation costs. However, when considering the potential benefits (energy recovery and resource substitution), we found that the net costs for PR_1 (59.1–79.2 CNY per kg P) and PR_2 (76.5–91.6 CNY per kg P) were

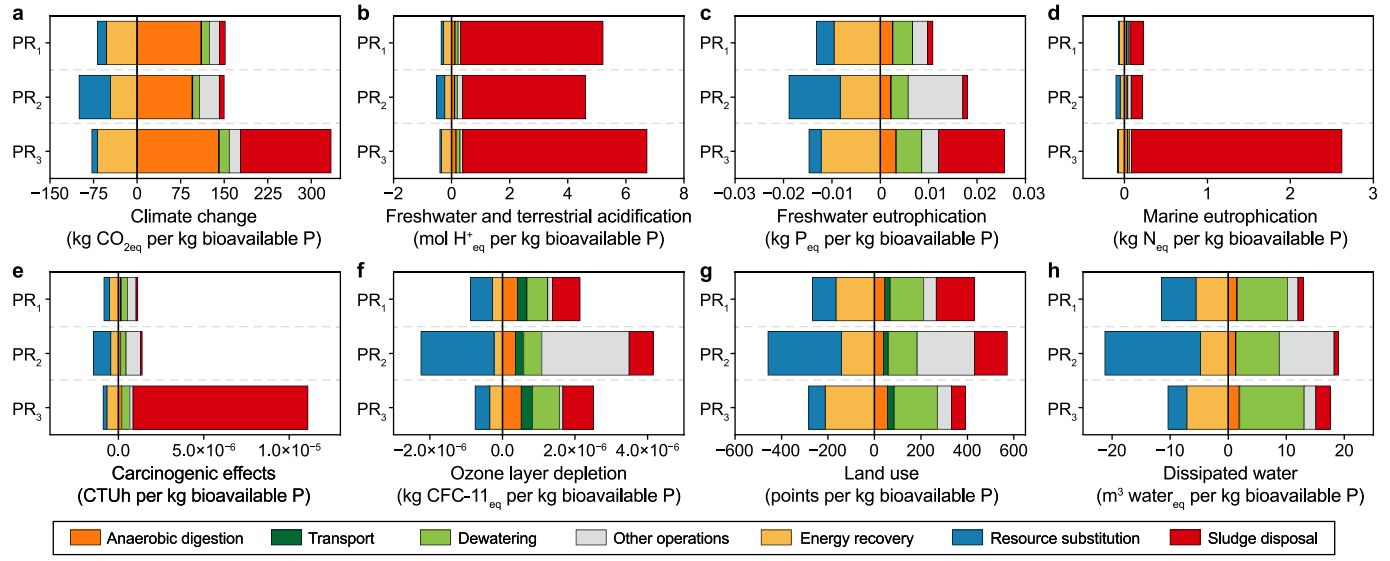

Fig. 4. Contributions of different processes on environmental and human health impacts under three P recovery scenarios. PR_1–PR_3: P recovery scenarios (struvite, vivianite, and treated sludge). **a**–**h**: Environmental and human health impacts for climate change (**a**), freshwater and terrestrial acidification (**b**), freshwater eutrophication (**c**), marine eutrophication (**d**), carcinogenic effects (**e**), ozone layer depletion (**f**), land use (**g**), and dissipated water (**h**). Different color bars represent different processes (anaerobic digestion, dewatering, transport, sludge disposal, energy recovery, resource substitution, and other operations, respectively). Other operations include WASSTRIP®/thickening, precipitation, land application, and other processes. The area on the left side of the zero axis indicates environmental benefit; the area on the right side of the zero axis indicates environmental burden. Eq: Equivalent; CTUh: Comparative toxic units for humans.

Table 2
The internal costs and benefits under different scenarios.

Scenario	Functional unit	Fixed cost	Operating costs			Transport cost	Internal cost	Benefits		Net cost		Unit
			Chemicals	Energy	Disposal			Energy	Resources	Min	Max	
PR_1	1 kg bioavailable P	37.4–50.2	15.0–20.0	24.9–33.4	20.5–27.5	3.7–5.0	101.5–136.1	30.0–40.2	12.4–16.7	59.1	79.2	CNY
PR_2	1 kg bioavailable P	34.5–41.3	51.9–62.2	24.7–29.6	18.8–22.6	3.5–4.2	133.5–159.9	27.6–33.0	29.4–35.3	76.5	91.6	CNY
PR_3	1 kg bioavailable P	39.2–54.8	7.7–10.7	30.8–43.0	5.2–7.2	4.4–6.1	87.2–121.9	37.5–52.4	10.6–14.8	39.1	54.7	CNY
CT_1	1 kg bioavailable P	31.9–42.8	9.2–12.3	20.3–27.2	20.5–27.5	3.5–4.7	85.4–114.5	30.0–40.2	0.0	55.4	74.3	CNY
CT_2	1 kg bioavailable P	29.4–35.2	8.5–10.1	18.6–22.3	18.8–22.6	3.2–3.8	78.5–94.0	27.6–33.0	0.0	50.9	60.9	CNY
CT_3	1 kg bioavailable P	39.2–54.8	11.5–16.1	26.9–37.5	25.6–35.9	4.3–6.1	107.5–150.4	37.5–52.4	0.0	70.0	97.9	CNY
CF	1 kg bioavailable P	-	-	-	-	-	8.2[a]	-	-	8.2[a]		CNY

[a] The cost of mining phosphate rock to produce and use 1 kg bioavailable P.

close to those for CT_1+CF (63.6–82.5 CNY per kg P) and CT_2+CF (59.1–69.1 CNY per kg P), respectively, while the net cost for PR_3 (39.1–54.7 CNY per kg P) was much lower than that for CT_3+CF (78.2–106.1 CNY per kg P). Compared with the current-day scenarios, both PR_1 (PR_1: 59.1–79.2 CNY per kg P < CT_1+CF: 63.6–82.5 CNY per kg P; that is $PR_1-CT_1-CF < 0$) and PR_3 (PR_3: 39.1–54.7 CNY per kg P < CT_3+CF: 78.2–106.1 CNY per kg P; that is $PR_3-CT_3-CF < 0$) had net benefits, which means they could be feasible scenarios.

As shown in Fig. 5, we further monetized the environmental impacts for different P recovery scenarios based on the information in Table S4. External costs for PR_1–PR_3 ranged from 366.8 to 924.3 CNY per kg P, which were also much higher than that of CF (11.7 CNY per kg P) (Fig. 5). PR_2 had the lowest external costs (366.8 CNY per kg P) among the three P recovery scenarios, whereas PR_3 had the highest external costs (924.3 CNY per kg P), mainly because of the large environmental burdens in sludge spreading. Compared with the P recovery scenarios, the current-day scenarios usually had higher external costs except CT_3+CF (CT_1+CF: 471.1 CNY per kg P; CT_2+CF: 408.2 CNY per kg P and CT_3+CF: 591.6 CNY per kg P). For almost all scenarios, external costs mainly came from the two impact groups of climate change and ecosystem quality (Fig. 5), which could account for more than 95% of the total costs. By contrast, external costs related to human health and resources were much smaller (Fig. 5). Compared with the current-day scenarios, the results indicated that PR_1 (PR_1-CT_1-CF: −13.6 CNY per kg P) and PR_2 (PR_2-CT_2-CF: −41.4 CNY per kg P) could be feasible scenarios.

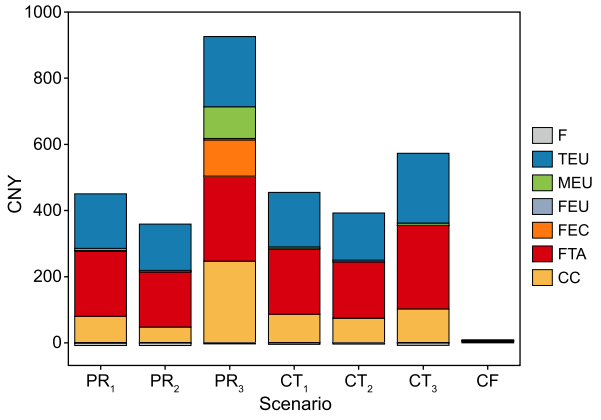

Fig. 5. External costs caused by environmental impacts for each scenario. Only the environmental impacts with higher external costs are listed. PR_1–PR_3: P recovery scenarios (struvite, vivianite, and treated sludge); CT_1–CT_3: Current-day treatments (sludge treatment and landfill, corresponding to the P recovery scenarios); CF: P chemical fertilizer application. CC: Climate change; FTA: Freshwater and terrestrial acidification; FEC: Freshwater ecotoxicity; FEU: Freshwater eutrophication; MEU: Marine eutrophication; TEU: Terrestrial eutrophication; F: Fossils.

When summing the internal costs, benefits, and external costs, the societal costs for PR_1 (526.7 CNY per kg P) and PR_2 (450.9 CNY per kg P) were lower than or comparable with those for CT_1+CF (544.2 CNY per kg P) and CT_2+CF (472.3 CNY per kg P). However, the societal cost for PR_3 was much higher (971.2 CNY per kg P) than CT_3+CF (683.7 CNY per kg P). In all cases, the societal cost of CF was the lowest. This result suggests that if only the replacement of P chemical fertilizer was considered, P recovery might have fairly high societal costs. However, if the benefits of replacing current-day treatments are included, the societal costs would be lower for the P recovery scenarios. Among the three P recovery scenarios, more benefits can be expected by P recovery through struvite and vivianite relative to the treated sludge. At the same time, considering the scenario feasibility of the net and external costs, it is clear that recovering struvite as P fertilizer has the highest societal feasibility.

3.4. Sensitivity analysis

Major sensitive inventory parameters that significantly affected the environmental indicators when the input variables varied by ±10% are listed in Table S9 [86]. Fig. 6 displays the sensitivity coefficients of the chemicals, energy, and other consumption in eight different environmental impact categories. Chemicals and energy consumption were sensitive factors for some environmental impacts such as dissipated water, climate change, and eutrophication. Some of the sensitivity coefficients were close to 80%. By contrast, the sensitivity of other consumption was much lower, mainly ranging between 5% and 25%. Overall, the results showed that all the sensitivity coefficients were positive, indicating that environmental impacts change in the same direction as the input parameters (Table S9). Furthermore, electricity was the parameter that caused the largest variation in results; for example, the sensitivity coefficient for electricity consumption on climate change was 66.3% (PR_1), 51.9% (PR_2), and 71.6% (PR_3) (Table S9). Moreover, changes in energy input parameters had larger impacts on climate change, acidification, and eutrophication (Fig. 6). In addition, the chemicals in PR_1 and PR_2 were sensitive to some environmental impacts, such as carcinogenic effects and freshwater eutrophication. A 10% increase in chemicals resulted in sensitivity coefficients of 61.0% (PR_1), 70.7% (PR_2), and 21.3% (PR_3) for the carcinogenic effects category (Fig. 6).

4. Discussion

4.1. P recovery potential from sewage sludge in China

As a byproduct of biological wastewater treatment, sewage sludge contains putrescible organic matter, nutrients, and pollutants derived from human waste; therefore, it displays dual attributes of “pollutant” and “resource” [44]. Compared with other

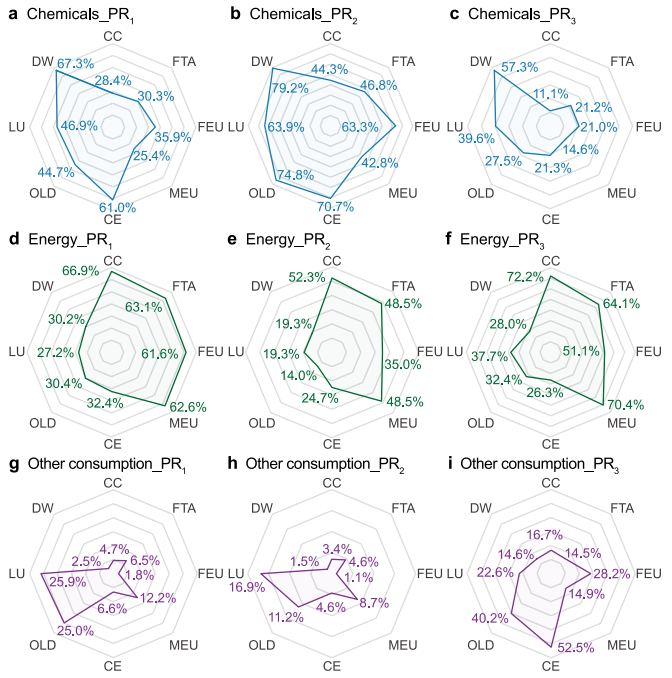

Fig. 6. Sensitivity analysis of the key parameters in the input inventory. PR_1–PR_3: P recovery scenarios (struvite, vivianite, and treated sludge). Chemicals, energy, and other consumption refer to chemical consumption, energy consumption, and other categories of consumption in the whole process, respectively. The sensitivity increases gradually from the center to the periphery. CC: Climate change; FTA: Freshwater and terrestrial acidification; FEU: Freshwater eutrophication; MEU: Marine eutrophication; CE: Carcinogenic effects; OLD: Ozone layer depletion; LU: Land use; DW: Dissipated water.

developed countries, sludge treatment and disposal in China started much later, and there are still issues regarding the safe disposal and resource recovery of sludge [89]. For technical or economic reasons, landfill is still the dominant method of sludge disposal in most provinces [90,91]. Furthermore, the large amount of sludge from WWTPs returns to the environment without appropriate disposal [92]. However, in the past decade, a series of advances in technologies and processing equipment have emerged in the areas of biological stabilization, resource utilization, dewatering, and co-digestion [93–96]. For example, advanced anaerobic digestion demonstration projects are already operating in Beijing, Zhengzhou, and Shanghai [44].

It is estimated that the residual P in sludge is equivalent to about 15% of the national consumption of agricultural P fertilizers in China. Currently, there are over 6000 WWTPs in operation, which are widely distributed across the country (Fig. S3). With wastewater treatments, around 6.0×10^{10} kg of sewage sludge is produced every year [97]. The P loading in sewage sludge in different areas in China is shown in Fig. S4. Based on influent and effluent P concentrations and the volume of wastewater treated at each facility [97], we estimate that about 2.2×10^8 kg of P remains in sludge each year. However, the amount of P remaining in sludge can vary substantially among different facilities or areas because of differences in the treatment capacity and local dietary habits [98]. Based on the P recovery efficiency of three scenarios (i.e., 45–55% in struvite [39], 50–60% in vivianite [56], 45–55% in treated sludge [35], Fig. 2), we estimated that 9.7×10^7–1.3×10^8 kg of P could be recovered from sludge on a national scale. However, on a local scale, the potential for P recovery relative to the agricultural P demand can fluctuate significantly and depends on the extent of the "co-location" of urban nutrient supplies and agricultural demands [17,99]. It is clear that areas with high P recovery potential always include an urban center with a large wastewater treatment facility, but the extent of local agricultural production is relatively limited [99]. For example, in Beijing, about 4.4×10^6–5.8×10^6 kg of P could be recovered from sludge, whereas agricultural P demand is only 3.0×10^6 kg (Fig. S5). Co-location of P recovery and consumption would indicate a higher likelihood of adding recovered P onto croplands [17,100]. Therefore, spatial planning would be necessary to increase the viability of P recovery from sewage sludge [4].

4.2. Selection of scenarios for P recovery from sewage sludge

Among the three potential scenarios discussed in this study (i.e., struvite, vivianite, and treated sludge), treated sludge was identified as the scenario with the lowest internal and net costs (Table 2), but its external cost was the highest (Fig. 5). In fact, using recycled human excreta on croplands was a common practice in China before 1980 [101]. The use of a special "dung stamp" was once quite popular, whereby farmers exchanged dung from urban residents and applied them to crops after some simple treatments [101]. In some African and European countries, the spreading of treated sludge had also been practiced for a long period [23]. Compared with P recovery through struvite (59.1–79.2 CNY per kg P) or vivianite (76.5–91.6 CNY per kg P), the net cost for treated sludge (39.1–54.7 CNY per kg P) is much lower (Table 2). Previous concerns around sludge application were primarily based on the presence of pathogens and heavy metals in sludge [67]. However, emerging studies have suggested that these risks could be low after some elementary treatment [102,103]. It is undeniable that sludge application still causes significant damage to the environment even now, and this is reflected by its high external costs. In the future, attention should be paid to emerging pollutants that are not destroyed or degraded, because this can greatly increase the externality of pollution, even if the magnitude is very low.

Despite progress in treatment technologies, the net costs for P recovery from sludge are still much higher than that for the use of P chemical fertilizer (e.g., 59.1–79.2 CNY per kg P for the struvite vs. 8.2 CNY per kg P for P chemical fertilizer). This result is consistent with many current international studies, which conclude that the economic costs of most P recovery pathways from sewage sludge are higher relative to mined P fertilizers [36,104]. However, we found that the societal costs for P recovery scenarios are not as high as expected when we considered the benefits of replacing the current-day treatments. There is a standpoint that the positive benefit of P recovery from sludge cannot offset the environmental burden and operational costs in some studies [35,53,105]. However, when we considered the societal costs, we found that the costs for P recovery in the form of struvite or vivianite are much lower than the current-day scenarios. Moreover, China has a large sludge production volume relative to other countries, which has huge potential for P recovery from sludge and the possibility of economies of scale to further reduce costs. Considering the large investment required for facilities such as anaerobic digesters and WASSTRIP® and Pearl® equipment [31], it is not feasible for all WWTPs to implement P recovery, especially for small or medium-sized WWTPs [106]. For small WWTPs, transporting sewage sludge to larger plants or taking conventional sludge treatment may be more economical.

4.3. Implications for sustainable P management in China

Sustainable P management is crucial to food security and environmental protection [107,108]. China has an important role in achieving the planetary boundaries of nutrient flows [109],

considering that China has accounted for about one-third of the global P fertilizer consumption during the past decade [110,111]. China also faces serious water pollution issues (e.g., eutrophication, and algal blooms) caused by cropland nutrient losses [47,112]. As a key part of the UN SDGs, a primary objective of wastewater treatment is to remove contaminants such as pathogens, nutrients, and organic matter [98,113]. Unlike N removal, which can proceed by conversion to a gaseous form, P removal from the wastewater is a process that involves coagulation, flocculation, and sedimentation [97]. As a result, the majority of P in wastewater (up to 90%) remains in the sludge, which provides opportunity for P recovery. In recent years, several strategies have been proposed to help reduce the use of P chemical fertilizers, including refined management of animal feces [114] and reuse of crop residues [115] and food waste [116]. Compared with these measures, P recovery from sewage sludge has the following advantages. First, WWTPs are highly centralized and urban sewer systems provide a means of collecting human waste for treatment in WWTPs. This configuration avoids the additional costs of collection and transport of biogenic materials. Second, P recovery technologies from sludge have been in development for over a decade and some technologies have matured or been put into practice (e.g., precipitation after anaerobic or aerobic digestion [117], acid leaching from incinerated sludge ash [118], and chemical extraction from sludge-based biochar prepared by pyrolysis or using the biochar as a soil amendment [119]). Third, wastewater treatment has been highly capitalized, which should assist local companies in identifying business opportunities. It should be noted that P recovery from sludge will not replace all P fertilizers in agriculture, and the recovery potential depends on cropland distribution and the capacity of surrounding WWTPs. Spatial mismatch between supply and demand would increase transport costs and reduce the feasibility of recovering P from the sludge. Transport over long distances would make the reuse of products more expensive, placing greater pressure on cities to recover highly concentrated products by applying more complicated processes. P recovery from sludge will not be a “one-size-fits-all solution” that can meet all the local agricultural P demands in China, but it has the potential to mitigate the “P crisis” in some areas. Furthermore, incentives through policy reform and financial support should be implemented to increase the feasibility and opportunity for P recovery and utilization. There is a critical need for policy reform to introduce a regulatory framework that boosts the use of recovered P products as alternatives to phosphate rock [4].

4.4. Limitations of this study

There are several limitations in this study. First, although techniques to recover P from sludge (e.g., struvite precipitation) have been developed and implemented in many recovery facilities, similar techniques have not yet been implemented in China. In addition, although P recovery as vivianite has been well studied in laboratory studies and pilot tests, it has not been implemented on a large scale [9]. This circumstance may lead to modifications of production processes in localizing technologies in China. Second, when estimating the external costs for different impacts, the lack of data in China required shadow prices on environmental impacts to be referenced from European studies. Third, some input variables (e.g., diesel, lime, and transport) were derived from European datasets, which might affect the estimations of environmental impacts and societal costs.

5. Conclusions

A large reserve of P remains in sewage sludge in China, with the amount estimated to be equivalent to about 15% of the national consumption of agricultural P fertilizers. Disposal of sludge in landfill is common in China and is a practice that neglects the potential benefits of P recovery from sewage sludge. We found that P recovery as struvite and vivianite have low environmental impacts, whereas P recovery as treated sludge has higher impacts in most categories. By comparing the impacts of different processes in the three P recovery scenarios, the key contributing processes can be identified. In terms of costs, we found that societal costs for production and land use of 1 kg P by P recovery from sewage sludge are much higher than those of P chemical fertilizer. However, when considering the costs of the current-day treatments, the societal costs of P recovery scenarios are close to or slightly lower than those of current-day scenarios. Among the three P recovery scenarios, we found that P recovery as struvite and use as P fertilizer has the highest societal feasibility. The feasibility of P recovery as vivianite, which has high added value, could be enhanced through technological advances. In China, further experiments and studies are needed on the application of treated sludge because the treated sludge scenario has the lowest internal costs but has the highest societal costs if considering the negative impacts on the environment. Although applications of P recovery from sewage sludge may be constrained by economic or technical conditions, we found that the societal costs of P recovery scenarios are not as high as expected when including the benefits of replacing the current-day treatments.

CRediT authorship contribution statement

Jiawen Xie: Conceptualization, Methodology, Software, Validation, Formal analysis, Writing - Original Draft, Visualization. **Xingchen Zhuge**: Software, Formal analysis, Investigation. **Xixi Liu**: Validation, Investigation. **Qian Zhang**: Methodology, Resources. **Yiwen Liu**: Supervision. **Peizhe Sun**: Supervision. **Yingxin Zhao**: Writing - Review & Editing, Funding acquisition. **Yindong Tong**: Conceptualization, Methodology, Writing - Review & Editing, Funding acquisition.

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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