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Trapped Urban Phosphorus: An Overlooked and Inaccessible Stock in the Anthropogenic Phosphorus Cycle

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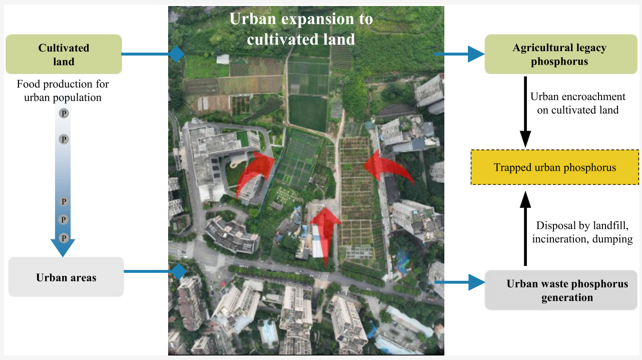

ABSTRACT: Urban landscapes are high phosphorus (P) consumption areas and consequently generate substantial P-containing urban solid waste (domestic kitchen wastes, animal bones, and municipal sludge), due to large population. However, urbanization can also trap P through cultivated land loss and urban solid waste disposal. Trapped urban P is an overlooked and inaccessible P stock. Here, we studied how urbanization contributes to trapped urban P and how it affects the P cycle. We take China as a case study. Our results showed that China generated a total of 13 (±0.9) Tg urban trapped P between 1992−2019. This amounts to 6 (±0.5) % of the total consumed P and 9 (±0.6) % of the chemical fertilizer P used in China over that period. The loss of cultivated land accounted for 15% of the trapped urban P, and half of this was concentrated in three provinces: Shandong, Henan, and Hebei. This is primarily since nearly one-third of the newly expanded urban areas are located within these provinces. The remaining 85% of trapped urban P was associated with urban solid waste disposal. Our findings call for more actions to preserve fertile cultivated land and promote P recovery from urban solid waste through sound waste classification and recycling systems to minimize P trapped in urban areas.

KEYWORDS: *urban expansion, waste disposal, legacy phosphorus, recycling, crop production*

■ INTRODUCTION

Urbanization, the ongoing migration of people from rural to urban areas, is a worldwide phenomenon. Urbanization usually implies urban land expansion and population growth in cities. Globally, the urban population has increased from 2.3 to 4.3 billion between 1992 and 2019,[1] with an additional 2.5 billion projected by 2050. Urban land has expanded, yet by 2020, it accounted for only about 1% of the world's land area.[2] Despite occupying a small percentage of the total land area, urban expansion remained a significant diver of global land use change. Between 1992 and 2015, global urban land had more than doubled.[3] Urbanization has notably altered the anthropogenic phosphorus (P) cycle,[4] much like its considerable impact on habitat loss[5] and climate change,[5,6] both of which have long been issues of public concern.

The anthropogenic P cycles involves the extraction of P from rocks and its subsequent distribution in fertilizers, animal feeds, agricultural crops, animal products, various industrial goods, and the environment.[7] Most of the P is first consumed in cropland and specifically used to produce food for urban populations. Urban areas emerge as significant consumers of P, particularly through the consumption of food and detergents, generating a substantial amount of P waste, such as human excreta and food waste. Multiple P stocks exist in the urbanization-driven anthropogenic P cycle, which are seen as potential reserves for crop production. Considerable advances in P stocks in the anthropogenic P cycle have been documented.[8] For example, MacDonald et al.[9] evaluated the global agricultural soil P balance, revealing a median soil P surplus of 26 kg year^{-1} in East Asia, Western and Southern Europe, and the coastal United States in 2000. This soil P surplus continued to rise in certain regions, notably in China.[10] Le Noë, et al.[11] indicated that the P stock in French agricultural soil could support crop production for up to 60 years even without mineral P fertilization. Waste from human consumption (human excreta, solid waste, and wastewater) in China was estimated to be 0.6 Tg in 2015.[12] However, not all P stocks are readily recoverable for crop production. Inaccessible P stock will challenge the sustainability of the anthropogenic P cycle. Yet, there is limited understanding as to how urbanization contributes to an inaccessible P stock.

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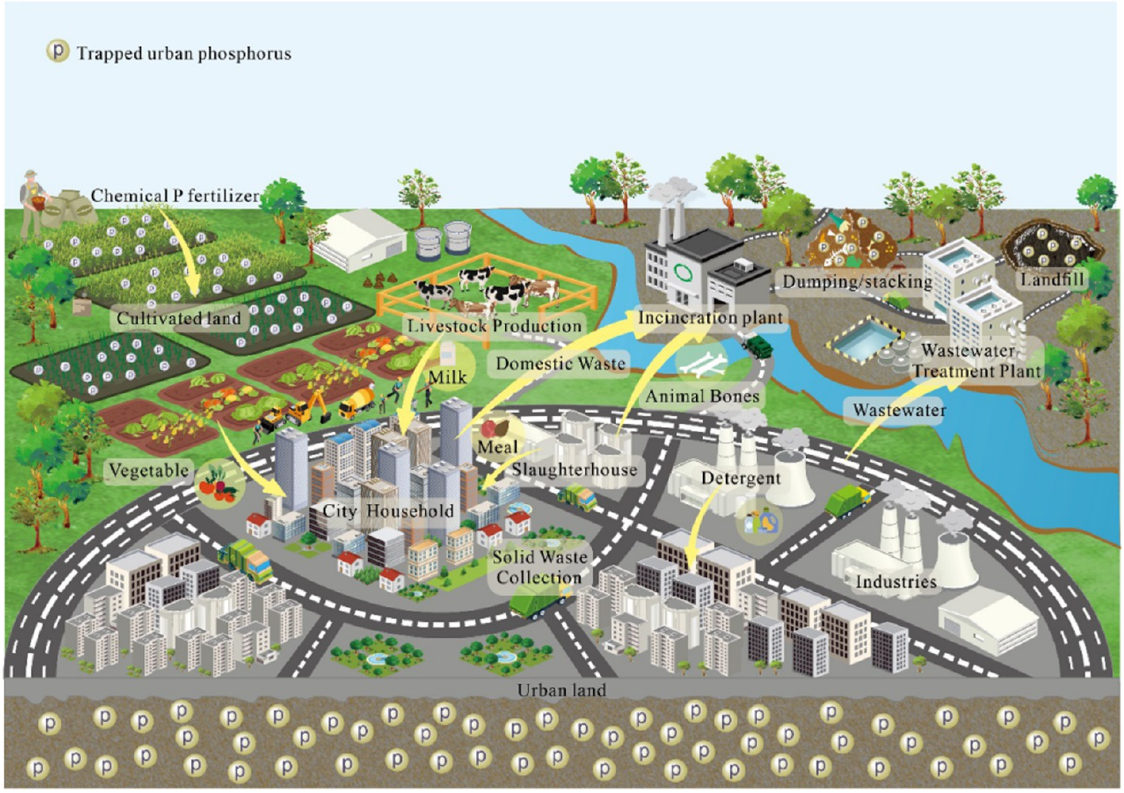

Figure 1. Trapped urban phosphorus (P): a conceptual framework illustrating how P accumulates in urban stocks (in both land and solid waste). We define trapped urban P as the P that accumulates in urban stocks (P in the urban land soil, and P in incinerated or dumped waste often discarded in landfills), which cannot be readily recovered in the short term.

Urbanization can lead to P being trapped and becoming an inaccessible stock in two ways. First, modern intensive agriculture relies heavily on P fertilizers sourced from phosphate rocks and recycled P to maximize crop yield. In many regions (e.g., East Asia, Southern Europe, and Southern Brazil), overuse of P fertilizers has led to its substantial P accumulation in cultivated agricultural land.[9,13] This overloading was particularly common in peri-urban farming systems, where agricultural practices and P fertilizer overuse have persisted for many years.[14] However, urban expansion often occurred at the expense of cultivated land. Approximately 70% of global urban land expansion has taken place on former cultivated land.[15] This phenomenon was particularly acute in Asia and Africa,[16] where population growth was much more rapid than in other regions. When cultivated land becomes urban land, the accumulated P in the cultivated land is trapped in urban P stocks. This part of P is inaccessible for crop production, thus breaking the P cycle, rendering sustainable P use challenging (Figure 1).

A second way of urban P trapping occurs through the disposal of P-containing urban solid waste, such as domestic solid waste, animal bones, and municipal sludge. Recovering P from these urban solid wastes and transforming them into marketable fertilizers for reuse in crop production is technically demanding but feasible.[17] Except for certain European nations like Switzerland and Germany, very few countries have regulations mandating the recovery and recycling of P wastewater treatment facilities. In many countries globally, P from urban solid waste usually ends up in underground landfills,[18,19] or after incineration, and is incorporated into construction materials, rendering a significant amount of P being locked into buildings and other artificial urban structures. Collectively, modern techniques for managing urban solid waste lead to substantial quantities of P being trapped in urban stocks (Figure S1). Consequently, these trapped P resources in urban become an inaccessible stock of the P cycle that is needed for crop production (Figure 1). It remains unclear to what extent the urban expansion and urban solid waste disposal are causing P to be trapped.

In this study, we define *trapped urban P* as the P that accumulates in urban stocks (P in soil under the city, and P in incinerated or dumped waste often discarded in landfills ultimately), which cannot be readily recovered in the short term. Here, we studied how urbanization generates trapped urban P and how it affects the P cycle, taking China as a case study. China is a representative case of emerging countries that have experienced fast urban expansion and large rural-urban migration at an unprecedented scale in the last decades. In addition, China's cultivated land experienced a large cumulative P surplus due to widespread and excessive fertilization.[13] To this end, we performed a time-series analysis of trapped urban P by cultivated land loss and urban solid waste disposal in a spatially explicit way from 1992 to 2019 in China. The year 1992 was a turning point in China's economic reform and urban development.[20] This study thus greatly advanced our quantitative understanding of the significance of trapped urban P in the anthropogenic P cycle.

■ MATERIALS AND METHODS

The trapped urban P arose from cultivated land loss and urban solid waste disposal. This section presented the description of the implemented modeling approach, quantifying trapped

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urban P from 1992 to 2019 in China. First, we produced high-resolution (300 m × 300 m) maps of legacy P in China's cultivated land for the years 1992–2019. Legacy P is cumulative P surplus in the cultivated land, without considering P accumulation before 1992 and native soil P. P surplus is based on P inputs in the form of fertilizer and manure and P outflows in the form of crop harvesting. Second, the annual legacy P maps were overlaid on the urban land expansion maps to quantify trapped urban P from cultivated land loss caused by the expansion of urban areas. The land cover maps with a spatial resolution of 300 m (1992–2019) provided by the European Space Agency-Climate Change Initiative (ESA-CCI), were used to determine urban land expansion onto cultivated land. Third, we implemented the Hierarchical national P Cycle model (HPC model)[12] to quantify the trapped urban P through urban solid waste disposal at the province scale in China from 1992 to 2019. The data sources and the methodology of analysis were described below.

Quantification of Trapped Urban P from Cultivated Land Loss. In this study, we quantified trapped urban P from cultivated land loss at a spatial resolution of 300 m for each year from 1992 to 2019 in China. For each grid (300 m × 300 m), we calculated the amount of trapped urban P by multiplying the area of cultivated land loss and the legacy P per unit area. Thus, this calculation was built on the high-resolution (300 m × 300 m) maps of cultivated land loss due to urban expansion and legacy P in the cultivated land affected. The steps to make high-resolution maps of cultivated land loss were as follows (Figure S2).

Cultivated Land Loss due to Urban Expansion. We analyzed the yearly change in urban land expansion between 1992 to 2019, using the continuous land cover data sets from the European Space Agency (ESA).[21] The land cover maps for the years 1992–2015 were produced by the ESA-CCI Land Cover Project. The land cover map for 2016–2019 was produced in the context of the Copernicus Climate Change Service (C3S). The C3S global Land Cover maps between 2016 and 2019 ensured continuity with the existing ESA global annual Land Cover maps from 1992 to 2015. These land cover maps were derived from multiple sensors and presented at a spatial resolution of 300 m for each year. The original land cover maps were presented with 22 classes using the United Nations (UN) Land Cover Classification System (LCCS). We grouped them into six categories: cultivated land, forest, grassland, urban land, wetland, and others (e.g., water bodies), following the Intergovernmental Panel on Climate Change (IPCC) land categories. The correspondence between the IPCC land categories and the LCCS land class can be found in the Land Cover CCI Product User Guide.[22] We overlaid each year's land cover map with the previous year's map to define maps of the urban cover change. All grids that were classified as cultivated land in the previous year and converted into urban land in the study year were treated as the loss of cultivated land due to urban expansion. The total area by each land cover or land change map per year was quantified by considering different cell sizes based on latitude and longitude (km^2).

Legacy P in the Cultivated Land. To be consistent with the land cover analysis, we built legacy P maps with a spatial resolution of 300 m between 1992 and 2019 in China. These maps were built in three steps. First, we assessed the total annual P surplus (kg) for cultivated land for all counties (2000–2019) or provinces (1992–1999) of China. The inconsistency in spatial analysis of P surplus during the different periods was mainly due to the availability of data. We calculated the P surplus (kg) for cultivated land as the sum of chemical (P_{fert}) and manure P (P_{man}) fertilizer minus P removal by crop harvested (P_{remo}). The equation to quantify P surplus for each county or province is shown below.

$$P_{surplus} = P_{fert} + P_{man} - P_{remo} \tag{1}$$

The data on chemical P fertilizer used from 1992 to 1999 were based on the agriculture consumption obtained from provincial statistics, while the application of chemical P fertilizer from 2000 to 2019 was collected from various municipal or county-level statistics. The P inputs from manure were estimated with animal excretion P (Table S6) and the recycling rate of manure (Table S4). We included animal manure of cattle, pigs, sheep, and poultry. P removal from cultivated land by crop harvesting constitutes the P in crop grain and the crop straw removed from the cultivated land (Figure S3). For calculating P removal from cultivated land by crop harvesting, statistical crop yield of counties of China and some parameters (e.g., P content in grain and straw among different crops (Table S3)) were used. In our calculations, we considered rice, maize, wheat, beans, cotton, flax, oil crops, sugar crops, potatoes, vegetables, and fruits. The details of the calculation and relevant parameters used were presented in Section 2 and Tables S1–S6 in the Supporting Information.

Second, we quantified the annual P surplus per unit cultivated land (kg ha^{-1}) for each county (2000–2019) or province (1992–1999) in China, using the total P surplus divided by the total cultivated land area of the respective county or province. The cultivated land areas were calculated using land cover maps from the ESA. We extrapolated the annual high-resolution (300 m × 300 m) data set of P surplus per unit cultivated land from the province and county scale calculations, assuming homogeneous P distribution within a province (1992–1999) or county (2000–2019). Finally, annual high-resolution (300 m × 300 m) legacy P maps (kg ha^{-1}) were generated from the cumulative P surplus starting from 1992. The cultivated land with a negative cumulative P did not produce trapped urban P.

Quantification of Trapped Urban P through Urban Solid Waste Disposal. The HPC model used for trapped urban P estimation in this study was based on the model described by Liu et al.[23] This model was built to quantify P flows and P stock changes across 14 compartments at the national scale based on substance flow analysis (Figure S4). In this study, HPC was used to quantify the continuously trapped urban P through urban solid waste disposal. However, we extended the results of the HPC model (P flow and stock) to the provinces of China and updated input data until the year 2019. In the HPC model, the P waste was generated by three anthropogenic compartments, including domestic solid waste, animal bones, and municipal sludge (Figure S4). The P waste in the form of domestic solid waste mainly depended on the annual collected weight of urban solid waste. The P waste in animal bones was based on the amount of bones treated for further disposal in the form of solid waste from animal slaughtering. The animal bones considered in this study were those generated from pig, cattle, sheep, poultry, horse, mule, donkey, and rabbit production. The P in the animal bone for disposal was quantified as total P in the animal body minus P in the edible meat, P in the animal body used to produce feed, and P in wastewater generation from slaughtering. To quantify

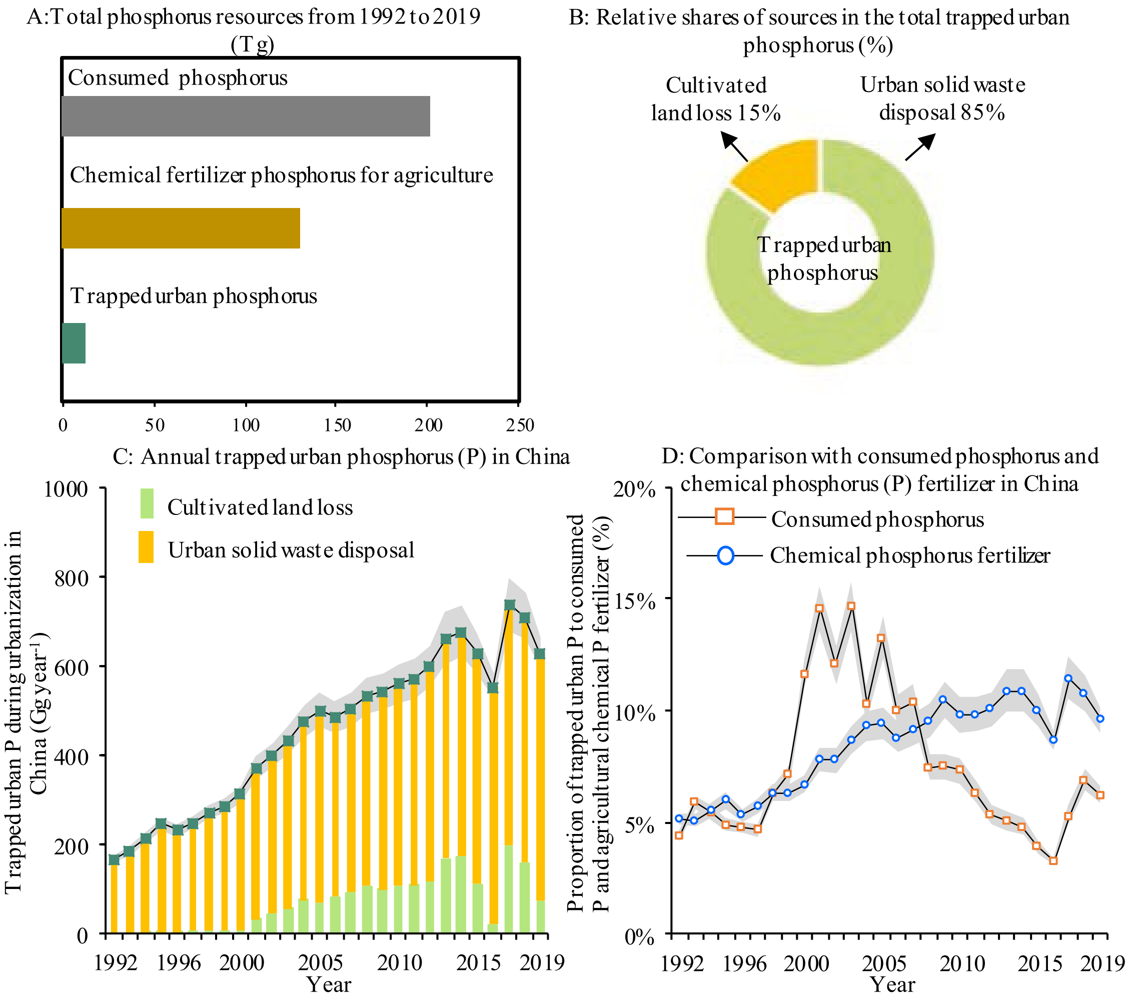

Figure 2. Trapped urban phosphorus (P) in China from 1992 to 2019. Panel A: Comparison of cumulative consumed P, total chemical fertilizer P used for agriculture, and total trapped urban P in China (sum of annual flows between 1992 and 2019 in Tg). Panel B: Relative contribution of cultivated land loss and urban solid waste disposal to the total (cumulative) trapped urban P shown in Panel A. Panel C: Annual trapped urban P (bar chart; Gg). Panel D: Proportion of trapped urban P to annual consumed P and chemical P fertilizer in China (line chart; %). Uncertainty of trapped urban P and its proportion to consumed P and chemical P fertilizer are shown in the gray shaded areas in Panel C and D. The proportion of cultivated land loss and urban solid waste disposal contributing to the total trapped urban P is also shown in the bar chart.

P waste generation in municipal sludge, wastewater from nine production processes involved in P flow was quantified (Figure S4). They were generated from the production of fertilizers, feed additives, elemental P, organophosphorus pesticides, soap and detergents, oil, meat, and urban municipal wastewater. All urban solid wastes were collected and then treated with different disposal pathways. The disposal pathways included landfill, incineration, dumping, or composting (Figure S4). This study considered the waste P to landfills, incineration, and dumping as trapped urban P. Furthermore, annual consumed P in China in this study was also quantified using the HPC model.

In this study, annual consumed P (P^{Consumed}, Gg year^{-1}) was defined as the net input of P in China, which was the sum of total P from locally mined phosphate rocks and net export P of different products (rock, fertilizer, feed additive, elemental P, pesticide, and detergent) by China to the world. The equation to quantify consumed P is as follows.

$$\begin{aligned} P^{\mathrm{Consumed}} = {} & D^{\mathrm{PR}} \times P^{\mathrm{PR}} - D^{\mathrm{PR,ne}} \times P^{\mathrm{PR}} - D^{\mathrm{FT,ne}} \times P^{\mathrm{FT}} \\ & \times \frac{62}{142} - D^{\mathrm{FA,ne}} \times P^{\mathrm{FA}} - D^{\mathrm{EP,ne}} \\ & - D^{\mathrm{P,ne}} \times P^{\mathrm{OP,1}} \times P^{\mathrm{OP,2}} - D^{\mathrm{DG,ne}} \\ & \times (P^{\mathrm{DG,1}} \times P^{\mathrm{DG,2}} + (1 - P^{\mathrm{DG,1}}) \times P^{\mathrm{DG,3}}) \end{aligned} \tag{2}$$

Where D^{PR} is the production of mined phosphate rocks (Gg year^{-1}). P^{PR} is the P content of mined phosphate rocks (0−1). $D^{\mathrm{PR,ne}}$ is the net export of mined phosphate rocks. $D^{\mathrm{FT,ne}}$ is the net export of P fertilizer (P_2O_5) (Gg year^{-1}). P^{FT} is the P_2O_5 content in P fertilizers produced (0−1). $D^{\mathrm{FA,ne}}$ is the net export of feed additives (Gg year^{-1}). P^{FA} is the P content in feed additives (0−1). $D^{\mathrm{EP,ne}}$ is the net export of element P (Gg year^{-1}). $D^{\mathrm{P,ne}}$ is the net export of pesticides (Gg year^{-1}). $P^{\mathrm{OP,1}}$ is the fraction of organophosphorus pesticides in the production of pesticides (0−1). $P^{\mathrm{OP,2}}$ is the P content of organophosphorus pesticides (0−1). $D^{\mathrm{DG,ne}}$ is the net export of detergents including soap (Gg year^{-1}). $P^{\mathrm{DG,1}}$ is the fraction of P-free detergent in the detergent production (0−1). P-free detergent is an alternative detergent containing sodium

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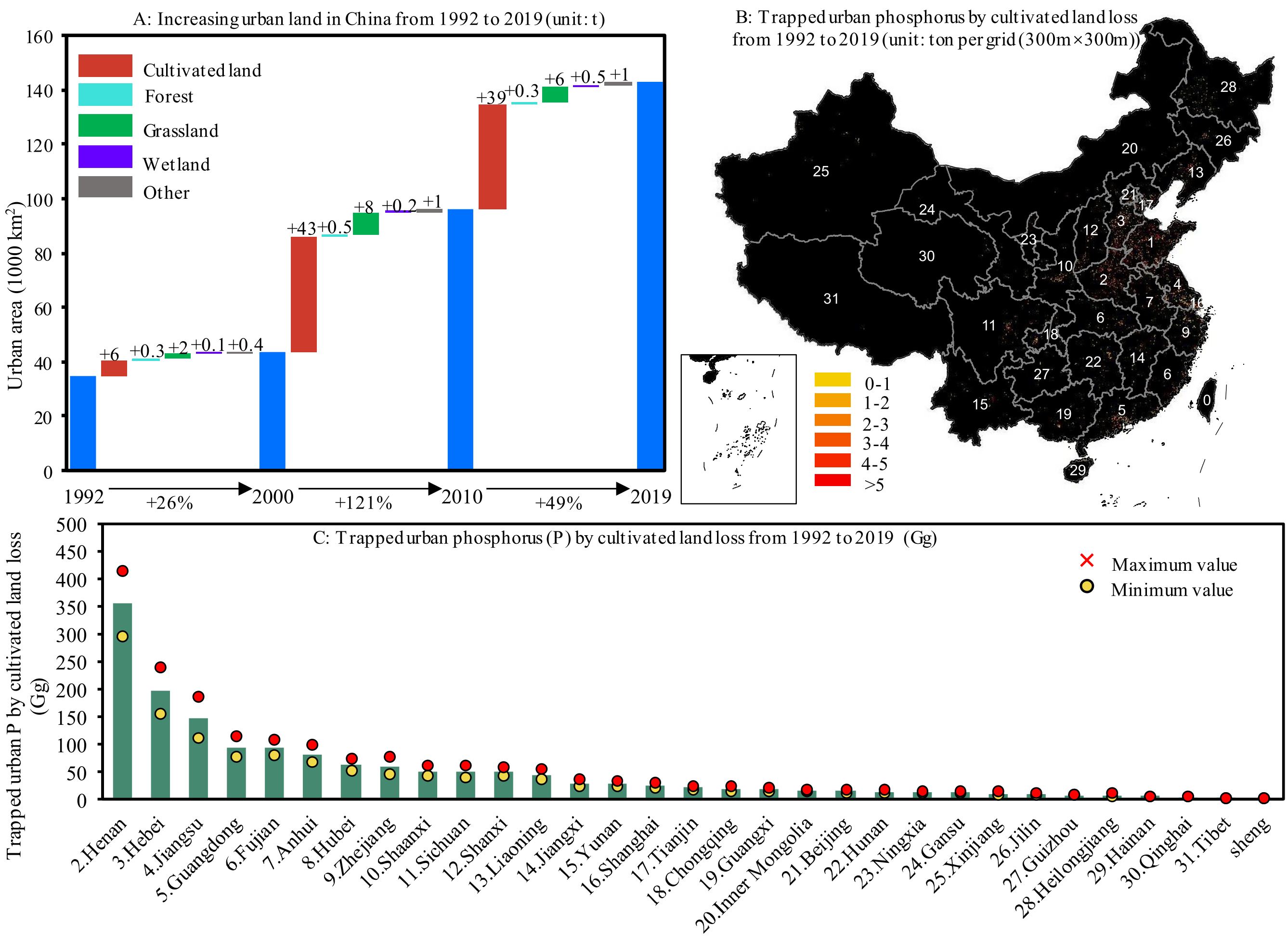

Figure 3. Urban expansion and trapped urban phosphorus (P) associated with cultivated land loss between 1992 and 2019 in China. Panel A: Urban land cover from 1992 to 2019 in China (km^2). Panel B: High-resolution (300 m × 300 m) mapping of trapped urban P caused by cultivated land loss in China during 1992−2019. Panel C: Total trapped urban P caused by urban land expansion in provinces of China during 1992−2019 (Gg). The yellow and red circular plots in the Panel C represent the minimum and maximum value of trapped urban P caused by urban land expansion among different provinces.

tripolyphosphate.[24] $P^{DG,2}$ is the P content of P-free detergent (0−1). $P^{DG,3}$ is the P content in the conventional detergents (0−1). The activity results of total P in the mined phosphate rocks and net export P across various products are illustrated in Figure S5. The calculated parameters used in eq 2 are detailed in Table S7.

Model Evaluation and Uncertainty Analysis. To increase the confidence in our results, we further evaluated the model in several ways. First, we quantified trapped urban P in China by integrating models of P budgets in cultivated land and urban solid waste. The modeling approach we employed was based on the widely accepted and previously validated methods (mass balance approach and the HPC model).[9,15,25] However, there are uncertainties in our quantification of trapped urban P and they arise from the approach, activity data, and parameters. We acknowledged them in Supporting Information S3. Second, in order to validate the reliability of our results, we compared our model outputs regarding P surplus in cultivated land, land cover, and P flow in urban solid waste with findings from other studies. Detailed information regarding these comparisons can be found in Supporting Information S5. Third, we performed a sensitivity analysis to test the response of anthropogenic activities and parameters and to changes in trapped urban P caused by cultivated land loss and urban solid waste disposal, respectively. For this analysis, we selected ten important model inputs and increased them by 10%, following the approach of the one-factor-at-a-time method.[26] Subsequently, we compared the model outcomes between the original model run and alternative model runs resulting from the sensitivity analysis. The chosen model inputs and the corresponding results from the sensitivity analysis can be found in Figure S6. Sensitivity analysis showed that P in the chemical P fertilizer and animal bones are the most sensitive sources of trapped urban P (Figure S6). In this study, we calculated the uncertainties (maximum and minimum values) of trapped urban P through cultivated land loss and urban solid waste disposal by adjusting them with ±10%.

RESULTS

Urban Areas Trapped 6% of Consumed P. Between 1992 to 2019, China generated a total of 13 (±0.9) Tg of trapped urban P (Figure 2A). Cultivated land loss and urban solid waste disposal contributed 15% and 85% of the total trapped urban P, respectively (Figure 2B). We found that the annual trapped urban P in China increased substantially

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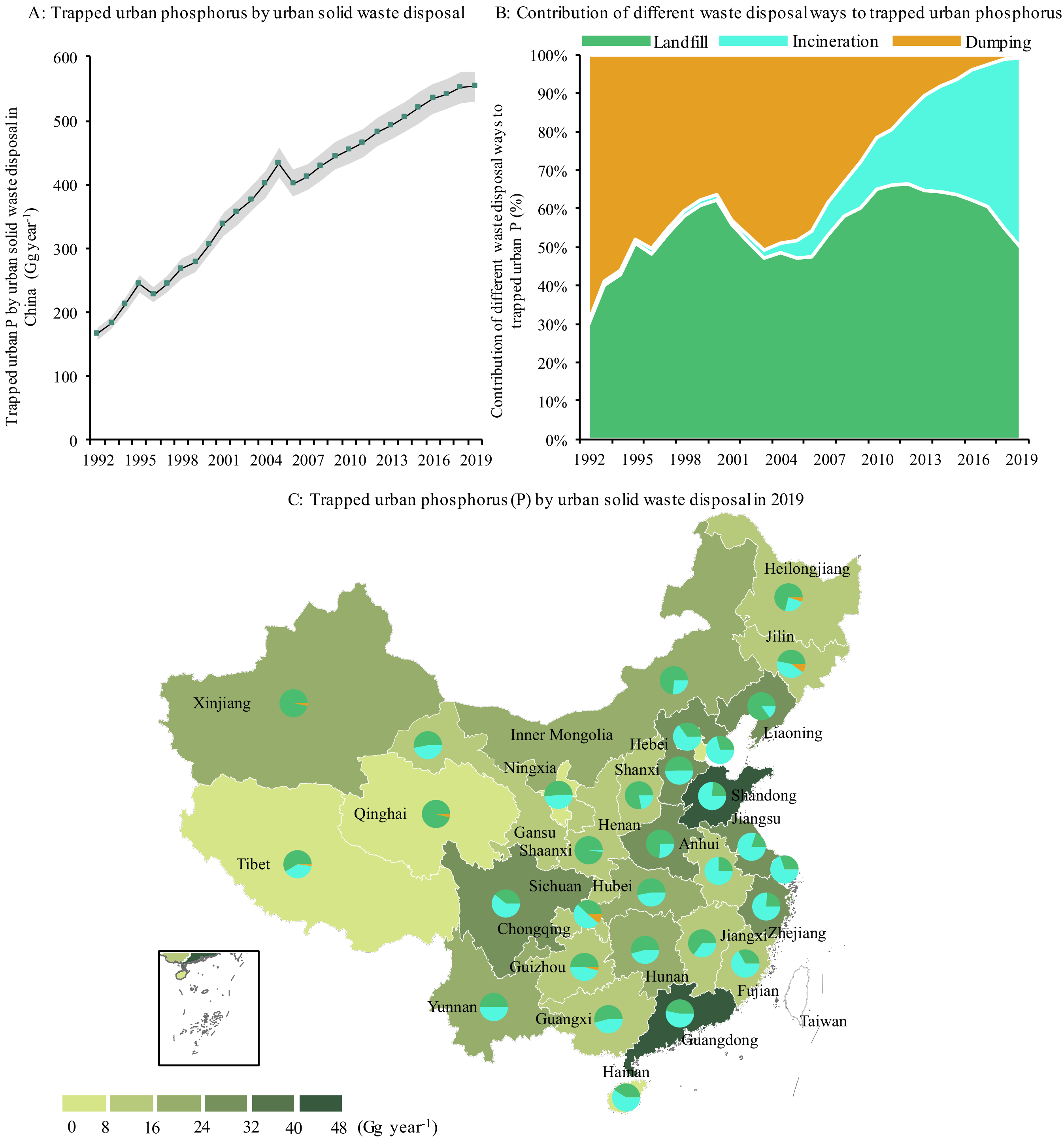

Figure 4. Spatiotemporal changes in trapped urban phosphorus (P) through urban solid waste disposal in China. Panel A: Temporal change in trapped urban P through urban solid waste disposal through landfilling, incineration, and dumping from 1992 to 2019 in China (Gg year^{-1}). Panel B: Trends in the contribution of landfill, incineration, and dumping to Trapped urban P through urban solid waste (%). Panel C: Trapped urban P through urban solid waste disposal across different provinces of China in 2019 (geographic map, Gg year^{-1}), with disposal methods including landfill, incineration, and dumping (pie charts, %). Uncertainty of trapped urban P through urban solid waste disposal is shown in the gray shaded areas in Panel A.

between 1992 to 2017 (Figure 2C), from 166 (±9) Gg year^{-1} in 1992 to 737 (±60) Gg year^{-1} in 2017, and then declined to 625 (±37) Gg year^{-1} in 2019 (Figure 2C). The sudden drop in trapped urban P in 2016 was attributed to a sharp decrease in urban expansion at that time. Since 2014, the annual trapped urban P in China has displayed a fluctuating trend, mainly following the pattern of cultivated land loss (Figure 2C). The contribution of cultivated land loss to annual trapped urban P in China varied over the years (Figure 2C). It showed an upward trend of ∼1% until 2000, 10−20% between 2001−2010, and more than 20% after 2010 (Figure 2C). This periodic upward trend can be attributed to two reasons. First, the annual urban expansion has been erratic (Figure S7). The annual urban expansion ranged 13803−15203 km^2 year^{-1}

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(Figure S7) prior to 2000 and it rose by 4−5 times thereafter during the study period. In 2017, the contribution of cultivated land loss to trapped urban P generation peaked at 26%, mainly because the urban expansion reached its maximum by that time. Second, the later the urban encroachment on cultivated land, the higher the P surplus in the cultivated land (Figure S8). In contrast, the growth of the trapped urban P through urban solid waste disposal has been relatively stable.

The total trapped urban P between 1992−2019 was equivalent to 6 (±0.5) % of the total consumed P (13 ± 0.9 Tg) (Figure 2A). Consumed P represented the net input of the anthropogenic P cycle. Between 1992 and 2019, the percentage of trapped urban P to annual consumed P ranged 4 (±0.2) −15 (±1) %, peaking in 2001 (Figure 2C). This indicated that trapped urban P was an important stock in the anthropogenic P cycle of China. Consumed P in China has increased from 4 Tg/year in 1992 to 10 Tg/year in 2019, with a total accumulation of 200 Tg during this period (Figure S9). It is worth noting that China's annual consumed P accounts for over 30% of total consumed P in the world (Figure S9). This suggests that approximately 2 (±0.1) % of global consumed P was trapped in urban areas of China every year.

The total trapped urban P also represented 9 (±0.6) % of the chemical fertilizer P used in China between 1992 to 2019 (Figure 2A). From 1992 to 2010, the proportion of annual trapped urban P to annual chemical fertilizer P used in China rose from 5 (±0.3) % to 10 (±1.0) %, stabilizing at 10 (±1.0) % post-2010 (Figure 2A). China now steadily consumes one-third of agricultural chemical P fertilizer used globally (Figure S9). This means that the trapped urban P in China accounts for 3 (±0.2) % of global chemical P fertilizer consumption. In summary, the trapped urban P in China is not only a significant component of the local anthropogenic P cycle, but it also poses a significant challenge to global P sustainability.

Urban Expansion on Cultivated Land Led to P Being Trapped. Urban land cover in China has increased from 34,544 to 142,847 km^2 from 1992 to 2019. This represents a remarkable net total expansion of 313%, with an average annual growth rate of 12% (Figure 3A). This annual urban growth rate was nearly four times greater than the global average for the period 1985−2015.[15] In China, urban land area increased slightly between 1992 and 2000, but it expanded massively thereafter. Thus, more than 90% of China's urban land expansion from 1992 and 2019 occurred after the year 2000 (Figure 3A). These temporal patterns of urban expansion were largely influenced by changes in public policy (Figure S10 and Section 1 in Supporting Information).[27]

Urban expansion led to 1885 (±377) Gg of trapped urban P associated with cultivated land loss between 1992 and 2019 in China (Figure 3B). Given the fact that a massive urban expansion occurred after 2000 (Figure 3A), nearly 99% of trapped urban P generated between 1992 and 2019, which was attributed to cultivated land loss, was formed since 2000 (Figure 3C). Two main reasons underpinned the issue of trapped urban P occurring from urban expansion. First, around 80% of the expanded urban land in China has come at the cost of cultivated land (Figure 3A). Over the period of 1992−2019, approximately 87,000 km^2 of cultivated land has been converted into urban land. This equated to 3% of China's cultivated land in 1992. Second, more than 80% of the counties in China had considerable legacy P in their cultivated land, which could be attributed to historical excessive fertilization (Figure S8). In this study, we considered the top 20% quantiles of P surplus (>20 kg ha^{-1} year^{-1}) of all counties from 2000 to 2019 as hotspot counties. We found that the hotspot counties with P surplus in China have increased by 25%, from 379 to 643, between 2000 and 2019. Moreover, the average P surplus in the cultivated land among counties of China has increased from 9.8 ± 14.2 kg ha^{-1} year^{-1} in 2000 to 14.9 ± 26.7 kg ha^{-1} year^{-1} in 2019.

Half of the trapped urban P associated with cultivated land loss was found in three provinces (Figure 3C). They were Shandong, Henan, and Hebei, collectively accounting for 25% of urban areas in China. This concentration of trapped urban P was associated with large losses of cultivated land. From 1992 to 2019, approximately 33% of the new urban area development in China took place in Shandong, Henan, and Hebei provinces, and that too at the expense of adjacent cultivated land in all these three provinces (Figure S11). Moreover, these provinces mostly have high-quality croplands.[28] The hotspot counties with P surplus in these three provinces have increased from 159 in 2000 to 222 in 2019, which amounts to half of the total number of counties in the three provinces or 35% of the total hotspot counties with P surplus in China.

Urban Solid Waste Disposal Accounts for 85% of Trapped Urban P in China. Urban solid waste disposal in China between 1992 and 2019 led to 10.8 (±0.5) Tg of P trapped in urban systems. This accounted for 85% of the total trapped urban P in China since 1992 (Figure 2B). We found that trapped urban P through urban solid waste disposal has increased substantially, from 166 (±9) Gg year^{-1} in 1992 to 553 (±23) Gg year^{-1} in 2019 (Figure 4A). This represented a net increase of 233% from 1992 to 2019 with a substantially increased rate of 14 (±0.5) Gg P per year. This net increase was primarily attributed to the increase in urban population from 322 million to 884 million in China during the same period (Figure S12). Moreover, the overall economic growth of the country also accelerated domestic waste accumulation.[29]

The pathways generating trapped urban P through urban solid waste disposal in China changed over time (Figure 4A). Dumping urban solid waste was the main reason for P trapping in urban areas, which accounted for 70% of trapped urban P in 1992 (Figure 4A). Later on, other pathways involving nonhazardous treatments of urban solid waste, such as landfills and incineration became more important (Figure 4A). Since 1995, landfills have become the dominant pathway to treat urban solid waste in China. This pathway accounted for 47−66% of trapped urban P during 1992−2019 (Figure 4A). Since 2005, the importance of incineration to treat urban solid waste has increased (Figure 4A). In 2019, half of the urban solid waste was incinerated in China. The implementation of relevant policies and laws drove the change in disposal pathways.[29] Waste incineration is an attractive alternative to landfilling. Incineration produces electricity and heat and reduces the land needed for waste disposal.[30] However, slag from waste incineration would go to landfills or be used as construction materials. This means that effectively P in incinerated waste will still be trapped in urban areas and will not return to the anthropogenic P cycle.

The spatial distribution of trapped urban P through urban solid waste disposal and their pathways showed great variability (Figure 4B). As an example, in 2019, Guangdong, Hebei, Shandong, Sichuan, and Henan were the top five provinces with the highest trapped urban P through urban solid waste disposal. They contributed to 33% of trapped urban P through urban solid waste disposal in China (Figure 4B). It is

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important to note that 34% of China's urban population in 2019 was also concentrated in these provinces (Figure S11). We also found that the contribution of incineration to trapped urban P in more urbanized provinces (e.g., Tianjin, Shanghai, Jiangsu, and Zhejiang) was higher than 70% (Figures 4B and S11). This could infer that incineration tends to be the main path of trapped urban P in China currently.

DISCUSSION

Significance of Trapped Urban P. *Trapped Urban P Is an Overlooked and Inaccessible P Stock Challenging the Sustainability of the Anthropogenic P Cycle.* While some studies have implicitly considered trapped urban P in their examination of P stocks, distinguishing it as an independent stock for further investigation remains lacking. For example, Lun et al.[31] revealed that China witnessed the most substantial cumulative increase in soil P between 2002 to 2010, totaling 34.6 Tg P. Following closely behind were India (11.4 Tg P), Brazil (3.6 Tg P), Pakistan (1.8 Tg P), the United States (1.8 Tg P), and New Zealand (1.8 Tg P). Liu et al.[32] showed that P waste (2.5 Tg year^{-1}) would be the largest P stock in China, second only to cultivated land (3.5 Tg year^{-1}). However, none of these studies have explicitly addressed the negative effects of trapped urban P to sustainable P cycle, highlighting the need for its independent evaluation. Traditionally, accumulated P stock in cultivated land, resulting from the historical net annual P input, has long been viewed as an important P reserve for agriculture production.[11] However, once urbanization encroaches upon cultivated land, the accumulated P becomes inaccessible for agriculture, undermining the sustainability of the P cycle. Modern disposal options for solid waste and byproducts, such as landfills or incineration prioritize environmental concerns but inadvertently lead to a loss of valuable secondary P resources. Previous research underscored the significance of sustainable P management in mitigating P losses throughout the P cycle, thereby mitigating water pollution.[18,33] The current study first shows that the trapped urban P deserves more attention as an inaccessible P resource for agriculture. The continuous accumulation of trapped urban P risks perpetuating the increasing reliance on external phosphate rock inputs and diminishing the internal recycling rate within the anthropogenic P cycle.

Although Trapped Urban P Accounted for a Relatively Small Proportion of the Total Consumed P, Its Absolute Amount in China Was Substantial. With a larger urban area and higher urbanization in the future, the trapped urban P will be more. The total trapped urban P in China between 1992–2019 (13 ± 0.9 Tg) corresponded to the total amount of P fertilizer used in the entire African agriculture for the past 18 years (Figure S13). And, the enormity of the trapped urban P problem can be readily recognized from the fact that in 2019 the amount of trapped urban P in China (625 ± 39 Gg year^{-1}) is about 36 (±2) %, 132 (±8) %, and 36 (±2) % of agricultural P fertilizer used in Europe, Canada, and the US, respectively (Figure S13), which shows the importance of this less studied and potentially recoverable P stock. Furthermore, increasing demand for animal-based food in urbanized areas will undoubtedly cause further pressure on P reserves. Globally, the agricultural P demand is projected to reach 22–27 Tg yr^{-1} in 2050 (19 Tg yr^{-1} in 2020).[34] With the scarcity of mineable P reserves in China and America, the demand for phosphate rock for most countries may ultimately depend on reserves in Morocco or Norway by 2100.[7] Therefore, preventing, or at least greatly reducing the trapped urban P will help meet a significant proportion of agriculture P demand.

Although phosphate rocks are relatively abundant in China in the short term, this does not mean that significant P reserves have been assured for centuries.[35] China is feeding 22% of the world population with less than 5% of mineral P reserves while supplying P rocks to meet more than 50% of the global demand.[36] Although the production of phosphate rock in China is projected to peak between 2035 and 2045,[35] it will remain high with the massive food and feed market.[37] Given this backdrop, it is particularly alarming considering the sheer scale of urbanization and the magnitude of P getting trapped in the urban areas occurring in China. Thus, preventing the accumulation of trapped urban P is an important step in closing the anthropogenic P cycle and ensuring sustainable crop production systems. A better understanding of trapped urban P therefore holds profound practical implications, particularly for urbanized and developing nations with limited access to rock phosphates (e.g., India).[38]

Uncertainties and Limitations. Our study has limitations and uncertainties associated with the approach and inputs used for modeling. To calculate legacy P in cultivated land, we adopted a mass balance approach. This approach is commonly used to identify and quantify legacy P (cumulative P surplus) in cultivated land at national and continental scale.[9] Nevertheless, it is important to be aware of the limitations and uncertainties in the assumptions and calculations used here. For example, atmospheric P deposition, P loss by leaching and runoff, and P inputs by seeds and irrigation were not included in this analysis. This was because these factors were considered to have negligible impact on P inputs and outputs compared to fertilizer and manure.[39] For instance, P inputs of atmospheric deposition in China were measured to be 0.92 kg ha^{-1} year^{-1}.[40] The observed P loss rate from upland and paddy fields were 1.24 and 0.8 kg ha^{-1} year^{-1}.[41] Therefore, the uncertainties in the P inputs and outputs to P surplus in cultivated land counterbalanced mutually. Furthermore, we developed high-resolution maps of legacy P in cultivated land for China from 1992 to 2019, using estimates at the provincial (1992–1999) and county scales (2000–2019). However, not all inputs were specified with sufficient details in all counties due to data limitations, such as the recycling rate of crop straw and animal manure (Table S4 and Figure S3). In such instances, we assumed homogeneous properties within counties or provinces. This assumption may not accurately reflect the spatial variation of legacy P, yet best estimates were used where required. In future studies, it is important to improve data quality on the variability of model inputs and related parameters.

Another limitation of our study is the challenge of accurately quantifying different land cover areas using data sets with a spatial resolution of 300 m. The ESA-CCI Land Cover data sets we used in this study had the highest spatial resolution and most consistent historically (1992 to 2019). Furthermore, it is worth noting that the land cover maps until 2015 (v207) were generated under the ESA CCI umbrella, while from 2016, the maps were operationally generated under the EC Copernicus Climate Change Service. This change resulted in a sudden drop in the trend of urban expansion in 2016, which in turn led to an abnormal change in results of annul trapped urban P at that time. This uncertainty inherent in the land use data set may underestimate the trapped urban P associated with cultivated land loss. However, we believe this uncertainty does

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not influence the main findings of this work, as it contributes relatively little to the total trapped urban P in China.

In this study, we did not run the HPC model to quantify total P in the municipal sludge at the provincial scale, as we did for animal bone and domestic solid waste. Instead, we allocated P in the municipal sludge to each province based on the share of dry sludge generation. This allocation assumed that the P content in dried sludge is the same across all provinces, which assumption may lead to uncertainties in the spatial variation of trapped urban P through urban solid waste disposal. However, we believe that this assumption does not influence the main messages of our study. Municipal sludge made a relatively small contribution to trapped urban P, compared to animal bone and domestic solid waste. Furthermore, domestic kitchen waste, a solid waste, is a P-rich material in urban daily life.[18] The HPC model does not explicitly quantify P in kitchen waste alone but instead incorporates it indirectly through municipal solid waste collection. This is because kitchen waste in China is not sorted and separately treated, and most of it is disposed in landfills.[42] Finally, we would highlight that the robustness of the HPC model has been tested by Liu et al.,[32] Liu et al.[12] and Liu et al.[23]

Policy Implication for P Management. Our novel findings shed light on the need for managing a currently inaccessible P stock caused by urbanization, a crucial aspect that has been overlooked in prior research. Building upon our novel results, we have developed two complementary policy recommendations aimed at minimizing the accumulation of P in urban stock as trapped urban P. First, we argue that strict policies to preserve high fertility cultivated land during urban land expansion need to be implemented. Cultivated land loss by urban expansion was estimated to contribute to 15% of trapped urban P. Cultivated land loss caused by urban expansion not only traps P directly, but also leads to a potential avoidable extra consumption of P recourse. China set a policy to retain at least 120 million hectares (1.8 billion mu) of cultivated land.[43] To maintain the set area of total cultivated land, a compensation system for cultivated land use for urbanization was established. This system aimed to strike a land use balance to ensure the target cultivated land area nationally. However, an unintended consequence of this compensation was cropland redistribution to marginal lands (e.g., shrubland and other land with poor soil), which spurred increased consumption of fertilizer.[3,28] Therefore, extra consumption of P recourse refers to the newly cultivated land requiring more P fertilizer to compensate for P deficiency. From 1992 to 2015, reclaimed cultivated lands in China were estimated to be 13×10^4 km^2, of which 74% were nutrient-poor, requiring substantial input of P fertilizers.[28] Therefore, governments should actively protect high-quality cultivated land and limit urban expansion.[44] Beginning in 2017, the State Council of China required the local governments to provide the same quantity and quality of cultivated land as compensation for urban expansion. However, to our knowledge, it is a challenge to compensate for cultivated land in terms of equal legacy P and soil fertility. Either direct or indirect P recourse loss from the urban expansion will pose a risk to food security and resource sustainability. If urban construction planning gives priority to other factors over fertile cultivated land, we appeal for engineering measures of stripping topsoil from cultivated land to reserve P fertility.

Second, it is highly recommended to promote the recovery of trapped P through urban solid waste recycling, with the ultimate goal of recovering P from the majority of urban solid waste. This will effectively reduce trapped urban P and enhance the resilience of the anthropogenic P cycle. We estimated that urban solid waste disposal contributed to 85% of the total trapped urban P in China (Figure 2B). P in urban solid waste consisted of 25, 40, and 35% municipal sludge, animal bones, and domestic waste, respectively (Figure S14). In the future, urban solid waste will become a more prominent source of trapped urban P. There are two reasons for this outcome. First, the urban population will continue to grow in Chinese cities and their rising standard of living will increase meat consumption. The demand for animal products per capita in China is projected to increase by 23% in 2050 compared with that of 2020.[37] Thus, more P is bound to be trapped in the urban solid waste. Second, the decline in urban expansion and protection of cultivated land will also lead to a further increase in the contribution of urban solid waste disposal to trapped P. Now, these P-containing resources have limited use in crop production due to multiple reasons. For example, the direct use of municipal sludge in cultivated land is restricted by legislation or scarcely practiced due to environmental risks posed by heavy metals and pathogens.[45] The technologies for recovering P from sludge or animal bones are still under development and the relevant market is very limited in developing countries.[46,47] Most of the P in domestic waste comes from food waste. Many studies have highlighted the implication of reducing food waste for food security and resource sustainability.[37,44] Composting is an effective way to recover P from food waste. Composting technology has been diversified to meet different requirements and capacities.[48] However, compost treatment must be built on sound waste classification and recycling systems in cities, especially the separation of kitchen waste from domestic waste. The garbage classification and recycling system for kitchen waste in China is still in its infancy. In 2017, mandatory waste classification and processing were instituted in 46 Chinese cities (e.g., Beijing and Shanghai). However, this mandatory policy failed to motivate the public toward waste reduction and waste separation for disposal.[42]

The key obstacle to P recovery from waste is the lack of effective collaboration and coordination among stakeholders, along with inadequate supporting measures and technologies.[46,49] In this context, stakeholders can include sewage treatment departments that act as suppliers of secondary phosphates, business entities as buyers of phosphate and producers of waste-derived fertilizers, researchers contributing improved technology and circular resource use model, and farmers as consumers of waste-derived fertilizers. For example, policymakers have yet to formulate appropriate P recovery policies and plans, hindering technology development, implementation of developed technologies and waste management commercialization. In addition, farmers do not fully recognize the environmental and ecological benefits of waste-derived fertilizers, which are indeed more expensive compared to conventional fertilizers currently. Several factors cause insufficient government support measures that promote P recovery. First, the implementation of P recovery solutions in waste treatment plants is crucial. But the current single-purpose infrastructure in developing countries (e.g., China) is for just nutrient load reduction rather than a multipurpose system capable of nutrient recovery and production of waste-

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derived products such as fertilizers. Also, wastewater treatment plants designed to recover multiple valuable products have not matured yet. Second, many waste-derived fertilizers are not competitive in the market due to high recycling costs. At present, the cost of recovering P from wastewater is several times higher than the market price of phosphate rock.[46] As a result, most P recovery technologies have not yet scaled up and value chains are only just emerging. By linking the benefits of resource recovery to the residents' personal income, as well as health and environmental well-being, the classification and recycling of waste at its primary source itself should be accelerated. For example, the local government can increase investment in the recycling industry for recovering waste P, creating numerous direct and indirect socio-economic benefits. This chain involves various stages, such as collection, sorting, extraction and separation, and manufacturing and marketing of waste-derived commercial products.[17] In such a scenario, waste P can be recognized as a value-added raw material, enabling residents to earn additional income through collection and trade. A market-driven waste recycling makes the P recovery chain more commonplace. However, implementation of systems for P recovery from waste will become easier in practice only when sourcing of mineral resources is limited by objective (e.g., mine depletion) or subjective reasons (e.g., trade difficulties). We believe that such a scenario may arise, and recycling of waste P become a necessity in the future.

Contributions to Knowledge Gaps on Anthropogenic P Cycles. To conclude, we here report the first study to define and quantify trapped urban P in China and show the magnitude and significance of the amount of trapped urban P relative to total consumed P and the chemical fertilizer P used locally. Our findings reveal that 6 ($\pm$0.5) % of consumed P would end up as trapped urban P in China. Our results also provide compelling evidence that trapped urban P is an often overlooked and currently inaccessible anthropogenic P stock in the cities. Inaction to recover and reduce its loss will lead to lasting dependence on P derived from finite phosphate rock reserves for agricultural production. A slowdown of urban expansion in the future will reduce the growth of trapped urban P. However, as the population further gathers in cities, the disposal of urban solid waste will still generate a substantial amount of trapped urban P. This situation is likely to place countries with limited or no mineable P reserves under more challenging conditions to sustain food production. Our study thus distinguishes itself from earlier P flow analyses in urban areas by incorporating the amount of P trapped in cities and through urban land expansion. This study is also a wake-up call for other urbanizing countries (e.g., in India, Southeast and African countries) with little P reserves and long-term dependence on imports. Trapped urban P generated during urban land expansion and urban P consumption poses a serious threat to global food security. Our study calls for a comprehensive P management system that minimizes trapped urban P and closes the loop of the anthropogenic P cycle needed for sustainable food production and to realize Sustainable Development Goals 2 and 11.

■ ASSOCIATED CONTENT

Data Availability Statement

All data are available in the main text or the Supporting Information. Data that support the findings presented in this study are available from the author upon reasonable request. R Scripts used for analysis in this study are available from the author upon reasonable request.

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.4c08078.

Detailed description for driving forces of changes in trapped urban P (Text S1), calculation of legacy phosphorus in the cultivated land (Text S2), spatial variability of P legacy in cultivated land in China (Text S3), description of the HPC model (Text S4) and model evaluation and uncertainties (Text S5). Supporting figures for the diagram of P flow in urban systems (Figures S1 and S4), the diagram of legacy P calculation (Figure S2), the fraction of crop straw removal (Figure S3), natural phosphate rocks in China (Figure S5), results of sensitivity analysis (Figure S6), urban land cover (Figure S7), county P surplus (Figure S8), consumed P in the world (Figure S9), policies associated with urbanization (Figure S10), provincial urban land cover (Figure S11), urban population in China (Figure S12), comparison of trapped urban P to fertilizer P (Figure S13), P in the urban solid waste (Figure S14) and relationship between urbanization and trapped urban P (Figure S15). Supporting tables for P content in the compound fertilizer (Table S1), P contents in grain and straw (Table S2), percentage of manure recycling to the cultivated land (Table S3), feeding cycle of animals (Table S4), excretion P from animal (Table S5) and sources of the HPC model inputs (Table S6) (PDF)

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Author Contributions

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Notes

The authors declare no competing financial interest.

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