Intensification of phosphorus cycling in China since the 1600s

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Phosphorus (P) is an essential nutrient for living systems with emerging sustainability challenges related to supply uncertainty and aquatic eutrophication. However, its long-term temporal dynamics and subsequent effects on freshwater ecosystems are still unclear. Here, we quantify the P pathways across China over the past four centuries with a life cycle process-balanced model and evaluate the concomitant potential for eutrophication with a spatial resolution of 5 arc-minutes in 2012. We find that P cycling in China has been artificially intensified during this period to sustain the increasing population and its demand for animal protein-based diets, with continuous accumulations in inland waters and lands. In the past decade, China's international trade of P involves net exports of P chemicals and net imports of downstream crops, specifically soybeans from the United States, Brazil, and Argentina. The contribution of crop products to per capita food P demand, namely, the P directly consumed by humans, declined from over 98% before the 1950s to 76% in 2012, even though there was little change in per capita food P demand. Anthropogenic P losses to freshwater and their eutrophication potential clustered in wealthy coastal regions with dense populations. We estimate that Chinese P reserve depletion could be postponed for over 20 y by more efficient life cycle P management. Our results highlight the importance of closing the P cycle to achieve the cobenefits of P resource conservation and eutrophication mitigation in the world's most rapidly developing economy.

sustainability | phosphorus cycling | eutrophication | food production | industrial ecology

The cycling of phosphorus (P), an essential nutrient for living systems, has been massively altered by human activities, with emerging concerns about long-term supplies of affordable P for food production (1, 2). The one-way journey of P has also increased eutrophication, contributing to significant disruptions, such as water quality degradation and biodiversity losses in aquatic ecosystems (3, 4). Amplified production of phosphate rock, largely for fertilizer production, reached 225 million tons globally in 2013 (5), leading to the conclusion that anthropogenic P use is operating well beyond planetary boundaries (6, 7). Among all countries, China, with its rapid increase in population and affluence, faces perhaps the greatest sustainability challenges in its P sector. Although China is now the world's largest producer of phosphate rock, contributing 48% of total production in 2013 (5), concerns about scarcity of Chinese P reserves have begun to emerge. Although this possibility has not been as widely recognized for P as for other critical mineral resources (*SI Appendix*, Fig. S1), reliance of Chinese agriculture on imported P would have major ramifications for global fertilizer markets. Reflecting this amplified P use, most Chinese freshwaters have experienced excessive total P loading for years (8), triggering high-profile events, such as the cyanobacteria bloom in Lake Taihu in 2007 that cut off the drinking water supply for 2 million people in the city of Wuxi for more than 1 wk (9). These pressing sustainability issues, as well as China's major role in the global P economy, highlight the importance of quantifying P pathways across China and assessing their regional impacts (10–12). However, previous studies in China have had limited temporal scale and incomplete assessment of P-associated activities and products (*SI Appendix*, Table S1). The temporal scales of these investigations are often limited to one specific year or to recent decades when human activities had already become intensive. Most previous studies also focus on agricultural production rather than the entire P cycle, which includes both key natural and anthropogenic activities. Furthermore, previous studies have not provided close spatial analysis of the potential eutrophication effects that P losses have on freshwater ecosystems in China. Understanding P's long-term temporal dynamics and the regional disparity of the eutrophication impact can provide a basis for P management strategies that establish a more sustainable P cycle in China, and indeed globally.

This study quantifies, for the first time to our knowledge, the entire P cycle across China over the past four centuries and evaluates the spatial distribution of the concomitant potential for eutrophication in 2012. Details of our analytical approach and the underlying data are described in *Materials and Methods* and *SI Appendix*. In general, we build a hierarchical national P cycle model (*SI Appendix*, Fig. S2) to analyze the dynamic changes of Chinese P cycling during 1600–2012 using time series of activity data and parameters. We further characterize the geographical patterns of anthropogenic P losses to mainland freshwaters and evaluate freshwater eutrophication potential (EP) with a spatial resolution of 5 arc-minutes in 2012.

Results and Discussion

We find that P cycling in mainland China intensified from rather simple, nature-dominated stable situations in the earlier three

Significance

The biogeochemical cycle of phosphorus (P) has been massively altered in China, challenging its food security and causing eutrophication of freshwaters. This study shows, for the first time to our knowledge, how P cycling in China was intensified in the past four centuries to sustain the increasing population and its demand for animal protein. Our analysis also reveals the spatial disparity of its concomitant eutrophication impact. The findings advance the knowledge base needed for closing the P cycle to sustain future food production and maintain healthy rivers, lakes, and oceans.

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SUSTAINABILITY SCIENCE

centuries to complex, human-dominated scenarios in the past six decades, especially after China's economic "reform and opening-up" policy was adopted in the late 1970s (Fig. 1). Changes of natural P flows over the past four centuries are multifarious. Natural P flow through atmospheric deposition was ~0.3 teragrams (Tg)-P·y^{-1} before the 1950s, but dramatically increased fourfold during the past half-century, larger than previous estimates because of higher experimental bulk P deposition rates (Fig. 2*A* and *SI Appendix*, section 4 and Fig. S4*A*). Atmospheric P from wind erosion and sea spray had no significant changes and was about 40% lower than atmospheric P from combustion sources (13). Natural P runoff into inland freshwaters decreased from 0.35 Tg-P·y^{-1} in 1600 to 0.25 Tg-P·y^{-1} in 2012. We estimated less than 20% of riverine P was transported to the open oceans annually. P entering the oceanic cycle is probably largely in the dissolved phase because most of the particulate P, which accounts for more than 75% of the total riverine P, is retained in the inland and coastal waters via sedimentation due to relatively flat terrains as well as artificial dams (14–16).

Before the 20th century, human mobilization of P in China was enhanced slowly through expanding agricultural activities associated with the growing population (Fig. 2*A* and *SI Appendix*, Fig. S4*A*). For example, from 1600 to 1900, P contained in crops increased from 0.44 Tg-P·y^{-1} to 0.81 Tg-P·y^{-1}, and P contained in animals increased from 4.7 gigagrams (Gg)-P·y^{-1} to 13.3 Gg-P·y^{-1}. However, these two P flows both decreased during the first half of the 17th century and the second half of 19th century. These declines were largely due to agricultural yield reductions when China suffered heavy population losses from decades of turmoil and wars during dynasty change and the later Taiping Rebellion (peasant revolts against the feudal monarchy). Furthermore, China isolated itself, and little P-associated international trade occurred until the first Opium War broke out in 1840. Following its defeat by the British, China was forced in the Treaty of Nanking to open four additional domestic port cities for foreign trade alongside Guangzhou. This expansion of trade led to a large-scale increase in the import of various goods into China, including the reintroduction of not only opium but also P-containing foodstuff.

This pattern changed dramatically after the 1910s, when domestic phosphate rock began to be extracted for export. Nevertheless, China had to import P-containing fertilizers to increase crop yields, and thus became a net P importer. From the late 1950s onward, domestic P extraction increased considerably and reached 12.50 Tg-P in 2012, comprising over 40% of the global P production (5) (Fig. 2*B*). Synthetic fertilizer production consumed about 70% of the domestically exploited P in 2012, followed by elemental P and feed additives, responsible for 8% and 4%, respectively. Annual P intake by crops surged to the recent peak of 3.31 Tg-P in 2012, similar to trends in animal products and human food demands (Fig. 2 *A* and *C* and *SI Appendix*, Fig. S4 *A* and *B*); all three, however, declined during the Great Chinese Famine (1959–1962), when millions of Chinese starved to death (17). Since 2003, China has become a net P exporter, and the export pattern shifted from high-quality (P_2O_5 grade: 32–34%) but cheap (~20% lower than the world average price) rock in the early 1990s to downstream value-added fertilizers and fine chemicals (Fig. 2*D*). Net P import in crop products increased dramatically from 0.04 Tg-P in 1990 to 0.46 Tg-P in 2012, mainly in the form of soybeans from the United States, Brazil, and Argentina (United Nations COMTRADE database at comtrade.un.org).

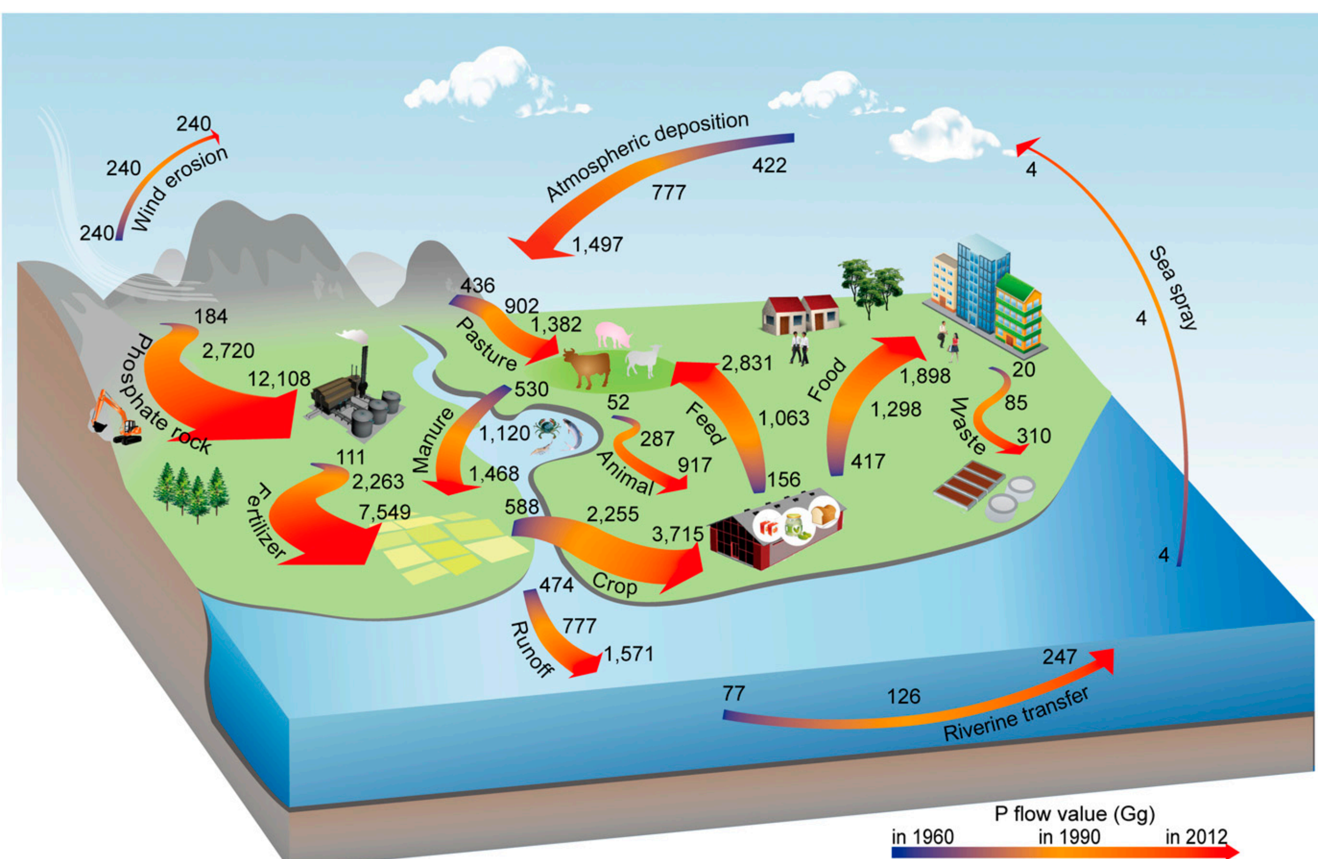

Fig. 1. Schematic model of P cycles in mainland China during 1960–2012. Systems here may be imbalanced because only aggregated key P flows balanced with import/export data are displayed for the sake of easy comprehension. The thickness of the arrows denotes the historical intensity of key P flows in 1960, 1990, and 2012.

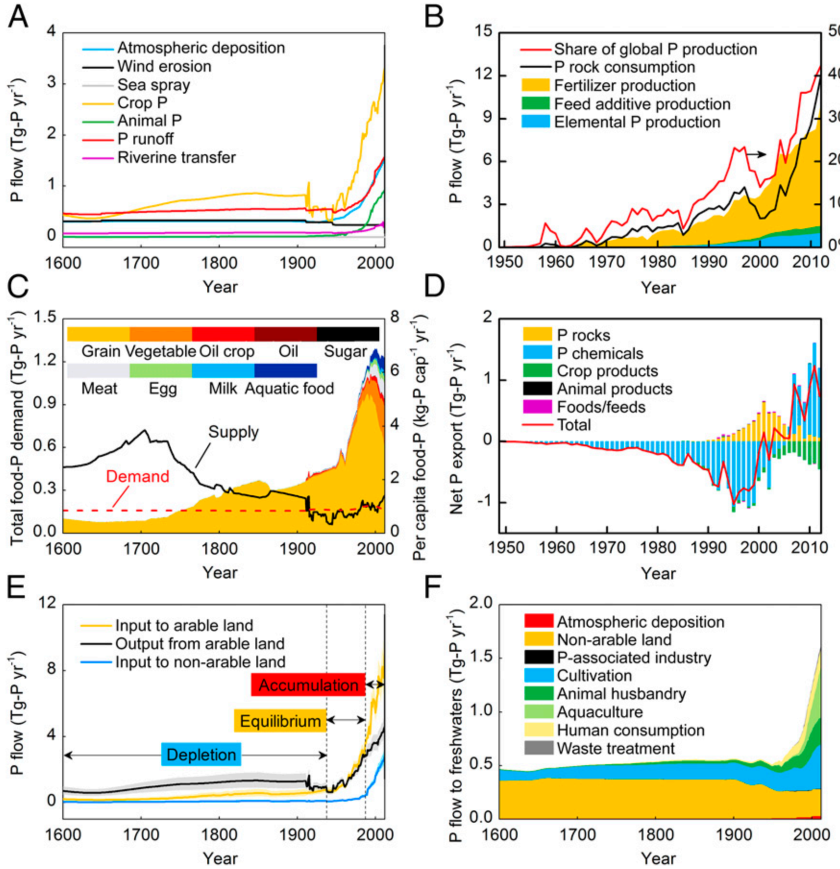

Fig. 2. Temporal changes of P cycles in mainland China. (*A*) Variations of key P flows excluding trade information, for which uncertainties are presented in *SI Appendix*, Fig. S3. (*B*) China's consumption of P rocks (left axis) and share of global P production (right axis). Domestic consumption of P rocks is estimated using two methods: One is summed up from the production of downstream P chemicals (stack area), whereas the other is the aggregate of domestic rock production and international trade statistics (black line). Data gaps between them during 1998–2007 could reflect the exclusion of a considerable number of small-scale mining operations. (*C*) Composition of food-P demand (left axis) and per capita P supply and demand (right axis). The solid black line corresponds to per capita food-P supply, whereas the dotted black line corresponds to per capita food-P demand. (*D*) P flows embedded in internationally traded commodities from alternative human activities. (*E*) P balance in arable land and scattered P wastes to nonarable land, with 90% confidence interval coverage (shaded area). (*F*) P losses to inland waters. Due to the lack of long-term observed P deposition, runoff, and leaching parameters, temporal characteristics are smoothed in Origin (Electronic Arts, Inc.) using the Savitzky–Golay method to show the overall escalating trend. P flows after a $\log_{10}$ transform on the *y* axis of *A*, *C*, *E*, and *F* are shown in *SI Appendix*, Fig. S4 to illustrate the earlier fluctuations.

Chinese industrial policies have played an important role in the entire P supply chain, especially after 1949, when the People's Republic of China was established, thus implementing a centralized regime. In 1983, the prices of several forms of P fertilizer (e.g., di-ammonium phosphate, calcium superphosphate) were raised by the government, but not in proportion to their content of P_2O_5. P-associated industries were stimulated to produce low-quality but more profitable types of P fertilizers. However, on the demand side, farmers had to pay high prices for relatively ineffective fertilizer, and thus reduced their fertilizer use. This condition led to an oversupply of P fertilizers in the mid-1980s. To cope with this situation, companies had to reduce their P fertilizer production, resulting in sudden decreases of P production (Fig. 2*B*). Recent fluctuations of net P export may be ascribed to the changeable policies (particularly export quotas and high tariffs) imposed on phosphate rock and fertilizers to restrict P export. For example, there was a sharp drop of net P export after the introduction of seasonal export tariffs for fertilizers in 2007 (Fig. 2*D*).

P use in Chinese crop production has also changed dramatically during the most recent six decades. Although Chinese farming has achieved yields close to the highest attainable for most crops (18), P overapplication has continued (19). Crop production maintained an average input intensity of 80 kg-P·ha^{-1} in 2012, more than double the input intensity that crops can generally assimilate. As shown in *SI Appendix*, Table S2, the situation is similar in other Asian countries (South Korea, Japan, and Thailand) but different from most European countries, such as Austria, Sweden, and the United Kingdom. Furthermore, unlike developed countries, which are beginning to recycle P-containing wastes, including composts and sewage sludge, emerging economies like China rely heavily on conventional commercial fertilizer P (over 60% of the total P inputs in 2012) to maintain agricultural yields.

We find that 4% of natural P resources were eventually ingested by Chinese residents in 2012, slightly lower than the 5% in the United States (20). Although per capita food-P demand, defined here as the P directly consumed by humans and independently estimated from national statistics, did not change significantly in China, its composition has varied markedly over the past four centuries, especially as seen in a decline in the share of crop-P from 98% in the 1950s to 76% in 2012 (Fig. 2*C* and *SI Appendix*, Fig. S4*B*). These dynamics indicate that the Chinese people expanded their demand for animal protein while still relying on plant-based P nutrition. The difference between per capita food-P demand and per capita food-P supply, defined here as the P available for human consumption derived from upstream flow balances, could be attributed to the direct food-P consumption of various processed products, such as bread, alcohol, and condiments, and the concomitant P losses (indirect food-P consumption) during the processing stages of these products. Changes in national strategic stockpiles, much more significant during earlier times when China stayed in seclusion, could broaden this gap as well. Crop products constitute similar percentages of food-P in Turkey, Zimbabwe, and Uganda, over 1.2-fold the percentages of food-P in Switzerland, the United States, and other developed countries (*SI Appendix*, Table S2). Because animal production has a much lower P use efficiency (PUE, defined here as useful P outputs in products divided by total P inputs of a compartment) than crop production, the growing demand for animal-based diets could worsen China's pressing P sustainability challenges. Considering the P losses in diet preparation, per capita P intake (P actually eaten by humans) in China is 30% lower than in the United States (21) but still exceeds the recommended dietary allowance (RDA) (22). Possible reductions of per capita P intake in both countries to the RDA level could save around 219 Gg-P nutrition in 2012, equivalent to the total P RDA of over 0.8 billion people worldwide. Our analyses also show that P wastes from human consumption shifted from rural areas to cities because of rapid urbanization, with a significant contribution of urban household solid wastes, especially organic food wastes (*SI Appendix*, Fig. S5). Increasingly large gaps in other socioeconomic factors between rural and urban areas, such as income levels and consumption habits (23), can also contribute to this shift.

The dynamics of total P mass balance in arable land over China's past four centuries can be broken into three stages: depletion (P input lags behind P output), equilibrium (P input equals P output), and accumulation (P input exceeds P output) periods (24) (Fig. 2*E* and *SI Appendix*, Fig. S4*C*). Only a small fraction of soil-accumulated P is liable to be chronically released to damage freshwater environments, whereas the rest, more than 85%, is likely immobilized as "legacy P," either as particulate inorganic P through chemical reaction and physical sorption or as organic P (e.g., phytin, nucleic acids, phospholipids) through biotic processes (25). However, under certain circumstances (influenced by pH, redox conditions, etc.) after entering freshwaters, some of these legacy materials may also release bioavailable P (26, 27). Furthermore, accumulated P stocks, mainly in the form of phosphogypsum tailings and uncollected excreta, dispersed into nonarable land could exacerbate this environmental risk.

P losses to Chinese freshwaters increased threefold over the past four centuries (Fig. 2*F* and *SI Appendix*, Fig. S4*D*). The anthropogenic contribution to the total exogenous P loading increased notably from ∼20% in the 1600s to 83% in 2012. P runoff and leaching losses from arable lands were sizable, as confirmed by other studies (e.g., ref. 11), whereas, perhaps surprisingly, ever-expanding freshwater aquaculture (28) became the largest constituent of P losses after 2009. Over 90% of the Chinese aquaculture feed-P remained in rearing waters as uneaten food and excretion, higher than levels observed in Finland (29) and Norway (30) (82% and 70%, respectively).

To ensure the robustness of model results, we conducted interactive cross-checks in P flow quantification and compared P flows with previous estimates from China and other countries (*SI Appendix*, section 4). In addition, Monte Carlo simulation was used to test the propagation of input uncertainties quantitatively, which may be caused by data unavailability, variability, and inconsistency, into the final results. After activity data and parameters were provided with continuous distributions (triangular or uniform) by considering data quality, the model was run for 10,000 trials using randomly selected values from the input distributions to generate ranges of outcomes. We find that P flows had smaller uncertainties in the past century than before the 1910s, thanks to the more sophisticated and publicly available data sources in recent times. Furthermore, uncertainties of most P flows are primarily associated with activity data rather than parameters (*SI Appendix*, section 6 and Fig. S3). Our analysis is based on the currently best available data, but prospective follow-up studies are necessary to substantiate these results further.

We next characterized the geographical patterns of anthropogenic P losses to mainland freshwaters, with a spatial resolution of 5 arc-minutes in 2012 (*SI Appendix*, Fig. S6), using spatial disaggregation methods. In brief, various geographically explicit activity datasets were used to calculate the grid-based distribution factors of individual human activities. High P emission densities were found in the eastern coastal provinces, which have relatively higher population density and per capita gross domestic products (*SI Appendix*, Fig. S7). The average P emission density in eastern China was 495 $kg \cdot km^{-2} \cdot y^{-1}$, eightfold higher than in western areas. Although the three megacities (Beijing, Shanghai, and Tianjin) were at the bottom of per capita P emissions, their impacts are manifested in their consumption of P-containing commodities produced by other provinces.

To assess the geographic distribution of likely water quality impacts of China's massive amplification of P cycling, we evaluated the freshwater EP of anthropogenic P losses using the globally differentiated EP characterization method (31, 32), an emerging regionalized impact assessment approach in the life cycle assessment domain (33). Although this approach cannot make specific inferences about the eutrophication status of particular water bodies, the regionalized EP characterization method can nevertheless provide deeper insight than P flow analysis results, which show only physical weights of environmental flows. Marked spatial disparities of EP were observed due to geographic variation in freshwater availability and in susceptibility to ecosystem damage, especially highlighting hotspots in the Yangtze River basin (Fig. 3). These regions, with their large anthropogenic P emissions and dense water networks connected with sensitive freshwater ecosystems, are at high risk of ecosystem damage. Note that water bodies in northern semiarid regions like Beijing and Tianjin are also prone to eutrophication (34). However, the current EP model is not suitable for such semiarid and arid areas; therefore, our analysis may underestimate the EPs in such environments. Furthermore, the EP model is only applicable for natural water bodies; artificial systems, such as the ongoing South-to-North Water Diversion Project in northern China, will need to be considered in future analyses. Nevertheless, this geographic analysis helps localize the impact of exceeding Earth's planetary boundaries for P utilization (6), identifying southeastern China as "ground zero" for P-accelerated eutrophication in freshwaters.

According to the 2012 US Geological Survey report, around 3,700 Tg of phosphate rock reserves (i.e., economically extractable resources) remain in China (5). If China maintains its current production rate, these domestic P reserves would be exhausted in the next ∼35 y (*SI Appendix*, section 5). This estimate is consistent with a P depletion model-based forecast (35) but much shorter than the world average longevity of several hundred years (36) based on a recent revision of global P reserve estimates that included an eightfold increase of estimated P reserves in Morocco (37). To address this emerging issue, China may opt to pursue more efficient and stage-based life cycle P management strategies under the combined action of multiple stakeholders (*SI Appendix*, section 5). Doing so will help China to achieve the cobenefits of P resource conservation and eutrophication mitigation, which will sustain future food production and attain a healthier water environment. For example, China could delay exhausting its P reserves by over 20 y by improving agronomic PUE to the average level of 80% in developed countries without impairing current crop yields (25). Measures that include improvement of aquaculture production, recovery of industrial and agronomic byproducts (e.g., phosphogypsum, animal excreta, crop straw), and shifts in human diets can also be undertaken (*SI Appendix*, Fig. S8). To

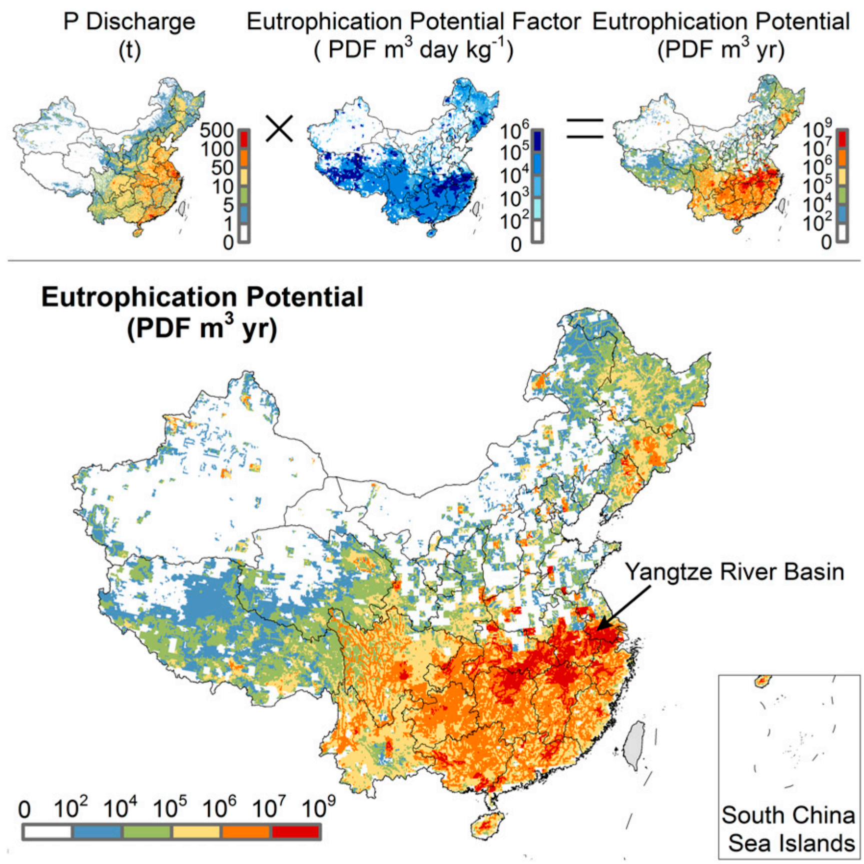

Fig. 3. EP induced by anthropogenic P losses in mainland China. EP values are computed as the number of potentially disappeared fraction (PDF) of species per cubic meter per year induced by eutrophication, meaning the fraction of species potentially lost from 1 m^3 of freshwater during 1 y as a function of freshwater availability and decreased biodiversity due to marginal increase in P concentration [as illustrated in the maps (*Inset*)]. Areas not included in this study due to insufficient data are shown in gray.

prolong the longevity of P reserves further, exploitation of legacy P reserves in soils, although still in preliminary stages (38), would provide an opportunity for improving China's P availability in the forthcoming generations (39). China is becoming increasingly aware of these issues, especially in response to water quality degradation. In 2015, the State Council of China announced a goal to reduce the environmental impacts of agricultural development via policies that will cap the use of P fertilizers by 2020 (40). Regardless of what future trajectory China follows, the past four centuries of change in P cycles are a record of one of the most rapid and profound biogeochemical disruptions that may someday mark a boundary event of the Anthropocene.

Materials and Methods

Temporal Pattern of P Cycle in China. We analyze the dynamics of P cycling in China over a four-century period with a hierarchical national P cycle model updated from the study by Yuan et al. (41), which includes 14 compartments as well as 102 separate P flows and stock changes. The model advances beyond previous socioeconomic P balances for China by including both major natural processes and anthropogenic metabolic activities with more detailed flow patterns over century time scales (a comparison with 15 other studies at the national scale of China is provided in *SI Appendix*, Table S1). P flows are simultaneously quantified in Microsoft Excel spreadsheets and R software to avoid miscalculations, with independent estimations as a first priority, although dependent calculations and process-based mass balances are used when information is limited. Data used in this study encompass time series of activity data and parameters. Activity datasets are collected from a number of international and national statistical databases, literature, and government documents, whereas most parameters are primarily derived from scientific research based on local field experiments (*SI Appendix*, Tables S3–S5 and Dataset S1).

Spatial Disaggregation of Anthropogenic P Runoff. Based on the assumption that anthropogenic P runoff is dependent on the intensity of relevant human activities, we further analyze the geographic patterns of anthropogenic P losses to mainland freshwaters with a high spatial resolution of 5 arc-minutes for the year 2012. We use various geographically explicit activity datasets to calculate the grid-based disaggregation factors, namely, the proportion of individual human activities (e.g., cultivation) in each grid cell relative to the aggregated national totals. The spatial disaggregation method can be represented by the following equation:

$$PL_{(i,j)} = PL_{total} \times DF_{(i,j)},$$

where $PL_{(i,j)}$ is P loss to surface waters from the grid cell with the longitude index i and the latitude index j, PL_{total} is the national total P losses to surface waters, and $DF_{(i,j)}$ is the spatially explicit disaggregation factor with the longitude index i and the latitude index j. It should be noted that the time domains differ among the original activity datasets. However, the spatial patterns are assumed to remain unchanged in 2012 regardless of the reference dates.

Freshwater EP of Anthropogenic P Runoff. Grid-based eutrophication potential factors (EPFs) are determined to evaluate the freshwater EPs induced by anthropogenic P runoff. The resultant environmentally relevant impact potential indicators, rather than estimations of actual eutrophication effects, provide more meaning to the P flow analysis results, which show only physical weights of environmental flows. An EPF is calculated by multiplying the spatially explicit fate factor (FF), describing P pathways through environmental media, with the effect factor (EF), representing the effects of a marginal P increase on freshwater ecosystems. Using global lake and reservoir databases, we update the spatially explicit FF model of P emissions to freshwaters from the study by Helmes et al. (31), which identifies advection, retention, and water use as three important processes that resulted in P removal. EF is determined from the empirical log-logistic relationships between the potentially disappeared fraction of temperate freshwater heterotrophic species and P concentration in two types of water bodies (lakes and streams), as developed by Azevedo et al. (32), with P concentrations in local freshwaters (*SI Appendix*, Fig. S9 and Table S6).

Uncertainty Analysis. To test the robustness of results, we conduct interactive cross-checks in P flow quantification and compare the results with previous estimates from China and other countries (*SI Appendix*, section 4). Furthermore, Monte Carlo simulation is applied to test the propagation of input uncertainties into the final results quantitatively. After activity data and parameters are provided with continuous distributions (triangular or uniform) after considering data quality, the model is run on 10,000 trials using randomly selected values from the input distributions to generate ranges of outcomes for 102 P flows (Dataset S2).

A fuller methodological description of this study is given in *SI Appendix*.

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