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Key Points:

- We describe the spatial variation of phosphorus (P) budgets in a series of nested watersheds
- We propose an empirical model for predicting riverine P export in similarly nested catchments
- In-stream retention is important in buffering P loading to the downstream, although the capacity is minor compared with landscape retention

Supporting Information:

- Supporting Information S1

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Anthropogenic Phosphorus Inputs to a River Basin and Their Impacts on Phosphorus Fluxes Along Its Upstream-Downstream Continuum

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Abstract The increasing trend in riverine phosphorus (P) loads resulting from anthropogenic inputs has gained wide attention because of the well-known role of P in eutrophication. So far, however, there is still limited scientific understanding of anthropogenic P inputs and their impacts on riverine flux in river reaches along the upstream-to-downstream continuum. Here we investigated P budgets in a series of nested watersheds draining into Hongze Lake of China and developed an empirical function to describe the relationship between anthropogenic inputs and riverine P fluxes. Our results indicated that there are obvious gradients regarding P budgets in response to changes in human activities. Fertilizer application and food and feed P import was always the dominant source of P inputs in all sections, followed by nonfood P. Further interpretation using the model revealed the processes of P loading to the lake. About 2%–9% of anthropogenic P inputs are transported from the various sections into the corresponding tributaries of the river systems, depending upon local precipitation rates. Of this amount, around 41%–95% is delivered to the main stem of the Huai River after in-stream attenuation in its tributaries. Ultimately, 55%–86% of the P loads delivered to different locations of the main stem are transported into the receiving lake of the downstream, due to additional losses in the main stem. An integrated P management strategy that considers the gradients of P loss along the upstream-to-downstream continuum is required to assess and optimize P management to protect the region's freshwater resource.

1. Introduction

Phosphorus (P) originating from anthropogenic sources as a pollutant of surface waters has been an environmental issue for decades because of the well-known role of P in eutrophication (Reed-Andersen et al., 2000). Human activities, such as food production and rapid urbanization, have been linked to increased terrestrial P inputs (Y. Han et al., 2011, 2013), which are often accompanied by corresponding increases in riverine P load (H. Han et al., 2012, 2010; Hong et al., 2013; Russell et al., 2008). Studies from Lake Erie (Scavia et al., 2014; Watson et al., 2016), Lake Taihu (Xu et al., 2010), and many other lakes (Havens, 2003) showed that such a rapid increase of riverine P inflow can significantly disrupt the original ecological structure of receiving waters, increasing the risk of algal blooms. It is also important for decision-makers to recognize the importance of the distribution of anthropogenic P inputs along nested watersheds from the headwaters to downstream reaches, because this significantly affects the overall watershed contribution to the riverine P fluxes of a receiving water body (Schwarz et al., 2006; Strokal et al., 2015) and corresponding environmental impacts.

Several researchers have used watershed-budget methods to study the negative impact of human activities (Groffman et al., 2004; Howarth et al., 2006; Sobota et al., 2011). The watershed approaches that account for all major inputs and outputs provide us with an estimate of net nutrient additions, which in turn can be used to assess the risk of potential pollution (H. Han et al., 2010; Russell et al., 2008). The well-known net anthropogenic nitrogen input (NANI) methodology is a relatively simple approach—originally proposed by Howarth et al. (1996), that has been used to estimate human-induced nitrogen (N) inputs to terrestrial and aquatic ecosystems (Alexander et al., 2002). A similar accounting approach has recently been developed for P, resulting in the calculation of NAPI (net anthropogenic phosphorus input) (Hong et al., 2012; Russell et al., 2008). NAPI can be calculated as the sum of its major components, which include P fertilizer application, nonfood P and P in net food and feed imports (Hong et al., 2012). Evidence from Lake Erie (H. Han et al., 2012), the Baltic Sea (Hong et al., 2012), Lake Michigan (H. Han et al., 2010), and some watersheds in China (Chen, Hu, Guo, et al., 2015; Chen et al., 2017; Zhang, Swaney, Hong, et al., 2015) showed that NAPI has advantages not only in predicting riverine P fluxes but also in determining the extent of P accumulation and surplus (Y. Han et al., 2011), in assessing riverine P pollution (Chen, Hu, Guo, et al., 2015; H. Han et al., 2012; Russell et al., 2008), and in evaluating the pattern of P retention (Hong et al., 2012).

Anthropogenic P inputs and in-stream P transport are two of the most important factors influencing P loading to the receiving water bodies (Withers & Jarvie, 2008). While most N transport is of dissolved forms, in-stream P transport is usually associated with particulate matter, which can settle out of the water column to become buried and unavailable (Lu et al., 2006; Roehl, 1962; Walling, 1983). Thus, hydrological factors (i.e., transport distance and flow rate) have potentially important controls on the degree of in-stream P retention (Withers & Jarvie, 2008), while anthropogenic P input represents the potential mass of P available for transport (Zhang, Swaney, Hong, et al., 2015). These two factors act together in determining the level of P in rivers. However, P input can itself have strong spatial heterogeneity along the length of a river, contributing substantially different P loads from headwaters to river mouth (Chen, Guo, et al., 2015; Russell et al., 2008). Under the joint impacts of P inputs and in-stream P transport along the continuum, the relationship between catchment sources and P loading to the receiving waters can become complex. However, most current P-budgeting studies that have been undertaken have focused on in-stream processing of nutrients (uptake and "spiraling," Doyle et al., 2003; Mulholland & Wollheim, 2002; Newbold et al., 1983, 1981; Runkel, 2007; Stream Solute Workshop, 1990). There remains an inadequate scientific understanding of the sequential variations of P budgets influenced by the hydrological and anthropogenic conditions as they vary along an upstream-to-downstream continuum. Such information is critical to improving our understanding of the behavior of anthropogenic P at the watershed level, given that there can be a significant gradient of climate, landscape topography, and human activity along the river in its basin, influencing P inputs and exports along the same gradient.

In this paper, we examine the spatial variability of NAPI and its impacts on riverine P fluxes along the river in the drainage basin of a large lake, the Hongze Lake Basin of China. Specifically, we investigate a sequence of eight monitoring sites along the main stem of the Huai River draining to Hongze Lake. This can be seen as a cascade of linked catchments, providing a framework for studying how P inputs and their impacts on riverine P fluxes at each site are interrelated from upstream to the downstream. To evaluate the long-term dynamics of P budgets, annual (2003–2010) data were used to calculate P inputs and outputs. We develop an empirical model, expressed by nonlinear regression of upstream riverine P flows, distance of hydrological transportation, watershed attributes, and contributions from P inputs to determine the influences of anthropogenic inputs on riverine P loading to the lake. The approach is broadly similar to, but simpler than, other spatially referenced statistical modeling approaches, notably SPARROW (Alexander et al., 2007; Schwarz et al., 2006; Smith et al., 1997), which has seen many applications in the United States and elsewhere (Alexander et al., 2008; García et al., 2011).

2. Methods

2.1. Study Area

Hongze Lake is the fourth largest freshwater lake in China. It was first formed at the end of the seventeenth century (Yin et al., 2013), when the Yellow River changed its course and merged with the Huai River. Sediment from the Yellow River blocked the course of the Huai River and diverted the water from the Huai, creating Hongze Lake. More than 70% of water storage in Hongze Lake comes from the main stem of the Huai River. As a part of the Huai River Basin, the Hongze Lake Basin is located between the Yangtze and the Yellow River Basins (Figure 1). The population in the basin is about 100 million. It is the most densely inhabited river basin and the main agricultural area of China, with nearly 630 persons per square kilometer. Precipitation is highly variable from year to year (the annual average for the period 2003–2010 was 963 mm), and more than 50% of annual precipitation is concentrated during the June–September period (Xia et al., 2011).

Eight incremental "sections" were delineated based on the eight corresponding monitoring sites along the main stem of the Huai River, such that the sum of all sections above a monitoring point comprises the drainage basin to that point. The total area of these sections is 123,861 km^2, accounting for 77% of Hongze Lake Basin. The sections encompass a wide variety of population density, land use types, and

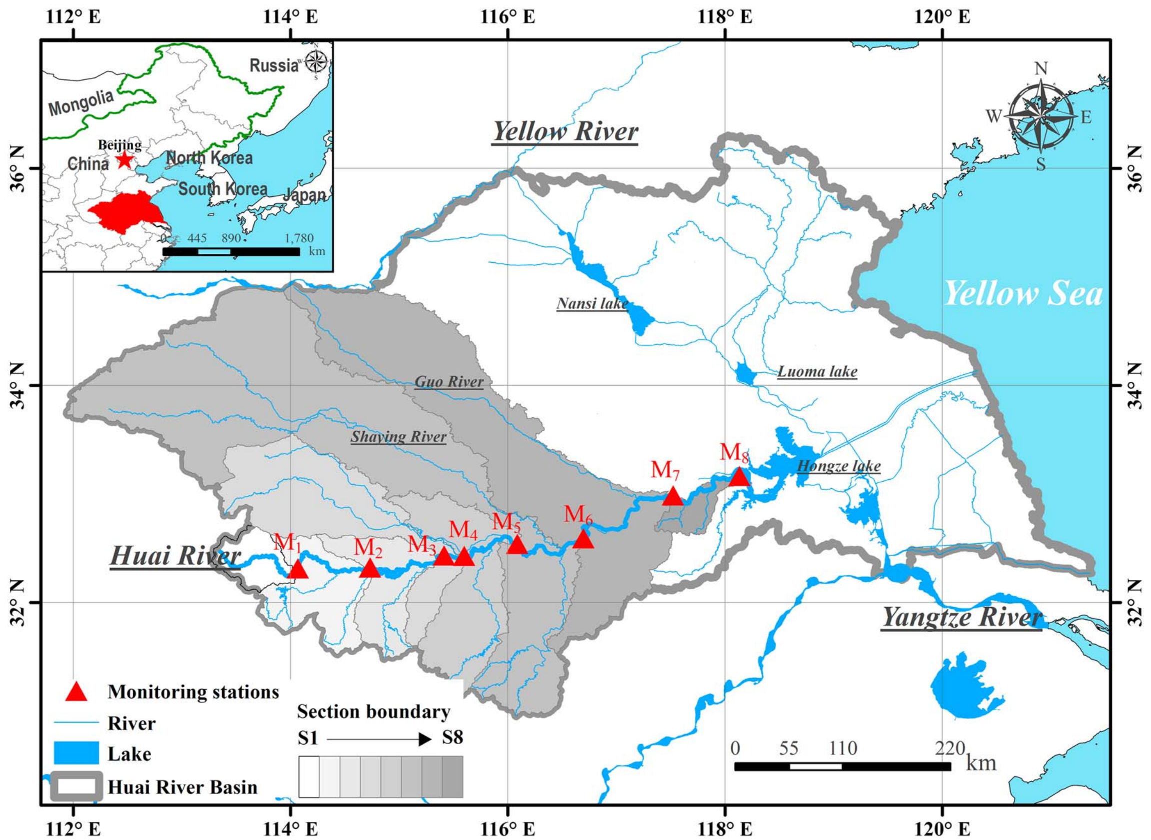

Figure 1. Study area location and boundaries of the eight watersheds used in constructing P budgets. These watersheds were delineated based on the water quality gaging stations (represented by triangles) of the main stem of the Huai River.

human activities (Table 1). There are substantial riverine P loads due to intensive agricultural production and rapid urbanization growth in recent years, causing lake eutrophication (Wang et al., 2015).

2.2. Estimation of P Inputs

For each section, we calculated components of a P budget using the NAPI approach (Howarth et al., 1996; Russell et al., 2008) by estimating the sum of P fertilizer application, P in net food and feed import, and import

Table 1
General Characteristics of Eight Sections (Incremental Watersheds) in Hongze Lake Basin for 2003–2010

ID	Area (km^2)	Length of the Huai River reach (km)	Total length of main tributaries (km)	Annual mean precipitation (mm)	Mean catchment elevation (m)	Annual mean air temperature (°C)	Average population density (km^{-2})	Forest + grass (%)	Wetland (%)	Agriculture + urban (%)
S_1	3,042	119	214	1,053	202	15.88	136	54.01	2.12	43.66
S_2	6,728	108	791	1,095	142	16.03	352	31.17	3.33	65.07
S_3	6,001	130	498	1,104	89	16.27	478	14.68	2.52	82.58
S_4	14,430	20	1,467	1,008	89	15.46	569	10.36	2.47	85.03
S_5	10,815	78	1,575	1,236	151	16.62	544	14.52	2.89	81.99
S_6	48,657	90	5,157	917	179	15.11	736	10.90	2.59	86.32
S_7	32,142	160	3,180	896	43	15.41	720	1.14	4.46	94.34
S_8	2,045	201	72	976	45	15.41	411	10.18	11.87	77.83
Entire watershed	123,861	905	12,954	973	122	15.49	642	10.95	3.27	85.35

Note. Eight sections (incremental watersheds) from the headwaters (S_1) to the farthest downstream, draining into Hongze Lake (S_8) were considered. Note that the data refer to the averages or totals corresponding to the incremental watershed between adjacent monitoring stations along the river (Figure 1). Total length of main tributaries here refers the length of the first-order and second-order tributaries of the Huai River. Annual average data of precipitation and air temperature were derived from China Meteorological Data Serive Center (http://data.cma.cn/en). Mean catchment elevation was calculated from GDEM 30 m data set, which were provided by Geospatial Data Cloud site (http://www.gscloud.cn). Land use data for each section were obtained from Chinese ecosystem assessment and ecological security database (http://www.ecosystem.csdb.cn/). Population density data (2003–2010) were collected from the annual Statistical Yearbook.

of nonfood P and seeding P. Seeding P (i.e., P embedded in seeds in agricultural planting operations) is often omitted from NAPI, because its contribution is usually negligible compared with other sources, but it has nevertheless been included in some previous studies (Y. Han et al., 2013). As our research area is one of the main agricultural areas in China (Bai & Shi, 2006), seeding P could be potentially significant, so we decided to include it in our analysis. Atmospheric deposition of P, which has been suggested to be negligible for this area (Mahowald et al., 2008), is difficult to quantify and so was not included, as in several other studies (Y. Han et al., 2011, 2013; Hong et al., 2012). The methodology used here in constructing NAPI is similar to that reported in Russell et al. (2008) and Y. Han et al. (2013), which was in turn adapted from the well-established N-budgeting methodology (i.e., NANI, Howarth et al., 1996).

2.2.1. P Fertilizer Application (P_{fert})

We obtained chemical P fertilization rates in each county from the Prefectural or Provincial Bureau of Statistics. Because P fertilizer is usually reported as P_2O_5 (Gao et al., 2001), we converted P fertilizer application to kg P by multiplying by 436.4 kg P per ton P_2O_5 applied and aggregated county-level fertilizer application to the watershed-level using the land use weighting method (H. Han & Allan, 2008; Zhang, Swaney, Hong, et al., 2015) (see the supporting information (SI)).

2.2.2. Seeding P (P_{seed})

We included vegetables and seven other primary agricultural crops in calculating seeding P input in each watershed and estimated seeding P by multiplying cultivated area for each crop type by the corresponding P input of seed per unit area in each county. We obtained statistical data on cultivated areas of crops from the Prefectural Bureau of Statistics of China and obtained rates of seeding P per unit area for each agricultural crop from Y. Han et al. (2013). Since the value of seeding P is only slightly different for different kinds of vegetable (Yang, 2004), we used cabbage for the vegetable category. Finally, we aggregated this county-level estimate to the watershed-level using the method of land use weighting (see SI).

2.2.3. Net Food and Feed Import (P_{im})

Net food and feed import is based on the assumption that imports and exports are determined by the balance of local consumption and production and is thus defined as P consumption (by animals and humans) minus P production (by crops and animal products) (Zhang, Swaney, Hong, et al., 2015). This value will be negative (representing an net export) if P production exceeds consumption. The amount of net food and feed import (P_{im}) can be calculated as follows:

$$P_{im} = P_{anim} + P_{hum} - P_{crop} - P_{pro,} \tag{1}$$

where P_{anim} and P_{hum} represent P consumption by animals and human, respectively. P_{crop} refers P in crops, and P_{pro} refers P in animal products.

We estimated human P consumption (P_{hum}) by multiplying the total population in each section by per capita intake of 0.52 kg P per year (Y. Han et al., 2011, 2013). Animals are usually fed according to straightforward dietary prescriptions designed for maintaining or gaining weight, so animal consumption of P in feed (P_{anim}) was calculated as the product of P consumption per individual and the number of each animal for every type in each section. We used the values of consumption reported by Zhang, Swaney, Hong, et al. (2015) and the values for the percentage P excreted reported by Van Horn (1998).

Animal products include meat, eggs, and milk. We estimated animal P production (P_{pro}) from the difference between animal excretion (waste production) and animal feed consumption (intake) (Zhang, Swaney, Hong, et al., 2015). We assumed that inedible and spoilage components caused about 7.3% loss of animal products available for human consumption based on the results of Liu et al. (2013). We estimated P in crops (P_{crop}) from the total mass of products and their P contents. P contents for different kinds of food were obtained from the book "China Food Ingredient Tables" (Yang, 2004).

2.2.4. Nonfood P (P_{non})

Nonfood P here mainly refers to the P-based detergents used for household cleaning. We used data for the period 2007–2009 from a national pollution census by the Chinese Ministry of Environmental Protection (Census, 2011). The census provides a per capita emission coefficient for P in detergents, which in our research area ranged from 0.23 to 0.42 kg P per person per year depending on the level of social development (Zhang, Swaney, Hong, et al., 2015). We then estimated nonfood P as the product of P input per person and the total population in each section.

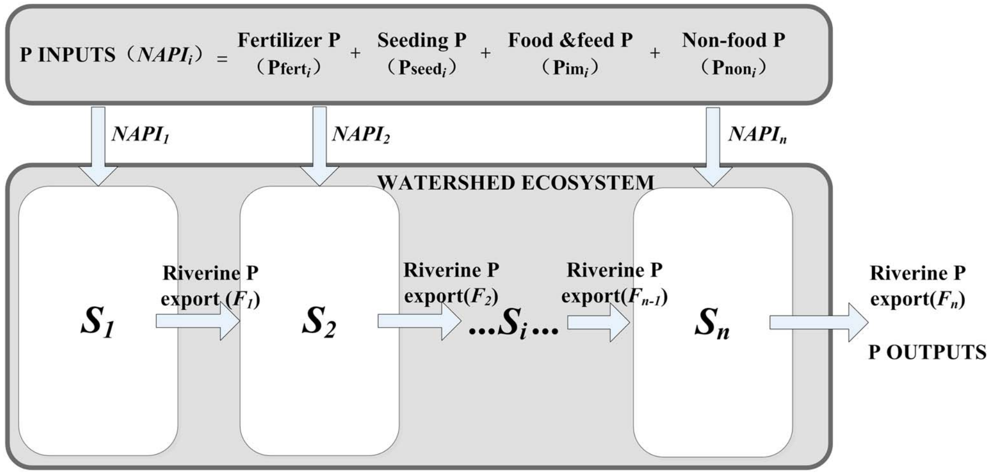

Figure 2. Conceptual diagram for P budget including inputs, system, and outputs in a series of incremental watersheds (i = 1, 2, …n).

2.3. Riverine TP Flux Estimate

We calculated TP flux (t P yr^{-1}) from the outlets of the ith section (F_i) using stream discharge and phosphorus concentration data with the LOADEST regression model (Runkel et al., 2004). Daily stream discharges were collected automatically at the eight hydrometric stations of the main stem of the Huai River from 2003 to 2010 (Figure 1). The water quality sampling data were obtained at the same stations. All of the above data were provided by the Huai River Commission (http://www.hrc.gov.cn/). The Huai River Commission follows strict data collection guidelines, namely, automatic monitoring of stream discharges follows the River Flow Monitoring Specification of China (MER, 1993) and water quality sampling follows the Regulation for Water Environmental Monitoring (MER, 1998). Flow data are accurate to within 90%–100%, with systematic error less than 2% (MER, 1993). TP (particulate + dissolved P) was determined by the K_2SO_4 degradation-ammonium molybdate spectrophotometric method (MEP, 2002). The measurement of TP concentration was controlled by calibrations using standard solutions. P concentration data are accurate to within 98.1%–100% (MER, 1998). For the years 2003–2006, all P concentration data were reported at a bimonthly time scale, while after 2007, these data were reported at a monthly scale. Some water samples failed to be documented because they were below the detection limit of the TP analytical method. In this study, we assumed that missing values are equal to the limit of detection (0.01 mg P L^{-1}). More detail about the estimate of riverine TP flux is provided in the SI.

2.4. Relating NAPI to Riverine TP Export via Empirical Modeling

We partitioned the entire watershed draining into Hongze Lake into adjacent, nonoverlapping sections, S_i (also called incremental watersheds), corresponding to the reaches between monitoring sites along the river (Figure 2). Thus, an nth nested watershed contained incremental sections S_1, S_2 …S_i …S_n (i = 1, 2, …, n, Figure 2). Conceptually, riverine TP fluxes (F_i) leaving the section S_i are derived from two sources: upstream P inflow (F_{i-1}) and sectional P inputs ($NAPI_i$). We assume that F_{i-1} is subject to retention in the main stem of the river, while $NAPI_i$ is subject to land-to-water processing, including landscape retention on and below the land surface and processing in tributary streams.

To determine the quantitative response of riverine TP export from each section i (F_i, t P yr^{-1}) to NAPI in that section ($NAPI_i$) and upstream P inflows (F_{i-1}), we evaluated the following relationship:

$$F_i = \exp(-\alpha * D_i) \cdot F_{i-1} + \left[\beta \cdot \exp\left(\gamma \cdot Z_{m,i}\right) \cdot \mathrm{NAPI}_i \cdot\right] \cdot \exp\left(-\delta * D_i^{'}\right), \tag{2}$$

where α (km^{-1}) is the first-order decay coefficient for P losses with respect to distance along the river and D_i (km) refers to the length of the corresponding river reach that hydrologically links S_{i-1} to S_i. F_i and F_{i-1} are riverine TP fluxes from the S_i and S_{i-1}, respectively. $Z_{m,i}$ represents driving variable m in S_i, β is the delivery coefficient, and γ the corresponding parameter relating change in delivery of NAPI to the driving variable. $D_i^{'}$ refers the total length of the first-order and second-order tributaries of the main stem lying within S_i. δ is the inverse retention length for the tributary retention. D_i and $D_i^{'}$ are presented in Table 1.

This first summation term represents P flux that derived from upstream P inflows after riverine retention, and if $i = 1$, then $F_{i-1} = 0$. Generally, the coefficient of river P delivery was often assumed to follow first-order reaction kinetics associated with hydraulic retention time (Bosch et al., 2009; Nixon et al., 1996; Smith et al., 1997). However, the hydraulic retention time, expressed by riverine transport distance divided by the flow velocity, is highly variable across space and time (Worrall et al., 2014). In order to reduce such variability and also to simplify the model estimates, we adopted a simple function as presented above to consider riverine retention, which assumed riverine delivery of P followed first-order decay associated with delivery distance (Reddy et al., 1999).

The second summation term represents riverine P flux derived from P inputs in that section after landscape retention, which is influenced by two stages of delivery: land-to-water delivery and in-stream delivery. $\beta \cdot \exp(\gamma \cdot Z_{m,i}) \cdot \exp(-\delta * D_i^{'})$ could be interpreted as the proportion of NAPI exported as riverine P fluxes. Specifically, $\beta \cdot \exp(\gamma \cdot Z_{m,i})$ is the proportion of NAPI_i that is delivered to receiving rivers as an exponential function of driving variables, m, denoted by $Z_{m,i}$. Variable m here could refer slope, stream density, wetland, precipitation, temperature, or disturbed land all of which have been demonstrated as important factors of land-to-water delivery (Schwarz et al., 2006). To determine the most efficient variable m for the model, potentially influential factors were added to the model one by one, independently. The factors retained in the model were those providing the best agreement between the model and the data (see Table S5 in SI). The term $\exp(-\delta * D_i^{'})$ is the proportion of mass in tributary channels that is transported to watershed outlets as a function of tributary channel length. This term assumed that in-stream delivery of NAPI followed first-order decay associated with the length of main tributaries. This is similar in form to the stream delivery factor defined in the first summation term of the flux equation, both of which assumed that in-stream delivery is associated with the channel length. Similar assumptions were also adopted by other models when flow data were not available (Schwarz et al., 2006).

Overall, the model structure presented in equation (2) is broadly similar to but simpler than SPARROW model (Alexander et al., 2007; Schwarz et al., 2006). SPARROW has been applied in China (e.g., Li et al., 2015; Zhou et al., 2017), and other similar models have also been developed for use in Chinese watersheds (e.g., Chen, Hu, Guo, et al., 2015; Zhang, Swaney, Hong, et al., 2015). A summary of the performance of the model structures considered here is presented in the SI.

Using equation (2), we can determine the contributions of riverine P loads from different individual sections to Hongze Lake. The equation can be expanded for each section, so that

$$F_1 = \beta \cdot \exp\left(\gamma \cdot Z_{m,1} - \delta \cdot D_1{'}\right) \cdot \mathrm{NAPI}_1 \tag{3}$$

$$\begin{aligned} F_2 &= e^{(-\alpha \cdot D_2)} \cdot F_1 + \beta \cdot \exp\left(\gamma \cdot Z_{m,2} - \delta \cdot D_2{'}\right) \cdot \mathrm{NAPI}_2 \\ &= \beta \cdot \exp\left(\gamma \cdot Z_{m,1} - \alpha \cdot D_2 - \delta \cdot D_1{'}\right) \cdot \mathrm{NAPI}_1 + \beta \cdot \exp\left(\gamma \cdot Z_{m,2} - \delta \cdot D_2{'}\right) \cdot \mathrm{NAPI}_2 \end{aligned} \tag{4}$$

Or in general,

$$F_n = \beta \cdot \sum\nolimits_{i=1}^{n} \left(e^{\gamma Z_{m,i} - \delta D_i{'} - \alpha \sum_{j=i+1}^{n} D_j} \cdot \mathrm{NAPI}_i \right). \tag{5}$$

Here α, β, γ, and δ are regression parameters. To determine the relative contributions of different individual sections to F_n, the TP flux to Hongze Lake, along the upstream-to-downstream continuum, we used the following function:

$$R_i = \beta \cdot e^{\gamma Z_{m,i} - \delta D_i{'} - \alpha \sum_{j=i+1}^{n} D_j} \cdot \mathrm{NAPI}_i \tag{6}$$

where $F_n = \sum_{i=1}^{n} R_i$. R_i is the P loading to Hongze Lake from the ith section, which represented the mass of riverine P load from the ith anthropogenic P inputs after retention in the landscape of section i as well as in successive river reaches below section i to the outlet.

Table 2
Summary Statistics for the Regression Model to Predict Riverine TP Flux

Parameters	Value	SD error	95% confidence interval Lower bound	Upper bound	t value	p value
α	0.00078	2.30×10^{-4}	3.17×10^{-4}	1.24×10^{-3}	3.38	<0.01
β	0.0059	0.0032	-4.63×10^{-4}	0.012	314.83	<0.01
γ	0.0016	3.95×10^{-4}	0.0024	8.07×10^{-4}	4.04	<0.01
δ	0.00017	5.69×10^{-5}	6.06×10^{-5}	2.88×10^{-4}	3.07	<0.01

2.5. Statistical Methods

In this study, we used Pearson correlation analysis to assess the strength of the relationships between P inputs and human activity, and used the p value (p) to evaluate the statistical significance of their relationships, using a criterion of $p < 0.01$ to establish significance. Annual data (2003–2010) for eight sections were used to fit the parameters of variants of equation (2). It should be noted that equation (2) is a nonlinear function requiring nonlinear fitting methods. The Levenberg-Marquardt algorithm was adopted to obtain the best values of the parameters. All model parameters were assigned initial values of 0 in the iteration algorithm, which proceeded until the convergence criterion was met (i.e., chi-square tolerance value $<10^{-9}$). Alternative versions of the models were assessed and compared by evaluating R^2 and Bayesian information criterion (BIC), an information theory-based measure that incorporates residual sums of squares and a penalty related to the number of model parameters (Schwarz, 1978). Descriptive statistics showing the ranges of parameters and their significance is shown in Table 2 below. A more detailed discussion of the methodology can be found in the supporting information (Cavanaugh, 2012; Kass & Raftery, 1995; Sinha & Michalak, 2016; Zhang, Swaney, Li, et al., 2015). Predicted and monitored TP fluxes were compared using the paired t test. All of the above analyses were performed using OriginPro software (OriginPro version 9.1, OriginLab Corp., USA, 2013). Nonlinear model fits were also checked using the Statistica software package, v 13.3 (Tibco Software, 2017).

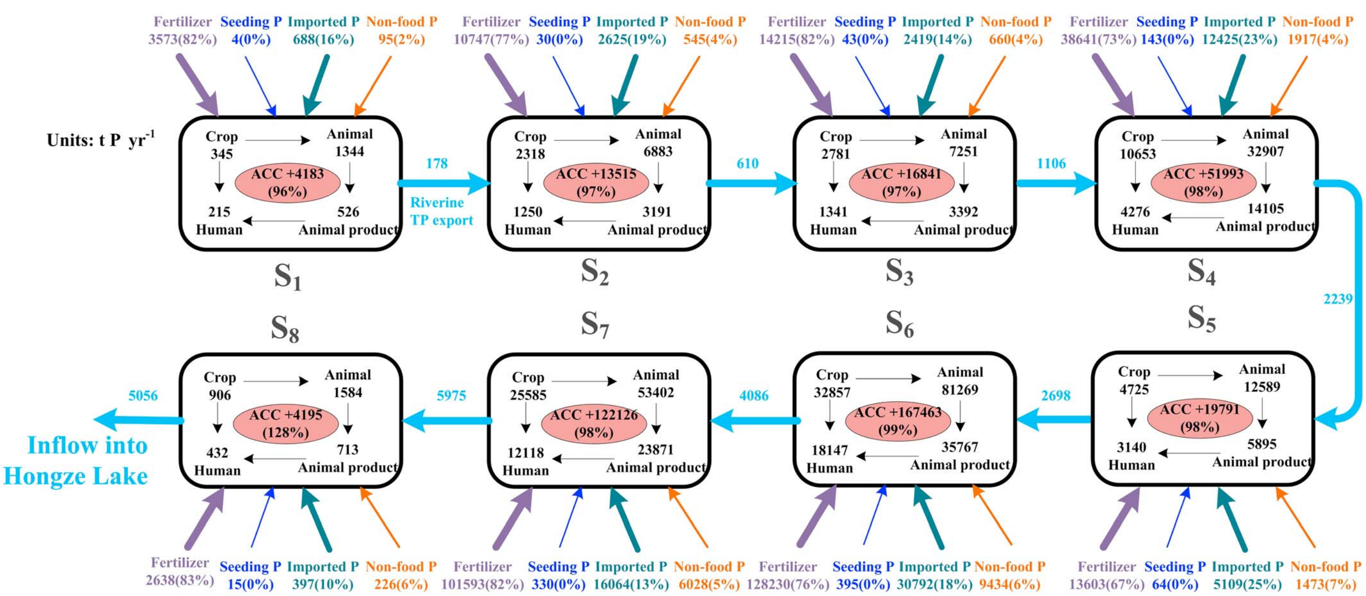

Figure 3. Distribution of section P budgets along the upstream-to-downstream gradient. Net anthropogenic P accumulation was estimated from the difference between P inputs and P outputs. The arrows pointing into the box refer the input items, while the arrows cycling inside the box refer P consumption (animal and human) and P products (crops and animal products).

3. Results

3.1. Sectional P Budgets

We used the annual NAPI and riverine TP fluxes for the 8 year record (2003–2010) to estimate the average annual P budget of each section along the river (Figure 3). P inputs to these sections ranged from 3267 to 168,851 t P yr^{-1}, with a total of 405,164 t P yr^{-1} for the whole basin. P inputs increased moving downriver from S_1 to S_8. Fertilizer P was always the largest source in all watersheds (accounting for 67~83% of NAPI) and had the highest percentage contribution in the eighth section (S_8 in Figure 3), but the lowest percentage contribution in the fifth section (S_5). The input of nonfood P made a slightly higher contribution in the sections downstream because larger populations tend to settle in these regions. Despite the agricultural character of the region, seeding P inputs were negligible compared with other sources across all sections (less than 1% of P inputs). As previous studies clearly documented (H. Han et al., 2010; Y. Han et al., 2011; Russell et al., 2008), the spatial distribution of P inputs was closely related to human activity, such as population, and the percentage of cropland and urban land ($p < 0.01$).

Annual riverine P exports varied widely across watersheds (Figure 3). The highest P export was 5975 t P yr^{-1} from the seventh station of the main stem, followed by the eighth station. There is a clearly increasing trend of P export from S_1 to S_7, corresponding to the cumulative effect of anthropogenic P inputs across sections. In contrast, P export from S_8 is obviously lower than that of S_7 indicating that retention of P exceeds incremental inputs from NAPI in S_8, implying that riverine P retention is a process of primary importance there. It is notable that the percentage of wetland in S_8 is significantly higher than other sections (see Table 1), which could account for the large amount of P retained.

High riverine retention in S_8 indicates that the equivalent of over 100% of P inputs from this section was retained, though some of it could have originated within it (Figure 3). Overall, about 4,000–17,000 t P yr^{-1} of anthropogenic P accumulated along the eight sections from upstream to downstream (Figure 3), with an overall 0.4×10^6 t P yr^{-1} accumulated for the whole basin. As with P inputs, the pattern of P accumulation was also associated with human activity, such as population, and the percentage of cropland and urban land ($p < 0.01$).

3.2. Empirical Model for Predicting Riverine TP Export

The calibrated results for models incorporating precipitation (mm yr^{-1}) explained annual riverine TP export with the highest adj-R^2 and with lowest BIC compared with the models incorporating other factors such as slope, wetland area, and temperature (see Table S5). This suggested that precipitation was one of the most important controlling factors of NAPI loss along the upstream-and-downstream continuum. Thus, riverine TP fluxes were estimated as

$$F_i = e^{-0.00078 \cdot \mathrm{D}_i} F_{i-1} + 0.0059 \cdot e^{0.0016 \cdot Z_{m,i} - 0.00017 \mathrm{D}_i'} \cdot \mathrm{NAPI}_i, \tag{7}$$

where $Z_{m,i}$ refers to the section-scale annual precipitation (mm) in S_i. The model explained 92% of the variation of riverine loads (Figure 4). All of the parameters in the function are statistically significant (Table 2).

In addition to predicting riverine P loads, the models evaluated in this study enable us to analyze the important processes affecting P loss in watersheds. The reliability of estimates of land-water delivery factors and in-stream transport can be evaluated to a limited extent in terms of the estimation error, 95% confidence interval, and the overall accuracy. The highest adj-R^2 and lowest BIC reported here (see Table S5) therefore encourage tentative use of the models in process interpretation. In this model, the value of the P assimilation coefficient α, 0.00078 km^{-1}, and corresponding "decay rate," suggest that, on average, riverine P loads entering a 100 km reach decrease by 7.5% over the length of the reach. Loads entering the river headwaters and traveling the entire 905 km of the river lose 50% of their mass.

The term, $0059 \cdot e^{0.0016 \cdot Z_{m,i}}$, suggested that only 2%–9% of yearly NAPI_i can be delivered into the river systems in the absence of the effects of tributary retention. Of this, around 41%–95% of the P mass present in the tributaries can be eventually transported to the watershed outlets of each section, as we further interpret the term $e^{-0.000171 \mathrm{D}_i'}$ as the component of retention along the length of the tributaries. The combined coefficient of NAPI_i, $0.0059 \cdot e^{0.0016 \cdot Z_m - 0.00017 \mathrm{D}_i'}$, exhibits spatial and temporal variation, with an overall average value of

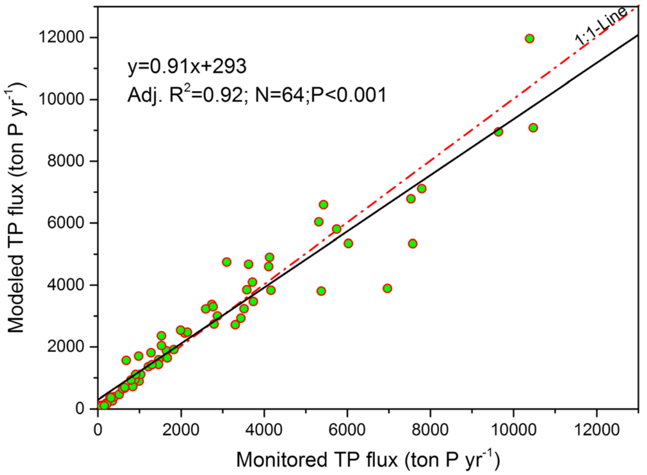

Figure 4. Modeled (*y* axis) versus monitored annual riverine TP exports (*x* axis) for 8 years from eight sections generated from simple regression. Paired *t* test showed that there is no significant difference at 99% confidence level between modeled and monitored values.

2.8% ± 1.3% (mean ± SD). For example, the fraction of NAPI exported as riverine TP fluxes in the third section (S_3) is highest, with a multiyear averaged value of 3.8%, and similar proportions (3.5%) occurring in the first section (S_1). The proportion of NAPI exported in rivers from S_6 is the lowest (1.1%). This may be largely due to the difference in precipitation and river length, both of which were interpreted as important controlling factors of P loss. Across the 8 years studied, the percentage of NAPI exported as riverine loads in a wet year (2003) is obviously higher than that of a dry year (2009), with average values of 4.3% and 1.9%, respectively. This is consistent with previous findings that higher precipitation will favor more P loss (Chen, Hu, Guo, et al., 2015; Sharpley et al., 2009).

Results of relative sensitivity analysis (see Table S3) revealed that the most sensitive model inputs include precipitation, F_{i-1}, fertilizer P (P_{fert}), and animal P consumption (P_{anim}). Hence, the parameters selected to calculate these inputs should be chosen most carefully as the uncertainties of parameters may influence model performance.

3.3. Contributions of Sectional P Inputs to Lake TP Loads

The model can provide valid information about managing the distributed upstream sources of P to Hongze Lake, which is sensitive to P loads because of the role of phosphorus as a limiting nutrient for freshwater eutrophication (Carpenter, 2005). The water quality of the river reach lying just upstream of Hongze Lake is a critical control point for load reduction.

As seen in equation (5), main stem retention is related to the total river length between each section and the lake, so the farthest upstream section delivers about 54% of the P load it receives and the farthest downstream section delivers all of it. Equation (6) allows us to estimate the contributions from the upstream sectional P inputs. The contributions derived from different sections showed large variation. The largest contributor of P inflows to Hongze Lake was the seventh section, accounting for 31% of P inflows to the lake (see Table S4). The sixth section contributed about 1,360 t P yr^{-1}, accounting for 26% of total flux to the lake. The trends were highly consistent with anthropogenic inputs (Figure 5a), suggesting that human activity was the major driver of riverine P loads. Because the sixth and seventh sections have the greatest P inputs and also because these two sections were located near the inlet of Lake Hongze without strong wetland interception as in S_8, more of their P is delivered into the lake. Excessive P loads from these sections could cause much more direct damage to the lake ecosystem than others. Sensitivity analysis of model components indicates

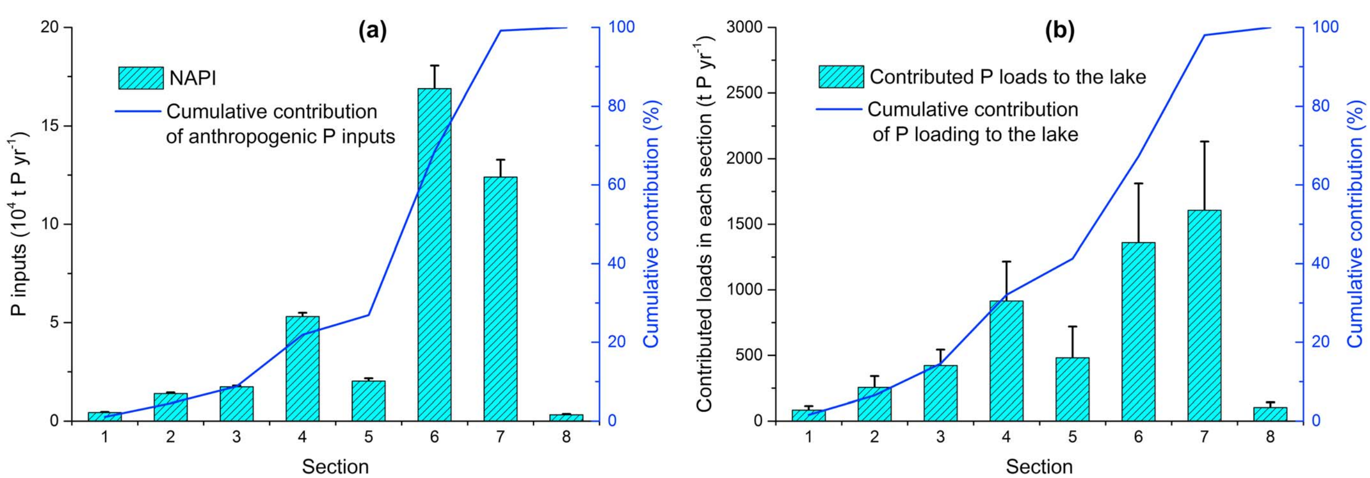

Figure 5. (a) Net anthropogenic P inputs and (b) the contributions of inputs sources to P loading to the Hongze Lake from different sections along the upstream-to-downstream continuum. The blue line in the figure indicates the cumulative P contributions moving upstream to downstream. P inputs in the figure (represented by stacked column) were affiliated with left *y* axis, while the cumulative P contributions (represented by the blue line) were affiliated with right *y* axis.

that riverine P exports are most sensitive to fertilizer P and animal P consumption, suggesting that the control of these sources should be a priority strategy of P management (see Table S3).

4. Discussion

4.1. Urgency of Reducing P Inputs

Our results show that P inputs to these sections ranged from 3,267 to 168,851 t P yr^{-1}, with an area-weighted average of 3,270 kg P km^{-2} yr^{-1} for the whole basin. This value is about 3 to 70 times that reported for watersheds in the Baltic Sea Basin (38–1,142 kg P km^{-2} yr^{-1} in the 2000s, Hong et al., 2012), and 6 times the average intensity of NAPI reported for mainland China (465 kg P km^{-2} yr^{-1} in 2009, Y. Han et al., 2014).

Such high levels of P inputs could impair the riverine ecosystem, because there are clear and direct relationships between anthropogenic inputs and riverine TP loads (Chen, Guo, et al., 2015; Chen, Hu, Yi, et al., 2015; Zhang, Swaney, Hong, et al., 2015). Low efficiency of P use is likely responsible for the high P loads to the Hongze Lake. Current crop P captures only 7.9%–25.8% of total agricultural P inputs (agricultural P = fertilizer P + manure P, manure P is estimated by animal P consumption minus P in animal products, Figure 3). The rest could potentially contribute to riverine P (Sharpley et al., 2001).

Our study reported that on average 2.8% of NAPI from each section enters the river as riverine loads, suggesting that the remaining 97.2% is retained in its watershed landscape. This kind of intensive P accumulation in the watershed landscape has fostered both a widespread buildup of P in soils and the potential future transport of these soils to aquatic systems (Stanley & Doyle, 2002; Zhang, Swaney, Hong, et al., 2015), resulting in higher P loss especially during storm events (Howarth et al., 1996; Zhang, Swaney, Hong, et al., 2015). Hence, to alleviate P loss, a priority measure is source management to reduce P inputs.

4.2. Implications for P Retention

The interpretion of P retention based on the model indicated that NAPI loading to the lake was controlled by a series of attenuation processes, including land-to-water retention and in-stream retention.

Our study reported that around 2%–9% of yearly NAPI from each section can be transported from land to water, suggesting that the remaining 91%–98% is retained in its watershed landscape. This high percentage of P retention, as well as similarly high retention shown by several studies in the Chesapeake Bay region (Russell et al., 2008), Lake Michigan (H. Han et al., 2010), Lake Erie (H. Han et al., 2010), and the Yongan River watershed (Chen, Hu, Guo, et al., 2015), consistently demonstrated that landscape P storage is the major fate of anthropogenic P inputs. Our results also indicated that the transport of NAPI was most sensitive to precipitation. Similar results were also reported by other modeling studies (Alexander et al., 2007; Chen, Guo, et al., 2015; Seitzinger et al., 2005). We believe that this may be attributable to the feature of P transport, which is usually associated with erosion of soil particles and subsequent sediment transport, as opposed to transport of dissolved forms (Withers & Jarvie, 2008). It is well known that erosion and movement of soil particles from land to water is highly dependent on precipitation (Sharpley et al., 2013, 2001).

As noted in section 3.2, around 5%–59% of NANI the river systems could be attenuated during transport from the tributaries to the main stem of the Huai River. This implies that the stream network has an important role in intercepting P inflows, a result consistent with previous findings. Némery and Garnier (2007) showed that P retention within a river network can be highly significant, with 15–30% of the riverine TP loads retained in the aquatic ecosystem. House (2003) reported that, in some instances, more than 30% of riverine TP loads can be stored in rivers, although riverine P retention was largely variable with streamflow and season. Based on long-term monitoring results, Mulholland (2004) also concluded that in-stream processes can remove about 30% of the riverine P loads annually.

In addition to the in-stream retention in the river network, main stem attenuation reduces upstream TP concentrations and exports, albeit to a lesser degree, and thus further lowers the risk of eutrophication of receiving water bodies. The percentage of P retained by riverine processes increases with the length of river reach because longer reaches allow more opportunity for P retention (Withers & Jarvie, 2008). As noted earlier, only 54% of P loads from the headwaters section are delivered into Hongze Lake. In contrast, about 86% of riverine TP loads from the adjacent-upstream sections (i.e., S_7) are transported to the lake. The mass of riverine P retention in the main stem is relatively small compared to small-stream studies (Mulholland, 2004; Newbold et al., 1983; Withers & Jarvie, 2008) but still yields an average P retention along the main stem of the Huai River of 2.21 t P km^{-1} yr^{-1}. It is worth noting that the in-stream retention estimated by our model (equation (7)) mainly represents the mean annual rates of P removal in the stream reach of each section. The mean annual retention reflects the steady state form and is insensitive to annual variation of streamflow, because we adopted riverine length as the response variable, but is sensitive to the magnitude of annual precipitation (and implicitly, annual flow variation), for which we have sufficient data to adequately fit the model. In addition, in-stream P processes considered in this study mainly include P sedimentation, which can remove P from the water and sequester it over annual and longer time scales (Stanley & Doyle, 2002; Walling et al., 2003; Withers & Jarvie, 2008). Seasonal removal processes (e.g., algal uptake) were not considered since these processes would not cause significant net removal of P over an annual period due to the seasonal recycling of nutrients in stream biota (Schwarz et al., 2006).

It is worth noting that the riverine P retention that we report in this study did not consider the influence of dams, which have been identified as an important controlling factor of riverine P retention (Nilsson et al., 2005; Stanley & Doyle, 2002).

4.3. Alternative P Model Structures Considered

In order to qualitatively explore alternative options, we tested several other possible models (see Text S5). These include an empirical model of the direct relationship between water quality and human actions (see Model variants I and III in Text S5), models with source-specific parameters for different delivery efficiencies for independent sources (see Model variant III), and models with explicit expressions showing the dynamic contribution from legacy P loss (see Model variant IV). All of these models or similar forms have been used widely in previous studies (Billen et al., 2009; Chen, Hu, Guo, et al., 2015; Schwarz et al., 2006; Zhang, Swaney, Hong, et al., 2015). Overall, most of the models we examined showed either marginally better results, but with statistically nonsignificant parameters, or marginally worse results, compared to the model presented here.

We also tested models introducing other spatial attributes (such as slope) in addition to precipitation. However, adding more spatial attributes that have been shown to be predictive in models of varying complexity (Mayorga et al., 2010; Smith et al., 1997) has not shown strong advantages in this case. We believe that introducing more factors can improve the physical basis of the model but simultaneously adds additional uncertainty in characterizing the complex processes of P delivery, especially given the limited data available for validation in this study.

We believe that the ultimate test of a model is how well it can simulate the system of interest subject to available data, usually assessed through validation. The relative performance of different model structures in validation depends to some extent on data availability—more detailed data permit more rigorous testing, potentially allowing identification of more complex model structures (Jackson-Blake et al., 2017). A related question is that of model resolution and scale: models of greater complexity may be necessary to describe data sets of finer resolution designed to assess management questions more appropriately addressed at finer scales of resolution (Alexander et al., 2008; Schwarz et al., 2006). Although our efforts to explore several other possible models have not shown better results than equation (7), we advocate continued attention to model development as additional data become available, with the aim of better parameterizing the complicated processes of P loss and the improvement of P management in these watersheds.

4.4. The Limitations and Potential Improvement of This Study

The basic framework of constructing the P model used here has been called a “black box” approach and has been widely adopted to predict P loads (Alexander et al., 2007; Chen, Hu, Guo, et al., 2015; H. Han et al., 2010; Hong et al., 2012). The terminology refers to a research unit (for example, a watershed) regarded as a black box in which the flow of P inputs and outputs are highlighted, but the details of P flows within the unit are not taken into account. Thus, the constructed models are very simple and more transparent in portraying the direct relationship between water quality and human actions, which in turn could promote simple policies to address them.

As with any black box approach, the negative aspect of the method is that it may be incapable of considering spatially distributed or regionalized information, which may be crucial for P prediction. For example, most P loss from a watershed is usually associated with soil particles as opposed to dissolved forms (Withers & Jarvie, 2008). The delivery of P from land to water is much more sensitive to spatially distributed factors (Zhang, Swaney, Hong, et al., 2015), such as precipitation, vegetation cover, and/or soil permeability, in relation to source locations (Sharpley et al., 2001). Much research has documented that P loss originates primarily from small sensitive areas (i.e., critical source areas or "hot spots," White et al., 2009; Kovacs et al., 2012) within watersheds during a few storms (Sharpley et al., 2003, 2001). These hot spots occur where high levels of P coincide with high precipitation or low vegetation interception and infiltration. Sharpley et al. (2009) documented that these hot spots (~20% of catchment areas) contributed the majority (~80%) of P losses, indicating their critical roles in environmental management (Sharpley et al., 2009). However, most of current black box methodology can only allow us to consider the generalized factors in catchment scale (for example, watershed-averaged precipitation) (Alexander et al., 2007; Chen, Hu, Guo, et al., 2015; H. Han et al., 2010; Hong et al., 2012), overlooking the relationship between critical source areas and associated control factors. The prediction of P loads using the framework of the black box methodology is capable of improvement, for example, simply by adding more spatially detailed meaning, for example, formulating functional relationships based on spatial correlations of distributed variables. Critical source areas and P loss events, as well as their critical controlling factors, can be considered as adequate spatially and temporally resolved data become available and eventually more targeted environmental management can benefit from their analysis.

The model considered here failed to show significant influence of legacy P on riverine fluxes. While many other well-constructed models have also not considered such impacts (Mayorga et al., 2010; Schwarz et al., 2006), as documented by other researchers, legacy P can be remobilized or recycled, acting as a continuing source to downstream water bodies for years, decades, or even centuries (Sharpley et al., 2013). However, developing a model that has the ability to accurately predict the long-term dynamics of the contributions of legacy P requires a long-term data record. Considerable research has been devoted to quantifying the contributions of legacy P. For example, Chen, Hu, Guo, et al. (2015) analyzed more than 30 years data to estimate the contribution of legacy P. Mittelstet and Storm (2016) used 90 years of data to quantify legacy P. Similar research on N even used decades or centuries of data to assess the influence of legacy stocks of N (Van Meter & Basu, 2016; Van Meter et al., 2017). This suggests that constructing a model using relatively short annual time series to include the dynamic contribution of legacy P is challenging, but as more data become available, the approach may be able to provide better estimates of its contribution.

There are several other lines of work that could improve future modeling results and thus provide further insights for management. First, we did not introduce source-specific export coefficients for each component of NAPI, even though they could be useful for source apportionment, because they are not supported by available data. The current results provide us with a perspective on how NAPI influences riverine P exports. As more data become available, watershed management should benefit from corresponding refinements in model structure. Second, our model did not consider the impacts of dams, which increase hydraulic residence time and trap sediment, both of which can increase riverine P retention. Future models could be refined by considering the potential impact of damming on these factors. Third, most of the data for estimating NAPI components were derived from county-level data sets. Errors associated with conversion of county scale to watershed scale are inherent in the method, although a more accurate conversion method has already been adopted (see Text S2 in the SI). More important is ensuring the quality and reliability of the county-level data itself. Another area of potential improvement is the refinement of parameters used to estimate P content of agricultural commodities produced locally, which may be different from those in other regions. Estimates of riverine P exports are also subject to improvement, mainly by increasing the frequency of water quality sampling.

5. Conclusion

This paper has presented a watershed budget analysis to investigate P input and riverine P export in Hongze Lake Basin, China and constructed an empirical model to explore the relationship between anthropogenic P inputs and their impacts on riverine fluxes. Our results show how anthropogenic P inputs from different sections of this basin collectively contribute to riverine P loads. P inputs to these sections ranged from 3,267 to 168,851 t P yr^{-1}, with a total of 405,164 t P yr^{-1} for the whole basin. Of the total NAPI, only about 2.8% contributed to the main stem of the Huai River as riverine loads. In-stream processes further reduced the riverine

P loading to the downstream receiving lake. Of the small proportion of NAPI reaching the main stem, riverine P loads were attenuated at the rate of 7.5% per 100 km, on average, as they travel from the upstream reach to the downstream lake. An integrated P management strategy that considers the interaction of spatially varying anthropogenic inputs and hydrological factors is required to assess and optimize P management to protect fresh waters.

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