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Factors controlling phosphorus export from agricultural/forest and residential systems to rivers in eastern China, 1980–2011

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SUMMARY

This study quantified long-term response of riverine total phosphorus (TP) export to changes in land-use, climate, and net anthropogenic phosphorus inputs to agricultural/forest ($NAPI_{AF}$) and residential ($NAPI_R$) systems for the upper Jiaojiang watershed in eastern China. Annual $NAPI_{AF}$ rose by 73% in 1980–1999 followed by a 41% decline in 2000–2011, while $NAPI_R$ continuously increased by 122% over the 1980–2011 period. Land-use showed a 63% increase in developed land area (*D*%) and a 91% increase in use of efficient drainage systems on agricultural land area (*AD*%) over the study period. Although no significant trends were observed in annual river discharge or precipitation, the annual number of storm events rose by 90% along with a 34% increase in the coefficient of variation of daily rainfall. In response to changes of $NAPI_{AF}$, $NAPI_R$, land-use and precipitation patterns, riverine TP flux increased 16.0-fold over the 32-year record. Phosphorus export via erosion and leaching was the dominant pathway for P delivery to rivers. An empirical model incorporating annual $NAPI_{AF}$, $NAPI_R$, precipitation, *D*%, and *AD*% was developed ($R^2 = 0.96$) for apportioning riverine TP sources and predicting annual riverine TP fluxes. The model estimated that $NAPI_{AF}$, $NAPI_R$ and legacy P sources contributed 19–56%, 16–67% and 13–32% of annual riverine TP flux in 1980–2011, respectively. Compared to reduction of $NAPI_{AF}$, reduction of $NAPI_R$ was predicted to have a greater immediate impact on decreasing riverine TP fluxes. Changes in anthropogenic P input sources ($NAPI_{AF}$ vs. $NAPI_R$), land-use, and precipitation patterns as well as the legacy P source can amplify P export from landscapes to rivers and should be considered in developing P management strategies to reduce riverine P fluxes.

1. Introduction

Enrichment of phosphorus (P) in surface waters is of great concern worldwide given its role in eutrophication and subsequent potential for hypoxia, loss of aquatic biodiversity and harmful algal blooms (Bowes et al., 2014; Fitzgerald et al., 2015). The extent of riverine P export is dependent on various factors, such as anthropogenic P inputs (Russell et al., 2008; Hong et al., 2012), hydroclimate (Sharpley et al., 2008), and land-use/land-management practices (Sobota et al., 2011; Duan et al., 2012). Therefore, an integrated and quantitative understanding of the factors controlling P export from landscapes is required for predicting the response of riverine P export to changes of human activities and climate, and effectively guiding watershed P management measures to protect water quality.

A P budgeting approach is useful for evaluating impacts of human activities on the P cycle by relating anthropogenic P inputs to outputs. Calculation of net anthropogenic phosphorus input (NAPI) is a P budgeting approach that sums annual P contributions from atmospheric deposition, fertilizer application, non-food input (i.e., detergent P), seed input, and net import/export in animal feed and human food supplies (Han et al., 2013). The NAPI approach has been successfully applied to many watersheds across Asia (Han et al., 2011b, 2013; Chen et al., 2015a), the Americas (David and Gentry, 2000; Borbor-Cordova et al., 2006; Han et al., 2011a; Russell et al., 2008; Sobota et al., 2011), and Europe (Hong et al., 2012). It is a simple yet powerful approach to evaluate net P inputs from anthropogenic sources to terrestrial ecosystems, as well as an effective tool to explain among-watershed or among-year variations in riverine P exports. However, the relationship between

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NAPI and riverine P export is additionally influenced by variations in hydroclimate and land management activities, as well as legacy P sources. It is commonly observed that years with higher precipitation or river discharge export a higher fraction of NAPI via rivers (Borbor-Cordova et al., 2006; Russell et al., 2008; Han et al., 2011a; Sobota et al., 2011; Hong et al., 2012; Chen et al., 2015a), and agricultural watersheds with tile drainage generally export a higher fraction of NAPI (Gentry et al., 2007; Han et al., 2011a; Morse and Wollheim, 2014). In addition, legacy P (i.e., surplus P stored in watershed landscapes that is derived from anthropogenic P inputs in previous years) can be remobilized or recycled after land management change (Meals et al., 2010; Sharpley et al., 2013) or when the degree of landscape P saturation increases after long-term excessive P addition (Kleinman et al., 2011), yielding a considerable P flux to downstream waterbodies (Jarvie et al., 2013; Haygarth et al., 2014; Chen et al., 2015a). As a result, changes in hydroclimate and land management as well as legacy P sources have the potential to enhance riverine P flux. Obviously, their influence is difficult to explicitly detect from short-term (several years) records, instead requiring a long-term (several decades) time series analysis.

For a watershed, chemical fertilizer, recycled manure, atmospheric deposition, and seed input are the primary P inputs to forest and agricultural lands, while residential lands mainly receive P from human and animal wastes. Phosphorus export from forest and agricultural landscapes to the river network is mainly via runoff and leaching (non-point sources). In contrast, a considerable proportion of P from residential areas enters the river network via direct sewage discharge (point sources). Relative to agricultural systems, the greater impervious surface area in residential systems further enhances P delivery efficiency due to decreased P retention capacity and enhanced P flushing/transport during rain events (Duan et al., 2012; Roy and Bickerton, 2014). As a result, residential systems have a higher potential to export NAPI than agricultural/forest systems. This is especially true in developing countries where efficient treatment of residential wastewater/stormflows and agricultural subsurface drainage are both often lacking (Hou et al., 2013; Chen et al., 2015b); thus human and animal wastes usually act as a major source of riverine P load in several regions in eastern China (Hou et al., 2013; Yuan et al., 2014). Therefore, it is valuable to separate watershed agricultural/forest and residential P budgets to effectively identify their relative contributions to riverine P fluxes.

This study provides a long-term (1980–2011) analysis of riverine P export in response to changes in NAPI components ($NAPI_{AF}$ vs. $NAPI_{R}$), hydroclimate, and land-use in the upper Jiaojiang watershed in eastern China. Specifically, this study (i) examines temporal variation of NAPI to agricultural/forest ($NAPI_{AF}$) and residential ($NAPI_{R}$) systems; (ii) addresses response of riverine TP flux to changes of $NAPI_{AF}$, $NAPI_{R}$, hydroclimate and land-use; (iii) develops an empirical model for linking $NAPI_{AF}$ and $NAPI_{R}$ to riverine TP flux; (iv) identifies the individual contributions from $NAPI_{AF}$, $NAPI_{R}$ and legacy P to riverine TP flux; and (v) forecasts riverine TP fluxes for future $NAPI_{AF}$, $NAPI_{R}$, land-use, and climate change scenarios. Based on records of riverine chloride and total suspended solids, agricultural soil P level and domestic waste treatment, we further addressed the major pathway for P delivery from landscapes to the river and fates of surplus $NAPI_{AF}$ and $NAPI_{R}$. Novel aspects of this study include improvement of the NAPI budgeting methodology to separately estimate watershed $NAPI_{AF}$ and $NAPI_{R}$ budgets and identification of their contributions to annual riverine TP flux. Developing such a quantitative understanding of the inter-relationships between multiple factors and P fluxes is essential for managers to determine which factors and sources should be targeted for P reduction.

2. Materials and methods

2.1. Watershed characteristics

The upper Jiaojiang watershed [120.23–121.01°E and 28.47–29.04°N; elevation ~15–1000 m above mean seal level (MSL)] is located in the highly developed Taizhou City area of Zhejiang Province, China (Fig. 1). The Jiaojiang River is the third largest river of Zhejiang Province and flows into Taizhou Estuary and the East China Sea, a coastal area that commonly experiences hypoxia (Gao and Zhang, 2010). The sampling location (Fig. 1) for this study was 55 km upstream of Taizhou Estuary at an elevation of ~15 m above MSL. The river drains a total area of 2474 km^2 and has an average annual water depth of 5.42 m [9.09 m in high flow regime (0–10% interval) and 2.80 m in low flow regime (90–100% interval)] and discharge of 72.9 $m^3 s^{-1}$ (411.3 $m^3 s^{-1}$ in high flow regime and 2.1 $m^3 s^{-1}$ in low flow regime) at the sampling location. The climate is subtropical monsoon having an average annual temperature of 17.4 °C and average annual precipitation of 1400 mm (Fig. 2a). Rainfall mainly occurs in May–September (~62%) with a typhoon season in July–September, while winter (December–February) is a major dry season (~12%). Although no significant trends in annual precipitation amount or average river discharge occurred over the 1980–2011 period (Fig. 2a), this watershed experienced a ~34% increase in the coefficient of variation for daily rainfall and a ~90% increase in the number of annual storm events (>50 mm per 24 h) (Fig. 2b). Such significant changes in the precipitation pattern imply a great potential to enhance nutrient export from watershed landscapes to rivers.

Data for P sources and sinks used to determine P budgets, land-use, domestic waste treatment, and recycled animal and human excreta for fertilizing croplands were derived from annual local statistics yearbooks and an investigation of rural ecological and environment quality conducted by the local agricultural bureau (Supplementary material A). Agricultural land (including paddy field, garden plot, and dryland) averaged ~12% of total watershed area in 1980–2011, with developed land (including rural and urban residential lands, roads, and mining and industry lands) and woodland, and barren land (including water surface, swamp, rock, and natural reservation land) contributing ~3%, ~67%, and ~18%, respectively (Table 1). The economic role of agriculture has been increasingly replaced by industry since the 1990s, resulting in a large reduction in chemical P fertilizer application (~38%) and cultivated crop area (~20%) since 2000 (Table 1). In 1980, 93% of animal and human solid waste was recycled on agricultural lands; this figure declined to 25% in 2011. The agricultural land area irrigated and drained with cement channels and pipes rose by 91% since 1980. Total population within the watershed increased from ~590,000 (95% in rural and 5% in urban) in 1980 to ~745,000 (89% in rural and 11% in urban) in 2011. Over the 32-year study period, domestic livestock production (pig, cow, sheep, and rabbit) declined ~25%, poultry production (chicken and duck) rose 4.8-fold, and freshwater aquatic species production (fish and shrimp) increased 11.7-fold (Table 1).

2.2. Agricultural soil P measurement

The upper Jiaojiang watershed is dominated by highly weathered and acidic soils [Oxisols (65%) and Ultisols (15%), mean pH < 5.5, Supplementary material B]. Soil samples were collected from the upper 20-cm layer of agricultural lands (i.e., paddy field, dry land and garden plots) in 1984 (n = 542) and 2009 (n = 3288). Upper 20-cm layer soil Olsen-extractable P (Olsen-P), total P (organic + inorganic P), and bulk density measured at the same location in both years were adopted to evaluate the changes

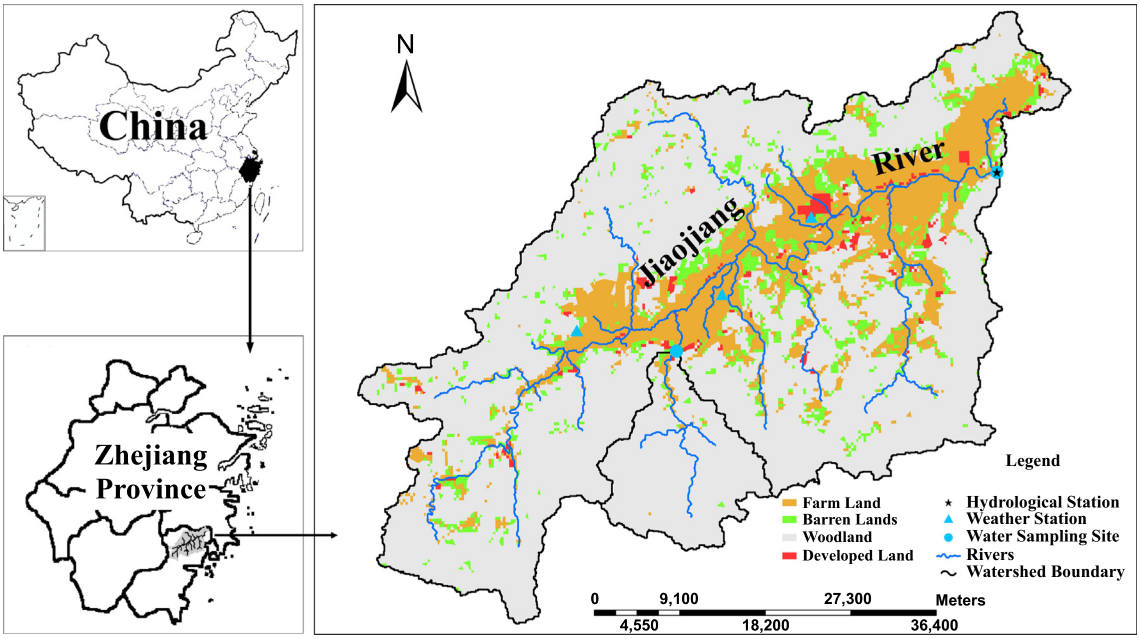

Fig. 1. Land use/land cover in the Jiaojiang watershed and the water quality sampling site and three weather stations.

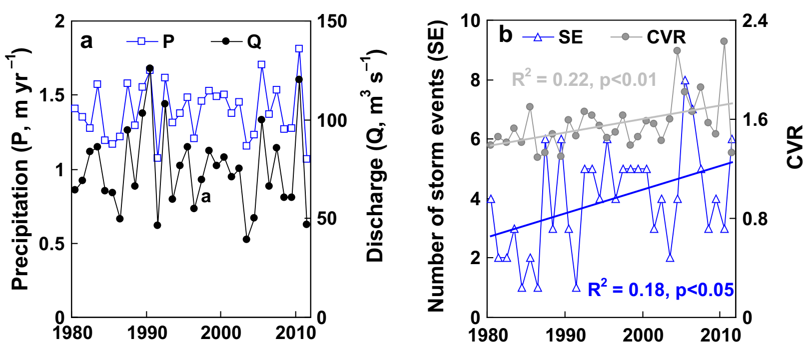

Fig. 2. Changes in (a) precipitation and discharge; and (b) number of storm events (>50 mm per 24 h), and coefficient of variation for daily rainfall (CVR) in the upper Jiaojiang watershed over the 1980–2011 study period.

between 1984 and 2009, resulting in time-paired Olsen-P content ($n = 460$), total P content ($n = 209$), and bulk density ($n = 195$). Net P accumulation in the upper 20-cm layer of agricultural soils between 1984 and 2009 was estimated using the measured changes in TP content and bulk density (Supplementary material B). Correlation analysis, regression analysis, and one-way analysis of variance in this study were performed using SPSS statistical software (version 16.0, SPSS Inc., Chicago, USA).

2.3. Riverine TP, chloride, and total suspended solids export estimates

Historical records (1980–2011) of river water quality (TP, $n = 183$; chloride, $n = 251$; and total suspended solids, $n = 170$) and daily river discharge at the watershed outlet (Fig. 1) were obtained from the local Environmental Protection Bureau and Hydrology Bureau, respectively (see Supplementary material C for field and laboratory protocols). Based on the discrete water quality records from 1980 to 2011, a regression model Load Estimator (LOADEST, Runkel et al., 2004) was applied to predict daily loads (estimated by multiplying pollutant concentrations and water discharge) of TP (dissolved + particle phosphorus), chloride (Cl^-), and total suspended solids (TSS), respectively (Supplementary material C). The calibrated LOADEST models had high R^2 values ($R^2 = 0.78$ for TP, $R^2 = 0.89$ for Cl^- and $R^2 = 0.96$ for TSS, $p < 0.001$) and low average relative errors (±4% to ±15%) between measured and modeled daily pollutant loads. Annual TP, Cl^-, and TSS fluxes (kg ha^{-1} yr^{-1}) were determined by dividing the annual TP, Cl^-, and TSS loads (calculated from the sum of daily loads for a corresponding year) by the watershed area. Annual average TP, Cl^-, and TSS concentrations were estimated as the ratio between annual load and cumulative river discharge. To address pathways of P delivery from landscapes to the river, changes in annual average riverine TP, Cl^- (as a conservative indicator for sewage, Jarvie et al., 2012) and TSS concentrations in high (0–10%) and low flow regimes (90–100%, Chen et al., 2013) were further examined. Average concentrations during the low and high flow regimes were estimated as the ratio between cumulative load and river discharge within the corresponding flow regime. To detect changes in trends for riverine TP concentration and flux as well as other variables (e.g., P source inputs, land use, and hydroclimate) over the period 1980–2011, regression analysis was adopted to establish the relationship between each variable and year number ($p < 0.05$ considered significant trend). The average increase in the percentage of each

Table 1
Characteristics of land-use distribution, population, domestic animal, and waste management for the upper Jiaojiang watershed over the 1980–2011 period.

Periods	1980s	1990s	2000s
Agricultural land (%)	11	11	13
Residential land (%)	2	3	3
Forest (%)	68	68	67
Barren land (%)	18	18	17
Annual area planted to crops (km^2)	637	601	470
Population density (capita km^{-2})	248	266	289
Livestock density (capita km^{-2})	100	86	62
Poultry density (capita km^{-2})	128	226	336
Freshwater aquatic products (ton yr^{-1})	3.2	13.1	26.8
Wastes treated in rural area (%)			
Solid	10	34	44
Liquid	0	10	15
Wastes treated in urban area (%)			
Solid	33	56	81
Liquid	15	35	60
Recycled animal excreta for fertilizing croplands (%)	95	75	57
Recycled rural/urban human excreta for fertilizing croplands (%)	90/80	65/40	25/10
Drained agricultural land area percentage (%)	19	18	29

The number of each type of livestock and poultry is converted into the number of pigs and chickens according to their phosphorus excretion rates as shown in Table C.5. All values are the average for each time period.

variable over time was estimated from the developed regression model.

2.4. Phosphorus budget calculations and uncertainty analysis

As in previous studies (Russell et al., 2008; Han et al., 2013), net anthropogenic P input budgets (NAPI, kg P ha^{-1} yr^{-1}) were calculated as the sum of five major components: atmospheric P deposition (APD), chemical fertilizer P application (CF), non-food input (NFI, i.e., detergent P), seed input (SI), and net food and feed input (NFFI):

$$\mathrm{NANI} = \mathrm{CF} + \mathrm{APD} + \mathrm{SI} + \mathrm{ABF} + \mathrm{NFFI} \qquad (1)$$

The NFFI (kg P ha^{-1} yr^{-1}) was composed of crop and livestock production and P consumption by livestock and human (Hong et al., 2012):

$$\mathrm{NFFI} = \mathrm{HC} + \mathrm{AC} - (G - \mathrm{GL} + \mathrm{RF}) - (\mathrm{AP} - \mathrm{APL}) \qquad (2)$$

where HC is human consumption, AC is animal consumption, *G* is harvested grain, GL is grain loss due to pests, spoilage and processing, RF is crop residual used as fodder, AP is animal products, and APL is spoilage and inedible components of animal products.

Phosphorus losses from forest and agricultural lands have similar delivery pathways (i.e., erosion and leaching), while P export from residential lands is associated with direct sewage discharge, erosion and leaching. Importantly, forest and agricultural lands (vegetation and soil at surface) have sharply contrasting surface characteristics compared to residential lands (highly impervious surface area). Therefore, we divided NAPI (kg P ha^{-1} yr^{-1}) into separate P budgets for agricultural/forest ($NAPI_{AF}$) and residential ($NAPI_R$) systems:

$$\mathrm{NAPI} = \mathrm{NAPI_{AF}} + \mathrm{NAPI_R} \qquad (3)$$

Furthermore, forest is the dominant land-use (>67%, Table 1) and only receives P from atmospheric deposition that accounts for <4% of annual NAPI in the watershed. Therefore, APD was fully considered as a new input for the agricultural/forest system:

$$\mathrm{NAPI_{AF}} = \mathrm{CF} + \mathrm{APD} + \mathrm{SI} + \mathrm{RAW} + \mathrm{RDW} - G - \mathrm{RF} \qquad (4)$$

where RAW is recycled animal excreta for fertilizing agricultural lands and RDW is recycled human excreta for fertilizing agricultural lands. $NAPI_R$ was then estimated as:

$$\mathrm{NAPI_R} = \mathrm{NFI} + \mathrm{HC} + \mathrm{AC} + \mathrm{GL} + \mathrm{APL} - \mathrm{RAW} - \mathrm{RDW} - \mathrm{AP} \qquad (5)$$

Detailed descriptions of individual P sources and sinks as well as the parameters used in estimating $NAPI_{AF}$ and $NAPI_R$ are available in Supplementary material A. To gain insight into the uncertainty of $NAPI_{AF}$ and $NAPI_R$ estimates, an uncertainty analysis was performed using Monte Carlo simulation (Supplementary material A). In performing the Monte Carlo simulation, we assumed that all the parameters used in the $NAPI_{AF}$ and $NAPI_R$ estimations followed a normal distribution with a 30% coefficient of variation for each parameter. A total of 10,000 Monte Carlo simulations were performed to obtain the mean and 95% confidence interval for annual $NAPI_{AF}$ and $NAPI_R$.

3. Results and discussion

3.1. Response of riverine TP export to changes in phosphorus budgets

In the upper Jiaojiang watershed, annual NAPI rapidly increased from 5.15 kg P ha^{-1} yr^{-1} in 1980 to 9.27 kg P ha^{-1} yr^{-1} in 2000 (Fig. 3), followed by a 15% decline from 9.03 kg P ha^{-1} yr^{-1} in 2000 to 7.68 kg P ha^{-1} yr^{-1} in 2011, a net increase of 49% over the 1980–2011 period. Considering the high percentage of forest and natural lands (>70%, Table 1), estimated NAPI was comparable with the average NAPI estimated for Zhejiang Province, China (7.14 kg P ha^{-1} yr^{-1} in 1981 to 9.72 kg P ha^{-1} yr^{-1} in 2009; Han et al., 2013). In terms of NAPI components, 42–63% was added to the agricultural/forest system ($NAPI_{AF}$), while the remaining 38–58% originated from residential systems ($NAPI_R$). Annual $NAPI_{AF}$ increased 75% in 1980–2000 and declined 38% over the 2000–2011 period. Annual $NAPI_R$ showed a continuous rise of 122% over the 32-yr study period due to a 13% growth in net food and feed P input and a 20-fold increase in non-food P input, as well as a 75% decline in recycled animal/human excreta for fertilizing croplands (Table 1). Increasing $NAPI_R$ implies that P flow from sewage and animal manures that originated as imported food/feed and detergent P was increased to support growing human population, poultry and aquaculture production (Table 1). The increasing sewage P discharge is supported by a 20.0-fold increase in Cl^-

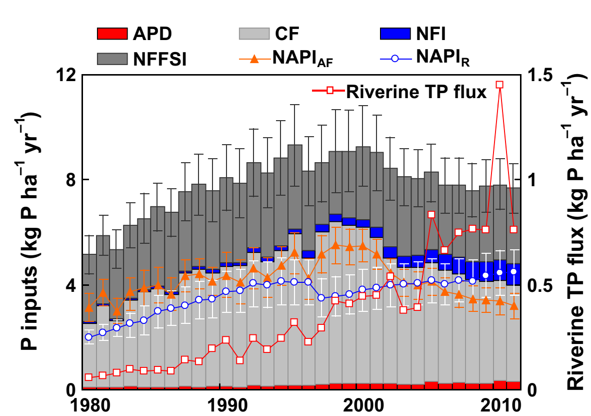

Fig. 3. Historical trends in net anthropogenic phosphorus inputs (NAPI) from atmospheric deposition (APD), chemical fertilizer (CF), non-food input (NFI), and net food, feed, and seed import (NFFIS) (seed input was <0.5% of NAPI and is not individually shown); NAPI to agricultural/forest ($NAPI_{AF}$) and residential ($NAPI_R$) lands; and riverine TP flux in the upper Jiaojiang watershed over the 1980–2011 study period. Error bars denote the 95% confidence intervals for NAPI, $NAPI_{AF}$, and $NAPI_R$ from Monte Carlo simulations.

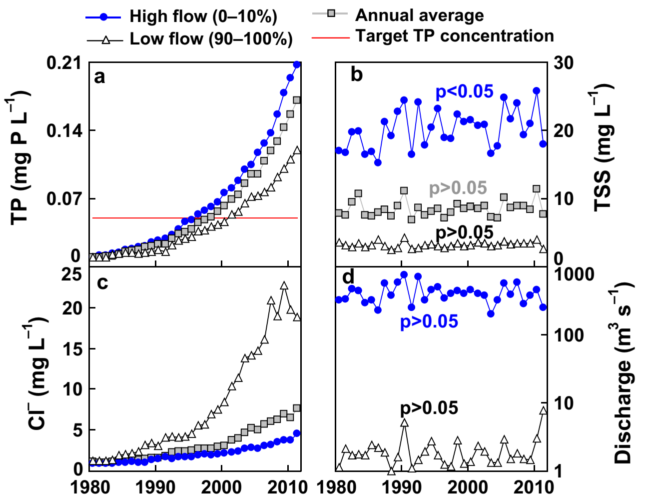

Fig. 4. Historical trends in annual average riverine TP, total suspended solids (TSS), and chloride (Cl^-) concentrations and water discharge during the low (90–100th interval) and high flow (0–10th interval) regimes in the upper Jiaojiang watershed over the 1980–2011 study period.

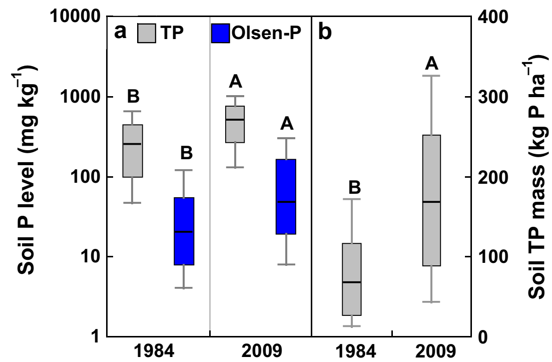

Fig. 5. Changes of total phosphorus (TP, $n = 209$) and Olsen-P ($n = 460$) contents (a) and TP mass in the upper 20 cm of agricultural soils between years 1984 and 2009 in the upper Jiaojiang watershed. Capital letters above bars denote significant differences ($p < 0.01$) based on one-way analysis of variance.

concentration and an 11-fold rise in TP concentration observed in the low flow regime over the past 32 years (Fig. 4a and c).

Over the 32-yr study period, annual riverine TP export continuously increased by 16.0-fold from an average 0.10 kg P ha^{-1} yr^{-1} in the 1980s, to 0.27 kg P ha^{-1} yr^{-1} in the 1990s, and 0.68 kg P ha^{-1} yr^{-1} in the 2000s (Figs. 3 and 4a). Since 1998, the river has exceeded the critical concentration of 0.05 mg P L^{-1} as an annual average suggested to reduce the risk of excessive algal growth (Li et al., 2010). This is especially significant during the high flow regime (Fig. 4a), since the high flow regime mainly occurs in the summer when high temperature and sunlight contribute to algae growth (Chen et al., 2013). As expected, annual riverine TP flux was significantly correlated with NAPI, consistent with previous studies (Borbor-Cordova et al., 2006; Russell et al., 2008; Han et al., 2011a; Hong et al., 2012). Similar to NAPI, both $NAPI_{AF}$ and $NAPI_R$ presented significant correlations with riverine TP flux (Table 2). Between 2000 and 2011, however, riverine TP fluxes continuously increased in spite of the decline or no significant change in NAPI or $NAPI_{AF}$ (Figs. 3 and 4a). Such contrasting trends imply a contribution of legacy P sources (Kleinman et al., 2011; Sharpley et al., 2015) or an increased fractional export of P inputs to rivers due to changes in land-use, hydroclimate, and NAPI components.

Table 2
Results of regression analysis between annual riverine TP flux (y) and various independent variables (x) in the upper Jiaojiang watershed in 1980–2011.

Independent	Regression equations	R^2	n
NAPI (kg P ha^{-1} yr^{-1})	$y = 0.0053\exp(0.50x)$	0.40**	32
NAPI to agricultural/forest land (kg P ha^{-1} yr^{-1})	$y = 0.13\exp(0.17x)$	0.12*	32
NAPI to residential land (kg P ha^{-1} yr^{-1})	$y = 0.0049\exp(1.10x)$	0.71**	32
Precipitation (m yr^{-1})	$y = 0.14x^{2.01}$	0.19**	32
River water discharge (m^3 s^{-1})	$y = 0.28x^{0.38}$	0.17**	32
Number of storm events (>50 mm per 24 h)	$y = 0.096x^{0.82}$	0.27**	32
The coefficient of variation for daily rainfall	$y = 0.0043x^{1.58}$	0.14**	32
Annual total suspended solid (mg L^{-1})	$y = 0.0022x^{2.26}$	0.13*	32
Drained agricultural land percentage	$y = 8.23x^{2.24}$	0.47**	32
Developed land percentage	$y = 7584.8x^{3.41}$	0.46**	32

** Significant $p < 0.01$.
* Significant $p < 0.05$.

3.2. Response of riverine TP export to changes in land-use and hydroclimate

Although no significant trends were observed in annual precipitation or river discharge over the 32-year record ($p > 0.05$, Fig. 2a), annual riverine TP fluxes were positively correlated with precipitation and river discharge (Table 2). Higher precipitation and river discharge enhance erosion and leaching that subsequently increase the fractional export of P inputs by rivers (Borbor-Cordova et al., 2006; Morse and Wollheim, 2014). Eroded sediments are typically enriched with P relative to the bulk soil (up to a factor of five times) due to the preferential removal of fine soil particles having a greater P content (Sharpley et al., 2002). Importantly, the watershed experienced a 90% rise in the number of annual storm events (>50 mm per 24 h) and a 34% increase in variability of daily rainfall amounts over the past 32 years (Fig. 2b), which is consistent with previously reported results (PGZP, 2010). Increased prevalence of storm events resulted in a considerable increase in P export from watershed landscapes to rivers (Table 2). Increasing the number of storm events as well as the variability in daily rainfall implies a greater erosivity and greater depth of leaching, which enhance P loss by erosion and leaching (Shigaki et al., 2007; Sharpley et al., 2008). The influence of storm events on P export to rivers is supported by a 20% increase in total suspended solids (TSS) concentration during the high flow regime (Fig. 4b). There was also a significant correlation between annual average TSS concentration and riverine TP flux (Table 2). Phosphorus loss during storm events would be further enhanced for soils that have received high rates of P amendments for a long time period, since P retention is inversely correlated with the saturation of a soil's P sorption capacity, especially for the topsoil horizon (Kleinman et al., 2011; Sharpley et al., 2013). This premise is supported by a 2.0-fold rise in TP concentration and a 1.4-fold increase in Olsen-P concentration in the upper 20 cm of agricultural soils between 1984 and 2009 (Fig. 5a).

Although the developed land area only accounts for ~3% of the entire watershed area (Table 1), it received 46% of cumulative NAPI over the past 32 years. Therefore, increasing developed land area showed a significant impact on annual riverine TP flux (Table 2). Expanding developed land area decreases P retention capacity and enhances flushing (i.e., runoff) during rainfall events due to a greater impervious surface area (Roy and Bickerton, 2014). Also, more developed land area and population density implies more waste production (Table 1) and greater P input to rivers via wastewater discharge directly into rivers (Sobota et al., 2011). This is especially significant since centralized wastewater collection is limited (<30% on average, the remaining percentage is directly discharged to local water bodies and soils) in the study area (Table 1). However, the much larger difference between high and low flow river discharge regimes (196-fold, Fig. 4d) compared to Cl^- concentration (4.0-fold, Fig. 4c) indicates that riverine Cl^- is not merely derived from sewage discharge. Therefore, Cl^- as well as P loss from rural and urban landscapes (e.g., yards, roads, pools/parks, greenbelts and waste landfills) via erosion and leaching might be an important pathway in the residential system. The increasing percentage of agricultural land area drained with cement channels and pipes, which improves hydrologic connectivity and enhances P delivery from land to rivers (Gentry et al., 2007; Jarvie et al., 2013), also showed a significant positive correlation with annual riverine TP flux (Table 2). The improved agricultural drainage system (e.g., tile drainage system) can increase dissolved P leaching through the soil and into drain tiles since there was an available pool of soil P that readily desorbed during rain events. This is likely to play an important role in transporting legacy P to rivers (Kleinman et al., 2011). Additionally with tile drainage, particulate P losses may be enhanced due to transport of fine clay particles during periods of preferential flow through the soil into drain tiles (Gentry et al., 2007).

Overall, changes of land-use and hydroclimate increased the fraction of P exported via enhanced soil erosion and sediment transport as well as direct wastewater discharge, decreased water residence time, greater P leaching, and reduced stream sedimentation. However, the high flow regime presented higher TP concentration (32-yr average 0.068 vs 0.043 mg P L^{-1}) as well as a greater rate of increase (16.0-fold vs 10.0-fold) compared to the low flow regime (Fig. 4a). This implies that river water discharge has a positive relationship with TP concentration (i.e., nonpoint source pollution is the dominant delivery process, Chen et al., 2013 and Bowes et al., 2014). Furthermore, annual riverine TP flux was positively correlated with precipitation, water discharge, and TSS average concentration (Table 2). These results implied that P loss via erosion and leaching is the dominant pathway for P delivery from agricultural/forest and residential landscapes to the river compared to wastewater discharge (i.e., nonpoint vs. point source pollution).

3.3. Models for predicting riverine phosphorus flux

The analyses reported above indicate that year-to-year variation of riverine TP flux was related to NAPI, $NAPI_{AF}$, $NAPI_R$, land-use, and hydroclimate, suggesting that the integrated influence of these factors should be considered in developing predictive models. The relationship between riverine TP export and NAPI or $NAPI_{AF}$ or $NAPI_R$ is best described using an exponential function and the relationship between riverine TP export and land-use and hydroclimate variables is best described using a power function (Table 2). Thus we developed the following model to predict annual riverine TP export (F, kg P ha^{-1} yr^{-1}) over the 1980–2011 period:

$$\begin{aligned} F &= \alpha x_i^{\beta_i} y_j^{\beta_j} \exp(\beta_3 \mathrm{NAPI}) \text{ or} \\ F &= \alpha x_i^{\beta_i} [DA\%^{\beta_2} \exp(\beta_3 \mathrm{NAPI_{AF}}) + D\%^{\beta_4} \exp(\beta_5 \mathrm{NAPI_R})] \end{aligned} \tag{6}$$

where x_i represents hydroclimate variables [i.e., annual precipitation (P, m $year^{-1}$), river water discharge (Q, m^3 s^{-1}), number of storm events (SE), and variability of daily rainfall (CVR)], y_j represents land-use variables [i.e., developed land area percentage ($D\%$) and agricultural land area percentage with improved drainage systems ($DA\%$)], and α, β_i, β_j, β_3, β_4 and β_5 are fitting parameters.

The unknown parameters in Eq. (6) were calibrated for 24 potential combinations of the various influencing factors (Supplementary material D, Table D.1). Compared to models using NAPI (R^2 = 0.54–0.89), models that separately incorporated $NAPI_{AF}$ and $NAPI_R$ explained annual riverine TP flux with higher accuracy (R^2 = 0.78–0.96). This implies a significant difference in TP delivery efficiency to rivers between $NAPI_{AF}$ and $NAPI_R$. Compared to NAPI or $NAPI_{AF}$ and $NAPI_R$, further inclusion of hydroclimate or land-use variables improved the model accuracy. Among the various combinations of variables, the combination of $NAPI_{AF}$, $NIPI_R$, P, $D\%$, and $DA\%$ provided the best predictor of riverine TP export (Table D.1):

$$F = 0.66P^{1.30}[DA\%^{2.67} \exp(0.54\mathrm{NAPI_{AF}}) + D\%^{2.08} \exp(1.42\mathrm{NAPI_R})] \tag{7}$$

Eq. (7) explained 96% of the variation in annual TP fluxes over the 32-yr study period with a Nash–Sutcliffe coefficient of 0.95 and mean relative error of <10% between modeled TP flux and LOADEST-estimated values (Fig. 6a). Considering the complexities of P biogeochemistry and transport in terrestrial landscapes and streams, the performance of model Eq. (7) is very reasonable in comparison with physical-based models [e.g., SWAT, AGNPS, and HSPF, reviewed by Moriasi et al. (2007)]. Physical-based models typically require a large amount of data to calibrate the many model parameters, making their implementation difficult for many watersheds (Chen et al., 2013). Although our empirical model lacks the ability to predict seasonal or daily riverine TP export and differentiate among possible fates of P compared to more complex models, it has the advantage of simplicity (especially for large spatial scales) and can easily be applied to predict riverine TP export from watershed anthropogenic P inputs, hydroclimate, and land-use. It should be pointed out that model Eq. (7), which is supported by the observed relationships between riverine TP flux and each of the relevant influencing factors (Table 2), is site specific for the Jiaojiang watershed. The influencing factors and function types in Eq. (7) may change and require optimization for application to other watersheds. However, the methodology proposed in this study for linking P budgets and riverine TP flux should be widely applicable to other watersheds.

3.4. Riverine TP source apportionment

This study assumes that the component of riverine TP export that is not explained by $NAPI_{AF}$ and $NAPI_R$ in Eq. (7) represents the contribution of legacy P sources from the agricultural/forest system (F_{LA}) and the residential system (F_{LR}), if the contribution from natural rock weathering is negligible. The legacy contributions were estimated as:

$$F_{LA} = 0.66P^{1.30}DA\%^{2.67} \tag{8}$$

$$F_{LR} = 0.66P^{1.30}D\%^{2.08} \tag{9}$$

We further determined annual riverine TP flux derived from $NAPI_{AF}$ (F_{AF}, kg P ha^{-1} yr^{-1}) and $NAPI_R$ (F_R, kg P ha^{-1} yr^{-1}) as:

$$F_{AF} = 0.66P^{1.30}DA\%^{2.67}[\exp(0.54\mathrm{NAPI_{AF}}) - 1] \tag{10}$$

$$F_R = 0.66P^{1.30}D\%^{2.08}[\exp(1.42\mathrm{NAPI_R}) - 1] \tag{11}$$

Over the 1980–2011 period, the estimated annual $NAPI_{AF}$-contributed TP flux ranged from 0.03 to 0.32 kg P ha^{-1} yr^{-1} and continuously increased by 5.6-fold, which accounted for 19–56% of annual riverine TP flux (Fig. 6a). This estimate is comparable with the 0.2–1.2 kg P ha^{-1} yr^{-1} lost from agricultural lands over six major regions across China (Li et al., 2015). Since there was a significant decline in $NAPI_{AF}$ between 2000 and 2011 (38% decrease, Fig. 3), the 12% increase of $NAPI_{AF}$-contributed TP flux mainly resulted from an enhanced export fraction of $NAPI_{AF}$ due

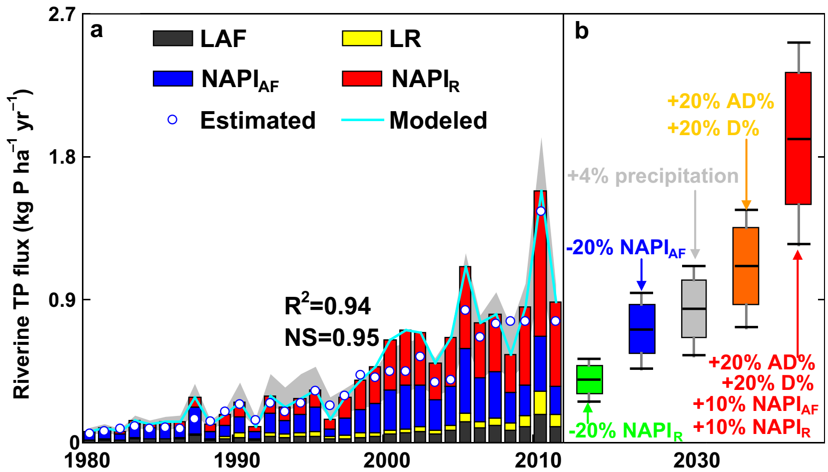

Fig. 6. (a) LOADEST-estimated versus modeled riverine TP fluxes using Eq. (7) and estimated contributions of $NAPI_{AF}$, $NAPI_R$, and legacy P sources in agricultural land (LAF) and residential land (LR) to annual riverine TP flux in 1980–2011 and (b) predicted TP flux changes in 2030 for future trends in $NAPI_{AF}$, $NAPI_R$, precipitation, developed land area percentage (*D*%) and percentage of agricultural lands with improved drainage (*AD*%) in the upper Jiaojiang watershed. Shadow area denotes the 95% confidence interval of the modeled TP fluxes. NS denotes Nash–Sutcliffe coefficient.

to a 69% rise in agricultural land area with improved drainage systems and an 8% increase in storm events during this time period. In comparison, the estimated $NAPI_R$-contributed TP flux ranged from 0.01 to 0.93 kg P ha^{-1} yr^{-1} and continuously increased by 29.4-fold, which accounted for 16–67% of the annual riverine TP flux (Fig. 6a). Such a rapid increase in $NAPI_R$-contributed TP flux resulted from a marked rise in $NAPI_R$ and an enhanced export fraction of $NAPI_R$ due to increasing developed land area and the number of storm events as well as direct waste discharge. However, the 29.4-fold increase observed in $NAPI_R$-contributed TP flux is somewhat higher than the 20.0-fold increase in Cl^- concentration during the low flow regime (Fig. 4c). This suggests that export of $NAPI_R$ to rivers is mainly associated with erosion and leaching processes in addition to direct sewage P discharge, consistent with results discussed above.

Compared to many physical-based models (in which legacy P source contributions to riverine TP flux are not well addressed, Kleinman et al., 2011), an important feature of the model developed in this study is its ability to easily quantify the contributions of legacy P sources to annual riverine TP flux. The legacy P sources in the agricultural/forest system comprised a considerable contribution (8–27%) to annual riverine TP export, while the legacy P sources in the residential system contributed 3–6%. Watershed legacy P sources in total contributed 13–32% of the annual riverine TP export (Fig. 6a). Annual legacy P contribution to the TP flux displayed an increasing trend ($p < 0.01$) with a 6.6-fold total increase, which was stimulated by changes in land-use and hydroclimate after long-term excess P inputs (Jarvie et al., 2013; Haygarth et al., 2014). Due to legacy P contributions, field studies indicated that even without additional fertilizer application, a decade or more of "P draw down" from agricultural soil P reserves would be required to substantially reduce P in runoff (Jarvie et al., 2013; Sharpley et al., 2013). Modeling results also indicated that in-stream remobilization of legacy P could account for 21–26% of annual riverine TP load in two Illinois Rivers (Jarvie et al., 2012) and 2–29% in the Yongan River in eastern China (Chen et al., 2015b). Several investigations conducted in urban streams suggested that enrichment of soluble-reactive P in groundwater due to sewage leaching can contribute a considerable proportion of riverine P flux, especially during the low flow season (Roy and Bickerton, 2014; Fitzgerald et al., 2015).

It should be pointed out that the method developed (Eqs. (8) and (9)) for estimating the legacy P contribution to the TP flux is based on the assumption that the amount of P released by rock chemical weathering is negligible. For the upper Jiaojiang watershed, the dominant soils are strongly weathered red and yellow soils (~90% of entire watershed area; Oxisols and Ultisols, Chen et al., 2015a) that are believed to contribute a very limited background P flux compared to anthropogenic P sources. For example, a study in a red soil dominated watershed of Lake Dianchi in southern China determined 0.006 kg P ha^{-1} yr^{-1} contributed by natural rock chemical weathering (Liu, 2005). This background chemical weathering rate for P is considerably less than the estimated contribution of legacy P to riverine TP fluxes observed in this study (Fig. 6a).

3.5. Fates of surplus phosphorus

Of the cumulative NAPI over the past 32 years for the upper Jiaojiang watershed, 4.8% was exported via river discharge. This estimate falls within the range reported for many studies in American, European and Chinese watersheds (i.e., 1–38%, David and Gentry, 2000; Borbor-Cordova et al., 2006; Russell et al., 2008; Han et al., 2011a; Sobota et al., 2011; Hong et al., 2012; Chen et al., 2015a). Of cumulative riverine TP flux (12.0 kg P ha^{-1}) over the 32-yr period, 48% was derived from the agricultural/forest system, while 52% originated from the residential system. This indicates that $NAPI_R$ (5.3%) had a higher export fraction via rivers than $NAPI_{AF}$ (4.3%), implying a need to examine the response of riverine P flux to changes in both $NAPI_{AF}$ and $NAPI_R$. The remaining ~96% of $NAPI_{AF}$ and ~95% of $NAPI_R$ not accounted for by riverine export is believed to be retained in agricultural/forest and residential landscapes or removed via waste treatment.

Our analysis suggests that the upper 20-cm layer of agricultural soils, which retained 92 kg P ha^{-1} and 69% of cumulative $NAPI_{AF}$ over the past 32 years, is the dominant sink for surplus $NAPI_{AF}$ (Fig. 5b). The observed increase in Olsen-P was 3.33 mg P kg^{-1} for each 100 kg P ha^{-1} applied as chemical and manure P amendments (Fig. 5a). These finding are consistent with those from long-term (1985–2000) field studies in seven typical farmlands across China that found increases in Olsen-P levels of 1.44–5.74 mg P kg^{-1} for each 100 kg ha^{-1} of P addition (Cao et al., 2012). The P accumulation observed in the upper 20-cm layer of agricultural soils (Fig. 5b) implies that deeper layers of agricultural soils also have the potential to retain additional P, suggesting that >23.6 kg P ha^{-1} yr^{-1} or >69% of the total $NAPI_{AF}$ was retained in agricultural soils. Similar results have been observed in previous studies in China where agricultural lands retained 10–44.7 kg P ha^{-1} yr^{-1} (35–82% of P addition) in the 1984–2008 period (Shen et al., 2005; Li et al., 2010; Ma et al., 2013).

In terms of $NAPI_R$, a major sink for P is removal of domestic solid waste and sewage from residential areas (Russell et al., 2008; Han et al., 2011b, 2013; Chen et al., 2015a). For the upper Jiaojiang watershed, there has been partial collection and transport of domestic wastes to landfills, incinerators, biogas digesters, septic-tanks or wastewater treatment plants (Table 1). Of cumulative $NAPI_R$ over the past 32 years, 38% and 62% was added to the urban and rural residential systems via food, feed, and detergent inputs, respectively. If we assume that $NAPI_R$ is comprised of 70% solid wastes and 30% liquid wastes as determined by the local agricultural bureau, $\sim$58 kg P ha^{-1} ($\sim$50% of total $NAPI_R$) was removed or buried via waste treatment over the past 32 year. This estimate is consistent with previous material flow analyses conducted in China where 38–48% of anthropogenic P inputs to residential system was removed or stored via landfill or incineration between 1985 and 2010 (Yuan et al., 2014; Cui et al., 2015; Li et al., 2015).

Phosphorus storage in wetlands, residential soils, and river sediments is a likely sink for the remaining 27% of total $NAPI_{AF}$ and 45% of total $NAPI_R$. There is a considerable percentage (18%) of barren land (i.e., sum of water surface, wetlands, rock, and natural vegetation land, Table 1) in the upper Jiaojiang watershed. Natural wetlands and riparian buffers are believed to retain a considerable proportion of the remaining $NAPI_{AF}$ and $NAPI_R$ (Yuan et al., 2014; Cui et al., 2015). In previous studies conducted in nearby regions, 19–26% of anthropogenic P inputs were retained in residential soils (Li et al., 2010, 2015; Yuan et al., 2014), which might be an important sink for the remaining $NAPI_R$. A field investigation conducted in the Nanjiang area showed that soils in urban greenbelts, roadways, parks, residential districts, and campuses have been enriched in P compared to agricultural soil (Yuan et al., 2007). In addition, groundwater might be an additional sink of transiently stored P in residential areas (Roy and Bickerton, 2014; Fitzgerald et al., 2015). Phosphorus sorption in bottom sediments of ditches, streams and rivers may be another important P sink, especially when water velocity is slow and the sediment P sorption capacity is not saturated (Sharpley et al., 2013; Jarvie et al., 2013; Chen et al., 2015b).

3.6. Forecasting future riverine phosphorus exports

To predict future riverine TP exports for 2030, we forecasted future trends for $NAPI_{AF}$, $NAPI_R$, precipitation, agricultural land area percentage with improved drainage (*AD*%), and developed land area percentage (*D*%) using five scenarios compared to the baseline conditions in 2011. The "developing" scenario projects a 10% increase in both $NAPI_{AF}$ and $NAPI_R$ (assuming a 0.5% increase of NAPI per year to support growing population and animal breeding) and a 20% increase in both *AD*% and *D*% (according to the annual average increasing percentages of *AD*% and *D*% observed in 2001–2011) by 2030 with no change in precipitation. Using the calibrated model Eq. (7), the "developing" scenario predicts an increase in riverine TP flux of 140% in 2030 (Fig. 6b). This scenario roughly characterizes the upper bound for future riverine P exports in response to anthropogenic activities, providing a baseline for assessing and adopting relevant watershed P management strategies.

The "climate change" scenario projects a 4% increase in precipitation by 2030 [i.e., $\sim$0.2% increase per year predicted by PGZP (2010)] with no change in $NAPI_{AF}$, $NAPI_R$, or land-use. This scenario predicted a 5% increase in future riverine TP flux by 2030 relative to baseline conditions (Fig. 6b). To demonstrate the influence of *AD*% and *D*% on riverine TP export, we used a "land-use change" scenario that projects a 20% increase in both *AD*% and *D*% according to the annual average percentage increase from 2001 to 2011 with no changes in $NAPI_{AF}$, $NAPI_R$, or precipitation. Changing land-use resulted in a 40% increase in riverine TP fluxes by 2030. These forecasts suggest that the influence of climate and land-use changes on riverine TP exports should be considered as part of management efforts to control P pollution. Given the enhanced P delivery efficiency by climate and land-use changes, it is advantageous to adopt and improve P interception measures along the hydrologic flowpath using constructed ecological ditches, flow-through wetlands, riparian buffer strips, and P sorbing materials (e.g., fly ash and steel slag) (Meals et al., 2010; Sharpley et al., 2013; Li et al., 2015) to mitigate P export from agricultural/forest and residential lands.

For the upper Jiaojiang watershed, average P fertilizer (chemical + manure) use efficiency is low and ranged from 20% to 31%, consistent with values from nearby regions (11–30%, Chen et al., 2008; Li et al., 2010). Therefore, the "the fist reduction" scenario utilizes a $\sim$30% reduction of chemical P fertilizer application (i.e., a 20% reduction in $NAPI_{AF}$), which would have no significant impact on crop yields. This scenario is predicted to decrease riverine TP export by 12% in 2030 (Fig. 6b). The "the second reduction" scenario utilizes a 20% reduction in $NAPI_R$ with no change in $NAPI_{AF}$, precipitation or land-use, based on an assumption that organic wastes applied as manure to agricultural land would increase from 20% in 2011 to 70% in 2030 to replace a corresponding P amount from chemical fertilizer application. This scenario is predicted to decrease riverine TP export by 51% in 2030. Therefore, the reduction of $NAPI_R$ would have greater impact on reducing future riverine TP fluxes than the reduction of $NAPI_{AF}$. To reduce $NAPI_R$, it would be advantageous to promote recycling of human and domestic animal solid wastes to replace chemical fertilizer use on croplands. Since P in wastewater effluent is generally in soluble and biologically-available forms (Sobota et al., 2011; Chen et al., 2015b), targeting human and domestic animal sewage (treated sludge can be used for fertilizing croplands) serves a potential role in reducing eutrophication and hypoxia.

4. Conclusion

This study highlights the importance of NAPI magnitude and components ($NAPI_{AF}$ vs. $NAPI_R$), land-use change, climate change, and legacy P sources in controlling long-term changes of riverine TP export. The model developed in this study incorporates these influencing factors and provides a simple and effective method for predicting TP fluxes derived from agricultural/forest and residential systems. For the upper Jiaojiang watershed, P loss via erosion and leaching is the dominant pathway for P delivery from landscapes to rivers. For annual riverine TP flux in 1980–2011, $NAPI_{AF}$ and $NAPI_R$ contributed 19–56% and 16–67%, respectively, while legacy P sources contributed 13–32%. There was a higher riverine export fraction for $NAPI_R$ (5.3%) than $NAPI_{AF}$ (4.3%), suggesting a need to examine the response of riverine P flux to changes in both $NAPI_{AF}$ and $NAPI_R$. The model predicted a 140%, 5%, and 40% increase in TP flux in 2030 compared to 2011 baseline conditions under the developing, climate change, and land-use change scenarios, respectively. In contrast, 12% and 51% reductions in riverine TP fluxes were predicted when NAP_{AF} and $NAPI_R$ declined by 20%, respectively. Changes in anthropogenic P input components, land-use, and precipitation patterns as well as legacy P source can amplify P export from watershed landscapes to rivers and should be considered in developing P management strategies.

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A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.jhydrol.2015.11.043.

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