

# Influence of legacy phosphorus, land use, and climate change on anthropogenic phosphorus inputs and riverine export dynamics

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**Abstract** A quantitative understanding of riverine phosphorus (P) export in response to changes in anthropogenic P inputs (NAPI), land use and climate is critical for developing effective watershed P control measures. This study indicated that annual riverine TP export for the six catchments of the Yongan River watershed in eastern China increased 4.1–30.3-fold over the 1980–2010 period. Increased riverine TP export resulted from a 61–85 % increase in NAPI and a 2.6–14.6-fold increase in riverine export fraction of NAPI due to 36–43, 30–125,

and 65–76 % increases in developed land area (D%), drained agricultural land area (DA%), and storm events, respectively. For the 31-year cumulative record, 1.6–14 % of NAPI was exported by rivers, 40–64 % was stored in the upper 20 cm of agricultural soils, and 30–55 % was retained in other landscape positions. An empirical model that incorporates annual NAPI, precipitation, D%, and DA% accounted for 94 % of the variation in annual riverine TP fluxes across the six catchments and 31 years. The model estimated that NAPI and legacy P contributed 42–92 % and 8–58 % of annual riverine TP flux, respectively. The model forecasts an 8–18 % increase in riverine TP flux by 2030 due to a 4 % increase in precipitation with no changes in NAPI and land use compared to the 2000–2010 baseline condition. Enhanced export of NAPI and legacy P by changes in land use and climate will delay the decrease in riverine P flux in response to NAPI reductions and should be considered in developing and assessing watershed P management strategies.

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## Introduction

Excess riverine phosphorus (P) from anthropogenic sources is a critical concern worldwide given its role in

eutrophication and subsequent potential for hypoxia, loss of aquatic biodiversity and harmful algal blooms (Han et al. 2011a; Roy and Bickerton 2014; Morse and Wollheim 2014). Due to differences in human activities, hydroclimate, land use/land cover, geology, and geomorphology, substantial spatio-temporal variability in anthropogenic P inputs and riverine P exports exists across regions and years (Némery et al. 2005; Russell et al. 2008; Han et al. 2011a; Kleinman et al. 2011; Hong et al. 2012; Bowes et al. 2014). To effectively guide P management for protecting water quality, it is important to quantify factors regulating anthropogenic P inputs and riverine export dynamics.

Net anthropogenic P input (NAPI) sums P contributions from atmospheric deposition, fertilizer application, non-food input, and net import/export of P in food, feed and seed to a watershed or region and provides a quantitative measure to assess anthropogenic impacts on P cycling (Russell et al. 2008; Han et al. 2011a; Hong et al. 2012; Han et al. 2013). Many studies indicate that among-catchment or among-year variation of riverine P flux can be largely explained by NAPI (Borbor-Cordova et al. 2006; Russell et al. 2008; Han et al. 2011a; Hong et al. 2012). Although the relationship between NAPI and riverine P export is generally strong, such a relationship is additionally influenced by variations in hydroclimate and land use. For example, it is commonly observed that years with higher precipitation or discharge result in a higher export fraction of multi-year averaged NAPI by rivers, whereas drier years result in a lower export fraction (Borbor-Cordova et al. 2006; Russell et al. 2008; Han et al. 2011a; Sobota et al. 2011; Hong et al. 2012). Furthermore, fractional export of multi-year averaged NAPI by rivers varies with land use, with higher fractional export generally observed in catchments having greater urban area (McMahon and Woodside 1997; Sobota et al. 2011; Roy and Bickerton 2014), as well as in catchments having more agricultural area with tile drainage (Gentry et al. 2007; Han et al. 2011a; Sharpley et al. 2013; Morse and Wollheim 2014). Although the export fraction is known to vary with hydroclimate and land use, there is a paucity of long-term studies that simultaneously consider their influences on spatio-temporal NAPI and riverine P export dynamics. Furthermore, the soil is a key interface linking anthropogenic P inputs to riverine P exports, since most P compounds tend to precipitate or adsorb to soil particles before delivery to surface waters

(Kleinman et al. 2011; Roy and Bickerton 2014). However, due to the unavailability of extensive soil characterization data, long-term changes in soil P levels are rarely incorporated in catchment P budgets to verify the assumption that a majority of P inputs are retained in soil.

Previous studies indicated that 1–38 % of NAPI was exported by rivers (David and Gentry 2000; Borbor-Cordova et al. 2006; Russell et al. 2008; Han et al. 2011a, b; Sobota et al. 2011), implying that the majority of NAPI was retained in soils, surface water sediments, and biomass (Sharpley et al. 2013; Jarvie et al. 2014), or even groundwater (Holman et al. 2008; Roy and Bickerton 2014). This accumulated P, referred to as legacy P (i.e., surplus P stored in watershed landscapes that is derived from anthropogenic P inputs in previous years), is not addressed in the NAPI estimation, as well as the majority of current watershed models (Kleinman et al. 2011). Legacy P can be remobilized or recycled, acting as a transient P source to downstream water bodies for years, decades, or even centuries (Zhang et al. 2004; Sharpley et al. 2009; Reddy et al. 2011). The contribution of legacy P has been recognized as a major cause for failure to achieve water quality targets 20–30 years after implementing nutrient management measures for the Chesapeake Bay watershed, Mississippi River basin, Florida's inland and coastal waters, and Lake Erie basin (Meals et al. 2010; Kleinman et al. 2011; Jarvie et al. 2013; Sharpley et al. 2013; Condon et al. 2013; Haygarth et al. 2014). Legacy P release and delivery to rivers is highly dependent on landscape P saturation (Kleinman et al. 2011; Jarvie et al. 2014), hydrological connectivity (Gentry et al. 2007; Han et al. 2011a), and storm runoff/erosion (Borbor-Cordova et al. 2006; Hong et al. 2012). Therefore, from the perspective of changes in climate and land management, legacy P release may be enhanced and subsequently has the potential to make a considerable contribution to the riverine P flux. However, limited knowledge is available concerning what proportion of riverine P export in a given year is derived from legacy P sources (Jarvie et al. 2013; Haygarth et al. 2014). Such quantitative information is critical for optimizing watershed P pollution control expenditures, strategies and schedules (Sharpley et al. 2013), as well as to set realistic expectations for the public (Kleinman et al. 2011).

Based on an extensive 31-year data record (1980–2010) for six catchments of the Yongan River

watershed in eastern China, this study demonstrates the integrated influence of legacy P, land use and climate change on spatio-temporal NAPI and riverine TP export dynamics. This study advances our understanding of catchment P budgets and riverine TP export dynamics by (i) examining the response of riverine TP flux and soil P levels to catchment NAPI, (ii) addressing the influence of hydroclimate and land use on riverine export fraction of NAPI, (iii) developing an empirical model that incorporates NAPI, land use, and hydroclimate for predicting riverine TP fluxes, (iv) identifying individual contributions from NAPI and legacy P sources to annual riverine TP flux, and (v) forecasting trends in P export based on scenarios for future (2011–2030) changes in human activities and climate. In so doing, this study provides a methodology for quantifying the contribution of legacy P to riverine TP flux and improves our understanding of long-term P fate and transport for informing watershed P management strategies.

## Materials and methods

### Watershed description

The Yongan watershed (120.2295°–121.0146°E and 28.4695°–29.0395°N; elevation ~15–1,000 m) is located in the highly developed Taizhou region of Zhejiang Province, China (Fig. 1). The Yongan River flows into Taizhou Estuary and the East China Sea, a coastal area that commonly experiences hypoxia. The river drains 2,474 km<sup>2</sup> and has an average water depth of 5.42 m and discharge of 72.9 m<sup>3</sup> s<sup>-1</sup> at the downstream BZA sampling location (Fig. 1). There are no major dams or water withdrawals in the river. Agricultural land (e.g., paddy field, garden plot and dry land) averaged ~12 % of total watershed area, with developed land (e.g., rural and urban residential, roads, mining and industrial), woodland, and natural lands contributing ~3, ~67, and ~18 %, respectively (supplementary material A, Fig. S1). Among the six catchments examined in this study, HG and ZK had the highest and lowest percentages of developed land (i.e., the sum of all rural and urban residential areas, roads, mining and industrial areas) and agricultural land (i.e., the sum of all paddy field, garden plots, and dry land), respectively (Table 1). The climate is subtropical monsoon having an average annual

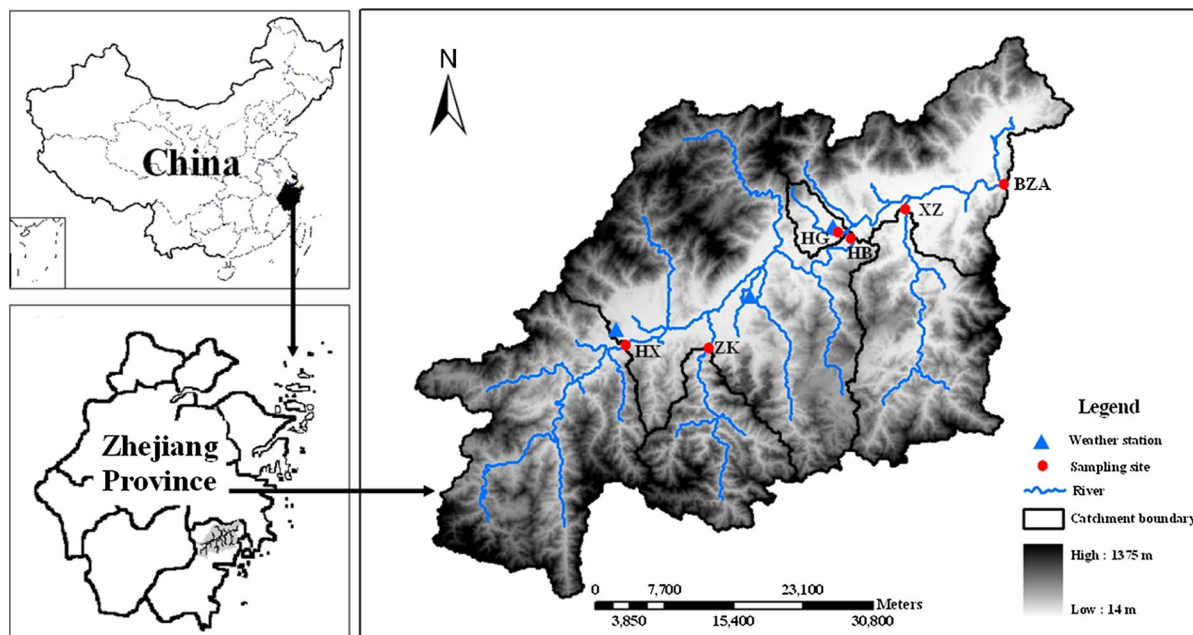
temperature of 17.4 °C and average annual precipitation of 1,400 mm (supplementary material A, Fig. S2). Rainfall mainly occurs in May–September with a typhoon season in July–September. Over the 1980–2010 period, the watershed experienced a ~22 day decrease in the number of rainy days and a 65–76 % increase in storm events (>50 mm per 24 h). Over the 31-year study period, human population within the watershed increased by ~25 %. Catchments HG and ZK have the highest and lowest human and domestic animal densities, respectively (Table 1). Since 2000, there was a remarkable reduction in cropland cultivation (38–50 %) as well as in chemical P fertilizer application (27–55 %). Due to recent replacement of the old agricultural irrigation and drainage system (constructed with stone and mud in the 1950s), the agricultural land area irrigated and drained with cement channels and pipes has increased by 150–310 % since 2000 (supplementary material C, Fig. S6).

### Riverine TP flux estimate

Water samples were collected once every 4–8 weeks at six sampling sites (Fig. 1) during the 1980–2010 study period (n = 151–183 sampling times). Data for river TP concentration (measured in non-filtered water samples following persulfate digestion using the ammonium molybdate spectrophotometric method) and daily discharge were obtained from the local Environmental Protection Bureau and Hydrology Bureau, respectively. To estimate annual TP flux based on the discrete TP concentrations for each sampling site, the LOADEST model (Runkel et al. 2004) was applied for predicting daily TP flux, resulting in high  $R^2$  values ( $R^2 = 0.72$ – $0.98$ ,  $p < 0.001$ ) and low average relative errors ( $\pm 4$  to  $\pm 15$  %) between measured and estimated values in each catchment (supplementary material B).

### Agricultural soil phosphorus measurement

The Yongan watershed is dominated by highly weathered and acidic soils (Oxisols (65 %) and Ultisols (15 %); mean pH < 5.5). Weathering inputs of P from these highly weathered soils were expected to be negligible relative to anthropogenic inputs. Soil samples (one composite sample per 15 ha for plain regions and one composite sample per 25 ha for hilly



**Fig. 1** The location the Yongan River watershed in China and Zhejiang Province and the six water quality sampling sites and three weather stations

regions) were collected from the upper 20-cm layer of agricultural lands (i.e., paddy field, dry land and garden plots) in the six catchments by the local Agriculture Bureau in 1984 and 2009. Olsen-extractable P (Olsen-P), total P (organic+inorganic P, measured by  $\text{H}_2\text{SO}_4\text{-HClO}_4$  degradation-molybdenum antimony colorimetric method), pH, and bulk density were measured in 1984 and 2009, resulting in time-paired Olsen-P ( $n = 36\text{--}126$ ) and total P ( $n = 12\text{--}95$ ) contents for each of the six catchments. 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 (supplementary material C).

#### NAPI estimation and uncertainty analysis

To examine spatio-temporal variation for the anthropogenic P budget, annual net anthropogenic P inputs (NAPI,  $\text{kg P ha}^{-1} \text{ year}^{-1}$ ) in each of the six catchments for the 1980–2010 study period were estimated as the sum of four major components: atmospheric P deposition (AD), commercial fertilizer P application (CF), net food, feed and seed P input (NFFSI), and detergent P or non-food P input (NFI) (Russell et al.

2008; Han et al. 2011b, 2013). Wastewater discharge was not considered explicitly in the NAPI analysis, since P in wastewater originates from food, feed and detergents (Hong et al. 2012). The net food, feed and seed P input was calculated as the sum of human and livestock P consumption and seed input, minus the sum of P in livestock and crop production. To gain insight into the uncertainty of NAPI estimates, an uncertainty analysis was performed using Monte Carlo simulation (supplementary material D). To examine temporal trends in the riverine export fraction of NAPI, we estimated the ratio between 5-year average riverine TP flux and NAPI for each catchment (i.e., riverine export fraction of 5-year average NAPI), as commonly applied in watershed studies (Russell et al. 2008; Han et al. 2011a) to account for a potential time lag between P input to watersheds and export by rivers.

#### Developing models for riverine TP export

To address the quantitative response of riverine TP flux ( $F$ ,  $\text{kg P ha}^{-1} \text{ year}^{-1}$ ) to changes in NAPI, hydroclimate, and land use, this study developed the following empirical model:

**Table 1** Characteristics of land-use distribution, population, domestic animals, and hydroclimate for the six catchments of the Yongan River watershed over the 1980–2010 period

Catchments	Periods	A (%)	D (%)	F (%)	N (%)	CA	P	D	PD	AD	DA (%)
HX (547 km <sup>2</sup> )	1980s	7	1	71	21	97.6	1,278	16.0	122	74	12
	1990s	7	1	71	21	92.0	1,331	17.1	130	70	10
	2000s	9	2	70	20	76.2	1,313	15.6	148	53	20
HB (1,650 km <sup>2</sup> )	1980s	10	3	66	18	382.0	1,337	48.2	194	96	16
	1990s	10	3	66	18	361.7	1,392	51.7	211	76	16
	2000s	11	4	65	17	291.4	1,396	47.1	228	89	25
BZA (2,474 km <sup>2</sup> )	1980s	11	2	68	18	636.6	1,365	72.2	248	107	19
	1990s	11	3	68	18	600.9	1,424	77.5	266	80	18
	2000s	13	3	67	17	469.5	1,424	70.7	288	99	29
ZK (218 km <sup>2</sup> )	1980s	3	1	75	21	19.5	1,388	6.4	68	42	10
	1990s	3	1	75	21	17.5	1,438	6.8	74	31	9
	2000s	3	1	76	20	15.8	1,483	6.2	79	35	16
HG (35 km <sup>2</sup> )	1980s	18	7	48	28	13.9	1,420	1.0	704	166	30
	1990s	17	7	49	27	13.3	1,488	1.1	744	159	27
	2000s	21	9	49	21	8.6	1,480	1.0	828	136	47
XZ (357 km <sup>2</sup> )	1980s	9	2	75	24	80.9	1,420	10.4	198	95	14
	1990s	9	2	75	24	75.1	1,488	11.2	215	64	14
	2000s	11	2	74	23	63.0	1,480	10.2	226	88	19

A (%) agricultural land; D (%) developed land; F (%) forest; N (%) water surface, wetlands, rock, and natural reservation land; CA cultivated crop land area (i.e., annual area planted to crops, km<sup>2</sup>), P precipitation (mm year<sup>-1</sup>), Q river water discharge (m<sup>3</sup> s<sup>-1</sup>), PD population density (capita km<sup>-2</sup>), AD domestic animal density (capita km<sup>-2</sup>, the number of each type of animal is converted into the equivalent number of pigs according to their P excretion rates as shown in Table S4), DA (%) percentage of agricultural lands with improved drainage. All values are the average for each time period. BZA denotes the entire watershed

$$F = a \prod_{i=1}^n x_i^{b_i} \exp(cNAPI), \quad (1)$$

where  $x_i$  is the factors (e.g., annual precipitation, number of storm events, developed land area percentage, and drained agricultural land area percentage) influencing P delivery from landscapes to rivers, and  $a$ ,  $b_i$ , and  $c$  are unknown parameters.

To determine the most efficient influencing factors for the model, the potential influencing factors mentioned above were added one by one for  $x_i$  for calibrating unknown parameters. The best set of influencing factors was determined according to the highest model agreement. The agreement between annual TP fluxes estimated by LOADEST and modeled by Eq. (1) was evaluated using correlation ( $R^2$ ) and Nash–Sutcliffe coefficients (Chen et al. 2014). Regression analysis was applied to calibrate the unknown parameters ( $a$ ,  $b_i$ , and  $c$ ) after a logarithmic transformation of Eq. (1). In this study, regression

analysis and analysis of variance (ANOVA) were performed using SPSS statistical software (SPSS version 16.0, SPSS Inc., USA, 2002).

#### Riverine TP source apportionment method

Model Eq. (1) developed between year-to-year NAPI and riverine TP export implies a contribution of legacy P that was derived from NAPI in previous years and accumulated in various landscape positions (e.g., soils, river sediments and biomass). This study assumed that the component of riverine TP export that is not explained by NAPI in Eq. (1) represents the contribution of legacy P sources ( $F_{Legacy}$ ) if the contributed P flux from natural rock weathering is negligible. Therefore,  $F_{Legacy}$  was calculated by Eq. (1) when NAPI was set equal to zero:

$$F_{Legacy} = a \prod_{i=1}^n x_i^{b_i} \quad (2)$$

Then, riverine TP export derived from NAPI in the current year ( $F_{NAPI}$ ) can be estimated as:

$$F_{NAPI} = a \prod_{i=1}^n x_i^{b_i} [\exp(cNAPI) - 1] \quad (3)$$

## Results

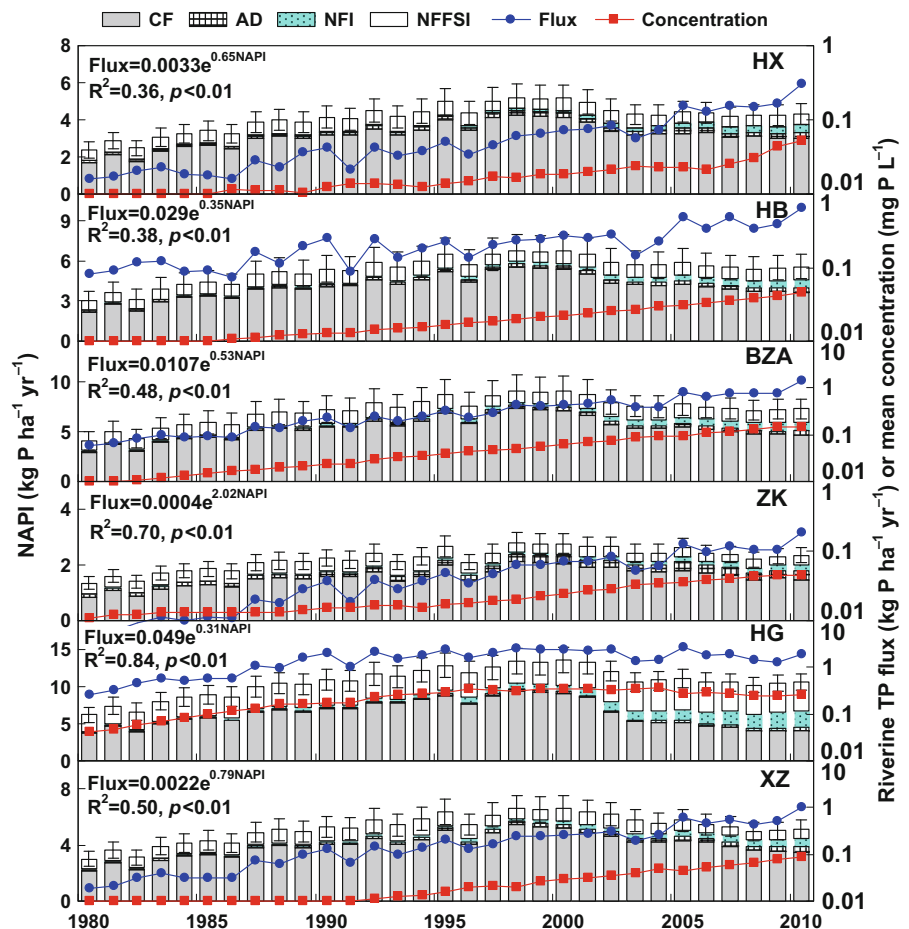
### Changes in NAPI and riverine TP export

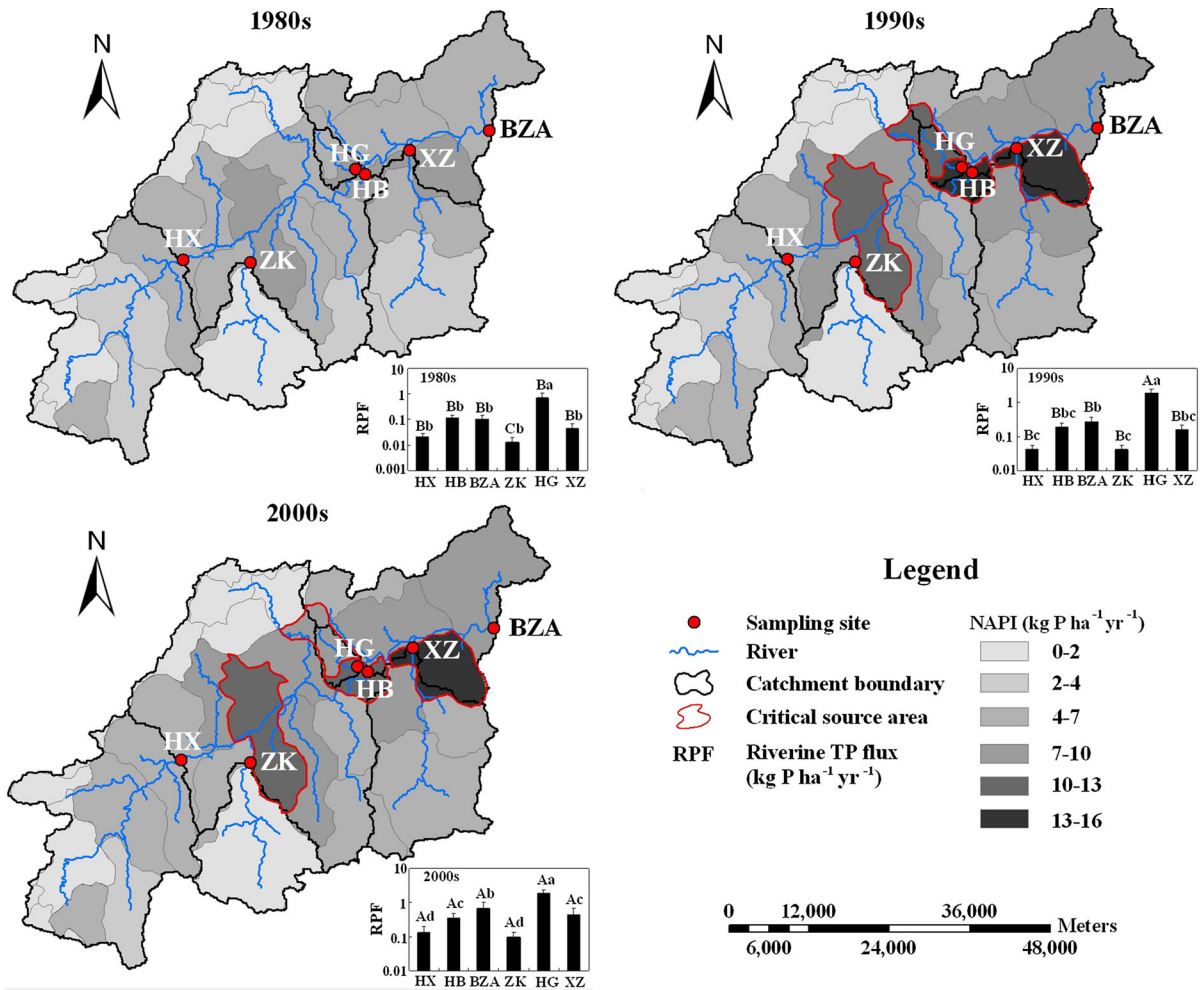
For the six catchments of the Yongan watershed, NAPI increased from 1.35–6.27 kg P ha<sup>-1</sup> year<sup>-1</sup> (watershed average = 4.06 kg P ha<sup>-1</sup> year<sup>-1</sup>) in 1980 to 2.76–13.6 kg P ha<sup>-1</sup> year<sup>-1</sup> (watershed average = 9.01 kg P ha<sup>-1</sup> year<sup>-1</sup>) in 2000 before declining to 2.34–10.7 kg P ha<sup>-1</sup> year<sup>-1</sup> (watershed average = 7.24 kg P ha<sup>-1</sup> year<sup>-1</sup>) in 2010, resulting in a net increase of 61–85 % over the 31-year study period (Fig. 2). Considering the high forest and natural

land percentage in this watershed (>70 %, Table 1), estimated NAPI for the entire watershed was comparable with the average NAPI estimated for Zhejiang Province, China (7.14 kg P ha<sup>-1</sup> year<sup>-1</sup> in 1981 to 9.72 kg P ha<sup>-1</sup> year<sup>-1</sup> in 2009; Han et al. 2013). Among the six catchments, ZK with greater than 96 % natural land cover (Table 1) had the lowest 31-year average NAPI of 2.22 kg P ha<sup>-1</sup> year<sup>-1</sup> (Fig. 3). Catchment HG with greater than 17 % agricultural land and 7 % developed land had the largest average NAPI of 10.7 kg P ha<sup>-1</sup> year<sup>-1</sup>. For the entire watershed, the region near the HB, HG and XZ sampling sites (delineated with red lines in Fig. 3) has the highest animal (>150 pig km<sup>-2</sup>) and human (>400 people km<sup>-2</sup>) densities and occupies only 11 % of the watershed area, but received 23 % of the total NAPI in the 1990s and 20 % of total NAPI in the 2000s.

Fertilizer application was the largest individual source of P (Fig. 2) accounting for 38–76 % of NAPI

**Fig. 2** Historical trends of NAPI from chemical fertilizer (CF), atmospheric deposition (AD), net food, feed and seed P input (NFFSI), and non-food input (NFI); and annual riverine TP flux and mean TP concentration in the six catchments of the Yongan River watershed over the 1980–2010 period. Error bars denote the 95 % confidence interval of NAPI. BZA denotes the entire watershed





**Fig. 3** Spatial distribution of average NAPI and riverine TP flux in the 1980s, 1990s, and 2000s in the Yongan River watershed. Capital and small letters within riverine TP flux

histograms denote significant differences ( $p < 0.05$ ) among periods and catchments, respectively. BZA denotes the entire watershed

in each of the six catchments over the 31-year study period; however, it decreased to 27–55 % since 2000 due to a decrease (38–50 %) in cropland cultivation (Table 1). Although atmospheric P deposition only accounted for 2–8 % of NAPI (5.4–6.0 kg P ha<sup>-1</sup>), its contribution continuously increased from 1980 to 2010. Similarly, net food, feed and seed import and non-food input increased by 31–74 % and 18–19-fold from 1980 to 2010, respectively. Catchment HG with the greatest animal (>100 pig km<sup>-2</sup>) and human (>300 people km<sup>-2</sup>) densities had the largest average net food, feed and seed P import and non-food P input, which in total accounted for 39 % of NAPI on average. Catchment ZK with the lowest animal (<50 pig km<sup>-2</sup>)

and human (<100 people km<sup>-2</sup>) densities had the lowest average net food, feed and seed import and non-food input.

Annual riverine TP exports at the HX, HB, BZA, ZK and XZ catchment outlets steadily increased by 4.1- to 30.3-fold over the 31-year study (Fig. 2). Riverine fluxes ranged from 0.01–0.11 kg P ha<sup>-1</sup> year<sup>-1</sup> (0.01–0.02 mg P L<sup>-1</sup>) in the 1980s, to 0.04–0.27 kg P ha<sup>-1</sup> year<sup>-1</sup> (0.02–0.04 mg P L<sup>-1</sup>) in the 1990s, and to 0.10–0.67 kg P ha<sup>-1</sup> year<sup>-1</sup> (0.03–0.11 mg P L<sup>-1</sup>) in the 2000s (Fig. 3). For catchment HG, annual riverine TP exports rapidly increased from 0.25 kg P ha<sup>-1</sup> year<sup>-1</sup> (0.04 mg P L<sup>-1</sup>) in 1980 to 2.44 kg P ha<sup>-1</sup> year<sup>-1</sup> (0.34 mg P L<sup>-1</sup>) in 2000, while there was

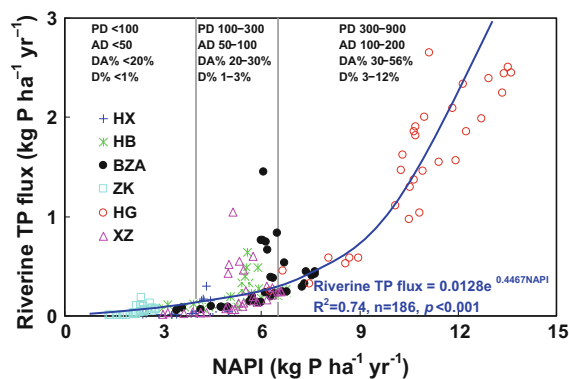
a decrease to  $1.91 \text{ kg P ha}^{-1} \text{ year}^{-1}$  ( $0.26 \text{ mg P L}^{-1}$ ) between 2000 and 2010, resulting in a net 6.5-fold increase over the 31-year study (Fig. 2). Spatially, catchment HG presented the highest TP export, while catchment ZK had the lowest TP export (Fig. 3).

### Response of riverine TP export to NAPI

For the six catchments of the Yongan watershed, NAPI accounted for 36–84 % of the temporal variability in annual riverine TP exports over the 31-year study (Fig. 2). For catchments HX, HB, BZA, ZK and XZ, despite a 31–41 % increase of NAPI, riverine TP flux/concentration did not increase between 1980 and 1986. Beginning in 1987, riverine TP flux/concentration increased through 2010 in spite of a 15–26 % decline in NAPI between 2000 and 2010. Such contrasting trends observed between NAPI and riverine TP fluxes (Fig. 2) over the 2000–2010 period implies a contribution from legacy P and/or increased fractional delivery of NAPI due to changes in hydro-climate, land use and/or watershed P saturation. Catchment HG was an exception, where both NAPI and TP export displayed decreasing trends over the 2000–2010 period, resulting in the highest correlation between NAPI and TP export across the 31-year record ( $R^2 = 0.84$ ,  $p < 0.01$ ). Averaging the 31-year records for all six catchments resulted in a strong spatial relationship between NAPI and riverine TP export:  $\text{Riverine TP flux} = 0.0214\text{exp}(0.4147\text{NAPI})$ ,  $R^2 = 0.92$ ,  $p < 0.01$ .

When the 31-year records for all six catchments were pooled, spatial and temporal riverine TP export was exponentially related to NAPI (Fig. 4). The exponential regression, explaining 74 % of spatio-temporal variation in riverine TP exports, indicates a disproportionate response of riverine TP export to increasing NAPI. The ratio between average riverine TP flux and NAPI for the categorical groups with NAPI  $< 4.00 \text{ kg P ha}^{-1} \text{ year}^{-1}$ ,  $4.00\text{--}6.50 \text{ kg P ha}^{-1} \text{ year}^{-1}$ , and  $> 6.50 \text{ kg P ha}^{-1} \text{ year}^{-1}$  were 1.7, 3.2, and 8.4 %, respectively (Fig. 4). Over the past 31 years, cumulative riverine TP export accounted for 1.6–14.2 % of total NAPI, with the highest and lowest export fractions occurring in catchments HB and ZK, respectively.

In terms of a long-term trend, estimated riverine export fractions of 5-year average NAPI for the six catchments rapidly increased from 0.7–5.8 % in



**Fig. 4** Spatio-temporal NAPI and riverine TP flux dynamics in the six catchments of the Yongan River watershed over the 1980–2010 period. PD, population density (people  $\text{km}^{-2}$ ); AD, domestic animal density (pig  $\text{km}^{-2}$ , the number of each type of animal is converted into the equivalent number of pigs according to their P excretion rates as shown in Table S4); AD%, drained agricultural land area percentage; and D%, developed land percentage. The breakpoints for NAPI were identified by changing ranges in PD, AD, DA% and D%. BZA denotes the entire watershed

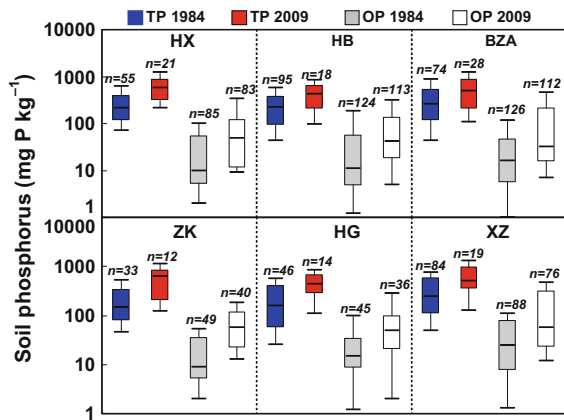
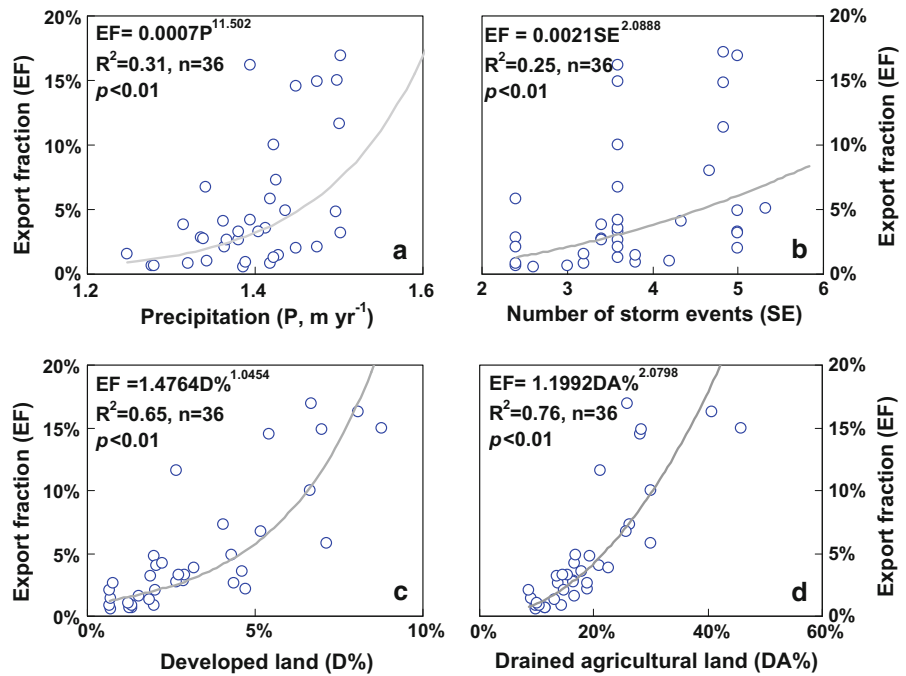
1980–1984 to 2.8–15 % in 2000–2010, resulting in a 2.6–14.6-fold increase over the 31-year study period. When all riverine export fractions of 5-year average NAPI for the six catchments were pooled, it was nonlinearly, positively and significantly related to average precipitation, number of storm events ( $> 50 \text{ mm per 24 h}$ ), developed land area percentage, and drained agricultural land area percentage (Fig. 5,  $p < 0.01$ ). The 5-year average export fraction had greater correlations with percentages of drained agricultural land and developed land than with precipitation and storm events (Fig. 5).

### Response of agricultural soil phosphorus levels to NAPI

For the six catchments, Olsen-P levels in the upper 20-cm layer of agricultural soils significantly increased from 8.8–24.2 to 32.0–57.2  $\text{mg P kg}^{-1}$  between 1984 and 2009, while TP levels increased from 144–244 to 419–629  $\text{mg P kg}^{-1}$  (Fig. 6). Although no site-specific data were available for the Yongan watershed, an investigation conducted for Zhejiang province indicated that  $\sim 50 \%$  of animal wastes were applied as manure to agricultural lands in 2010 (Mo et al. 2011). This implies a considerable proportion of net food and feed P was transported to



**Fig. 5** Response of riverine export fraction of 5-year averaged NAPI to changes in hydroclimate and land use for the six catchments of the Yongan River watershed in 1980–2010



**Fig. 6** Changes in total phosphorus (TP) and Olsen phosphorus (OP) levels in the upper 20-cm layer of agricultural soil between 1984 and 2009 in the six catchments of the Yongan River watershed. Measured TP or OP levels between 1984 and 2009 in each catchment were significant difference ( $p < 0.01$ ). Box plot denotes 97.5, 75, 50, 25, and 2.5 % confidence interval of soil P levels. BZA denotes the remaining watershed area independent of the other five catchment areas

agricultural lands. Therefore, we believe that agricultural soils received more than 61–81 % of cumulative NAPI in the six catchments over the past 31 years, resulting in the significantly elevated soil P levels. Increases in soil TP and Olsen-P contents resulting

from each 100 kg P ha<sup>-1</sup> of agricultural applied NAPI (i.e., fertilizer + seed input + atmospheric deposition—crop production) were estimated to be 25.5–44.8 and 2.4–4.4 mg P kg<sup>-1</sup> for the six catchments, respectively. Based on the changes in TP level, an estimated 17.7–43.7 kg P ha<sup>-1</sup> year<sup>-1</sup> was accumulated in the upper 20-cm layer of agricultural soils, accounting for 40–64 % of total NAPI applied to each of the six catchments during the past 31 years (Table 2).

#### Models for riverine TP export

The analyses reported above indicate that the relationship between riverine TP export and NAPI was best described using an exponential function (Figs. 2 and 4), while the relationships between riverine export fraction of NAPI and precipitation, number of storm events, developed land area and drained agricultural land area were best described using a power function (Fig. 5). These results suggested that the adopted power and exponent functions in Eq. (1) are appropriate for characterizing the relationship of riverine TP fluxes with NAPI and the other influencing factors.

The calibrated results for models incorporating various influencing factors indicated that the

**Table 2** Cumulative phosphorus sources and sinks over the 1980–2010 period for the six catchments of the Yongan River watershed (kg P ha<sup>-1</sup>)

Catchments	HX	HB	BZA	ZK	HG	XZ
<i>Sources</i>						
Atmospheric deposition	5.4	5.7	5.8	5.9	6.0	6.0
Chemical fertilizer	97.5	125.8	138.0	48.7	198.5	123.3
Seed input	0.8	0.9	0.9	0.3	1.2	0.9
Non-food input	4.4	6.9	8.7	2.4	25.0	6.9
Net food and feed input	19.3	27.0	33.2	11.6	101.4	24.7
Total	127.4	166.3	186.6	69.0	332.1	161.8
<i>Sinks</i>						
Riverine export	2.1	6.9	11.2	1.6	47.3	6.9
Upper 20 cm of agricultural soils	65.7	70.9	119.9	39.3	143.3	65.4
Other landscape storage	59.6	88.5	55.5	28.1	141.5	89.5
Total	127.4	166.3	186.6	69.0	332.1	161.8

BZA denotes the entire watershed

combined NAPI and any one of annual precipitation ( $P$ , m year<sup>-1</sup>), number of storm events ( $SE$ ), developed land area percentage ( $D\%$ ) and drained agricultural land area percentage ( $DA\%$ ) explained annual riverine TP export with reasonably high accuracy ( $R^2 = 0.77$ – $0.86$ ) compared to individual variables alone (supplementary material E, Table S6). As expected, the predictive capability was further improved by combining NAPI,  $D\%$ ,  $DA\%$ , and either  $P$  or  $SE$ , ( $R^2 = 0.84$ – $0.94$ ). Among them, the combination of NAPI, precipitation, developed land area percentage and drained agricultural land area percentage best described spatio-temporal variability of riverine TP export:

$$F = 0.379P^{1.779}DA\%^{1.404}D\%^{0.109}\exp(0.245NAPI) \quad (4)$$

Equation (4) accounted for 94 % of the variation in annual riverine TP flux over the 1980–2010 period for the six catchments (Nash–Sutcliffe coefficient = 0.92 between modeled TP flux and LOADEST-estimated values; Fig. 7a). The mean relative error of prediction accuracy for individual years and catchments was

<10 % and the interquartile range for each year was relatively symmetrical, ranging from  $-28$  to  $+28$  %. Plots of predicted values and residuals indicated that the residual interval was not correlated with the variance of the mean TP flux (Fig. 7b,  $p > 0.05$ ).

#### Source apportionment for riverine TP

Based on Eq. (4), riverine TP fluxes derived from legacy P sources ( $F_{Legacy}$ ) and NAPI in the current year ( $F_{NAPI}$ ) were estimated as:

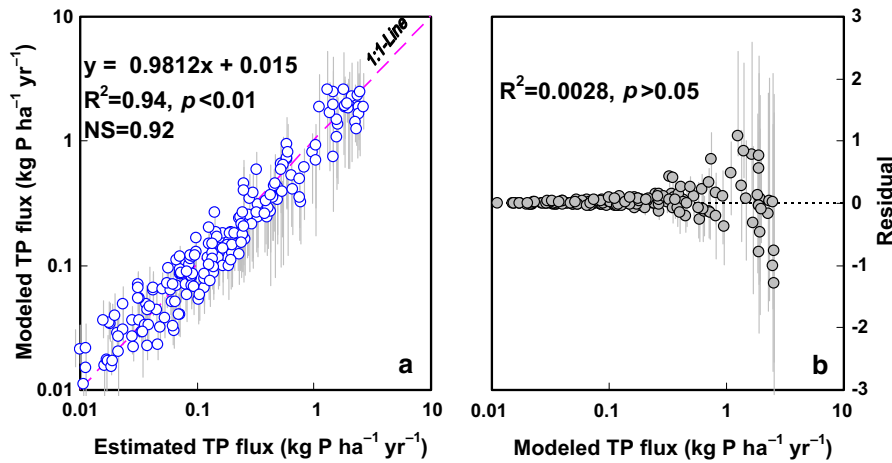
$$F_{Legacy} = 0.379P^{1.779}DA\%^{1.404}D\%^{0.109} \quad (5)$$

$$F_{NAPI} = 0.379P^{1.779}DA\%^{1.404}D\%^{0.109}[\exp(0.245NAPI) - 1] \quad (6)$$

Over the 1980–2010 period, current year's NAPI contributions to annual riverine TP exports followed (kg P ha<sup>-1</sup> year<sup>-1</sup>): HX = 0.043 (63 %), HB = 0.17 (72 %), BZA = 0.28 (76 %), ZK = 0.23 (42 %), HG = 1.43 (92 %) and XZ = 0.16 (71 %) (Fig. 8). Catchment HG with the highest percentages of developed land area and drained agricultural land area had the highest contribution of annual NAPI to riverine TP export. With the exception of catchment HG, annual riverine TP flux derived from current year NAPI presented a significant increasing trend ( $p < 0.001$ ) over time. As expected, the legacy P sources comprised a considerable contribution (8–58 %) to annual riverine TP export from the six catchments over the past 31 years (kg P ha<sup>-1</sup> year<sup>-1</sup>): HX = 0.023 (37 %), HB = 0.058 (28 %), BZA = 0.083 (24 %), ZK = 0.029 (58 %), HG = 0.098 (8 %) and XZ = 0.058 (29 %) (Fig. 8). Catchments ZK and HG presented the highest and lowest contribution of legacy P to riverine TP export, respectively. Annual riverine TP flux derived from legacy P sources displayed a significant increasing trend ( $p < 0.001$ ) with time in each of the six catchments.

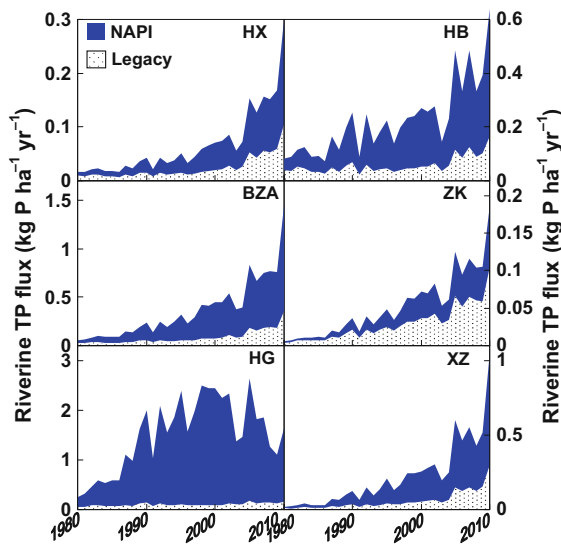
#### Forecasting future riverine TP export

To forecast riverine TP exports for the 2011–2030 period using Eq. (4), we took average NAPI and precipitation in 2000–2010 and the developed land area and drained agricultural land area in 2010 as baseline conditions. We forecasted future trends of



**Fig. 7** LOADEST-estimated versus modeled riverine TP flux using model Eq. (4) that incorporates annual NAPI, precipitation, developed land area and drained agricultural land area (a) and model residual values plotted against the modeled mean

riverine TP flux (b) for the six catchments of the Yongan River watershed in 1980–2010 ( $n = 186$ ). Vertical lines denote the 95 % confidence interval of the fluxes or residuals. NS denotes Nash–Sutcliffe coefficient



**Fig. 8** The contributions of current year NAPI and legacy P sources to observed annual riverine TP export in the six catchments of the Yongan River watershed in 1980–2010. BZA denotes the entire watershed

NAPI, precipitation, developed land area and drained agricultural land area based on four scenarios. The “climate change” scenario projects a 4 % increase of precipitation by 2030 (i.e., 0.2 % increase per year, based on “The Response Program for Climate Change of Zhejiang Province in China”, PGZP 2010) with no change in NAPI or land use. The climate change

scenario predicted an 8–18 % increase of future riverine TP flux by 2030 relative to baseline conditions (Table 3). Catchment ZK presented the greatest response of riverine TP export to increasing precipitation. Under the “developing I” scenario, riverine TP fluxes are expected to increase 94–145 % in 2030 due to 20 % increases in NAPI, drained agricultural land area, and developed land area. Under the “developing II” scenario, riverine TP fluxes are expected to increase 52–95 % in 2030 due to 20 % increases in NAPI and developed land area. Under the “adaptive management” scenario, riverine TP export in 2030 decreased by 8–58 % due to a 20 % reduction of NAPI by 2030 in each catchment. Predicted riverine TP fluxes for catchments HG and ZK presented the highest and lowest response to changes in future NAPI and land use, respectively.

**Discussion**

Long-term fate of anthropogenic P inputs

For cumulative NAPI ( $69.0\text{--}332.1 \text{ kg P ha}^{-1}$ ) in the six catchments over the past 31 years, riverine discharge and retention in landscapes (i.e., soils, biomass and groundwater) are believed to be the two major sinks for P. Of the total NAPI, 1.6–14.2 % was lost via river export across the six catchments

**Table 3** Forecasted changes in riverine TP fluxes for 2030 resulting from changes in precipitation, NAPI, developed land area percentage (D%), and drained agricultural land area percentage (DA%) in the six catchments of the Yongan River watershed

Scenarios	Climate change (%)	Developing I (%)	Developing II (%)	Adaptive management (%)
	+4 % precipitation	0 % precipitation	0 % precipitation	0 % precipitation
	0 % NAPI	+20 % NAPI	+20 % NAPI	-20 % NAPI
	0 % D%	+20 % D%	+20 % D%	0 % D%
	0 % DA%	+20 % DA%	0 % DA%	0 % DA%
HX	15	94	57	-20
HB	8	99	61	-31
BZA	16	95	58	-33
ZK	18	88	52	-8
HG	15	145	95	-58
XZ	13	101	71	-14

BZA denotes the entire watershed

(Table 2). This estimate falls within the range reported for several studies in American, European, and Guayas 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). Higher export fractions occurred in regions dominated by human and animal waste discharge (e.g., 35 % for Illinois, David and Gentry 2000) and intensive soil erosion (e.g., 38 % for Guayas watersheds, Borbor-Cordova et al. 2006).

Our analysis suggests that the upper 20-cm layer of agricultural soils, which retained 40–64 % of cumulative NAPI over the past 31 years, is the dominant P sink (Table 2). Observed increase in Olsen-P for each 100 kg P ha<sup>-1</sup> of agricultural applied NAPI (i.e., 2.4–4.4 mg P kg<sup>-1</sup>, Fig. 6) was consistent with results from long-term (1985–2000) field observations in seven typical farmlands across China that found increases in Olsen-P levels of 1.44–5.74 mg P kg<sup>-1</sup> for each 100 kg ha<sup>-1</sup> P addition (Cao et al. 2012). Oxisols and Ultisols, which are the dominant soils types (90 % of soil area) in the Yongan watershed, have high Fe/Al oxide/hydroxide content with a high capacity for P retention (Shen et al. 2005) and are responsible for the high percentage of NAPI retention in the upper 20 cm of agricultural soils. The significant P accumulation observed in the upper 20-cm layer of agricultural soil (Fig. 6) implies that deeper layers of agricultural soils also have the potential to retain additional P, suggesting that from >17.7 to >43.7 kg P ha<sup>-1</sup> year<sup>-1</sup> or from >40 to >64 % of total NAPI was retained in agricultural soils. This estimate is consistent with previous studies in eastern China where P

retention in agricultural land ranged from 10 to 44.7 kg P ha<sup>-1</sup> year<sup>-1</sup> (35–82 % of NAPI) on average in the 1984–2008 period (Shen et al. 2005; Ma et al. 2013), as well as retention of 16–50 % of NAPI in Wisconsin agricultural soils between 1970 and 1995 (Bundy and Sturgul 2001) and 68 % for Europe between 1965 and 2007 (Sattari et al. 2012).

In addition to the deeper layers of agricultural soils, P storage in wetlands, forest biomass and river sediments is a likely location for accumulation of the remaining 30–55 % of total NAPI (Table 2). Considering the considerable percentage (17–28 %) of natural land (i.e., the sum of water surface, wetlands, rock, and natural reservation land, Table 1), natural wetlands, which can retain 5–20 % of inflowing P load within soils, vegetation and plant litter (Fisher and Acreman 2005), are believed to retain a considerable proportion of the remaining NAPI for the Yongan watershed. In areas with riparian buffers up to 80 % of P inputs can be retained (Russell et al. 2008). Given the high percentage of forests in the six catchments (53–74 %), ~0.5 to ~2 % of NAPI (or ~25 % of atmospheric P deposition) was potentially retained by forest biomass and understory vegetation storage as observed in eastern China (Xu et al. 2002). Although groundwater is not commonly considered an important sink for storing P due to its low mobility through soils, some investigations have found that dissolved P concentrations in groundwater can be elevated and contribute to eutrophication of surface waters (Holman et al. 2008; Roy and Bickerton 2014). Thus, groundwater might be an additional source of transiently stored P in some watersheds. 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 (David and Gentry 2000; Sharpley et al. 2013; Jarvie et al. 2013). Furthermore, due to the unavailability of public records we could not account for the amount of municipal waste and sewage sludge that was removed from residential areas and transported to landfills, incinerators or farmlands, which is a potential fate for a portion of the NAPI (Russell et al. 2008; Han et al. 2011b).

#### Influence of changes in hydroclimate and land use on NAPI and riverine TP export

For the six catchments of the Yongan watershed, changes in hydroclimate are an important driver for temporal NAPI and riverine TP export dynamics over the 1980–2010 period, while changes in land use are believed to be responsible for both temporal and spatial dynamics. This results in greater correlations for the 5-year average export fraction with percentages of drained agricultural land and developed land than with precipitation and storm events (Fig. 5). Particle P loss via erosion, dissolved P leaching (diffuse source pollution) and waste discharge (point source pollution) are believed to be the major pathways for P entering rivers (Némery et al. 2005; Gentry et al. 2007; Sobota et al. 2011). Higher precipitation enhances erosion and leaching and subsequently increases fractional export of NAPI by rivers (Borbor-Cordova et al. 2006; Morse and Wollheim 2014, Fig. 5a). 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; Puustinen et al. 2007). Fractional export of NAPI by rivers is also dependent on the number of storm events (>50 mm per 24 h, Fig. 5b), due to the greater erosivity of high intensity storms and greater depth of leaching from larger rainfall events, which enhance P loss by erosion and leaching (Kleinman et al. 2006; Shigaki et al. 2007; Sharpley et al. 2008). P loss by greater precipitation and storm events would be further enhanced for soils that have received high rates of P 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). Although there were no significant trends in total annual precipitation over the past 31 years (supplementary material A, Fig. S2), the observed 1.0–5.0-fold increase in Olsen-P and 0.9–3.4-fold increase in TP concentrations (Fig. 6) as well as a 65–76 % increase in storm events (Fig. S2) would certainly contribute to an increasing riverine TP export fraction of NAPI and legacy P, implying that climate change (i.e., increased precipitation and increased storm events) will enhance P loss to rivers.

The significant positive correlation between agricultural land area drained with cement channels and pipes and riverine export fraction (Fig. 5c) implies that an increase in hydrologic connectivity via efficient drainage enhances P delivery from land to river (Gentry et al. 2007; Jarvie et al. 2013). Increasing drained agricultural land area as well as increased precipitation and storm events increase the P export fraction via increased soil erosion and sediment transport, decreased water residence time in the watershed, enhanced P leaching, and decreased stream sedimentation. The high dependence of riverine export fraction on the developed land area (Fig. 5d) is due to decreased P retention capacity and enhanced flushing during rainfall events, since developed lands have a greater impervious surface area resulting in greater runoff to surface waters and less infiltration to promote P retention by soils (McMahon and Woodside 1997; Sobota et al. 2011; Roy and Bickerton 2014). Also, years or catchments with more developed land area are generally accompanied with larger human and animal densities (Fig. 4), which implies more animal waste production and greater P input to rivers via waste discharge directly into rivers (point source pollution, Sobota et al. 2011). As a result, the catchment (e.g., HG) and period (e.g., 2005–2010) with greater drained agricultural land and developed land had a larger response in riverine TP export from NAPI (Fig. 4). These results imply that changes in land use (along with changing P pollution sources) play an important role in enhancing fractional export of P to rivers.

#### Efficacy and implication of the model for predicting riverine TP flux

The calibrated model Eq. (4) that incorporates NAPI, current year's NAPI, precipitation, drained agricultural land area percentage, and developed land area percentage for the Yongan watershed demonstrated

high accuracy for predicting annual riverine TP fluxes, as well as constant variance of model residuals (Fig. 7), reflecting the reliability and robustness of the model (Chen et al. 2014). Although this model lacks the ability to predict seasonal or daily riverine P export and differentiate among possible fates of TP compared to more complex mechanistic models, it has the advantage of simplicity and is easily applied to predict riverine TP export from watershed anthropogenic P inputs, hydroclimate and land use. Another important feature of this model is its ability to quantify the contributions of current year's NAPI and legacy P sources to annual riverine TP flux. The model Eq. (1), which is supported by the observed relationships between riverine TP flux and each of the relevant influencing factors for the Yongan watershed (Figs. 2 and 5), might be directly applicable in other watersheds with similar characteristics. For watersheds with different characteristics, the influencing factors and function types in Eq. (1) may change and require optimization to apply in different watersheds. However, the methodology proposed in this study should be widely applicable to other watersheds.

Using model Eq. (2), the method (i.e., Eqs. (2) and (3)) developed for identifying the contributions of current year's NAPI and legacy P sources to annual riverine TP flux is based on the assumption that the amount of P released by rock weathering is negligible. For the Yongan watershed, the dominant soils are highly weathered red and yellow soils (~90 % of entire watershed area; Oxisols and Ultisols) that are believed to contribute a very limited background P flux compared to NAPI sources (Zhang et al. 2005). This hypothesis is supported by a study in the Lake Dianchi watershed in southern China where red soils were found to contribute  $0.006 \text{ kg P ha}^{-1} \text{ year}^{-1}$  via natural rock weathering (Liu 2005). Similarly, the low TP export coefficients observed in several natural ecosystems ( $0.0085 \text{ kg P ha}^{-1} \text{ year}^{-1}$ , USEPA 2002) indicate weathering rates that are considerably less than the legacy P contribution to riverine TP fluxes in the Yongan watershed (Fig. 8). Due to the inverse relationship between contributions from current year's NAPI and legacy P sources, the contribution of legacy P sources would be overestimated if NAPI was underestimated (and vice versa). Therefore, improving the accuracy of NAPI estimates (i.e., parameters and P source/sink data, supplementary material D) would reduce uncertainty associated with estimates of

riverine TP sources. Since the riverine TP source apportionment method (i.e., Eqs. (2) and (3)) is derived from model Eq. (1), the model error has a potentially large influence on the reliability of source apportionment results. Therefore, determining the model with the highest prediction accuracy for a given watershed is necessary to reduce the uncertainty in apportioning riverine TP sources.

#### Influence of NAPI and legacy P on riverine TP export dynamics

In the Yongan watershed, the high contribution (92 %) of current year NAPI to annual riverine TP flux in catchment HG (Fig. 8) implies a highly efficient delivery pathway, such as wastewater discharge directly into the river. This finding is supported by a significant negative correlation between river TP concentration and water discharge in the HG catchment ( $p < 0.01$ , supplementary material B, Fig. S6). Since wastewater discharge to the river is relatively constant and hydrologically independent, it is subsequently diluted during the high flow regime (Chen et al. 2013; Bowes et al. 2014). In contrast, the lack of significant correlations in catchments HX, HB, BZA and XZ (Fig. S10) indicates that both hydrologically-dependent processes (erosion and leaching) and hydrologically-independent processes (sewage discharge) contributed to riverine TP fluxes in these catchments (Bowes et al. 2014).

Current year's NAPI contributions to annual riverine TP fluxes continuously increased in catchments HX, HB, BZA and XZ (Fig. 8), even when NAPI began to decline in 2000 (Fig. 2), which mainly resulted from the enhanced fractional export of NAPI due to changes in hydroclimate and land use (2.6–14.6-fold increase since 1980, Fig. 5). The small decrease in the current year NAPI contribution to riverine TP flux between 2000 and 2010 in catchment HG is mainly attributed to the 25 % decrease of domestic animal density since 2000 (Table 1), implying a decrease in animal waste discharged to the river (Borbor-Cordova et al. 2006). Wastewater treatment plant construction in this highly population catchment is another possible cause of decreasing riverine TP flux since 2000. Furthermore, the high correlation between riverine TP flux and NAPI in the form of an exponential relationship for catchment HG ( $R^2 = 0.84$ ,  $p < 0.01$ , Fig. 2) suggests that small

changes in NAPI can lead to relatively large changes in riverine TP flux (Fig. 4). This relationship perhaps results from P saturation of watershed storage pools (soils, wetlands, riparian buffers and sediments) for retaining P after long-term excessive P inputs (Dunne et al. 2011; Reddy et al. 2012).

Legacy P sources comprised a considerable proportion (8–58 %) of the annual riverine TP export in the six catchments (Fig. 8). Such legacy P contribution partially explains the increasing riverine TP flux observed in the 2000s, despite a decline in NAPI since 2000 in catchments HX, HB, BZA, ZK and XZ (Fig. 2). Legacy P can be remobilized or recycled as landscape storage capacity becomes P-saturated, or after changes in land use, land management and hydroclimate (Jarvie et al. 2013; Haygarth et al. 2014). Thus, legacy P sources can provide a continuing source of P to downstream water bodies (Zhang et al. 2004; Kleinman et al. 2011; Hamilton 2012; Reddy et al. 2012; Sharpley et al. 2013). The highest contribution (58 %) of legacy P to annual riverine TP flux was observed in catchment ZK (Fig. 8) and suggests a slow delivery of P from land to river and a dominant role from nonpoint sources (e.g., erosion and leaching). This hypothesis is supported by the positive relationship between riverine TP concentration ( $C$ , mg P L<sup>-1</sup>) and river discharge ( $Q$ , m<sup>3</sup> s<sup>-1</sup>):  $C = 0.0118Q^{0.0395}$ ,  $p < 0.01$  (supplementary material B, Fig. S6), since nonpoint source P input to rivers via erosion and leaching increases with increasing runoff volume (Chen et al. 2013; Bowes et al. 2014). Over the past 31 years, legacy P contributions to riverine TP flux continuously increased in all six catchments (Fig. 8), implying that legacy P contributions mask the impacts of contemporary P source reduction measures. If we assume that only 0.65 % of the P accumulated over the past 31 years (Table 2) is active and available for loss to rivers via erosion and leaching as determined in the Lake Okeechobee Basin by Reddy et al. (2011), the six catchments in the Yongan watershed will continue to yield average riverine TP fluxes of 0.04–1.71 kg P ha<sup>-1</sup> year<sup>-1</sup> from legacy P sources (average of the 2000–2010 period) for the next 16–100 years. This highlights the impact of the long lag time associated with legacy P mobilization which hinders riverine TP reductions following watershed P source reduction strategies (Jarvie et al. 2013; Sharpley et al. 2013; Haygarth et al. 2014).

## Implications for watershed phosphorus management

The forecasted results represented by the “adaptive management” and “developing I” scenarios roughly characterize the upper and lower bounds for future riverine P exports in response to anthropogenic activities and climate change (Table 3). These forecasted bounds provide a baseline for assessing and adopting relevant watershed P management strategies. Increasing precipitation is predicted to have an impact on increasing TP export from the watershed due to the enhanced export fraction of NAPI and legacy P, which will be further enhanced by an increasing prevalence of storm events (Fig. 5b). This forecast suggests that the influence of climate change on riverine TP exports should be considered as part of management efforts to control P water pollution. The application of intermediate or cover crops (Meals et al. 2010; Sharpley et al. 2013) is a beneficial practice for decreasing P loss via enhanced runoff and leaching due to climate change and legacy P release.

In terms of P source management in the Yongan watershed, reducing chemical fertilizer application would likely have the greatest impact on reducing riverine TP fluxes given the dominant contribution of P fertilizer to NAPI (Fig. 2). Increasing net food/feed P input and non-food P input (Fig. 2) implies increasing human and animal wastes. Enhanced sewage collection and treatment are required to reduce domestic waste disposal to rivers, while application of animal waste to agricultural lands requires attention to balance application rates with plant uptake requirements. For the entire watershed, P source reduction should be primarily focused on the areas located in the middle and downstream portions of the watershed as delineated with red lines in Fig. 3. Targeting these critical source areas, which only occupy 11 % of the entire watershed area but contribute 29 % of riverine TP load at the BZA sampling site in 1990–2010 (as predicted by Eq. (4)), is considerably more cost effective than mitigation approaches that blanket the entire watershed (Kleinman et al. 2011).

Legacy P provided a considerable contribution to riverine TP fluxes in 1980–2010 (Fig. 8) and has the potential to increase contributions with changes in climate and land use in the future (Table 3). Therefore, future management programs designed to reduce P exports to rivers must consider the large amount of P transiently stored in the watershed (Bennett et al. 1999;

Meals et al. 2010; Sharpley et al. 2013; Haygarth et al. 2014). In the Yongan watershed, Olsen P levels in the upper 20-cm layer of agricultural soils in 2009 (32–57.2 mg P kg<sup>-1</sup>, Fig. 4) exceeded the optimal range (e.g., 25–30 mg P kg<sup>-1</sup>) for crop production recommended in several developed countries (Ma et al. 2013). This results in a required reduction of 6–65 % in Olsen-P levels (5.3–75.6 kg P ha<sup>-1</sup>) for the upper 20-cm layer of agricultural soils to attain the optimum range for crops. This is especially important for catchment XZ, which requires a 65 % (75.6 kg P ha<sup>-1</sup>) reduction, while catchment ZK requires a 63 % (70.5 kg P ha<sup>-1</sup>) reduction (Fig. 6). According to the average P removal rate by crop production of 12.7 kg P ha<sup>-1</sup> year<sup>-1</sup> for catchment ZK and 10.5 kg P ha<sup>-1</sup> year<sup>-1</sup> for catchment XZ, it would require 6–7 years of crop production with no additional fertilizer application to remove the excess P without seriously comprising crop yields. This prediction is supported by several field observations, where a 48–90 % reduction in Olsen-P levels required 8–14 years (Sharpley et al. 2013) and subsequently required a decade or more to “draw down” soil P reserves to levels where dissolved P in runoff was substantially reduced (Jarvie et al. 2013). Bioavailability and utilization of legacy inorganic and organic P resources in soil can be enhanced using biological methods (e.g., plant breeding and genetic modification to enhance P uptake rates, Condon et al. 2013). Given the enhanced dissolved P leaching contributed by improved agricultural drainage systems, subsurface drainage from agricultural lands will play an increasing role in transporting legacy P to rivers (Kleinman et al. 2011). Intercepting P during hydrologic transport using constructing ecological ditches, flow-through wetlands and P sorbing materials (e.g., fly ash and steel slag) would be beneficial for reducing dissolved P loss from these drainage waters (Penn et al. 2014). Deep tillage may also be used to promote mixing of highly P-saturated surface soils with subsoil horizons that have a higher P sorption capacity (e.g., higher Fe/Al oxides/hydroxides) to improve agricultural soil retention of P (Kleinman et al. 2011).

## Conclusion

This study highlights that spatio-temporal variations in riverine TP flux are not only dependent on changes

in NAPI, but are also influenced by changes in hydroclimate and land use as they determine the export fraction of NAPI and legacy P to rivers. The model developed in this study incorporates these influencing factors and provides a simple and efficient method for predicting riverine TP export and identifying the contribution of legacy P sources to annual riverine TP flux. Legacy P that has accumulated in watershed storage pools (e.g., soils, vegetation, river sediments) from historical practices, especially in agricultural soils, is a considerable potential source of riverine TP and should be considered in addressing the relationship between catchment P budgets and riverine P export. Anthropogenic activities have increased P inputs available for export and the fractional export of P inputs, as well as the contribution from legacy P sources, while climate change can further enhance P export to rivers. Enhanced export of legacy P and NAPI by changes in land use and climate are predicted to delay reductions in riverine P flux in response to reductions in NAPI and should be considered in developing watershed P management strategies.

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**Conflict of interest** This study has no conflict of interest with any persons or affiliations.

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