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Long-term (1980–2010) changes in cropland phosphorus budgets, use efficiency and legacy pools across townships in the Yongan watershed, eastern China

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ABSTRACT

Quantitative information on cropland phosphorus (P) flows at the township scale is critical for developing sustainable P management measures under the smallholder farming system. This study addressed changes in cropland soil surface P budgets (i.e., net of P inputs and crop outputs), use efficiencies (i.e., the ratio between crop P uptake and total P input) and legacy P pools across 21 townships in the Yongan watershed of eastern China in 1980–2010. For the entire watershed, total P input (>98% from synthetic fertilizer and farmyard manure), crop uptake and budgets per cropland area increased from 50.4, 17.3 and 33.1 kg P ha^{-1} yr^{-1} in 1980 to 74.6, 20.5 and 55.1 kg P ha^{-1} yr^{-1} in 1995, and then sharply declined to 39.6, 11.4 and 28.2 kg P ha^{-1} yr^{-1} in 2010, respectively. Estimated P use efficiency decreased from 34% in 1980 to 26% in 1999 before slightly increasing to 28% in 2010. Although the 21 townships had similar temporal variations over the 1980–2010 period, P budgets and use efficiency showed 2–3-fold spatial variability among townships within a given year. Spatio-temporal variations in the P budget and use efficiency were mainly related to changes in P fertilization rates and patterns (i.e., ratio of applied synthetic fertilizer P and farmyard manure P) and cropland types. The 20 townships having soil data had 87–720% and 113–395% increases of Olsen-P and total P contents in the upper 20 cm of cropland soils between 1984 and 2009, respectively. Increased soil TP level between 1984 and 2009 suggested that more than 53–79% of the cumulative P budget accumulated as legacy P pools in cropland soils. Based on regression analyses, legacy soil P contribution to annual crop P uptake was estimated to increase from 0.47 kg P ha^{-1} yr^{-1} (3%) in 1980 to 3.45 kg P ha^{-1} yr^{-1} (31%) in 2010, with 52–80% from synthetic fertilizer and 2–46% from farmyard manure. Improved utilization of soil legacy P pools for crop production and increasing P use efficiency are necessary to minimize P inputs and reduce nonpoint source P pollution load. The high spatial heterogeneity in P budgets and use efficiencies across townships, as well as considerable legacy soil P pools after long-term over-application, should be considered in developing P management strategies under smallholder farm systems.

1. Introduction

Phosphorus (P) fertilization is an important measure for ensuring soil fertility, improving crop yield, and meeting food needs for a rapidly increasing global population (Haygarth et al., 2014; Liu et al., 2016). However, excessive P application has induced P surplus in croplands, decreasing agronomic P use efficiency as well as increasing P loss to surface waters resulting in eutrophication (Sharpley et al., 2013; Wang et al., 2014). Therefore, efficient use of non-renewable P resources for crop production has become a global concern from agronomic, economic and environmental perspectives (Li et al., 2015; Sharpley et al.,

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2015). To improve P use efficiency and reduce P loss to surface waters, quantitative knowledge concerning P inputs and outputs is required for developing sustainable agricultural P management measures.

Budgeting approaches that compare inputs and outputs of P are used to assess P use efficiency and a P surplus indicates excessive P application and soil P build-up, as well as increased potential for P loss to surface waters (Oenema et al., 2003; Chen et al., 2015). There are three basic agricultural P budgeting approaches: farm gate, soil surface budget and soil system budget (Oenema et al., 2003; Wang et al., 2014). Although each approach has its specific function and advantage, the availability of input data often determines selection of methods. The soil surface budget approach, which addresses the P that enters the soil via the surface and leaves the soil via crop uptake (Ouyang et al., 2013), has been widely applied at different geographical scales (e.g., field plot, catchment, regional, and national, MacDonald et al., 2011; Han et al., 2012; Sattari et al., 2012; Cao et al., 2012). However, fewer studies have conducted cropland soil surface P budgets at the township scale. Worldwide there are about 450–500 million smallholder farms that manage up to 2 ha of cropland, with Asian countries having a particularly high percentage (Anthony and Ferroni, 2012). As a major smallholder farm dominant country, China has 240 million smallholder farmers with each household having limited cropland area (e.g., 0.01–0.50 ha) (Miao et al., 2011; Ouyang et al., 2013). In China, current scientific and technological extension services for agriculture are offered by county-level "Agricultural Technical Extension Centers" and township-level stations (Miao et al., 2011). The township acts as the grass-roots government unit for managing such smallholder crop production systems, resulting in considerable heterogeneity in applied P fertilizer rates and cropland types among townships even within a county. From an environmental perspective, the issues associated with agricultural nonpoint source P pollution are often best approached by considering management options on a watershed scale due to its high dependence on hydrological processes (Haith, 2003). Therefore, information on spatial variations of cropland P budgets and use efficiency across townships within a watershed is critical for guiding efficient P management from both agronomic and environmental perspectives under the smallholder farming system.

Previous cropland P budget studies commonly show that only 10–20% of input P is used by the first crop after application and a substantial fraction of applied P accumulates in the soil as residual P (Sattari et al., 2012; Haygarth et al., 2014; Powers et al., 2016). Globally, 71% of the cropland area was estimated to have overall P surpluses in 2000, including most of East Asia, sizeable tracts of Western and Southern Europe, coastal United States, and southern Brazil (MacDonald et al., 2011). Such surplus or legacy P in soils derived from anthropogenic P inputs in previous years can be remobilized or recycled, acting as a continuous P source to crop production as well as to P pollution in surface waters (Sharpley et al., 2013; Jiang and Yuan, 2015; Liu et al., 2016). Although legacy P has received increasing attention from agronomic and environmental perspectives (Sattari et al., 2012; Sharpley et al., 2015; Rowe et al., 2016), limited knowledge is available on what proportion of crop P uptake is derived from legacy P compared to synthetic fertilizer and farmyard manure P. Such quantitative information is required for assessing the potential utilization of legacy P pools as an alternative P source compared to non-renewable rock phosphate resources for crop production. Legacy P is particularly significant in many Chinese croplands (Jiang and Yuan, 2015; Powers et al., 2016; Liu et al., 2016), since most smallholder farmers lack information concerning the appropriate amount and timing of P fertilizer applications to match crop requirements. This often leads to excessive P application rates as an "insurance" to reach maximum yields (Miao et al., 2011). In eastern China (identified as having the highest eutrophication potential in freshwaters due to excessive P in China, Liu et al., 2016), P loss from croplands due to over-applied P is a primary source of P to surface waters (Hou et al., 2013; Li et al., 2016). Our previous studies indicated that legacy P pools could contribute 13–32% (with ~80% derived from croplands) of annual riverine total P flux in 1980–2010 in the Yongan watershed of eastern China (Chen et al., 2015, 2016). Accordingly, it is important to provide quantitative information concerning accumulation of the legacy P pool in croplands and its contribution to crop P uptake for watersheds in eastern China.

We hypothesized that there is a considerable spatial heterogeneity in cropland P budgets and use efficiency across townships within a watershed, as well as a significant legacy soil P effect after long-term crop cultivation under smallholder land management. Based on an extensive 31-year data record (1980–2010) for 21 townships in the Yongan River watershed of eastern China, this study (i) evaluates spatio-temporal variations of P input, crop uptake, budgets and use efficiency as well as soil P levels; (ii) addresses the factors influencing P budgets and use efficiency, and (iii) quantifies contributions of legacy soil P, synthetic fertilizer, and farmyard manure to crop P uptake. Results of this study improve our understanding of cropland P flows under smallholder cropping systems to guide efficient P management for achieving the co-benefits of P resource conservation and eutrophication mitigation.

2. Materials and methods

2.1. Watershed description

The Yongan watershed (120.2295°–121.0146° E and 28.4695°–29.0395° N) is located in the developed Taizhou region of Zhejiang Province, China (Fig. 1). The watershed area (2474 km^2) covers 20 townships within Xianju County and one township within Linhai City. Total population within the watershed increased from ~590,000 in 1980 to ~740,000 in 2010. Over the 31-year study period, domestic livestock production (pig, cow, sheep and rabbit) decreased by ~25%, while poultry production (chicken and duck) increased by 4.8-fold (**Supplementary material A, Fig. A.1**). Cropland (e.g., paddy field, garden plot and dryland) averaged ~12% of total watershed area in 1980–2010 (**Supplementary material A, Fig. A.1**). Recycled animal and human excreta for fertilizing croplands (e.g., farmyard manure) decreased from ~93% in 1980 to ~21% in 2010 due to increasing availability of synthetic P fertilizer.

Considering the availability of relevant long-term data, this study selected the Yongan watershed as a representative watershed in eastern China, as it was subjected to rapid changes in cropland P input rates and patterns as well as crop rotation patterns over the past three decades under the smallholder land policy. The cropland policy named "household responsibility system" (i.e., smallholder land policy) was implemented in 1978. Under the smallholder land policy, a single household farm generally cultivates a farmland area of about 0.01–0.20 ha with different crop types and field management practices. The cropland types, crop yields, and fertilization quantities are highly heterogeneous across the 21 townships (Table 1). On average over the 1980–2010 period, paddy field (e.g., rice and wheat) and garden plot (e.g., tea and fruits) area contributed 43–70% and 18–48% of total agricultural area, respectively, while dryland (e.g., vegetables and potato) accounted for 4–16% across the 21 townships. Over the 1980–2010 period, the ratio between annual synthetic fertilizer and farmyard manure P application rates ranged from 1.2:1 to 2.7:1 across the 21 townships. Since the 1950s, smallholder farms have typically implemented a rotation cropping pattern that

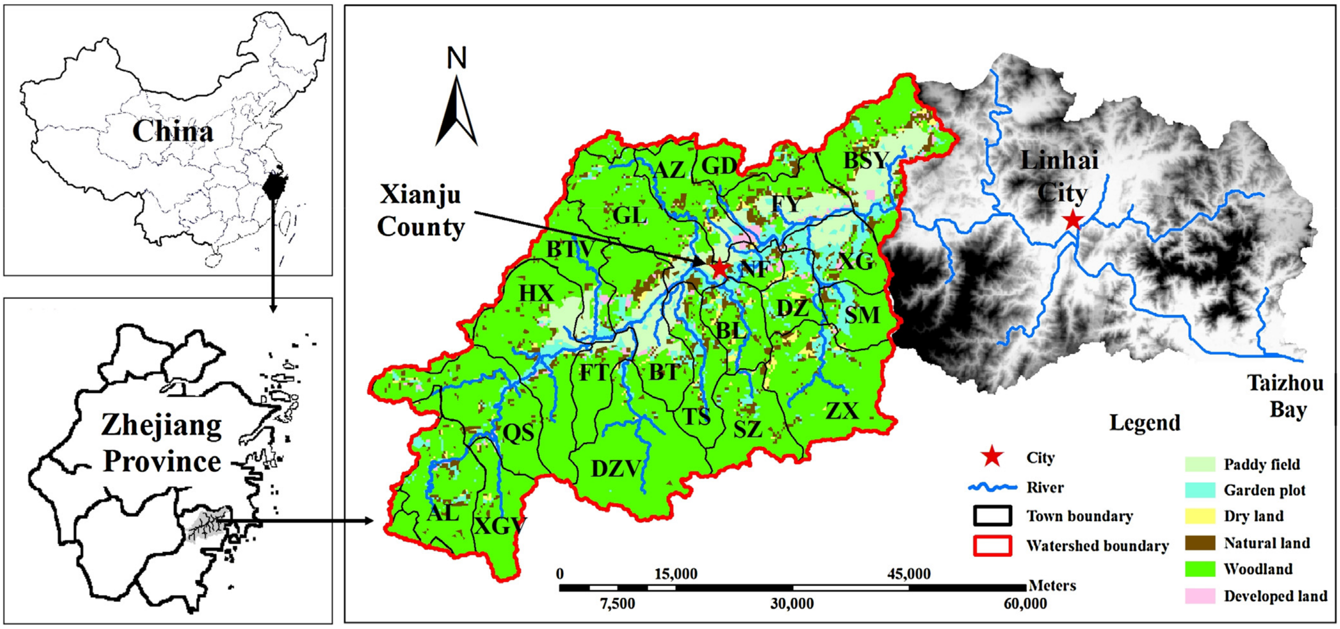

Fig. 1. The location of the Yongan River watershed in China and Zhejiang Province and the 21 townships.

continuously cultivates two or more crops on the same field in a given year. Typical rotation cropping systems, such as wheat–rice–rice, rape–rice–rice, wheat–rice–vegetables and wheat–rice–maize in paddy fields and leafy vegetables–melons, leafy vegetables–fruity vegetables, and maize–root vegetables in dryland fields, were widely applied in each township. Although there was a slight increase in cropland area (**Supplementary material A, Fig. A.1**), annual cultivated cropland areas (i.e., cumulative area planted to crops in a given year) have experienced a large reduction (25–66%) including a large decline in synthetic fertilizer application (24–73%) since 2000.

2.2. Cropland P budget and use efficiency estimations and uncertainty analysis

Due to the unavailability of some relevant data (e.g., annual synthetic fertilizer and farmyard manure P application rates as well as crop yields for each cropland type), this study was unable to separately estimate P budgets and use efficiency for each individual cropland type. Instead, we made estimates for the average P budget and use efficiency of all croplands in each township. Considering no grazing activities in the study watershed, the annual cropland P budget was estimated as the difference

Table 1
Characteristics of cropland types, population (P), domestic animals (DA), P application rates of synthetic fertilizer and farmyard manure, and crop yields for the 21 townships of the Yongan watershed over the 1980–2010 period.

Township	Paddy field (ha)	Dry land	Garden plot	P (capita ha^{-1})	DA	Synthetic fertilizer (kg P ha^{-1} yr^{-1})	Farmyard manure	Grain crop yield (t ha^{-1} yr^{-1})	Cash crop yield
AZ	438	57	209	35	10	45.9	26.0	19.5	23.1
NF	358	47	260	42	14	34.9	28.8	21.5	18.8
FY	1184	137	581	24	7	36.0	18.4	20.1	15.0
HX	1479	110	869	17	6	37.3	16.4	12.9	14.0
BTV	586	48	263	19	9	48.3	21.1	22.6	17.6
BT	1426	126	473	19	9	39.5	21.5	20.3	13.6
TS	877	97	333	20	11	47.4	26.3	23.8	18.6
GL	630	61	261	21	10	46.3	26.6	17.5	15.2
XG	1615	375	430	19	7	42.3	19.0	18.0	13.4
ZX	859	113	465	20	8	42.5	20.7	14.4	16.6
AL	218	51	223	23	8	44.8	22.7	9.0	15.7
XGV	292	68	314	16	8	32.0	19.4	7.1	14.8
QS	471	81	359	18	8	40.9	21.2	14.9	12.2
DZV	324	75	127	23	12	45.2	27.4	17.2	12.3
FT	502	94	231	17	6	36.4	27.3	21.0	13.9
SZ	421	53	283	17	7	37.5	19.3	10.8	11.4
BL	482	66	369	16	7	35.0	17.6	11.1	15.4
GD	369	53	360	13	15	47.6	32.2	13.0	23.5
DZ	562	82	196	11	9	46.8	19.9	21.7	13.0
SM	484	78	135	13	7	47.6	17.4	19.3	15.3
BSY	2309	394	1504	22	5	35.6	15.7	18.2	12.9

The number of each type of domestic animal is converted into the equivalent number of pigs according to their P excretion rates as shown in Table 2. Grain crop (i.e., rice, wheat and maize) yield is dry weight, while garden crop (i.e., all kinds of vegetables and fruits) yield is fresh weight. The value for each variable in a column denotes the average over the 1980–2010 period.

between total P input to croplands and P removal via crop harvest (Shen et al., 2005; Maguire et al., 2009; Schröer et al., 2011; Li et al., 2015). The soil surface P budget may be overestimated by not considering P loss via runoff from croplands (which is one of the largest contributors to total P pollution loads to surface waters in many regions) due to lack of relevant data. However, such overestimates are considered small since annual P loss to surface waters accounts for <5% of the cropland P budget (Zhang et al., 2015; Chen et al., 2016), with cropland loss rates of <1.5 kg P ha^{-1} yr^{-1} observed in China (Hou et al., 2013; Ma et al., 2013; Li et al., 2015). Therefore, runoff losses of P would have limited impact from an agronomic perspective. In this study, synthetic fertilizer, farmyard manure, atmospheric deposition, irrigation water and seed were considered as P inputs. Pesticide P input was neglected since it represented <0.001% of P fertilizer input (Chen et al., 2015). Annual P use efficiency was then estimated as the ratio between crop P uptake and total P input.

This study made several modifications to the conventional soil surface P budget approach by i) considering the change in annual human P consumption rate on P excreta rate over the past 31 years (Chen et al., 2016) for estimating recycled human P excreta to croplands, ii) adjusting the influence of breeding time for each domestic animal type on annual P excreta rate (Chen et al., 2015) in estimating recycled animal P excreta to croplands; and iii) considering the removal of crop residues for feeding domestic animals and for cooking fuel in rural areas (Yan et al., 2006) in estimating P output via crop harvest (i.e., grain/vegetable/fruit harvest and crop residue removal). Detailed descriptions of methods for estimating P input and output via crop harvest as well as data and reference sources for the parameters used in the budgets are listed in **Supplementary material B, Table B.1**.

To gain insight into the uncertainty in the estimates for each township, an uncertainty analysis was performed using Monte Carlo simulation (Chen et al., 2015). In accordance with previous studies (Yuan et al., 2011; Chen et al., 2016), we assumed that all parameters (**Supplementary material B, Table B.1**) used in the total P input and crop uptake estimations followed a normal distribution with a coefficient of variation equal to 30%. The Monte Carlo sampling method randomly generated 10,000 sets of model parameters according to their normal distribution functions, resulting in 10,000 iterations for total P input, crop uptake, budgets, and use efficiency simulations for each year in each township to obtain a mean and 95% confidence interval for these values.

2.3. Cropland soil P measurement

The Yongan watershed is dominated by highly weathered and acidic soils (Oxisols (65%) and Ultisols (15%), **Supplementary material C**). The local Agriculture Bureau provided data on soil Olsen-extractable P (Olsen-P), total P (organic + inorganic P), and bulk density measured at the same location in 1984 and 2009 along with data on each cropland type in each township. As a result, we compiled time-paired Olsen-P (n = 13–57), total P (n = 7–20), and bulk density (n = 5–20) data sets for the 20 townships in Xianju County to evaluate changes in soil P pools between 1984 and 2009 (soil data for township BSY in Linhai City was not available). Soil bulk density ranged within 0.72–1.82 g cm^{-3} in 1984 and 0.70–1.80 g cm^{-3} in 2009 with no significant difference between years (Chen et al., 2015). The TP or Olsen-P densities in the upper 20-cm soil layer of each cropland type in 1984 or 2009 were estimated from average soil TP or Olsen-P contents and bulk density. The areal average increase (AI) in TP or Olsen-P content (mg P kg^{-1}) or density (kg P ha^{-1}) for each township between 1984 and 2009 was estimated as:

$$AI = \frac{IP \times AP + IG \times AG + ID \times AD}{AP + AG + AD} \quad (1)$$

where IP, IG and ID are increased average P content or density in paddy field, garden plot and dry land soils between 1984 and 2009, respectively; AP AG and AD are paddy field, garden plot and dryland areas, respectively. Net P accumulation amounts in the upper 20 cm soil layer of each cropland and each township between 1984 and 2009 were estimated as the average increase in TP density and the areal average increase in TP density, respectively.

2.4. Data processing and analysis

Data compilations and relevant figures were generated with Excel 2007 software (Microsoft Corp., Seattle, WA, USA). To address uncertainty, the estimation procedures for total P input, crop uptake, budget and use efficiency were individually formulated in Excel 2007 embedded with Crystal Ball software (Professional Edition 2000, Oracle Ltd., Redwood Shores, CA, USA) to run Monte Carlo simulations. Correlation analysis, regression analysis, and one-way analysis of variance were performed using SPSS statistical software (version 16.0, SPSS Inc. Chicago, USA). Significance of changing trends for cropland P budget and P use efficiency over time period was determined by regression analysis between each parameter and year number.

3. Results

3.1. Spatio-temporal variations of cropland P inputs, crop uptake and budgets

For the 21 townships of the Yongan watershed, total P input to croplands increased from 39.7–79.6 kg P ha^{-1} yr^{-1} (50.4 kg P ha^{-1} yr^{-1} for entire watershed) in 1980 to 52.9–117.1 kg P ha^{-1} yr^{-1}

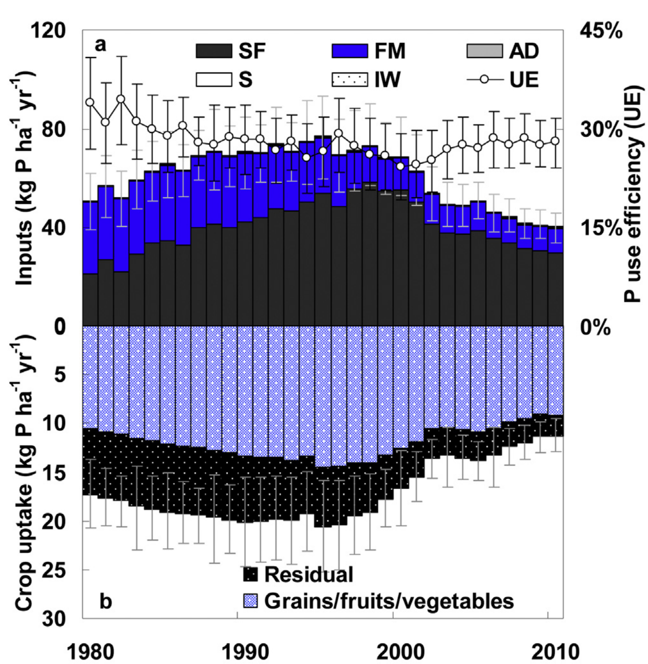

Fig. 2. Historical changes in P input to croplands from synthetic fertilizer (SF), farmyard manure (FM), atmospheric deposition (AD), seed (S) and irrigation water (IW); P use efficiency (UE); and crop P uptake in the Yongan watershed over the 1980–2010 study period. Error bars denote the 95% confidence intervals of total input and crop uptake from Monte Carlo simulations.

Table 2
The minimum and maximum values of cropland P input, crop uptake, budget and use efficiency among 21 townships of the Yongan watershed in each year over the 1980–2010 period.

	Total input ($kg\,P\,ha^{-1}\,yr^{-1}$)		Crop uptake ($kg\,P\,ha^{-1}\,yr^{-1}$)		Budget ($kg\,P\,ha^{-1}\,yr^{-1}$)		Use efficiency (%)	
Years	Min.	Max.	Min.	Max.	Min.	Max.	Min.	Max.
1980	39.7	79.6	11.4	23.0	26.7	63.6	20	40
1981	44.1	89.3	11.5	23.1	27.3	72.6	19	38
1982	40.1	81.3	11.4	23.3	22.8	64.7	20	43
1983	47.8	92.0	11.4	23.5	29.2	75.5	18	39
1984	51.1	96.1	11.3	23.6	34.9	79.6	17	36
1985	53.0	100.2	11.4	23.9	37.5	83.5	17	35
1986	50.5	95.9	11.2	24.1	35.9	79.1	18	36
1987	54.5	106.7	11.0	24.2	38.5	90.1	16	33
1988	55.3	110.4	10.8	24.6	38.5	93.7	15	34
1989	53.5	106.1	11.7	24.9	37.3	89.5	16	35
1990	54.4	109.4	12.5	24.8	37.1	91.5	16	35
1991	54.8	106.3	12.4	25.2	40.6	89.6	16	34
1992	57.3	114.0	12.8	24.8	42.0	97.2	15	32
1993	63.4	91.6	12.0	26.5	44.3	79.6	13	33
1994	65.2	98.7	12.2	26.2	48.7	86.5	12	30
1995	52.9	117.1	13.1	26.3	37.6	99.0	15	32
1996	58.5	83.3	12.4	26.7	39.4	66.6	19	35
1997	60.8	91.4	10.6	25.8	42.9	74.4	16	33
1998	49.0	91.3	9.9	23.7	39.1	73.3	16	31
1999	51.3	89.7	10.4	23.4	36.1	67.2	14	33
2000	54.6	95.0	9.6	22.6	43.3	72.4	12	29
2001	53.6	90.2	8.7	22.0	40.9	68.3	14	29
2002	40.7	73.7	8.6	17.9	29.9	56.5	17	31
2003	31.7	73.5	6.8	19.7	24.0	53.9	16	37
2004	33.0	75.8	7.0	19.9	23.8	55.9	17	35
2005	34.6	73.9	6.9	18.8	25.3	56.7	17	34
2006	33.7	68.9	7.5	20.0	22.7	48.9	19	34
2007	34.1	63.5	6.7	17.6	23.8	45.9	19	35
2008	32.2	58.0	7.0	17.2	21.1	42.8	19	35
2009	30.2	58.1	6.4	16.4	20.4	45.0	19	34
2010	29.3	62.2	6.9	17.4	18.8	46.8	19	36

(74.6 $kg\,P\,ha^{-1}\,yr^{-1}$ for entire watershed) in 1995 before sharply declining to 29.3–62.2 $kg\,P\,ha^{-1}\,yr^{-1}$ (39.6 $kg\,P\,ha^{-1}\,yr^{-1}$ for entire watershed) in 2010 (Fig. 2a and Table 2). Of annual total P input for each township, 98–99% was derived from synthetic fertilizer and farmyard manure applications, implying that other sources (i.e., irrigation water, seed input, and atmospheric deposition) had a very limited contribution and could be neglected (Fig. 2a). For the 21 townships, synthetic fertilizer P application rates increased by 29–47 $kg\,P\,ha^{-1}\,yr^{-1}$ from 1980 to 1999 followed by a 13–36 $kg\,P\,ha^{-1}\,yr^{-1}$ decline from 2000 to 2010.

Coincident with changes in total P input between 1980 and 2010, cropland P outputs via harvest and residue removal increased from 11.4–23.0 $kg\,P\,ha^{-1}\,yr^{-1}$ (17.3 $kg\,P\,ha^{-1}\,yr^{-1}$ for entire watershed) in 1980 to 13.1–26.3 $kg\,P\,ha^{-1}\,yr^{-1}$ (20.5 $kg\,P\,ha^{-1}\,yr^{-1}$ for entire watershed) in 1995 before declining to 6.9–17.4 $kg\,P\,ha^{-1}\,yr^{-1}$ (11.4 $kg\,P\,ha^{-1}\,yr^{-1}$ for entire watershed) in 2010 (Figs. 2 b and 3 b and Table 2). Cropland P output via crop residue harvest continuously decreased by 50–81% (67% for entire watershed) over the past 31 years (Figs. 2 b and 3 b), which mainly resulted from increasing domestic animal feed imported from other regions (Chen et al., 2016) and decreasing use of crop residues for cooking fuel in rural areas (**Supplementary material B, Table B.1**).

Estimated as the difference between total P input and crop P uptake, the annual P budget increased from 26.7–63.6 $kg\,P\,ha^{-1}\,yr^{-1}$ (33.1 $kg\,P\,ha^{-1}\,yr^{-1}$ for entire watershed) in 1980 to 37.1–101.9 $kg\,P\,ha^{-1}\,yr^{-1}$ (55.8 $kg\,P\,ha^{-1}\,yr^{-1}$ for entire watershed) in 1995 before declining to 18.8–47.8 $kg\,P\,ha^{-1}\,yr^{-1}$ (28.3 $kg\,P\,ha^{-1}\,yr^{-1}$ for entire watershed) in 2010 (Fig. 4a and Table 2). This implies that P input rates largely exceeded crop demands. Although the 21 townships generally exhibited similar temporal trends in 1980–2010, there was ~2–3-fold spatial variability in total P input, crop P uptake and the net P budget across the 21 townships in a given year (Tables 2 and 3). Townships GD and BSY had the largest (31-year average: 66.3 ±22.9 $kg\,P\,ha^{-1}\,yr^{-1}$) and smallest (31-year average: 35.0 ±21.1 $kg\,P\,ha^{-1}\,yr^{-1}$) P budgets, respectively (Table 3).

3.2. Spatio-temporal variations in cropland P use efficiency

For the 21 townships in the Yongan watershed, the estimated cropland P use efficiency (ratio between crop P uptake and total P input) significantly decreased ($P < 0.05$) from 23 to 38% (34% for entire watershed) in 1980 to 16–30% (26% for entire watershed) in 2000 before slightly increasing ($P < 0.05$) to 19–36% (28% for entire watershed) in 2010 (Figs. 2 a, 4 b and Table 2). Although the 21 townships generally exhibited similar temporal trends, they displayed ~2-fold spatial variability in P use efficiency in a given year (Fig. 4b and Table 2). Townships BSY and GD had the highest

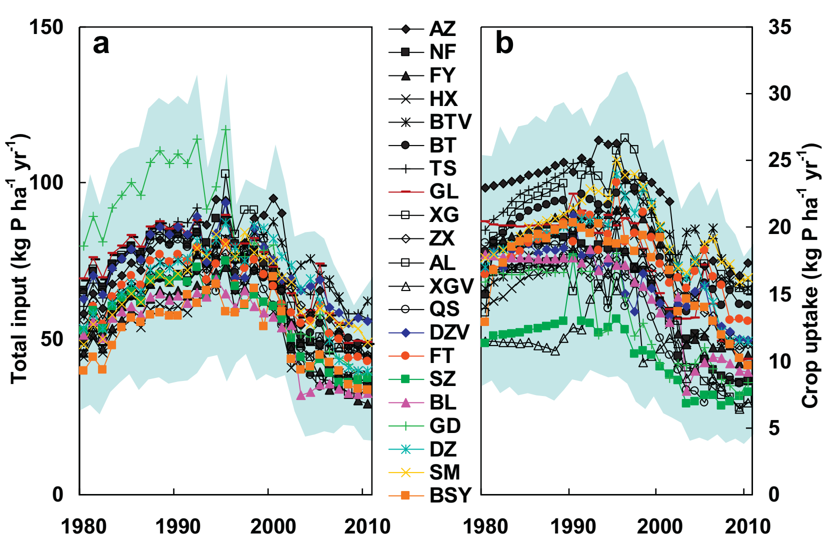

Fig. 3. Historical changes in total P input to croplands and crop P uptake across 21 townships in the Yongan watershed over the 1980–2010 study period. Shadow areas denote the largest 95% confidence intervals of annual total input and crop uptake among the 21 townships for each year from Monte Carlo simulations.

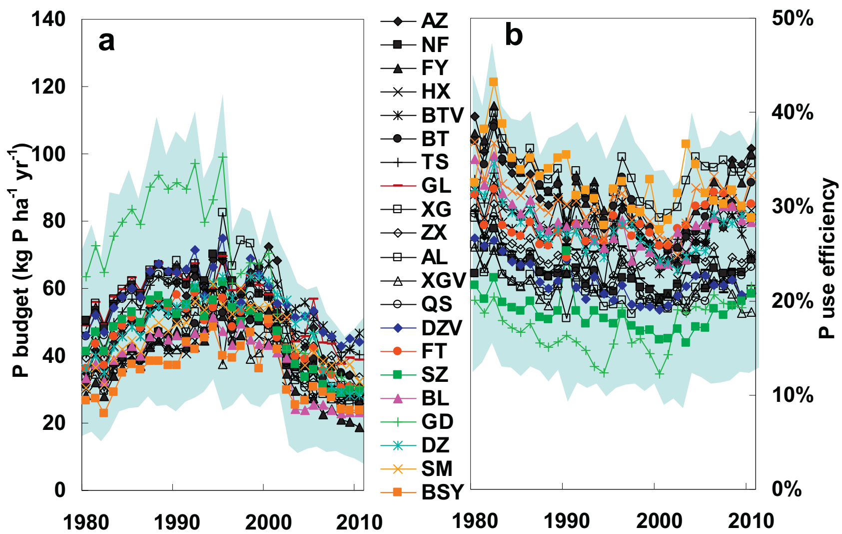

Fig. 4. Historical changes in cropland P budget and use efficiency across 21 townships in the Yongan watershed over the 1980–2010 study period. Shadow areas denote the largest 95% confidence intervals of annual total input and crop uptake among 21 townships for each year from Monte Carlo simulations.

Table 3
Average (±SD) cropland P input, crop uptake, budget and use efficiency over the 1980−2010 period for each of 21 townships in the Yongan watershed.

Township	Total input ($kg\,P\,ha^{-1}\,yr^{-1}$)	Crop uptake ($kg\,P\,ha^{-1}\,yr^{-1}$)	Budget ($kg\,P\,ha^{-1}\,yr^{-1}$)	Use efficiency (%)
AZ	72.9 ± 15.2	17.4 ± 3.7	51.1 ± 12.6	30 ± 4
NF	67.6 ± 17.8	10.7 ± 4.0	52.1 ± 14.0	23 ± 2
FY	55.2 ± 14.4	12.7 ± 4.2	37.9 ± 10.8	32 ± 4
HX	54.2 ± 10.5	12.0 ± 3.1	38.8 ± 7.8	28 ± 2
BTV	70.0 ± 9.8	18.4 ± 2.2	51.1 ± 8.0	27 ± 2
BT	61.5 ± 9.7	15.1 ± 3.3	42.6 ± 7.3	31 ± 3
TS	74.3 ± 10.7	17.9 ± 2.8	53.4 ± 8.3	28 ± 2
GL	73.4 ± 12.5	13.3 ± 3.8	55.8 ± 9.2	24 ± 2
XG	61.9 ± 12.2	16.8 ± 3.7	41.7 ± 9.2	33 ± 3
ZX	63.7 ± 9.0	13.9 ± 2.1	47.9 ± 7.3	25 ± 2
AL	68.0 ± 20.3	9.7 ± 4.1	53.7 ± 16.5	21 ± 2
XGV	52.0 ± 11.1	9.3 ± 2.9	40.5 ± 8.5	22 ± 2
QS	62.6 ± 17.2	8.8 ± 4.6	48.2 ± 12.9	23 ± 3
DZV	73.1 ± 10.6	13.6 ± 2.5	57.0 ± 8.7	22 ± 2
FT	64.3 ± 11.1	15.8 ± 2.4	46.4 ± 8.8	28 ± 2
SZ	57.3 ± 12.4	7.7 ± 2.6	46.6 ± 10.1	19 ± 2
BL	53.0 ± 12.5	10.8 ± 3.6	38.1 ± 9.4	28 ± 3
GD	79.6 ± 25.0	9.3 ± 3.6	66.3 ± 21.9	17 ± 4
DZ	67.4 ± 14.2	15.4 ± 3.3	49.0 ± 11.3	28 ± 4
SM	65.7 ± 10.8	17.8 ± 2.5	45.8 ± 8.7	31 ± 3
BSY	51.9 ± 10.9	13.9 ± 3.3	35.0 ± 8.1	33 ± 5

(31-year average: 33 ± 3%) and lowest (31-year average: 17 ± 3%) P use efficiencies, respectively (Table 3). Spatio-temporal variations of cropland P use efficiency (RE) were primarily attributable to differences in total P input rates (TI) and paddy field area percentage (PD): $RE = -0.0022TI + 0.29PD + 0.23$ ($R^2 = 0.68$, $n = 651$). A two-segment linear regression analysis demonstrates that there is a critical total P input value of ~60 $kg\,P\,ha^{-1}\,yr^{-1}$ at which the slope for P use efficiency shows a distinct change (Fig. 5a). P use efficiency declines rapidly at total P input ≥60 $kg\,P\,ha^{-1}\,yr^{-1}$, while there was no significant trend when total P input was <60 $kg\,P\,ha^{-1}\,yr^{-1}$. A two-segment linear regression analysis further indicates that there is a critical paddy field area percentage of ~50% associated with changing P use efficiency at the township scale (Fig. 5b). This implies that P use efficiency increases significantly at paddy field area ≥50%, while there was no significant trend when paddy field area was <50%.

3.3. Spatio-temporal cropland p budgets and soil P dynamics

For the 20 townships in the Yongan watershed having paired soil P data for 1984 and 2009, average Olsen-P levels in the upper 20 cm layer of paddy field, dryland and garden soils significantly increased by 13–42 $mg\,P\,kg^{-1}$ (38–420%), 27–82 $mg\,P\,kg^{-1}$ (132–1025%) and 24–68 $mg\,P\,kg^{-1}$ (93–1113%) between 1984 and 2009; correspondingly, TP levels increased by 187–440 $mg\,P\,kg^{-1}$ (82–309%), 273–640 $mg\,P\,kg^{-1}$ (155–464%) and 107–510 $mg\,P\,kg^{-1}$ (63–459%), respectively (Fig. 6a and b). As an areal average for all cropland in each township, soil Olsen-P and TP contents increased by 22–64 $mg\,P\,kg^{-1}$ (87–720%) and 245–530 $mg\,P\,kg^{-1}$ (113–395%) within each township, respectively (Fig. 6a and b). Increases in TP and Olsen-P contents resulting from addition of 100 $kg\,P\,ha^{-1}$ were estimated to be 23–34 and 1.8–3.8 $mg\,P\,kg^{-1}$ across the 20 townships, respectively. Spatial variation of increased Olsen-P contents in response to a 100 $kg\,P\,ha^{-1}$ addition among the 20

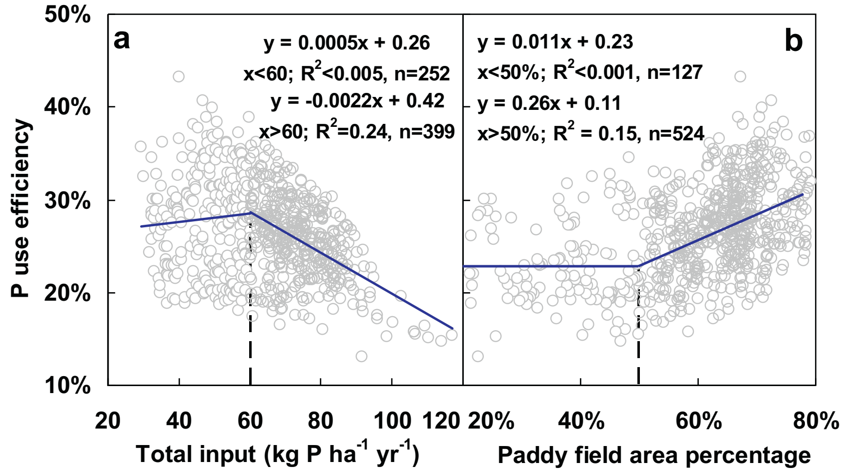

Fig. 5. Responses of P use efficiency to changes in total P input rate (a) and paddy field area percentage (b) across 21 townships in the Yongan watershed over the 1980–2010 study period.

townships may be mainly related to differences in the ratio between applied farmyard manure and synthetic fertilizer P rates (Table 1, $r = 0.78$, $P < 0.01$, $n = 20$). Consequently, township GD having the highest farmyard manure P/synthetic fertilizer P ratio (Table 1) had the largest (3.8 mg P kg^{-1}) increase in Olsen-P levels for each 100 kg P ha^{-1} addition.

Based on the changes in TP levels, an estimated 416–1264, 660–1811 and 263–1443 kg P ha^{-1} was accumulated in the upper 20 cm layer of paddy field, dryland and garden soils, respectively (Fig. 6c). The areal average for each township indicates an estimated 514–1329 kg P ha^{-1} was accumulated in the upper 20 cm layer of cropland soils (Fig. 6c), accounting for 53–79% of the cumulative P budget (i.e., 955–1700 kg P ha^{-1}) for each of the 20 townships during the 1984–2009 study period. The significant correlation between estimated areal average net TP accumulation in the upper 20-cm layer of soils (y) and cumulative P budget (x) ($y = 0.65\times - 236$, $R^2 = 0.70$, $n = 20$) implies that over-application of P was the major cause of legacy P accumulation in cropland soils. As a result, township GD having the highest P budget (Table 3) exhibited the greatest build-up of the soil legacy P pool (Fig. 7c). The significant P accumulation observed in the upper 20 cm layer of soils implies that deeper soil layers also have the potential to retain additional P, suggesting that more than 53–79% of the P budget was accumulated in cropland soils. In addition to the deeper layers of cropland soils, P storage in wetlands, drainage

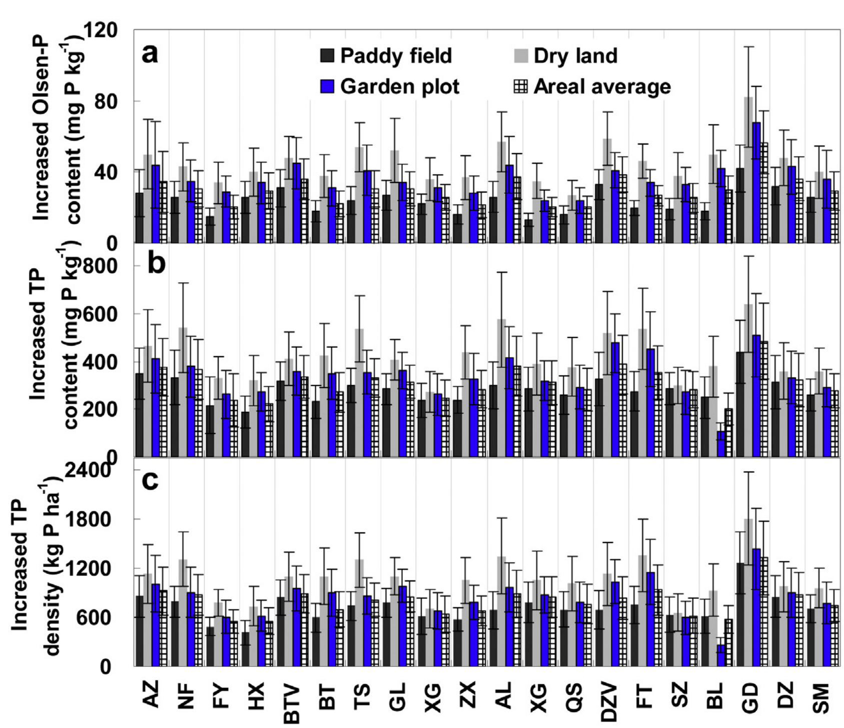

Fig. 6. Increased contents of Olsen-P (a) and TP (b) and increased TP density (c) in the upper 20-cm layer of cropland soils between 1984 and 2009 in 20 townships of the Yongan watershed. Error bar denotes standard deviation.

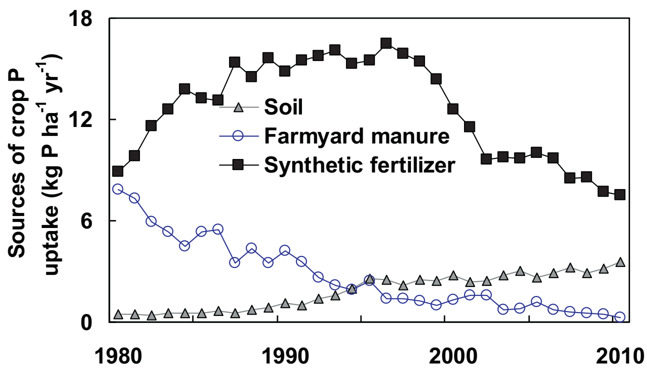

Fig. 7. Historical changes in estimated contributions of annual applied synthetic fertilizer, farmyard manure and soil legacy P to annual crop P uptake for the Yongan watershed over the 1980–2010 study period.

ways and river sediments, and P export by rivers are likely fates for the remaining cumulative cropland P budgets in the watershed.

3.4. Sources apportionment of annual crop P uptake

For each of the 21 townships, there was a close correlation between annual total P input and crop P uptake (r = 0.81–0.94, P < 0.01, n = 31), as well as between annual total P input and total crop yield (r = 0.86–0.97, P < 0.01, n = 31) over the 1980–2010 period. This indicates that application of synthetic fertilizer and farmyard manure P to croplands contributed to crop yields. Spatially, we regressed crop P uptake and synthetic P input to croplands in a given year across the 21 townships from 1980 to 2010 and found a generally linear relationship (R^2 = 0.57–0.80, n = 21, Table 4). For a given year, the slope of the line can be viewed as the average fraction of applied synthetic P taken up by crops for the entire watershed in that year, while the intercept can be viewed as the P uptake by crops from farmyard manure and soil P since other P inputs from atmospheric deposition, seed input and irrigation water are negligible (≤2%, Fig. 2a). To distinguish contributions of soil P from farmyard manure to annual crop P uptake, we further regressed crop P uptake against total P input to croplands in different townships for each year. Again, there was a generally linear relationship between crop P uptake and total P input rate (R^2 = 0.60–0.81, n = 21, Table 4). The regression line slope can be viewed as the average fraction of total P input taken up by crops (i.e., P use efficiency) for the entire watershed in a given year, while the intercept is the average P uptake by crops from soil pools.

Table 4
Spatial relationships between crop P uptake (U, kg P ha^{-1} yr^{-1}) and synthetic P fertilizer (SF, kg P ha^{-1} yr^{-1}) or total P (TI, kg P ha^{-1} yr^{-1}) input rates across 21 townships of the Yongan watershed in each year of the 1980–2010 period.

Years	Relationship between U and SF	Relationship between U and TI
1980	U = 0.50SF + 8.34 (R^2 = 0.78)	U = 0.33TI + 0.47 (R^2 = 0.77)
1981	U = 0.48SF + 7.80 (R^2 = 0.72)	U = 0.31TI + 0.48 (R^2 = 0.80)
1982	U = 0.50SF + 6.33 (R^2 = 0.80)	U = 0.33TI + 0.37 (R^2 = 0.79)
1983	U = 0.47SF + 5.89 (R^2 = 0.73)	U = 0.30TI + 0.54 (R^2 = 0.80)
1984	U = 0.48SF + 5.02 (R^2 = 0.70)	U = 0.29TI + 0.54 (R^2 = 0.81)
1985	U = 0.43SF + 5.87 (R^2 = 0.78)	U = 0.28TI + 0.53 (R^2 = 0.78)
1986	U = 0.41SF + 6.09 (R^2 = 0.64)	U = 0.30TI + 0.64 (R^2 = 0.63)
1987	U = 0.43SF + 4.01 (R^2 = 0.75)	U = 0.28TI + 0.51 (R^2 = 0.74)
1988	U = 0.40SF + 5.09 (R^2 = 0.56)	U = 0.27TI + 0.75 (R^2 = 0.65)
1989	U = 0.37SF + 4.34 (R^2 = 0.77)	U = 0.30TI + 0.47 (R^2 = 0.76)
1990	U = 0.35SF + 5.34 (R^2 = 0.68)	U = 0.25TI + 1.09 (R^2 = 0.67)
1991	U = 0.36SF + 4.53 (R^2 = 0.79)	U = 0.27TI + 1.01 (R^2 = 0.78)
1992	U = 0.37SF + 4.01 (R^2 = 0.80)	U = 0.25TI + 1.36 (R^2 = 0.79)
1993	U = 0.38SF + 3.80 (R^2 = 0.61)	U = 0.27TI + 1.60 (R^2 = 0.60)
1994	U = 0.33SF + 3.92 (R2 = 0.62)	U = 0.26TI + 2.00 (R^2 = 0.61)
1995	U = 0.32SF + 5.06 (R^2 = 0.75)	U = 0.25TI + 2.60 (R^2 = 0.78)
1996	U = 0.37SF + 3.89 (R^2 = 0.74)	U = 0.26TI + 2.52 (R^2 = 0.73)
1997	U = 0.35SF + 3.61 (R^2 = 0.75)	U = 0.25TI + 2.21 (R^2 = 0.74)
1998	U = 0.30SF + 3.77 (R^2 = 0.76)	U = 0.26TI + 2.51 (R^2 = 0.65)
1999	U = 0.31SF + 3.40 (R^2 = 0.71)	U = 0.24TI + 2.44 (R^2 = 0.76)
2000	U = 0.28SF + 4.07 (R^2 = 0.74)	U = 0.22TI + 2.76 (R^2 = 0.71)
2001	U = 0.29SF + 3.97 (R^2 = 0.69)	U = 0.23TI + 2.37 (R^2 = 0.65)
2002	U = 0.30SF + 4.01 (R^2 = 0.70)	U = 0.25TI + 2.46 (R^2 = 0.79)
2003	U = 0.33SF + 3.51 (R^2 = 0.72)	U = 0.27TI + 2.76 (R^2 = 0.67)
2004	U = 0.30SF + 3.84 (R^2 = 0.71)	U = 0.27TI + 3.06 (R^2 = 0.81)
2005	U = 0.31SF + 3.78 (R^2 = 0.63)	U = 0.26TI + 2.61 (R^2 = 0.67)
2006	U = 0.33SF + 3.62 (R^2 = 0.74)	U = 0.28TI + 2.89 (R^2 = 0.71)
2007	U = 0.34SF + 3.79 (R^2 = 0.65)	U = 0.25TI + 3.21 (R^2 = 0.70)
2008	U = 0.31SF + 3.42 (R^2 = 0.66)	U = 0.27TI + 2.87 (R^2 = 0.68)
2009	U = 0.32SF + 3.58 (R^2 = 0.57)	U = 0.27TI + 3.14 (R^2 = 0.64)
2010	U = 0.32SF + 3.81 (R^2 = 0.66)	U = 0.28TI + 3.45 (R^2 = 0.67)

Based on the intercepts from these two regression lines for each year (Table 4), we apportioned the annual average contributions of synthetic fertilizer, farmyard manure and soil P to annual crop P uptake for the entire watershed (Fig. 7). Estimated crop P uptake from applied synthetic fertilizer (i.e., difference between total crop P uptake shown in Fig. 2b and the intercept of synthetic P input-crop P uptake line shown in Table 4) increased from 8.93 kg P ha^{-1} yr^{-1} (52%) in 1980 to 16.45 kg P ha−1 yr−1 (80%) in 1995 before declining to 7.54 kg P ha^{-1} yr^{-1} (66%) in 2010. These results are consistent with the changing trends observed in synthetic P fertilizer application rates over the past 31 years (Fig. 2a). Estimated crop P uptake from annually applied farmyard manure (i.e., difference between the intercepts of the two regressions, Table 4) rapidly and continously decreased from 7.87 kg P ha^{-1} yr^{-1} in 1980 (46%) to 0.27 kg P ha^{-1} yr^{-1} (2%) in 2010. Although annual farmyard manure P input rates were higher than synthetic fertilizer P inputs in 1980–1982 (Fig. 2a), the contribution of synthetic fertilizer P to annual crop P uptake was higher than that of farmyard manure P (Fig. 7), implying that synthetic P fertilizer is more readily available to crops than farmyard manure P (Yan et al., 2013). Estimated crop P uptake from soil P pools increased continuously from 0.47 kg P ha^{-1} yr^{-1} (3%) in 1980 to 3.45 kg P ha^{-1} yr^{-1}(31%) in 2010 (Fig. 7). There were significant increases in soil TP levels in the upper 20 cm layer of cropland soils between 1984 and 2009 (Fig. 6a), implying a net increase in soil P despite crop uptake and removal by harvest. Therefore, the increase of crop P uptake from soils resulted primarily from legacy P inputs from previous years, especially from synthetic fertilizer and farmyard manure.

4. Discussion

4.1. Factors influencing cropland P budgets and use efficiency

Annual cropland P budgets (18.8–99.0 kg P ha^{-1} yr^{-1}) and use efficiency (12–43%) exhibited high spatio-temporal variability across the 21 townships of the Yongan watershed over the past 31 years (Tables 2 and 3). Although these estimates are comparable with results observed in China (i.e., 46–65 kg P ha^{-1} yr^{-1} and 26–45%, Chen et al., 2008; Ma et al., 2012; Hou et al., 2013; Li et al., 2015, 2016; Jiang and Yuan, 2015), the P budgets are much higher and P use efficiencies much lower than results found in USA and Europe (i.e., −3–21 kg P ha^{-1} yr^{-1} and ∼60%, Maguire et al., 2009; Schröer et al., 2011; Ma et al., 2013) in recent decades. Such large spatio-temporal variations might be explained by differences in P fertilization patterns and cropland types. There was a positive correlation observed between the annual P budget and applied farmyard manure P rates (r = 0.77, P < 0.001, n = 651) as well as dryland areas (r = 0.64, P < 0.001, n = 651) across the 21 townships and 31 years. Smallholder farmers in the study area, as well as in other regions of China, usually apply farmyard manure without considering the nutrients contained in the manure and residual soil pools (Miao et al., 2011; Han et al., 2012; Yan et al., 2013). This results in excessive P application when coupled with synthetic fertilizer P additions to croplands. In addition, the majority of synthetic fertilizer and farmyard manure is applied pre-planting or at planting by smallholder farmers in China, while the peak of crop uptake is later in the growing season (Miao et al., 2011). Such a lack of temporal synchronization between crop growth demand and fertilizer application further increases P excess. Compared to paddy field and garden plot crops, dryland crops generally received >40% of P from farmyard manure according to field investigations in the studied watershed, as well as in other regions of China (Chadwick et al., 2015; Yan et al., 2013). In addition, there is relatively lower immediate availability of P from farmyard manure than synthetic fertilizer P (Yan et al., 2013). Thus, there are higher net increases in TP contents and densities in the upper 20 cm layer of dryland soils compared to paddy field and garden plot soils between 1984 and 2009 (Fig. 6 and Table C.1).

In general, increasing P input decreased annual P use efficiency across the 21 townships and 31 years ($r = -0.58$, $P < 0.001$, $n = 651$), consistent with results observed in previous studies for croplands (MacDonald et al., 2011; Nziguheba et al., 2016). However, there was a critical total P input value of ~60 kg P ha^{-1} yr^{-1} influencing P use efficiency (Fig. 5a), which may occur because the crop assimilation capacity for P becomes saturated due to continuous excess P inputs (i.e., crop biomass plateaus when P largely exceeds crop requirement, Simpson et al., 2011; Ouyang et al., 2013). The critical P input value of 60 kg P ha^{-1} yr^{-1} suggested in this study is consistent with the recommended P application rate of 61 kg P ha^{-1} yr^{-1} for soils with Olsen-P levels lower than 7 mg P kg^{-1} for achieving optimal cereal yield in China (Li et al., 2011). In terms of cropland types, a positive correlation was found with paddy field area percentage ($r = 0.67$, $P < 0.001$, $n = 651$) and negative correlations with garden plot area percentage ($r = -0.59$, $P < 0.01$, $n = 651$) and dryland area percentage ($r = -0.42$, $P < 0.01$, $n = 651$). The reduction phase associated with paddy field flooding enhances P availability for crops by converting Fe^{3+}-P minerals to more soluble Fe^{2+}-P minerals in acidic paddy soils, improving P uptake efficiency (Huang et al., 2013). The critical paddy field area of 50% observed for influencing P use efficiency (Fig. 5b) implies that at paddy field area >50%, the paddy fields receive the majority of the total P inputs (>60%) for each township or year and thus largely determines the annual P use efficiency. Due to perennial plants (e.g., fruit trees) cultivated in garden plots, a considerable proportion of annual P inputs to garden plots is used to support plant biomass growth, while only a small proportion is exported as harvested crop products resulting in lower apparent P use efficiency for garden plots (Lu et al., 2016). Previous studies in China also indicated that P use efficiencies for cereal (e.g., rice and wheat) cultivation in paddy fields (25–63% with average of 43%, Zhang et al., 2008; Han et al., 2012; Yuan et al., 2011) is usually higher than those for non-cereal crops (e.g., vegetables and fruits) in dryland and garden plots (8–53% with average of 19%, Li et al., 2011; Han et al., 2012; Yan et al., 2013; Ji et al., 2008).

4.2. Accumulation and role of legacy soil P pools

Corresponding to high P budgets and low P use efficiency, significant increases in Olsen-P (87–720%) and TP (113–395%) levels were observed in the upper 20 cm layer of cropland soils between 1984 and 2009 (Fig. 6a and b). Similar results were found in a national cropland assessment in China with average Olsen-P increasing by 17 mg P kg^{-1} (234%) between 1980 and 2007 (Li et al., 2011). Estimated increases in soil Olsen-P contents resulting from addition of 100 kg P ha^{-1} of synthetic P fertilizer (1.8–3.8 mg P kg^{-1}) agreed well with results from long-term (>15-year) observations in typical croplands across China that found increases in Olsen-P levels of 1.1–5.7 mg P kg^{-1} for each additional 100 kg P ha^{-1} (Tang et al., 2008; Cao et al., 2012; Hua et al., 2016). Compared to paddy fields, dryland and garden soils had larger increases in P levels in general (Fig. 6a and b), which is consistent with the positive correlation observed between P use efficiency and paddy field area percentage as previously discussed. Similarly, a recent *meta*-analysis found average Olsen-P levels increased by 16 mg P kg^{-1} (145%) in cereal cultivated soils and 61 mg P kg^{-1} (1114%) in non-cereal crop cultivated soils in China between 1990 and 2012 (Ma et al., 2016). An estimated areal average of more than 514–1329 kg P ha^{-1} of accumulated legacy soil pools (Fig. 6c) was consistent with the average cumulative cropland soil pool of 160–1115 kg P ha^{-1} observed across six continents between 1965 and 2007 (Sattari et al., 2012), as well as accumulated cropland soil legacy P pools of 504–953 kg P ha^{-1} observed in the China over the past three decades (Hou et al., 2013; Ma et al., 2013; Li et al., 2015; Jiang and Yuan, 2015). Previous P flow analyses conducted in China indicated that cropland soils were the major location for accumulation of legacy P pools as they receive the majority of anthropogenic P inputs (Chen et al., 2008; Ma et al., 2012, 2013; Jiang and Yuan, 2015). As compared to nitrogen, the large accumulation of cropland soil P pools is associated with the lack of a gaseous phase loss and limited mobility through soil profiles due to high adsorption capacity of P to soil particles (Sattari et al., 2012; Sharpley et al., 2013). Considering the large spatial variability in soil P contents, we acknowledge that the estimated net P accumulations of legacy P pools in the upper 20 cm soil layer for each cropland type and township (Fig. 6c) imply a considerable uncertainty due to the limited number of observation points (**Supplementary material B, Table B.1**). Thus, more extensive and rigorous long-term time series data for soil P dynamics in different soil layers are required to improve estimation of net P accumulation in cropland soil profiles.

The large accumulation of legacy P pools (Fig. 6c) in cropland soils has considerable agronomic and environmental implications (Sharpley et al., 2013; Liu et al., 2015; Rowe et al., 2016). Our previous modeling results suggested that the annual P pollution load derived from cropland legacy soil P has rapidly increased from 0.15 to 1.41 kg P ha^{-1} yr^{-1} between 1980 and 2010 in the Yongan watershed (Chen et al., 2016). 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 in many watersheds (Kleinman et al., 2011; Sharpley et al., 2013). This study also suggests that legacy soil P pools have a considerable potential for supporting crop growth compared to synthetic fertilizer and farmyard manure, especially in recent years (Fig. 7). Some long-term field studies indicate that crops can recover the legacy P from long-term over-application of fertilizer and manure P with no reduction in crop yields for several years to decades after cessation of P fertilization (Sharpley et al., 2013; Jarvie et al., 2013; Liu et al., 2015; Rowe et al., 2016). Therefore, enhancing bioavailability and utilization of legacy soil P pools for crop production through adopting biological (e.g., plant breeding and genetic modification to enhance P uptake rates, Condron et al., 2013) and physical (e.g., deep tillage to favor an increased rooting depth, Kleinman et al., 2011) measures are warranted to mitigate depletion of rock P resources and attenuate nonpoint source P pollution.

The statistical approach utilized in this study was based on regression analyses (Table 4) and provides a simple but efficient tool to estimate the contributions of applied synthetic fertilizer P, farmyard manure P and legacy soil P to annual crop P uptake at the watershed scale (Fig. 7). Due to lack of relevant field observations, it is difficult to directly validate the estimated results. However, the estimated changes in annual synthetic fertilizer, farmyard manure and legacy soil P contributions to crop P uptake rates by this statistical approach (Fig. 7) closely matched the trends observed for synthetic fertilizer and farmyard manure P input rates over the

1980–2010 period (Fig. 2a) and were further supported by the significant increase in soil Olsen-P levels between 1984 and 2009 (Fig. 6b). Estimated average P use efficiency from this regresion analysis (22–33%, Table 4) was very close (R^2 = 0.95, relative error ± 5%) to that estimated directly from the ratio between crop P uptake and total P input for the entire watershed (Fig. 2a), further supporting the efficacy of this approach. In the future, more detailed data for crop P uptake and cropland soil P dynamics from long-term comparative field trials would be valuable for validating the results estimated from our statistical approach.

4.3. Implications for cropland P management under smallholder farm systems

Across the 21 townships of the Yongan watershed, there was a 2–3-fold spatial heterogeneity in cropland P budgets and P use efficiency within a given year (Table 2 and Table 3), as well as in legacy P pools (Fig. 6c), highlighting the importance of township-scale cropland P management under smallholder farm systems. In China, however, the current agricultural technical extension system usually determines what technologies should be transferred at the central, provincial or county level without sufficient involvement from township and smallholder farm stakeholders (Miao et al., 2011). As a result, it remains a challenge for adopting specific technologies and sustainable management strategies for improving P use efficiency and enhancing utilization of the legacy soil P pool in many townships. The agricultural extension system should incorporate the spatial heterogeneity across townships for guiding agricultural P management. For example, township GD has the largest cropland P budget, the lowest P use efficiency (Table 3) and the largest legacy soil P pool (Fig. 6c) within the watershed. Thus, reduction of synthetic P fertilizer application rate and enhanced utilization of the legacy soil P pool for crop production should be particularly effective for township GD. Given the high domestic animal population densities (implying high organic waste quantity) in townships GD and NF (Table 1), they should promote recycling of human and animal waste to partially replace synthetic P fertilizer use on croplands, as well as to reduce domestic and animal P loads that are often directly discharged to surface waters (Hou et al., 2013; Chadwick et al., 2015). In terms of cropland types, expanding paddy field cultivation through conversion of dryland and garden plots in townships AL, XG and GD that have low paddy field area percentages (<50%, Table 1) would be beneficial for improving P use efficiency. In the future, it is necessary to improve the township-scale agricultural technical extension system to enhance adoption of relevant technologies and techniques to customized soil P management strategies by individual townships under smallholder farm systems.

5. Conclusion

This study highlights the large spatial heterogeneity in cropland P budgets and P use efficiency across townships and further documents a considerable accumulation of legacy soil P after long-term excessive P addition under smallholder farm management. All 21 townships of the Yongan watershed demonstrated similar temporal trends in P total input, crop uptake and budgets for croplands, as well as P use efficiency in 1980–2010. However, there was 2–3-fold spatial variability among townships within a given year. Spatio-temporal variation in P budgets was mainly related to farmyard manure P application rates as a result of neglecting the P content of the applied farmyard manure, while the P use efficiency was primarily influenced by the total P input rate and paddy field area percentage. Rapid increases of soil P contents observed in the upper 20 cm of cropland soils between 1984 and 2009 imply that more than 53–79% of the cumulative P budget accumulated as legacy sources in cropland soils. Compared to synthetic fertilizer (52–80%) and farmyard manure (2–46%) inputs, legacy soil P contribution to annual crop P uptake rapidly increased from 0.47 kg P ha^{-1} yr^{-1} (3%) in 1980 to 3.45 kg P ha^{-1} yr^{-1} (31%) in 2010. Improved utilization of soil legacy P pools for crop production and increasing P use efficiency are necessary to conserve rock P resources and control nonpoint source P pollution. The high spatial heterogeneity in P budgets and use efficiency across townships, as well as the considerable legacy P pools, should be incorporated in agricultural P management strategies for the smallholder farm systems.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.agee.2016.12.003.

References

Anthony, V.M., Ferroni, M., 2012. Agricultural biotechnology and smallholder farmers in developing countries. Curr. Opin. Biotech. 23, 278–285.
Cao, N., Chen, X., Cui, Z., Zhang, F., 2012. Change in soil available phosphorus in relation to the phosphorus budget in China. Nutr. Cycl. Agroecosys. 94, 161–170.
Chadwick, D., Jia, W., Tong, Y., Yu, G., Shen, Q., Chen, Q., 2015. Improving manure nutrient management towards sustainable agricultural intensification in China. Agric. Ecosyst. Environ. 209, 34–46.
Chen, M., Chen, J., Sun, F., 2008. Agricultural phosphorus flow and its environmental impacts in China. Sci. Total Environ. 405, 140–152.
Chen, D.J., Hu, M.P., Guo, Y., Dahlgren, R.A., 2015. Influence of legacy phosphorus, land use, and climate change on anthropogenic phosphorus inputs and riverine export dynamics. Biogeochemistry 123, 99–116.
Chen, D.J., Hu, M.P., Wang, J.H., Guo, Y., Dahlgren, R.A., 2016. Factors controlling phosphorus export from agricultural/forest and residential systems to rivers in eastern China, 1980–2011. J. Hydrol. 533, 53–61.
Condron, L.M., Spears, B.M., Haygarth, P.M., Turner, B.L., Richardson, A.E., 2013. Role of legacy phosphorus in improving global phosphorus-use efficiency. Environ. Dev. 8, 147–148.
Haith, D.A., 2003. Systems analysis, TMDLs and watershed approach. J. Water Resour. Plan. Manage. 129, 257–260.
Han, Y., Li, H.P., Nie, X.F., Xu, X.B., 2012. Nitrogen and phosphorus budget of different land use types in hilly area of Lake Taihu upper-river basin. J. Lake Sci. 24, 829–837 (in Chinese with English abstract).
Haygarth, P.M., Jarvie, H.P., Powers, S.M., Sharpley, A.N., Elser, J.J., Shen, J.B., Peterson, H.M., Chan, N.I., Howden, N.J.K., Burt, T., Worrall, F., Zhang, F.S., Liu, X.J., 2014. Sustainable phosphorus management and the need for a long-term perspective: the legacy hypothesis. Environ. Sci. Technol. 48, 8417–8419.
Hou, Y., Ma, L., Gao, Z., Wang, F., Sims, J., Ma, W., Zhang, F., 2013. The driving forces for nitrogen and phosphorus flows in the food chain of China, 1980 to 2010. J. Environ. Qual. 42, 962–971.
Hua, K.K., Zhang, W.J., Guo, Z.B., Wang, D.Z., Oenema, O., 2016. Evaluating crop response and environmental impact of the accumulation of phosphorus due to long-term manuring of vertisol soil in northern China. Agric. Ecosyst. Environ. 219, 101–110.
Huang, L., Zhang, G., Thompson, A., Rossiter, D.G., 2013. Pedogenic transformation of phosphorus during paddy soil development on calcareous and acid parent materials. Soil Sci. Soc. Am. J. 77, 2078–2088.
Ji, H.J., Zhang, R.L., Wu, S.X., Zhang, H.Z., Zhang, W.L., 2008. Analysis of fertilizer input and nutrient balance of farmland in Taihu watershed. Soil Fert. Sci. China 5, 70–75 (in Chinese with English abstract).
Jiang, S.Y., Yuan, Z.W., 2015. Phosphorus flow pattern in the Chaohu watershed from 1978 to 2012. Environ. Sci. Technol. 49, 1397–3982.
Kleinman, P.J.A., Sharpley, A.N., McDowell, R.W., Flaten, D.N., Buda, A.R., Liang, T., Bergström, L., Zhu, Q., 2011. Managing agricultural phosphorus for water quality protection: principles for progress. Plant Soil 349, 169–182.
Li, H., Huang, G., Meng, Q., Ma, L., Yuan, L., Wang, F., Zhang, W., Cui, Z., Shen, J., Chen, X., Jiang, R., Zhang, F., 2011. Integrated soil and plant phosphorus management for crop and environment in China. A review. Plant Soil 349, 157–167.
Li, H., Liu, J., Li, G., Shen, J., Bergström, L., Zhang, F., 2015. Past, present and future use of phosphorus in Chinese agriculture and its influence on phosphorus losses. AMBIO 44, 274–285.

Li, G.H., van Ittersum, M.K., Leffelaar, P.A., Sattari, S.Z., Li, H.G., Huang, G.Q., Zhang, F.S., 2016. A multi-level analysis of China's phosphorus flows to identify options for improved management in agriculture. Agr. Syst. 144, 87–100.
Liu, J., Hu, Y., Yang, J., Abdi, D., Cade-Menun, B.J., 2015. Investigation of soil legacy phosphorus transformation in long-term agricultural fields using sequential fractionation, P K-edge XANES and solution P NMR spectroscopy. Environ. Sci. Technol. 49, 168–176.
Liu, X., Sheng, H., Jiang, S.Y., Yuan, Z.W., Zhang, C.S., Elser, J.J., 2016. Intensification of phosphorus cycling in China since the 1600. Proc. Natl Acad. Sci. U. S. A. 113, 2609–2614.
Lu, Y.L., Chen, Z.J., Kang, T.T., Zhang, X.J., Bellarby, J., Zhou, J.B., 2016. Land-use changes from arable crop to kiwi-orchard increased nutrient surpluses and accumulation in soils. Agric. Ecosyst. Environ. 223, 270–277.
Ma, L., Velthof, G., Wang, F., Qin, W., Zhang, W., Liu, Z., Zhang, Y., Wei, J., Lesschen, J., Ma, W., 2012. Nitrogen and phosphorus use efficiencies and losses in the food chain in China at regional scales in 1980 and 2005. Sci. Total Environ. 434, 51–61.
Ma, D.C., Hu, S.Y., Chen, D.J., Li, Y.R., 2013. The temporal evolution of anthropogenic phosphorus consumption in China and its environmental implications. J. Ind. Ecol. 17, 566–577.
Ma, J.C., He, P., Xu, X.P., He, W.T., Liu, Y.X., Yang, F.Q., Chen, F., Li, S.T., Tu, S.H., Jin, J.Y., Johnston, A.M., Zhou, W., 2016. Temporal and spatial changes in soil available phosphorus in China (1990–2012). Field Crops Res. 192, 13–20.
MacDonald, G.K., Bennett, E.M., Potter, P.A., Ramankutty, N., 2011. Agronomic phosphorus imbalances across the world's croplands. Proc. Natl Acad. Sci. U. S. A. 108, 3086–3091.
Maguire, R.O., Rubæk, G.H., Haggard, B.E., Foy, B.H., 2009. Critical evaluation of the implementation of mitigation options for phosphorus from field to catchment scales. J. Environ. Qual. 38, 1989–1997.
Miao, Y.X., Stewart, B.A., Zhang, F.S., 2011. Long-term experiments for sustainable nutrient management in China. A review. Agron. Sustain. Dev. 31, 397–414.
Nziguheba, G., Zingore, S., Kihara, J., Merckx, R., Njoroge, S., Otinga, A., Vandamme, E., Vanlauwe, B., 2016. Phosphorus in smallholder farming systems of sub-Sahara Africa: implications for agricultural intensification. Nutr. Cycl. Agroecosyst. 104, 321–340.
Oenema, O., Kros, H., de Vries, W., 2003. Approaches and uncertainties in nutrient budgets: implications for nutrient management and environmental policies. Eur. J. Agron. 20, 3–16.
Ouyang, W., Wei, X.F., Hao, F.H., 2013. Long-term soil nutrient dynamics comparison under smallholding land and farmland policy in Northeast of China. Sci. Total Environ. 450 (-451), 129–139.
Powers, S.M., Bruulsema, T.W., Burt, T.P., Chan, N.I., Haygarth, P.M., Howden, N.J.K., Jarvie, H.P., Lyu, Y., Peterson, H.D., Sharpley, A.N., She, J.B., Worrall, F., Zhang, F.S., 2016. Long-term accumulation and transport of anthropogenic phosphorus in three river basins. Nature Geosci. 29, 353–356.
Rowe, H., Withers, P.J.A., Baas, P., Chan, N.I., Doody, D., Holiman, J., Jacobs, B., Li, H.G., MacDonald, G.K., McDowell, R., Sharpley, A.N., Shen, J.B., Taheri, W., Wallenstein, M., Weintraub, M.N., 2016. Integrating legacy soil phosphorus into sustainable nutrient management strategies for future food, bioenergy and water security. Nutr. Cycl. Agroecosys. 104, 393–412.
Sattari, S.Z., Bouwman, A.F., Giller, K.E., van Ittersum, M.K., 2012. Residual soil phosphorus as the missing piece in the global phosphorus crisis puzzle. Proc. Natl. Acad. Sci. 109, 6348–6353.
Schröer, J.J., Smit, A.L., Cordell, D., Rosemarin, A., 2011. Improved phosphorus use efficiency in agriculture: a key requirement for its sustainable use. Chemosphere 84, 822–831.
Sharpley, A.N., Jarvie, H.P., Buda, A., May, L., Spears, B., Kleinman, P., 2013. Phosphorus legacy: overcoming the effects of past management practices to mitigate future water quality impairment. J. Environ. Qual. 42, 1308–1326.
Sharpley, A.N., Bergström, L., Aronsson, H., Bechmann, M., Bolster, C.H., Börling, K., Djodjic, F., Jarvie, H.P., Schoumans, O.F., Stamm, C., Tonderski, K.S., Ulén, B., Uusitalo, R., Withers, P.J., 2015. Future agriculture with minimized phosphorus losses to waters: research needs and direction. AMBIO 44, 163–179.
Shen, R.P., Sun, B., Zhao, Q.G., 2005. Spatial and temporal variability of N, P and K balances for agroecosystems in China. Pedosphere 15, 347–355.
Simpson, R.J., Oberson, A., Culvenor, R.A., Ryan, M.H., Veneklaas, E.J., Lambers, H., Lynch, J.P., Ryan, P.R., Delhaize, E., Smith, F.A., Smith, S.E., Harvey, P.R., Richardson, A.E., 2011. Strategies and agronomic interventions to improve the phosphorus-use efficiency of farming systems. Plant Soil 349, 89–120.
Tang, X., Li, J.M., Ma, Y.B., Hao, X.Y., Li, X.Y., 2008. Phosphorus efficiency in long-term (15 years) wheat-maize cropping systems with various soil and climate conditions. Field Crops Res. 108, 231–237.
Wang, X.L., Feng, A.P., Wang, Q., Wu, C.Q., Liu, Z., Ma, Z.S., Wei, X.F., 2014. Spatial variability of the nutrient balance and related NPS P risk analysis for agro-ecosystems in China in 2010. Agric. Ecosyst. Environ. 193, 42–52.
Yan, X.Y., Ohara, T., Akimoto, H., 2006. Bottom-up estimate of biomass burning in mainland China. Atmos. Environ. 40, 5262–5273.
Yan, Z.J., Liu, P.P., Li, Y.H., Ma, L., Alva, A., Dou, Z.X., Chen, Q., Zhang, F.S., 2013. Phosphorus in China's intensive vegetable production systems: overfertilization, soil enrichment, and environmental implications. J. Environ. Qual. 42, 982–989.
Yuan, Z.W., Liu, X., Wu, H.J., Zhang, L., Bi, J., 2011. Anthropogenic phosphorus flow analysis of Lujiang county, Anhui Province, central China. Ecol. Model. 222, 1534–1543.
Zhang, F.S., Wang, J.Q., Zhang, W.F., Cui, Z.L., Ma, W.Q., Chen, X.P., Jiang, R.F., 2008. Nutrient use efficiencies of major cereal crops in China and measures for improvement. Acta Pedolog. Sin. 45, 915–924 (in Chinese with English abstract).
Zhang, W.S., Swaney, D.P., Hong, B., Howarth, R.W., Han, H., Li, X.Y., 2015. Net anthropogenic phosphorus inputs and riverine phosphorus fluxes in highly populated headwater watersheds in China. Biogeochemistry 126, 269–283.