Science of the Total Environment 805 (2022) 150441

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Leached phosphorus apportionment and future management strategies across the main soil areas and cropping system types in northern China

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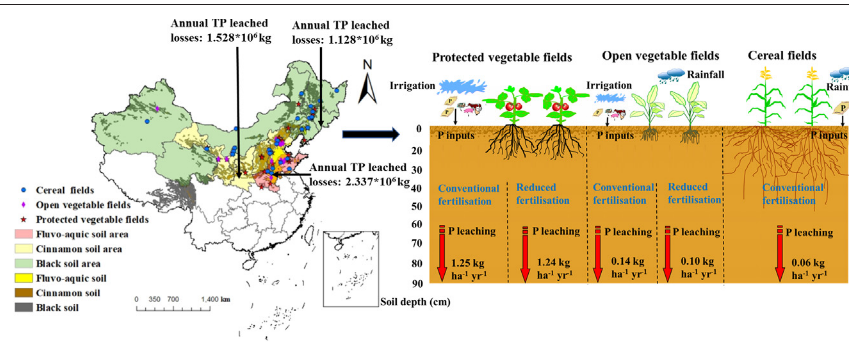
HIGHLIGHTS

- Annual total phosphorus (TP) leached losses were 4.99×10^6 kg in northern China.
- Protected vegetable fields (PVFs) contributed to 48.5% of TP leached losses.
- The percentage of TP leached losses as a fraction of P inputs was 0.3% in PVFs.
- The fluvo-aquic and cinnamon soil areas combined accounted for 77.5% of the TP leached losses.
- A reduction of 10–30% of P input was feasible in vegetable cropping systems.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 13 July 2021
Received in revised form 15 September 2021
Accepted 15 September 2021
Available online xxxx

Editor: Ouyang Wei

Keywords:
Northern China
TP leached losses
Soil areas
Cropping system types
P management strategies

ABSTRACT

Excess phosphorus (P) leached from high fertiliser input cropping systems in northern China is having detrimental effects on water quality. Before improved management can be directed at specific soils and cropping system types estimates of P leached loss apportionment and mitigation potentials across the main soil (fluvo-aquic soil, FAS; cinnamon soil, CS; black soil, BS) areas and cropping systems (protected vegetable fields, PVFs; open vegetable fields, OVFs; cereal fields, CFs) are needed. The present study designed and implemented conventional fertilisation and low input system trials at 75 sites inclusive of these main soils and cropping system types in northern China. At all sites, a uniform lysimeter design (to 0.9 m depth) enabled the collection and analysis of leachate samples from 7578 individual events between 2008 and 2018. In addition, site-specific static and dynamic activity data were recorded. Results showed that annual total phosphorus (TP) leached losses across the main soil areas and cropping systems were 4.99×10^6 kg in northern China. A major finding was PVFs contributed to 48.5% of the TP leached losses but only accounted for 5.7% of the total cropping areas. The CFs and OVFs accounted for 40.3% and 11.2% of the TP leached losses, respectively. Across northern China, the TP leached losses in PVFs and OVFs were greatest in FAS areas followed by CS and BS areas. The higher TP leached losses in FAS areas were closely correlated with greater P fertiliser inputs and irrigation practices. From a management perspective in PVFs and OVFs systems, a decrease of P inputs by 10–30% would not negatively affect yields while protecting water quality. The present study highlights the importance of decreasing P inputs in PVFs and OVFs and supporting soil P nutrient advocacy for farmers in China.

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https://doi.org/10.1016/j.scitotenv.2021.150441

1. Introduction

Phosphorus (P) is an essential element for food production and living organisms (Withers et al., 2014). Improvements with respect to P inputs from inorganic and organic fertilisers have led to increasing crop productivity in arable lands globally (Li et al., 2015), but excessive fertilisation has also enhanced soil P accumulation and corresponding P losses along surface and subsurface ways. This situation is more concentrated in China as around one-third of global P fertilisers were consumed in China during the past decade (FAO, 2019; Jin et al., 2020; MacDonald et al., 2011), resulting in serious problems of aquatic eutrophication in inland and coastal waters (Xia et al., 2020). For example, a study by Liu et al. (2018) showed an increase in riverine export total P from 58 Gg P yr^{-1} to 381 Gg P yr^{-1} along the Yangtze River basin to the East China Sea and Yellow Sea during 2004–2010. This increased P loss is one of the reasons for the rapid increase of harmful algal blooms. Focusing on China, some studies have estimated and predicted past and future P losses from agricultural systems using long term experimental data and through modelling (Hou et al., 2018; Hu et al., 2020; Liang et al., 2020; Powers et al., 2016). For example results from the first China Pollution Source Census (CPSC) showed that the P losses from crop systems in 2007 were 1.1×10^8 kg, with the net P losses highest (0.03–0.31 kg P ha^{-1}) in the Huang-huai-hai plain and lowest (0–0.05 kg P ha^{-1}) in the Northeast China Plain (Huang et al., 2017). The second CSPS included data from 2017 to 2020 and showed P losses from crop systems decreased to 0.76×10^8 kg. Both of these studies showed that the dominant source of P losses was from agriculture. However, the CPSC database does not differentiate between surface and subsurface P load losses or apportion losses to particular soil or cropping system types. This makes it difficult to concentrate management and resources on specific areas to minimise future P losses to groundwater (i.e. leached losses of P) with subsequent positive effects on surface waters.

In the literature P losses along surface pathways have received more attention and this bias is evident within studies conducted in China (Lian et al., 2020; McDowell et al., 2020; Wu et al., 2020). For example, in a meta-analysis (151 studies across 13 Chinese provinces), Wang et al. (2019) estimated P runoff losses from vegetable systems was 3.45 kg ha^{-1}. However, until the present study equivalent estimates in the leached pathway are not possible due to a sparsity of data (Han et al., 2016). Recent studies by Yan et al. (2016) and Fei et al. (2019) identify the leached pathway as the main loss pathway for P in vegetable cropping systems of northern China. In fields that received 10 years of P inputs from 260 to 478 kg P ha^{-1} yr^{-1}, annual TP leached losses at 0.9 m ranged from 1.38 to 2.03 kg P ha^{-1}. Such losses recharge to the underlying groundwater body quickly within land areas such as Tianjin municipality in northern China due to shallow water tables (0.7 to 4 m below ground level) (Pan et al., 2017). In 2015, China's Central government set ambitious targets to improve subsurface water quality over the coming decades (Han et al., 2016). Herein, estimates of P leached loss apportionment across different soil areas and cropping system types at a regional scale are needed.

Therefore, the present study collated data from 78 field leaching experiments conducted across the main soil areas and cropping systems of northern China carried out from 2008 to 2018. The main objectives were to (1) quantify and apportion annual TP leached losses to these main soil areas and cropping systems, (2) identify the factors of importance that create differences between soil and cropping systems, and (3) estimate the potential to mitigate P losses in these systems. Objectives 2 and 3 will help future management of these systems and guide future leached losses.

2. Materials and methods

2.1. Field site selection

To select and identify the sites to install the lysimeter monitoring equipment several soil types and cropping systems were targeted where annual TP leached losses are known to be problematic. Three soil types which widely distributed in northern China were selected: (a) fluvo-aquic soil, FAS, (b) cinnamon soil, CS and (c) black soil, BS. The main areas in northern China that have such soils are as follows: (a) FAS areas - Shandong and Henan provinces, (b) CS areas - Beijing and Tianjin municipalities and Hebei, Shanxi, Shaanxi, Ningxia and Gansu provinces and (c) BS areas - Heilongjiang, Jilin, Liaoning, Inner Mongolia, Xinjiang and Qinghai provinces (Fig. 1).

Three cropping systems were selected as follows: (a) protected vegetable fields system, (b) open vegetable fields system and (c) cereal fields system. Usually, mono-cropping cereal system (e.g. maize or wheat) are predominant in BS areas, while multi-cropping cereal system (e.g. wheat-maize rotation) are prevailing in CS and FAS areas. In addition, vegetable cropping systems including protected vegetable fields and open vegetable fields are also indispensable in China. The total cropping area of open vegetable fields and protected vegetable fields in China were 22.6 million ha and 5.35 million ha, respectively (NCBS, 2017). Generally, the crops planted in the protected vegetable fields and open vegetable fields are similar, but the multiple crop index (the ratio of annual planting areas to arable land areas), irrigation water and fertiliser inputs are significant higher in the protected vegetable fields (Wang et al., 2019).

2.2. Field site experimental design

Sites ($n = 78$) corresponding with the main soil and cropping systems identified for TP annual apportionment were now selected and plots were delineated for various treatments. Further details pertaining to each of the sites including location, land use and plot area are available in Table S1. Across 75 sites (Table S2), two fertiliser (control (CK) and reduced (RS)) input systems existed. Other on-site activity data such as fertiliser inputs (N and P), planting regime, crop yields and rainfall and irrigation amounts were recorded. Soil physicochemical properties inclusive of soil texture, bulk density, organic matter, total P, soil Olsen-P and pH were analysed after crops harvested every year (Table 1). Generally, the BS areas have relatively greater soil organic matter while lower soil pH values.

Fig. S1 outlines the design of the lysimeter monitoring system. Treatment plots at each location were identified and lysimeters installed as follows: soil pits for installing leaching disks were excavated outside each plot and rectangular-shaped PVC leaching disks, 0.2 m^2 or 1 m^2 were installed tightly into the prepared soil space. Leachate at 0.9 m collected after 48 h of irrigation or natural rainfall was transferred to a sampling bottle using a vacuum pump. Next, the total volume of the leachate was determined and a 100 mL sample transported to the laboratory for further analysis. The TP concentration in the leachate were analysed after autoclave persulfate digestion (0.9 mol L^{-1} H_2SO_4 and 1 g $K_2S_2O_8$) and measured by colorimetric analysis (Murphy and Riley, 1962). Another sample of leachate was filtered through 0.45 μm filter membrane to measure the total dissolved P (Murphy and Riley, 1962).

2.3. Estimation of TP leached losses

In summary, 33, 16 and 29 experimental sites were used to estimate the annual TP leached losses in protected vegetable fields, open vegetable fields, and cereal fields (Table S1). During the period of 2008–2018, 708, 640 and 330 TP leaching events occurred in the FAS areas, CS areas and BS areas in the protected vegetable fields system; 721, 389 and 93 TP leaching events occurred in the FAS areas, CS areas and BS areas in the open vegetable fields system; 226, 294 and 388 TP leaching events occurred in the FAS areas, CS areas and BS areas in the cereal fields system.

The total leaching volume, P leached losses and P losses coefficient was calculated according to the following equation (Li et al., 2021):

$$\text{Leaching volume} = \textit{water depth} \times \textit{leaching disk area} \quad (1)$$

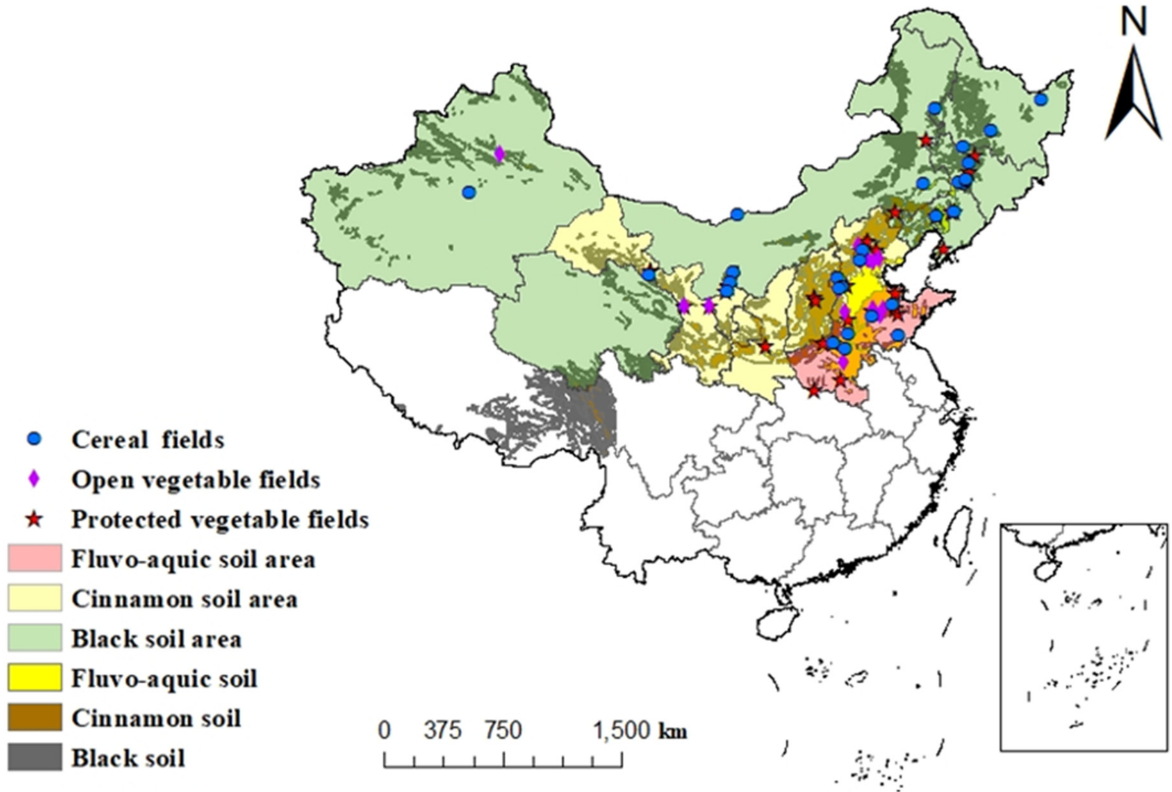

Fig. 1. Geographical distribution of the 78 monitoring sites (33 sites, 16 sites and 29 sites in PVFs, OVFs and CFs, respectively) used to estimate the TP leached losses in northern China. Fluvo-aquic soil areas include the Shandong and Henan provinces; Cinnamon soil areas include the Beijing municipality, Tianjin municipality, Hebei, Shanxi, Shaanxi, Ningxia and Gansu provinces; Black soil areas include the Heilongjiang, Jilin, Liaoning, Inner Mongolia, Xinjiang and Qinghai provinces. Fluvo-aquic, cinnamon and black soil are clarified based on the Chinese soil taxonomy.

$$\text{P leached losses} = P\ concentration \times leaching\ volume \quad (2)$$

$$\text{P losses coefficient} = \frac{\text{P leached losses}}{\text{P inputs}} \times 100\% \quad (3)$$

The amounts of annual TP leached losses (kg ha^{-1} yr^{-1}) at each site was evaluated using the following equation:

$$TPY = \overline{TP} \times N \quad (4)$$

where $\overline{TP}$ represents the average TP leached losses (kg ha^{-1}) at each site during one year, while N represents the total numbers of TP leaching events.

The amounts of TP leached losses in the protected vegetable fields, open vegetable fields and cereal fields across the main soil areas in northern China were calculated use the following equation:

$$TP_x = A_x \times \overline{TPY_x} \quad (5)$$

where A represent the cropping areas. $\overline{TPY}$ represents the annual TP leached losses (kg ha^{-1} yr^{-1}) from 2008 to 2018. x denotes the protected vegetable fields, open vegetable fields and cereal fields in FAS areas, CS areas and BS areas respectively.

The cropping area of the protected vegetable fields, open vegetable fields and cereal fields in the FAS areas, CS areas and BS areas used the

Table 1
Physicochemical properties (mean ± standard deviation) of the experimental soils (0–20 cm) under conventional fertilisation of farmers' practice from 2008 to 2018.

Cropping systems/soil areas[1]	Soil types	Soil texture	Bulk density (g cm$^{3-1}$)	Organic matter (g kg^{-1})	Total P (g kg^{-1})	Olsen-P (mg kg^{-1})	pH
PVFs/soil areas							
FAS areas	Luvisols	Sandy loam, Loam	1.33 ± 0.2a[2]	22.48 ± 5.4b	1.73 ± 0.8a	141.7 ± 81.9a	7.03 ± 1.3b
CS areas	Luvisols, Cambisol	Sandy loam, Loam	1.31 ± 0.1a	26.82 ± 15.2a	1.72 ± 0.8a	129.6 ± 94.7b	8.00 ± 0.6a
BS areas	Luvic Phaeozem	Clay loam	1.33 ± 0.1a	28.67 ± 8.6a	1.70 ± 0.7a	115.0 ± 88.0c	7.03 ± 1.0b
OVFs/soil areas							
FAS areas	Luvisols	Loam, Clay	1.42 ± 0.1a	15.31 ± 4.5a	0.94 ± 0.6a	28.31 ± 24.5b	8.31 ± 0.4a
CS areas	Luvisols, Cambisol	Sandy loam, Loam	1.23 ± 0.1b	17.14 ± 4.7a	1.05 ± 0.4a	64.60 ± 50.0a	8.25 ± 0.4a
BS areas	Podzol	Loam	1.31 ± 0.1b	13.77 ± 7.8a	0.78 ± 0.4b	35.89 ± 69.1b	8.31 ± 0.7a
CFs/soil areas							
FAS areas	Luvisols	Loam, Clay	1.24 ± 0.1b	17.19 ± 3.8b	0.93 ± 0.4a	27.80 ± 17.1b	8.11 ± 0.3a
CS areas	Luvisols, Cambisol	Sandy loam, Loam	1.35 ± 0.1a	16.80 ± 3.7b	0.91 ± 0.3a	20.17 ± 13.6b	8.08 ± 0.4a
BS areas	Luvic Phaeozem	Clay loam	1.32 ± 0.2a	26.63 ± 15.2a	0.80 ± 0.3b	55.71 ± 37a	6.40 ± 1.0b

[1] FAS areas: Fluvo-aquic soil areas include the Shandong and Henan provinces; CS areas: Cinnamon soil areas include the Beijing municipality, Tianjin municipality, Hebei, Shanxi, Shaanxi, Ningxia and Gansu provinces; BS areas: Black soil areas include the Heilongjiang, Jilin, Liaoning, Inner Mongolia, Xinjiang and Qinghai provinces.
[2] The small letters followed the standard deviation means significant difference ($p < 0.05$) among the different soil areas in PVFs, OVFs and CFs respectively.

dataset 2016 from the China Statistical Yearbook (NCBS, 2017). The total TP leached losses in northern China were estimated by the sum of the TP leached losses from the FAS, CS and BS areas.

2.4. Chemical analysis

Soil pH was measured using a pH meter (MP522 version 3, SANXIN, China) with 1: 2.5 ratio of soil to deionised water. Soil organic matter was determined using the potassium dichromate oxidation method (Skjemstad and Baldock, 2007). Soil TP was extracted using a 1.000 g air dried soil with H_2SO_4-$HClO_4$ digestion (Thomas et al., 1967) and then determined by continuous-flow analyser (AA3, BRAN+LUEBBE, Germany). Soil Olsen-P was extracted using a 2.5 g sample shaken with 50 mL 0.5 M $NaHCO_3$ (buffered at pH 8.5 with NaOH) for 30 min (Olsen, 1954) followed by colorimetric analysis (AA3, BRAN+LUEBBE, Germany) (Murphy and Riley, 1962).

2.5. Statistical analysis

Microsoft Excel 2016 was used to analyse the statistical data and Origin 2018 was used to create figures. Phosphorus inputs, manure P inputs, chemical P inputs and sum of precipitation and irrigation inputs which have significant effects ($P < 0.05$) on the variation of TP leached losses were subjected to ANOVA analysis of the *adonis* function (vegan package, R 3.6.3) and the parameters that significantly explained the variance in TP leached losses were calculated.

3. Results

3.1. Apportionment of P losses to main soil areas and cropping systems

The annual TP leached losses from 2008 to 2018 was 4.99×10^6 kg in northern China (Table 2). The annual P leaching from protected vegetable fields, open vegetable fields and cereal fields accounted for 48.5%, 11.2% and 40.3% of the TP leached losses. A major finding was the cropping areas of the protected vegetable fields in northern China was only 1.846×10^6 ha, which accounted for 5.7% of the total cropping areas, while the annual TP leached losses in this system was 2.422×10^6 kg (Table 2). The annual TP leached losses of 2.011×10^6 kg were estimated in the total cropping areas of 2.78×10^7 ha of the cereal fields in northern China. It was calculated that the P losses coefficient (percentage of TP leached losses as a fraction of P inputs) were 0.3%, 0.07% and 0.1% in the protected vegetable fields, open vegetable fields and cereal fields respectively (Table 2).

The cropping areas in FAS, CS and BS areas were responsible of 29.7%, 26.0% and 44.3% of the total cropping areas. However, the TP leached losses from FAS area, CS areas and BS areas accounted for 46.8%, 30.6% and 22.6% of the TP leached losses in northern China (Table 2).

3.2. Phosphorus leaching characteristics across soil areas and cropping systems

During 2008–2018, the average TP concentrations in the leachate changed from 0.14–0.23 mg L^{-1} in the cereal fields and 0.26–0.28 mg L^{-1} in the open vegetable fields. In the protected vegetable fields, the TP concentrations increased to 0.69–1.1 mg L^{-1}, which was 5–6 folds higher than those in the open vegetable fields and cereal fields (Fig. 2). In addition, results showed that the total dissolved P was the dominant P leaching form, which accounted for 63.4% to 72.3% in these cropping systems.

The average TP leached losses in the FAS, CS and BS areas from 2008 to 2018 were 0.251, 0.131 and 0.101 kg ha^{-1} in the protected vegetable fields system, respectively (Fig. 3). In the open vegetable fields and cereal fields systems, the average TP leached losses in the FAS, CS and BS areas were 0.016 and 0.018, 0.038 and 0.028, 0.011 and 0.013 kg ha^{-1}. Additionally, the numbers of TP leaching in the FAS areas was the highest in the vegetable systems (Fig. 3a'-b'), which inextricably linked with the highest irrigation inputs (Table 2). It was noteworthy that average TP leached losses in the open vegetable fields and cereal fields was close, while the TP leaching events were greater in the open vegetable fields (Fig. 3b'-c'). In summary, the annual TP leached losses in the protected vegetable fields systems were 2.51, 0.895 and 0.589 kg ha^{-1} yr^{-1} in the FAS, CS and BS areas (Fig. 3a'). In the open vegetable fields system, the annual TP leached losses were 0.252, 0.179 and 0.072 kg ha^{-1} yr^{-1} in the FAS, CS and BS areas (Fig. 3b'). In the cereal fields system, the annual TP leached losses were 0.074, 0.098 and 0.053 kg ha^{-1} yr^{-1} respectively in the FAS, CS and BS areas (Fig. 3c').

In the protected vegetable fields system, the trend of TP leached losses decreased from 2008 to 2018, with the decreasing rate highest in the CS areas and lowest in the BS areas (Fig. 4). In the open vegetable fields system, it was also found that TP leached losses decreased in the CS areas while they maintained their levels in the FAS and BS areas. In the cereal fields system, the TP leached losses increased slightly in the FAS and BS areas (Fig. 4).

Table 2
Annual TP leached losses from the fluvo-aquic, cinnamon and black soil areas of PVFs (protected vegetable fields), OVFs (open vegetable fields) and CFs (cropland fields) under conventional fertilisation of farmers' practice from 2008 to 2018 in northern China.

Cropping systems/soil areas[2]	Crop planting Areas (ha)	P inputs (kg ha^{-1} yr^{-1})			Irrigation inputs and precipitation[3] (mm)	TP leached loss (kg ha^{-1} yr^{-1})	Annual TP leached losses (10^6 kg)
		Total P	Chemical P	Manure P			
PVFs/soil areas [3]							
FAS areas	573411[1]	608±285a[4]	300 ± 274a	308 ± 248a	870 ± 681a	2.510 ± 2.876a	1.439a
CS areas	761,752	417 ± 441b	203 ± 160b	214 ± 452a	510 ± 335b	0.895 ± 1.908b	0.682b
BS areas	510,601	241 ± 282c	193 ± 267b	48 ± 74b	397 ± 291c	0.589 ± 0.849c	0.301c
OVFs/soil areas							
FAS areas	1,255,784	304 ± 77a	265 ± 72a	39 ± 42a	841 ± 424a	0.252 ± 0.346a	0.316a
CS areas	1,089,236	215 ± 176a	160 ± 137b	55 ± 104a	580 ± 296b	0.179 ± 0.413a	0.195b
BS areas	676,796	187 ± 81b	145 ± 13b	42 ± 24a	705 ± 170a	0.072 ± 0.131b	0.049c
CFs/soil areas							
FAS areas	7,863,765	76 ± 29a	76 ± 29a	0	602 ± 158a	0.074 ± 0.062a	0.582b
CS areas	6,640,786	76 ± 18a	76 ± 18a	0	505 ± 232a	0.098 ± 0.103a	0.651b
BS areas	13,247,228	51 ± 26a	51 ± 26a	0	476 ± 297a	0.059 ± 0.074a	0.778a

[1] The crop planting area in 2016 was sourced from the China Statistical Yearbook (2017).
[2] FAS areas: Fluvo-aquic soil areas include the Shandong and Henan provinces; CS areas: Cinnamon soil areas include the Beijing municipality, Tianjin municipality, Hebei, Shanxi, Shaanxi, Ningxia and Gansu provinces; BS areas: Black soil areas include the Heilongjiang, Jilin, Liaoning, Inner Mongolia, Xinjiang and Qinghai provinces.
[3] PVFs systems only have irrigation inputs as PVFs are covered by polyethylene foil.
[4] The small letters followed the standard deviation means significant difference ($p < 0.05$) among the different soil areas in PVFs, OVFs and CFs respectively.

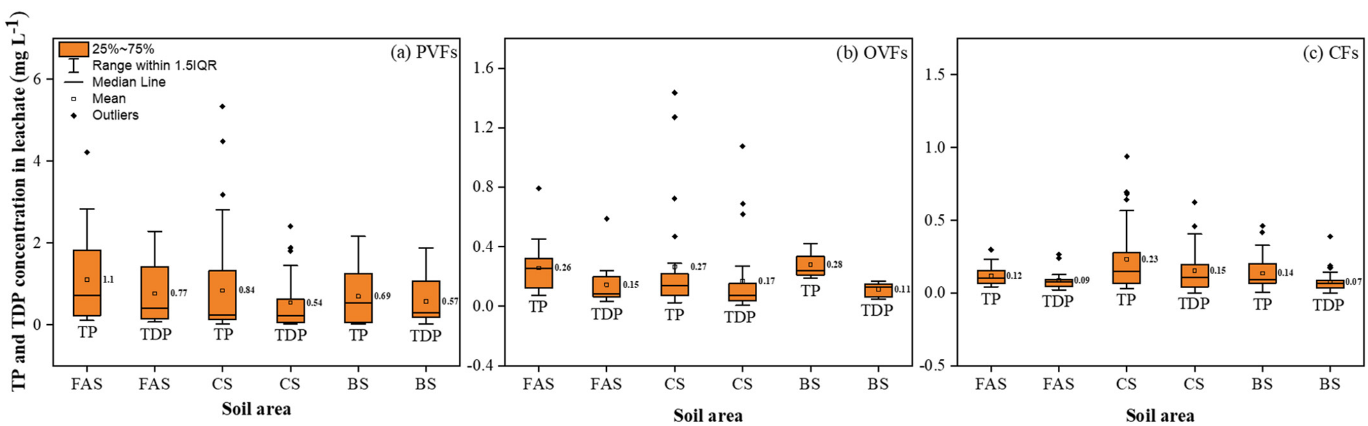

Fig. 2. Total phosphorus (TP) and total dissolved phosphorus (TDP) concentration (mg L^{-1}) in leachate, in PVFs (protected vegetable fields), OVFs (open vegetable fields) and CFs (cereal fields) of the fluvo-aquic, cinnamon and black soil areas from 2008 to 2018 in northern China.

3.3. Variation of TP leached losses with varying P inputs

Across 75 sites (Table S2), the TP leached losses from two fertiliser (control (CK) and reduced (RS)) input systems were compared. Results showed that in the protected vegetable fields, a decrease of TP inputs by 28% merely resulted in a 0.8% decline of TP leached losses (Fig. 5). Considering the soil areas and cropping systems, a decrease of TP inputs by 25% and 30% in the FAS and CS areas only resulted in a 3% and 2% decrease of TP leached losses in the protected vegetable fields. Interestingly, the crop yields increased by 1.8%. However, in the open vegetable fields, a decrease of P inputs by 29% led to a reduction of TP leached losses by 27% (Fig. 5). The detailed results were a decrease of P inputs by 29% and 29.5% in the FAS and CS area led to a reduction of TP leached losses by 21% and 28%. In the cereal fields, the correlation of P inputs and TP leached losses was negligible due to the slight change of the P inputs in the FAS, CS and BS areas (Fig. 5). With aim to interpret the differential change of TP leached losses in these cropping systems, soil Olsen-P contents in the CK and RS treatments were ascertained (Table S3) and the relationship between soil Olsen-P contents and TP leached losses were established (Fig. S2). Results showed that in the PVFs and OVFs system, TP leached losses increased gradually with an increase in soil Olsen-P. However, TP leached losses retained its levels in the CFs system (Fig. S2). Focusing on the changes of soil Olsen-P, we found that soil Olsen-P were decreased in the RS treatment when compared with the CK treatment. However, soil Olsen-P contents still exceeded 100 mg kg^{-1} in the RS treatment of the protected vegetable fields (Table S3).

In addition, the P inputs in the control treatment decreased significantly in the protected vegetable fields after 2013. Therefore, the database of P inputs in the CK treatment was divided into the period of 2008–2013 compared to the period of 2014–2018 in this study. A decline of TP leached amounts from 0.337 kg ha^{-1} to 0.173 kg ha^{-1} was found when the P inputs decreased from 857 kg ha^{-1} to 585 kg ha^{-1} in the FAS areas (Table 3). Likewise, TP leached losses reduced from

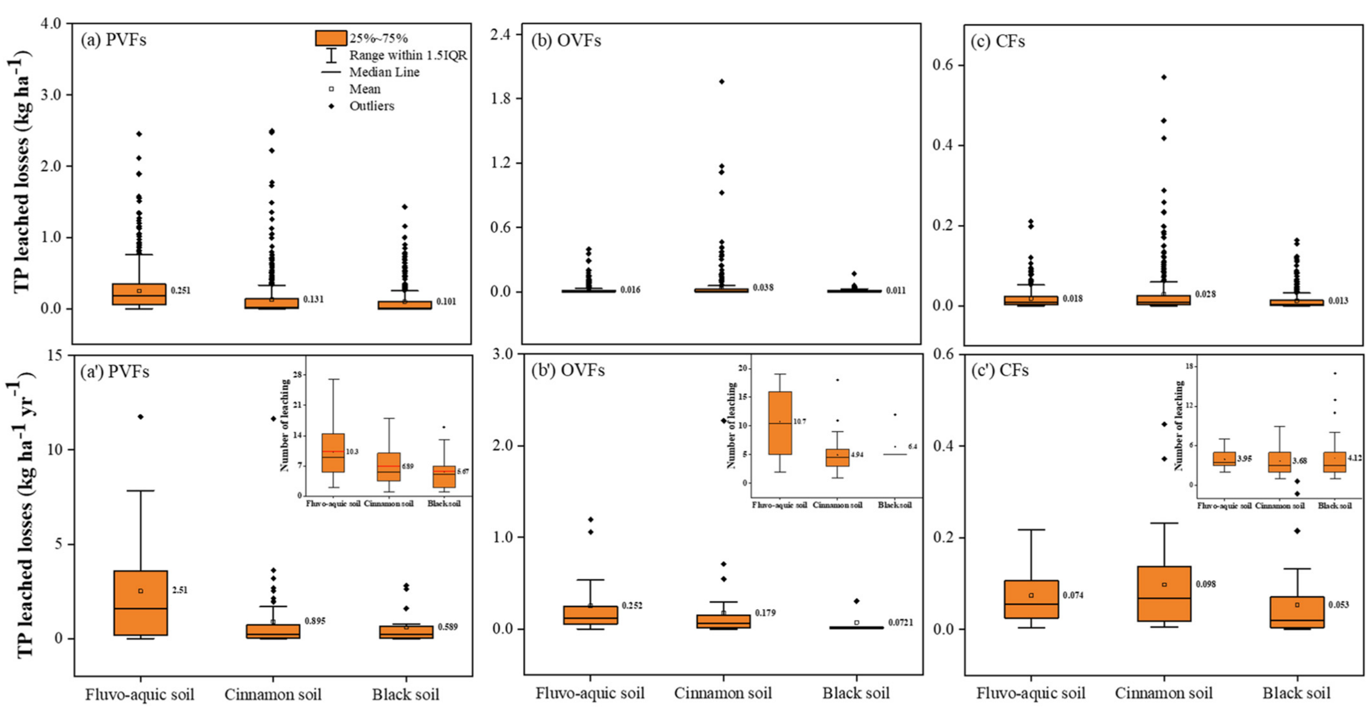

Fig. 3. Total phosphorus leached losses (kg ha^{-1}, a-c), numbers of TP leaching events and annual TP leached losses (kg ha^{-1} yr^{-1}, a'-c') in PVFs (protected vegetable fields), OVFs (open vegetable fields) and CFs (cereal fields) of the fluvo-aquic, cinnamon and black soil areas from 2008 to 2018 in northern China.

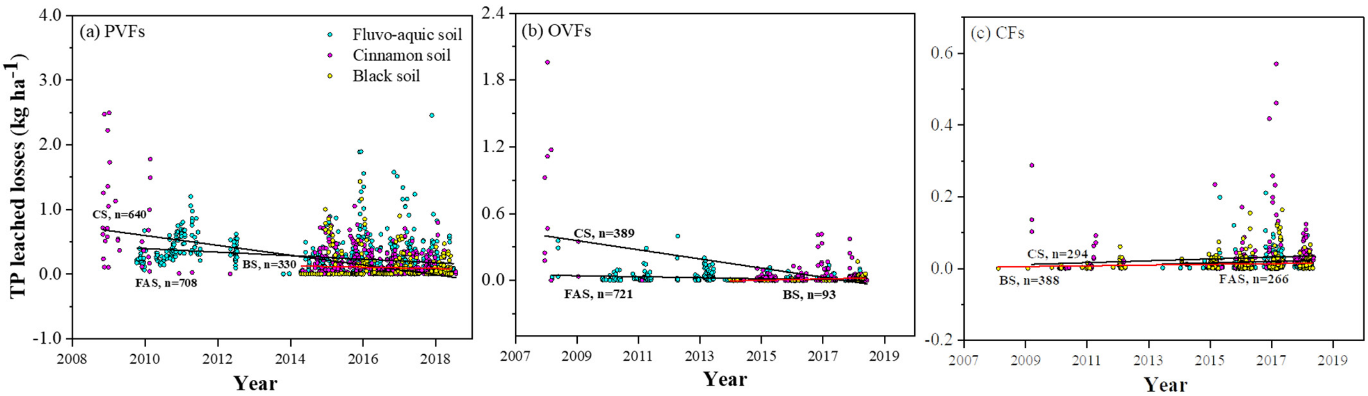

Fig. 4. Total phosphorus losses in leached pathway in the fluvo-aquic, cinnamon and black soil areas for (a) PVFs (protected vegetable fields), (b) OVFs (open vegetable fields) and (c) CFs (cereal fields) from 2008 to 2018 in northern China. With 708, 640 and 330 data points illustrated in the fluvo-aquic, cinnamon and black soil areas in the PVFs system, respectively; With 721, 389 and 93 data points illustrated in the fluvo-aquic, cinnamon and black soil areas in the OVFs system, respectively; With 226, 294 and 388 data points illustrated in the fluvo-aquic, cinnamon and black areas in the CFs system, respectively.

0.408 kg ha^{-1} to 0.096 kg ha^{-1} when the P inputs decreased from 1186 kg ha^{-1} to 336 kg ha^{-1} in the CS areas. However, in the open vegetable fields and cereal fields systems, the comparison of TP leached losses and P inputs between 2008 and 2013 and 2014–2018 was vague and imprecise (Table 3). This finding was in line with the relationship between P fertiliser inputs and TP leached losses, in which it was found that TP leached losses had no positive relationship with the P inputs in the open vegetable fields and cereal fields (Fig. 6). However, the TP leached losses increased in a linear way ($p < 0.001$) when the P inputs increased in the protected vegetable fields.

4. Discussion

4.1. Explanation of P leached losses and concentrations

In northern China, some studies have demonstrated a well-documented deterioration in P concentration at a soil depth of 90–180 cm (Fei et al., 2019; Yan et al., 2016). Recent years, China is becoming increasingly aware of the environmental impacts from agricultural production and has been implementing many major policies, e.g. "zero increase target for chemical fertilisers", "10-point Water Plan", "10-point soil Plan" etc. to mitigate P pollution (Jin et al., 2020). Tied closely to these policies, this study also indicated that the TP leached losses have been decreasing from 2008 to 2018 (Fig. 4). And, with the gradual decrease of P fertiliser inputs, the TP leached loads losses in vegetable cropping systems decreased significantly from 2008 to 2013 to 2014–2018 (Table 3). However, the P inputs in these areas in 2014–2018 were still several folds higher than the crop offtake (Yan et al., 2013). In addition, despite the knowledge that protecting groundwater is of great importance, how much P leached losses out of the rootzone is not clear in northern China.

In the present study, it was estimated that the annual TP leached losses were 4.99 × 10^6 kg during 2008–2018 in northern China (Table 2), which accounted for 6.5% of the total TP losses (0.76 × 10^8 kg) based on the data given in the second China Pollution Source Census. The TP leached losses from vegetable cropping systems, specifically in protected vegetable fields, were the predominant P lost to groundwater in northern China. This result is in accordance with many other studies which also demonstrated that the main P losses were sourced from vegetable cropping systems in China (Wang et al., 2019; Yan et al., 2013). It is noteworthy that the TP leached losses reached up to 2.5 kg ha^{-1} in protected vegetable fields in the present study (Fig. 3), in accordance with the results of Yan et al. (2016) and higher than the national scale results of 0 to 0.95 kg ha^{-1} reported by Huang et al. (2017). However, the TP leached losses from cereals fields system should not be neglected due to the relatively high cropping areas (Table 2).

The TP concentration in the leachate to 0.9 m depth all exceeded 0.1 mg L^{-1} (Fig. 2). The total dissolved P concentrations in protected vegetable fields even reached up to 0.6–0.8 mg L^{-1} (Fig. 2), which was significantly higher than the P threshold of 0.01–0.1 mg L^{-1} for the eutrophication of water bodies (Sharpley et al., 1996). In addition, we found that the total dissolved P is the main P form of P lost in agricultural production areas, in line with the finding of Hua et al. (2019).These suggested TP leached losses should be adequately addressed in northern China.

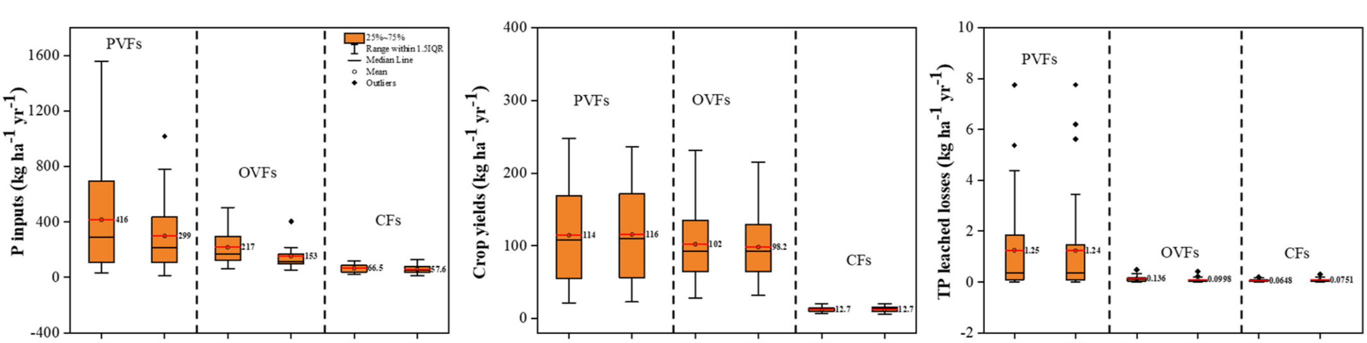

Fig. 5. Average of P inputs, crop yields and TP leached losses (kg ha^{-1}) in the CK (control) treatment and RS (reduced fertilisation) treatment in the PVFs (protected vegetable fields), OVFs (open vegetable fields) and CFs (cereal fields) from 2008 to 2018 in northern China.

Table 3
The decrease of TP leached losses from the fluvo-aquic, cinnamon and black soil areas of PVFs (protected vegetable fields), OVFs (open vegetable fields) and CFs (cropland fields) under conventional fertilisation of farmers' practice in the period of 2008–2013 versus 2014–2018.

	2008–2013			2014–2018		
	P inputs (kg ha^{-1} yr^{-1})	TP leached rates (kg ha^{-1})	TP leached amounts (kg ha^{-1} yr^{-1})	P inputs (kg ha^{-1} yr^{-1})	TP leached rates (kg ha^{-1})	TP leached amounts (kg ha^{-1} yr^{-1})
PVFs/soil areas						
FAS areas [1]	857±190a[3]	0.337 ± 0.128a	7.443 ± 3.398a	556 ± 271b	0.173 ± 0.181b	1.691 ± 1.810b
CS areas	1186 ± 1046a	0.408 ± 0.478b	3.720 ± 5.490a	336 ± 237b	0.096 ± 0.131a	0.613 ± 0.892b
BS areas	NS[2]	NS	NS	241 ± 282	0.124 ± 0.16	0.589 ± 0.85
OVFs/soil areas						
FAS areas	231 ± 57a	0.061 ± 0.046a	0.595 ± 0.430a	341 ± 58a	0.010 ± 0.009b	0.081 ± 0.071b
CS areas	101.1 ± 57b	0.239 ± 0.357a	0.619 ± 1.107a	239 ± 179a	0.022 ± 0.027b	0.116 ± 0.163b
BS areas	NS	NS	NS	187 ± 81	0.007 ± 0.010	0.072 ± 0.131
CFs/soil areas						
FAS areas	NS	NS	NS	76 ± 29	0.020 ± 0.017	0.074 ± 0.062
CS areas	79 ± 25a	0.037 ± 0.055a	0.079 ± 0.076a	75 ± 15a	0.038 ± 0.047a	0.104 ± 0.113a
BS areas	58 ± 34a	0.004 ± 0.004a	0.026 ± 0.044b	49 ± 23a	0.015 ± 0.020a	0.060 ± 0.079a

[1] FAS areas: Fluvo-aquic soil areas include the Shandong and Henan provinces; CS areas: Cinnamon soil areas include the Beijing municipality, Tianjin municipality, Hebei, Shanxi, Shaanxi, Ningxia and Gansu provinces; BS areas: Black soil areas include the Heilongjiang, Jilin, Liaoning, Inner Mongolia, Xinjiang and Qinghai provinces.
[2] NS: Data not shown.
[3] The small letters followed the standard deviation means significant difference ($p < 0.05$) of the P inputs, TP leached rates and TP leached amounts between 2008 and 2013 and 2014–2018.

4.2. Factors of importance that inform TP leached losses apportionment

In the present study, the TP leached losses in the vegetable cropping systems were significantly greater in the FAS areas (Shandong and Henan provinces) than those in CS and BS areas (Table 2). In line with the findings in many other studies, the higher TP leached losses were caused by higher P inputs, especially manure P, and frequent irrigation (Bai et al., 2020; Chen et al., 2019a; Fan et al., 2020). The present statistical analysis showed that the manure P inputs, chemical P inputs, irrigation inputs contributed to 24.2%, 2% and 23.3% of the TP leached losses in the present study.

The reason of the high TP leached losses in FAS areas can be ascribed as follows: (1) The North China plain is one of the most intensive agricultural region in China (Ju et al., 2009), and FAS areas are the breadbasket in the North China plain, which received more P fertiliser inputs and corresponding irrigation inputs (Table 2). In the present study, the P inputs in the vegetable systems of FAS areas were 1.3–3 folds greater than those in CS and BS areas. And, the percentage of manure P inputs in the protected vegetable fields occupied 51% of the P inputs in FAS areas. Additionally, the irrigation inputs in FAS areas were 1.5–2 folds higher than the CS and BS areas. This greater P fertiliser and irrigation inputs would inextricably increase the TP leached losses. (2) Current heavier N inputs have further accelerated the decrease of soil pH (Guo et al., 2010), which promoted soil P solubility and mobility. Results from the present study showed that the soil pH in the PVFs system of FAS areas was lower than the CFs system (Table 1). A study of Kianpoor Kalkhajeh et al. (2021) also reported the soil pH in FAS areas decreased 1–2 units over 10 years of vegetable production. Consequently, soil P leached losses increased with the acidifying of alkaline soil which distributed widely in northern China (Chen et al., 2019b). (3) the variation of soil properties and climatic conditions. Studies specifically explored the response of soil labile P influenced by soil properties indicated that the fluvo-aquic soil in the FAS areas has lower organic matter while greater Ca-bearing minerals and sandy contents compared with the black soil (Wang et al., 2015; Zhang et al., 2020b). Generally, the organic matter had a positive effect on soil labile P while the influence of Ca-bearing minerals was negative (Luo et al., 2017). Herein, inherently productive and fertile black soil in northern China are a large soil organic matter pool which contributes to increasing P availability when compared with the fluvo-aquic soil (Li et al., 2020). Consequently, more fertilisers needed to be applied in the FAS areas to safeguard crop production but corresponding P accumulation and losses raised. As this study mentioned above, owing to the relatively high soil fertility in the BS areas (Table 1), the P inputs was the lowest in this areas and P leached load losses were also the lowest compared with FAS and CS areas (Table 2).

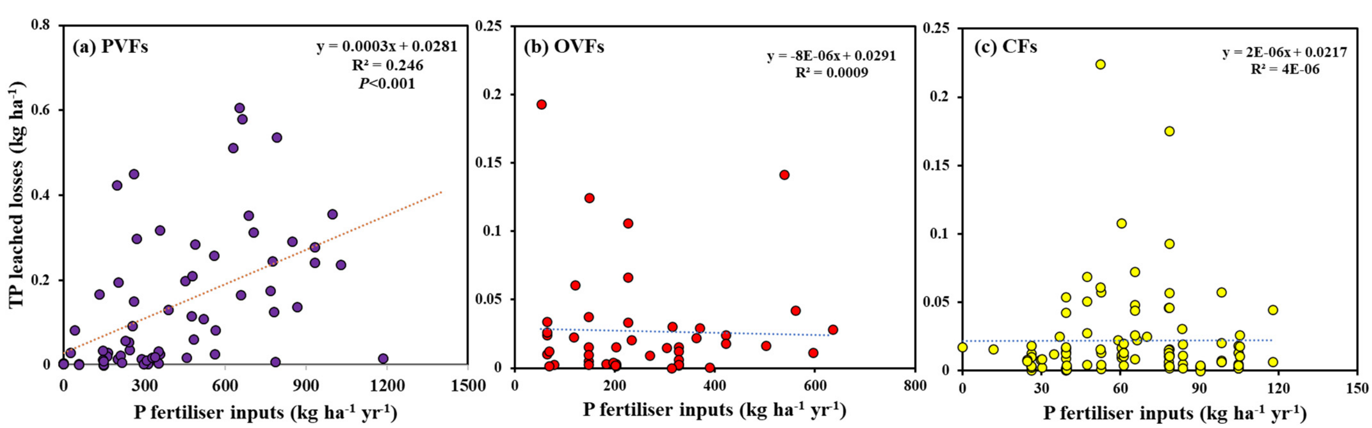

Fig. 6. Relationship between average TP leached losses and P inputs in the fluvo-aquic, cinnamon and black areas for (a) PVFs (protected vegetable fields), (b) OVFs (open vegetable fields) and (c) CFs (cereal fields) from 2008 to 2018 in northern China.

4.3. Management options to mitigate P losses

Generally, soil clay mineral, Fe/Al and Ca/Mg render most of the exogenous phosphate ion from fertiliser unavailable (Fan et al., 2021; Gérard, 2016; Yan et al., 2018). However, a large fraction of P in soils and bedrocks might be leaching as dissolved and particulate forms in excessively fertilized soils with increasing P availability (MacDonald et al., 2011; Withers et al., 2017). In the present study, it was found that TP leached losses changed slightly with the increase of P inputs in the cereal fields and open vegetable fields, while it increased abruptly in a linear way in protected vegetable fields with an increase in P inputs (Fig. 6a). This result was in accordance with the findings of Qin et al. (2020), who reported that the water extracted P increased rapidly under 26 years of consecutive manure P inputs.

More importantly, this study demonstrated the decrease of the TP leached losses was difficult to fulfil in the protected vegetable fields (Fig. 5). Compared with the control treatment under the farmers' practices, a decline of 29% of P inputs in the open vegetable fields resulted in a decrease of nearly 30% of TP leached losses while a reduction of 28% of P inputs in protected vegetable fields only decreased the TP leached losses by 0.8%. When compared the soil Olsen-P contents in the control and reduced fertiliser trials, we found that the soil Olsen-P contents in the RS treatment still higher than 100 mg kg^{-1} in the PVFs system (Table S3). Although the P inputs in RS treatment decreased by 10–30% in PVFs, it was still 4–5 folds in excess of crop removal. This might be the reason that the TP leached losses did not decrease significantly with the reduction of P inputs in the PVFs. These results implied that the easier built up of soil legacy P with heavy P inputs, but a longer lag time to decrease the P losses in the P enriched soil, in accordance with the findings of other studies that soil P from excessive to optimum agronomic P levels can take long period due to biogeochemical lag time (Sattari et al., 2012; Schulte et al., 2010). However, the present study verified a decline of 20–30% of P inputs in the protected vegetable fields (Fig. 5), and that a reduction of 10–30% of P inputs in the open vegetable fields of FAS and BS areas had no effect on crop yields yet (Fig. 5), while mitigating TP leached losses. So in existing protected vegetable fields systems the future management advice is to reduce the P inputs to decrease the P stores in soil and in new lands being planted the advice is to avoid the occurrence of the high legacy P soil.

As with cereal production fields, vegetable cultivation relied heavily on a greater degree management, larger fertiliser inputs and irrigation. Growers of the protected vegetable fields are usually the elderly and have little knowledge about precise fertilisation in China, but China has no national/regional guide or legislation for fertiliser application to prevent nutrient losses from cropping systems (Bai et al., 2020). It was highlighted that the decrease of the P inputs should be recommended in China. In the cereal fields, the P inputs is chemical fertilisers and maintained around 40–90 kg P ha^{-1}, the crop production and TP leached losses did not change significantly between the control and low P inputs systems (Fig. 5). However, Chinese government is focusing on replacing chemical fertiliser by manure in the cereal fields these years due to the excessive manure production and the decline of soil fertility (Jin et al., 2020; Zhang et al., 2020a). It is also noteworthy that the TP leached losses in the future will increase as the manure contributes more to P mobility than chemical P fertiliser (Liu et al., 2020; Ma et al., 2020; Zhang et al., 2020a).

5. Conclusions

The present study estimated and apportioned annual TP leached losses across the main soil areas and cropping systems in northern China. It also examined difference between different P input systems in terms of leached mitigation potential across this matrix of soil areas and cropping systems. An estimate of annual TP leached losses from 2008 to 2018 was 4.99×10^6 kg in northern China. The FAS and CS areas combined accounted for 77.5% of the TP leached losses in northern China. Protected vegetable fields contributed to 48.5% of the total TP leached losses. A reduction of P inputs in P enriched soils will see some improvements in leached loads and reductions from 10 to 30% in P inputs in vegetable cropping systems could still maintain yields while lowering P leached losses. The results of this study are pivotal to provide guidance for the precise fertilisation in China. Further studies are recommended to exploring the influences of equivalent N inputs alongside different P inputs on the TP leached losses and crop yields.

CRediT authorship contribution statement

Bingqian Fan: Data curation, Investigation, Methodology, Writing – original draft. **Hongyuan Wang:** Conceptualization, Funding acquisition, Project administration, Resources, Supervision. **Limei Zhai:** Writing – review & editing, Validation. **Jungai Li:** Software, Formal analysis, Writing – review & editing. **Owen Fenton:** Writing – review & editing, Validation. **Karen Daly:** Writing – review & editing, Visualization. **Qiuliang Lei:** Writing – review & editing, Visualization. **Shuxia Wu:** Writing – review & editing, Visualization. **Hongbin Liu:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported by the National Key Research and Development Program of China (2016YFD0800101), the Agricultural Science and Technology Innovation Program of Chinese Academy of Agricultural Sciences and Fundamental Research Funds for Central Non-profit Scientific Institution.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.150441.

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