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Potentially toxic trace element pollution in long-term fertilized agricultural soils in China: A meta-analysis

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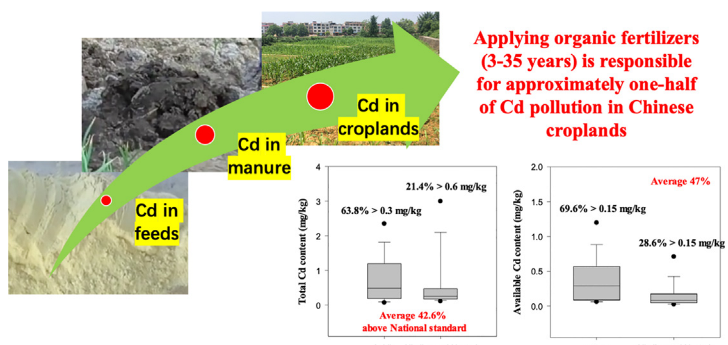
HIGHLIGHTS

- Long-term organic fertilization increased the relative availability of Zn by 96–128.6%
- Cd pollution in organic fertilized croplands (approximately one-half) is the most important issue
- Type and application rate of organic fertilizers and soil type were dominant factors influencing variation of PTTEs
- Organic fertilizers containing Cd less than 1 mg kg^{-1} were recommended
- Lower amounts of P fertilizers were proposed for Alfisols and Semi-hydromorphic soils

GRAPHICAL ABSTRACT



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ABSTRACT

Fertilization results in potentially toxic trace element (PTTE) pollution in agricultural soils. However, it is unclear which factors determine the effect sizes of fertilization on PTTEs at the multiple spatial-temporal scale. This work synthesized 379 observations in 78 field sites (3–35 years) across China's main grain producing areas, and showed that long-term organic fertilization significantly enhanced total Cu, Zn and Cd by 25.7%, 18.9% and 66.6%, and soil available Cu, Zn and Cd by 60.5%, 155.3% and 83.6%, respectively; whereas long-term inorganic fertilization increased only available Cu, Zn and Cd by an average of 6.3%. Organic fertilizer (OF) type and application rate dominated the variation of PTTE concentrations, where approximately one-half of Cd pollution (42.6% of total Cd and 47% of available Cd) was observed. Furthermore, OFs containing Cd less than 1 mg kg^{-1} were recommended to be safely applied to agricultural soils. Soil type was main factor under long-term inorganic fertilization determining available PTTE variation, resulted in higher pollution risk in some soils such as Alfisols and Semi-hydromorphic soils, where we suggested the use of lower amounts of P fertilizers or the application of ones having small amounts of PTTEs. In short, long-term organic fertilization caused serious pollution of PTTEs especially Cd in Chinese croplands, and some strategies with a focus towards reducing the pollution risk must be developed, e.g., promoting straw return, forbidding Cd addition to feeds and feed additives, and improving carbon sequestration efficiency (CSE) of OFs and thus soil organic matter (SOM).

1. Introduction

Fertilizer plays an important role in increasing cereal yields, the global use of which has increased by 30.8% (N), 31.4% (P_2O_5) and

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61.3% (K_2O), respectively, from 2002 to 2017 (FAOSTAT, 2020). For the next 30 years, more fertilizers would be used to obtain more products to feed the projected global population of over 9 billion (Godfray et al., 2010; Thompson, 2016). However, their excessive use caused farmland pollution due to a large input of potentially toxic trace elements (PTTEs) from fertilizers (Mortvedt, 1995; Atafar et al., 2010; Ramos et al., 2020).

Most researchers in America and Europe have generally accepted that application of inorganic phosphate fertilizers has inadvertently added Cd and other toxic elements to the soil (Mortvedt, 1995; Nziguheba and Smolders, 2008; Atafar et al., 2010). For example, even only 2-year-period of fertilization could result in PTTE concentration increasing 1.1–2.9 times after harvesting (Atafar et al., 2010). In China its application, even at very high levels (100–120 kg P_2O_5/ha), has insignificantly increased the soil total PTTE contents (Zhou et al., 2015; Rao et al., 2018). However, it is still necessary to regulate the utilization of phosphate fertilizers in North China, especially in Henan, due to having the highest Cd input from phosphate fertilizers, also due to the increase in the content of bioavailable soil Cd after phosphate fertilizer application, which causes a higher content of Cd in agricultural products (Dharma-Wardana, 2018; Li et al., 2020). It is known that the risk of Cd accumulation by crops is more related to its availability than the total Cd present in the soil, therefore, more efforts should be put to evaluate the effect of long-term phosphate fertilization on available Cd levels in soils. Yet, only a limited number of studies have focused on it (Rao et al., 2018; Wei et al., 2020), and there is a big knowledge gap in understanding which factors have the greatest effect on availability.

A large number of studies on the influence of organic fertilization have been published, and most have only focused on high inputs of PTTEs and the subsequent pollution from animal manure, without paying attention to availability with long-term manure application (Luo et al., 2009). Both soil organic matter (SOM) and dissolved organic matter (DOM) increased markedly after long-term organic fertilization, where SOM could immobilize PTTEs and reduce their bioavailability; while DOM could enhance the mobilization of PTTEs from soil (Antoniadis and Alloway, 2002; Karlsson et al., 2006; Ondrasek and Rengel, 2012; Laurent et al., 2020). Thus, it is still unclear whether the availability increases or decreases. Additionally, there are many kinds of organic fertilizers (OFs) with different content of PTTEs that could be used in farmland, resulting in considerable variations in accumulation in these PTTEs in soil. It is also unclear which fertilizer should be preferred for environment safety. For example, swine manure was frequently considered as a primary contributor of PTTEs especially Cu and Zn to farmland (Novak et al., 2004; Shi et al., 2011; Liu et al., 2020). However, our previous study showed only the content of Cd in long-term swine manure-fertilized soils exceeded (by 3-fold) the national standard (Zhou et al., 2015). Similar results indicated swine and chicken manures were mainly responsible for Cd contamination of farmland soils (Li et al., 2010). Also, some studies showed that total soil Cu and Zn concentrations, as well as available soil Cd and Zn concentrations, were elevated significantly due to the long-term application of higher rates of cattle manure (Benke et al., 2008), which supply about 64% of total Cd inputs in soils in England and Wales (Nicholson et al., 2003).

Soil, crop, fertilizer, and climate all influenced the load, distribution, accumulation, and availability of PTTEs in soil-plant systems (Hou et al., 2017; Zupancic, 2017). We need a better understanding of what drives these variations of PTTE contents in agricultural soil, in order to both improve fertilizer efficiency and reduce environmental pollution. Soil PTTE pollution derived from fertilizer use is a slow process occurring over decades, which can be described only by relying on long-term field experiments. However, there is a lack of studies conducted to determine effect size of fertilization at multiple spatial-temporal scales. In this study, we conducted a synthesis of 379 experimental observations in 78 field sites in 22 provinces across China's main grain producing areas, which represented the typical farming systems in this country (Fig. S1 and Dataset 1). The questions we addressed were (1) Which element is the most strongly associated with fertilizer application among Cu, Zn, Cd, and Pb? (2) Which variable affects the heterogeneity of soil PTTEs the most? (3) If so, what measures can be taken to minimize environmental pollution caused by fertilizer overuse?

2. Materials and methods

2.1. Sampling and chemical analysis

After the autumn harvest (2012–2014), soil samples were collected from the 0–20 cm soil layer at each experiment plots from 39 long-term fertilization sites across China (Fig. S1 and Dataset 1). The fresh soil samples were air-dried and ground to pass through a 1 mm sieve for the determination of available Cu, Zn, Cd and Pb, and a 0.15 mm sieve for the analysis of their respective total content. Animal manure, green manure, and straw were also collected from some of the above sites. After collection, they were oven-dried at 70 °C, ground and passed through a 0.25 mm sieve for the determination of total metal concentrations.

Available PTTEs in acidic soils were extracted with 0.1 M HCl; while those in neutral and alkaline soils were extracted with 0.005 M diethylenetriaminepentaacetic acid (DTPA) in 0.01 M $CaCa_2$, buffered at pH 7.3 with 0.1 M triethanolamine (TEA) (Lindsay and Norvell, 1978; Qian et al., 1996). The detailed procedures were as follows: 5.00 g air-dried soil passed through a 1 mm sieve was mixed with 25 mL DTPA (or HCl) and continuously stirred at 25 °C for 2 h. The suspension was centrifugated at 4500 rpm for 10 min and kept at 4 °C for subsequent determination. To measure the contents of PTTEs in soil, manure and straw, soil samples were digested by HNO_3-HF, and manure and straw samples were digested by HNO_3 in microwave (Wu et al., 1997; Sandroni et al., 2003). The detailed procedures were as follows: air-dried soil (0.5000 g) passed through a 0.15 mm sieve was weighted in a digestion vessel, into which 3 mL HF and 9 mL HNO_3 were added and allowed to stand for 24 h at room temperature, then digested in a microwave digester (Anton Paar Multiwave 3000, Graz, Austria), following a gradient temperature program: from ambience to 120 °C (increasing at the rate of 20 °C min^{-1}), then 120 °C for 3 min, and then 7 °C min^{-1} to 190 °C, followed by a final hold at 190 °C for 20 min. The digested sample was transferred into Teflon crucible and then heated on an electric hot plate to drive away the acids. Finally, it was diluted with 0.5% HNO_3 to 25 mL, filtrated and stored at 4 °C for later analysis. All digestion and extraction solutions were finally analyzed for Cu, Zn, and Pb by inductively coupled plasma optical emission spectrometry (ICP-OES, Varian 715-ES, Varian Medical Systems, USA), and for Cd by atomic absorption spectrometry (AAS, ZEEnit 700, Analytik Jena AG, Germany). For each sample, three repetitions were performed, and the results were presented as means $\pm$ SD.

In addition, several reference soil and plant samples (GBW07451 and GBW07449 for soil digestion; GBW07458, GBW07416a and GBW07413a for soil extraction; GBW07603 for manure and straw digestion) and reagent blanks were used as the quality control samples during the analyses, where total Cu, Zn, Cd, Pb recoveries were 101.9%, 98.7%, 98.2%, and 95.3%; whereas available PTTE recoveries were 105.9%, 95.0%, 100.9%, and 97.6%, respectively. Analytic reagents purchased from Sinopham Chemical Reagent Co., China, and ultra-pure water (18 M$\Omega \cdot$cm) supplied from Pall Co., USA, were used throughout the whole experiments.

2.2. Data collection and dataset construction

An extensive literature survey was conducted through China National Knowledge Infrastructure (CNKI), China Wanfang Database, China Weipu Database, National library of China, Web of Science, and Google Scholar until June 30, 2019. The search keywords were ("long-term fertilization" OR "long-term fertilizer" OR "long-term experiment") AND ("heavy metal" OR "microelement" OR "trace element"). The following criteria were used to select appropriate studies: (1) only those long-term fertilization field experiments located in China were selected; (2) field experiments were conducted for not less than 3 years; (3) for each treatment, total and available contents of at least one of these potentially toxic trace elements (Cu, Zn, Pb and Cd) were determined; (4) experiments were designed to include a control group (no fertilization) and one or more treatment group (inorganic and/or organic fertilization); (5) OFs used for the experiments are animal manure, green manure, straw, and their compost, but not including sewage sludge; (6) only the results from the last sampling period were collected if multiple measurements were performed throughout the experimental period in the same study site, because meta-analysis required that datasets are independent (Hedges et al., 1999). Using these criteria, 28 studies at 39 experimental sites were found to be suitable for the meta-analysis (Fig. S1, Reference list and Dataset 1).

The data were directly gathered from tables and from figures by the GetData Graph Digitizer (Version 2.26.0.20, Russia). If only standard error (SE) was provided, it was converted to SD: $SD = SE\sqrt{n}$ (n is the sample size). In the cases where no SD or SE were reported, 1/10 of means was determined as SD (Luo et al., 2006).

As a result, this dataset included data measured by ourselves (39 sites) as well as data collected through a literature survey (39 sites). That is, a total of 379 paired observations covering 78 experimental sites were included in this study, which was divided into Cu (231 observations), Zn (233 observations), Cd (207 observations) and Pb (207 observations) in inorganic fertilization treatments, Cu (311 observations), Zn (315 observations), Cd (275 observations) and Pb (274 observations) in organic fertilization treatments (Dataset 1).

The dataset also included location (i.e., latitude and longitude) and climate factors (mean annual temperature (MAT), mean annual precipitation (MAP), annual evaporation, ≥10 °C accumulated temperature, and aridity index (AI)), initial soil factors (soil type, parent material, texture, pH, SOM, Olsen-P, available N and K), cultivation factors (crop type, cultivated land type, rotation, multi-cropping), and fertilization factors (OF type, OF application rate, PTTE content in OFs, inorganic P fertilization) in each site. Overall, the dataset covered broad variations in climate, soil, cultivation and fertilization. All data except those measured by ourselves were extracted from 52 publications (Dataset 1 and Reference list).

2.3. Meta-analysis

Effect sizes of fertilization on total and available potentially toxic trace element concentrations in soils were expressed as the natural log-transformed response ratio (ln(RR)), and were calculated by Eq. (1):

$$\ln(RR) = \ln\left(\frac{\overline{X_t}}{\overline{X_c}}\right) \tag{1}$$

where $\overline{X_t}$ and $\overline{X_c}$ are the means of treatment (with fertilizer application) and control (without fertilizer application), respectively.

Variance (Var) of ln(RR) was computed by Eq. (2):

$$Var(\ln RR) = \frac{SD_t^2}{n_t\overline{X_t^2}} + \frac{SD_c^2}{n_c\overline{X_c^2}} \tag{2}$$

where n_t and n_c were the sample size of the treatment and control groups, respectively; SD_t and SD_c were the SDs of the treatment and control groups, respectively (Luo et al., 2006).

ln(RR) was used as an effect size metric for the meta-analysis. The use of the natural logarithm linearizes the metric, ensuring the approximate normal distribution with a mean equal to the true response ratio (Luo et al., 2006; Seufert et al., 2012). For interpretation of the results, mean effects and 95% confidence intervals (CI) were back-transformed, using the formula: $(e^{\ln(RR)}-1)\times100\%$ and reported as the percentage changes between control and fertilizer application. A significant response could be considered if its 95% CI did not overlap zero (Seufert et al., 2012).

Due to the higher heterogeneities among the subgroups, observations with the same or similar management or system characteristics were grouped together (Table S1), and the grouped effects were computed using a categorical meta-analysis with a random model (MetaWin 2.1, Sinauer Associates Inc., USA). To test for differences in the effect sizes between groups, the total heterogeneity of the sample was partitioned into the within group (Q_W) and between group heterogeneity (Q_B), and the significance of Q_B was tested by comparing it against the critical value of the χ^2 distribution. A significant Q_B ($p < 0.05$) indicated that there are differences among cumulative effect sizes between groups, so, only those effects showing a significant Q_B are presented in graphs (Seufert et al., 2012). Moreover, all of these variables with significant Q_B were selected and combined for multivariable meta-regression analysis, so as to determine dominant factors controlling soil PTTE variation under long-term fertilization. Stata/SE 15.0 (Stata Corp., College Station, TX, USA) was used to build a regression model through analysis of the influence of the *t*-value and *p*-value to determine heterogeneity of variables, where statistical significance was assumed at $p < 0.05$ (Thompson and Higgins, 2002; Yu et al., 2018).

3. Results

3.1. Overall effects of long-term fertilization on total and available potentially toxic trace element content

Long-term organic fertilization increased total Cu, Zn and Cd by 25.7% (21.4–30.0%), 18.9% (16.0–21.8%) and 66.6% (52.7–80.5%) and soil available Cu, Zn and Cd by 60.5% (50.1–70.9%), 155.3% (128.2–182.4%) and 83.6% (62.5–104.6%), respectively; whereas long-term inorganic fertilization increased only available Cu, Zn and Cd by an average of 6.3% (Fig. 1). There was no difference in Pb content and availability between fertilization (whether inorganic or organic) and control (without fertilizer application).

From Fig. 1 it could be seen that organic fertilization caused much higher risk of PTTE pollution than inorganic fertilization, where the biggest concern would be the highest increase in soil total Cd and available Zn. Generally, a more significant increase in available metal amounts in fertilized soils was observed. For this reason, relative availability (RA) as the ratio of available PTTE (DTPA or HCl) to total PTTE concentration was calculated and shown in Fig. S2 (Inaba and Takenaka, 2005; Karalić et al., 2013; Ouyang et al., 2020), which demonstrated that OF application resulted in the highest increase in RA_{Zn} (96.0–128.6%), and the smallest increase in RA_{Cd} (4.5–15.8%). Because using RA as an indicator may minimize measurement errors among different researchers, compared to an absolute value (total or available metal content) (Su et al., 2015); more importantly, RA made it possible to compare PTTE availability from different extraction procedures such as HCl-, DTPA-, and DGT-extractability, and Exchangeable forms (Su et al., 2015; Dataset 1), it is clear that RA is a better indicator of risk of PTTE pollution than those total and available fractions.

3.2. Dominant factors determining the variation in soil PTTEs under long-term OF application

The content and availability of PTTEs in organically fertilized soils were controlled by many soil, climate and fertilization factors such as soil type, texture and parent material, site location, OF type and application rate (Tables S2–S3, Figs. S3–S4). However, multivariate meta-regression model confirmed that OF type and application rate (equivalent C input) appeared to be the most important factors, where OF type mainly determined the variation of total and available Cu and Zn, with variation explained (R^2) being 21.66% and 19.91% for total Cu and Zn, 26.96% and 16.72% for available Cu and Zn, respectively; while soil Cd

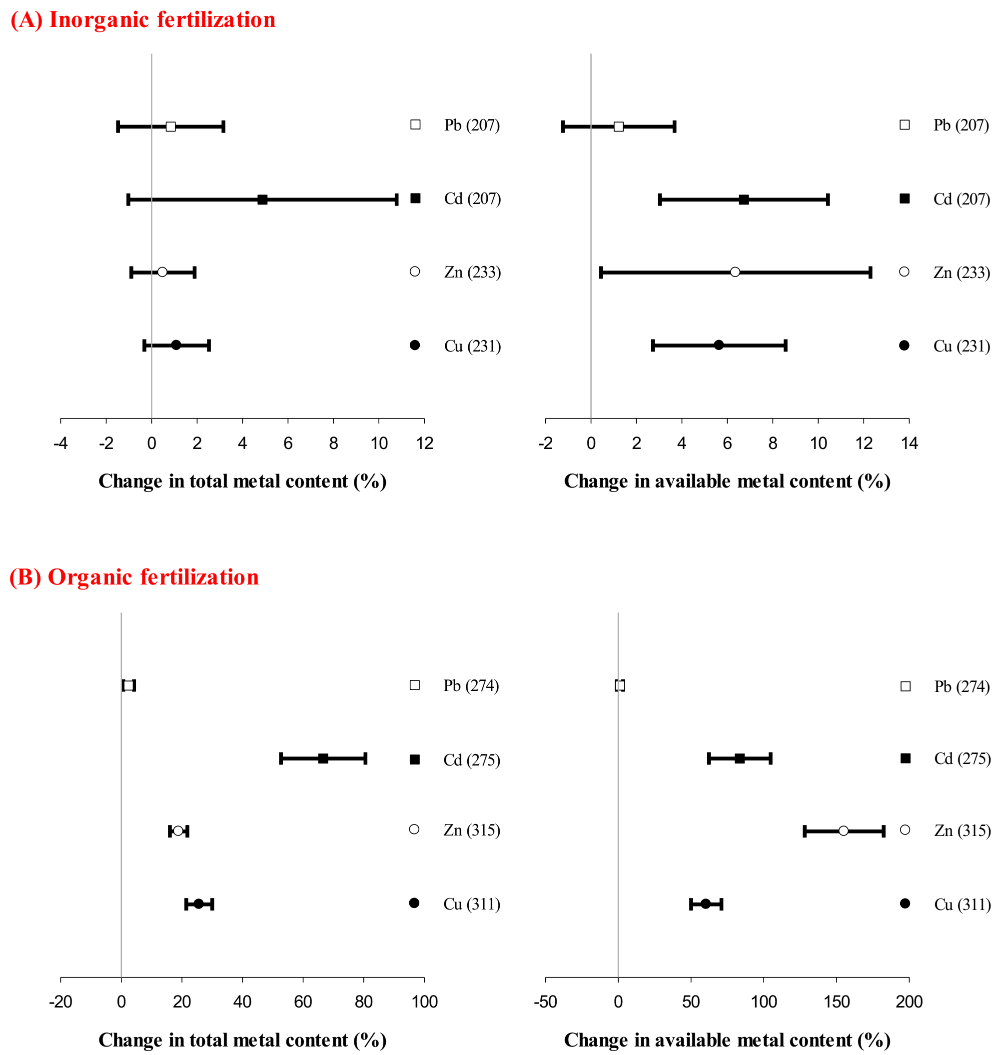

Fig. 1. Percentage changes in total and available potentially toxic trace elements for inorganic (A) and organic (B) fertilization compared to control (no fertilization). Values are mean effect sizes with 95% confidence intervals (CI). A significant response is when CI does not overlap 0. The number of observations in each class is shown in parentheses.

was dominantly determined by OF application rate, with $R^2 = 16.07\%$ and 14.88% for total and available Cd, respectively (Tables S4-S9).

OF type exhibited a higher effect on the accumulation of PTTEs comparing to its application rate (Fig. 2), where swine manure and high application rate of other animal manure significantly enhanced the accumulation of Cd (by 508.8%), Zn (by 53.4%) and Cu (by 84.7%) in soils (3–7 times higher than average); whereas long-term application of straw and green manure did not increase soil PTTEs.

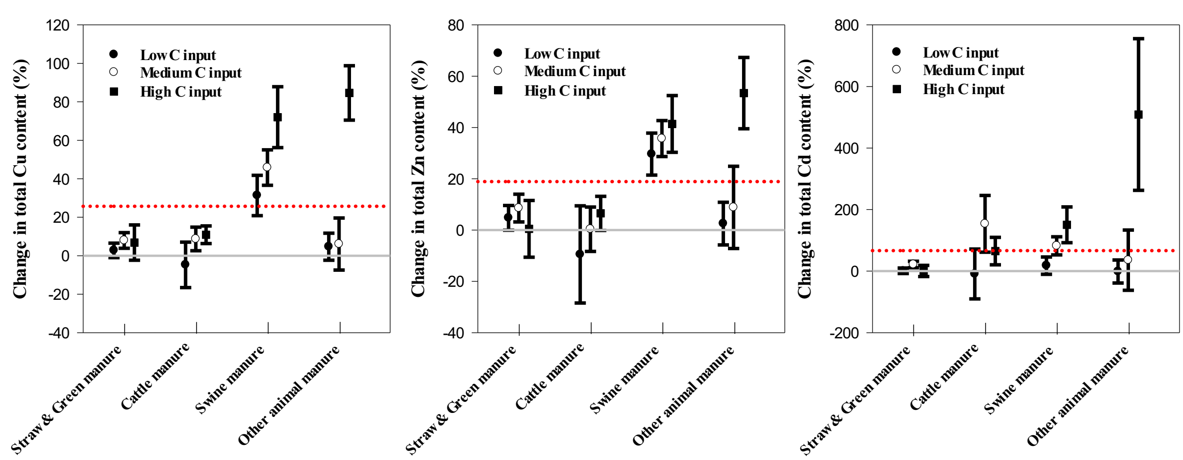

Fig. 2. The influence of manure type and equivalent C input on organic fertilization effect sizes for soil total Cu, Zn and Cd, respectively, compared to no fertilization. Values are mean effect sizes with 95% confidence intervals. The dotted line indicates the cumulative effect size across all classes.

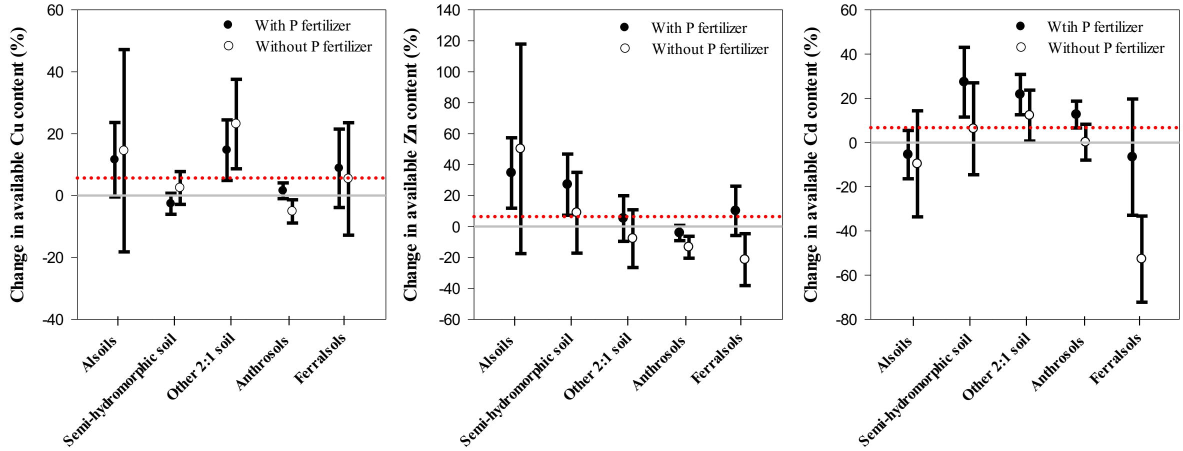

Fig. 3. The influence of soil type and P fertilizer on inorganic fertilization effect sizes for soil available Cu, Zn and Cd, respectively, compared to no fertilization. Values are mean effect sizes with 95% confidence intervals. The dotted line indicates the cumulative effect size across all classes.

3.3. Main factors explaining the variation of available PTTEs under long-term inorganic application

Although soil available Cu, Zn and Cd increased slightly in response to inorganic fertilizer application, higher significant variation was also observed among different factors such as soil type, soil texture, temperature zone, location, land type, etc. (Tables S10-S11 and Fig. S5). The result of multivariate meta-regression showed that soil type contributed most to the heterogeneity, because it was the only significant variable explaining 15.11% variability in soil available Cu, and played a more important role in determining variation in soil available Zn and Cd, explaining 14.38% and 13.43% of the variation, respectively, which was far higher than that of P fertilizer, another of the two only significant variables (2.47% and 3.96%, respectively) (Tables S12-S14).

Our data further showed that the increase in available PTTEs in some soil types (such as Alfisols, Semi-hydromorphic soils and other 2:1 soils) under P fertilization was significantly higher than the average effect sizes (Fig. 3). For example, available Cd in Semi-hydromorphic soils with P fertilization increased by 27% (13–44%); whereas there was a slight but statistically insignificant increase in available Cd without P fertilization. So, one needs to be careful to apply inorganic fertilizers (in particular P fertilizers) to these soils.

3.4. PTTE pollution in long-term organically fertilized soils

There was a close linear relation between the response of available and total PTTEs to long-term (3–35 years) organic fertilization, with a slope of 3.68 for Cu, 11.15 for Zn, and 0.75 for Cd, respectively (Fig. 4), indicating that the potential risk of Cd came from the near-synchronous increase in total and available Cd in soils (up to 25 times higher), whereas Zn's risk exists in a 35-times increase in its availability. However, the box-plot showed that Cd pollution was the most serious in long-term organically fertilized soils, with 42.6% of total Cd and 47% of available Cd content exceeding the national standards (GB15618-2018 and DB35/T 859-2016) (Fig. 5). By contrast, Zn pollution was not serious, for the percentages exceeding the national standard limit were only 1.57% and 7.9%, respectively. Moreover, it should be noted from Fig. 5 that long-term application of OFs resulted in much more PTTE pollution in acidic soils than in alkaline and neutral soils.

A stronger linear relation was observed between soil Cd accumulation and Cd inputs from OFs ($R^2 = 0.81$, $p < 0.0001$) (Fig. 6). To ensure safe application of OFs to farmlands within the next 100 years, we proposed a threshold limit value of 1.0 mg Cd per kg dry matter based on the linear relationship and the background and risk control values of soil Cd (0.097 and 0.3 mg kg^{-1}, respectively) (Wei et al., 1991), where a conventional application rate of 380 kg N/ha/yr (20% N was substituted by dry OFs having a 2.08% average N content) was used as the basis for calculation (NAESC, 1999; Saikia et al., 2015; Zhang et al., 2018). The recommended value was in excellent agreement with the EU permissible level in compost (Kulikowska et al., 2015), but much less than the limit value (3 mg kg^{-1}) of the Chinese standard for OFs (NY 525-2012).

4. Discussion

4.1. Risk of PTTE pollution of soils during long-term fertilization

Fertilizers were responsible for only 6–9% of Pb inputs to Chinese croplands (Luo et al., 2009), where Pb accumulation in soils was probably just offset by its loss through crop harvesting. So, applying fertilizers

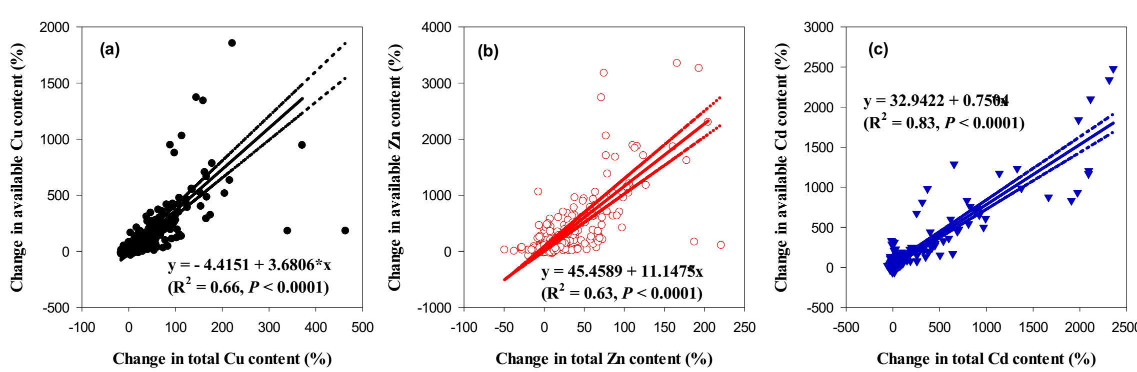

Fig. 4. The relationships between response of soil total and available potentially toxic trace elements (a: Cu, b: Zn, c: Cd) to long-term organic fertilization. The solid lines were the linear fits that best fitted the measurements, while the dotted lines were their 95% confidence intervals.

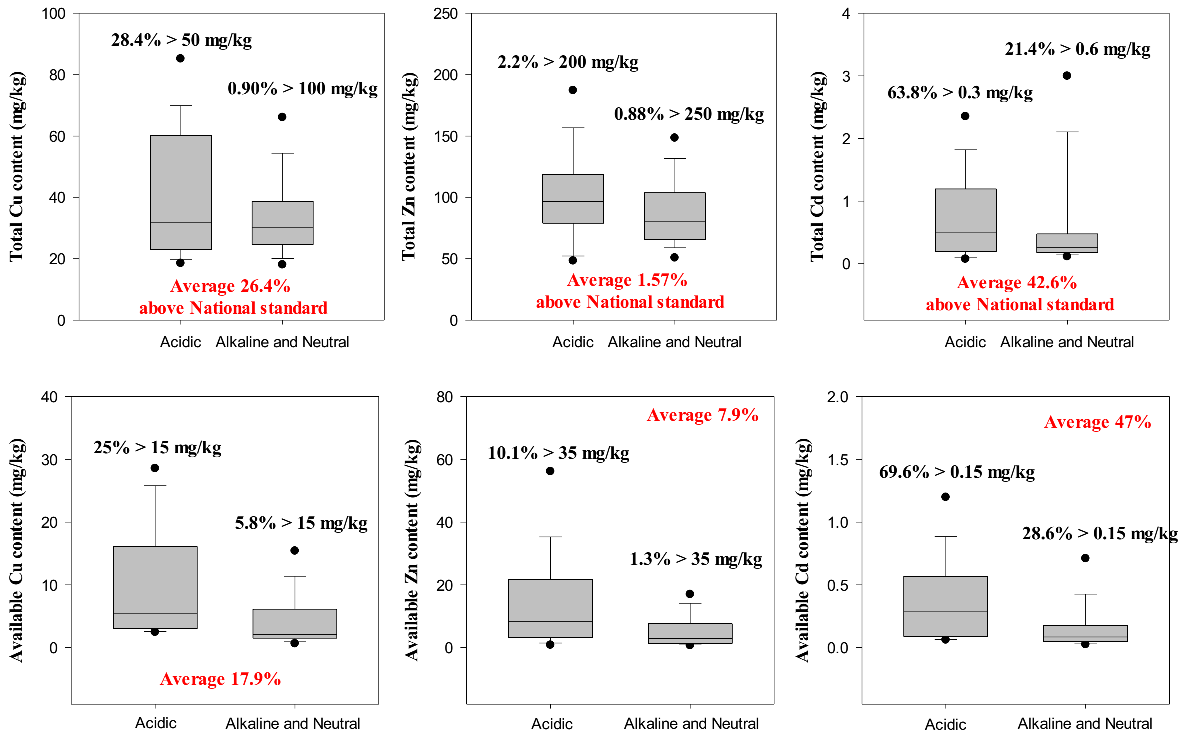

Fig. 5. Box-plot of total and available Cu, Zn and Cd in long-term (3–35 years) organically fertilized soils.

did not contribute to Pb pollution. Similarly, PTTE input via inorganic fertilizers (in particular P fertilizers) in Chinese croplands was much less than in those of some other nations, where inorganic P fertilizer was responsible for only 1.7% of Cd inputs in China, whereas it contributed to 25.0% in England and Wales (Nicholson et al., 2003; Luo et al., 2009). Thus, it was considered that inorganic fertilizers (including P fertilizers) were not important pollution sources of PTTEs in Chinese croplands. The results were inconsistent with some researches in US and European countries, which showed that P-based fertilizers were likely to be the leading source of Cd soil pollution (Ali et al., 2020; Gupta et al., 2014; Nziguheba and Smolders, 2008). In conclusion, there is a low risk of PTTE pollution from long-term inorganic fertilizer application. However, the overuse of inorganic fertilizers had caused a decrease in soil pH (soil acidification), which potentially caused the increase in PTTE availability (Bolan et al., 2003; Wei et al., 2020). For example, P fertilization could increase available Cd in Semi-hydromorphic soils by 44% (Fig. 3). This means that reducing the application rate of P fertilizers, or applying P fertilizers containing relatively small amounts of PTTEs, would be practical solutions for reducing the potential pollution risk in some soils (such as Alfisols, Semi-hydromorphic soils and other 2:1 soils).

The findings of this study revealed that inputs of PTTEs (especially Cd) from animal manure application are the main sources of PTTE pollution in agricultural soils, for their contribution to the inputs of Cu, Zn and Cd to Chinese croplands were 68.5%, 51.0% and 54.9%, respectively, significantly higher than that of inorganic fertilizers (3.8%, 4.2% and 8.0%, respectively) (Luo et al., 2009). Numerous studies have shown that the majority of PTTEs in feeds was excreted in feces and urine by livestock and poultry without being metabolized, which became a primary contributor to pollution in farmlands (Cang et al., 2004; Li et al.,

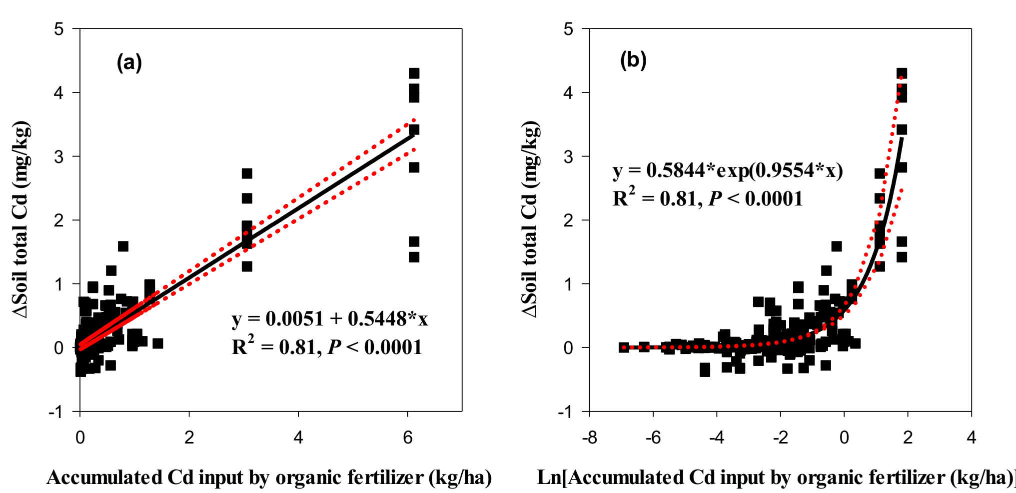

Fig. 6. The relationships between soil Cd accumulation and its inputs from organic fertilization (a: Cd input; b: Ln(Cd input)). The symbol (Δ) means the difference in soil total Cd content between organic fertilization treatment and control (no fertilization). Solid lines were the model fits, and dotted lines were the associated 95% confidence intervals.

2010; Li et al., 2019; Zhang et al., 2012; Wang et al., 2013). For example, Li et al. (2010) reported that higher residue of Cd in feeds (31 mg kg^{-1}) resulted in higher levels of Cd in manures (129.8 mg kg^{-1}), more than 430 times higher than China's maximum permissible limit of Cd in OFs (NY 525-2012). Hance, much effort should be devoted to stopping the adding PTTEs to feeds and feed additives.

In particular, it should be noted that long-term organic fertilization led to a significant increase in PTTE availability or RA in agricultural soils (Fig. 1 and Fig. S2). Obviously, this risk of PTTE pollution from applying OFs would substantially increase. Some researches have shown that DOM increased greatly after OF application due to its rapid decomposition, which would induce a prominent increase in available PTTEs (Antoniadis and Alloway, 2002; Laurent et al., 2020). To reduce the availability of toxic metals such as Cd in soils, some strategies for enhancing carbon sequestration efficiency (CSE) of OFs could be adopted.

4.2. Strategies for safely applying OFs

OFs have been highlighted as a major pollution source of Cd, Cu and Zn in agricultural soils, contributing to approximately one-half of soil Cd pollution (42.6% of total Cd and 47% of available Cd, respectively) (Fig. 5). Therefore, strategies with a focus towards reducing both metal content in OFs during the manufacturing processes and metal availability in soils during the decomposition of OFs must be developed. The following strategies have the potential to improve future OF application.

Firstly, we should pay attention to promoting straw and green manure return to field, or to implementing composting of various straw resources. They not only have low pollution risk potential to the surrounding environment (Fig. 2), but also show advantages in promoting soil fertility and thus offsetting partly the need for inorganic fertilizers and reducing PTTE availability in soils (Li et al., 2018). Secondly, laws and regulation were proposed to control Cu and Zn content in feeds and to forbid to specially add Cd to feeds and feed additives. Cd as an impurity is often present in Zn-mineral supplements (such as zinc sulfate and phosphate, and Zn oxide). Thus, due to the abuse of Zn-mineral additives to animal feeds for stimulating animal growth, higher residue of Cd in feeds and corresponding manure (31 mg kg^{-1} and 129.8 mg kg^{-1}, respectively) have been reported (Li et al., 2010). Meanwhile, the government should promote the utilization of natural feed additives and plant-derived products for livestock and poultry production. Finally, the nation needs to continue to advance SOM Improvement Project since 2005, because SOM played an important role in reducing available soil PTTEs (Karlsson et al., 2006; Ondrasek and Rengel, 2012). However, the increase in SOM is a very slow process, where the CSE of manures in Chinese croplands was as low as 9.6% so that SOM had an increase less than 5 g kg^{-1} during 30 years (Yang et al., 2017; Jiang et al., 2018). Therefore, more studies focusing on the improving CSE in China were needed.

5. Conclusion

The meta-analysis results showed that long-term organic fertilization resulted in serious potentially toxic trace element pollution (especially Cd) in Chinese croplands, where more significant increase in available Cu, Zn and Cd than in their total contents was found; whereas only a slight but significant increase in available Cu, Zn and Cd was observed under long-term inorganic fertilization. Organic fertilizer type and application rate dominated the variation of potentially toxic trace elements, where animal manures such as swine and cattle manures containing Cd less than 1 mg kg^{-1} were recommended to be safely applied to agricultural soils. Soil type was main factor under long-term inorganic fertilization determining the variation of available potentially toxic trace elements, suggested that in some soils such as Alfisols and Semi-hydromorphic soils, lower amounts of P fertilizers or ones having small amounts of potentially toxic trace elements should be applied in order to reduce the potential pollution risk. In the organic fertilization practice, some strategies with a focus towards reducing the pollution risk must be developed, e.g., promoting straw return to field, forbidding Cd addition to feeds and feed additives, and improving carbon sequestration efficiency and thus soil organic matter. Overall, the findings of this study clearly gave a warning that organic fertilizer for the needs of agricultural production in China was not fully secured, and that governments, farmers and scientists needs to take timely measures to reduce its pollution risk.

CRediT authorship contribution statement

Shiwei Zhou: Conceptualization, Writing – original draft, Investigation, Validation, Writing – review & editing. **Shu Su:** Formal analysis, Resources, Data curation, Validation, Writing – review & editing. **Ling Meng:** Investigation, Validation, Writing – review & editing. **Xiao Liu:** Formal analysis, Resources, Data curation, Validation, Writing – review & editing. **Hongyuan Zhang:** Validation, Writing – review & editing. **Xiaoli Bi:** Conceptualization, Writing – original draft, Investigation, Validation, Writing – review & editing.

Declaration of competing interest

The authors declare no competing financial interests.

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Appendix A. Supplementary data

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