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Accumulation of heavy metals in soil and maize after 17-year pig manure application in Northeast China

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Abstract: Application of composted pig manure (PM) is a traditional practice to improve soil fertility, whereas generally leads to some environmental questions. The effects of PM application on Cd, Cu and Zn accumulation in soil and maize were investigated based on a long-term field trial in Northeast China, including control (CK), PM_L, PM_M, and PM_H, receiving 0, 100, 250 and 500 kg total N/ha/year from 2002 to 2008 and 0, 10, 25 and 50 t fresh weight/ha/year from 2009 to 2018, respectively. Results showed that long-term soil application of PM increased maize grain yield, soil organic carbon (SOC) contents, coupled with significant accumulation and availability of Cd, Cu, and Zn in soil (0–15 cm). Compared with CK, the soil total Cd, Cu and Zn concentrations significantly increased by 105, 287 and 108% at high PM rate, respectively. Notably, the increments enhanced these heavy metals storage in maize roots rather than in grains. Moreover, the application of PM confirmed vertical transport of heavy metals in the tested soil, particularly for Cd and Cu in PM_H treatment. Overall, the repeated application of PM can cause the accumulation and leaching of Cd, Cu and Zn in soil.

Keywords: *Zea mays* L.; toxicity; organic fertilisation; crop yield; toxic elements; correlation

Northeast China serves as one of the main food production areas. Recent studies showed that soil organic carbon (SOC) in farmlands as the corn of soil fertility only in this area have decreased gradually due to intensive cultivation since 1980 in China (Zhao et al. 2018). Because the application of organic manure can improve soil fertility and increase crop yield, it is a traditional fertilisation regime in the world, and consequently is encouraged for remaining and increasing SOC in Northeast China (Li et al. 2011). However, the feeds and additive contain heavy metal that are widely used in stockbreeding, which lead to high heavy metals contents in manures due to their incomplete metabolism in the organism (Cang et al. 2004). A large part of those metals is released into soil and water with animal manures (Nicholson et al. 2003), which may raise environmental risks.

Application of animal manures to agro-ecosystems led to heavy metal accumulation in soil, especially for Cd, Cu, and Zn (Nicholson et al. 2003, Shi et al. 2011). Livestock manures accounted for approximately 55, 69 and 51% of the total Cd, Cu and Zn inputs in agricultural soils of China, respectively (Shi et al. 2019). In addition, some of the heavy metals transported vertically to deeper soil layers or were leached out of the soil. Cadet et al. (2012) found that the metal contents in soil profile significantly increased relative to those in un-amended soils after continuous poultry litter application, coupling with the increments in bioavailable and phytotoxic of metal elements. Increasing the risk of heavy metal uptake by crops, such as rice and wheat, was also observed in soils fertilised with manures (Zhou et al. 2015). A wide range of studies that have investigated

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the heavy metal accumulation in soils and crops after manure application, and have been conducted in soil-rice/wheat systems or a short time (Li et al. 2011, Shi et al. 2011, Huang et al. 2018). However, long-term field trials generally provide more reliable results of the fertilisation effects on soil ecological environment (Rasmussen et al. 1998). Maize is also a major cereal crop and food source for both humans and animals, and is the dietary staple for > 200 million people, contributing to 20% of the world's calories (Nuss and Tanumihardjo 2010). So heavy metals accumulation and distribution in the soil-maize system should be focused on especially under the long-term organic manure application.

The application of organic manure is believed to an efficient way to improve SOC, as well as enhance heavy metals accumulation in soil (Li et al. 2011). Meanwhile, the interactions of SOC with heavy metals are complex. On the one hand, SOC could reduce the availability of heavy metals by enhancing the surface area of soil adsorption sites or increase their availability by activating soil-bound heavy metals during decomposition of organic matter (Guan et al. 2018). On the other hand, the soil metals availability may be further modified over time after manure application because the changes in soil properties manure-caused may activate more soil-bound metals (Zhao et al. 2014). In addition, manure application doses impact the accumulation and fractionation of heavy metals, thereby changing the metal availability in soil (Zhao et al. 2014, Huang et al. 2018). Meta-analysis showed that the application rate of organic manure was one of the significant factors to minimize some undesirable effects on the soil environment (Chen et al. 2018). It is interesting and important to understand the effects of long-term application of organic manure at different rates on crop yield, the availability of heavy metal in soil and food security. Yet, less information is known regarding the effect of pig manure (PM) application at different dosages on the availability of heavy metals in the soil-crop system, based on long-term field trials. Thus, the goals of the present study are to determine the effects of long-term PM application on heavy metal Cd, Cu, and Zn availability, accumulation, and distribution in the soil-maize system.

MATERIAL AND METHODS

Study site and experiment design. The long-term field trial was started in 2002 at the National Field Research Station of Shenyang Agro-ecosystems (41°32'N, 123°23'E), Chinese Academy of Sciences in Liaoning province, Northeast China. This site has a temperate sub-humid mainland climate and 147–164 days of the frost-free period. The annual average temperature and precipitation are 7.5 °C and 520 mm. The test soil is classified as Aquic Brown Soil (FAO 2006).

Four treatments were designed: including control (CK), PM_L, PM_M, and PM_H, receiving 0, 100, 250 and 500 kg total N/ha/year from 2002 to 2008 and 0, 10, 25 and 50 t fresh weight/ha/year from 2009 to 2018, respectively. The compost produced from pig manure was mainly applied prior to crop cultivation. Each year, the fertilisers were incorporated into the surface soil (0–15 cm) by rotary tiller before planting in the spring. The crops were sown in May, relying on the weather and soil conditions. A three-course rotation with soybean (*Glycine max* L.) – maize (*Zea mays* L.) – maize from 2002 to 2011 and maize monoculture from 2012 were conducted in each experimental plots. No other fertiliser, irrigation and herbicide were input. Each treatment is arranged by a complete-block design with three replicates (main plot and size 108 m^2). The maize was manually harvested and measured the grain yield at maturity in mid-October. The basic characteristics and heavy metal contents of tested soil and PM were shown in Table 1.

Sampling and analysis. Soil samples were collected from surface soil (0–15 cm) and depth of 0–10, 10–20, 20–40 and 40–60 cm profile after the maize harvest in October 2018. Each soil sample comprised five

Table 1. Basic characteristics and heavy metal contents of tested soil and pig manure (PM)

	pH	Total C	Total N	Total P	Total K	Olsen-P	Available K	Total Cd	Total Cu	Total Zn
		(g/kg)				(mg/kg)				
Soil	6.40	11.55	1.00	0.50	17.57	10.4	103.0	0.20	27.7	66.9
PM*	–	212.54	15.60	12.58	13.29	–	–	1.00	438.1	624.1

*On average PM contained the concentrations of nutrients and heavy metals based on a dry weight basis

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random soil cores, which were air-dried and sieved (< 2 mm) for further analysis. The plant samples were collected together with soil samples, dividing them into root, stalk, and grain. Plant samples were dried in an oven at 65 °C and then ground into a powder with a stainless steel pulverizer. The basic characteristics of tested soil and PM were measured by the methods of Lu (1999). The total Cd, Cu and Zn in soil and plant samples were digested using HF-HNO_3-$HClO_4$ (2:4:1) and HNO_3-$HClO_4$ (4:1), respectively, and were determined by ICP-OES (Optima 7000DV, PerkinElmer Inc., USA). The available Cd, Cu and Zn in soil were assessed with DTPA (diethylenetriaminepentaacetic acid) extraction (Zhu et al. 2012) and were analysed by ICP-OES.

Statistical analysis. The date is shown as the means ± standard deviation (SD, n = 3). Significances and Duncan's tests were performed by SPSS 23.0 (SPSS; Chicago, USA). The correlation coefficients (r) based on Spearman's equations were calculated between SOC, the metals in soil and plant.

RESULTS AND DISCUSSION

Soil organic C. The SOC concentration in surface soil was significantly (P < 0.05) increased by 35.8–106.3% for the effect of PM application (Figure 1). This is owing to PM is a rich source of carbon, likely providing more substrates for carbon accumulation in soil (Chen et al. 2018). Moreover, the accumulative effect of SOC caused by PM application primarily appeared in 0–20 cm rather than in 20–60 cm profiles because the PM was mainly incorporated into the surface soil (Zhao et al. 2014).

Soil Cd, Cu, and Zn accumulation and availability. The soil total Cd, Cu and Zn concentrations increased as PM application rates increased. Compared with CK, the soil total Cd, Cu and Zn concentrations were increased by 3.7–105.5, 68.1–287.1 and 9.1–108.1% for the application of PM, respectively (Figure 2). Zhao et al. (2014) found that the increase in soil heavy metals was affected significantly by manure application, which was similar to our results. The total Cd and Cu in PM_H treatment were higher than the maximum permissible limit (1 and 100 mg/kg, respectively) in Chinese agricultural soils (MEP 1995). This may be due to the high Cd, Cu and Zn input levels from the pig manure (1.0, 438.1 and 624.1 mg/kg in this study, respectively). These metals could still accumulate in surface soil if their input exceeds the output (Cang et al. 2004, Guan et al. 2018). Thus, the content of the metals in manure before application should be treated for minimizing soil pollution risk.

The DTPA-extractable Cu, Zn, and Cd were generally regarded as predictors of Cu, Zn and Cd availability, respectively (Zhu et al. 2012). The DTPA Cd, Cu and Zn accounted for 11.2–25.0, 9.1–27.0 and 1.8–19.3% of the total soil Cd, Cu and Zn, respectively (Figure 2). The highest proportion was obtained in the PM_H, irrespective of the different heavy metals. In addition, the DTPA Cd, Cu and Zn concentrations significantly (P < 0.05) increased by 0.9–4.0, 2.7–10.6 and 7.0–21.9 folds in the treatments receiving PM over that of the CK (Figure 2). These results could be ascribed to the fact that the increment of total heavy metals caused by the application of PM may convert into available heavy metals in soil (Shi et al. 2011, Zhou et al. 2015). Additionally, the SOC may supply organic chemicals to the soil solution that can serve as chelates and increase metal availability (Finžgar et al. 2007). In this study, the total Cd, Cu and Zn were significantly positively correlated with

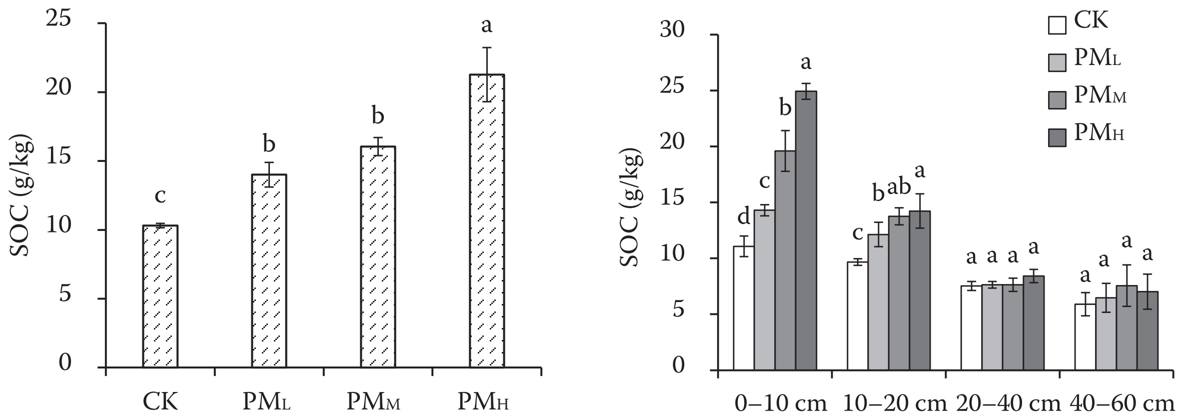

Figure 1. Effects of long-term pig manure (PM) application on soil organic carbon (SOC) in surface soils and 0–60 cm profiles. CK – 0, PM_L – 100, PM_M – 250, and PM_H – 500 kg total N/ha/year

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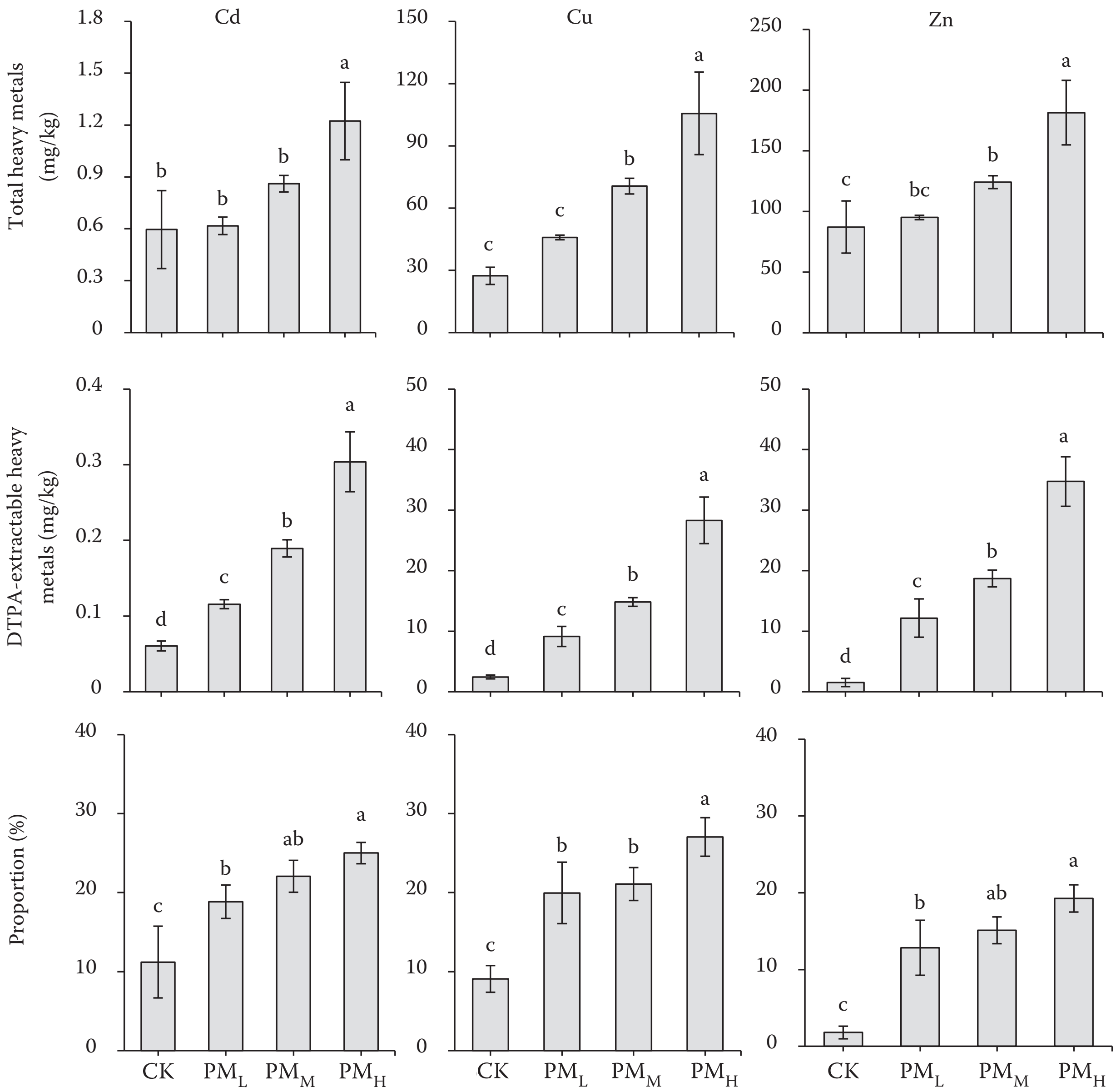

Figure 2. Effects of long-term pig manure (PM) application on soil Cd, Cu and Zn accumulation, availability and the proportion of the available heavy metals to the total soil heavy metals. CK – 0, PM_L – 100, PM_M – 250, and PM_H – 500 kg total N/ha/year

available Cd, Cu and Zn in surface soil, and all *r* values exceeded to 0.8 (Table 2). This also showed that these metals have a high synchronisation from pig manure. In summary, the application of PM enhanced SOC and heavy metals accumulation in soil, thereby likely increasing soil metals availability.

Cd, Cu, and Zn distribution in the soil profile. The total soil Cd, Cu and Zn concentrations in 0–60 cm profile decreased with increasing depth after 17-year addition of PM (Table 3). Compared with CK, the total Cd, Cu and Zn concentrations in 0–20 cm soil layers significantly ($P < 0.05$) increased with increasing levels of PM application. Zhou et al. (2015) pointed out that the soil total Cd, Cu and Zn concentrations

Table 2. Correlation correlations (*r*) between total-metals and DTPA-metals in the surface soil (0–15 cm) ($n = 12$)

	Total Cd	Total Cu	Total Zn
DTPA-Cd	0.846**	0.958**	0.860**
DTPA-Cu	0.811**	0.944**	0.846**
DTPA-Zn	0.811**	0.944**	0.846**

**$P < 0.01$

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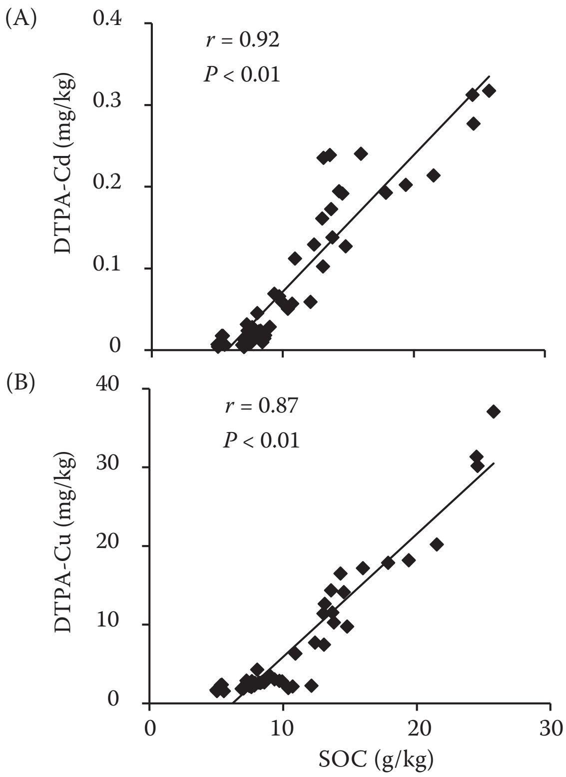
Table 3. Distributions of total and DTPA Cd, Cu and Zn at a profile of 0–60 cm in the tested soil of long-term pig manure (PM) application

Treatment		Total (mg/kg)			DTPA extractable (mg/kg)		
		Cd	Cu	Zn	Cd	Cu	Zn
0–10 cm	CK	0.421 ± 0.048^{d}	23.2 ± 0.8^{d}	59.8 ± 2.0^{d}	0.055 ± 0.004^{d}	2.1 ± 0.1^{d}	1.3 ± 0.2^{d}
	PM_L	0.634 ± 0.012^{c}	49.3 ± 1.1^{c}	95.6 ± 2.9^{c}	0.153 ± 0.036^{c}	12.2 ± 3.8^{c}	16.0 ± 3.7^{c}
	PM_M	0.856 ± 0.025^{b}	72.8 ± 0.6^{b}	134.1 ± 1.2^{b}	0.203 ± 0.011^{b}	18.7 ± 1.3^{b}	25.6 ± 2.5^{b}
	PM_H	1.126 ± 0.061^{a}	111.7 ± 2.9^{a}	196.5 ± 6.8^{a}	0.302 ± 0.022^{a}	32.8 ± 3.7^{a}	42.7 ± 3.7^{a}
10–20 cm	CK	0.439 ± 0.026^{c}	23.9 ± 0.5^{d}	57.1 ± 1.4^{b}	0.065 ± 0.005^{d}	2.9 ± 0.1^{c}	1.1 ± 0.2^{c}
	PM_L	0.516 ± 0.063^{c}	37.2 ± 1.2^{c}	70.5 ± 3.0^{b}	0.115 ± 0.014^{c}	7.1 ± 0.8^{b}	7.3 ± 1.2^{b}
	PM_M	0.685 ± 0.040^{b}	53.4 ± 2.9^{b}	93.4 ± 7.3^{a}	0.175 ± 0.015^{b}	12.3 ± 1.6^{a}	15.4 ± 2.4^{a}
	PM_H	0.896 ± 0.040^{a}	64.8 ± 6.9^{a}	101.9 ± 12.1^{a}	0.238 ± 0.003^{a}	14.7 ± 2.3^{a}	16.4 ± 3.4^{a}
20–40 cm	CK	0.336 ± 0.019^{b}	34.9 ± 2.8^{a}	57.8 ± 1.3^{a}	0.012 ± 0.002^{b}	2.1 ± 0.2^{b}	0.7 ± 0.4^{b}
	PM_L	0.315 ± 0.021^{b}	30.8 ± 2.1^{ab}	55.8 ± 1.1^{a}	0.023 ± 0.006^{ab}	2.6 ± 0.2^{b}	1.3 ± 0.2^{ab}
	PM_M	0.359 ± 0.026^{ab}	27.6 ± 2.3^{b}	54.8 ± 1.7^{a}	0.026 ± 0.005^{a}	2.7 ± 0.2^{ab}	1.6 ± 0.3^{a}
	PM_H	0.411 ± 0.052^{a}	32.7 ± 5.3^{ab}	59.9 ± 5.6^{a}	0.032 ± 0.012^{a}	3.5 ± 0.8^{a}	2.1 ± 0.6^{a}
40–60 cm	CK	0.304 ± 0.021^{a}	32.9 ± 2.9^{c}	61.7 ± 2.0^{a}	0.005 ± 0.002^{b}	1.6 ± 0.2^{b}	2.3 ± 0.8^{a}
	PM_L	0.365 ± 0.066^{a}	36.2 ± 1.0^{bc}	67.6 ± 5.1^{a}	0.007 ± 0.001^{b}	1.9 ± 0.3^{b}	2.4 ± 0.3^{a}
	PM_M	0.350 ± 0.019^{a}	45.7 ± 7.1^{a}	62.1 ± 8.7^{a}	0.017 ± 0.002^{a}	2.6 ± 0.3^{a}	2.0 ± 0.3^{a}
	PM_H	0.340 ± 0.050^{a}	42.9 ± 5.3^{ab}	62.8 ± 7.9^{a}	0.014 ± 0.004^{a}	2.4 ± 0.2^{a}	1.8 ± 0.5^{a}

Different letters within the same column indicate significant differences of heavy metals in the same soil profiles among treatments. CK – 0, PM_L – 100, PM_M – 250, and PM_H – 500 kg total N/ha/year

among PM treatments were relatively higher in the 0–20 cm profiles than in the 20–60 cm profiles, showing that the metals mainly accumulated in the surface soil. The total Cd, Cu and Zn concentrations



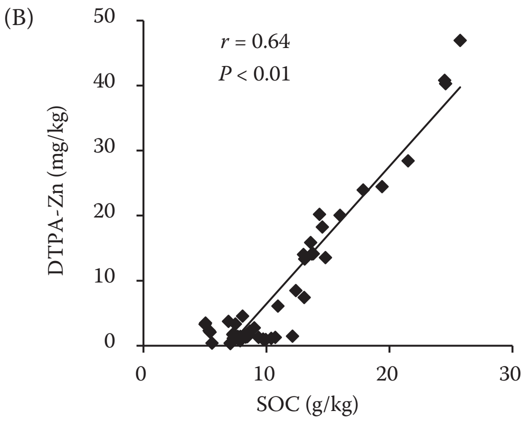


Figure 3. Correlation analysis of soil organic carbon (SOC) and DTPA (A) -Cd; (B) -Cu and (C) -Zn in the 0–60 cm profiles (n = 48)

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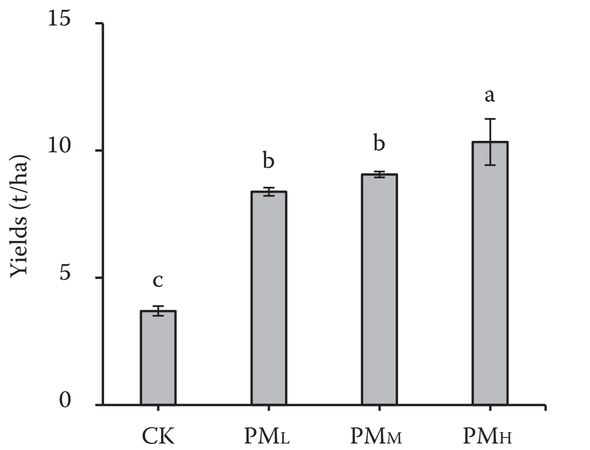

Figure 4. Average yields of maize for the period of the last 5 years (2014–2018). CK – 0, PM_L – 100, PM_M – 250, and PM_H – 500 kg total N/ha/year

in the deeper soil layers have no great fluctuation between CK and PM amended soils, likely because the PM is applied mainly at the surface of the soil (Berenguer et al. 2008, Zhao et al. 2014).

The DTPA Cd, Cu, and Zn concentrations in 0–20 cm profiles were increased as the increased PM application rates (Table 3), likely due to the activation effect of PM application on heavy metals (Huang et al. 2018). Compared with CK, PM_H treatments significantly (P < 0.05) increased the soil DTPA-Cd and -Cu in 20–60 cm profiles, as well as the DTPA-Zn in 20–40 cm profiles. Rao et al. (2018) and Zhou et al. (2015) pointed out that long-term applying PM led to the transport of heavy metals in the soil profile, and the mobility of Cd and Cu may be higher than Zn. Because the application of PM activates more soil-bound metals and surface soil absorption reach to the threshold, the mobility of the metals in profile was enhanced and leached into the deeper soil layers (Rao et al. 2018). Thus, long-term PM application aggravated the leaching risk of heavy metal in soil, which may further contaminate groundwater.

The DTPA-Cd, -Cu, and -Zn concentrations were significantly (P < 0.05) positively correlated with SOC across the 0–60 cm profiles (Figure 3). This was in line with the fact that SOC may be closely related to soil Cd, Cu and Zn availability (Finžgar et al. 2007). Improvement of soil fertility caused by PM application generally coupled with the increase of the amount and availability of heavy metals (Guan et al. 2018).

Grain yield and Cd, Cu, Zn concentrations in different parts of maize. The average yield of maize over the period of the last 5 years (2014–2018) was significantly (P < 0.05) increased by 126–179% for long-term PM application (Figure 4). Berenguer et al. (2008) found that the yield of maize was significantly raised with the soil liquid swine manure application, likely due to the improvement of soil fertility (Li et al. 2011). The Cd, Cu, and Zn concentrations in root were significantly (P < 0.05) increased by 31.1–177.8, 49–270 and 20.6–186.0% under long-term PM application, respectively (Figure 5). Compared with

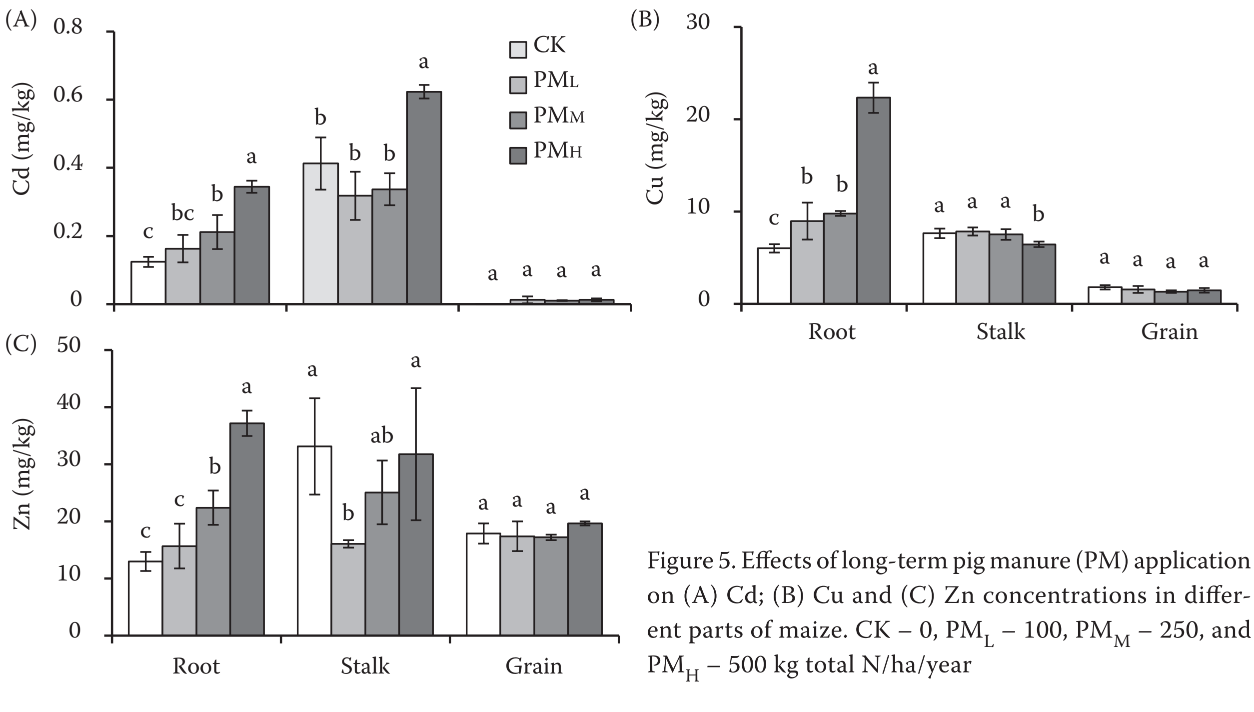

Figure 5. Effects of long-term pig manure (PM) application on (A) Cd; (B) Cu and (C) Zn concentrations in different parts of maize. CK – 0, PM_L – 100, PM_M – 250, and PM_H – 500 kg total N/ha/year

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CK, the PM_H treatment significantly increased stalk Cd while obviously reduced stalk Cu, likely because the high Cd uptake by stalk inhibited transformation for Cu (Jalil et al. 1994). Moreover, the PM application and the accumulation of Cd, Cu and Zn in soil showed no impact on the corresponding metals in grain (Figure 5), and their contents were below the threshold levels of the Chinese food safety standards (0.1, 10 and 50 mg/kg, respectively). Li et al. (2009) and Zhou et al. (2015) indicated that long-term application of PM alone enhanced in a significant accumulation of heavy metals in wheat grains and rice biomass rather than in maize grain. The differences among crops for heavy metal accumulation and distribution were ascribed to the lower accumulation ability for heavy metals in maize than in wheat and rice (Grant et al. 2008). It is possibly due to the decrease in the internal transport of trace elements from root to grain avoid metals toxicity in maize (Berenguer et al. 2008). In addition, the "dilution effect" caused by increased maize yield is also believed to be a central reason for our results to some extent (Jarrell and Beverly 1981). Based on the above results, advocating planting maize may be a viable way to reduce the risk of soil heavy metal accumulation in local staple food.

Correlation analysis of the metals in soil and maize plants. The Cd, Cu and Zn in maize root significantly positively correlated with the total and available metals in soil (Table 4), indicating their importance in accumulation of Cd, Cu and Zn in root. Huang et al. (2018) reported that the accumulation of heavy metals in plant root was associated with the soil's total and available metals. In addition, the total soil Cu was significantly negatively related to the Cu concentration in stalk and grain, while there were no significant correlations between Cd and Zn in soil and the metals in maize grain and stalk.

Table 4. Correlation analysis of the metals in soil and maize plant (n = 12)

		Root	Stalk	Grain
Cd	total	0.643*	ns	ns
	DTPA	0.916**	ns	ns
Cu	total	0.804**	–0.706*	ns
	DTPA	0.881**	–0.629*	–0.594*
Zn	total	0.909**	ns	ns
	DTPA	0.909**	ns	ns

ns – not significant; *P < 0.05; **P < 0.01

These results were consistent with the other work (Berenguer et al. 2008, Zhao et al. 2014), showing that the accumulation of the metals caused by long-term PM application mainly concentrated in the maize root, but not in grain. So removing the roots from the soil after plant harvest may be a feasible way to reduce the accumulation of heavy metals in soil receiving organic manure.

In conclusion, the application of PM significantly increased maize grain yield and SOC, as well as the concentrations and availability of Cd, Cu, and Zn in soil. The increments enhanced these heavy metals storage in maize roots rather than in grains. Although the inputs of heavy metals with PM application primarily accumulated in the depth of 0–20 cm soil, leaching of these metals in deeper soil layers was observed, particularly for Cd and Cu in PM_H treatment, the environmental risk of heavy metals accumulation caused by PM application in soil-plant-water system should be further assessed by mass balance assessment using data from this long-term field trial.

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