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Review

Comparisons of pollution characteristics, emission situations, and mass loads for heavy metals in the manures of different livestock and poultry in China

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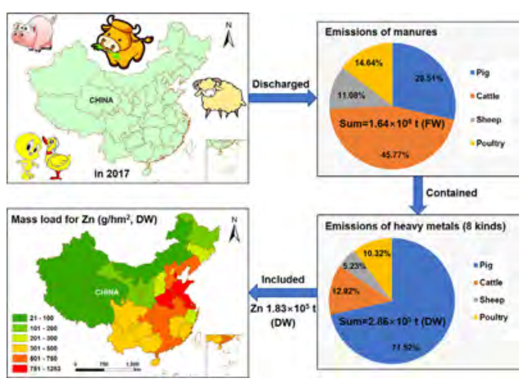
HIGHLIGHTS

- Zn, Cu, Cd, and As occur as fairly serious pollution in the pig manure of China.
- Similar concentrations to other countries, but higher heavy metal emissions in China.
- Heavy metals emission in China from livestock and poultry manures was up to 2.86×10^5 t.
- Southeastern provinces of China presented high mass loads of manures and heavy metals.
- Zn and Cu in agricultural soils principally contributed by livestock and poultry manures.

GRAPHICAL ABSTRACT



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ABSTRACT

The application of livestock and poultry manures was the predominant source of heavy metals in agricultural soils, particularly in China. It is important to systematically compare the pollution characteristics, emission situations and mass loads for heavy metals in the manures of different livestock and poultry in China. According to analysis and estimation based on the reported concentration levels of eight heavy metals (Zn, Cu, Pb, Cd, Cr, Hg, As, and Ni) and the feed quantities of livestock (pig, cattle, and sheep) and poultry in 2017, the concentrations of Zn and Cu and the over-standard frequencies of Zn, Cu, Cd, and As were much higher than those of other heavy metals, especially in pig manure. In 2017, the total emission of livestock and poultry manure in China was 1.64×10^9 t (FW), which was mainly excreted from cattle (45.77%); while the total emission of heavy metals sourced from manures was 2.86×10^5 t (DW), with the predominant contribution originating from pig manure (71.52%). The highest mass loads of manures and heavy metals were observed in Shandong, Tianjin, Henan, and Shanghai, where heavy metal contamination may be occurring (especially for Zn and Cu). The heavy metal concentrations in livestock and poultry manures of China were similar to other countries; however, more heavy metals were discharged into agricultural land through manure (especially for Zn and Cu). For many countries,

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abundant Zn and Cu exist in agricultural soils, principally contributed by livestock and poultry manures. These heavy metals originate from their addition to livestock and poultry feeds. Therefore, reducing the addition of Zn and Cu in feeds is an effective measure to lower their input into agricultural soils.

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Contents

1. Introduction

In recent decades, the contamination of agricultural soils by heavy metals, including Zn, Cu, Pb, Cd, Cr, Hg, As, and Ni, has become the focus of attention in China (Leclerc and Laurent, 2017; Luo et al., 2009; Peng et al., 2019). Many studies have reported the pollution levels and corresponding ecological health risks of heavy metals in agricultural soils (Guan et al., 2018a; Jiang et al., 2017). Heavy metals have been found pervasively in the agricultural soils of China, with the average concentration of several mg/kg to tens of mg/kg [dry weight (DW)] for Zn, Cu, Pb, Cr, As, and Ni, while Cd and Hg occurred at concentrations of μg/kg (Shao et al., 2016; Song et al., 2013). Individually, the average concentrations of Zn and Cu could be up to hundreds of mg/kg in some heavily polluted farmlands, which may lead to high ecological health risks (Zhuang et al., 2009). To reduce the ecological health risks of heavy metals in the soil environment, their concentration thresholds in agricultural soils have been stipulated in the Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land (GB15618-2018) (MEEPRC, 2018). According to the national survey of soil contamination conducted by the Ministry of Environmental Protection (MEP) and Ministry of Land and Resources of China in 2005–2013, the concentrations of typical pollutants in 19.4% of sampling sites of agricultural soils exceeded their corresponding standard limits, and the dominating pollutants were heavy metals, with the order of frequency of above desired concentrations as Cd > Ni > As > Cu > Hg > Pb (MEPPRC and MLRPRC, 2014). The heavy metals exist in agricultural soils with long residence times (usually exceeding decades) and persistent bioavailability (Ding et al., 2017; Xiao et al., 2017). Many of these heavy metals can pose high ecological risks to soil organisms and crops due to their toxicity at low concentration levels (Abdu et al., 2017; Zhang et al., 2019), even threatening animal and human health through transmission and accumulation in the food chain (Zhang et al., 2018a; Zhang et al., 2015). Moreover, heavy metals could promote the occurrence of metal resistance genes (MRGs), and participate in the co-selection of antibiotic resistance genes (ARGs) (Guo et al., 2018; Zhou et al., 2016).

Given the risks associated with heavy metals, Pb, Cd, Cr^{6+}, Hg, As, and their compounds have been identified as priority control chemicals by the MEP and other two related ministries of China (MEPPRC, 2017). Chinese governments have also developed many management policies and treatment technologies to control heavy metal pollution of agricultural soils. One such policy promotes effective control through reducing the input of heavy metals from the source (PRC, 2018; SCPRC, 2016). The sources of heavy metals in agricultural soils are extensive, including atmospheric deposition, sewage irrigation and the application of sewage sludge, agrochemicals, fertilizers, and livestock and poultry manures (Luo et al., 2009; Peng et al., 2019; Shi et al., 2018). Previous studies have proven that the application of livestock and poultry manures was a dominant source (after the atmospheric deposition) for most heavy metals in agricultural soils (Guan et al., 2018b; Luo et al., 2009; Shi et al., 2018). Due to the nutrient rich nature of livestock and poultry manures, including nitrogen, phosphorus, and potassium, most manures were directly (no treatment) or indirectly (after general treatment) applied to agricultural lands (Qiu et al., 2012; Wang et al., 2017). However, general treatment (e.g., anaerobic digester and composting) could not effectively reduce the concentrations or bioavailability of heavy metals (Yang et al., 2017), even displaying enriched concentrations during digestate storage (Li et al., 2018). Therefore, long-term or excessive application of livestock and poultry manures could lead to heavy metal pollution of agricultural soils (Liu et al., 2014; Qian et al., 2018).

China has a large population and consumes large volumes of meat and other animal products. It is, therefore, a veritable breeding superpower. Livestock and poultry production have increased dramatically since the beginning of the 21st century. Nationwide, 7.02×10^8, 4.34×10^7, 3.08×10^8 and 1.30×10^{10} heads of fattened pig, cattle, sheep, and poultry were slaughtered in 2017, which increased by 35.36%, 14.01%, 56.70%, and 57.63% from 2000, respectively (NBSC, 2018). In the process of livestock and poultry breeding, a large amount of manures were produced, and most of them were applied to agricultural lands to improve soil fertility and increase crop productivity (Niu and Ju, 2017; Wu et al., 2018). To some extent, the popularity of utilizing livestock and poultry manure generated an increasing accumulation of some heavy metals (such as Cd and Hg) in agricultural soils over the last decade (Guo et al., 2018; Huang et al., 2019; Leclerc and Laurent, 2017). Due to the different diets and breeding of different livestock and poultry, the concentrations of heavy metals differ significantly in their manures (Shen et al., 2015; Wang et al., 2013). Many previous investigations focused on pig, chicken and cattle manure, while ignoring sheep and other poultry, which are consumed in large amounts in China (Qian et al., 2018; Zhang et al., 2012). In addition, many studies of the heavy metal residue in manures and the corresponding contamination of agricultural soils were focused on specific heavy metals (Li et al., 2010; Xiong et al., 2010), or studied limited regions of China (Guan et al., 2018a; Hou et al., 2014; Shi et al., 2019). Therefore, it was necessary to systematically investigate the discharge of eight hazardous heavy metals from the manure of different livestock and poultry across China, and then evaluate the effects on the quality of agricultural soils.

In this study, the concentration levels and pollution characteristics of heavy metals in the different livestock and poultry manures of China were assessed according to data collected from relevant literature. The manure emission and mass load in every province of China were estimated based on the updated feeding numbers of different livestock and poultry in 2017. Then the concentration data of heavy metals and emissions of manures were combined to estimate the emissions and mass loads for heavy metals in the manures of different livestock and poultry. In addition, the concentration levels, emissions, and mass loads of heavy metals in/from livestock and poultry manures of China and other countries were compared during the analysis and assessment. The results of this study will provide an important reference for researchers and regulators, helping them develop effective measures to alleviate heavy metal pollution of agricultural soils resulting from usage of livestock and poultry manures.

2. Materials and methods

2.1. Data sources

Basic data used in this study included the feeding numbers of different livestock (pig, cattle, and sheep) and poultry, the area of agricultural land and average concentrations of heavy metals in the manures of different livestock and poultry in China. The feeding number of livestock and poultry and the area of agricultural land in every province of China (Table S1, Supplementary Material) were cited from the China Rural Statistical Yearbook (2018), compiled by the National Bureau of Statistics of China (NBSC, 2018). The average concentrations of heavy metals in the manures of different livestock and poultry were cited from the investigations undertaken in the different provinces of China. Out of 31 provinces, data were obtained for 29 provinces for pigs (Table S2), 28 for poultry (Table S3), 28 for cattle (Table S4), and 24 for sheep (Table S5). The data were sourced from 47 references from 2004–2018, and collected from Web of Science, Science Direct, and China National Knowledge Internet (CNKI).

2.2. Estimation methods

2.2.1. Estimation for the emission and mass load of manure

The manure emissions of different livestock and poultry can help to reveal the potential pollution profile of each breeding industry, and they are the basic data used for estimating the emissions and mass loads for heavy metals from the manures of different livestock and poultry. Based on the method of excretion coefficient, the manure emissions of different livestock and poultry in every province of China were estimated using the following equation:

$$M = (N \times K \times D \times f) \times P \quad (1)$$

where, M [t, fresh weight (FW)] and N ($\times 10^4$ heads) represent the manure emissions and the feeding number of certain livestock (pig, cattle, and sheep) or poultry in a certain province of China in 2017; K (kg/d/head, FW) is the excretion coefficient of the manure for per unit animal; D (d) stands for the feeding period of animal; f is a conversion factor for unifying the units, with the value of 10; P (%) is the percentage of animal manure returned into agricultural land. Given that the feeding numbers of different livestock and poultry vary, the slaughtered number during a year or feeding numbers at year-end were used as stable feeding numbers for different livestock and poultry. For cattle (including beef and cow), the feeding period lasted longer than a year, and the feeding numbers at year-end were greater than the number slaughtered during a year for most provinces of China; therefore, the feeding number at year-end was used to estimate eq. (1). However, for mutton sheep, pigs, and poultry, the feeding period was shorter than a year, and their feeding numbers at year-end were lower than the number slaughtered during a year for most provinces of China; therefore, the number slaughtered during a year were used to estimate eq. (1). The corresponding statistical data of 2017 is listed in Table S1. The excretion coefficient of the animal manure (K), feeding period of animal (D), and percentage of animal manure returned into agricultural land (P) were taken as the mean of the reported values sourced from the relevant references (Table 1).

The mass load of manure (t/hm^2, FW) from livestock and poultry in a certain province of China was calculated through the total manure emissions ($\sum M$, t, FW) of all the livestock (pig, cattle, and sheep) and poultry divided by the area of agricultural land (hm^2) in the corresponding province of China in 2017 (Table S1).

2.2.2. Estimation for the emissions and mass loads of heavy metals in manure

The emissions for heavy metals in the manures of different livestock and poultry were estimated based on the emissions and water content of the manure, and the mean concentrations of heavy metals in the manure, as follows:

$$H = M \times (1 - W) \times C \times f' \quad (2)$$

where, H (t, DW) represents the emission of a particular heavy metal in the manure of certain livestock (pig, cattle, and sheep) and poultry in a certain province of China in 2017, and M (t, FW) represents the

Table 1
Parameters used for the estimation of the emissions for the manure and heavy metals among manures for different livestock and poultry.

Category	Pig	Cattle	Sheep	Poultry	Reference
Excretion coefficient of the manure for per unit animal (kg/d/head, FW) [a]	3.34	25.33	2.16	0.13	Niu and Ju, 2017
Feeding period for animal (d) [a]	222	365	304	203	Niu and Ju, 2017
Water content of animal manure (%)	73	85	70	75	Peng et al., 2019
Percentage of animal manure returned into agricultural land (%) [b]	90	like pig	like pig	70	Qiu et al., 2012

[a] The parameters in these lines were the mean of the reported value in the relevant references;
[b] These parameters were the mean of investigation results in 2005 and 2010.

Table 2
Descriptive statistics for the average concentrations (mg/kg, DW) of heavy metals in different livestock and poultry manures.

Manure source	Category	Zn	Cu	Pb	Cd	Cr	Hg	As	Ni
Pig	Minimum	100.26	72.66	0.27	0.04	3.53	0.00	0.01	4.67
(n = 2352)	Maximum	4638.72	1288.00	22.88	59.66	85.23	0.31	89.30	18.97
	Mean	1019.98	531.37	8.63	2.97	26.80	0.10	16.47	10.69
	Sample size [a]	>2209	>2319	>1929	>1984	>1845	>1248	>1686	>770
Chicken	Minimum	165.68	18.24	2.99	0.03	4.00	0.02	0.05	5.21
(n = 629)	Maximum	578.00	314.00	32.58	4.09	250.61	0.54	23.26	39.31
	Mean	346.17	96.05	12.43	1.34	58.93	0.13	6.88	13.93
	Sample size	>629	>625	>578	>581	>572	>446	>550	>278
Duck	Minimum	97.82	34.68	4.51	0.29	6.60	0.03	0.01	8.37
(n = 63)	Maximum	682.10	198.76	40.79	2.53	63.61	0.07	6.83	16.12
	Mean	339.11	71.78	14.47	0.97	30.37	0.05	3.99	10.77
	Sample size	63	63	60	60	60	40	50	50
Poultry [b]	Minimum	77.42	14.71	2.04	0.03	2.50	0.02	0.01	5.21
(n = 879)	Maximum	682.10	314.00	40.79	4.09	250.61	0.54	23.26	39.31
	Mean	322.20	84.42	12.48	1.19	50.60	0.11	5.99	13.88
	Sample size	>879	>875	>750	>753	>734	>573	>694	>433
Cattle	Minimum	48.72	12.28	1.64	0.04	0.76	0.02	0.01	4.19
(n = 565)	Maximum	816.24	173.60	32.31	3.40	79.38	0.60	6.33	18.86
	Mean	221.27	56.93	11.72	0.84	24.53	0.12	2.09	10.10
	Sample size	>565	>561	>479	>466	>452	>363	>435	>246
Sheep	Minimum	42.38	8.37	1.74	0.28	8.00	0.19	0.59	1.22
(n = 116)	Maximum	431.70	214.70	19.80	1.40	22.19	2.39	2.60	12.40
	Mean	178.83	60.89	11.14	1.09	12.27	0.78	1.53	7.62
	Sample size	116	116	67	67	59	59	59	43

[a] Represent the sample size with measure concentration for this heavy metal among the total sample size (n);
[b] Except above chicken and duck manures, including 10 goose manures (2 average concentrations), 9 dove manures (2 average concentrations) and 168 poultry manures (3 average concentrations).

corresponding manure emission, sourced from the results of 2.2.1; W (%) is the average water content of certain livestock and poultry manures, taken from previous reports (Table 1); C (mg/kg, DW) is the mean concentration of a particular heavy metal in the manure of certain livestock and poultry in China (Table 2); and f is a conversion factor for unifying the units, with the value of 10^{-6}.

Similarly, the mass load of heavy metals (g/hm^2, DW) from livestock and poultry manure in a certain province of China was calculated through the total emission ($\sum$H, g, DW) for a particular heavy metal in the manure of all the livestock (pig, cattle and sheep) and poultry divided by the area of agricultural land(hm^2)in the corresponding province of China in 2017 (Table S1).

3. Results and discussion

3.1. Concentration levels for heavy metals in the manures of different livestock and poultry

The original data and references for the average concentration of heavy metals in different livestock and poultry manure in China are listed in Table S2 ~ S5. Considering the vast pig feeding number and manure emission in China, the study on the occurrence of heavy metals in pig manure was more popular than those of other livestock and poultry. Many relevant investigations were conducted by Chinese scholars, with principal focus on provinces with plentiful pig breeding, such as Zhejiang, Jiangsu, Guangdong, and Shandong. The total sample size of pig manure from all these studies was 2352 samples, which was a significantly larger dataset compared to other manures (63–879). For all the manure samples of different livestock and poultry, the measured concentrations of Zn and Cu were greater than those of other heavy metals, indicating that the residual problems of Zn and Cu in manures requires additional attention.

The average concentrations listed in Table S2 ~ S5 are used to determine the summarized concentration levels (minimum, maximum, and mean), as shown in Table 2. Eight heavy metals (Zn, Cu, Pb, Cd, Cr, Hg, As, and Ni) occurred extensively in the manure of livestock (pig, cattle, and sheep) and poultry (chicken, duck, goose, and dove), and their mean concentrations were 178.83–1019.98 mg/kg for Zn, 56.93–531.37 mg/kg for Cu, 8.63–14.47 mg/kg for Pb, 0.84–2.97 mg/kg for Cd, 12.27–58.93 mg/kg for Cr, 0.05–0.78 mg/kg for Hg, 1.53–16.47 mg/kg for As, and 7.62–13.93 mg/kg for Ni. For pig manure, the mean concentrations of heavy metals decreased in the order of Zn>Cu>Cr>As>Ni>Pb>Cd>Hg, while those for cattle and sheep was Zn>Cu>Cr>Pb>Ni>As>Cd>Hg, and for poultry was Zn>Cu>Cr>Ni>Pb>As>Cd>Hg. Overall, the concentrations of Zn and Cu were significantly higher than those of the other six heavy metals studied (especially in pig manure), with the reported highest average concentrations of 4638.72 mg/kg and 1288.00 mg/kg, respectively, both occurring in pig manure.

The majority of the literature listed in Table S2 ~ S5 only reported detailed concentrations of heavy metals in the various livestock and poultry manures of China, without systematically comparing these concentration levels with other countries. We have therefore supplemented this with data collected from literature (listed in Table S6). The results are illustrated in Fig. 1. The average concentrations of heavy metals in the Chinese manure samples of China lay at median levels when compared with those of other countries, for the majority of manures of livestock and poultry (except for Zn, Cu, and Cr in sheep manure). In general, the concentrations of Zn and Cu in animal manures were higher than other heavy metals, and followed the order of pig>poultry>cattle>sheep. This phenomenon was observed in the majority of countries, such as China, Austria, Canada, England & Wales, and Germany. The major focus of manure studies in other countries was cattle, whereas in China it was pigs. This difference may be attributed to differences in farming structures of livestock and poultry and dietary habits of people in different countries.

The high concentrations of Zn and Cu observed in the manure of livestock and poultry were found to be related to their abundant addition in feeds (Jensen et al., 2018; Jiang et al., 2011). A number of studies revealed that the average concentrations of Zn and Cu in the various feeds were high, at tens to hundreds of mg/kg (DW), while the highest values were up to thousands of mg/kg, significantly higher than other heavy metals (Wang et al., 2013; Zhang et al., 2012). Compared to cattle and sheep, the feeds for pig and poultry contained more Zn and Cu and sold more in the market (especially in China) (CIDRN, 2017; Ni, 2017; Sager, 2007), suggesting that the farming of pig and poultry required more feed than cattle and sheep. This result may explain why the

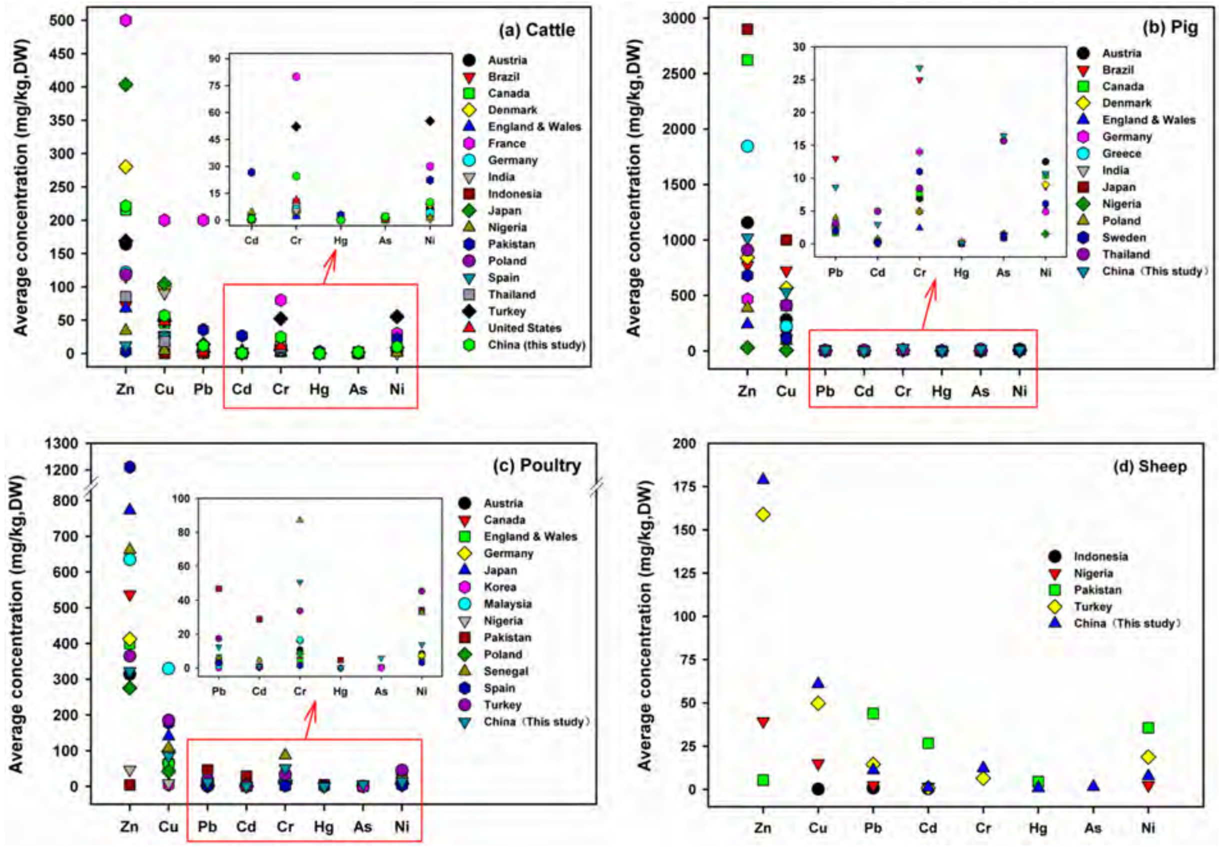


Fig. 1. Concentration comparisons of heavy metals in the different livestock and poultry manures of China and other countries [(a) Cattle including cow, beef, buffalo; (b) Pig including feeder, nursery, and sow pig; (c) Poultry including broiler, layer, turkey, duck etc.; (d) Sheep including goat].

concentrations of Zn and Cu in the manures of pig and poultry were higher than those of cattle and sheep. Overall, the occurrence of Zn and Cu in livestock and poultry manures (especially in pig manure) were more extensively investigated than other heavy metals in China, and their concentrations were also higher than others, implying that the Zn and Cu pollution in agricultural soils may be caused by the discharge or application of livestock and poultry manures.

3.2. Pollution characteristics for heavy metals in the manures of different livestock and poultry

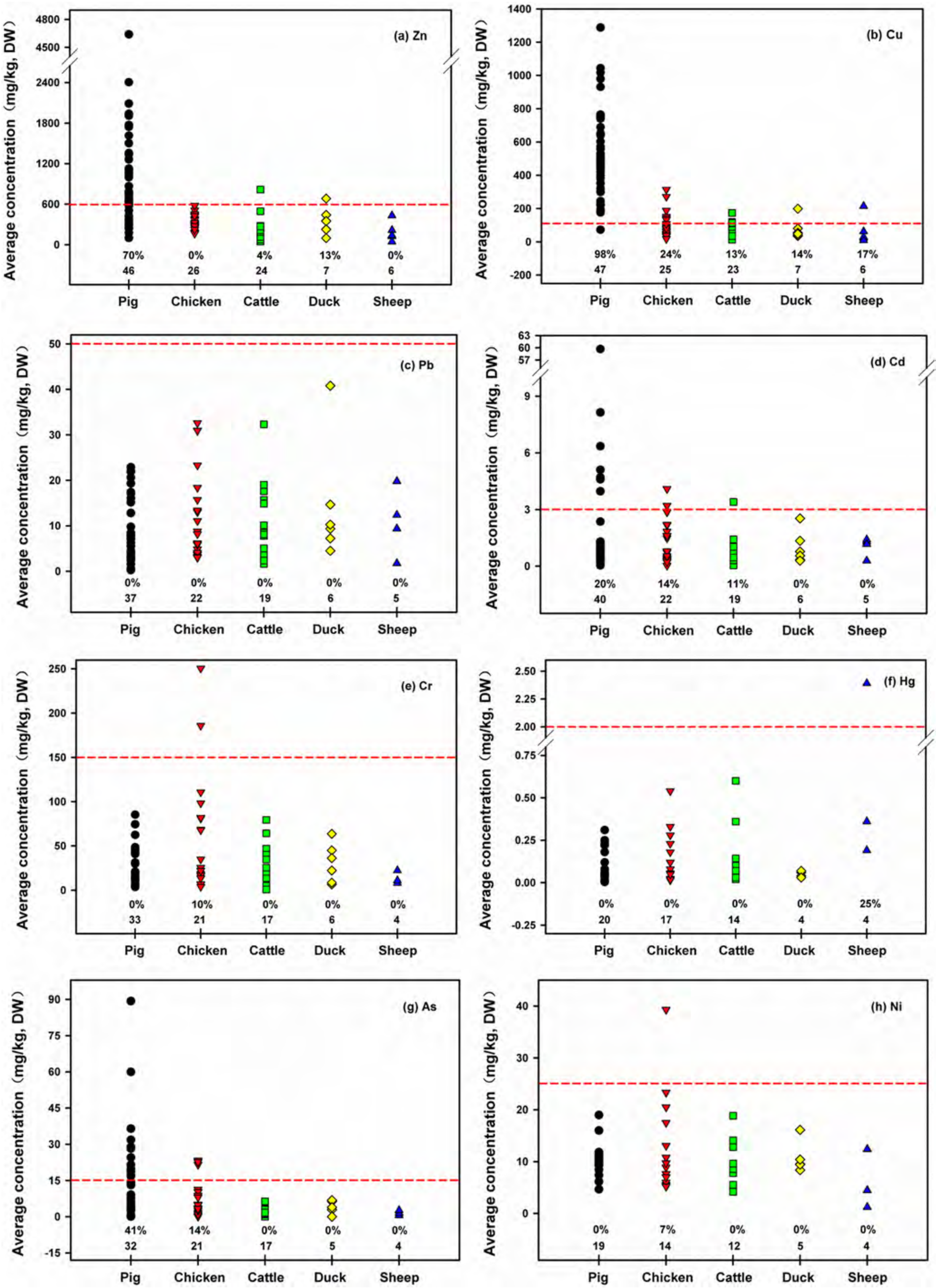
The distribution of the average concentrations of eight heavy metals (Zn, Cu, Pb, Cd, Cr, Hg, As, and Ni) in the manures of livestock (pig, cattle, and sheep) and poultry (chicken and duck) in China are shown in Fig. 2, and the over-standard situation of these heavy metals were assessed based on the Chinese agricultural industry standard of organic fertilizer (NY 525–2012, for Pb, Cd, Cr, Hg, and As) and the standard of organic fertilizer in Taiwan province, China (for Zn, Cu, and Ni) (MAPRC, 2012; Zhu et al., 2013). As a result, seven eighths of heavy metals were observed to be higher than allowed values in the manure of these livestock and poultry. For Zn [Fig. 2 (a)], the over-standard phenomenon of average concentrations occurred in the manure of pig, cattle, and duck, with the over-standard frequencies of 70%, 4%, and 13%, respectively. For Cu [Fig. 2 (b)], the average concentration in the manure of pig, chicken, cattle, duck, and sheep were all presented over-standard phenomenon, and the over-standard frequency in pig manure was 98%, which was dramatically higher than those of other livestock and poultry (13% ~ 24%). For Cd [Fig. 2 (d)], average concentrations in the manure of pig, chicken, and cattle exceeded the standard value, and the over-standard frequencies ranged from 11% to 20%. For As [Fig. 2 (g)], 41% and 14% average concentrations in the manure of pig and chicken were distributed over the line of standard value. For Cr and Ni [Fig. 2 (e and h)], the over-standard phenomenon of average concentrations both occurred in the manure of chicken, and the over-standard frequencies were 10% and 7%. For Hg [Fig. 2 (f)], 25% average concentrations were over-standard in the manure of sheep. Specially, no over-standard phenomenon was displayed for Pb [Fig. 2 (c)].

Among eight heavy metals, the heavy metals with high over-standard frequencies were Zn, Cu, Cd, and As, which primarily occurred in pig manure. Overall, the pollution profile of heavy metals in pig manure was more serious than those of other livestock and poultry manures in China. These results are consistent with previous reports (Bo et al., 2018; Zhu et al., 2013). Furthermore, the over-standard of heavy metals in livestock and poultry manures directly led to their over-standard in commercial organic fertilizers made from these manures (Ding et al., 2017; Yang et al., 2017). The concentration thresholds of five heavy metals (Pb, Cd, Cr, Hg and As) with high toxicity were set up in the agricultural industry standard about the organic fertilizer (NY 525–2012), which published by the Ministry of Agriculture of the PRC (MAPRC, 2012). However, this standard did not consider three other heavy metals (Zn, Cu, and Ni), which were detected frequently with high concentrations both in manures and commercial organic fertilizers (Ding et al., 2017; Liu et al., 2005). Therefore, it was important to assess systematically the ecological health risks of Zn, Cu, and Ni to agroecosystems and humans, in order to establish suitable concentration thresholds in the revised national standard of organic fertilizer (Zhang et al., 2018b).

3.3. Emission situation and mass load for the manures of different livestock and poultry

3.3.1. Emission situation for the manure

The emission situation for the manures of different livestock and poultry is shown in [Fig. 3 (a)]. Across China (31 provinces), the


(a) Zn
Average concentration (mg/kg, DW)
70% 0% 4% 13% 0%
46 26 24 7 6
Pig Chicken Cattle Duck Sheep
(b) Cu
98% 24% 13% 14% 17%
47 25 23 7 6
(c) Pb
0% 0% 0% 0% 0%
37 22 19 6 5
(d) Cd
20% 14% 11% 0% 0%
40 22 19 6 5
(e) Cr
0% 10% 0% 0% 0%
33 21 17 6 4
(f) Hg
0% 0% 0% 0% 25%
20 17 14 4 4
(g) As
41% 14% 0% 0% 0%
32 21 17 5 4
(h) Ni
0% 7% 0% 0% 0%
19 14 12 5 4

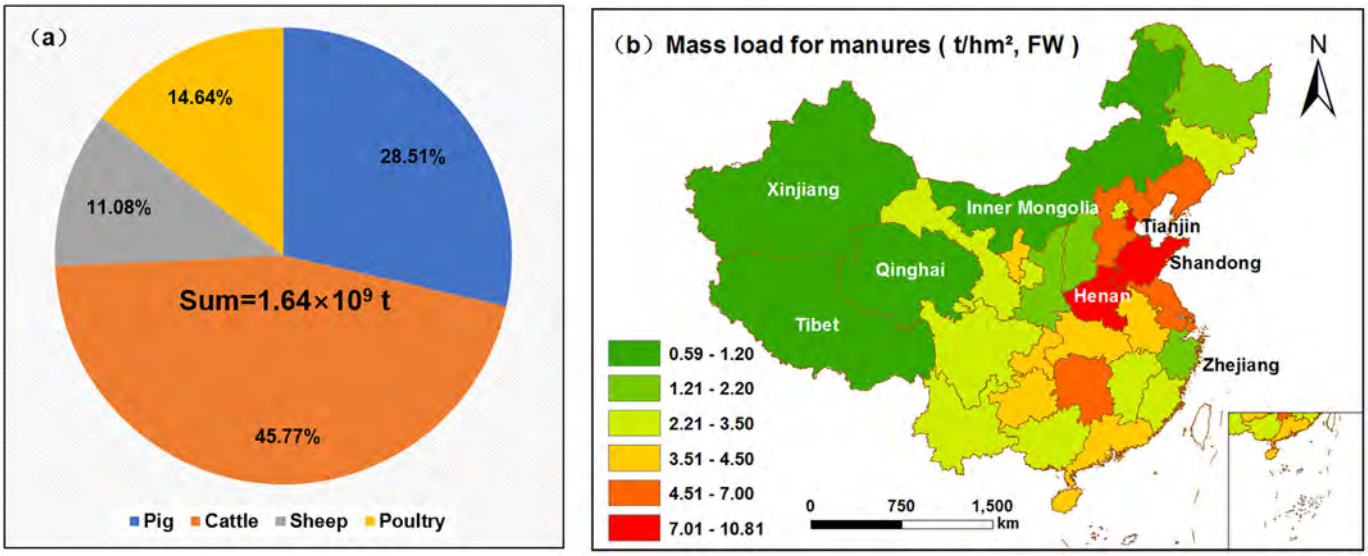


Fig. 3. Emission situation and mass load (FW) for the manures of different livestock and poultry in the whole of China in 2017 [(a) the total emission of manures and its contribution proportion from different livestock and poultry; (b) the distribution of mass load for manures in 31 provinces].

cumulative total emission for the manures of different livestock (pig, cattle and sheep) and poultry in 2017 was as high as 1.64×10^9 t (FW), and the emission from cattle accounted for 45.77%, higher than the contributions of pig (28.51%), poultry (14.64%), and sheep (11.08%). According to another estimation under the same statistical range in a similar study (no heavy metals involvement), the total emission for the manures and urine of different livestock (pig, cattle, and sheep) and poultry was 1.91×10^9 t in 2015, and this value for manure alone was 1.019×10^9 t (Wu et al., 2018), which was slightly lower than this study (1.64×10^9 t, 2017). This similar study revealed that the contribution proportion of manure and urine from different livestock and poultry conformed to the following order: cattle (48.3%) > pig (33.9%) > sheep (13.1%) > poultry (4.7%) (Wu et al., 2018). The proportions for the individual manure contributions were not mentioned. If a larger contribution from the urine of sheep is deducted when compared to poultry, the above order may be in line with this study. Generally, the manure emissions of different livestock and poultry are effected by the feeding numbers, manure excretion coefficients, and feeding periods of different animals (Luo et al., 2009; Peng et al., 2019). The total feeding numbers of livestock and poultry were stable from 2015 to 2017 in China (NBSC, 2016, 2018); therefore, the slow rise of total manure emission from 2015 to 2017 was probably caused by the difference of manure excretion coefficients and feeding periods adopted in the two estimation. The manure excretion coefficient and feeding period of cattle were much higher than those of other livestock and poultry (Table 1); therefore, cattle manure presented the highest contribution to the total manure emission.

3.3.2. Mass load for the manure

In 2017, the mass loads for the manures of different livestock and poultry in the 31 provinces of China ranged from 0.59–10.81 t/hm^2 (FW) [Fig. 3 (b)], with a mean value of 3.81 t/hm^2 (Table S7). The mass loads of Shandong (10.81 t/hm^2), Tianjin (8.08 t/hm^2), Henan (8.06 t/hm^2), and 10 other provinces (3.85–6.94 t/hm^2) were higher than this mean value; while the 18 remaining provinces were lower than this mean value (0.59–3.74 t/hm^2). Xinjiang, Inner Mongolia, Qinghai, and Tibet incurred relatively low mass loads, with the value of 1.20 t/hm^2, 1.20 t/hm^2, 1.12 t/hm^2, and 0.59 t/hm^2, respectively. Theoretically, the mass load for the manure was directly proportional to the feeding number of livestock and poultry, while inversely proportional to the area of agricultural land (Zhang et al., 2009). Shandong and Henan fed a great number of livestock and poultry, and Tianjin has limited agricultural land (Table S1), resulting in high mass loads for manure. Conversely, Xinjiang, Inner Mongolia, Qinghai, and Tibet possess capacious agricultural land area and relatively small feeding numbers of livestock and poultry (Table S1), resulting in low mass loads for manure. Across China, the mass load of livestock and poultry manures in the southeastern region was greater than those of the northwestern region. Individually, a relatively low manure mass load was found in the southern province Zhejiang, which may be related to the rapid decrease of the feeding number for livestock and poultry, due to the designation of vast zones where breeding was prohibited in order to avoid the pollution of livestock and poultry breeding in recent years (ZJGOV, 2015).

To date, the Chinese government has not set application limits for livestock and poultry manures in agricultural land; however, some scholars regarded 30 t/hm^2 as the maximum mass load for the manures (Wang et al., 2006; Wu et al., 2018). This maximum (30 t/hm^2) was much higher than the estimated mass loads for the manures in 31 provinces of this study (0.59–10.81 t/hm^2), which did not indicate that there remained capacious application space for the livestock and poultry manures in the agricultural land of China. The maximum (30 t/hm^2) was estimated based on the nutrient supply for nitrogen and phosphorus (Wang et al., 2006; Wu et al., 2018), and did not consider the residual of antibiotics, heavy metals, pathogens and other hazardous substances in livestock and poultry manures (Guo et al., 2018; Leclerc and Laurent, 2017). In addition, the feeding numbers (slaughtered or year-end) of livestock and poultry adopted in this study were lower than the factual numbers due to animals being slaughtered (during a year) and stockpiled (in year-end). Animals were fed at the same time (unavailable), and other livestock (except pig, cattle, and sheep) were ignored in this study since their feeding numbers were unavailable in the statistical yearbook. The above two reasons could lead to larger emissions and mass loads for manures than the estimation in this study. Therefore,

Fig. 2. Distribution and over-standard situation for the average concentration (DW) of heavy metals in the manures of different livestock and poultry. In this figure, the dotted lines represent the limited standards of heavy metals in organic fertilizer, the standard value for Pb, Cd, Cr, Hg and As come from the agricultural industry standard for organic fertilizer (NY 525–2012) applied in China (MAPRC, 2012), while the standard value for Zn, Cu and Ni come from the standard of organic fertilizer in Taiwan province, China (Zhu et al., 2013); The Arabic numbers above the abscissa represent the amount of the average concentrations, while the percentages represent the over-standard frequencies of these average concentrations.

a further study on the suitable mass loads of manures in agricultural land should be performed based on the primary hazardous substances in all livestock and poultry manures.

3.4. Emission situations and mass loads for heavy metals in the manures of different livestock and poultry

3.4.1. Emission situations for heavy metals

The discharge of heavy metals into the receiving environment through livestock and poultry manures in China is shown in Fig. 4. In 2017, the emission for eight heavy metals (Zn, Cu, Pb, Cd, Cr, Hg, As, and Ni) derived from the manures of pig, cattle, sheep, and poultry obeyed the following order: Zn (1.83×10^5 t)>Cu (8.20×10^4 t)>Cr (9.87×10^3 t)>Pb (3.77×10^3 t)>Ni (3.74×10^3 t)>As (2.76×10^3 t)> Cd (602 t)>Hg (75 t) (DW), and the sum emission of eight heavy metals ($\sum$8HMs) was up to 2.86×10^5 t (DW). Owing to the difference of manure emissions and heavy metals concentrations in manures, a conspicuous diversity for the contribution of heavy metals from pig, cattle, sheep, and poultry was observed. For Zn, Cu, Cd, and As, their emission was principally contributed by pig manure, whose contributions were as high as 70.45%, 81.93%, 62.45%, and 75.40%, respectively [Fig. 4 (a, b, d and g)]. For Pb, the contribution proportions from different animal manures were similar (16.12% ~ 35.05%), with the maximum originating from cattle manure [Fig. 4 (c)]. For Cr, those contribution proportions occurred in the order of pig (34.34%) > poultry (30.83%) > cattle (28.04%) > sheep (6.79%) [Fig. 4 (e)]. Although the proportions were closer, the contributions for Ni were: pig (36.13%) > cattle (30.45%) > poultry (22.30%) > sheep (11.12%) [Fig. 4 (h)]. A distinct difference from other heavy metals was that the biggest contribution of Hg was derived from sheep manure, with a proportion of 54.69% [Fig. 4 (f)]. Overall, the contributions from different livestock and poultry manures to the $\sum$8HMs were ranked as pig (71.52%) >> cattle (12.92%) > poultry (10.32%) > sheep (5.23%) [Fig. 4 (i)]. These data suggest that agricultural land pollution of heavy metals was mostly contributed by pig manure input (except for Pb and Hg), compared to cattle, sheep, and poultry manures.

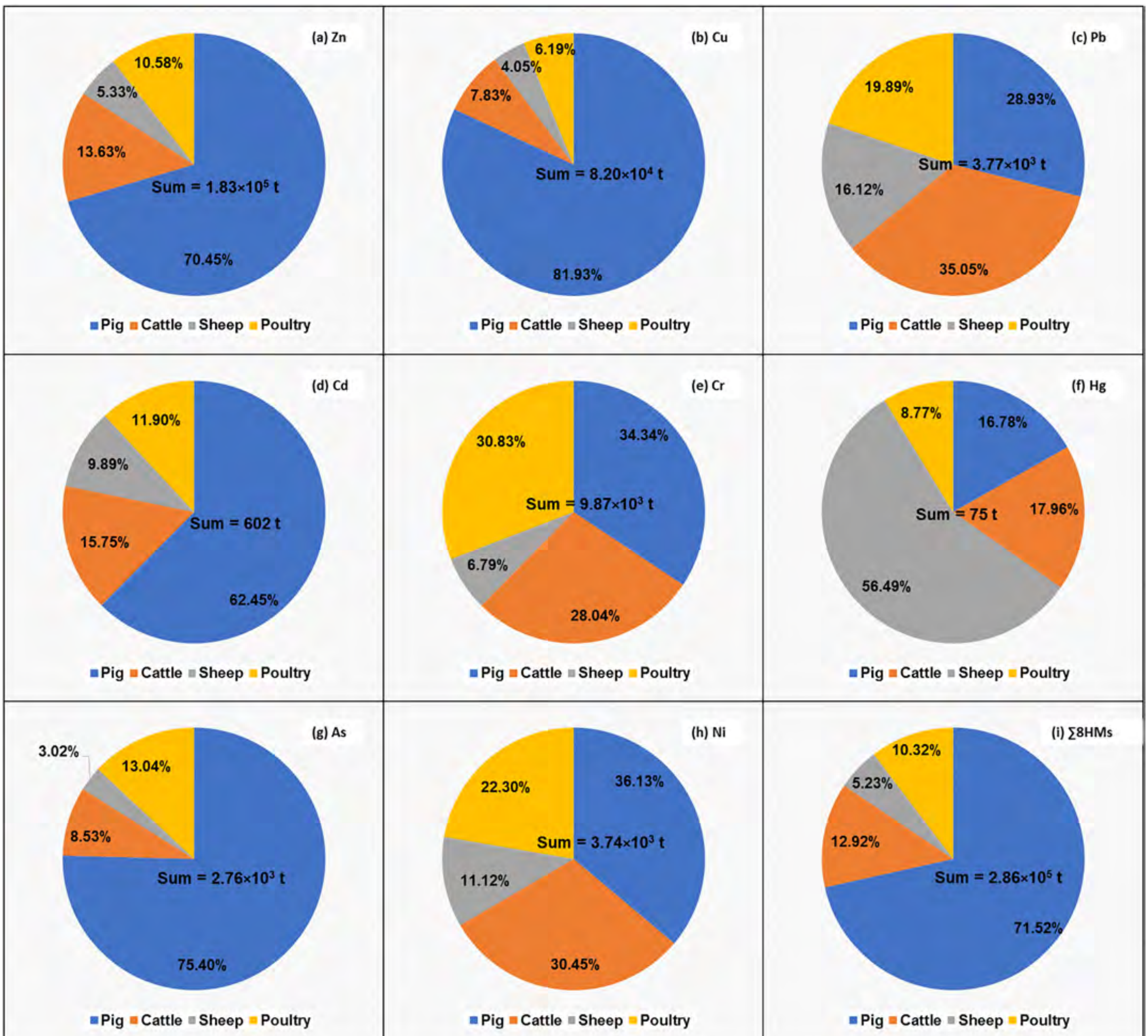


Fig. 4. Emission (DW) of each heavy metal from different livestock and poultry manures in the whole of China in 2017.

Table 3
Emission comparisons of heavy metals from the manures of different livestock and poultry between China and other countries.

Country	Year of data	Manure source	Zn	Cu	Pb	Cd	Cr	Hg	As	Ni	Reference
China	2017	Pig, cattle, sheep, poultry	183,122	82,037	3773	602	9870	75	2763	3742	This study
China	2016	Pig, cattle, sheep, poultry, horse, donkey, mule	135,667	64,030	1800	225	3964	36	2221	2051	Peng et al., 2019
China	2005	Pig, cattle, sheep, poultry, horse, donkey, mule	95,668	49,229	2594	778	6113	23	1412	2643	Luo et al., 2009
France	2012	Pig, cattle, sheep, poultry, horse	15,190	4869	696	54	1001	12	134	568	Belon et al., 2012
England & Wales	2000	Pig, cattle, sheep, poultry	1858	643	48	4.2	36	0.3	16	53	Nicholson et al., 2003
United Kingdom	2000	Pig, cattle, sheep, poultry	4200	909	97	6.5	203	2.1	18	119	Leclerc and Laurent, 2017
Poland	2003	Pig, cattle, chicken	1681	393	137	7	382	–	–	240	Calculate based on Dach and Starmans, 2005 [a]
Netherlands	2003	Pig, cattle, chicken	1250	425	–	3	–	–	–	–	Dach and Starmans, 2005

[a] Calculate through the heavy metal mass load multiplied by the agricultural land area of Poland in 2002, which reported in this literature.

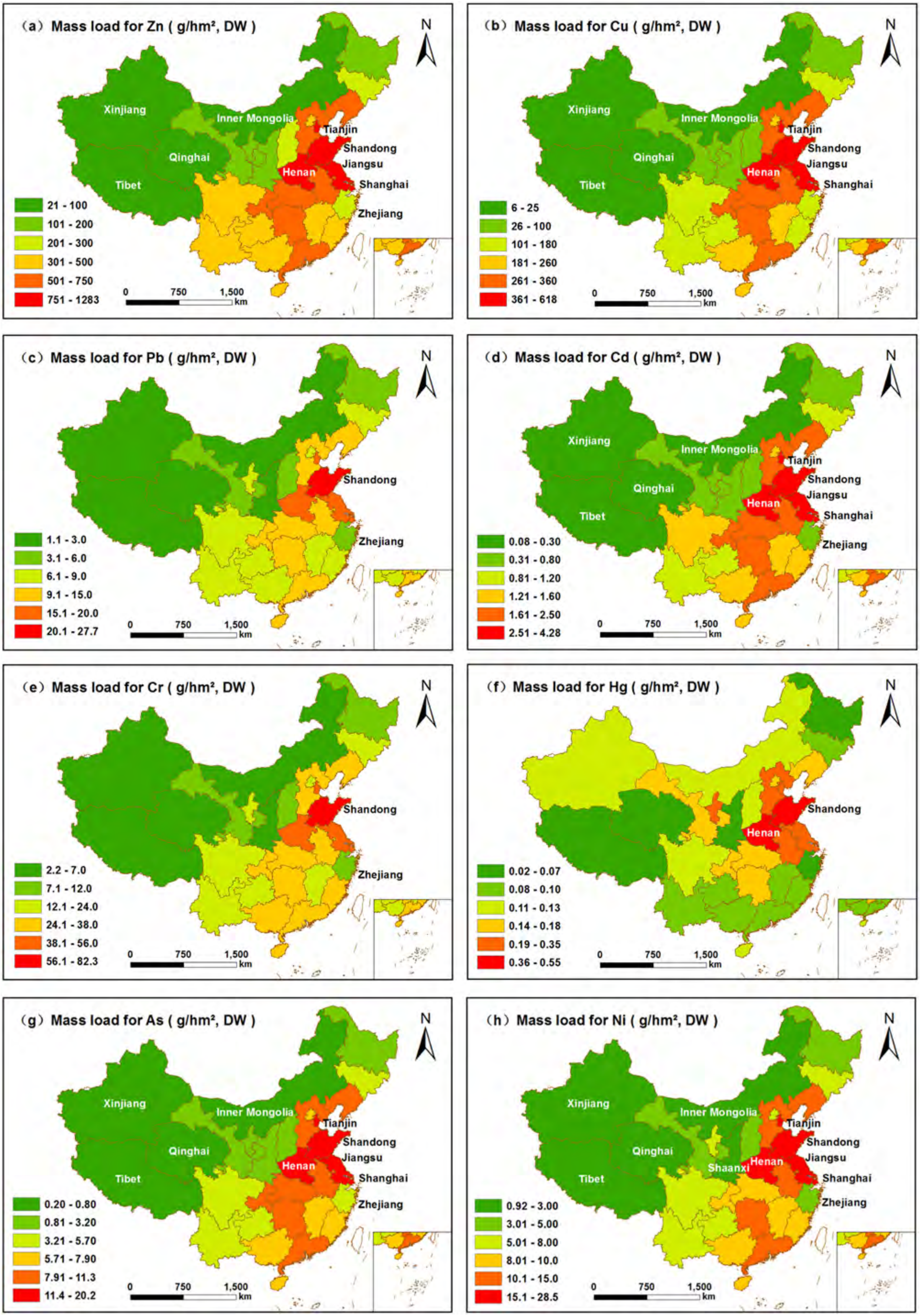
Previous literature also reported heavy metal emissions from livestock and poultry manures at home and abroad. As shown in Table 3, these reports from different countries included data from different years and different animals, using different estimated parameters and methods, which might lead to weak comparisons of the heavy metal emissions between different countries. However, clear observations could be made from these emissions: (1) the heavy metal emissions in China were much higher than those of other countries, which was mainly because the feeding number of livestock and poultry in China was largely greater than other countries (Dach and Starmans, 2005; Luo et al., 2009). (2) For the majority of countries, the heavy metal emissions occurred with the following order: Zn > Cu > Cr > Ni ≈ Pb > As > Cd > Hg. For China (Luo et al., 2009; Peng et al., 2019), the heavy metal emissions increased from 2005 to 2017 (except for Cd), mainly due to the augmentation of livestock and poultry feeding number and heavy metal concentrations in manures (NBSC, 2006; NBSC, 2018; Peng et al., 2019). Even though horse, donkey, and mule manures were not included in this study, the emissions of eight heavy metals from these manures in 2017 were higher than those of 2016 in the reports of Peng et al. (2019), which might be caused by the difference of estimated parameters used in these two studies, mainly including the excretion coefficients for animal manures, feeding periods for animals, and heavy metal concentrations in manures.

The heavy metals in manures originate mainly from the daily diets of livestock and poultry (Jiang et al., 2010; Zhang et al., 2012). Many investigation have confirmed that some heavy metals (especially for Zn, Cu, and As) were artificially added in commercial feeds to help animals by promoting growth and improving disease resistance (Hu et al., 2018; Jiang et al., 2011). However, these heavy metals cannot be fully absorbed by the animals and some is discharged through manures (Jensen et al., 2018; Li and Chen, 2005). It was reported that the usage amount of trace elements in China was up to 300,000 to 380,000 t every year; however, approximately 95% remained as an unabsorbed portion and was excreted with manures and urine due to the low bioavailability (Wang et al., 2016). With the continuous expansion of intensive breeding, more heavy metals might be added into feeds for achieving greater short term economic benefits (Wang et al., 2013). This behavior might be one of the explanations for the gradual increase of Zn, Cu, and As concentrations in livestock and poultry manures from 1999–2018 (Peng et al., 2019). Moreover, intensive breeding (especially for pigs) required more feed than family breeding, resulting in increased emissions of heavy metals (especially for Zn, Cu, and As) through manures (Shan and Zhang, 2012; Xie et al., 2014). As verified above in this study, pig manures contributed more emissions of Zn, Cu, and As (>70%) than cattle, sheep, and poultry manures [Fig. 4 (a, b, and g)]. Therefore, reducing the additions of heavy metals in feeds and the usage of feeds in intensive breeding (especially for pig) would be an effective method to cut down the emission of heavy metals from livestock and poultry manures (Xu et al., 2019).

Table 4
Mass load comparisons of the heavy metals from the manures of different livestock and poultry between China and other countries.

Country [a]	Year of data	Manure source	Zn	Cu	Pb	Cd	Cr	Hg	As	Ni	Reference
China	2017	Pig, cattle, sheep, poultry	283.97	127.22	5.85	0.93	15.31	0.12	4.28	5.80	This study
China	2016	Pig, cattle, sheep, poultry, horse, donkey, mule	210.30	99.25	2.79	0.35	6.14	0.06	3.44	3.18	Calculate based on Peng et al., 2019
China	2005	Pig, cattle, sheep, poultry, Horse, Donkey, Mule	735.69	378.57	19.95	5.98	47.01	0.18	10.86	20.32	Calculate based on Luo et al., 2009
France [b]	2012	Pig, cattle, sheep, poultry, horse	1828.32	369.15	90.38	1.59	54.09	0.75	18.91	43.42	Calculate based on Belon et al., 2012 [c]
Canada	2009	Livestock and poultry	1500.00	300.00	1.30	0.34	–	–	0.00	–	Sheppard et al., 2009
Poland	2003	Pig, cattle, chicken	91.64	21.44	7.47	0.38	20.82	–	–	13.07	Dach and Starmans, 2005
Netherlands	2003	Pig, cattle, chicken	641.35	218.06	–	1.54	–	–	–	–	Dach and Starmans, 2005
Switzerland	2001	Pig, cattle, poultry	9495.40	5.00	–	–	–	–	–	–	Calculate based on Keller et al., 2001 [d]
China	2017	Pig	200.07	104.23	1.69	0.58	5.26	0.02	3.23	2.10	This study
England & Wales	2000	Pig	2210.00	1488.00	27.00	2.00	22.00	0.10	8.70	48.00	Nicholson et al., 2003
China	2017	Cattle	38.71	9.96	2.05	0.15	4.29	0.02	0.37	1.77	This study
China	2017	Sheep	15.14	5.16	0.94	0.09	1.04	0.07	0.13	0.65	This study
England & Wales	2000	Cattle & Sheep	718.00	168.00	27.00	2.70	20.00	0.20	11.90	28.00	Nicholson et al., 2003
China	2017	Poultry	30.05	7.87	1.16	0.11	4.72	0.01	0.56	1.29	This study
England & Wales	2000	Layer	2734.00	422.00	42.00	6.10	27.00	0.10	2.20	47.00	Nicholson et al., 2003
England & Wales	2000	Other poultry	1142.00	175.00	18.00	2.60	11.00	0.10	1.90	20.00	Nicholson et al., 2003

[a] The mass loads for China and France were calculated through the heavy metal emission of whole nation divided by the agricultural land area of whole nation in the corresponding year [2005:130039.2; 2016: 645126.6; 2017: 644863.6 (unit: $\times 10^3$ hm^2)];
[b] Except for manure, the mass loads for France also including slurry and emitted in the field;
[c] This literature did not reported the single emission of livestock and poultry, so these mass loads were calculated based on the total mass load (maximum) of heavy metals from different sources and the corresponding contributed ratio (estimated according to the Fig. 1) of livestock and poultry to the total;
[d] These two values were the mean input flux of different livestock and poultry and farming scale in this literature.


(a) Mass load for Zn (g/hm², DW)
N
Xinjiang
Inner Mongolia
Tianjin
Qinghai
Shandong
Jiangsu
Henan
Tibet
Shanghai
Zhejiang
21 - 100
101 - 200
201 - 300
301 - 500
501 - 750
751 - 1283
0 750 1,500 km
(b) Mass load for Cu (g/hm², DW)
N
Xinjiang
Inner Mongolia
Tianjin
Qinghai
Shandong
Jiangsu
Henan
Tibet
Shanghai
6 - 25
26 - 100
101 - 180
181 - 260
261 - 360
361 - 618
0 750 1,500 km
(c) Mass load for Pb (g/hm², DW)
N
Shandong
Zhejiang
1.1 - 3.0
3.1 - 6.0
6.1 - 9.0
9.1 - 15.0
15.1 - 20.0
20.1 - 27.7
0 750 1,500 km
(d) Mass load for Cd (g/hm², DW)
N
Xinjiang
Inner Mongolia
Tianjin
Qinghai
Shandong
Jiangsu
Henan
Tibet
Shanghai
Zhejiang
0.08 - 0.30
0.31 - 0.80
0.81 - 1.20
1.21 - 1.60
1.61 - 2.50
2.51 - 4.28
0 750 1,500 km
(e) Mass load for Cr (g/hm², DW)
N
Shandong
Zhejiang
2.2 - 7.0
7.1 - 12.0
12.1 - 24.0
24.1 - 38.0
38.1 - 56.0
56.1 - 82.3
0 750 1,500 km
(f) Mass load for Hg (g/hm², DW)
N
Shandong
Henan
0.02 - 0.07
0.08 - 0.10
0.11 - 0.13
0.14 - 0.18
0.19 - 0.35
0.36 - 0.55
0 750 1,500 km
(g) Mass load for As (g/hm², DW)
N
Xinjiang
Inner Mongolia
Tianjin
Qinghai
Shandong
Jiangsu
Henan
Tibet
Shanghai
Zhejiang
0.20 - 0.80
0.81 - 3.20
3.21 - 5.70
5.71 - 7.90
7.91 - 11.3
11.4 - 20.2
0 750 1,500 km
(h) Mass load for Ni (g/hm², DW)
N
Xinjiang
Inner Mongolia
Tianjin
Qinghai
Shandong
Jiangsu
Shaanxi
Henan
Tibet
Shanghai
Zhejiang
0.92 - 3.00
3.01 - 5.00
5.01 - 8.00
8.01 - 10.0
10.1 - 15.0
15.1 - 28.5
0 750 1,500 km

3.4.2. Mass loads for heavy metals

As a nation, the mass loads for heavy metals from the livestock and poultry manures of China were calculated based on the total heavy metals emission and agricultural land area in 2017, and the results were compared with previous similar studies (Table 4). Similar to the emission results (Table 3), the mass loads of heavy metals from the manures are difficult to compare across different countries, due to these values being estimated using different parameters and methods based on data from different years and different animals. Nevertheless, according to these mass loads, two trends could be extracted as follows: (1) the mass loads of most heavy metals in China were lower than those of France, Canada, Netherlands, and England & Wales, which may result from the smaller agricultural land area of these countries when compared to China. Differences in the estimated parameters and methods may be another important reason. (2) Irrespective of the country, the mass loads of Zn and Cu were significantly higher than other heavy metals, consistent with the concentration levels and emissions of heavy metals for livestock and poultry manures (Fig. 1, Table 3). This observation suggests that livestock and poultry manures are the predominant input sources of Zn and Cu in agricultural land. For China (Luo et al., 2009; Peng et al., 2019), the mass loads of heavy metals decreased distinctly from 2005 to 2017, mainly due to the remarkable increase of agricultural land area from 130,039.2 to 644,863.6 (unit: $\times 10^3$ hm^2); while the minor difference of the mass loads of heavy metals between 2016 and 2017 probably results from the different parameters for the estimation of heavy metal emissions.

Due to the lack of detailed mass loads for every province in the studies of Luo et al. (2009) and Peng et al. (2019), the mass load of every heavy metal from the livestock and poultry manures of 31 provinces in 2017 were estimated in this study, and the results are shown in Fig. 5 (Table S7 listed the detailed data). The geographical distribution of mass loads for Zn, Cu, Cd, As, and Ni were very similar [Fig. 5 (a, b, d, g, and h)], with higher concentrations in the southeast than in the northwest. For individual provinces, Shandong, Tianjin, Henan, Jiangsu, and Shanghai occurred with high mass loads, while Xinjiang, Inner Mongolia, Qinghai, and Tibet with low mass loads, and a relatively low mass load was observed in Zhejiang compared to other southern provinces. The above phenomenon was consistent with the distribution of manure emissions [Fig. 3 (b)], revealing a co-occurrence relationship of Zn, Cu, Cd, As, and Ni in the livestock and poultry manures of most provinces (particularly in the southeast). Similarly, a recent report indicated that the livestock and poultry manures of south China contributed more heavy metal inputs to agricultural land than those of north China, due to its widespread breeding and planting industries (Peng et al., 2019). Another co-occurrence phenomenon was found for the distribution of Pb and Cr in almost all the provinces, with the highest mass load in Shandong and relatively low mass load in southern province Zhejiang [Fig. 5 (c and e)]. The distribution of manure emission for Pb differed from other heavy metals, with no significant region law [Fig. 5 (f)], which may be related to its very low mass loads (mean: 0.17 g/hm^2, DW), which were significantly lower than other heavy metals (mean: 1.57–487.33 g/hm^2, DW) (Table S7).

Among eight heavy metals, the mass loads of Zn and Cu were distinctly higher (by up to three orders of magnitude) than other heavy metals (Fig. 5). This may be explained as the livestock and poultry manures accounted for the main inputs of Zn and Cu to agricultural soils. A previous estimation result based on the national scale of China showed that the livestock and poultry manures were respectively responsible for approximately 69%, 55%, and 51% of the total Cu, Cd, and Zn inputs in agricultural soils from different pollution sources (Luo et al., 2009). Another similar study reported that the livestock and poultry manures were the major contributors for Cu (up to 76%) in the agricultural soils of China (Peng et al., 2019). The excessive application of livestock and poultry manures not only directly polluted the agricultural soils, but also indirectly contaminated the surface water or even groundwater through eluviation and surface runoff (Chen et al., 2018; Khan et al., 2018). Moreover, it could lead to more serious pollution problems, if livestock and poultry manures were mixed with sewage and directly discharged into the water environment (Hooda et al., 2000). Given that livestock and poultry manures contained plentiful nutrients, the resource utilization of manures was vigorously advocated by the Chinese government (Chadwick et al., 2015). Therefore, to ensure the safe application of livestock and poultry manures, and promote the sustainable development of agricultural production, some valid measures should be taken, such as balancing the structure of planting and breeding, formulating an acceptable usage of manures, reducing the additions of heavy metals in feeds, and improving the passivation technologies of heavy metals during manure treatment.

4. Conclusions

The residue of eight heavy metals (Zn, Cu, Pb, Cd, Cr, Hg, As, and Ni) in livestock and poultry manures (especially in pig manure) garnered much attention from Chinese scholars. The pollution profile of heavy metals (especially for Zn, Cu, Cd, and As) in pig manure is more serious than those of other livestock and poultry in China. Cattle excreted more manure than pig, sheep and poultry, while pig manure contributed more heavy metal emissions than other manures. Some southeastern provinces (such as Shandong, Tianjin, Henan, and Shanghai) presented high mass loads of manures and heavy metals, and, therefore, their agricultural soils might suffer from contamination of heavy metals (especially for Zn and Cu). Compared with foreign countries, the heavy metal concentrations in the livestock and poultry manures of China were located in the median levels, showing higher emissions (especially for Zn and Cu), with relatively lower mass loads. For most countries, abundant Zn and Cu exist in agricultural soils, principally contributed by the livestock and poultry manures, and finally sourced from their addition in the feeds of livestock and poultry. Balancing the structure of planting and breeding, formulating an acceptable usage of manures, reducing the additions of heavy metals in feeds and improving the passivation technologies of heavy metals during manure treatment would be valid measures to promote the sustainable development of agricultural production.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.139023.

Fig. 5. Distribution of mass load (DW) for each heavy metal from different livestock and poultry manures in the 31 provinces of China in 2017.

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