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 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.

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

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 frequen-
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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.

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

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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