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Cadmium in animal production and its potential hazard on Beijing and Fuxin farmlands

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ABSTRACT

A random sample of pairs of animal feeds and manures were collected from 215 animal barns in Beijing and Fuxin regions of China. The concentrations of Cd in manures and feeds ranged from non-detectable to 129.8 mg/kg dry weight and non-detectable to 31 mg/kg dry weight, respectively. The concentrations of Cd in pig, dairy cow and chicken manures were positively correlated to those in their feeds. About 30% of the manure samples contained Cd concentrations higher than the upper limit for use in farmlands, and pig and chicken manures might be the primary contributors of Cd to farmlands. The farmlands in Beijing and around the Fuxin Downtown areas would exceed the soil quality criteria within several decades according to current manure Cd loading rates. Undoubtedly, more scientific animal production and manure management practices to minimize soil pollution risks are necessary for the two regions.

1. Introduction

Domestic feed safety issues and international green trade barriers for agricultural products, combined with public awareness of the impact of environmental pollution on crops and general environmental issues, has provoked major considerations on soil and water contamination in China. Soil is a pool for most solid wastes such as the animal manure, the municipal sewage sludge and agricultural wastes, resulting in soil pollution by heavy metals, etc. [1]. It is estimated that more than 12 million tons of toxic metal-polluted crops have been produced, causing an annual economic loss of over 20 billion Chinese Renminbi (RMB) because of trade limitations and the food safety consideration [2].

As we know, cadmium (Cd), a nonessential trace transition metal, is a carcinogen and a possible mutagen [3]. The impact of Cd on human and animal health is now increasingly recognized [4–6]. Cadmium pollution of farmlands has been investigated since the end of the 1970s in China. It is estimated that more than 13,000 ha of farmland are contaminated by Cd in the 11 provinces of China, and more than 50 million kg/year of Cd-rich rice are produced in these farmlands [7]. In addition, Cd is often detected in vegetables and some animal haslets were observed to contain Cd levels above the national food hygiene standards in China [8–10].

It is common practice to add mineral additives to animal feeds to meet the demands of the animal body for essential micro-nutrients and to stimulate their growth. Copper, zinc, and phosphate are usually added in animal production [11]. Feed supplements are permitted for use in animal production at reasonable levels in China [12]. However, due to abuse of mineral additives, high residues of Cu, As, and Zn in animal manure have been reported, this has caused scientists and the general public to worry about food safety and the potential risks on farmlands where animal manures are used [13,14]. Moreover, the toxic metal of cadmium (Cd) has attracted our attention, because several out of the ordinary values were reported by some studies. The pig manure samples collected in Jilin province were detected with mean value of 59.66 mg/kg Cd and in the big range of 0.25–120.13 mg/kg [14]. Furthermore, a large scale investigation in 14 Chinese provinces concluded that 66.0%, 51.7%, 38.1% and 20.0% of the commercial organic fertilizers made from chicken manure, pig manure, cattle manure and sheep manure contained Cd exceeding the limitation, the highest values were 23.1, 42.7, 51.5 and 4.7 mg/kg, respectively [15].

Because of its environmental hazards and biological toxicity, the studies about the environmental behaviors of Cd and the influence factors have been carried out for longtime. For example, the utilization of poultry litter in metal-contaminated soils could accelerate the movement of Cd in soil profiles, it might be closely related to the soluble or colloidal organics derived from animal excreta which are able to mobilize metals, enhance the risk of heavy metals leaching and possibly deteriorate groundwater quality even increase

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the plant absorption [16–18]. The high annual rainfall will transfer Cd from the cultivated layer into deeper layer in a silt loam soil [19]. Certainly, many simple or complicated models were developed to predict the potential environmental impact on soil based on the toxic metals imports and exports [6,20]. However, the previous regional study proved that not much is known about feed and manure management in China, for example, on Chinese dairy farms [21,22], although Chinese farmers are accustomed to using animal manure as a soil nutrient source for more than thousands years, but people never compared its risks to its benefits because it has always been done this way. Hence, it is necessary to understand the situation of Cd in animal feeds and its residue in manures, and its potential risk on farmlands in order to ensure the reasonable use of animal manure in agriculture.

A large scale investigation was conducted in animal farms in the regions of Beijing and Fuxin in order to identify the range of Cd concentrations in animal feeds and feces, to investigate the source of manure Cd, and to estimate the potential pollution risk of Cd from animal manure application in the two cities.

2. Materials and methods

2.1. Sampling areas

Beijing, the capital of China has more environmental protection and public health measures than any other province. Although concentrated animal farms are forbidden within the Sixth Ring of the city, animal production is still the predominant income for Beijing farmers, and currently accounts for 60% of their annual income. In 2004, more than 1250 million pigs, cattle and sheep and 3248 million poultry were produced according to Beijing official statistics [23].

Table 1
Numbers of animal manure and feed samples.

	Pig	Chicken	Cattle	Sheep
Manure	114	18	71	12
Feed[a]	113	18	71	8

[a] Five feed samples were lost.

Fuxin City, located in Liaoning Province, has more than one hundred years of mining history. Due to high unemployment rates, the local government has encouraged unemployed workers to rear livestock. Pig, dairy cattle and poultry production has increased more than 2–3-fold in the last 5 years. Per capita, animal production in Fuxin is currently the highest among the 14 cities of Liaoning province.

In summary, animal production is an important economic aspect in these two cities. Hence, environmental problems relating to animal production in these cities need to be investigated.

Six of ten counties in Beijing and four of seven districts and counties in Fuxin were selected to collect animal feed and manure samples for this study. The sampling areas are shown in Fig. 1.

2.2. Sampling, pretreatment and analysis

A total of 210 animal feedingstuff samples and 215 manure samples were randomly sampled in 2005 in Beijing and Fuxin. Detailed sample numbers for each type of animal are shown in Table 1.

Owing to privatization of the animal farms and the hygienic consideration, visiting animal farms is exceedingly difficult in the two cities. With the consent of farm owners, the feed and manure were sampled, where possible, accompanying with the short interviews with the farmers, about farm operation, including herd size and composition, livestock facilities, feeding practices and manure

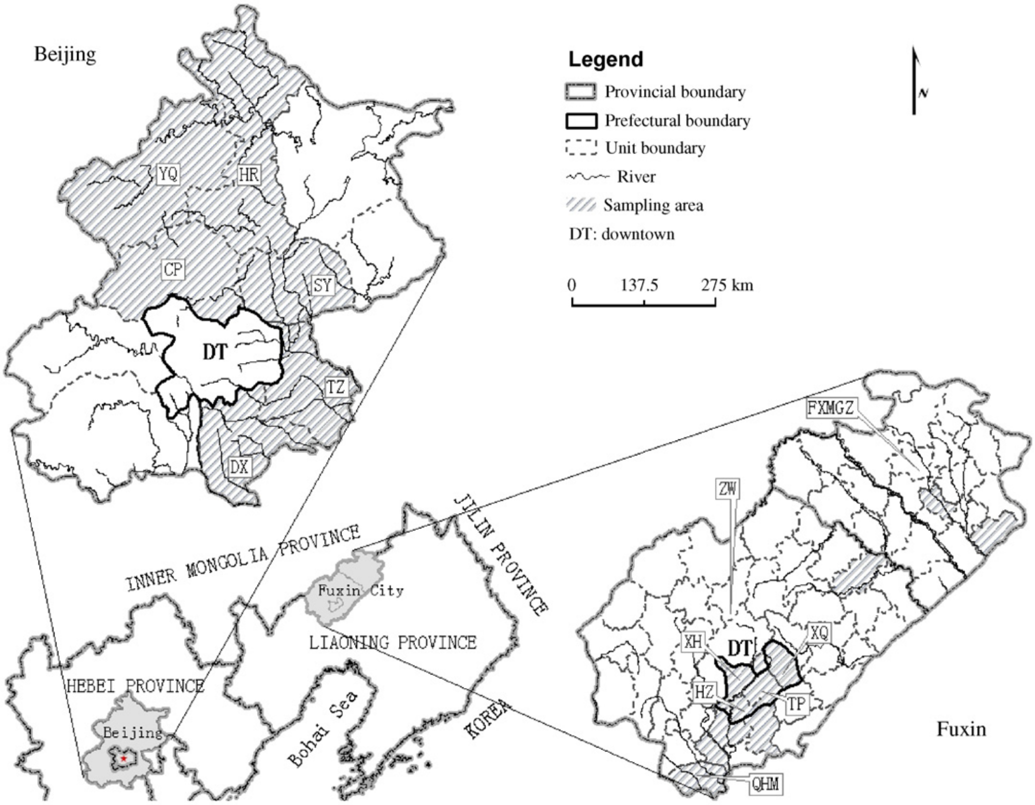


Fig. 1. The study areas and the sampling sites.

Table 2
Descriptive statistics of cadmium concentration in animal feeds (mg/kg, dm).

	N. of sample	Minimum	Maximum	Mean	Median	Std. deviation	Kurtosis	Skewness
Pig	113	nd[a]	27.60	2.29	0.26bc[b]	5.48	9.38	3.10
Dairy cattle	71	nd	31.00	2.79	0.45b	5.06	17.27	3.95
Chicken	18	0.02	21.92	8.13	4.54a	8.25	−1.48	0.50
Sheep	8	nd	0.92	0.39	0.20c	0.37	−1.48	0.38

Hygienical standard for feeds GB 13078-2001, feed Cd < 0.5 mg/kg, dm [12].

[a] nd: not detectable.

[b] Different letters in the line indicate the significant difference ($p < 0.05$) by Kruskal–Wallis *H* test.

Table 3
Descriptive statistics of cadmium concentration in animal manures (mg/kg, dm).

	N. of sample	Minimum	Maximum	Mean	Median	Std. deviation	Kurtosis	Skewness
Pig	114	nd[a,b]	129.76	12.05	1.85ab	25.27	6.73	2.65
Cattle	71	nd	35.50	3.75	0.94b	8.29	4.82	2.4
Chicken	18	0.63	63.64	15.38	3.26a	21.11	0.76	1.47
Sheep	12	nd	8.39	1.42	0.19b	2.41	6.18	2.32

Control standards for urban wastes for agricultural use GB8172-87 standard, Cd < 3.0 mg/kg, dm [29].

[a] nd: not detectable.

[b] Different letters in the line indicate the significant difference ($p < 0.05$) by Kruskal–Wallis *H* test.

management practices. In this survey, the animal operations were comprised of cooperative association of many families or single operator, which are all intensive operation. The sizes of the farms ranged from 140 to 12,000 for pig, 60 to 32,000 for dairy cattle, 1200 to 32,000 for chicken and 300 to 1300 for sheep, respectively. All animals are fed with compound feeds which were purchased commercially or home mixes. The formula of the home mixes are directed by professionally agricultural consultants or followed experiences from other successful farmers. In China, crop farming and livestock farming are separated usually. Most of the animal farmers do not have their own farmlands to grow crops or recycle the animal manure in this survey, hence, the individual feed components has to be purchased through markets, some of the feed ingredients are imported from other native cities even overseas markets. The animals were usually stall-fed, only partial dairy farms let the animals spent several hours in the unvegetated barnyards daily in this investigation.

At each farm, 3 or 4 feed samples along with 3 or 4 corresponding fecal samples were taken individually from the different groups of animals. The animal groups were usually stratified by stage of growth or animal function depending on the classification by the farm operators. For the consistency principle, each of the feed sample acquisitions was the final mixture comprising of every ingredients and the mixture sample must be going to feed animals on the day of sampling. Each feed sample was collected by taking a minimum of 20 sub-samples from different positions within the feed tanks. Sub-samples were then bulked together and thoroughly mixed to provide one representative sample of approximately 2 kg for analysis. The fresh solid droppings (manure, excluding liquid droppings, cleaning water and bedding materials) of animal were taken directly from the floor of each barn. A minimum of 30 fecal sub-samples were taken from different positions on the unit floor. Sub-samples were then bulked and thoroughly mixed to provide one representative sample of approximately 2 kg for analysis.

The fresh fecal and feed samples were air-dried in the shade, then ground and passed through a 0.25 mm mesh PVC sieve. A 0.5 g sub-sample of the dry powder was weighed and digested in heated, concentrated HNO_3 and H_2O_2 [24]. The Cd concentration in the filtered supernate was determined by graphite furnace atomic absorption spectrometry (AAS, Vario 6, Jena Co. Ltd., Germany). The accuracy of the analysis was checked with samples of wheat and soil with certified concentrations (GSS-1, GBW-08501, respectively, China National Center for Standard Materials). Recovery of Cd was 97.2% and 98.4% in GSS-1 and GBW-08501, respectively. A 15% paralleled replication of samples was also used as the quality control procedure.

2.3. Statistical analysis

The correlation analysis between manure and feeds was conducted using Spearman method. The differences between different feed or animal manure were tested using non-parametric method (Kruskal–Wallis *H* test) since no normal distribution was found for the feed and manure data. All statistical analysis mentioned above was fulfilled with SPSS software 11.0.

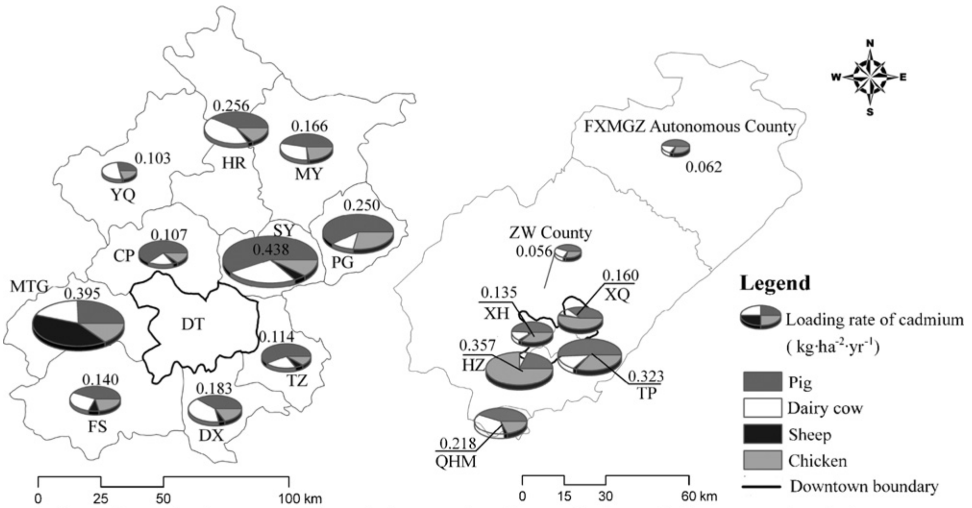
3. Results and discussion

3.1. The concentrations of cadmium in the animal feedingstuffs and manure

The large range and big dispersion of feed Cd and manure Cd data sets (Tables 2 and 3) in this survey are completely different from the international reports such as Nicholson et al. in England and Wales [25] and Sager in Austria [26], which showed a unique features of toxic metal Cd in China animal production.

Although Cd is not necessary for animal growth, over 88.6% of the 210 feed samples were detected to contain this toxic metal in the survey (Table 2). Median Cd concentration in feeds was in the order of chicken > dairy cattle > pig > sheep, and it was significantly higher in chicken feeds than that in the pig, dairy cattle, and sheep feeds ($p < 0.05$), indicating that the dietary strategy for chickens might result in more Cd in the mixture feed. Although dairy cattle and sheep are chiefly fed with fodder, the Cd content in the dairy cow feed was significantly higher than that in the sheep feed ($p < 0.05$), suggesting that the non-fodder diet ingredients probably contained more Cd and thus increased its content in dairy cattle feeds.

The concentrations of Cd in animal feeds clearly featured a large range from non-detectable (nd) to 31.0 mg/kg, dm, which is far beyond the range and mean values reported in England and Wales [25]. Previous investigations also presented the high level of Cd appearance in animal feeds. Zhou and Xu [27] found that 5% of premixed feeds for pigs purchased in the market contained 150–370 mg/kg of Cd, while 73.6 mg/kg of Cd was observed in home-mix feeds [28]. Besides high level of Cd in animal feeds, the Cd accumulation in animal edible offals exceeding the food hygiene criterion of <0.10 mg/kg was reported in Beijing and



Beijing: YQ: Yanqing County, HR: Huairou District, MY: Miyun County, PG: Pinggu District, TZ: Tongzhou District, DX: Daxing District. FS: Fangshan District, MTG: Mentougou District, CP: Changping District, SY: Shunyi District; **Fuxin:** FXMGZ: Fuxin Mongolian Autonomous County, ZW: Zhangwu County, XH: Xihe District, XQ:Xinqiu District, HZ: Haizhou District, TP: Taiping District, QHM: Qinghemen District.

Fig. 2. Loading rates of cadmium with animal manures to Beijing and Fuxin farmlands and the proportions of four animal manure contribution.

Jiangsu markets, it further confirmed the occurrences of Cd in animal production and might closely relate to the animal feeds [8,10,27]. Compared the hygiene standard for animal feeds (GB 13078-2001, feed Cd < 0.5 mg/kg, dm) [12], a total of 84 of the 210 feed samples in this survey contained Cd above this limit. Especially, 64% of chicken feed samples contained Cd higher than 0.5 mg/kg, the proportion was nearly three times more than that of dairy cow feeds and four times more than that of pig feeds. Our results revealed that Cd contamination in animal feeds is serious, and more attention should be paid to chicken feeds.

Consistent with the results of animal feeds, a wide range of Cd (nd–129.8 mg/kg, dm) was observed in the four types of manure (Table 3). The Cd concentrations were in the order of chicken manure > pig manure > dairy cow manure > sheep manure, the median of Cd concentrations in the chicken manures were significantly higher than those in the dairy cow and sheep manures ($p < 0.05$). No significant differences were observed between the chicken and pig manures, or the dairy cow and sheep manures. An extremely high Cd concentration of 129.8 mg/kg was detected in the pig manure, coincidentally, 120.1 mg Cd/kg in pig manure was reported by other research in Jilin Province too [14]. The limit of Cd content for animal manure land application should follow the GB8172-87 standard (Cd < 3.0 mg/kg, dm) [29]. A total of 64 of 215 manure samples exceeded the limitation in the study. In detail, Cd concentrations in 56%, 33%, 18% and 8% of chicken, pig, dairy cow and sheep manures were over this limit, respectively. Sager [26] reported Cd concentrations in animal manures from less than 0.02 to 0.93 mg/kg, and Nicholson [25] measured the similar range of manure Cd concentrations to Sager's. These concentrations of Cd are far lower than those found in the present study, which displayed a unique feature of Cd appearance in Chinese animal manure compared to international studies.

3.2. The sources of Cd in animal feeds

There were significant correlations between Cd concentrations in the feeds and manures of pig, dairy cow and chicken ($p < 0.01$, Spearman test), except sheep. The results indicated that the high level of Cd in three types of animal manures may probably originate from the animal feeds. Furthermore, Cd concentrations of the animal manures were much higher than those of the animal feeds, showing a concentrated effect through animal metabolic process.

According to current knowledge, cadmium could not be added as feed additives for animal growth. Usually, Cd as an impurity is often present in mineral supplements such as phosphates, Zn sulfate and Zn oxide. Thus Cd may enter the animal production process with these feed ingredients, and these supplements might contribute significantly to dietary contamination [30]. Some surveys, in China provinces of Guangxi, Hubei and Hunan, proved that the added zinc sulfate and phosphate were the main sources of Cd in the animal compound feeds, with 1–3.6% of Cd in the Zn sulfate additive [31–33]. Another source of Cd in the feed may be forage and grain materials, where the Cd contents of these materials could be influenced by soil contamination. Commonly, the background content of Cd in Chinese soils is relatively low (about 0.1 mg/kg)[34]. Atmospheric deposition and fertilizer use (phosphates, sewage sludge, etc.) may be primary sources to influence the Cd contents of soils [35]. High concentrations of Cd (up to 10 mg/kg) were found in the forages grown in fields near industrial zinc-plating sites, or where urban sludge was used as fertilizer [36]. Cai et al. [37] reported that cattle tissues, especially kidneys, were contaminated and closely correlated to high concentration of Cd (10 mg/kg) in the forage. In addition, the other sources of Cd possibly entering animal production such as drinking water or dust in the barn are needed to further investigate for more reasonably explain the data.

3.3. Potential Cd soil pollution by application of animal manures

Animal manure spread onto arable lands is a common agricultural practice, and is used as a greenhouse medium for planting vegetables in Beijing and Fuxin. In order to understand the potential risk of manure Cd at a worse case situation (assuming the animal manure will be entirely and evenly applied into farmlands in the two cites, without plant uptake and Cd loss in soil), the loading rates of Cd in Beijing and Fuxin farmlands were estimated using the following Eq. (1):

$$L_j = \frac{\sum C_i \cdot M_{i \cdot j}}{A_j} \times 10^{-6} \qquad (1)$$

Table 4
Time scale needed to increase cadmium content in the farmland soils from current level to the maximum permissible limit, due to animal manures' use in farmlands of Beijing and Fuxin cities (in unit of year).

Soil criteria	Beijing area sites									
	SY	CP	PG	HR	MTG	MY	FS	TZ	DX	YQ
Grade II	5	8	9	10	11	11	17	19	22	24
Grade III	26	36	43	49	52	52	81	90	105	118

	Fuxin area sites						
	TP	QHM	HZ	XH	XQ	FXMGZ	ZW
Grade II	13	23	31	32	36	72	88
Grade III	66	114	157	163	183	364	446

GB15618-1995 environmental quality standards for soils-Grade II: Cd $\leq$ 0.3 mg/kg, at soil pH $\geq$ 6.5.
GB15618-1995 environmental quality standards for soils-Grade III: 0.3 < Cd $\leq$ 1.0 mg/kg, at soil pH $\geq$ 6.5. The soil cannot be used for agricultural cultivation.

where L_j is the loading rate of Cd in j county (kg/ha year), C_i is the mean concentration of Cd in i animal's manure (mg/kg), $M_{i\cdot j}$ is i animal's manure production in j county (kg/year), and A_j is the farmland area in j county (ha).

Fig. 2 illustrates the Cd loading rates of animal manure use in Beijing and Fuxin farmlands. The Cd loading rate was 0.438, 0.395, 0.256, 0.250, 0.183, 0.166, 0.140, 0.114, 0.107 and 0.103 kg/ha year for SY (Shunyi), MTG (Mentougou), HR (Huairou), PG (Pinggu), DX (Daxing), MY (Miyun), FS (Fangshan), TZ (Tongzhou), CP (Changping) and YQ (Yanqing) farmlands in the Beijing area, respectively. The highest loading rate was found in SY County, the largest animal production county of Beijing. The SY County is located in northern Beijing, the local government has encouraged the animal industry for around 40 years, resulting in a population of 2.5 million pig, cattle and sheep and 43.8 million poultry. The second highest loading rate of manure Cd was in the mountainous county-MTG. Although MTG County has the lowest animal production, the extremely limited farmland area of 77.4 km^2 in this mountainous county resulted in the high loading rate of Cd. The smallest input of Cd and the lowest loading rate was in YQ County of Beijing, because of its relatively low chicken and pig population and enough farmland area to receive the animal manure. Based on the inventory of Cd input to soils (Fig. 2), pig manure was the primary contributor of Cd in the six counties of Beijing, but according to Nicholson's study [38], cattle could supply 64% of the total manure Cd inputs to England and Wales. The two exceptions were MTG and YQ counties in Beijing, where sheep and dairy cow manures were the largest sources of Cd input accounting for about 45% and 50%, respectively.

To reduce the long-term pollution risk of hazardous elements in sewage sludge, the Council Directive 86/278/EEC [39] recommended the upper loading rate of 0.10 kg/ha year of Cd to farmlands. Farmlands in the all counties of Beijing, received Cd loading rates over this limit, though the Cd loading rate in the farmlands of CP and YQ closely approached this limit. Similarly, the Cd loading rates in the farmlands of the five districts in Fuxin city exceeded this upper limit, and the most heavily loaded region was confined to the areas surrounding the downtown region, especially HZ (Haizhou) and TP (Taiping) districts, where there are not only several coal mining industries but also a few intensive animal farms. Therefore, this might pose more of a pollution risk to local residents. In contrast to that in Beijing, chicken manure was the major contributor of Cd in addition to pig manure. Owing to very large areas of farmland and less intensive animal production, the countryside areas such as FXMGZ (Fuxin Mongolian Autonomous County) and ZW (Zhangwu County) did not receive high levels of animal manure Cd.

Assuming that arable lands solely received Cd from animal manure and had no Cd loss and plant uptake, the estimation of time-scale to increase topsoil concentrations of Cd from the current level to the maximum permissible limit was calculated by Eq. (2) and is listed in Table 4.

$$T_j = \frac{(C_k - C_{0k}) \cdot m \times 10^{-6}}{p \cdot L_j} \tag{2}$$

where T_j is the time for Cd content in soils to exceed the standard of GB15618-1995 [40] in j county's farmland (year); C_k is the criteria concentration of Cd in soils indicated by GB15618-1995 (mg/kg); C_{0k} is the current concentration of Cd in j county's farmland, cited from the reports by Zheng et al. [41] and Li et al. [42] (mg/kg); m is the mass of the top 0–20 cm soil in farmland (kg/ha); p is the supposed proportion of manure applied to farmland, $p = 1$; L_j is the loading rate of Cd in j county (kg/ha year).

Based on the Grade II level (Cd $\leq$ 0.3 mg/kg), it was calculated to be in the range of 5–24 years for farmland in the ten counties of Beijing, and 13–88 years for the farmlands in the seven counties and districts of Fuxin to reach the Cd limit (Table 4). Even when we adopted the Grade III level of 1.0 mg/kg as the ceiling limit, Cd content in the farmland soils of SY, CP, PG and HR counties in Beijing might be higher than the soil Cd criteria after 26–50 years. All counties in Beijing and two districts (HZ and TP) surrounding the Fuxin downtown area require further attention, considering that the farmland Cd build-up from animal manure requires less than 30 years to reach the Grade II level limit. These results suggested that Beijing farmlands might have more Cd input than those of Fuxin, due to more animal production and fewer arable lands to recycle the animal manure. Huckabee and Blaylock [43] concluded that 4–6% of the Cd in soil could be transferred to water, but once in water, Cd accumulated in the sediments more quickly than in biota, thus retaining the long-term potential hazard to underground water. Therefore, we strongly suggest that the local government, especially in the northern counties of Beijing, where there is over 60% of the surface water resources for the city such as the Miyun and Guanting reservoirs, take into account a reasonable animal production strategy and manure disposal policy.

This estimation may provide an elementary evaluation of the pressure caused by animal manure containing Cd in Beijing and Fuxin, and is a guide to local governments to ensure reasonable management of animal production and effective animal manure disposal. However, it should be noted that (1) soil Cd loading rates might be underestimated due to Cd inputs from other sources such as atmospheric deposition, sewage sludge application, chemical fertilizers, irrigation water, etc.; (2) the leaching loss and crop uptake may alleviate the accumulation of Cd in farmlands; (3) the soil loss and plant uptake of Cd may be affected by factors such as Cd speciation, the physical and chemical characteristics of the soil, and climate etc. Further studies on the risks of metals contained in manure on the environment should take these factors into account.

4. Conclusion

The concentration of Cd in animal feeds was in the range of nd–31.0 mg/kg; while the Cd concentration in animal manures was in the range of nd–129.8 mg/kg. The concentrations of Cd in more than 30% of the animal manure samples were above the upper limit for manure Cd concentrations for use in farmlands. The average concentrations of Cd in the four types of animal manures followed the order: chicken > pig > dairy cow > sheep. The concentrations of Cd in pig, dairy cow and chicken manures were positively correlated to those in pig, dairy cow and chicken feeds at the 0.01 level, respectively. Pig and chicken manures would be the important contributors to soil Cd in Beijing and Fuxin farmlands, respectively, when they are used in farmlands. As a whole, the farmlands in Beijing and around the Fuxin Downtown area received overloaded rates of manure Cd. Therefore, Cd concentrations in the farmland soils would exceed the soil quality criteria for Cd in several decades time according to current Cd loading rates. Undoubtedly, more scientific animal production and manure management practices to minimize soil pollution risks are necessary for these two cities.

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