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Heavy metals pollution in poultry and livestock feeds and manures under intensive farming in Jiangsu Province, China

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Abstract: The heavy metals pollution in poultry and livestock feeds and manures under intensive farming in Jiangsu Province was investigated. 97 feed and manure samples were sampled from 31 farming plants in 10 major cities of Jiangsu. 14 metals, including Zn, Cu, Pb, Cd, Cr, Ni, Mo, Mn, Ba, Co, Sr, Ti, As and Hg, were analyzed after samples acid digestion. The results showed that the most feed samples contained high concentration of metals exceeding National Hygienical Standard for Feeds. Meanwhile, it was found that Cu, Zn, Pb, Cd and Cr concentrations in animal manures were also high, for example, Cu concentration in a manure sample reached to as much as 1726.3 mg/kg. Heavy metals loading quantities in soil per year were then calculated when metals contaminated organic fertilizers were applied, and its effects on soil environmental quality were further evaluated. Keywords: heavy metal; intensive farming; feed; manure; Jiangsu Province

Introduction

Poultry and livestock manures have been widely applied as important organic fertilizer sources in agriculture (Huang, 2001). However, feed additives and animal medicines, which were used to facilitate weight increase and disease prevention for poultry and livestock under intensive farming, will easily cause environmental problem due to medicine residue in and metal uptake by these animals (Zhang, 1995; Xing, 2001). Recently, nitrogen and P pollution as well as how to control wastewater and pathogeny contamination from breeding plants has been concerned in China (Dong, 1998; Deng, 1999; Liu, 1997; Ding, 2000; Xu, 2002), but attention was not yet paid to heavy metals pollution in poultry and livestock feeds and manures (Sims, 1994; Rong, 1998; Yu, 2001).

At present, more and more people in China care about food quality over quantity, when their living level gets to be improved significantly compared with decades ago. So, to develop green and safe food is very necessary. Replacing chemical fertilizer by organic fertilizer may be an important alternative for green agriculture.

In this study, we chose Jiangsu Province as a typical research area. It contained relatively developed economy system and large numbers of intensive farming plants in China. About 31 poultry and livestock farming plants in 10 major cities were investigated related to its heavy metals pollution status. The results will possibly be helpful for people to understand environmental situation of poultry and livestock feeds and manures in Jiangsu Province, and correspondingly regulate effective countermeasures to control their pollution.

1 Materials and methods

1.1 Sampling

97 poultry and livestock feeds and manures in total were sampled in 31 intensive farming plants in 10 cities in Jiangsu

Province, which consisted of 47 feed samples, 48 manure samples and 2 organic fertilizer samples. These poultries and livestocks included chickens, pigs, milch cows, ducks, gooses and doves. The detailed sampling information is listed in Table 1.

Table 1 Sampling information for poultry and livestock feeds and manures samples

City (farming plant numbers)	Feed sampling numbers	Manure samplin	ng Organic fertilizer sampling numbers	Total
Nanjing (2)	7	6	1	14
Suzhou (2)	4	5	1	10
Wuxi (2)	3	4		7
Changzhou (2)	2	2		4
Nantong (5)	6	7		13
Yangzhou (4)	5	5		10
Taizhou (2)	4	4		8
Yancheng (4)	8	8		16
Huaian (5)	5	4		9
Lianyungang (3)	3	3		6
Total (31)	47	48	2	97

Manure samples were dried at room temperature, and feed samples were dried at 70 ℃ in an oven. All manure and feed samples were then ground in a mill and pass through a 60 mesh sieve.

1.2 Analysis and statistics

About 1.5 g dried feed or manure samples were weighed, put into a flask, and digested using HNO₃-HClO₄ until complete decompose of each sample (Soil Science Society of China, 2000). Each digestion solution was filtrated through a filtrate paper into a 10 ml flask, water was then added into the flask to a fixed volume. Zinc, Cu, Pb, Cd, Cr, Ni, Ba, Co, Mo, Mn, Sr and Ti concentrations in filtrates were determined using an ICP-AAS. With regard to As and Hg analysis, another digestion method with HNO₃-H₂SO₄-HClO₄ was used (Cui, 2001), and filtrates were analyzed with a HG-AFS. Two standard plant samples were used to guarantee date accuracy.

2 Results and discussion

2.1 Heavy metals pollution in poultry and livestock feeds

Table 2 shows the average concentrations of heavy metals and their ranges in feeds of chickens, pigs, milch cows, ducks, gooses and doves. Average Zn concentration in chicken feeds was 150 mg/kg, which is almost equal to that in pig and duck feeds. Milch cow feeds included a higher average Zn concentration than that in goose and dove feeds.

Average Zn concentrations were 109.24 mg/kg, 96.69 mg/kg and 29.06 mg/kg in milch cow feeds, gooses feeds and dove feeds, respectively.

Average Cu concentration in pig feeds was 105.36 mg/kg, which was the highest average Cu concentration among all examined feed samples. Chicken feeds, milch cow feeds and goose feeds contained almost a same average Cu concentration as 22.56 mg/kg, 19.09 mg/kg and 12.79 mg/kg, respectively. In the same way as Zn, average Cu concentration in dove feeds was the lowest as only 5.15 mg/kg.

Table 2 Heavy metals concentration in poultry and livestock feeds

				Feed from			
Heavy metal		Chicken(16)	Pig(7)	Milch cow(7)	Duck(2)	Goose(2)	Dove(3)
Zn	Ave.	153.78	144.17	109,24	155.9	96.69	29.06
	Range	35.76-399.36	35.54-320,20	27.64-378.31	138.5-173.3	33.79-159.59	25.32-31.64
Cu	Ave.	22.56	105.36	19.09	20.86	12.79	5.15
	Range	6.09-50.50	4.91-392.1	4.51-29.92	18.21-23.5	7.67-17.90	4.06-6.75
Pb	Ave.	7.21	10.68	5.76	6.64	8.14	3.64
	Range	3.07—14.41	1.04-60,19	4.42-6.67	2.38-10.90	6.99-9.29	3.16-4.01
Cd	Ave.	0.64	0.57	0.22	0.51	0.53	0.09
	Range	0.08-2.81	0.04-3.22	0.15-0.43	0.04-0.97	0.31-0.75	0.04-0.16
Cr	Ave.	29.96	25.71	13.67	12.50	5.46	7.61
	Range	6.57—162.36	5.94-97.71	7.56-19.93	11.56-13.44	3.76-7.16	6.16-9.77
Ni	Ave.	12.60	7.85	2.30	2.36	1.25	3.37
	Range	0.12-91.80	0.67-29.63	0.335.13	1.57-3.14	0.99-1.50	1.99-4.77
Ba	Ave.	19.14	12.96	11.78	11.48	4.70	5.47
	Range	4.58-36.74	5.65-46.71	8.01-22.47	11.32-11.64	4.50-4.90	1.49-8.87
Co	Ave.	0.52	0.59	0.36	0.46	01.0	0.22
	Range	0.11-1.31	< 0.01-1.59	0.09-1.41	0.40-0.53	0.024-0.172	< 0.01-0.49
Mo	Ave.	1.34	0.94	1.01	0.38	1.03	0.57
	Range	0.26-5.56	0.20-2	0.69-1.43	0.13-0.64	< 0.1-1.956	< 0.1-1.33
Mп	Ave.	190.31	133.64	85.41	157.50	84.52	30.19
	Range	57.29-472.25	55.08-335.97	20.43-268.83	128.53—186.47	32.75-136.28	15.49-57.35
Sr	Ave.	44.12	19.30	23.99	10.11	7.66	2.22
	Range	12.15-75.63	4.02-53.53	8.45-61.93	9.10-11.13	3.14-12.17	1.49-3.01
Ti	Ave.	24.15	21.69	17.88	10.93	7.34	3.77
	Range	6.49-48.55	7.67-55.16	5.90-43.72	10.71-11.15	2.89-11.79	2.845.29
As,	Ave.	134	90	22	5	32	2
μg/kg	Range	< 4-138.0	<4-111.1	< 4—83	< 48	< 4—61	< 4
Hg,	Ave.	5.3	5.8	1.1	13.6	0.2	0.9
μg/kg	Range	< 0.2-33.4	< 0.2-290	< 0.2-6.8	9.6—17.5	< 0.2-0.3	< 0.2-2.4

Notes: where samples were below the limit of detection (LOD), a value of $0.5 \times LOD$ was used to calculate means; the unit is mg/kg except As and Hg

According to above analysis, it can be concluded that average Zn and Cu concentrations in feeds changed significantly with sorts of poultry and livestock. More Cu was supplied in pig feeds, but less used in dove feeds. Except in dove feeds, zinc was also widely used as additives in all examined feeds.

Nicholson et al. (Nicholson, 1999) recently reported that in England average Zn and Cu concentrations in pig feeds were obviously higher than that in milch cow feeds, which is in agreement with our results. However, zinc concentration in pig feeds changed significantly in the range of 216—2920 mg/kg and the average Zn concentration reached to 2300 mg/kg, which was higher than ours. Average Cu concentration in Nicholson result was 175 mg/kg, which was also higher than the data reported in this paper. National Hygienical Standard for Feeds (GB13078-2001) did not show any information on Cu and Zn maximum concentrations allowed in feeds, as

shown in Table 3. So, to regulate metal limitation concentrations in feeds will be very important not only for improving animal quality but also for guaranteeing environmental security. Compared with Russian Federal Standard for Feeds, zinc and Cu concentrations in these feed samples exceeded the allowance value except that in goose and dove feeds.

Table 3 Heavy metal limitation concentration (mg/kg) in feeds (data came from Xie, 2001)

	Zn	Cu	Pb	Cd	Cr	As	1Ig
China		_	5	0.5	10	2.0	0.1
Russian	100	8.0	5	0.4	-	_	0.1
England	-	-	5	-	-	2.0	0.2
Germany	_	_	6	_	-	2.0	0.2
American	-	-	5	0.5	-	2.0	0.2
Јарап			3	1.0			0.4

Lead, Cd, Cr, As and Hg concentrations in feed are

limited in GB13078-2001 and also in many countries' standard. It was found that average Pb concentration was satisfied with the requirement in GB13078-2001 only in dove feeds. Average Pb concentration in pig feeds was 11.68 mg/kg, and some samples contained Pb with a concentration even up to 61.19 mg/kg. Moreover, average Pb concentration in goose feeds also reached to 8.14 mg/kg.

Dove and goose feeds contained less average Cr concentration than the standard. But, it was found that average Cr concentrations in chicken and pig feeds were as two times as the national standard. One chicken sample has Cr concentration even as high as 162.36 mg/kg. Average Cd concentrations in chicken feeds, pig feeds, duck feeds and goose feeds were higher than the National Hygienical Standard for Feeds.

So, it suggests that metal concentration in minerals, which was used as metal additives in feeds, should be paid more attention during feed production.

Arsenic and Hg concentration in most feeds were lower than National Hygienical Standard for Feeds (As ≤ 2.0 mg/kg, Hg ≤ 1.1 mg/kg) except in several chicken and pig feed samples.

According to above results, it can be concluded that heavy metals pollution in chicken and pig feeds in investigated area was serious. Not only were Cu and Zn concentrations in feeds very high, but also Pb, Cr and Cd concentration exceeded the National Hygienical Standard for Feeds.

2.2 Heavy metals pollution in poultry and livestock manures as well as organic fertilizer samples

Table 4 shows heavy metals concentrations in poultry and livestock manures as well as 2 organic fertilizer samples. It is obviously observed that poultry and livestock manures contained more heavy metals than feeds. Average Zn concentration in manures was 1—6.6 times that of feeds.

Table 4 Heavy metals concentration in poultry and livestock manures and 2 organic fertilizer samples (Unit:mg/kg except As and Hg)

			concentration ii	· pourse in				(0,111,111,8,11,8)	except 113 and 116
Heavy metal		Chicken(17)	Pig(16)	Milch cow(8)	Manure Duck(2)	from Goose(2)	Dove(3)	Fertili . 1	Ferthili .]]
Zn	Ave.	417.0	505.9	185.52	440.62	77.42	192.44	203.37	377.16
	Range	175.3-964.7	113.6-1505.6	98.9-305.9	434.8-446.4	66.33-88.51	142.16-242.2	203.37	377.10
Cu	Ave.	89.14	399.0	45.99	46.95	14.71	22.59	34.16	102.85
	Range	20.7-569.7	35.7-1726.3	22.7-91.0	41.69-52.21	11.78-17.64	18.15-26.51	34.10	
Pb	Aye.	11.08	12.80	9.74	4.51	18.20	14.39	12.19	33.90
	Range	2.92-20.31	4.22-82.91	5.31-12.89	4.27-4.74	16.86-19.55	0.53 - 37.57	12.19	33.90
Cd	Ave.	1.84	0.80	0.70	0.29	1.40	0.36	0.20	0.60
	Range	0.31-11.32	1.13-4.35	0.30-1.23	0.27 - 0.29	1.05-1.75	0.23 - 0.42	0.38	0.60
Cr	Ave.	81.15	46.23	46.90	44.85	54.75	100.73	49.35	25.41
	Range	38.76-269,11	23.21-64.67	37.86-57.73	44,74-44.96	45,14-64.35	56.02-168.58	48.25	25.41
Ni	Ave.	17.51	9.51	8.91	10.40	32.49	17.92	9.20	20.94
	Range	6.9131.4	3.62-22.10	4.30-11.66	10.22-10.59	32.23-32.74	10.96-21.8	8.39	
. Ba	Ave.	89.31	45.23	66.49	81.98	202.6	38.09	(4.05	221.32
	Range	28.21-180.24	28.36-78.79	34.63-122.82	79.53-84.44	187.7-217.4	34.29-43.21	64.37	
Co	Ave.	2.29	2.11	1.67	1.37	9.44	1.21	1 45	
	Range	0.24 - 5.71	0.10-7.98	1.97-2.09	0.95-1.79	8.37-10.51	0.01 - 2.06	1.42	5.64
Mo	Ave.	3.80	1.60	2.91	1.81	1.65	2.69	0.73	
	Range	1.20-6.75	< 0.1-3.42	1.03-4.34	1.17-2.44	0.21-1.08	1.54-3.49	0.63	2.61
Mn	Ave.	623.7	451.95	472.2	576.1	511.6	193.3	(00.41	740 OO
	Range	207.1-1117.3	192.7892.34	195.3-1167.3	574.2-578.1	421.7-601.5	111.5-287.0	688.41	540.88
Sr	Ave.	145.76	63.16	77.75	48.29	62.10	88.24	00.00	404.40
	Range	48.24-299.21	9.94-171.07	64.27-94.87	47.14-49.45	59.19-65.01	80.34-95.00	92.73	185.62
Ti	Ave.	139.92	76 .18	100.25	50.06	1042.9	49.58	108.83	736.24
	Range	31.6-351.89	31.34-183.75	74.54-134.03	45.07-55.05	821.4-1264.6	46.27-53.74		
As,	Ave .	47	12	13	6	7	14	40	10
µg/kg	Range	< 4553	< 478	4-42	6—6	6—7	6-21	48	12
Hg,	Ave.	24.2	33.0	39.0	29.0	22.4	51.7	24.7	.0.2
μg/kg	Range	01.2-77.3	< 0.2-118.3	< 0.2-72.6	7.5-50.5	< 0.2-44.7	19.0-118.3	34.7	< 0.2

Dove manures had the highest Zn accumulation times, which increased from 29.06 mg/kg in feeds to 192.33 mg/kg in manures. Additionally, zinc accumulation times in pig and chicken manures were 3.5 and 2.7, respectively.

Similar to Zn, copper was also obviously accumulated in manures, which was 1—4.4 times that of feeds. Dove manures had the highest accumulation coefficient, in which average Zn concentration increased from 5.15 mg/kg in feeds to 22.59 mg/kg in manures. Chicken manures and pig manures also had high Zn accumulation times as 4.0 and 3.8, respectively.

Nicholson (Nicholson, 1998) reported that Zn and Cu concentrations in poultry manures were 2—5 times that of poultry feeds. Kunkle *et al.* (Kunkle, 1981) also showed

that Cu levels in broiler litter were linearly related to Cu added in the diet and were concentrated about 3.25 times.

So, it can be concluded that heavy metal accumulation mechanism changed with sorts of poultry and livestock. In view of environmental protection point, metal concentration in animal feeds must be strictly controlled as soon as possible, and heavy metal accumulation ability should be cared at the same time.

In Table 4, average Cu and Zn concentrations in pig manures were 505.9 mg/kg and 399.0 mg/kg, respectively. Copper concentrations in other poultry feeds were relatively low. In deed, average Cu concentrations in Belgian poultry manures were also found to be only 59 mg/kg (Fleming, 1991). Median Cu concentration in Swiss poultry manures

was 35-44 mg/kg (Menzi, 1998).

Lead, Cd, Cr, As and Hg concentrations in poultry and livestock manures were also higher than that in feeds. Manures included as 1—3 times high Pb as in feeds. For Cd and Cr, they were 2—4 times and 2—10 times, respectively. Some other elements, including Ni, Ba, Co, Mo, Sr and Ti, were also found to accumulate in poultry and livestock manures obviously.

Organic fertilizers I and II were obtained from two organic fertilizer plants, which were made from cow manures and combined manures of pigs, chickens and ducks, respectively. Zinc, Cu and Pb concentrations in 2 organic fertilizer samples were high. For organic fertilizer II, zinc, Cu and Pb concentrations were 377.16 mg/kg, 102.85 mg/kg and 33.90 mg/kg, respectively. Therefore, once this organic fertilizer is applied in practice it will possibly cause environmental pollution.

2.3 Heavy metals loading quantities in soil per year after application of organic fertilizers made from poultry and livestock manures

Generally speaking, applying organic fertilizer or biological technologies in agriculture is much better for food safety rather than using chemical fertilizer and toxic pesticide. However, heavy metals concentration is usually high in poultry and livestock manures, also shown in our results. So, once such manures are applied, heavy metals included in these manures will easily be accumulated in soil and correspondingly cause soil and water pollution (Bomke, 1991; Han, 2000).

Heavy metals loading quantities in soil per year are calculated after these two organic fertilizers application. In general, nitrogen is applied in field at a rate of 250 kg/hm². So, if nitrogen concentration in organic fertilizer is 28 g/kg, organic fertilizer application quantity will be 8930 kg/hm². Heavy metals loading quantities in soil per year (HMLQ) can be calculated using following equation:

 $HMLQ = (application quantity of fertilizer) \times (metal concentration in organic fertilizer).$

Table 5 shows the heavy metals loading quantities in soil per year after using organic fertilizers I and II. If surface soil depth is assumed to be 1.2 m, and soil specific gravity to be 1.3 kg/dm³, soil Zn, Cu, Pb, Cd, Cr, As and Hg concentrations will increase 0.70 mg/kg, 0.12 mg/kg, 0.042 mg/kg, 0.0013 mg/kg, 0.17 mg/kg, 0.16 μg/kg and 0.12 μg/kg, respectively, each year when fertilizer I is used. When fertilizer II is applied, soil Zn, Cu, Pb, Cd, Cr, As and Hg concentrations will increase 1.30 mg/kg, 0.35 mg/kg, 0.12 mg/kg, 0.002 mg/kg, 0.087 mg/kg, 0.041 μg/kg and 0.001 μg/kg, respectively, per year. So, it is obvious that long-term application of organic fertilizer including high levels of heavy metals will easily result in metal accumulation in soil.

Table 5 Heavy metals loading quantities in soil per year after application of organic fertilizers

Organic fertiliz er s	Heavy metals loading quantities in soil per year, g/hm²								
made from	Zn	 Cu	Pb	Cd	Cr	As	Hg		
Milch cow manure	1816	305.0	3.801	3.40	430.	8 0.425	0.300		
Combined manures	3367	918.3	302.3	3 5.35	226.	8 0.108	0.003		

3 Conclusions

According to statistic results from 31 poultry and livestock farming plants, it was found that Cu, Zn, Pb, Cd, and Cr concentrations in feeds were high; meanwhile, animal manures also contained high concentration of heavy metals. So, it suggested that maximum concentration of feed additives in poultry and livestock farming should be regulated and enforced in China as soon as possible. Additionally, basic and application research on poultry and livestock manures need to be performed and much perfect standard should be established. If so, poultry and livestock manures will become a cost-effective and environment-friendly organic fertilizer source in green agriculture production.

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