

# Heavy metal content of manures in Switzerland

*Teneurs en métaux lourds dans les déjections animales en Suisse.*

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## Abstract

*Manures contribute considerably to heavy metal loads on agricultural soils. To assess the importance of heavy metals in manures, we analyzed the content of copper (Cu), zinc (Zn), cadmium (Cd) and lead (Pb) in about 1100 manure samples. Special attention was given to manures from pigs and beef cattle (600 and 250 samples respectively; balance experiments on different farms). The range of heavy metal contents was large for all animal categories, manure types and elements. Differences could mostly be explained by the content of the rations. On a dry matter basis, the Cu and Zn content was higher for pig manures than for cattle and poultry manures. These differences almost disappeared when calculated per unit phosphorus. The Cd and Pb content was higher in cattle manures than in pig and poultry manures due to the grass based rations. The mean (median) content of Cu and Zn in pig manures and the Cd and Pb content in cattle, pig and poultry manure have decreased considerably between 1990 and 1995 (Cu 28 %, Zn 17 %, Cd 56 %, Pb 49 %). This demonstrates the effect of air protection measures (Cd and Pb) and of recommendations to limit heavy metals in animal feed according to animal needs. At average animal density and mean heavy metal contents, heavy metal inputs from manures clearly exceed plant uptake only for Zn in pig and poultry production. At maximum heavy metal contents, inputs from manure can even exceed maximum heavy metal loads allowed for sewage sludge use in agriculture.*

Key words : manure, slurry, heavy metals, environment, feeding, pigs

## Résumé

Les déjections animales contribuent considérablement à la charge en métaux lourds apportés aux sols. Afin de vérifier l'importance des teneurs en métaux lourds, nous avons analysé la teneur en cuivre (Cu), zinc (Zn), cadmium (Cd) et plomb (Pb) dans environ 1100 échantillons d'effluents d'élevage. Une attention particulière a été portée aux déjections porcines et bovines viande (600 et 250 échantillons respectivement). La teneur en métaux lourds varie considérablement pour toutes les catégories d'animaux, types de déjections. Les différences s'expliquent notamment par le contenu de l'alimentation. Sur une base de matière sèche, la teneur en cuivre et zinc

est supérieure pour les lisiers et fumiers de porcs que pour les bovins et déjections avicoles. La teneur en cadmium et plomb était supérieure pour les déjections bovines que pour les porcs et volailles à cause de l'alimentation à base d'herbe/fourrage.

Les concentrations moyennes (médianes) en Cu et Zn dans les déjections porcines et en Cd, et Pb dans les déjections bovines, porcines et avicoles, ont diminué considérablement entre 1990 et 1995. Cela démontre l'effet des mesures de protection de l'air (pour Pb et Cd) et des préconisations pour adapter les métaux lourds dans l'aliment en fonction des besoins des animaux. En prenant en compte les densités moyennes animales et la moyenne en métaux lourds, les apports de métaux par les déjections excèdent les exportations par les plantes seulement pour Zn en production porcine et avicole.

Aux doses maximales rencontrées, les apports en métaux lourds par les déjections dépassent les charges en métaux autorisées pour l'épandage de boues de stations d'épuration en agriculture.

Mots-clés : déjections, lisier, métaux lourds, environnement, alimentation, porcs.

## **1. Introduction**

Fertilizers, pesticides and deposition from the atmosphere introduce heavy metals to agricultural land. If the input surpasses plant uptake these heavy metals will accumulate in the soil. There is a growing awareness, that the heavy metal content of the soil is increasing at many locations and that critical levels could be reached on some surfaces within the foreseeable future if inputs are not reduced (e.g. Keller and Desales, 1997). Heavy metal loads on agricultural surfaces in Switzerland were studied for example by Buwal (1993), von Steiger and Baccini (1990), von Steiger and Obrist (1993), Frei et al. (1993) or Studer et al. (1995). Apart from some measurements on a small number of farms, these calculations used available databases to estimate the heavy metal input by agricultural management. Whilst quite detailed knowledge is available of the heavy metal content of mineral fertilizers (Buwal, 1991) and sewage sludge (Candinas et al., 1991), the data on manure is rather scarce. A first survey showed that heavy metal contents vary considerably between manure types, animal categories and farms (Menzi et al., 1993). This study also suggested, that the highest loads could be expected on pig farms. To get more reliable information about this variability and its causes, a more detailed study of the heavy metal content of different types of manure was organized which included a number of samples from other experiments as well as detailed balance experiments on pig and beef cattle farms.

## **2. Material and methods**

Between 1991 and 1997 a total of about 1100 slurry and solid manure samples from cattle, pig and poultry production were analyzed for their content of copper (Cu), zinc (Zn), cadmium (Cd) and lead (Pb; table 1). Other metals could only be analyzed since 1996 and will therefore not be presented here. The content of dry matter (DM), total

nitrogen (N), ammonium nitrogen ( $N_{NH_4}$ ), phosphorous (P), potassium (K), magnesium (Mg) and calcium (Ca) were also analyzed in all samples.

Animal category	Manure type	Cu + Zn	Cd + Pb
dairy cattle	liquid manure (all excrements)	48	48
	slurry (urine, some dung)	90	67
	solid manure stacked (dung, some urine, straw)	94	82
beef cattle	liquid manure	200	197
	farmyard manure (all excrements, straw)	54	53
fattening pigs	pig slurry	194	198
dry sows	pig slurry	81	75
sows with piglets/weaners	pig slurry	198	189
pigs undefined	pig slurry	47	26
	solid manure	41	42
laying hens	litter from belts	10	10
	litter deep pits	43	36
broilers, turkeys	litter	59	56
Total		1159	1079

*Table 1*

*Number of samples of different manure types which were analyzed for their content of copper (Cu), zinc (Zn), cadmium (Cd) and lead (Pb) from 1992 till 1997.*

For dairy cattle and poultry the samples were mostly taken from other projects working under farm conditions. These observations usually consisted of one to three replicate samples. For pigs most samples came from detailed balance experiments on 13 farms with fatteners and 11 farms with breeding sows. For breeding sows the following two groups were distinguished: "dry sows" and "sows with piglets/weaners". Weaners could not be distinguished as a special group because many farms remove the sow after weaning and leave the weaners in the same compartment. The balance experiments usually consisted of two to four periods of one to several weeks, depending on conditions and technical possibilities on the farms. Three to eight replicate samples were taken per observation period from different locations in the slurry channel, the slurry pit or of the deep litter solid manure at the time of removal. The quantity of slurry or solid manure produced during the experiment period was also determined. At least three times per farm the mineral and heavy metal content of the ration and of individual feed components were analyzed and the feed consumption measured. Thus, the measured excretion could be compared to the one calculated from feed uptake and retention in animals. Similar balance experiments were done on 12 beef fattening farms.

Special attention was given to a good homogenization of the slurry and to the taking of representative manure samples. Preliminary balance experiments had shown that especially pig slurry samples taken with a scoop tended to have a lower content of

elements excreted primarily in the feces (e.g. P content up to 40 % too low) than expected according to the balance calculations. A special probe which takes a sample over the whole depth of the slurry was therefore usually used and samples were taken with running homogenization equipment wherever possible.

Except for poultry, where the number of samples was relatively low, the number of samples analyzed and the selection of farms from which the samples originated can be considered to be quite representative for Swiss production.

After drying at 105 °C, 2.5 g of each sample were pulverized and ashed at 500 °C. The ash was extracted with 80 ml concentrated hydrochloric acid and diluted to 200 ml with water (final dilution 1:80). For the analysis this solution was further diluted by 1:10. The mineral content was analyzed by ICP-AES; Cu, Zn (until 1993), Cd and Pb (until 1995) by AAS. Starting in 1994 Cu and Zn were also analyzed by ICP-AES and starting in 1996 all other heavy metals could be analyzed by ICP-MS. Comparative analyses of selected samples showed that these changes in method did not have a statistically significant effect on the results. We used the average contents of replicate samples for further calculations on specific farms and median values for the mean concentrations of different farms. Heavy metal contents are presented relative to the DM or P content of the manure to facilitate the interpretation of the results in spite of the inevitable variability of the dilution of the slurries and the different DM content of slurry and solid manure.

To estimate the total heavy metal load in Swiss manure we multiplied the metal content per unit P of cattle liquid manure (dairy and beef cattle), pig slurry and poultry manure (belt litter hens, broiler litter) with the quantity of P excreted by the corresponding livestock category. This was calculated from animal census data and standard P excretion per animal (Walther et al., 1994). For horses, sheep and goats the metal content of cattle manure was used. The thus calculated heavy metal load does not include the quantity introduced by straw and other litter material.

### **3. Results and discussion**

#### **3.1. Heavy metal content of manure per unit dry matter**

On a dry matter basis, Cu- and Zn-contents were clearly higher for pigs (especially those of fattening pigs and of sows with piglets/weaners) than for other animals (table 2). This can be explained by the Cu and Zn added to pig feed for their antimicrobial and growth stimulating effect (Kessler et al., 1994). A high Cu content in the slurry was often linked with a high Zn content. As expected, the balance experiments clearly demonstrated a close dependence between the heavy metal content of the ration and the manure produced. As Cu and Zn are mainly used for weaners, the highest values

were measured for this animal category (where it could be analyzed separately from the sows), followed by sows with piglets/weaners and fattening pigs (figure 1).

		Cu	Zn	Cd	Pb	Cu	Zn	Cd	Pb
Animal category	Manure type	$\mu\text{g g}^{-1}$ DM				$\text{mg g}^{-1}$ P			
dairy cattle	liquid manure	37.1	162.2	0.178	3.77	4.1	17.6	0.019	0.40
	slurry	19.1	123.3	0.160	2.92	2.7	16.6	0.022	0.42
	solid manure	23.9	117.7	0.172	3.77	3.2	16.1	0.025	0.49
beef cattle	liquid manure	52.5	244.7	0.168	2.98	5.1	20.9	0.016	0.31
	solid manure	22.0	91.1	0.151	2.81	5.1	17.3	0.033	0.50
fattening pigs	liquid manure	115.3	746.5	0.210	1.76	4.6	30.4	0.008	0.07
dry sows	liquid manure	71.1	517.5	0.170	2.53	3.7	22.9	0.009	0.11
sows with piglets/weaners	liquid manure	119.2	553.8	0.230	2.55	6.3	31.7	0.013	0.13
laying hens	litter from belts	35.2	425.3	0.310	2.22	1.8	23.6	0.014	0.10
	litter deep pits	44.0	511.5	0.195	2.25	1.5	19.3	0.008	0.08
broilers, turkeys	litter	43.8	349.2	0.292	2.92	3.9	27.0	0.024	0.24

*Table 2*

*Median values of the Cu, Zn, Cd and Pb analyzed in different types of manure.  
For number of samples see table 1.*

The lowest Cu- and Zn-values were found for dairy cattle, where heavy metals are only added to the ration according to animal requirements. For beef cattle somewhat higher median values were found, mainly due to some high values. Poultry manure had a slightly higher mean Cu- and mainly Zn-content than cattle manure. The lower content of solid manure as compared to that of liquid manure of dairy and beef cattle as well as pigs can be explained by the "dilution" achieved by adding straw and probably also by the different origin of the samples and the small number of solid manure samples analyzed.

a) breeding pigs

b) fatteners

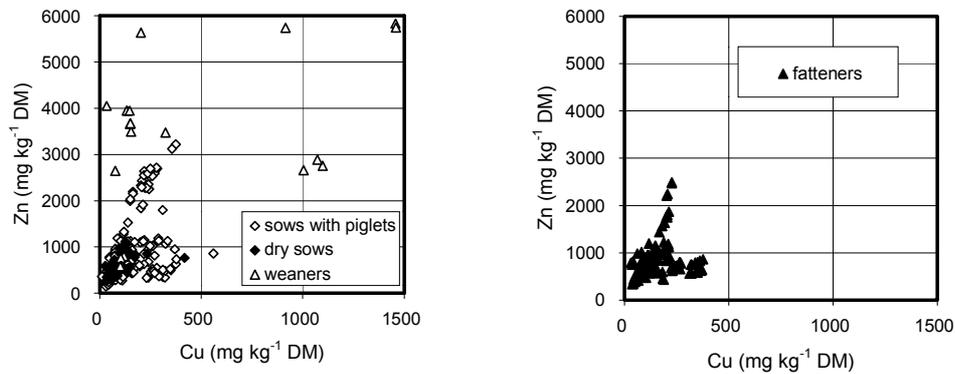


Figure 1  
*Cu and Zn content (mg kg<sup>-1</sup> DM) of pig slurries for breeding pigs*  
 a) and for fatteners b). Weaners could only be considered in a separate category if they were moved to a separate compartment after weaning

The Pb content per unit DM was highest for cattle. This can be explained by the fact that Pb is introduced into the agricultural cycle mainly by deposition from the atmosphere. Animals with a high proportion of roughage will therefore take up higher amounts of Pb than those fed mainly on grains. For Cd there was no clear difference between manure types and animal categories.

For all heavy metals studied, the range between the highest and the lowest values found was considerable (table 3). The fact, that except for poultry, where the number of samples was too low, the median value was always clearly in the lower half of the total range, demonstrates that a few exceptionally high values were responsible for the wide range (see also fig. 1). In the balance experiments, these few high values could always be traced back to high inputs in the feed. Especially for weaners, the Cu and Zn content in the ration often surpassed the recommended value considerably (recommended: 6 mg Cu and 95 mg Zn per kg feed; average content 88 mg kg<sup>-1</sup> Cu, 794 mg kg<sup>-1</sup> Zn; maximum values 169 mg kg<sup>-1</sup> Cu, 2525 mg kg<sup>-1</sup> Zn). Relative to the median value, the range of contents in the manure was highest for Pb. This was probably mainly due to regional differences in Pb-deposition.

In general the heavy metal contents of different manures agreed well with those recently reported for other countries (Chambers et al., 1998; Unwin, 1998). Where Cu and Zn use in feeding is known to differ between countries, this also showed in the manure. Compared to the values cited from different authors by Wilcke and Döhler (1995) the mean heavy metal content of manures in our study were generally lower. The difference is considerable for Cd and Pb in all categories and for Cu and Zn in pig slurry. It is probably mainly due to the rapid reduction of Cd and Pb emissions in the past years and to somewhat lower metal contents in pig rations in Switzerland.

### 3.2. Heavy metal content per unit P

If the heavy metal content is expressed per unit P, all animal species had about the

same median value for Cu, and the Zn-content of pig slurry was only slightly higher than that of cattle slurry. This can mainly be explained by the higher P-content of pig slurry. As long as manure is applied in doses adjusted to crop needs, this should insure that heavy metal loads per hectare are not higher for pig than for cattle slurry. Nevertheless, the metal content per unit P also differed considerably from farm to farm, and the maximum values were clearly higher in pig than in cattle slurry.

Animal category	Manure type	Cu	Zn	Cd	Pb
		$\mu\text{g g}^{-1}$ DM			
dairy cattle	liquid manure	13-160	102-395	<0.08-3.2	1.3-50
	slurry	0.5-188	44-716	0.08-1.5	1-17.3
	solid manure	2.5-80	40-412	0.04-3.1	0.09-15.6
beef cattle	liquid manure	12-267	88-938	<0.08-0.80	0.3-14.2
	solid manure	15-51	49-448	<0.08-0.62	1.3-11.9
fattening pigs	liquid manure	30-376	337-2490	<0.08-0.51	0.9-15.8
dry sows	liquid manure	28-418	269-1112	0.09-0.56	1.2-23.9
sows with piglets/weaners	liquid manure	12-1459	146-5832	0.06-1.3	0.34-12.8
laying hens	litter from belts	24-119	379-533	0.25-0.38	1.5-3.0
	litter deep pits	17-486	237-789	0.09-0.42	1.5-4.1
broilers, turkeys	litter	8.4-88	52-437	0.11-0.76	1.7-23.7

*Table 3*  
*Range (minimum and maximum) of values found for the Cu, Zn, Cd and Pb content of different types of manure. For number of samples see table 1.*

### 3.3. Heavy metal load per hectare

To assess, if heavy metals in manure are an ecological problem or not, the load applied to the land must be compared with the typical plant uptake. Only if this balance is clearly positive, an accumulation will take place, which in the long run might impair soil fertility. With the ecological restrictions Swiss farmers have to fulfill, it can be assumed that manure will not usually be applied in quantities far exceeding the nutrient demand of crops. On the other hand, it is quite common that farmers cover the whole nutrient demand of the crop with manure. The heavy metal load applied with manure can therefore normally be assumed not to exceed the amount applied with the manure dose covering the P demand of the crop. Table 4 therefore shows the metal load in a manure dose equivalent to 70 kg P<sub>2</sub>O<sub>5</sub> which is assumed as a mean crop demand (manure content and crop demand according to Walther et al., 1994). As a simplification only the data for the most important manure types (liquid manure for cattle and pigs) are presented. As can be deduced from the metal content per unit P in table 2, other manure type would not differ much.

	Cu	Zn	Cd	Pb
$\text{g ha}^{-1} \text{ year}^{-1}$				
crop uptake				

meadow 10 t DM year <sup>-1</sup>	55-210	270-580	0.3-1.5	20-62
wheat grain	20-29	164-232	0.21-0.52	0.53-11.1
wheat straw	35-73	79-119	0.86-1.39	18.5-26.4
load with 70 kg P <sub>2</sub> O <sub>5</sub> ha <sup>-1</sup> year <sup>-1</sup>				
liquid manure dairy cattle	124	537	0.57	12.3
liquid manure beef cattle	156	639	0.49	9.5
pig slurry (fatteners)	139	929	0.25	2.2
pig slurry (breeding pigs) <sup>1)</sup>	161	861	0.35	3.7
poultry manure (hens, belts)	56	724	0.43	3.2
poultry manure broilers	118	824	0.73	7.4

<sup>1)</sup> Weighted mean of dry sows (0.4) and lactating sows with piglets/weaners (0.6).

Table 4.

*Comparison of the heavy metal uptake of an intensive meadow (yield: 10 t DM) or wheat (yield: 6 t grains, 7.5 t straw) with the heavy metal load in the dose of different manures equivalent to 70 kg ha<sup>-1</sup> P<sub>2</sub>O<sub>5</sub>. Range of heavy metal content of grass and wheat according to von Steiger and Baccini (1990).*

If the total phosphorus demand of the crop is covered by manure of average metal content, the heavy metal load would only clearly surpass crop uptake in the case of Zn from pig and poultry manure.

If the maximum heavy metal content found in our study would be assumed, the Cu and Zn load would by far exceed crop uptake and even maximum loads allowed for sewage sludge (1000 g ha<sup>-1</sup> Cu and 3333 g ha<sup>-1</sup> per year). The Cd and Pb dose per hectare with mean manure contents lies clearly below crop uptake for all animal categories.

A dose of 70 kg P<sub>2</sub>O<sub>5</sub> is equivalent to 2.0 Swiss livestock units (LU). At the average Swiss livestock density of about 1.3 LU ha<sup>-1</sup> the load per hectare at mean manure content would substantially surpass crop uptake only in the case of Zn in pig and poultry manure. Provided that the heavy metal content of the manure is not above average, animal husbandry at such animal density can therefore be assumed not to cause metal loads that harm soil fertility. Nevertheless, the total heavy metal load can only be assessed if the heavy metal deposition and the input due to other fertilizers and pesticides is also known. The median value of five deposition monitoring sites in agricultural regions of Switzerland (1990-1994) was 41 g ha<sup>-1</sup> Cu, 159 g ha<sup>-1</sup> Zn, 1.1 g ha<sup>-1</sup> Cd and 37 g ha<sup>-1</sup> Pb (BUWAL, 1995; for Cu Studer, personal communication). The average metal load per hectare of agricultural land (excluding alpine pastures) from mineral fertilizers in Switzerland in 1990 (Frei et al., 1993) can be calculated at 6.5 g ha<sup>-1</sup> Cu, 42 g ha<sup>-1</sup> Zn, 1.0 g ha<sup>-1</sup> Cd and 4.0 g ha<sup>-1</sup> Pb. The metal input due to sewage sludge, compost and pesticides can be assumed to be of local importance only, because they are used only on a small proportion of the land. Using these data it can be estimated that normal agricultural management at average animal density and without high heavy metal inputs in sewage sludge, compost or pesticides does not lead to excessive heavy metal loads to the land. A slight surplus exists for Zn on pig and poultry farms and for Cd on farms using mineral P fertilizers. The latter can be

assumed to have decreased recently because of new limits for the Cd content of fertilizers introduced in 1992 (50 g Cd per t P).

### 3.4. Total heavy metal load in manures

In 1995 the total heavy metal load in manures in Switzerland amounted to 94 t Cu, 453 t Zn, 0.375 t Cd and 7.43 t Pb (table 5). Of this load 64 % (Zn) to 87 % (Pb) were in cattle manure. This is not surprising, as cattle also contributed 70-80 % of the N and P excretion. Compared to the heavy metal load in manure calculated for 1990 with heavy metal contents found in the preliminary study between 1988 and 1991 (Menzi et al., 1993), the load decreased by 28 % for Cu, 17 % for Zn, 56 % for Cd and 49 % for Pb. Only 6 % (Cu and Zn) and 4 % (Cd and Pb) of this decrease (% of load 1990) was due to the reduction of the animal number. This illustrates a noticeable success of the efforts to reduce the Cu- and Zn-content in animal feed and of air protection measures in the case of Cd and Pb. It can be expected, that the reduction will continue thanks to new limits imposed on the Cu and Zn content of feed and continuing air protection measures.

A comparison of the heavy metal load in manure with that in other fertilizers shows that manure is responsible for about two thirds of the Cu and Zn load in fertilizers and for about 20 % of the Cd and Pb load. For the Cd and Pb load mineral fertilizers and sewage sludge respectively are the most important sources.

	Cu	Zn	Cd	Pb
	t year <sup>-1</sup> (% of total load from animal husbandry)			
1995 (animal numbers 1995, metal contents see table 2)				
cattle	67.5 (71)	291 (64)	0.301 (80)	6.458 (87)
pigs	19.7 (21)	122 (27)	0.038 (10)	0.364 (5)
poultry	1.9 (2)	18 (4)	0.013 (3)	0.112 (2)
horses, sheep, goats	5.0 (5)	22 (5)	0.023 (6)	0.497 (7)
total	94.1 (100)	453 (100)	0.375 (100)	7.431 (100)

1990 (animal numbers 1990, metal contents according to Menzi et al. 1993)				
cattle	73.0 (56)	344 (63)	0.697 (83)	12.822 (88)
pigs	50.2 (38)	164 (30)	0.086 (10)	0.784 (5)
poultry	2.3 (2)	16 (3)	0.016 (2)	0.133 (1)
horses, sheep, goats	4.7 (4)	22 (4)	0.045 (5)	0.832 (6)
total	130.2 (100)	547 (100)	0.843 (100)	14.570 (100)
load in other fertilizers in 1990 (Frei et al. 1993)				
mineral fertilizers	7	45	1.010	4.3
sewage sludge, compost	44	160	0.490	30.6
Reduction of load in manure between 1990 and 1995 (% , negative values = increase)				
cattle	7.5 %	15.4 %	56.8 %	49.6 %
pigs	60.7 %	26.0 %	55.5 %	53.5 %
poultry	20.0 %	-11.7	17.1 %	15.5 %
horses, sheep, goats	-6.1 %	2.4 %	49.2 %	40.3 %
total	27.7 %	17.2 %	55.5 %	49.0 %

*Table 5  
Heavy metal load in manures of different animal species in Switzerland in 1995 and comparison with the load in manures and other fertilizers in 1990.*

#### 4. Conclusions

Heavy metal contents vary considerably between manures from different animal types and from different farms. The highest median values were found in pig slurry, especially that of weaners or sows with piglets/weaners. Maximum values were up to ten times higher than median values. The great variability between farms shows that management (especially feed content) is of major importance for the heavy metal content in manures. Results from many farms show that animal production with low heavy metal turnover is possible. Therefore, the extremely high values found on some farms should be reduced immediately. Pig production (especially weaners) would have the biggest potential for improvements.

Manure is the most important cause of Cu and Zn input to Swiss crop production. Even though the content of these elements in manure has gone back considerably in the past years, the load per hectare is still near or above crop uptake at average animal density. Thus, an accumulation will take place. To avoid this and to secure long term soil fertility, the heavy metal content in animal feed should not surpass animal requirements. This limitation is especially justified and feasible for Cu, where contents in pig feed are often below the range where a clear positive influence on animal growth can be expected but far above official recommendations and animal requirements.

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