SLUDGE QUALITY IN SWEDEN – INQUIRY RESULTS FOR YEAR 1995 TO 1997

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ABSTRACT

Sludge use on agricultural land is at present often considered to be the preferred alternative for the recovery of nutrients in sludge. This work on evaluation of sludge quality is based on the responses from Swedish municipalities to an inquiry on sludge quality and disposal. There are regulations with limits for maximum permissible concentration in sludge for Pb, Cd, Cu, Cr, Hg, Ni, Zn, nonylphenol, toluen, PAH and PCB. For the metals there are restrictions for addition of metals to agricultural soil then fertilising with sludge. Based on the inquiry, mean and median values for all restricted pollutants and the nutrient phosphorus have been calculated. The reported concentrations were in general agreement with concentrations reported by SNV and SCB (2000). The evaluated regression coefficients among the metals in sludge did not show any significant correlation. To evaluate sludge quality, the pollutant contents were compared to the limit for maximum permissible concentration. The reported sludge contents indicated that 49 % of the sludges had some pollutant with a concentration higher than the limit, in spite of the mean value of analysis over the limit being only 6.3 %. From the analysis of phosphorus and metal contents, the amount of metal added to soil at fertilising with sludge equivalent to 10, 20 and 30 kg phosphorus per hectare and year were calculated. The largest per cent of sludges exceeding the permissible limit is 96% for cadmium followed by 75% for lead at 30 kg P/ha, year. For the restrictions imposed from year 2000 and a sludge amount comparative to 10 kg P/ha, year 26 % of the sludges had some metal with a concentration larger than the permissible limit. Whereas at 20 kg P/ha, year 81 % of the sludges had a metal with a concentration larger than the permissible limit and for 30 kg P/ha, year as much as 96 % of the sludges had a metal with a concentration larger than the permissible limit.

KEYWORDS

Sewage sludge, metal content, inorganic pollutant content

INTRODUCTION

Sustainable sludge handling has as an important goal to recycle resources without supply of harmful substances to humans or the environment (Hultman and Levlin, 1997 and Hultman et al., 1997). Sludge use on agricultural land is at present the main alternative for the use of sludge as a resource. The main nutrients in sewage sludge are potassium, calcium, phosphate and nitrogen, of which phosphorus is most important to recover. Swedish agriculture uses annually 50 000 tonnes phosphorus as fertiliser, of which 2 500 tonnes are in form of sewage sludge and 20 000 tonnes in form of stable dung (Tideström et al., 2000). The amount of phosphorus in sewage sludge is about 9 000 tonnes per year. The goal for SNV (Swedish Environmental Protection Agency) is that 90 % if the sludge shall be used as fertiliser on agricultural land year 2010. However, increasingly more stringent requirements, resistance from farmers, food industry or the public have made the use of sludge for agriculture less possible. Sludge fertilising cause accumulation in the soil of metals originating from the sludge (Kelly et al., 1999, Tideström et al., 2000, Walter and Cuevas, 1999), which may cause higher uptake of metals to crops and thus create a human health risk. However, field tests with sludge fertilising by Andersson and Nilsson (1999) showed no increased metal uptake by the plants. Sludge fertilising increases the precipitated metal fraction and the soluble metal fraction remains the same

(Walter and Cuevas, 1999). Efforts must be taken to reduce metal contents in sludge and thereby prevent metal increase of soil when fertilising with sewage sludge.

In Sweden, there are regulations with limits for maximum concentration in sludge for the metals lead, cadmium, copper, chromium, mercury, nickel and zinc (Swedish legislation SFS 1998:944) and also for the addition of metals to agricultural soil then fertilising with sludge (Swedish legislation SNFS 1994:2). The restrictions of addition of metals were made more stringent from year 2000. Table 1 shows the restrictions for metals and the year from which the restrictions are in force. Based on an agreement between LRF (Swedish Farmers Association), VAV (Swedish Water and Wastewater Association) and SNV, there are also restrictions for some organic pollutants (SNV 1995 and 1996). The restricted pollutants are nonylphenol, toluen, PAH (sum of 6 species of polyaromatic hydrocarbons) and PCB (sum of 7 species of polychlorinated biphenyls). The limit for permissible maximum organic pollutant in sludge are for nonylphenol, 50 mg/kg DS, toluen, 5 mg/kg DS, PAH, 3 mg/kg DS, and PCB 0.4 mg/kg DS.

Table 1. Limits for permissible maximum metal concentration in sludge and limits for maximum addition of metals to agricultural soil then fertilising with sludge.

Limit		In force from	Cd	Cr	Cu	Hg	Ni	Pb	Zn
In sludge,	mg/kg DS	August 1999	2.00	100	600	2.5	50	100	800
Addition to soil,	g/ha,year	1995	1.75	100	600	2.5	50	100	800
Addition to soil,	g/ha,year	2000	0.75	40	300	1.5	25	25	600

This work on evaluation of sludge quality is based on the response to an inquiry to the Swedish municipalities made by VAV. The inquiry included among other questions the content of the restricted pollutant and phosphorus in the sludge. The evaluation will help to estimate the possibility to use sludge for fertilising agricultural soil. The response of the inquiry was 176 answers from 171 municipalities for year 1995, 227 answers from 216 municipalities for year 1996 and 192 answers from 171 municipalities for year 1997. There are totally 283 municipalities in Sweden. The number of municipalities answering the enquiry do not correspond to the number of sludge analysis. Some municipalities with many sewage treatment plants have reported analysis for each treatment plant, while other as Stockholm with three treatment plant has reported mean values for all three plants together. Some municipalities especially in the suburbs of Stockholm, having a treatment plant together with other municipalities, have not given analysis of the sewage sludge.

REPORTED POLLUTANT CONTENTS

Table 2 shows the reported content of phosphorus, metals and organic pollutants in sewage sludge for year 1995 to 1997 as mean values, median values and the standard deviation. Log-normal distribution diagrams for analysis between $Q_{5\%}$ and $Q_{95\%}$ for year 1997 are shown by figure 1 for metals and phosphorus and by figure 2 for the organic pollutants. If the difference in analyses is caused randomly it will be shown by a straight line in a log-normal distribution diagram. The figures show that there is a larger variation in inorganic pollutant concentrations than for metal concentrations. A large variation in concentrations were obtained for toluen there the $Q_{95\%}$ value is 200 times larger than the $Q_{5\%}$ value.

Table 3 shows pollutant concentrations in sewage sludge for year 1995, 1996 and 1997, obtained by dividing the total amount of each pollutant by the total sludge amount. The total amount of each pollutant was obtained by adding the amount of pollutant in each sludge, which is the concentration multiplied with amount of sludge. The thus calculated concentration corresponds to the concentration achieved by mixing all sludges together. The concentrations are a little larger than in table 2, which shows that sludges from larger treatment plants, which have a larger influence on the values of table 3, have larger pollutant contents than sludge from small treatment plants. From table 4 which shows the regressions coefficients between metal contents, some correlation can be noted to occur between Zn and the other inorganic species except for Ni. The estimated regression coefficient between Pb and Zn is largest, about 0.58, and the coefficient for Pb and Cd is 0.51. No significant negative correlation was found between any of the reported analysis.

Table 2 Reported concentrations (of DS, dry substance) of phosphorus, metals and organic pollutants contents in sewage sludge for year 1995 to 1997 as mean values, median values and the standard deviation.

	1995				1996				1997			
	Answ.	Mean	Med.	St. dev	Answ.	Mean	Med.	St. dev	Answ.	Mean	Med.	St. dev
Phosphorus. %	122*	2.35	2.25	1.15	167*	2.54	2.55	0.891	161*	2.86	2.5	3.34
Metals. mg/kg												
Cadmium, Cd	142*	2.08	1.1	8.52	191*	1.22	1.01	1.02	183*	1.42	1.1	2.51
Chromium, Cr	142*	32.4	24.75	34.8	190*	38.2	38.3	131	187*	28.2	23	18.2
Copper, Cu	142*	311	251	245	191*	318	273	222	185*	323	269	247
Lead, Pb	142*	36.2	31	30.8	190*	32.3	27	28.0	187*	32.3	27	34.2
Mercury, Hg	142*	10.0	0.94	104	191*	0.963	0.968	0.604	186*	1.27	0.79	3.63
Nickel, Ni	141*	14.1	11.4	10.8	191*	13.4	13.4	10.8	187*	15.8	12	16.7
Zink, Zn	142*	462	431.5	193	191*	448	450	174	187*	460	450	168
Organic, mg/kg												
Nonylphenol	135*	37.7	34	27.0	183*	34.9	35.1	33.0	182*	23.8	16	24.7
Toluen	137*	4.03	0.9	10.6	183*	3.60	3.62	7.96	181*	4.18	1	10.7
PAH	120*	1.24	1.045	0.893	170*	2.27	2.28	12.3	171*	1.45	1.22	1.87
PCB	131*	1.06	0.07	0.179	177*	0.455	0.457	4.66	177*	0.482	0.0585	4.89
*Of total:	142 answers				192 answers				187 answers			

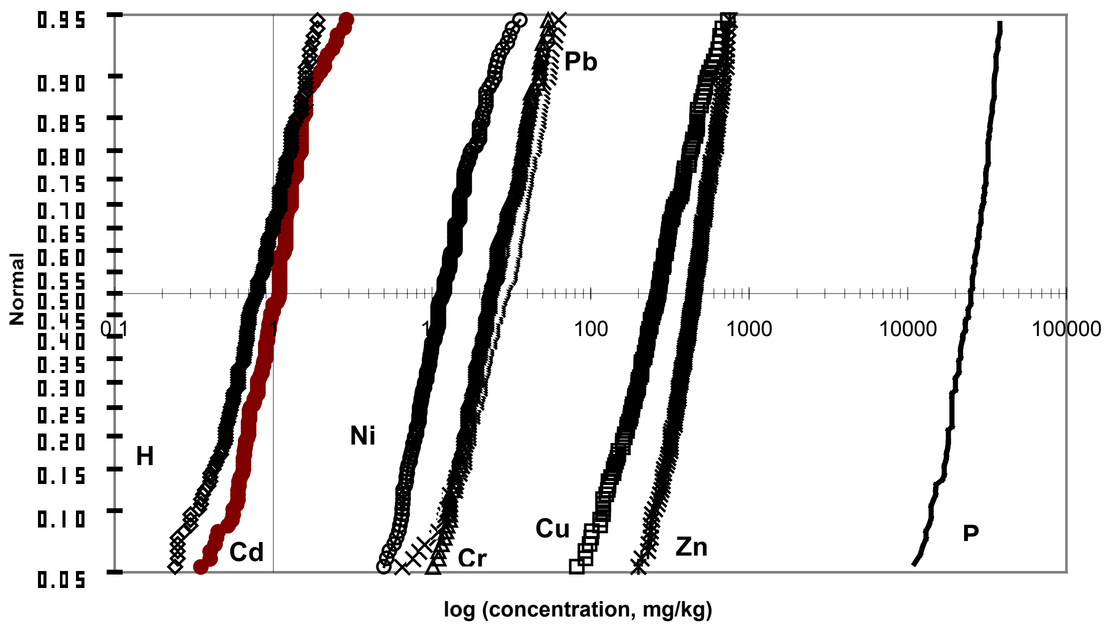

Figure 1. Log-normal distribution diagram for metals and phosphorus for sludge analysis between $Q_{5\%}$ and $Q_{95\%}$ for year 1997.

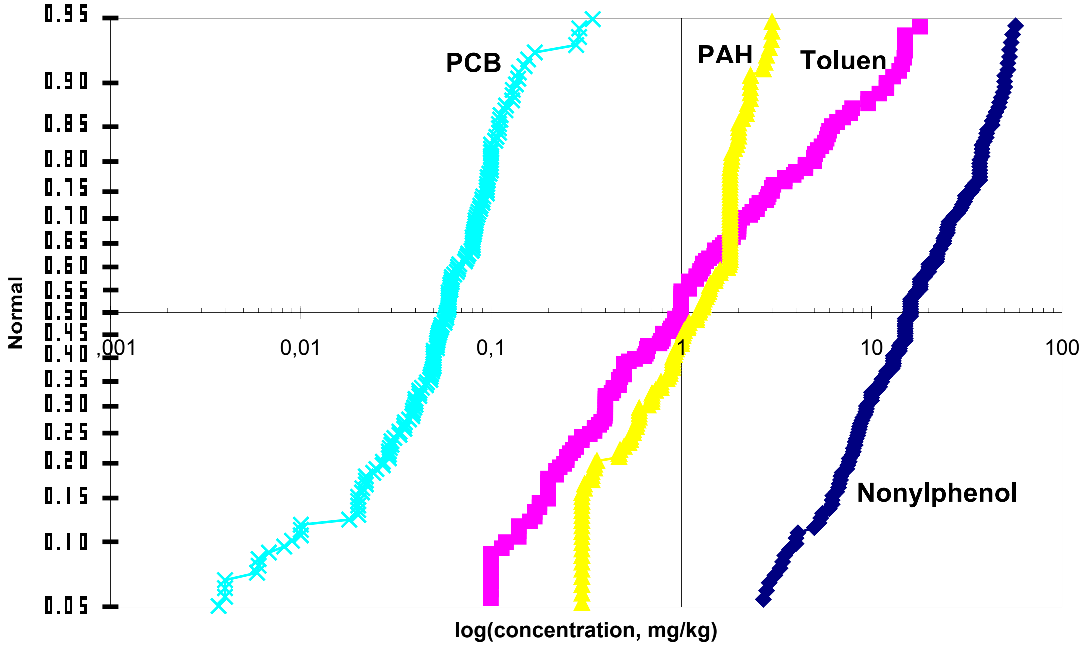

Figure 2. Log-normal distribution diagram for organic pollutants for sludge analysis between $Q_{5\%}$ and $Q_{95\%}$ for year 1997.

Table 3. Pollutant contents in sewage sludge for year 1995, 1996 and 1997, calculated by adding concentrations multiplied with sludge amount and dividing the sum with the total sludge amount.

		1995	1996	1997
Phosphorus, P	%	2.02	2.71	2.66
Lead, Pb	mg/kg DS	49.77	35.39	43.26
Cadmium, Cd	mg/kg DS	2.36	1.31	1.91
Copper, Cu	mg/kg DS	399.4	386.5	433.3
Chromium, Cr	mg/kg DS	38.01	34.13	28.82
Mercury, Hg	mg/kg DS	4.65	1.25	1.53
Nickel, Ni	mg/kg DS	17.79	16.10	15.87
Zinc, Zn	mg/kg DS	540.6	493.9	505.3
Nonylphenol	mg/kg DS	51.92	42.68	28.98
Toluen	mg/kg DS	2.32	2.68	3.10
Sum PAH	mg/kg DS	1.65	2.60	5.09
Sum PCB	mg/kg DS	0.137	0.476	0.126

Table 4. Calculated regression coefficients between metal contents in sewage sludge for year 1997.

	P	Cd	Cr	Cu	Pb	Hg	Ni	Zn
Phosphorus, P	1	0.1857	0.2188	0.2692	0.1962	0.3428	0.2047	0.4654
Cadmium, Cd		1	0.1380	0.1300	0.5118	0.1913	0.0905	0.4709
Chromium, Cr			1	-0.0131	0.1896	0.0738	0.2678	0.2177
Copper, Cu				1	0.3009	0.3719	0.0197	0.4489
Lead, Pb					1	0.2790	0.0957	0.5793
Mercury, Hg						1	0.0617	0.3990
Nickel, Ni							1	0.0707
Zinc, Zn								1

SLUDGE QUALITY EVALUATION

To evaluate the quality of sludge, the sludge analysis for year 1997, were compared with the maximum permissible pollutant concentration limits. Table 5 shows the permissible limit of metals and organic pollutions, per cent analyses larger than the limit and the quotient concentration divided by limit. The percent of analysis values larger than the limit is highest for toluen (20%) and nonylphenol (10%) and in case of the metals cadmium is highest (9.1%) followed by copper (8.6%). Since table 4 shows no significant correlation between the pollutant contents, 49 % of the reported sludges had some of the 11 restricted pollutant with a concentration higher than the permissible limit for maximum concentration, in spite of a mean value of only 6.3 % of the analysis over the permissible limit. Of 187 reported sludges analysis, 96 sludges (51 %) had no pollutant above limit, 65 sludges (35 %) one pollutant above the limit, 17 sludges (9 %) two pollutants above limit, 9 sludges (5 %) three pollutants above limit and no sludges had more than 3 pollutants above limit.

Table 5. Limits for maximum concentration of metals and organic pollutions, per cent analyses larger than limit and the quota concentration/limit for year 1997.

Specie	Limit, mg/kg DS	Per cent > limit	Quota concentration/limit 10-percentil	Median	90-percentil
Cadmium, Cd	2	9.1 %	0.295	0.55	0.94
Chromium, Cr	100	1.1 %	0.13	0.23	0.48
Copper, Cu	600	8.6 %	0.20	0.45	0.91
Lead, Pb	100	1.1 %	0.13	0.27	0.54
Mercury, Hg	2.5	2.7 %	0.14	0.32	0.64
Nickel, Ni	50	3.2 %	0.13	0.24	0.50
Zink, Zn	800	3.8 %	0.33	0.56	0.87
Nonylphenol	50	10.4 %	0.08	0.32	1.00
Toluen	5	20.0 %	0.028	0.20	2.40
Sum PAH	3	5.8 %	0.10	0.42	0.77
Sum PCB	0.4	4.8 %	0.06	0.15	0.35
Mean value		6.3 %	0.147	0.337	0.854
(Sum of quota concentration/limit)/reported analysis			0.234	0.407	0.734

When using sludge as fertiliser, the amount of metals added into the soil is calculated from the phosphorus need of the soil, the phosphorus content in the sludge and the metal content in the sludge:

$$\text{Added metal to soil (g/ha,year)} = \frac{\text{Phosphorus need (kg P/ha, year)}}{\text{Phosphorus content (\%P)/100}} \times \frac{\text{Metal content (mg/kg)}}{1000}$$

From the analysis of phosphorus and metal contents for year 1997 the amount of added metal to soil per hectare and year at fertilising with sludge equivalent to 10, 20 and 30 kg phosphorus per hectare and year was calculated. Table 6 shows analysis in number and per cent, where the added metal exceeds the limit for maximum addition valid from year 2000 and 1995. The table shows that more sludges exceed the limit at larger phosphorus requirements. The largest percent of sludges exceeding the limit is for cadmium (96% at 30 kg P/ha, year) followed by lead (75% at 30 kg P/ha, year).

Table 7 shows sludges divided by how many of the seven metals Pb, Cd, Cu, Cr, Hg, Ni and Zn, there the amount added metal to soil exceeds the limit. For the restrictions imposed from year 2000 and a sludge amount comparative to 10 kg P/ha, year 26 % of the sludges had some metal with a concentration larger than the permissible limit. For 20 kg P/ha, year 81 % of the sludges had a metal with a concentration larger than the permissible limit and for 30 kg P/ha, year as much as 96 % of the sludges had a metal with a concentration larger than the permissible limit.

Table 6. Analysis in number and per cent from 161 municipalities, there the metal concentration exceeds the limit for addition of metals to agricultural soil when fertilising with an amount of sludge equivalent to 10, 20 and 30 kg phosphorus per hectare and year valid from 1995 and from 2000.

	Metal	Limit g/ha, year	10 kg P/ha, year Number	10 kg P/ha, year Percent	20 kg P/ha, year Number	20 kg P/ha, year Percent	30 kg P/ha, year Number	30 kg P/ha, year Percent
2000	Cd	0.75	20	12 %	110	68 %	154	96 %
	Cr	40	3	1.9 %	17	11 %	45	28 %
	Cu	300	14	9 %	49	30 %	94	58 %
	Pb	25	17	11 %	68	42 %	121	75 %
	Hg	1.5	6	3.7 %	10	6 %	30	19 %
	Ni	25	6	3.7 %	14	9 %	25	16 %
	Zn	600	3	1.9 %	12	7 %	61	38 %
1995	Cd	1.75	9	5.6 %	16	9.9 %	35	22 %
	Cr	100	1	0.6 %	1	0.6 %	3	1.9 %
	Cu	600	3	1.9 %	14	8.7 %	27	17 %
	Pb	100	3	1.9 %	6	3.7 %	10	6.2 %
	Hg	2.5	3	1.9 %	7	4.3 %	9	5.6 %
	Ni	50	2	1.2 %	6	3.7 %	11	6.8 %
	Zn	800	3	1.9 %	5	3.1 %	23	14 %

Table 7. Sludge analysis from 161 municipalities, divided by how many of the 7 metals Pb, Cd, Cu, Cr, Hg, Ni and Zn, where the concentration exceeds the limit for addition of metals to agricultural soil when fertilising with an amount of sludge equivalent to 10, 20 and 30 kg phosphorus per hectare and year valid from 1995 and from 2000.

	Number of metals	10 kg P/ha, year Number	10 kg P/ha, year Per cent	20 kg P/ha, year Number	20 kg P/ha, year Per cent	30 kg P/ha, year Number	30 kg P/ha, year Per cent
2000	0	119	74 %	31	19 %	6	3.7 %
	1	29	18 %	44	27 %	10	6.2 %
	2	9	5.6 %	49	30 %	32	20 %
	3	1	0.6 %	22	14 %	41	25 %
	4	0	0 %	10	6.2 %	41	25 %
	5	0	0 %	1	0.6 %	21	13 %
	6	2	1.2 %	1	0.6 %	6	3.7 %
	7	1	0.6 %	3	1.9 %	4	2.5 %
1995	0	146	91 %	127	79 %	96	60 %
	1	10	6.2 %	26	16 %	36	22 %
	2	3	1.9 %	4	2.5 %	19	12 %
	3	1	0.6 %	1	0.6 %	6	3.7 %
	4	0	0 %	0	0 %	0	0 %
	5	1	0.6 %	0	0 %	1	0.6 %
	6	0	0 %	3	1.9 %	0	0 %
	7	0	0 %	0	0 %	3	1.8 %

DISCUSSION

The reported concentrations for phosphorus, metals and organic pollutants in sewage sludge can be compared with SNV and SCB (2000) statistics for year 1998. Table 8 shows median and mean values for nutrients, metals and organic pollutants in sludge divided in three classes depending on the size of the treatment plant in number of connected persons. Except for toluen and PCB the pollutant content appear to increase with the size of the treatment plant. The concentrations reported by SNV and SCB (2000) for year 1998 are in the same order of magnitude as the concentrations achieved from the VAV inquiry (table 2). About 60% of the sludges had all pollutants below the permissible limit for maximum concentration (Tideström et al., 2000).

Table 8. Median and mean values for nutrients, metals and organic pollutants in sludge for year 1998 divided in three classes depending on the number of persons connected to the treatment plant and total mean values for year 1998 and 1995 (SNV and SCB, 2000).

	Median values, mg/kg DS			Mean values, mg/kg DS			Total 1998	Total 1995
	Less than 20 000	20 001 – 100 000	More than 100 000	Less than 20 000	20 001 – 100 000	More than 100 000		
Phosphorus	21 500	28 000	32 500	21 177	27 739	31 108	27 702	27 600
Nitrogen	36 000	35 500	40 000	35 205	37 220	40 379	38 112	34 200
Lead	24	30.5	33.2	26.6	36.9	39.2	35.4	41.8
Cadmium	0.9	1.1	1.2	1	1.3	1.3	1.2	1.5
Copper	220	280	378	266.9	349.6	502.7	394.1	393.7
Chromium	21	26	28.5	26.3	30.8	35.7	35.7	33.6
Mercury	0.6	0.9	1.1	0.8	0.9	1.3	1.1	1.3
Nickel	10.1	12	19.6	12.6	19.9	19.7	18.2	16.5
Zinc	415	530	590	423.1	554.4	606.4	545.4	537.2
Nonylphenol	10.1	18.34	25.8	13.4	23.1	27	22.8	46.6
Toluen	0.9	0.7	0.4	12.3	4.1	1.1	4.5	2.3
PAH	0.7	1.1	1.4	1.2	2.2	1.8	1.8	1.8
PCB	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1

Figure 3 shows how median values for heavy metal content in sludge for a medium size treatment plant with 20 000 to 100 000 connected persons have changed during the years 1987 to 1998 (SNV and SCB, 2000). During the 80:ies where were a sizeable decrease for all metals in sludge. Storm water from roads contributes to a large part of the cadmium (53 % of total Cd) and lead (48 % of total Pb) in sewage sludge (Boller, 1997). The prohibition against the use of cadmium and mercury have given a decrease of these substances in sludge concentrations. However, cadmium is still used as for instance as stabilizer in plastic. Prohibition on the use of leaded fuel has given a successive decrease of lead concentration in sludge. The contents of chromium and nickel in sludge decreased until year 1998, but increased again between 1995 and 1998 with 6 % (table 8). This increase may be caused by increased use of stainless steel, which contains 18 % chromium and 8 % nickel. A large contribution to the copper content is dissolution of copper from copper pipes. Municipalities with hard drinking water often have large copper contents in the sludge. The dissolution of copper can be prevented by raising the pH-level, which must be combined with softening to prevent carbonate precipitation (Levlin, 1991 and 1993). Today brake coatings which have substituted asbestos coating, are large contributors to copper in road storm water (SNV, 2000).

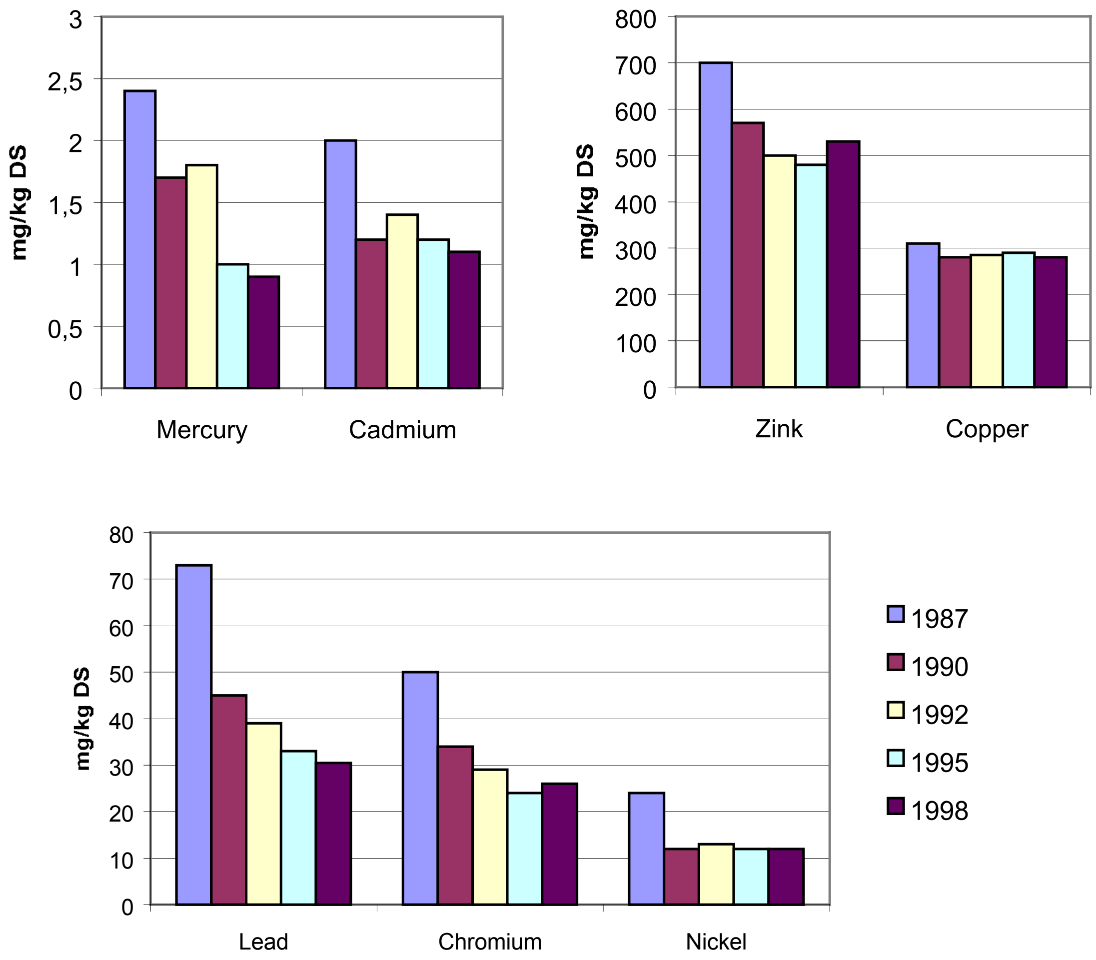

Figure 3. Heavy metal content in sludge for year 1987 to 1998 (SNV and SCB 2000).

The pollutant contents in Swedish sludge can also be compared with statistics from other countries. Table 9 shows pollutant concentrations in sludge from treatment plants in Denmark for year 1997 and 1987 (Miljøstyrelsen, 1999) and Germany (Werther and Ogada, 1999). Metal concentrations in Danish sludge have decreased from year 1987 to 1997. The concentrations in German sludge, which are from the end of the 80:ies, are equal to the concentrations in Danish sludge from 1987. The table also shows metal content in stable dung and wastes from compost toilets (VAV, 1989), which are smaller than the concentrations in sewage sludge.

Table 9. Metal contents in mg/kg DS in sludge from Denmark (Miljøstyrelsen, 1999), Germany (Werther and Ogada, 1999), wastes from compost toilets and stable dung (VAV, 1989).

	Cd	Cr	Cu	Hg	Ni	Pb	Zn
Denmark 1997	1.97	32.0	262	1.41	22.9	71.4	760
1987	4.99			3.76	48.0	174	
Germany	3.8	91	330	2.7	39	159	1318
Mould from compost toilets	0.5	1.4	17	0.3	3	3	120
Stable dung	0.3	4.5	37	0.08	7.1	5.6	190

Miljøstyrelsen (1999) have also reported the content of some organic pollutants in Danish sludge for year 1997. Table 10 shows the concentrations of LAS (linear alkylbenzensulfonathe), PAH (polyaromatic hydrocarbons), NPE (nonylphenol) and DEHP (dietylhexylphthalat). Since aerobic stabilisation of sludge decomposes organic pollutants, the mean concentrations for aerobic stabilised sludge and anaerobic stabilised (digested) sludge are also reported. The share of aerobic stabilised sludge was 43.7 % and the share of anaerobic digested sludge was 43.2 %.

Table 10. Inorganic pollutants in Danish sludge (Miljøstyrelsen, 1999).

		Mean value	Aerobic stabilised Median value	Anaerobic digested Median value
LAS	mg/kg DS	714.0	79.0	795.0
PAH	mg/kg DS	2.1	1.3	2.3
Nonylphenol	mg/kg DS	30.1	5.8	38.0
DEHP	mg/kg DS	26.3	22.0	25.0

CONCLUSIONS

Sludge use on agricultural land is often considered to be the preferred main alternative for the recovery of nutrients in sludge. However, increasing resistance from the farmers, food industries or public have, due to its content of metal and organic pollutants, made sewage sludge less attractive, as a source of fertiliser on agricultural soil.

The reported concentrations based on an inquiry to the Swedish municipalities, were in the same order of magnitude as the concentrations reported by SNV and SCB (2000).

In spite of only 6.3 % of the reported concentration above the permissible limit for maximum concentration in sludge for Pb, Cd, Cu, Cr, Hg, Ni, Zn, nonylphenol, toluen, PAH and PCB, 49 % of the reported sludges had some pollutant with a concentration higher than the limit.

The prohibitive restrictions, imposed from year 2000 on addition of metal to soil, have caused that at fertilising with sludge amount comparative to 10 kg P/ha, year 26 % of the sludges had some metal with a concentration larger than the permissible limit. Whereas, for 20 kg P/ha, year 81 % of the sludges had a metal with a concentration larger than the limit and for 30 kg P/ha, year as much as 96 % of the sludges had a metal with a concentration larger than the limit.

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