Cadmium balance in soils under different fertilization managements including sewage sludge application

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ABSTRACT

Simple balance of Cd input by different treatments and removal by agricultural crops was investigated in an ongoing precise long-term field experiment with application of sewage sludge (SS), farmyard manure (FYM), and mineral fertilizers (NPK). Potatoes, wheat and barley were grown in a rotation at 4 experimental sites of the Czech Republic with the aim to assess the risk of Cd accumulation in soil and plants under different soil and climate conditions. The results showed significant differences in Cd content of the input materials used, and in Cd inputs to soils under different fertilization managements. Three applications of sewage sludge during 1996–2005 resulted in total addition of 110 g Cd/ha into soil, which was by one order of magnitude higher than Cd addition in FYM or NPK treatments. From the total amount of sludge-borne Cd, only small portion was removed by harvests of crops (approximately 3.5%). Soil conditions significantly affected Cd input-removal balance. The highest Cd removals were obtained on soils with the lowest pH. The highest Cd removal was achieved by potato tubers followed by wheat and barley plants. The results showed that the risk of Cd accumulation in soils is high, especially with repeated sludge applications.

Keywords: sewage sludge; cadmium; input-removal balance; long-term field experiment

Sewage sludge is a solid waste produced during wastewater treatment. Its production is increasing due to growing pressure on water quality. Possible ways of sludge disposal encounter mainly application on farmland, land filling and incineration (Smith 1996). Since sewage sludge is a source of organic matter, macronutrients as well as trace elements and can improve physical, chemical and biological characteristics of soils, its recycling in agriculture is a preferable way of disposal. However, sludge application on farmland also represents a potential risk. Sludge usually contains environmentally hazardous substances as heavy metals, organic pollutants and pathogens. Application of sludge on farmland can therefore lead to accumulation of contaminants in soil and subsequent decrease of soil fertility, reduction of crop quality and yield, contamination of surface and ground water and entry of risky compounds into the food chain. Unlike organic compounds, heavy metals do not degrade in soil and they accumulate in organisms and in the environment and affect the health of plants and their consumers. When evaluating the sludge use on farmland, long-term research is needed because many of its effects, e.g. organic matter enrichment and the possible build up of toxic elements in soil, evolve slowly and are difficult to predict (Bergkvist et al. 2003).

Cadmium is one of the most toxic and available heavy metals found in sewage sludge and may limit its suitability for use in agriculture. However, cadmium is also added to soil by manure, phosphorus fertilizers, lime products and atmospheric deposition.

When sludge is applied into soils, Cd appears to remain in the zone of application and can increase Cd accumulation in plants grown on sludge-treated soils. Compared to phosphate fertilizers, the Cd

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Table 1. Soil characteristics

Site	Hněvčeves	Humpolec	Lukavec	Suchdol
Soil type	Luvisol	Cambisol	Cambisol	Chernozem
Soil texture	loam	loam	loam	clay-loam
pH_{KCl}	5.7	4.5	6.1	7.0
C_{ox} (%)	1.8	2	1.8	2.3
CEC (mval/kg)	179	159	128	255
$P_{Mehlich\ III}$ (mg/kg)	84	123	124	112
$K_{Mehlich\ III}$ (mg/kg)	251	286	245	223
$Mg_{Mehlich\ III}$ (mg/kg)	157	137	114	259
Cd_{total} (mg/kg)	0.270	0.377	0.375	0.428

Table 2. Characteristics of the sewage sludge

Year of application	Application rate of fresh matter (t/ha)	N content in fresh matter (%)	DM (%)	Cd content in dry matter (mg/kg)		Ammount of Cd added (g/ha)
				x	SD	
1996	28.45	1.16	24.39	4.87	0.16	33.81
1999	30.56	1.08	30.32	5.98	0.28	55.41
2002	23.58	1.40	30.00	2.88	0.03	20.35

content in the sewage sludge is roughly 10 × lower than that of phosphate fertilizer, but it is applied at a rate roughly 100 × higher than a P-fertilizer (Hutton 1996). P fertilizers may also influence Cd availability through their effects on soil pH, ionic strength of the soil solution and crop growth (Grant et al. 1999).

Nitrogen fertilizers can increase phytoavailability of Cd for plants although the fertilizer does not normally contain significant levels of Cd. The increasing crop uptake of Cd with N fertilizers application may be due to the increase in ion-exchange reactions or soil acidification. Effect of N fertilizers on Cd concentration in crops may differ with the N source. Fertilizers with high NH_4^+ content can decrease soil pH (exchange with H^+) and subsequently increase the Cd crop uptake (Grant et al. 1999).

The aim of the study was to investigate the balance of Cd input caused by different long-term fertilization managements (including application of sewage sludge, farmyard manure or mineral fertilizers) and removal depending on different soil conditions and crops planted, and to assess the risk of Cd accumulation in soil.

MATERIAL AND METHODS

An ongoing precise long-term field experiment was used for the study. The experiment was conducted since 1996 at four sites of the Czech Republic (Hněvčeves, Humpolec, Lukavec, Praha-Suchdol) with different soil and climate characteristics. Soil characteristics measured prior to the establishment of the experiment are described in Table 1. The content of Cd in the experimental soils did not exceed the limit value set up in the Czech Republic for sewage sludge use on agricultural soil (0.5 mg/kg) (MŽP 2001).

The experiment was designed in order to simulate common agricultural practices as much as possible. Potatoes (*Solanum tuberosum* L.), winter wheat (*Triticum aestivum* L.) and spring barley (*Hordeum vulgare* L.) were grown in a three-year rotation, which enabled investigation of direct as well as subsequent effects of different treatments. The experimental fields were divided into plots (60 m^2) with 9 or, at some localities, 12 treatments. Following treatments were chosen for this study: (1) zero treatment as control, (2) NPK mineral fertilization (the doses of N-P-K nutrients were

Table 3. Characteristics of the farmyard manure

Year of application	Site	Application rate of fresh matter (t/ha)	N content in fresh matter (%)	DM (%)	Cd content in dry matter (mg/kg)		Ammount of Cd added (g/ha)
					x	SD	
1996	Hněvčeves	53.23	0.62	18.53	0.19	0.01	1.82
	Humpolec	80.48	0.41	28.30	0.27	0.04	6.04
	Lukavec	68.75	0.48	36.96	0.41	0.03	10.34
	Suchdol	51.57	0.64	21.70	0.18	0.07	1.97
1999	Hněvčeves	62.27	0.53	23.13	0.26	0.00	3.69
	Humpolec	45.20	0.73	23.23	0.63	0.02	6.65
	Lukavec	66.00	0.50	23.79	0.72	0.07	11.31
	Suchdol	61.11	0.54	45.64	0.44	0.01	12.16
2002	Hněvčeves	55.00	0.60	21.89	0.15	0.04	1.81
	Humpolec	63.45	0.52	19.44	0.06	0.02	0.73
	Lukavec	66.00	0.50	17.97	0.29	0.00	3.44
	Suchdol	50.78	0.65	32.47	0.33	0.09	5.47

330–90–300 kg/ha for the whole three-year period), (3) sewage sludge (SS) – application rate corresponding to 330 kg N/ha (approximately 9 t dry matter of sludge/ha), (4) farmyard manure (FYM) (the rate corresponding to 330 kg N/ha).

The organic fertilizers (SS, FYM) were applied only before potatoes plantation, i.e. once in a three-year period. They were applied in the autumn before ploughing. Samples of SS as well as FYM were analyzed for the content of nitrogen and heavy metals. The application rate of organic fertilizers was always counted on their nitrogen content basis. Identical anaerobically treated SS originating from the same wastewater treatment plant was used at all experimental sites in one period. Organically fertilized treatments (SS, FYM) did not receive any additional mineral fertilization.

The characteristics of SS and FYM are presented in Tables 2 and 3, respectively.

The mineral fertilizers were applied in rates shown in Table 4 and were also analyzed for the content of heavy metals.

Potato tubers as well as wheat and barley were harvested in full maturity. The yields of the crops were recorded. Plant samples were dried, homogenized and mineralized by the dry ashing procedure (Mader et al. 1990). The ash was dissolved in 1.5% HNO_3 and the content of metals was detected by AAS flame technique on Varian SpectrAA-400 equipment. Reference materials RM 12-02-03 Lucerne and RM 12-03-12 Sludge were regularly used to evaluate the quality of analysis.

In our experiment Cd removal by plants was represented by the harvests of potato tubers and grain and straw of wheat and barley. Potato haulms were left in the field and ploughed in. Hence Cd contained in the haulms was returned into soil and was not implemented into the balance.

This paper summarizes the results from the years 1996–2005, i.e. three completed rotation cycles. The crops were planted as follows: potatoes 1997, 2000, 2003, wheat 1998, 2001, 2004 and barley 1999, 2002, 2005. SS and FYM were applied in the years 1996, 1999, 2002.

Table 4. Application rates and characteristics of the mineral fertilizers (3-year cycle)

	Applied amount of the nutrient (kg/ha)			Nutrient content in the fertilizer (%)	Cd content in the fertilizer (mg/kg)	Ammount of Cd added (g/ha)
	potatoes	wheat	barley			
N-fertlizer	120	140	70	27	0.02 ± 0.01	0.024
P-fertilizer	30	30	30	21	6.08 ± 2.31	2.606
K-fertilizer	100	100	100	50	< 0.005	–

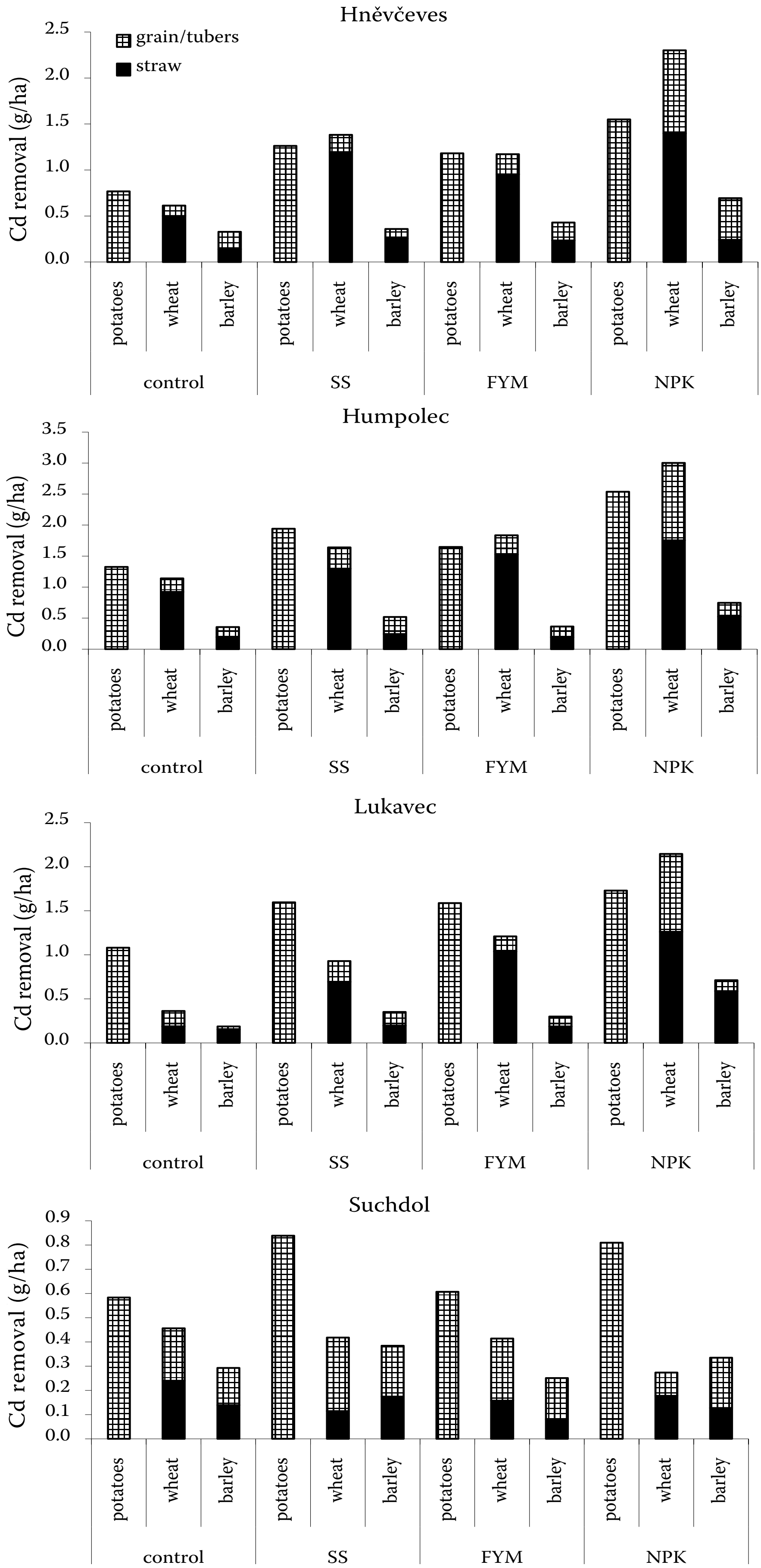

Figure 1. Average annual Cd removals by plants on different sites (g/ha)

RESULTS AND DISCUSSION

Cadmium content in the fertilizers

From the comparison of Cd content in both organic fertilizers used (Tables 2 and 3), it is obvious that the content of Cd in SS is by one order higher than the content in FYM. The standard deviation is higher in SS showing that SS is more heterogeneous material. A high difference of Cd content in SS as well as FYM among particular years was shown. In the Czech Republic, the limit value for Cd content in SS applied into soil is 5 mg Cd/kg of DM. In the experimental sludge, this limit was exceeded in the year 1999. Also during other years of the long-term experiment, which are out of scope of this paper, the limit value was exceeded quite often. This implies that meeting the limits regularly would be problematic for sludges originating from wastewater treatment plants of large cities.

Sewage sludge caused the highest Cd input into the soil among the treatments, ranging between 20.35 and 55.41 g/ha per three-year period (Table 2). Chang et al. (1982) reported much higher Cd inputs from four-year repeated applications of SS. Total amounts of sludge in their experiment ranged from 33–146 t/ha for the whole period and Cd input ranged from 1.4 to 6.2 kg/ha.

Cadmium content in FYM varied depending on the year of application and the site and was in the range 0.06–0.72 mg/kg DM, showing a great influence of the origin of FYM. The Cd input from FYM was in the range of 0.73–12.16 g/ha per three-year period (Table 3). Alloway (1995) reported values of 0.3–1.8 mg/kg DM for Cd content in the farmyard manure. Repeated dumping of large quantities of farm wastes into soil can therefore add significant amounts of Cd.

Among the mineral fertilizers, the highest concentration of Cd was found in P-fertilizer, as it was expected. Cadmium content in N-fertilizer was very low, and it was under the detection limit in K-fertilizer. Phosphate fertilizers are the most important source of Cd contamination of agricultural soils among mineral fertilizers. These fertilizers are manufactured from phosphorites (phosphate rocks), which can contain relatively high concentration of Cd. The enrichment is thought to be due to the substitution of Cd^{2+} for Ca^{2+} in mineral structure and the cadmium concentrations vary significantly depending on the origin of the phosphorite raw material (Alloway 1995). Cadmium content in phosphate fertilizers ranges between 0.15 and 130 mg Cd/kg (Johnston and Jones 1996). An illustration of the significance of different Cd contents of source rock is provided by the data from two long-term field experiments in the USA. P-fertilizers made from Florida phosphorite containing < 10 mg/kg contributed 0.3–1.2 g Cd/ha/year, whereas P-fertilizers manufactured from western phosphorite deposits (California) containing an average of 174 mg Cd/kg contributed 100 g Cd/ha/year (McLaughlin and Singh 1999). Cadmium input from mineral fertilizers in our experiment (2.63 g/ha per three-year period) was more than one order lower than the input caused by SS.

Cadmium removal by plants

The removal of elements depends on the yield of the harvested biomass and the content of the element in it. Total accumulation of elements in plant biomass depends on soil properties, the crop grown and also on the distribution in growing plant (Keefer et al. 1986, Kim et al. 1988, Tiller et al. 1989).

We investigated the total Cd removal (from the soil and the added materials) and the net removal (from the added Cd only), which was counted as a difference between the removal by plants grown on the treated plots and the removal by plants grown on the control plots. Comparing the total removals performed by particular plants (Figure 1), the highest total removal was performed by potatoes, followed by wheat and barley. The exception was the NPK treatment, where the highest total removal was achieved by wheat. Potatoes showed the highest accumulation among the crops planted, which proved the reported results that most dicotyledoneous plants absorb more heavy metals than the monocots do (Sauerbeck 1991).

The net removal of Cd differed in dependence on the treatment. On the FYM and NPK treated plots, higher net removal was achieved by wheat (except Suchdol), while it was not consistent on the SS treated plots. The lowest net removal from all treatments was achieved by barley.

We compared the removal achieved by grain and straw of the cereals. In the case of wheat much higher total removal was achieved by straw than by grain, caused by the fact that Cd content in the straw from all treatments was higher than in grain and the yield of straw was similar or slightly higher than the yield of grain. The only exception was found at Suchdol site, where on SS and FYM treated plots, higher Cd removal was achieved by grain due to its markedly higher yield. As was proved by several studies (Sauerbeck 1991, Moreno et al. 1996, Chaudhuri et al. 2003), vegetative parts of plants usually accumulate more Cd than generative ones, which indicates a physiological barrier protecting the generative organs. In our experiment, straw was removed from the field. However, in cases when straw is incorporated into the soil, much higher amount of Cd is returned to the soil increasing the risk of Cd accumulation in soil. In case of barley, Cd removals by straw and grain were similar. The yield of straw was usually lower than the yield of grain and the Cd content was higher in the straw. The removals of Cd by plant parts on different localities are shown in Figure 1.

Soil conditions affected the removal of Cd markedly and pH showed to be the key factor because of its influence on Cd uptake by plants. The highest total removal was found at Humpolec site – soil with the lowest pH, whilst the lowest removal was found at Suchdol site – soil with the highest pH value and the highest CEC, but on the other hand soil with the highest total content of Cd. Our experiment confirmed that the total content of the element did not have the major influence on plant uptake, and the soil characteristics have to be considered.

The priority of soil pH in affecting the uptake of Cd by plants was proved by several studies (Eriksson 1989). Tiller et al. (1984) reported that an increase of soil pH from 5.0 to 7.0 significantly influenced the content of available Cd in soil, which decreased from 75% to 15%. Hansen and Tjell (1983) reported that a fall of 0.5 units of pH leads to a 20–40% increase in the uptake of Cd.

The Cd removal did not correlate with Cd input. The highest removal was found on NPK treatment although the Cd input by mineral fertilizers was significantly lower than on plots treated with SS or FYM.

Long-term balance of Cd addition and removal

Table 5 shows the Cd amount, which was added by different treatments, and the amount, which was removed by harvested plants during the period of 9 years. Sludge and mineral fertilizers applications resulted in total addition of 109.58 g Cd/ha and 7.89 g Cd/ha, respectively. The total addition of Cd from FYM differed on the localities and ranged between 7.32 and 25.09 g Cd/ha. High differences in Cd removal by plants among the localities were found, caused by different uptake of Cd by plants as well as different yields. The removal on the soil with high pH was markedly lower indicating lower risk of contamination of crops by Cd but on the other hand higher risk for Cd accumulation in soil.

From the sludge-borne Cd, harvested plants removed only very small portion – approximately 3.5% on sites Hněvčeves, Humpolec and Lukavec (Luvisol and Cambisols) and only 0.84% on Suchdol site (Chernozem), which is the site with the highest pH value. The risk of Cd accumulation in soil is therefore high. Also McGrath (1987) and Adriano (2001) reported that metal removal by plants usually represents only small portion (less than 1%) of the total amount added by sewage sludge. However Tlustoš et al. (2000) investigated Cd removal in a pot experiment with spinach, oat and maize. Spinach achieved the maximum removal more than 10% at acid Cambisols and less than 2% on Luvisols and Chernozems, oat and maize removed less than 4% of applied Cd. In another study by Tlustoš et al. (1997) the results were similar to ours. They planted oat, maize, barley and poppy subsequently in a four-year pot experiment. The total Cd removal by the aboveground biomass during the experiment was 3.2% from the total content in soil. Balík et al. (1998) reported mean removal by plants of 12% and 23%, respectively, from the amount of Cd added by two different sewage sludges in their pot experiment with poppy, barley, maize and oat. In pot experiments, however, the elements accumulation in plants and their removal are higher than in field conditions. This is due to higher density of plants and plant roots per amount of soil and optimal soil moisture.

Chang et al. (1984) reported inputs of 8–30 kg Cd/ha after 6 years of application with cumulative doses 137–548 t/ha of composted sludge, and removal by plants (barley and sorghum) in the range of 15–55.7 g/ha on sandy loam and 9.5–41.1 g/ha on loamy soils; the removal at the control treatment was 5.6 and 4.0 g/ha, respectively. Mullins and Sommers (1986) reported an addition of 19 kg Cd/ha with a single application of 100 t/ha of anaerobically treated SS. Sewage sludge application in these two experiments resulted in much higher Cd inputs than in our experiment. The removal of Cd by plants on treated plots reported by Chang et al. (1984) was also higher compared to our results but represented even much lower portion of the inputs from sludge application. Since plant

Table 5. Balance of Cd addition by sludge application and removal by harvested plants

Period	Treat-ment	Cd added (g/ha)	Hněvčeves			Humpolec			Lukavec			Suchdol		
			total removal (g/ha)	net removal		total removal	net removal		total removal (g/ha)	net removal		total removal (g/ha)	net removal	
				(g/ha)	(%)	(g/ha)	(g/ha)	(%)		(g/ha)	(%)		(g/ha)	(%)
1997–1999	control	–	0.71	0.00	–	3.22	0.00	–	1.14	0.00	–	1.08	0.00	–
	sludge	33.81	1.72	1.01	2.98	5.05	1.83	5.41	2.36	1.22	3.62	1.56	0.48	1.42
	FYM	*	1.44	0.73	39.97	3.98	0.76	12.56	1.49	0.35	3.39	1.17	0.09	4.78
	NPK	2.63	2.79	2.08	79.19	6.90	3.68	140.02	2.60	1.47	55.73	1.79	0.72	27.19
2000–2002	control	–	2.04	0.00	–	3.38	0.00	–	1.53	0.00	–	1.38	0.00	–
	sludge	55.41	4.63	2.59	4.68	3.94	0.56	1.01	2.94	1.41	2.55	0.99	–0.38	–0.69
	FYM	*	4.44	2.40	65.01	4.35	0.97	14.59	4.24	2.71	23.95	0.94	–0.44	–3.64
	NPK	2.63	5.56	3.52	133.77	5.58	2.20	83.58	5.15	3.62	137.72	1.04	–0.34	–12.82
2003–2005	control	–	2.45	0.00	–	1.88	0.00	–	2.22	0.00	–	1.54	0.00	–
	sludge	20.35	2.77	0.33	1.60	3.32	1.45	7.10	3.32	1.10	5.42	2.37	0.83	4.06
	FYM	*	2.58	0.13	7.20	3.23	1.35	185.69	3.56	1.34	38.93	1.71	0.17	3.04
	NPK	2.63	5.40	2.95	112.16	6.39	4.51	171.67	6.00	3.78	143.75	1.42	–0.12	–4.59
Total 1997–2005	control	–	5.19	0.00	–	8.47	0.00	–	4.89	0.00	–	4.00	0.00	–
	sludge	109.58	9.12	3.93	3.58	12.31	3.84	3.50	8.63	3.74	3.41	4.92	0.92	0.84
	FYM	**	8.45	3.26	44.49	11.55	3.08	22.95	9.28	4.40	17.53	3.82	–0.18	–0.93
	NPK	7.89	13.74	8.55	108.37	18.87	10.39	131.76	13.75	8.87	112.40	4.25	0.26	3.26

*see Table 3, **Hněvčeves 7.32, Humpolec 13.41, Lukavec 25.09, Suchdol 19.60

absorption of metals was insignificant compared with the total metal inputs in their experiment, all applied Cd remained in the soil.

The results also showed that the amount of added Cd did not have a major effect on the Cd removal. Although the Cd addition by sludge in the second period of our experiment (2000–2002) was significantly higher than in the other two periods, increased removal was found only at Hněvčeves and Lukavec sites. Repeated application did not lead to a growing trend in Cd removal.

The addition of organic matter into soils led in majority of cases to similar or lower total Cd removal compared to control and NPK treatments. Application of organic matter influences many soil properties including CEC and pH values and leads to changes in the intensity of metal sorption (Grant et al. 1999). Karapanagiotis et al. (1991) reported that organic matter applied in the sludge could significantly increase adsorption capacity of soil for metals. Ram and Verloo (1985) reported reduced mobility of metals in soil due to increased complexation capacity of soil and the formation of stable organo-metallic complexes after the application of sludge organic matter. Elliot et al. (1986) agreed that increased soil organic matter content should restrict mobility and availability of Cd, at least under acidic conditions (pH 5.0) where soluble metal complex formation is limited. Application of sludge organic matter also leads to increase in microbial biomass. The capacity of microorganisms to immobilize heavy metals is well recognized (Gadd and White 1989). The content of microbial biomass in our long-term experiment was studied by Černý et al. (2008). They found higher contents of microbial biomass in treatments with organic fertilizers, being the highest on the plots treated with sewage sludge.

The highest removal of Cd was found on NPK-treated plots, although the Cd addition by NPK was more than ten times lower than by SS. The net removal by plants on mineral fertilizers-treated plots was very often higher than the amount added by the fertilizers. It is probably due to Cd release into soil solution after the application of ammonia and potassium in NPK fertilizers and local acidification, which both led to increased availability of Cd. The removal was low only at Suchdol site. Results showed that when phosphate fertilizers do not contain extremely high amounts of Cd, the removals of Cd by harvested plants are usually higher than the inputs.

High sorption capacity of Suchdol soil led to the lowest Cd accumulation in plant biomass and the lowest net removal, if any, from all tested treatments. Removal of Cd from the control plot at Suchdol site was in some cases higher than from the treated ones. Suchdol site showed also the smallest differences in Cd removals among the treatments. All these results imply that high sorption ability of Suchdol soil restricted the availability of Cd to plants.

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