Contemporary Problems of Management and Environmental Protection
Vol. 4, 2009. Sewages and Waste Materials in Environment: 95-105

CHAPTER VI

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THE EFFECT OF THE MUNICIPAL WASTE LANDFILL ON THE HEAVY METALS CONTENT IN SOIL

Introduction

The factors responsible for the degradation of the soil environment include an excessive cumulation of heavy metals which contain all metal elements with atomic mass higher than that of calcium and density higher than 5 $g \cdot cm^{-3}$ when exceeding toxic concentrations (KOC 1994, SANECKI 1995, KABATA-PENDIAS, PENDIAS 1999, ROSIK-DULEWSKA 2007). They occur as natural components in nature, but belong to particularly dangerous elements, which create a potential hazard to the biological environment and affect human health. The elements of a very high degree of risk include, but are not limited to, cadmium, lead, copper and zinc, and those of a medium degree of risk – arsenic (KOC 1994, KABATA-PENDIAS, PENDIAS 1999, GAMBUŚ, GORLACH 2001a). Harmfulness of heavy metals occurs sooner in animal organisms than in plants, because the safe level for a plant is often toxic in the case of its use as fodder or human food (KOC 1994, GAMBUŚ, GORLACH 2001a, ZGNILICKA 2002). In addition, in the environment they are often susceptible to bioaccumulation, and in living organisms they are easily absorbable from the alimentary canal, permeate the biological barrier which is the blood and brain, form connections with sulphohydryl groups of proteins and damage the nucleic acids chain.

One of the major and still topical issues is the estimation of the effect of various factors and processes on the soil quality. Unfavourable changes in the physical, chemical or biological soil properties may result not only in a decrease of its fertility, but they can even totally exclude it from production. Their cumulation in the soil surface layer is particularly noteworthy (BIERNACKA, MAŁUSZYŃSKI 2007; NIEDŹWIECKI et al. 2007). The presence of metals such as lead, cadmium, and mercury plays a great role, particularly in the case of their washing out to ground waters in an amount that would threaten the quality of potable water and create a hazard to human and animal health (TERELAK et al. 1995, GAMBUŚ, GORLACH 2001a, KARWACZYŃSKA et al. 2005).

In terms of the contents of some compounds and heavy metals, human activity has the highest effect on soil formation, natural conditions (mother-rock, climate, landform features) being of secondary importance (DOMSKA et al. 2005, OLEŚKÓW 2007).

The spreading of heavy metals such as copper, zinc, arsenic, cadmium and lead, among others, is caused particularly by the chemical industry (Cu, Zn, As), artificial fertilizers industry (Cu, Zn, Cd, Pb), cellulose and paper industry (Cu, Zn, Pb), petroleum refineries as well as metallurgy and ferrous metallurgy (Cu, Zn, Cd, Pb), glass-making, ceramic and cement industry (Cu, Pb) and power stations (elements occurring in fuels). A great influence on the cumulation of heavy metals in soils is also made by the location of industrial plants, motorization and herbicides, fertilizers and waste used for soil fertilization (SANECKI 1995, SIUTA 2000, GAMBUŚ, GORLACH 2001b, ZGNILICKA 2002, KUSZA, CIESIELCZUK 2007, ROSIK-DULEWSKA 2005, NIEWIADOMSKI, TOŁOCZKO 2005, KARCZEWSKA, KRÓL 2007, MEDYŃSKA, KABAŁA 2007). Soil contamination with heavy metals has also been reported in urban areas of a high degree of urbanization, located close to industrial plants and transport routes (LASKOWSKI, TOŁOCZKO 1995, KARCZEWSKA 2002, KUSZA, CIESIELCZUK 2007, OLEŚKÓW 2007, PLAK 2007).

Although soil contamination with heavy metals is mostly caused by industrial activity and coal or oil burning (SIUTA 2000, ZGNILICKA 2002, KUSZA, CIESIELCZUK 2007), solid, liquid or gaseous contaminants can also get into the soil from post-flotation and municipal waste landfill and mining activity (SANECKI 1995, GAMBUŚ, GORLACH 2001b, ROSIK - DULEWSKA 2007, NIEWIADOMSKI, TOŁOCZKO 2005, KARCZEWSKA, KRÓL 2007, MEDYŃSKA, KABAŁA 2007). However, some authors (OWCZARZAK, MOCEK 2004, DOMSKA, RACZKOWSKI 2008) do not indicate any unfavourable effect of brown coal opencast mines located on autogenic soils on the availability to plants or on the content of nutrients in the soil.

As the civilisation develops and large population centres increase, the process of continuous cumulation of waste in industrial and municipal landfills progresses. This waste can also be a hazard to the environment as a result of a release of numerous components from them, including heavy metals, through dusting, flow, washing out or ignition and smoking (KOC 1994, KABATA-PENDIAS, PENDIAS 1999, ROSIK-DULEWSKA et al. 2008). The qualitative composition of municipal waste consists of flammable and non-flammable waste. The former comprises organic waste, paper, fabrics, plastics, leather and rubber, while the latter – metals, glass and ceramic goods. Municipal landfills located outside urbanized areas and illegal rubbish dumps can be a source of contamination not only of adjoining farm land but also of water poisoning in addition to the fact that they occupy another area, often at the cost of agriculture or forestry (SZYMAŃSKA, PULIKOWSKA 2003, KARCZEWSKA, KRÓL 2007, ROSIK-DULEWSKA et al. 2008).

Waste is one of the major problems of environmental protection because

it creates hazard to all environmental spheres – lithosphere, hydrosphere, atmosphere and biosphere (KARWACZYŃSKA 2001, SZYMAŃSKA-PULIKOWSKA 2003, NIEDŹWIECKI et al. 2007, ROSIK-DULEWSKA 2007, ROSIK-DULEWSKA et al. 2008). A lot of waste management regulations have been introduced in Poland recently, but its state is still unsatisfactory. It results, among others, from the uncontrolled composition of dumping grounds, in which one can come across not only debris, household appliances or electronic equipment but also hazardous waste (remains of electrolytes, paint, lacquer, pigments, anti-corrosion agents, seed dressings, solvents, herbicides, batteries and overdue pharmacological agents, ash from individual heating systems as well as organic fractions showing considerable abilities to cumulate heavy metals (GAMBUŚ, GORLACH 2001A; KARCZEWSKA 2002, NIEDŹWIECKI et al. 2007).

The purpose of this research was to determine the effect of a municipal waste landfill on the cumulation of copper, cadmium, lead and arsenic in the soil of an adjoining area and to estimate risks connected with their contents and distribution.

Research conditions

The investigation was carried out in 2008 in the area around the municipal waste landfill of the town and commune of Węgorzewo near Czerwony Dwór. It occupies a territory of the total area of 3.6 hectares and has been exploited since 1996. The basic technical and exploitation parameters of the landfill amount to: upper area - 4950 m^2, bottom area - 1700 m^2, total volume – 20000 m^3, target dumping datum – 158,20 m above sea level. The quantity of the waste cumulated so far is estimated at 99 thousand m^3. The waste deposited at the landfill is not separated and mostly consists of household and building waste (ORYŃCZAK 2008). The area adjoining the landfill has been additionally secured by a 10-metre wide tree planting strip.

Soil was sampled from the surface soil layer on the southern side of the stockpile at a distance of 5, 10, 20 and 30 m. In mean soil samples (formed by mixing 10 individual samples), the granulometric composition was determined with the Bouyoucos aerometric method modified by Casagrande and Prószyński, pH – electrometrically in 1 $mol \cdot dm^{-3}$ KCl, the organic carbon content – according to Tiurin, the phosphorus and potassium content – with the Egner-Riehm method, magnesium – with the Schachtschabel method, and the contents of copper, zinc, cadmium, lead and arsenic – with the atomic absorption spectrometry technology after a sample mineralization using nitric acid and hydrochloric acid.

The data from particular sampling sites did not show any significant variations, therefore the findings are presented in tables as mean values.

The significance of variations has been calculated using the Tuckey's test, at the level of $p=0,05$.

Physical and chemical properties of the soils under study

It has been found out that the soil samples taken from the area adjoining the landfill had similar physical and chemical properties typical of soils of good agricultural usefulness. It was proved by their granulometric composition of light and medium loam, acidity of pH values from 7.0 do 7.2, and the humus content from 0.4 to 0.5% (Tab.1).

The soil conditions prevailing in the neighbourhood of the landfill did not show properties which would favour excess cumulation of heavy metals.

Decisive role not only in the contents of mineral components in the soil, but also their mobility and availability to plants is played by not only the soil acidity, but also the type and properties of the soil, including the granulometric cmposition and humus content (KABATA-PENDIAS, PENDIAS 1999, KARCZEWSKA 2002). Relatively high soil acidity (close to neutral) during the investigation, like in previous investigations by DOMSKA et al. (1996) and DOMSKA and WOJTKOWIAK (2000) was probably conditioned by its granulometric composition and applied agricultural technology. This acidity was not favourable for a high mobility of heavy metals, thus limiting the penetration of contaminants into the plants' root system. Setting in motion of the forms which are easily available to plants occurs with acid reaction; under such conditions, there is generally a larger content of heavy metals (GAMBUŚ, GORLACH 2001a). Soil graining typical of sands and sandy clay indicates a possibility of an occurrence of water permeability and easy migration of contaminants into the soil profile. In soils with a very small fraction of clay and a high content of organic carbon there are favourable conditions for potential accumulation of contaminants only in organic and mineral complexes (KUSZA, CIESIELCZUK 2007). Humus, in turn, shows high abilities of heavy metals absorption, which makes it more difficult to wash them out of the soil (MEDYŃSKA, KABAŁA 2007).

Table 1

Some physical and chemical soil properties

Place of sampling	Acidity (pH in 1n KCl)	Granulometric composition	Humus %
1	7.2	light loam	0.5
2	7.2	light loam	0.5
3	7.2	light loam	0.4
4	7.0	medium loam	0.4

Distance from landfill: 1 – 5m, 2 – 10 m, 3 – 20 m, 4 – 30 m.

The contents of available phosphorus (54.4-61.1 mg·kg^{-1}), potassium (124.5-132.8 mg·kg^{-1}) and magnesium (57.7-61.5 mg·kg^{-1}) around the landfill corresponded to the average soil abundance in relation to these nutrients (Tab.2).

Table 2

Available phosphorus, potassium and magnesium content (mg·kg^{-1} d.m.)

Place of sampling	Phosphorus	Potassium	Magnesium
1	54.5	124.5	57.7
2	56.7	128.6	60.1
3	56.7	124.5	57.9
4	61.1	132.8	61.5
LSD $_{p=0,05}$	2.5	4.6	3.9

∗see tab. 1

A slightly larger cumulation of the analysed forms (by 4.4-6.6, 4.2-8.3 and 1.4-3.8 mg·kg^{-1}, respectively) occurred in the margin of the area under study in comparison with the terrain located directly next to the landfill. It was probably connected with a variation of the granulometric composition and absorbing capacity of the soils studied (light and medium loam). In the available literature, phosphorus is pointed to as a factor which may contribute to changes in zinc or cadmium cumulation in soil, but only in the case of applying large doses of phosphorus fertilizers (TERELAK et al. 1995).

Copper and zinc

Copper and zinc belong to components of high biological importance, necessary for a proper functioning of an organism. We count them among heavy metals, affecting unfavourably the growth and yield of plants, when toxic concentrations are exceeded (KOC 1994, GAMBUŚ, GORLACH 2001A, KABATA-PENDIAS, PENDIAS 1999). In Polish soils, copper occurs in amounts from 0.2 to 293.3 mg·kg^{-1} forming low-mobile connections in the form of carbonate and sulphate and with organic matter and clay minerals. In the case of zinc, its content ranges from 0.5 to 1725.0 mg·kg^{-1} and is closely connected with the reaction, because it forms compounds of high solubility, which grows with acidification and decreasing soil absorption ability (TRELAK et al. 1995). According to research conducted by the Institute of Cultivation, Fertilization and Soil Science in Puławy together with Regional Chemical and Agricultural Stations, the permissible total content in the surface soil layer in relation to copper amounts to 25 to 74 mg·kg^{-1}, and that of zinc – from 80 to180 mg·kg^{-1} (GAMBUŚ, GORLACH 2001b). No unfavourable effect on particular ecosystems is revealed below these numbers.

The copper content in the studied area's soil near the municipal waste landfill was of little variation (showed no significant differences) and ranged within the values corresponding to good soil abundance for this element from 4.93 to 6.15 mg·kg^{-1} of dry matter (Tab.3).

Table 3

Copper and zinc content in landfill site area (mg·kg^{-1} s.m.)

Place of sampling*	Cu	Zn
1	5.02	45.71
2	6.15	44.25
3	4.93	43.28
4	5.61	38.24
LSD $_{p=0,05}$	1.30	2.56

* see tab. 1

More varied results, but not exceeding (just like in the case of copper) a good soil abundance, were obtained when analyzing the cumulation of zinc in the studied area. More zinc, i.e. from 43.28 to 45.71 mg·kg^{-1} of dry matter occurred in the vicinity of the landfill at the distance of 5, 10 and 20 m, while at a farther distance (30 m) there was only 38.24 mg·kg^{-1}. The variations ranged from 5.04 to 7.47 mg·kg^{-1} of dry matter, therefore, they were low and probably resulted from the difference in the granulometric composition of the soil and a higher content of phosphorus at a farther distance of the sampling place from the landfill. In comparison with the investigation of BIENIEK (2005) conducted in soils of a similar granulometric composition (light loam) in the vicinity of Olsztyn, the cumulation of copper and zinc near the municipal waste landfill in the vicinity of Czerwony Dwór was much lower. However, like in the quoted research, the values concerning the soil contents of the mentioned metals stayed within the limits of their natural contents in the soil and significantly below the quality standards of soils and land specified for agricultural land in the regulation of the Minister of Environment (2002).

According to NIEDŹWIECKI et al. (2007), uncontrolled waste landfills located in sandy areas may contaminate the soil's surface layers with heavy metals, particularly copper and zinc. However, the quoted authors, as well as SZYMAŃSKA and PULIKOWSKA (2003), maintain that municipal waste is characterized by a very varied content of heavy metals, and the intensity of environmental changes occurring under their influence is connected with the quality of the waste, frequency and time of storage and supply of the dump with illegal domestic sewage discharge, particularly on uncontrolled dumps.

In the research of OLEŚKÓW (2007) conducted in the allotments of Wrocław in the vicinity of industrial plants and transport routes, a higher contamination of soils, among others – with zinc and copper, was proved, most of the soil being contaminated with zinc (around 90%), while the contamination with copper was higher (corresponding to scale I-V). Investigations of other authors (GAMBUŚ, GORLACH 2001a) show that the excess of copper in the soil occurs mostly in areas contaminated by the copper industry and as a result of contamination with herbicides containing copper, and in the case of zinc – as a result of coal and waste burning and due to a storage of metal industry goods, or, like in the case of copper, is caused by herbicides.

Lead, cadmium and arsenic

The lead content in the Polish soils amounts to 0.1 to 992.5 $mg \cdot kg^{-1}$ of dry matter and is mostly dependent on the mineralogical and granulometric composition and the origin of mother rocks. Its availability is also dependent on the soil's reaction and, to a lower degree, on humus and the soil absorption ability (TERELAK et al. 1995). Besides, it is less mobile than zinc and cadmium, because it is included in slightly soluble minerals. Environmental conditions, factor analysis (mother rock, the character and causes of regional content differentiation and some soil properties) and the dependence between their actual content in soils and the expected range including numerical values after an exclusion of extreme observations resulting from significant analytical errors or accidental contamination are also noteworthy, particularly in the case of lead, cadmium and arsenic (DUDKA 1992).

The lead content in the surface soil layer in the area under study ranged from 10.54 to 15.57 $mg \cdot kg^{-1}$ of dry matter, therefore, not only did it exceed the natural values but it was also lower than the permissible limit of 40 mg (GAMBUŚ, GORLACH 2001b).

The most lead was in the immediate vicinity of the landfill, i.e. at the distance of 5 m (Tab. 4). As the distance from the landfill increased, the content of lead in the soil decreased, corresponding to the values of 14.23, 12.71 and 10.54 $mg \cdot kg^{-1}$ of dry matter. Although the main source of environmental contamination with lead is metallurgy and transport, it can also be released from the waste in the form of utensils, packages and production equipment (KOC 1994). In the investigation of KABAŁA (1995), variations in the properties of the analysed soils had a limited range, while significant correlations between the contents of lead, zinc and copper and the content of organic carbon and soil acidity occurred. In the findings of BIENIEK (2005), in soils of physico-chemical properties similar to those in the vicinity of Czerwony Dwór there was also a very low content of lead. Contrary to this investigation, OLEŚKÓW (2007) proved that soils in the vicinity of Wrocław exposed to the impact of industrial plants and transport routes were medium contaminated with lead in 70% in the scale from degree I to III. In the research of LASKOWSKI and TOŁOCZKO (1995), carried out near urban and industrial agglomerations, it was proved that the concentration of lead showed a larger dependence on the type of mother rock, granulometric composition or the content of organic substance than on the location of research sites in the field. However, the authors concluded that even a low content of heavy metals can be dangerous with a severe acidification of soils due to a large share of soluble forms in their total content.

Table 4

Cadmium, lead and aresenic content in landfill site area ($mg \cdot kg^{-1}$ s.m.)

Place of sampling*	Pb	Cd	As
1	15,57	0,14	1,00
2	14,23	0,12	1,35
3	12,71	0,15	0,65
4	10,54	0,13	0,75
LSD $_{p=0,05}$	1,51	0,03	0,30

*see tab. 1

In Polish soils the content of cadmium ranges from 0.01-24.75 $mg \cdot kg^{-1}$ of dry matter, 0.22 $mg \cdot kg^{-1}$ on average, while the permissible content in soil surface layers ranges from 1 to 3 $mg \cdot kg^{-1}$ of dry matter (TERELAK et al. 1995, GAMBUŚ, GORLACH 2001b). At the same time, this element shows a high mobility as a soil environment component which is easily taken in by plants (TERELAK et al. 1995). Its cumulation in soil, exceeding natural values, can be connected with the character of the basement complex, sewage sludge application, or overfertilization with phosphorus, etc. However, dust emissions from non-ferrous metallurgical plants and dust from scrap materials dumps, carried by wind, constitute the major source of soil contamination with cadmium. Environmental contamination with cadmium can also be caused by the impact of municipal landfills containing industrial and energetic waste, paint and lacquer residues (KOC 1994, GAMBUŚ, GORLACH 2001a).

The content of cadmium in the soil of the studied landfill site near Czerwony Dwór ranged within the limits of the natural (O°) content and much below the soil and land quality standards determined for agriculturally used land in the regulation of the Minister of Environment (2002). It was very similar in the whole area under study and ranged from 0.12 to 0.15 mg·kg^{-1} of dry matter (tab.4). Contrary to these findings, in the investigations by OLEŚKÓW (2007) carried out in allotments in Wrocław near large industrial plants and transport routes, soil contamination was proved not only with zinc, copper and lead, mentioned before, but also with cadmium in 87% of the studied soil, the contamination level being determined as severe (degree I-V).

In the investigation of ROSIK-DULEWSKA and KARWACZYŃSKA (2004), attention was focused on contamination with heavy metals in black earth limestone soil lying within the reach of the impact of the "Grundman" waste landfill in Opole. It was found out that after a 50-year period of the use of the landfill, the content of cadmium and lead was higher than that regarded as natural by IUNG (class I) and stayed within the class II standard. The highest contents of heavy metals occurred in the soil at the depth of up to 30 cm in the form of chemically stable and biologically inactive compounds – bounded with ferric oxide, manganese oxide and organic substance. This means that their availability to plants, soils and waters is considerably limited.

Arsenic belongs to elements which are very common in the environment (PLAK 2007). It is used in various branches of industry and in agriculture as a component of pesticides. Besides, it appears in small amounts in all food agents, and in larger amounts in sea products (KOC 1994). Anthropogenic sources include, apart from pesticides containing arsenic, also agents for wood conservation or production of paints and lacquers, but it is non-ferrous metallurgy, particularly copper metallurgy and liquid and solid fuel burning, which creates the greatest hazard (KABATA-PENDIAS, PENDIAS 1999, PLAK 2007, ROSIK-DULEWSKA 2007). In the soil, arsenic is absorbed by organic substances, ferric oxides, aluminium hydroxides and manganese compounds, and its content is highly variable and ranges from 0.1 to 95 mg·kg^{-1}. In soils originating from sedimentary rocks it remains at the level of 20-30 mg·kg^{-1}. Arsenic appears in larger amounts in clayey soils and soils rich in organic components, ferric, aluminium and phosphorus compounds, and in the region of the metallurgic and chemical industry, and in large urban agglomerations its concentration in soil can reach the values of as much as 2500 mg·kg^{-1} (KABATA-PENDIAS, PENDIAS 1999).

The content of arsenic in the soil near the landfill under study stayed within the permissible standards from 0.65 to 1.35 mg·kg^{-1} of dry matter (tab.4). However, there was much more arsenic (around 1.5 to 2 times) at the distance of 5 and 10 m in comparison with the remaining area under study (20 and 30 m from the landfill). The findings indicate an impact of some waste contained in the landfill, according to the observations of other authors (SANECKI 1995, GAMBUŚ, GORLACH 2001b, PLAK 2007), but it does not create such a hazard as that in the investigation of MEINHARDT

(1995) carried out in Wrocław province and in the city of Wrocław. The author assessed the degree of heavy metals contamination of soils with the granulometric composition of light loamy sand with the humus content from 2.1 to 4.9% and proved an increased content of zinc, lead, cadmium, nickel, mercury, arsenic, sulphur, fluorine and PAH in the immediate vicinity of industrial plants, transport routes and waste landfills (Czechnica thermal-electric power station and Siechnice steel mill). According to this research, a high soil contamination with zinc and lead resulted from, among others, a long-term impact of the steel mill on the environment. In the investigation of KUSZA and CIESIELCZUK (2007), in turn, the content of heavy metals, such as chromium, zinc, cadmium, copper, nickel, lead and mercury in the areas of industrial plants of the Opole region showed their low effect on the state of soils of the adjoining land. It was only in one case that a content of lead exceeding the permissible value (56.83 $mg \cdot kg^{-1}$) according to soil and land quality standards was found out. A low threat of external factors was also indicated by GAMBUŚ and GORLACH (2001b), who, on the basis of 3337 samples taken in the province of Warmia and Mazury proved that in the total area of arable land the share of soils with a natural content of heavy metals amounts to 91.5%, while soils with the contamination degree from II to V constitute only 0.5%.

Summary

Based on the obtained data, after a 12-year's period of the use of the municipal waste landfill near Czerwony Dwór, and in the light of standards and legal regulations in force (Ordinance of the Minister of Environment of 9 September 2002 concerning soil quality standards and land quality standards, Journal of Law No. 165, item 1359 of 4 October 2002) and an assessment of heavy metals content in the soil surface layer according to KABATA-PENDIAS (1999), it has been found out that there was no exceedance of standards concerning the permissible content of the studied elements (copper, zinc, lead, cadmium and arsenic) in the soil utilization group B, including agriculturally used soils. The content of copper and cadmium in the analyzed area was similar and ranged from 4.93 to 6.15 and from 0.12 to 0.15 $mg \cdot kg^{-1}$ of dry matter, respectively. A little more zinc (from 43.28 to 45.71 $mg \cdot kg^{-1}$ of dry matter) and lead (from 12.71 to 15.57 $mg \cdot kg^{-1}$ of dry matter) occurred in the soil at the distance up to 20 m from the landfill, and arsenic (from 1.00 to 1.35 $mg \cdot kg^{-1}$ of dry matter) – closer to the landfill (at the distance up to 10 m). The findings do not indicate any threat to the environment due to a cumulation of heavy metals. In addition, they do not suggest any necessity of introducing restrictions in farm production near a waste landfill.

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