AES BIOFLUX

Advances in Environmental Sciences - International Journal of the Bioflux Society

Heavy metals inside and in the vicinity of contaminated sites - case study of industrial and municipal waste landfills Bistriţa

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Abstract. Landfills, domestic and industrial, are an example of contaminated sites that have negative effects on the environment and human health both during operation and closure. This paper analyzes the industrial and municipal waste landfills in Bistriţa, which were closed (the first in 2006, the second phased in 2006-2010) and in recent years have been surrounded by houses with individual gardens and orchards of fruit trees. The objective of the study was to determine and compare the concentrations of heavy metals in soils and vegetation, both in the storage area and in the adjacent area of the landfills for the period 2007-2011. Monitoring data shows that while heavy metals from contaminated sites migrate to surrounding areas and are found in the vegetation covering the surrounding area. In many cases it was found that concentrations of heavy metals in soil and vegetation surrounding waste landfills are even greater than those inside warehouses. The study also highlights that the transfer from soil to plants depends on both the type of vegetation (analyzes were made on leaves of apple trees, grass and burdock leaves) and on the season (transfer is strongest during the spring months).
Key Words: heavy metals, soil, plants, contaminated sites.

Introduction. Heavy metals are an important part of environmental pollutants with major negative effects on human health. They come mainly from human activities and have received more attention in recent years because of their bioaccumulative and serious medical implications and long term they produce (Islam et al 2007; Duruibe et al 2007). Studies have shown that an important input of heavy metals in living organisms occurs by ingestion of foods prepared from products grown or cultivated in areas with polluted soils (Islam et al 2007; Lăcătuşu et al 1996). Also accumulation of heavy metals in the food chain can be extremely dangerous to human health. A special case is the cultivation of soil in urban and periurban areas, with higher levels of pollutants compared to soils in rural agricultural areas (Öborn & Linde 2001; Bretzeli & Calderisi 2005). The situation is even more problematic when crops are grown surrounding polluted soils areas.

Landfills are among the main pollution sources of soil (Ashworth & Alloway 2004; Zhang et al 2008; Howari 2004). Storage in landfills without waterproofing barrier systems or without previous neutralization of waste may results in heavy pollution of the surrounding environment. In addition to the direct action of waste, infiltration of rains water and physical-chemical processes in the waste, lead to the formation of leachate that seeps into the soil more quickly carrying dissolved elements (Zhang et al 2008; Ashworth 2003; Öborn & Linde 2001).

Material and Method

Site description. Two landfilling sites: one used for the storage of municipal wastes and one for the storage of industrial wastes located in the unincorporated area of the Bistriţa town, in full development of its urban area was studied. Both sites are presented in

Figure 1. The satellite view reveals households and gardens in the vicinity of this landfilling site, especially near the industrial landfill.

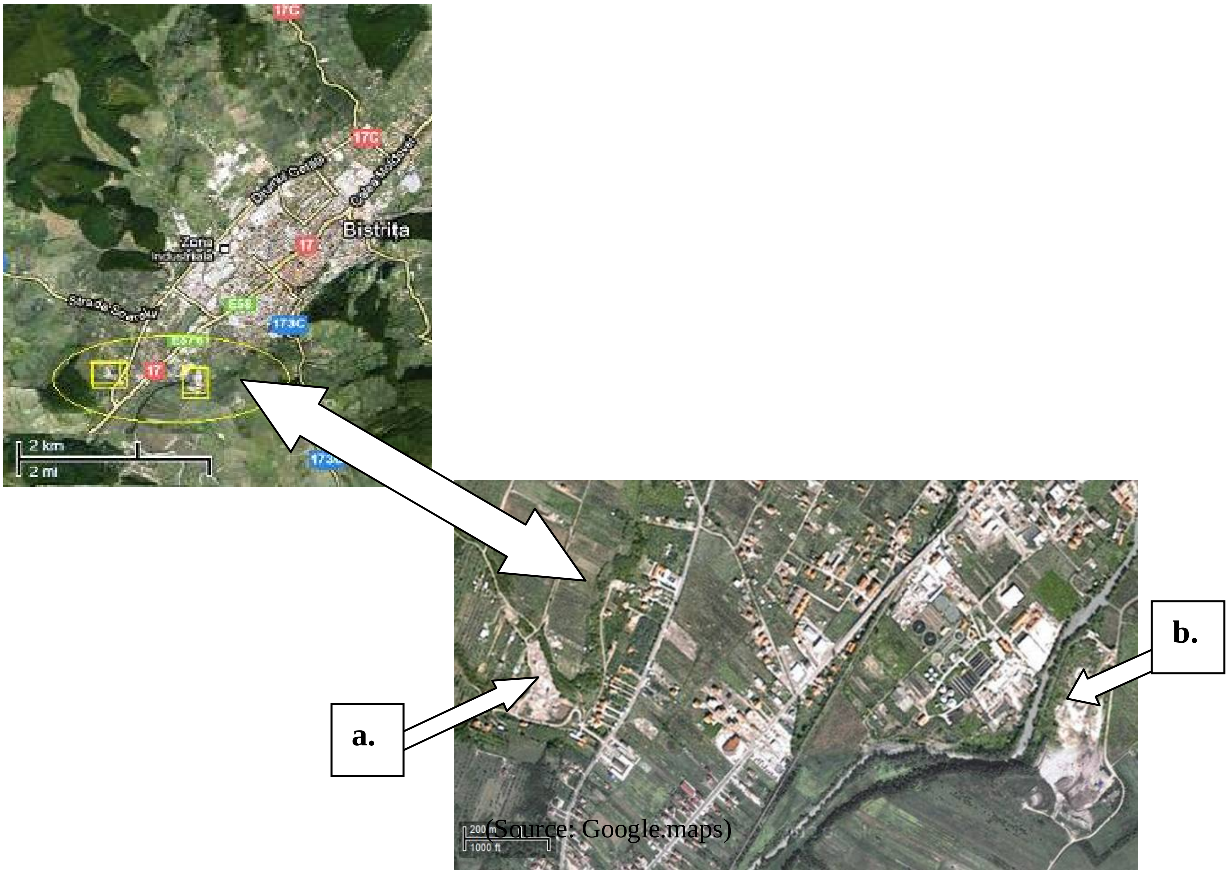


Figure 1. Location of the two landfilling sites: a). industrial landfill, b). municipal waste landfill.

The industrial landfill is located in the SE of Bistriţa town and it covers an area of 2.4 ha and it was used from 1981 until 2006 for storage of industrial wastes from the iron and steel foundry and the glass factory.

The municipal waste landfill is located in the southwest of Bistriţa town, occupying an area of approx. 7 ha. Opened about 50 years ago, came to the attention of store only just in 1992. It was managed by several companies and was closed in three stages between 2006-2010. Since 2011 the landfill greening started.

Sample collection and analysis. To assess the level of heavy metal pollution of the municipal and industrial waste landfills a long-term monitoring program of soil and vegetation within the landfill and in its vicinity was carried out during 2006-2012.

For this study 92 soil samples and 46 vegetation samples from the municipal landfill site and 90 soil samples and 45 vegetation samples from industrial landfill were collected. Soil analysis was performed on surface (0-5 cm) and deep (20-30 cm) samples. The determined metals were zinc, copper, lead, cadmium and chromium. All heavy metal analyses were performed by atomic absorption method using atomic absorption spectrometer GBC AVANTA PM, type 932G Plus, dual beam, oxy-acetylene burner.

Results and Discussion. The analysis of these data found that between metal concentrations in surface soil - soil depth – vegetables there is a growing interdependence, as shown in Figure 2, where is the evolution of chromium concentrations in samples collected in industrial landfill is presented. It is noted that high levels of metal in the soil found in vegetation.

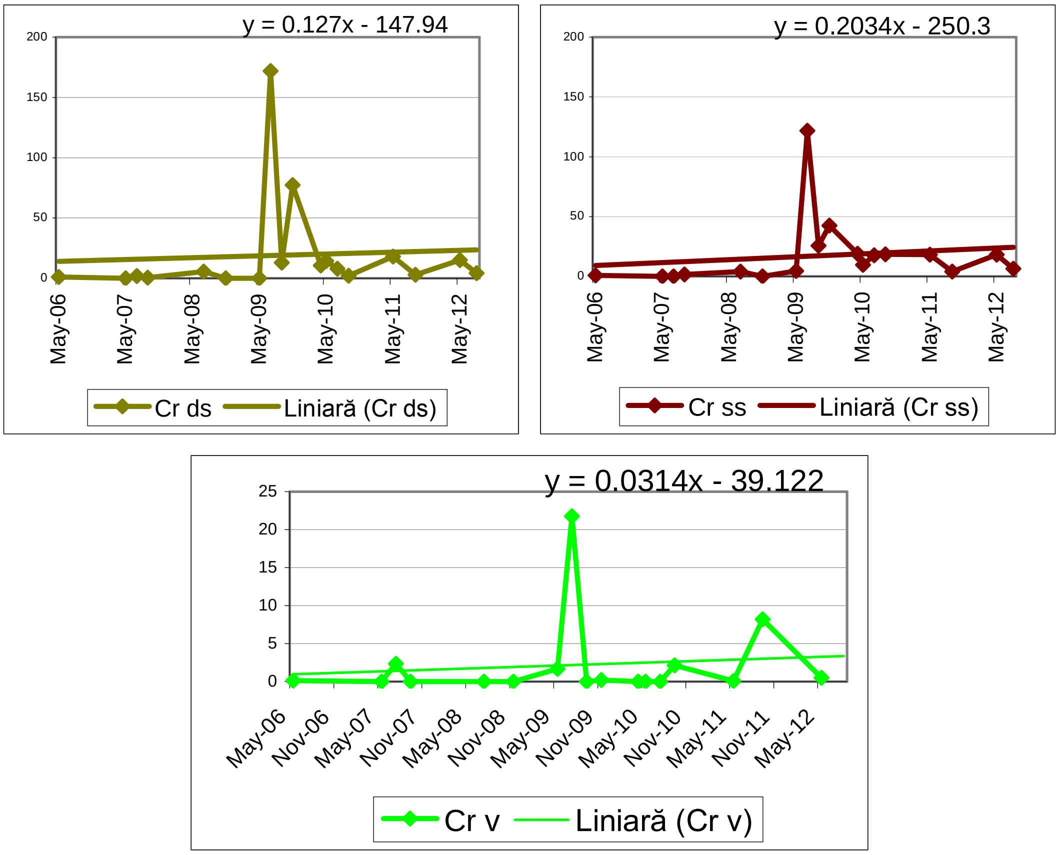


Figure 2. Evolution and trend line of chromium in soil and vegetation of industrial landfill (mg Cr kg^{-1} dry sample) (Note: ds=depth soil sample, ss=surface soil sample, v=vegetables sample).

An increasing trend of chromium concentrations was found although storage in this area has ceased. The same trend was observed for zinc, copper and lead in industrial landfill for both soil and vegetation.

The metal contents in the two studies landfills were compared using for comparison the maximum and average concentrations of the monitored metals. After analyzing the data it was found that while we have an industrial landfill, in which uncontrolled wastes from industrial processes were stored, concentration levels of zinc, cadmium and chromium were generally higher in landfill waste than in industrial landfill, as shown in Table 1. For examples: average concentrations of zinc outside waste landfill are 64 mg kg^{-1} dry soil and outside industrial landfill are 55 mg kg^{-1}.

Basically, the concentrations are higher in industrial waste for specific items of relatively intense activities in the industrial area of the city, such as metal plating (galvanization) or crystal industry, where they used rich heavy metals color pigments.

As specified in some of specialized papers (Ashworth & Alloway 2004; Howari 2004), metals migrate slowly, using leachate as a carrier, and they can advance deposits in neighboring areas. This table also shows that although, in general, both averages and maximums are higher in landfill than in the surrounding area, there are exceptions, such as cadmium and chromium that are higher in soils and vegetation in the periphery.

Table 1

Average and maximum values recorded inside and limit (outside) municipal and industrial waste landfill, Bistriţa, 2006-2012 period, in mg kg^{-1} dry sample

elements	*Average*				*Maximum*			
	outside landfill area		*inside landfill area*		*outside landfill area*		*inside landfill area*	
	waste landfill	*industrial landfill*	*waste landfill*	*industrial landfill*	*waste landfill*	*industrial landfill*	*waste landfill*	*industrial landfill*
pH s	7.396	7.639	7.221	8.053	8.390	8.360	8.150	8.900
Zn s	64.055	55.249	55.078	65.789	251.250	247.000	196.000	226.510
Cu s	24.290	16.928	41.150	24.855	133.725	50.125	226.500	106.425
Pb s	50.737	31.475	72.187	37.750	444.500	308.000	1140.750	158.680
Cd s	0.363	0.194	0.196	0.614	4.150	2.500	2.750	5.100
Cr s	13.415	7.416	8.148	29.802	120.050	44.025	92.775	134.000
pH d	7.361	7.737	7.212	8.163	8.310	8.410	8.520	8.970
Zn d	89.183	72.029	93.950	74.611	409.739	412.250	541.560	539.500
Cu d	40.180	14.375	43.292	25.247	202.800	37.125	264.250	91.250
Pb d	44.458	17.291	63.105	39.732	290.000	85.500	797.500	291.750
Cd d	0.444	0.341	0.109	0.544	6.800	3.750	1.150	6.150
Cr d	16.875	6.842	16.275	33.390	102.825	47.650	191.450	261.250
Zn v	22.707	15.144	77.733	18.234	88.029	51.925	528.195	65.885
Cu v	6.895	5.811	14.218	9.471	22.475	27.925	67.075	79.500
Pb v	3.013	6.299	3.029	4.770	31.1	46.630	32.200	72.525
Cd v	0.262	0.227	0.717	0.206	1.850	1.625	7.400	1.300
Cr v	0.531	1.220	0.456	2.187	2.875	16.750	2.875	21.775

Note: s = surface sampling, d = depth sampling, v = vegetation sampling.

Interesting is the allure of trend line for some of the studied elements. In Table 2 are presented the trend line equations for heavy metals in soil and vegetation, inside and outside of municipal waste landfill. It can be seen from these equations an increasing trend of concentrations for zinc and cadmium in the soil outside the.

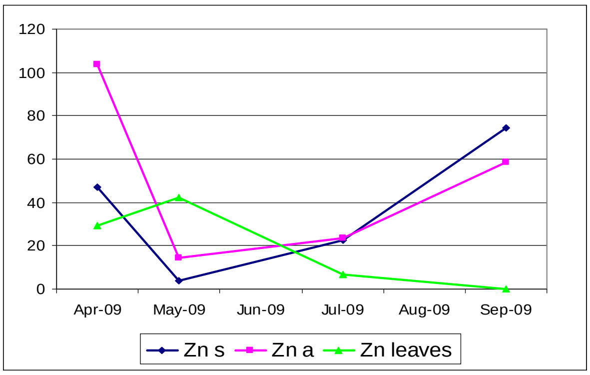
Table 2

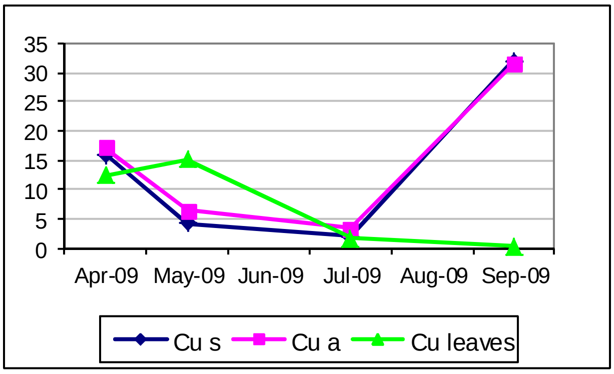
Trends lines of heavy metals concentration inside and outside the municipal waste landfill of Bistriţa

Element		*Inside*	*Outside*
Zn	s	$y = 0.8965x - 1124$	$y = 0.8965x - 1124$
	d	$y = 2.1193x - 2693.4$	$y = 2.1193x - 2693.4$
	v	$y = 2.2853x - 2932.1$	$y = 2.2853x - 2932.1$
Cu	s	$y = 0.0892x - 76.107$	$y = 0.0892x - 76.107$
	d	$y = 0.145x - 147.38$	$y = 0.145x - 147.38$
	v	$y = 0.227x - 284.79$	$y = 0.227x - 284.79$
Pb	s	$y = -1.658x + 2252.8$	$y = -1.658x + 2252.8$
	d	$y = -1.0212x + 1406.1$	$y = -1.0212x + 1406.1$
	v	$y = 0.0345x - 42.4$	$y = 0.0345x - 42.4$
Cd	s	$y = -0.0102x + 13.551$	$y = -0.0102x + 13.551$
	d	$y = 0.002x - 2.5466$	$y = 0.002x - 2.5466$
	v	$y = 0.0037x - 4.1465$	$y = 0.0037x - 4.1465$
Cr	s	$y = -0.1386x + 190.49$	$y = -0.1386x + 190.49$
	d	$y = -0.417x + 564.76$	$y = -0.417x + 564.76$
	v	$y = 0.0036x - 4.3421$	$y = 0.0036x - 4.3421$

Note: s = surface soil, d = deep soil, v = vegetation.

It was also observed that the transfer of heavy metals in vegetation is dependent not only on that metal concentrations in soil but also the season in which is a sample was collected. In Figure 3 is presented the evolution of zinc concentration in leaves of apple trees, during several consecutive months.



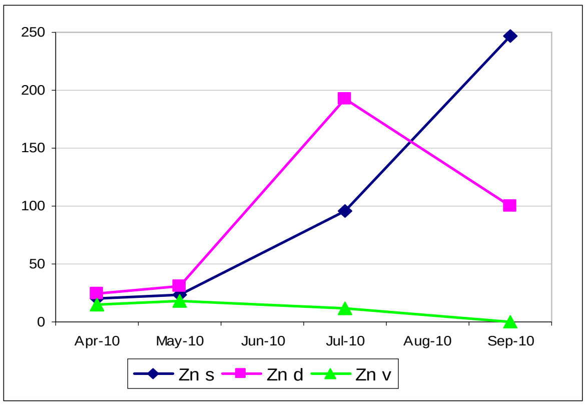


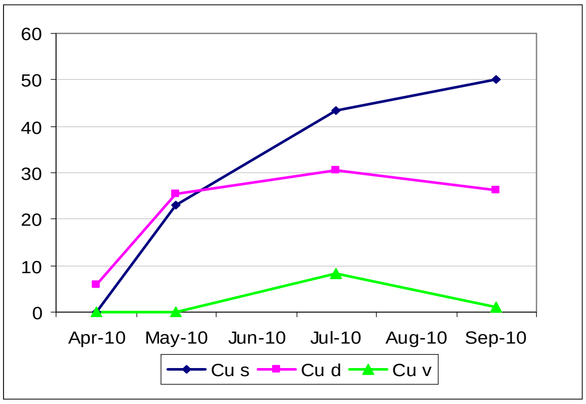
(a) **(b)**

Figure 3. Evolution of zinc (a) and copper (b) concentrations in the leaves of apple trees (concentration in mg metal kg^{-1} dry sample).

It was found that in the spring, especially in May, the transfer of some metals is mach stronger in the leaves of apple trees.

For the same elements in grass and burdock, allure of graph is rathes similar to that of soil development without increases in the spring (Figure 4).





(a) **(b)**

Figure 4. Evolution of zinc (a) and copper (b) concentration in grass and burdock leaves (concentration in mg metal kg^{-1} dry sample).

Conclusions. The study shows that contaminated sites are a serious source of environmental pollution. Although heavy metals migration is relatively slow, as specified in some specialty papers, they can advance to vicinal areas, especially using leachate as a carrier. Thus they are found in consistent in the surroundings of the contaminated sites. This study revealed that cadmium and chromium averages and maximum concentrations are higher outside than inside their storage area, which show a trend of metals to migrate. These higher values are found not only in soil but also in vegetable which shows the trend of accumulation of these heavy metals in vegetation.

It should be noted that both in the vicinity of industrial landfill and the municipal waste landfill in recent years developed a strong urban area consisting of many individual households with gardens and orchards. Under these conditions, the growing trend of evolution of heavy metal concentrations in vegetation, especially metals such as lead, chromium, cadmium, recognized as having high toxicity to living organisms, require a least monthly monitoring frequency, long-term, of these elements from soil and vegetation. Even they have been closed these deposits must be followed and if monitoring results will show an increase over the concentration limits will be taken whatever action will be imposed to avoid endangering health of local people and environment.

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Received: 20 February 2013. Accepted: 25 February 2013. Published online: 13 April 2013.

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How to cite this article:
Cordoş A. A., Ristoiu D., 2013 Heavy metals inside and in the vicinity of contaminated sites - case study of industrial and municipal waste landfills Bistriţa. AES Bioflux 5(2):135-140.