
Soil contamination by heavy metal mobile forms near landfills

Vitalii Ishchenko

Department of Ecology and Environmental Safety,
Vinnytsia National Technical University,
21021, Khmelnytske shose 95,
Vinnytsia, Ukraine
Email: ischenko.v.a@vntu.edu.ua

Abstract: The dissemination of heavy metals (Cr, Pb, Cd and Ni) near the landfills was analysed by measuring the concentrations of their mobile forms in soil on different distances from the place of leachate outflow at Stadnytsia landfill (Vinnytsia region, Ukraine). Exceeding the limit was not observed for any heavy metal. The highest levels are about 0.5 of limit values for Cr and Pb. A strong correlation between the distance from the landfill and the concentration is observed only for Cd. The dependence is ambiguous for Pb and Cr, perhaps due to the spatial proximity of sampling points and the existence of predominant migration routes of mobile forms of these metals. The landfill influence on the content of Ni mobile forms in nearby soils is most likely minimal. The research also confirms that migration of heavy metals depends on the conditions at landfill and on its compliance with requirements.

Keywords: heavy metals; contamination; landfill; soil; heavy metal mobile forms; Ukraine.

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Biographical notes: Vitalii Ishchenko holds a PhD in Devices and Methods of Control and Definition of Substances from Vinnytsia National Technical University, Ukraine. He has over ten years of experience in the field of environment protection. Currently, his research addresses waste management, especially on waste impact on the environment. He has co-authored over 50 scientific papers in peer-reviewed journals and conference proceedings. He has also worked on many funded projects related to waste management.

1 Introduction

Landfilling is still the main method of municipal solid waste (MSW) handling in developing countries. For example, 95% of MSW in Ukraine accumulates at landfills. Besides, these countries mostly do not have waste separation system. That is why a lot of hazardous substances are sent to landfills as well. Taking into account the poor conditions at landfills, there is a real threat of such substances migration into the environment. Usually, some part of the mobile forms of hazardous substances migrates from the landfill to the nearby soils.

Vergara and Tchobanoglous (2012) consider heavy metals as main soil polluters near landfills. In many countries it is obligatory to control some parameters, including heavy metals, in soil near landfills. For example in Ukraine according to (Ministry of Municipal and Communal Service of Ukraine, 2008, 2010) there are control distances of 50, 100, 200 and 500 m from a landfill. However, such measurements in Ukraine are carried out only partly at landfills of major cities. Therefore, a research of hazardous substances concentrations in soil near landfills is the important task. Such heavy metals as Pb, Cd, Ni and Cr have to be controlled according to Ukrainian legislation, so they are analysed in this paper.

Soil contamination by heavy metals near landfills was studied by many authors, e.g., (Kasassi et al., 2008; Nikolaev, 2014; Vojtzihovska and Butin, 2008). Authors (Abidemi and Theresa, 2015; Kanmani and Gandhimathi, 2013) have defined heavy metals concentration in soils at different directions from the landfill. There are few studies, where authors analyse the changes of contamination level depending on the distance from a landfill. For example, Xie et al. (2009) have investigated the spatial distribution of heavy metals in soils within the landfill body. Bahaa-Eldin et al. (2008) have compared heavy metals concentration within and outside the landfill. Authors (Liu et al., 2013; Tumuklu et al., 2007] have studied the changes of heavy metals concentrations at different distances from the landfill and have got different results. All above mentioned researches had deal with total concentration of heavy metals. For studying the heavy metals dissemination it is more informatively to know the content of mobile forms of heavy metals. That is because they determine the contamination level outside landfill body, near landfilling area. Authors (European Commission, 2000) have also concluded that long-term impact of landfills on the environment (including the potential heavy metals release from landfill body) is not completely known. At the same time in many countries, where landfills are very common, contamination becomes more considerable with increasing the duration and the strength of landfill impact on the environment. It is noted, for example, by Abidemi and Theresa (2015). Such a situation is at Stadnytsia landfill (regional centre Vinnytsia, Ukraine). The case study of this landfill will be considered in the work as well as the soil contamination by heavy metals at different distances from the place of landfill leachate outflow, which can be the greatest source of heavy metals in adjacent soil (see details in study area section). This is essential due to favourable conditions for heavy metals migration outside the landfill as consequence of its non-compliance with requirements. Besides, the proximity of farmlands to investigated landfill and possible crop contamination is another reason for such a research. Therefore, study of heavy metals at different distances from the landfill could help to understand the possible landfill impact on adjacent soils.

2 Materials and methods

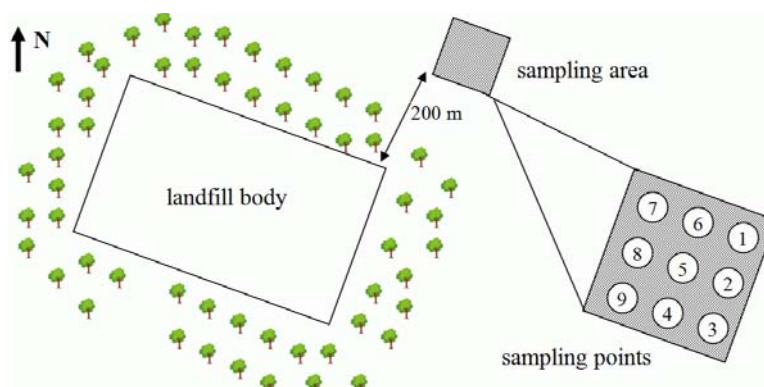
2.1 Study area

Stadnytsia landfill is the greatest waste disposal site in Vinnytsia region with about 1.5 million m³ household waste delivered every year. As most landfills in Ukraine, this landfill does not meet many requirements. Unless the landfill is situated in the area with more or less waterproof rocks and surrounded by ground barrier of several metres height, it is unlined and has no any leachate collection system.

The study area is adjacent to northeast part of Stadnytsia landfill chosen for investigation. This area has very well conditions for substances migration in soils. The northeast part of the landfill has natural leachate outflow. This is due to terrain features. Here some kind of leachate removal facility is situated, which provides leachate flow to nearby (less than 50 metres) leachate ponds (without any treatment). Thus, this place is potential source of adjacent soil contamination by heavy metals. Although there are no obvious leachate flows from landfill to sampling area, but there are other factors, which could conduce to leachate flowing into sampling area (e.g., ground or underground water). To study the local level of migration of hazardous substances from the landfill in a specific direction, samples were taken at a short distance from each other. Soil sampling was carried out at one time at a distance of 200 metres north-east from the landfill, at nine points (depth 10 cm), which are located at a distance of 20 m from each other (Figure 1). Each sample was obtained by mixing five samples from one location (sampled in four corners and centre of 30 cm * 30 cm square). Both sampling area and landfill are situated at the same terrain (according to GPS measurement) and therefore topographic situation in both cases is the same.

It should be noted that very close to the sampling area (no more than 20 m) are farmlands.

Figure 1 Sampling points near Stadnytsia landfill (see online version for colours)



2.2 Instrumental analysis

Soil preparation was performed on the same day after sampling: it was ground and sieved through a 1 mm mesh sieve. For heavy metals extraction, ammonium-acetate buffer (pH 4.8) was used in appropriate proportion (5 g of soil sample and 50 ml of extractant). After that, the mixtures were closed in flasks and shaken off during one hour and then filtered. The concentration of mobile forms of heavy metals (below we consider the concentration of heavy metals as concentration of their mobile forms) in the filtered solutions was determined using atomic-absorption spectrophotometer with flame ionisation detector Selmi C-115. Heavy metals concentration in each soil sample were measured three times. Then mean values were calculated.

To make the research more complete the results of measurement of Pb, Cd and Ni concentrations in soils near other landfills in Vinnytsia region were provided by the State Environmental Inspection in Vinnytsia region.

3 Results and discussions

The heavy metals concentrations (Pb, Cd, Cr and Ni) measured near Stadnytsia landfill are shown at Figures 2–5. It is known that there is a high possibility for insoluble heavy metal compounds to react with other substances in or out of landfills and to form soluble compounds. Thus, such processes increase the concentration of soluble forms of heavy metals and raise environmental risks.

Figure 2 Concentration of Cd mobile forms in soil

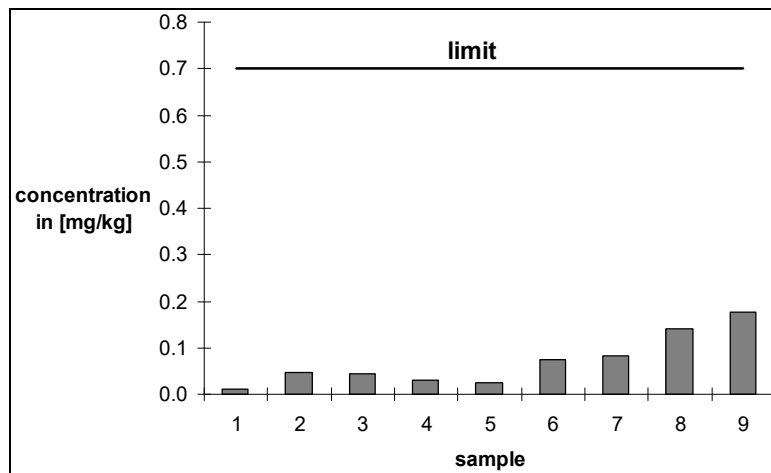


Figure 3 Concentration of Cr mobile forms in soil

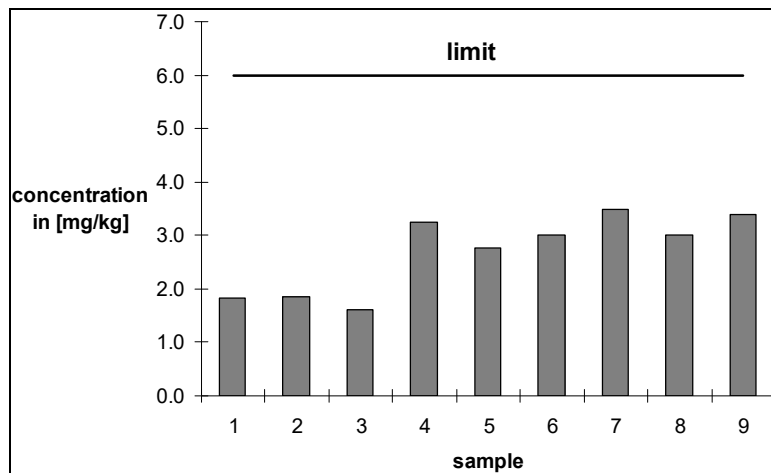
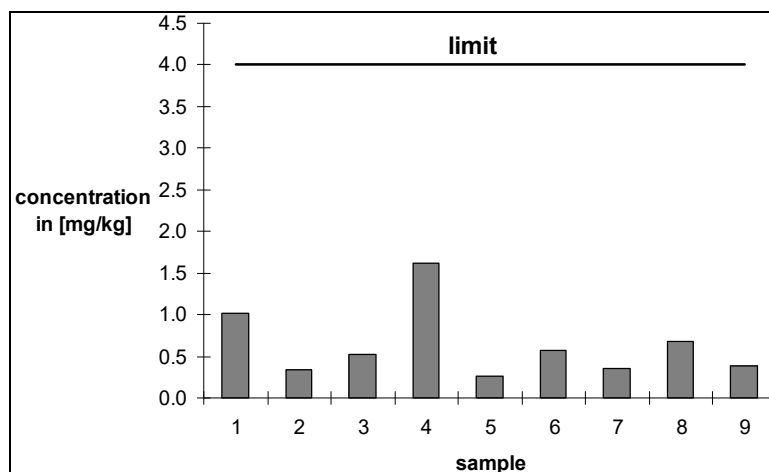
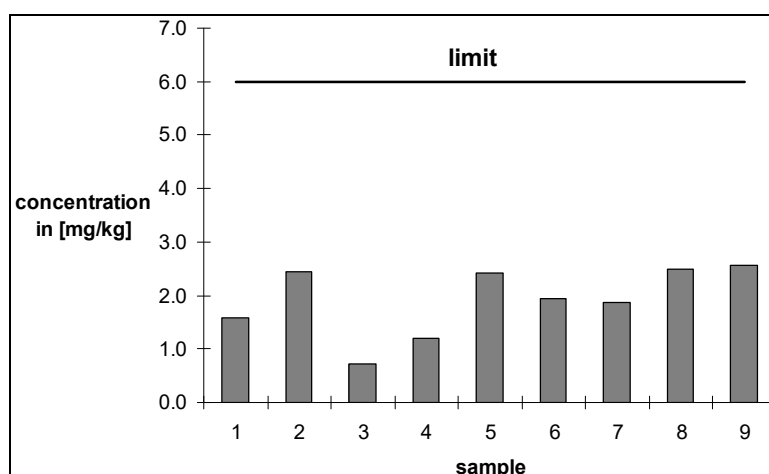


Figure 4 Concentration of Ni mobile forms in soil**Figure 5** Concentration of Pb mobile forms in soil

The results show that the excess of limit concentrations is not observed for any heavy metal. The concentrations of Cr and Pb in majority of samples (except samples 1, 2 and 3 for Cr and samples 3 and 4 for Pb) are near twice lower than permissible value, but are high enough to concern about. At the same time, Cd and Ni concentrations in all samples are significantly below their limits. Only samples 8 and 9 in the case of Cd and samples 1 and 4 in the case of Ni have relatively high content of these metals.

Analysis of metals concentrations distribution depending on the distance from the landfill leachate outflow results in next conclusions. Cd content gradually decreases with increasing a distance to the landfill (except sample 4). Also, Cd concentration decreases towards the northeast, that corresponds to the location of sampling points (Northeast from the landfill) and previous researches (Liu et al., 2013).

One cannot conclude something valuable from spatial distribution of Cr concentrations. On the one hand, the concentrations of Cr in the samples 1, 6 and 7

(far row from the landfill, see Figure 1) are very similar but still slightly higher if compare to some neighbour points (samples 3, 4 and 9). A similar result was obtained by Liu et al. (2013), which have defined Cr concentration in the soil (at the same depth of 20 cm) at distances of 3–6 km from a landfill higher in comparison to samples taken near the landfill (unsorted household waste is also delivered, the climate is little wetter in comparison to landfill under research). There was no any Cr concentration dependence on the distance from landfill in other similar research (Tumuklu et al., 2007) as well despite of the fact that sampling was performed downhill the landfill, which could conduce to chromium dissemination from the landfill. Besides, in that study samples were also taken from area situated in the direction of leachate outflow from landfill. On the other hand, as well as in the case of Cd, there were lower Cr concentrations in eastern part of the sampling area (samples 1, 2 and 3). Therefore, we can assume relatively active Cr migration in the soil, and places with higher concentrations probably have better conditions for migration. This assumption is partly confirmed by Flyhammar (1997), who has concluded that significant portion of Cr at landfills exists in oxidised form. For the insoluble $\text{Cr}(\text{OH})_3$ formation it is required a neutral or slightly alkaline environment. It is not usual for Ukrainian landfills, where anaerobic phase and acidic environment last a long time due to high organic content. Another explanation for the uncertain distribution of Cr in the soil may be the low effect of the landfill on its concentration in our case, so other sources of Cr may have a stronger influence.

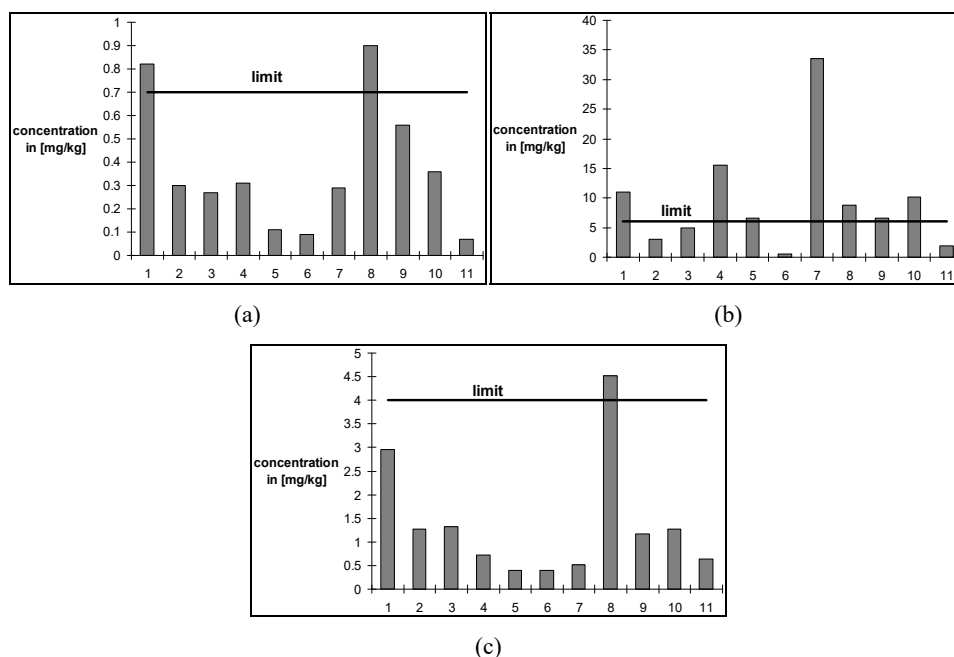
Pb concentrations are higher in western part of the sampling area in comparison to its Eastern part. Thus, the dynamics of Pb and Cr concentrations are similar. In the case of Pb, dynamic is less clear and there are several points which do not correspond to general trend. This may indicate little impact of landfill on Pb concentration in soil at the study area. For example, Yanful et al. (1988) have investigated that 80% of Pb at landfills are found in the form of insoluble carbonates and do not migrate intensively from landfills to adjacent soils.

The spatial distribution of Ni concentrations at investigated area has no any evident trend. There is research (Tumuklu et al., 2007) where concentration of Ni in soil was investigated to be decreased sharply during the first 100 metres from a landfill (also in the direction of leachate outflow from landfill as in present research), and then its concentration varied little. We can therefore assume that in our case, at a distance of 200 metres from the landfill leachate outflow, its impact on Ni concentration in soil is minimal.

For other Ukrainian landfill, Vojtzihovska and Butin (2008) have shown heavy metals concentrations in soil at the area of landfills influence not exceeding the permissible values (sampling depths were 1–6 m). The concentration ranges of Pb, Cr, Ni and Cd were 0.2–1.6, 0.06–0.97, 0.06–0.44 and less than 0.2 mg/kg respectively. There are significantly higher Pb concentrations in our research, while Cr and Ni concentrations are only slightly higher. Besides, Nikolaev (2014) has determined the total contents of heavy metals (Pb, Cr and Ni) in soil near landfill, which were 1.5–3 times higher in comparison to their background concentrations. According to Nikolaev (2014), Cr concentration ranges from 100 to 120 mg/kg, Ni – from 62 to 92 mg/kg, Pb – from 25 to 60 mg/kg. Besides, Mor et al. (2006) have found Pb, Cd, Cr and Ni concentration in groundwater near the landfill lower the detection limit and have suggested these metals to be largely absorbed by soil. Comparison of these data with our research lets to suggest that majority of the heavy metals considered exist in soils in bound form after migration from landfill. However, this suggestion has to be approved by additional investigations.

Some heavy metals concentrations (Pb, Cd and Ni) in soil at the area not impacted by leachate outflow (flat terrain, 300 m north to Stadnytsia landfill and approximately 300 m from sampling area) are obtained from unpublished data of State Environment Inspection in Vinnytsia region. These concentrations are as follows: Pb – 2.2 mg/kg, Cd – 0.05 mg/kg, Ni – 0.48 mg/kg. Comparing these values to our results, we may suggest the next. Pb and Ni concentrations are more or less similar in both areas (except few points). This proves before mentioned suggestion about little impact of Stadnytsia landfill on Pb and Ni concentration in nearby soil. Cd concentration at the area not impacted by leachate outflow is similar to concentrations in sampling points situated further from the landfill. This also corresponds to previously considered Cd concentration dynamics. Thus, we can suggest that after decreasing at first 200–250 metres from the landfill Cd concentration is suppose to be stabilised.

Figure 6 Heavy metals concentrations in soils near landfills in Vinnytsia region: (a) Cd; (b) Pb; (c) Ni (1 – Orativ, 2 – Koziatyn, 3 – Yampil, 4 – Chernivtzi, 5 – Tyvriv, 6 – Kalynivka, 7 – Bar, 8 – Gajsyn, 9 – Tomashpil, 10 – Zhmerynka, 11 – Stadnytsia (investigated landfill))



Comparing the heavy metals concentrations in soils near the investigated landfill to those measured near other landfills in Vinnytsia region, one should note much lower concentrations at the investigated territory [Figures 6(a), 6(b) and 6(c)]. This is even despite the landfill near Stadnytsia is much larger than other landfills and has much more potential for soil contamination. These results indicate strong negative consequences of the wrong landfill site selection and failure to comply with the rules of landfill construction and operation. In the case of Vinnytsia region, landfills do not meet with the environmental requirements and heavy metals concentrations in soils nearby are several times higher in comparison to those in soils near Stadnytsia landfill (there are also

environmental problems, but this landfill meets the requirements much more). This is despite the waste volume at other landfills is few times or even ten times smaller.

Generally, there is a potential risk of soil contamination by Cr and Pb (to a lesser extent) in the impact zone of investigated landfill, while the pollution levels of Cd and Ni are negligible. Also, despite the proximity of the sampling points to each other, one should note significant changes of the heavy metals concentrations, especially Pb and Ni.

4 Conclusions

The analysis of concentrations of heavy metal mobile forms in soil near the place of leachate outflow at one of the Ukrainian landfills shows that allowable limits are not exceeded. The highest levels are about 0.5 of limit (Cr and Pb) while the concentrations of Cd and Ni are negligible. Besides, the heavy metals concentrations in soil at the study area are lower compared to those in soils near other landfills in Vinnytsia region (Ukraine). This confirms that the wrong site selection and failure to comply with the requirements of landfill construction and operation could have a significant impact on soil contamination with heavy metals. Comparing obtained results with results of other authors, we can conclude that mobile forms of heavy metals present only a small part of their total content in soils near landfills. These are mobile forms which most likely come to the soil from landfill. Clear dependence of heavy metals concentration on the distance from landfill leachate outflow can be found only for Cd (its concentration decreases slowly while increasing the distance from the landfill site). For Pb and Cr such dependence is not obvious, that may be due to the proximity of sampling points to each other and the existing of predominant migration routes of mobile forms of these metals. The landfill impact on the concentration of Ni mobile forms in soil is likely minimal, taking into account chaotic changes in its concentrations at different sampling points. Thus, the ratio of heavy metal total content and its mobile form concentration needs to be investigated more for clearer understanding of soil contamination near landfills by heavy metals.

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