The Accumulation of Heavy Metals Pb, Cu and Cd at Roadside Forest Soil

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Letter to Editor

The Accumulation of Heavy Metals Pb, Cu and Cd at Roadside Forest Soil

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Abstract

The accumulation of heavy metals at the roadside has been studied by the analysis of topsoil samples. Concentrations of the elements Pb, Cu and Cd were measured on 81 topsoil samples collected from three similar sites located near the Vilnius-Klaipėda highway.

The soluble and total content of heavy metals was determined in fractions >2mm by atomic absorption spectrophotometer employing an air–acetylene flame. Different chemical extractants AAAc-EDTA and DTPA were used for determination of extractable elements. The comparison of the results indicates that AAAc-EDTA extraction solution is the most efficient and well suited for determination of Pb, Cu and Cd.

The highest heavy metals concentration was found at a distance of 5 meters from the road and it exceeded the local background in the forest soils for all metals. Contents of metals tended to decrease with increasing distance from the highway. The results showed that the soil near the highway had significant enrichment, particularly in Pb, whose easily mobile amount exceeded maximal limited concentrations at a distance of 5-40 meters, and to a lesser extent with Cd and Cu.

Keywords: heavy metals, forest roadside soils, environmental pollution

Introduction

In our century of advanced technologies and technical progress soil contamination by various pollutants is one of the most significant environmental problems, which is likely to become more serious and more widespread in the relative near future.

The main sources of heavy metals (Cu, Pb, Cr, Cd and Zn) in the air are traffic, domestic heating and long-range transport [1-4]. Pollutants emitted into the atmosphere from mobile sources comprise about 70% of pollutants (almost 440,000 tons per year) in Lithuania [5]. Although heavy metals constitute an insignificant portion among transport pollutants, they play a potential role in assessing the quality of the roadside environment. It is determined that plants, snow cover and soil near highways in Lithuania accumulate heavy metals, mostly Pb, also Cr, Ni, Cd, Sr as well as Ti, V, Fe, Co, Cu, Zn and Zr [6-10].

The persistence of heavy metals in soils is a big environmental problem. This could have long-term implications for the biological, chemical and physical properties of agricultural and forest soil and its fertility as well as productivity [11]. The heavy metals Cr, Cd, Pb, Ni, Cu, Zn take part in the biological turnover and their excess or lack of disturbance of the metabolism and inhibited vegetation [12, 13]. Metals may bioaccumulate in living organisms and be transferred into the food chain [14, 6]. It is a confirmed fact that the major parts (75-80%) of heavy metals get into human organisms with vegetable diet when plants take it from the soil [15, 16].

The dispersion of heavy metals and their migration in arable land of Lithuania have been investigated by researchers by evaluating the total amount [17-21], yet there is little data concerning the amount of soluble fractions of heavy metals and distribution in the roadside soils in

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Table 1. Characteristics of the soil.

Geomorphology	Forest type	Habitat	Soil group	Organic matter, %	Soil texture, %
Lymnoglacialic plain	Vaccinio- myrtilo Pinetus (Pv-m)	Poor fertility, dry soil (Nbl)	Dystri-Haplic Arenosol: Arh-dy	2.36 ± 0.59	Sand - 83.7 ± 4.5 Silt – 11.9 ± 4.3 Clay – 4.5 ± 0.4

forest ecosystems. In examining environmental pollution knowledge of the total amount of an element is not sufficient, as it indicates the degree of pollution, but doesn't provide information about bioavailability and toxicity with respect to specific biotic components. In this way it is important to know the easily mobile amounts of heavy metals which are immediately available for biological and geochemical process.

The purpose of the current study was:

1) investigation of forest soil contamination of the Vilnius-Klaipėda highway by Pb, Cu, Cd and accumullation of heavy metals depending on different distances from the highway
2) determination of total and soluble contents of heavy metals (Pb, Cd and Cu) in the forest roadside soils with various determination methods.

Material and Methods

Study Site and Sampling

For the present study soil samples were collected from three forest roadside areas located at a distance of 5-100 meters from the Vilnius-Klaipėda highway (see Fig.1). This highway is the most intensive traffic road in Lithuania with daily traffic volumes ranging from <12,500 to 15,000 vehicles per day [22]. The places were chosen similar in geomorphology, type of forest, habitat, and soil texture. Some characteristics of these places are described in Table 1.

Sampling plots were marked in each area at 5, 10, 15, 20, 25, 30, 35 and 40 meters distance from the highway. Three soil samples were taken from every distance in all selected sites and analyzed independently. Each soil sample consists of about 4-5 sub-samples collected from the sampling plot of about 6 m^2. At each sampling point, the top 10 cm layer of the soil profile was taken using a stainless steel hand trowel, which was cleaned between samples to avoid cross contamination. In total, 81 soil samples were collected.

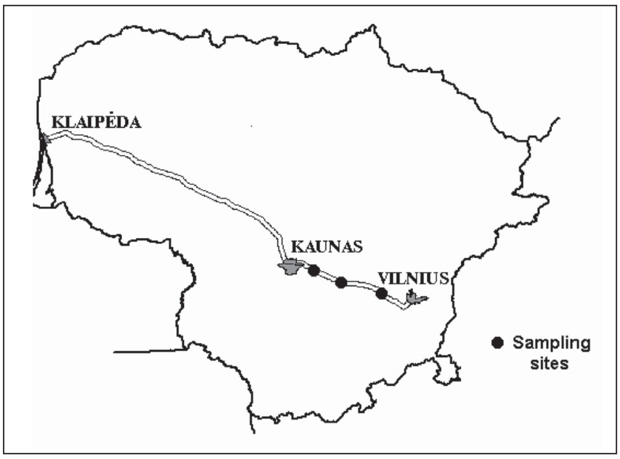

Fig 1. Sampling sites at the highway Vilnius-Klaipėda.

Chemical Analyses

Soil samples were air-dried, ground and passed through a 2mm sieve. Organic matter was estimated by the Walkley-Black titration method [23]. Particle size distribution was analyzed following a pipette method [24]. Soil pH was measured potentiometrically in a suspension of 10 g soil in 50 ml 1M KCl solution [25]. The pH (KCl) value was measured after 2 h from a re-stirred suspension.

For the chemical extraction of heavy metals two types of reagents are generally used: acids and complexants. Strong acid extractants such as concentrated HNO_3 and aqua regia are often used to determine "total" metals in contaminated soils.

Total concentrations of Cd, Pb and Cu in the soils were analyzed according to aqua regia method [26]. Briefly, 3 g of dry soil was weighed in the reaction vessel, 21 ml of HCl followed by 7 ml of HNO_3 was added and left to stand overnight. The suspension was boiled slowly for 2 h under reflux condenser.

The soluble trace elements contents were determined using different extractants, namely 0.02 M ethylenediaminetetraacetic acid (EDTA) (pH 4.65) solution and 0.005 M diethylenetriaminepentaacetic acid (DTPA) (pH 7.31) solution for the determination of soluble heavy metals content.

Extraction with specific extracting agents, especially containing chelating agents, allows the examination of the distribution of soluble and exchangeable forms. EDTA as extracting agent for many elements has been widely applied in soil science and environment chemistry [27-31]. It was found that Cd, Cu and Pb concentrations in plants growing on contaminated soils were related to the EDTA-extracted metals from the soils. This extractant was used in order to compare obtained results with literature data as well as to find a correlation between extractable fraction of heavy metals in the forest soil and plants growing in that soil.

The detailed procedure of EDTA extraction outlined by Lakanen and Ervio [32] was followed in this study. The AAAc-EDTA extraction solution was made by diluting 57.1 ml 100% acetic acid, 37.3 ml NH_4OH and 7.44 g

Na_2EDTA to 1 L with distilled water. The pH was adjusted to 4.65. Soil and extracting solution (1:10) were shaken for 1 h.

Extraction of trace elements by DTPA solution also mainly applies to the estimation of the availability of Cu, Cd and Pb to plants growing in the contaminated soils. The DTPA extraction method was followed according to ISO standard [33]. The soil sample (10 g) is extracted under shaking conditions, with a mixed buffered solution (pH 7.31) of triethanolamine (0.1 mol·l^{-1}) with calcium chloride (0.01mol·l^{-1}) and DTPA (diethylenetriaminepentaacetic acid 0.005mol·l^{-1}) [33].

Total and soluble metals concentrations were determined by air-acetylene flame (Cu) and graphite furnace (Pb, Cd) atomic absorption spectrophotometry employing background correction facilities to eliminate all non-specific absorption (Perkin-Elmer, AANALYST-100). External standards prepared in the corresponding extraction solution were used for calibration for each extract.

Statistical evaluation was applied in data analyzing study.

Results

Total Heavy Metals Fraction

The total amounts of Pb, Cu and Cd extracted by aqua regia in forest roadside soils are given in Fig.2. Their amount varies from standard-background level to harmful exceeding maximal limited concentration [34]. The highest concentrations for all metals were found at a distance of 5 meters from the highway and they tended to decrease with the increasing distance from the highway.

The total concentrations of Pb ranged from 7.88 to 54.27 mg·kg^{-1} and at a distance of 5 m the total amount of Pb was 4 times higher than the one obtained from forest soils at a distance of 40 m from the highway. At a distance of 5-10 meters from the road total concentrations of Pb exceeded maximal limited concentration (32 mg·kg^{-1} according [34]) 1.4 times. At a distance of 5-20 meters from the highway Pb accumulation, as compared to the local background in forest soils (17.6 ± 0.8 mg·kg^{-1} according [35]), was 2.1 times greater, while elsewhere it did not exceed local background.

The accumulation of Pb content in forest soils with the distance from the highway was evaluated by logarithmic relationships. It was expressed by following regression equation: $y = -17.741\ Ln(x) + 73.668$, $R^2 = 0.8891$.

The total content of Cu in forest roadside soils varied from 1.32 to 11.25 mg·kg^{-1}. The highest total Cu concentration was determined at a distance of 5 m from the highway and it was 3.3 times greater than total Cu concentration at a distance of 40 meters. The total amount of Cu at a distance of 5 meters in forest soils exceeded the local background concentrations [35] 1.8 times, while with the increasing distances from the highway the total concentration of Cu decreased and didn't exceed local background.

The accumulation of Cu content in soils with the distance from the highway was expressed by following regression equation: $y = -2.581\ Ln(x) +11.288$, $R^2 =0.6614$.

The total amount of Cd in forest soils at selected areas ranged from 0.096 to 1.19 mg·kg^{-1} and Cd values did not exceed maximal limited concentration (3 mg·kg^{-1}, according [34]). The highest total content of Cd was determined at a distance of 5 meters and it was 4.5 times higher than at a distance of 40 meters from the highway. At a distance of 5-10 meters from the highway the accumulation of Cd, as compared to the local background in the forest soils (0.72 ± 0.06 mg·kg^{-1}, according [38]), was 1.4 times greater, while with the increasing distance from the highway the total content of Cd decreased.

The relationships between the distance from the highway and content of Cd were expressed by the equation: $y = - 0.3637\ Ln (x) + 1.5286$, $R^2 = 0.8002$.

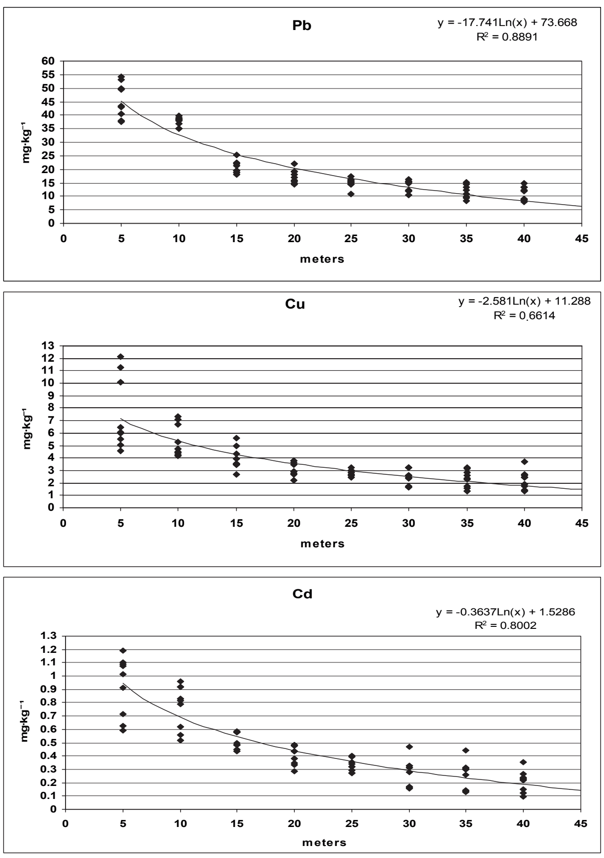

Fig. 2. Aqua regia extractable heavy metals (Pb, Cu, Cd) concentration at various distance from the highway.

Extracted Heavy Metals Fractions

The concentration of heavy metals Pb, Cu and Cd soluble forms extracted with DTPA and AAAc-EDTA complexants is shown in Fig.3. The content of heavy metals Pb, Cu and Cd extracted in AAAc-EDTA solution had a wide range of values; however, the tendency of accumulation is similar as for the total amount of heavy metals. With the increasing distance from the highway extractable heavy metals concentrations significantly decreased.

The AAAc-EDTA extractable content of Pb in forest soils is relatively high compared with Cu and Cd and values varied from 4.92 to 37.5 mg·kg^{-1}. The highest content of AAAc-EDTA extractable Pb was determined at a distance of 5 meters from the highway and it was 7.5 times greater than at a distance of 40 meters, although at all selected distances from the highway AAAc-EDTA extractable Pb concentrations in forest soils exceeded maximal limited concentration (6 mg·kg^{-1}, according [34]). DTPA extractable Pb concentration in forest soils varied from 2.4 to 14 mg·kg^{-1} and as compared to AAAc-EDTA extractable Pb content, was smaller, especially at a distance of 5 m from the highway. However, DTPA extractable Pb concentrations also exceeded the maximal limited concentration at a distance of 5 to 35 m from the highway.

The concentration of Cu extracted in AAAc–EDTA solution ranged from 0.68 to 4.69 mg·kg^{-1}. At a distance of 5 m from the highway the highest Cu concentration was determined and it was 6 times greater than at a distance of 40 m. DTPA extractable Cu concentration was 2 times smaller, as compared to AAAc-EDTA extractable content of Cu, and values ranged from 0.2 to 3.05 mg·kg^{-1} in the forest roadside soils.

Determined AAAc-EDTA extractable Cd concentrations in soils varied from 0.13 to 0.66 mg·kg^{-1}. The highest Cd content was determined at a distance of 5 m and the average value was 5 times greater than at a distance of 40 m from the highway. DTPA extractable concentration of Cd in the forest soils at selected areas ranged from 0.05 to 0.36 mg·kg^{-1} and as compared to AAAc–EDTA extractable Cd concentrations, was 1.6 times smaller.

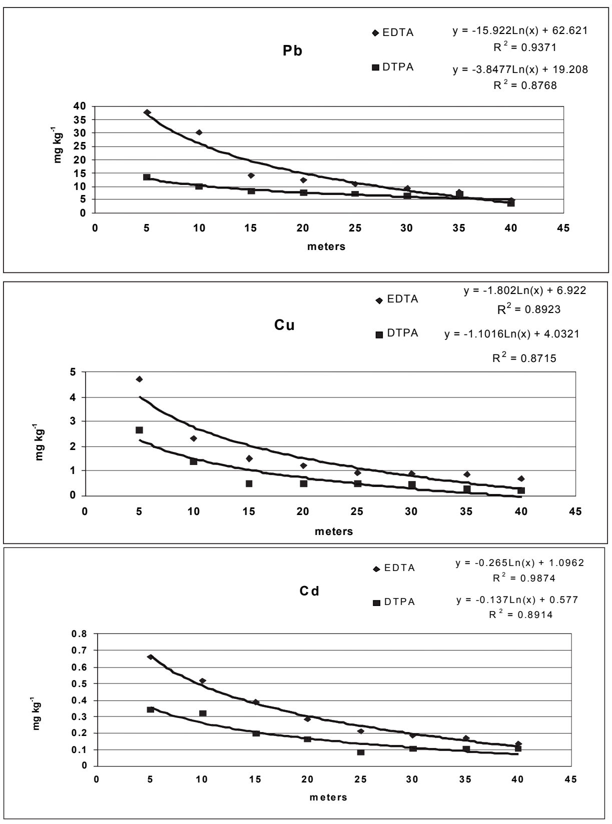

Fig. 3. AAAc - EDTA and DTPA extractable heavy metals fractions at various distances from the highway.

Table 2. Correlation coefficients (r) between heavy metals content of the soil as determined with different extractants.

Heavy metal / Extractant	Pb	Cd	Cu
Aqua regia – AAAc-EDTA	0.977	0.952	0.92
Aqua regia – DTPA	0.819	0.756	0.957
AAAc-EDTA - DTPA	0.863	0.865	0.964

The data for AAAc – EDTA and DTPA extractable heavy metals (Pb, Cu and Cd) was examined for the accumulation of heavy metals depending on distance from the highway by logarithmic relationships (see Fig. 3).

The statistical evaluation obtained from linear regression analysis (see Table 2) revealed that AAAc-EDTA extractable metals content was positively correlated with DTPA-extractable metals, giving r values of 0.863 (Pb), 0.964 (Cu) and 0.865 (Cd). Significant positive correlations of heavy metals total content with extractable fractions were obtained for all elements. The obtained results showed that a similar tendency of accumulation was observed for AAAc-EDTA and DTPA extractable fraction of heavy metals (Pb, Cu and Cu) as well as for total digestion amount.

The relationship between AAAc-EDTA extractable fraction of heavy metals from the total amount with the distance from the highway is shown in Fig. 4. The AAAc-EDTA extractable content of Pb, Cd and Cu in roadside soils was on average 69.26% ± 15.1, 67.95% ± 12.968, 41.541% ± 13.33 respectively, of their total digestion concentrations, which suggested that the mobility of three metals is different. The statistical evaluation showed that there are no correlations between relative percentage of extractable fraction from the total amount with the distance from the highway for Cd and very week correlation obtained for Cu and Pb. The relative percentage value of extractable fraction from the total amount depends on pH. Our analysis have indicated that pH of soils varies greatly. The greatest pH value, 7.4, was determined at a distance of 5 m from the highway and in the distance pH value gradually declined. At a distance of 10, 15 and 20 m pH value was 7.1, 6.5 and 6.2 respectively, and at a distance of 25 to 40 m it decreased slightly from 4.9 to 4.7. Contrary to expectation, decreases in pH of soils have not been accompanied by increases in the relative percentage value of extractable heavy metal fraction from the total amount. This may be a consequence of the factor that pH of EDTA extraction solution (pH 4.65) could affect pH of soil samples during analysis.

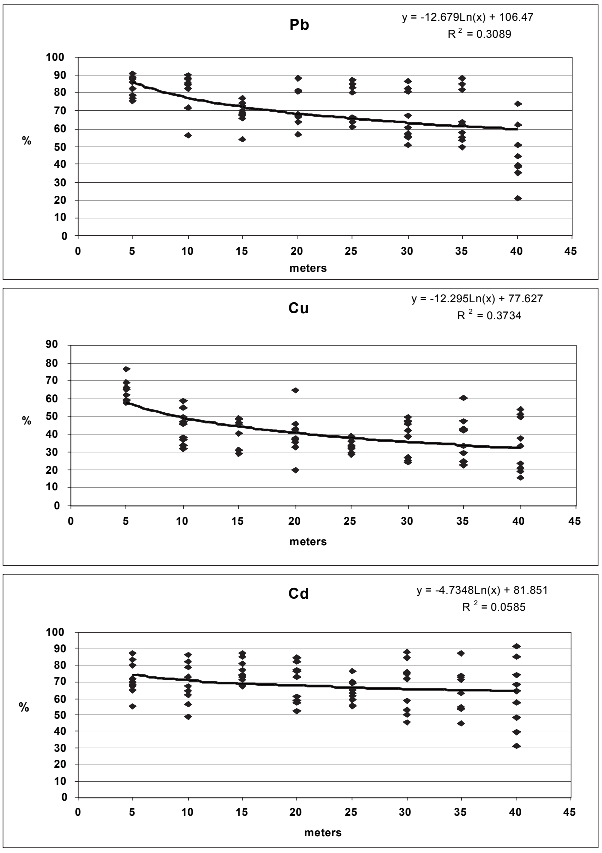

Fig. 4. Extractable heavy metals (Pb, Cu, Cd) fractions of the total amount (%) at various distances from the highway.

Discussion

In summarizing the obtained results it has emerged that the total amounts of heavy metals (Pb, Cd and Cu) in the roadside forest soils at a distance of 5-10 meters from the highway in comparison with the long-time forest monitoring data [35] are considerably higher and it reveals that the forest soil near this highway is polluted. Comparison of our obtained results at the forest soil to the results of soil analysis at an open area conducted at the same highway [9] has revealed similar trends of heavy metal accumulation. However, in comparison to the heavy metals contamination in open area soils near other highway roads, heavy metal concentrations near the selected Vilnius-Klaipėda highway are significantly higher. Research data reveal that recently this highway has been an intensive source of pollution and heavy metals, especially Pb, which are constantly accumulated in roadside soils.

Obtained data of easily mobile amounts of heavy metals in roadside soils indicate that the results depend on many factors such as metal and soil qualities, pH, distance from the highway, extraction method as well as forest vegetation biodiversity.

Soil pH acts significantly in the soil oxidation-reduction reactions and also in the formation of mobile metal forms. Soil pH value set by us varied and was the highest close to the highway and in the distance pH value declined possibly because of the impact of forest flora. It is important to evaluate soil pH in order to choose relevant solutions for extraction of heavy metals. Current research indicates that mobile forms of Cd relative value varied significantly when using different solutions: in extraction with EDTA complex solution with pH 4.65, double amount of Cd has been obtained than with the DTPA solution with pH 7.31. As it was noted, different easily mobile amounts of heavy metals in soil were obtained when using different extractants. Greater amounts of all metals (Pb, Cu and Cd) have been obtained while extracting with the EDTA than DTPA complex solution. According to the literature data, Cd, Cu and Pb concentrations in plants growing on contaminated soils are related to the EDTA-extracted metals from the soils [27, 29]. Thus, summarizing the obtained EDTA and DTPA extraction results, it can be stated that EDTA solution is more pertinent for measuring easily mobile fractions of heavy metals in forest soils as pH values of soil and extractants are similar and extracted amounts are several times greater than using DTPA complex solution.

The results of this investigation showed that the soils near the Vilnius-Klaipėda highway had significant enrichment, particularly in Pb and to a lesser extent with Cd and Cu. The rate of heavy metals contents at the same distance from the road followed the order: Pb > Cd > Cu. The greater concentrations in soils near the highway could represent long-term contamination of heavy metal from transport in a roadside environment. In the selected areas forest ecosystems act as biogeochemical barriers which attenuate intensity of Pb, Cd and Cu migration in the soils horizontally and vertically. The heavy metals (Pb, Cd and Cu) accumulate in the soil mostly at a distance of 5-10 m from the highway, where grass and small bushes predominate. Moving further, where higher bushes and trees grow, heavy metals content tended to decrease with increasing distance from the highway.

Further detailed research is required for more accurate evaluation of the heavy metals accumulation in soils and environmental pollution impact for the quality of roadside soils in forest ecosystems.

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