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THE ROLE OF PH IN HEAVY METAL CONTAMINATION OF URBAN SOIL

Asta KAZLAUSKAITĖ-JADZEVIČĖ[a], Jonas VOLUNGEVIČIUS[b], Virginija GREGORAUSKIENĖ[c], Saulius MARCINKONIS[d]

[a]*Traku Voke Branch of the Lithuanian Research Centre for Agriculture and Forestry, Žalioji a. 2, Trakų Vokė, LT-02232 Vilnius, Lithuania*
[b]*Faculty of Environmental Sciences, Vilnius University, M. K. Čiurlionio g. 21/27, LT-03101 Vilnius, Lithuania*
[c]*Lithuania Geological Survey, S. Konarskio g. 35, LT-03123 Vilnius, Lithuania*
[d]*Vilniaus kolegija/University of Applied Sciences, Saltoniškių 58, LT-08105 Vilnius, Lithuania*

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Abstract. The aim of this study to assess interdependence between urban soil pH and its accumulation of heavy metals. The article meant to be a contribution to a better knowledge of peculiarities and diagnostics of urban soil and its anthropogenic transformation. The hypothesis assumes that relationship between urban soil pH and its accumulation of heavy metals may be determined by the origin and age of parent material as well as the nature and degree of the anthropogenic impact. The spatial variability of topsoil pH level was performed in 100 points in eldership of Šnipiškės of the city Vilnius. Laboratory analysis was based on ISO 10390:2005. Samples were collected from 20 cm topsoil layer in the same sampling points where have been analysed concentrations of topsoil chemical elements using optical atomic emission spectrophotometry. The contamination of urban soils exhibits somewhat different compared to agricultural soils. In contradiction to earlier studies in Lithuanian agricultural soils where strong correlation between soil pH and Cr, Cd, Pb, Ni, Cu and Z_d found, the conducted analysis shows a statistically reliable, but very weak (<0.3) correlation between the soil pH and concentration of contaminants. The proof to this correlation is provided by an existing relationship between pH and the concentration of copper (r = 0.20), mercury (r = 0.15), strontium (r = –0.12) and the overall contamination index (r = 0.12). The applied statistical analysis, however, failed to reveal the nature of interdependence between the soil pH and its contamination with studied heavy metals there concentration of contaminant chemical elements depends on the pH range of the soil and, conversely, the chemical reactivity of the soil changes affect on the concentration of studied chemical elements.

Keywords: soil pH, soil contamination, correlation analysis.

Introduction

One of the many environmental problems is a growth in extent to which the soil is being anthropogenised, subsequently causing the soil properties and its chemical and physical composition to change. The ever-growing rate of soil anthropogenesis (human impact on the environment, in this case resulting from the use of soil for human needs) expands the gap between the chemical, physical and biological properties of the soil found in urban territories and more natural soils. Nowadays Europe is facing an on-going loss of healthy soils reporting approximately 3.5 million sites potentially contaminated, resulting in failure to provide ecosystem services (Thematic Strategy ... 2006). In search of the ways how the contamination problem of the urban territories can be solved, the constant monitoring of the soil state as well as documentation of any changes observed in it are highly efficient. An assessment of the environmental risk due to soil pollution is of particular importance for urban areas, because of heavy metals direct and indirect effects on human health (Grzebisz *et al.* 2002). The inclusion of ecological risk assessment in soil quality standards shows an increasing interest in many EU Member States (Swartjes *et al.* 2008).

Individual concentration rates of chemical elements K_k (HN 60:2004; Taraškevičius 1996) as well as the overall index of contamination with these chemical elements Z_d (HN 60:2004; Taraškevičius 1996) and criteria of the

Corresponding author: Asta Kazlauskaitė-Jadzevičė
E-mail: asta.kaz@gmail.com

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maximum permissible contents usually are used for soil pollution assessment. Studies of trace element dynamics in long-term field experiments have been shown to be useful complementary tool to assess soil accumulation as well (Jones *et al.* 1987).

Within last decades in Lithuania, significant research interest has been shown in the problem of soil contamination with heavy metals in agricultural soils (Marcinkonis 2008; Sunkieji metalai … 2001). The extensive overview on long-term agricultural impact on accumulation of heavy metals in topsoil and subsoil using selected reference elements and reference horizons has been reported by S. Marcinkonis (2008). The sorption of the heavy metals from polluted after and their migration in the system soil-tree has been provided by D. Butkus *et al.* (2004). Overview for the analysis of the correlation between Lithuanian soils pH and concentration of soluble heavy metals found in the agricultural soils has been provided in monograph by Sunkieji metalai… (2001).

The extensive studies of urban soil contamination with heavy metals been reported by different Lithuanian scientists (Taraškevičius, Zinkutė 2011; Zinkutė *et al.* 2011; Kumpienė *et al.* 2011; Jankauskaitė *et al.* 2008; Gregorauskienė 2006; Gregorauskienė, Kadūnas 2006; Taraškevičius 1996). But due to high variation of heavy metals in urban soils mainly long-term statistically significant changes are detectable not only in Lithuania (Vasarevičius, Greičiūtė 2004; Sunkieji metalai … 2001) but in other countries as well (Reimann, Caritat 2012; Irha *et al.* 2009; Lado *et al.* 2008; Imperato *et al.* 2003).

Up to the present, there are limited studies of interactions and interdependence of soil pH and heavy metals not only in Lithuania (Eidukevičienė *et al.* 2010) but in other countries as well (Paartel *et al.* 2004). Soil resistance/sensitivity to contamination with chemical elements depending its physicochemical properties including pH range been overviewed at European scale by R. G. Burau and R. J. Zasoski (2002), by Kadūnas *et al.* (2005). In general the scientific papers providing the relevant information on the role of pH in heavy metal contamination are often regionally fragmented. From various countries both, the heavy metals found in soil and its pH range reported by V. Cappuyns and R. Swennen (2008), Tong-Bin Chen *et al.* (2005), T. L. Woodard *et al.* (2007).

Up to the present, there are limited Pedological studies of soil contamination with heavy metals in urban Lithuanian soils. A possible relationship between the soil pH and urban soil contamination (in Vilnius city) with heavy metals is represented in this article. This study contributes to the body of applied sciences by investigating issues of plants in urban territories, it is important for specific planting work and selection of recreation sites. It represents more comprehensive study with the aim to assess interdependence between urban soil pH and its accumulation of heavy metals and meant to be a contribution to a better knowledge of peculiarities and diagnostics of urban soil and its anthropogenic transformation; as well as the basis provided for the geochemical index of assessing the soil state.

1. Materials and methods

1.1. Site

The eldership of Šnipiškės, chosen for the analysis, is one of the oldest elderships found in the city of Vilnius. Eldership occupies part of the city centre and the north-western part of Vilnius (Fig. 1). From the geomorphological point of view, the eldership of Šnipiškės lies in the sandy terraces of the river Neris valley. In its central part, low-rise private houses are found predominant. In the northern, eastern and north-eastern districts of the eldership, however, the most dominant are multi-storey blocks of flats, and the eldership is surrounded by flows of heavy traffic on its western, northern and eastern boundaries.

It well represents the diversity of anthropogenic impact on the soil that is observed in Vilnius as well as the most typical state of anthropogenised soil that is characteristic of urban territories. The most common source of pollution results from traffic flows and industrial activity. Therefore this eldership can best represent the common state of the anthropogenised soil found in the city of Vilnius. The distinctive feature of the eldership is a large number of former and present pollution sources: organizations of construction sector, transport maintaining companies, boiler-houses, repair workshops, the Institute of Thermal Insulation, toy factory, companies producing metal equipment, furniture factories, garages used for car repairs, marketplace, residential houses (multi-storey blocks of flats and quarters of poor and neglected low-rise houses) and streets of heavy traffic.

1.2. Sampling

The soil samples were collected in an "envelope's" principle, taking 5 samples in equal amounts from the soil surface horizon A on specific locations as shown in Figure 1. The topsoil horizon A was chosen for the analysis because it undergoes the biggest impact of versatile origin – mechanical, physical, chemical and biological. Due to the extent of the chemical impact, soil obtains its unique composition and its properties that are only typical to it (e.g. a shift in the pH range found in the surface of the soil).

Soil pH samples were collected in the same locations as the soil samples for chemical element concentration detection. Sampling points were decided by the Vilnius city municipality monitoring study "Urban and recreational territories soil pollution level evaluation" (Taraškevičius 2007). Sampling locations were chosen with consideration for the territories identified by Vilnius city municipality for being areas of intense pollution and differing pollution sources: transport hubs, marketplaces, teaching institutions, areas of mass assembly or residential areas. Therefore the choosing of sampling locations was based on three principles: technogenic pollution diversity, accessibility to data about soil pollution by chemical elements and possibility of comparison, comparing results of the soil pH measurements with results of soil pollution by chemical elements.

The analysed attribute meanings meet the normal eligibility requirements.

The sampling was formed to reflect the diversity of technogenisation patterns of the chosen territories, as well as to incorporate all geomorphological structures in the territory. Therefore while formulating the sampling it was aimed at being representative of the technogenisation diversity and the geomorphological situation.

Samples were collected in standard Urban conditions, during summer time (June 2009). For collection of samples a day without rain was chosen, in meteorological conditions which are common for Vilnius.

Sampling area fully represents regular approbation network. Soil samples for the pH analysis were collected in 2009 in the same soil sampling area, in Šnipiškės neighbourhood, Vilnius. The same soil sampling area was used in 1996 for determining the chemical element concentration coefficients.

1.3. Laboratory analyses

The collection and preparation of the soil samples for the laboratory analysis were based on methods as they are found in recommendations provided in international standards (ISO 10390:2005) and widely employed by Lithuanian geologists in their geochemical research. The analysis was conducted in the laboratory of General and Inorganic Chemistry Department, at Vilnius University, for which the pH meter of "Mettler Toledo MP220 pH Meter" was used.

Taking the measurements of the pH range in soil, the samples were prepared in ratio of 1 part of soil surface material (2 g) to 5 parts (10 ml) of boiled distilled water. In an attempt to obtain a more precise estimation of pH range, the solution was filtrated through a usual neutral filter paper. For calibration, the standard buffer solutions (pH 7.00 and pH 4.00) were used. The samples were analysed three times.

1.4. Data mining and statistical procedures

All of the data on sample collection and analysis were stored into the geographic database (GIS). The statistical analysis of contamination rate of individual chemical elements was carried out taking the data of Vilnius Municipality, provided in R. Taraškevičius's report of 1996. Along with the pH data, individual concentration rates of chemical elements K_k (HN 60:2004; Taraškevičius 1996) as well as the overall index of contamination with these

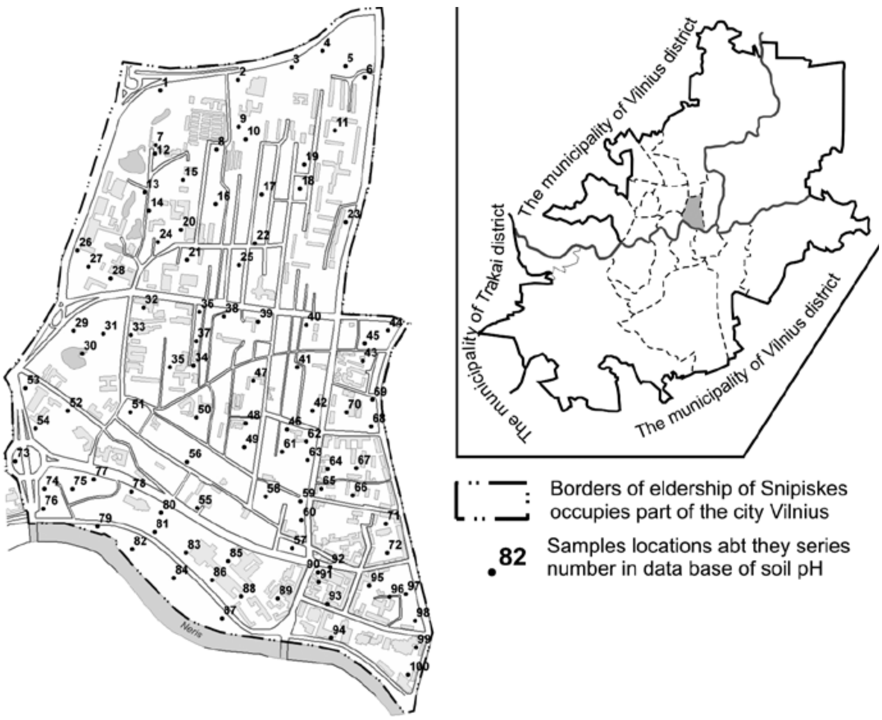

Fig. 1. Soil sampling specific locations (100 points) in eldership of Šnipiškės of the city Vilnius

chemical elements Z_d (HN 60:2004; Taraškevičius 1996) and background individual concentration rates of chemical elements (HN 60:2004; Taraškevičius 1996) were used. According to the data submitted in 2007 by Vilnius city council (Taraškevičius 2007) the major part of the soil chemical element concentration during the timeframe from 1996 to 2006 did not change and remained the same.

The statistical analyses (Fig. 3) were executed using the *"Statistica 6"* program. In reference to the collected data of the investigated samples, contour maps of topsoil pH range (Fig. 2) were prepared with the use of *"ArcGis 9.1"* program.

2. Results and discussion

What sets apart the investigated territory of the eldership of Šnipiškės from other neighbouring more natural territories is that it is only distinguished by the intensity of the conducted anthropogenic activity, which in the long term has caused the pH of the soil to increase (Fig. 2), i.e. the surface horizon of the soil has turned alkaline. As much as 98.04% of the surface horizon A of the investigated soil turned out to have the pH which is typically found in the alkaline surface of soils (≥7.0). Only 1.96% of the territory could be described as having the close to neutral pH (6.6–6.9).

Undoubtedly, one of the most significant factors influencing the reactivity of the soil surface is the geomorphological features of the location. Background values are important when analysing anthropogenic soil pH. The pH of the soil depends on physical, chemical and microbiological soil qualities a more visible reaction change during a relatively short soil formation period is caused only by liming of soil or soil dumping. The fluvio-glacial (Flg) and fluvial (Fl) deposits that shape the landform of the eldership of Šnipiškės had strong influence on the formation of the background values of the soil pH found in the territory.

The tendency that was observed, while investigating the territory, suggests that greater pH values (pH > 8) are found as typical of younger deposited sediment (Fl s-I) rather than the older deposits (Flg II, Flg III), where the value of pH ranges from 7 to 7.5 (Fig. 2). Within the eldership of Šnipiškės, the topsoil have a tendency of being more acid with their samples taken from the younger fluvial deposits (Fl s-I) in the south and less acid

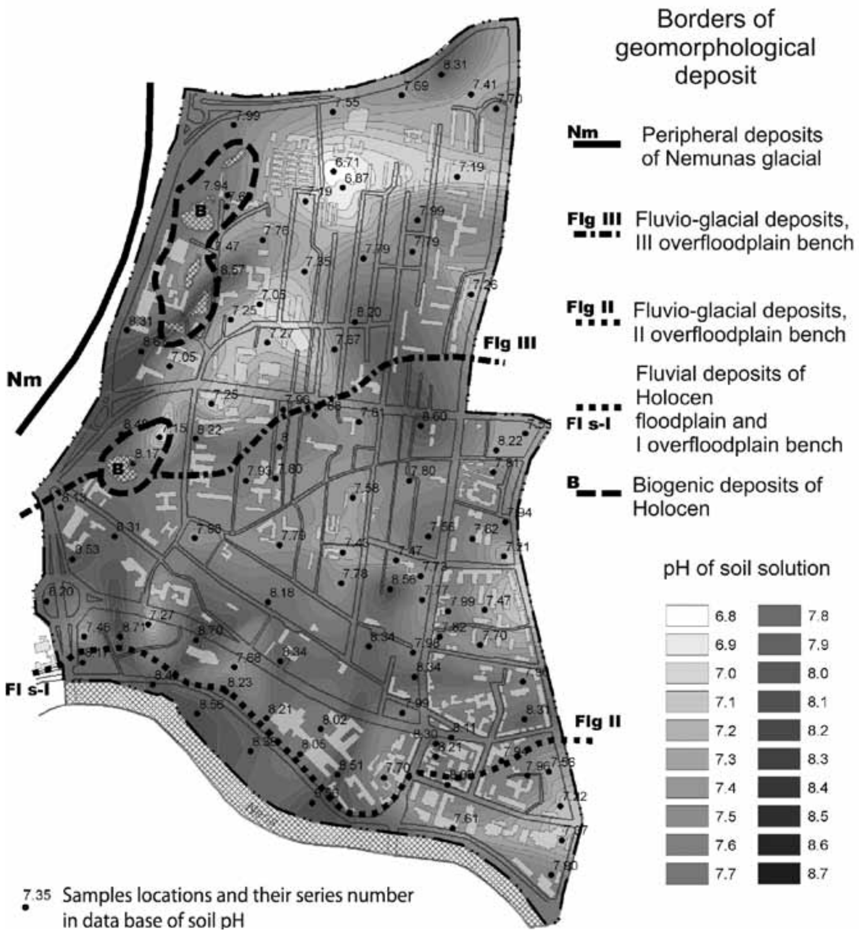

Fig. 2. Specific soil samples locations and soil pH values in eldership of Šnipiškės of the city Vilnius

with samples taken from the older fluvio-glacial deposits (Flg II, Flg III) in the north.

I–III terrace as well as B biogenic structure pH measurement massifs: for Flg I the average pH is 7.98, Flg II the average pH is 7.98 as well. The average for Flg III pH is 7.67 and for B the average pH is 7.72.

In the north-western part of the eldership of Šnipiškės, there are a few small lakes of thermal karst origin that have turned into a wetland. Because of the fenny edges of the lakes, biogenic deposited sediment (B) is found prevailing in the lowland. These deposits commonly have a greater value of topsoil pH due to the strong sorption of the alkaline contaminant elements caused by organic matter.

The north-western part of the investigated territory clings to the slope of the river Nėris valley formed by the peripheral deposit (Nm) brought from the river Nemunas in the glacial period. This deposit is found beyond the borders of the investigated territory and therefore has no significant impact on reactivity of topsoil.

According to the pH map of background soils published by J. Grybauskas (1978), the predominant soils in the territory of Vilnius were soils with the pH value ranging between 4.5 and 6.1. However, under the influence of morphological processes that took place in Vilnius and the slowdown of these processes, – the environment of the time was gradually shaped by human action and anthropogenic contamination that came along.

Studies in different environments (agricultural, urban and transition land-use zones) demonstrate that in the acid soils, heavy metals are more mobile than in the alkaline one, and the alkaline environment better sustains metals (Sunkieji metalai … 2001). The solubility of heavy metals decreases with increasing pH and vice versa, therefore accumulation of heavy metals often observed in the alkaline environment. On other hand, contamination of soil with heavy metals can stimulate rising of soil pH level.

The results achieved in the cluster analysis of soil contamination with heavy metals (Fig. 3) showed that the strongest association within the investigated territory is made up of Ba-Co-Ni-Zn, and the weaker one by Ag-Cr-Pb and Cd-Hg-Sn, which in most cases appear in the soil because of air pollution and, taken together, form the index of the overall contamination Z_d .

Certain regularities emerge when analysing the soil contamination with individual chemical elements in topsoil. Statistically, the strongest accumulation association is formed by Ba-Co-Ni-Zn, among which a strong linear interdependence is observed and which can be related with both clay minerals of organic origin and pollution levels in the city. In this association, two elements: barium (Ba) and cobalt (Co) are closely linked. The weakest genetic links of these metals connect them with zinc (Zn) that is found in their association.

The accumulation association of boron (B) and

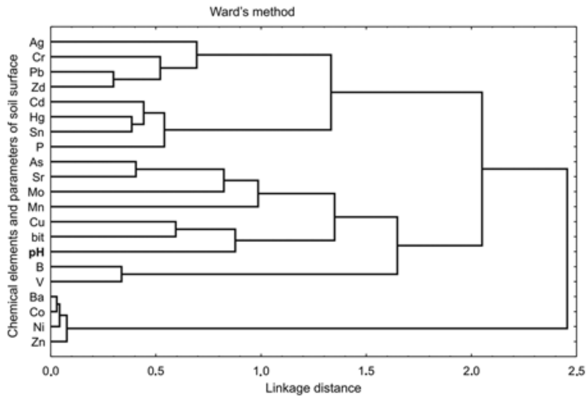

Fig. 3. Relationships and linkages of heavy metals and rare elements in soil (Šnipiškės eldership, Vilnius) by cluster analysis Ward's minimum variance method

vanadium (V) is related to associations of Cu-Bi and As-Sr-Mo-Mn via correlative links. Molybdenum (Mo) and manganese (Mn) are closely related to clayey soils, and an interdependent association between them usually shows up in areas of high pollution levels caused by traffic. Except for B-V, which are related to clay minerals found in the soil and commonly regarded as typical elements of power plat pollution, and Sr-As, which are linked to carbonaceous soil material of large scale broken rock, these elements and parameters form pseudo-associations based on weak links.

With reference to the book "Soil and water chemistry" (Burau, Zasoski 2002), it becomes possible to explain a stronger correlation between lithogenous elements of arsenic (As), strontium (Sr), molybdenum (Mo), manganese (Mn) and the pH value of the topsoil. Copper (Cu) is yet another lithogenous trace element found in soil, but the increased levels of it, as they are observed in the topsoil, can be put down on anthropogenic activity due to the intensive use of copper in industry. In terms of the city's environment, nickel (Ni) and zinc (Zn) are more likely to be taken as metals of technogenic origin and their concentration in soil is related to carbonaceous factors. They poorly correlate with the pH value, although they are known to have strong correlative links between themselves (Ni-Zn).

Results of correlation analysis suggest that the topsoil pH has weakest impact on technogenic origin metals.

The interdependence between soil reactivity and its contamination with heavy metals is hypothetical. A possible interdependence between the two issues suggests that the concentration of pollutant elements found in the soil depends on the pH value of the soil and, likewise, the soil reactivity changes affection the concentration of the pollutant elements. In the course of investigation, the conducted correlation analysis between the topsoil pH and the concentration of contaminant elements (Ag, Pb, Sn, Zn, Hg, Cd, Sr, Co, Ba, As, Ni, Cr, Cu, Mn, Mo, V, B, P) found in the soil showed very weak linear interdependence ($r < 0.3$). High variation in the content often reported to make it difficult to analyse their degree of contamination without taking into account the spatial variability of metals and their content in the parent materials of soils, which is the natural background (Grzebisz *et al*. 2002). Studies in Moscow region showed that distribution of heavy metals by the fractions primarily depended on the chemical properties of the heavy metals themselves; the pollution level; and, to a lesser degree, on the soil properties (Plyaskina, Ladonin 2009).

Copper (Cu) concentration found having very weak linear correlation with pH ($r = 0.2$), but that mainly based on the three ultimate points (out of 100 possible ones) (Fig. 4).

The mercury (Hg) linear correlation with pH $r = 0.15$ was found even weaker than that of copper (Cu) (Fig. 5). The weak negative, but statistically reliable association was indicated between pH and strontium (Sr), $r = -0.12$ (Fig. 6).

In the course of investigation, it was also established that there is a very weak direct relationship between topsoil pH value and the index of the overall contamination Z_d ($r = 0.12$) (Fig. 7).

The conducted analysis determined the interdependence between the index (Z_d) of overall contamination with some contaminants (Cu, Hg, Sr) and the topsoil pH value.

The observed results confirmed that the contamination of urban soils exhibits somewhat different compared to agricultural soils. In contradiction to earlier studies in Lithuanian agricultural soils where strong correlation between soil pH and Cr, Cd, Pb, Ni, Cu and Z_d found (Sunkieji metalai … 2001), the conducted urban soils analysis shows a statistically reliable, but very weak (<0.3) correlation between the soil pH and concentration of contaminants (Figs 4, 5, 6, 7).

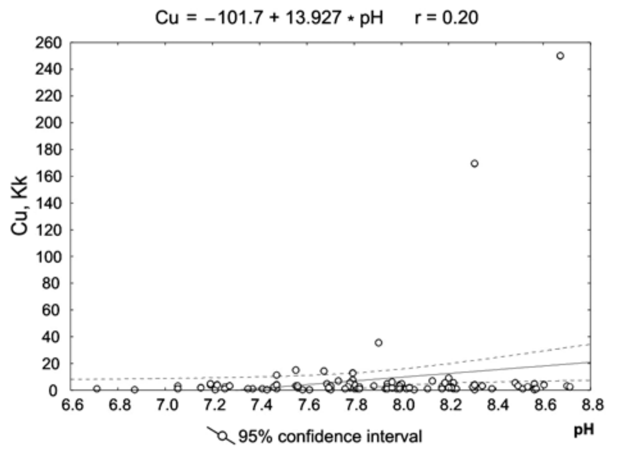

Fig. 4. Correlation (r) between concentration coefficient of copper (Cu) and topsoil pH in eldership of Šnipiškės of the city Vilnius

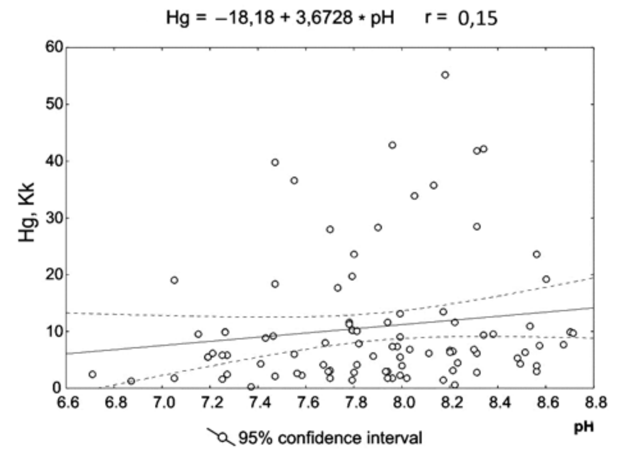

Fig. 5. Correlation (r) between concentration coefficient of mercury (Hg) and topsoil pH in eldership of Šnipiškės of the city Vilnius

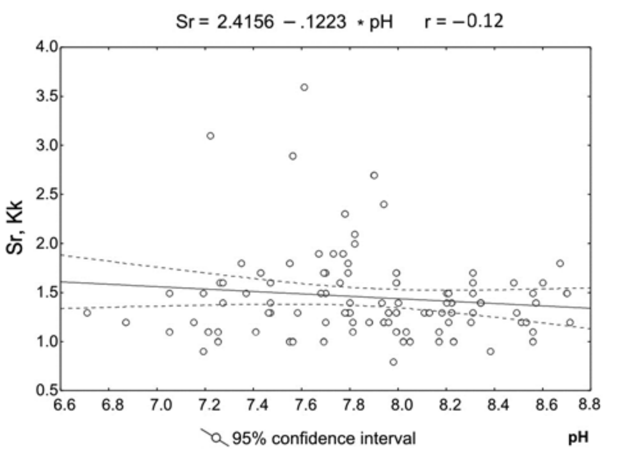

Fig. 6. Correlation (r) between concentration coeficient of stroncium (Sr) and topsoil pH value in eldership of Šnipiškės of the city Vilnius

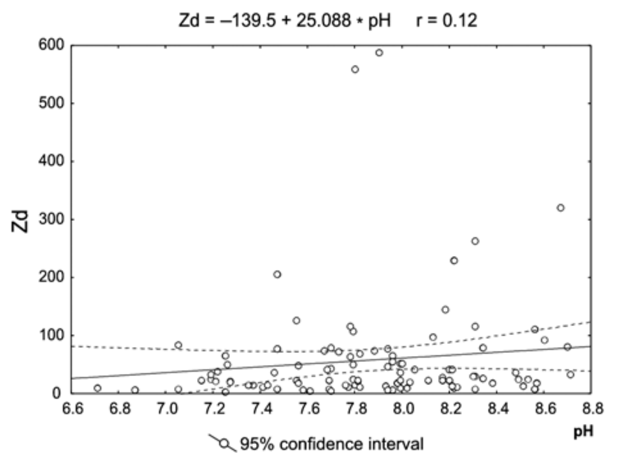

Fig. 7. Correlation (r) between the index of overall contamination (Z_d) and topsoil pH value in eldership of Šnipiškės of the city Vilnius

Conclusions

Anthropogenic impact determines the alkalinity of the urban soil (pH → 9) and, simultaneously, the intensity of sorption of specific contaminants (silver (Ag), lead (Pb), tin (Sn), zinc (Zn), mercury (Hg), bitumoide, cadmium (Cd), strontium (Sr), cobalt (Co), barium (Ba), arsenic (As), nickel (Ni), chromium (Cr), copper (Cu), manganese (Mn), molybdenum (Mo), vanadium (V), boron (B), phosphorus (P) the overall contamination (Z_d).

The morphological features of the territory influence the pH value of the soil. Greater pH values (pH > 8) are more characteristic of younger deposited sediment than of the older deposits.

The analysed spatial premises, by which the pH shift in the soil and concentration of heavy metals in contaminated areas as well as the interdependence of the two issues are determined, confirm the claim for the existence of this interdependence.

According to analysis results specific heavy metals form specific accumulation associations that indicate different contamination sources.

The conducted analysis showed a statistically reliable, yet very weak correlation between the soil pH and concentration of heavy metals (Ag, Pb, Sn, Zn, Hg, Cd, Sr, Co, Ba, As, Ni, Cr, Cu, Mn, Mo, V, B, P) found in the soil ($r < 0.3$). Between specific heavy metals and the topsoil pH, a strongest statistically reliable relationships were observed between the soil pH and copper (Cu) as well as the soil pH and the index of overall contamination (Z_d).

The conducted analysis does not reveal the nature of the interdependence between the soil pH and its contamination with heavy metals, i.e. it remains unproven if the soil reactivity influences its contamination or, conversely, if the soil contamination causes changes to its reactivity.

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Asta KAZLAUSKAITĖ-JADZEVIČĖ. She is a PhD student, working at the Lithuanian Research Centre for Agriculture and Forestry. Her research interests include renaturation, GIS, and soil carbon modelling.

Jonas VOLUNGEVIČIUS. He is a Doctor of Physical Sciences (Geography) and works as a Docent at the Department of Geography and Land Management, Faculty of Natural Sciences in Vilnius University, and also is a President of Lithuanian Soil Science Society at the Lithuanian Academy of Science Agricultural and Forest Sciences Department. He is the author and co-author of ~14 scientific papers and other scientific publications and has attended 2 international and 11 local conferences. His research interests include geography of landscape management, soil geography, environmental policy, and application of GIS in geography.

Virgilija GREGORAUSKIENĖ. She is a Doctor of Physical Sciences (Geography), works as a Senior technician at the Subdivision of Engineering Geology and Processes, Geological Survey of Lithuania, and is also a Lector at Mykolas Riomeris University. Her research interests include environmental geochemistry, soil science, soil contamination, soil quality standards, air borne contamination, methods of geochemical mapping at different scales, and medical geology.

Saulius MARCINKONIS. He is a Doctor of Biomedical Sciences (Agronomy), working as a Head of Strategic Development Department at the University of Applied Sciences (Vilniaus kolegija). He is the author and co-author of ~33 scientific papers and has attended 40 conferences. His research interests include soil chemical degradation, liming, fertilization, soil pollution, renaturation, and GIS.