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Contamination and health risk assessment of potentially toxic elements in soils of different kinds of land use in Lithuania

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Using the case of five different kinds of land use from different territories of Lithuania, this study assesses the level of contamination and human health risk assessment of arsenic (As), and heavy metals (HMs) such as cadmium (Cd), copper (Cu), zinc (Zn) and chromium (Cr) in the surface soil of the study areas. Geo-accumulation index (I_{geo}) analysis indicated that heavy Cd contamination occurred in agricultural territory (AT), while for As, no contamination to mild contamination occurred in all territories. For living territory (LT), green territory (GT) and technical territory (TT), I_{geo} readings for Cu showed no pollution to moderate pollution, while there was no pollution for natural territory (NT) and AT. For AT, there is no contamination from Zn or Cr. By contrast, I_{geo} values for Zn and Cu represent minimal to no pollution in the remaining territories. As in LT and Cd in AT, two of the HMs discovered, were deemed to be of medium risk, whereas other components fell into the permitted range. Among three different routes to exposure, it was discovered that the ingestion pathway was the main health risk. The Hazard quotient (HQ) and hazard index (HI) values for As, Cd, Cu, Zn, and Cr were lower than the suggested limit (HI = 1), indicating minimal non-carcinogenic risk to inhabitants in the study regions. The carcinogenic risk values for As (1.12E-04 children), Cd (2.20E-04 children), and Cr (2.35E-04 children) in AT pose a risk to children's health when ingested. The GT's carcinogenic readings for Cr (1.02 E+00 adult), put adults at risk of developing cancer, whereas As (1.89E-04) and Cr (2.28E-04) in LT put children at risk of cancer and for TT, both adults and children were at risk due to Cr's higher carcinogenic values (1.93E-04 for adults and 5.21E-04 for children).

Key words: heavy metals, contamination, soil, health risk assessment.

INTRODUCTION

Soil is a complex and changing system, a key component of the living environment of people and ecosystems. It consists of accumulated materials of natural and anthropogenic origin. These materials are found in soil profiles and can accumulate for hundreds or thousands of years (Manta et al., 2002; Pasieczna, 2003; Li et al., 2013; Kowalska et al., 2016).

Anthropogenic processes are usually accompanied by negative effects, manifested as pollution. Pollutants that fall out of the atmosphere and enter with industrial, construction, agriculture, transport, sewage or household waste can accumulate in the soil. Most pollution sources are concentrated in cities where densely populated areas prevail (Lu et al., 2010; Taraškevičius and Zinkutė, 2011; Tomassi-Morawiec et al., 2016; Ferreira et al., 2017). Due to intensive anthropogenic activity, urban soils are technogenically affected and lose their natural soil properties (Nriagu and Pacyna, 1988; Taraškevičius and Zinkutė, 2003). Soil pollution by heavy metals (HMs) is one of the main problems that can cause major ecological problems (Adomaitis et al., 2003; Mikalajūnė and Jasulaitytė, 2011).

Heavy metals are not biodegradable, so they remain in the environment for a long time and are difficult to remove from it. Soil contaminated with HMs has a negative impact on the envi-

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ronment and people (Gregorauskienė, 2006; Jankauskaitė et al., 2007). The effect of As and HMs can be carcinogenic, mutagenic and teratogenic; an excess of these metals in the soil can cause endemic diseases, affect the growth and development of biological organisms, damage reproductive functions, and weaken immunity. An excess of HMs is detrimental to the entire ecosystem. Soil pollution with HMs can go unnoticed for a long time, and the results of their effects are difficult to predict in the future (Mažvila, 2001; Zvilnaitė and Tričys, 2009).

Around the world, there have been considerable research on the contamination of soil with HMs. Much research has looked at the quantity of HMs in soil and their possible damage to human health throughout this time. There are, however, few studies of soil HM content in different territories of Lithuania from different land use. Previous studies have found that the main cause of environmental pollution problems, particularly in living areas and technical areas, is thought to be anthropogenic sources of HMs pollution. Traffic emissions, industrial emissions (power plants, coal combustion, metallurgical industry, auto repair shops, chemical plants, etc.), domestic emissions, weathering of buildings and pavement surfaces, as well as particles deposited from the atmosphere are just a few examples of anthropogenic HM sources. In agricultural territory the main source of HM contamination is usage of chemical fertilizers, pesticides, herbicides and so on. Since soil is the main place where people are exposed to pesticides directly, soil contamination is a major worry, especially in areas with agricultural production (Gu et al., 2023).

This study examined concentrations, pollution levels and health hazards associated with five potentially toxic elements (As, Cd, Cu, Zn, and Cr) at five sampling sites of different land use in Lithuania. The study brings some new results, because pollution levels and health risks at these territories were not previously assessed. So, the objectives of this study are:

- to determine the concentration of As and HMs (Cd, Cu, Zn, and Cr) from five different land use areas;
- to determine the level of pollution using the geo-accumulation index, pollution co-efficient and contamination factor;
- assess the health risks (non-carcinogenic and carcinogenic) for humans with the help of the USEPA method.

MATERIALS AND METHOD

STUDY AREAS

One of the most densely populated districts in the city of Vilnius is the Šnipiškes microdistrict (LT) (Fig. 1). About 20 thousand people live there within an area of 3.1 km^2, the resulting density being 4,900 people/km^2. Šnipiškes has long been considered a comfortable and popular suburb. In recent years, this microdistrict is often called the new centre of the capital: many business centres with a tendency to expand are located here, the streets are constantly busy with traffic, and the infrastructure is expanding. There has been little research into the soils of the Šnipiškes microdistrict, and in particular there is a lack of reliable, comprehensive and detailed data on the soil pollution of the microdistrict with HMs. Samples were therefore collected to analyse HM and As pollution and assess the health risk of the residents. The current wave of industrialization, urbanization, and economic diversification has contaminated the environment's natural resources. The capital city (Vilnius) of Lithuania is currently a rapid growing city and a major commercial hub. Additionally, the number of automobile and metal works is growing quickly, and while in the city, a person can find a automobile parking area at a maximum distance of 3 km apart. This implies that different types of pollutants are released by vehicle parking area, whether they are large or small, when they carry out various operational tasks, potentially significantly negative impacting the environment (Olukanni and Adeoye, 2012; Ololade, 2014). In order to understand the effects on people and the environment, it is important to measure the degree of As and HM pollution in soil in vehicle parking area and automechanical workshops. In this study, we also collected soil samples from metal vehicle parking area of six different microdistricts located in Vilnius city such as Pashilaičiai, Karoliniškes, Pilaite, Zirnau, Rasu and a neighbouring parking area, regarded as technical territory (TT) (Fig. 1), to estimate HM pollution. Urban green parks or recreational areas now serve as a critical barometer for assessing both the urban levels of the surrounding metropolis and the quality of life of its citizens following the recent economic boom. Likewise, Vilnius's old town green park (GT) is considered as a significant recreational place for the residents of this city, and we collected samples to assess the pollution level of its soil.

Čepkeliai is the largest nature reserve in Lithuania, and is considered as natural territory (NT) (Fig. 1) for this research; it is located in the Varna District Municipality, in the southern part of Lithuania. The Kotra River, which runs along the Belarus-Lithuania border in this region, is north of Marcinkonys Village and south of Čepkeliai it. We choose Raseiniai-Betygala region as agricultural territory (AT) (Fig. 1) where land is used for crop cultivation, and we collected samples from contaminated sites.

Another study area (Raseiniai-Betygala region) is a sub-urban territory of Raseiniai district situated in the central part of Lithuania, covering an area of ~56 km^2 of mostly agricultural land, having a population of ~12,000, with an average annual rainfall of 660–668 mm. Very limited research has previously been conducted in this study area, as regards the concentrations and source of HMs, with none determining soil contamination levels in this region. This area comprises a cluster of local settlements, grazing land, farmland, abandoned industrial facilities and small wetlands; it is few kilometres north of the Kaunas-Klaipėda highway. Number of samples and sources of contamination in soil of five different land uses from five territories of Lithuania are given in Table 1.

SAMPLING PREPARATION

Sampling took place during summer to avoid snowfall and runoff, soil samples being taken from different depths (up to 0.3 m) using a stainless steel shovel, and placed in medium-size registered polyethylene bags to avoid cross-contamination and other damage in transport from site area to laboratory.

The steps of sample preparation for analysis were as follows:

- collected samples were taken to the laboratory and placed in petri dishes;
- samples were dried in a drying oven keeping temperature at (105 ±10°C) for 24 hours;
- in a porcelain mortar, dry samples were ground to a uniform mass;
- the sample's uniform mass was passed through a 2.00 mm filter, with sieving of the soil to reduce the particle size to <0.250 mm (or preferably to <0.125 mm to ensure separation of coarser from finer particles);

Fig. 1. Locations of the five different study areas in Lithuania

Table 1

Number of samples and sources of contamination in soil from five different territories of Lithuania

Territory	No. of samples	Source of contamination
Natural	72	natural
Green	30	anthropogenic
Living	103	anthropogenic
Agricultural	20	anthropogenic
Technical	72	anthropogenic

• the dry filtered mass was placed in XRF (X-ray fluorescence) sample cup, and inserted into a spectrometer for analysis;

• An X-ray fluorescence spectrometer (*Thermo Scientific Niton® XL2* series) was used to determine HM and As concentrations. In the 30 to 600 second measurement time range, the maximum 600-second analysis time interval was selected for optimum quality of the results.

Wet-chemical techniques like atomic absorption spectrometry (AAS), atomic emission spectrometry (ICP-AES or DCP-AES), or inductively coupled plasma mass spectrometry (ICP-MS) have frequently been employed for soil analyses. However, X-ray fluorescence (XRF) is a reliable, economical approach for the detection of a wide range of elements in soil. For the *in situ* detection of heavy metal ions in soils, portable (hand-held) XRF equipment have become widely employed in recent years (Valskys et al., 2022). The capability to evaluate a variety of metals is clearly a strength, despite the lesser sensitivities. The ability to analyse both solid and liquid samples offers significant sampling diversity as well.

GEO-ACCUMULATION INDEX

Muller's (1969) geo-accumulation index (I_{geo}), has become widely used to evaluate metal pollution in European trace metal investigations. By contrasting the current and background concentrations, I_{geo} allows evaluation the level of heavy metal pollution in soils. This equation is used to calculate I_{geo}:

$$I_{geo} \quad \log_2 \frac{C_n}{1.5B_n} \qquad [1]$$

where: C_n – metal's soil concentration, B_n – geological background value (n).

The correction value for the backdrop matrix resulting from atmospheric effects is 1.5. With the aid of the constant 1.5, we were able to examine normal variations in the amount of a given material present in the environment as well as very minute anthropogenic influences (Yaqin et al., 2008).

Seven categories are used to categorize the data based on the geo-accumulation index (Table 2). Higher values for class seven are unconstrained and may be hundreds of times greater than baseline values.

Table 2

Index of geo-accumulation

Class	Value	Pollution level
0	$I_{geo} \leq 0$	no contamination
1	$0 < I_{geo} < 1$	no contamination to moderate contamination
2	$1 < I_{geo} < 2$	moderate contamination
3	$2 < I_{geo} < 3$	moderate contamination to heavy contamination
4	$3 < I_{geo} < 4$	heavy contamination
5	$4 < I_{geo} < 5$	heavy to extreme contamination
6	$I_{geo} > 5$	extreme contamination

POLLUTION COEFFICIENT

To compare the concentration of the chemical substance in the soil with the maximum permissible concentration (MPC), the soil contamination coefficient K_0 is determined, which is equal to:

$$K_0 = C / MPC \quad [2]$$

where: C – concentration of the chemical substance in the soil sample analysed (mg/kg); MPC – the maximum permissible concentration of a chemical substance in soil (mg/kg).

Based on the results obtained, the degree of soil hazard is determined: permissible when $K_0 < 1$; medium risk when $1 < K_0 < 3$; dangerous when $3 < K_0 < 10$; very dangerous when $K_0 > 10$.

CONTAMINATION FACTOR (CF)

The CF is the result of dividing the mean concentration of each element in the soil by the background value (concentration in unpolluted soil) using equation [3]. Contamination levels can be grouped on a scale from 1 to 6 (Table 3; Muller, 1969).

Table 3

The intensity of the contamination levels categorized on a scale from 0 to 6

CF values	Pollution category, level
0	no pollution
1	zero to medium pollution
2	moderate pollution
3	moderate to strong pollution
4	strong pollution
5	strong to very strong pollution
6	extremely high pollution

RISK ASSESSMENT OF HUMAN HEALTH

DOSE OF EXPOSURE

Assessment of dose exposure is a useful tool for identifying risks to human health. Humans are typically exposed to pollution through three primary routes: ingestion, skin contact, and breathing (inhalation). The average daily dose (ADD) (mg/kg/day) of three exposure pathways (ingestion, skin contact, and inhalation) can be calculated by using equations [4–6].

$$ADD_{ing} \quad \frac{C \; R_{ing} \; CF \; ED \; EF}{AT \; BW} \quad [4]$$

$$ADD_{ing} \quad \frac{C \; R_{inh} \; EF \; ED}{AT \; BW \; PEF} \quad [5]$$

$$ADD_{derm} \quad \frac{C \; SA \; SAF \; DAF \; ED \; EF \; CF}{AT \; BW} \quad [6]$$

where: C is the element concentration in soil (mg/kg); the terms ADD_{ing} ADD_{inh} and ADD_{derm} stand for average daily dose of metals via ingesting (mg · kg^{-1}/day), inhalation (mg · kg^{-1}/day), and dermal inhalation (mg · kg^{-1}/day), respectively (Sonomdagva et al., 2019).

Table 4 includes a list of all exposure variables and values used to calculate intake values and risk.

Table 4

Parameters with reference value for the assessment of health risk

Parameters	Unit	Adult	Children	Reference
R_{ing}	mg/kg	100	200	USEPA (1989)
R_{inh}	m^3/day	12.8	7.63	Adimalla (2019b)
ED	years	30	6	Adimalla (2019b)
BW	kilogram	70	20	Adimalla (2018, 2019a)
AT	years	8760	2190	Adimalla (2019b)
EF	days/year	365	365	Adimalla (2019b)
CF	kg/mg	1×10^{-6}	1×10^{-6}	Adimalla (2019b)
SAF	mg/cm^3	0.7	0.2	Adimalla (2019b)
PEF	m^3/kg	1.36×10^9	1.36×10^9	Adimalla (2019b)
SA	cm^3	4350	1600	Adimalla (2019b)
DAF	–	0.001	0.001	Adimalla (2019b)

R_{ing} – rate of ingestion, R_{inh} – rate of inhalation, ED – duration of exposure, BW – body weight (average), AT – average time, EF – exposure frequency, CF – conversion factor, SAF – skin adherence factor, PEF – particle emission factor, SA – skin surface area (exposed), DAF – dermal adsorption factor

ASSESSMENT OF NON-CARCINOGENIC RISK

The hazard quotient (HQ), which is computed by dividing the daily dose by a certain reference dose (Rfd), is used to depict the non-carcinogenic risk after the calculation of ADD and the three exposure routes (RfD) (according to USEPA, RfD values vary depending on the element), as in equation [7]:

$$HQ \quad \frac{ADD}{Rfd} \qquad [7]$$

A value of HQ less than one indicates absence of adverse health effects; whereas, an HQ value greater than one means there is a probability of adverse health effects (USEPA, 1989). The total risk of non-carcinogenic elements for a particular element is represented by the hazard index (HI), which is the sum of HQ (Adimalla, 2019b).

$$HI \quad HQ(HQ_{ingestion} \quad HQ_{inhalation} \quad HQ_{dermal}) \qquad [8]$$

If the value of HI ≤1, it represents "no significant risk" of non-carcinogenic effects, whereas when HI >1, there is a possibility of non-carcinogenic adverse effects, and the probability increases with a rising value of HI (Sonomdagva et al., 2019). HI will be utilized in this research to evaluate the danger to human health from exposure to five HMs in the soil study areas.

ASSESSMENT OF CARCINOGENIC RISK

The lifetime risk of contracting cancer as a result of exposure to carcinogenic hazards is known as the carcinogenic risk (Chen et al., 2019; Zhaoyong et al., 2019). The following formulae can be used to determine a heavy metal's lifelong carcinogenic risks, via equations [9] and [10] (USEPA, 1989, 2002):

$$CR = ADD \cdot SF \qquad [9]$$

$$TCR = \Sigma\, CR = CR_{ing} + CR_{inh} + CR_{derm} \qquad [10]$$

where: CR and TCR stand for carcinogenic risk and total carcinogenic risk, respectively. Slope factor (SF) and reference dose (RfD) as listed in Table 5. CR and TCR values less than $1 \cdot 10^{-6}$ are considered as insignificant. On the other hand, CR and TCR values beyond $1 \cdot 10^{-4}$ are likely to be detrimental to human health (USEPA, 1989).

Table 5

Reference dose (Rfd) and slope factor (SF) for non-carcinogenic and carcinogenic metals, respectively

Elements	Reference dose (Rfd)			Slope factor (SF)		
	Ingestion	Inhalation	Dermal	Ingestion	Inhalation	Dermal
As	3.00E-04	1.23E-04	1.23E-04	1.50E+00	4.30E-03	3.66E+00
Cd	1.00E-03	1.00E-03	2.50E-05	6.30E+00	6.30E+00	6.30E+00
Cu	4.00E-02	4.00E-02	1.20E-02	–	–	–
Zn	3.00E-01	3.00E-01	6.00E-02	–	–	–
Cr	3.00E-03	2.86E-05	3.00E-03	5.01E-01	4.20E+01	2.00E+01

RESULTS

The geo-accumulation index, pollution coefficient, contamination factor (CF), enrichment factor (EF) and human health risk assessment (non-cancer and cancer risk) were used and calculated to show the results in this section. The exposure doses were computed using the heavy metal concentrations to determine the health risk assessment, and the non-carcinogenic risk was assessed for each HM examined, while carcinogenic risk was assessed for As, Cd and Cr.

CONCENTRATIONS OF HEAVY METALS

Tables 6–10, represent the concentrations of five elements (As, Cd, Cu, Zn and Cr), along with their background values, and maximum allowable limits for natural territory (NT), agricultural territory (AT), green recreational territory (GT), living territory (LT) and technical territory (TT), respectively.

In the surface soil samples of NT, the amounts of As, Cd, Cu, Zn and Cr are 98, 0, 15.5, 66 and 12.5%, respectively. Cd was not detected in natural territory (Table 6 and Fig. 2). A decreasing order was observed for the mean concentrations of HMs in the surface soils of NT, i.e. Zn > As > Cr > Cu > Cd. However, the HMs' coefficient of variation (CV%) in surface soils showed an ascending trend of Cd (0%) < As (29%) < Zn (65%) < Cr (276%) < Cu (787%). The CV value of Cu and Cr indicated large differences in concentrations of these HMs.

Table 6

Concentrations (mg/kg) of HMs in soil samples of NT

Metals	As	Cd	Cu	Zn	Cr
Minimum, mg/kg	0.00	0.00	0.00	0.00	0.00
Maximum, mg/kg	9.64	0	10.04	74.88	45.24
Mean, mg/kg	6.56	0	0.16	25.01	3.95
Standard deviation	1.95	0	1.27	16.38	10.92
CV%	29	0	787	65	276
MAC[1] (in soil), mg/kg	10	3	100	300	100
BC[2] (in soil), mg/kg	3.6	0.2	11	36	44

[1] – maximum allowable concentrations (Lithuanian Hygienic Norm HN 60: 2004);
[2] – background concentrations in soils (Lithuanian Hygienic Norm HN 60: 2004)

In the case of AT, surface soil contains 60% of As, 25% of Cd and 60% of Cu. On the other hand, Zn and Cr were found in almost all soil samples. The average concentration of As, Cd, Cu, Zn and Cr in the investigated area were 7.60, 3.50, 11.30, 40.60 and 44.30 mg/kg, respectively. Only the maximum value of Cd reached the MAC limit. Other metals' maximum value was below MAC (Table 7 and Fig. 2). A decreasing order was observed for the mean content of HMs in the surface soils of AT, i.e. Cr > Zn > As > Cu > Cd. This demonstrates clear changes in the levels of HMs in the study region's surface soils. However, the HMs' coefficients of variation (CV%) in surface soils showed an ascending trend of Cr (27.70%) < Zn (37.80%) < Cu (94.10%) < As (174.60%) < Cd (181.40%). The very large CV for As, Cd and Cu showed that there were significant differences in the concentrations of these elements between different sampling locations. Additionally, it was discovered that the CVs of Cr and Zn had values of >20 but <50%, suggesting a moderate degree of variation; however, the CVs of As, Cd, and Cu showed a large variation (Qing et al., 2015; Zhou et al., 2016).

In GT, As was identified in 34% of samples of the soil, however, Cd was not found in any samples. Cu was found in 90% of soil samples, Zn in all samples, whereas Cr in 30% of samples. Maximum values of HMs did not reach the MAC limit in the territory investigated (Table 8 and Fig. 2). A decreasing order was observed for the mean content of As and HMs in the surface soils

Table 7

Concentrations (mg/kg) of HMs in soil samples of AT

Metals	As	Cd	Cu	Zn	Cr
Minimum, mg/kg	0.00	0.00	0.00	0.00	21.90
Maximum, mg/kg	61.00	18.20	25.10	63.90	72.80
Mean, mg/kg	7.60	3.50	11.30	40.60	44.30
Standard deviation	13.30	6.40	10.60	15.30	12.30
CV%	174.6	181.4	94.10	37.80	27.70
MAC[1] (in soil), mg/kg	10	3	100	300	100
BC[2] (in soil), mg/kg	3.6	0.2	11	36	44

For explanations see Table 6

Table 8

Concentrations (mg/kg) of HMs in soil samples of GT

Metals	As	Cd	Cu	Zn	Cr
Minimum, mg/kg	0.00	0.00	0.00	35.88	0.00
Maximum, mg/kg	9.96	0.00	73.85	396.67	119.09
Mean	3.01	0.00	33.82	135.77	11.50
Standard deviation	4.08	0	19.78	80.18	24.37
CV%	135	0	58	59	211
MAC[1] (in soil), mg/kg	10	3	100	300	100
BC[2] (in soil), mg/kg	3.6	0.2	11	36	44

For explanations see Table 6

Table 9

Concentrations (mg/kg) of HMs in soil samples of LT

Metals	As	Cd	Cu	Zn	Cr
Minimum, mg/kg	0	0	0	36.13	0
Maximum, mg/kg	36.05	0	212.40	2853.86	217.03
Mean	2.95	0	41.30	287.05	20.85
Standard deviation	6.237	0	33.707	358.29	30.545
CV%	211	0	81.6	124	146
MAC[1] (in soil), mg/kg	10	3	100	300	100
BC[2] (in soil), mg/kg	3.6	0.2	11	36	44

For explanations see Table 6

Table 10

Concentrations (mg/kg) of HMs in soil samples of TT

Metals	As	Cd	Cu	Zn	Cr
Minimum, mg/kg	0	0	0	22.77	0
Maximum, mg/kg	10.9	11.5	2777.99	965.81	1135.07
Mean	1.44	0.75	69.34	205.9	97.82
Standard deviation	3.04	2.77	324.64	213.07	214.24
CV%	210	369	468	103	219
MAC[1] (in soil), mg/kg	10	3	100	300	100
BC[2] (in soil), mg/kg	3.6	0.2	11	36	44

For explanations see Table 6

of the research locations, i.e. Cr > Zn > As > Cu > Cd. As and the trend of CV was Cr (211%) > As (135%) > Zn (59%) > Cu (58%) > Cd (0%). Cr had large CV values indicating significant differences in concentrations at different sampling points of GT.

For LT, As found in samples only for 23.5%, while Cd was not found in any samples, whereas Cu and Cr were found in 77.5 and 47.5% of soil samples, respectively. Zn was found in all samples. Maximum values of As, Cu, Zn and Cr surpass the MAC limit 3.6, 2.1, 9.51 and 2.17 times, respectively. The decreasing order of mean content of HMs in LT was Zn > Cr > Cu > As > Cd. Except for Cd, the value of CVs of As, Zn, Cr and Cu represented a large variation and the order was As (211%) > Cr (146%) > Zn (124%) > Cu (81.6%) > Cd (0%).

In the case of TT, As was found in 12 samples out of 72, whereas Cu, Zn and Cr were found almost all samples. All elements reached the MAC limit of Lithuania (Table 10 and Fig. 2). The decreasing order of mean content of As and HMs observed in TT was Zn > Cu > Cr > As > Cd which is similar to LT. The trend of CV values found in TT was Cu (468%) > Cd (369%) > As (210%) > Cr (219%) > Zn (103%).

ESTIMATION OF LEVEL OF CONCENTRATION

GEO-ACCUMULATION INDEX AND POLLUTION CO-EFFICIENT

I_{geo} values of elements for different territories of Lithuania showed the level of soil contamination. As shown in Table 11, I_{geo} values for As represent no contamination to moderate con-

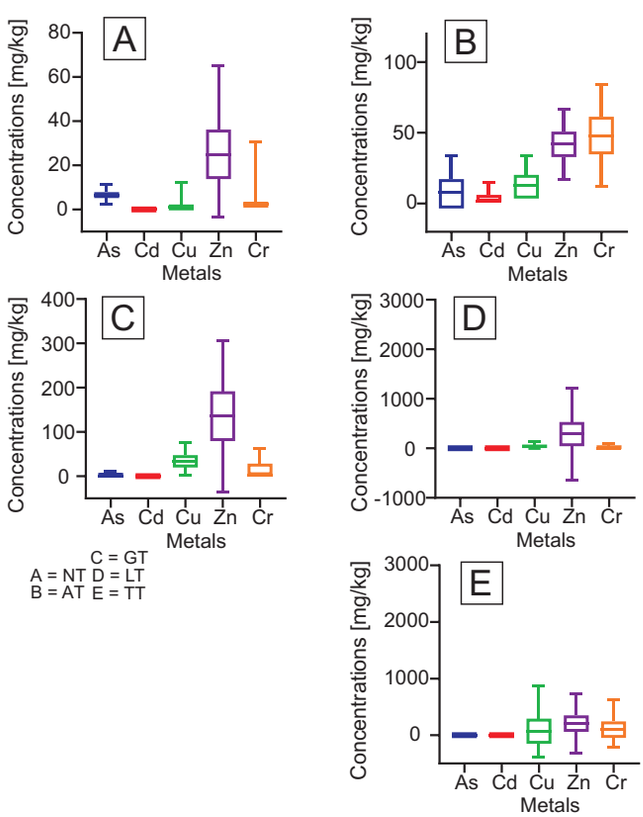

Fig. 2. Boxplot of the distribution of HMs in different territories of Lithuania

Table 11

I_{geo} estimation in the soil of different territories of Lithuania

	LT	GT	AT	NT	TT
As	0.28	0.29	0.51	0.50	0.09
Cd	0.00	0.00	3.54	0.00	–1.38
Cu	0.34	0.30	–0.54	–0.16	0.37
Zn	0.15	0.13	–0.41	0.08	0.14
Cr	0.07	0.05	–0.57	0.03	0.10

tamination in all territories and there is no Cd contamination in the various territories except for AT (heavily contaminated).

I_{geo} values for Cu showed no contamination to moderate contamination for LT, GT and TT, and no pollution for NT and AT. There is no pollution with Zn and Cr for AT. On the other hand, I_{geo} values for Zn and Cu represent no contamination to moderate contamination for the rest of the territories (Table 11 and Fig. 3).

Among the elements detected, only Cd in AT is attributed as medium risk ($1< K_0 < 3$), while other elements showed the permissible category ($K_0 < 1$) (Table 12).

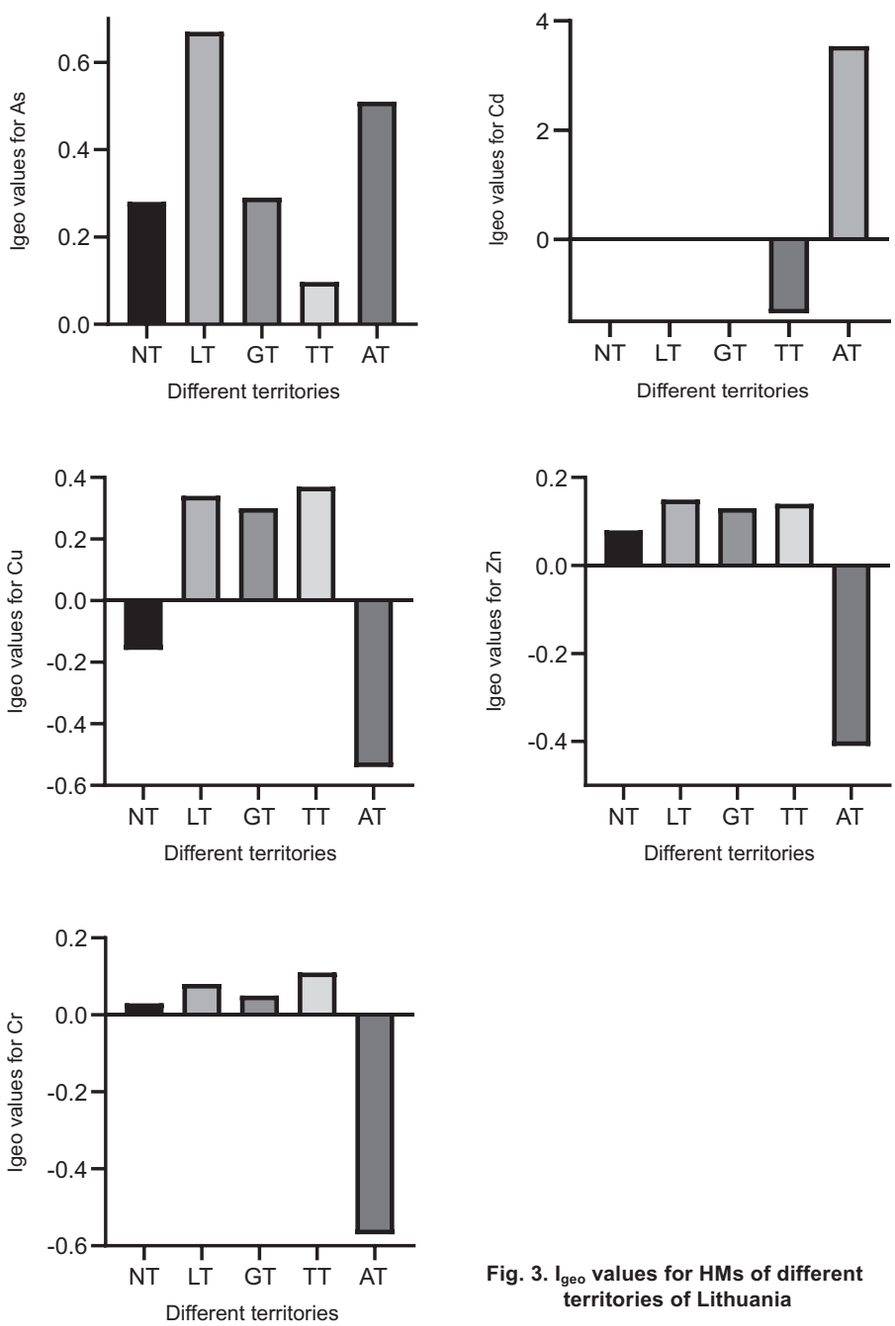

Fig. 3. I_{geo} values for HMs of different territories of Lithuania

Table 12

Estimation of pollution coefficient (K_0)

	NT	AT	GT	LT	TT
As	0.65	0.76	0.30	0.29	0.14
Cd	0	1.16	0	0	0.25
Cu	0.001	0.11	0.33	0.41	0.68
Zn	0.08	0.13	0.45	0.95	0.67

CALCULATION OF CF

CF values of As in different territories were in the following order TT (0.40) < LT (0.81) < GT (0.83) < NT (1.82) < AT (2.11). GT, LT and TT showed no pollution, whereas AT and NT showed moderate and moderate to strong levels of pollution, respectively. In the case of Cd, AT and TT showed extremely high and moderate to strong levels of contamination, respectively. However, no pollution of Cd was identified in NT, GT and LT. For Cu, CF values for different territories were in the order: NT (no pollution) < AT (zero to medium) < GT (moderate to strong pollution) < LT (moderate to strong pollution) < TT (extremely high pollution). For Zn, NT (no pollution) < AT (zero to medium) < GT (moderate to strong) < TT (strong to very strong) < LT (extremely high). For Cr, the CF values for NT, GT and LT showed no pollution, whereas AT and TT showed zero to medium and moderate levels of pollution, respectively (Tables 3 and 13).

According to the CF values, extremely high pollution were identified in LT (7.97) and TT (5.71) with Zn, and with Cu in TT (6.30). Extremely high contamination also found in AT with Cd (17.5). Moderate to strong contamination by Cd, Cu and Zn was recorded in TT, LT and GT, respectively. The following order reflected the CF value of As in various territories: TT < LT < GT < NT < AT. In the case of Cd, the contamination levels for AT and TT were extremely high and moderate to strong, respectively. However, no Cd contamination was found in NT, GT and LT. Cu contamination levels in different territories were in the order NT < AT < GT < LT < TT, whereas for Zn the order was NT < AT < GT < LT < TT. CF values for Cr contamination did not show any pollution for NT, GT and LT, while pollution levels on AT and TT were negligible to moderate and medium respectively.

CORRELATION MATRIX

A strong positive correlation was observed for Cu-Cr (r = 0.56) and Cr-Zn (r = 0.67) in the soil of TT (Fig. 4). This strong correlation suggests that the HMs in the soil have a common source/origin. This indicated a mutual concentration dependence of the metals in the soil system under investigation. In the case of GT, Cu shows weak correlation with Zn (r = 0.32) (Fig. 4). For LT, Cu shows strong relation with Zn (r = 0.52) (Fig. 4); however, the HMs in AT show very weak correlation between each other (Fig. 4). Natural territory also showed very weak correlation within metals (Fig. 4).

Table 13

Estimation of CF of As, Cd, Cu, Zn and Cr in soil of five different territories

	CF_{As}	CF_{Cd}	CF_{Cu}	CF_{Zn}	CF_{Cr}
NT	1.82	0	0.01	0.70	0.08
AT	2.11	17.5	1.02	1.12	1.00
GT	0.83	0	3.07	3.77	0.26
LT	0.81	0	3.75	7.97	0.47
TT	0.40	3.75	6.30	5.71	2.22

HUMAN HEALTH RISK ASSESSMENT

NON-CARCINOGENIC RISK

Table 14 suggests that the HQ values for adults and children via different exposure routes in different study areas were found below one (HQ <1), indicating negligible non-carcinogenic risks in the study areas. The highest health risk (highest HQ values) related to the As content was observed in LT (42.0E-02 children and 7.50E-02 adult), where the risk values were closest to permissible ones, and the lowest health risk was observed in TT (1.96E-06 children and 1.96E-06 adult) (Table 14). The highest HQ for children, associated with the presence of Cd, was recorded in AT (3.50E-02 children, 6.23E-03 adult), and the lowest in NT, GT and LT (HQ = 0).

The highest health risk related to the Cu content was observed in GT (2.00E-02 children) and TT (3.07E-03 adults), where the risk values were closest to permissible ones, and the lowest health risk was observed in NT (6.72E-10 adult, 1.12E-09 children) (Table 14). In the case of Zn, the highest value of non-carcinogenic risk was determined in LT for children (9.56E-03), and the lowest in NT (2.33E-08) and for adults were highest in LT (1.70E-03) and lowest in AT (2.27E-08). Highest and lowest HQ values for Cr were found in TT (32.0E-02 children) and NT (2.10E-05 children), and for adults were highest in TT (58.0E-03) and lowest in AT (8.00E-06) (Table 14).

CARCINOGENIC HEALTH RISK

Carcinogenic substances such as As, Cd, and Cr may increase the risk of cancer in populations that are exposed to them. TCR and CR levels below 1.0E-06 should be regarded as minimal, whilst those exceeding 1.0E-04 are probably detrimental to individuals (USEPA, 1989, 2002).

In the case of NT, the carcinogenic risks associated with As, Cd and Cr in the soil were at an acceptable level (<1.0E-04) (Table 15). For AT, the carcinogenic risk values for As (1.12E-04 children), Cd (2.20E-04 children) and Cr (2.35E-04 children) all surpassed the tolerance level when ingested, indicating that these substances pose a serious threat to children's health when consumed. For GT, carcinogenic values for Cr (1.02 E+00 adult) exceeded the threshold limit, and therefore posed a health risk to adults. In LT, children were under carcinogenic health risk since the values of As (1.89E-04) and Cr (2.28E-04) exceeded the safe level. For TT, both adults and children were under the threat of carcinogenic health risk, as the carcinogenic values for Cr (1.93E-04 adult and 5.21E-04 children) exceeded the acceptable limit. To sum up, it can be said that the assessment of health hazards brought on by HM soil pollution will be useful in deciding how to manage soil quality.

DISCUSSION

HM contamination in soil of five different land uses from five different territories in Lithuania are discussed in this study. For

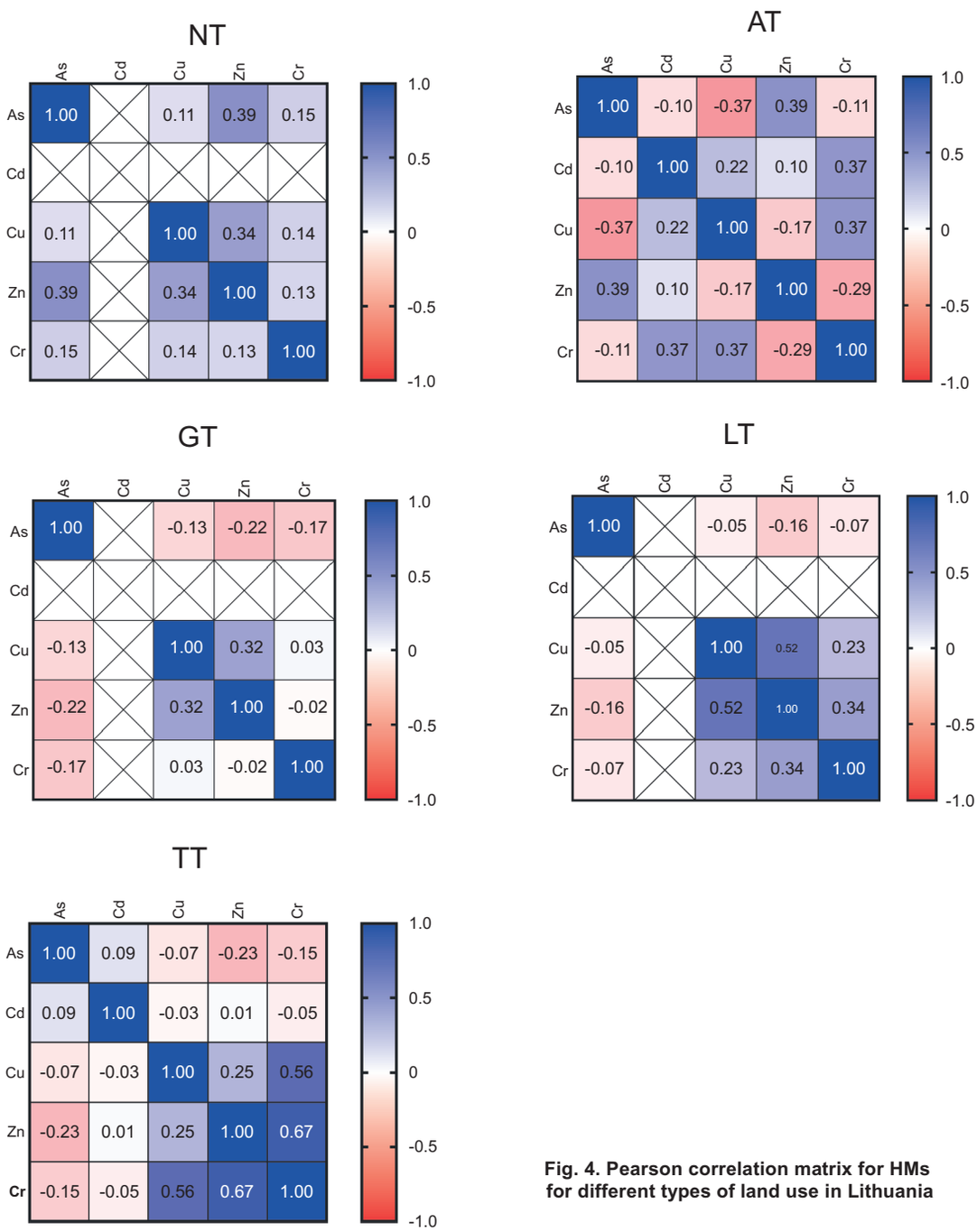

Fig. 4. Pearson correlation matrix for HMs for different types of land use in Lithuania

territories such as LT and TT, the average concentration of the elements declined in the following order Zn > Cr > Cu > As > Cd, whereas for GT, NT and AT, the orders were Zn > Cu > Cr > As > Cd, Zn > As > Cr > Cu > Cd and Cr > Zn > Cu > As > Cd, respectively. Mean concentration were higher than background values in Lithuania. Similarities were observed in Polish soil, though in terms of urban soil, the mean content of Cd, Zn, Cu were higher than the background values of the Polish soils (Skorbiłowicz et al., 2021). In Hungary, on average, the HM concentrations were below the pollution limit values in urban green spaces (Tóth et al., 2023), whereas in green territories of Lithuania Cu and Zn were found above the BC limit values. Living territories such as cities all across the world differ from one another in terms of population density and rate of urbanization. The various HM sources are widespread in both urban and industrial settings. Each city's natural characteristics affect the distributions and concentrations of HMs. Most results indicate that the levels of HMs exceeded the upper limit. Concentrations of HMs in different urban living areas around the world are given in Table 16.

Zn (287.05 mg/kg) concentration was detected as highest in LT compared to other territories, being designated as populated residential area in Lithuania, and lowest in NT. As concentrations were lowest in TT, these being mostly used in metal alloy production, vehicle repair and parking lots, and highest in AT.

The total amount of Cd in a soil is made up of inputs from external sources, the majority of which are human in nature, as

Table 14

Results of HQ and HI of each element for estimation of non-carcinogenic risk

	HQ_{ing}		HQ_{inh}		HQ_{derm}		HI	
	Natural territory							
HMs	Adults	Children	Adults	Children	Adults	Children	Adults	Children
As	3.90E-02	21.0E-02	8.94E-06	1.50E-05	2.89E-03	8.53E-04	4.18E-02	2.10E-01
Cd	0	0	0	0	0	0	0	0
Cu	7.12E-06	4.00E-05	6.72E-10	1.12E-09	7.24E-07	2.13E-07	7.84E-06	4.00E-04
Zn	1.48E-04	8.33E-04	1.40E-08	2.33E-08	2.26E-05	6.67E-06	1.70E-04	8.39E-04
Cr	2.34E-03	1.00E-02	2.32E-05	3.88E-05	7.13E-05	2.10E-05	2.43E-03	1.00E-02
	Agricultural territory							
As	4.50E-02	2.50E-01	1.03E-05	1.72E-05	3.34E-03	9.83E-04	4.83E-02	2.51E-02
Cd	6.23E-03	3.50E-02	5.88E-07	9.80E-07	7.60E-03	2.24E-03	1.38E-02	3.72E-02
Cu	5.02E-04	2.82E-03	4.72E-08	7.90E-08	5.10E-05	1.50E-05	5.53E-04	2.83E-03
Zn	2.40E-04	1.35E-03	2.27E-08	3.76E-08	3.66E-05	1.08E-05	2.76E-04	1.36E-03
Cr	2.63E-02	1.47E-01	2.60E-04	4.33E-04	8.00E-06	2.36E-04	2.65E-02	1.47E-02
	Green recreational territory							
As	18.0E-03	100E-03	4.10E-06	6.85E-06	1.33E-03	3.91E-04	1.93E-02	100E-03
Cd	0	0	0	0	0	0	0	0
Cu	1.50E-03	8.45E-03	1.42E-07	2.37E-07	1.53E-04	2.00E-02	1.65E-03	2.84E-02
Zn	8.03E-04	4.50E-03	7.60E-08	1.27E-07	1.23E-04	3.62E-05	9.26E-04	4.53E-03
Cr	6.80E-03	38.0E-03	6.75E-05	1.13E-04	2.08E-04	6.13E-05	7.07E-03	3.81E-02
	Living territory							
As	7.50E-02	42.0E-02	1.71E-05	2.89E-05	5.59E-03	1.73E-03	8.06E-02	4.21E-01
Cd	0	0	0	0	0	0	0	0
Cu	2.30E-03	1.20E-02	2.16E-07	3.60E-07	2.32E-04	7.21E-05	2.53E-03	1.20E-02
Zn	1.70E-03	9.56E-03	1.61E-07	2.68E-07	2.58E-04	8.03E-05	1.95E-03	9.64E-03
Cr	2.50E-02	14.3E-02	2.52E-04	4.19E-04	7.77E-04	2.40E-04	2.60E-02	1.43E-01
	Technical territory							
As	8.53E-03	48.0E-03	1.96E-06	3.27E-06	6.34E-04	1.86E-04	9.16E-03	4.81E-02
Cd	1.33E-03	7.50E-03	1.26E-07	2.10E-07	1.62E-03	4.80E-04	2.95E-03	7.98E-03
Cu	3.07E-03	17.0E-03	2.90E-07	4.85E-07	3.14E-04	9.16E-05	3.38E-03	1.70E-02
Zn	1.22E-03	6.83E-03	1.15E-07	1.92E-07	1.85E-04	5.48E-05	1.40E-03	6.88E-03
Cr	58.0E-03	32.0E-02	5.73E-04	9.58E-04	1.77E-03	5.20E-04	6.03E-02	3.21E-01

HQ_{ing} – hazard quotient-ingestion; HQ_{inh} – hazard quotient-inhalation; HQ_{derm} – hazard quotient-dermal contact

well as contributions from the parent geological material. The levels of Cd that environmental pollution causes to build up in soil will depend on the volume of emissions from each source, the movement of the metal from the source to the site, and the retention of the metal once it has entered the soil. In this study the highest mean value of Cd (3.50 mg/kg) was found in AT: since AT is mostly used for agricultural purposes, various chemical fertilizers, pesticides, herbicides, raw water were used in the soil for better harvesting of crops, which could raise the amount of Cd and associated HMs. The potential for mobilization of Cd is the largest when compared to other HMs, which means faster release from soil into groundwater than for other HMs that pose a health risk to humans.

The various sources of Cu in the environment are industrial Cu use, pesticides, vehicle fluids leakage and dumping, vehicle brake pads and architectural and marine antifouling coatings of Cu *etc*. The highest average content of Cu (69.34 mg/kg) was discovered in TT. Elevated amounts of Cu (41.30 mg/kg) were also observed in LT. It is assumed that construction, usage of various vehicle lubricants, welding materials, metal-containing petroleum, fuel content, battery and metallurgical sources may be responsible for this Cu pollution.

Agricultural material, sewage sludge, municipality discharge, metallurgical and industrial waste are common sources of Cr pollution. The highest mean amount of Cr (97.82 mg/kg) was detected in TT. Considerable amounts of Cr were also observed in the AT soils. All of the elements studied showed values that were significantly below their corresponding MAC values. This MAC was developed after extensive geochemical research and consideration of every type of soil in Lithuania; it likely can be used as a generic benchmark for assessing contamination.

In the case of NT, the maximum value of Zn and Cr surpassed the background limit but did not exceed the maximum allowable concentrations. Therefore, no environmental hazads were detected in this region. For AT, the analysed concentra-

Table 15

Estimation of CR and TCR for adults and children via three different routes of exposure

	Adults				Children			
Carcinogenic	CR_{ing}	CR_{inh}	CR_{derm}	TCR	CR_{ing}	CR_{inh}	CR_{derm}	TCR
	Natural territory							
As	1.75E-05	4.73E-12	1.30E-06	1.88E-05	9.84E-05	7.91E-12	3.84E-07	9.87E-05
Cd	0	0	0	0	0	0	0	0
Cr	3.52E-06	2.78E-08	4.28E-06	7.82E-06	1.97E-05	4.66E-08	1.26E-06	2.10E-05
	Agricultural territory							
As	2.02E-05	5.46E-12	1.50E-06	2.17E-05	1.12E-04	9.11E-12	4.42E-07	1.12E-04
Cd	3.92E-05	3.70E-09	1.20E-06	4.04E-05	2.20E-04	6.17E-09	3.52E-07	2.20E-04
Cr	3.94E-05	3.12E-07	4.80E-07	4.01E-05	2.21E-04	5.20E-07	1.41E-06	2.35E-04
	Green recreational territory							
As	8.02E-06	2.17E-12	5.96E-07	8.61E-06	4.51E-05	3.62E-12	1.76E-07	4.52E-05
Cd	0	0	0	0	0	0	0	0
Cr	1.02E+00	8.10E-08	1.24E-05	1.02+00	5.76E-05	1.35E-07	3.6E-06	6.13E-05
	Living territory							
As	3.37E-05	9.03E-12	2.51E-06	3.62E-05	1.89E-04	1.52E-11	7.79E-07	1.89E-04
Cd	0	0	0	0	0	0	0	0
Cr	3.82E-05	3.02E-07	4.66E-05	8.51E-05	2.14E-04	5.04E-07	1.44E-05	2.28E-04
	Technical territory							
As	3.84E-06	1.04E-12	2.85E-07	4.12E-06	2.16E-05	1.73E-12	8.41E-08	2.16E-05
Cd	8.37E-06	7.93E-10	2.56E-07	8.62E-06	4.72E-05	1.32E-09	7.56E-08	4.72E-05
Cr	8.71E-05	6.88E-07	1.06E-04	1.93E-04	4.89E-04	1.15E-06	3.12E-05	5.21E-04

CR_{ing} – carcinogenic risk-ingestion, CR_{inh} – carcinogenic risk-inhalation, CR_{derm} – carcinogenic risk-dermal

Table 16

HM concentrations (mg/kg) in urban soil of different cities around the world

Country	City	Cd	Cu	Zn	Cr	References
United Kingdom	Birmingham	1.62	466.9	534	NC*	Sezgin et al. (2004)
India	Gorimedu	6.54	202.24	222.46	0.85	Khan and Kathi (2014)
Greece	Kavala	0.2	124	272	196	Christoforidis and Stamatis (2009)
Mexico	Tijuana	0.1	50.2	NC*	17.1	Quińonez-Plaza et al. (2017)
South Korea	Ulsan	1.5	148	NC*	NC*	Duong and Lee (2011)

*NC – not counted

tions of As, Cd, Zn, Cu and Cr exceeded background concentrations; moreover, maximum concentrations of As and Cd approached the maximum allowable concentrations, and so there may be a serious hazard to the soil ecosystem and even to human health. In GT, the maximum concentrations of Zn and Cr exceeded the MAC, although average concentrations of Cu and Zn approached the BC limit, which may cause a threat to the environment. In the soil of LT, the average content of As, Cu, Zn approached to the BC limit; however, the maximum amount of As, Cu, Zn and Cr surpassed the MAC limit which may cause serious detrimental effects to the environment. The maximum concentrations of As, Cd, Cu, Zn and Cd exceeded the MAC limit manifolds which may pose a serious threat to the surroundings as well as to humans.

The level of soil pollution was indicated by the I_{geo} values of the elements for various Lithuanian regions. Since these I_{geo} values were mostly in the 0–1 range, this indicates Class 1 and no contamination to moderate contamination. I_{geo} values for As represent no pollution to moderate contamination across all areas, while there is no Cd contamination in any other region than AT (heavily contaminated). For LT, GT, and TT, I_{geo} readings for Cu showed no pollution to mild pollution, but there was no pollution for NT and AT. For AT, there is no contamination from Zn or Cr. I_{geo} readings for Zn and Cu, on the other hand, represent no pollution to moderate contamination for the remaining territories.

According to the CF values, extremely high pollution was identified in LT and TT with Zn, and with Cu in TT. Extreme contamination also found in AT with Cd. Moderate to strong contamination by Cd, Cu and Zn were recorded in TT, LT and GT, respectively. The following order reflected the CF value of As in various territories: TT < LT < GT < NT < AT. In the case of Cd, the contamination levels for AT and TT were extremely high and moderate to strong, respectively. However, no Cd contamination was found in NT, GT and LT. The Cu contamination levels in different territories were in the order NT < AT < GT < LT < TT whereas, for Zn the order was NT < AT < GT < LT < TT. CF val-

ues for Cr contamination did not show any pollution for NT, GT and LT, while pollution levels on AT and TT were negligible to moderate and medium, respectively.

CONCLUSIONS

Determination was made of the concentration of potentially toxic elements such as of arsenic, chromium, cadmium, copper and zinc, in the soil of five different kinds of land use in Lithuania. The I_{geo}, level of HMs contamination measured for five different land use from five territories in Lithuania showed that no Cd contamination in any study area other than AT (heavily contaminated). I_{geo} readings for As showed no pollution to moderate contamination across all study areas. I_{geo} measurements for Cu for LT, GT, and TT revealed no contamination to minor pollution. In the soil of TT, there was a significant positive correlation for both Cu-Cr (r = 0.56) and Cr-Zn (r = 0.67). Furthermore, Cu and Zn show a strong relationship with LT (r = 0.52). This study also assessed the human health risk caused by As and HMs. The carcinogenic risk of As, Cd, and Cr in the soil were at an acceptable level (<1.0E-04) in the case of NT. In the case of AT, the carcinogenic risk values for As, Cd and Cr all exceeded the tolerance level when ingested, showing that these compounds pose a substantial risk to childrens' health when consumed. Adults were at risk of developing cancer due to GT's carcinogenic values for Cr, which were above the threshold level. Children in LT were at risk of developing cancer because the values of As and Cr were higher than the acceptable limit. Because the carcinogenic values for Cr were higher than the allowable limit for TT, both adults and children were at carcinogenic health risk. In conclusion, it can be said that determining how to maintain soil quality will benefit from an evaluation of health risks caused by HM soil contamination.

The findings of this research can be utilized to decrease the presence of HMs and As in the soil among five different land use from different territories of Lithuania and enhance the quality of soil, which is crucial for the well-being of both adults and children.

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