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VARIATION OF ACCUMULATING ASSOCIATIONS IN TOPSOIL OF THE OLDEST PART OF VILNIUS

ĶĪMISKO ELEMENTU ASOCIĀCIJU AKUMULĒŠANAS AUGSNĒ VARIĀCIJAS VIĻŅAS VECĀKAJĀ DAĻĀ

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Abstract. *The aim of research was to analyse and explain the variation of accumulating associations of 13 chemical elements Ag, B, Ba, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sn, V, Zn in topsoil of the oldest part of Vilnius. These elements characterise soil quality and are included in the check list of hygienic norm HN 60-2004 of Lithuania. According to adjusted accumulating associations ordinary and unusual associations have been distinguished and distribution of the types or unusual associations has been analysed. They are often near present or former industrial sites, railway or streets with intensive traffic.*

Keywords: *accumulating associations, heavy metals, unusual associations, urban centre topsoil.*

Introduction

Heavy metals and other potentially hazardous chemical elements are usual objects of investigation in different environmental components, especially in urban territories, which are characterised by multiple sources of these contaminants. As a rule, urban topsoil is the main sampling media to reveal anthropogenic geochemical anomalies. On one hand, this is because it is able to accumulate contaminants and they are better revealed [1], on the other hand, this is because urban topsoil is related to different routes of heavy metal exposure [2].

Topsoil geochemical mapping in Vilnius in the XXth century was done several times: 1985–1988 (scale 1: 50000–75000), 1991–1993 (scale 1:25000–50000), 1995–1999 (central districts, scale 1:10000) [3]. Mapping was continued in XXIst century: sampling of peripheral districts was in 2002 (scale 1:100000) [4, 5, 6] and central districts in 2006 [7]. Comparison of topsoil quality in four urban land use groups of four Lithuanian towns revealed the highest contamination level in Vilnius, especially in its industrial-infrastructural zones [4].

Trace element contents in topsoil depend on many factors, both natural and anthropogenic. Basing on 2006 sampling in 4 central districts of Vilnius, it has been determined that 3 main groups of urban land-use – infrastructural-industrial, residential or public-residential and recreational – mostly differ in topsoil contamination and that in residential and public-residential sites the main variability is related to the time-span of urbanisation [7].

Topsoil from urban centres, which are usually characterised by longer time span of urbanisation, are especially important for investigation. Chen et al. noticed higher contamination of soil in urban parks in the densely populated historic center districts and explain this by the highest road density and long duration of traffic pollution [8]. Geochemical mapping in different Lithuanian towns including Vilnius also confirmed higher topsoil contamination in their central parts [4]. Besides, in central districts of Vilnius the absolute growth of topsoil total contamination index from 1985 to 2002 was higher than in peripheral [5]. However, contamination level of various central districts of Vilnius also differs: older Senamiestis and Naujamiestis are much more contaminated than younger Žvėrynas, Šnipiškės, Antakalnis and Žirmūnai [3].

For interpretation of the reasons of topsoil contamination trace element associations are very useful. Both accumulating and paragenetic associations of different industrial enterprises were analysed in detail in Panevėžys [9] basing on sampling results in 1995 [10], also in industrial part of Žirmūnai district in Vilnius [11]. However, presently most of the enterprises in urban territories are rather related to trade and marketing, so traffic actually is the main factor responsible for topsoil pollution. With the help of statistical methods the following main traffic and road construction related and soil accumulated elements have been distinguished near the highway Vilnius-Kaunas: Pb, Zn, Cu, Sn, Mo and Sr [12]. Most of them (Pb, Zn, Cu, Sn) are related to non-ferrous metals. Therefore in different land-use sites of the four younger central districts of Vilnius the generalised additive contamination index calculated according to concentration coefficients of Ag, Cu, Pb, Sn, Zn is higher than analogous index calculated according to concentration coefficients of Mo, Ni, Cr, Co, V [7]. Increase of Naujamiestis topsoil contamination in 1997 compared to 1992 was also mainly to the same group of elements [13]. However, topsoil in the old central districts of Vilnius can still contain the contaminants from the former industrial enterprises, which were especially abundant in Naujamiestis [12], to lower extent also in Senamiestis [3] and which were quite close to residential districts. Therefore it might be supposed that topsoil contamination in these districts is caused not only by traffic, but also by present or former industry. Analysis of accumulating associations can help to interprete the main reasons of topsoil contamination.

Therefore the aim of this research was to analyse and explain the variation of accumulating associations of 13 chemical elements Ag, B, Ba, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sn, V, Zn in topsoil of the oldest part of Vilnius – Senamiestis and Naujamiestis districts. These elements characterise soil quality and potential risk to human health and environment and are included in the check list of hygienic norm HN 60-2004 of Lithuania, which is based on their real total contents [14].

Methods

Sampling was done in 2006 in the oldest part of Vilnius, on the left bank of the Neris River. The methodology of sampling was aimed at reducing the number of samples and making them more composite to characterise each sampling site in general. In total, there were 59 sampling sites, 30 of them in Senamiestis district and 29 – in Naujamiestis district (Fig. 1). They were from 3 land-use groups. Each bulk sample of about 8-10 kg was gathered by zigzag crossing the site area and collecting 20-25 similar mass increments from the upper soil layer (0-10 cm).

Before further treatment sampled material was homogenised and composite sample reduced to about 1 kg. Total contents of Ag, B, Ba, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sn, V, Zn were determined in fraction <1 mm by optical atomic emission spectrophotometry by spectrophotometer DFS13.

In each site the concentration coefficients K_k were calculated by dividing the element contents by the same background values, which were used for 4 younger central districts [7]. They are given in parentheses in mg/kg: Ag(0.08), B(26.1), Ba(400), Co(4.8), Cr(32.9), Cu(8.8), Mn(578), Mo(0.71), Ni(12.3), Pb(16), Sn(2.07), V(29.7), Zn(30.9). After this the values of topsoil additive contamination index Z_d were calculated according to above-mentioned 13 elements as well as their maximum coefficient of danger K_o and general evaluation of soil quality in each site was obtained basing on Lithuanian hygienic norm [14] by choosing more dangerous of two categories: either according to Z_d, or according to K_o. To interprete the reasons of contamination, *point accumulating associations* [9] were determined, which included elements with $K_k>2$ listed in descending order of K_k values. To avoid some influence of the background variation on the territory on these associations, they were adjusted according to the intervals of accumulation of elements by separating them by dashes into groups. The following K_k intervals indicating the level of accumulation were used: 2-4 – low (L), 4-8 – medium (M), 8-16 – high (H) and >16 – extremely high (E). To analyse lateral variation of adjusted accumulating associations AAA, their types were distinguished taking into account the following principles: 1) the elements in AAA, 2) the element with maximum K_k, 3) the interval, to which the element with maximum K_k belongs, i.e. *level of association*. The regularities of the distribution of AAA types were analysed in three main urban land-use groups and two districts of the study area.

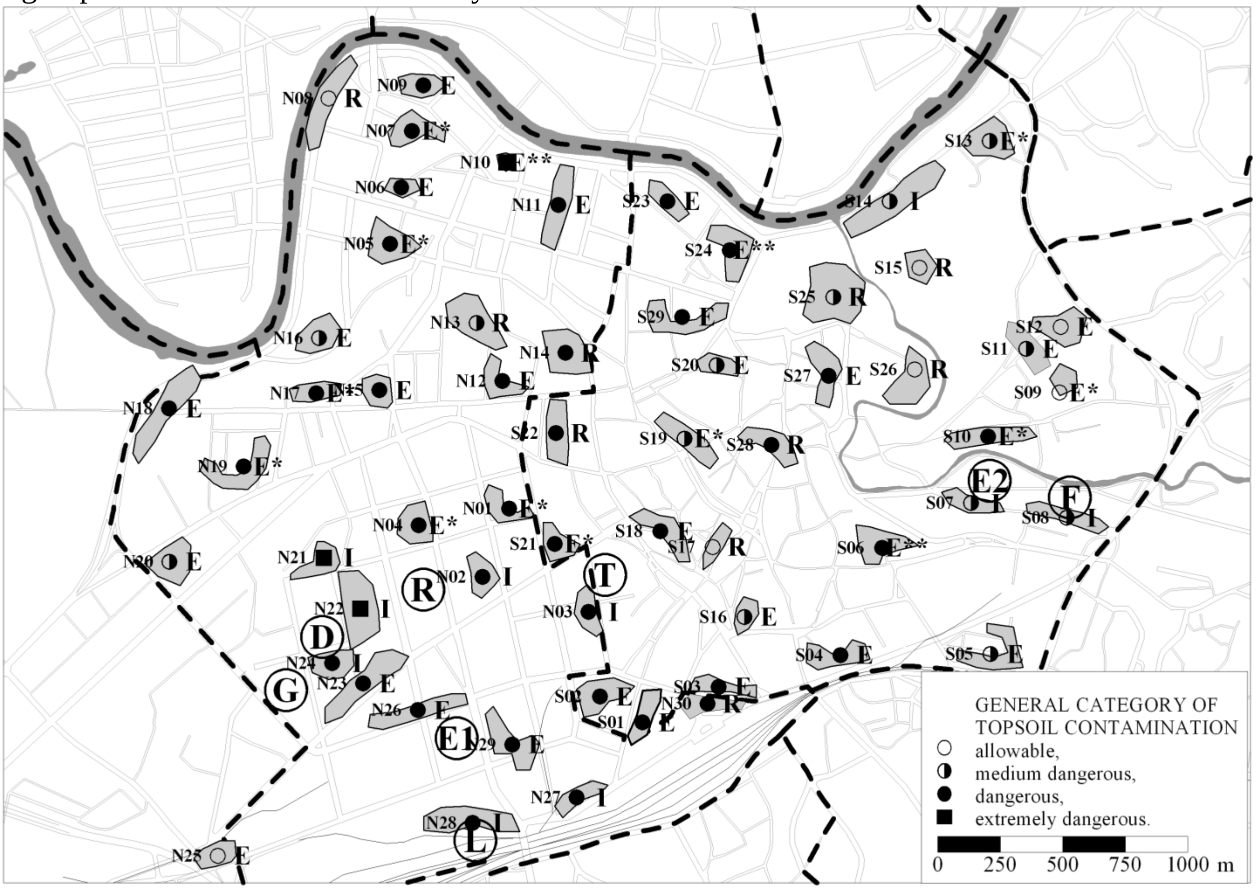

Fig.1. **Topsoil sampling sites, their land-use and general contamination category**

Notes. Land-use code of site and number of sites in parentheses: I (10) – industrial-infrastructural, i. e. near present or former industrial enterprise, railway or main traffic road, E (39) – residential or public-residential (E* – includes educational institution, E** – includes health care institution), R (10) – recreational. Industrial enterprises (in circles): D – drill plant, G – former grinding machine plant, R – former radio engineering works, T – former turning lathe plant, L – locomotive depot, E1, E2 – former electrical engineering works, F – fur plant. Sites from Naujamiestis are indicated by N and number and from Senamiestis – by S and number.

Results and discussion

Allowable category according to K_o is in 48.3% sites of Naujamiestis and 30.5% sites of Senamiestis. The percentages of this category according to Z_d in Naujamiestis and Senamiestis are lower: 6.7% and 17.2%, respectively. General category of soil quality is predetermined by Z_d. Naujamiestis is characterised even by extremely dangerously contaminated sites. The percentages of sites with dangerous and medium dangerous contamination in this district are also higher than respective percentages in Senamiestis district. Unlike younger districts, medium dangerous or dangerous contamination level is not only near the present or former industrial sites, railway or streets with intensive traffic, but also in public-residential and even recreational sites (Fig. 1). Median value of additive contamination index $\mathbf{Z_d}$ calculated according to above-mentioned elements belongs to the dangerous category.

The accumulating association of the whole study area is the following: Zn(9.42)>Ag(9.17)>Cu(5.15)>Pb(4.69)>Sn(3.77)>Mo(1.73)>Ni(1.35)>Co(1.34)>V(1.31).
It indicates much higher input to $\boldsymbol{Z_d}$ of elements related to non-ferrous metals (Zn, Cu, Pb, Sn, Ag) compared to respective input of elements related to ferrous metals (Mo, Ni, Cr, V, Co). Therefore the elements related to non-ferrous metals are usual members of AAA. According to percentage of sites, where the elements belong to AAA, they can be arranged in the following sequence: Ag(100%)> Zn(98.3%)> Pb(96.6%)> Cu, Sn(91.5%)> Mo(37.3)> Ni(6.8%)> Cr(5.1%)> V(5.1%)> Co(3.4%)> Ba(1.7%). It is obvious that Ag and Zn are the most widely spread contaminants. This is in agreement with investigations in Russia, which have shown that these two elements are the first indicators of the beginning of urbanisation [15].
The value of $\boldsymbol{Z_d}$ greatly depends on the value of maximum $\boldsymbol{K_k}$: according to the data of the study area the correlation coefficient between them equals to 0.98 and is significant at $p<0.001$. The linear relationship between these two indices is especially obvious, when maximum $\boldsymbol{K_k > 16}$. It can be presumed that in comparison with other elements Ag and Zn have often the highest accumulation level. The results confirm this: in 42.4% of sites Ag has maximum $\boldsymbol{K_k}$ and in 42.4% of sites – Zn. Other elements are characterised by maximum $\boldsymbol{K_k}$ only in rare cases: Mo – in 6.8% of sites, Pb – in 5.1%, Sn and Cu – in 1.7%. So in urban territories AAA with the highest accumulation of Ag or Zn are quite usual. The highest percentage of sites with maximum $\boldsymbol{K_k}$ of Ag is in recreational sites, of Zn – in public residential and residential sites and of other elements – in industrial-infrastructural sites (Fig. 2). Silver and zinc are usual atmospheric contaminants, most probably associated with different incineration processes. They are widely spread on the whole urban territory and even beyond it. The sites of their high accumulation largely depend on the content of organic matter in soil, especially of Ag, which has high coefficient of uptake by plants. This explains why the highest percentage of sites with maximum $\boldsymbol{K_k}$ of Ag is in recreational territories. In other land-use sites Ag might be related to de-icing salts. Some investigators suppose that Ag has domestic sources [16]. The highest percentage of sites with maximum $\boldsymbol{K_k}$ of Zn in public-residential or residential territories can be explained by intensive traffic and corrosion of roofs of the buildings.

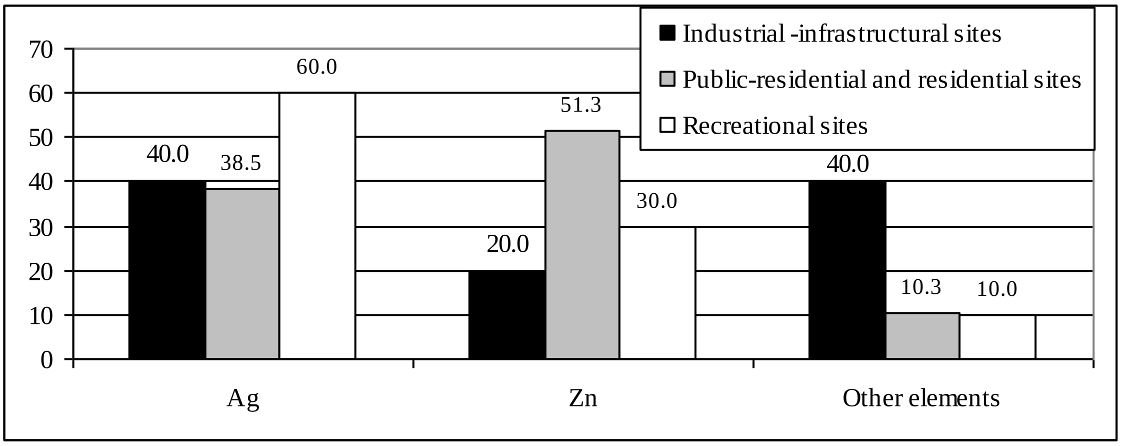

Fig. 2. **Percentage of sites with maximum accumulation level of different elements in various land-use sites**

To reveal the influence of specific contamination sources, mainly AAAs with maximum $\boldsymbol{K_k}$ of other elements should be analysed. The number of sites, where this is observed, is low. However, part of AAAs with maximum $\boldsymbol{K_k}$ of Ag or Zn includes not only elements related to non-ferrous metals, but also other elements. Such AAAs are also rare and usually more multielement. Two above-mentioned groups can be named *unusual associations (U).* AAAs only with elements related to non-ferrous metals and with the highest $\boldsymbol{K_k}$ of Ag or Zn can be named *ordinary associations (O)*. Taking into account also the level of association, i.e. interval of maximum $\boldsymbol{K_k}$, 8 groups of AAA can be formally distinguished, 7 of them are found in the study area (Table 1.).

Table 1.

Number of sites from different districts or land-use zones belonging to various groups of adjusted accumulating associations

Group	*District*		*Land-use*			*Total number*
	Naujamiestis	*Senamiestis*	*Industrial-infrastructural*	*Residential or public-residential*	*Recreational*	
E-O	2	**9**	0	**11**	0	11
H-O	4	**7**	1	**8**	2	11
M-O		**7**	2	1	**4**	7
L-O	1	**2**	0	**2**	1	3
E-U	**11**	1	**6**	5	1	12
H-U	**10**	3	1	**10**	2	13
M-U	**2**	0	0	**2**	0	2

Note. Ordinary associations of different level: E-O – extremely high, H-O – high, M-O – medium, L-O – low. Unusual associations of different level: E-O – extremely high, H-O – high, M-O – medium.

There is 54.2% of ordinary and 45.8% of unusual associations in the study area. Ordinary associations are more abundant in Senamiestis, while unusual – in Naujamiestis, which has been more industrialised [3]. When the level of association is M, associations are usually ordinary, when it is L, only ordinary. This is because ordinary associations can include maximum 5 elements (Ag, Zn, Pb, Sn and Cu). When the level of association is H or E, there are slightly more sites with unusual associations than with ordinary (Table 1). This is in accordance with the fact that more multielement pollution results in higher contamination of topsoil [9]. When the level of association is E, the main part of unusual associations is observed in industrial-infrastructural sites, meanwhile of ordinary associations – in residential or public-residential sites. This regularity indicates that extremely dangerously contaminated sites, where topsoil has been polluted by industrial activity, can often be traced according unusual associations. When the level of association is H or M, the main part of unusual associations is in residential or public-residential districts, though in the first case some of them can be found even in recreational sites. These land-use zones can be affected by industry or infrastructural objects. Unusual associations can be subdivided into simple or complex types (Table 2). There are four simple types distinguished taking into account more rare members of AAAs, which belong to 3 different groups: N – related to non-ferrous metals (Pb, Cu, Sn), F – related to ferrous metals (Mo, Ni, Cr, V, Co) and C – related to carbonates (Ba). Associations with maximum $\boldsymbol{K_k}$ of elements from N group are attributed to type N1 and with maximum $\boldsymbol{K_k}$ of elements from F group – to type F1. As Ba has nowhere maximum $\boldsymbol{K_k}$, the respective type is absent. However, type C2, which is characterised by Ba having not maximum $\boldsymbol{K_k}$, is present in site N11, where maximum permitted concentration (MPC) of Ba is exceeded (Table 2). Type F2, which is characterised by elements from group F having not maximum $\boldsymbol{K_k}$, is also found on the study area. Meanwhile elements from group N with not maximum $\boldsymbol{K_k}$ are quite usual in AAA, they are in ordinary AAA. There are also 2 mixed types of unusual AAA: F1-F2 and N1-F2. Types of unusual associations characterise the influence of specific urban pollution sources (Fig. 3). Type F1-F2 is found in the closest neighbourhood to the drill plant (sites N21, N22, N24), type F1 – in residential site N23 at some distance from this plant, while type F2 – at greater distances. All these types are characterised by accumulation of Mo. Though part of Mo can be from traffic (e.g. probably in site N06), the fact that F2 type is almost absent in Senamiestis (except S19), but is widely spread in Naujamiestis confirms great influence of the drill plant on the latter district.

Table 2.

Unusual accumulating associations and their types

Site	*L*	*Max K_k*	*LA*	*Adjusted accumulating association (AAA)*	*Type*	*Rarer members of AAA*	*Exceed MPC*	*Zd*	*G*
N21	I	250	E	**Mo**----ZnCrAgCuVPb-CoNiSn	F1-F2	Mo+Cr,V,Co,Ni	MoCr	282	4
N22	I	147	E	**Mo**---Zn-CuCrAgPbV-SnNiCo	F1-F2	Mo+Cr,V,Ni,Co	MoZnCr	189	4
N24	I	14.0	H	MoZn-CuAgPb-SnNi	F1-F2	Mo+Ni	MoZn	42.4	3
N23	E	10.3	H	Mo-ZnAgPbCu-Sn	F1	Mo	Mo	35.1	3
N27	I	17.3	E	**Zn**-CuAgPb-Sn-MoCr	F2	Mo,Cr	ZnPb	46.1	3
N02	I	18.4	E	**AgZn**-SnPb-CuMo-	F2	Mo	SnZnPb	60.0	3
N03	I	16.0	E	**Ag**-Zn-SnCuPb-Mo	F2	Mo	ZnSn	50.4	3
N14	R	31.1	E	**Ag**-CuZn-PbSn-Mo	F2	Mo	AgSnPb	63.4	3
N04	E*	19.6	E	**Zn**-Ag-MoCuPb-Sn	F2	Mo	Zn	48.0	3
N17	E*	32.5	E	**Zn**---AgPb-MoSnCuNi	F2	MoNi	Zn	52.1	3
N01	E*	39.1	E	**Ag**--Zn-SnCuPb-Mo	F2	Mo	AgSnZn	69.0	3
N19	E*	10.4	H	AgZn-MoPb-CuSn	F2	Mo	Zn	33.8	3
N18	E	10.1	H	ZnAg-Pb-MoCuSnV	F2	Mo,V	Zn	32.1	3
N26	E	12.8	H	ZnAg-CuMoPb-Sn	F2	Mo	Zn	40.4	3
N12	E	12.8	H	ZnAg-PbCuSn-Mo	F2	Mo	ZnPbSn	45.2	3
N06	E	13.9	H	ZnAg-Cu-PbMoSn	F2	Mo	Zn	35.0	3
N16	E	10.5	H	Zn-Ag-PbCuMoSn	F2	Mo	Zn	24.0	2
S19	E*	15.9	H	Ag--ZnMoSnPb	F2	Mo		24.9	2
N13	R	8.7	H	Ag-Zn-CuMoSn	F2	Mo		22.4	2
N20	E	4.8	M	AgPbMoZn-CuSn	F2	Mo		18.5	2
N25	E	5.1	M	Zn-AgPbCuMoSn	F2	Mo		15.8	1
N28	I	24.1	E	**Sn**-AgCuZn-Pb-Mo	N1-F2	Sn+Mo	SnPb	64.5	3
N30	R	8.1	H	Cu-ZnAgSnPb-	N1	Cu	SnPb	36.4	3
S04	E	57.1	E	**Pb**--CuAg-Sn-Zn	N1	Pb	PbCu	85.7	3
S21	E*	14.1	H	PbZn-AgCuSn-	N1	Pb	PbSn	39.6	3
S05	E	11.7	H	Pb-ZnAgCu-Sn	N1	Pb	Pb	27.1	2
N11	E	21.2	E	**ZnAg**-CuPb-SnBa	C2	Ba	ZnBa	53.5	3

Notes: L – land-use code, (explanation is in Fig. 1), LA – level of association, MPC – maximum permitted concentration given in HN 60-2004 [14], G – general category of topsoil contamination according to HN 60-2004 [14]: 1 – allowable, 2 – medium dangerous, 3 – dangerous, 4 – extremely dangerous.

In 4 sites of F1-F2 or F1 types near the plant, MPC of Mo is exceeded, in two of them also of Cr. Earlier research has shown their extremely high accumulation on the territory of the drill plant (median K_k of Mo is 497 and of Cr is 46.2), they are followed by Co, Cu (median K_k 21.2 and 18.3, respectively) and V, Ni (median K_k in the interval 8-16) [12]. In most sites with F2 type associations only Mo adds to AAA, its level of accumulation is usually low or medium. In site N18 V adds to Mo in association, while in site N17 – Ni. As their K_k are low (only slightly exceed 2), their origin can be from thermoelectric power station, which is to south-west (the prevailing winds are also from this direction). Ni and V are specific elements of this type of pollution [9]. Association of site N28 near locomotive depot belongs to type N1-F2, as it is characterised by the highest K_k of Sn due to metal processing, while Mo has low accumulation. In this site the origin of Mo can be both from the plant of drills and from locomotive depot. Earlier research has shown that median K_k values of Mo and Sn on its territory are elevated –2.9 and 2.4 [12]. Type F2 site N27, which is characterised by Mo and Cr in AAA, can be influenced not only by the drill plant, but also by the railway pollution. In site N03 there might be some remnants of Mo pollution from the former turning lathe plant, which is now demolished. The content of Mo on its former was high: median K_k was 11.4 [12]. Type N1 is found in site N30, where Cu has maximum K_k. The reason is that the site is near trolleybus ring. Earlier research has shown that trolley-bus parking places are characterised by elevated Cu content in topsoil [7] and that it is specific element of trolleybus traffic pollution [17]. Type N1 with the highest accumulation of Pb is found in sites S04, S05 and S21. Topsoil contamination in the first two sites seems to originate mainly from the railway pollution, meanwhile in the last one – from intensive motor transport.

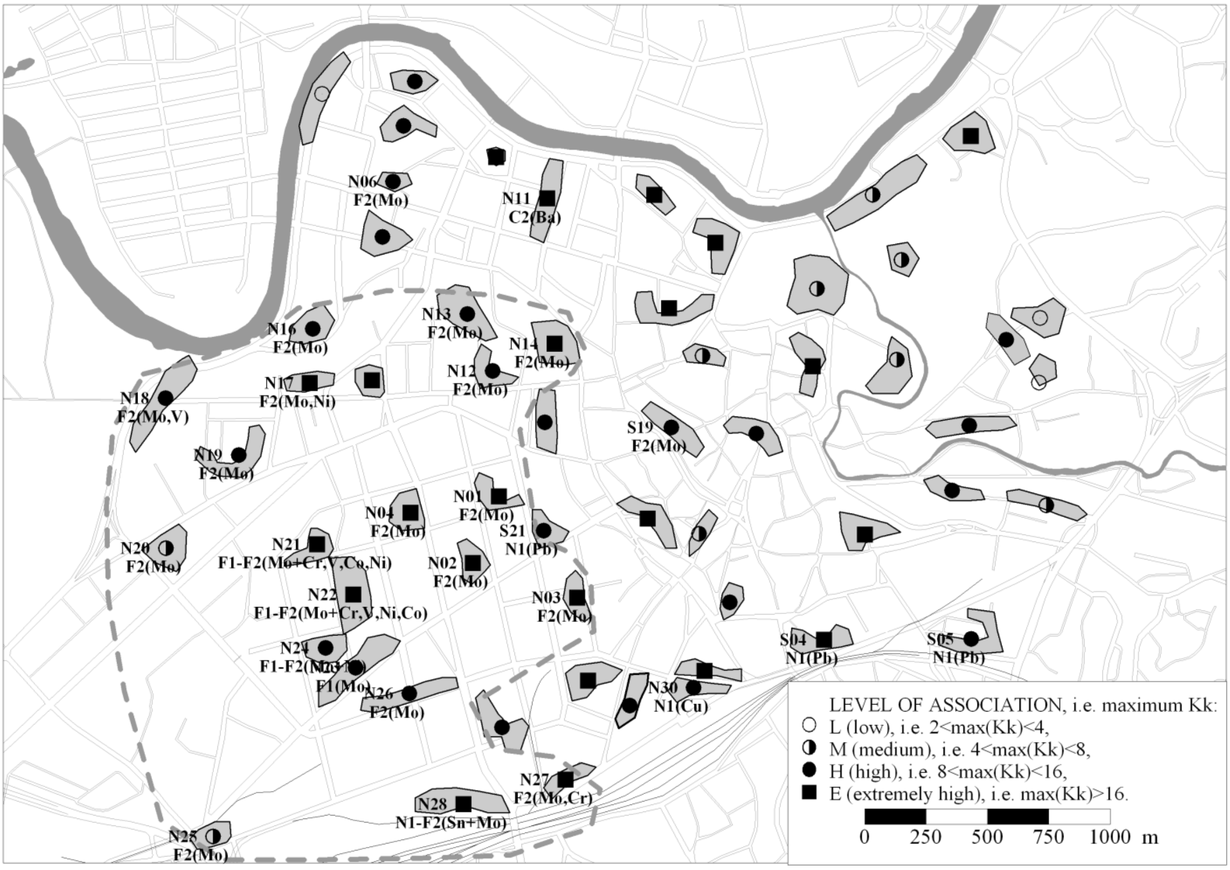

Fig. 3. **Variation of the level of ordinary and unusual associations and different types of unusual associations**

Note. Each unusual association is indicated by its type and more rare members of adjusted accumulating association. The dashed line indicates probable influence zone of the drill plant.

Conclusions

Topsoil quality in old urban centres mainly depends on the additive index Z_d of contamination by Ag, B, Ba, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sn, V, Zn and is related to accumulating associations, especially to the level of association, i.e. maximum concentration coefficient K_k of elements in it. Maximum K_k is significantly positively correlated with Z_d. Urban topsoil, especially in the old centres of towns is usually characterised by higher contamination by elements related to non-ferrous metals, i.e. Ag, Zn, Pb, Cu, Sn, compared to other elements. Accumulating associations including only these five elements with the highest K_k of Zn or Ag are ordinary. Unusual associations are either characterised by the highest K_k of other above-mentioned elements or include wider spectrum of elements, mainly Mo, Ni, Cr, V, Co, which are related to ferrous metals, or Ba, which is related to carbonates. When the level of association is extremely high, i.e. the highest K_k exceeds 16, the main part of unusual associations is observed in industrial-infrastructural sites, meanwhile of ordinary associations – in residential or public-residential sites. Extremely dangerously or dangerously contaminated sites, where topsoil has been polluted by industrial activity, can often be traced according to unusual associations.

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