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CHANGES OF ECOLOGICAL-GEOCHEMICAL STATE OF TOPSOIL AND RIVER SEDIMENTS IN VILNIUS
EKOLOĢISKI- ĢEOĶĪMISKĀ GRUNTS UN UPES NOGULŠŅU STĀVOKĻA IZMAIŅAS VIĻŅAS PILSĒTĀ

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Abstract. *The changes of environmental quality were revealed according to temporal differences of additive contamination indices calculated for topsoil and sediments of Neris and Vokė rivers in Vilnius. Topsoil contamination indices were calculated on the basis of Ag, Ba, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sn, V and Zn contents for 2 different periods (1985 and 1996-2002) and their comparison was realised on 237.86 sq.km territory of the first sampling. The areas, where topsoil ecological-geochemical state has worsened have been determined. Part of them with unallowable topsoil pollution level indicates the quarters, where population health risk increase is possible. Monitoring is necessary there, also health protection measures. The tendency of improvement of ecological-geochemical state of river sediments is observed, but potential sources of secondary pollution in sediments can be hazardous for downstream segments.*

Keywords: *ecological-geochemical state, heavy metals, topsoil, stream sediments, urban quality.*

Introduction

Due to intensive traffic, fuel and waste incineration and industrial activity, urban territories are polluted by heavy metals. They are hazardous to human health and the whole environment. Not only the geochemical quality of air and water is important, but also of soil and stream sediments. The investigation of topsoil is especially important, because heavy metals accumulate there and the routes of human exposure to them can be direct (accidental ingestion of contaminated soil or dermal contact with it) or indirect (drinking of water contaminated due to migration of hazardous substances from soil, inhalation of air contaminated by soil particles with heavy metals, eating of contaminated fruits, vegetables, fish, meat). Similar routes of human exposure are from contaminated sediments. Besides, their research enables to analyse the migration of pollutants. Therefore geochemical mapping of urban topsoil is done in various countries [1, 2]. It is often supplemented by investigations of bed sediments of streams crossing urban territories. This enables to find out the input of each town to stream pollution.

Geochemical investigations of Vilnius were started in the ninth decade of the XXth century by geochemists of the Institute of Geology. The first topsoil sampling was done in 1985 in the area covering large part of Vilnius territory and part of its environs. However, regular geochemical monitoring of topsoil in Vilnius was not done. So further sampling was for different aims and not in the same sites. Repeated sampling of topsoil was done in 1995-1998 in 6 central districts of Vilnius and Valakampiai. It revealed that Naujamiestis and Senamiestis districts were contaminated most of all [3]. Regular monitoring of stream sediments was also not done. Their early sampling was in the ninth decade of the XXth century, e.g. samples from Neris and Vokė were taken in 1981-1983. Some samples from these streams were taken also in 1991. The last sampling of topsoil and stream sediments was in 2002 during implementation of the program "Urban environmental quality and its change" aimed at complex evaluation of Joniškis, Mažeikiai, Šiauliai and Vilnius. Topsoil samples were taken from the territory of 15 peripheral districts of Vilnius and sediment samples from Neris, Vokė and Vilnelė. The quality of topsoil and stream sediments was compared in these towns, in Šiauliai lake sediments were also analysed [4]. Comparison of topsoil quality in four urban land use groups (public, residential,

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industrial-infra-structural and recreational) revealed the highest pollution level in Vilnius, especially in its industrial-infra-structural zones [5].
The aim of present research was to reveal the changes of environmental quality in Vilnius from the ninth decade of the XXth century till the end of XXth century according to temporal differences of additive contamination indices (Z13) calculated for topsoil and sediments of Neris and Vokė rivers taking into account the following potentially hazardous elements: Ag, B, Ba, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sn, V, Zn.

Materials and methods

Complex samples consisting from several sub-samples were taken from the upper (0-10 cm depth) soil layer. In 1985 topsoil samples (652) were taken at scale 1:50000, in 1995-1998 from central districts of Vilnius and Valakampiai (2237) at scale 1:7500-1:15000 and in 2002 from peripheral districts at scale 1:100000. Due to great difference in scales all samples were classified into 2 groups: 6 central districts, which were mapped in more detail, and 15 peripheral. Stream sediment samples were taken with a scoop. In 2002 they were taken from Nėris and its tributaries Vilnelė and Vokė. Comparison of changes in topsoil ecological-geochemical state was realised on 237.86 sq. km territory of the first sampling, while for stream sediments of Neris and Vokė in segments indicated by large circles (Fig. 1).

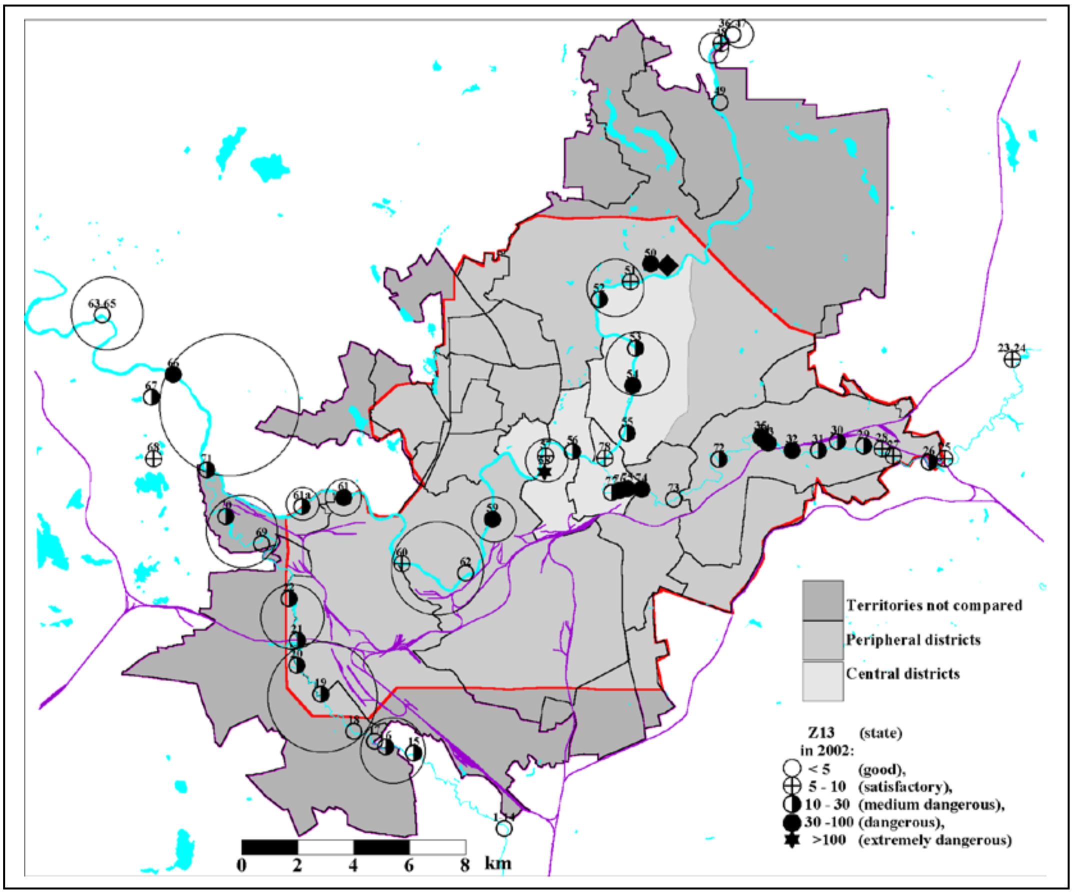

Fig. 1. **Study areas of topsoil contamination and stream sediment last sampling sites**

Note. Large circles indicate river segments, where contamination level in two periods was compared.
All samples were air-dried, sieved through nylon sieves (choosing fraction <1 mm). After burning at 450^{0}C and mechanical pulverisation they were analysed by DC arc emission spectrophotometry for determination of the total contents of Ag, B, Ba, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sn, V, Zn. The international reference materials OOKO 153 and OOKO 151 were used for quality control. They were analysed in each batch of 10 samples. Since 1997 for quality assurance of analytical results the laboratory of the Institute of Geology and Geography

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participates in “International Soil analytical exchange” subprogram organised by Wageningen University and international programs of inter-comparison [6].
Additive contamination indices Z13 for topsoil and stream sediments were calculated on the basis of Ag, Ba, Co, Cr, Cu, Mn, Mo, Ni, Pb, Sn, V and Zn contents for 2 different periods. Regional background values of these elements in soil were taken as the first approximation [7]. They were adjusted in a way described elsewhere [8]. Table 1 shows that for greater part of the elements, except Co, Sn, Pb, Ni, the adjusted local background values of the second sampling were slightly lower than for the first sampling. Median values of elements in bed sediments of Lithuanian streams depend on soil in the watershed [9]. Lithuanian streams draining dominant areas of sandy-loamy soil were chosen and anomalies of elements eliminated (values greater than 2*Md and lower than 0.5*Md, where Md is median value). The new medians after elimination of anomalies were taken as the background for element content in sediments of Neris, Vilnelė and Vokė. Table 1 shows that for most elements, except Pb, these background values are higher than for soil.

Table 1.

Background values (ppm) of elements in soil and stream sediments

Elements	B	B_1	B_2	$RB=B_1/B_2$	BSS
Ag	0.064	0.093	0.072	0.78	0.086
B	21.3	23.2	20.1	0.87	28.1
Ba	305	297	287	0.97	431
Co	3.70	2.64	2.88	1.09	4.9
Cr	24.3	25.8	24.3	0.94	34.9
Cu	6.80	7.64	7.60	0.99	10.8
Mn	436	544	395	0.73	888
Mo	0.64	0.74	0.64	0.86	0.84
Ni	9.6	9.4	9.4	1.00	12.2
Pb	15.0	15.1	15.4	1.02	13.7
Sn	1.95	1.93	2.05	1.06	2.3
V	25.7	25.0	24.3	0.97	30.0
Zn	24.9	27.2	23.1	0.85	47.2

B – regional soil background (South Lithuanian Plain), B1 – adjusted local background of the first sampling, B2 – adjusted local background of the second sampling, BSS – background for stream sediments**.**

Results and discussion

Table 2 indicates that during the first sampling the accumulating association of all investigated territory included only 5 non-ferrous metals. In central area it was wider and besides them contained also Ba, Cr, Ni and Mo, meanwhile in peripheral area it was narrower and consisted of the only element Zn. So during the first sampling the central part of Vilnius was affected not only by transport, corrosion of building material and household pollution, but also by industrial pollution of metal working plants (plants of boring machines, fuel equipment, etc.), meanwhile the pollution of peripheral part was actually only beginning. Therefore the median concentration coefficients of almost all elements, except B, Mn and V, were higher in the central part than in peripheral. The same concerns Z13 index: its median value in the central area was 2.5 times higher than in peripheral part and was already approaching the limit for allowable contamination (16).
During the period of the second sampling the same elements remained in accumulating association of central part, but the growth of contamination was observed only for part of them: Zn, Pb, Cu, Ag, Ba and Cr, i.e. mainly for those related to non-ferrous metals. Contamination level of Ni remained the same and of Sn and Mo even slightly decreased probably due to the fact than a lot of industrial enterprises (including metal processing) stopped functioning, part of

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accumulated pollutants could be washed out of topsoil or removed with soil cover during reconstruction of separate quarters of the central districts. Despite decrease of accumulation level of Sn and Mo, the additive contamination in the central part has grown up and exceeded the limit for allowable contamination (16) indicating that the greater part of central districts are characterized by unfavourable conditions for life, most probably due to intensification of traffic, construction works, corrosion of building materials and growth of household pollution and further accumulation of pollutants in topsoil. New centres of pollution appeared near parking places and crossings with intensive traffic.

Table 2.

Temporal change of contamination indices in topsoil

In all investigated area (N_1=617, N_2=2453)					**In central area (N_1=117, N_2=2238)**					**In peripheral area (N_1=500, N_2=215)**				
Var.	**M_1**	**M_2**	**R**	**D**	**Var.**	**M_1**	**M_2**	**R**	**D**	**Var.**	**M_1**	**M_2**	**R**	**D**
Z13	5.28	18.24	3.45	12.96	Z13	12.22	19.61	1.60	7.39	***Z13***	4.88	9.32	1.91	4.44
Zn	**1.39**	**5.10**	3.68	3.71	**Zn**	**3.13**	**5.21**	1.67	2.08	**Zn**	**1.30**	**2.87**	2.21	1.57
Ag	**1.36**	**3.22**	2.37	1.86	**Pb**	**2.17**	**3.20**	1.48	1.04	***Cu***	1.24	**1.72**	1.39	0.48
Pb	**1.29**	**3.04**	2.35	1.75	**Cu**	**1.78**	**2.32**	1.30	0.54	***Pb***	1.21	**1.55**	1.28	0.34
Cu	**1.34**	**2.26**	1.68	0.92	**Ag**	**2.74**	**3.50**	1.28	0.76	*Ba*	1.21	**1.42**	1.18	0.22
Sn	**1.32**	**1.94**	1.47	0.62	**Ba**	**1.30**	**1.44**	1.11	0.14	***Ag***	1.28	**1.50**	1.17	0.22
Mo	1.17	**1.50**	1.29	0.33	**Cr**	**1.33**	**1.37**	1.03	0.04	*Mo*	1.13	**1.30**	1.14	0.16
Ni	1.20	**1.48**	1.23	0.28	Co	1.27	1.28	1.01	0.01	*Co*	1.16	**1.32**	1.14	0.16
Ba	1.22	**1.44**	1.18	0.22	**Ni**	**1.50**	**1.50**	1.00	0.00	*Cr*	1.21	**1.34**	1.11	0.13
Cr	1.24	**1.36**	1.10	0.13	**Mo**	**1.55**	**1.54**	0.99	-0.01	Mn	1.21	**1.34**	1.11	0.13
Co	1.18	1.28	1.09	0.10	Mn	1.09	1.08	0.99	-0.01	V	1.20	**1.31**	1.10	0.12
V	1.19	1.10	0.93	-0.08	B	1.08	1.05	0.98	-0.03	Ni	1.17	1.27	1.08	0.10
B	1.15	1.06	0.93	-0.09	V	1.15	1.09	0.94	-0.07	***Sn***	1.28	**1.34**	1.05	0.07
Mn	1.18	1.09	0.92	-0.09	**Sn**	**2.16**	**2.01**	0.93	-0.15	B	1.16	1.16	1.00	0.00

Note. Var. – variable (element or additive contamination index of all 13 elements), N_1 and N_2, – number of samples, M_1 and M_2 – median values of concentration coefficients of elements or their additive contamination index Z13 during the first and the second sampling, respectively. $R=M_2/M_1$, $D=M_2-M_1$. Elements belonging to accumulating associations (with median concentration coefficient higher than 1.3) are in bold.

The accumulating association in peripheral part became even wider and included not only all elements related to non-ferrous metals, but also to ferrous metals: Zn>Cu>Pb>Ag>Ba>Cr>Mn>Sn>Co>V>Mo. The highest rate of contamination increase is characteristic of Zn, Cu, Pb, Ba, Ag, i.e. elements related to transport and household pollution. The median value of additive contamination index in peripheral area of Vilnius exceeded 8, i.e. started to approach to the limit of allowable contamination. The relative growth of Z13 in peripheral districts (1.91) was even higher than in the central districts (1.60), but the absolute growth in the latter districts (7.39) exceeded the absolute increase of Z13 in periphery (4.44). Some of administrative districts in periphery are highly industrialized and therefore are characterized by high median contamination level. They are located mainly in the southwestern and southern part of Vilnius (Fig. 2), also in Naujoji Vilnia, which is in the eastern part. The sites, where ecological-geochemical state of topsoil has worsened are mainly related to motor and railway transport, industry and household pollution.

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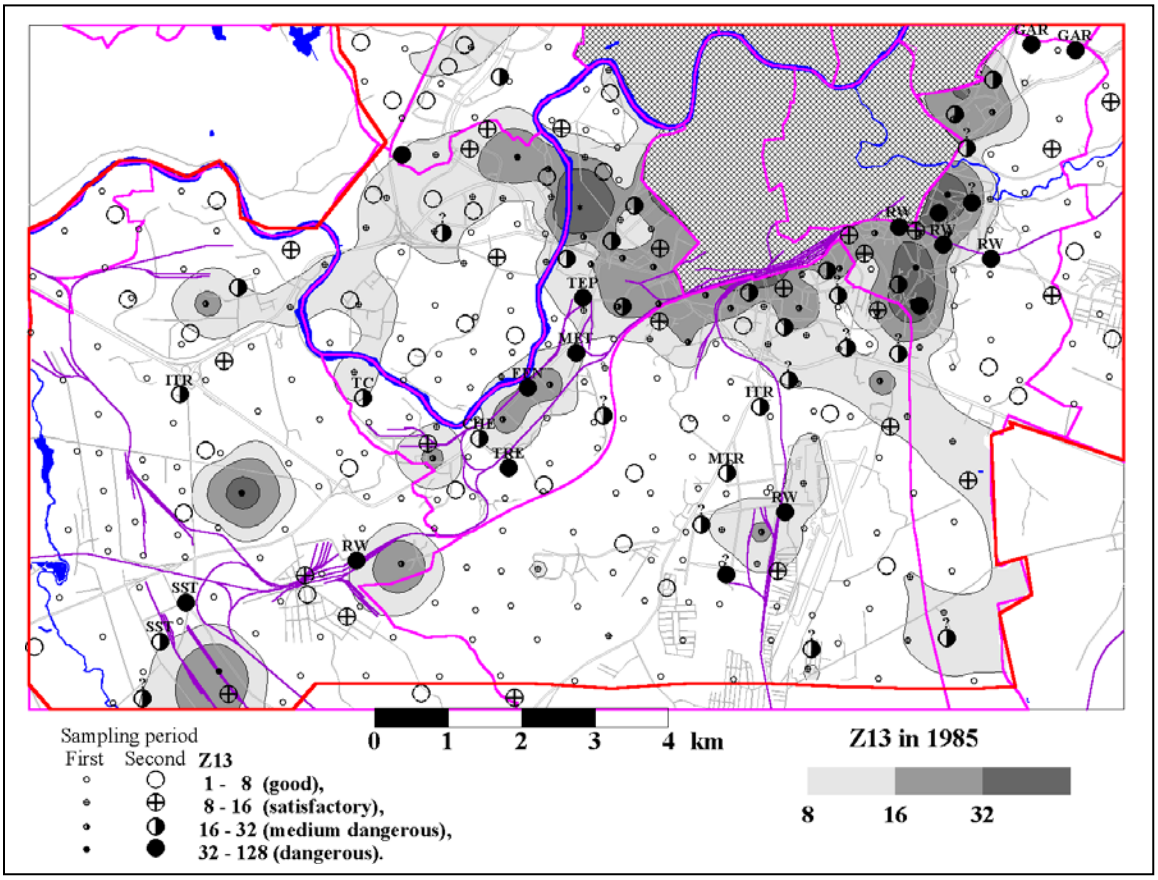

Fig. 2. **Temporal variability of topsoil contamination in southwestern part of Vilnius**

Note. New sites, which were revealed during the second period of sampling, where topsoil contamination exceeds the allowable level (16) and where the ecological-geochemical state became less favourable for life, are indicated by label, which explains the possible reason of contamination increase: ITR – intensive transport, GAR – garages, SST – service stations, RW – railway, MET – metal processing, MTR – metal trade, EEN – electrical engineering, CHE – chemical industry, TEP – thermoelectric power plant, HHP – household pollution.

The relative growth of topsoil contamination in all investigated territory was 3.45, while the absolute one was 12.96. The highest accumulation level was always characteristic of Zn due to variety of its different sources (metal processing, other industry, fuel burning, traffic, fertilizers, corrosion of roofs, etc.).

Median Z13 of Neris sediments in the whole Vilnius has 3.8 times decreased from 26 in 1981–1983 to 6.8 in 2002, i.e. from medium dangerous to satisfactory state. The number of accumulating elements with concentration coefficient (CC) exceeding 1.3 decreased from 9 in 1981–1983 to 4 in 2002, for most of them, except Mn and Pb, the accumulation level (CC) decreased from $Ag_{16}Cu_{3.6}Sn_{3.3}Cr_{2.8}Zn_{2.7}Mn_{1.5}Mo_{1.5}Pb_{1.4}Ni_{1.4}$ in 1981–1983 to $Ag_{4.5}Mn_{1.9}Pb_{1.5}Cu_{1.4}$ in 2002. Table 3 indicates that in most of the segments of Neris and Vokė Z13 index decreased. Almost everywhere (except segments V and VI, which are heavily polluted) the highest accumulation level (CC>2) is characteristic of non-ferrous metals (Ag, Cu, Zn, Sn and Pb), also Mn and Cr.

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Table 3.

Temporal change of contamination and accumulating elements in stream sediments

River segment	Year	Z13	Accumulating elements	R	Increase of
I. Neris upstream of Vilnius (No 36-47)	1982	6.7	$Ag_{2.6}$CuNiMnZnCrSn	0.85	**Mn**Mo
	2002	5.7	$Mn_{4.3}$AgMo		
II. Neris downstream of Žalesa (No 48)	1983	3.0	$Mn_{2.7}$	1.22	Sn
	2002	3.6	$Mn_{2.5}$Sn		
III. Neris near Verkiai (No 51-52)	1981	1.2	-	2.14	MoMnCu
	2002	2.5	MnMoCu		
IV. Neris near Žirmūnai (No 53-54)	1981	4.6	$Ag_{3.3}$Mn	0.90	MoCu Pb**Mn**
	2002	4.2	$Mn_{2.1}$CuPbAgMo		
V. Neris near Žvėrynas (No 57-58)	1981	65	$Ag_{40}Zn_{7.6}Sn_{6.1}Cu_{5.9}Pb_{3.0}Cr_{2.8}Mo_{2.7}Ba_{2.3}$MnNi	0.35	Mn
	2002	23	$Ag_{17}Pb_{2.6}Zn_{2.5}$CuMnSnMo		
VI. Neris near „VELGA" (No 59)	1981	76	$Sn_{24}Cu_{18}Ag_{18}Zn_{8.2}Cr_{5.0}Mo_{3.3}Pb_{3.1}Mn_{2.2}$Ni	0.40	**CrNi**V
	2002	31	$Cr_{13}Ni_{7.2}Ag_{6.1}Cu_{4.4}$ZnPbVSn		
VII. Neris in Bukčiai (No 60, 62)	1981	21	$Ag_{10}Cu_{8.3}Zn_{2.1}$CrSnPb	0.20	Mn
	2002	4.1	$Ag_{3.3}$Mn		
VIII. Neris down-stream of wastewater treatment pl. (No 61)	1983	30	$Ag_{17}Cr_{4.2}Cu_{3.4}Zn_{3.3}Sn_{2.5}$MnMoPbNiBaCo	0.52	**MnPb**
	2002	16	$Ag_{11}Mn_{2.4}Pb_{2.1}$CuZn		
IX. Neris near Sudervėlė (No 61a)	1983	10	$Ag_{4.6}Cu_{2.4}Cr_{2.3}Mn_{2.0}$ZnNiSn	1.28	**PbAg**
	2002	14	$Ag_{9.6}Pb_{2.3}$ZnMnCu		
X. Neris dowstream of Vokė (No 66, 71)	1981	34	$Ag_{22}Cu_{3.8}Sn_{3.8}Cr_{3.6}Zn_{2.5}$MoMnNiPb	0.40	PbMn **Zn**
	2002	13	$Ag_{7.5}Cu_{2.9}Zn_{2.7}$PbMnNiSn		
XI. Neris near Verkš-nionys (No 63-65)	1981	34	$Ag_{23}Cr_{4.0}Cu_{3.5}Sn_{2.7}Zn_{2.4}$MnMoNi	0.11	–
	2002	3.7	$Ag_{3.7}$		
XII. Vokė in Vaidotai (No 15-17)	1984	2.9	AgSnZn	0.58	Mn
	2002	1.7	Mn		
XIII. Vokė in Nauja-kiemis (No 18-20)	1984	7.5	$Ag_{4.8}$ZnSnMo	0.80	MnCu **Pb**
	2002	6.0	$Zn_{2.2}Pb_{2.1}$CuAgMnSn		
XIV. Vokė in Trakų Vokė (No 21-22)	1984	7.3	$Ag_{3.6}Zn_{2.2}$MoSnMnPb	1.61	**SnCuPb** NiMn
	2002	12	$Sn_{6.2}Pb_{2.9}Cu_{2.2}$MnZnNiAgMo		
XV. Vokė in Grigiškės (No 69-70)	1981	3.1	AgSnMn	1.02	**Zn**
	2002	3.1	$Zn_{2.7}$Ag		

Note. River segments are shown in Fig. 1. Accumulating elements have CC>1.3. The subscript of element indicates its CC (only CC>2 are shown). R is the ratio of Z13 in the second period to Z13 in the first period. Elements with high increase are in bold.

In segment V near former tannery and leather processing plant Cr, Mo and Ba added to other common pollutants in 1981. However, when the plant was transferred to another place, the content of these elements decreased in sediments taken in 2002 and the ecological-geochemical state improved from dangerous to medium dangerous category.

It also improved in segment VI, where surface runoff from welding equipment plant comes: the Z13 decreased due to lower CC of Sn (tinning technology is no more used in the plant).

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However, the ecological-geochemical state of the segment remained dangerous, because Cr and Ni are still used in technological processes and their pollution increases.

High water migration coefficient explains variability of Ag in bed sediments. Accumulation of Ag and Mn can be related to geochemical barriers (oxidation and sorption processes) in sediments in the places of groundwater discharge and inflow of unpolluted surface water. Increase of Pb and Ag downstream of wastewater treatment plant can be explained by general increase of transport and household pollution in the whole town. This can be the reason of Z13 increase in segment IX.

Paper mill in Grigiškės, where waste paper is processed, can explain the growth of Zn in Vokė (segment XV) and in Neris downstream of Vokė (segment X). However, Z13 increase is not essential in Vokė (segment XV). So the influence of this tributary on Neris pollution is not great and ecological-geochemical state of Neris downstream of Vokė has improved greatly.

Meanwhile the growth of Z13 and Sn, Cu, Pb pollution in Vokė sediments within segment XIV as well as increase of Pb accumulation in segment XIII is related to intensive railway and motor transport pollution in vicinity of storehouses and enterprises of southwestern industrial district. The relative growth of Z13 in Neris near Verkiai (segment III) is related to geochemical barriers, because Z13 indices of both periods are low.

Ecological-geochemical state of Vilnelė, which is other tributary of Neris, was analysed only in 2002 and differs in various segments. In some of them (i.e. in segment following metal processing plants in Naujoji Vilnia or near tannery and electrical engineering plant near central districts of Vilnius) the ecological-geochemical state of sediments is still dangerous (Fig. 1).

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Conclusions

During the period from 1985 to 2002 topsoil total contamination index has mainly increased in Vilnius indicating that this medium is accumulating pollutants over long period. Besides, new polluted areas have appeared in peripheral districts and especially near parking places. The growth of topsoil contamination by non-ferrous metals (Zn, Pb, Cu, Sn and Ag) was more intensive than by elements related to ferrous metals and can be explained mainly by increase of traffic intensity and household pollution. The relative growth of additive contamination level of topsoil in peripheral districts of town is higher than in the central districts, partly due to location of industry, railway and streets with intensive transport in peripheral areas. However, both the levels and absolute growth of total contamination index in central districts, which are older and more densely populated, is higher. These regularities can be observed also in other towns. Decrease of topsoil anomalies on local areas is possible due to disturbances of soil cover.

The tendency of improvement of ecological-geochemical state of river sediments is observed due to decrease of industrial emissions of the territories of industrial enterprises, lower enrichment of surface runoff with heavy metals, better treatment of wastewater and migration of pollutants with water flow. The highest accumulation level in stream sediments is characteristic of non-ferrous metals (Ag, Cu, Zn, Sn and Pb), also Mn and Cr, but their accumulation mostly decreases, except several segments near local emission sources of pollutants and downstream of them, also in places of geochemical barriers. Increase of Pb contamination can be related to rain sewage discharge to streams.

Geochemical monitoring is necessary in the areas, where topsoil ecological-geochemical state has worsened and exceeds the allowable level of topsoil contamination. The same concerns stream sediments, not only because it is necessary to monitor the influence of effluents, but also

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because the sites heavily contaminated until now are potential secondary pollution sources for downstream segments.

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