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Chemical composition of soil and lake sediments — an indicator of geological processes in Lithuania

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Long-term multipurpose geochemical studies in Lithuania show that the chemical composition of surface sediment can be used as an indicator of geological process: duration of weathering, soil formation and thickness of sediment. The chemical composition of upper and lower soil layers and of lake sediments gives information on past sedimentation: the type and age of Quaternary deposits and the location and depth of Pleistocene glaciolacustrine basins. We therefore have a baseline to predict changes in surface chemistry provoked by current anthropogenic pressures.

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INTRODUCTION

The chemical composition of surface sediment is a complex indicator of various geological processes: weathering, soil formation, sedimentation. It has long been used, though not always successfully, as a means to help separate Quaternary deposits of the same genesis but of different age (Late Weichselian and Late Saalian) in eastern Lithuania (Baltrūnas, 1995). More recent geochemical research on the soils formed on these sediments show that soil chemical composition may, rather be an important geoindicator, differentiating Quaternary deposits of the same genesis which have been affected by weathering and soil forming processes of different duration (Righi *et al*., 1997).

The trace element content of lake sediments depends on many factors, one being the depth at which sediment accumulated (see Salonen *et al*., 1993; Itkonen and Olander, 1997 for data from other countries). This dependence of trace element accumulation on lake depth can help evaluate the distribution of pollutants in reservoirs. It can also help to reconstruct the bathymetry of Pleistocene glaciolacustrine basins.

METHODS

This study is based mainly on data from the *Geochemical Atlas of Lithuania* and lake sediment investigations (Kadūnas *et al*., 1999; Kadūnas and Budavičius, 1999).

Samples of four soil types (sandy, sandy-loamy, loamy-clayey and peat) were collected from the topsoil horizon (A) during the geochemical mapping of soil in Lithuania at a scale of 1:1,000,000. In the eastern Lithuanian highland 75 sandy soil, 80 sandy loam and 82 loam-clay samples were taken, while in the neighbouring Ašmena–Medininkai highland 16, 17 and 15 samples respectively were collected. This geochemical data has been used to more precisely define the age and limits of glaciations in these highlands.

To understand the influence of soil forming processes on the regional chemistry, 249 samples were collected throughout 53 complete soil profile sites (Fig. 1).

For the lake sediment investigation, the four deepest lakes in the eastern Lithuanian highland — Smalvas, Bebrusai, Luokesai and Dusia — were selected. The 484 lake sediment samples were taken in profiles crossing the deepest zones of the lakes (Fig. 2). Samples with loss on ignition above 50% were termed organic sediments, while samples reacting with HCl

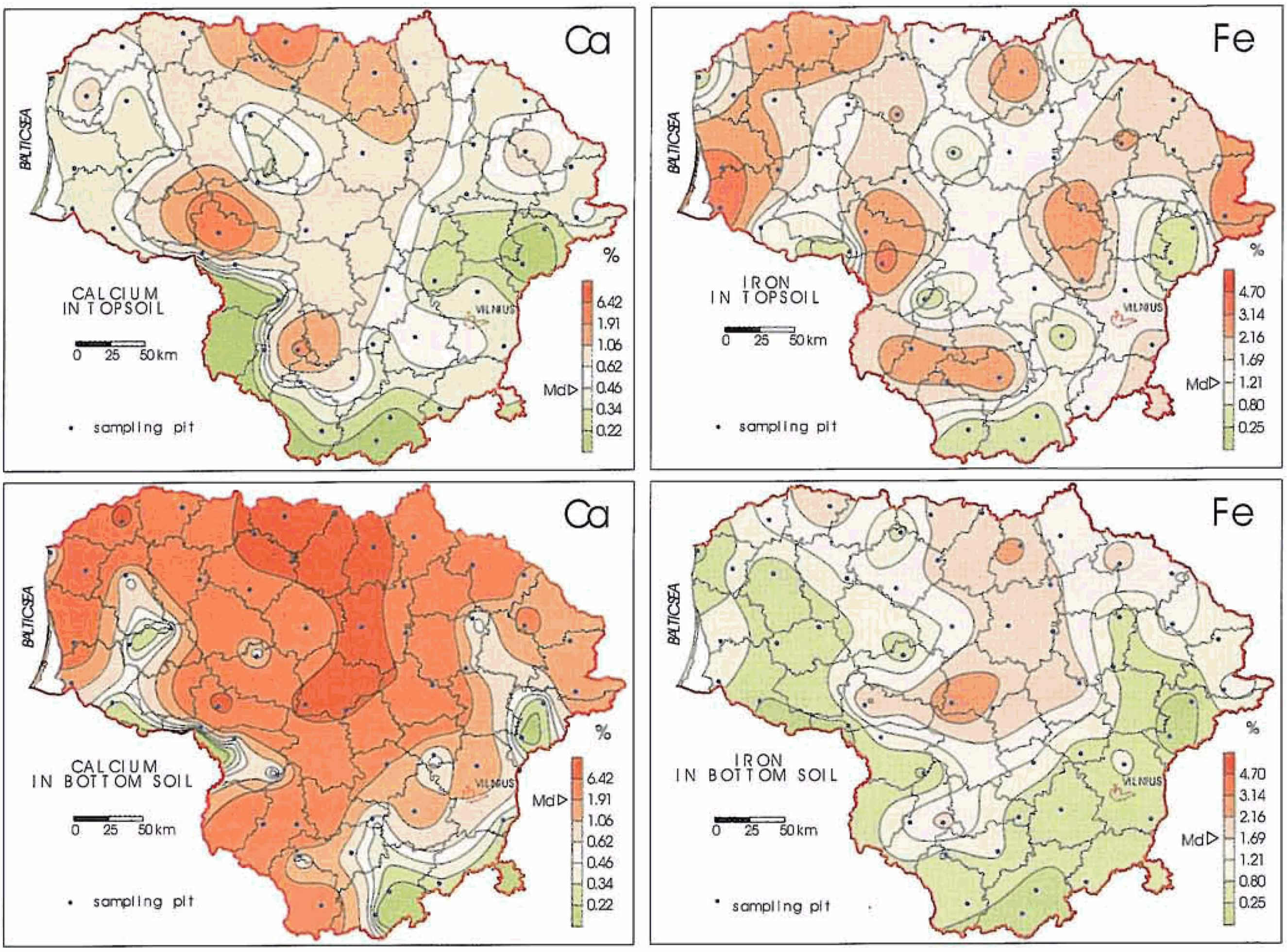

Fig. 1. Regional distribution of calcium and iron in topsoil and soil parent material in Lithuania

and having a Sr content > 200 ppm were categorised as calcareous sediments. A content of Ga > 7 ppm (the median value of Ga in clay and loam in Lithuania) was an additional index to distinguish terrigenous clayey sediments. Median values of elements were calculated for sediment from three different lake zones: littoral < 5 m, sublittoral 10–20 m, and profundal > 20 m. The following element associations in sediment were distinguished: authigenic elements related to carbonates, phosphates, hydroxides that formed *in situ*; allochthonous — relating to primary detrital minerals; and soluble biogenic elements that are biologically cycled and deposited mainly with organic matter. Part of the biogenic elements may be locally anthropogenic in origin and so this element association is referred to as biogenic-anthropogenic. Allochthonous trace elements additionally have been disjoint to allochthonous association — the trace elements of the common minerals (quartz, feldspar, mica and clay minerals) and allochthonous-accessory elements related to the weathering-resistant minerals (zircon, rutile, ilmenite, tourmaline *etc.*). Element associations are named according to element state and mineralogy.

Median values of trace elements

Highland	*	Ag	B	Ba	Co	Cr	Cu	Ga	Y	Yb	La	Li
Eastern Lithuania (Late Weichselian)	I	0.064	21.8	293	3.3	21.5	5.8	4.6	12	1.6	19.3	10.4
	II	0.068	28.1	369	4.8	31.0	8.9	5.6	17	2.0	23.2	11.9
	III	0.064	37.7	413	6.6	42.3	11.5	7.4	20	2.4	31.7	15.5
Ašmena–Medininkai (Late Saalian)	I	0.079	22.0	296	3.4	23.1	6.8	5.3	12.4	1.8	18.3	11.3
	II	0.087	24.8	367	4.5	26.2	7.7	5.4	11.6	1.9	21.3	11.6
	III	0.083	24.8	438	4.6	32.6	8.6	6.7	13.8	2.0	22.4	14.3

* — type of topsoil: I — sand, II — sandy loam, III — loam-clay

Microelement content was determined by DC Arc Emission Spectrometry in the < 1 mm fraction and recalculated to air-dried material. Analytical and mapping methods are described in Gregorauskienė and Kadūnas (1999).

In data processing, standard statistical methods (nonparametric, descriptive statistics, correlation and factor analysis) and software (EXCEL, SPSS and STATISTICA) were used. Concentration (accumulation) coefficients (K_k) were determined by dividing median microelement values in each sediment type by their median value in all sediments not differentiated according to their types.

CHEMICAL COMPOSITION OF TOPSOIL AS AN INDICATOR OF SEDIMENT AGE

Soil in the eastern Lithuanian highland that formed during the last Nemunas Glaciation (Late Weichselian) is rather different in trace element composition from soil in the neighbouring Ašmena–Medininkai highland (Tab. 1) that formed during the penultimate Medininkai Glaciation (Late Saalian) (Kudaba, 1983; Baltrūnas, 1995; Bitinas and Satkūnas, 1995). This shows the influence of weathering and duration of soil forming processes on the distribution of the trace elements.

The longer duration of these processes (about 100 thousand years) and the periglacial reworking of sediment (Basalykas *et al.*, 1976) produced a characteristic trace element composition of soils in the Ašmena–Medininkai highland. Some trace elements (P, Mn, Ag, Zr) are present in higher concentrations than in other Lithuanian regions, while others (V, Ni, Cr, Co, Sc) are scarce. Trace elements related to clay minerals tend to have been removed from these older soils. Increased values of Zr, Nb, Y, Yb and P reflect a relative increase in the amount of accessory minerals (e.g. zircon, apatite, tourmaline, staurolite) during weathering. The increased values of P and Mn can also be related to the formation of authigenic phosphate (vivianite) and Mn hydroxides. Increased values of Mn, Ag, Zr and partly P and decreased values of Cr, Co, Ni, V as well as other clay mineral-related trace elements in the soils above the Medininkai glacial deposits can also be linked with periglacial processes: these tend to form loamy sandy soils with depleted in clay minerals and enriched in silt and sand. This chemical signature occurs widely in loamy sand in the country.

These soils of different ages also show differences in the correlations between trace elements (Tab. 2). More trace elements show strong correlation in soils of the Ašmena–Medininkai highland ($R > 0.8$–0.9) than in soils of the eastern Lithuanian highland. In the Ašmena highland trace elements related to weathering-resistant minerals (Ti-Zr-Nb-Y-Yb-La) prevail, while in the eastern Lithuanian highland the trace elements are related to clay minerals (Ga-Li-Cr-V-Ni-Co-Sc). The long periods of weathering in effect reveal the primary relations of the weathering-resistant minerals from the time of their formation. As the composition of the top soil horizon differs little from the trace element composition of the parent material (horizon C) (Gregorauskienė and Kadūnas, 1999), these peculiarities can be used as an indi-

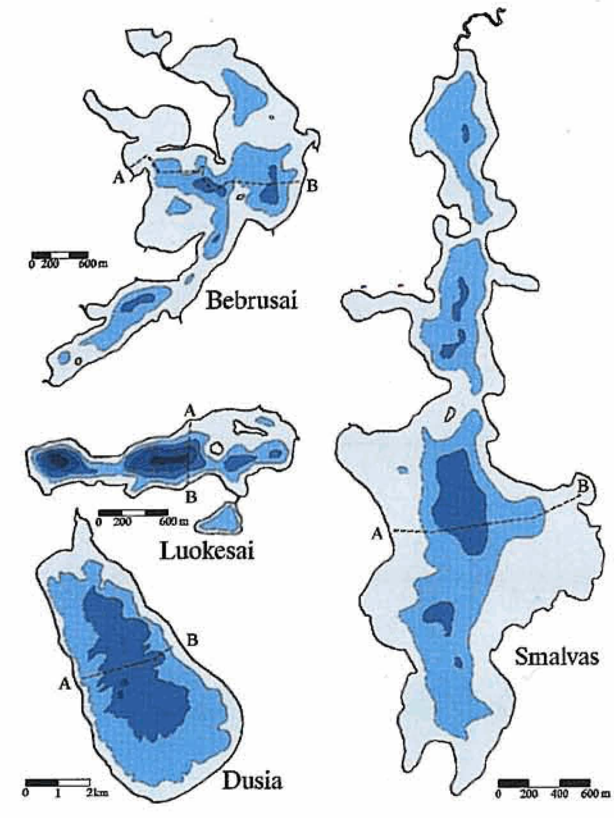

Fig. 2. Lake sediment sampling sites

Dotted lines indicate the location of bathymetric profiles with trace element distribution; isobaths are every 10 m

Table 1

in different age soils of Lithuania

Mn	Mo	Nb	Ni	P	Pb	Rb	Sc	Sn	Sr	Ti	V	Zn	Zr
407	0.59	13.6	8.6	547	13.9	52.2	2.5	1.94	73.9	1841	25.4	17.6	323
473	0.66	14.5	12.7	482	13.1	68.6	4.8	2.01	84.9	2650	33.8	25.6	344
474	0.75	14.4	18.3	462	13.4	89.2	7.7	2.22	94.5	3199	49.2	34.9	322
598	0.62	14.6	8.6	847	15.0	46.3	2.5	2.07	68.3	2121	23.9	18.9	351
679	0.62	15.4	8.5	851	14.4	49.5	3.9	2.00	87.8	2499	26.2	28.3	369
605	0.66	14.6	10.7	897	14.6	55.8	5.5	1.95	90.0	2341	31.2	32.2	310

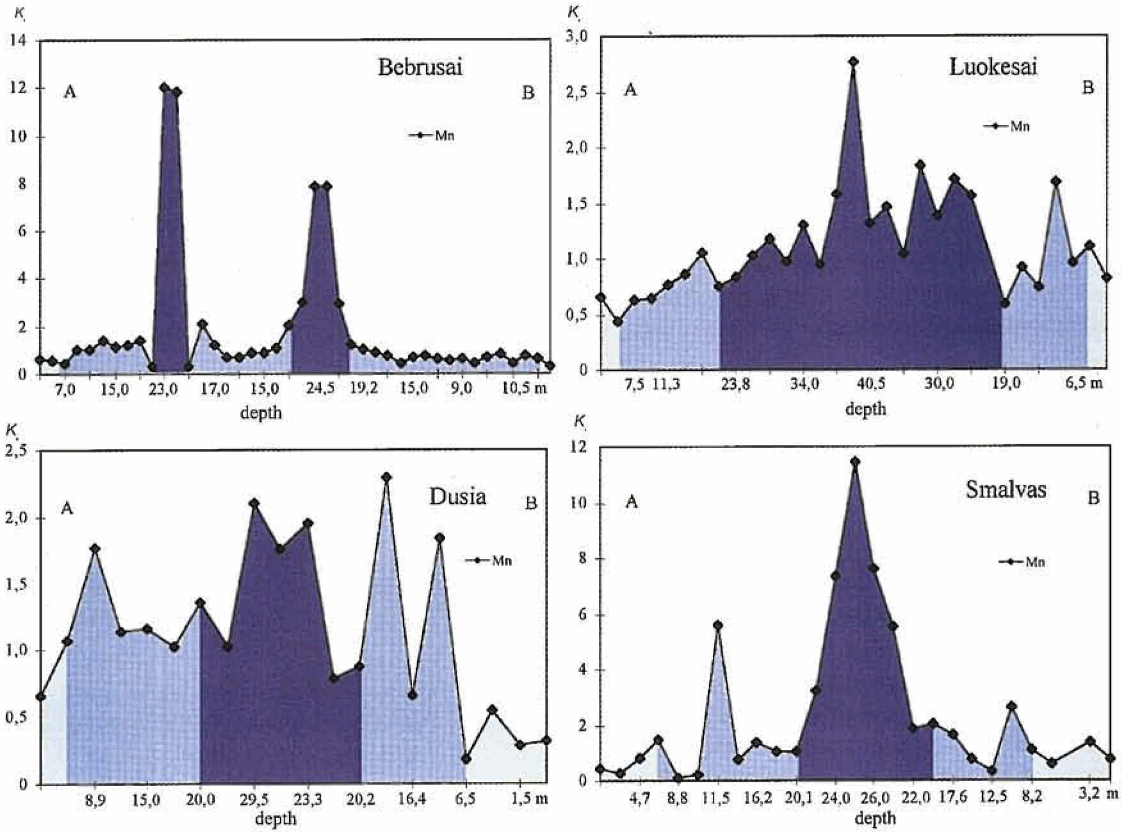

Fig. 3. Distribution of Mn in bathymetric profiles (sections A–B) of lakes Bebrusai, Luokesai, Dusia and Smalvas. Element values are shown as coefficients of concentration K_k

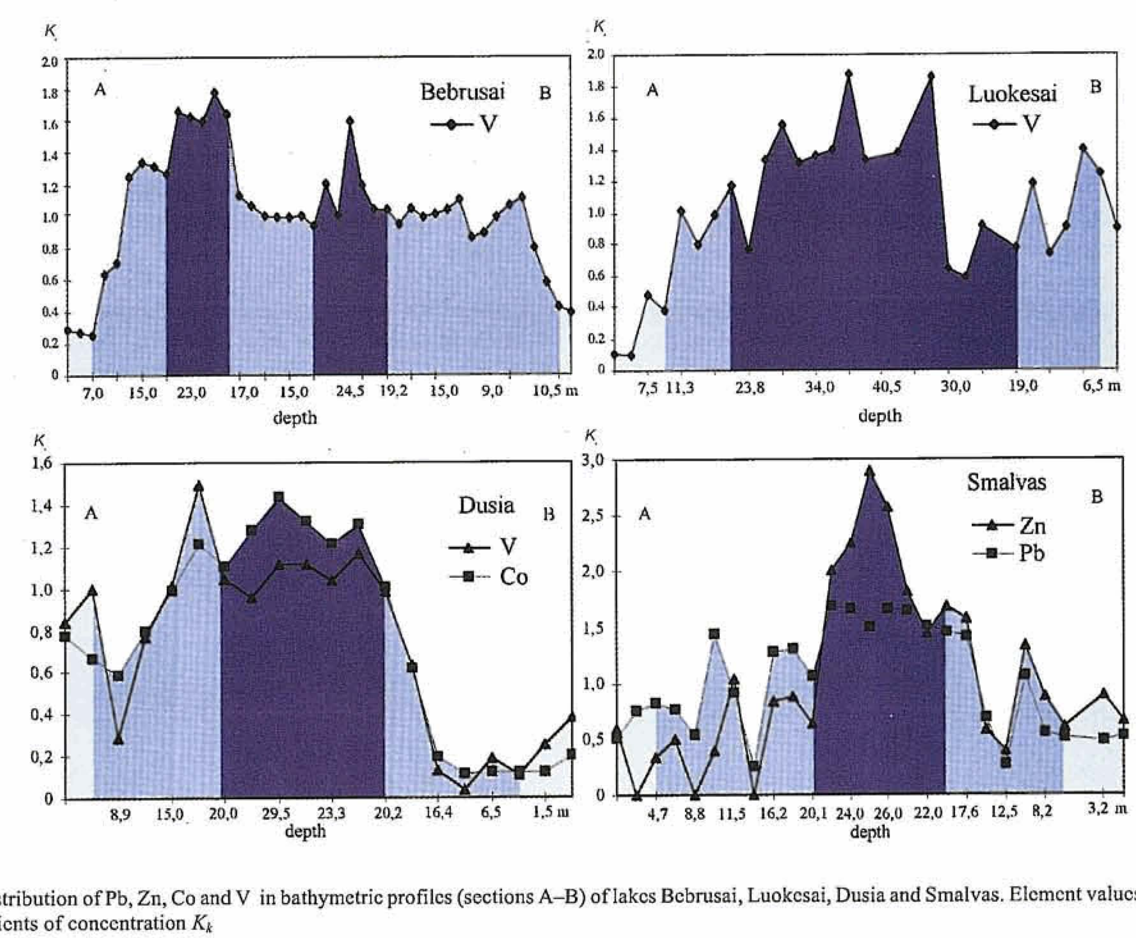

Fig. 4. Distribution of Pb, Zn, Co and V in bathymetric profiles (sections A–B) of lakes Bebrusai, Luokesai, Dusia and Smalvas. Element values are shown as coefficients of concentration K_k

Table 2

Correlation and associations of trace elements in topsoil of eastern Lithuanian and Ašmena–Medininkai highlands

Parvaise correlation	$R = 1–0.9$	$R = 0.89–0.8$	$R = 0.79–0.7$
Sand			
East Lithuania	–	Y–Yb	Cr–(Ga, Ni) La–(Y, Yb) Sc–Sr
Ašmena–Medininkai	–	Cr–(Ba, La, V) Cu–Co–Ni–V Y–Yb Ti–Zr	Ni–(Cr, Cu, B) Ti–(B, La, Cr, Y) Y–(V, Co, La) Yb–Co
Sandy loam			
East Lithuania	–	Li–Ga Co–(V, Cr, Ni)	(B, Ga)–(Co, Cr, V) Ni–(B,V) Co–Zn
Ašmena–Medininkai	V–(Cr, Co) Ga–Pb Cr–Yb	(B, Ga)–(V, Cr) Y–B–Cr V–(Zn, Mo, Yb) Ti–(Cr, Co)	Ga–(B, Zn, Yb, Sc, Li) Ag–Co–Mo Ti–(Yb, La, Mn) Cr–(Pb, Nb, Rb)
Loam-clay			
East Lithuania	Ga–Co	Ga–V–Cr–Co	Ga–Cu Cr–Zn Li-(Co,V)
Ašmena–Medininkai	B–(V, Ni) Cr–(Co, V, Rb) Ni–(Cu, Cr) (Ga, V)–(Co, Cu)	Ga–(B, Li, Ni) Co–Cr–Li Cr–(Y, Yb, B) Cr–V–Sc	Li–(V, Cr, Ni) Sr–Ba Rb–(B, La, Yb, Zn, Cu) Zr–Nb

Factors	F1	F2	F3
Sand			
East Lithuania	Sr–Sc–Rb–V–Ni–Y–La–Cr–Yb–Cu–Ba–Mn	Ag–Li–Mo–Pb–Ga	Zr–Ti–Nb
Ašmena–Medininkai	Yb–Y–Ti–Zr–La–Cr–Mn–Co	Ba–Li–Ga–Cu	V–Mo–Ni
Sandy loam			
East Lithuania	Ga–Cr–Ni–Co–V–Li–B–Ti–Mo–Sc	P–Nb–Pb–Mn–Zn	Sr–Rb–La
Ašmena–Medininkai	La–Zr–Ni–Mn–Ti–Nb–Ba–Co–Sr	Sn–Li–Ga–Pb	Zn–Y–Rb–Ag
Loam-clay			
East Lithuania	Ga–Cr–V–Co–Zn–Li–Pb–Mo–Cu	Sr–La–Y	Zr–Nb–Ti–Rb
Ašmena–Medininkai	Ga–Li–Cu–V–Co–Ni–Pb–Cr–Zn–B–Sn–Yb	Sr–Sc–Ba	Zr–Ti–La

Remark: elements in the parenthesis correlate with element before the parenthesis but not between itself

cator of glacial sediment of similar primary composition but different age.

CHEMICAL COMPOSITION OF SOIL PARENT MATERIAL AS AN INDICATOR OF GLACIATION AGE AND LIMITS

The chemical composition of the soil parent material (horizon C) is less affected by soil forming processes (humification, leaching, translocation) and weathering than top horizon and therefore reflects the primary composition of the Quaternary deposits more clearly. The regional distribution of elements, particularly the major elements in the subsoil reflects the limits of the glaciations (Guobytė, 1998). Calcium, magnesium, iron and trace elements related to clay minerals (Co, V, Ti, and Ni) in subsoil most clearly reflect the glaciation limits, especially as regards phases of the last Late Weichselian glaciation (Fig. 1).

Comparing the values of elements in the top (A) soil horizon and soil parent material (horizon C), it becomes apparent that leaching effects dominate the soils of Lithuania. Heavy precipitation, moderate winters without soil frost and the coarse soil texture has led to rapid decomposition of primary non-silicate minerals such as calcite, dolomite and hematite forming secondary clay minerals and oxyhydroxides, translocated from upper to lower soil horizons (Berrow and Mitchell, 1991). In spite of intensive agriculture and input of trace elements *via* fertilisers, pesticides and pesticides into topsoil, only a few elements show the higher concentrations than in soil the parent material (Tab. 3). Increased levels of silver, lead and zinc in the upper soil layer by comparision with parent material may be anthropogenic in origin, particularly in organic-rich soil. This can be related to regional and transregional pollution (Gregorauskienė *et al.*, 2000). The top of horizon A represents the oldest part of the soil profile, and so usually contains the most weathered material. Therefore, relatively higher concentrations of Zr, Ba, Nb related to weathering-resistant

Table 3

Median values of trace and macroelements in soil parent material (horizon C) and topsoil (horizon A)

Elements	Median values		Ratio of medians	
ppm	horizon C	horizon A	A/C	C/A
Ag	0.066	0.096	1.45	0.69
B	45.8	30.3	0.66	1.51
Ba	291	341	1.17	0.85
Co	8.4	6.7	0.80	1.25
Cr	49.9	40.9	0.82	1.22
Cu	10.7	7.8	0.73	1.38
Ga	9.6	7.8	0.81	1.24
Y	19	15	0.80	1.25
Yb	2.6	2.2	0.84	1.18
La	30.8	24.9	0.81	1.24
Li	19.2	17.2	0.90	1.11
Mn	457	469	1.03	0.97
Mo	0.94	0.75	0.80	1.25
Nb	12.9	13.3	1.03	0.97
Ni	24.4	14.2	0.58	1.71
P	536	495	0.92	1.08
Pb	15.0	18.9	1.26	0.79
Rb	75	65	0.86	1.16
Sc	6.8	6.0	0.87	1.15
Sn	2.11	2.31	1.09	0.91
Sr	96.5	81.5	0.84	1.18
Ti	2733	2705	0.99	1.01
V	60.0	40.6	0.68	1.48
Zn	35.0	32.8	0.94	1.07
Zr	190	256	1.35	0.74
%				
Ca	2.37	0.46	0.19	5.21
Mg	1.01	0.30	0.29	3.42
Na	0.42	0.43	1.01	0.99
K	2.05	1.80	0.88	1.14
Fe	1.59	1.23	0.77	1.30
Al	3.77	3.09	0.82	1.22

minerals occur here. Such chemical ratios between soil top layer and parent material may be used as indicator of the character and intensity of soil forming processes.

TRACE ELEMENT COMPOSITION OF LAKE SEDIMENTS AS AN INDICATOR OF THE DEPTH BATHYMETRY

Investigation of the trace element composition of sediments in deep (> 20 m) Lithuanian lakes (Fig. 2) has shown that the depth of sedimentation relates to the distribution of trace elements: there are depth-related increases in the concentrations of elements such as Mn, Pb, Co, Zn, V, Sn, Ni, Ag, Mo (Tab. 4).

Trace elements values are lowest in the littoral (to 5 m deep) sediments, except for Sr and Nb. In the nearshore zones of some lakes, increased levels of Zr and La reflect natural heavy mineral concentrates in the wave zone. Various trace elements accumulate in sediment of the sublittoral zone (5–20 m deep): allochthonous Sc, Ga, B, Cr, Li, allochthonous-accessory Ti, Zr, Y, La and Ba of carbonates. Most trace elements accumulate in profundal (> 20 m deep) sediment and biogenic-anthropogenic Mo, Mn, Pb, Zn, Sn, Ag clearly prevail here (Fig. 3). Allochthonous and mobile Co and Ni are also present (Fig. 4). Levels of these elements directly correlate with the depth of sedimentation. The trace elements accumulate differently in different sedimentary facies: in calcareous and organic-rich sediment they mostly occur in the deepest zones, and in clayey sediment in shallow and transitional zones. The greater the depth of sedimentation the higher the levels of mobile trace elements sorbed on to the sediment.

Allochthonous-accessory trace elements Ti-Zr-Nb-Y-Yb-La show the strongest correlation in all sediments while the allochthonous trace elements V-Ni-Cr-Ga-Sc show weaker correlation. Trace elements show the strongest correlation (correlation coefficient of > 0.8–0.9) in calcareous sediment, and the least correlation in clayey sediment. Thus, authigenesis reveals the most stable primary relations, in weathering-resistant minerals, and among closely connected trace elements those which are mineral-forming or are an isomorphous admixture. The deeper the sedimentation, the stronger the correlation among trace elements, particularly in organic-rich and calcareous sediment. This reflects the importance of allochthonous-accessory trace elements in organic-rich sediment, and allochthonous-accessory and biogenic-anthropogenic trace elements in calcareous sediment. In clayey sediment this pattern is not so distinct.

The trace element compositions and ratios can, on this basis, be used to reconstruct glaciolacustrine basin bathymetry. Elevated values of Zr, Nb, Y and Yb (Kadūnas *et al.*, 1999) relate to sediments deposited at shallow depths, while increased values of Cu, Zn, Mo, Mn indicate deeper zones.

CONCLUSIONS

Trace element composition of topsoil reflects the duration of soil forming processes, and thus the age of Quaternary deposits. Young soils of eastern Lithuanian highland that formed on the Late Weichselian marginal deposits possess a relatively high content of trace elements in relation to clay minerals. By comparison, the older soils of the Ašmena–Medininkai highland which rest on the Late Saalian genetically similar deposits contain higher amounts of trace elements, related to weathering-resistant minerals, and elements related to secondary phosphates and manganese hydroxides. This is also a consequence of long-term periglacial processes, which changed the mineralogical and textural composition of surface sediment. Therefore the dominant soils in the Ašmena–Medininkai highland are monotonous in texture (sandy loam) with larger amounts of quartz, zircon, apatite, tourmaline and staurolite.

Table 4

Accumulative associations of trace elements in different type and depth of lake sediments

Organic sediments			Calcareous sediments			Clayey sediments			All sediments		
depth [m]											
< 5	5–20	> 20	< 5	5–20	> 20	< 5	5–20	> 20	< 5	5–20	> 20
$Sr_{1.1*}$	$Li_{1.26}$	$Ag_{1.51}$	$Sr_{1.78}$	$Mn_{1.13}$	$Sn_{1.89}$	$Zn_{1.27}$	$Cr_{1.15}$	$Mn_{2.43}$	$Sr_{1.13}$	$Sc_{1.35}$	$Mn_{1.58}$
	$Cr_{1.22}$	$Pb_{1.34}$	$Nb_{1.1}$	$Ba_{1.1}$	$Pb_{1.72}$	$Ga_{1.26}$	$Ni_{1.1}$	$V_{1.17}$	$Nb_{1.1}$	$Cr_{1.34}$	$Pb_{1.41}$
	$Sc_{1.17}$	$Zr_{1.32}$			$Co_{1.66}$	$Ag_{1.13}$	$Sc_{1.1}$	$B_{1.12}$		$Li_{1.28}$	$Co_{1.4}$
	$Ba_{1.14}$	$V_{1.28}$			$Zn_{1.64}$	$La_{1.13}$	$Li_{1.1}$			$Y_{1.22}$	$Zn_{1.38}$
	$Ga_{1.1}$	$Mn_{1.26}$			$Y_{1.55}$	$Mo_{1.1}$				$Yb_{1.16}$	$Sn_{1.32}$
	$Y_{1.1}$	$Mo_{1.24}$			$B_{1.47}$	$Ba_{1.1}$				$Ba_{1.14}$	$Ni_{1.31}$
	$Co_{1.1}$	$B_{1.2}$			$Ti_{1.35}$	$Nb_{1.1}$				$Ga_{1.2}$	$Ag_{1.3}$
		$Ni_{1.19}$			$V_{1.35}$	$Zr_{1.1}$				$La_{1.1}$	$Mo_{1.28}$
		$Yb_{1.14}$			$Zr_{1.33}$					$Zr_{1.1}$	$V_{1.25}$
		$Ti_{1.12}$			$Ni_{1.28}$						$B_{1.16}$
		$Sn_{1.12}$			$Mo_{1.26}$						
					$Sc_{1.21}$						
					$Li_{1.23}$						
					$Yb_{1.21}$						
					$Cr_{1.17}$						
					$Ag_{1.13}$						
					$Cu_{1.12}$						

Coefficients of concentration of trace elements presented in subscript. They are calculated by formula: $K_k = C_i/C_f$, where: C_i — median values of trace element in different type and depth of lake sediments, C_f — exponent median value of trace element in the four investigated lakes sediments

The chemical composition of the soil parent material can precisely reflect the age of Quaternary deposits and so be useful in defining glaciation limits. Comparing the chemical composition of topsoil and of parent material helps ascertain the dominant washout-leaching process in the soils of Lithuania: concentrations of trace elements in topsoil are 8% lower on average, and of macroelements are 34% lower, than in parent material. Only concentrations of trace elements related to weathering-resistant minerals (Nb, Zr, Ba) and biogenic-anthropogenic elements (Mn, Ag, Pb, Sn) are higher in topsoil than in subsoil, indicating the impact of airborne pollutants.

Trace element contents in lake sediments depend on sediment type and correspondingly on the depositional environment. Therefore patterns of chemical variation in modern lake sediments can be used to reconstruct Pleistocene glaciolacustrine basin bathymetry. Increased values of Zr, Nb, Y and Yb may be related to the sedimentation in shallow zone, while the increased levels of Cu, Zn, Mo, Mn characterize the sedimentation of deeper zones.

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