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Considerations regarding the definition of a geochemical baseline of elements in the surficial materials in areas differing in basic geology

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Abstract

The problem of a geochemical baseline in areas differing in basic geology was studied by comparing the results of regional geochemical mapping programmes undertaken in Finland and Lithuania using different sample media and a variety of analytical methods. The authors discuss issues relevant to the definition of a geochemical baseline.

Most of the differences in geochemical baselines between Lithuania and Finland are due to the dissimilarities in basic geology, which in Lithuania is controlled by Palaeozoic and Mesozoic sedimentary rocks, but in Finland by Archaean and Proterozoic metamorphic and intrusive rocks. Other important factors are the geological processes that after the last glaciation created the present surficial materials. In Finland, for instance, the abundances of potentially harmful elements derived from crystalline bedrock tend to be higher than in Lithuania. However, in both countries element concentrations are higher in finely-grained marine and lacustrine sediments than in glacial till, and they are also higher in the finer grain size fractions than in the coarser fractions of till samples. Only a small proportion of the total heavy metal concentrations is bioavailable. Thus the baseline concentrations depend not only on the basic geology, but also on sample material collected, its grain size and the extraction method. © 2000 Elsevier Science Ltd. All rights reserved.

1. Introduction

The term 'geochemical baseline' was introduced in the International Geological Correlation Programme (IGCP) projects 259 and 360 (Darnley et al., 1995) in order to create a global reference network for national regional geochemical data sets and as international background data for environmental legislation. The recommendations of IGCP 259 and 360 projects include the concept of multimedia multipurpose regional geochemical mapping. However, the term geochemical baseline was not well defined.

Tidball and Ebens (1976) used the term regional geochemical baseline in order to describe the natural background in areas of a heavy anthropogenic impact from the coal based electric generating industry in the Powder River Basin in Montana–Wyoming. Their definition was that a baseline encompasses the central 95% of the observed concentrations. The basis of this definition has been discussed earlier by Tidball et al. (1974) and it was

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applied in wide regional geochemical mapping programs in Wyoming and Montana during the 1970s (Severson, 1979; Severson and Tidball, 1979; Ebens and Connor, 1980).

The term geochemical background, in contrast, has been defined in many different ways in textbooks of geochemistry (cf. e.g. Levinson, 1974; Beus and Grigorian, 1977; Rose et al., 1979). The most common definition is the natural abundance of an element in a particular material (e.g. soil, sediment, rock) with reference to a particular area or data set. It is usually expressed as a single value showing the limit (threshold) between anomalous and background concentrations. This definition is used mainly in exploration, and is not appropriate for environmental purposes. A new concept including an anthropogenic factor was therefore developed in Germany (LABO, 1995). According to this definition, the background content of a soil is composed of its geogenic basic content and the ubiquitous substances distributed as a consequence of diffuse entry into the soil. Another definition is that of the International Standardisation Organisation (ISO 11074-1, 1996): "geogeneous or pedogeneous average concentration of a substance in an examined soil". This definition, however, leaves many questions unanswered.

Yet another way to determine the natural abundance of a particular element is to calculate its Clarke value, which is the average concentration of an element in the Earth's crust. The concentrations of elements differ so much, however, from one geologic unit to another that use of the Clarke value of an element in national or local contexts does not permit a sufficiently thorough study of variations in element distributions caused by mineralization or contamination in a particular sampling medium.

Geochemical baseline refers to the prevailing variation in the concentration of an element in the surficial environment. Geochemical baselines are very important in environmental legislation, which prescribes limits for heavy metals in contaminated land and other surficial materials as defined by environmental authorities. In certain areas in Finland, for instance, natural concentrations of several heavy metals exceed the recommended or limit values designated for contaminated land. Thus baselines must always be verified in any assessment of sites for contamination (Gregorauskienė and Kadūnas, 1997; Salminen and Tarvainen, 1997).

The authors present some thoughts on how to establish geochemical baselines with the aid of regional geochemical mapping data from Finland and Lithuania and highlight the problems associated with differences in basic geology on both regional and local variation. In addition, attention is paid to sampling material, grain size fraction and choice of analytical methods.

2. Main features of the basic geology in Lithuania and Finland

The bedrock from which the glacier abraded material is derived, is very different in Finland compared to Lithuania. In Finland, the bedrock consists mainly of intensely metamorphosed Archaean and Proterozoic crystalline rocks varying widely in their original chemical and mineralogical make up. Among the prevailing rock types are greenstones, gneisses and schists of various composition, and granitic rocks. The greenstones occur in large belts in which ultrabasic and basic rocks are common. In Lithuania the bedrock consists of Devonian (clay stones, sandstones) and Cretaceous (chalk, marl) sedimentary rocks and the chemical composition of the bedrock remains the same over extensive areas. It can be assumed that the composition of till derived from such bedrock is likewise rather homogeneous having a characteristic geochemical signature, and very different from that abraded by the icesheet from the bedrock in Finland.

The main constituents of Finnish and Lithuanian surficial material are till, glaciofluvial sediments, marine and limnoglacial clay and organic matter, either alone (peat) or mixed with minerogenic material (mud, gyttja). During the last glaciation the ice divide was situated in Finland, but the marginal area of the glacier was in Lithuania. This resulted, in addition to the basic geology, in the till being more homogeneous in Lithuania than in Finland. As all of the minerogenic constituents of till originally were derived from bedrock during glaciogenic processes, till should represent the average composition of the bedrock. However, changes in the physical composition in the course of geological processes cause major changes in element abundances.

3. Influence of basic geology on the geochemical baselines of elements

The results of regional geochemical mapping of the most common constituents of surficial material from Finland and Lithuania are shown in Table 1. The Finnish and Lithuanian geochemical data are from the unaltered C-horizon of till. Although the difference in the analysed grain size fraction may cause some shift in the concentration level, as is discussed later, the concentration of heavy metals and P are generally twice as high in Finland. However, in contrast, the concentrations of the major elements are similar. HowTable 1

	Si (%)	Ca (%)	Mg (%)	P (ppm)	S (ppm)	Cu (ppm)	Ni (ppm)	V (ppm)	Zn (ppm)
Finnish till ^a	30.2	1.8	1.0	650	140	20	24	79	57
Range	23.7-37.2	0.0-3.1	0.0-3.3	< 70-6460	6-1980	< 3–255	< 1–187	1-217	< 1-224
Lithuanian soil ^b	_	1.9	1.0	360	_	13	14	38	33
Range		0.1-16.3	0.0 - 2.8	100-1830	_	2-26	2-40	5-118	5-102
Lithosphere (Clarke value) ^c	29.5	2.96	1.87	930	320	47	58	90	83
Earth's upper crust ^d	31.0	3.0	1.3	900	300	30	40	100	80

Average abundances (total analysis by XRF) of selected elements in Finnish till and Lithuanian soil. The average element concentrations of the Earth's crust is also presented

^a Median values of < 0.06 mm grain size fraction (Koljonen, 1992).

^b Median values of 249 soil samples (C-horizon) from Lithuania (<1.0 mm grain size fraction).

^c Concentration in the Litosphere (two parts acid rock and one part basic rock) (Rösler and Lange, 1976).

^d Upper continental crust, compiled from various sources (Koljonen, 1992).

ever, compared with the average element concentrations of the Earth's crust, all concentrations (excluding Si) are much lower in both of these countries. Thus the geochemical baseline of the elements, when defined as the average concentration in a certain area, differs remarkably in the area of metamorphosed crystalline rocks from that of sedimentary rocks.

The distribution of element abundances is more homogeneous in glacial drift than in the bedrock from which the till was derived. Glaciofluvial processes have further homogenised the material, and the abundances of many heavy metals in particular, are strongly diluted compared with their abundance in the bedrock. On the other hand, the finest clay fraction washed out from the drift by fluvial processes differs significantly from the coarser fractions. The mineralogical composition has changed and easily soluble micaceous minerals, with their higher abundances of heavy metals, have been enriched relative to quartz and feldspars. Owing to the larger surface area and surface activity of clays, heavy metal ions are adsorbed on the surfaces of the mineral grains and thus are also enriched. There is an essential difference in element enrichment in Finnish marine clays compared with Lithuanian limnoglacial clays. The differences of element abundances in parent material (till) explain the main part of the differences observed in clays but partly it is also due to variation in geological processes prevailing during the sedimentation phase. The enrichment is, in fact, so pronounced, and the finely-grained sediments are often covering such wide areas, that geochemical baselines should be defined separately for the marine clays (see Table 2).

A widespread and common influence of human activities on the geochemical baselines are observed in the information collected from arable soils. During the Baltic Soil Survey (Tarvainen and Kuusisto, 1999) samples were collected from arable soils from both Lithuania and Finland. A top soil sample representing the cultivated layer (0-25 cm) and a subsoil sample representing the natural layer of the surficial material (50-75 cm) were collected from every sampling site. Element abundances were measured from the <2 mm fraction following a hot aqua regia leach. Although the sampling material mainly consists of till, marine, glaciofluvial and glaciolacustrine materials, the typical geochemical features based on the basic geology can be seen in the subsoil samples (Table 3). Because of limestone and dolomite prevalence in bedrock, the Ca and Mg abundances are much higher in Lithuania but the heavy metal and phosphorus concentrations tend to be higher in Finland. Agricultural activities together with soil forming processes are the reason for different element abundances in the top soil layer. During past years the use of fertilisers in Lithuania

Table 2					
Average total element abundances	(XRF) of fine	grained se	diments in	Finland and	Lithuania

	Si (%)	Ca (%)	Mg (%)	P (ppm)	S (ppm)	Cu (ppm)	Ni (ppm)	V (ppm)	Zn (ppm)
Marine clay ^a Limnoglacial clay ^b	26.9	1.1 _	1.8	971 550	340	59 13	56 22	136 56	156 44

^a Median values of 66 clay samples from SW Finland (Salminen et al., 1997).

^b Means of 49 glaciolacustrine clay samples from central Lithuania (Baltakis, 1993).

Table 3

	Ca (%)	Mg (%)	S (ppm)	P (ppm)	Cu (ppm)	Ni (ppm)	V (ppm)	Zn (ppm)	n
Topsoil									
Lithuania	0.30	0.19	174	471	10	7	15	20	27
Finland	0.24	0.28	219	669	12	10	30	26	80
Subsoil									
Lithuania	1.46	0.75	< 50	310	15	11	19	19	27
Finland	0.25	0.43	< 50	471	12	14	35	26	80

Median values of aqua regia leachable element abundances (ICP-AES) in the $\leq 2 \text{ mm}$ fraction of arable minerogenic soils in Lithuania and Finland. *n* is the number of samples

has been very limited compared with that in Finland. Therefore the concentration of the main nutrients (Ca, Mg, Cu) has decreased in the top soil in relation to the subsoil. In Finland, instead, the use of fertilisers has replaced the loss caused by the uptake of plants. Higher P and S abundances in the top soil layer in both countries are due to organic material in the ploughed layer. On the other hand elements which plants use as nutrients only in minor amounts (Ni, V, Zn) have remained nearly unchanged in the top soil and subsoil layers in both countries.

Pure organic matter does not generally contain high abundances of heavy metals. Differences in the data between Lithuania and Finland (Table 4) are minor for most elements, only V and Zn concentrations of stream sediment show slight differences, and these can be related to the differences in basic geology. Thus the basic geology on the regional scale does not have a marked influence on element concentrations of organic material. Such matter does, however, have an exceptional capacity to collect ions (e.g. due to contamination) from water solution by ion exchange or chemical bonding (see e.g. Salminen and Sipilä, 1996; Reimann et al., 1998) and may locally contain a strong anthropogenic influence. The geochemical baseline of an organic-rich constituent of the surficial material, therefore, often differs from that of other constituents.

4. Relevance of the sampling material and analysed grain size fraction

Soil forming processes cause variation in the distribution of elements in the uppermost part of the surficial material. In addition, selected grain size fractions and analytical methods are factors which give varying results and they should be taken into account when defining geochemical baselines.

Although the variation caused by soil-forming processes is a result of natural development, the changes in element concentrations are remarkable

 Table 4

 Total element abundances of different organic constituents of surficial material

	Si (%)	Ca (%)	Mg (ppm)	S (ppm)	P (ppm)	Cu (ppm)	Ni (ppm)	V (ppm)	Zn (ppm)
Stream sediment ^a	0.21	0.6	0.38	1260	870	12	14	44	46
Stream sediment ^b	-	_	_	_	779	8	13	33	42
Peat ^c	-	0.3	0.1	_	606	5	3	6	6
Peat ^d	-	_	_	_	528	6	5	12	12
Humus ^e	0.02	0.3	0.1	1700	900	8	6	6	45
Humus ^f	0.05	0.3	0.08	1530	930	10	9	5	46

^a Median values of element abundances (in dry matter) of 1268 organic stream sediment samples from all of Finland (Lahermo et al., 1996).

^b Median values of element abundances (in dry matter) of 1765 organic rich stream sediment samples from all of Lithuania (Kadūnas et al., 1999).

^c Median values (in dry matter) of 641 peat samples from W-Finland (Virtanen, in preparation).

^d Median values of 64 unpolluted peat samples from the Šiauliai area, central Lithuania.

^e Mean values of element abundances (in dry matter) of uncontaminated humus in Finland, compiled from various sources.

^f Median values of element abundances (in dry matter) of 617 humus samples from the Ecogeochemistry Kola project area (Reimann et al., 1998).

Median values of element concentrations (total analysis, XRF) in soil horizons of the surficial material in Lithuania (1) and in the Kola Ecogeochemistry area (Reimann et al., 1998) (2). The data include samples from different soil types. The analysed grain size in the Lithuanian data was < 1.0 mm and in the Kola Ecogeochemistry data < 2.0 mm. *n* is the number of samples

	Ca (%)		Mg (%)		Cu (ppm)		Ni (ppm)		Zn (ppm)		Р		п
	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	
A horizon	0.46	0.30	0.30	0.08	8	9.7	9	9	32	46	470	930	54
E horizon	0.31	_	0.30	_	8	_	10	-	26	-	220	_	34
B horizon	0.42	1.93	0.46	1.04	9	10	12	16	30	26	320	436	50
C horizon	1.93	2.18	0.95	1.15	13	16	13	19	32	66	350	393	63

(Table 5), so much so that every soil horizon actually has its own geochemical baseline. The variation is different if comparing data from Lithuania and northern Finland (the Kola Ecogeochemistry area, Reimann et al., 1998). The different mineral composition and differences in climatic conditions are factors causing differences in the variation of element concentrations.

The focus of geochemical mapping is often on exploration, and analyses are therefore made very often on the finest fraction (<0.06 mm) of a minerogenic sample. This fraction contains trace element concentrations that are relatively high and the anomaly/background contrast usually is at its highest. Environmental researchers and soil scientists, however, often prefer to analyse a coarser fraction (<2 mm) that contains more feldspar, quartz and rock fragments and thus better represents the lithology of the surficial material (see Salminen and Tarvainen, 1997). In this coarser fraction the heavy metal concentrations are often lower, and there are clear differences in element concentrations between the two grain size fractions (Table 6). This effect does not depend on the basic geology where the abundances of heavy metals are concerned (Tables 6 and 7).

The concentrations of some elements depend on the lithologic composition of the surficial material, i.e. during geological processes these elements remain in the mineral lattices. Such elements include Zr, Ti, Nb, Yb and Y (Baltakis, 1993). Other elements, e.g. Cu,

Ni, Zn, Co, V and Li, are partly leached from the minerals and precipitated as colloidal particles; they are then either bound on the surfaces of minerals or they form chemical compounds with organic matter. The abundances of elements tightly bound in mineral lattices do not depend so much on the mechanical composition of the soils. Their abundances in soils rich in organic matter are relatively low, whereas those of other elements are nearly twice as high in loamy soils and in organic soils (Table 7). Thus, the baseline of an element also depends on the mechanical composition of the surficial material.

Several analytical methods are available for regional geochemical mapping, the ultimate choice depending on the purpose of the survey. Total analysis (e.g. XRF) gives absolute element abundances and a true picture of natural relations of elements in the sample analysed. For many practical purposes, however, methods based on a partial leach in which only a soluble part of the element is extracted and measured, are preferred. Such extractable abundances are often used in prospecting and in the mapping of contaminated land. A standard method for determining element concentrations in contaminated land is hot aqua regia leach (ISO/DIS 11466, 1992). Much weaker leaches are used in studies of nutrient concentrations in the surficial material when an assessment of the bioavailable concentrations of elements is needed. The geochemical baseline of elements can thus be determined in many different ways, the analytical methods chosen depending on the final objective (Table 8).

Table 6

Median values of fine (<0.06 mm) and coarse (<2 mm) till fractions. Total concentrations (total dissolution, ICP-MS) of 638 till samples from southern and central Finland (Tarvainen, 1995)

	Si (%)	Ca (%)	Mg (%)	Cu (ppm)	Ni (ppm)	V (ppm)	Zn (ppm)
< 0.06 mm	31.1	1.72	0.80	16.7	13.6	68.2	54
< 2 mm	34.2	1.43	0.56	11.5	9.7	42.4	40

Table 7

Median values of total element abundances (XRF) in soils (<1.0 mm fraction) from all of Lithuania differing in mechanical composition. *n* is the number of samples. (Kadūnas et al., 1999)

	Sn (ppm)	Nb (ppm)	Zr (ppm)	P (ppm)	Cu (ppm)	Ni (ppm)	V (ppm)	Zn (ppm)	п
Sand	1.9	12.5	222	622	6.5	9.1	26	21	676
Sandy loam	2.1	13.7	280	552	9.4	13.3	37	28	699
Loam	2.3	13.6	263	527	11.3	16.9	45	33	678
Organogenic (mean of loss on ignition 46%)	1.5	6.8	142	745	8.7	10.9	27	29	630

5. Discussion

In the secondary environment the geochemical background changes regionally with the basic geology and locally with the type and genesis of the surficial material. Whereas geochemical baselines in areas of sedimentary rocks (as in Lithuania) may remain constant over large areas, in shield areas (as in Finland) they exhibit considerable variation.

The geochemical baseline of an element in the surficial material describes the natural variation of this element in a particular natural material, thus taking into account the geochemical variation in basic geology and the different geological units. With existing geochemical mapping data in mind, the authors are of the opinion that the geochemical baseline should be considered not as a single value, but as a fluctuating reference surface over a wide area, to which local geochemical mapping data sets could then be referred. However, if single values are used to describe the baseline, the arithmetic mean is appropriate only for normally distributed populations. Because the lognormal distribution of elements in geological materials is more common, the median value is more correct than the mean. Tidball and Ebens (1976) used the geometric mean in their calculations of the geochemical baseline, which again is a much better basis for the geochemical baseline.

However, in fact the geochemical baseline should be defined in some other way, taking into account the variation of element concentrations in natural geological materials. An attempt in this direction was made by Tidball et al. (1974) and Tidball and Ebens (1976), when they proposed that a baseline encompass the central 95% of the observed concentrations. The expected range sets limits on the frequency distribution so that values inside the limits are defined as common and values outside are defined as uncommon. This kind of calculation takes into account the natural variation of elements and it is a correct way to evaluate natural background when the legislation for contaminated land is discussed. However, for a global geochemical baseline to which the local data sets could be levelled this definition cannot be applied.

The picture is further complicated by regional geochemical mapping data, which indicate that natural background concentrations vary widely due to differences in basic geology; moreover, several different backgrounds can be established, depending on the sampled constituent of the surficial material, the grain size, the analytical method used and so on. Correct determination of the background value is not just a scientific problem; it also has economic implications. Consider, for instance, the financial loss of a landowner whose soil is declared contaminated.

The geochemical baseline of an element in surficial material, such as stream and flood plain sediment, top soil or surface water, describes the prevailing situation in light of a particular analytical method. The geochemical baseline takes into account the geochemical variation in basic geology, the different geologic units, and the influence of soil-forming processes, but it also includes factors such as the influence of mineralization and anthropogenic influences. There are, however, some sampling media (e.g. subsoil samples of residual surficial material, pristine layers of overbank and flood

Table 8

Total (XRF), extractable (hot aqua regia leachable) and bioavailable (NH_4 -acetate leachable) element concentrations in marine clays from SW Finland, median values of 66 samples (Salminen et al., 1997)

	Si (%)	Ca (%)	Mg (%)	P (ppm)	Cu (ppm)	Ni (ppm)	V (ppm)	Zn (ppm)
Total	26.9	1.1	1.76	971	59	56	136	156
Extractable	0.07	0.59	0.83	970	54	41	85	92
Bioavailable	0.01	0.16	0.08	12	1	< 1	< 1	1

plain sediment) that allow the natural geochemical baseline to be determined without the influence of anthropogenic or soil-forming processes.

An attempt to determine global or continental geochemical baselines according to the guidelines given by Darnley et al. (1995) is currently being undertaken by the IUGS Working Group on the Global Geochemical Baseline set up in Europe by the FOREGS Geochemistry Task Force (Salminen et al., 1998). Even so, we still need a more precise definition for the term 'geochemical baseline'. It would seem reasonable, especially for political decision-making, to determine many separate baselines. Thus there could be have local and regional baselines, different baselines for the various constituents of the surficial material, and yet others depending on the analytical method used. These could then serve as points of reference by which local geochemical data sets could be levelled.

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