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Original Research

Different Approaches in Using and Understanding the Term “Geochemical Background” – Practical Implications for Environmental Studies

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Abstract

This article presents various aspects of the use and evaluation of geochemical background for different environmental media with a brief discussion on the origins, historical and present context, and application of this and other related terms. This subject has become one of the most crucial issues in geochemistry and environmental sciences during the last few years because of its application to environmental protection, land management, ecotoxicology and geochemistry. Three approaches (methods) of geochemical background evaluation are discussed: (i) direct, (ii) indirect, and (iii) integrated. In response to a strong need to reduce a large number of methods applied to background assessment, a plausible methodology based on archival data, including the results derived from determinations of Mn, Pb, Zn for quartzites, soils and *Pinus sylvestris* needles, from the Holy Cross Mountains (Świętokrzyskie Mountains, south-central Poland) has been proposed. This methodology belongs to the integrated approach to the background evaluation. Samples were collected in relatively pristine areas and statistical data analysis (iterative 2σ-technique) was used to eliminate outliers from the original datasets.

Keywords: geochemical background, geochemical methods, statistical methods, integrated approach

Introduction

The term “geochemical background” was originally introduced by exploration geochemists in the mid 20th century to differentiate between the abundance of an element in unmineralized and mineralized rock formations. During the last few decades “geochemical background” has become one of the most crucial terms in environmental sciences. It is sometimes used to distinguish anthropogenic input (pollution) from natural (geogenic and/or biogenic) concentrations of elements in different environmental samples [1-3].

The process of adopting the term “geochemical background” to environmental sciences has resulted in broadening its meaning and application to different materials. At present the term “geochemical background” is applied not only to rocks, minerals and sediments, but also to water, air, and even to plants despite many constraints put on the selection of specific samples [4, 5].

Different aspects of the evaluation and application of geochemical background have recently been discussed in many scientific journals [4-6], but still both geochemists and environmentalists have problems with a precise definition, as well as a coherent method for assessing geochemical background in surface or near-surface environments [4, 5, 7–9]. The discrepancy in assumptions, approaches, and methods is a problem which can only be

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eliminated by accepting a specific definition and method of background evaluation. Is it possible to achieve this purpose when scientists are still far away from reaching a consensus after at least 50 years of using the term "geochemical background"? There is a Latin citation from Terentius that is adequate for the present situation "*Quot capita, tot sensus*" ("There are as many opinions as there are heads").

Evaluation of geochemical background is important because it has many implications for environmental risk assessment and setting regulatory levels of pollutants (with their financial consequences). In Poland, for example, the accepted quality criteria for soils recognizes high concentrations of pollutants derived only from anthropogenic sources. However, soil quality standards say nothing about distinguishing natural from non-natural levels of a given substance [10]. Such imprecise criteria in environmental law give opportunities to environmentally dishonest individuals and corporations responsible for causing pollution and for accomplishing very expensive land reclamation. On the other hand, no reclamation is needed for the land showing high levels of natural substance of geogenic origin; moreover, this may jeopardize local biota (including plant bioaccumulators), which is adapted to high element concentrations [11]. Reimann and Garrett [5] give the example of Austria as a country that belongs to an "As-geochemical province" and claim that remediation of arsenic may well worsen the environmental situation there. Another example is the Holy Cross Mountains, which may generally be identified as a "Pb-geochemical province"; however, scattered vein mineralization containing galena does not jeopardize the environment due to the prevailing carbonate host rocks, *i.e.* limestones and subordinate dolomites [12].

Three groups (methods) of background evaluation have recently been used: (i) direct (geochemical), (ii) indirect (statistical) and (iii) integrated (for details see the following sections). Each of them requires different assumptions and may be criticized for different vulnerable points. The principal objective of this article is to present and discuss the features of the method that could be accepted for plausible background evaluation.

Review of "Geochemical Background" Definitions and Related Terms

The term "geochemical background" has been used by many authors mostly without providing a precise definition. The most common meaning of this term refers to a natural range of values for a given medium not impacted by anthropogenic activities [13]. In its original meaning (in exploration geochemistry and geology) it is understood as "the normal concentration of a given element in a material under investigation such as rock, soil, plants, and water" [14, 15]. In other words, it is a reference level above which element concentrations are likely to represent mineralization in the study area [16].

Matschullat et al. [4] presented a definition of geochemical background used in environmental sciences: "a relative measure to distinguish between natural element or compound concentrations and anthropologically-influenced concentrations in real sample collectives." In another definition proposed by Gałuszka [17] "geochemical background is a theoretical 'natural' concentration of a substance in a specific environmental sample (or medium), considering the spatial and temporal variables, which may be determined with direct, indirect, and integrated methods."

As indicated in the previous paragraphs, the same term is differently understood by geochemists and environmentalists. In this context, it would be necessary to discuss whether "geochemical background" should be commonly used only in its original meaning or be substituted for "background concentrations," more specifically in environmental sciences. This simple action would also end up a long debate on the application of "geochemical background" to waters, air or bioindicators.

Table 1 presents different approaches to the term "geochemical background" in selected environmental studies. A brief look through the meaning of the different variants shown in Table 1 leads to the conclusion that the terms "ambient background," "anthropogenic background" and "area background" are in fact synonyms that refer to both natural substances and ubiquitous pollutants released from unidentified regional-scale sources. The term "baseline" ("geochemical baseline") seems to be more proper than "ambient," "area" or "anthropogenic background" (see the next paragraph). Consequently, the terms: "natural background," "naturally occurring background" and "pre-industrial background" ought to be substituted for "geochemical background" or "background concentrations."

The term "geochemical baseline" is often used as a synonym of "geochemical background" [21]. Originally, it was used by Tidball et al. [22] in a study on the influence of a coal-fired powerplant on the environment in the Powder River Basin (Montana and Wyoming) and it was defined as "a natural background in a heavy anthropogenically polluted area." Gough and Erdman [23] used an eight-level analysis-of-variance design to take into account the predominant scales of geochemical variability inherent in nature. Gough and Crock used the term "baseline" for element concentrations determined outside the zone of measurable industrial influence (about 6 km from a coal-fired power plant) in a study in Alaska near Denali National Park [24]. The general opinion of these studies is that "baselines" are best expressed as expected ranges of element concentrations, such as the central 95-percent range. Another definition of "baseline" is given by Lee and Helsel [25]: "a summary of existing conditions over some time frame for some environmental system, or material of interest."

There is no agreement among scientists whether the term "baseline" should be used. According to Darnley [26], "geochemical baselines" are needed for "documenting the pres-

Table 1. Different variants of "geochemical background" used in the literature of the subject.

Variant	Meaning	References
Ambient background	"The concentrations of naturally occurring inorganic substances and ubiquitous anthropogenic inorganic substances in the environment that are representative of the region surrounding the site and not attributable to an identifiable release".	[18]
Anthropogenic background	"Concentrations typically observed in a region that are the result of human activities but that are not associated with a specific contamination activity". "Chemicals present in the environment due to human activities that are not related to specific point sources or site releases".	[8] [19]
Area background	"The concentrations of hazardous substances that are consistently present in the environment in the vicinity of a site which are the result of human activities unrelated to releases from that site".	[20]
Natural background	"The amount of naturally occurring substances in the environment, exclusive of those from anthropogenic sources". "The concentration of hazardous substance consistently present in the environment that has not been influenced by localized human activities".	[18] [20]
Naturally occurring background	"Ambient concentrations of chemicals present in the environment that have not been influenced by human activity".	[19]
Pedogeochemical background	"Natural concentrations of elements in soils".	[1]
Pre-industrial background	"… is sometimes used when data either come from age-dated materials or are collected from areas believed to represent survey/study area in its supposed 'preindustrialization' state".	[5]

Table 2. Different approaches to the background evaluation with short characterization.

Approach	Aspect/technique	Background expression	Principal requirements
Direct (geochemical)	Historical aspect; Contemporary aspect	Mean or median (single values)	Not anthropogenically influenced samples
Indirect (statistical)	Regression analysis; Fractal method; Probability plots; Techniques used to eliminate the outliers	Range of values	Large datasets
Integrated	–	Upper limit of the range of values	Pristine areas for sample collection. Expert knowledge

ent state of the surface environment and to provide datum against which any changes can be measured". Reimann and Garrett [5] do not support the use of this term because they stress that it is not possible to find a single concentration that separates natural from non-natural concentrations in a heterogeneous environment. However, it should be stressed that "geochemical baselines" (or "baseline concentrations") have commonly been used as reference levels for future monitoring in many environmental studies conducted in the USA, Germany and Poland [*e.g.* 23, 24, 27–30].

Review of Methods for Assessing Geochemical Background

The methods of background evaluation can be divided into (i) direct, (ii) indirect and (iii) integrated (Table 2) [17, 31]. The direct (geochemical) ones are based on the analysis of samples recognized as not anthropogenically influenced. In these studies, the background concentrations are usually presented as mean or median values. Two aspects may be distinguished here:

i. Historical aspect – study of samples collected before or dated back to pre-industrial times, *i.e.* prior to the mid 19th century (*e.g.* dated sediments, archival plants from herbaria, glacial ice cores, etc.);
ii. Contemporary aspect – the samples are collected from relatively pristine sites.

The vulnerable point of the first approach is that in prehistoric times with the beginning of primitive metal smelting, the heat processing of metal ore as well as manufacturing and burning charcoal released substantial amounts of pollutants to the environment. It is well known that raised levels of lead dating back to the ancient Greek and Roman periods were found in Greenland ice cores [32], as well as copper dating from Roman and medieval times [33]. However, collecting pre-industrial samples and dated materials does not guarantee that they are not anthropogenically impacted. The second aspect of this approach is often criticized for subjective decision criteria of selecting unpolluted study area as well as for subjective sample selection criteria, high costs, and heavy laboratory workload.

Although a contemporary aspect of the direct approach to background evaluation is controversial from the geochemical point of view (differences in element concentrations between remote areas may not be linked to their anthropogenic influence, but to variations in natural factors such as geological setting, mineralization zones), it is often used for setting a reference level of substances in different environmental studies [34, 35, 36]. The other examples of the contemporary aspect used in geochemical background evaluation are the studies carried out in mineralized areas by Kelley and others in northwest Alaska [37] and Kelley and others in northern Chile [38]. The geochemical background was established directly in undisturbed (unmined) areas.

There are also vulnerable points in the historical aspect of the direct approach to background evaluation. Many environmental studies were performed to evaluate anthropogenic influence on the basis of comparison between the upper and bottom layers of sediments [2, 39], peat cores [40], deep and surface soil horizons [1, 41]. As shown by Reimann and de Caritat [3] enrichment factors, commonly calculated for soils reflect their natural soil properties (*e.g.* the presence of natural sorbents – Fe- and Mn-hydroxides, organic matter), not historical changes in element concentrations.

The indirect (statistical) methods encompass various techniques, *i.e.* regression analysis (partial least squares regression analysis) [42], fractal method [9], probability plots [7], but most of all, techniques used to eliminate the outliers that are considered to be anthropogenically influenced [4]. Statistical methods of background evaluation are criticized by geochemists for neglecting the significance of natural processes that influence distribution of elements or chemical compounds in environmental materials and for not considering uncertainty of sample treatment stages, including sampling, sample preparation and chemical analysis [43]. In other words, supporters of this approach are mostly interested in numbers, not in a large variety of factors that influence concentrations of substances in environmental samples.

Various statistical methods are used to evaluate geochemical background [4, 6]; however, it is impossible to describe them in detail in one article. In general, the statistical approach to background evaluation takes into account the distribution (presented as histograms, boxplots, probablility plots etc.) and techniques for identification and elimination of the outliers from original datasets. The outliers are recognized as anthropogenically influenced. The detailed revision of statistical methods of background (and threshold) evaluation was published by Reimann and others [6]. Their view is that constructing boxplots and cumulative probability plots together with information gained from geochemical maps are the best way of background evaluation.

The integrated method of geochemical background evaluation combines both the direct and indirect approach. However, the prerequisite for this is that the samples must be collected in relatively pristine areas (*e.g.* national parks, nature preserves, forest ecosystems), restricting purposely the range of obtained data that are subjected to statistical analysis [31, 44]. Selecting the study area in mature forest ecosystems guarantees no direct anthropogenic influence. This method is just now being introduced to assess background concentrations in some regions of Poland [17, 31, 44], and Luxembourg [45] and it seems to be very promising for geochemical and environmental studies. The most important advantages of this method are: (i) it is simple and clear to understand and perform; (ii) it takes into account expert knowledge on the behavior of elements in the environment, affecting the higher robustness of background evaluation; (iii) it uses a statistical approach, influencing the higher precision of background evaluation; (iv) it provides no direct anthropogenic impact on the study area; (v) it allows us to evaluate background ranges on regional and local scales, including the specificity of the region.

No matter what method of background evaluation is chosen, there are some rules to obey. When selecting the area for background evaluation, a historical context of this area (industrial, residential, commercial) should be studied, and archive sources (historical maps, documents, manuscripts) used. In the author's opinion the historical industrial use of the area exclude it from further background studies. The most important natural factor that influences the raised concentrations of elements in environmental materials is the geologic setting of a given area. It should be stressed that geologic settings are the same for the pristine and contaminated sites, but without geologic knowledge the raised concentrations of elements in mineralized areas may be attributed to environmental pollution. When positive geochemical anomalies are highlighted by zones of metalliferous mineralization, an assessment of geochemical background must be employed with caution and must include detailed preliminary investigations. Natural variability in the calculation of element concentrations in environmental materials and complexity of geochemical processes (*e.g.* weathering, transport, mobility, deposition, remobilization of elements) must also be considered.

Helpful tools in geochemical background evaluation are geochemical maps of the study areas. These maps are prepared on regional-, national- and sub-continental scales. In Poland, for example, geochemical maps of the entire country [46] and several regions or cities have been produced [*e.g.* 47, 48]. The information gained from both geochemical and geological maps may be decisive, particularly in indirect approach of geochemical background evaluation and in classification of outliers (anthropogenic or natural). In addition, this prevents the misinterpretation of statistical analysis of original datasets.

It is important to realize that it is impossible to determine background with an absolute certainty. As a matter of fact the geochemical background [17] represents a theoretical range of values which may be obtained with different methods. Nonetheless, to obtain a comparable and reliable geochemical background one specific method should be used.

Method Proposal for Background Evaluation

This section presents methodology for geochemical background evaluation based on the archival data derived from geochemical studies conducted in the Holy Cross Mountains of south-central Poland [49]. Background concentrations have been assessed for Mn, Pb and Zn in different environmental materials, *i.e.* Cambrian quartzites (bedrocks), the soil horizons-A and -B, and one-year old Scots pine (*Pinus sylvestris* L.) needles. The samples were collected in the Main Range of the Holy Cross Mountains. The results (including a much wider range of chemical and isotopic determinations) were published and interpreted, but not in terms of geochemical background assessment. The detailed description of the study area, sampling, sample preparation and analytical methods were presented by Migaszewski and Pasławski [29]. All the results used for background calculations were obtained from the sites of identical geologic settings and soil taxonomy.

Of the statistical techniques, the iterative 2σ-technique was used for background calculation. This technique, presented in detail by Matschullat and others [4] is based on the assumption that all values in a dataset beyond the mean ±2σ are iteratively omitted until all the values lie within this range (approaching a normal distribution). This technique of background evaluation was selected because it is plausible and realistic [4, 17, 31, 44]. Geochemists used the sole formula: mean ±2σ to distinguish background from anomalies since the mid 20th century [16]. Although this approach has lately been criticized by Reimann and others [6], its application to background evaluation in the form of the iterative 2σ-technique seems to be reasonable. As originally shown by Matschullat and others [4] as well as by Gałuszka [17], the ranges of geochemical background obtained with this technique lie in the class of the highest frequency of the distribution function and they do not differ from those established on the basis of cumulative probability plots.

The results of background evaluation with some additional data (descriptive statistics) are presented in Table 3. The lowest background values were obtained for the rock and soil horizon-B samples. The highest background concentrations of Mn and Zn (important trace elements for plant metabolism) were found in Scots pine needles. In contrast, Pb (non-essential element) was scarce in this bioindicator, mostly below detection limit (<3 mg kg^{-1}). It is interesting to note that the outliers were detected for Mn (quartzites – 8 values, soil horizon-A – 5 values, soil horizon-B – 6 values); Pb (horizons-A and -B – 2 values) and Zn (quartzites – 2 values, horizon-A – 1 value). In statistical methods outliers are considered as anthropogenically influenced, but here, the presence of outliers in Cambrian quartzites represents natural variability of element concentrations that may be attributed to geochemical

Table 3. Background concentrations and descriptive statistics for Mn, Pb and Zn in different environmental materials from the Holy Cross Mountains (south-central Poland).

Sample	Range	Mean	Median	Standard deviation	Upper limit of background range	Number of samples	Number of eliminated values
Mn (mg·kg^{-1})							
Quartzites	5 – 420	51	20	90	**26**	22	8
Soil/ horizon-B	11 – 1040	255	112	315	**183**	19	6
Soil/ horizon-A	20 – 964	261	207	220	**330**	24	5
Pine needles	110 – 837	426	383	215	**856**	16	0
Pb (mg·kg^{-1})							
Quartzites	3 – 91	39	39	28	**94**	22	0
Soil/ horizon-B	8 – 285	50	34	63	**64**	21	2
Soil/ horizon-A	33 – 466	202	202	105	**335**	22	2
Pine needles*	<3 – 4	–	–	–	–	16	–
Zn (mg·kg^{-1})							
Quartzites	5 – 49	14	11	10	**22**	22	2
Soil/ horizon-B	9 – 47	31	35	12	**55**	21	0
Soil/ horizon-A	30 – 157	85	85	29	**130**	22	1
Pine needles	36 – 45	43	41	8	**59**	16	0

* background was not calculated due to a large number of censored values

processes occurring during deposition and diagenesis. Much higher background concentrations in the horizon-A compared to the horizon-B are also linked to natural features of these two soil horizons. The horizon-A is distinctly enriched in organic matter, clay minerals, and oxides and hydroxides. Many elements tend to remain immobilized in this horizon by chelation with organic matter, adsorption on clay minerals, and precipitation as oxides and hydroxides [50]. The TOP/BOT ratios or Enrichment Factors (EF) frequently used as an anthropogenic imprint in soils have lately been criticized by Reimann and Garrett [5] and Reimann and de Caritat [3]. Considering this, without knowledge of geochemical factors and processes it is easy to misinterpret the results obtained.

Conclusions

The following conclusions can be drawn from discussion on the use and evaluation of geochemical background and empirical data obtained from the study in the Holy Cross Mountains:

1. There is a strong need for providing a terminological arrangement to establish a precise definition and a range of application for the term "geochemical background" in environmental sciences versus exploration geochemistry. The author suggests that "geochemical background" be used only in exploration geochemistry, whereas "background concentrations" should be used in environmental sciences.
2. In order to avoid confusion, selected methods should be officially proposed for background evaluation. Of the different methods, the integrated one seems to be feasible and reliable. This method may be used in areas of little anthropogenic imprint; however, geochemical knowledge and a holistic approach to the environment is required.
3. The present study shows that the background ranges are variable for different environmental samples collected at the same site. This indicates that samples must be collected from the same soil horizon, plant species, rock formations. Calculation of geochemical background requires using the same methods of sampling, sample preparation (*e.g.* grinding, digestion, ashing, washing), and chemical analyses.

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