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Correlation Analysis of Trace Element Content in Caustobioliths: The Deep Fluid Involvement, Conjunction of Features of Organic and Inorganic Models of Oil Genesis

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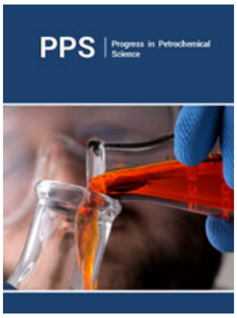

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Abstract

The database has been compiled containing the Trace Elements (TE) content of coals, shales, mud-volcanic and carbonic waters, crude oils, and oil degradation products, more than 300 analyzes in total. A correlation analysis of the TE content of these samples with the model compositions of the upper, middle, and the lower continental crust and with different types of biotas (terrestrial and marine, plants and animals) was carried out. For coals, shales, and clays the expected maximum correlation with the upper crust content was found (correlation coefficient R_Upper). On the contrary, for the overwhelming majority of oils, the TE content correlates better with the chemical content of the lower crust (R_Lower>R_Upper). A reliable trend was found when a decrease in the correlation with biota (maxR_biota) corresponds to an increase in the difference between the correlation coefficients with the lower and the upper crust, R_Lower -R_Upper. The results of calculations for oil degradation products (asphalts) are characterized by a large spread of values, and even larger values of R_Lower -R_Upper, and lower maxR_biota values. In some cases, the calculation of correlation coefficients makes it possible to indicate the dominant type of initial organic matter, viz., terrestrial or aquatic. The obtained results are interpreted as evidence in favor of the necessity of existence of an ascending flow of the deep fluid in the process of oil formation. Seismological evidence in support of the existence of such ascending deep fluid flows are given. The results testify for conjunction of important features of organic and inorganic models of oil genesis in the real process of oil genesis.

Keywords: Caustobioliths; Trace elements; Database; Hydrocarbons; Mud volcanic; Carbonic fluids; Correlation analysis; Biota

Abbreviations: TE: Trace Element; OM: Organic Matter; CC: Correlation Coefficients; DB: Data Base; CMW: Caucasian Mineral Waters

Introduction

The debate on the organic or inorganic origins of oil has lasted for about 300 years. Both of these models have their own factual basis and arguments, and both models encounter serious objections [1-6]. Despite the fact that no commercial oil and gas fields of definitely inorganic origin have been found so far, the inorganic origin model continues to be widely discussed [5,6]. As a possible compromise, the possibility of oil polygenesis was suggested [3,6]. It was hypothesized that inorganic oil migrates from the deep strata and permeates into the oil fields reservoirs where the organic genesis oil, however, can be dominant. An important aspect of the discussion on oil genesis is connected with the study of the Trace Element (TE) composition of oils. Unlike hydrocarbons, the polygenesis of which remains a hypothesis, the polygenesis of the TE content of oils appears to be firmly established [7-15]. Main part of TEs in oils is inherited from an Organic Matter (OM), that is testified by the predominance of biogenic elements in bulk TE content, and a high correlation between TE concentrations in oils and the mean chemical composition of OM; other TEs were imported into oil from host rocks and formation water. Some TEs in oils indicate, however, the presence of a deep-seated source [8-12,15]. This inference has, however, merely qualitative character; it was not clear what is the relationship between the deep and the biogenic factor, as well as what is the change in their relative contributions under different conditions. Answers to some of these questions were obtained by the correlation analysis of the Trace Element (TE) composition in oils and other caustobiolites on the one hand and, on the other hand, the model chemical composition of the upper, middle, and the lower continental crust [16], and the chemical composition of biota [17] of different types (terrestrial and marine, plants and animals).

The Method and Data Base

The method used in geochemical studies of TE content consists usually in comparing the concentrations of certain characteristic elements and their ratios. However, the concentrations of many (rather nearly all) TEs in different samples frequently show great variability being dependent on numerous local factors that are frequently unknown or poorly known. It follows that the ensuing results are frequently unstable, being occasionally ambiguous and contradictory. Correlation analysis, which uses data on the totality of all known trace elements can provide more stable results, even though at the cost of poorer sensitivity [13,18]. The calculations of Correlation Coefficients (CC) to be reported below were conducted for samples with the number of determinations of the concentrations of at least 30 different trace elements. The calculation of the CCs was performed on a log scale, thus enabling us to take into account data on elements with very low concentrations. Besides this peculiarity, the standard method of correlation analysis was used. No preprocess procedures of data corrections were used. To obtain more representative results we had compiled a Data Base (DB) for trace element concentrations in different naturally occurring objects. For these data we calculated correlations of TEs content in samples with model compositions of the upper, middle, and the lower continental crust [16], and with four types of biotas, aquatic and terrestrial organisms, animals and plants [17]. In addition to data for crude oil, the DB contains results of TE analyses of carbonic and mud-volcanic waters of the Greater Caucasus region, data from analyses of the resin-asphaltene component of oils, and data on natural bitumen (asphalts). The data in DB are more complete for territory of Russia. The analytical material used was borrowed from [9-12,15,19-25], with a total of over 300 analyses. Besides a few data sets for the territories of United States and Trinidad are taken from [26,27]. Analyzes only with a large number of concentration determinations of elements (as a rule, at least 30 different elements) were incorporated into the database. In the figures below only part of the information from the database is used. Preference was given to later works, preferably with a large number of analyses from the same team of authors. These limitations presumably provide better homogeneity and accuracy of the results of the analysis. Other data sets display similar regularities in the change of the TEs composition with those shown below, but with a larger scatter. No additional data rejections (e.g., rejections of concentration values sharply deviated from the rest similar values) were performed. More details concerning the DB can be found in [28]. Some results of calculations presented below are described in more detail in [13,14,28].

Results

The trend in change in the trace element composition in process of oil genesis

The correlation coefficients for mean TE content of a few rock types (averaged TE concentrations for these rock types) are presented in (Table 1) (upper lines). They indicate an expected higher correlation of the TE composition of clays, coals, and shales (all substances are of upper crustal origin) with the chemical content of the upper continental crust. In contrast, the mean TE composition of oil is closely correlated with the chemical content of the lower continental crust. Note also that the correlation coefficients typical of mean oil are substantially below those of clays, coals, and shales (Table 1). This probably reveals a more complex and multifactor process formation of oils. The obtained results show presumably that the deep crustal factors play an important role in the genesis of hydrocarbons, and that the process of oil generation is more complex than that of generation of coals and oil shales. Note also a higher correlation with the chemical content in plants than with the chemical content in animals; this seems to be due to the predomination of plant biomass. The results of the examination of the mean TE data on the main oil-and-gas bearing basins of Russia are also presented in (Table 1). As can be seen, in all of these cases the correlation with the lower continental crust is also higher than with the upper and the middle crust. An exception from this tendency was found for the Kamchatka region, both for small oil fields in Kamchatka and for oil seeps in the caldera Uzon volcano [21,29]. For these oils the correlation coefficient with the chemical content of the upper crust is similar to or even sometimes higher than with the lower crust (Table 1). Note also that the correlation with biota for the Kamchatka oils is higher than in most other cases. These peculiarities are probably due to higher deep temperatures in Kamchatka, and with the younger age of Kamchatka oils. Based on the above discussion, we will estimate the relative contributions from the lower and the upper continental crust by the difference in correlation coefficients for a given specimen with model chemical content for the lower and the upper crusts, R_lower-R_upper. The strength of the relationship to biota will be estimated by the maximum correlation coefficient for biota, maxR_biota. Figure 1 shows results from a comparison of (R_lower-R_upper) and maxR_biota values for different regions in USA, and for Trinidad [26,27,30]. The cluster of points is strongly elongated. The upper left tip corresponds to a higher correlation with biota and with the upper crust. The lower right end of the field of data points corresponds to a lower correlation with the chemical composition of biota and a higher correlation with the lower crust compared with that for the upper crust. Similarly, to the case for the data presented above, we see a strong predominance of cases of closer correlation of TE composition of oils with the lower continental crust than with the upper crust. The field of data points for certainly upper crustal rock types (clays, coals, shales) is adjacent to the upper left tip of area of the data points for crude oils and is characterized by higher correlation with the upper crust chemical content. Figure 2 shows in a similar manner the data for naphthides for the different areas of Russia. For a few data sets for Russia the scatter in the data points is rather high. We had presented data sets with lower scatter. The same tendency as in (Figure 1). can be seen. The products of oil degradation (asphalts) are represented by the wide field of data points adjacent to the lower right corner of data point cluster for crude oils and resin-asphaltene fraction of oils. The data points for resin-asphaltene fraction of oils are displaced in comparison with those for crude oil to higher correlation with the lower crust.

Table 1: Correlation coefficients between mean TE composition of different caustobioliths, oils including and the chemical composition of the Earth's crust and the types of biota.

Rock Type/ Region	Continental Earth Crust			Biota			
	Upper	Middle	Lower	Aquatic Plants	Terrestrial Plants	Aquatic Animals	Terrestrial Animals
Mean TE Content for Different Substances, Data from [19]							
Clays	0.90	0.85	0.84	0.77	0.72	0.53	0.46
Coals	0.84	0.76	0.79	0.78	0.71	0.48	0.50
Oil shale	0.83	0.76	0.78	0.78	0.74	0.54	0.55
Black shale	0.82	0.84	0.80	0.78	0.75	0.57	0.56
Mean oil	0.60	0.58	0.63	0.61	0.58	0.59	0.54
Different Main Oil-and Gas Bearing Basins of Russia, Data from [11]							
Dnipro-Donetsk	0.54	0.51	0.58	0.45	0.37	0.54	0.37
Timano-Pechorsky	0.57	0.55	0.62	0.57	0.57	0.53	0.50
Volga-Ural	0.59	0.60	0.63	0.54	0.60	0.61	0.52
Western Siberia	0.69	0.68	0.73	0.71	0.73	0.60	0.65
Eastern Siberia	0.57	0.54	0.60	0.57	0.54	0.55	0.54
Kamchatka, Caldera Uzon, Data from [21]							
Data 2011 year	0.64	0.60	0.64	0.78	0.74	0.70	0.78
Data 2012 year	0.64	0.60	0.65	0.74	0.69	0.67	0.76
[29]	0.73	0.73	0.72	0.75	0.68	0.60	0.67

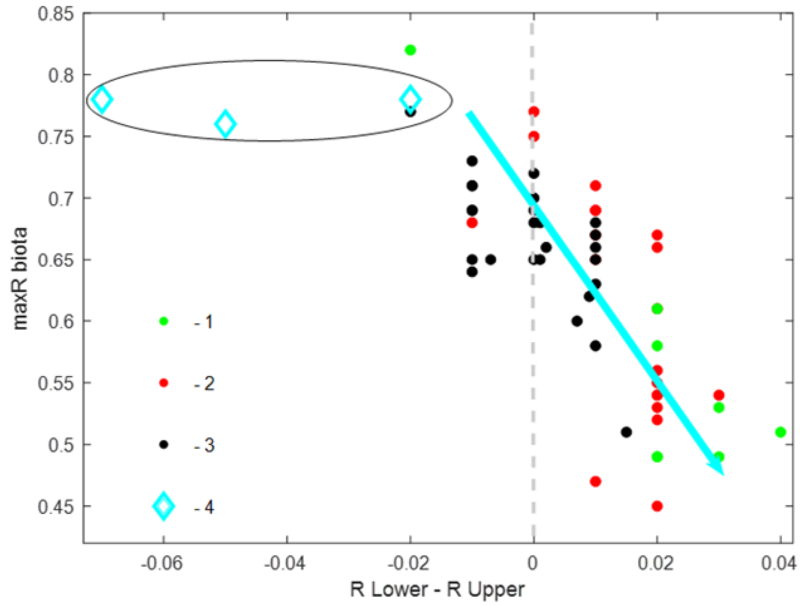

Figure 1: The relationship between the difference in correlation coefficients of specimen with the chemical content of the lower and the upper crust (R_Lower –R_Upper) and the maximum correlation value with biota (maxR_biota): 1-for Trinidad, 2-for data from California, 3-for different USA regions, data from [26,30]; the ellipse and blue diamonds (4) represent mean values for clays, coals, and shales, data from [19]; the vertical dashed line separates the regions where the relationship with the upper or with the lower crust dominates; the arrow indicates the tendency of change from shallow rocks to mature oils.

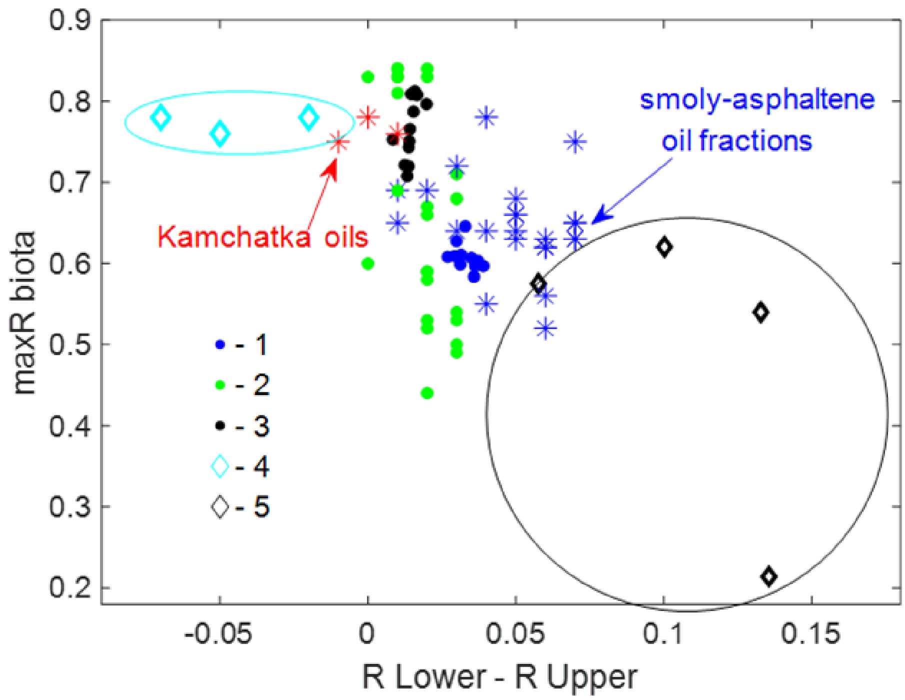

Figure 2: The tendency of the relationship between the difference in correlation coefficients with the model chemical content of the lower and the upper crust (R_Lower –R_Upper) and the maximum correlation with biota (maxR_biota) based on data for the area of Russia: 1-for data from [25], 2-from [15], 3-from [23] the ellipse and blue diamonds (4) represent the values for clays, coals, and shale [19]; the ellipse and black diamonds (5) represent the values for asphalts [25,28]. The results for Kamchatka oils (red stars) are based on data from [21,29] and those for resin-asphaltene fraction (blue stars) are from [10,12] as indicated in the figure.

The mud-volcanic and carbonic waters of the great caucasus

The close connection of the mud volcanism and oil-and-gas bearing regions is well known. It is commonly thought that mud volcanism provides evidence of the generation of HC gases in the interior of a region in question. The data on TE composition in the mud volcanic waters of the Caucasian region are examined in relation to more numerous, hence capable of yielding more valid statistical results, data on carbonic spring waters in the mega anticlinorium of the Greater Caucasus [20,22]. The mean correlation coefficient values for TE concentration in samples with the upper, middle, and the lower continental crust and with different types of biotas are listed in (Table 2) where data for several river basins that have been sampled to a greater extent are presented; due to larger statistics the errors of correlation coefficient values can be estimated. For spring carbonic waters of high-mountain regions in the Greater Caucasus (Elbrus and Kazbek regions) the maximum correlation is observed with the middle crust chemical composition and with the composition of terrestrial plants or animals. The differences between mean Correlation Coefficients (CC) for different crustal horizons and with different biota types are statistically significant. For the TE composition of samples from the Caucasian Mineral Waters (CMW) area the maximum of correlation also takes place with the middle crust, but the CC values with the mean chemical composition of marine and terrestrial biota are statistically insignificant. It seems that remnants of both terrestrial and marine organisms provided comparable contributions into the formation of TE composition of deep waters here. Note that roots of the carbonic spring waters systems and mud-volcanic fluid systems were found above to be deeper than in [20,22], where they are suggested to be located in the upper crust. However, the deeper depth estimates obtained here agree with the seismological data on the location of seismic roots of some mud volcanoes and the main magmatic chambers beneath the Elbrus and Kazbek volcanoes [31-34]. The effect of erosion should also be taken into account; in the highlands, initially deeper-seated rocks may be located closer to the surface.

Table 2: Correlation coefficients between the compositions of carbonic spring waters (the Greater Caucasus high mountain area) with the Earth crust and biota.

Region (Number of Analyses)	Continental Earth Crust			Biota			
				Plants		Animals	
	Upper	Middle	Lower	Marine	Terrestrial	Marine	Terrestrial
Kuban R (41)	0.75±0.01	0.76±0.01	0.71±0.01	0.80±0.005	0.82±0.01	0.78±0.005	0.75±0.005
Baksan R (15)	0.75±0.01	0.76±0.01	0.72±0.01	0.81±0.01	0.83±0.01	0.81±0.01	0.75±0.01

Ardon R (16)	0.75±0.01	0.77±0.01	0.73±0.01	0.81±0.01	0.84±0.01	0.80±0.005	0.77±0.005
South Osetia (17)	0.76±0.01	0.77±0.01	0.74±0.01	0.82±0.005	0.84±0.01	0.79±0.01	0.75±0.01
CMW region (14)	0.70±0.01	0.73±0.01	0.68±0.01	0.83±0.01	0.83±0.01	0.83±0.01	0.81±0.01

The mud-volcanic waters of the surroundings of the great caucasus

The results of CC calculation based on TE concentrations in the mud volcanic waters sampled in Azerbaijan, the Taman Peninsula, and eastern Georgia are listed in (Table 3). Similarly, as to the samples of spring geothermal waters in the Greater Caucasus, the highest correlation occurs between the TE composition of mud volcanic waters and the middle crust composition. However, the CC values for mud volcanic waters are below those for high-mountain carbonic thermal waters. We have also, that for the mud volcanoes in the Taman, Azerbaijan, and Georgia, in contrast to the waters sampled in the high mountain areas of the Great Caucasus, the highest correlation is observed systematically with the chemical composition of marine plants and animals. The resulting spatial confinement of the areas under study with greater correlation with marine or with terrestrial biotas is in good agreement with the orography and geological evolution of the Caucasian region. This agreement testifies in favor that the used method can be applied both to estimation of the depths for a given fluid system and to determinations of the corresponding dominant type of biota. Note, that the method for determining the dominant initial type of biota using all set of data on TE concentrations can appear to be more stable than other methods, for example, using the data on the concentration of V and Ni only [35].

Table 3: Correlation coefficients between the compositions of water sampled at mud volcanoes in Azerbaijan, the Taman Peninsula, and Georgia with the Earth's crust and biota.

Region (Number of Analyses)	Continental Earth Crust			Biota			
				Plants		Animals	
	Upper	Middle	Lower	Marine	Terrestrial	Marine	Terrestrial
Taman (17)	0.61±0.01	0.68±0.01	0.58±0.01	0.82±0.01	0.74±0.01	0.76±0.01	0.71±0.01
Georgia (5)	0.66±0.03	0.68±0.03	0.64±0.03	0.81±0.01	0.75±0.01	0.76±0.01	0.72±0.01
Apsheron (5)	0.42±0.04	0.48±0.04	0.44±0.04	0.72±0.04	0.67±0.02	0.77±0.02	0.65±0.03
Shemakha-Gobustan (23)	0.42±0.01	0.48±0.01	0.44±0.01	0.75±0.01	0.66±0.01	0.76±0.01	0.67±0.01
Kura R (12)	0.47±0.03	0.55±0.03	0.50±0.03	0.76±0.02	0.70±0.01	0.79±0.01	0.69±0.01

Discussion

The results of the foregoing correlation analysis have confirmed a polygenic character of the Trace Elements (TE) composition in oils associated both with organic matter from sedimentary strata and with the host rocks and waters and the deep fluid. We have identified a persistent tendency in the change of TE composition in oils during the transformation of hydrocarbons under different geodynamic settings. A trend of a decrease in the correlation with biota and an increase in the difference between the correlation coefficients with the lower and the upper crust takes place for different samples in sequence from the organic matter to crude oil and resin-asphaltene fraction of oil, and further to products of oil degradation (asphalts). This trend appears to be connected with reworking of organic matter and oils by the ascending deeper-crust fluid flow. Thus, in all examined cases the oil genesis process appears to be closely connected with a fluid flow ascending from the lower or middle crust level. An absence of alternative situations seems to indicate that the presence of such a flow is not of secondary importance or an accident, but it is a necessary condition of the process of oil genesis. The less deep middle or even the upper crust roots of the oil genesis connected fluid systems in Kamchatka correlates with the higher deep temperatures occurring here, so the dehydration is occurring at shallower depths, and the ascending fluid flow carries its TE marker from the shallower depths. The existence of ascending fluid flows in the lithosphere finds a strong support in seismological data [36,37]. It has been found that earthquake rupture zones have a tendency of an upward development of the rupture at depths from the middle crust to the uppermost mantle [38]. Besides, aftershock swarms also typically exhibit a dominant tendency of occurrence of events with shallower depths. Besides, later aftershocks and earlier aftershocks also turn out to be systematically shallower than the background earthquakes. All of these persistent statistical tendencies can be quite naturally explained by the existence of ascending water-gas (mostly supercritical) fluid flows. Provided the data set is large enough, the method of correlation analysis can be used to find the depth to the roots of the fluid system under study and the dominant type of biota (marine or terrestrial) corresponding to this area. Such possibilities appear to be useful in the exploration and exploitation of hydrocarbon fields.

Oil genesis occurring on the basis of an organic matter, but with the obligatory presence of the ascending deep fluid flow, acquires some features that are proper to the abiogenic oil genesis model. Thus, this model combines positive features from both the biogenic and the abiogenic models of oil genesis. We conclude by emphasizing, however, that, while providing convincing evidence in favor of a polygenetic source for trace elements in oils, the method of correlation analysis still does not give unambiguous indication in favor of a considerable deep abiogenic component in the main hydrocarbon component of oils. The existence of input of deep hydrocarbons along with a supercritical fluid flow discussed above remains debatable.

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