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COMPARING THE RESULTS OF LINEAMENT ANALYSIS WITH ISOTOPE GEOCHEMISTRY DATA

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Abstract

This article presents the results of a geochemical survey carried out in the southwestern part of the Siberian platform, within the Sayan-Yenisei (Angara) syncline (a superorder Riphean-Middle Paleozoic structure). The object of research was hydrocarbon gases contained in the subsoil rocks (clays). The subsoil samples were taken from the bottom of boreholes (40 mm in diameter) made with an electric drill. The sampling depth was 0.6–1 m. Further laboratory studies included chromatographic and isotope analysis. Lineament analysis of the digital elevation model was carried out as a complementary study. One of the lineament analysis results was a lineament density map, which reflects the permeability (macro-fracture density) of the sedimentary cover. This allowed a comparison of the macro-fracture density with the gas content and isotopic composition. The study revealed that gases with a high content of heavy isotopes tend to gather in the low permeability areas. This can be explained by the fact that the gases disperse quickly within fractured zones, and deep gases should be expected only in the areas with strong cap rocks, i.e. in the areas with low macro-fracture density where stable hydrocarbon deposits have already formed.

Keywords:

Hydrocarbons;
Geochemical survey;
Isotope geochemistry;
Lineament analysis.

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1. Introduction

Methane is the major component of natural gas (hydrocarbon gas, damp, marsh gas, etc.). There are also microorganisms that produce methane as a metabolic byproduct in anaerobic conditions.

Natural gas is enriched (relative to the oil carbon and organic matter) in the C¹² isotope and is liable to changes in the isotopic composition. It was subsequently discovered that carbon isotopic composition distinguishes three types of methane [1]:

1. Biochemical (bacterial) methane is the lightest one. δC^{13} values usually range from -60 ‰ to -90 ‰ . This type of methane is generated in a strictly anaerobic environment by certain bacteria. It happens only after the complete depletion of free and bound oxygen (the termination of bacterial sulfate reduction).

2. In thermogenic (catagenetic) methane, δC^{13} values usually range from -20 ‰ to -50 ‰ . This type of methane is formed during the thermocatalytic decomposition of organic matter.

3. Deep methane (abiogenic) is the heaviest one with δC^{13} values over -20 ‰ .

To sum up, δC^{13} can be used for genetic typing of natural gas. Isotope analysis reveals the origin of natural gas encoded within its isotopic composition.

It should be stressed that one should think of genetic typing of natural gas rather than morphological typing, since the isotopic composition is controlled primarily by genetic factors. The secondary factors such as interaction with the environment and mode of occurrence play a much smaller role.

In nature, methane of different genotypes can mix together and acquire a combined isotopic composition determined by the proportion of the mixture. In addition, the isotopic composition of natural methane can be affected by the mixture of its precursor (CO₂) with different isotopic compositions.

Therefore, given the insignificance of isotopic effects associated with the gas movement, it is possible to use isotope data for the genetic identification of gases, and hence for studying the migrations and their nature.

The genetic scheme described above directly relates the isotopic composition to the nature of

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the initial source, the degree of its transformation, and the conditions of gas generation. However, the whole picture is greatly complicated by subsequent migration and dissipative processes. As a result, gases may be found in conditions different from the original ones, and therefore the isotopic composition will contrast with the characteristics of the enclosing sediments. This contrast can be used as the basis for studying gas migration.

Hydrocarbon gases that underwent thermal metamorphism have a peculiar isotopic composition. Isotopic exchange in the $\text{CH}_4\text{--C}_2\text{H}_6\text{--C}_3\text{H}_8\text{--C}_4\text{H}_{10}$ system, which takes a geologically short time and occurs at a temperature of 150–200 °C, brings all the hydrocarbons to a similar isotopic composition.

Hydrocarbon gases contain large amounts of ethane and propane in addition to methane enriched in heavy isotopes. Coal (anthracite) gases contain primarily heavy methane, but lack its higher homologues. Thermal gases are enriched in carbon dioxide [2].

With the genetic type identified, it is possible to indicate the source of the gas (based on the geological structure of the region and the facies composition of the deposits) and therefore to determine the direction and sometimes the time of migration.

However, migration and dissipative processes can release the hydrocarbon reservoir from genetically related gas (containing heavy methane, which is characteristic of the oil-forming processes) and fill it with gas of a different origin, for example, with light methane from early-stage organic metamorphism. When studying migration and accumulation of hydrocarbons, it is always necessary to take into account the role of seismic activity and dissipative processes reflected in isotope data.

Oxidation processes, which occur continuously during migration of gases, often determine the changes in their isotopic composition.

In the subsoil, the biodegradation of alkanes usually leads to a number of O-substituted compounds: hydroxylation produces short-chain primary and secondary alcohols, which are oxidized to ketones and aldehydes, respectively (the latter form organic acids) [3-5]. Esters can also form during these reactions [4, 6]. The formation of ketones, aldehydes, esters and organic acids strictly depends on the biodegradation type. Aerobic decomposers activate alkane molecules using O_2 as an oxidizer to form CO_2 . On the other hand, monooxygenases activating alkanes overcome their low reactivity by producing reactive O-containing particles. For example, CH_4 is first oxidized to methanol (CH_3OH), then it is converted to formaldehyde (CH_2O) and finally it is transformed into formic acid (HCOOH). In general, decomposition of n-alkanes begins with oxidation of the terminal methyl group. In this case, primary alcohols form, which undergo further oxidation into the corresponding aldehydes, which in turn are converted into fatty acids (Wentzel et al., 2007 [7]). Severe degradation of alkanes in soil is clearly indicated by a decrease in their

concentration with an increase in molecular weight. This confirms that biodegradation in the subsoil becomes increasingly effective as the alkane chain grows [8]. It is worth noting that n-hexane showed higher concentration than other alkanes. The latter observation, also recorded by Capaccioni et al. [9] and Darling [10] in relation to volcanic and hydrothermal fluids can be attributed to the stability of the C_6 structure.

Degradation processes strongly depend on the sediment's age and the residence time of gases in the soil. The degree of degradation is controlled by several parameters such as soil permeability, air pressure and pore water content. All this governs the decomposition processes that determine the composition of VOCs [11].

Various products are formed during oxidation of methane and its homologues. These are mainly alcohols and aldehydes (ketones in case there are tertiary carbon atoms present). Often, there are more oxidation products in the subsoil than methane and its homologues: various microorganisms make a significant contribution to the content of oxidation products.

2. Geological setting

The study area is located in the southwestern part of the Siberian platform, within the superorder Riphean-Middle Paleozoic structure – the Sayan-Yenisei (Angara) syncline (fig.1). In the south, the study area is bordered by the folds of the Sayan-Altai region; in the west, there is the Yenisei folded area (which includes the Angara-Kan massif); in the north, the border between the study area and the Baikitskaya antecline and the Irkineevsky wedge runs along the regional faults and the Angara folded zone; in the east, the border follows the Kovinsko-Vikhorevskaya fold and the Angara-Vilyui fault separating the study area from the Angara-Lena plateau [12]. There are two levels in the geological structure of the syncline: 1) the

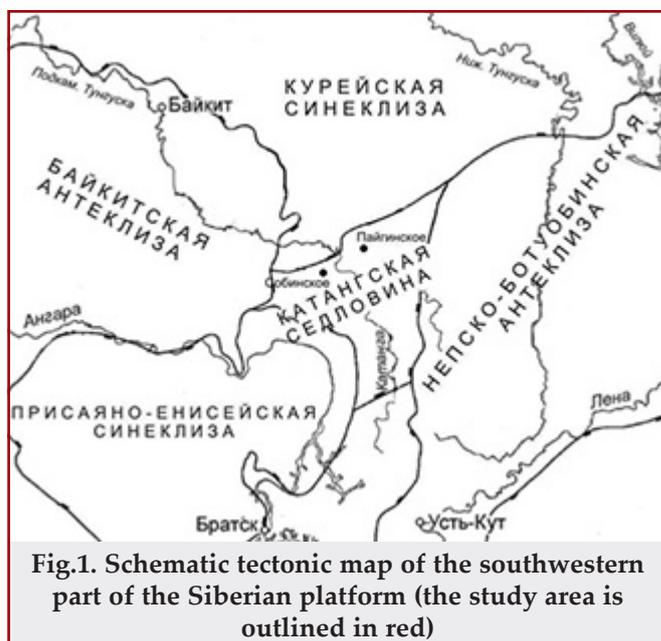


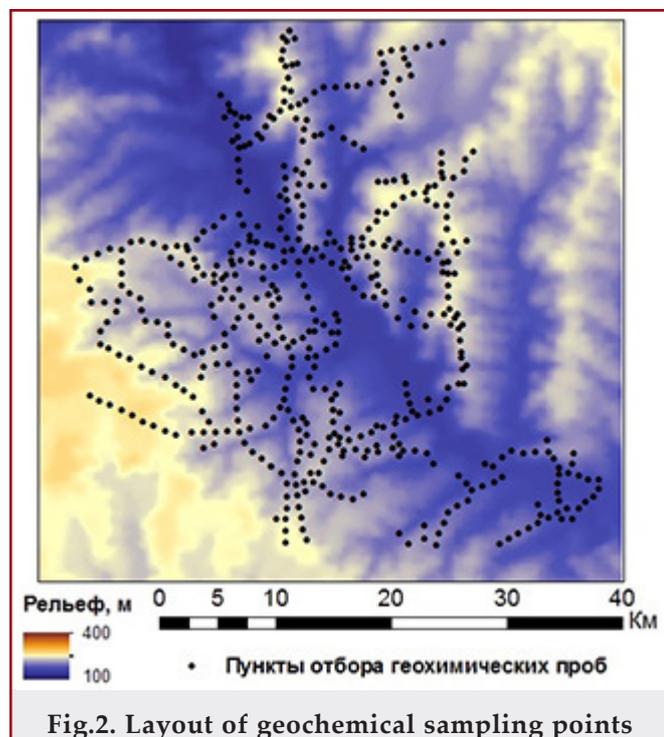
Fig.1. Schematic tectonic map of the southwestern part of the Siberian platform (the study area is outlined in red)

basement which is represented by crystalline rocks of the Archean and Proterozoic; 2) the sedimentary cover formed during the Riphean. Deep-hole prospecting data and seismic exploration showed that the lithological and stratigraphic composition of the study area includes rocks of the heterogeneous basement and the sedimentary cover, which is represented by deposits of the Vendian, Cambrian, Ordovician, Carboniferous, Permian, Triassic, Jurassic, Cretaceous and Paleogene. On the surface of the crystalline basement, the Chuno-Biryusinsky arch (inversion zone) can be seen, within which the Boguchano-Mazinsky wedge (the actual study object) can be outlined along the -6000 m isohypse [12]. The Boguchano-Manzinsky wegde is an ancient structure composed of oil source rocks (rocks enriched with organic matter), which is considered an area of high oil and gas potential [13].

3. Methods.

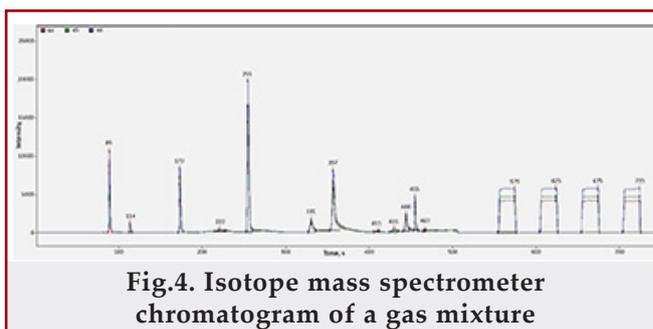
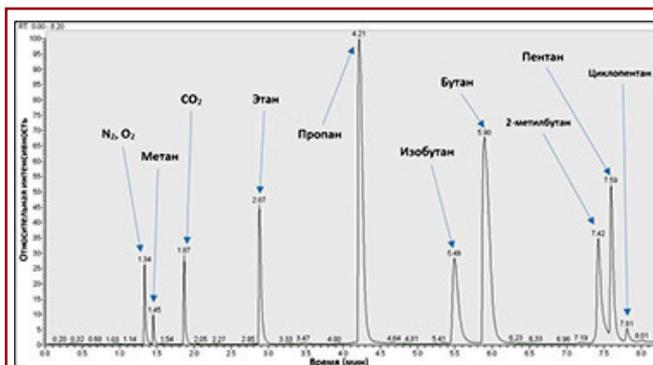
Geochemical sampling technique

The subsoil samples were taken from the bottom of boreholes (40 mm in diameter) made with an electric drill. The sampling depth was 0.6–1 m. The samples were placed in airtight containers marked with the sample number, and then transported to the geochemical laboratory for further analysis. 500 routine samples and 25 control samples (525 samples in total) were analyzed (fig.2). Each sample weighed not less than 0.5 kg.



Chromatographic and isotope analysis

$^{13}\text{C}/^{12}\text{C}$ ratios in gas mixtures were determined with the TRACE 1310 gas chromatograph (ThermoFisherScientific, Germany) attached to the ICQ quadrupole mass spectrometer (ThermoFisherScientific, USA) through the GC Isolink interface unit in the Delta V Plus isotope mass



spectrometer (ThermoFisherScientific, Germany). The headspace technique was used to obtain data on gas mixtures. The subsoil sample was placed in a vial with a septa cap and heated to 130 °C for 12 minutes. Then 2.5 ml of the gas sampled with the TriPlusRSH microsyringe (CTC Analytica AG, Switzerland) was injected into an injector heated to 140 °C.

Components of the gas mixture were separated using the TracePLOT TG-BOND Q column (ThermoFisher Scientific, Germany) with a nonpolar, 100% divinyl benzene phase. The chromatograph operated in a split mode with a helium flow rate of 4 ml/min and a 5-fold dilution. The temperature program consisted of several stages that are optimal for the separation of methane homologs (up to pentane).

Individual compounds separated in the column (fig.3) were transformed into simple gases at temperatures up to 1000 °C in an oxidizing reactor. The GC-MS system can be used to determine the isotopic composition of individual components of a complex mixture.

Analysis of each sample concluded with mass spectrometry. First, several portions of standard carbon dioxide (with a known isotopic ratio) were injected into a mass spectrometer. Then, the mass spectrometer was calibrated against the standard gas in order to properly determine the isotopic ratio of the sample (fig.4). For the control purposes, the measurements were made against the IAEA-600, which is caffeine with a known isotopic ratio dissolved in acetone. The Alkane-MixC14-C15-C16 dissolved in isooctane (with concentrations of 200ng/mcL and 15ng/mcL) was used for the mass spectrometer calibration.

Lineament analysis

The analysis focuses on lineaments. The WinLESSA software was used, which is designed to automate the analysis of various types of data such as images, diagrams, digital elevation models (DEMs) in geological studies. This is a tool that allows getting a uniform numerical description of the data pattern [14-18]. The distribution and orientation of small linear elements are described.

4. Results.

Chromatographic and isotope analysis

Chromatography data showed extremely low (background level) content of methane and its homologues in the study area. This is due to the fact that such concentrations are at the very sensitivity limit of the flame ionization detector. In addition, there is a large amount of carbon dioxide, which releases almost simultaneously with the studied

components. Therefore, the quantitative assessment of gas concentrations would be inaccurate. The isotopic ratio of the gas mixture was also indeterminable. Oxidized forms of methane and its homologues have been detected in significant concentrations. This is due to the fact that these gases are better absorbed by the soil: the highest concentrations were observed in samples containing clay soils. Thus, clays appear to be great natural adsorbents of methane oxidation products. When analyzing chromatograms, the following substances were identified as the most characteristic and evidential (arranged in order of increasing number of carbon atoms in the molecule):

CH₃OH (methanol) – CH₃CHO (acetaldehyde) – CH₃C(O)CH₃ (acetone) – CH₃CH(CH₃)CHO (2-methylpropanal) – CH₃CH₂CH₂CH₂CHO (pentanal).

Table 1 shows the statistical characteristics of δC¹³ for methane oxidation products.

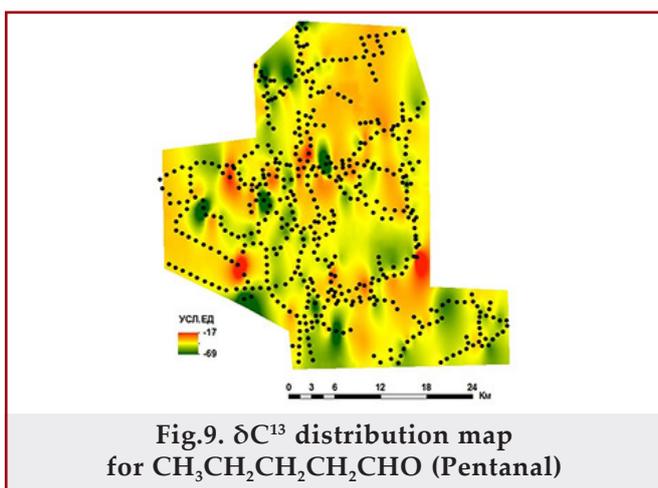
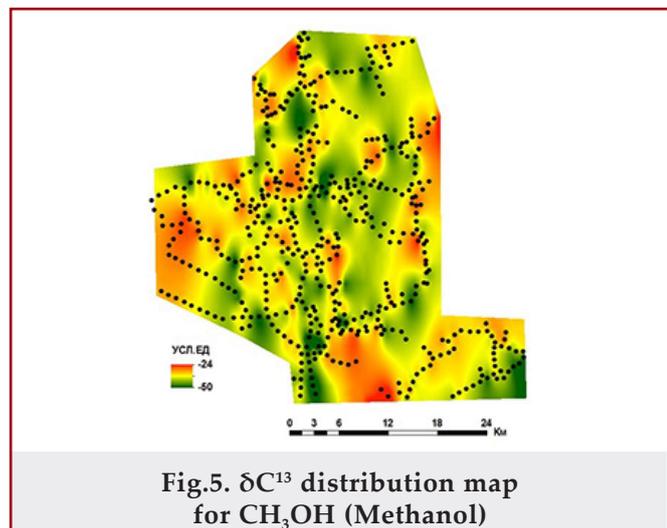
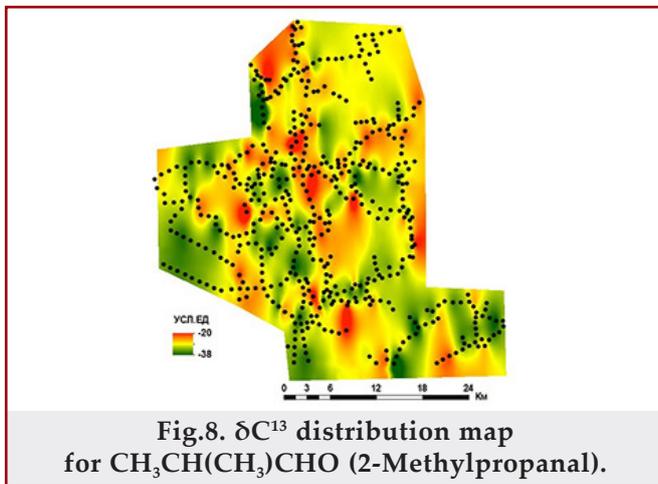
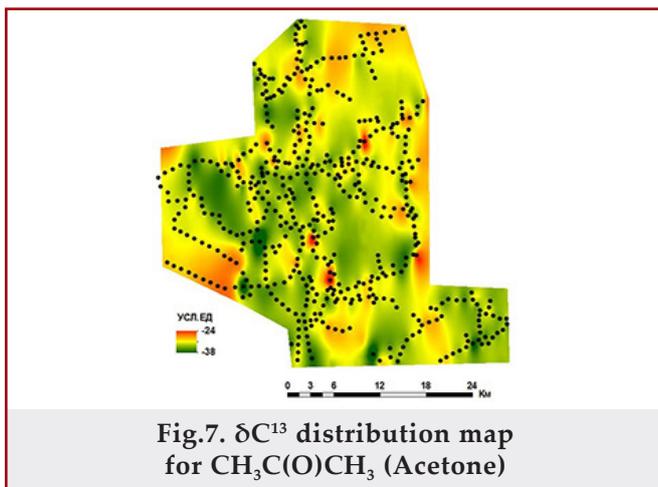
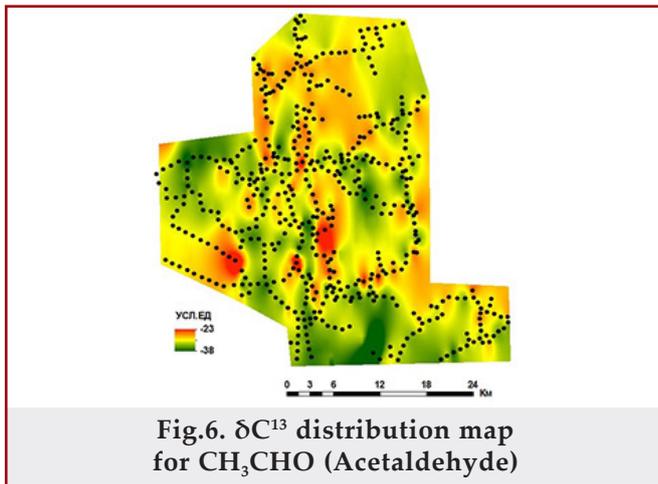
Table 1		
δC¹³ values obtained for methane (and its homologues) oxidation products		
Component	Statistics (values in ‰)	Distribution density *
Methanol	Number of measurements: 363 Minimum: -53.75 Maximum: -22.38 Average: -34.70 Standard deviation: 4.34	
Acetaldehyde	Number of measurements: 403 Minimum: -39.00 Maximum: -18.80 Average: -27.83 Standard deviation: 1.53	
Acetone	Number of measurements: 424 Minimum: -39.06 Maximum: -22.03 Average: -31.26 Standard deviation: 1.58	
2-Methylpropanal	Number of measurements: 266 Minimum: -39.12 Maximum: -17.73 Average: -29.53 Standard deviation: 3.21	
Pentanal	Number of measurements: 320 Minimum: -51.97 Maximum: -13.18 Average: -32.85 Standard deviation: 4.19	
<p>*Note: the red line on the diagrams indicates the δC¹³ threshold value (see explanation below).</p>		

These substances can be metabolic byproducts of microorganisms depleted in heavy carbon isotope (δC^{13} ranging from -60 ‰ to -90 ‰) or they can be transformed from methane and its homologues under thermal action and various conditions, i.e. they can be enriched in heavy carbon isotope (δC^{13} ranging from -10 ‰ to -40 ‰). However, the environmental conditions in this case will affect the isotopic ratio to a lesser extent than the genetic nature of these substances. Therefore, δC^{13} values in methane oxidation products can be used for the genetic classification of natural gases.

Given the patterns and δC^{13} ratios described above (which are characteristic of hydrocarbon gases), the δC^{13} values closest to those of the reservoir gases were selected (tbl.2). When choosing the δC^{13} range for each substance, the distribution density was also taken into account: the maximum value in the δC^{13} range corresponds to the maximum value on the diagram; the minimum value in the δC^{13} range corresponds to the threshold value (marked with a red line) where the distribution density is equal to $1/2$ of the maximum distribution density.

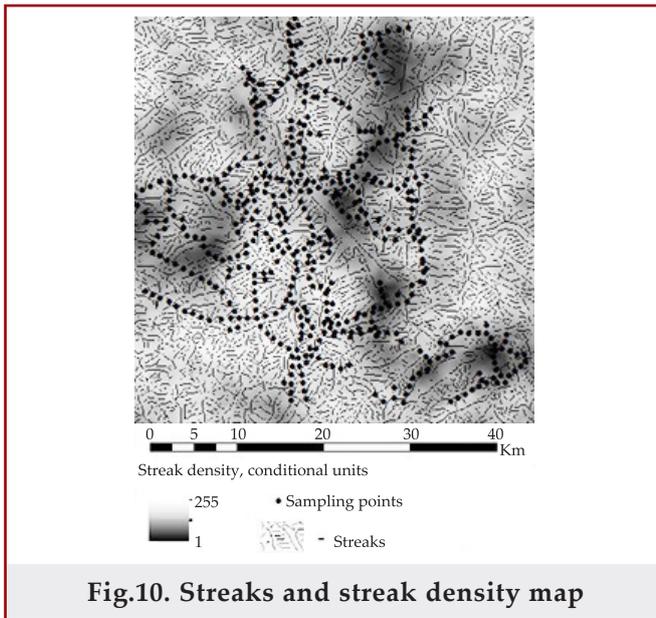
Component	δC^{13} range, ‰	Genotype
Methanol	$-30.35 \div -22.38$	Thermogenic, belongs to a hydrocarbon reservoir
Acetaldehyde	$-26.78 \div -18.80$	
Acetone	$-29.07 \div -22.03$	
2-Methylpropanal	$-27.47 \div -17.73$	
Pentanal	$-29.47 \div -13.18$	

Figures 5–9 show the δC^{13} distribution maps covering the study area for the following compounds: CH_3OH (Methanol) – CH_3CHO (Acetaldehyde) – $CH_3C(O)CH_3$ (Acetone) – $CH_3CH(CH_3)CHO$ (2-methylpropanal) – $CH_3CH_2CH_2CH_2CHO$ (Pentanal).



Lineament analysis of DEM

One of the lineament analysis results is the map of straight extended lineaments. The DEM processing results include streaks (dashes, short linear elements), lineaments and streak density (the number of streaks per unit area) maps. Lineament maps reflect features of the sedimentary cover associated with modern geodynamics. Streak density maps reflect permeability (macro-fractures) of the sedimentary cover: the higher the stroke density, the higher the permeability of the sedimentary cover, and vice versa (fig.10).



5. Discussion

Gases enriched in light carbon isotopes are generated in the upper part of the sedimentary cover. Presence of heavy gases in surface sediments could indicate productive layers at a greater depth [2]. It was also safe to assume that areas with high δC^{13} values coincide with areas of high permeability. In reality, the reverse was the case.

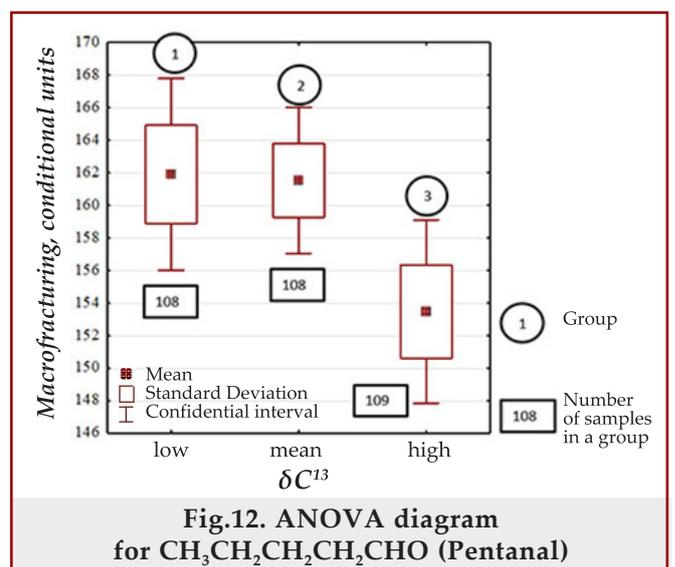
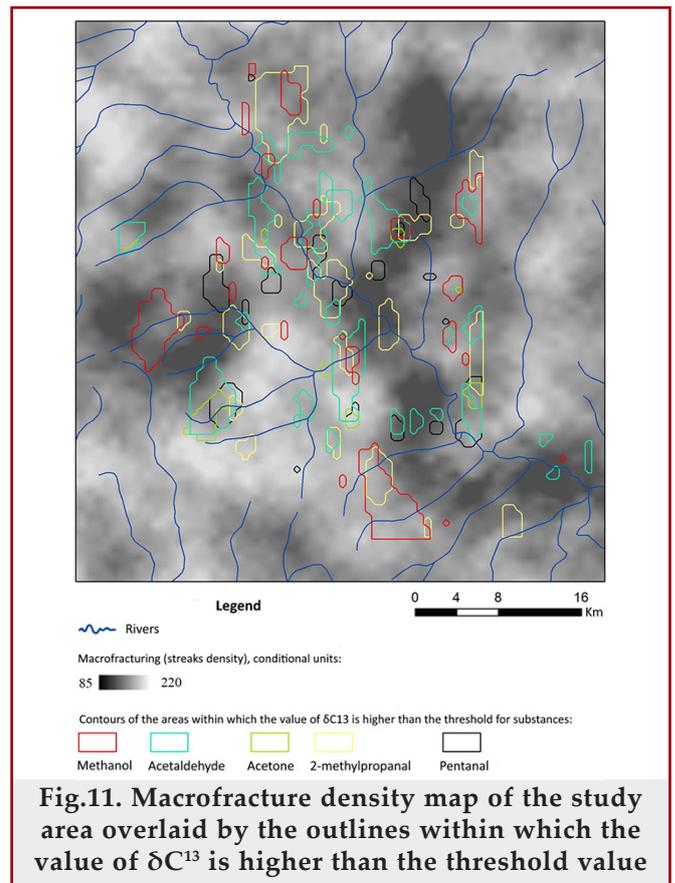
Figure 11 shows that areas with high δC^{13} values tend towards areas with medium and low fracture density (reflected in streaks density).

In order to test the hypothesis about the relationship between isotopic composition and macro-fractures, one-way ANOVA test [19] was carried out, where macro-fracture density was the dependent variable, and δC^{13} was the grouping variable. This test shows whether there are statistically significant differences in the average values of variable groups. If so, the objects within the group behave similarly (have similar properties), but differ from the objects in other groups.

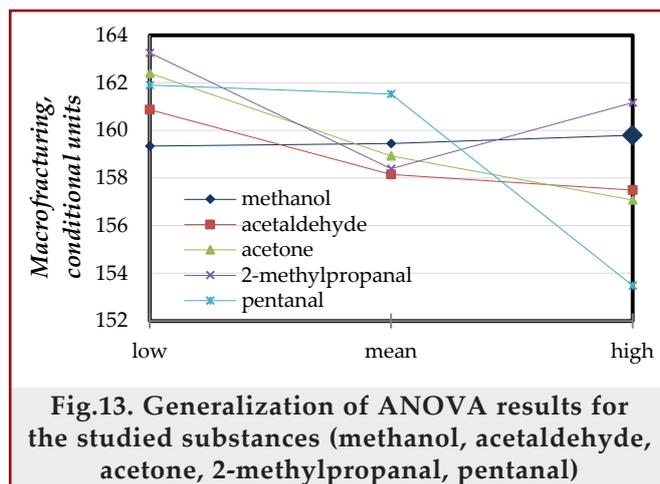
Macrofracture density values were extracted from the streak density raster using the ArcGIS SpatialAnalyst geoprocessing tools and bound to the geochemical sampling points. Thus, the initial data set included δC^{13} values and streaks density for each sampling point. For each of the substances (methanol, acetaldehyde, acetone, 2-methylpropanal, pentanal), the δC^{13} values were divided into 3 classes. Quantile

[20] was the classification method. Quantile assigns the same number of data values to each class. The first class contains sampling points with the lowest δC^{13} values (class 1), the second class contains points with average δC^{13} values (class 2) and the third class contains points with the highest δC^{13} values (class 3). Further, the data was exported to the STATISTICA 6.

The ANOVA test was carried out separately for each of the studied substances. For example, figure 12 shows the analysis results for $CH_3CH_2CH_2CH_2CHO$ (Pentanal). The analysis showed significant differences in the average values (at the p-level equal to 0.05). For other substances, the results were similar.



Next, the averages for all groups were compiled in one diagram (fig.13). In general, there is the following tendency: the higher the heavy gas content, the lower the streak density (with the exception of class 3 2-methylpropanal). In other words, heavy gases were found in areas with low macro-permeability of rocks. This dependence can be explained by the rates of gas diffusion and migration. Gas diffusion rates [2] and isotope analysis results indicate that most of the gas formed in the pre-Paleogene has left the host rocks [2]. Through open fractures, gases can escape in an even shorter time. Therefore, in fractured zones, the rate of gas volatilization is very high, and deep gases should be expected only in the areas with strong cap rocks, i.e. in the areas with low macro-fracture density.



Conclusion

The results obtained during this study lead to several conclusions:

- Natural sorbents of hydrocarbon gases (clays, loams, etc.) are promising objects for geochemical surveys.
- The study of isotopic composition of hydrocarbon gases contained in the subsoil air significantly increases the information content of geochemical studies.
- Geomorphological analysis (lineament analysis in particular) increases the reliability of geochemical data.
- The study revealed that gases with a high content of heavy isotopes tend to gather in the low permeability areas. This can be explained by the fact that the gases disperse quickly within fractured zones, and deep gases should be expected only in the areas with strong cap rocks, i.e. in the areas with low macro-fracture density where stable hydrocarbon deposits have already formed.

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Результаты сопоставления данных изотопной геохимии и линеamentного анализа рельефа

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Реферат

В статье представлены результаты геохимической съемки проведенной на площади расположенной в юго-западной части Сибирской платформы, пределах надпорядковой рифейско-среднепалеозойской структуры, Присаяно-Енисейской (Ангарской) синеклизы. Объектом исследований являлись газообразные углеводороды содержащиеся в породах подпочвенного слоя (глинах). Пробы грунта отбирались из подпочвенного слоя с призабойной части шпуров диаметром 40 мм, пробуренных при помощи электробура. Глубина пробоотбора составляла 0.6-1 м. Лабораторные исследования включали в себя хроматографический и изотопный анализ. В качестве дополнительных исследований был проведен линеamentный анализ цифровой модели рельефа. Одним из результатов анализа является карта плотности штрихов, отражающая степень проницаемости (макротрещиноватости) осадочного чехла. Это позволило сопоставить данные по макротрещиноватости с содержанием газов и их изотопным составом. По результатам работ было установлено, что на данной территории области распространения газов с высоким содержанием тяжелых изотопов совпадают с областями низкой макропроницаемости пород. Данный факт может быть объяснен тем, что скорость рассеяния газов в трещинных зонах очень высокая, и наличие глубинных газов следует ожидать только на тех участках, где свойства покрышек наилучшие, т.е. на участках с низкой макротрещиноватостью пород осадочного чехла где уже сформировались стабильные углеводородные залежи.

Ключевые слова: углеводороды; геохимическая съемка; изотопная геохимия; линеamentный анализ рельефа.

İzotop geokimyası və relyefin lineament analizi məlumatlarının müqayisəsinin nəticələri

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Xülasə

Məqalədə Sibir platformasının cənub-qərb hissəsində yerləşən ərazidə, Sayan-Yenisey (Anqara) sineklizinin Rifey-Orta Paleozoy yuxarı strukturu hüdudlarında aparılmış geokimyəvi tədqiqatın nəticələri təqdim olunmuşdur. Tədqiqat obyektı torpaqaltı qatın süxurlarında (gillərdə) yerləşən qaz halında olan karbohidrogenlər olmuşdur. Torpaq nümunələri elektrobubr vasitəsilə qazılmış, diametri 40 mm olan şpurların quyudubi hissəsinin torpaqaltı qatdan götürülmüşdür. Nümunənin götürülmə dərinliyi 0.6-1 m təşkil etmişdir. Laboratoriya tədqiqatlarına xromatoqrafik və izotop analiz daxildir. Əlavə tədqiqat olaraq rəqəmsal relyef modelinin lineament analizi aparılmışdır. Analiz nəticələrindən biri çöküntü çexolunun (örtüyünün) keçiricilik dərəcəsini (makroçatlılığı) əks etdirən ştrixlərin sıxlıq xəritəsidir. Bu, makroçatlılıq məlumatlarını qazlar həcmi və onların izotop tərkibi ilə müqayisə etməyə imkan vermişdir. Aparılan işlərin nəticələrinə əsasən müəyyən edilmişdir ki, həmin ərazidə tərkibində ağır izotopların çox olduğu qazların yayılma sahələri süxurların makrokeçiricilinin aşağı olduğu sahələrə təsadüf edir. Bu fakt onunla izah oluna bilər ki, çatlı zonalarda qazların yayılma sürəti çox yüksəkdir və dərindən gələn qazların mövcudluğu yalnız örtük xassələrinin yaxşı olduğu yerlərdə (yəni artıq stabil karbohidrogen yataqlarının formalaşdığı, çöküntü çexolu süxurlarının makroçatlılığı aşağı olan hissələrində) gözlənilə bilər.

Açar sözlər: karbohidrogenlər; geokimyəvi tədqiqat; izotop geokimyası; relyefin lineament analizi.