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Gas-hydrogeochemical conditions of the Bilche-Volytsia oil-gas-bearing area (Carpathian foredeep, Ukraine)

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ABSTRACT

Introduction. Water-dissolved gases in exploratory oil and gas hydrogeology are the fundamental criteria of oil and gas potential. Their quantitative and qualitative characteristics allow to study the conditions of formation and preservation of the hydrocarbon deposits and to identify the perspective searching areas.

The purpose of article was to determine the vertical and lateral gas-hydrogeochemical zonation of the Bilche-Volytsia oil and gas zone, to assess the role of water-dissolved gases in the formation of gas fields with the prospect of predicting new hydrocarbon deposits.

The research methods base on the calculation and interpretation of the following parameters: gas saturation of water, saturation pressure, gas saturation coefficient. Graphs of dependences based on experimental studies from published scientific publications were used to determine the solubility of methane.

The results of research. Nitrogen-methane composition of water-dissolved gases of the Upper Jurassic and Upper Cretaceous aquifers in combination with specific geochemical characteristics of groundwater of the north-western and central parts of the Bilche-Volytsia OGBA indicate the open hydrodynamic conditions, which, in general, are unfavourable for the formation and preservation of hydrocarbon deposits. In deep-submerged reservoirs of the south-eastern part of this area, water-dissolved gases of the Upper Jurassic and Upper Cretaceous aquifers are characterized by high contents of methane homologues.

In the Upper Badenian aquifer the lateral distribution of water-dissolved methane is presented in the growth of its portions from the West and East European platforms in the direction of the sub-submerge of the Carpathians, which is due to an increase in the degree of hydrogeological closure of structures. Increased methane contents also spatially tend to transverse tectonic faults, which determines their role in the vertical migration of water-hydrocarbon mixtures.

Water-dissolved gases of the highly productive Lower Sarmatian aquifer are mainly methane, occasionally nitrogen-methane. The gas saturation of the waters directly correlates with the proximity to gas deposits. Laterally, the portion of water-dissolved methane is directly correlated with TDS of water, the high values of which accordingly reflect the structures of a high degree of hydrogeological stagnation.

Conclusions. The portions of water-dissolved methane increase from the West and East European platforms in the direction of the sub-submerge of the Carpathians, which is effect of higher degree of hydrogeological closure of structures. It has been established that water-dissolved methane, nitrogen and carbon dioxide have different sources of origin and different spatiotemporal mechanisms of water saturation. The hydrodynamically closed structures (favourable for the formation and preservation of hydrocarbon deposits) characterized by high relative and absolute contents of dissolved methane. Increased methane contents also spatially tend to transverse tectonic faults, which determines their role in the vertical transportation of water-hydrocarbon mixtures. Deep hydrodynamically closed aquifers of the Bilche-Volytsia OGBA often are marked by high nitrogen contents of non-air origin. Its source can be rock organic matter, bound rock nitrogen released during metamorphism, nitrogen of deep genesis.

Keywords*: Bilche-Volytsia oil-gas-bearing area, water-dissolved gases, gas saturation of water.*

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Introduction. Water-dissolved gases in exploratory oil and gas hydrogeology are the fundamental criteria of oil and gas potential. Their quantitative and qualitative characteristics allow to study the conditions of formation and preservation of the hydrocarbon deposits and to identify the perspective searching areas. The amount and composition of water-dissolved gases depend mainly on the presence of hydrocarbon deposits, organic matter in rocks, tectonic, lithological and hydrogeological conditions of the region [1, 2, 3]. The area of gas scattering of hydrocarbon deposits, which can be located in both adjacent and upper aquifers, plays the main role. The gas scattered aureole occurs due to the destruction of hydro-carbon deposits and diffusion-filtration movement of components from deposits into formation waters [4]. According to Kartsev A. [5], the most favourable for the saturation of groundwater with hydrocarbon gases are the reducing conditions on the exfiltration stages of the hydrogeological history of the artesian basin.

Studies of natural dissolved gases and their impact on the formation of gas fields performed in the Republic of China found that Weiyuan gas field in the Sichuan River basin was formed by degassing and accumulation of dissolved gases due to the decrease of formation pressures and temperatures in aquifers during the rise of tectonic structures of the Himalayas [6]. The following conclusions were expressed on the results of studying of geochemical and isotopic features of formation waters, and also isotopic characteristics of carbon of free and water-dissolved gases.

Also, the results of carbon isotopes studies of free and water-dissolved gases of the Central, South-Western and Southern gas-bearing areas of Sichuan confirmed the mechanism of formation of deposits due to degassing of hydrocarbons from formation waters [7].

The mechanism of gas fields formation in Western Siberia (Russia) is seen in the release of dissolved gas from the Cretaceous aquifer complex due to lower formation pressures and temperatures during its geodynamic rise in the Quaternary period [8]. Such conclusions were made by researchers in the analysis of petrophysical, hydrodynamic, hydrochemical characteristics of aquifers and isotopic studies of hydrocarbon components of free and water- dissolved gases.

In Ukraine, based on the study of hydrogeochemical characteristics, qualitative and quantitative characteristics of dissolved gases, the criteria of gas potential of the Black Sea artesian basin were clarified [9, 10].

For the Carpathian Oil and Gas-Bearing Province, it was found that the high gas saturation of waters and high *Pg/Pf* (gas pressure/formation pressure) ratio indicate the prospects of Sarmatian sediments of the Outer Zone of the Carpathian Foredeep in terms of gas potential. The sediments of the Upper Badenian of the central part of the Outer Zone are characterized by unfavourable conditions for the preservation of hydrocarbon deposits, due to the significant content of water-dissolved nitrogen. Based on the qualitative and quantitative characteristics of the water-dissolved gases, reservoir thicknesses and their porosity parameters, the resources of water-dissolved gases of the Outer Zone were quantified. For the Neogene complex, they are 100, for the Cretaceous - 6, for the Jurassic - 28 billion m^3 [11].

According to isotope studies of methane carbon and carbon dioxide carbon [12], the methane genera-tion temperatures of the Outer Zone of the Carpathian Foredeep are 40-135 °C. This allowed the researchers to predict the generation of methane at depths of 2.3-4.3 km. For the Inner Zone of the Carpathian Foredeep, the calculated generation temperatures are 170-355 °C, which corresponds to depths of 8-17 km.

The values of $\delta^{13}C$ in methane of free gases of the Carpathian region vary in the range of -70 - -18 ‰ [13]. The distribution is displayed in the growth of $\delta^{13}C$ with depth increasing, and laterally – from the Outer Zone through the Inner Zone and Folded Carpathians to Transcarpathia. According to this researcher, methane and carbon dioxide gases have thermocatagenetic and thermometamorphic origins, and their sites do not correspond to the sites of formation.

Pavliukh [14] sees the formation of Bilche-Volytsia OGBA gas deposits in “the presence of supplying deep faults, the characteristic of their contact with permeable layers through which gas migrated into the raised structures; the presence of the formations with good reservoir properties, covered by aquitards, and the presence of the traps with favorable conditions for the accumulation of hydrocarbons and their long-term storage”.

Maievskii and Kurovets [15] consider the mechanism of formation of hydrocarbon deposits both in the Carpathian Foredeep and other oil and gas regions, as pulsation-sequential subvertical (vertical) migration of fluid hydrocarbon systems of deep genesis by zones of intersection of tectonic faults.

Despite a certain number of researches on water-dissolved gases of Bilche-Volytsia OGBA, the role of these gases in the mechanism of formation of hydrocarbon deposits and their oil-gas-bearing criteria are still not fully clarified.

The study aim was to determine the vertical and lateral gas-hydrogeochemical zonation of the Bilche-Volytsia oil and gas zone, to assess the role of water-dissolved gases in the formation of gas fields with the prospect of predicting new hydrocarbon deposits.

Geological structure and oil-gas-bearingness. Bilche-Volytsia OGBA is confined to the Outer Zone of the Carpathian Foredeep (Fig. 1). The Outer Zone from the north-east is separated from the old East European platform and the young West European platform by a system of regional tectonic faults. From the south-west, the formations of the Inner Zone of the Carpathian Foredeep cover the Outer Zone. The north-western part of the Outer Zone is formed by Badenian and Sarmatian sandy-clay deposits up to 4000-4500 m thick. Most of the gas fields of Bilche-Volytsia OGBA are connected with this part of these formations.

Neogene, Cretaceous and Jurassic rock complexes are regionally oil-gas-bearing here. The gas deposits are associated with Miocene sandstone reservoirs, oil deposits – with Upper Cretaceous sandstone reservoirs and Upper Jurassic limestones. The porosity of sandstones is 18–23 %. The depth of gas deposits varies from 500 to 1500 m, their thickness - from several tens of meters up to 200 m. The absolute free gas flow rate currently reaches 7 million m^3/day [17]. In the Bilche-Volytsia OGBA 72 hydrocarbon fields were explored, most of which are gas fields and only 7 - gas condensate, oil and oil-gas fields.

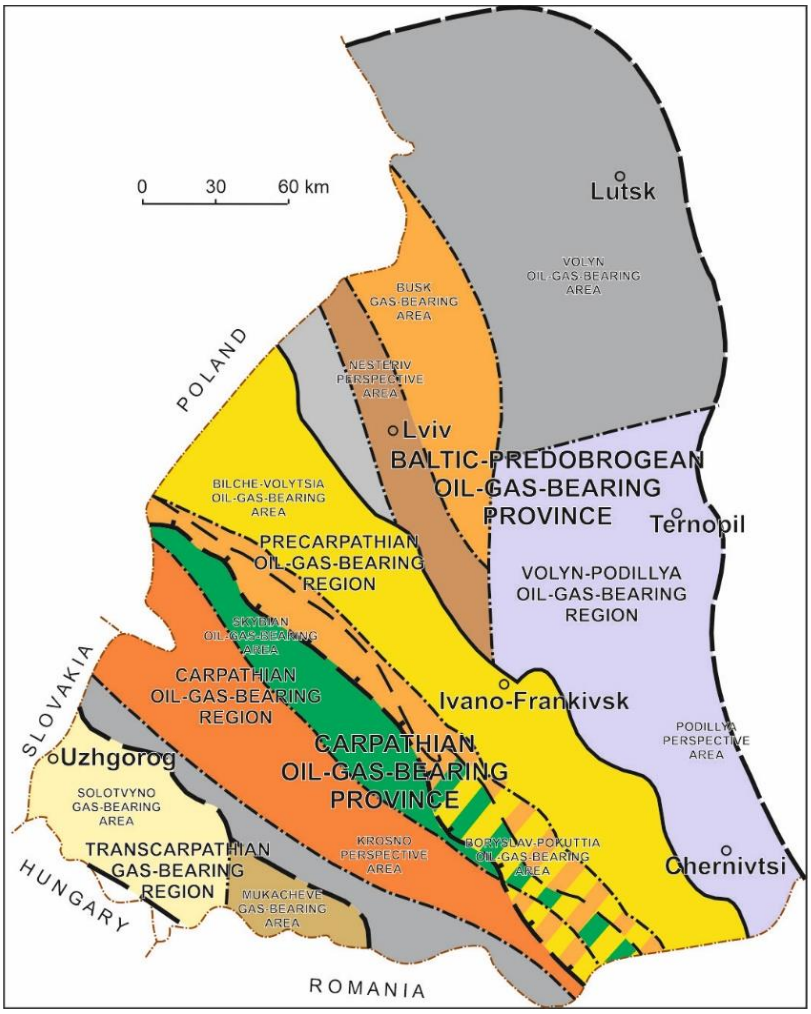

Fig. 1. Scheme of oil-gas-geological zoning of western Ukraine [16]

Data and research methods. Data on hydrogeochemical, geothermal, hydrodynamic parameters of the aquifers, qualitative and quantitative characteristics of water-dissolved and free gases of Bilche-Volytsia OGBA were obtained from the data funds of SE "Lvivgazvydobuvannia" and SE "Zakhidukrgeolo-giia", which were determined during testing of the boreholes (sampling of formation waters, measurements of formation pressures and temperatures, degassing of samplers). The total number of analyzed samples of water-dissolved gases used in this work is 255.

In this work, the following groundwater parameters are calculated and used:

Gas saturation of water – the volume of gas, dissolved in a unit of water volume under normal conditions (P = 760 mm Hg, T = 20°C). Gas saturation is the sum of all water-dissolved gases. The solubility of gases in water depends on the composition of the gas, the pressure of gas and water, temperature and TDS (total dissolved solids) in water. At pressures up to 5 MPa, the solubility of gases is calculating according to Henry's law. In oil and gas hydrogeology, this method is not used due to much higher pressures.

Saturation pressure – the pressure at which all the gas is in a dissolved state. If the formation water pressure is greater than the saturation pressure, the water is able to dissolve an additional amount of gas. If the water pressure is less than the saturation pressure, then part of the gas is released into the free phase and under favourable conditions can form deposits.

Gas saturation coefficient – is equal to the volume of water-dissolved gas divided by its solubility value under appropriate conditions (temperature, pressure, TDS in water) [18].

To determine the solubility of methane (the maximum possible content under certain PT conditions) were used graphs of its solubility, based on expe-rimental studies [19]. First, the solubility of methane in distilled water (S_{dw}) was determined at the appropriate formation pressure and temperature, then the correction coefficient (α) for TDS in water was determined. The general formula is as follows:

$$S = S_{dw}\alpha, \quad (1)$$

This method makes it possible to determine the solubility of methane basing on the same pressure of water and gas. The results of the calculations performed according to this method generally coincide with the methane solubility data calculated [20] based on the equation of state and Pitzer's theory [21].

The diagrams and tables [1] calculated on the basis of experimental data [22, 23] were also used to estimate the solubility of methane in formation waters at different methane pressures, TDS and water temperatures. To present the chemical composition of natural gases, which are the mixtures of several components, the classification by their prevalent components [13] was used.

Results and discussion. Water-dissolved gases of the ***Upper Jurassic aquifer (J₃)*** were disclosed by the boreholes in the north-western, central and south-eastern parts of the Bilche-Volytsia OGBA and taken in the number of 20 samples.

In the north-western and central parts, water-dissolved gases were found in the depth interval of 1131–3307 m (the Kokhanivka, Nyklovychi, Rudky, Hrushiv, Medenychi, Letnia, and Bohorodchany fields). They had mostly nitrogen-methane composition. The parts of methane were 76-95 % vol. (% of volume), nitrogen – 1-11 % vol., homologues of methane – 0-3 % vol., carbon dioxide – 0.1-3 % vol. There is a regularity of reducing the portions of methane by increasing the amount of nitrogen at a distance from gas deposits and deconsolidated areas of tectonic faults.

In the well intervals far from gas deposits (boreholes Nyklovychi-27, Letnia-41), water-dissolved gases with methane content of 20 and 24 % vol. accordingly, nitrogen – 41 and 51 % vol., carbon dioxide – 37 and 24% vol., homologues – up to 3.16% vol. were discovered.

In the south-eastern part of the Bilche-Volytsia OGBA, water-dissolved gases were sampled in the depth interval of 1595-5421 m at the Krasnoilsk gas field and Lopushna oil field. In the deep reservoirs of the Lopushnya field, the contents of methane are 65-94 % vol., nitrogen – 1-33 % vol., carbon dioxide – 0.3-3.6 % vol., homologues – 0.4-27 % vol. In the borehole Krasnoilsk-1 (the interval of 1570-1620 m, 400 m deeper than the existing gas deposit) water-dissolved gases with a methane content of 52 % vol., nitrogen – 46% vol., carbon dioxide - 1% vol., homologues – 1% vol. were found.

The gas saturation of the waters of the Upper Jurassic aquifer in the borehole Lopushna-8 within the productive horizon was 0.31 m^3/m^3. In the Krasnoilsk gas field, the gas saturation of the unproductive Upper Jurassic horizon was 0.29 m^3/m^3.

Water-dissolved gases of the ***Upper Cretaceous aquifer (K_2)*** were discovered by boreholes in the north-western and south-eastern parts of the Bilche-Volytsia OGBA and tested in the amount of 8 samples.

In the north-western part, water-dissolved gases were disclosed in the depth interval of 1129-2500 m by boreholes of Opary and Uhersko gas fields. They had nitrogen-methane composition. The content of methane was 90-93 % vol., nitrogen – 3.8-9.5% vol., carbon dioxide – 0-2.4% vol., homologues – 0-0.1% vol.

In the south-eastern part, water-dissolved gases were discovered by the boreholes of Kovalivka, Serhii, Lopushna, and Petrivetska areas at depths of 2290-5430 m. The gases had homologues-nitrogen-methane composition. In Kovalivka field (the interval of borehole perforation 2290-2300 m, which is deeper than 400 m from the gas deposit), the content of methane was 70 % vol., nitrogen – 27 % vol., carbon dioxide – 1.6 % vol., homologues were absent. In fact, such composition of water-dissolved gas, probably, can be considered as a background for such depths of the Upper Cretaceous aquifer.

In the deep structures of the Carpathian Foredeep in the boreholes that did not disclose hydrocarbon deposits (Sergii, Petrivetska areas, depths 5047-5410 m), the contents of water-dissolved methane were 80-81 % vol., nitrogen – 14-18 % vol., carbon dioxide – 0.4-2% vol., homologues – 0.9-2 % vol.

In the boreholes of Lopushna oil field (oil deposits with a gas cap, depth of perforation 4254-5430 m) the contents of methane (depending on the proximity to the deposit) were 80-94% vol., nitrogen – 1-4 % vol., carbon dioxide – 0.2-0.6% vol., homologues – 0.3-18 % vol.

Data on gas saturation of the Upper Cretaceous aquifer are absent.

The Lower Badenian aquifer (N_1b_1) is sporadically distributed in north-western and south-eastern parts) of the Bilche-Volytsia OGBA. It was studied for the content of water-dissolved gases in the north-western parts by boreholes of the areas of Novosilky, Rudky, and Pivdennograbynska. The methane content was 86-98 % vol., nitrogen – 2-9.8% vol., carbon dioxide – 0.2-4.3% vol., methane homologues – 0-4.2 % vol. There is a clear pattern of methane increasing in the approach to gas deposits. Data on gas saturation of this aquifer are absent.

The Upper Badenian aquifer (N_1b_2) is widespread in the central and south-eastern parts of the Bilche-Volytsia OGBA. In the central part, the aquifer for the content of water-dissolved gases was tested in the interval of depths of 677-1823 m in the areas of Gai, Kadobno, Hrynivka in the amount of 11 samples. The gases had mainly methane composition: methane – 89-99% vol., nitrogen – 0.6-10 % vol., carbon dioxide – 0.16-0.93 % vol., homologues – 0.11-5.29 % vol.

In the south-eastern part, water-dissolved gases were taken in the amount of 50 samples in the areas of Pylypiv, Debeslavtsi, Kovalivka, Krasnoilsk in the depth interval of 189-2040 m. The gases had also methane, occasionally nitrogen-methane composition. The relative content of methane was 62-99 % vol., nitrogen – 0.4-34 % vol., carbon dioxide – 0.03-2.2 % vol., homologues of methane – 0.06-0.69 % vol. It is also confirmed a clear regularity of methane content increasing in the approach to a gas deposit, as well as an increase in the relative contents of methane homologues with depth.

The lateral distribution of water-dissolved methane in the Upper Badenian aquifer is reflected in the increasing of its relative portion from the East European platform in the direction of the Carpathian deep structures (Fig. 2) with the deepening of the aquifers and increasing of hydrogeological isolation es. The increase of methane portion is due to the decrease of nitrogen and carbon dioxide portions. The high methane contents are also spatially correlated with transverse tectonic faults, which may detect them as ways of vertical migration of water-hydrocarbon mixtures.

The gas saturation of the waters varies in the range of 0.2-1.3 m^3/m^3, in some cases reaching 6 m^3/m^3 (Krasnoilsk gas field).

The Lower Sarmatian aquifer (N_1s_1) is distributed in the north-western and central parts of the Bilche-Volytsia OGBA.

This aquifer was tested for the content of water-dissolved gases in the areas of Dashava, Uhersko, Grudiv, Letnia Medenychi, Grushiv, Dovhiv, Tyniv, Susoliv, Mainychi, Gorodok, Novosilky, Zaluzhany, Pyniany, Sadkovychi, Khidnovychi, Vyshnia, Vyzhomlia, Kokhanivka, Svydnytsia, Bohorodchany in the amount of 141 samples in the interval of depths 310-2445 m.

The gases had mainly methane, occasionally nitrogen-methane composition. The portion content of methane was 69-99 % vol., nitrogen – 0.2-28 % vol., carbon dioxide – 0.1-4 % vol., homologues – 0.03-60 % vol. The gas saturation of waters of the Vyzhomlia field was 0.20-0.34 m^3/m^3, the gas saturation was increasing from 0.5 to 2 m^3/m^3 in the Khidnovychi field with depth and approaching to the deposit.

In the lateral plan of the north-western part of Bilche-Volytsia OGBA, the content of water-dissolved methane spatially clearly correlates with TDS in water (Fig. 3, Fig. 4), the high values of which shows the structures of high hydrogeological isolation. More open hydrogeological structures are characterized by smaller amounts of methane due to increased nitrogen content (Fig. 5).

The statistical analysis of the gas-hydrogeochemical system of the set of aquifers of Bilche-Volytsia OGBA (Table 1) did not reveal the correlations of methane, nitrogen and carbon dioxide either with the depth of the aquifer or with the geochemical characteristics of formation waters. High inverse correlations exist between methane and nitrogen and carbon dioxide. This situation indicates different nature of gases and different ways of gas saturation. Obviously, the spatial-temporal mechanisms of penetration of these gases into water also are different.

The contents of methane homologues are directly correlated with the depth of the aquifer, TDS in water, the contents of Na^+, Ca^{2+}, Cl^-, Br^- ions. The highest amounts of water-dissolved methane homologues are typical for the Jurassic and Cretaceous aquifers of the submerged south-eastern part of the Outer zone of Carpathian Foredeep. In fact, these aquifers are characterized by high values of TDS in water (up to 300 g/dm^3) due to high concentrations of Na^+, Ca^{2+}, Cl^-, SO_4^{2-} and other ions [25].

These aquifers lie here at depths of 4000-5500 m. The contents of methane homologues in the formation waters which contact with oil or gas deposits (Lopushna oil field) are 6-27% vol. and directly correlate with proximity to the deposit. With the distance from the deposit, and in boreholes that have not opened hydrocarbon deposits, the proportion of methane does not exceed 80 % vol., homologues – 2% vol. However, the nitrogen content here reaches 30 % vol., carbon dioxide – 2 % vol. [26].

According to research [27], the correlation of water-dissolved methane homologues contents with depth in comparison both between individual fields of Bilche-Volytsia OGBA and between deposits within one field is a consequence of vertical gas mixtures migration and their gravitational differentiation on the ways of traps filling.

Statistical pairwise relationships between the components of water-dissolved gas and the depth of the aquifer in the Bilche-Volytsia OGBA also confirm the correlation of depth only with the contents of methane homologues. The linear dependence of the contents of methane homologues with the values of formation pressures and temperatures, which are

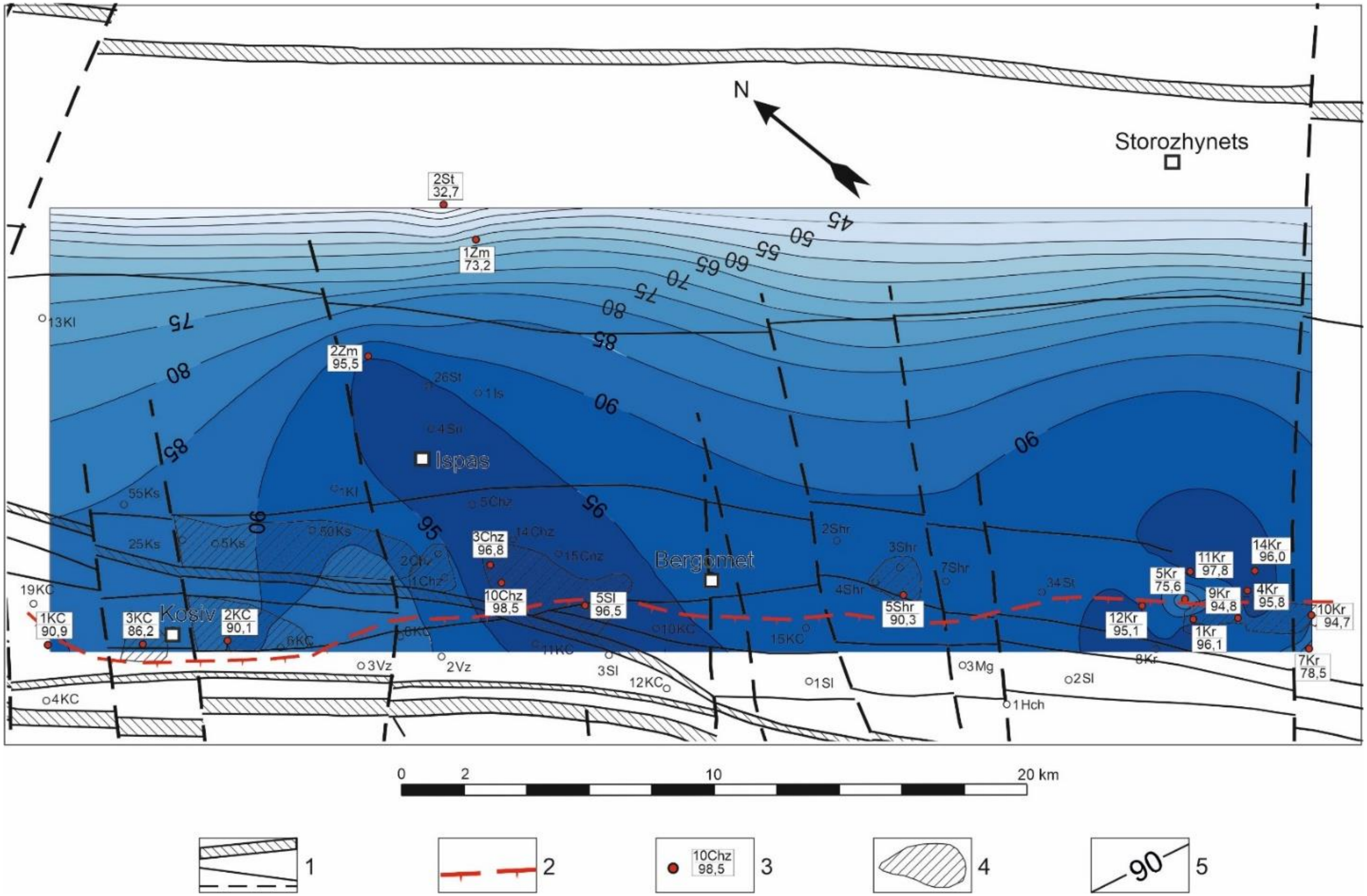

Fig. 2. Map-scheme of the distribution of the water-dissolved methane portion in the aquifer N_1b_2 of the south-eastern part of the Bilche-Volytsia OGBA (structural basis ([24]): 1 - tectonic faults; 2 - Stebnytsia cover line; 3 – the well, numerator - its name, denominator - the portion of water-dissolved methane, % vol.; 4 - gas deposit; 5 - isoline of the methane portion

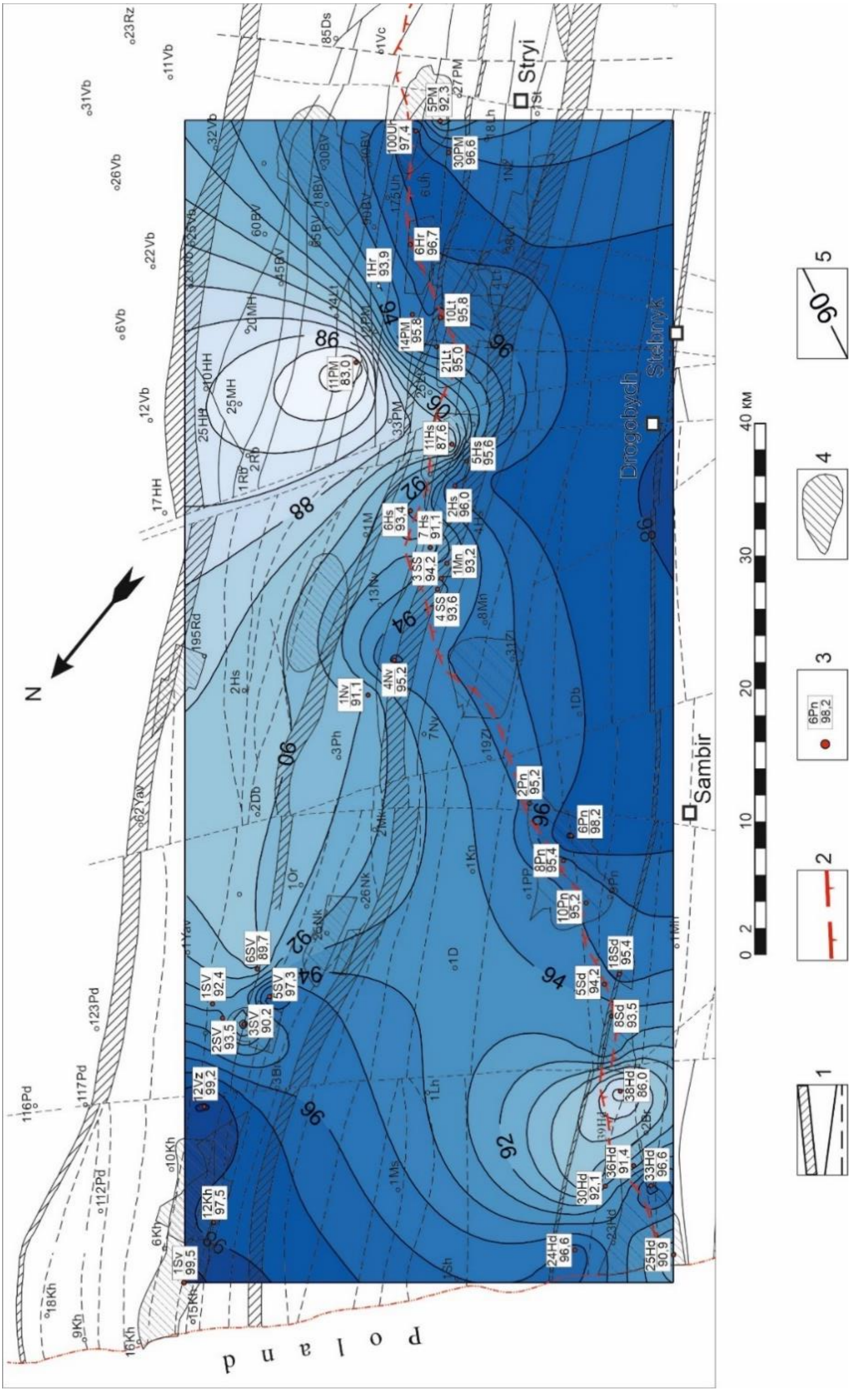

Fig. 3. Map-scheme of the distribution of the water-dissolved methane portion in the aquifer N_1S_1 of the north-western part of the Bilche-Volytsia OGBA (structural basis [24]): Legend – as in Figure 2

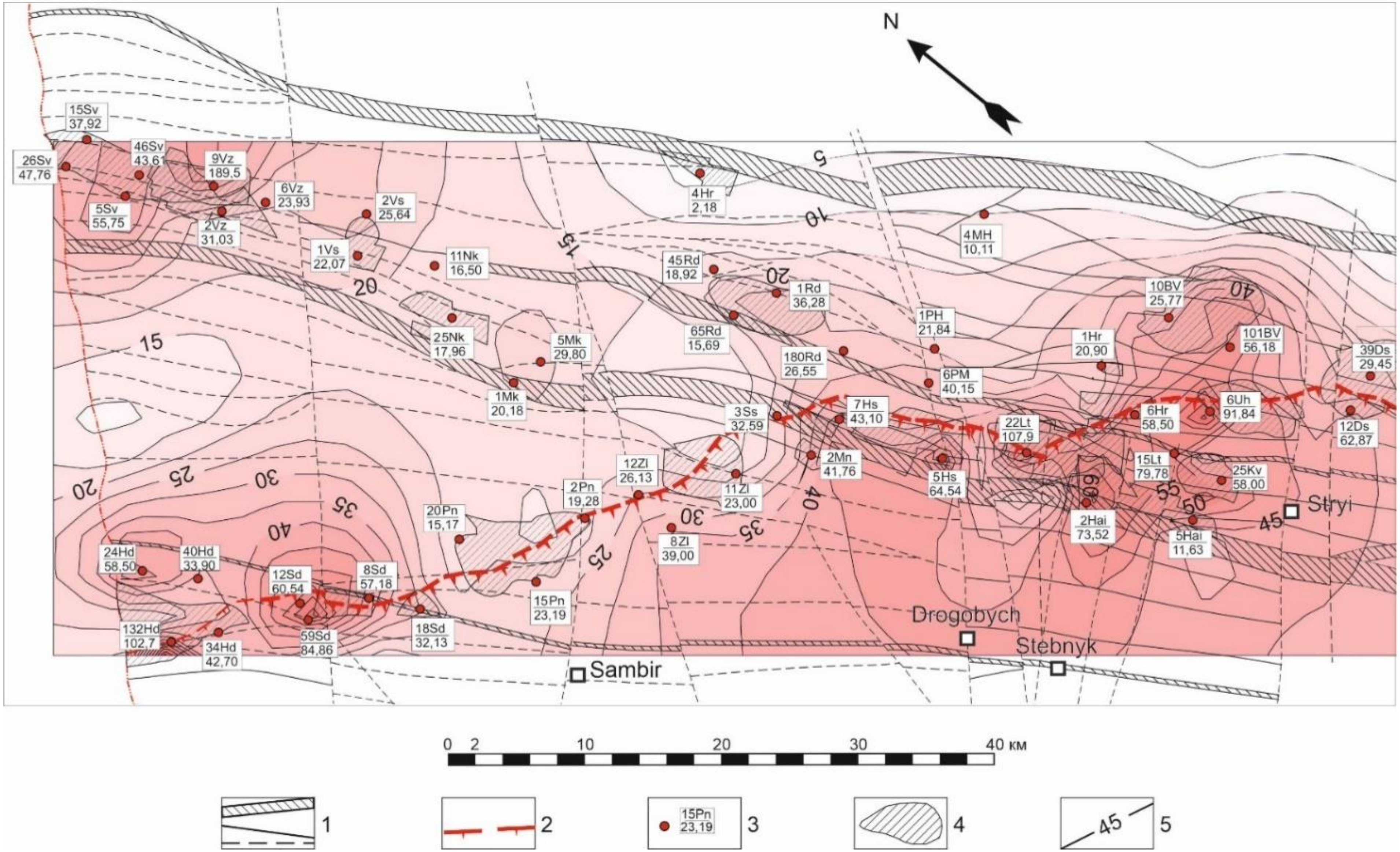

Fig. 4. Map-scheme of TDS values in the waters of the aquifer N_1s_1 of the north-western part of the Bilche-Volytsia OGBA (structural basis [24]):
1 - tectonic faults; 2 - Stebnytsia cover line; 3 – the well, numerator - its name, denominator – TDS in the water, mg/dm³; 4 - gas deposit; 5 - isoline of TDS

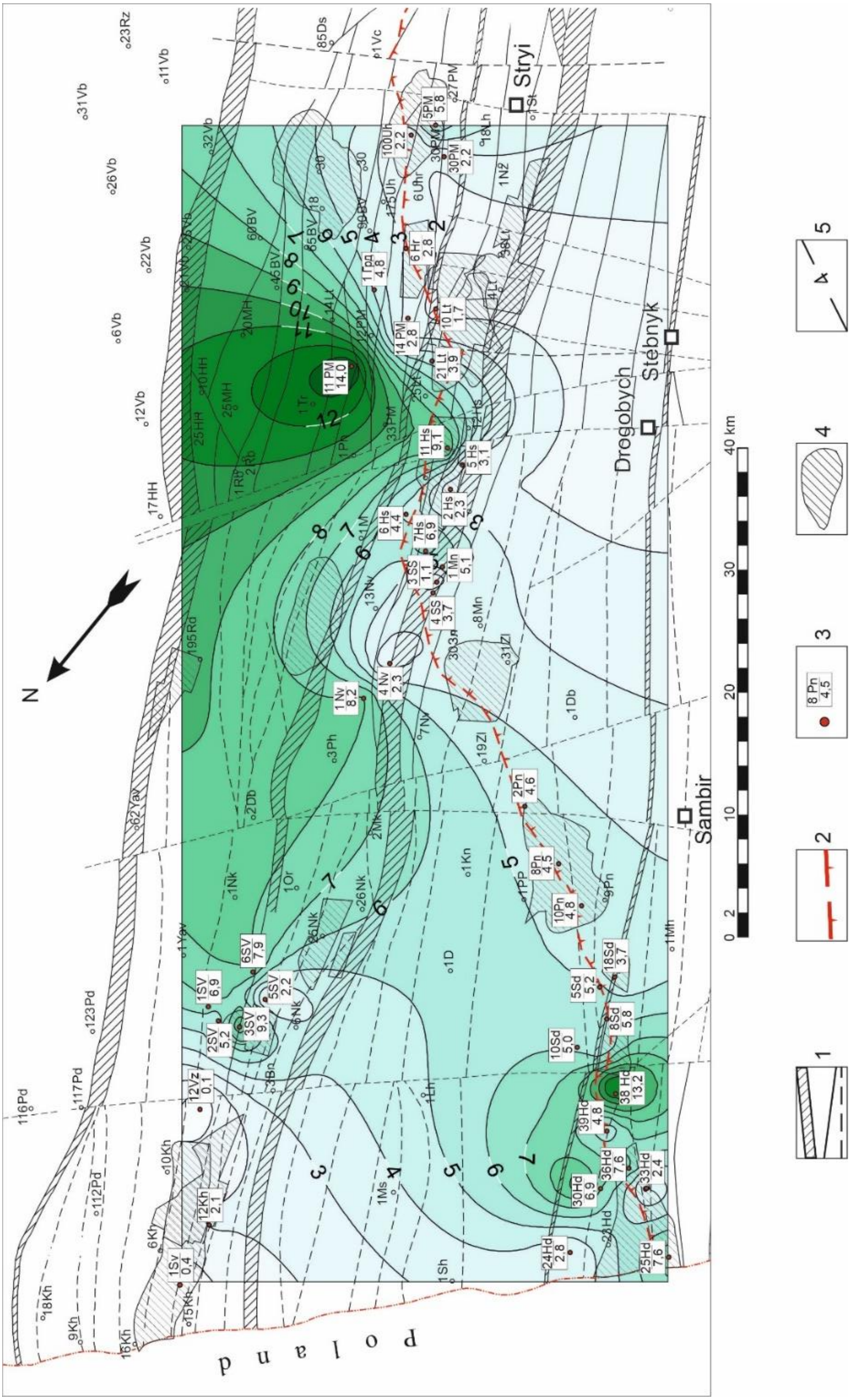

Fig. 5. Map-scheme of the distribution of the water-dissolved nitrogene portion in the aquifer N_1s_1 of the north-western part of the Bilche-Volytsia OGBA (structural basis [24]): Legend – as in Figure 2

Table 1

Correlation matrix of parameters of the gas-hydrogeochemical system
of the set of aquifers of Bilche-Volytsia OGBA (R threshold value = 0.22, N = 177)

	D*	**CH_4**	**N_2**	**CO_2**	**H****	**TDS**	**Na+K**	**Ca**	**Mg**	**NH_4**	**Cl**	**SO_4**	**HCO_3**	**Br**	**J**	**rNa/ rCl**
D*	1,00	-0,13	0,03	0,10	**0,58**	**0,71**	**0,71**	**0,73**	**0,27**	**0,32**	**0,71**	0,04	0,17	**0,33**	0,04	-0,01
CH_4	-0,13	1,00	**-0,95**	**-0,72**	-0,11	-0,16	-0,15	-0,16	-0,15	-0,08	-0,16	-0,06	0,11	-0,15	-0,02	0,12
N_2	0,03	**-0,95**	1,00	**0,53**	-0,02	0,10	0,09	0,12	0,14	0,05	0,11	0,06	-0,13	0,08	0,04	-0,12
CO_2	0,10	**-0,72**	**0,53**	1,00	0,08	0,04	0,04	0,00	0,09	0,12	0,04	0,08	-0,05	0,03	-0,04	-0,06
H**	**0,58**	-0,11	-0,02	0,08	1,00	**0,56**	**0,57**	**0,55**	0,12	0,14	**0,56**	-0,01	-0,03	**0,26**	-0,04	-0,11
TDS	**0,71**	-0,16	0,10	0,04	**0,56**	1,00	**0,99**	**0,92**	**0,63**	**0,51**	**1,00**	0,16	**-0,25**	**0,63**	**0,22**	**-0,47**
Na+K	**0,71**	-0,15	0,09	0,04	**0,57**	**0,99**	1,00	**0,88**	**0,58**	**0,48**	**0,99**	0,12	**-0,25**	**0,65**	**0,26**	**-0,46**
Ca	**0,73**	-0,16	0,12	0,00	**0,55**	**0,92**	**0,88**	1,00	**0,55**	**0,49**	**0,93**	0,03	**-0,22**	**0,44**	0,06	**-0,45**
Mg	**0,27**	-0,15	0,14	0,09	0,12	**0,63**	**0,58**	**0,55**	1,00	**0,56**	**0,62**	**0,70**	**-0,27**	**0,49**	0,14	**-0,48**
NH_4	**0,32**	-0,08	0,05	0,12	0,14	**0,51**	**0,48**	**0,49**	**0,56**	1,00	**0,51**	**0,24**	-0,18	**0,30**	-0,05	**-0,43**
Cl	**0,71**	-0,16	0,11	0,04	**0,56**	**1,00**	**0,99**	**0,93**	**0,62**	0,51	1,00	0,13	**-0,27**	**0,63**	**0,22**	**-0,49**
SO_4	0,04	-0,06	0,06	0,08	-0,01	0,16	0,12	0,03	**0,70**	0,24	0,13	1,00	-0,08	0,13	-0,01	-0,05
HCO_3	0,17	0,11	-0,13	-0,05	-0,03	**-0,25**	**-0,25**	**-0,22**	**-0,27**	-0,18	**-0,27**	-0,08	1,00	**-0,23**	-0,14	**0,65**
Br	0,33	-0,15	0,08	0,03	**0,26**	**0,63**	**0,65**	**0,44**	**0,49**	0,30	**0,63**	0,13	**-0,23**	1,00	**0,58**	**-0,39**
J	0,04	-0,02	0,04	-0,04	-0,04	**0,22**	**0,26**	0,06	0,14	-0,05	**0,22**	-0,01	-0,14	**0,58**	1,00	-0,09
rNa/ rCl	-0,01	0,12	-0,12	-0,06	-0,11	**-0,47**	**-0,46**	**-0,45**	**-0,48**	-0,43	**-0,49**	-0,05	**0,65**	**-0,39**	-0,09	1,00

*D - depth; **H - methane homologues

generally functions of the depth of the aquifer, has been recognized. There are no clear dependences of gas saturation on the depth, pressure and temperature of aquifers.

The growing of gas saturation of deep groundwaters in the Bilche-Volytsia OGBA is primarily due to the increase of the methane content, which is confirmed by a clear linear relationship between these parameters. At the same time, clear inverse linear dependences of water gas saturation with rNa/rCl values and sulfate contents in waters aredisplayed. It is known that reduced values of rNa/rCl and sulfates in water indicate a closed hydrodynamic system, which is most favourable for the preservation of hydrocarbon accumulations [5].

The distribution of free gas contents with the depth of the gas deposits in the Bilche-Volytsia OGBA reflects the predominance of methane, the amount of which is 90-99% vol. in the range of depths of 300-2000 m. In the interval of depths of 4000-5000 m its contents decrease to 65-90% vol. by increasing the contents of its homologues to 30% vol. The contents of nitrogen and carbon dioxide do not change significantly with depth increasing.

The coefficient $Ar \cdot 100/N_2$, proposed Savchenko [28] to determine the portion of "non-air" nitrogen, in most samples of water-dissolved gases is less than or close to 2.68 (for fresh water in surface conditions it is 2.68). Moreover, the distribution of this coefficient values does not correlate with the depth of gas-saturated aquifers. At the same time, the values of $Ar \cdot 100/N_2$ are quite closely inversely correlated with the values of rNa/rCl, the reduced values of which reflect long-term ion exchange processes between waters and water-bearing rocks in conditions of stagnant hydrodynamic regime.

Taking into account that the main amount of argon in gases of air origin, we can talk about the enrichment of the deep groundwaters of the Bilche-Volytsia OGBA with nitrogen of non-air origin. The source of nitrogen in water of the stagnant hydrodynamic regime areas is considered to be organic matter of rocks, bound nitrogen of rocks released during metamorphism, nitrogen of deep genesis [29,30].

Conclusions. 1. Nitrogen-methane composition of water-dissolved gases of the Upper Jurassic and Upper Cretaceous aquifers in combination with specific geochemical characteristics of groundwater of the north-western and central parts of the Bilche-Volytsia OGBA indicate the open hydrodynamic conditions, which, in general, are unfavourable for the formation and preservation of hydrocarbon deposits. Only within certain local closed structures there is a possibility to preserve the deposits.

2. In deep-submerged reservoirs of the south-eastern part of this area, water-dissolved gases of the Upper Jurassic and Upper Cretaceous aquifers are characterized by high contents of methane homo-

logues, the portions of which increase with approaching to oil deposits (Lopushna field). Also, when approaching the deposits, the gas saturation of the waters increases.

Unproductive aquifers are characterized by low gas saturation and high nitrogen content.

3. In the Upper Badenian aquifer, water-dissolved gases are mainly methane, occasionally nitrogen-methane. A clear regularity in the growth of methane contents as it approaches the gas deposit, as well as an increase in the relative contents of methane homologues with depth.

The lateral distribution of water-dissolved methane is presented in the growth of its portions from the West and East European platforms in the direction of the sub-submerge of the Carpathians, which is due to an increase in the degree of hydrogeological closure of structures. Increased methane contents also spatially tend to transverse tectonic faults, which determines their role in the vertical migration of water-hydrocarbon mixtures.

4. Water-dissolved gases of the highly productive Lower Sarmatian aquifer are mainly methane, occasionally nitrogen-methane. The gas saturation of the waters directly correlates with the proximity to gas deposits.

Laterally, the portion of water-dissolved methane is directly correlated with TDS of water, the high values of which accordingly reflect the structures of a high degree of hydrogeological stagnation.

5. Statistical analysis of the system of dissolved gases of the Bilche-Volytsia OGBA revealed high inverse correlations between methane and nitrogen, methane and carbon dioxide, which indicates their different sources of origin and spatio-temporal mechanisms of water saturation with these gases.

6. Deep hydrodynamically closed aquifers of the Bilche-Volytsia OGBA often are marked by high nitrogen contents of non-air origin. Its source can be rock organic matter, bound rock nitrogen released during metamorphism, nitrogen of deep genesis.

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Бильче-Волицкий нефтегазоносный район (НГР) является крупнейшим по запасам углеводородов в Карпатской нефтегазоносной провинции Украины. Здесь открыто 72 месторождения газа и газового конденсата и 3 месторождения нефти. Растворенные в воде газы в разведывательной нефтегазовой гидрогеологии являются основными критериями нефтегазового потенциала. Их количественные и качественные характеристики позволяют изучить условия образования и сохранения месторождений углеводородов и определить перспективные районы поиска. Целью исследования было установить вертикальную и латеральную газо-гидрогеохимическую зональность Бильче-Волицкого НГР, оценить роль растворенных в воде газов в формировании газовых месторождений с перспективой прогнозирования новых залежей углеводородов. Методы исследования базировались на интерпретации компонентного состава водорастворенных газов, расчета и интерпретации газнасыщения воды, давления насыщения, коэффициента насыщения воды газом. Для определения растворимости метана были использованы графики на основе экспериментальных исследований и расчетов с научных публикаций. Исходные данные по гидрогеохимических, геотермических, гидродинамических параметрах водоносных горизонтов, качественных и количественных характеристик, растворенных в воде и свободных газов Бильче-Волицкого НГР были определены при испытании скважин. Результаты исследований показали, что частицы растворенного в воде метана увеличиваются со стороны Западно- и Восточноевропейской платформ в направлении погружения Карпат, что является следствием более высокой степени гидрогеологической закрытости структур. Установлено, что растворенный в воде метан, азот и углекислый газ имеют различные источники происхождения и различные пространственно-временные механизмы насыщения воды. Гидродинамически закрытые структуры (благоприятные для образования и сохранения месторождений углеводородов) характеризуются высоким относительным и абсолютным содержанием растворенного метана. Повышенное содержание метана также пространственно тяготеет к зонам поперечных тектонических разломов, что указывает на их роль в вертикальном транспортировании водно-углеводородных смесей.

Ключевые слова: *Бильче-Волицкий нефтегазоносный район, водорастворенные газы, газонасыщенность воды.*

Газо-гідрогеохімічні умови Більче-Волицького нафтогазоносного району (Передкарпатський прогин, Україна)

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Більче-Волицький нафтогазоносний район (НГР) є найбільшим за запасами вуглеводнів у Карпатській нафтогазоносній провінції України. Тут відкрито 72 родовища газу та газового конденсату та 3 родовища нафти. Розчинені у воді гази в розвідувальній нафтогазовій гідрогеології є основними критеріями нафтогазового потенціалу. Їх кількісні та якісні характеристики дозволяють вивчити умови утворення та збереження родовищ вуглеводнів та визначити перспективні райони пошуку. Метою дослідження було виявити вертикальну та латеральну газо-гідрогеохімічну зональність Більче-Волицького НГР, оцінити роль розчинених у воді газів у формуванні газових родовищ з перспективою прогнозування нових покладів вуглеводнів. Методи дослідження базувалися на інтерпретації компонентного складу водорозчинених газів, розрахунку та інтерпретації газонасичення води, тиску насичення, коефіцієнта насичення води газом. Для визначення розчинності метану були використані графіки на основі експериментальних досліджень та розрахунків з наукових публікацій. Початкові дані щодо гідрогеохімічних, геотермічних, гідродинамічних параметрів водоносних горизонтів, якісних та кількісних характеристик розчинених у воді та вільних газів Більче-Волицького НГР були визначені під час випробування свердловин. Результати досліджень показали, що частки розчиненого у воді метану збільшуються зі сторони Західно- та Східноєвропейської платформ у напрямку занурення Карпат, що є наслідком більш високого ступеня гідрогеологічної закритості структур. Встановлено, що розчинений у воді метан, азот та вуглекислий газ мають різні джерела походження та різні просторово-часові механізми насичення води. Гідродинамічно замкнуті структури (сприятливі для утворення та збереження родовищ вуглеводнів) характеризуються високим відносним та абсолютним вмістом розчиненого метану. Підвищений вміст метану також просторово тяжіє до зон поперечних тектонічних розломів, що вказує на їх роль у вертикальному транспортуванні водно-вуглеводневих сумішей.

***Ключові слова**: Більче-Волицький нафтогазоносний район, водорозчинені гази, газонасиченість води.*

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