

Methane Concentration and $\delta^{13}\text{C}$ Isotopic Signature in Methane over Arctic Seas in Summer and Autumn 2020

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Abstract—We have studied the assessment of methane content in the near atmospheric layer of the Arctic seas and the sources of its emissions. Continuous atmospheric measurements of methane concentration and $\delta^{13}\text{C}$ isotope signature in methane were carried out during two expeditions aboard the R/V *Akademik Mstislav Keldysh* in the summer and autumn of 2020. The minimum CH_4 concentration (mean 1.937 ± 0.010 ppm) were obtained over the North and Norwegian Seas. The maximum CH_4 concentration took place above the Kara (1.998 ± 0.030 ppm on average, maximum values are up to 2.000–2.040 ppm) and White (2.006 ± 0.030 ppm on average) seas. The average values of $\delta^{13}\text{C}$ in the studied region were close to that typical for the Northern Hemisphere: -47.5% . Regional changes in $\delta^{13}\text{C}$ allowed CH_4 sources to be identified and showed that the methane concentration in the surface atmospheric level is mainly affected by the direction of air-mass advection.

Keywords: Arctic, methane, shipboard measurements, stable isotope, air masses, trajectory analysis

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INTRODUCTION

Significant methane reserves are concentrated in the Arctic region, but their quantitative assessment and emission rates are poorly understood and have significant uncertainties. So, for example, methane emissions from the sea surface at high latitudes, according to various estimates, can differ by an order of magnitude: from 4 to 25 Tg CH_4 /year [10, 17–20, 25]. The sources of methane in the water column can be gas seeps at the bottom (thermogenic or fossil methane), its generation in bottom sediments and the water column, and lateral transport with river runoff. Information for determining the sources of methane can be obtained from analysis of data from direct measurements of the carbon isotopic composition of methane [3, 6, 11]. Isotopically lightened CH_4 is mainly a product of biological generation, while the value of $\delta^{13}\text{C}$ varies markedly with the degree of oxidation—in particular, during transport in soil or water. Isotopically weighted CH_4 usually comes from pyrogenic and thermogenic sources such as biomass burning and coal mines. However, even with careful measurement of methane from a particular source, there are significant uncertainties that are associated, for example, with temperature, soil moisture, geographical location, and season. For swamps in Finland, depending on the sea-

son, the value of $\delta^{13}\text{C}$ can vary from -60 to -78% , with the highest repeatability of values being from -68 to -74% [5]. Methane entering the atmosphere due to the decomposition of gas hydrates corresponds on average to $\delta^{13}\text{C} -55\%$ [7]; from fires, -26% [4]; and from gas leaks from deposits and gas pipelines, -42% [8, 16, 21]. At the same time, it is not always possible to distinguish the contribution of certain sources to methane generation far from them by the isotopic marker alone due to the complexity of measurements and significant mixing of air with distance from the sources [1, 2, 13], as a result of which the values of the isotopic signature change towards average values of 47.5% .

For more accurate analysis of the isotopic composition of carbon in CH_4 , it is necessary to use the Keeling plot method [9, 14]. Using the Keeling method yields the maximum effect in the case of large variations in methane concentration. The difficulty in identifying sources during ship-based measurements in the near layer of the atmosphere consists in that the air flow is inhomogeneous and, passing over different areas, accumulates methane from several sources.

Additional information about the origin of air masses in the areas of measurements and about regional variations in the concentration of methane in the atmosphere can be obtained from the analysis of

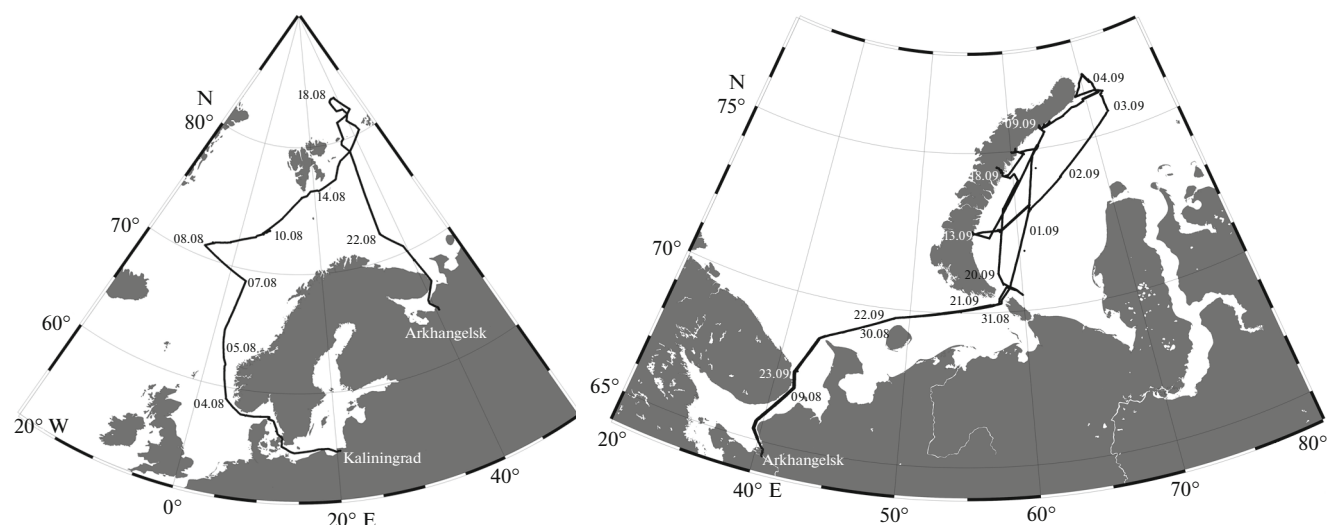


Fig. 1. Routes of expeditions AMK-80 (left) and AMK-81 (right); the dates of passing waypoints are specified in the format day.month.

backward trajectories. They can be used to identify the spatial inhomogeneity of the distribution of methane concentration in the lower layers of the atmosphere and to identify areas with the maximum and minimum contribution of different sources to the observed values of CH_4 concentration.

This paper presents the results of onboard measurements of the concentration of methane and its isotope signature carried out in the marine Arctic in 2020. All expeditions were carried out during the period when the seas were ice-free, and, according to [12, 25], emissions in the ocean–atmosphere system were insignificant. This allows us to consider air mass advection as the main factor in the variability of methane concentration over the sea surface during the observation period. Work results are of interest not only, in fact, for assessing the regional values of the concentration of CH_4 in the Arctic atmosphere, but also to analyze the regional transport of air masses and its influence on the dynamics of methane concentration in the Arctic air.

MEASUREMENTS

In the periods from July 31 to August 25 and from August 27 to September 23, 2020, continuous measurements of methane concentrations and the value of $\delta^{13}\text{C}$ in methane in driving air. Flight AMK-80 ran from the port of Kaliningrad through the Baltic Sea and the Danish Straits to the North and Norwegian Seas, then to the Barents Sea to 83°N , and further across the White Sea to the port of Arkhangelsk. The route of cruise AMK-81 passed from the port of Arkhangelsk through the White and Barents seas to the Kara Sea, where observations were made in the areas of the St. Anna Trough and the Novaya Zemlya archipelago. Flight route schemes are shown in Fig. 1.

For measurements, a specialized measuring complex based on a methane isotope composition analyzer (hereinafter referred to as the “complex”) of the G2132-i type manufactured by Picarro Inc. was used (United States). The complex and its periodic calibrations immediately before and after sea expeditions are described in detail in [1, 2, 13]. On cruise AMK-80, the complex was located in the ship’s meteorological laboratory, and on cruise AMK-81, in the pilot room. The air intake was carried out on board the ship at a height of 18.5 m above sea level. Air was supplied to the device by a 10-m-long pipeline at an air flow rate of 1.5 L/min.

As a result of the observations, almost continuous series of data were obtained on the concentration of CH_4 and $\delta^{13}\text{C}$ with a time resolution of 1 min for 26 days during 80-AMC and 28 days during 81-AMC.

RESULTS

For analysis of observational data, sections were allocated for individual seas: Baltic, North and Norwegian, White, Barents, and Kara. For these areas, the statistical characteristics of the methane concentration and $\delta^{13}\text{C}$ (Table 1), as well as the distribution functions of CH_4 content, are plotted and $\delta^{13}\text{C}$ (Figs. 2a, 2b).

The resulting data set allows us to identify some regularities in the spatial and temporal variability of the methane concentration in the lower atmosphere. During cruise AMK-80, the minimum values of methane concentration were observed in the North and Norwegian seas. On some sections of the route, it decreased to 1.913 ppm, while the average value was fixed at the level of 1.937 ± 0.010 ppm (Table 1); the methane-concentration distribution is close to normal (Fig. 2a). Also, reduced values of methane concentration in near-water air were observed over the Baltic

Table 1. Statistical characteristics of minute series of measurements of $\delta^{13}\text{C}$ and CH_4 concentrations in a near-water atmosphere for the expeditions AMK-80 and AMK-81

Region (dates)	min	max	min	max	Mean \pm std. deviation	
	$\delta^{13}\text{C}$, ‰		CH_4 , ppm		$\delta^{13}\text{C}$, ‰	CH_4 , ppm
Cruise AMK-80						
Port of Kaliningrad (July 28–31)	–57.5	–46.3	1.967	4.728	-50.8 ± 1.3	2.058 ± 0.090
Baltic Sea (July 31–Aug. 2)	–53.9	–47.4	1.932	2.018	-50.3 ± 1	1.949 ± 0.010
North, Norwegian seas (Aug. 2–13)	–54.5	–47.1	1.913	2.110	-51.4 ± 1	1.937 ± 0.010
Barents Sea and Arctic Ocean (Aug. 13–23)	–54.5	–47.2	1.943	2.166	-50.8 ± 1.2	1.980 ± 0.030
White Sea (Aug. 24–25)	–54.0	–49.4	1.964	2.082	-51.5 ± 0.7	2.006 ± 0.030
Port of Arkhangelsk (Aug. 25)	–52.4	–49.9	1.972	2.005	-51.3 ± 0.6	1.984 ± 0.010
Cruise AMK-81						
Port of Arkhangelsk (Aug. 26–28 and Sep. 23)	–54.0	–48.6	1.979	2.631	-51.2 ± 1	2.065 ± 0.090
White Sea (Aug. 27–29 and 22–23)	–56.1	–48.3	1.958	2.170	-51.5 ± 1.1	1.989 ± 0.020
Barents Sea (Aug. 29–31 and Sep. 20–22)	–53.6	–44.5	1.958	2.130	-49.6 ± 1.3	1.987 ± 0.020
Kara Sea (Aug. 31–Sep. 20)	–55.9	–44.1	1.955	2.216	-50.3 ± 1.7	1.998 ± 0.030

Sea (1.949 ± 0.010 ppm). $\delta^{13}\text{C}$ values from the Baltic to the Norwegian Sea varied on average from -51.5 to -50.3‰ ; the maximum values were in the range from -47.4 to -47.1‰ , and the minimum values were from -54.5 to -53.9‰ .

As the scientific vessel moved eastward, the content of methane in the atmosphere increased, and natural CH_4 sources, the main ones of which, most likely, are wetlands [18], the emissions from which are most intense during the observation period [23]. At the same time, the methane-concentration values had a bimodal distribution, which is associated with a change in the direction of air-mass transfer (Fig. 2a). The average values of the maximum concentrations of CH_4 in the atmosphere during the cruise AMK-80, excluding the territory of ports, reaching 2.006 ppm, were observed over the White Sea, where emissions from swamps and anthropogenic sources are combined. $\delta^{13}\text{C}$ in this region also changed towards lighter values and ranged from -54.0 to -49.4‰ .

During the cruise AMK-81, the lowest average methane concentrations were observed in the White and Barents seas (average 1.987–1.989 ppm, minimum 1.958 ppm) (Table 1, Fig. 2b). At the same time, in the White Sea, the values of the isotopic signature turned out to be lower than in the eastern regions and varied from -56.1 to -48.3‰ . Similar variability of $\delta^{13}\text{C}$ during this period may indicate a gradual change from west to east of the proportion of methane in the atmosphere from various sources. The longest measurements during the cruise AMK-81 were carried out in the Kara Sea, to the east of the Novaya Zemlya archipelago, where the vessel was often drifting during expedition work. The average concentration of methane in the atmospheric near layer of this region was at

the level of 1.998 ± 0.030 ppm, and that of CH_4 was from 2.000 to 2.040 ppm (Fig. 2b).

The ports of Arkhangelsk and Kaliningrad, where many anthropogenic sources are concentrated, had the highest concentrations of methane in the atmosphere (Table 1). The maximum at a single value of up to 4.728 ppm was recorded in Kaliningrad. However, due to the significant influence of strong local sources of methane in ports, these data were not analyzed in detail in this work.

Reverse Trajectory Analysis

For all expeditions, 5-day reverse trajectories of air flows were analyzed that were constructed on the basis of the NOAA Hysplit Lagrangian model [15, 22]. To construct the trajectories, GDAS $1^\circ \times 1^\circ$ meteorological fields were used, and the trajectories were calculated with a step of 6 h for 00.00, 06.00, 12.00, and 18.00 UTC with an end point of 500 m a.s.l. Figure 3 shows the series of methane concentration and $\delta^{13}\text{C}$ with a generalized direction of transfer and the location of the R/V.

The AMK-80 expedition was carried out in late July–early August during the period of the seasonal minimum of methane content in the near-water air. According to analysis of the return trajectories, the low content of methane corresponds to the removal of air masses from the southwest from the North Atlantic. During this period, the concentration of CH_4 reached 1.913 ppm. The reduced values of methane content also correspond to the removal from the west and from the northwest, in conditions in which the flow did not cross the Scandinavian Peninsula. So, during the movement of the ship through the North and Norwegian seas, there was an inflow into the region of air

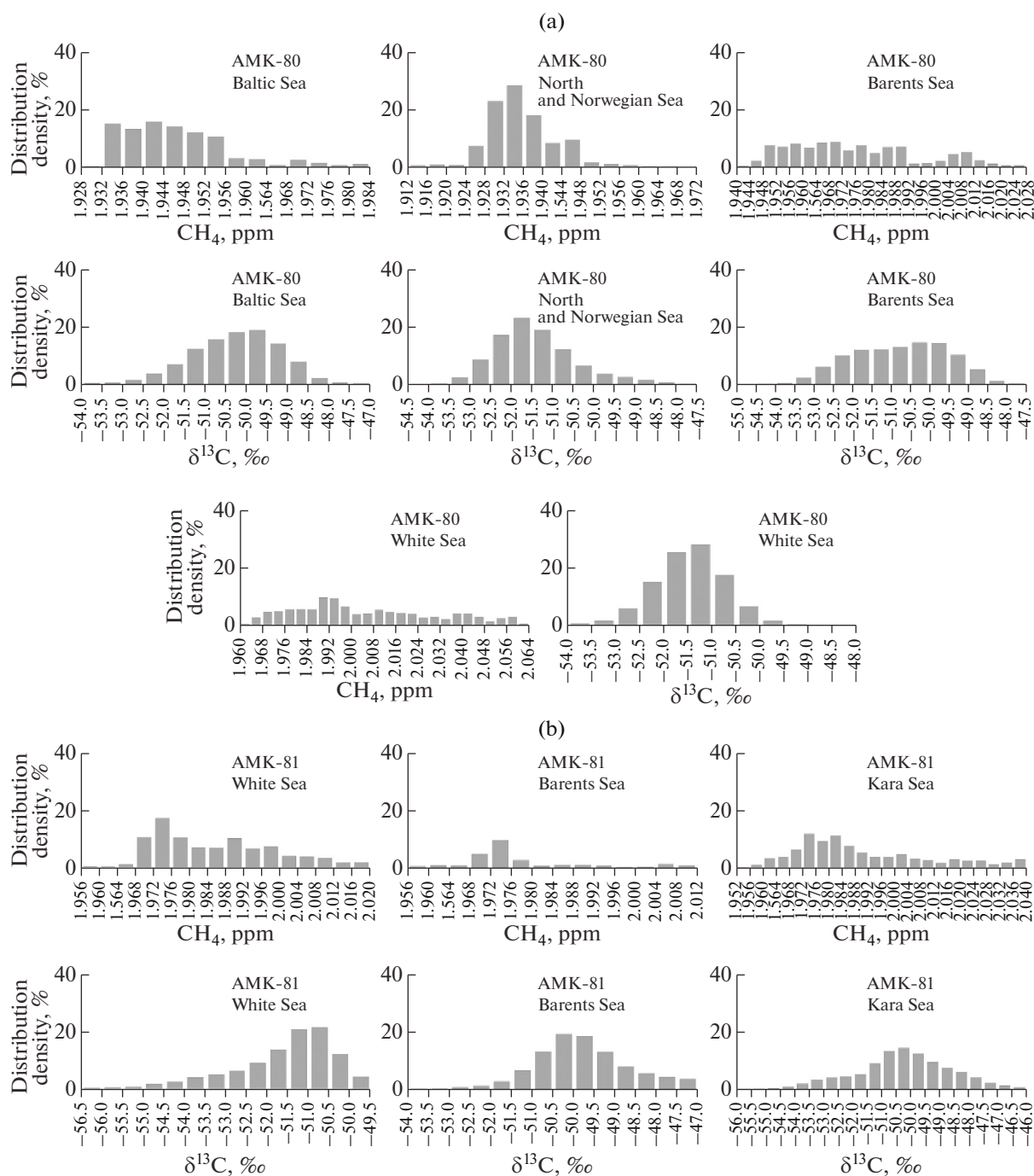


Fig. 2. CH_4 concentration distribution and $\delta^{13}\text{C}$ for various seas according to (a) AMK-80 and (b) AMK-81 expedition data (data with minute averaging were used).

from the south and west, advection was noted from the north, while the air flow did not cross the mainland. As a result, the mean methane concentration was 1.937 ppm, and in some cases it decreased to 1.913–1.920 ppm. When the air flow crossed Scandinavia, the methane concentration increased on average to ~1.970 ppm, which may be due to the active emission of CH_4 from wetlands. The maximum average value of methane concentration (2.006 ± 0.030 ppm) is

observed in the White Sea during advection of air masses from the south of the European Territory of Russia (ETR). For comparison, already a few days later, when the direction of the air flow changed to a western one (advection from Scandinavia and the Kola Peninsula) during the AMK-81 expedition in the White Sea, the average CH_4 concentration in the atmosphere dropped to 1.989 ppm. On the whole, during the AMK-81 expedition, the same regularities

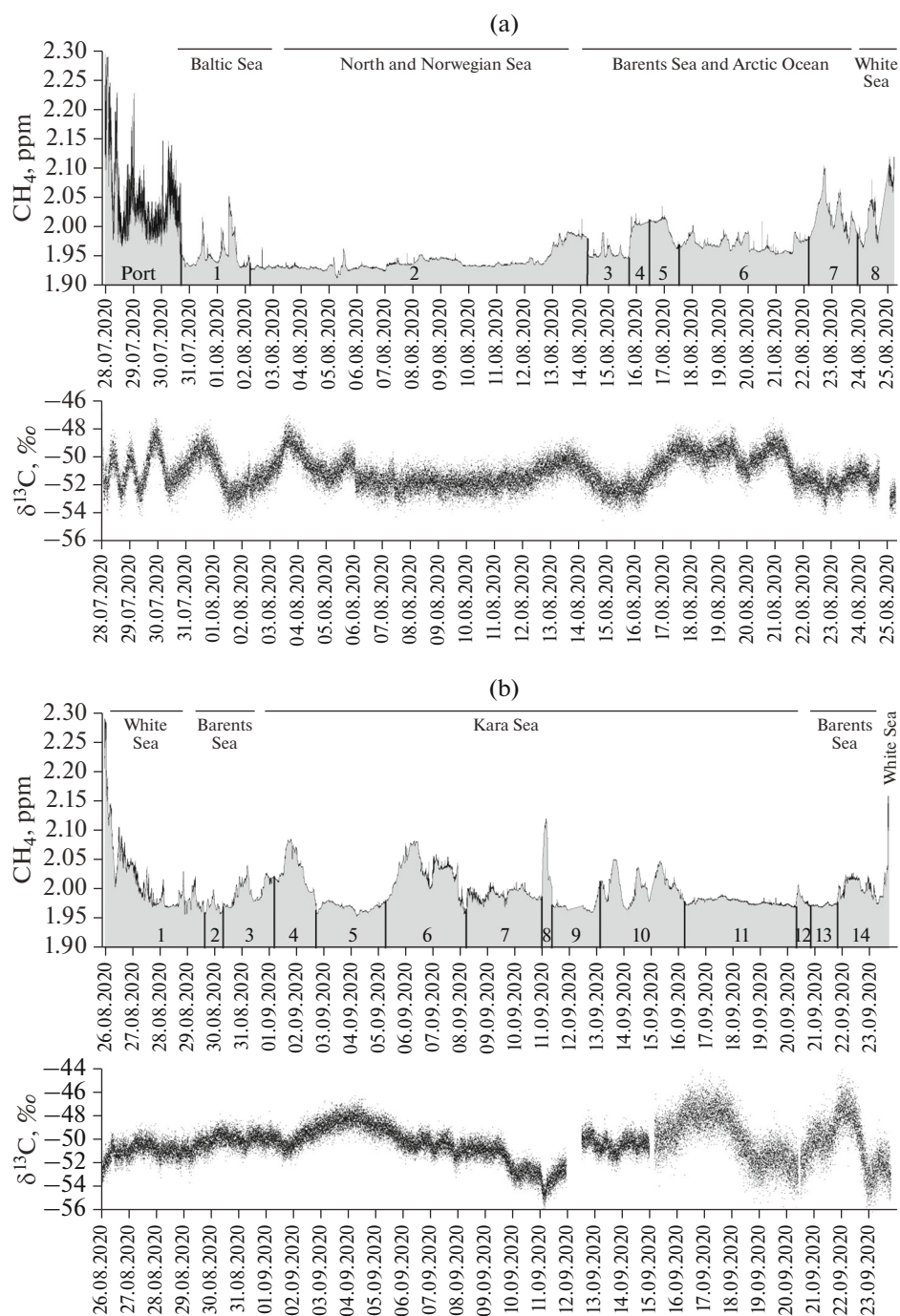


Fig. 3. CH_4 concentrations and values of $\delta^{13}\text{C}$ for (a) AMK-80 and (b) AMK-81 cruises (data with minute averaging were used). The areas where the vessel is located are above; the numbers indicate the direction of the removal of air masses. (a) AMK-80: (1) from Scandinavia; (2) from the regions of the North Atlantic (without crossing the mainland regions); (3) from the north and northeast (from the regions of the Arctic Ocean); (4) from the west, southwest, and North Atlantic; (5) from Scandinavia and the north of the European territory of Russia (ETR); (6) from the Barents Sea and the North Atlantic; (7) from northern Scandinavia, Karelia, and northwest of the ETR; and (8) from the North Atlantic and southern Scandinavia; (b) AMK-81: (1) from the North Atlantic, north of Scandinavia, the Kola Peninsula; (2) from the North Atlantic, crossing the northeast of the ETR; (3) from the North Atlantic, crossing the northwest of Siberia; (4) from the east, crossing the north of Siberia and the Laptev Sea areas; (5) from the North Atlantic, the Barents Sea; (6) from the southwest through Scandinavia and the North Atlantic; (7) from the Barents Sea, Arctic Ocean; (8) from the south of the ETR; (9) from the north of Scandinavia and the North Atlantic; (10) from the north of the ETR, north west of Siberia; (11) Arctic Ocean; (12) from the northwest of Siberia (through the areas of production and transportation of natural gas); (13) from the Kara and Barents seas and the AO; and (14) from the west with crossing of Scandinavia.

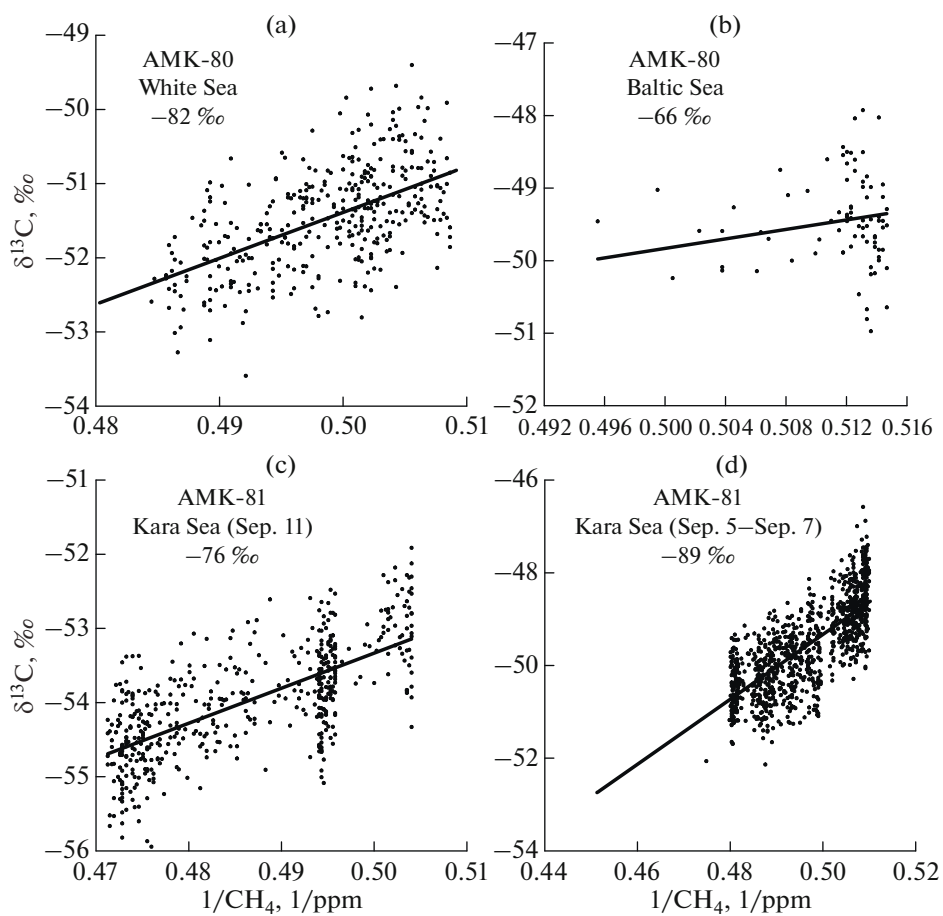


Fig. 4. Keeling plot analysis for sections of the AMK-80 and AMK-81 routes. The captions for each graph indicate the cruise number, the area where the measurements were taken (dates are given in parentheses (c, d)), and the value of coefficient B of the linear interpolation.

are noted as in the previous voyage. The minimum content of methane was noted during the removal of air masses from the west and north, as a rule, without crossing the mainland areas by the air flow. During this period, the methane concentration in the Kara Sea decreased to 1.955 ppm, which was the minimum for the entire period of the AMK-81 expedition. In the Barents Sea, when air was removed from the North Atlantic, the minimum of methane corresponded to 1.958 ppm. Elevated values of CH_4 content were observed during eastern air transport and advection from the south of the ETR, when the concentration of methane often exceeded 2 ppm (September 11, 2020).

Determination of Methane Sources

In all likelihood, the vessel on the routes of both expeditions was far from methane sources; as a result, a certain $\delta^{13}\text{C}$ was close to the average value for the Northern Hemisphere of -47.5‰ [23]. Figure 4 shows examples of Keeling plots constructed for various sections of the AMK-80 and AMK-81 cruises.

The values of $\delta^{13}\text{C}$ given in Figs. 4a and 4d are lowered and are outside the limits characteristic of wetlands. Such a picture is typical in most cases when carrying out shipboard measurements in the Arctic seas. Figure 4b shows a graph for the Baltic Sea. According to the results of the trajectory analysis, during the measurement period, there was an influx of air from Scandinavia. The $\delta^{13}\text{C}$ value corresponded to -66‰ , which is typical for methane emission from swamps. In Fig. 4c, the value of $\delta^{13}\text{C}$ corresponds to -76‰ , which is within the limits typical for wetlands, although here, as in the case of the graphs presented in Figs. 4a and 4d, it is shifted towards lighter values.

No methane emissions from the surface of the ocean appeared during the analysis of the data of the 2020 expeditions, which, apparently, is due to the fact that the studies were carried out during the formation of a rigid pycnocline that blocked vertical CH_4 fluxes in the water column and, accordingly, its flows at the ocean–atmosphere boundary. Also, there were no clear manifestations of anthropogenic methane emissions outside the zone of influence of ports.

CONCLUSIONS

The results of measurements of methane concentration and $\delta^{13}\text{C}$ in the atmosphere of the Arctic seas in summer and early autumn 2020 showed that the influence exerted on the content of CH_4 in the near layer of the atmosphere was mainly by the direction of the transfer of air masses. The minimum content of methane in the surface air was observed in the southern and western regions—over the Baltic, North, and Norwegian seas. Such a result is consistent with the results of the marine expedition cited in [24]. A minimum is also recorded in the western seas of the Arctic. The maximum concentration of methane in the near layer of the atmosphere is observed during the advection of air masses from the east from the regions of the north of Siberia and the water area of the Laptev Sea.

Analysis of the values of $\delta^{13}\text{C}$ in the studied areas suggests a mixed-type source. At the same time, over the Baltic, when advection from Scandinavia was noted, the Keeling analysis clearly showed the presence of emissions from swamps. Also, the values of $\delta^{13}\text{C}$ were noted during the AMK-81 expedition when the ship was moving along some parts of the Kara Sea. In general, due to significant air mixing over the studied marine areas, the methane isotopic signature in our summer–autumn measurements was close to the average values typical for the Northern Hemisphere: -47.5‰ [23].

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REFERENCES

1. N. V. Pankratova, I. B. Belikov, V. A. Belousov, et al., "Concentration and isotopic composition of methane, associated gases, and black carbon over Russian Arctic seas (shipborne measurements)," *Oceanology* **60**, 593–602 (2020).
2. A. I. Skorokhod, N. V. Pankratova, I. B. Belikov, et al., "Observations of atmospheric methane and its stable isotope ratio ($\delta^{13}\text{C}$) over the Russian Arctic seas from ship cruises in the summer and autumn of 2015," *Dokl. Earth Sci.* **470**, 1081–1085 (2016). <https://doi.org/10.7868/S0869565216290247>
3. P. Bousquet, P. Ciais, J. Miller, et al., "Contribution of anthropogenic and natural sources to atmospheric methane variability," *Nature* **443**, 439–443 (2006). <https://doi.org/10.1038/Nature05132>
4. E. J. Dlugokencky, E. G. Nisbet, R. Fisher, et al., "Global atmospheric methane: budget, changes and dangers," *Philos. Trans. A Math. Phys. Eng. Sci.* **369**, 2058–2072 (2011). <https://doi.org/10.1098/rsta.2010.0341>
5. R. E. Fisher, J. L. France, D. Lowry, et al., "Measurement of the ^{13}C isotopic signature of methane emissions from northern European wetlands," *Global Biogeochem. Cyc.* **31**, 605–623 (2017). <https://doi.org/10.1002/2016GB005504>
6. R. E. Fisher, S. Sriskantharajah, D. Lowry, et al., "Arctic methane sources: Isotopic evidence for atmospheric inputs," *Geophys. Res. Lett.* **38**, L21803 (2011). <https://doi.org/10.1029/2011GL049319>
7. M. Gupta, S. Tyler, and R. Cicerone, "Modeling atmospheric $^{13}\text{CH}_4$ and the causes of recent changes in atmospheric CH_4 amounts," *J. Geophys. Res.* **101**, 22923–22932 (1996). <https://doi.org/10.1029/96JD02386>
8. R. Jackson, M. Saunio, P. Bousquet, et al., "Increasing anthropogenic methane emissions arise equally from agricultural and fossil fuel sources," *Environ. Res. Lett.* **15**, JRC121045 (2020).
9. C. D. Keeling, "The concentration and isotopic abundances of atmospheric carbon dioxide in rural areas," *Geochim. Cosmochim. Acta.* **13**, 322–334 (1958).
10. S. Kirschke, P. Bousquet, P. Ciais, et al., "Three decades of global methane sources and sinks," *Nature Geosci.* **6**, 813–823 (2013).
11. L. Levin, C. Veidt, B. H. Vaughn, et al., "No inter-hemispheric delta (CH_4)-C-13 trend observed," *Nature* **486**, E3–E4 (2012). <https://doi.org/10.1038/Nature11175>
12. C. L. Myhre, B. Ferré, S. M. Platt, et al., "Extensive release of methane from Arctic seabed west of Svalbard during summer 2014 does not influence the atmosphere," *Geophys. Res. Lett.* **43**, 4624–4631 (2016).

13. N. Pankratova, A. Skorokhod, I. Belikov, et al., “Evidence of atmospheric response to methane emissions from the East Siberian Arctic shelf,” *Geography, Environment, Sustainability* **11**, 85–92 (2018).
<https://doi.org/10.24057/2071-9388-2018-11-1-85-92>
14. D. E. Pataki, J. R. Ehleringer, L. B. Flanagan, et al., “The application and interpretation of Keeling plots in terrestrial carbon cycle research,” *Global Biogeochem. Cycles* **17**, 1022 (2003).
<https://doi.org/10.1029/2001GB001850>
15. G. Rolph, A. Stein, and B. Stunder, “Real-time Environmental Applications and Display sYstem: READY,” *Environmental Modelling & Software* **95**, 210–228 (2017).
<https://doi.org/10.1016/j.envsoft.2017.06.025>
16. C. J. Sapart, N. Shakhova, I. Semiletov, et al., “The origin of methane in the East Siberian Arctic Shelf unraveled with triple isotope analysis,” *Biogeosciences*. **14**, 2283–2292 (2017).
<https://doi.org/10.5194/bg-14-2283-2017>
17. M. Saunio, P. Bousquet, B. Poulter, et al., “The global methane budget 2000–2012,” *Earth. System Sci. Data* **8**, 697–751 (2016).
<https://doi.org/doi.10.5194/essd-8-697-2016>.
18. M. Saunio, A. R. Stavert, B. Poulter, et al., “The global methane budget 2000–2017,” *Earth System Science Data* **12**, 1561–1623 (2020).
<https://doi.org/doi.10.5194/essd-12-1561-2020>.
19. N. N. Shakhova, I. I. Semiletov, A. A. Salyuk, et al., “Extensive methane venting to the atmosphere from sediments of the East Siberian Arctic Shelf,” *Science* **327**, 1246–1250 (2010).
20. N. Shakhova, I. Semiletov, I. Leifer, et al., “Ebullition and storm-induced methane release from the East Siberian Arctic Shelf,” *Nature Geosci.* **7**, 64–70 (2014).
<https://doi.org/doi10.1038/ngeo2007>
21. O. A. Sherwood, S. Schwietzke, V. A. Arling, et al., “Global inventory of gas geochemistry data from fossil fuel, microbial and burning sources, version 2017,” *Earth Syst. Sci. Data* **9**, 639–656 (2017).
<https://doi.org/10.5194/essd-9-639-2017>
22. A. F. Stein, R. R. Draxler, G. D. Rolph, et al., “NOAA’s HYSPLIT atmospheric transport and dispersion modeling system,” *Bull. Amer. Meteor. Soc.* **96**, 2059–2077 (2015).
<https://doi.org/10.1175/BAMS-D-14-00110.1>
23. N. J. Warwick, M. L. Cain, R. Fisher, et al., “Using $\delta^{13}\text{C}-\text{CH}_4$ and $\delta\text{D}-\text{CH}_4$ to constrain Arctic methane emissions,” *Atmos. Chem. Phys.* **16**, 14891–14908 (2016).
<https://doi.org/10.5194/acp-16-14891-2016>
24. J. Yu, Z. Xie, L. Sun, et al., “ $\delta^{13}\text{C}-\text{CH}_4$ reveals CH_4 variations over oceans from mid-latitudes to the Arctic,” *Sci. Rep.* **5**, 13760 (2015).
<https://doi.org/10.1038/srep13760>
25. L. Yurganov, D. Carroll, and H. Zhang, “Ocean stratification and sea-ice cover in Barents and Kara seas modulate sea-air methane flux: satellite evidence,” *Earth and Space Science* 2020.
<https://doi.org/10.1002/essoar.10504047.1>