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Air Composition over the Russian Arctic: 2—Carbon Dioxide

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Abstract—We analyze the spatial distribution of carbon dioxide over the seas of the Russian Arctic based on the results of the comprehensive experiment conducted in September 2020. It turned out that during the experiment, the concentration of CO_2 increased from west to east. The minimum of 396 ppm was over the Barents Sea, and the maximum of 4106 ppm was over the Chukchi Sea. The difference between the concentrations at an altitude of 200 m and in the free troposphere attained 156 ppm over the Barents Sea and decreased to 56 ppm over the Laptev Sea. Over the eastern seas, the difference became generally positive, which was associated with the air transfer from Alaska. Above the waters of most seas, the distribution of carbon dioxide was horizontally heterogeneous, which showed the regional features of its assimilation by the ocean and transfer from the continent.

Keywords: Arctic, atmosphere, air, vertical distribution, carbon dioxide, greenhouse gases, transport, impurities, composition

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INTRODUCTION

The current global climate change, which, according to the conclusion of the Intergovernmental Panel on Climate Change (IPCC), is due to an increase in the concentrations of greenhouse gases in the atmosphere, leads to the need in monitoring of their content around the globe [1], especially in regions where warming is faster, for example, in the Arctic, where the rate of increase in the air temperature is 2–3 times higher than in other regions [2, 3]. Among greenhouse gases, the largest contribution to additional air heating, after water vapor, is made by carbon dioxide, more precisely, its anthropogenic additive [1].

As early as 1960, Ch.D. Keeling drew attention to the possible role of CO_2 in global warming. He started systematic measurements of carbon dioxide concentrations at the Mauna Loa Observatory (United States) and in Antarctica. In his work [4], he showed that the rate of increase in the concentration of CO_2 at the South Pole corresponds to the amount of fossil fuels burned worldwide per year. Recognition came to the author much later (in the early 1970s), after which the CO_2 long-term variability curve was named after him (the Keeling curve). From that time, active studies of the content of carbon dioxide in the atmosphere and its exchange with the biosphere and the underlying surface began.

Despite the studies, the generalization of which is available in [5, 6], this problem is still far from being solved. The main absorber of CO_2 on the land surface is vegetation, which, according to estimates [7, 8], compensates for about a third of its anthropogenic emissions. The estimates of CO_2 sinks are very different [9]. It is also unclear how long the land biosphere will be capable of coping with the growing excess of CO_2 [10].

A significant part of the excessive carbon dioxide from the atmosphere is absorbed by the ocean [11, 12]. However, the issues of the land–ocean interaction during sink processes remain unclear, in particular, how much CO_2 enters the ocean, which is carbon storage, from land with river waters [13, 14]. The estimates CO_2 runoff into the ocean by different authors [15–17] also markedly differ, for example, by a factor of two [18].

Considering the Arctic region, one should also mention the exchange of CO_2 between the atmosphere and the surface of the many thermokarst lakes. The fact of this exchange has been established [19, 20], but there are no reliable estimates of the power of CO_2 fluxes yet.

Studies of carbon dioxide in the Arctic are most often carried out in the surface air layer along the coast of the Arctic Ocean [21–25] or in the near-water layer from vessels [26, 27]. The data acquired in such studies on the dynamics of CO_2 in coastal areas allow estimating the power of sources and sinks of impurities controlled. In the Russian Arctic, studies were carried out in the surface air layer [28–30], in the near-water layer at drifting stations [31], and from research vessels [32, 33].

Aircraft laboratories are widely used worldwide to study the vertical distribution of CO_2 [34–37]. In Russia, this type of studies was rare in recent decades. We carried out only two small flight campaigns in the Arctic Russia within the international projects [38, 39].

This work continues the series of papers with the detailed analysis of the features of the air composition over Arctic seas on the basis of the experiment on sounding the atmosphere and water surface over all seas in the Russian Arctic with the TU-134 *Optik* aircraft laboratory in September 2020.

Work [40] gives a complete description of the experiment, including a description of the equipment used, sounding schemes, synoptic conditions, and average concentrations of gases and aerosol, we will not repeat them in this article. We only point out that the concentrations of CO_2 was measured with a G2301-*m* (Picarro Inc., United States) CRDS gas analyzer. This allows detecting the CO_2 concentration in the range 0–1000 ppm with an error $< \pm 0.2$ ppm at a frequency of 1 Hz. The gas analyzer was calibrated before the start of the aircraft campaign using calibration gas mixtures of CO_2 and CH_4 (CGM) manufactured by DEUSTE Steininger GmbH (Germany), the exact concentrations of which were determined at the Laboratory of Climate and Environmental Sciences (LSCE UMR 8212, France) using primary standards on the WMO-CO_2-X2007 and WMO-CH_4-X2004A scales. After the campaign, the G2301-*m* gas analyzer was retested against the aforementioned CGMs for performance drift over this period; no drift was found. The CO_2 profiles were retrieved from the data of every second measurement using a moving average over 15 points and subsequent interpolation with an altitude step of 10 m.

As was shown in [40], the differences in CO_2 over different seas are small above 5000 m; therefore, we do not consider the range above this altitude.

1. VERTICAL DISTRIBUTION

1.1. Barents Sea and Coastal Areas

Sounding of the atmosphere over the Barents Sea was carried out from 12:58 to 15:38 on September 4, 2020. Hereinafter, the time is given in GMT because the experiment was carried out in nine time zones in several large administrative districts, where the local and astronomical time differ.

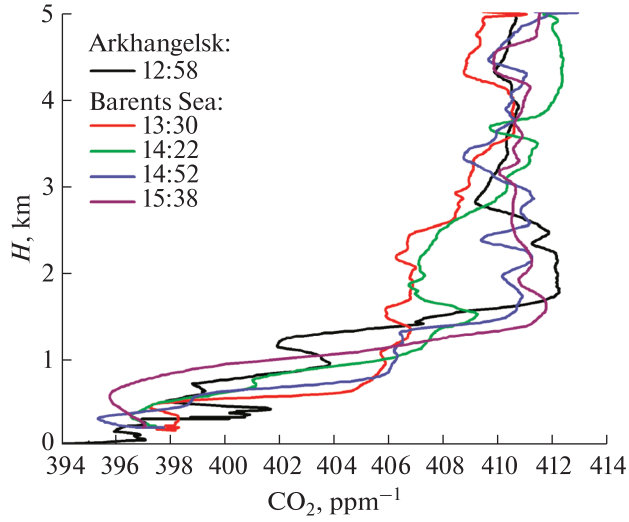

Fig. 1. Vertical carbon dioxide distribution over the Barents Sea on September 4, 2020.

The measurement results over the Barents Sea are shown in Fig. 1. The vertical CO_2 profile measured over the coastal area during departure from Arkhangelsk airport is also shown for comparison.

It can be seen that the CO_2 concentration, in contrast to methane [41], significantly increases from the near-water layer to the upper boundary of the ABL (H_{ABL}). The value of H_{ABL} has already been given in [41] and amounted to 1.9 ± 0.5 km on that day. Depending on the region of the Barents Sea water area, the difference in the concentration (ΔCO_2) can range from −10 to −16 ppm. Above the boundary layer, the CO_2 concentration either slowly increased or insignificantly changed. At a level of 3.5–3.8 km, the CO_2 concentrations are similar over all regions of the Barents Sea. Above this level, the profiles again diverge. Apparently, the processes of formation of the carbon dioxide vertical distribution change at this altitude. In the lower troposphere, including the ABL, the main processes are its absorption by the ocean and its transfer from adjacent territories. Above 4 km, the main role is played by the general atmospheric circulation. The resulting vertical distribution of CO_2 shows its absorption by both the ocean and continental ecosystems (profile for the Arkhangelsk region).

In the first part of this work [41], the analysis of methane transfer to the Barents Sea water area revealed that, air was supplied from the southwest direction, i.e., from the land to the sea, during the measurements. Moreover, the concentration of methane over land was higher than in the near-water air layer [40, 41]. The comparison between CO_2 concen-

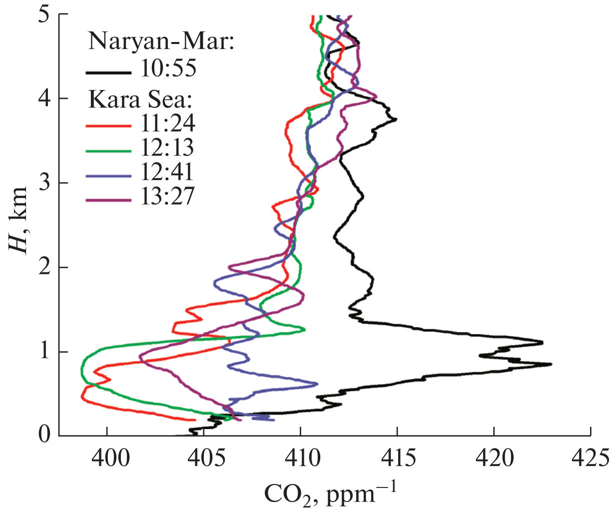

Fig. 2. Vertical carbon dioxide distribution over the Kara Sea on September 6, 2020.

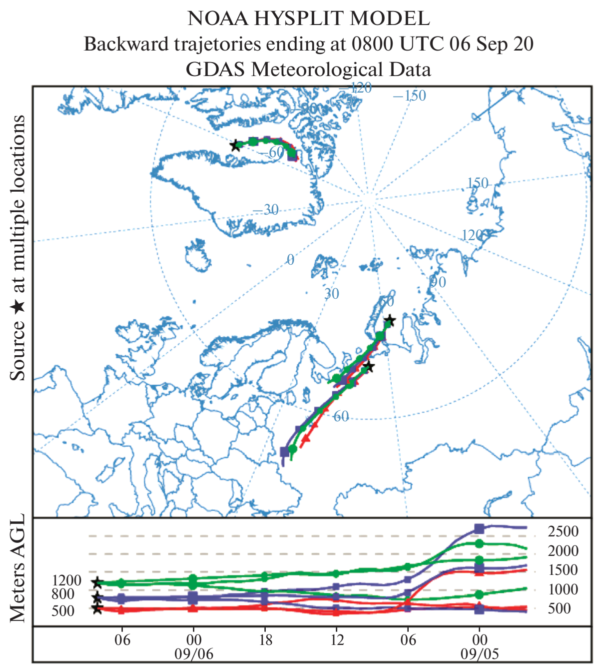

Fig. 3. Back air mass trajectories to the Naryan-Mar region and to the Kara Sea on September 6, 2020.

trations over the land and sea shows (Fig. 1) that there is no such excess for carbon dioxide, since September 2020 turned out to be abnormally warm for this region and photosynthesis was still effective. Figure 1 shows that CO_2 concentrations were approximately equal at all altitudes considered.

1.2. Kara Sea and Coastal Areas

Atmospheric sounding over the Kara Sea was carried out on September 6, 2020. All parameters of this part of the experiment were described in [40] (the same applies to other seas). Figure 2 shows CO_2 profiles over the sea and the coastal territory (Naryan-Mar).

Three of the four profiles show the same trend as over the Barents Sea: a significant negative difference in the concentrations in the ABL and a slow increase above H_{ABL} up to a level of 3 km; ΔCO_2 over the Kara Sea changed from −6 to −11 ppm. The fourth profile (12:41) in the ABL behaves near neutrally. This might well reflect the transfer of an additional amount of CO_2 from the mainland. This assumption is also supported by the CO_2 concentration profile measured over the coastal area. The comparison shows that the CO_2 concentration in the ABL was 18 ppm higher over land than over the sea.

To analyze the possible transfer in the ABL, let us consider the back air mass trajectories for Naryan-Mar and the farthest point of the flight (Fig. 3).

The air was transported to the Kara Sea water area from the Naryan-Mar region, where it, in turn, came from the western part of the European Russia, where there are many industrial facilities which are potential sources of CO_2. Thus, the increase in the carbon dioxide concentration over the Kara Sea in the ABL is probably associated with the anthropogenic factor or with a decrease in CO_2 uptake by vegetation on land due to a decrease in vegetation activity during the experiment.

It is important to dwell on one more aspect. In the first part of the work [41], we found that the Kara Sea was an additional source of methane under the southwestern transport and the high content of methane over the coast during the measurement period. However, the concentration of CO_2 over the ocean was much lower than over land. This indicates that the ocean absorbs a significant part of CO_2.

1.3. Laptev Sea and Coastal Areas

The air composition over the Laptev Sea was measured on September 9, 2020. The results of aircraft sounding are shown in Fig. 4.

It can be seen that the difference between the CO_2 concentrations in the ABL and in the free troposphere decreases from west to east. Over the Laptev Sea, ΔCO_2 varied from −3 to −6 ppm depending on the sounding region. Hence, either the water area of this sea less absorbed carbon dioxide, or air enriched with carbon dioxide came here. The second option is supported by the concentrations measured over the Tiksi airport.

To verify this conclusion, let us consider Fig. 5, which shows the back trajectories for the Tiksi settlement and the Laptev Sea water area.

The air arrived to different areas of the Laptev Sea and to Tiksi along different trajectories. In Tiksi and to the south of the Laptev Sea, air was transported from

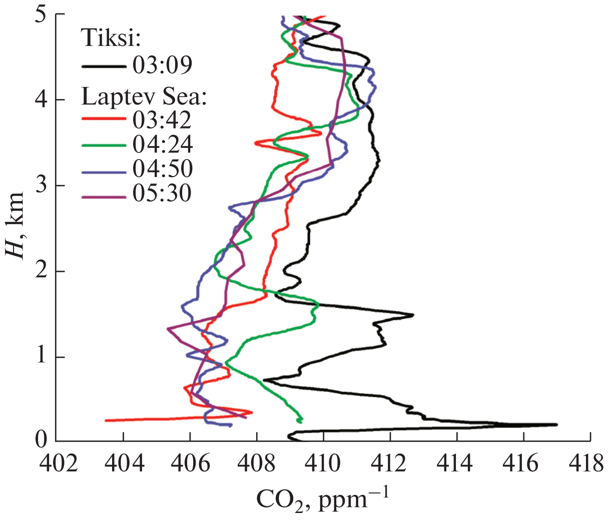

Fig. 4. Vertical carbon dioxide distribution over the Laptev Sea on September 9, 2020.

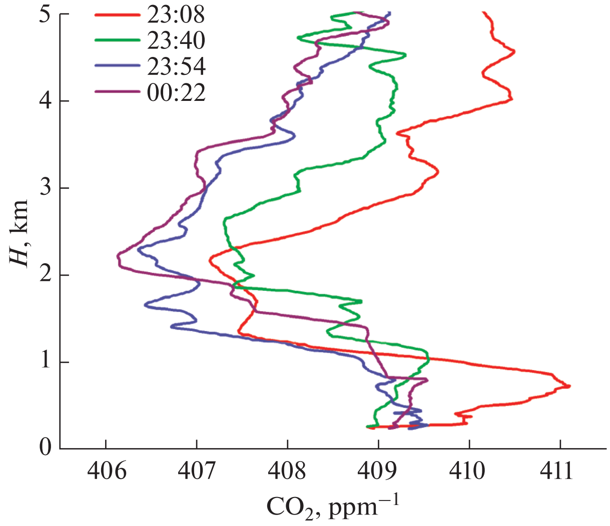

Fig. 6. Vertical carbon dioxide distribution over the East Siberian Sea on September 15 and 16, 2020.

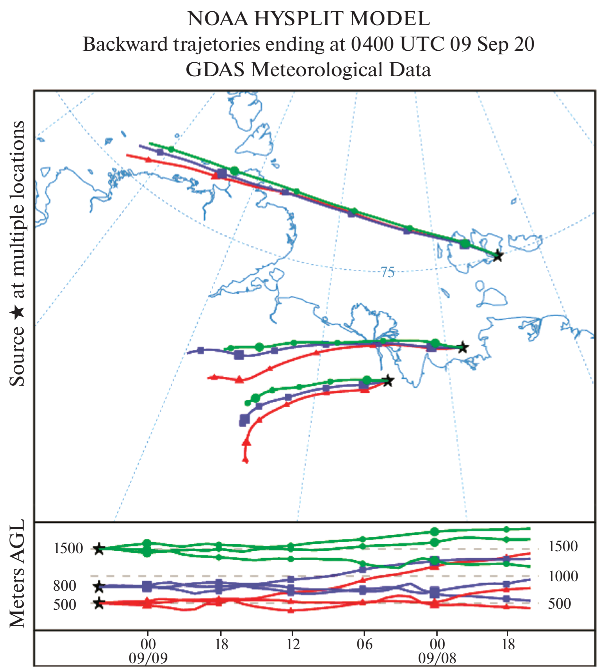

Fig. 5. Back air mass trajectories to the Tiksi region and to the Laptev Sea water area on September 9, 2020.

the continent, and to the northern part of the sea, from the Arctic Ocean. The lower CO_2 concentration over the water area as compared to land indicates the absorption of part of CO_2 by the ocean.

1.4. East Siberian Sea

As was already mentioned in [40, 41] sounding over the East Siberian and Chukchi Seas was carried out from the Anadyr airport, which is located quite far from the Arctic Ocean. Therefore, the comparison of data over the water areas of the seas with the coastal territory is incorrect. Taking into account the 12-hour zone difference with Greenwich Mean Time, the study was carried out on September 15 and 16, 2020. The measurement results are shown in Fig. 6. One can see that over this sea the distribution of carbon dioxide is essentially different from all the seas considered above. The CO_2 concentration was minimal near the upper boundary of the ABL ($H_{ABL} = 1.8 \pm 0.5$ km) [41]. The difference between the concentrations in the near-water air layer and in the free troposphere over the East Siberian Sea was positive and varied from +2 to +3.5 ppm. At the same time, we cannot state that the ocean does not absorb CO_2. If we consider the lower part of the profiles, we can see that the concentration decreases with altitude; ΔCO_2 becomes negative in the lower part and it fluctuates from −0.5 to 2.5 ppm over the water area.

The back trajectories (Fig. 7) show that in the lower level, where the CO_2 concentrations are low, the air came from the water area of the western sector of the Arctic Ocean. In the ABL with a high CO_2 content, the air was transported from the territory of Alaska. Carbon dioxide rose from the ABL to a level of 2200 m, where the CO_2 concentration were minimal. The difference in the concentrations could be due to the difference in the power of the sources in different areas of Alaska, from where air masses arrived.

1.5. Chukchi Sea

Due to the difficult synoptic conditions over the East Siberian Sea, aircraft sounding of the air composition on September 14 and 15, 2020 was carried out

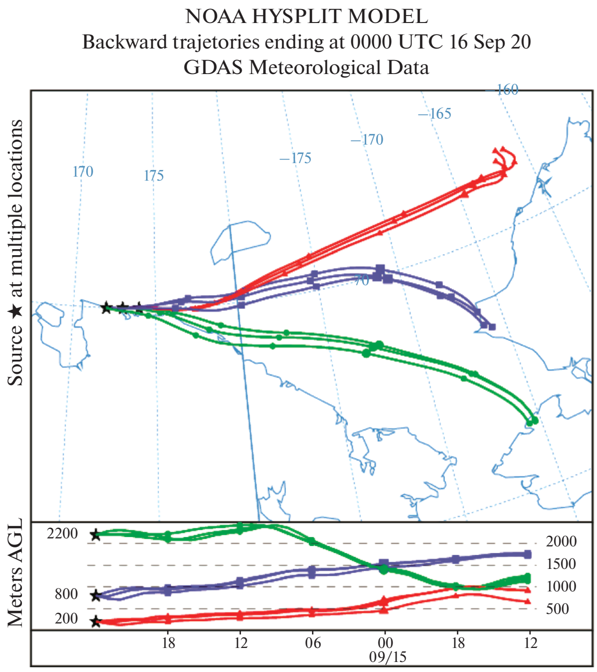

Fig. 7. Back air mass trajectories to the East Siberian Sea water area on September 15 and 16, 2020.

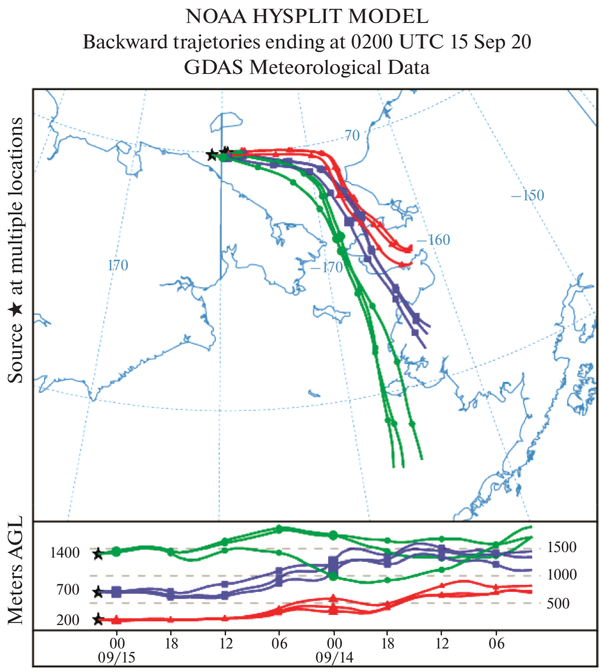

Fig. 9. Back air mass trajectories to the Chukchi Sea water area on September 14 and 15, 2020.

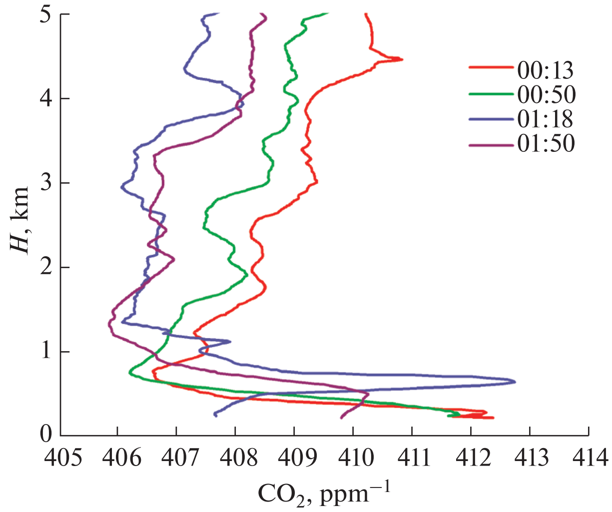

Fig. 8. Vertical carbon dioxide distribution over the Chukchi Sea on September 15, 2020.

over the Chukchi Sea. The CO_2 concentration measurement results are shown in Fig. 8. The vertical distribution of CO_2 over the Chukchi Sea is obviously similar to that observed over the East Siberian. However, two differences can be noted.

The first is that the minimal concentrations dropped to a level of 0.8–1.2 km. This is not surprising since, according to [41], H_{PSA} was the lowest here (1.3 ± 0.3 km). This change in the stratification of the atmosphere resulted in a slight increase in ΔCO_2 between the ABL and the free troposphere slightly, it varied from +4 to +6 ppm. The negative difference in CO_2 also increased in the bottom layer to −5.1 ppm.

The second difference is the higher concentrations of CO_2 in the ABL.

Figure 9 shows that air masses arrived to both the East Siberian and Chukchi Seas from the southern regions of Alaska.

The anomalous distribution of methane, described in [41], and carbon dioxide led to a need in additional sounding of the air composition over the Bering Sea, which was not included in the original schedule of the experiment.

1.6. Bering Sea and Coastal Areas

Atmospheric sounding over the Bering Sea was carried out on September 16, 2020, immediately after the East Siberian Sea. The purpose of this additional part of the experiment was to analyze the discovered features of the distribution of carbon dioxide and methane over the Chukchi and East Siberian Seas.

The measurement data are shown in Fig. 10. It can be seen that the vertical distribution of CO_2 over the Bering Sea generally repeats the profiles over the East Siberian and Chukchi Seas. Quantitatively, it occupies an intermediate position. Thus, ΔCO_2 between the ABL and the free troposphere was in the range from +1 to +4 ppm, and in the lower, near-water, part of the ABL, from −0.5 to −2.8 ppm. The ABL height, according to [41], was 1.7 ± 0.2 km. This is also an intermediate value.

Figure 11 shows how the vertical distribution of CO_2 was formed: the air was transported to the sounding areas from the Alaska or along its coast.

Thus, the features of the vertical distribution of carbon dioxide over the eastern sector of the Russian Arctic are regional in nature and are due to air transport from Alaska. This is also confirmed by the vertical profile of CO_2 measured over the coastal region (Anadyr). It can be seen that the carbon dioxide concentration over the Russian Arctic was significantly lower than over the American Arctic.

2. HORIZONTAL INHOMOGENEITIES OF CARBON DIOXIDE DISTRIBUTION

The horizontal inhomogeneities in CO_2 distribution over the waters of the Arctic seas are analyzed on the basis of data from flights performed at an altitude of 200 m over different regions. Recall that during such flights, the concentrations of trace atmospheric gases were measured with a frequency of 1 Hz. The measurement areas are shown in Fig. 15 in [41]. We assumed that the inhomogeneity of CO_2 uptake by the ocean surface should be pronounced at this altitude. The measurement results are shown in Fig. 12 for the comparison of changes in the CO_2 concentrations over different seas.

One can see quite homogeneous distributions of carbon dioxide concentrations over the Bering and East Siberian Seas. Over the Kara, Laptev, and Chukchi Seas, it abruptly changed when moving from one region of a water area to another. The Barents Sea is characterized by a concentration gradient with one burst 14 km long. Most likely that this is a plume from a high-power continental source.

The variability of the CO_2 concentration can be estimated from Table 1. ΔCO_2 is significantly different over different seas, which is obviously due to the heterogeneity of the CO_2 sink to the ocean and its supply from the continent.

3. DISCUSSION

During the aircraft sounding, two types of carbon dioxide vertical profiles derived retrieved: typical for marine regions, where the CO_2 concentration of decreased with altitude due to absorption by the ocean [42–44], and atypical, where the concentration in the ABL was higher than in the near-water layer and free troposphere. The atypical profile can be associated both with the seasonal ratio of the power of sources and sinks [45, 46] and with regional features.

As is shown above during the analysis of back trajectories, atypical profiles were observed over the seas, where air masses were transported from Alaska.

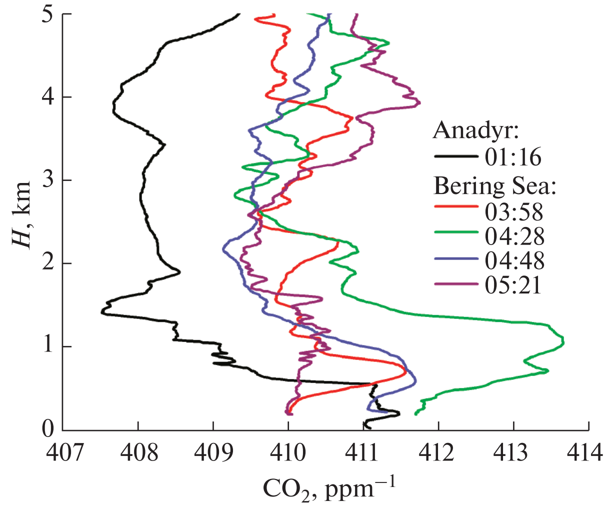

Fig. 10. Vertical carbon dioxide distribution over the Bering Sea on September 16, 2020.

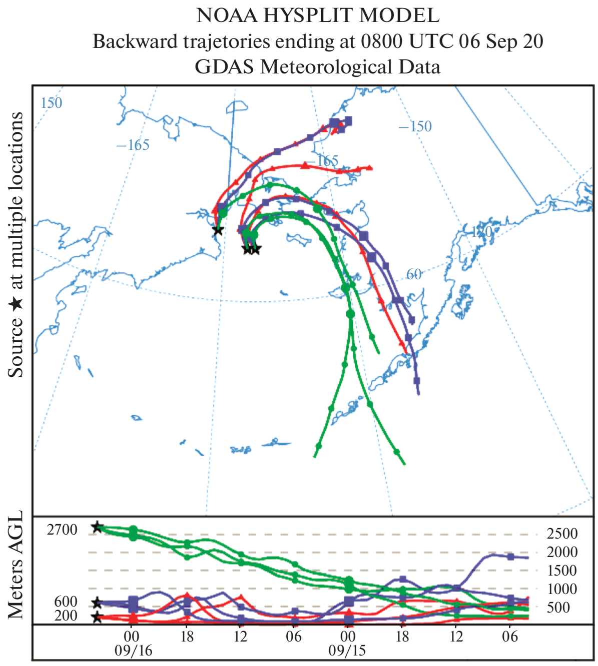

Fig. 11. Back air mass trajectories to the Bering Sea water area on September 16, 2020.

Table 1. Range of change in carbon dioxide concentration at a height of 200 m in horizontal sections

Sea	ΔCO_2, ppm
Barents	6
Kara	9
Laptev	11
East Siberian	3
Chukchi	6
Bering	2

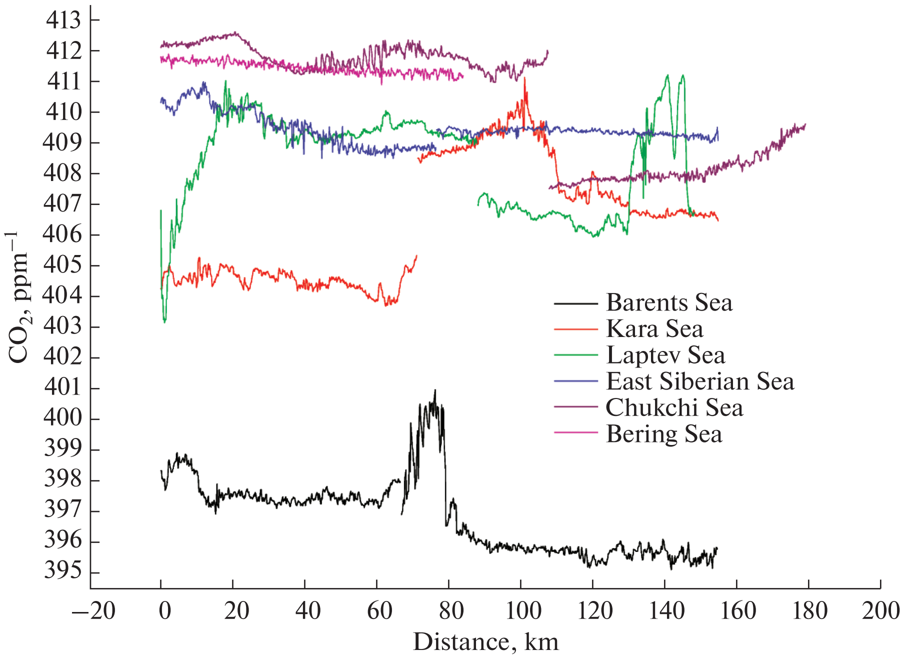

Fig. 12. Carbon dioxide concentration at an altitude of 200 m above the seas of the Russian Arctic and Pacific Ocean.

Therefore, this region can be a source of high CO_2 concentrations. This is confirmed by the results of works [47–49]. According to [47], the uptake of CO_2 at ocean shelf in the Western Hemisphere is noticeably lower than in the neighboring Chukchi Sea. The study of the inland regions of Alaska, where there were fires [48], revealed that those regions emitted the third part of all carbon dioxide in Alaska. In [49], CO_2 concentrations up to 425 ppm were measured from aircraft as early as in 2017. Taking into account the long-term global trend towards an increase in the CO_2 concentrations by about 2.4 ppm per year [50], we can assume that it could be higher than 430 ppm during our experiment in 2020. Though it could be diluted while being transported, it could lead to such high values over the East Siberian, Chukchi, and Bering Seas.

Let us compare the carbon dioxide concentrations over the seas under study (Table 2).

According to Table 2, the average CO_2 concentration increased from west to east in September 4–17, 2020: it was 396.9 ppm over the Barents Sea and 411.5 ppm over the Bering Sea, i.e., 15 ppm higher. This change in the carbon dioxide concentration in the Arctic atmosphere is directly opposite to that for methane [41]. Moreover, it does not correspond to the arrangement of anthropogenic sources of CO_2, located mainly in Europe, and sites of sink—boreal forests in the Russian Asia.

There was also a tendency towards a decrease in the power of the CO_2 sink from the atmosphere to the ocean from west to east. Thus, ΔCO_2 = 13.5 ppm over the Barents Sea, 9.3 ppm over the Kara Sea, and

Table 2. Average characteristics of carbon dioxide distribution and its variability

Parameter	Sea					
	Barents	Kara	Laptev	East Siberian	Chukchi	Bering
CO_2 concentration at an altitude of 200 m, ppm	396.9 ± 1.2	406.4 ± 1.9	408.4 ± 1.6	409.4 ± 0.5	410.4 ± 1.9	411.5 ± 0.2
ΔCO_2 between 200 m and the free troposphere, ppm	−13.5 ± 2.6	−9.3 ± 2.4	−4.8 ± 1.3	+3.1 ± 0.9	+5.3 ± 1.0	+3.2 ± 1.5
ΔCO_2 between the maximum in the ABL and in the near-water layer, ppm^{-1}	–	–	–	−1.1 ± 1.0	−1.9 ± 2.1	−1.4 ± 1.3
Air temperature at an altitude of 200 m, °C	14.7 ± 1.9	12.1 ± 0.6	7.2 ± 1.7	3.6 ± 0.4	5.0 ± 0.4	6.9 ± 0.3
Sea temperature (coastal stations), °C	11	8	7	5	4	8

Table 3. Maximum and minimum carbon dioxide concentrations over the seas and coastal regions of the Arctic

Sea	CO_2, ppm		Shore	CO_2, ppm	
	max	min		max	min
Barents	401	395	Arkhangelsk	420	394
Kara	412	403	Naryan-Mar	420	401
Laptev	414	403	Tiksi	414	405
Chukchi	413	407	Anadyr	415	404
Bering	412	409			
East Siberian	411	408			

4.8 ppm over the Laptev Sea. It became positive to the east due to the air mass transport from Alaska, although the ocean continued to absorb carbon dioxide, as evidenced by the difference between the concentration maximum in the ABL and in the near-water layer. It was −1.1, −1.9, and −1.4 ppm for the East Siberian, Chukchi, and Bering Seas, respectively.

Let us consider one more aspect. If the ocean assimilates carbon dioxide by dissolving, then, on the one hand, there should be a negative dependence of its absorption on the water temperature [51]. On the other hand, plankton in the upper ocean layer feed on CO_2. The metabolic rate of many of its species positively and nonlinearly depends on the water temperature [52]. A natural question arises: which of these processes predominates? Table 2 shows that the higher the water temperature, the lower the CO_2 concentration in the air, therefore, the more CO_2 is absorbed by the ocean. Thus, the process of carbon dioxide uptake by microorganisms in the upper ocean layer is predominant for the seas of the Russian Arctic.

Considering the change in methane concentration along the Russian Arctic coast in [41], we made an assessment of the possible effect of CH_4 emissions on the coast when they are transferred to the water area. Following this approach, we compiled Table 3 for carbon dioxide. Here, it is important to take into account that in addition to anthropogenic emissions, a certain amount of CO_2 is added by terrestrial vegetation, which respires at night [53].

Table 3 data show the spread of CO_2 concentrations on the coast to be higher than over the water areas. Hence, when being transferred from land to a sea, both an increase and a decrease in the carbon dioxide concentration are possible over the sea. The comparison of the minimal concentrations over the coast and the seas allows us to conclude that land vegetation can uptake more than the ocean.

CONCLUSIONS

The results of the aircraft experiment carried out for a quite short time made it possible to compare the concentrations of carbon dioxide over all the seas of the Russian the Arctic. We have ascertained that the CO_2 content increased from west to east in September 2020. It was the minimal over the Barents Sea and maximal over the Chukchi and Bering Seas. The last flight was a control flight for the eastern region.

As the concentration increased from west to east, its difference between the near-water layer and the free troposphere also decreased, which reflected the degree of absorption of carbon dioxide by the ocean. Over the East Siberian, Chukchi, and Bering Seas, that difference became positive, which was caused by the air mass transfer from the territory of Alaska or from the water areas of the adjacent seas.

The distributions of CO_2 over the water areas of most seas were heterogeneous, most likely because of the difference in its absorption by the ocean and features of its transfer from the continent.

The CO_2 absorption by the ocean depended on the water temperature during the experiment. That relationship was positive, which showed the predominance of CO_2 uptake by plankton over its dissolution in water.

Since this kind of experiment is the first in the Russian history of studying the composition of the atmosphere in the Arctic, it is natural that the results should be considered as preliminary. It is necessary to carry out such sounding in other seasons, when some sources and sinks can be "turned off," for example, in winter when the ocean is frozen and there is no vegetation activity.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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