ATMOSPHERIC RADIATION, OPTICAL WEATHER, AND CLIMATE

# Vertical Distribution of Greenhouse Gases above Western Siberia by the Long-Term Measurement Data

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**Abstract**—By the results of long-term (1997–2007) airborne sounding, the vertical distribution of three greenhouse gases such as  $CO_2$ ,  $CH_4$ , and  $N_2O$  above the south of Western Siberia is investigated. The average monthly profiles of the distribution of these components in height and the long-term change in gas concentration at different heights are presented. The climatic characteristics of the vertical distribution of these gases are determined.

Key words: greenhouse gas, carbon dioxide, methane, nitrous oxide, vertical distribution of gases.

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#### INTRODUCTION

The global change in climate and environment is one of the urgent problems today, since the consequences of such changes may not only adversely affect the habitation area, but also cause natural disasters. The growing concentration of greenhouse gases recorded in many regions is considered crucial to this problem, leading to an increase in the average global temperature due to the additional absorption of infrared radiation. As a rule, a prognosis of the climate change is based on the calculations by regional models or by models of general atmospheric circulation, where the measured concentration of greenhouse gases and the tendencies of their change serve as initial data [1–3].

Such calculations show that the contribution of certain regions to the global balance of greenhouse gases is rather different due to their physical-geographical features. For example, Russia has vast undisturbed or weakly disturbed ecosystems that occupy 65% of its territory. This country is a "blind spot" in scientific research, as there are almost no measurement data of greenhouse gases on its territory. It is not fortuitous that the completed estimates have an opposite character in the absence of such data. Thus, by the calculations [4], the Siberian forests release about 0.035 Pg C/yr. The authors [5, 6] proved, on the contrary, that the Siberian forests absorb  $0.42 \pm 0.07$  Pg C/yr. We should mention that the last value is comparable with carbon absorption by all of the Northern Hemisphere according to data [7]. Consequently, in order to eliminate such uncertainty, we should correctly measure the concentration of greenhouse gases above little-studied regions.

The purpose of this paper is to investigate the vertical distribution of greenhouse gases above Western Siberia, where these components were not studied systematically.

#### METHODS AND EQUIPMENT

To perform measurements, the AN-30 Optic-E aircraft laboratory was used. It is described in detail in [8]. During the flight, air was taken in glass bottles at heights of 0.5, 1, 1.5, 2.0, 3.0, 4.0, 5.5, and 7 km. To fill in the bottles with the outboard air, a GAST DOA-P108 oil-free diaphragm pump maintaining a pressure of 2 atm was installed. Then, the air from the bottles was analyzed by a chromatographic method in the laboratory of the National Institute for Environmental Studies. The measurement ranges and errors of concentration were:  $(340-450 \pm 0.03)$  ppm for CO<sub>2</sub>,  $(1500-2500 \pm 1.7)$  ppb for CH<sub>4</sub>, and  $(250-450 \pm 0.3)$  ppb for N<sub>2</sub>O.

Air sampling and the measurement of the gas composition of the air were performed after the 20th day of each month in clear weather starting from July 1997 until the present time. Sounding was carried out at the same place in the southwest of Novosibirsk, as to not allow the influence of the city. The route of the flights went above the pine forest along the right bank of the Ob reservoir near Zyryanka and Ordynskoye settlements with initial coordinates 54°35′ N, 82°40′ E. The flight map is shown in [9].

By the present time, the continuous measurements have been collected for more than a ten-year period. Even taking into account unsuccessful flights, the series analyzed below exceeds 120 vertical profiles of  $CO_2$ ,  $CH_4$ , and  $N_2O$  distribution.

#### **RESULTS AND DISCUSSION**

First, we consider Fig. 1, which presents concentrations of  $CO_2$ ,  $CH_4$ , and  $N_2O$  by the data of all measurements at all controlled heights. It is seen that two greenhouse gases,  $CO_2$  and  $N_2O$ , have a marked tendency to concentration growth during the whole measurement period at each height, despite the modulation in the annual behavior.  $CH_4$  does not show such a tendency during the same period. At first sight, its concentration behavior seems chaotic.

The amplitudes of variations and some peaks of concentrations are greater at heights of 0.5 and 1.0 km, i.e., in the boundary layer. In our opinion, this fact proves that sources and sinks of the gases in question are located on the underlying surface [10-12].

It is also clear from Fig. 1 that for each gas, the differences in concentration in height and amplitude of the variations are significantly different. This is evidence of two circumstances: the power of the mechanisms of generation and sink of these compounds in the region and the difference in concentrations for the global and regional backgrounds. Thus, if concentrations of gases at a height of 0.5–1.5 km are considered the regional background and the global background is considered to be at a height of 7 km, then the bigger their difference, the greater the differences between such scales.

Figure 2 presents average long-term vertical profiles for each month of the year. It is seen that the minimum concentration of  $CO_2$  above the region is recorded in August and at all heights. It may seem strange that the maximum concentration of  $CO_2$  is reached in April and also at all heights. The greatest amplitude of variations in the annual behavior of  $CO_2$  concentration is recorded in the boundary layer (0.5–2 km), amounting to 14–20 ppm. The modulation in the annual behavior of  $CO_2$  concentration remains in the free atmosphere too, although the amplitude here is smaller (7–8 ppm).

We draw one more conclusion from Fig. 2 for the vertical change in  $CO_2$  concentration. This gas is assimilated by vegetation only during four months in May–August. In the other part of the year,  $CO_2$  is generated due to breathing plants.

The methane behavior (Fig. 2) discloses the features of Western Siberia, which has the world's biggest swamp systems. It is seen that the methane concentration in the boundary layer is almost 100 ppb greater than in the free atmosphere. This fact evidences that a powerful source of  $CH_4$  is situated on the underlying surface. The methane does not release into the region in this case, as the content of  $CH_4$  is higher in the boundary layer than in the free atmosphere, even at the minimum concentration (August). The maximum concentration is recorded in January–February in all atmospheric depths, including the boundary layer.

The differences in the concentration of  $N_2O$  between the boundary layer and the free atmosphere do not exceed 1 ppb (Fig. 2). This means that in the area in question, there are no powerful sources and sinks of nitrous oxide. Figure 2 shows one more interesting fact. The gas concentrations are almost identical during 10 months of the year. Their differences increase only near the minimum (July) and the maximum (December). The absence of powerful sources and sinks results in that the average annual amplitude of the variation in N<sub>2</sub>O concentration is not over 2 ppb.

To make sure that our conclusions are correct, we plotted Fig. 3 by the same data. It is seen that there are very powerful mechanisms of  $CO_2$  generation and sink in Western Siberia which change their character during a year. In September–April, the generation of  $CO_2$  exceeds the sink. In May–August, the sink capacity exceeds the generation. This fact is proven by the conversion of concentration curves at different heights.

All year generation is typical to methane (Fig. 3). At heights of 0.5–1.5 km, two maximums and two minimums recorded by the monitoring data in the surface air can be distinguished [13]. This fact may be explained by two mechanisms or cycles of methane formation [14, 15]. It can be generated by anaerobic bacteria in soil or can emit from the swamp surface. Therefore, the winter maximum is influenced by bacteria activity, and the summer maximum by the release from the swamps.

We do not make additional comments regarding the annual behavior of  $N_2O$ , since it is not significant.

Close monitoring measurement conditions (every month at the same time of the day (at 1:00 p.m local time) and the same place of observation) may assign equal statistical weight to each profile in the first approximation. Then, it is possible to determine not only the average monthly, but also the average annual concentrations of the gases in question (Fig. 4).

Figure 4 shows that regardless of the short variation period (1997–1999), the steady-state and almost linear increase in  $CO_2$  concentration is recorded at all heights every year. The growth rate was 21 ppm for 11 years or 1.9 ppm/year, which is close to the value determined by WMO using the global monitoring network [16].

For methane, such an accurate estimate cannot be made from Fig. 4. The data of the global network do not provide an exact answer about  $CH_4$  behavior either. By the



Fig. 1. The change in CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O concentrations above Western Siberia at different heights since 1997.



Fig. 2. The vertical distribution of greenhouse gases for each month of the year.

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Fig. 3. The annual behavior of a change in gas concentrations at different heights.



Fig. 4. The annual average concentrations of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O above Western Siberia at different heights.

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**Fig. 5.** The long-term behavior of  $CO_2$  in July at different heights above Western Siberia.

data in [16],  $CH_4$  decreased by 1 ppb/year in 2005–2006 and by 2.4 ppb/year on average for the last 10 years.

According to Fig. 4, the concentration of nitrous oxide grew at a rate of 0.73 ppb per year at all heights, which is somewhat less than in [16].

We showed earlier in [17] that at the total growth of  $CO_2$  above Western Siberia, its concentration in the boundary layer was almost constant during the summer period. Therefore, we made the conclusion that the vegetation of the region in the summer dealt with the releasing  $CO_2$ , i.e., assimilated it completely. The continuous measurements made us doubt the correctness of the conclusion. It follows from Fig. 5 that in 2005–2007, the  $CO_2$  concentration increased sharply in the boundary layer (0.5 and 1 km). Such behavior does not have obvious reasons. There was neither large-scale deforestation nor strong fires. Therefore, it is still difficult to answer what periods—2002–2004 or 2005–2007—were abnormal.

Figure 6 represents how the concentration of greenhouse gases increased in all atmospheric depths.

It is seen that the concentrations of  $CO_2$  and  $N_2O$  grow at a different rate but in all studied atmospheric depths, the average annual profiles of concentration do not intersect. This is not true for methane. Except for 1997, which is represented by an incomplete series of profiles, all the others are located in a narrow passage with numerous intersections of curves at different heights. Such behavior probably shows local weather conditions that were rather diverse during this period [18].

To complete our analysis, we present Fig. 7, which illustrates the long-term average profiles of the vertical distribution of  $CO_2$ ,  $CH_4$ , and  $N_2O$  concentrations above Western Siberia plotted by all sounding records (over 120 cases).

The horizontal sections designate the mean square deviations of concentrations ( $\pm 1$  MSD) with the absolute maximum and minimum concentrations indicated



**Fig. 6.** The long-term change in the vertical distribution of greenhouse gases above Western Siberia.

at each height for the entire sampling on the right and on the left.

We believe that the data presented in Fig. 7 may be useful in the selection of modeling scenarios for regional models and in other climate applications.



**Fig. 7.** The long-term average vertical distributions and mean square deviations of  $CO_2$ ,  $CH_4$ , and  $N_2O$  concentrations above Western Siberia; absolute maximums and minimums of concentrations at different heights.

# CONCLUSIONS

Our investigation proves that there is a long-term trend of  $CO_2$  and  $N_2O$  concentration in the atmospheric depth above Western Siberia at a rate of 1.9 ppm/year

and 0.73 ppb/year, respectively. For  $CH_4$ , such unambiguity is not established.

The region varies in time from a powerful source to sink for carbon dioxide. In September–April, Western

Siberia is a  $CO_2$  donor; in May–August, it assimilates  $CO_2$ . For methane, it is a constant donor that changes its intensity during the year. The small variation in the N<sub>2</sub>O concentration at all heights points to the absence of significant sources and sinks of this component of the air in the region.

The greatest variation of all three greenhouse gases is recorded in the boundary layer, where it is higher than in the free atmosphere by 2–3 times.

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## REFERENCES

- A.V. Eliseev, I.I. Mokhov, M.M. Arzhanov, et al., "Interaction of Methane Cycle and Processes in Swamp Ecosystems in a Climatic Model of Intermediate Complexity," Izv. RAN, Fiz atmos. i okeana, No. 2, 44, 147–162 (2008) [Atm and ocean physics, No. 2, 44 (2008)].
- E.M. Volodin, "Cycle of Methane in a Climate Model of RAS, Izv RAN," Fiz atmos. i okeana, No. 2, 44, 163–170 (2008) [Atm and ocean physics, No. 2, 44, (2008)].
- V.A. Grabar and M.L. Gitarskii, "Estimate of Formation of Atmospheric Carbon in Forest Products," Meteorol i gidrol., No. 4, 23–29 (2008).
- 4. R.A. Houghton, "Revised Estimates of the Annual Flux of Carbon to the Atmosphere from Changes in Land Use and Land Management," Tellus, No. 2, 55B, 378–390 (2003).
- A. Shvidenko and S.A. Nilson, "A Synthesis of the Impact of Russian Forests on the Global Carbon Budget for 1961–1998," No. 2, 55B, 391–415 (2003).
- S.A. Nilson, A. Shvidenko, M. Jonas, et al., "Uncertainties of a Regional Terrestrial Biota Full Carbon Acrount:

A Systems Analysis, Water, Air, and Soil Pollution," Focus, Nos. 4–5, 7, 425–441 (2007).

- C. Rodenbek, S. Houweling, M. Gloor, et al., CO<sub>2</sub> Flux History 1982–2001 Interred from Atmospheric Data Using a Global Inversion of Atmospheric Transport," Atmos. Chem. Phys., No. 6, 3, 1919–1964 (2003).
- V.E. Zuev, B.D. Belan, D.M. Kabanov, et al., "AN-30 "Optic-E" Aircraft-Laboratory for Ecological Investigations," Optika atmos. i okeana, No. 10, 5, 1012–1021 (1992) [Atm and ocean optics, No. 10, 5 (1992)].
- A.N. Ankilov, A.M. Baklanov, B.D. Belan, et al., "Annual Change in Concentration of Protein in Biogenic Component of Atmospheric Aerosol in the South of Western Siberia," Optika atmos. i okeana, Nos. 6–7, 14, 520–525 (2001) [Atm and ocean optics, Nos. 6–7, 14, (2001)].
- F. Apadula, A. Gotti, A. Pigine, et al., "Localization of Source and Sink Regions of Carbon Dioxide through the Method of the Synoptic Air Trajectory Statistics," Atmos. Environ., No. 27, 37, 3757–3770 (2003).
- M.A.K. Khalil and R.A. Ramussen, "Sources, Sink, and Seasonal Cycles of Atmospheric Methane," J. Geophys. Res., No. 9, 88, 5131–5144 (1983).
- 12. B.D. Belan, "Tropospheric Ozone. Components of Main Cycles," Optika atmos. i okeana, No. 4, **22**, (2009) (in press).
- M.Yu. Arshinov, B.D. Belan, D.K. Davydov, et al., "Spatial and Time Variation of CO<sub>2</sub> and CH<sub>4</sub> Concentrations in the Ground Air Layer of Western Siberia," Optika atmos. i okeana, No. 2, **22** 183–192 (2009).
- D.H. En halt, "The Atmospheric Cycle of Methane," Tellus, Nos. 1–2, 26, 58–70 (1974).
- 15. G.A. Zavarzin, "Microbal Cycle of Methane under Cold Conditions," Priroda, No.6, 3-14, (1995).
- 16. WMO Greenhouse Gas Bulletin, No. 3, 4, (2007).
- M.Yu. Arshinov, B.D. Belan, G.Inoue, et al., "Dynamics of the Verical Distribution of CO<sub>2</sub> and CO Concentration over Western Siberia (1997-2003), Advances in the Geological Storage of Carbon Dioxide," Kluwer Academic Publishers, New-York, 65, 11–15 (2006).
- B.D. Belan, T.M.Rasskazchikova, and T.K. Sklyadneva, "Synoptic Regime of Tomsk 1993–2004," Optika atmos. i okeana, No. 10, 18 897–902 (2005).