### Organization of monitoring of the greenhouse gases and of the components oxidizing the atmosphere over Siberia and some results obtained. 1. Gas composition

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We describe the multilevel system for monitoring of the greenhouse atmospheric components as well as of the components oxidizing the atmosphere and present some results obtained. The system has been developed under the international research program. The monitoring is being carried out in three research areas: 1) determination of the emission of greenhouse gases at the Iksinskoye (Bakcharskoye) marshland; 2) study of the fluxes in the ground atmospheric layer (monitoring of the vertical distribution of the greenhouse gases is performed using tall instrumented towers in Tomsk Region, Yamalo-Nenetskii Autonomous District, Tyumen Region, and Khanty-Mansiysk Autonomous District; 3) study of the spatial distribution. Measurements of the vertical distribution of the greenhouse gases in the boundary layer of the atmosphere are being performed weekly from onboard an AN-2 aircraft, while the measurements aimed at estimating the regional contribution to the global balance from onboard an AN-30 aircraft. In 2006 studies under YAK-1 Project have been started. The studies under this project are being carried out from onboard an AN-30 aircraft, equipped with the same instrumentation, in flights along the route from Novosibirsk to Yakutsk and back to Novosibirsk at different flight heights along the route.

#### Introduction

The problem of possible global changes of the environment and climate due to increase of greenhouse gas concentration related to the anthropogenic pollution is among the main presentday problems. The second aspect of this problem, also related to anthropogenic activity, refers to noncarbonaceous greenhouse gases and secondary atmospheric admixtures determining the oxidizing characteristics of the atmosphere. They are primarily the tropospheric ozone, nitrogen oxides, sulfur oxides, and aerosol.

The formation of air composition in a specific region is determined by the sum of several factors. Among those there are the background content of impurities in the air mass arrived or located at the moment over the area under study as well as the balance between local sources and surface sinks and also the transformation of impurities, taking place constantly though significantly changing the mechanisms and intensity during a day.

Under conditions of west-to-east air mass transport, prevailing on the Earth, the export of pollutants from Europe to Siberia is observed. These regions differ considerably by the emissions of the precursors of secondary impurities. In Europe the emissions of  $NO_x$  and CO from industries and

transportation are of the primary importance, while in Siberia the arrival of hydrocarbons from the forest ecosystems plays an important part. Therefore it is vital to determine the budget of pollutants in the Siberian atmosphere and also from what part of Europe (south, central or north) and what amount of pollutants is exported to Siberia. For Siberia the problem of studying the components of carbon cycle and oxidizing components is dramatized by a nearly complete absence of the observation network.

We present in this paper a multilevel system of monitoring of greenhouse gases and pollutants oxidizing the atmosphere that has been developed under the international cooperation. Also we present some results obtained by now.

### 1. Measurements of greenhouse gases emission

The determination of gas emission is made at Iksinskoye (Bakcharskoye) marshland (56°49'35"N, 82°51′27″E) at about 16 km to the southwest from Plotnikovo settlement, Bakcharskii District, Tomsk Region, at the stationary field research base of the Institute of Agricultural Chemistry and Soil Science (IACSS) SB RAS (Novosibirsk). This field base was chosen because this marshland is typical for the Vasyugan area and a long series of observations had already been carried out there by the scientists from IACSS SB RAS.

Twelve plastic chambers of  $0.9 \times 0.9 \times 0.6$  m<sup>3</sup> size were mounted there, the air from which was pumped through pipes to two sensors, sensitive to the methane and CO<sub>2</sub> concentration. Figure 1 shows the map with disposition of the chambers installed.



Fig. 1. Schematic of location of the measurement chambers at the polygon in Plotnikovo: (*a*) the marsh remained in the primitive form; (*b*) the marsh recultivated during the Soviet period.

Besides, a standard meteorological mast was installed at this field base with the following instruments:

– temperature and humidity sensors (HMD45A/D, Vaisala, Finland);

- sensors of wind velocity and direction (Model 05103, Joung Wind Monitor, USA);

– ultrasonic anemometer (CSAT Campbell Scientific, Inc., Canada);

- sensor of soil temperature (QMT103, Vaisala, Finland);

sensor of pressure PTB210, the firm Vaisala, Finland);

- sensor of water level (PTX1830/1730, Druck LTD, Great Britain).

The operation of the above-mentioned complexes is as follows. The methane and carbon dioxide concentrations in a closed chamber are measured every hour, during 15 min whole day round. After taking measurements, the chamber is ventilated. The measurement data are recorded into a CR10X datalogger in 10-min intervals.

Figure 2 shows the results of measurement of methane fluxes in each of the six chambers in both parts of the marshland.



Fig. 2. Methane fluxes measured at the two areas.

One can notice that at the primitive marsh the methane fluxes can exceed by several times the emission from the recultivated marsh. The differences are also evident at separate parts of one and the same measurement site. They can reach three times.

# 2. Measurements in the atmospheric boundary layer

The air composition monitoring may be subdivided historically into two stages. At the first stage the monitoring was performed in the suburbs of Tomsk. For this purpose an automated station named the TOR Station has been created (according to the name of the project Tropospheric Ozone Research under the program EUROTRAC). This station was put into operation in December 1992 and it has been permanently operated until now. The TOR Station is described in the literature.<sup>1</sup> Since 2006 one more station has been put into operation at the "Fonovyi" (background) polygon near Kireevsk settlement, Tomsk Region. The functioning of these two stations has been described in the literature.<sup>2-5</sup> Under the project Russian-Japanese research joint the investigation of spatial distribution of greenhouse

gases and components oxidizing the atmosphere have been extended into other geographical regions.

The measurements are being conducted using tall towers located at the following points: the station Karasevoe, Tomsk Region; Berezorechka settlement, Tomsk Region; Noyabrsk city, Yamalo-Nenetskii Autonomous District; the Dem'yanskoe settlement, Tyumen Region; Igrim settlement, Khanty-Mansiyskii Autonomous District. Each of the towers is equipped with identical sets of instruments, including an LI-820 nondispersive infrared analyzer of  $CO_2$ , an HMD45A/D (2 pcs) temperature and humidity sensitive element; a wind velocity and detector – Young Model direction 05103; an atmospheric precipitation meter – Young Model 52202; a Kipp&Zonen Model CM3 pyranometer; a CR10X datalogger in a complex with a COM210 telephone modem. Figure 3 shows the location of sensors on the tower.



Fig. 3. The positioning of sensors at the tower.

The measurements are carried out continuously with the intakes located at two heights being successively switched. One of the intakes is located at the top of a tower, the other at the level to 0.5 of the distance between the height of surrounding trees and the tower top. The measurement data are acquired with a datalogger at a rate of 1 Hz. The compiled arrays of data are every month transmitted, through a modem, to the Institute of Atmospheric Optics SB RAS.

At present 5 stations have been in operation in the routine monitoring mode. To extend the research geography, a permission of Federal Service on Technical and Expert Control was received to mount the equipment at 2 stations more and an application has been sent there for the permission to equip the eighth tower. Figures 4 and 5 show time behavior of the  $CO_2$ and methane concentration at three stations. It is evident that for  $CO_2$ , regardless of the location, the time dynamics is determined by one and the same atmospheric process.

There are differences only in the amplitude of variations. The methane behavior at these points is of great diversity. Because the methane flux is mainly directed from the bottom upwards, i.e., its source is the underlying surface shown in Fig. 6, one can draw a preliminary conclusion that the mechanisms of the formation of spatial fields of these gases are different.







Fig. 6. Time dynamics of methane at two levels at the station Karasevoe.

# 3. Measurements in the atmospheric boundary layer

The measurements of the vertical distribution of greenhouse gases in the atmospheric boundary layer are performed from onboard an AN-2 aircraft belonging to the Tomsk regional agency ROSTO. An LI-800 gas analyzer is mounted onboard the airplane to measure the  $CO_2$  concentration, also "Vaisala" temperature and humidity sensors are mounted.

The aircraft is based at the ROSTO airport (the Golovino settlement, Tomsk District, Tomsk Region). The monitoring of air was made over the Berezorechka settlement, Tomsk District, Tomsk Region close to the point at 56°08'N, 84°20'50"E. This region has been chosen for airborne sensing to enable referencing the measurement data, obtained using the tower, to data obtained in the free troposphere from onboard an AN-30 aircraft. During warm season the flights are carried out in the altitude range from 150 m to 3000 m, while during cold season – from 150 m to 2000 m. The flights are carried out every week given cloudless weather conditions.

Figure 7 is an example of the data measured, which shows the vertical profiles of  $CO_2$  in different seasons obtained using the complex of equipment mounted onboard an AN-2 aircraft.

Figure 7 shows that the greatest variations of  $CO_2$  concentration are observed in the atmospheric boundary layer from 345 ppm to 400 ppm. By the top of the atmospheric boundary layer the dynamics decreases from 352 ppm to 382 ppm. Taking into

account that the region of measurements is surrounded by woodlands, such a result seems to be quite natural.

# 4. The measurement of vertical distribution of components

The experiments on estimating the regional contribution to the global balance are carried out from onboard AN-30 aircrafts from "Severnyi" airport in Novosibirsk. The description of the set of instrumentation this aircraft is equipped with can be found in the literature.<sup>6</sup>

Air sampling is performed with a GAST DOA-P108 (Trademark of Gast Manufacturing INC., USA) oil-free diaphragm pump into the glass flasks, which then are used in the gaschromatographic analysis.

The air sampling and analysis of the gas composition are performed the third decade of each month, depending on the weather conditions, during 3 hours, including flight time to the measurement polygon and back. The region for monitoring was chosen to the southwest of Novosibirsk, lest the city industrial plume would affect the instrumental readouts. The operation route passes over the pine forest, close to Zyryanka and Ordynskoye



Fig. 7. The vertical  $CO_2$  distribution in the atmospheric boundary layer near the Berezorechka settlement.

settlements. The airborne measurements start at the point with the coordinates 54°05′N, 81°50′E and stop at the point with the coordinates 54°35′N, 82°40′E. The flight height ranges from 500 to 7000 m.

Below we present some results of this type of monitoring, started in July 1997 and lasting up to now.

In the first approximation one may consider that in the figures given below the gas concentration at a height of 500 m shows the action of local sources, at a height of 2000 m it shows the action of the area of West Siberia and the north of Kazakhstan, at the height of 7000 m – global content at this latitude. It follows from Fig. 8*a* that the CO<sub>2</sub> concentration increases everywhere. Besides, we notice that over the period from 1997 to 1999 and in 2003 the CO<sub>2</sub> content over the southern part of Western Siberia was higher than the global content.



345 340 335 1997 1998 1999 2000 2001 2002 2003 2004 2005 *C* 

Fig. 8. The long-term behavior of gas concentrations over the area of Western Siberia surveyed.

Methane at all heights behaves differently (Fig. 8b). First an increase of the methane content was observed (1997–1998), then during the period

from 1999 to 2002 the increase stopped, and after that its behavior became close to neutral.

If the increased methane concentrations at low heights can obviously be explained by the contribution from of marshes that are vast in West Siberia, the growth  $CO_2$ , in the region with the abundant vegetation accumulating  $CO_2$ , calls for certain questions. The absorption of  $CO_2$  by Siberian forests is shown in Fig. 8*c*, where one can observe the long-term variations of  $CO_2$  concentration during summer period.

It follows from this figure that in the period of intense summer vegetation the southern areas of West Siberia and Russia, as a whole, are the area of sink for  $CO_2$  from the atmosphere. The  $CO_2$  concentration at heights of 500 and 2000 m is much lower than at the level of 7000 m. Moreover, Fig. 8*c* shows that the  $CO_2$  sink remains relatively constant over the period of nine years.

Evidently, such differences in the long-term variations of  $CO_2$  concentration (annually mean, Fig. 8*a*, and summer one, Fig. 8*c*) are due to the short vegetation period in Siberia. To confirm this hypothesis, the annual behavior of  $CO_2$  concentration, obtained at the TOR Station in the atmospheric boundary layer is shown in Fig. 9.



Fig. 9. The annual behavior of  $CO_2$  concentration in the ground layer of the atmosphere near Tomsk.

Figure 9 shows that only during three months the  $CO_2$  sink exceeds its generation. Consequently, in the annual mean profile this territory is rather producing than accumulating  $CO_2$ .

Carbon monoxide is a precursor of the carbon dioxide developing into the latter in the course of oxidizing processes. Therefore, the knowledge of its spatiotemporal variability can help in the predicting the  $CO_2$  concentration. Figure 10*a* shows that carbon monoxide concentration decreases during the period under study if the burst in 2003 is not considered. A comparison of concentrations at heights of 0.5 and 7 km (that can be taken as the global background) indicates that the area of Western Siberia is the sink for this air component.

Nitrous oxide is the precursor of nitric oxides oxidizing the atmosphere. The variation of its concentration, shown in Fig. 10*b*, exhibits a tendency toward the growth during the entire period of measurements.



**Fig. 10.** Long-term behavior of CO over Western Siberia (*a*). The same for the nitrous oxide (*b*).

Thus, from all the above-mentioned components the two components have a tendency toward the concentration stabilization or even toward a decrease, while two other components have a tendency toward the concentration increase. This is in a good agreement with data of different measurements generalized in the literature.<sup>7</sup>

# 5. The measurement of spatial distribution in the free atmosphere

This type of monitoring is performed in the framework of the YAK-1 Russian-French research project. The goal of this project is the analysis of seasonal and interannual variations of sources and sinks of  $CO_2$ , the transport of ozone forming substances from the Western Europe to Asia (Siberia) by determining the monthly vertical profiles of CO,  $CO_2$ , and  $O_3$  and calculating their west-to-east gradients (Europe–Asia); vertical gradients between the atmospheric boundary layer and the troposphere at different parts of the path along the latitude circle.

To achieve the goals of the project the investigations are carried out along the route from Novosibirsk to Yakutsk and back to Novosibirsk. The schematic diagram of the flight is shown in Fig. 11.

The experiments on estimating the regional contribution to the global carbon balance are being carried out from onboard an instrumented AN-30 "Optik-E." In addition to standard set of equipment,<sup>6</sup> the following equipment has been installed onboard the aircraft, namely, an ultraviolet ozonometer (49C model), and a correlation CO gas analyzer (48CTL model), manufactured by Thermo Environmental Instruments, USA; a nondispersion infrared gas analyzer of series LI-6262, from LICOR Company, USA, which is intended for measuring CO<sub>2</sub> and water vapor in the atmosphere.



Fig. 11. The diagram of the flight routes under the YAK-1 project.



Fig. 12. Vertical atmospheric section along the flight from Novosibirsk to Yakutsk.

It is now planned that in the future an automated "MPI.BCG modular Flask Sampler" system, produced by Max-Planck-Institute für Biogeochemie, will be mounted to fill the glass flasks with outboard air, which will be used in the gaschromatographic analysis.

To obtain the vertical profile of the atmosphere over the entire route, the flight height varied from the minimal possible height of 500 m over the terrain and 100 m in the airport zone and up to 7000 m along the flight route. Seven such descents are performed along both flight legs (Novosibirsk–Yakutsk and Yakutsk–Novosibirsk). The flights to and back were performed using different routes: the flight to along the north route and back along the south route. As a result, during each round trip 14 profiles are acquired spaced along a horizontal (depending on the flight height) by 50 to 250 km. Such a flight route allows one to assess the gradients of the measured parameters with a step of 50 to 500 km.

By now the first flight has been performed under this project. Figure 12 shows the vertical section of the  $CO_2$  concentration along the route. It can be seen from the figure that its distribution is inhomogeneous and the maximum values are in the middle part of the route. Preliminary analysis of the route has shown that it was just in the region where the arctic air invaded from the northern regions where  $CO_2$  concentration is, as a rule, higher.<sup>7</sup> It is planned that in the future the time of flight should be chosen at the west-to-east transport to achieve the goal stated above.

#### Conclusion

The integration of resources and efforts of international cooperation has made it possible to create the system of monitoring the greenhouse gases and components oxidizing the atmosphere in the range of heights from 0 to 7000 m over a considerable part of Siberian region.

The results obtained by now have shown a significant variability of the measured components both in time and in space, contradicting in some cases the earlier formed conceptions.

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