### Sites for monitoring of greenhouse gases and gases oxidizing the atmosphere

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We describe two sites intended for monitoring of CO,  $CO_2$ , NO,  $NO_2$ ,  $SO_2$ ,  $O_3$ , aerosol disperse composition, and meteorological quantities. One of the sites is located in the background zone and the other one in urban area of Tomsk Region. We present characteristics of automatic calibration of gas analyzers using test mixtures or microflow capsules. Observations at the sites are being preformed hourly round the clock. The measurement data are available at the Internet.

### Introduction

One of the primary modern challenges is the problem of possible global environmental and climate changes which, given their extreme manifestation, may lead to natural global catastrophes, whose consequences will not only be large economic but, unfortunately, also human losses. The reason for its appearance lies in possible climate change due to the growth of the concentration of greenhouse gases associated with anthropogenic activity.

Considerable uncertainty present in climate estimates of possible anthropogenic changes, obtained by numerical modeling, is associated with the modern state of the studies into the interrelation between the climate change and abundance of the greenhouse gases in the atmosphere and, in particular, the components of carbon cycle. This problem only starts to be investigated in the leading research centers of the world.

On the territory of Russia there are Siberian forests containing about 15% of the world carbon recourses. It is noteworthy that their role as carbon sources and sinks is still poorly accounted for in the current global and regional models because of the extreme paucity of the initial data and their insufficient spatial representativeness. As a result, the estimates of the contribution of Siberian forests to the global carbon balance are quite ambiguous and contradictory. For instance, Houghton<sup>1</sup> estimates suggest that these forests are the carbon source with production of carbon at about 0.035 PgC/year. On the contrary, Shvidenko and Nilsson<sup>2</sup> have shown that the Siberian forests mainly uptake carbon at the rate of  $(0.42 \pm 0.07)$  PgC/year. The quantity determined by them is comparable with the carbon uptake over the entire Northern Hemisphere.<sup>3</sup> Therefore, to eliminate this ambiguity, it is necessary to perform correct direct measurements of carbon fluxes over the entire region.

The second aspect of the problem is associated with the non-carbon greenhouse gases and secondary atmospheric gases determining the oxidizing properties of the atmosphere and primarily with the tropospheric ozone. Given the predominant west-to-east transport on the globe, there is export of pollutants from Europe to Siberia.<sup>4</sup> These regions substantially differ by the emissions of the ozone precursors. In Europe the major role is played by  $NO_x$  and CO emissions by industry and motor transport, while in Siberia it is the delivery of hydrocarbons from forest ecosystems. Therefore, it is important to determine the budget of pollutants in the atmosphere of Siberia, and in particular what (south, central, or north) part of Europe exports species to Siberia and what are the amounts.

For Russia, the problem of studying the components of carbon cycle is complicated by almost total absence of network of observations of greenhouse gases.

At present, regular measurements of concentration of greenhouse gases, including components of carbon cycle, are performed at the following locations: 11 sites in Africa, 28 sites in Asia (excluding Russia), 5 sites in south America, 46 sites in North and Central America, 31 sites in the region of Pacific Ocean, 87 sites in Europe (excluding Russia), and 19 sites in Antarctic. Most of these sites are equipped with modern gasanalytic instrumentation manufactured in the USA, Great Britain, and France. As a rule, the monitoring stations operate in an automated mode, attended periodically (once or twice a month) by operators. The performance of devices for such short time intervals is corrected through daily calibration using test mixtures or reference microflows. It should be noted that the calibration of devices on foreign network of the stations takes about 60% of time and about same fraction in the measurements costs.

In Russia (Russian Hydrometeorological Agency) there are only 3 stations measuring the carbon dioxide, two located on islands and one being oceanic. Thus, on the territory of Russia *there are no domestic stations for monitoring of greenhouse gases*. Naturally, in the absence of data their role as carbon sources and sinks is poorly accounted for in the current global and regional models.

This situation forces the world community to fill this gap at least partly. The measurements of greenhouse gases are performed either as a part of European projects (episodically) under international grants or as part of long-term (from 1994 to the present time) Russian-Japanese program on studying the greenhouse gases over Siberia.

For solution of this problem, it is necessary to organize monitoring of greenhouse gases. This paper describes two sites created at the Institute of Atmospheric Optics SB RAS for monitoring of greenhouse gases and gases oxidizing the atmosphere.

### 1. Site location

The Institute of Atmospheric Optics SB RAS owns two polygons: (1) the base experimental complex (BEC) located in Akademgorodok (56°29'N, 85°04'E) at 170 m asl, and (2) a "Background" site located near Kireevsk village (56°25'N, 84°04'E) 60 km to

the west of the city on the bank of Ob river at 80 m asl (Fig. 1). It is seen that, given the prevailing west-to-east transport the air will sequentially pass over the background polygon, Tomsk, and BEC.

The two polygons are equipped with identical automated complexes for measurement of meteorological quantities in the near-ground atmospheric layer. Both of the complexes are Unzha-2 guyed masts equipped, at four levels, with the air temperature and humidity sensors and M-127 anemometers of electromechanical type. Structure features of the complex have been described in Refs. 5 to 7. Wind sensors are installed at the heights of 10, 20, 30, and 36 m above the ground at the beam holders oriented toward the south and north directions. Such an arrangement makes it possible to take into account the perturbation of air flows by the mast frame. At these same levels, the electronic temperature and humidity sensors are also installed. Some results of exploitation of these complexes can be found in Ref. 8.



Fig. 1. Location of sites at polygons: "Background" (1) and BEC (2).

In constructing the measurement sites, we used already existing meteorological complexes, installed at both polygons of the institute.

For arrangement of gas analyzers, aerosol analyzers, controlling, recording, and transmitting instrumentation, at the "Background" polygon we used a building made from bricks, located in immediate vicinity of the mast. At the BEC we installed, for these purposes, a 20-foot container with the internal partition, separating it into tambour and instrumentation module.

In both rooms, we undertook measures of thermal insulation of instrumentation modules, and to provide comfortable operation of the instrumentation at low/high ambient temperatures, we installed climate installations, operated in automated mode and being capable of keeping the temperature and humidity in the workroom within the required range.

Around the "Background" polygon, there are no industrial plants, and the data obtained with it will characterize background conditions. The second site is located in north-east suburbs of Tomsk Akademgorodok. Considering the prevailing west-to-east air transport in the region, we can say that the second site will be overburdened by the anthropogenic effect because the air mass, past "Background" site, will pass through the territory of Tomsk, and then will arrive at Akademgorodok. Thus, the obtained data will not only make it possible to monitor the air composition under the background conditions, but also to identify the contribution of big industrial center to its variations.

## 2. Composition and structure of sites and measured parameters

This section describes both sites because they are identical in the principles of arrangement of measurements, but different in the number of parameters measured.

Because the "Background" polygon is separated from the city by the distance of 60 km, the devices installed there cannot be attended continuously. Therefore, we included in it only reliable, tested devices. At the base experimental complex, servicing personnel resides continuously. Here, we plan to perform validation of all new methods and new instruments. Consequently, the two polygons will have different measurement complexes (Table 1).

For equipment at the sites we used the gas analyzers with detection thresholds at the level of background concentrations. These gas analyzers, fabricated by ZAO "OPTEK" (Saint Petersburg), have been exploited for a long time as part of TOR station<sup>9</sup> and proved the possibility of their use in the longterm monitoring regime.

Table 1. Measurement site at the base experimental complex at the "Background" polygon

	1	0 100	
Instrument	Parameter	Range, error	Note
	Base experimental compl	lex	
	Temperature	-50+50°C, ±0.2°C	4 levels
Meteorological complex	Relative humidity	10100%, ±5%	4 levels
of IAO SB RAS	Wind speed	$0.560 \text{ m/s}, \pm 5\%$	4 levels
	Wind direction	0360°, ±10°	4 levels
S310	SO <sub>2</sub> $02000 \ \mu g/m^3, \pm 25\%$		2 levels
D 2404	NO	$01000 \ \mu g/m^3, \pm 25\%$	2 levels
R-310A	NO <sub>2</sub>	$01000 \ \mu g/m^3, \pm 25\%$	
3-02P	O <sub>3</sub>	$01000 \ \mu g/m^3, \pm 15\%$	2 levels
SV-320	$H_2S$	$0200 \ \mu g/m^3, \pm 15\%$	2 levels
N-320	NH <sub>3</sub>	$01000 \ \mu g/m^3, \pm 15\%$	2 levels
L-061-02 (GMM222)	$CO_2$	010000 ppm, ±20%	2 levels
K-100	СО	$0400 \text{ mg/m}^3, \pm 20\%$	2 levels
NMHC-2000	$CH_4$ and $\Sigma CH$	$05$ ppm and $050$ ppm $\pm 20\%$	2 levels
Aerosol meter, GRIMM model 1108	Aerosol disperse composition	$0300 \text{ cm}^{-3}, \pm 25\%$	1 level
DAS	Same	$01000 \text{ cm}^{-3}, \pm 25\%$	1 level
	"Background"		
	Temperature	$-50+50^{\circ}C, \pm 0.2^{\circ}C$	4 levels
Meteorological Complex of IAO SB	Relative Humidity	10100%, ±5%	4 levels
RAS	Wind speed 0.560 m/s, ±5%		4 levels
	Wind velocity	0360°, ±10°	4 levels
S310	$SO_2$	$02000 \ \mu g/m^3, \ \pm 25\%$	2 levels
D 2404	NO	$01000 \ \mu g/m^3, \pm 25\%$	2 levels
R-310A	NO <sub>2</sub>	$01000 \ \mu g/m^3, \pm 25\%$	
3-02P	O <sub>3</sub>	$01000 \ \mu g/m^3, \pm 15\%$	2 levels
L-061-02 (GMM222)	$CO_2$	010000 ppm, ±20%	2 levels
K-100	СО	$0400 \text{ mg/m}^3, \pm 20\%$	2 levels
Aerosol meter, GRIMM model 1108	Aerosol disperse composition	$0300 \text{ cm}^{-3}, \pm 25\%$	1 level

Analyzers of ammonium and hydrogen sulfide are new and so they are installed only at BEC. Moreover, at this stage the instruments are not integrated into the measurement complex. Their data are not written into the output file, instead being saved into a separate storage. For these, we have selected the time cycle of schedule of work and calibration, and perform additional work for the case of possible failure of separate instruments. After testing, the second set of these instruments is planned to be installed at remote site and merge their readings into the common information-transmitting network of complexes.

For a more complete determination of carbon cycle we have installed, as part of the site at the BEC, a foreign NMHC-2000 analyzer (France). Since a part of carbon in the atmosphere is converted into the aerosol form, for closure of the atmospheric cycle of carbon balance a control of aerosol disperse composition is required. For this, aerosol meters of the GRIMM firm, Germany are included into site instrumentation. Finally, the diffusion aerosol spectrometer (DAS), fabricated at the Institute of Chemical Kinetics and Combustion SB RAS and designed for measurement of nanoparticles in the atmosphere, is the device of laboratory performance that requires permanent servicing, and so it is located only at BEC.



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At the available beam consoles of the masts (Fig. 2), at the altitudes of 30 and 10 m we have mounted air samplers, separating out the direct precipitation and biological constituent. Via pipes, the air intakes are connected with gas analyzers, installed in the instrumentation modules at the base of the

mast. For measurements, calibration, and transmission of the data, all the devices are united into a local network managed by the logger. For archival of the data, we use the principles formulated earlier during multiyear exploitation of TOR station. Location of intakes of gas constituents at two levels makes it possible to determine not only gas concentrations but also their fluxes by the gradient method.

Samples of atmospheric air for gas analyzers are collected at two altitudes. Moreover, they are calibrated hourly. We also have developed a general scheme of gas-distribution channels, constructed taking into account the specific features of calibration of each of the devices. As seen from Table 1 the gas analyzers in the complexes are, at the first stage, different, however, the developed gas distribution scheme makes it possible to complement the complex at the "Background" polygon and to make it identical to the complex at the BEC but, when necessary, to add new gas analyzers in it.

For the reason of different specifications the gas analyzers are separated into two groups, with analyzers of SO<sub>2</sub>, NO<sub>x</sub>, H<sub>2</sub>S, NH<sub>3</sub>, and O<sub>3</sub> in the first group, and analyzers of CO, CO<sub>2</sub>, and CH<sub>4</sub> in the second one. The gas commutation schemes themselves are presented in Figs. 3 and 4.



**Fig. 3.** Scheme of air release module for gas analyzers of the first group installed at the BEC.

Gas analyzers of the first group measure "more active" atmospheric gases. For this reason, all channels for gas analyzers are assembled from fluoroplastic tube and fluoroplastic valves and flow distributor. In addition, they have common principle of calibration, performed using "zero" gas and gases released by active capsules of microflows. Pumps, impelling the gas flow and delivering the air to analyzers, are placed after devices. These same pumps constantly purge air through the entire air channel ensuring permanently the presence of "fresh" air at the flow distributor entry.



Fig. 4. Scheme of air line for gas analyzers of the second group installed at BEC.

In contrast to the first group, analyzers of the second group are calibrated using standard mixtures and comparison gases calibrated against them. The scheme of delivery of sample to devices is based on the principle of excessive pressure. Purging of the channels is also performed continuously, and at the entry of the devices a constant pressure is maintained. The CO<sub>2</sub> and CH<sub>4</sub> analyzers are installed in series because gas cylinders with test mixtures simultaneously contain carbon dioxide and methane.

At the "Background" polygon, the transfer of the analyzed samples is organized absolutely in the same way and so the discussion of it is omitted here.

# 3. Automatic calibration of gas analyzers

The devices installed at both sites pass yearly testing at agencies of the Governmental Bureau of Standards. As experience of exploitation of the instruments for monitoring of air gas composition shows, they need frequent calibration. This procedure is performed using test mixtures or capsules with microflow, also tested by agencies of the Governmental Bureau of Standards. Since the list of measured gases is quite long and the test mixtures are available not for all of them, the sites use both types of the calibration, with the test mixtures and capsules of microflows.

The first task is completed using the system of two-position program driven air valves (Fig. 5), which admit air samples and calibration mixtures into the analyzers in four stages: air samples from heights 30 and 10 m and calibration mixtures, namely, test and standard gases. In this case, as an example, we consider carbon monoxide and dioxide. The sample is analyzed air, the standard gas is the gas with known concentration of mixture, and comparison gas is the atmospheric air pumped into the gas cylinder under the pressure on the order of 5 atm with known content of gas component to be determined (determined in the course of calibration using standard gases) and used as calibration gas for more frequent calibration of the gas analyzer.

We have implemented the following algorithm of system operation: calibration against standard gases is made twice a day: from 09:00 to 09:20 we inject low-concentration calibration gas, from 09:20 to 09:40 LT the high-concentration calibration gas, and from 09:40 to 10:00 LT the test gas, with same cycle repeated from 21:00 to 22:00 LT. In the other time, the air sample and test gas are admitted by turn every 20 min (from 10:00 to 10:20 LT we inject sample from height 30 m, from 10:20 to 10:40 LT the sample from 10 m, from 10:40 to 11:00 LT the test gas, etc.). This calibration scheme gives us two advantages: (1) it yields smaller measurement errors because we can rapidly correct for zero drift of the instruments, and (2) it is seriously saves the costly test mixtures. The scheme of site operation is presented in Table 2.

At rest, when no valve is on, all the air channels of the module of reference gases are shut off (see Fig. 5). Thus, in the case of switching off of the power supply, the gas will stay in the gas cylinders.



Fig. 5. Scheme of automatic calibration of the  $\mathrm{CO}_2$  and  $\mathrm{CO}$  gas analyzers.

The pressure in gas cylinders is maintained at the level from 1.5 to 4.5 atm. Refilling the cylinders from 1.5 to 4.5 atm takes 5 to 6 h, therefore, to ensure hourly calibration with the reference gas, two gas cylinders with comparison gas are used in the measurement channel. At the same time, while the first gas cylinder is used as a source of reference gas for hourly calibration, the second gas cylinder is refilled to the level of 4.5 atm. After a certain the least permissible pressure is reached in the first gas cylinder, the system switches them over so that the first gas cylinder is refilled up to the level of 4.5 atm, while the second one is used as a source of reference gas, and so on. As a rule, time of gas consumption during calibration much exceeds the refilling pump time the control system also checks that the switching over takes place immediately before the calibration of the reference gases against standard mixtures.

	09:00 LT		09:20 LT		09:40 LT	
	21:00 LT		21:20 LT		21:40 LT	
Standard gas 1						
Standard gas 2						
Comparison gas 3						
Measurements	XX:00 LT	XX:10 LT				
of gas composition						
of the atmosphere	blow-through	averaging	XX:20 LT			
at height 30 m	of gas analyzers	<b>↑</b>	data logging			
Measurements	[		XX:20 LT	XX:30 LT		
of gas composition			XX.20 L1	AA.50 EI		
of the atmosphere			blow-through	averaging	XX:40 LT	
at height 10 m			of gas analyzers	<b>f</b>	data logging	
Measurements					XX:40 LT	XX:50 LT
of						
gas mixtures	XX+1.0 hr				blow-through	averaging
	data logging				of gas analyzers	1

Table 2. Operation schedule of devices at the sites in the first scheme (CO<sub>2</sub>, CO)

Table 3. Operation schedule of devices at the sites in the second scheme (NO<sub>2</sub>, NO, SO<sub>2</sub>)

Measurements of gas composition of the atmosphere at height 30 m		XX:15 LT data logging		
Measurements of gas composition of the atmosphere at height 10 m		XX:15 LT averaging from 20 min	XX:30 LT data logging	
Delivery of zero gas to gas analyzers			XX:30 LT averaging from 35 min	XX:45 LT data logging
Delivery of calibration gas to gas analyzers	XX + 1:00 hr data logging			XX:45 LT averaging from 55 min

The second task is suggested to be achieved using analogous (see Fig. 5) system (Fig. 6), but with zero gas taken instead of the comparison gas. The standard gas is obtained from the capsule of the microflow, and zero gas is nitrogen.

The system is operated according to the following schedule: the first 15-min interval of every hour we inject the sample from the height 30 m, the second 15-min interval the sample from height 10 m, the third 15-min interval the zero gas, the fourth 15-min interval the standard gas, and so forth. The scheme of the site operation is presented in Table 3.

In addition to the automatic calibration, periodic instrument calibration is to be performed by the operators (1-2 times a month).



Fig. 6. Scheme of automatic calibration of  $SO_2$  and  $NO_2$  gas analyzers.

For this purpose, we have purchased a 667 GR-03M generator-diluent (OPTEK Closed JSC), allowing

us to test the instruments in a wider range of concentrations.

In addition, operators have a GS-024a ozone generator of the same firm, certified by the Governmental Bureau of Standards. We plan to use this generator for calibration of ozonometers and disinfection of supply channels of other analyzers.

In accordance with the manual of manufacturer, the instruments must be yearly tested by the agencies of the Governmental Bureau of Standards of the Russian Federation. This procedure is performed by OPTEK Closed JSC having the authorization certificate of the Governmental Bureau of Standards.

# 4. Arrangement of site operation, and data collection and transfer

Information is collected according to the scheme (Fig. 7), which has been successfully used for already 2 years during operation of the complexes in the regime of monitoring of meteorological quantities and suffered no major modifications during the increase of the number of parameters to be measured because it had sufficient reserve for the bulk of data to be transmitted. After the initial processing and creation of the files describing each measurement, the data are transferred to the center of data acquisition located at the Institute. It is noteworthy that for transferring data from BEC to the Institute we use the optical fiber channel, and from "Background" polygon to the Institute the radiotelephone line.

For query of the sensors, storage, and preliminary processing of the data, we have developed and fabricated special devices and software making it possible to rapidly obtain readouts and control the operation of the control system.

The system of the query of the sensors is based on the PIC16F877 microcontroller. It is worthy to note that we use an original method of switching on the anemometers allowing us to refuse from intermediate AD conversions. Here, we measure time interval between pulses of the main and additional series. The frequency of the pulses of the main series corresponds the speed of rotation of screw-propeller (one pulse per one rotation) and, via the dependence found, it determines the wind speed. The phase shift between pulses of the additional and main series corresponds to the orientation of the anemometer relative to the base and determines the wind direction. Internal timer of the controller fixes the time intervals between the pulses of the main series and time between the pulses of the main and additional series of the eight anemometers, each time renewing these values in the internal memory. Upon the request from a computer, the newest data will be sent.

Through the signal selector, the analog signals from the temperature and humidity sensors are fed to the built-in 10-digit ADC controller, whose reference voltage is determined by the external REF192 precision source.

In addition, this controller traces the temperature inside the container or building, maintaining it at a given level. Upon requests from the computer, the controller drives the state of aspiration of the temperature and humidity sensors, switching it on for the period of blow-through and measurements.

Also, upon request from the computer, it receives current data from all the sensors. Thus, with the help of the computer, it is possible to arrange any measurement schedule. In our case, these are 10minute measurements by polling the sensors every second, and subsequent averaging of the accumulated data and rms calculation. Aspiration is switched on 10 min before every measurement. Each measurement starts at the beginning of each hour (Fig. 8).



Fig. 7. Block-diagram of gas analytic complex.

Standby	Blow-through (10 min)	Measurements (10 min)	Standby
Time	(xx-1):50	xx:00	xx:10

Fig. 8. Time diagram of the measurement cycle.

Initially, the computer and the device polled were separated by large distance (about 800 m), and they communicated via radio channel using radio BCC418 modems (Bluechip Communication AS, Norway, 433 MHz, FSK, 10 mW, RS-232) with directional antenna connected to them. Subsequently, during further increase of the measurement complex (installation of gas analyzers in immediate vicinity of the mast), it was decided to "dispose" the computer closer to the measurement site. Now, the data collecting device is connected to a computer via cable, without the use of a radio channel, and computer in its turn is connected to the central sever of the Institute via optical fiber cable.

An analogous mast with acquisition system is also installed at the "Background" polygon, with the only difference that the communication with the central server is accomplished by modems via radiotelephone line of an automatic telephone station.

On the control computers in Kireevsk and Tomsk, remote-access Dial-Up-servers are installed. Dial-up (remote communication via telephone line) connects the computers to the network environment of laboratory for the time of communication session. Thus, telephone lines and modems are used for access to the common resources of network group.

The server in Tomsk bridges, in automatic mode, communication with the server at the "Background" polygon via modem connection using Point-To-Point protocol (PPP), with a certain frequency, in accordance with the settings. Then, files with the newest data and servicing messages are copied.

In addition, a remote desktop server is installed on the computer at the "Background" polygon, which transfers the server interface to the client computer and communicates typing on the keyboard and mouse clicking from client computer to the server. Operator in Tomsk accomplishes distant access to the server, allowing him/her to service computer at the polygon from his working place. If necessary, the operator can change working parameters of the software of the data acquisition, valve control, etc.

Computer station of the data processing (SDP), located in Tomsk, uses built-in modem to "dial" the number of the telephone at the "Background" polygon, also with computer station of data acquisition (SDA) connected to it (again via a modem). In the case of successful connection, SDP sends a query.

In the monitoring mode we use in the case of undisturbed operation the SDP initiates exchange with SDA once a day (03:00 LT).

The SDA creates a file containing averaged measurement data, notes of the states of valves at the corresponding time interval, and comments. It is worth noting that this file is created taking into M.Yu. Arshinov et al.

consideration the convenience of its further processing (in Origin and Excel).

In the case of the failure of power supply of the computer, the SDA is restarted automatically, and a message is immediately sent to the SDP containing a note of the time of last record and time of turn on of the power supply.

Data for the day are sent to the station of data acquisition, where they are analyzed and prepared for publication in the Internet at http://www.iao.ru/ru/structure/juc/. If necessary, times and frequency of connections can be altered.

In each measurement cycle, a corrected value for each level (10 and 30 m), averaged over last 10 min of measurements is recorded into the file. During this time, about 200 values from gas analyzers are received.

### Conclusion

We have created and put into the monitoring mode of operation two automated complexes for measurement of fluxes of greenhouse gases and atmospheric oxidants, disperse aerosol composition, and meteorological quantities. The complexes are located in urbanized and background regions of Tomsk Region.

For improvement of the metrological characteristics of the instruments used under field conditions and saving consumables, we have developed an original system of their calibration by use of comparison mixtures and calibration capsules of a microflow or a standard gas.

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