MR-32/MGC SETUP FOR MONITORING GAS COMPOSITION OF ATMOSPHERE

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ABSTRACT

A brief description of MR-32/MGC setup for monitoring gas composition of the atmosphere is presented. The complex allows to determine the content of greenhouse gases (methane, carbon dioxide and nitrous oxide) and carbon monoxide in samples of surface air, as well as the total content of listed components in the atmospheric column, including additionally ozone and water vapor. In the first case, IR absorption spectra of sample in a multi-pass cell with optical path length of about 30 m are registered. In the second case, IR spectra of direct solar radiation passed through the entire atmosphere are recorded. Some results of measurements at Obninsk (55°06'38" N, 36°35'54'' E) in 2015 – 2020 are given.

1. INTRODUCTION

The atmospheric greenhouse gases with the most significant impact on the earth's radiation balance include water vapor (H_2O), carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O). Available data show that in the industrial era, from 1750 to 2000, the concentration of CO_2 increased from 280 to 368 ppm, CH_4 from 700 to 1750 ppb, N_2O from 270 to 316 ppb [1]. Over the past 2 decades, the globally averaged concentrations of CO_2, CH_4, N_2O at background monitoring stations continued to grow, reaching the present time 407 ppm, 1857 ppb, and 330 ppb, respectively [2]. At present, monitoring of radiation-active minor gas components (MGC) in the surface layer is carried out at GAW and ESRL NOAA stations. Measurements of the total content of a number of atmospheric gases using Bruker IFS-120/125 HR, EM27/SUN instruments are currently being carried out at the network of stations NDACC, TCCON and recently COCCON. The instruments and methods of gas analysis developed at RPA "Typhoon" and tested over decades [3] led to the concept of the gas analytical complex MR-32/MGC, which is a more modern analogue of the experimental installations previously used for regular and field measurements of water vapor, methane, carbon dioxide, nitrous oxide and carbon monoxide in the atmosphere column and in the surface air.

2. EXPERIMENTAL SETUP

A general view of the MR-32/MGC instrument consisting of an infrared Fourier spectrometer, multi-pass cell, sun-tracking and pumping systems is shown in Figure 1. The complex is designed to monitor trace gases variations in the atmosphere [4] and is used in two modes:

- recording the artificial source (globar) radiation passed through multi-pass cell,

- registration of solar radiation passed through the atmosphere.

To switch from the measuring in multi-pass cell to the measuring of total gas content in the atmospheric column, the position of the movable mirrors (8) is changed. The multi-pass optical cell ensures the absorbing gas layers up to 30 m thick. The technological system is used to pump out and fill the cell with air from a sampler located at a height of 3 m from the earth's surface. In the second mode a tracking system (3) located on the roof of the building scans the position of the Sun and directs a focused solar radiation flux to the input of the spectrometer.

a) b) c)

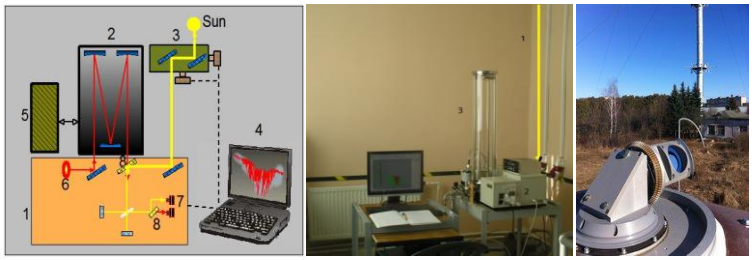

Figure 1. a) - Block diagram of the MR-32/MGC complex. b) - general view, c) - tracking system
1- spectrometer IFS-M, 2- cell, 3- tracker (sun-tracking system), 4- control computer, 5- system for pumping out and filling the cell, 6- IR source-globar, 7- radiation receivers, 8- movable mirrors.

The spectral range of the channel for recording radiation transmitted through the cell is 1800 - 5000 cm^{-1}. The channel for recording solar radiation initially was 1800 - 7700 cm^{-1}, after renovation in 2018 the range was extended to 800-7700 cm^{-1}. The effective spectral width of the instrumental contour at the 50% level is 0.12 - 0.15 cm^{-1}, the limit of the main measurement errors of the intensity of solar radiation and an artificial radiation source is no more than 3.0%. The transmission spectrum of the surface air sample is calculated as ratio of the air sample spectra to the averaged spectrum before and after sampling [5]. For this, 10 baseline measurements are carried out with the evacuated cell, 20 measurements of the air sample and 10 baseline measurements again with the evacuated cell. The temperature inside and outside the cell is recorded simultaneously. The measurement period is about 20 minutes. Two spectral intervals are used to determine the concentrations of CO_2, CH_4, N_2O and CO (see table). To determine the content of water vapor and referencing the spectrum, a single H_2O line was used in the region 3059–3061 cm^{-1}. To assess systematic errors, calibration measurements are carried out with standard mixtures of VNIIM Mendeleev. Gas concentrations are determined by comparing the experimental spectrum of the sample with the spectrum calculated from the HITRAN spectral lines database [6]. The random error in gas concentration found from the scatter of results obtained from repeated analyzes of the same air sample does not exceed 1 ppm for CO_2, 20 ppb for CH_4, 5 ppb for N_2O, and 6 ppb for CO.

Table – Spectral microwindows, cm^{-1}

Gas	Total column	Multi-pass cell
H_2O	4685-4710	3059–3061
CO_2	6173-6390; 6225-6227	2243-2250; 2251-2255
O_3	2126-2129; 2130-2132	-
N_2O	2155-2160; 2525-2535	2209-2211; 2215-2218
CH_4	2920-2922; 5897-6145	2998-3000; 3037-3041
CO	2126-2129; 2155-2160	2144-2150; 2164-2181

The SFIT4 V0.9.4.4 software package and HITRAN database were used as a basis for calculating the total gas content. The priori profiles of temperature, pressure, and gas profiles were borrowed from the Whole Atmosphere Community Climate Model (https://www2.acom.ucar.edu/gcm/waccm). The micro-windows used to determine the total content of CO_2, CH_4, N_2O and CO are shown in the table. Ozone overtone lines in 2126- 2132 cm^{-1} allow to estimate the total ozone content. To assess the contribution to the absorption of water vapor the spectral range of 4680 - 4710 cm^{-1} was chosen, containing a single H_2O line with a frequency of 4699.7507 cm^{-1}. The results of determining water vapor total column were used in calculating carbon dioxide and methane to exclude the effect of water vapor and additionally to control the frequency scale of the spectrometer.

Comparison of the MR-32/MGC data for methane and carbon dioxide with the measurements from the GOSAT satellite, as well as with the data of the nearest European NDACC and TCCON stations, showed that the MR-32/MGC data have a constant systematic excess. Correction factors were introduced to compensate for the constant offset and to be bound to the data of the NDACC and TCCON stations. Correction factors are 0.97 for methane and 0.98 for carbon dioxide.

3. MEASUREMENT RESULTS

The observed global growth of greenhouse gases in the surface layer is also typical for Obninsk. Thus the average annual concentration of CO_2 has increased by 11 ppm, CH_4 by 11 ppm, and N_2O by 7 ppm over the past 5 years. The results of measurements of surface concentrations of methane and carbon dioxide are available on the website (http://www.rpatyphoon.ru/activities/climate-monitoring/LAST/measurements.php). The main results of measurements of greenhouse gases at Obninsk are also contained in the annual summary reports of Rosgidromet on the state of the climate [7].

The column-averaged dry-air mole fractions XCO_2 , XCH_4 and the total column H_2O are compared in Figure 2 with the results of measurements by the GOSAT (The Greenhouse Gases Observing Satellite) and AIRS (Atmospheric Infrared Sounder, precipitable water vapor) satellite instruments. Satellite data with gaps in time and space were averaged for the region (53° - 57°) N and (34° - 38°) E. Data from the Karlsruhe station (49.1° N, 8.44° E) were used to compare the results of measurements of XN_2O. The comparison shows a fairly good agreement between MR-32/MGC data and satellite data and measurements at the nearest European stations for both seasonal variation and long-term variability (trends).

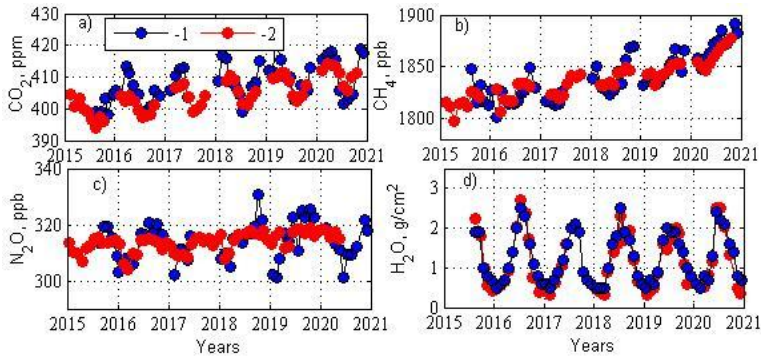

Figure 2. Column-averaged XCO_2 (a), XCH_4 (b), XN_2O (c) and total column H2O (d).
1 - Obninsk, 2 - GOSAT (https://data2.gosat.nies.go.jp), AIRS (https://giovanni.gsfc.nasa.gov) and Karlsruhe station (https://tccondata.org), see text for more details.

Simultaneous observations of greenhouse gases in the surface layer and throughout the entire atmosphere make it possible to assess the influence of local sources. The seasonal variation of greenhouse gases CH_4, CO_2 and N_2O in the surface layer and throughout the entire atmosphere averaged over the observation period from 2015 to 2020 is shown in Figure 3.

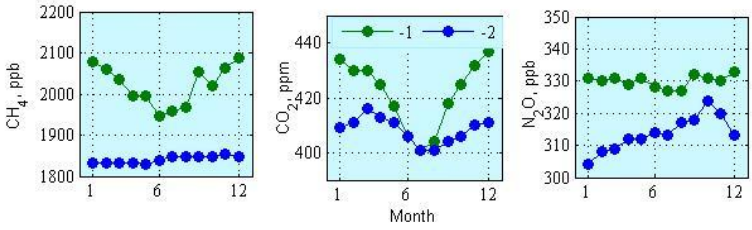

Figure 3. Averaged seasonal variation at surface layer (1) and throughout the entire atmosphere (2)

The greatest excess of surface concentrations over concentrations in the entire atmosphere is observed in winter. If for carbon dioxide the minimum concentrations in summer coincide, then for methane the excess persists for all seasons. Possibly this indicates noticeable surface sources or tropospheric methane sinks. The results of measurements of nitrous oxide at Obninsk are presented for the first time and are preliminary.

4. CONCLUSION

Operation of the complex in 2015-2020 showed the possibility of using the MR-32/MGC and the developed software for monitoring greenhouse and other gases in the surface layer and throughout the entire atmosphere. The possibility of using the 800-1800 cm^{-1} region to determine other trace gases is currently being tested.

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