ISSN 1024-8560, Atmospheric and Oceanic Optics, 2022, Vol. 35, No. 4, pp. 341–344. © Pleiades Publishing, Ltd., 2022.
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SPECTROSCOPY OF AMBIENT MEDIUM

Comparison of Stratospheric CO_2 Measurements by Ground- and Satellite-Based Methods

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Received December 6, 2021; revised December 20, 2021; accepted February 7, 2022

Abstract—We compared the measurements of the CO_2 content in the lower stratosphere (12–18 km altitude layer) using a ground-based Bruker 125HR Fourier-transform spectrometer and ACE satellite measurements in 2009–2019. The analysis of the two types of measurements showed a good agreement between them. On the average, the ground-based CO_2 measurements exceed the satellite data by 2.8 ppm (less than 1%), the standard deviations being ~5.0 ppm. The correlation coefficient between the measurements by the two methods is 0.77. The ground-based and satellite CO_2 measurements show weak seasonal variations, opposite to those in tropospheric CO_2: the CO_2 content in the lower stratosphere is maximal in summer and minimal in winter.

Keywords: CO_2 measurements in the stratosphere, carbon dioxide monitoring, ground-based spectroscopic measurements, satellite measurements, Bruker 125HR, ACE-FTS

DOI: 10.1134/S1024856022040145

INTRODUCTION

Current changes in the Earth's climate caused by the growth of greenhouse gases and, primarily, carbon dioxide [1] stimulate permanent monitoring of the CO_2 content in the atmosphere by different methods [2]. At present, ground-based local measurements of the near-surface CO_2 concentrations and different remote measurements of the total CO_2 content are actively used for monitoring. Among the remote methods, measurements of the atmospheric transparency in different spectral regions are most often used, as well as the scattering method. In recent years, different satellite methods are increasingly actively used, as well as the methods of transparency and reflection of solar radiation in the near-infrared (NIR) and thermal infrared (IR) regions, making it possible to acquire the global data on the total CO_2 content and certain information concerning the vertical CO_2 distribution [3].

The high-resolution ground-based spectroscopic measurements of solar IR spectra make it possible to obtain certain information on the vertical structure of the CO_2 content [4]. For instance, the authors of works [5–7] analyze a possibility of retrieving the CO_2 content in the troposphere and stratosphere. An important role in studying the vertical structure of CO_2 and calibrating the ground-based spectroscopic measurements is played by aircraft and balloon measurements (see, e.g., [8, 9]).

Carbon dioxide measurements in the stratosphere are important for studying the troposphere-stratosphere exchange, determining the CO_2 lifetime in the atmosphere, and validating numerical atmospheric models. The validation and intercalibration of different methods and instruments make it possible to create high-accuracy databases on different spatiotemporal scales used in climatologic studies [10].

In this work, we compared CO_2 content in the lower stratosphere retrieved from the spectra of IR solar radiation measured by a ground-based high-resolution Bluker 125HR Fourier-transform spectrometer and by the Atmospheric Chemistry Experiment Fourier-transform spectrometer (ACE-FTS) onboard a satellite.

METHODS FOR MEASUREMENTS OF CO_2 CONTENT IN THE STRATOSPHERE

The ground-based spectroscopic measurements of the CO_2 total content (TC) have been used at stations of the International Network for the Detection of Atmospheric Composition Change (NDACC, in the mid-IR spectral region) and Total Carbon Column Observing Network (TCCON, in near-IR region) for quite long time. The CO_2 measurements have been initiated at St. Petersburg State University (SPSU) in 2009; and since 2016, the SPSU station has officially been a part of the NDACC international network [11].

The authors of work [7] suggested a technique for interpreting the measurements, making it possible to determine not only the CO_2 TC, but also the CO_2 content in the troposphere and stratosphere. The problem of optimizing the retrieval of average carbon dioxide mixing ratio (XCO_2) from Bruker 125HR Fourier-transform spectrometer measurements was solved based on the PROFFIT software [12], used at a number of NDACC stations. The spectral scheme of CO_2 retrieval, suggested in [13] for selecting quality spectral measurements at NDACC stations was taken as a basis for the method for the inverse problem solution, which was optimized for the CO_2 TC determination at the St. Petersburg station in [14, 15]. In the present work, this method was modified for the XCO_2 retrieval in two atmospheric layers: "the conventional troposphere" (0–12 km) and "the conventional stratosphere" (12–55 km). In contrast to the above-mentioned works, which were aimed at determining the total carbon dioxide content and where the inverse problem was solved through profile scaling, we used the Tikhonov–Phillips regularization to determine XCO_2 in two layers.

All the spectra measured by the Bruker 125HR Fourier-transform spectrometer from 2009 to 2019 were analyzed using PROFFIT. The results were filtered according to the following criteria:

— The residual discrepancy in the spectral channels is less than 0.15, which approximately corresponds to a signal-to-noise ratio of ~700.

— The number of degrees of freedom for CO_2 in the spectral channels of the measurements is larger than 2.

— Individual XCO_2 measurements throughout atmospheric depth during a single day differ by no more than 2% of the daily average measurements, making it possible to sort out the bursts in the results caused by high measurement errors.

We note that the average random error of the XCO_2 retrieval in the stratosphere over the measurement ensemble is ~2 ppm ($0.41 \pm 0.06\%$), markedly less than the natural CO_2 variations. The authors of work [15] considered in detail the sources of errors in XCO_2 retrieval.

Satellite measurements of the gas composition of the atmosphere by the transparency method were first carried out as early as 1983 using the GRILLE-1 instrumentation. This method was then repeatedly used [4]. These measurements have been performed onboard the ACE satellite by the ACE-FTS spectrometer for more than 15 years [16]. The ACE-FTS measures the spectra of solar radiation in the region 2.2–13.3 μm (750–4400 cm^{-1}) with a resolution of 0.02 cm^{-1}. Special methods and algorithms for processing measurements were developed to determine the vertical profiles of the CO_2 content, in particular, in the stratosphere and troposphere [17]. For this work, we took the ACE-FTS satellite data available at [18]; the version 4.1/4.2 data were used [19].

To compare the two methods of measurements, we selected satellite (for the layer of 12–18 km) and ground-based data obtained on the same day at points spatially separated by no more than 500 km. Over 2009–2019, there were just 46 such measurements. This is primarily because ACE-FTS measures relatively rarely (~28 measurements per day) and has no spatial (horizontal) scanning; and ground-based measurements were only carried out under clear-sky conditions. The ACE-FTS measurement errors in the lower stratosphere (layer of 12–18 km) are ~4 ppm (~1.0%).

COMPARISON OF THE METHODS

Below, we present the statistical characteristics obtained from comparison of ACE-FTS satellite measurements (500 km) and ground-based Bruker 125HR Fourier-transform spectrometer measurements of the CO_2 content in the atmospheric layer of 12–18 km for 46 coinciding days: the standard deviation σ reaches 5.06 ppm, the root-mean-square discrepancy S is 5.72 ppm, the average discrepancy M is 2.78 ppm, with ground-based measurements exceeding the satellite ones. The correlation coefficient $R = 0.77 \pm 0.10$, indicating that the quantities compared are closely interrelated.

Figure 1 presents the stratospheric CO_2 content retrieved from ground-based and satellite measurements during 46 selected days considered here. The determination coefficients R^2 are 0.74 for ground-based Bruker 125HR measurements and 0.67 for ACE-FTS measurements. It can be seen that the trend line for the ground-based data overlies that for satellite ones;

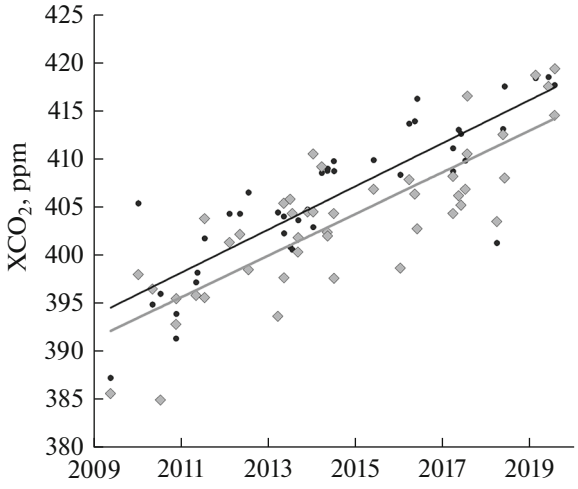

Fig. 1. The XCO_2 variations in the stratosphere (>12 km) derived from ground-based Bruker 125HR Fourier-transform spectrometer (circles) and ACE-FTS satellite (rhombs) measurements over 2009–2019 within the 500-km radius of St. Petersburg station; straight lines indicate a linear fit for each dataset.

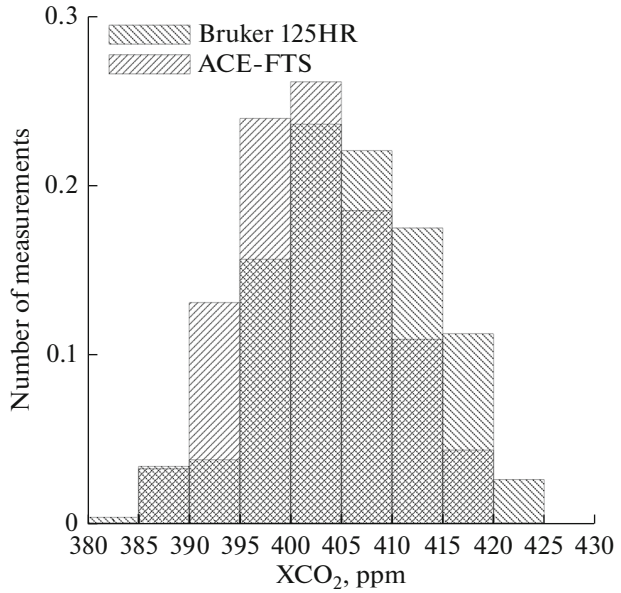

Fig. 2. Comparison of frequency histograms of the stratospheric CO_2 content from ground-based (Bruker 125HR) and satellite (ACE-FTS) measurements.

therefore, the ground-based data show a stronger and steadier CO_2 growth.

We note that satellite and ground-based measurements demonstrate weak seasonal variations in the stratospheric CO_2 content differing from variations in the total and tropospheric CO_2. In the lower stratosphere, the maximal values are observed in summer and minimal values in winter. These (opposite to tropospheric) seasonal variations in the lower stratosphere were also reported in [7, 20].

Figure 2 presents the frequency histograms of the CO_2 measurements in the lower stratosphere by the ground-based Bruker 125HR spectrometer and satellite ACE-FTS. It can be seen that the satellite-based (ground-based) CO_2 concentrations are most numerous in the range from 395 to 405 ppm (from 405 to 415 ppm).

Thus, the CO_2 content retrieved from the ground-based Bruker 125HR measurements exceed the values retrieved from the ACE-FTS measurements. However, despite these differences, the distributions of the numbers of measurements for different CO_2 ranges for both methods are similar in shape, making it possible to conclude that the CO_2 values in the lower atmosphere derived by the two methods are close in value.

CONCLUSIONS

We compared the satellite (ACE-FTS Fourier-transform spectrometer) and ground-based (Bruker 125HR Fourier-transform spectrometer) measurements of the CO_2 content in the lower stratosphere (layer of 12–18 km) carried out on the same days in the regions spatially separated by no more than 500 km (46 measurements over 2009–2019). These two types of measurements agree reasonably well. On the average, the ground-based measurements exceed the satellite values by 2.8 ppm (less than 1%), the standard deviations being ~5.0 ppm. The correlation coefficient between the two measurements is 0.77. Data from the two types of measurements showed weak seasonal variations in the stratospheric CO_2 content.

Validation and cross-calibration of different methods and instruments used for measurements play an important role in studying the spatiotemporal variations in the CO_2 content in the atmosphere. The information obtained from the comparison and, in particular, the information concerning the errors of measurements by different instruments makes it possible to analyze the variations in the CO_2 content, study the intensity of anthropogenic CO_2 emissions, and to create high-accuracy databases.

FUNDING

This work was supported by the Russian Foundation for Basic Research (grant no. 20-05-00627). The experimental data were obtained using the instrumentation from the SPSU resource center "Geomodel." The ACE satellite measurements are funded by the Canadian Space Agency.

CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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Translated by O. Bazhenov