ISSN 1024-8560, Atmospheric and Oceanic Optics, 2017, Vol. 30, No. 3, pp. 263–268. © Pleiades Publishing, Ltd., 2017.
Original Russian Text © Yu.M. Timofeyev, Ya.A. Virolainen, S.P. Smyshlyaev, M.A. Motsakov, 2017, published in Optika Atmosfery i Okeana.

ATMOSPHERIC RADIATION, OPTICAL WEATHER, AND CLIMATE

Ozone over St. Petersburg: Comparison of Experimental Data and Numerical Simulation

Yu. M. Timofeyev[a], *, **Ya. A. Virolainen**[a], **S. P. Smyshlyaev**[b], **and M. A. Motsakov**[b]
[a]*St. Petersburg State University, St. Petersburg, 199034 Russia*
[b]*Russian State Hydrometeorological University, St. Petersburg, 195196 Russia*
**e-mail: y.timofeev@spbu.ru*

Received March 16, 2016

Abstract—Comparison of numerically simulated ozone fields with various experimental data makes it possible to evaluate the quality of models for their use in reliable prediction of variations of the ozone layer. We compared the satellite (SBUV) and ground-based (IFS Bruker 125HR) measurements of ozone content in two atmospheric layers (0–25 and 25–60 km) with the numerical simulation data (obtained using the low- and middle-atmosphere models) over St. Petersburg for the period between 2011 and 2014, namely, the daily and monthly average values of ozone content for 3.5 years (June 2011—December 2014). In general, model describes the experimental ozone content with good or satisfactory accuracy in the two layers. Nevertheless, some systematic differences are found out between the satellite and ground-based data and the results of simulation. In the autumn–winter period, the model usually overestimates the ozone column in the 0–25 km layer as compared to the satellite measurements, and underestimates it in the 25–60 km layer. The same features are observed for daily and monthly average values. In some cases, the model shows strong and high-frequency oscillations in the ozone content, which are not observed in the measurements.

Keywords: atmospheric ozone, atmospheric models, remote sensing methods

DOI: 10.1134/S1024856017030149

INTRODUCTION

Comparisons of simulated spatial-temporal variations in ozone content with various measurements allow evaluating the quality of a model and its correction, which enables the use of the model for reliable forecasts of changes in the ozonosphere under different scenarios of anthropogenic and natural impacts. Similar comparisons with measurements of the total ozone content (TOC) were carried out repeatedly (see, e.g., [1–7]). Consideration of only TOC data does not allow revealing all characteristics of altitude ozone variability, as well as objectively identifying the photochemical and dynamic factors that define the characteristics of the observed seasonal and interannual variability. Comparison of individual atmospheric layers is of higher information content, because different processes of ozone formation and transformation prevail at different altitudes.

In this paper, we analyze the results of comparison of the measured and simulated data on ozone content in two atmospheric layers: 0–25 and 25–60 km. These comparisons were made for both daily average values (which is important, for example, for predicting the appearance of ozone miniholes and increase in the UV illumination of the surface) and monthly average values of ozone content (which is topical for climatological studies).

MEASUREMENTS

We analyze two types of ozone measurements: satellite and terrestrial. The satellite measurement data are taken from the Solar Backscatter Ultraviolet Instrument (SBUV) that allows measuring both TOC and ozone content in various altitudinal layers. Instruments, such as SBUV, measure the spectra of scattered and reflected solar radiation in the UV region [8]. Descriptions of the device and algorithms for interpretation and analysis of the measurement errors are given, for example, in [9]. Ten SBUV instruments on various satellites have been used since 1970 to monitor the ozonosphere, and the instrument calibration and interpretation methods are well developed. The SBUV instruments perform nadir measurements of spectra in the 250–340 nm region with a horizontal resolution of about 200 × 200 km. The accuracy of satellite measurements has been examined in many works (see, for example, [9, 10]), in which it was shown that the errors in determining the ozone content in thick layers do not significantly depend on the quality of a priori information, and amount to 5–15%.

Ground-based ozone measurements were carried out at the Peterhof station near St. Petersburg (59.88° N, 29.82° E) using a Bruker 125HR Fourier spectrometer (FS) [11]. These measurements are based on interpretation of high resolution direct solar radiation IR spectra. The TOC and elements of the vertical profile of ozone (the content in separate layers) were determined from the measurements in five micro-windows in the interval 991–1014 cm^{-1} (see [12] for more details). Comparisons of ground-based measurements with various independent ones, including satellite measurements, are given in [13, 14]. It is shown that ground-based and satellite measurements of ozone content in different atmospheric layers are consistent within 5–17%.

The spatial and temporal requirements for selection of the measurement data were rather soft in order to obtain sufficiently representative ensembles for comparisons. Ground-based IR measurements were carried out in the presence of direct solar radiation (cloudless atmosphere or large discontinuities in the clouds). Since the satellite measurements over St. Petersburg took place at 10:00–11:00 GMT, we took average data from 8 to 14 h for ground-based measurements. The satellite SBUV measurements of the MOD (Merged Ozone Data) used in the comparisons are integrated and interpolated to the coordinates of the Voyeikovo station (59.97° N, 30.30° E).

THE MODEL

Numerical experiments were carried out with the global model of composition of the lower and middle atmosphere (MCLMA) to study the altitudinal and temporal features of ozone variability. The model was created on the basis of the chemical and climatic model of the Institute of Numerical Mathematics, Russian Academy of Sciences, and the Russian State Hydrometeorological University [15]. It was previously successfully used to solve problems related to the interaction of physical and chemical processes in the atmosphere [16–18].

In contrast to the chemical and climatic model, the wind, temperature, humidity, and pressure fields in the MCLMA are not calculated, but are set according to the reanalysis data. In the present study, we used the MERRA (Modern-Era Retrospective analysis for Research and Applications) data [19]. The MCLMA covers the altitude range 0–60 km, and takes into account the variability of 74 trace atmospheric gases interacting in 174 chemical reactions and 46 photodissociation reactions. The use of reanalysis data [19], which present, by means of the hydrodynamic model, a correlated account of a large number of ground-based and satellite measurements, makes it possible to simulate the response of ozone content to the actually observed variability of the dynamic characteristics of the atmosphere.

Even though the subject of this study is the local variability of ozone, a long period of the analysis requires consideration of the processes that are close to the global scale. In this connection, the global version of the MCLMA was used for the analysis presented in this paper, which allowed us to consider the influence of global scale processes on ozone variability in the vicinity of St. Petersburg. However, due to rather coarse horizontal resolution of the global model (about 300–400 km for the region under study) this led to a substantial averaging of the simulation results. Meanwhile, the latter circumstance, as an additional result, helps to estimate how local ground-based measurements correspond to ozone variability that is typical for the region as a whole, especially since satellite SBUV measurements are also tied to the other point in the region under study.

Numerical experiments used in this paper relate to 2011–2014. Changes in the flows of gases from the underlying surface, aerosol content, ocean surface temperature, and ice coverage were determined based on the scenarios of the World Meteorological Organization, which are used to estimate changes in the Earth's ozone layer [20]. In this paper, we analyzed and compared daily average values of ozone in two atmospheric layers: 0–25 and 25–60 km.

COMPARISON OF SIMULATED AND MEASURED DATA

When comparing simulated and measured data, it is important to establish criteria of good, satisfactory, and unsatisfactory agreement. To some extent, these criteria depend on the error of measurements, the spatial-temporal variations of ozone fields, and differences arising from the spatial and temporal mismatch between different data. We assume that for the lower layer with an ozone content of ~200–300 DU, the good, satisfactory, and unsatisfactory agreement between model and measurements corresponds to 5, 10, and ≥ 15% (10–15, 20–30, and ≥ 30–45 DU), respectively. For the higher layer, with ozone content of ~100 DU, the corresponding values are 5, 10, and ≥ 15 DU.

For the layer 0–25 km, an unsatisfactory agreement between measurements and modeling is observed in the autumn–winter–spring period, in particular, November 2011–March 2012, January–March 2013, and December 2013–March 2014, December 2014. In these cases, simulation demonstrates, on average, higher values of ozone concentration than measurements. Beyond these periods, an agreement between measured and simulated values is good and satisfactory; as a rule, the error does not exceed 20–30 DU. It may be noted that in some periods, the general trends in variation in ozone content, including its short-term variations, agree very well. At the same time, significant deviations (up to 80 DU) are observed sometimes between the satellite measurements and modeling. However, additional analysis has shown that this is

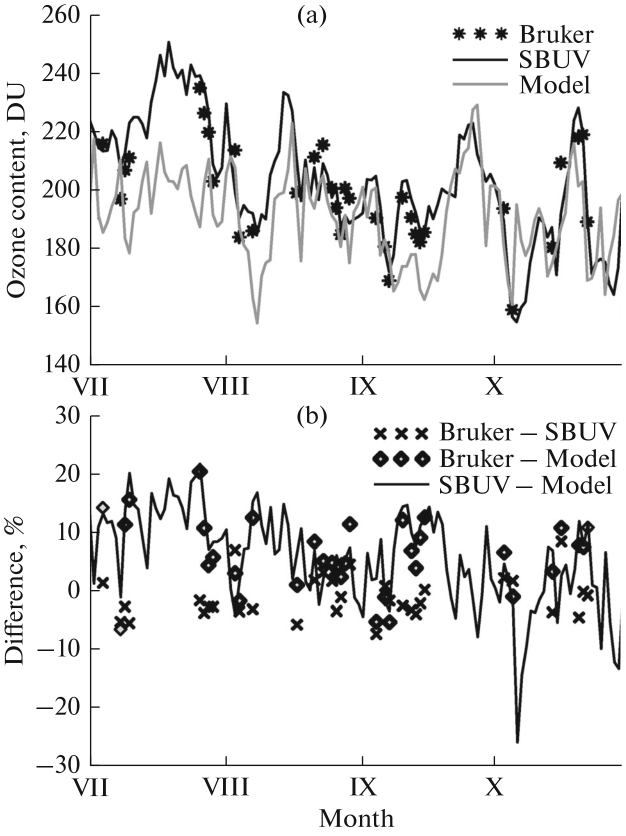

Fig. 1. (a) Daily average values of ozone content in the 0–25 km layer derived from measured and simulated data; (b) their difference.

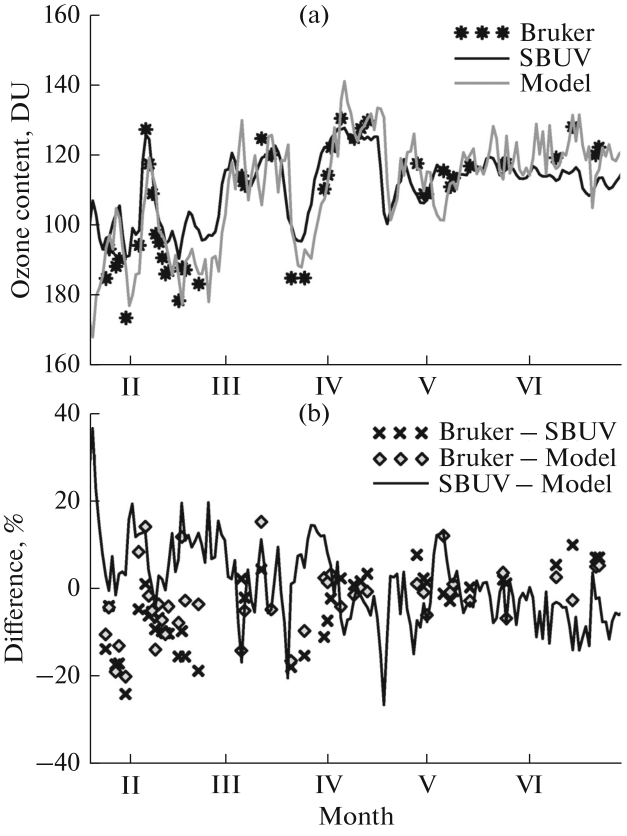

Fig. 2. (a) Daily average values of ozone content in the 25–60 km layer derived from measured and simulated data; (b) their difference.

caused by spatial and temporal mismatch of the data, which is confirmed by measurements performed with the other satellite instruments.

In some cases, there are significant differences between ground-based and satellite measurements, which may also indicate the impact of spatial-temporal mismatch between different data. In some periods, there are high-frequency variations in ozone content according to the both model data and, partly, satellite measurements. Some maxima and minima are well reproduced by different data, but not in all cases. Figure 1 shows an example of significant disagreement between simulation and measurements with the relative difference reaching 20–25% in the layer 0–25 km (July–October 2013). At the same time, the satellite and ground-based measurements agree fairly well with each other (within 7.5%).

For the 25–60 km layer, the differences between experimental and model values of ozone content are usually 5–15 DU. In most cases, all data describe well trends in changes in ozone content and, in many cases, the short-term variation of ozone. Unsatisfactory agreement was observed in the following periods of comparison: November 2011–January 2012, November–December 2012, December 2013–March 2014, and December 2014. As a rule, the model underestimates the ozone content in this layer within these periods. In the summer months (for example, June–July 2012), the model predicts larger values of ozone than the measured data. The difference between ground-based and satellite measurements is sometimes negative and other times positive. The measured amplitudes of ozone changes in the considered layer are often smaller than the results of simulation. The amplitude of high-frequency ozone variations reaches 20–25 DU in the model, which is not always observed in the measurements. Figure 2 shows one of the cases (February–June 2012) where the simulated values of ozone content in the 25–60 km layer are in a better agreement with the ground-based measurements than with the satellite data. This can also be due to the spatial inhomogeneity of the ozone field, since the ground-based and satellite measurements are separated in space, and the simulated data are horizontally averaged.

ANALYSIS OF THE RESULTS

Comparisons of ozone content according to the simulation and measurements (satellite and ground-based methods) have shown overall good and satisfactory agreement for the temporal changes and ozone

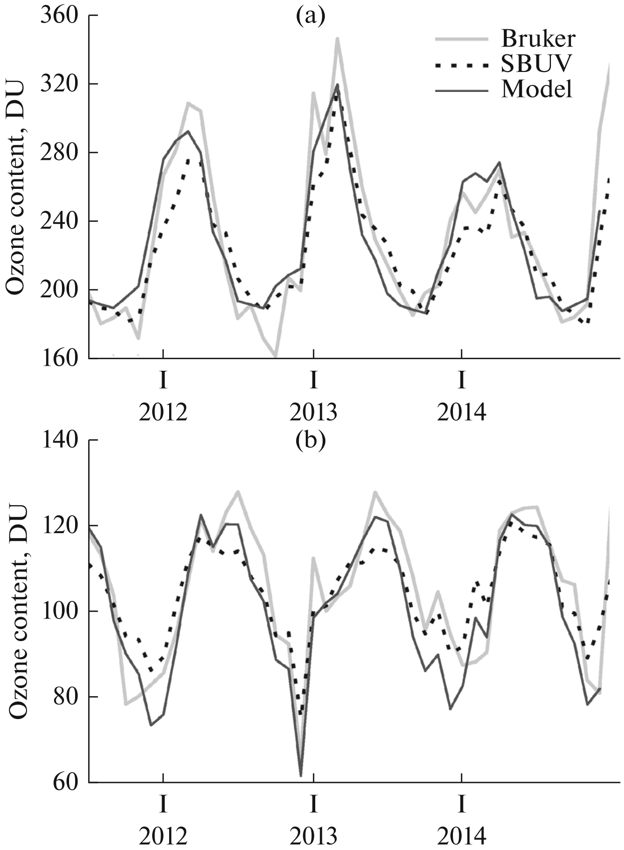

Fig. 3. Monthly average values of ozone content derived from the measured and simulated data in the layer (a) 0–25 km and (b) 25–60 km.

variations in the layers 0–25 and 25–60 km, except for certain periods. However, there are periods of well-pronounced systematic disagreements between satellite and model ozone values. A certain role in the detected significant differences can be played by rather soft selection criteria used for spatial-temporal adjustment of satellite and model data.

Compared to the satellite measurements, the model usually overestimates the values of ozone content in the layer 0–25 km in the autumn–winter period; it was observed in five out of the seven half-years considered. At the same time, sometimes, the model underestimates the ozone content in this layer, for example, in the summer of 2013 and 2014. In the 25–60 km layer, the model often underestimates the ozone content in the autumn and winter periods: it took place in six out of the seven half-years considered. Thus, in the autumn–winter period, the model overestimates ozone content in the 0–25 km layer and underestimates it in the layer 25–60 km. This is a clear example of the advantages of studying ozone in different atmospheric layers, as this effect is difficult to detect in the ozone column observations.

In some cases, the model provides strong and high-frequency ozone oscillations. Sometimes, they are recorded in satellite measurements, but not always. The cases of bad model description of satellite-observed ozone variations are as follows: for the 0–25 km layer, it was in November 2011, January 2012, May 2013, July 2013, January–April 2014, and November–December 2014; for the layer 25–60 km, it was in July 2012, October 2012, January–February 2013, August–September 2013, December 2013, and September 2014.

The above systematic differences are evident when comparing monthly average ozone contents in the layers studied (Fig. 3). It can be seen that the model overestimates considerably the satellite data for the layer 0–25 km in the winter months, and the situation is opposite for the layer 25–60 km. Furthermore, the model usually shows greater seasonal variability of ozone content compared to the satellite measurements. Due to peculiarity of ground-based measurements (only in sunny weather) there may be only a few days in some months when the data are available (usually in the autumn and winter). That is why the monthly average values show greater variability than the rest of the data in this period, and do not fully describe the seasonal behavior of the ozone cycle. At the same time, in the spring and summer months, the number of measurement days is statistically significant for obtaining the monthly average values; that is why the results in these periods are similar to the data from the other ensembles.

Let us consider some statistical parameters of the comparisons made. Table 1 shows characteristics of comparison of different ozone measurements with each other and with the results of simulation for the layer 0–25 km.

All three ensembles are consistent in variability of ozone in the layer (about 20%). Bruker FS overestimates the average ozone concentration by 4% (10 DU) as compared to the SBUV, and by 1.7% (4 DU) as compared to the model. The standard deviation from the average mismatch between the two types of measurement lies within measurement accuracy of each method. There is a very high correlation coefficient between the two types of measurements (0.954). Disagreements exceed the level that is normally reached in the programs of validation of satellites and ground-based measurements (see, e.g., [12–14]), but it should be taken into account that rather soft requirements for spatial-temporal agreement between different data were used to form ensembles of comparison and analysis.

On average, there is a very good agreement between satellite measurements (SBUV) and results of simulation. The model overestimates the satellite measurements by 2% (4 DU); the standard deviation (10.6%) is not much larger than the standard deviation (7.3%) between two types of measurements. There is about the same agreement between the results of ground-based measurements and simulation, but the correlation coefficient between them is slightly higher (0.90 vs. 0.85).

Table 1. Statistical characteristics for comparison of ensembles of ozone content measured in the 0–25 km layer (lower atmosphere) using Bruker FS (St. Petersburg State University) and SBUV, as well as the simulated data

Gauge	Number of comparisons	Average content, DU	Variability, %	Difference, %	Correlation coefficient *R*
Bruker SBUV	291	243 233	23 20	3.9 ± 7.3	0.954 ± 0.005
SBUV Model	2191	229 233	18 19	−1.9 ± 10.6	0.847 ± 0.006
Bruker Model	263	240 236	23 22	1.7 ± 10.0	0.90 ± 0.01

Table 2. Statistical parameters for comparison of ensembles of ozone content measured in the 25–60 km layer (lower atmosphere) using Bruker FS (St. Petersburg State University) and SBUV, as well as the simulated data

Gauge	Number of comparisons	Average content, DU	Variability, %	Difference, %	Correlation coefficient *R*
Bruker SBUV	291	110 108	14 10	2.1 ± 7.0	0.89 ± 0.01
SBUV Model	2191	104 101	12 18	2.3 ± 8.6	0.911 ± 0.004
Bruker Model	263	109 106	14 15	2.4 ± 7.7	0.86 ± 0.02

Table 2 shows similar characteristics of comparison of different ensembles, but for the 25–60 km layer. The analysis of the data in Table 2 allows drawing about the same conclusions as the analysis of Table 1. The Bruker FS demonstrates somewhat greater ozone variability compared to satellite observations, which may be due to the less accurate ground-based ozone measurements in the layer in question compared to the satellite measurements. Similarly, the simulated ozone contents also exhibit greater variability than the SBUV data. For all comparisons, systematic mismatches are small and standard deviations are approximately the same. The maximum correlation coefficient is observed when comparing the results of simulation and satellite measurements (0.911); in other comparisons, it is also close to 0.9.

In [5], ground-based measurements of TOC were compared with the results of simulation (EMAS model [21]). It was shown that in almost all cases the simulated TOC values exceed the measured data. The disagreement is especially significant in the winter–early-spring period. In addition, the model predicts a greater spread of TOC values than ground-based measurements. The conclusions of [5] basically coincide with the conclusions of our studies.

CONCLUSIONS

Satellite and ground-based measurements of ozone content in two atmospheric layers (0–25 and 25–60 km) were compared with numerically simulated data for St. Petersburg using the MCLMA. The model was created on the basis of the chemical and climatic model of the Institute of Numerical Mathematics, Russian Academy of Sciences, and the Russian State Hydrometeorological University. The daily and monthly averaged values of ozone content were compared for 3.5 years (June 2011–December 2014).

1. In general, there is good or satisfactory agreement between the experimental and model values of ozone content in the two layers. Nevertheless, systematic differences between satellite and terrestrial data have been found from the results of simulation. Most often, the model overestimates the ozone content in the 0–25 km layer during the autumn–winter period. An inverse picture is sometimes observed in the same layer: the model underestimates ozone (for example, in the summer 2013 and 2014). In the layer 25–60 km, the model very often underestimates ozone in the autumn–winter period.

2. The described systematic differences are clearly observed when comparing the monthly average ozone contents in the layers under study. In the autumn–winter period, the model overestimates ozone in the layer 0–25 km and underestimates it in the 25–60 km layer as compared to the satellite measurements.

3. Sometimes, the model demonstrates strong and high-frequency ozone fluctuations in different seasons, which are not recorded in satellite measurements.

4. The statistical characteristics of the comparisons show a good agreement between the measured and numerically simulated ozone values in the two atmospheric layers. Ground-based measurements exceed slightly both satellite and model data in both layers, while the model data, on average, are slightly higher

than satellite data in the 0–25 km layer and lower in the 25–60 km layer. The ground-based and satellite measurements of ozone are statistically more consistent: the highest correlations ($R = 0.95$) are observed in the 0–25 km layer, while the minimum values of the standard deviation (7%) occurred in the 25–60 km layer. For the remaining pairs of comparisons, the correlation coefficient is also close to 0.9, and the standard deviation does not exceed of 7–11%.

5. The conclusions of this paper coincide basically with the conclusions of earlier studies of TOC [5], where another three-dimensional model (EMAS) also overestimated TOC values as compared to the measurement results, especially in the winter period.

ACKNOWLEDGMENTS

The work was carried out at St. Petersburg State University. Experimental studies were performed using the experimental facility of the "Geomodel" Research Center at St. Petersburg State University. The analysis of the data was performed under financial support of the Russian Science Foundation (grant no. 14-17-00096).

REFERENCES

1. W. M. F. Wauben, J. P. F. Fortuin, P. F. J. van Velthoven, and H. M. Kelder, "Comparison of modeled ozone distributions with sonde and satellite observations," J. Geophys. Res., D **103** (3), 3511–3530 (1998).
2. *Scientific Assessment of Ozone Depletion: 2002. Global Ozone Research and Monitoring Project Rep. N 47* (WMO, Geneva, 2003).
3. *Scientific Assessment of Ozone Depletion: 2006. Global Ozone Research and Monitoring Project Rep. N 50* (WMO, Geneva, 2007).
4. T. Egorova, E. Rozanov, V. Zubov, E. Manzini, W. Schmutz, and T. Peter, "Chemistry-climate model SOCOL: A validation of the present-day climatology," Atmos. Chem. Phys. **5** (6), 1557–1576 (2005).
5. Ya. A. Virolainen, Yu. M. Timofeev, A. V. Polyakov, D. V. Ionov, O. Kirner, A. V. Poberovskii, and Kh. Imkhasin, "Comparing data obtained from ground-based measurements of the total contents of O_3, HNO_3, HCl, and NO_2 and from their numerical simulation," Izv., Atmos. Ocean. Phys. **52** (1), 57–65 (2016).
6. D. Pendlebury, D. Plummer, J. Scinocca, P. Sheese, K. Strong, K. Walker, and D. Degenstein, "Comparison of the CMAM30 data set with ACE-FTS and OSIRIS: Polar regions," Atmos. Chem. Phys. **15** (21), 12465–12485 (2015).
7. V. Eyring, N. R. P. Harris, M. Rex, T. G. Shepherd, D. W. Fahey, G. T. Amanatidis, J. Austin, M. P. Chipperfield, M. Dameris, P. M. Forster, A. Gettelman, H. F. Graf, T. Nagashima, P. A. Newman, S. Pawson, M. J. Prather, J. A. Pyle, R. J. Salawitch, B. D. Santer, and D. W. Waugh, "A strategy for process-oriented validation of coupled chemistry-climate models," Bull. Amer. Meteorol. Soc. **86** (8), 1117–1133 (2005).
8. P. K. Bhartia, R. D. McPeters, C. L. Mateer, L. E. Flynn, and C. G. Wellemeyer, "Algorithm for the estimation of vertical profiles from the backscattered ultraviolet technique," J. Geophys. Res., D **101** (13), 18793–18806 (1996).
9. P. K. Bhartia, R. D. McPeters, L. E. Flynn, S. Taylor, N. A. Kramarova, S. Frith, B. Fisher, and M. MeLand, "Solar backscatter UV (SBUV) total ozone and profile algorithm," Atmos. Measur. Technol, No. 6, 2533–2548 (2013).
10. P. K. Bhartia, R. D. McPeters, C. L. Mateer, L. E. Flynn, and C. Wellemeye, "Algorithm for the estimation of vertical ozone profiles from the backscattered ultraviolet technique," J. Geoph. Res., D **101** (13), 18793–18806 (1996).
11. A. V. Poberovskii, "High-resolution ground measurements of the IR spectra of solar radiation," Atmos. Ocean. Opt. **23** (2), 161–164 (2010).
12. Ya. A. Virolainen, Yu. M. Timofeev, A. V. Poberovskii, M. Eremenko, and G. Dyufor, "Evaluation of ozone content in different atmospheric layers using ground-based Fourier transform spectrometry," Izv., Atmos. Ocean. Phys. **51** (2), 167–176 (2015).
13. Ya. A. Virolainen, Yu. M. Timofeyev, and A. V. Poberovsky, "Intercomparison of satellite and ground-based ozone total column measurements," Izv., Atmos. Ocean. Phys. **49** (9), 993–1001 (2013).
14. Y. Virolainen, Y. Timofeyev, A. Polyakov, D. Ionov, and A. Poberovsky, "Intercomparison of satellite and ground-based measurements of ozone, NO_2, HF, and HCl near Saint Petersburg, Russia," Int. J. Remote Sens. **35** (15), 5677–5697 (2014).
15. V. Ya. Galin, S. P. Smyshlyaev, and E. M. Volodin, "Combined chemistry-climate model of the atmosphere," Izv., Atmos. Ocean. Phys. **43** (4), 399–412 (2007).
16. S. P. Smyshlyaev, V. Ya. Galin, P. A. Zimenko, and A. P. Kudryavtsev, "A model study of the atmospheric ozone sensitivity to solar flux spectral variations caused by solar activity," Rus. Meteorol. Hydrol., No. 8, 17–25 (2005).
17. S. P. Smyshlyaev, V. Ya. Galin, E. M. Atlaskin, and P. A. Blakitnaya, "Simulation of the indirect impact that the 11-year solar cycle has on the gas composition of the atmosphere," Izv., Atmos. Ocean. Phys. **46** (5), 623–634 (2010).
18. S. P. Smyshlyaev, E. A. Mareev, and V. Ya. Galin, "Simulation of the impact of thunderstorm activity on atmospheric gas composition," Izv., Atmos. Ocean. Phys. **46** (4), 451–467 (2010)
19. M. M. Rienecker, M. J. Suarez, R. Gelaro, R. Todling, J. Bacmeister, E. Liu, M. G. Bosilovich, S. D. Schubert, L. Takacs, G.-K. Kim, S. Bloom, J. Chen, D. Collins, A. Conaty, and A. Silva, "MERRA: NASA'S Modern-Era Retrospective Analysis for research and applications," J. Clim. **24** (14), 3624–3648 (2011).
20. *Scientific Assessment of Ozone Depletion: 2010. Global Ozone Research and Monitoring Project Rep. N 50* (WMO, Geneva, 2011).
21. J. Buchholz, L. Ganzeveld, P. Hoor, A. Kerkweg, M. G. Lawrence, R. Sander, B. Steil, G. Stiller, M. Tanarhte, D. Taraborrelli, J. van Aardenne, and J. Lelieveld, "The atmospheric chemistry general circulation model ECHAM5/MESSy1: Consistent simulation of ozone from the surface to the mesosphere," Atm. Chem. Phys **6** (12), 5067–5104 (2006).

Translated by I. Ptashnik