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OPTICAL INSTRUMENTATION

Empirical Assessment of Errors in Total Ozone Measurements with Different Instruments and Methods

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Abstract—Knowledge of measurement errors is one of the most important issues for assessing the quality of experimental data. In this paper, we compared various methods and instruments for measuring the total ozone content (TOC) near St. Petersburg in the period from 2009 to 2015. We considered the TOC datasets of ground-based measurements at Voyeykovo (Dobson spectrophotometer and M-124 ozonometer) and Peterhof (Bruker 125HR spectrometer), as well as OMI and IASI satellite measurements. To assess the errors intrinsic to each of these gages, three ensembles of the TOC measurements were formed containing different numbers of comparisons and based on different selection criteria. At the first stage, we determined the means and standard deviations between the ensembles of the TOC measurements. Then, assuming a horizontally homogeneous and stationary ozone field, the random and systematic errors of individual methods were evaluated. The average random errors of the TOC measurements for all tree ensembles were 2.9 ± 0.5%, 2.8 ± 0.7%, 1.2 ± 0.2%, and 1.4 ± 0.1% for IASI, M-124, OMI, and Bruker 125HR, respectively. The systematic error of the standard Dobson measurements is –1.7% and –2.1% for OMI and IASI, respectively, and amounts to + 0.5% and + 2.1% for M-124 and Bruker 125HR, respectively. The OMI and Bruker 125HR TOC measurement errors are most resistant to atmospheric conditions, whereas errors in IASI and M-124 TOC measurements depend to a large extent on the state of the atmosphere.

Keywords: atmospheric ozone, errors, validation, IASI, OMI, M-124, Bruker 125HR, Dobson
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INTRODUCTION

The unique role of ozone in the Earth's atmosphere and its impact on many atmospheric processes and the environment stimulated the creation of an extensive and diverse monitoring system for its content [1–3]. This system uses a variety of instrumentation and measurement methods, both ground-based and satellite. The successful operation of this system is due to the intercalibration of different methods, as well as the knowledge of the measurement errors of various instruments. Estimation of errors in measurements of the total ozone content (TOC) is carried out by various methods: laboratory studies, joint measurements and intercalibration, methodical and numerical calculations. Considerable attention is paid to the validation of various satellite data by means of independent measurements with known errors (see, for example, [4–9]). As a rule, such studies allow estimating systematic and random mismatches between different TOC measurements. Estimates of the random and systematic errors of particular methods and instruments, their dependence on the time and place of measurements, the state of the atmosphere, and so on, are also of considerable interest.

Certain possibilities for obtaining such estimates are available in the case of simultaneous measurements by different methods and instruments. In the presence of two TOC measurements, their difference is determined by the random and systematic errors of both methods and instruments and by temporal and spatial variations in the ozone content. If we have three or more ensembles of simultaneous TOC measurements with different instruments, which were performed in the same air mass (horizontally homogeneous, stationary atmosphere), the accuracy characteristics of each instrument can then be derived, for example, the random and systematic errors of these measurements. Such an approach is a solution of a specific inverse problem, i.e., determination of measurement errors from the statistical characteristics of mismatches between the data from different instruments. This approach was demonstrated in several papers (see, for example, [10, 11]) devoted to the determination of errors in the moisture content of the atmosphere. For TOC, such assessments have not previously been conducted.

In this paper, the problem of estimating the random and systematic errors in determining the TOC from a number of ground-based and satellite instruments is solved for the first time based on observations in Peterhof (59.88° N, 29.82° E) and Voyeykovo (59.95° N, 30.70° E).

1. INSTRUMENTS AND METHODS FOR THE TOC MEASUREMENTS

Various methods and instruments for measuring the TOC are compared at the Department of Atmospheric Physics of the Physics Faculty, St. Petersburg State University, including the validation of satellite data [5–7]. Let us briefly describe the methods and devices.

Ground-based IR measurements with the Bruker 125HR Fourier spectrometer. Measurements of high-resolution spectra of direct solar IR radiation are carried out using the Bruker 125HR Fourier spectrometer (FS Bruker) and are described in detail in [6, 7]. The spectral windows recommended by the NDACC International observational network and the PROFFIT interpretation program are used [12]. Measurements in stable atmospheric conditions give an error of about 2–3% for a single TOC measurement, to which a significant contribution comes from the systematic error of measurements caused by errors in specifying the spectroscopic information.

Measurements with Dobson spectrophotometer. The Dobson spectrophotometer No. 108 (the Voeikov Main Geophysical Observatory (MGO)) serves as a reference instrument for the Roshydromet ozonometric network and is regularly checked (once every 3–4 years) in international comparisons with the WMO reference instrument. Observations are mainly performed on the direct Sun; the error of a single TOC measurement does not exceed 2% [13]. Measurements and processing are carried out by the members of MGO based on a standardized methodology.

Measurements with M-124. Filter ozonometer M-124 was the main instrument in the Russian ozonometric network for a long time. These instruments are regularly calibrated in simultaneous measurements with Dobson spectrophotometer no. 108. Long-term measurements with an M-124 ozonometer have shown that the error of a single TOC measurement does not exceed 5–8%, and the error in determining the daily average TOC value is 3–4% [14]. Observations of the TOC with M-124 ozonometer no. 403, which is a working instrument at the Voeikovo ozonometric station, are conducted daily (8 sessions per day). On clear days, parallel observations are made with the Dobson spectrophotometer; the maximum discrepancies with this device, as a rule, do not exceed 10 DU.

Satellite measurements with the OMI instrument. OMI (Ozone Monitoring Experiment) [15] is a space-scanning spectrometer for nadir measurements of outgoing (reflected and scattered) radiation in the UV and visible spectral regions. The OMI device operates on board the US AURA satellite, launched in 2004 to a near-polar solar-synchronous orbit. The scheme of OMI scanning allows daily global mapping of the TOC field with a nadir spatial resolution of $\sim 13 \times 24$ km^2. The declared inaccuracy of the TOC measurements using the TOMS version of processing is less than 3% [16].

Satellite measurements by the IASI instrument. The IASI instrument measures the spectra of the outgoing thermal radiation of the atmosphere in a wide spectral range, from 645 to 2760 cm^{-1}, with a high spectral resolution. These measurements are used to determine the vertical profiles of temperature, humidity, ozone content, and other parameters of the atmosphere and surface [17]. The spatial resolution is about 12 km in diameter. The measurements are carried out twice a day: at 9:30 and 21:30 local astronomical time (LAV). When interpreting the IASI data to determine the ozone content, various methods and algorithms are used [18]. In our work, the LISA algorithm is used, which gives the average TOC error no higher than 3% [18]. Previously, this algorithm was tested at the Peterhof station when comparing the tropospheric ozone content obtained with this method with the Bruker FS data [19].

2. ARRAYS OF CONSISTENT TOC MEASUREMENTS

To compare different measurements, we selected the days in which the TOC was measured by all the indicated instruments. There were 125 such days for the period from 2009 to 2015. The number of measurements is relatively small since Dobson spectrophotometer no. 108 periodically takes part in intercalibration with other devices. The features of the ensembles of the TOC measurements are as follows:

—Bruker FS data (measurements at Peterhof) are averaged over the period from 7:00 to 13:00 LAT, or over a shorter period, depending on the weather.

—The IASI satellite data are averaged within a radius of 1° from Peterhof over measurements in the morning hours (9:30 LAT).

—The OMI satellite data are obtained using the TOMS processing version—averaged values falling within a radius of 200 km from Voyeykovo (measured at about 12:00–13:00 LAT).

—The data of the Dobson and M-124 surface instruments in Voyeykovo (measurements by the Sun) are averaged over the day (measurements are mainly carried out in the first half of the day).

To assess the errors of individual instruments, three different ensembles were formed. Ensemble 1 consists of 125 days of the TOC measurements by all five instruments. Ensemble 2 consists of 278 days of measurements (comparison of four instruments, without Dobson), and ensemble 3, of measurements by four

Table 1. Statistical parameters of the TOC measurement ensembles for different instruments

Instrument	Average and SD, DU		
	1	2	3
IASI	336 ± 44	334 ± 47	329 ± 46
OMI	338 ± 44	334 ± 47	330 ± 45
M-124	345 ± 45	339 ± 48	336 ± 46
Bruker FS	351 ± 46	348 ± 50	343 ± 48
Dobson	344 ± 44	–	–

instruments (without Dobson) for days of high temporal stability of the TOC (125 days).

Table 1 shows the average values of the TOC and their variability (standard deviations (SD) from the average) according to the measurements by different instruments for all three ensembles. Since the ground-based measurements were carried out on the direct Sun, most of the measurements in the samples refer to the spring-summer period. Ensemble 1 consists of 55 days of measurements in spring, 59 days in summer, and 11 days in autumn. Ensemble 2 consists of 91 days of measurements in spring, 115 days in summer, 45 days in autumn, and 27 days in winter. In ensemble 3, the most stable days were selected out of 278 days of measurements when the changes in the TOC, according to the Bruker FS data, did not exceed 1% for several hours. There were 125 such days, 43 of which were in spring, 50 in summer, 22 in autumn, and 10 in winter.

It can be seen from Table 1 that, firstly, the average values and SDs for all ensembles and instruments are very close (the differences between the averages are 2–4%); second, the ground-based measurements give slightly larger average TOC values than the satellite measurements, while the ensemble average value is the highest for the Bruker FS. At the same time, the variability of the TOC is approximately the same according to the measurements of all devices for all ensembles.

3. COMPARISON OF DIFFERENT TOC MEASUREMENTS

Table 2 shows the average mismatches M^{A-B} and the standard deviations σ^{A-B} from the average values, in pairs for all instruments, separately for each ensemble. The correlation coefficient for different ensembles is 0.97–0.99 (with an error of 0.01–0.02).

The data in Table 2 allow us to draw the following conclusions:

—Minimum average differences in the TOC measurements are observed when comparing Dobson and M-124 devices (–0.5%) and between OMI and IASI satellite measurements (from 0.0 to 0.4). In the first case, this can be explained by periodic calibration of M-124 with the data from the Dobson spectrophotometer measurements, and in the second case, it can be assumed that validation of the IASI data by OMI measurements was carried out while testing the techniques and algorithms for processing the IASI measurements.

—If we consider the TOC measurements with a Dobson device as the reference, then the satellite instruments underestimate the TOC by approximately 2%. Similar results were obtained in a number of other papers (see, for example, [9, 20]). At the same time, ground-based measurements with a Bruker FS, on the contrary, overestimate the TOC also by about 2%. A similar effect was observed earlier in [21] when comparing the UV and IR measurements of the TOC. It is explained by the inconsistency between the quantitative characteristics of molecular absorption in different spectral regions. Consequently, the ground-based measurements (M-124 and Bruker FS) give 2–4% higher TOC values as compared to the satellite measurements (OMI and IASI devices).

—Variability of the characteristics of disagreements between different instruments for different compared ensembles is small and, generally, amounts to about 1%. The maximum disagreements are observed for ensemble 2, the most representative ensemble of comparisons

Table 2. Average mismatches and standard deviations from the averages when comparing the three TOC ensembles obtained from measurements by different instruments

Instrument	Ensemble	IASI, %	OMI, %	Bruker FS, %	M-124, %
OMI	1	+0.4 ± 2.5			
	2	0.0 ± 3.4			
	3	0.3 ± 2.7			
Bruker FS	1	+4.3 ± 2.6	+3.8 ± 2.0		
	2	+4.0 ± 3.6	+4.1 ± 2.4		
	3	+4.2 ± 2.8	+4.0 ± 2.2		
M-124	1	+2.6 ± 3.1	+2.2 ± 2.5	–1.6 ± 2.4	
	2	+1.6 ± 5.2	+1.6 ± 3.4	–2.4 ± 3.6	
	3	+2.0 ± 4.2	+1.7 ± 3.1	–2.2 ± 2.9	
Dobson	1	+2.1 ± 2.9	+1.7 ± 1.9	–2.1 ± 2.3	–0.5 ± 2.1

covering all seasons of the year. This is especially true for standard deviations of the TOC under different comparisons.

—On average, the best agreement is observed when comparing Dobson and OMI (if ignoring the comparison of the Dobson and M-124 ensembles) measurements in the UV spectral region.

As a rule, the validation of various satellite measurements ends here, but with simultaneous measurements of the TOC with different instruments, it is possible to estimate the measurement errors of individual instruments.

4. EMPIRICAL ASSESSMENTS OF THE ERRORS IN TOC MEASUREMENTS WITH DIFFERENT INSTRUMENTS

We represent the individual measurement x_i^A by the device A in the form

$$x_i^A = x_{i,\text{true}}^A + M^A + \sigma_i^A,$$

where $x_{i,\text{true}}^A$ is the true magnitude of the measured value, M^A and σ_i^A are the systematic and random measurement errors, respectively. For sufficiently large samples (when the average random errors are zero), the ensemble average difference M^{A-B} between two types of measurements (with A and B instruments) is determined by the difference in the systematic errors of the individual measurements

$$M^{A-B} = M^A - M^B. \tag{1}$$

This model of comparison is valid for a horizontally homogeneous and stationary atmosphere.

The standard deviation from the average difference σ^{A-B} in the presence of temporal σ^{t} and spatial σ^{sp} mismatches between measurements, under the condition that the individual random errors are not correlated, and assuming that the mean random errors are zero, can be written in the general case as

$$\sigma^{A-B^2} = \overline{\sigma^{A^2}} + \overline{\sigma^{B^2}} + \overline{\sigma^{\text{t}^2}} + \overline{\sigma^{\text{sp}^2}}. \tag{2}$$

Since the data contains satellite measurements that are averaged over a certain radius around ground-based measurement stations, it can be assumed that for a sufficiently large sample size, the average spatial disagreement between ground-based and satellite measurements tends to zero. The distance between Peterhof and Voyeykovo is about 50 km, and, as a rule, an air mass with the same ozone content is observed in both points. With regard to the temporal mismatch, the ground-based measurements are averaged over a period of several hours to both sides from the time of the satellite measurement. As a rule, the TOC changes little during the day, except for the periods of active movement of air masses. Therefore, taking the averaging of ground-based measurements into account, it can be assumed that the mean temporal mismatches also tend to zero. It should be noted that ensemble 3 was formed specifically to investigate the possible impact of a time-varying nature of the TOC field.

Assuming that the unknowns in Eq. (2) related to the temporal and spatial mismatches between measurements are zero, we obtain the upper error estimates for each of the instruments. Writing Eq. 2 for each pair of instruments, we obtain a system of 10 linear equations, in which the free term is the square of the standard deviation σ^{A-B^2} from Table 2. The pseudosolution of an overdetermined system of linear equations $Ax = B$, i.e., the most plausible solution to this inverse problem, is the column x that ensures the minimum of the sum of squares of the differences between the left and right parts of the equations. The solution of the minimization problem is the pseudoinverse matrix to the matrix A, that is

$$x = (A^{\text{T}}A)^{-1}A^{\text{T}}B. \tag{3}$$

As a result of solving problem (3), we obtained experimental estimates of the random error of individual devices presented in Table 3 for different ensembles. For ensemble 1, the random errors in the TOC measurements are minimal for the measurements with OMI (1.3%), Dobson (1.5%), and Bruker FS (1.5%). For M-124 ozonometer, this error is 1.9%, and for IASI device, 2.4%. The significant extension of the ensemble of comparisons (ensemble 2) markedly increases the random errors for IASI and M-124 (up to 3.5%).

It should be noted that the errors of ground-based and satellite measurements by a number of instruments have a noticeable seasonal variation (see, for example, [6]), and an increase in the random errors in Table 3 for ensemble 2 is probably related to the inclusion of autumn and winter measurements. For the satellite OMI device, the random component of the errors decreased and amounted to 0.9%, which is possibly related to the suppression of random measurement errors due to a larger number of comparisons, as well as suppression of the contribution of random spatial and temporal mismatches. Finally, for ensemble 3 (minimum temporal variations in the ozone field), the random errors for M-124 and IASI devices decreased noticeably compared to ensemble 2, which indirectly indicates possible mismatches caused by temporal variations of ozone, which, according to our estimates, are 0.6–0.8% (the inset at the bottom of the 2nd column of Table 3).

The average error for the TOC measurement by the IASI instrument in all three ensembles is (2.9 ± 0.5)%; M-124, (2.8 ± 0.7)%; OMI, (1.2 ± 0.2)%; Bruker FS, (1.4 ± 0.1)%. The value of error variability characterizes its dependence on the measurement conditions. As can be seen, the TOC measurements with OMI and Bruker FS are the most stable; the accuracy of M-124 and IASI is more dependent on the state of the atmo-

Table 3. Estimations of the errors (%) in TOC measurement by different instruments. Systematic and total errors are calculated with respect to zero systematic error of the Dobson instrument

Instrument	σ_{rand}			σ_{sys}	σ_{total}			$\sigma_{\text{total indep}}$
	1	2	3	1	1	2	3	
IASI	2.4	3.5	2.7	–2.1	3.2	4.1	3.4	2.7 [18]
OMI	1.3	0.9	1.3	–1.7	2.1	1.9	2.1	3 [16]
M-124	1.9	3.5	2.9	+0.5	2.0	3.5	2.9	3 [14]
Bruker FS	1.5	1.5	1.2	+2.1	2.6	2.6	2.4	2–4 [6]
Dobson	1.5	$\sigma^{t} = 0.6–0.8$		0.0	1.5	$\sigma^{sp} = 0.9$		2 [13]

sphere. In general, the values obtained indicate stability in the solution of the inverse problem for determining the errors in the TOC measurements by individual devices.

Since the OMI data are related (centered) to the Voyeykovo station, and the IASI data, to the Peterhof station, we can use equations (2) to estimate the spatial error assuming that it is nonzero between devices belonging to different ground-based stations. In this case, the random errors for individual devices remained almost unchanged, and the spatial component of the error in the TOC measurement between Voyeykovo and Peterhof was estimated at 0.9% (the inset at the bottom of the 4th column of Table 3).

If we take the TOC measurements with the Dobson spectrophotometer as the reference and assume that their systematic errors are zero, then the 5th column shows estimates of the systematic measurement errors for the four instruments with respect to the Dobson measurement scale (see Eq. (1)). For ensemble 1, the systematic errors are maximal for IASI and Bruker FS, amounting to 2.1% in absolute value. As noted above, IASI measurements underestimate the TOC values and the Bruker FS overestimates them as compared to the Dobson instrument data. The systematic errors of the OMI satellite device are also quite significant, and they are minimal for M-124 ground-based measurements. The latter is explained by the regular calibration of M-124 using a Dobson spectrophotometer.

In the 6–8th columns of Table 3 we show estimates of the total measurement errors of different instruments for different ensembles under comparison. They are minimal for the OMI device and close to 2%. The total errors for the Bruker FS are 2.4–2.6%. For the IASI device, it is 3.2–4.1%, depending on the ensemble used for comparison. The last column of Table 3 shows the estimates of the total errors of TOC measurements by different instruments according to the works indicated in the table, which, overall, are in good agreement with the estimates obtained in our studies. The estimates of [18] for the IASI device are an exception, and are 0.5–1.4% smaller than our estimates. This may be due to differences in the ensembles used for comparison in these two works (as shown above, the measurement errors for IASI instrument are sensitive to the atmospheric conditions). At the same time, large errors for IASI instrument as compared to the other devices can be explained by the fact that they were obtained by interpreting the measurements of the outgoing thermal radiation of the atmosphere. There are significantly more factors in this remote method (the temperature gradients in the atmosphere and the accuracy of their determination [22]) that affect the error of the method than in the other methods used.

5. MAIN RESULTS AND CONCLUSIONS

Comparisons of different instruments and methods for measuring the TOC in the region of St. Petersburg in 2009–2015 were carried out. We compared the measurements with the Dobson and M-124 instruments at Voyeykovo, the Bruker FS at Peterhof, and satellite measurements using the OMI and IASI instruments. To estimate the errors of each of these devices, three ensembles were formed, coordinated with respect to the time of the TOC measurements. These ensembles contain different numbers of comparisons (measurement days), different selection criteria, and include measurements of five or four instruments (without the Dobson spectrophotometer, which is considered a reference instrument).

The averages and SDs of the TOC for all ensembles and instruments are very close (the differences between the averages are 2–4%). Minimum average differences in the TOC measurements are observed when comparing Dobson and M-124 (–0.5%) and OMI and IASI satellite measurements (from 0.0 to +0.4%). In the first case, this can be explained by periodic calibration of M-124 according to Dobson measurements; in the second one, it is assumed that validation of IASI data was performed using OMI measurements when testing the techniques and algorithms for interpreting IASI measurements.

The maximum standard deviations from the average mismatches are observed for the most representative ensemble of comparisons, which covers all seasons of the year. On average, the best agreement is observed when comparing the Dobson and OMI instruments (when ignoring the comparison between Dobson and M-124 measurement ensembles). The correlation coefficient between the data obtained using different devices is 0.95–0.98 (with an error of 0.01–0.02).

At the second stage of the research, the inverse problem was solved to determine the measurement errors of individual devices based on the previously calculated statistical characteristics of the discrepancies between these devices. Under the assumption of a horizontally homogeneous and stationary ozone field, the following results are obtained.

The average random error of the TOC measurement in all three ensembles is (2.9 ± 0.5)% for the IASI instrument, (2.8 ± 0.7)% for M-124, (1.2 ± 0.2)% for OMI, and (1.4 ± 0.1)% for the Bruker FS. The TOC measurements with the OMI and Bruker FS instruments are the most stable; the accuracy of the other two instruments is more dependent on the state of the atmosphere, getting worse with a substantial expansion of the ensemble of comparisons (ensemble 2). With a certainly stationary ozone field (ensemble 3), the random errors are noticeably smaller for the M-124 and IASI devices, which indirectly indicates possible discrepancies caused by time variations of ozone (~0.6–0.8%).

Assuming that the OMI data are centered on the Voyeykovo station and the IASI data are obtained at the Peterhof station, the spatial component of the TOC measurement error between Voyeykovo and Peterhof can be estimated to be 0.9%.

The systematic errors of four instruments were determined with respect to the Dobson spectrophotometer: they are maximal for the IASI and Bruker FS (2.1%). For the OMI device, the error is 1.7%. The satellite devices underestimate and the Bruker FS overestimates the TOC values as compared to the Dobson device. Minimal errors are observed for M-124, which is explained by the regular calibration of this device with a Dobson spectrophotometer.

The total errors of the TOC measurements with the different instruments and for different ensembles of comparison are determined. They are minimal for the OMI device (~2%). The Bruker FS measurements have the total errors of 2.4–2.6%; IASI, 3.2–4.1%, depending on the ensemble of comparisons used. The comparison of the estimates with previously published data shows their good agreement, except for the estimates made for the IASI satellite device, which are 0.5–1.4% higher than the estimates made in [18]. The latter is probably caused by the difference between the conditions in Peterhof and Voyeykovo from comparison conditions in [18].

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