
REMOTE SENSING OF ATMOSPHERE,
HYDROSPHERE, AND UNDERLYING SURFACE

Spectroscopic Measurements of O₃ and NO₂ Atmospheric Content: Correction of Ground-Based Method and Comparison with Satellite Data

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Abstract—The results of daily observations of O₃ and NO₂ atmospheric total content near St. Petersburg retrieved from the automated ground-based measurements of zenith-sky scattered visible solar radiation are presented. The measurements of 2009–2013 are compared with the data of satellite measurements with the GOME (ERS-2), SCIAMACHY (ENVISAT), and OMI (AURA) satellite spectrometers. The analysis of differences revealed between the data of satellite and ground-based measurements allowed us to improve the ground-based method (DOAS technique) and decrease the average difference down to ~2 and ~20% for O₃ and NO₂, respectively. Remaining differences may be further decreased if the seasonal variability of air mass factors calculated is taken into account in the ground-based method.

Keywords: DOAS measurements, O₃ atmospheric content, NO₂ atmospheric content

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INTRODUCTION

Long-term high-accuracy measurements of O₃ and NO₂ atmospheric content are of interest in connection with an urgent problem of determining possible natural and anthropogenic variations in the gas composition of the atmosphere. One of the methods for effective monitoring of these atmospheric gases in different regions and under different weather conditions is ground-based spectrometry of zenith-sky scattered UV and visible solar radiation.

In particular, within the NDACC international network (Network for the Detection of Atmospheric Composition Change, <http://www.ndacc.org>), the UV and visible measurement segment (UV-visible network) has been operating already for several decades; it consists of more than 35 certified ground-based spectrometers arranged around the globe and monitoring the O₃ and NO₂ atmospheric content. NDACC (former NDSC: Network for the Detection of Stratospheric Change) is a global network of stations for long-term monitoring of the gas composition of the atmosphere. The NDACC UV-visible network includes stations equipped with SAOZ (Système d'Analyse par Observation Zénithale [1]) instruments, which play the leading role in this group. SAOZ is a diffraction spectrometer with a multichannel radiation photodetector of CCD (charge-coupled device) type, which measures zenith-sky scattered UV and visible solar radiation. The measurements are mainly carried out in morning and evening hours, during sunrise and

sunset, in twilight. Beginning from the late 1980s—early 1990s, SAOZ spectrometers have been installed at many stations (see, e.g., <http://saoz.obs.uvsq.fr>), as well as other measuring instruments similar in geometry and measuring principles. In particular, similar routine measurements were started in the former USSR at Issyk Kul (Kyrgyzstan) [2], in Zvenigorod [3], and in Salekhard and Zhigansk [4].

Since 2004, atmospheric content of O₃ and NO₂ have been monitored on the basis of zenith measurements of scattered visible radiation in St. Petersburg, at the V.A. Fock Institute of Physics, St. Petersburg State University (Petrodvorets) [5]. In this work, we compare the results of daily ground-based measurements with concurrent satellite data on O₃ and NO₂ contents near St. Petersburg, estimate their agreement, and determine and eliminate some causes of differences between the data.

1. TECHNIQUES AND EQUIPMENT FOR GROUND-BASED MEASUREMENTS

The method used by the authors for measurements and key principles of the result processing technique for O₃ and NO₂ content retrieval have been described in detail in [5]. The spectral equipment described in [5] was then significantly improved. Below, we characterize the equipment that has been used for the monitoring since the beginning of 2009, and some features

of the results interpretation, which have not been considered earlier [5], i.e.:

1) simultaneous use of O₃ and NO₂ spectral absorption coefficients at two different temperatures (223 and 243 K for O₃ and 220 and 296 K for NO₂) in the algorithm. This procedure, being one of the standard options of the WinDOAS algorithm [6], assumes a partial account for the temperature dependence of the absorption coefficients with altitude with the aim of distinguishing the main contribution of the stratospheric content at a temperature of about 220 K (the coefficients that correspond to different temperatures are considered mutually orthogonal);

2) account for additional noises proportional to a signal measured (intensity) and induced by the so-called flare light and dark current (noise) of the detector. For this, an additional term (*offset*(λ)) is included in the main equation (Beer–Lambert–Bouguer law in exponential form):

$$I(\lambda) - \text{offset}(\lambda) = I_o(\lambda) \exp\left(-\sum q_i \sigma_i(\lambda) - P(\lambda)\right). \quad (1)$$

Here $I(\lambda)$ is the intensity measured at the wavelength; $I_o(\lambda)$ is the reference spectrum; q_i is the effective content of the i th components at the radiation propagation path; $\sigma_i(\lambda)$ is the spectral absorption coefficient of the i th component; $P(\lambda)$ is the polynomial in the form $\sum d_k \lambda^k$ for accounting the contributions of the Rayleigh and aerosol attenuations. The parameter *offset*(λ) is defined in the form of function

$$\text{offset}(\lambda) = \left(a + b(\lambda - \lambda_0) + c(\lambda - \lambda_0)^2\right) \bar{I},$$

where λ_0 is the central wavelength of the working spectral range; \bar{I} is the interval average intensity; a , b , and c are the parameters selected. The DOAS algorithm, developed for interpretation of data of measurements at the V.A. Fock Institute of Physics beginning from 2009, uses a polynomial of the zero order (with zero b and c coefficients) to describe the spectral dependence of the parameter *offset*(λ) and a fifth power polynomial as the function $P(\lambda)$.

Main specifications of the spectral equipment that has been used for routine measurements in St. Petersburg (Petrodvorets) since 2009 are given below.

Optical scheme	Asymmetric Czerny–Turner scheme
Radiation detector	CCD array of 3648 elements
ADC capacity	14 bits
Width of entrance slit	50 μm
Diffraction grating	1200 gr./mm
Spectrum recording time	3.8 ms–60 s
Spectral range	400–610 nm
Spectral resolution	0.6 nm
Angular field of view	10°

The device is based on a commercial HR4000 spectrometer (Ocean Optics Inc., <http://oceanoptics.com>). A holographic diffraction grating is used as a dispersion element in the spectrometer, and a CCD-based multi-channel radiation director, as a radiation receiver. As compared to equipment used before [5], new equipment is characterized by a wider spectral interval (400–610 instead of 428–515 nm), a higher spectral resolution (~0.6 instead of ~1.3 nm), and a much lower recording time (1–3 instead of 1 min). In general, main specifications of the device are close to SAOZ specifications (CCD-based photodetector, spectral range of ~300–600 nm, and spectral resolution of ~1 nm) [1].

For uniformity and homogeneity of the series of ground-based DOAS measurement at the V.A. Fock Institute of Physics, the effective O₃ and NO₂ content is determined in the same spectral range as that used during the processing of measurements with previous equipment in 2004–2009 (428–515 nm). The resulting effective (inclined) content is transformed in the sought vertical (total column) content with the use of model coefficients, the so-called air mass factors (AMF). AMFs are calculated in the atmospheric radiative transfer model (SCIATRAN [7]) with the use of characteristic annual average O₃ and NO₂ vertical distribution profiles, as well as air pressure and temperature. Series of vertical contents measured during sunrise and sunset are averaged over the range of zenith angles of Sun 90°–91° (the range of twilight measurements has been narrowed toward low Sun to minimize a possible contribution of anthropogenic pollution of the troposphere near St. Petersburg). To consider the daily cycle of NO₂ content in the stratosphere, the resulting morning and evening estimates of the vertical content are reduced to the local midday using a photochemical model similar to [5, 8, 9]. In general, errors of the ground-based measurements of the vertical O₃ and NO₂ content are determined not by their inclined content errors (which are 0.7–0.8 and 1.4–2.0% for O₃ and NO₂, respectively), but rather uncertainty of AMF used caused by the natural variability of the atmosphere state (mainly the vertical distribution of the O₃ and NO₂ contents). The total errors of the ground-based method are estimated (see, e.g., [10]) as 7 and 21% for O₃ and NO₂ contents, respectively. The estimate of the total column NO₂ increases up to 40% under conditions of strongly polluted atmosphere, i.e., increased NO₂ in the surface layer.

2. COMPARISON WITH SATELLITE DATA

The results of ground-based daily measurements of O₃ and NO₂ content from 2009 to 2013 have been compared with data received from the nearest satellites and measured with such instruments as GOME (Global Ozone Monitoring Experiment [11], ERS-2 satellite), SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CHartography [12],

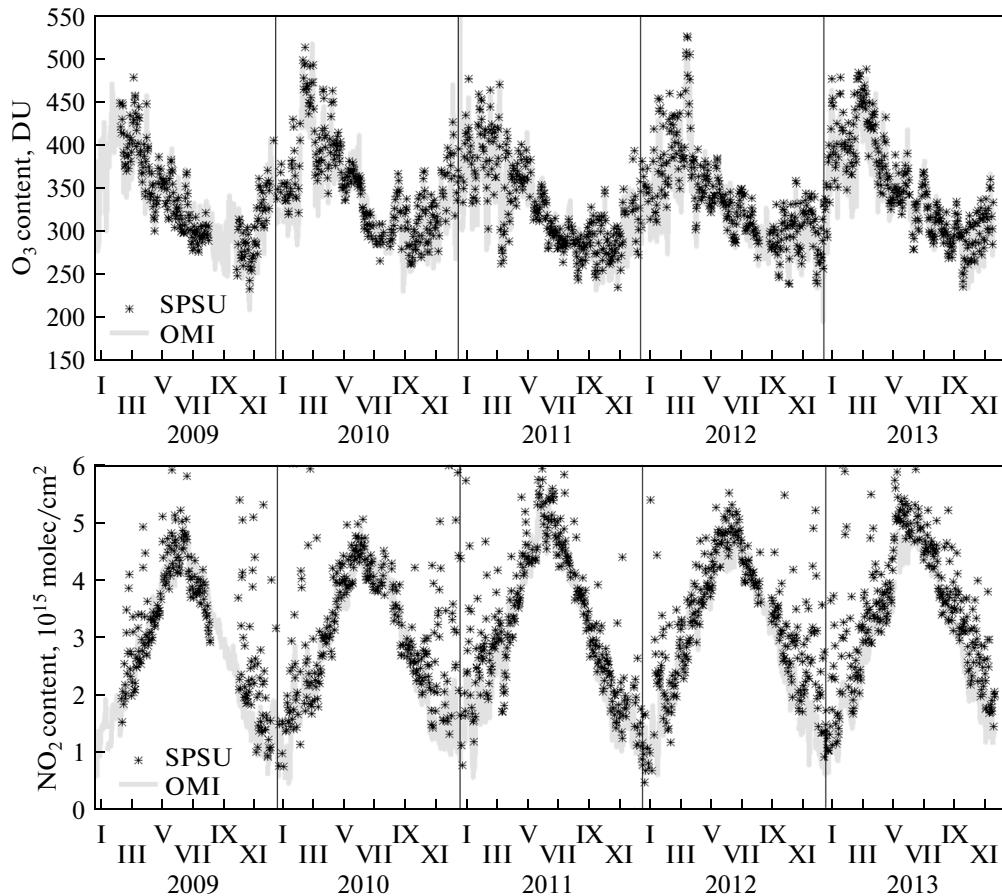


Fig. 1. Ground-based (St. Petersburg State University) and satellite (OMI) measurement data on O_3 and NO_2 near St. Petersburg (Petrodvorets) in 2009–2013 (the results have been reduced to the local midday to account the daily cycle of NO_2 content).

ENVISAT satellite), and OMI (Ozone Monitoring Experiment [13], AURA satellite). These scanning spectrometers allow global monitoring of the spatial distribution of O_3 and NO_2 atmospheric contents on the basis of nadir measurements of outgoing visible radiation with a horizontal resolution from ~ 300 km for GOME to ~ 60 km for SCIAMACHY and ~ 20 for OMI. In this work, we used data from ESA GOME (O_3 and NO_2 , <http://atmos.eoc.dlr.de/gome>), ESA SCIAMACHY (O_3 , <http://atmos.caf.dlr.de/sciamachy>) and IUPB SCIAMACHY (NO_2 , http://www.iup.uni-bremen.de/doas/scia_no2_data_acve.htm), and NASA OMI (O_3 and NO_2 , <http://avdc.gsfc.nasa.gov>) above checkpoints the nearest to St. Petersburg.

These spectrometers measured at the St. Petersburg latitude almost every day (GOME measurements ceased in July, 2011, SCIAMACHY, in April, 2012, and OMI is currently measuring). For the satellite data selected, the average checkpoint time passage and distance between a subsatellite point and St. Petersburg were $12:30 \pm 0:30$ and (140 ± 110) km for GOME, $12:00 \pm 0:30$ and (40 ± 25) km for SCIAMACHY, and $13:20 \pm 0:45$ and (25 ± 25) km for OMI. The NO_2 con-

tent error has been estimated as 2–5% for GOME [14], SCIAMACHY [16], and OMI [17]; it can attain 50% in the polluted troposphere [17].

Figure 1 exemplifies the comparison of simultaneous ground-based (St. Petersburg State University) and satellite (OMI) measurements near St. Petersburg in 2009–2013 in the form of series of daily average O_3 and NO_2 contents.

Both measurement systems show seasonal variability of the O_3 and NO_2 stratospheric contents typical for this latitude: O_3 content is maximal (~ 450 DU) in spring and minimal (~ 250 DU) in autumn; the NO_2 content is maximal ($\sim 5 \times 10^{15}$ molec/cm²) in the mid-summer and minimal ($\sim 1 \times 10^{15}$ molec/cm²) in the midwinter. Cases of significant differences are seen in the NO_2 measurement data, i.e., significantly higher ground-based measured values as compared to satellite data. These cases are connected with an increased NO_2 content in the surface air layer, sometimes observed despite the twilight geometry of ground-based measurements and a significant distance from the main sources of anthropogenic pollution in the central part of St. Petersburg.

Table 1. Average deviation Δ and standard deviation from the mean σ for the satellite–ground-based spectrometer difference in relative units (%) and correlation coefficient R

Gas	Satellite spectrometer	Δ		σ		R	
		version 1	version 2	version 1	version 2	version 1	version 2
O ₃	GOME	+13.3	−0.6	4.8	5.4	0.93	0.94
	SCIAMACHY	+13.0	−0.8	3.9	4.8	0.95	0.95
	OMI	+11.5	−2.7	5.4	6.2	0.91	0.92
NO ₂	GOME	−19.4	−16.6	77.7	78.0	0.47	0.46
	SCIAMACHY	−17.1	−14.0	51.4	52.9	0.65	0.64
	OMI	−30.7	−26.4	66.7	61.8	0.69	0.69

In contrast to ground-based measurement results, which are estimations of the total column NO₂ (though mainly in the stratosphere), the OMI measurement data used in this work characterize more the stratospheric part of the total NO₂ due to special algorithms for data correction accounting for the tropospheric contribution [18].

3. ANALYSIS OF DIFFERENCES AND CORRECTION OF GROUND-BASED METHOD

The results of comparison of satellite and ground-based measurement results in the form of estimates of deviations and correlation coefficients are given in Table 1. The GOME, SCIAMACHY, and OMI spectrometer data were used for the comparison. For uniformity, taking into account the cessation of the GOME and SCIAMACHY spectrometers, the array of data compared is bounded by the period from January, 2009, to July, 2011. According to estimates in Table 1, the ground-based results for ozone are lower than the satellite data by 12–13% on the average for all the spectrometers considered (see “version 1” column in Table 1). The standard deviations from the average difference are 4–5%, and the correlation coefficients are 0.91–0.95. The behavior of average differences common for the three different satellite spectrometers, relatively low standard deviations, and high correlation coefficients point out to a systematic error in the ground-based measurements of ozone content. In contrast to O₃, the data of ground-based measurements of NO₂ are much higher than satellite data, by 20–30% on the average (see “version 1” column in Table 1). The standard deviations from the average differences are quite high in this case (50–80%) and the correlation coefficients are relatively low (0.5–0.7). The above-mentioned abnormally high NO₂ content recorded from the ground in the surface air layer mainly contributes to the differences in the NO₂ measurement data compared.

A more detail analysis of the comparison results has revealed several parameters of the algorithm for

ground-based measurement data interpretation that contribute a systematic error in the estimation of the ozone content and require corresponding correction:

1. Characteristic temperature of the spectral coefficients of O₃ and NO₂ absorption. The ground-based measurement results are closer to the satellite data when using only “cold” absorption coefficients, i.e., those that corresponds to the mean stratospheric temperature (~220 K). It has been ascertained that consideration of both coefficients (“warm” and “cold”) simultaneously results in an increase in the difference between satellite and ground-based measurement results.

2. Degree of a polynomial that describes the effects of Rayleigh and aerosol attenuations [$P(\lambda)$ in Eq. (1)]. The use of a cubic polynomial instead of quintic, common in the interpretation of ground-based measurement results, decreases the difference with satellite data.

3. Degree of a polynomial that describes the instrumental noises of spectral measurements [$offset(\lambda)$ in Eq. (1)]. The use of the second degree instead of zero one decreases the difference with satellite data.

To illustrate the influence of each of the modifications of the algorithm for ground-based measurement result interpretation, we give below (Table 2) the corresponding variations in the effective (inclined) O₃ and NO₂ contents. These estimates have been found from the analysis of one measurement during the sunset (zenith angle was 90°) on February 17, 2011.

The initially selected parameters are designated hereinafter as “version 1”, and the parameters and working results of the modified algorithm, as “version 2”. Table 2 also presents the results of calculations of the effective O₃ and NO₂ contents (version 1) and their relative variations accounting each modification separately and all the modifications ((version 2 – version 1)/version 1). These modifications affect the resulting O₃ and NO₂ contents oppositely. Thus, the O₃ content found increases (+13%) when using only the “cold” O₃ absorption coefficient and decreases (−8%) when using the polynomial

Table 2. Effect of parameters of the algorithm for ground-based measurement result interpretation on estimates of the O₃ and NO₂ atmospheric content (q): temperature of the absorption coefficients ($T_{\sigma(\text{O}_3)}$, $T_{\sigma(\text{NO}_2)}$) and degrees of $P(\lambda)$ and $offset(\lambda)$ polynomials

Gas	q , molec/cm ²	$T_{\sigma(\text{O}_3)}$	$T_{\sigma(\text{NO}_2)}$	$P(\lambda)$	$offset(\lambda)$	Σ^*
O ₃	$13.3 \pm 0.5 \times 10^{19}$	+13%	−1%	−8%	−4%	+21%
NO ₂	$6.5 \pm 0.4 \times 10^{16}$	−3%	−2%	+6%	0%	−15%

* All algorithm modifications are considered.

$P(\lambda)$ of a lower degree. Vice versa, the NO₂ content increases (+6%) with a decrease in the degree of the polynomial $P(\lambda)$ and decreases (−3%) when using the “cold” O₃ absorption coefficient. In general, when changing from version 1 to version 2 of the algorithm for the measurement considered, the O₃ content estimate increases by 21% and the NO₂ content estimate decreases by 15%.

Table 1 presents the estimates of differences between the satellite and ground-based measurement data on O₃ and NO₂ contents and ground-based measurement results processed with the use of the algorithm modified (“version 2” column) in 2009–2011. New ground-based data on O₃ content are significantly closer to satellite data: average differences are 1–3% instead of 12–13%. The ground-based data on NO₂ content also become closer to satellite data: the average difference decreases from 17–31% to 14–26%. However, the standard deviations of the average differences and correlation coefficients between ground-based and satellite data remain almost the same.

Figure 2 shows seasonal variations in the differences in the form of their monthly average values calculated for comparison with OMI spectrometer data for 200–2012. The standard deviation Δ , standard deviation from the mean σ , and correlation coefficient R are estimated as −2.8%, 5.5%, and 0.92 and −32.0%, 63.5%, and 0.77 for comparison with data on O₃ and NO₂, respectively. Average differences between the satellite and ground-based data (version 2) significantly depend on the season and show significant excesses of the ground-based measurement results over satellite data in winter: up to 5% for O₃ and up to 45% for NO₂. High winter values for NO₂ in the ground-based measurements can be explained mainly by the contribution of anthropogenic pollution of the troposphere (excluded in satellite data). The average difference does not exceed 5% from April to September (see Fig. 2). At the same time, characteristic seasonal variations in the differences observed in O₃ and NO₂ measurements can be also connected with ignorance of seasonal variability (AMF) in the ground-based techniques (in contrast to satellite algorithms, where this dependence is considered). Accounting for the seasonal variability of AMF in the ground-based

DOAS measurements of O₃ and NO₂ can significantly improve agreement with the satellite data, which, in particular, has been recommended for NDACC network stations. Thus, similar comparison of satellite data with ground-based DOAS measurement results for O₃ at some SAOZ stations has shown significantly lower differences when using the AMF climatological data array recommended for NDACC [19]. We should note that the seasonal variability of the differences revealed between satellite and ground-based measurement data on O₃ (see Fig. 2) are generally similar to the results of analogous comparisons in [19], where a possibility of a significant decrease in this variability with the use of the AMF, recommended above, has been also shown.

CONCLUSIONS

Ground-based zenith measurements of spectra of scattered solar radiation allow implementation of a quite simple method for automated all-weather measurements of O₃ and NO₂ atmospheric (mainly stratospheric) contents. Such measurements have been carried out routinely near St. Petersburg (Petrodvorets) since 2004; they provide for O₃ and NO₂ atmospheric content twice a day (during sunrise and sunset). A part of the ground-based data array (2009–2013) received from measurements of currently operating equipment has been compared with data of satellite measurements near St. Petersburg. The comparison analysis has revealed systematic differences, which were partly eliminated after a correction of the algorithm for interpretation of ground-based DOAS measurements. Thus, the mean deviation from the satellite data decreased from 12–13% to 1–3% for O₃ and from 17–31 to 14–26% for NO₂ in the corrected ground-based measurement results. The decrease has been attained mainly due to the choice of the characteristic temperature of the spectral coefficient of ozone absorption and degree of the polynomial that is responsible for nonselective radiation attenuation in the DOAS technique. The remaining difference has pronounced seasonal variability and can be eliminated by consideration of air mass factors in the ground-based method (e.g., recommended for the NDACC international network [19]). The results allow further processing of

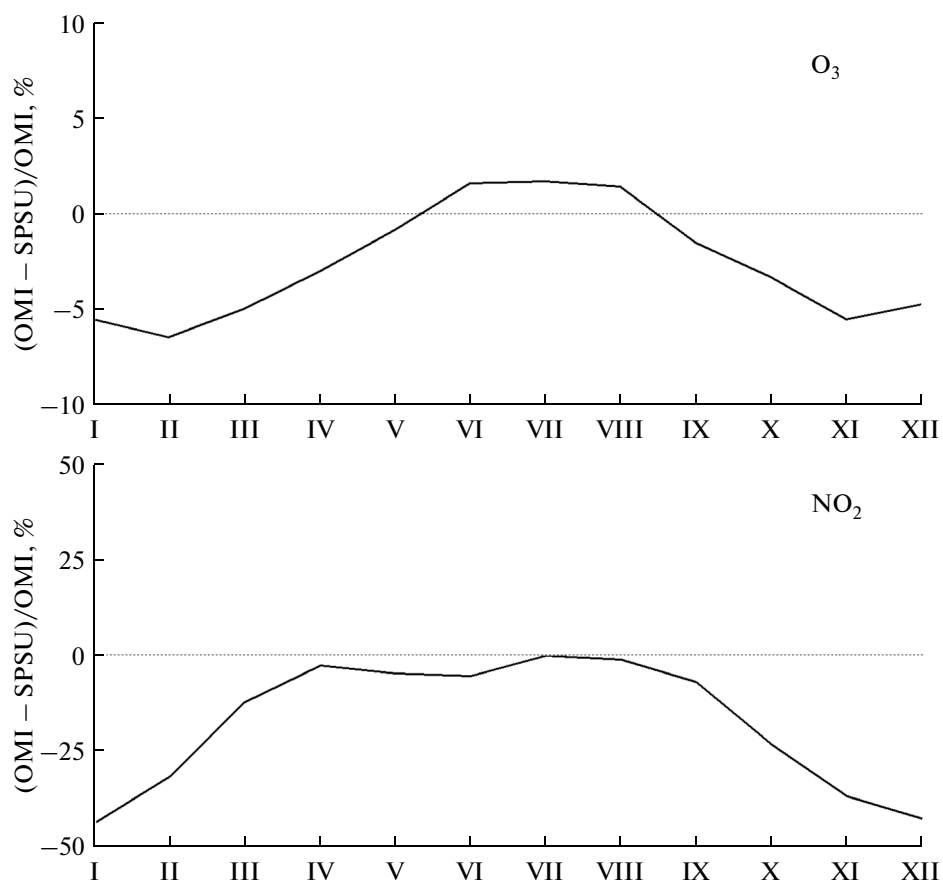


Fig. 2. Seasonal variations in differences between satellite (OMI) and ground-based (St. Petersburg State University, version 2) measurements of O₃ and NO₂ in the form of monthly average values, calculated for the period of comparison (2009–2013).

the whole array of DOAS measurements and generation of a continuous uniform series of observation data on O₃ and NO₂ content near St. Petersburg from 2004 up to the present.

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