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NO_2 Content Variations near St. Petersburg as Inferred from Ground-Based and Satellite Measurements of Scattered Solar Radiation

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Abstract—An automatic spectral complex developed at the Institute of Physics, St. Petersburg State University, is described. This complex is used for regular ground-based spectroscopic measurements of the total NO_2 content in the vertical column of the atmosphere during the twilight and daylight hours of the day near St. Petersburg (Petrodvorets). In 2004–2006, a number of ground-based twilight measurements of the total NO_2 content were obtained near St. Petersburg, and variations in the NO_2 content in the troposphere were estimated from the results of daytime ground-based measurements. An example of the spatial annual mean distribution of the NO_2 content (central and northern Europe, northwestern Russia) based on the data of satellite measurements over the period 2003–2005 is presented. This example demonstrates the main sources of anthropogenic pollution. An increase in the mean annual contents of tropospheric NO_2 near Moscow and St. Petersburg is preliminarily estimated for the entire period of satellite observations with the GOME instrument at about 30–40% over ten years.

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1. INTRODUCTION

The changes in the thermal regime of the atmosphere at different heights, ultraviolet illuminance of the surface, atmospheric dynamics, precipitation regime, etc., that are associated with variations in the gas composition of the atmosphere already have and will have progressively enhanced negative ecological, economical, and social consequences [1–4]. In this regard, monitoring of the gas composition of the atmosphere (in particular, global continuous observations of the content of greenhouse and ozone-destroying minor gas constituents) is one of the central problems of modern physics and chemistry [5]. Nitrogen dioxide (NO_2), one of the key gas constituents in atmospheric chemistry, belongs to such substances. In the stratosphere, NO_2 takes part in the cycle of reactions destroying atmospheric ozone, whereas in the troposphere, the photolysis of NO_2 leads to the formation of O_3.

At present, the main information about characteristics of the gas composition of the atmosphere is supplied by different observation systems—ground-based, aircraft, balloon, and satellite. The satellite system of monitoring atmospheric characteristics is undoubtedly the most important component of the global system of observations. However, regular satellite measurements of the NO_2 content in the atmosphere have become possible relatively recently with the appearance of space-scanning high-resolution spectrometers, which perform nadir measurements of the outgoing (reflected and scattered) radiation in the visible region of the spectrum (~400–500 nm). Such instruments as the GOME [6] in 1996 (the ERS-2 satellite), SCIAMACHY [7] in 2002 (the ENVISAT satellite), and OMI [8] in 2004 (the AURA satellite) were the first to allow a virtually daily global mapping of the NO_2 content in a vertical column of the atmosphere. At the same time, owing to considerable difficulties with the interpretation of such satellite measurements, the question concerning the accuracy of these data still remains open, especially if the NO_2 content in the troposphere is increased [9]. Additionally, the time series of these observations is insufficiently long to obtain reliable estimates of the corresponding trends. Thus, the problem of simultaneous ground-based measurements of the NO_2 content in the atmosphere remains important for both the validation of the developing system of satellite observations and the estimation of long-term variations in the NO_2 content.

The Network for the Detection of Atmospheric Composition Change (NDACC), the former NDSC, is the main element of the global system of ground-based observations of the atmospheric gas composition. The NDACC network is an international program unifying the stations equipped with a standardized set

† Deceased.

Main characteristics of measuring instruments

Characteristic	Value
Angle of view, deg	10
Spectral range of spectrometer, nm	428.4–515.3
Scanning pitch, nm	0.05
Spectral resolution, nm	1.3
Photometric band center, nm	440
Photometric band half-width, nm	5

of instruments for remote sounding of the atmosphere from the Earth's surface. These instruments are intercompared and intercalibrated [10–13]. Ground-based spectrometers recording the spectra of the visible solar radiation scattered into the zenith are used for measuring the total content of NO_2 at the NDACC stations. In Russia and CIS countries, the total NO_2 content is observed at a number of stations, some of which are certified as complementary sites of the NDACC network (Zvenigorod [14], Salekhard, Zhigansk [15], Issyk Kul [16, 17]). Since the middle of 2002, the Institute of Physics of St. Peterburg State University (Petrodvorets, (IOP SPBSU)) has regularly measured the total NO_2 content near St. Petersburg (about 30 km southwest of the city center) by the method of spectrometry of the solar radiation scattered from the zenith. These observations are performed both under the conditions of twilight and daylight. This paper describes the instruments used and the procedures of data measurement and interpretation and gives examples of the results of observations of the NO_2 content near St. Petersburg.

2. INSTRUMENTS AND METHODS OF INTERPRETATION OF MEASUREMENTS

An automatic spectral complex (ASC) developed at the IOP SPBSU is used for measurements of the solar radiation scattered in the zenith direction. This complex consists of a spectrometer (an MDR-12 monochromator and an FEU-100 photoreceiver), a photometer with an interference filter (to control the temporal variability of the zenith brightness), and a recording system. The main characteristics of the ASC are presented in the table. Measurements are performed continuously in an automatic regime at zenith angles of the Sun smaller than 100°. During daylight hours (from sunrise to sunset), depending on its duration, up to 1200 measurements of intensity spectra of the solar radiation scattered in the zenith are obtained at different positions of the Sun. The measurements during the entire daylight hours and not only in the period of morning or evening twilight allow us to determine diurnal variations in the NO_2 content and reveal the cases of tropospheric pollution.

The NO_2 content on the path of radiation formation (the slant content) is retrieved from the results of ground-based spectroscopic measurements with the use of the differential absorption method [18]. The differential optical absorption spectroscopy (DOAS) method lies in the minimization of the mismatch between the results of measurements and a linear combination of the known molecular absorption spectra (O_3, NO_2, H_2O, O_4) after subtracting the smoothly varying attenuation components (surface albedo, molecular scattering, aerosol scattering, etc.) described by a polynomial. At the first stage, the NO_2 content for the path of the outgoing radiation (the so-called slant content) is determined by the least-squares method and, after that, the total NO_2 content in the vertical atmospheric column is determined with the use of the air-mass fsctors (the modeled ratio of the slant content to the vertical one) calculated beforehand for different measurement conditions. Since 2004, ground-based measurements of the total NO_2 content at the IOP SPBSU have been interpreted on the basis of the standard procedure developed for the NDACC international network of stations (WinDOAS [19]).

The spectrum of solar radiation scattered into the zenith is usually represented in the form of the Bouguer–Lambert–Beer law:

$$I(\lambda_l, z) = I_{\text{ref}}(\lambda_l)\exp\left(-\sum_{i=1}^{N} c_i\alpha_i(\lambda_l)m_i(z)\right), \quad (1)$$

where $I(\lambda_l, z)$ is the measured intensity of the solar radiation scattered into the zenith at the wavelength λ_l and at the zenith angle of the Sun z, $\alpha_i(\lambda_l)$ is the spectral attenuation coefficient for the ith optically active component, c_i is its total content, and $m_i(z)$ is the related air-mass factor. The extrapolated value of $I(\lambda_l, z)$ at $m_i(z) = 0$ obtained during a day with a stable optical weather is used as the reference intensity of the extraatmospheric radiation.

In this case, the observed optical thickness is

$$\tau(\lambda_l, z) = \ln(I_{\text{ref}}(\lambda_l)) - \ln I(\lambda_l, z) = \sum_{i=1}^{N}\alpha_i(\lambda_l)x_i(z) + \varepsilon_l, \quad (2)$$

where $x_i(z) = c_i m_i(z)$ is the slant atmospheric content of the ith component and ε_l are the effects of smoothly varying attenuation components and a possible contribution of the stray light of the spectrometer or the dark current of the detector. The typical optical densities of the main gas constituents (O_3, NO_2, H_2O, O_4) observed in the ground-based measurements on July 5, 2004, at sunrise (the zenith angle is ~90°) under the conditions of increased humidity are presented in Fig. 1. This figure shows the contribution of each of the four constitu-

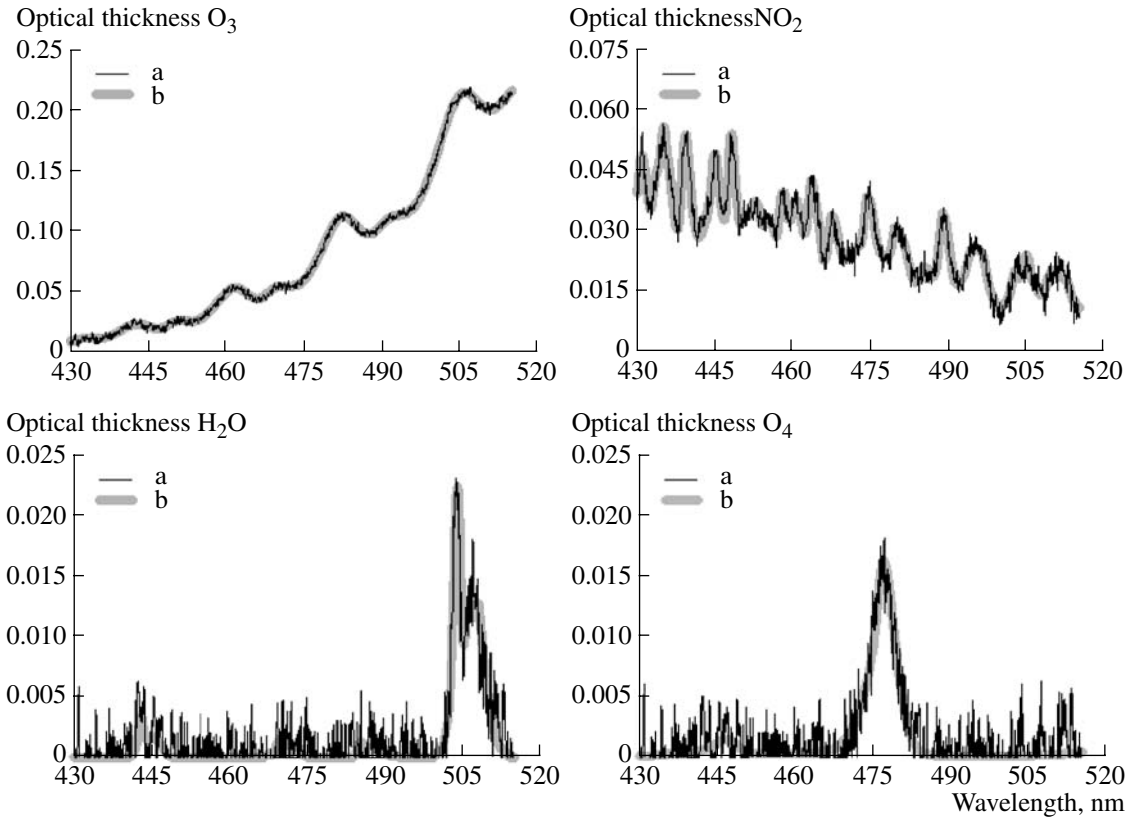

Fig. 1. Optical densities of the main gas constituents (O_3, NO_2, H_2O, and O_4) from the data of ground-based measurements on July 5, 2004, at sunrise (the zenith angle ~90°) under the conditions of increased humidity: (a) measured optical thickness and (b) optical thickness calculated for the reconstructed slant content.

ents to the spectral optical thickness for the data of measurements and the data calculated for the reconstructed slant content. As is seen from the figure, the optical thicknesses of H_2O and O_4 (<0.025) are nearly an order of magnitude smaller than that for ozone (up to 0.2) and several times smaller than the corresponding value for NO_2 (up to 0.055). At the same time, the errors of the method characterized by the noise of the measurement curve (~0.002) are fairly small, which makes it possible to reveal with certainty characteristic spectral features of all gases and separate their contributions to the absorption.

System of N equations (2) in x_i for a large number of wavelengths is solved by the least-squares method with the use of an optical model of measurements, which includes the following components: (i) the molecular absorption coefficients of O_3 [25], NO_2 [26], H_2O [27] and O_4 [28] and the absorption coefficients of O_3 and NO_2 taken at the temperatures 223 and 220 K, respectively; (ii) the 5th-degree polynomial taking into account the contributions of Rayleigh and aerosol attenuation and possible variations in the spectral sensitivity; (iii) the parameters of shift and stretch/squeeze of the spectral scale; (iv) the Ring–Shefov effect, which is included as an additional component with the absorption coefficient calculated in accordance with [29]; and (v) the spectral dependence of the degree of light polarization in the ASC.

To check the accuracy of NO_2 content measurements by the developed ASC, comparative measurements were performed at the Zvenigorod Scientific Station during the period from September 17 through September 27, 2003, simultaneously with the observations at the Institute of Atmospheric Physics, Russian Academy of Sciences (IAP RAS) [14]. An example of comparison of the slant NO_2 contents measured with two instruments (developed at the IOP SPBSU and IAP RAS) in the interval of zenith angles 84°–96° with the step 0.5° is presented in Fig. 2. For this comparison, we used the results of 259 simultaneous measurements during the period September 22–27 (at sunrise and sunset). Analysis of the discrepancies demonstrates good agreement of the data obtained (the rms discrepancy is 9.6%) characteristic of analogous comparisons of similar instruments [10–13].

3. EXAMPLES OF THE RESULTS OF OBSERVATIONS

The main results of measuring total NO_2 content at the IOP SPBSU in 2004–2006 are presented in Fig. 3 as the total NO_2 contents averaged for each day of twi-

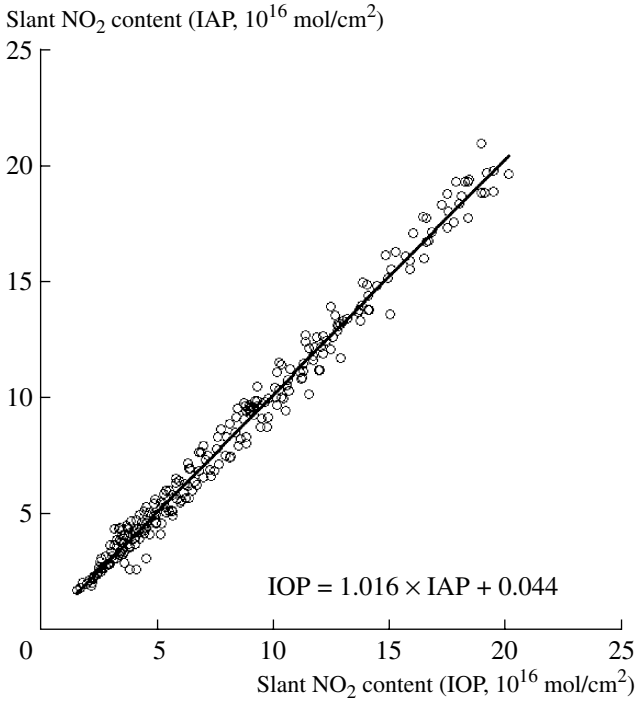

Fig. 2. Comparison of the slant NO_2 contents with the two IOP SPBSU and IAP RAS instruments at the Zvenigorod Scientific Station from September 22 to September 27, 2003.

light observations at sunrise and sunset. We used the NO_2 air-mass factor calculated for midlatitudes on the basis of the radiative-transfer model developed at the Belgian Institute for Space Aeronomy (the calculated air-mass factor is used at the majority of NDACC stations). On the whole, the seasonal variation of the total NO_2 content yielded by the results of measurements corresponds to the climatic data: this content is maximum in summer ($4–5 \times 10^{15}$ mol/cm^2) and minimum in winter (1×10^{15} mol/cm^2) because of the duration of daylight. At the same time, against the background of a smooth seasonal variation, one can note abrupt bursts of high values of the total NO_2 content (up to 10^{16} mol/cm^2), apparently caused by an increased NO_2 content in the troposphere under conditions of anthropogenic pollution. Indeed, if we separately consider the total NO_2 content measurements averaged over the time 10:00–14:00 (see Fig. 4), the results will demonstrate high values of the total NO_2 content (up to 5×10^{16} mol/cm^2), sharply changing during the year without any noticeable seasonal variation. As a rule, in the absence of tropospheric pollution, it is midday measurements of the solar radiation scattered into the nadir that are used as the reference extra-atmospheric spectrum, when the effect of NO_2 absorption at a high Sun is negligible. Evidently, owing to a relative nearness of the measuring instruments to the center of St. Petersburg, this condition is often not fulfilled, and the corresponding daylight measurements give an idea of the value and temporal variability of tropospheric NO_2.

3.1. Comparison with Satellite Measurements

At present, satellite measurements of the total NO_2 content are performed with three instruments: the ERS-2 GOME, the ENVISAT SCIAMACHY, and the AURA OMI. Because the GOME data have a low spatial resolution (~300 km) and the OMI data have been available only since September 2004, for comparison, we used the SCIAMACHY data, which are characterized by a relatively high spatial resolution (~40 km) and encompass the entire period of observations (2004–2006). The SCIAMACHY (Scanning Imaging Absorption Spectrometer for Chartography) instrument is mounted aboard the ENVISAT European satellite launched by the ESA in March 2002. The instrument can perform the nadir and limb soundings of the atmosphere by measuring the solar or lunar radiation in the ultraviolet, visible, and near infrared regions of the spectrum (240–2380 nm). The main goal of the experiment is to determine the global distributions of different minor gas constituents in the troposphere and stratosphere, such as O_3, NO_2, BrO, N_2O, CO, CO_2, H_2O, and CH_4 [7]. In this study, we used the results of interpretation of the SCIAMACHY nadir measurements obtained by the algorithm for retrieving the stratospheric NO_2 content that was developed at the University of Bremen (Germany) [20]. These data correspond to the SCIAMACHY measurement, nearest to the point of ground-based measurements located in a radius of 200 km from St. Petersburg, and are presented in Fig. 3. It is seen that the results of satellite and ground-based measurements of the total NO_2 content agree well with each other. It is known from the experience of validation of the data of SCIAMACHY and analogous satellite instruments (GOME, OMI) located on solar-synchronous orbits that the results of midday satellite measurements of the total NO_2 content are closer to the results of morning ground-based measurements than to the results of evening measurements [21]. This situation is also valid in the case of comparison of the SCIAMACHY data with the IOP SPBSU ground-based measurements near St. Petersburg (see Fig. 3). In this case, the mean discrepancy between satellite and morning ground-based measurements of the total NO_2 content was 0.07×10^{15} mol/cm^2 (~12%).

3.2. Variations in the Tropospheric NO_2 Content

It is important that the increased NO_2 content in the surface atmosphere is a direct threat to man and the environment because it produces photochemical smog and acid rains. Note that up to 70% of NO_2 emissions

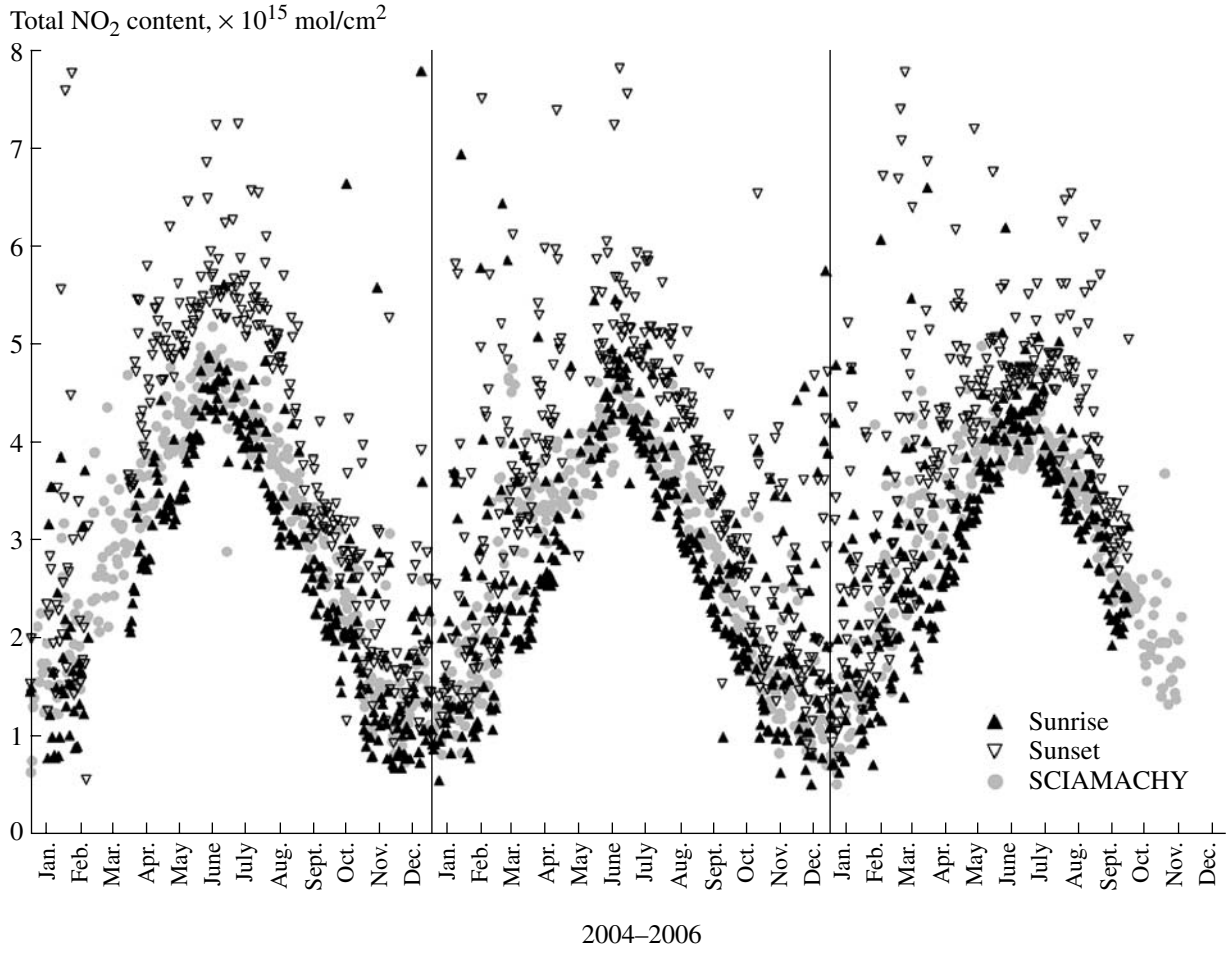

Fig. 3. Results of twilight ground-based measurements of the total NO_2 content by the IOP SPBSU and the data of satellite measurements near St. Petersburg (SCIAMACHY) in 2004–2006.

into the troposphere result from fuel combustion by motor transport and industrial plants. The anthropogenic contribution to the NO_2 content in the entire thickness of the atmosphere is so large that it often exceeds its natural level and becomes noticeable in observational data of satellite systems of global monitoring. In turn, this circumstance makes it possible to use the results of satellite measurements of the NO_2 as an indicator of anthropogenic pollution [22]. Analysis of satellite measurements of the total NO_2 content in the atmosphere of remote regions of the world ocean, where anthropogenic pollution is absent, allows the estimation of the stratospheric component in the total vertical NO_2 content. Under the assumption of a zonal structure of the global NO_2 distribution in the stratosphere, this quantity is subsequently used for extraction of the stratospheric component from the total NO_2 content at a given latitude and, accordingly, for determining the NO_2 content in the troposphere [23]. For example, the annual mean distribution of the NO_2 content in the troposphere of the Baltic region (central and northern Europe and northwestern Russia) in 2003–2005 was obtained in such a manner. This distribution is presented in Fig. 5. The map is constructed on the basis of the joint interpretation of SCIAMACHY measurements at the Belgian Institute for Space Aeronomy and the Royal National Institute of the Netherlands (http://www.temis.nl/). As is seen from Fig. 5, the increased (up to 1.9×10^{16} mol/cm^2) NO_2 content in the southwestern part of the region (Belgium, Holland, Germany) and local maxima near Moscow (2.4×10^{16} mol/cm^2) and St. Petersburg (1.0×10^{16} mol/cm^2) are the main features of the field of tropospheric NO_2 in this territory. Analysis of analogous maps constructed by us on the basis of long-term GOME measurements likewise displays relatively high contents of tropospheric NO_2 near Moscow and St. Petersburg. We considered the annual mean contents of tropospheric NO_2 near both cities for the entire period of GOME satellite measurements in order to reveal a possible temporal trend (Fig. 6). Although, as is seen from Fig. 6, the series of contin-

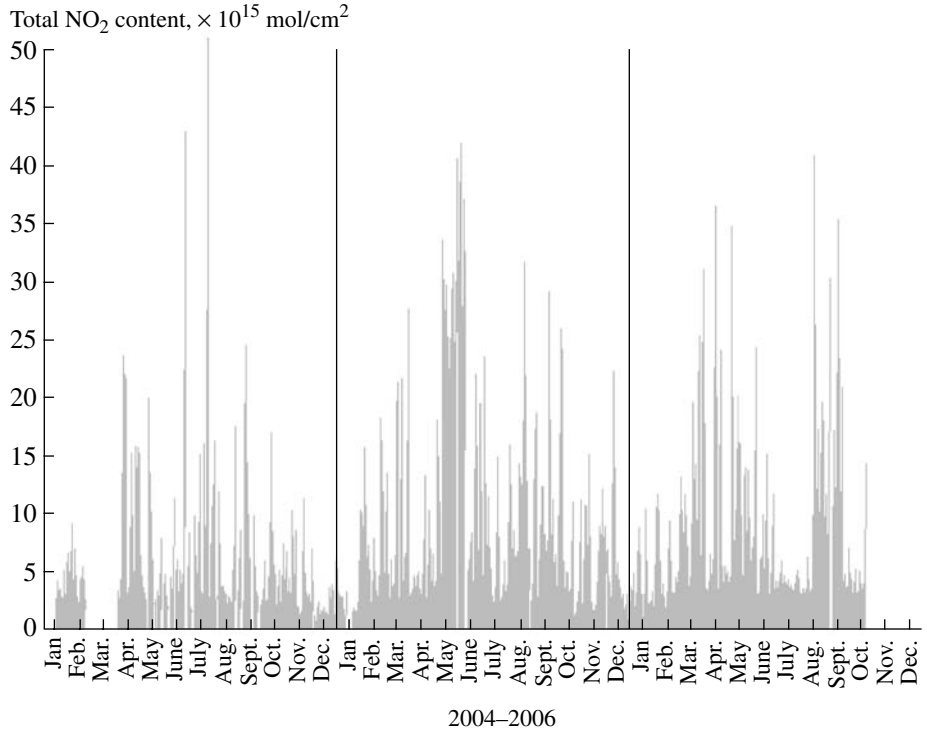

Fig. 4. Results of daytime measurements of the total NO_2 content by the IOP SPBSU averaged over the time 10:00–14:00.

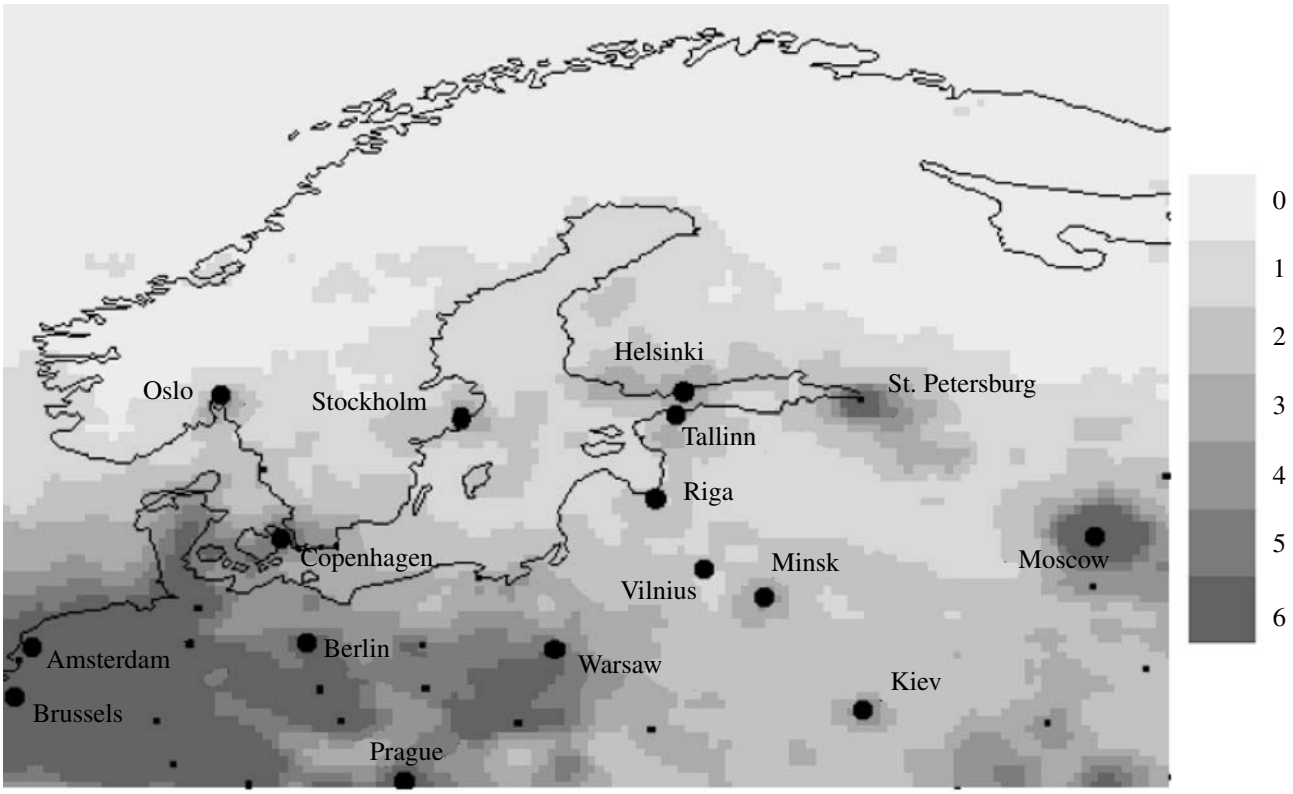

Fig. 5. Annual mean distribution of the NO_2 content (10^{15} mol/cm^2) in the troposphere of the Baltic region (central and northern Europe, northwestern Russia) based on the interpretation of SCIAMACHY satellite measurements in 2003–2005 (http://www.temis.nl).

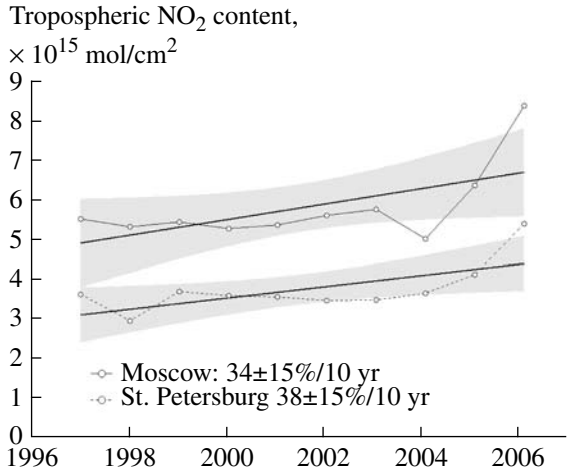

Fig. 6. Annual mean values of tropospheric NO_2 near Moscow and St. Petersburg during the period of GOME satellite observations. The straight line is the linear approximation of data by the least-squares method; the confidence level of the approximation (95%) is shown in gray.

uous observations available to date is insufficiently long for reliable estimations, noticeable positive trends (about 34 ± 15% and 38 ± 15% over ten years for the Moscow and St. Petersburg regions, respectively) are present in these data. These estimates are very approximate because the accuracy of identification of the tropospheric contribution in the results of satellite measurements of the total NO_2 content depends on a number of factors that are disregarded here: the feasibility of the assumption on a strictly zonal distribution of stratospheric NO_2 and the error in determination of the tropospheric air-mass factor, which is necessary for the conversion of the slant content of tropospheric NO_2 to the desired vertical content. In turn, an accurate calculation of the tropospheric air-mass factor calls for the knowledge of the cloud-cover amount and height, surface albedo, and an a priori NO_2 profile in the troposphere. In this study, we used both the minimum total NO_2 content at a given latitude for estimating the stratospheric contribution and the fixed tropospheric air mass equal to 1.

In this regard, independent measurements of the NO_2 content under the conditions of anthropogenic pollution of the troposphere are of particular interest for the validation of satellite data. Thus, an analogue of the method described above has been realized in the algorithm for operational processing of the OMI (Ozone Monitoring Instrument) data and makes it possible to supply users not only with data on the total NO_2 content but also with data on the tropospheric NO_2 content. An example of comparison of the OMI measurements near St. Petersburg with the simultaneous ground-based observations at the IOP SPBSU (Petrodvorets) is shown in Fig. 7. The results of ground-based measurements are presented as the daylight NO_2 contents averaged over the time 10:00–14:00, which are close in time to satellite measurements. The results of measurements of the surface

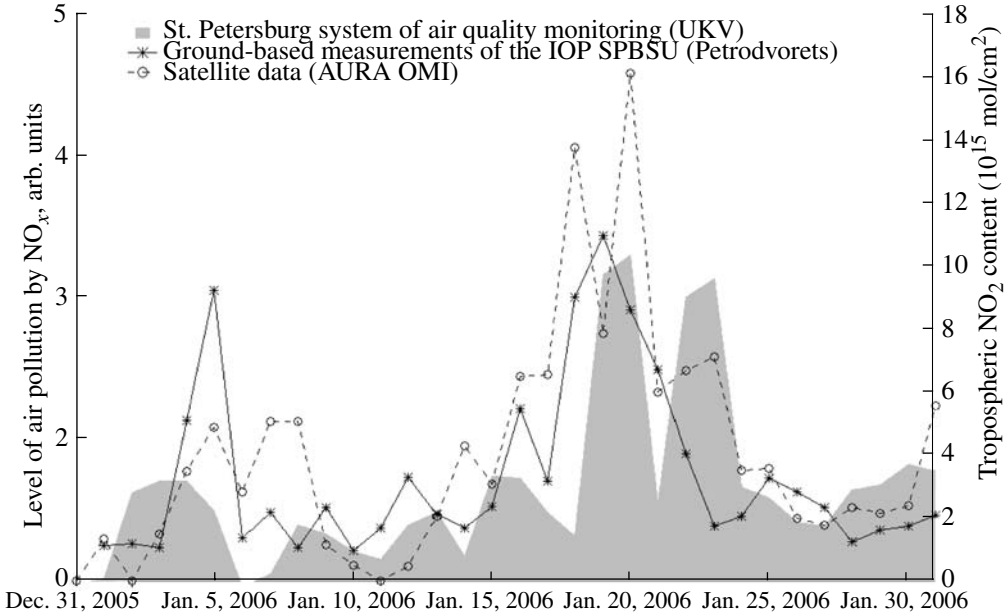

Fig. 7. Comparison of satellite measurements of the tropospheric NO_2 content (OMI) near St. Petersburg with the results of simultaneous ground-based observations (IOP SPBSU, Petrodvorets) and direct measurements of the surface NO_x concentrations at the stations of the urban monitoring of air quality (http://gov.spb.ru).

NO_x ($NO + NO_2$) concentrations at the stations of the Automatic System of Atmospheric Air Quality Control in St. Petersburg (UKV system [24]) are shown in the plot on a separate scale. The UKV automatic stations for monitoring atmospheric air pollution function continuously and ensure a regular (every 20 min) rapid supply of information about the level of pollution of the St. Petersburg air by the main pollutants: CO, NO, NO_2, SO_2, O_3, and NH_3. The UKV system includes 14 stations of the pavilion type equipped with chemiluminescent gas analyzers of nitrogen oxides. For estimating the air-pollution level, it is customary to present the summarized concentration $NO + NO_2$ (or NO_x), because these constituents are closely related via mutual photochemical transitions (oxidation and photodissociation), and their sum is stable under ordinary conditions. The UKV data are presented as the average (over all city stations) daylight value of the NO_x concentration, which is called the conditional index of pollution and varies from 0 to 10 (the index values 2.0–2.5 correspond to the average pollution over all Russian cities). These data are available at the official internet site of the Committee on Nature Management, Environmental Protection, and Environmental Safety of the St. Petersburg Goverment (http://gov.spb.ru). According to the information of the Committee of February 2, 2006, adverse meteorological conditions favoring an increase in pollutant concentrations were observed in the surface atmospheric layers as a result of intense anticyclonic activity in the period of cooling from January 17 to January 25, 2006. The situation that took place in St. Petersburg during these days was recorded at 17 automatic stations of air quality control. A low wind speed and the presence of moderate and strong temperature inversions led to a significant pollution of atmospheric air on January 20 and 23. As is seen from Fig. 7, satellite and ground-based measurements of the NO_2 content in the troposphere agree well with each other and qualitatively reproduce variations fixed in the results of observation of surface concentrations (the UKV system).

4. CONCLUSIONS

An automatic spectral complex developed at the IOP SPBSU has been described. This complex is used for regular ground-based spectroscopic measurements of the total NO_2 content in a vertical column of the atmosphere during the twilight and daylight hours of the day near St. Petersburg (Petrodvorets). The developed instruments allow almost continuous measurements of the spectra of solar radiation scattered into the zenith in the spectral region 428–515 nm (with the resolution 1.3 nm) during both twilight and daylight hours. The slant and vertical NO_2 contents are determined on the basis of the standard algorithm of DOAS measurements developed for the NDACC international network of stations. A number of ground-based twilight measurements of the total NO_2 content were obtained near St. Petersburg in 2004–2006, and variations in the tropospheric NO_2 content were estimated from the results of daytime ground-based measurements.

Examples of comparison of the results of ground-based spectroscopic measurements of the NO_2 content in 2004–2006 near St. Petersburg with the data of satellite observations and the results of direct measurements of the NO_2 concentration in the surface air are presented. It is shown that the IOP SPBSU ground-based measurements near St. Petersburg (Petrodvorets) are subject to a considerable anthropogenic pollution from this megalopolis, which is recorded in daytime spectroscopic measurements, sensitive to variations in the tropospheric NO_2 content. At the same time, the twilight ground-based measurements of the total NO_2 content agree relatively closely with the results of satellite measurements (for example, SCIAMACHY)—their average discrepancy is 0.07×10^{15} mol/cm^2 (~12%). On the basis of satellite measurements over the period 2003–2005, an example of the spatial annual mean distribution of the NO^2 content (central and northern Europe, northwestern Russia) is presented to demonstrate the main sources of anthropogenic pollution. An increase in the annual mean values of the tropospheric NO_2 content near Moscow and St. Petersburg for the entire period of satellite observations with the GOME instrument are preliminarily estimated at about 30–40% over ten years.

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