

Analysis of Variability of the CO, NO₂, and O₃ Contents in the Troposphere near St. Petersburg

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Abstract—The space–time variability of the fields of CO, NO₂, and O₃ concentrations and contents in the troposphere of northwestern Russia is analyzed on the basis of experimental data and the results of numerical modeling. The influence that the St. Petersburg emission has on the concentrations and contents of CO, NO₂, and O₃ in the troposphere is estimated for March 2006. A comparison of the measurements of the total CO content and the tropospheric NO₂ content with the results of modeling showed a qualitative and, in some cases, quantitative agreement between the results of calculations and experimental data. When synoptic conditions are determined, the St. Petersburg train can be detected at a distance of more than 300 km, which can affect the atmospheric air quality in adjacent countries.

Keywords: numerical modeling, air quality, spectral measurements, carbon monoxide, nitrogen dioxide.

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1. INTRODUCTION

An investigation into the thermal, dynamic, and chemical processes in the atmosphere affecting the climate of the earth is one of the central problems of modern atmospheric physics and chemistry [1]. The pollution of atmospheric air in industrial and urbanized regions leads to the formation of chemically active substances, which have a harmful influence on human health [2]. At present, the atmospheric air quality is investigated, controlled, and operationally predicted on the basis of the results of calculations of modern transport–photochemistry models (such as CHIMERE, MOCAGE, MM5-CAMx, BOLCHEM, CMAQ, and others), databases on the inventory of pollution sources, and experimental data on the concentrations of harmful substances in atmospheric air (national and international measurement networks and satellite measurements) [2–4]. Carbon monoxide (CO), nitrogen dioxide (NO₂), and ozone (O₃) are among the most important gaseous components of atmospheric chemistry that indicate anthropogenic air pollution. The northwestern region of the Russian Federation (RF) chosen for investigating the influence that anthropogenic emissions have on the NO₂, CO, and O₃ concentrations is one of the most densely populated industrial regions of Russia.

2. EXPERIMENTAL DATA

The results of spectral measurements of CO and NO₂ (no data on O₃ for the period under investigation

were available for us) obtained at the Fock Institute of Physics (FIP), Faculty of Physics, St. Petersburg State University (SPbSU) were invoked as experimental data. The measurements were conducted in Petrodvorets, which is a suburb of St. Petersburg located 30 km from the center of the megalopolis. The geographic coordinates of this point are 59.88° N and 29.83° E.

2.1. Carbon Monoxide

The CO content over the entire thickness of the atmosphere has been measured from ground-based spectra of direct solar radiation since 1995 [5]. The spectra are recorded in the presence of cloudiness breaks (~15 min are sufficient for recording a spectrum) or in conditions of cloudless weather with a solar IR spectrometer (SIKS) in the spectral region 2140–2180 cm⁻¹ with the resolution ~0.5 cm⁻¹. The error of daily mean values of the total CO content is 1–4% [5]. The available array of measurement results makes it possible to analyze the variability of the total CO content for different time scales—from the daily trend to a long-term component [5]. The variability of the total CO content in the atmosphere is rather large: the amplitude of its seasonal trend is 20–25% of the mean value; the presence of regional sources of powerful forest fires can thrice increase the total CO content. The variability of the total CO contents recorded for neighboring days of measurements is usually no more than 5%; however, in some cases, a substantially larger vari-

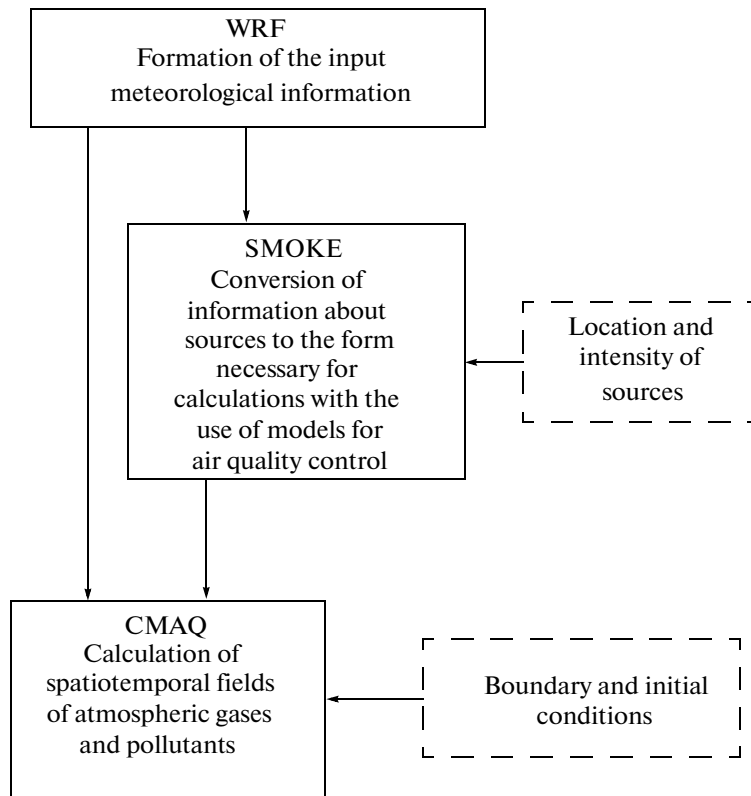


Fig. 1. Block diagram of the model calculations.

ability is observed. An analysis of the results of spectroscopic measurements for March 2006 showed the presence of significant variations in the total CO content, which took place for several days and were equal to ~40% of the monthly mean values in March. A study of such cases makes it possible to reveal the most important factors responsible for the observed variations in the total CO content.

2.2. Nitrogen Dioxide

The FIP (Faculty of Physics, SPbSU) started measuring the total NO₂ content in 2002 [6]. A ground-based automatic spectral complex recording the spectra of visible solar radiation scattered into the zenith is used for the purpose. The instrumentation used is a spectrometer based on the industrially manufactured Russian MDR-12 (LOMO) scanning monochromator with diffraction grating, ensuring a step of spectrum scanning of ~0.05 nm. The spectra in the region ~428–515 nm are measured with the spectral resolution ~1.3 nm. The NO₂ content in the troposphere is determined with the use of the DOAS (Differential Optical Absorption Spectroscopy) method of differential absorption from the results of zenith measurements in the visible region of the spectrum.

The measurement error of the effective NO₂ content on a path with a sufficient absorption (optical thickness >0.01) is 10–20% [6, 7]. In order to determine the true NO₂ content in the vertical column of the troposphere, it is necessary to specify a large amount of a priori information whose essential uncertainty considerably reduces the accuracy of these measurements. With allowance for the unavoidable influence of cloudiness, the total error of satellite measurements of this type can be 50–100% [8]; the preliminary estimates of the error of ground-based measurements yield analogous values. The accumulated multiyear series of virtually uninterrupted daily measurements makes it possible to reveal the characteristic features of the temporal variability of the total NO₂ content—the weekly cycle and the seasonal trend [9]. The NO₂ content in the troposphere is characterized by very sharp and significant daily variations, especially near sources of anthropogenic pollution. Depending on the wind direction and velocity, the NO₂ content can change several times. According to the measurements near St. Petersburg, the mean amplitude of the seasonal trend of the total NO₂ content can be about 70% [9].

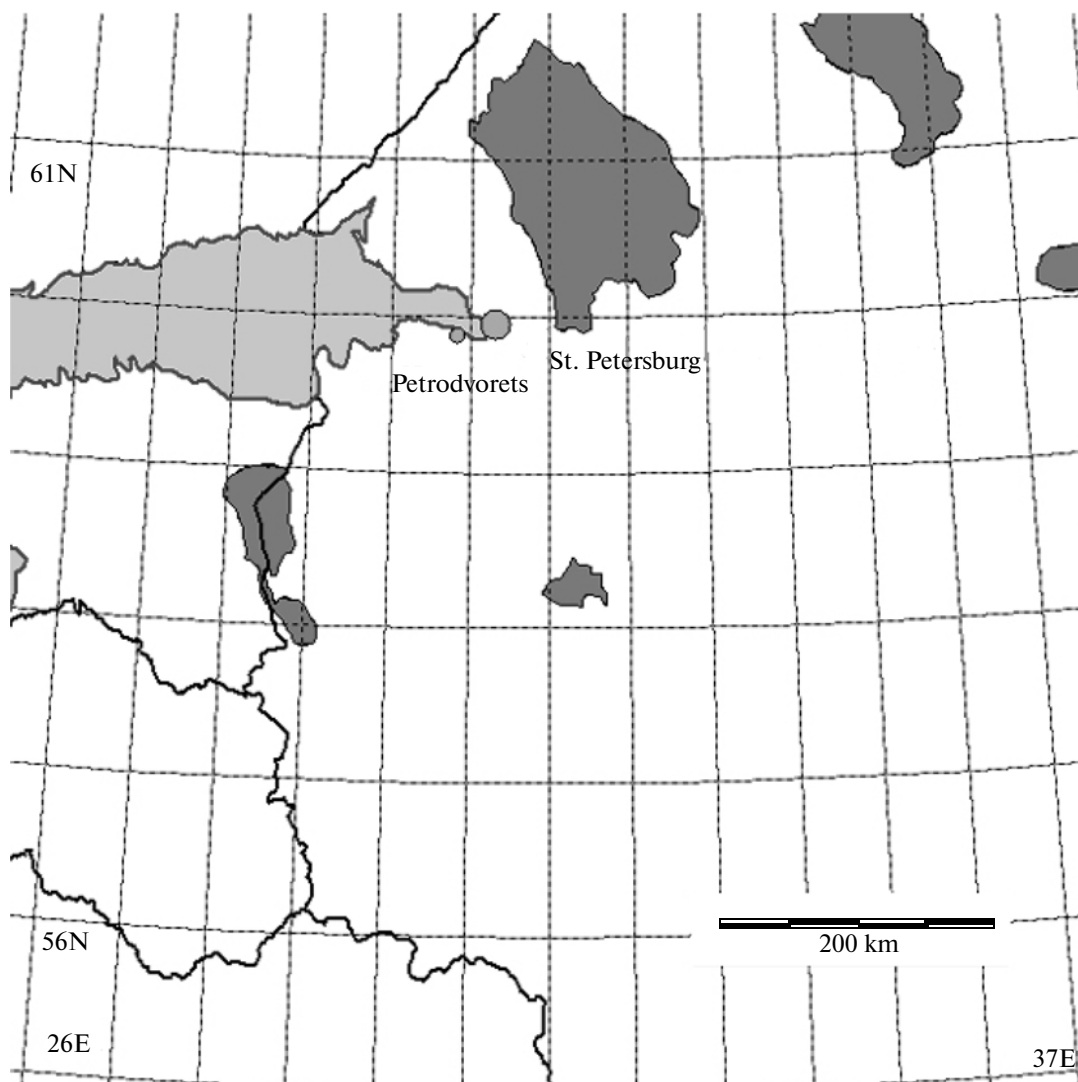


Fig. 2. Territory used for the modeling.

3. MEANS FOR MODELING THE FIELDS OF ADMIXTURE CONCENTRATIONS IN THE TROPOSPHERE

We used the Community Modeling and Analysis System (CMAS) for numerical modeling [10], whose

Data on the annual emissions of CO, NO_x, NH₃, SO₂, and volatile organic compounds (VOC) for St. Petersburg

Annual emission for St. Petersburg, thousands of tons

CO	NO _x	NH ₃	SO ₂	VOC
555	38.2	—	12.4	22.4

main module is the Community Multiscale Air Quality (CMAQ) transport–photochemistry model [11], in this work. Figure 1 illustrates the general structure of model calculations.

The input meteorological information for the CMAQ model is formed with the aid of the Weather Research and Forecasting (WRF) model, which is the mesoscale meteorological model of a new generation used for weather forecasting and investigations [12]. The NCEP FNL meteorological data measured every 6 h with a spatial resolution of 1.0° × 1.0° in latitude and longitude are used in the calculations [13].

The Sparse Matrix Operator Kernel Emissions (SMOKE) module is intended for converting the information about sources to the form necessary for calculations with the use of models controlling the air quality [14].

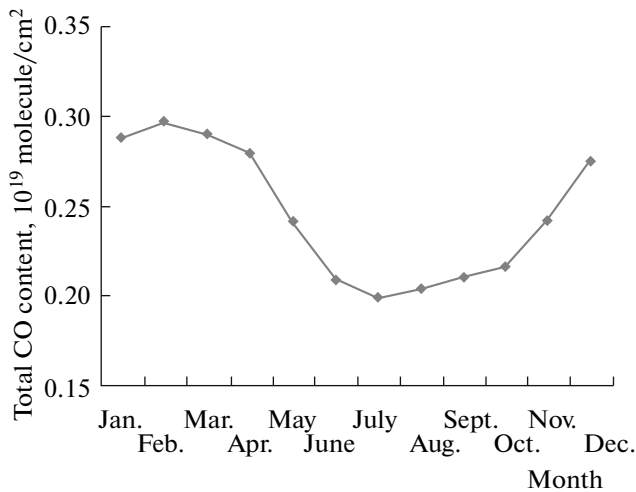


Fig. 3. Annual trend of the total CO content from the data of ground-based spectroscopic measurements at the FIP, Faculty of Physics, SPbSU.

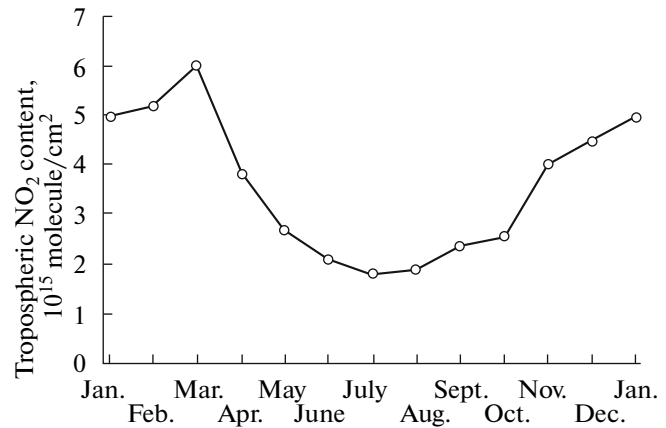


Fig. 4. Annual trend of the tropospheric NO₂ content near St. Petersburg (from the data of GOME, SCIAMACHY, and OMI satellite measurements in 1996–2009).

The CMAQ is a model of the third generation developed both for applied problems of controlling and predicting the atmosphere state and for scientific problems of investigating the complex of complicated chemical and physical processes in the atmosphere. This model works on different spatial scales (from local (1 km) to hemispheric) and makes it possible to calculate the concentration fields of different chemically active constituents in the troposphere, such as CO, NO₂, NO, O₃, volatile organic compounds, and aerosols (PM_{2.5}, fractions to 2.5 μm, and PM₁₀, fractions to 10 μm); the deterioration of visibility; and the fallout of acid precipitation. It also makes it possible to predict phenomena of the formation and destruction of some chemically active constituents of the troposphere (for example, ozone and nitrogen oxides).

The calculations were performed for two model domains for the period March 1–23, 2006, with the time resolution 1 h. The territory ~800 × 800 km (spatial resolution 15 × 15 km), including the northwestern region of the RF and parts of adjacent countries, formed the enclosed domain (Fig. 2).

3.1. Data on Emissions of Pollutants

Data on the emissions of the most powerful anthropogenic sources in the northwestern territory of the RF, i.e., cities and towns with more than 20000 people, were prepared as the initial information for the SMOKE module. Only the largest populated points, such as Helsinki and Tallinn, were taken into account outside Russia.

The official data over 2005 [15, 16] and the published data [17] were used as the initial data on the emissions of CO, NO_x, NH₃, SO₂, volatile (not methane) organic compounds (NMVOC), and aerosols (PM_{2.5} and PM₁₀) for the modeling domain. In the

absence of information about emission sources for a concrete populated point, we took as a basis the emission values for all of the components taken into account for the inhabitants of St. Petersburg and multiplied them by the number of dwellers of the populated point in question. The data on emissions for St. Petersburg, which were used as the initial information, are presented in the table.

Note that the intensities of pollutant emissions were specified with allowance for the annual (Figs. 3, 4), weekly (Fig. 5), and daily (Fig. 6) trends, which were determined from the results of CO and NO₂ measurements for St. Petersburg and its suburbs, as well as from the published data for megalopolises [9].

3.2. Initial and Boundary Conditions

The vertical profiles of the CO concentration used as the initial and boundary conditions for the external

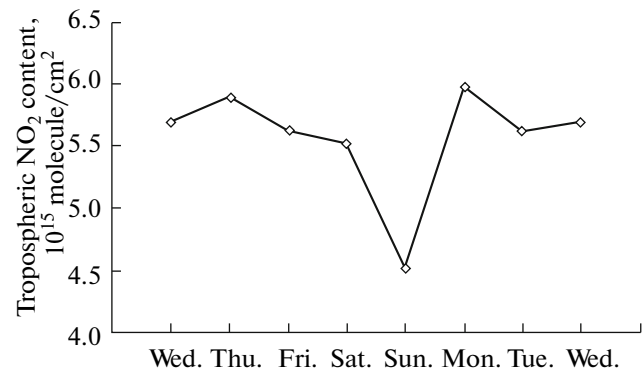


Fig. 5. Weekly trend of the tropospheric NO₂ content near St. Petersburg (from the data of OMI satellite measurements in 2004–2009).

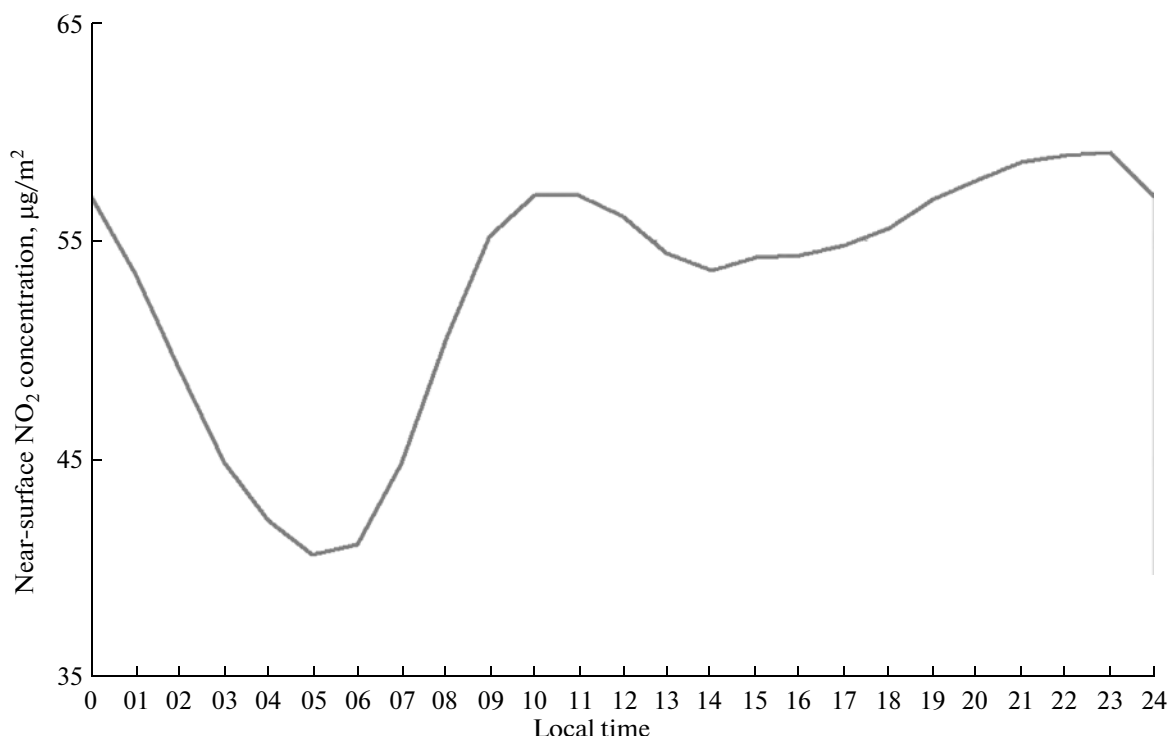


Fig. 6. Diurnal trend of the near-surface NO₂ concentration in St. Petersburg (from the data of the automatic system of atmospheric air monitoring in 2004–2007 [18]).

domain were constructed on the basis of an analysis of measurements of the CO total content and concentrations. In this case we used ground-based measurements in the NOAA (NOAA ESRL/GMD CCGG Cooperative Air Sampling Network) [20] and Network for Detection of Atmospheric Chemistry Composition (NDACC) [21] networks, as well as our own spectroscopic measurements (described above). The correspondence between the boundaries of the modeled territory and the network stations was as follows:

(i) the northern boundary: Kiruna (67.84° N, 20.41° E), Sweden, NDACC (total CO content); Pallas-Sammaltunturi (67.97° N, 24.12° E), Finland, NOAA (CO concentration);

(ii) the western boundary: Harestua (60.2° N, 10.8° E), Norway, NDACC (total CO content); Baltic Sea (55.35° N, 17.22° E), Poland, NOAA (CO concentration);

(iii) the southern and eastern boundaries: the results of an analysis of the total CO content at the FIP SPbSU, together with the HYSPLIT backward trajectories of motion of air masses (transboundary transport) [22].

The initial conditions for CO were Harestua, Norway, NDACC (total CO content); Baltic Sea, Poland, NOAA (CO concentration); and FIP SPbSU (total CO content).

The initial and boundary conditions for NO₂ in the troposphere were from the data of the two-dimensional transport–chemistry MPIC (Max-Planck-Institut für Chemie) model. The initial and boundary conditions for NH₃, SO₂, and volatile organic compounds were from climatic data available in the CMAQ model.

4. RESULTS AND DISCUSSION

Figure 7 shows examples of the spatial distributions of the CO, NO₂, and O₃ concentrations in the 0–25 m layer for 22:00 GMT on March 16, 2006, obtained with the aid of the CMAQ model. At the specified time, easterly and northeasterly winds prevailed and the southern coast of the Gulf of Finland (where the FIP is located) was in a cloud of urban emissions. The O₃ destruction during the interaction with nitrogen oxides (other processes affecting O₃ variations in the cold season have a substantially smaller influence) leads to the formation of a region of reduced concentrations of the surface ozone over St. Petersburg and its nearest vicinities [23]. The results of calculations of the temporal trends of the CO, NO₂, and O₃ concentrations in the 0–25 m layer for the geographic coordinates of the FIP are presented in Fig. 8. The periods of increased CO and NO₂ concentrations and reduced O₃ concentrations, corresponding to the location within the megalopolis train, are clearly recognizable

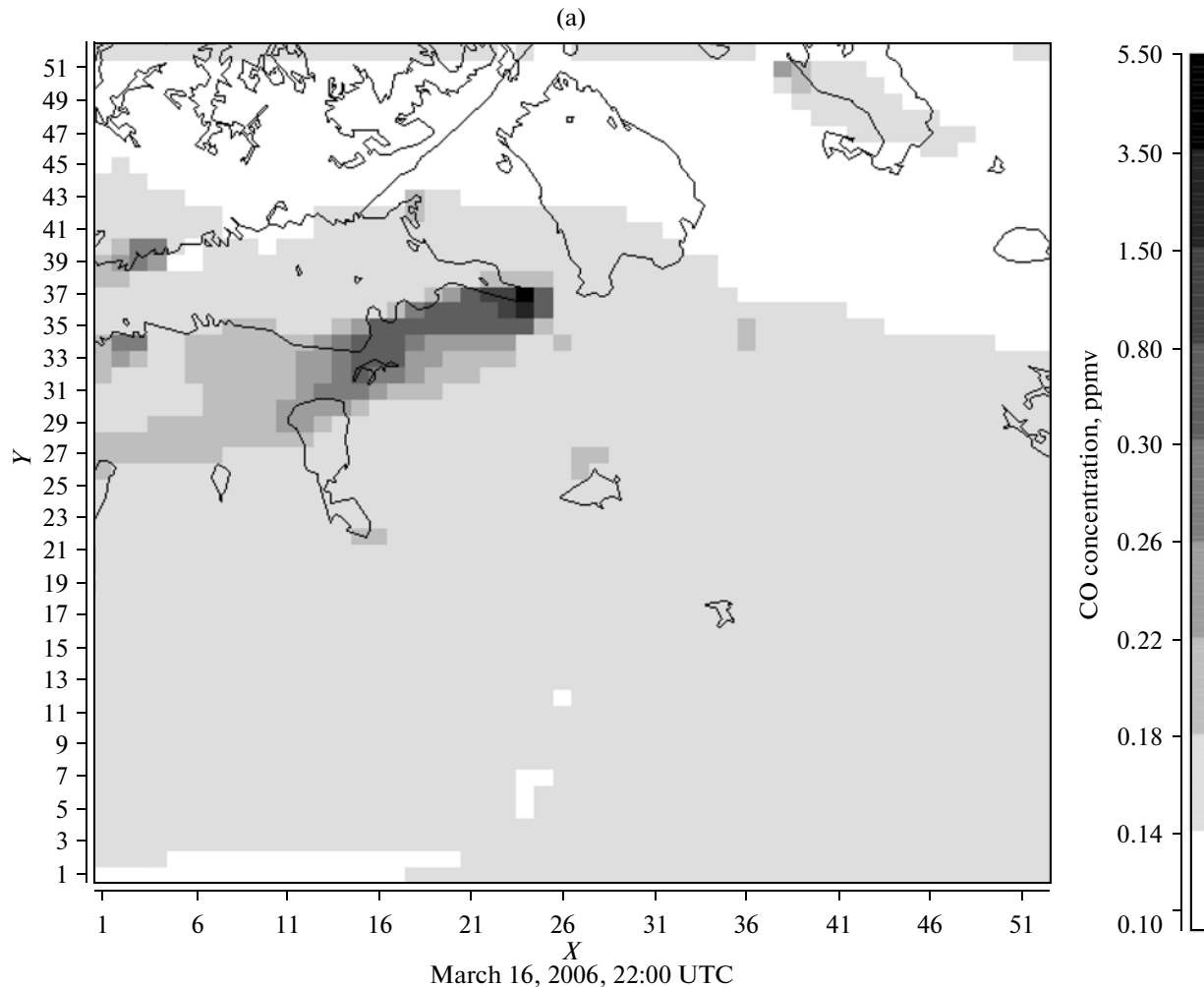


Fig. 7. Spatial distributions of the (a) CO, (b) NO₂, and (c) O₃ concentrations in the 0–25 m layer at 22:00 UTC on March 16, 2006, obtained with the aid of the CMAQ model.

in this figure. The points on the plots of concentrations corresponding to 22:00 on March 16, 2006, are marked by asterisks in Fig. 8; the values of the CO, NO₂, and O₃ concentrations themselves are also indicated there. According to the results of modeling, in the period of March 1–23, 2006, the maximal CO, NO₂, and O₃ concentrations for a suburb of St. Petersburg were 1.6 ppm (00:00 GMT on March 6), 42 ppb (07:00 GMT on March 9), and 40 ppb (13:00–14:00 GMT on March 23), respectively. In Fig. 8, the maximal concentrations are marked by triangles. The one-time maximum permissible concentrations (MPC) of these gases at the ambient air temperature 0°C are about 4 ppm (CO), 100 ppb (NO₂), and 75 ppb (O₃), which exceed the concentrations obtained by us in the modeling of the maximal CO, NO₂, and O₃ concentrations 2–3 times. The time variations in the O₃ and NO₂ concentrations are in antiphase, and higher O₃ concentrations comparable with seasonal mean

European concentrations (~40 ppb) are characteristic of “clean” air masses. As the results of modeling showed, in the lower layers of the troposphere, the influence of St. Petersburg emissions on the CO, NO₂, and O₃ concentrations is noticeable at a distance of ~300 km from the city center (which can be seen, for example, in Fig. 7). Therefore, when air masses are transported in the western and northwestern directions, the St. Petersburg train can be detected in the territories of adjacent countries (for example, Finland and Estonia). The occasional influence that St. Petersburg has on the air quality in neighboring territories will be manifested more strongly in the cold period, because pollutant emissions into the atmosphere increase and the air-mixing processes in the troposphere become weaker (anticyclonic conditions with a weak wind and the presence of inversions can lead to the formation of a train with high concentrations of pollutants).

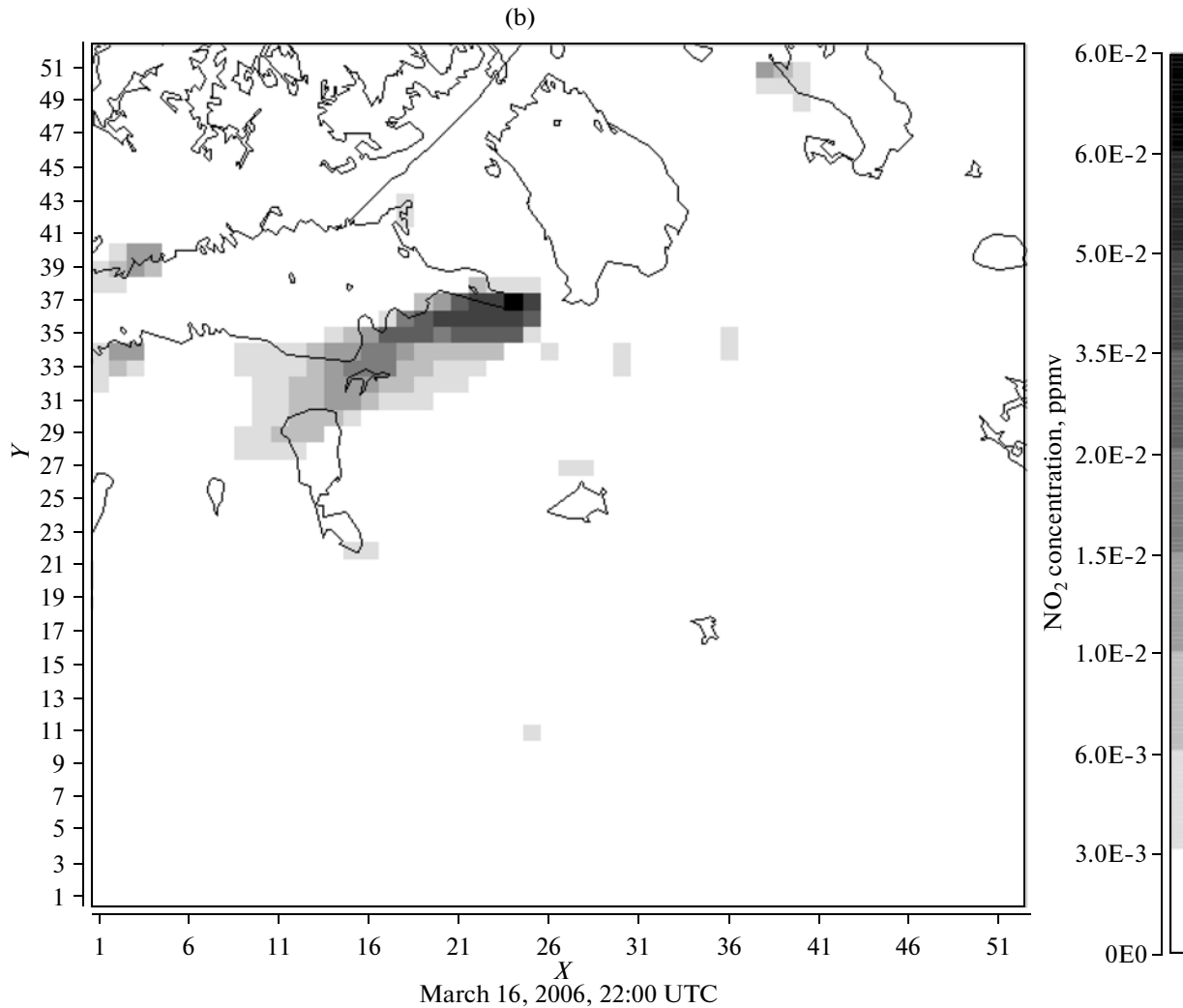


Fig. 7. (Contd.)

The reproduction of available experimental data by the model is one of the main components of analysis of the results of modeling concentrations of different constituents. For this purpose we compared the measurements of the total CO content and tropospheric NO₂ concentration (FIP, Faculty of Physics, SPbSU) with the results of calculations of the CMAQ model (for Petrodvorets). The qualitative and sometimes quantitative agreement between the results of calculations and the measurements (Fig. 9) points to the adequacy of the input information used for the CMAQ model (meteorological information; estimates of pollutant emissions for the domain of modeling; boundary conditions; and mechanisms and parametrizations used by the models). The input data on emissions did not include specific features of the spatial distribution of sources in the St. Petersburg territory (detailed information about the emission sources would proba-

bly allow us to obtain a better agreement between the model and the measurements). Such detailed data on emissions (unfortunately, we do not have such data at our disposal) become necessary for modeling with a high spatial resolution of the admixture concentration fields in St. Petersburg and its suburbs (i.e., in the immediate vicinity of sources). During the cold season, spectroscopic measurements of the total CO content in St. Petersburg are relatively rare (which can be seen from Fig. 9a) due to the small number of sunny days. Such measurements are usually accompanied by anticyclonic conditions (characterized by a weak wind, reduced temperatures, and the presence of inversions) and the arrival of continental air masses (with predominant easterly winds). In this case, the measurements are often conducted through “the hat” of admixtures formed over St. Petersburg. It can be seen from Fig. 9a that about half of the measurements

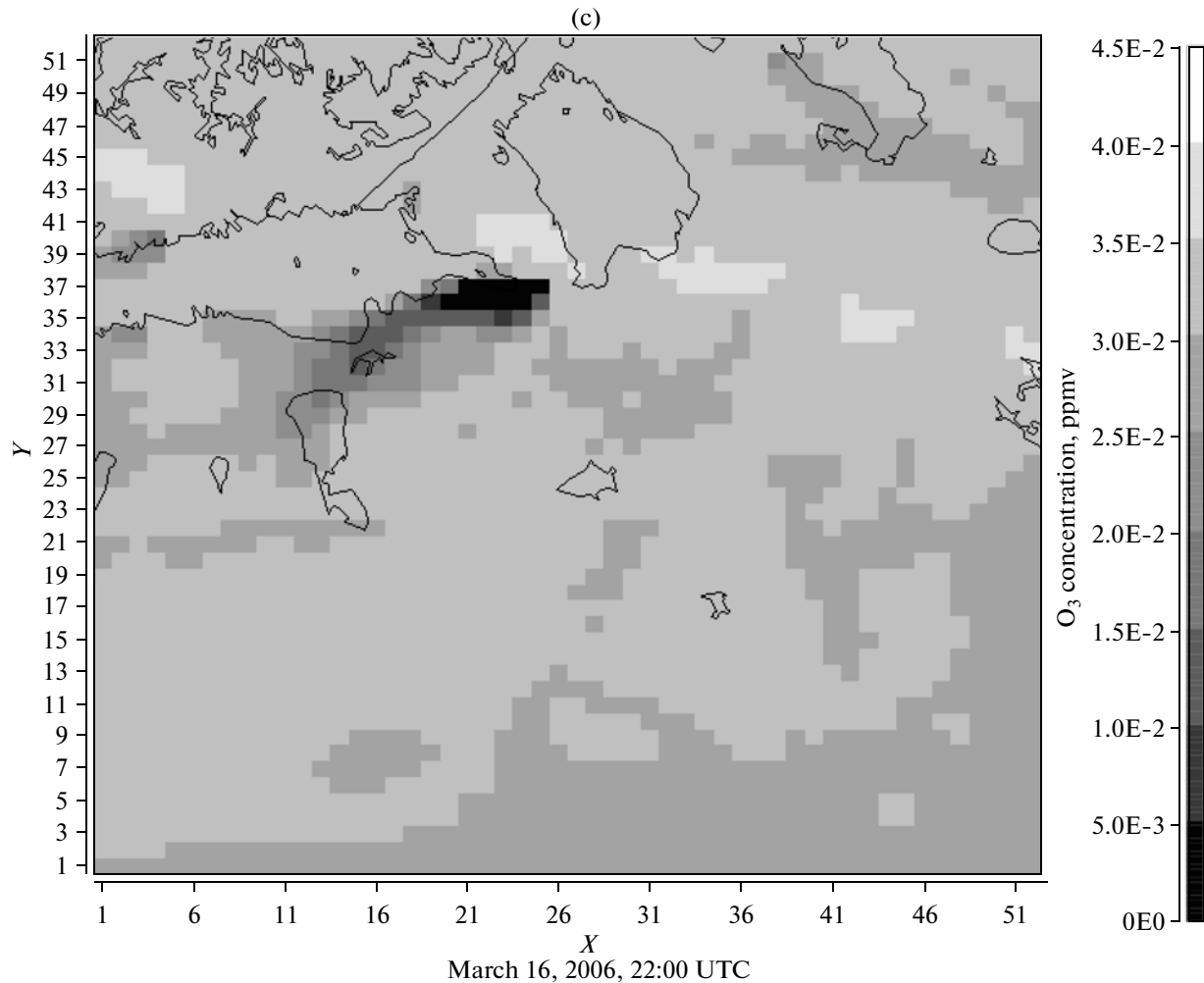


Fig. 7. (Contd.)

of the total CO content are precisely under such meteorological conditions. In the case of NO₂, the measurements of scattered solar radiation are much less restricted by weather conditions, and the periods of observations inside and outside the urban pollution cloud are more clearly recognizable in the series of values of the tropospheric NO₂ concentration (Fig. 9b).

A comparison of the results of modeling and experimental data shows that the contribution that the urban emission from St. Petersburg makes to variations in the total CO content can amount to 40% of the mean values. For the tropospheric NO₂ content, the influence of urban emissions is even more substantial: in the period under consideration, a 12-fold increase in the NO₂ content was observed, both from measurement data and from the results of modeling. Such a joint analysis of measurements and modeling results makes it possible to come to a conclusion on the correspondence (in a first approximation) between true

emissions and their estimates used in the model and to subsequently separate the arrays of measurements (in our case, the total CO content and the tropospheric NO₂ concentration) into the background part and the part disturbed by urban emissions, which is important for the correct investigation of space–time variations in the components under consideration. The latter must be taken into account if measurements are conducted near megalopolises such as St. Petersburg or Moscow [5].

The results of modeling can be of interest as a source of information about possible variations in these constituents in different layers of the troposphere directly near the point where spectral measurements are conducted (because the amount of data on the profiles of CO, NO₂, and O₃ concentrations is limited). Examples of the profiles of CO, NO₂, and O₃ concentrations are presented in Fig. 10 for the 0–2 km layer for the geographic coordinates of the FIP. In this

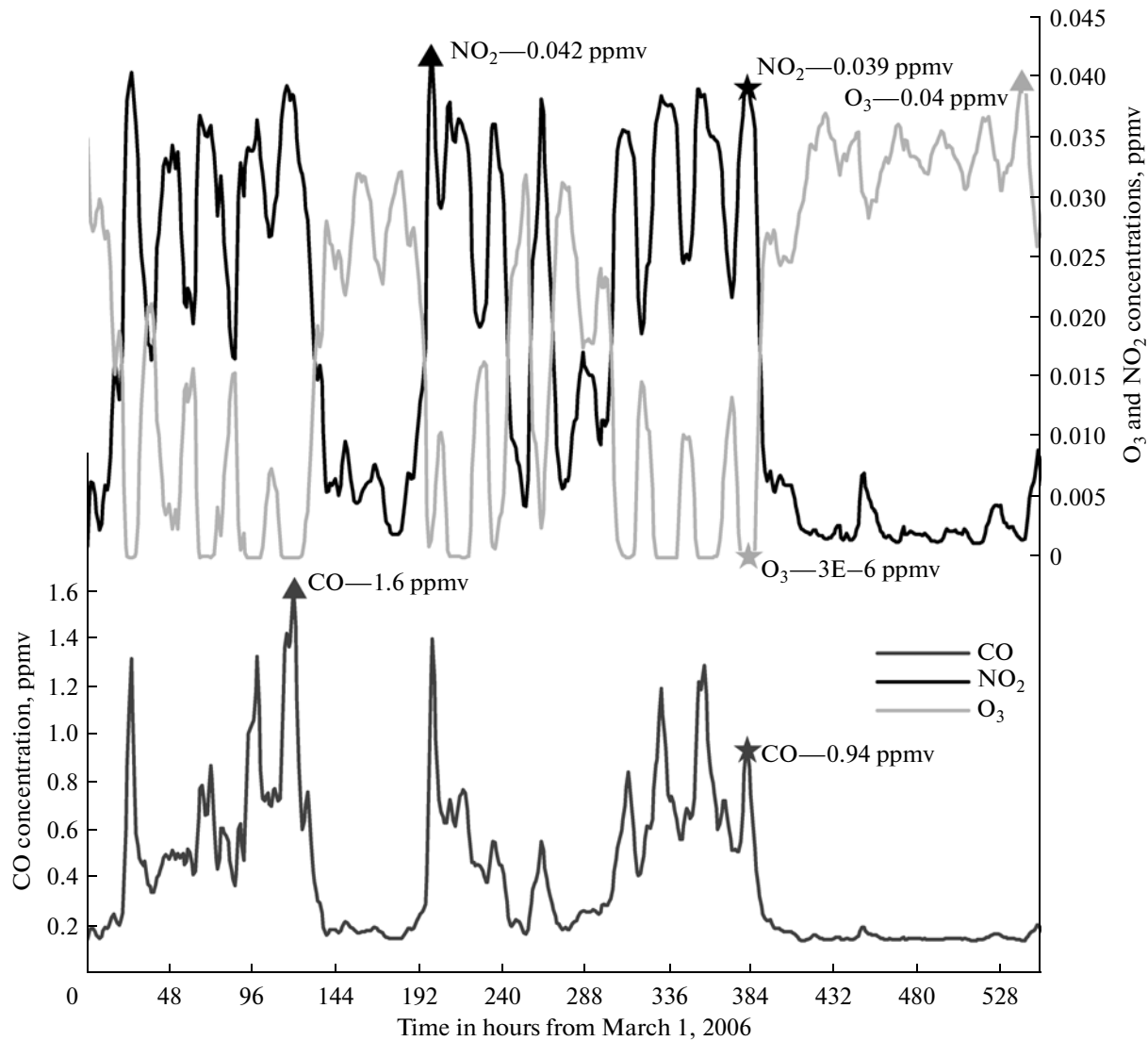


Fig. 8. Temporal trends of the CO, NO₂, and O₃ concentrations in the 0–25 m layer for Petrodvorets.

figure, the dashed lines indicate the profiles of gases for March 16, 2006 (corresponding to the situation reflected in Fig. 7), March 5, 2006 (when the highest concentrations of CO and NO₂ were observed), and March 19, 2006 (profiles of the CO, NO₂, and O₃ concentrations are close to the background ones). In the lower 500-m layer, the CO concentrations can sometimes increase the background values 10 times and the NO₂ concentrations can exceed the background values 20 times. As can be seen from Fig. 10c, high concentrations of nitrogen oxides almost completely destroy ozone in the lower layers of the troposphere. The influence of anthropogenic emissions from the megalopolis and specific chemical processes inside the train is restricted by the boundary-layer height, which usually does not exceed 1 km. The CO concentration in the free troposphere is mainly controlled by the remote

admixture transport (regional and global sources taken into account as the boundary conditions) and dynamic atmospheric processes. The influence of precisely these factors explains, for example, the observed distinctions between the CO profiles at heights exceeding 1 km (see Fig. 10a). For shorter lived chemically active gases (NO₂ and O₃), it is necessary to note photochemical processes (whose influence in winter is substantially smaller than in summer) along with the abovementioned factors. The obtained vertical distributions of gases can be used, for example, as a priori information on the problems of reconstructing the profiles of CO, NO₂, and O₃ concentrations or the total content of these gases obtained from the high-resolution IR spectra of direct solar radiation measured near St. Petersburg [24].

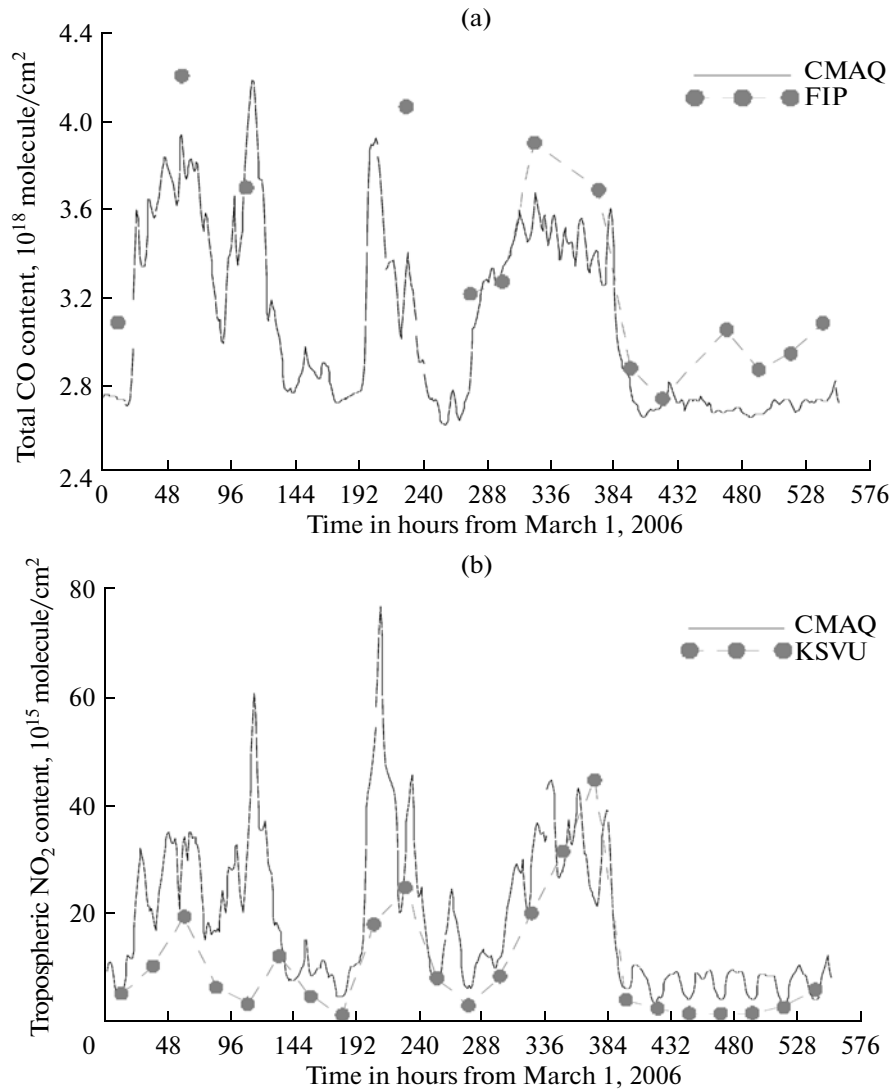


Fig. 9. Results of calculations of the CMAQ model and the measurements of (a) the total CO content and (b) the tropospheric NO₂ content.

5. CONCLUSIONS

An analysis of the spatiotemporal variability of CO, NO₂, and O₃ in the troposphere performed on the basis of measurements of the total CO content and the tropospheric NO₂ concentration near St. Petersburg and numerical modeling showed the following.

(1) Global sources and sinks control the observed annual trend of the total CO content, whose amplitude for northwestern RF is ~20–25%; minimal values are usually fixed in July–August, and maximal values are fixed in February–March. Against the background of the annual trend, variations in the total CO content of considerably smaller time scales (for example, one day or several days) are observed, which are caused by the influence of regional or local sources. In this case, the variability of total CO contents is usually no more

than 5%; however, in some cases, it can reach ~40–50% (March 2006).

(2) From the data of measurements near St. Petersburg, the mean amplitude of the seasonal trend of the tropospheric NO₂ concentration is ~70%. Significant sharp (by several times) variations in the tropospheric NO₂ concentration are observed near anthropogenic pollution sources depending on the meteorological situation (wind direction and velocity, the presence of inversions, etc.).

(3) A comparison of measurements of the total CO content and the tropospheric NO₂ concentration with the results of modeling showed a qualitative and, in some cases, quantitative agreement between the results of calculations and the experimental data.

(4) The contribution that the urban emission of St. Petersburg makes to variations in the total CO con-

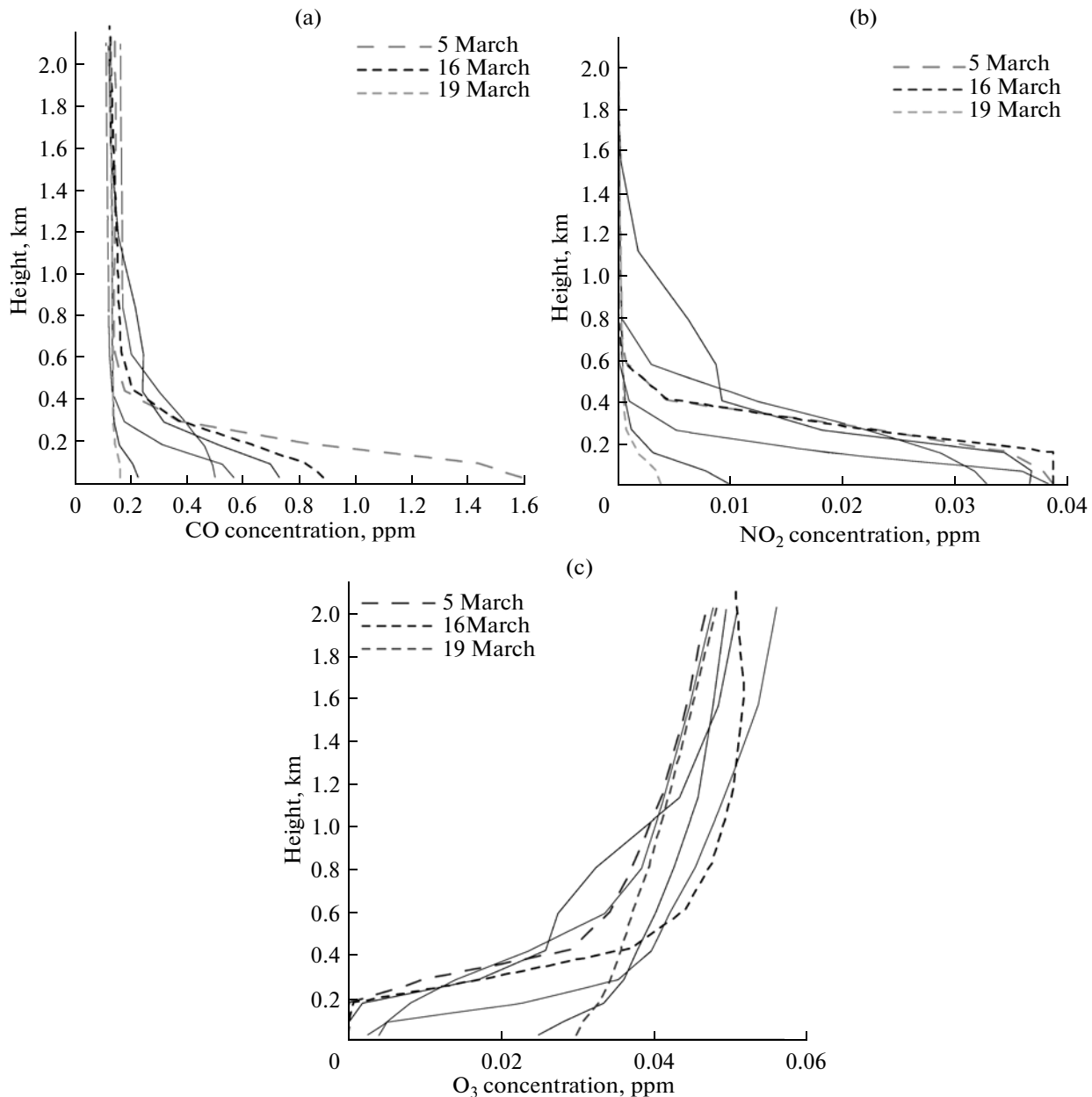


Fig. 10. Examples of model profiles of (a) CO, (b) NO₂, and (c) O₃ concentrations for Petrodvorets.

tent can amount to ~40% of the mean values. For NO₂, the influence of urban emissions can increase by twenty times its tropospheric concentration. This must be taken into account in an analysis of the results of measurements of total gas contents near megalopolises (such as St. Petersburg or Moscow).

(5) St. Petersburg is one of the main permanent disturbing factors (large cities such as Helsinki and Tallinn have smaller influences) responsible for the formation of heterogeneities in the field of CO, NO₂, and O₃ concentrations for northwestern RF. The results of modeling showed that emissions from

St. Petersburg can affect the air composition in lower layers of the troposphere at a distance of more than 300 km (including the territories of adjacent countries, for example, Finland and Estonia).

6. In the vicinity of the megalopolis (the nearest suburbs), in the lower 500-m layer, the CO and NO₂ concentrations can exceed the background values by ~10 and 20 times, respectively. The estimates of the minimal CO, NO₂, and O₃ concentrations for March 2006 obtained with the use of the CMAQ model for Petrodvorets were 1.6 ppm, 42 ppb, and 40 ppb, respectively, which are 2–3 times lower than the one-

time maximum permissible concentrations (MPC) for these gases.

(7) The model vertical distributions of CO can be used as a priori information in the problems of reconstructing the profiles of CO concentrations or total content from the high-resolution IR spectra of direct solar radiation measured near St. Petersburg.

(8) If the bases of data on sources of pollutant emissions into the atmosphere in the territory of Russia are available, the adapted (for northwestern RF) CMAQ model can be used as an effective tool for the analysis and prediction of the atmospheric air quality for different regions of the RF.

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