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Variation of Tropospheric NO_2 on the Territories of Saint Petersburg and Leningrad Region According to Remote Sensing Data

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Abstract—NO_2 is a reactive gas which is produced mainly due to man-made activities (burning of fossil fuels) and influences negatively people organisms and environment. Since the Earth population keeps increasing, the content of the gas in the atmosphere is expected to rise. Satellite monitoring is the most optimal method to observe the spatio-temporal distribution of NO_2 in the troposphere globally which cannot be achieved by ground-based measurements. However, there are several different satellite measurement systems which provide the information on tropospheric NO_2. In the current study we compared tropospheric NO_2 data for the more than 10-year period retrieved from the measurements of OMI and GOME/SCIAMACHY/GOME-2 satellite measurement systems for the territories of Saint Petersburg and Leningrad region (Russia). Also, we investigated correlation between the NO_2 tropospheric content by satellite measurements and near-surface NO_2 concentration by ground-based measurements in Saint Petersburg. The research demonstrated that OMI and GOME/SCIAMACHY/GOME-2 data on tropospheric NO_2 content possessed large discrepancies (approximately 100% relative to OMI data) for the area and period of interest. The datasets did not correlate well but some similarities in a seasonal variation of the tropospheric NO_2 content for Saint Petersburg and Leningrad region were found. In addition, we registered an obvious correlation in the trend of near-surface and tropospheric NO_2 content obtained by ground-based and OMI satellite measurements respectively.

Keywords: NO_2 content, tropospheric column, satellite measurements, OMI, GOME, SCIAMACHY, GOME-2, ground-based observations

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1. INTRODUCTION

The presence of some gases in the atmosphere influences the life of human beings significantly. They can have an impact on organisms and change an environment due to specific physical and chemical properties of some gases (e.g. dry and wet deposition, greenhouse effect, etc.) [1]. According to the data of the World Health Organization (WHO), approximately 7 million of people die every year due to exposure of polluted air. This is in the proximity of the population of Leningrad region and Saint Petersburg [2].

One of such gases is nitrogen dioxide (NO_2) which, together with nitrogen monoxide (NO), forms NO_x family. The changes in content of these gases in the troposphere and stratosphere depend on each other. NO and O molecules accordingly are formed as a result of NO_2 destruction due to photodissociation. In its turn, NO molecules are able to react with ozone (O_3) molecules producing NO_2 and O_2 [3]. NO_2 is a toxic reactive gas which is mainly produced in the troposphere as a result of anthropogenic activities such as the burning of fossil fuels (as well as NO). There are several known natural sources of the gas—biomass burning, lightning, and emissions from soil [4]. NO_2 influences organisms predominantly via respiratory pathways. According to the WHO report [5], the gas causes harmful effects on healthy human organisms from as low as 1 ppm concentration. However, people with chronic respiratory diseases can be under harmful influence even with smaller concentrations (0.2–0.3 ppm). The WHO recommends that maximum allowable NO_2 concentrations for 1 hour and 1-year exposure have to be under 0.1 and 0.02 ppm respectively. Yearly average NO_2 concentrations in urbanized areas are in a range 0.01–0.05 ppm while hourly average concentrations near automobile roads with heavy traffic can constitute 0.5 ppm. In addition to its toxic properties, NO_2 can influence the concentration of tropospheric ozone which is also a harmful gas [6, 7].

As a result of global population growth leading to an increase in fossil fuel consumption, the concentra-

Table 1. The main characteristics of satellite measurement systems monitoring tropospheric NO_2

Name	GOME	GOME-2	SCIAMACHY	OMI	TROPOMI
Max spatial resolution (km^2)	320 × 40	80 × 40	60 × 30	13 × 12	7 × 3.5 to 7 × 7
Swath (km)	960	1920	960	2600	2600
Spectral bands (nm)	240–790	240–790	240–2380	270–500	270–500, 675–775, 2305–2385
Time of full Earth coverage (days)	3	1.5	6	1	1
Period of measurements	1995–2011	From 2006	2002–2012	From 2004	From 2017

tion of NO_2 within the territories of large cities are expected to increase [1, 8]. For example, the amount of automobile transport in Saint Petersburg increased by a factor of 4 from 1990 to 2010 [9]**.** In addition, according to data from [10] the cargo turnover of Saint Petersburg seaport expanded approximately 2 times from 2000 to 2019. From the 1970s actions for regulating the amount of harmful pollutants emitted by automobile transport have been considering. Noticeable results in the reduction of NO_x emissions by automobile transport working on petrol engines had been reached by the beginning of 21th century. However, there was no significant reduction of the emissions of nitrogen oxides by transport working on diesel engines. In addition, the European emission standard Euro 5 was totally spread to cars in Russia only in 2016 when it was implemented in 2009. Even though, today a lot of developed countries move from polluting diesel engines to the cleaner petrol, electrical and hybrid ones. Therefore, it is most likely that increasing number of automobile transports will not be an explicit indicator of NO_2 increase in the atmosphere in the future [11].

Therefore, much attention in studies of atmospheric composition has to be devoted to the monitoring of the NO_2 spatio-temporal distribution. Since the main sources of gas are the man-made activities it may be assumed that the maximal concentrations are found in the troposphere. Today, NO_2 can be monitored by local and remote measurements [12–14]. Satellite remote observations are of prime interest since they, unlike local measurements, provide information on spatio-temporal variations of the pollutant and are available for more than ten-year period [15]. Analyzing the variation of NO_2 in the atmosphere it is maybe more reliable to use several sources of the satellite data. There are some known satellite measurement systems which provide global data of scattered solar radiation from which scientists are able to derive tropospheric NO_2. These are the Aura satellite (Ozone Monitoring Instrument or OMI, launched in 2004), ERS-2 (The Global Ozone Monitoring Experiment or GOME, 1995-2011), MetOp-A and MetOp-B (GOME-2, launched in 2006 and 2012 respectively) and ENVISAT (Scanning Imaging Absorption Spectrometer for Atmospheric Cartography or SCIAMACHY, 2002–2012). It is worthy to mention TROPOMI (TROPOspheric Monitoring Instrument), which has been launched on the Sentinel-5P satellite in 2017. However, its observation period is short and that is why it can be used for short special case analysis only. Some important characteristics of the mentioned satellite measurement systems are presented in Table 1 [16–27].

On the same method of the scattered solar radiation measurements by different satellites, the retrieved tropospheric NO_2 can differ. It can be related to the differences in spatial resolution and a spectral range of the instruments, mathematical algorithms of the tropospheric NO_2 retrieving, etc. [28].

In addition to the analysis of the NO_2 spatio-temporal distribution in the troposphere these data can have correlation with ground-based local measurements of near-surface NO_2 concentration. Modern investigations have already demonstrated that it is possible to retrieve near-surface NO_2 concentration using satellite data and numerical chemistry transport modelling. For example, it was shown in a study [29] that tropospheric NO_2 data according to OMI measurements can be used to estimate spatial variation of the NO_2 concentration near the Earth surface. This may be applied in ecological applications since the ground-based local measurements are irregular in space and cover only a relatively small volume of air when the satellites provide a global coverage.

The aim of this study is the comparison of tropospheric NO_2 data according to the satellite measurements of the OMI and GOME/SCIAMACHY/GOME-2 instruments for the territories of Saint Petersburg and Leningrad region during more than 10-year period. In addition, we assigned the task of the assessment of correlation between trends of tropospheric and near-surface NO_2 retrieved using satellite and local ground-based measurements respectively.

2. DATA AND METHODS

Satellite Data

We used two datasets of monthly average NO_2 content in the troposphere in this study. The data were based on satellite measurements by OMI (NASA Earth Observations, https://neo.sci.gsfc.nasa.gov) and GOME/SCIAMACHY/GOME-2 (GSG2, https://www.temis.n). The GSG2 is a combination of three different observation data. To retrieve NO_2 content in a tropospheric vertical column a method is used, according to which stratospheric NO_2 content is removed from the total NO_2 slant column. After that a produced tropospheric slant column is converted to the vertical tropospheric column of NO_2 [30, 31]. Spatial resolution of OMI and GSG2 datasets were 0.125° and 0.25° respectively. Both datasets covered a period from 2005 to 2017. Examples of the data are provided in Fig. 1.

To analyze the satellite data, we calculated spatial averages of the tropospheric NO_2 on the territories of Saint Petersburg (~1400 km^2) and Leningrad region (~84000 km^2) for every available time step. To find the average values and other statistical parameters (maximal and minimal values, standard deviation, etc.) we used ArcGIS tool "Zonal Statistics as Table". Discarded data were filtered out before the statistical analysis. We used data from May to September for every year only since it is assumed that the satellite measurements were the most accurate for these periods.

In situ Observations

The data of ground-based in situ measurements of near-surface NO_2 in Saint Petersburg were used in the current study. These data were presented in the report on ecological situation of the city in 2018 [32]. The measurements are carried out at 25 automatic stations located on the territory of Saint Petersburg (http://www.sc-mineral.ru/ru/p/air_rus/). A gas analyzer APNA-370 by HORIBA (https://www.horiba.com) is used at the stations for the NO_2 near-surface concentration observations. The instrument works on the basis of cross-flow modulated semi-decompression chemiluminescence method. It can provide continuous observations with a detection threshold equal to 0.5 ppb.

3. RESULTS AND DISCUSSIONS

3.1. Comparison of Tropospheric NO_2 Content According to OMI and GSG2 Data

Figure 2 presents a time series of monthly average tropospheric NO_2 content in Saint Petersburg area for 2005–2017 according to OMI (a) and GSG2 (b) data as well as their differences (c). The graphs depict maximal and minimal values, and standard deviations on the territory of interest for every time step. Significant differences between the datasets can be clearly seen in Fig. 2c. The OMI data on average were less than GSG2 on 280 molec. × 10^{13} cm^{-2} (or on approximately 100% related to OMI data). However, the differences raised up to 700 molec. × 10^{13} cm^{-2} during some periods when the differences between the maximal values constituted approximately about 900 molec. × 10^{13} cm^{-2}. The curve of differences (Fig. 2c) depicts some changes after 2013–2014 which started to vary with notable regularity having the maximal differences in May and minimal in June and July. Probably it was related to the completion of SCIAMACHY measurements in 2012 when the GOME and GOME-2 became the main instruments in GSG2 dataset. According to Figs. 2a, 2b, standard deviation of both datasets on average was quite similar (80–100 molec. × 10^{13} cm^{-2}).

Histograms in Fig. 3a characterize OMI and GSG2 monthly mean data distribution during 2005–2017 averaged for the territory of Saint Petersburg. Both histograms possess quite similar pattern of the data distribution. Nevertheless, a shift in the values can be seen. Perhaps it means that OMI and GSG2 data differ systematically. (approximately on 280 molec. × 10^{13} cm^{-2}).

NO_2 tropospheric content, according to the satellite observations, averaged for the territory of Leningrad region was significantly smaller than maximal values (especially for the GSG2 data) during the whole period (Figs. 4a, 4b). The differences between average and maximal values constituted approximately 700 molec. × 10^{13} cm^{-2} for OMI and more than 1000 molec. × 10^{13} cm^{-2} for GSG2 in some years. It was related to the fact that the area of Leningrad region is much larger than of Saint Petersburg area. Therefore, the averaged tropospheric NO_2 content was smoothed in comparison to the spatial mean values for Saint Petersburg (Fig. 3). Also, such a big difference between the spatial mean and maximal values could signify that Saint Petersburg is the only very large source of NO_2 in the Leningrad region. Differences between the OMI and GSG2 data (Fig. 4c) were on average 60 molec. × 10^{13} cm^{-2} for the spatial means and more than 300 molec. × 10^{13} cm^{-2} for the maximal values.

The histograms of the OMI and GSG2 data distribution in 2005–2017 averaged for the territory of Leningrad region can be seen in Fig. 3b. As for Saint Petersburg, similar patterns of the data distribution with shift in values were found. The shift can be minimized by, for instance, subtraction of 60 molec. × 10^{13} cm^{-2} from the GSG2 dataset. The distributions of OMI and GSG2 data, averaged for Leningrad region, are more similar to each other than those for the territory of Saint Petersburg. This is due to the averaging over the larger territory.

A linear correlation between the OMI and GSG2 data was weak with a correlation coefficient equal to

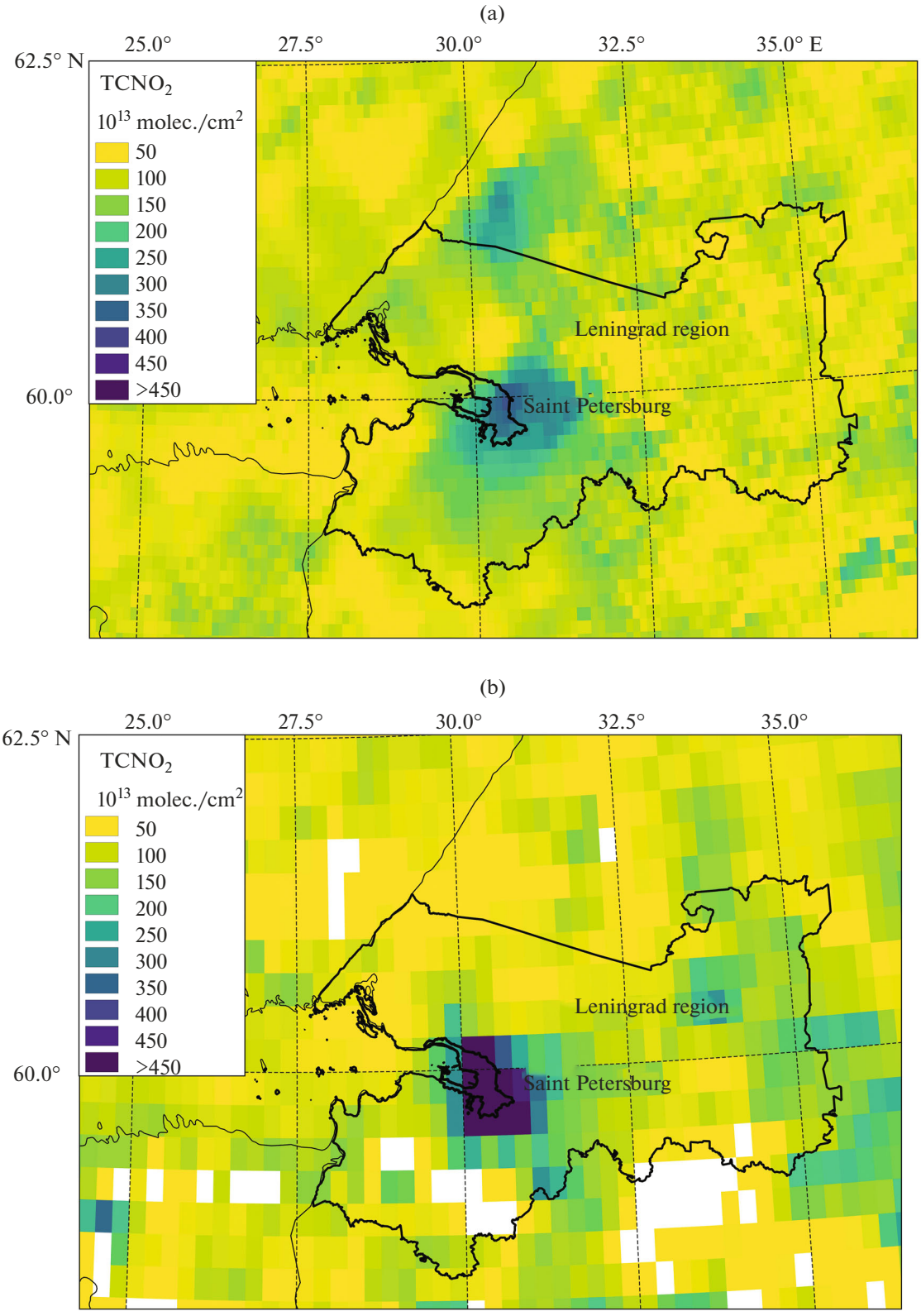

Fig. 1. Monthly average NO_2 content in the troposphere according to OMI (a) and GOME/SCIAMACHY/GOME-2 (b) measurements in July 2017 for the territories of Saint Petersburg and Leningrad region; $TCNO_2$—tropospheric column NO_2.

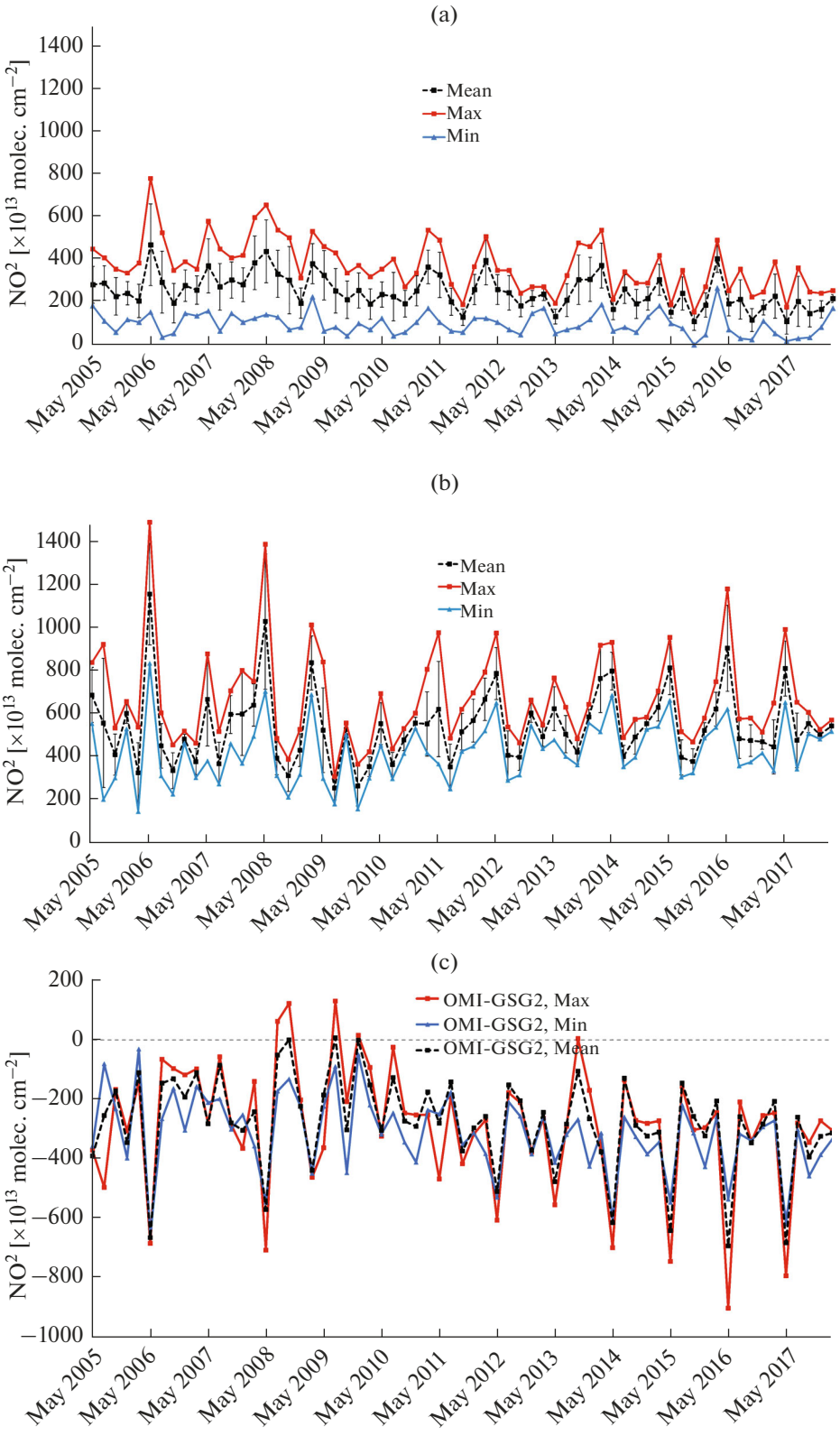

Fig. 2. Monthly average spatial mean, maximal and minimal tropospheric NO2 content on the territory of Saint Petersburg in 2005–2017 according to the OMI (a), GSG2 (b) data and their differences (c); bar charts (black vertical lines on "a" and "b") depict standard deviation of the data within the territory.

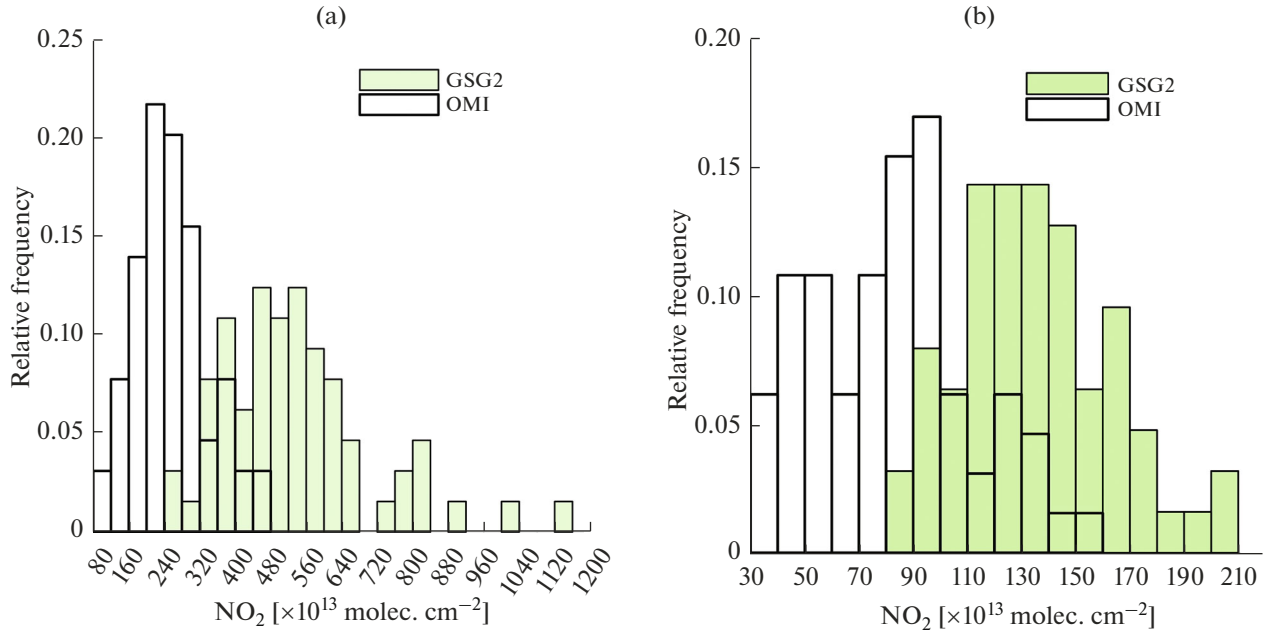

Fig. 3. Histograms of the distribution of monthly average tropospheric NO_2 content in 2005–2017 according to the satellite data averaged for the territory of Saint Petersburg (a) and Leningrad region (b).

~0.4 for both territories. However, a connection in the representation of seasonal variation was found. The graphs in Fig. 5 demonstrate temporal variation of the tropospheric NO_2 from 2005 to 2017 for every single month (from May to September). Figure 5 shows that the general tropospheric NO_2 content decreased from May to June–July and increased to September according to the both datasets in Saint Petersburg (Fig. 5a) and in Leningrad region (Fig. 5b).

3.2. Comparison of Tropospheric and Near-Surface NO_2 Concentration

Since NO_2 enter the atmosphere due to the anthropogenic activity, we can assume that variation of tropospheric NO_2 content and near-surface concentration may be correlated. To confirm this, we compared the trends of yearly average near-surface NO_2 concentration which was measured locally on Saint Petersburg ground-based stations [30] and tropospheric NO_2 content estimated according to the OMI and GSG2 data for the 2005–2017 (Fig. 6). The analysis revealed a correlation between the ground-based and OMI data. As it can be seen from Figs. 6a, 6b, both datasets presented an increase in NO_2 content from 2005 to 2007–2008 with the following decrease to 2017. Lines of linear regression fit the temporal variations of the local near-surface and OMI data relatively well with a coefficients of determination equal to ~0.6. The tropospheric NO_2 content according to the GSG2 dataset did not correlate with the near-surface concentration. Moreover, the trend of the NO_2 content according to the GSG2 data seems to be increasing. However, the line of linear regression has quite poor agreement with the GSG2 data (a coefficient of determination less than 0.1). It is worth noting that the connection between the yearly average measurements and approximating values based on the linear regression was statistically insignificant. However, it could be caused by quite small sample size (12 values). Furthermore, as it can be seen on graphs of monthly and yearly average NO_2 content in the troposphere, the gas varies from month to month and from year to year significantly. Therefore, interannual and interseasonal variation of the real NO_2 content could not be approximated relatively well by the straight lines of the linear regression. It also could influence the estimation of the data significance. To make further conclusions on the correlation between the NO_2 contents at the ground level and in the troposphere, additional comparison of the bigger datasets of the in situ and satellite measurements is needed.

4. CONCLUSIONS

The study demonstrated the following:

(1) Monthly average tropospheric NO_2 content according to the satellite observations by OMI and GOME/SCIAMACHY/GOME-2 differed significantly for the territory of Saint Petersburg (on average, approximately 100% relative to OMI data). Perhaps such discrepancies are related to the cruder spatial resolution of the GOME/SCIAMACHY/GOME-2 data, differences in tropospheric NO_2 retrieval algorithms

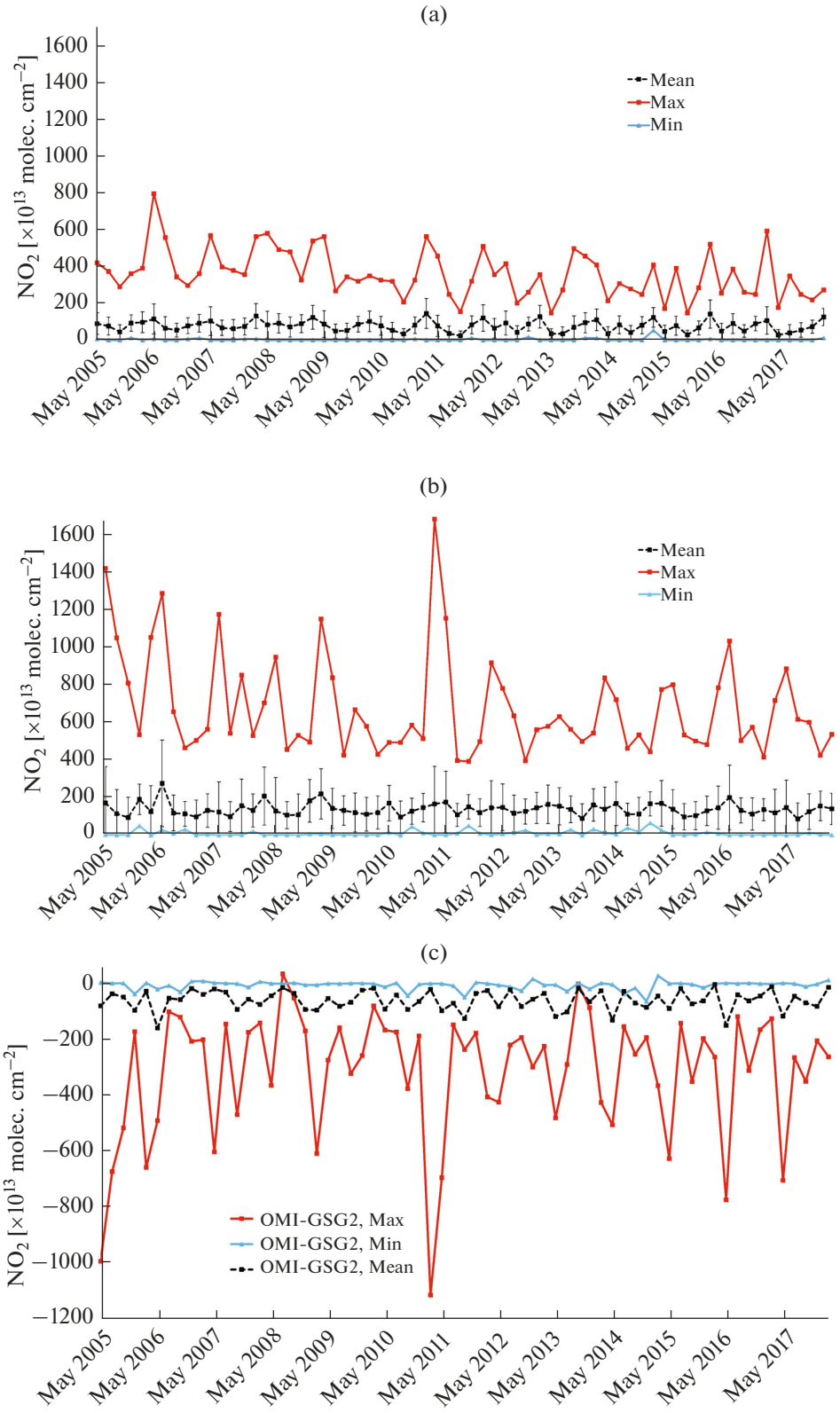

Fig. 4. Monthly average spatial mean, maximal and minimal tropospheric NO_2 content on the territory of Leningrad region in 2005–2017 according to the OMI (a), GSG2 (b) data and their differences (c); bar charts in the bottom (black vertical lines on "a" and "b") depict standard deviation of the data within the territory.

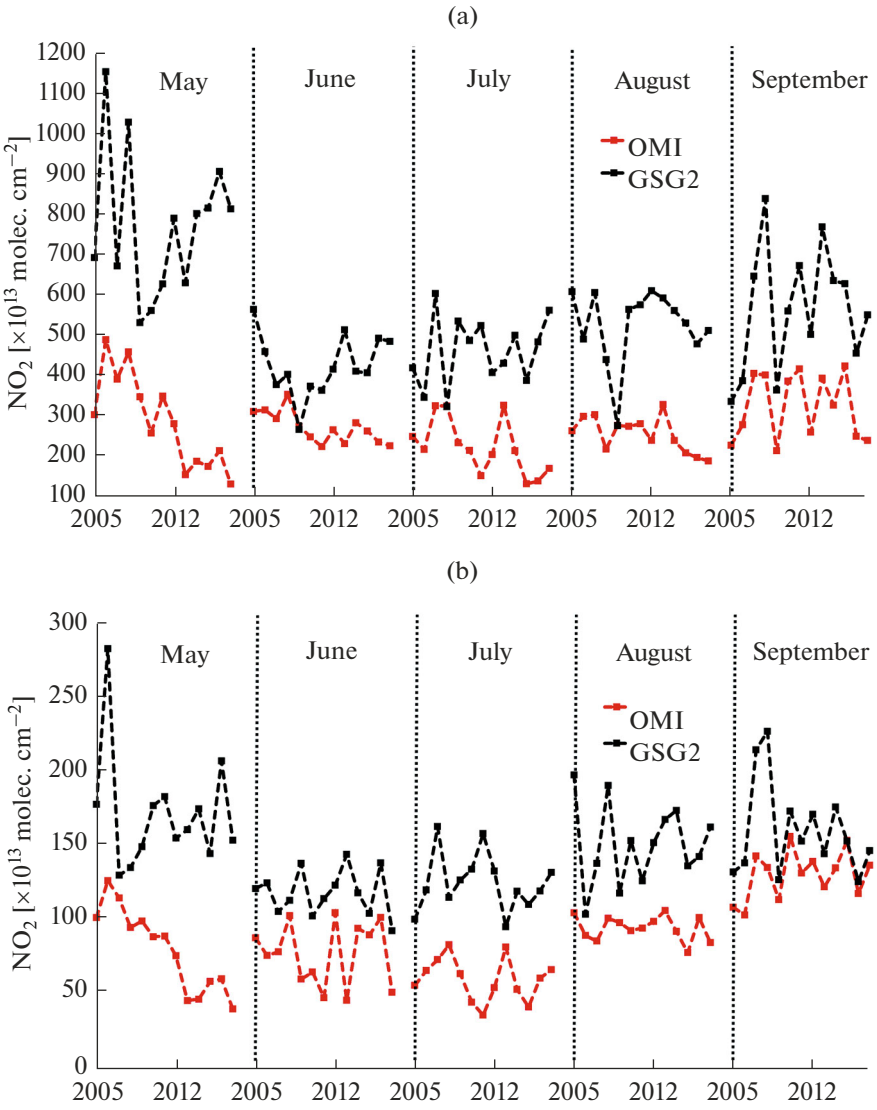

Fig. 5. Monthly average tropospheric NO_2 content according to OMI and GSG2 datasets for Saint Petersburg (a) and Leningrad region (b) in 2005–2017.

or with other sources. The discrepancies between two datasets revealed to be smaller for the territory of Leningrad region, since the spatial averaging for the larger area smoothed the analyzed values. The linear correlation between the OMI and GOME/SCIAMACHY/GOME-2 data was weak. However, both datasets presented similarities in the seasonal variation of tropospheric NO_2 content for Saint Petersburg and Leningrad region.

(2) We found noticeable correlation between trend of yearly average near-surface NO_2 concentration and tropospheric NO_2 content according to the ground-based and OMI measurements for Saint Petersburg, respectively. In both cases the NO_2 content decreased to 2017. The GOME/SCIAMACHY/GOME-2 data did not correlate with the near-surface NO_2 concentration.

In conclusion, it can be said that satellite-based estimation of tropospheric NO_2 content according to the OMI and GOME/SCIAMACHY/GOME-2 measurements for the territories of Saint Petersburg and Leningrad region are with distinctive systematic misfits. Therefore, we would recommend to analyze the discrepancies and correlation between these datasets before studying spatio-temporal variation of tropospheric NO_2 in the areas of other big cities. In addition, if it is impossible to verify the quality of the satellite-based data, the using of both datasets in the analysis would be an optimal solution. Our study demonstrated a potential in using the OMI data for interpreting near-surface NO_2 variation. However, deeper study is needed to estimate the correlation between tropospheric NO_2 and near-surface concentration of the gas.

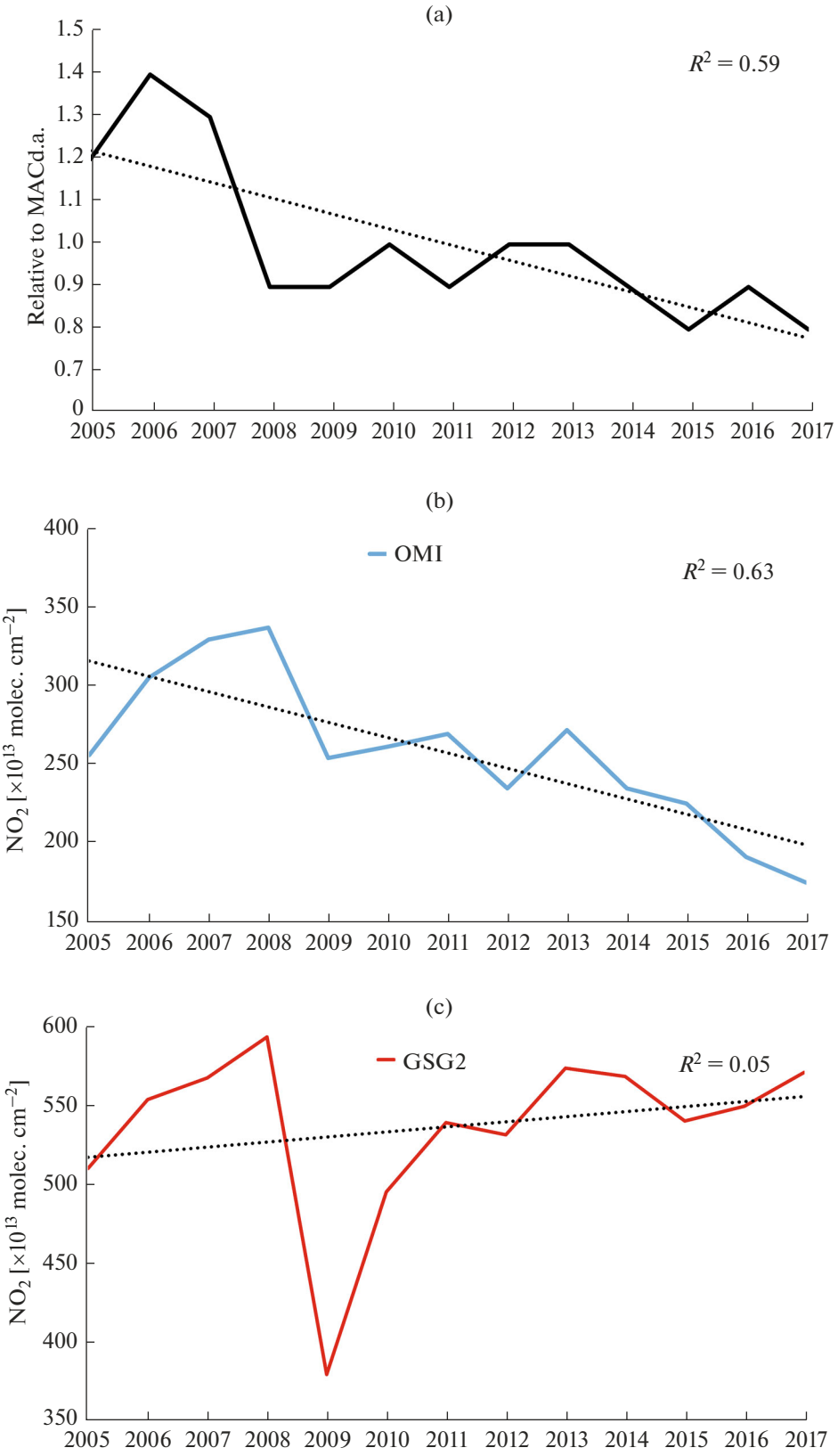

Fig. 6. Yearly average near-surface (a) and tropospheric (b, c) NO_2 content for Saint Petersburg in 2005–2017; $MAC_{d.a.}$—daily average maximum allowable concentration.

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