Tropospheric NO_2 trend over St. Petersburg (Russia) as measured from space

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Received 23 March 2010; accepted 7 April 2010; published 29 April 2010.

For the period 1996–2009, combined data set of tropospheric NO_2 measurements by satellite instruments GOME, SCIAMACHY and OMI has been analyzed to derive possible trend over the city of St. Petersburg in Russia. The time series of the monthly NO_2 columns for these 14 years have been fitted with a linear function superposed on an annual seasonal cycle. Resulting trend estimate provides a linear growth of $\sim 4\%$ per year, which generally correlates with an independent assessments done by NO_x inventory studies. *KEYWORDS: Atmospheric nitrogen dioxide; satellite remote sensing; tropospheric pollution.*

Citation: Ionov, D. V. (2010), Tropospheric NO_2 trend over St. Petersburg (Russia) as measured from space, *Russ. J. Earth. Sci., 11,* ES4004, doi:10.2205/2010ES000437.

Introduction

Nitrogen dioxides ($NO_x = NO + NO_2$) have a large impact on human health, climate and atmospheric chemistry. In the dense populated regions NO_x are intensively emitted by all combustion processes and contribute to photochemical smog production and nitric acid deposition. Starting from 1995 the global mapping of tropospheric NO_2 has become possible with help of satellite instruments GOME (1995–2003) [*Burrows et al.*, 1999] aboard ERS 2, SCIAMACHY (from 2002) aboard Envisat [*Bovensmann et al.*, 1999], OMI (from 2004) aboard EOS-Aura [*Levelt et al.*, 2006] and GOME 2 aboard MetOp [*Munro et al.*, 2006]. A number of studies have been devoted to detection of trends in tropospheric NO_2 over different regions of globe. To the first-order approximation the local changes in NO_2 columns can be assumed to be proportional to changes in local NO_x emission. Thus, persistent efforts to reduce emissions from traffic and industry, adopted in a number of developed countries (e.g. European Union and USA), are expected to show the decrease in tropospheric NO_2 column. On the contrary, the regions of rapid economic development must be characterized with tropospheric NO_2 growth (e.g. East Asia). Indeed, several studies have revealed statistically significant negative NO_2 trend over Western Europe and Eastern USA, and positive trend over China. *Richter et al.* [2005] analyzed tropospheric NO_2 columns retrieved from GOME and SCIAMACHY in 1996–2004 and found substantial reduction in NO_2 over Europe ($\sim 20\%$) and Central East Coast of the USA ($\sim 10\%$), but significant increase of $\sim 50\%$ over the industrial areas of China. *Van der A et al.* [2006] studied trend in NO_2 using the data of similar satellite measurements in 1996–2005, and showed large growth over eastern China – up to 20% per year (city of Shanghai). Further, *Van der A et al.* [2008] extended that study to the 10-years data set of satellite observations in 1996–2006 and revealed reductions in NO_2 over Europe and eastern USA (up to 7% per year), with a strong increase over Asia (up to 29% per year in China), but also some industrial regions of Siberia in Russia – up to 11% per year (city of Novosibirsk).

Here the 14-years combined data set of NO_2 measurements by GOME, SCIAMACHY and OMI in 1996–2009 is analyzed to quantify the trend in tropospheric NO_2 column over the city of St. Petersburg in Russia. First, a brief description of satellite instruments and tropospheric NO_2 retrieval is given. Further, the data analysis is described and an overview of long-term NO_2 time series, acquired over St.Petersburg in 1996–2009, is given. Then, accomplished trend analysis of tropospheric NO_2 is presented. Finally, the results of study are discussed and further work is proposed.

Tropospheric NO_2 Measurements From Space

Along with ozone, the global monitoring of NO_2 is a key objective of NASA's Ozone Monitoring Experiment (OMI). In this regard, OMI succeeds two other space experiments – ESA's GOME and SCIAMACHY. GOME (Global Ozone Monitoring Experiment) was launched in April 1995 onboard the ERS 2, into a sun-synchronous polar orbit [*Burrows et al.*, 1999]. It is a nadir-viewing UV-visible spectrometer that measures the solar irradiance and the solar radiation

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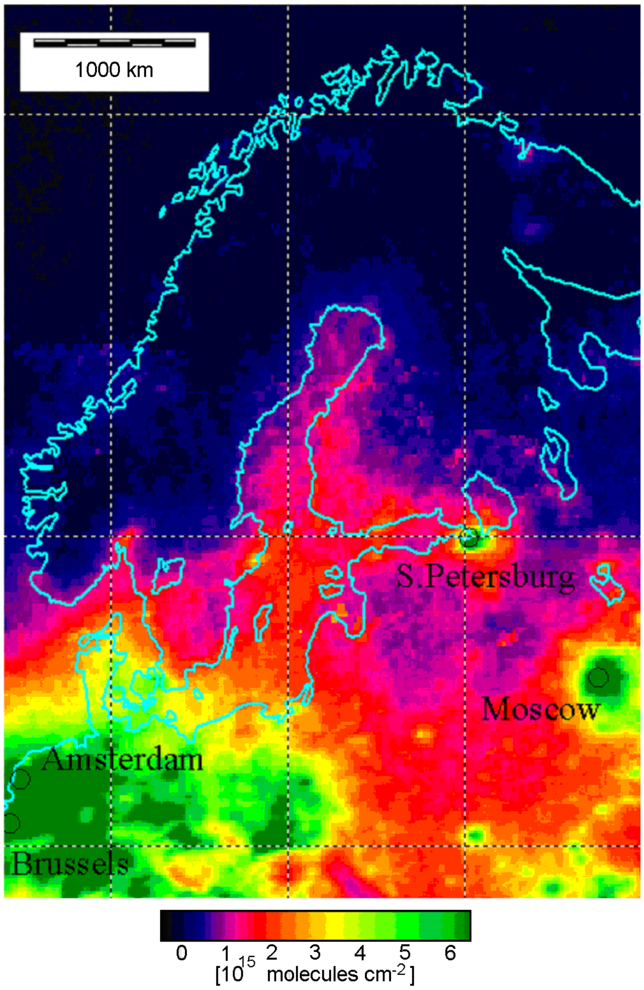

Figure 1. Annual map of tropospheric NO_2 vertical column over the Baltic Sea region, produced from the global data of Aura OMI monthly mean tropospheric NO_2 in 2004–2009 (available at http://www.temis.nl).

backscattered from both the atmosphere and the Earth's surface. Besides providing global maps of atmospheric ozone, GOME was also the first spaceborne instrument capable of measuring the total column amount of NO_2. The trace gas column densities given in the GOME data products (ozone and NO_2), are derived by the Differential Optical Absorption Spectroscopy (DOAS) technique [*Platt*, 1994]. GOME was followed by the SCanning ImAging spectroMeter for Atmospheric CartograpHY (SCIAMACHY) onboard ENVISAT in 2002, which could also measure NO_2 vertical columns [*Bovensmann et al.*, 1999].

The work presented here is based on tropospheric NO_2 columns retrieved from the satellite data by the Royal Netherlands Meteorological Institute [*Boersma et al.*, 2004, 2007]. The algorithm is based on the combined retrieval-assimilation-modelling approach. A chemistry-transport model, driven by high-quality meteorological fields and assimilated NO_2 information from previously observed orbits, provides best guess profiles of NO_2. These model forecast fields are collocated with satellite observations, and the radiative transfer modelling in the retrieval is performed based on the model trace gas profile and temperature profiles. The modelled stratospheric NO_2 distribution is employed to derive a tropospheric column by subtracting the modelled (assimilated) stratosphere from the measured column. The retrieval is coupled to cloud top height and cloud fraction retrievals derived from the satellite data, and also coupled to high quality albedo maps. The global dataset of such monthly averaged tropospheric NO_2 columns retrieved from the GOME, SCIAMACHY and OMI instruments is publicly available through (http://www.temis.nl).

For better accuracy, only measurements with an estimated cloud fraction less than ~ 0.2 are used in the calculation of monthly mean NO_2 fields. Detailed error budget of tropospheric NO_2 retrieval is presented in the work of *Boersma et al.* [2004]. According to that study, a meaningful estimate of the tropospheric vertical column over the heavily polluted continental areas can be given with a precision of 35–60%.

Long-Term NO_2 Data Over St. Petersburg

As an illustration of the opportunity to monitor urban emissions from space, an annual distribution of tropospheric NO_2 over the Baltic Sea region was averaged for the years 2004–2009 of OMI measurements (see Figure 1). The map shows almost no NO_2 in the troposphere of northern Europe ($< 1 \times 10^{15}$ molecules cm^{-2}), but an area of high values in the south-west of the region – Belgium, Netherlands and Germany (up to $\sim 18 \times 10^{15}$ molecules cm^{-2}), and a spots of polluted air over the cities of Moscow (up to $\sim 18 \times 10^{15}$ molecules cm^{-2}) and St. Petersburg (up to $\sim 9 \times 10^{15}$ molecules cm^{-2}).

To analyze NO_2 trend over St. Petersburg, the data records from 3 satellite instruments were assembled: the GOME data in 04/1996–06/2003, the SCIAMACHY data in 07/2002–09/2009 and the OMI data in 10/2004–09/2009. The monthly mean tropospheric NO_2 columns, available at TEMIS website are gridded on a different scale, depending on the instrument – $0.25° \times 0.25°$ for GOME and SCIAMACHY and $0.125° \times 0.125°$ for OMI. To filter out the effects of different spatial resolution, rectangle of 1° latitude by 2° longitude centered over the city of St. Petersburg (59.95°N, 30.32°E) was selected to average the individual grid pixels. Resulting time series of monthly mean data by GOME, SCIAMACHY and OMI over St. Petersburg are shown in Figure 2. The measurements demonstrate high month-to-month variation – tropospheric NO_2 column is changing from ~ 1 to $\sim 10 \times 10^{15}$ molecules cm^{-2}.

To achieve more significant results, both GOME and SCIAMACHY data are used in 2002–2003, and both SCIAMACHY and OMI are used since October 2004. However, to homogenise the full data record 1-year periods of overlap measurements (by SCIAMACHY and OMI in

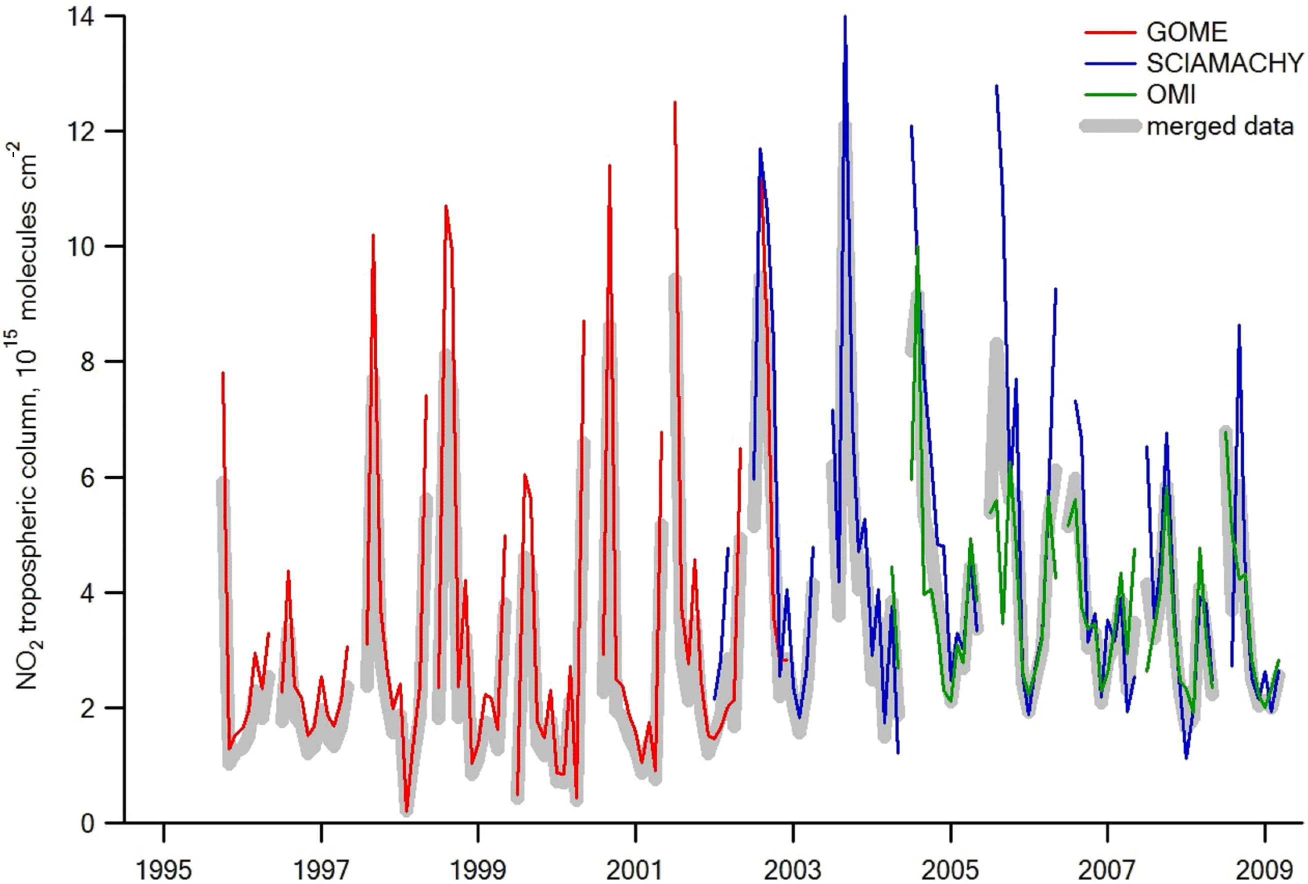

Figure 2. Time series of monthly mean tropospheric NO_2 column, as measured by ERS-2 GOME, ENVISAT SCIAMACHY and Aura OMI over St. Petersburg. The merged data for the full period of 1996–2009 are shown by grey curve.

10/2004–09/2005, GOME and SCIAMACHY in 07/2002–06/2003) were used to fit and apply the necessary adjustments. For this, the corresponding monthly data were spatially convolved from their original resolution to the uniform grid of 1°(latitude) ×2° (longitude), and the yearly averaged field of tropospheric NO_2 column over Eurasia (40–80°N, 0–180°E) was calculated for each instrument. Resulting maps – GOME and SCIAMACHY in 2002–2003, SCIAMACHY and OMI in 2004–2005 – are presented on Figure 3, demonstrating reasonable agreement between satellite instruments. Thus, the correlation coefficient for the pixel-to-pixel comparison results in 0.95 for GOME versus SCIAMACHY and 0.96 for SCIAMACHY versus OMI. Regression equations, computed from these comparisons, were used to adjust GOME data to SCIAMACHY, and SCIAMACHY to OMI. The average of two measurements, when available, was taken – GOME and SCIAMACHY in 2002–2003, SCIAMACHY and OMI since 2004. Resulting time series of tropospheric NO_2 column over St. Petersburg is plotted as a separate line on Figure 2 ("merged data").

Trend Analysis

Following similar studies [*Ionov et al.*, 2009; *Poberovskii et al.*, 2007], a simple statistical model was used to describe long-term variation of tropospheric NO_2 column:

$$Y_t = A + BX_t + C\mu_t + N_t \tag{1}$$

where Y_t represents the monthly NO_2 column of month t, X_t is the number of months after January 1996, N_t is the residual noise, A represents the NO_2 column in January 1996, B is the monthly trend in NO_2, and C is the amplitude of seasonal component μ_t. Thus, coefficients A, B and C are the fit parameters of the multiple linear regression.

The precision of the trend B is estimated from the variance σ_N of remainder, the data set length n (in years) and autocorrelation of N_t, [*Weatherhead et al.*, 1998]:

$$\sigma_B \approx \left[\frac{\sigma_N}{n^{3/2}} \sqrt{\frac{1+|\phi|}{1-|\phi|}} \right] \tag{2}$$

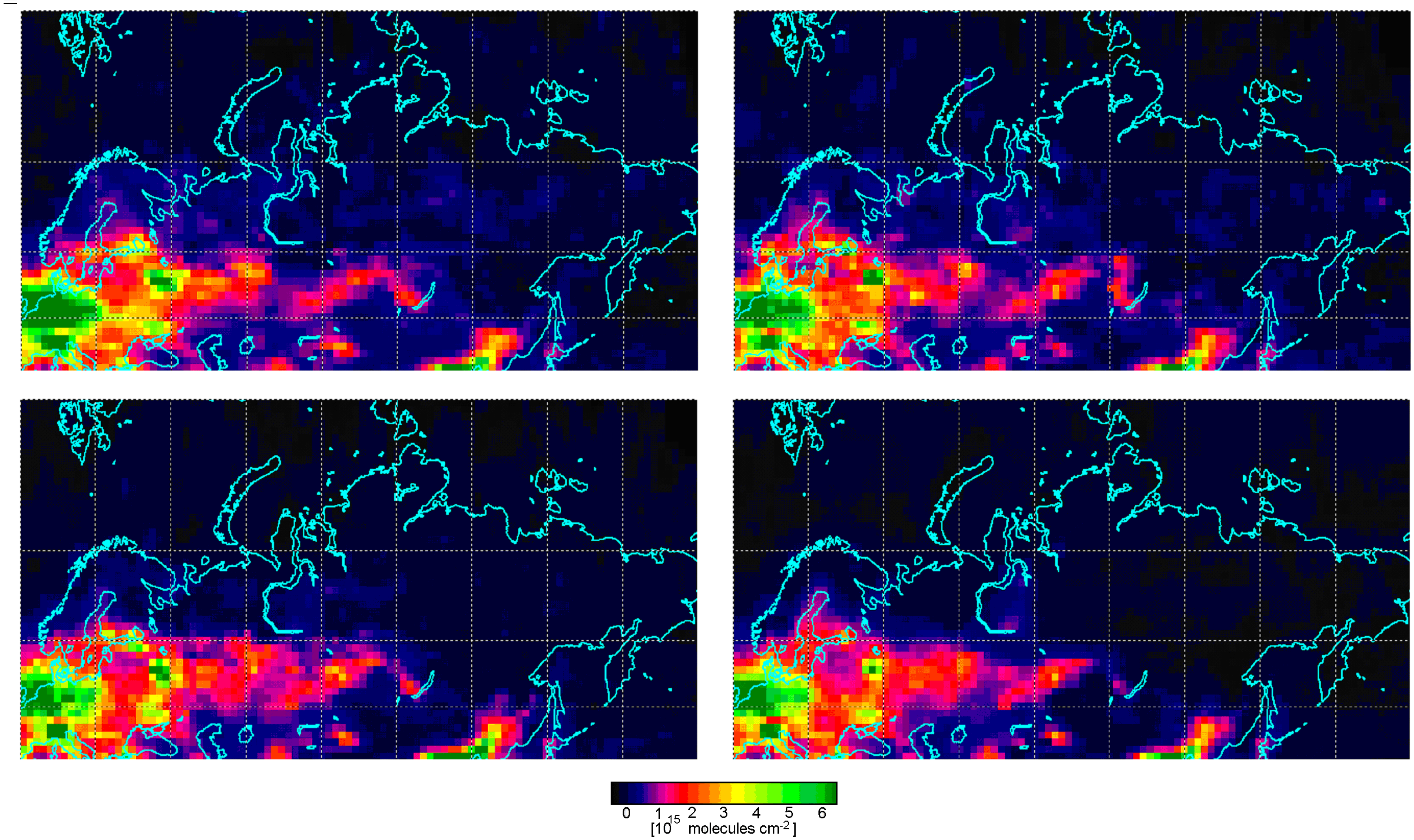

Figure 3. Comparison of annual tropospheric NO_2 by satellites, spatially convolved from their original resolution to the uniform grid of 1°(latitude)×2°(longitude): GOME (left top) versus SCIAMACHY (right top) in 2002–2003, SCIAMACHY (left bottom) versus OMI (right bottom) in 2004–2005.

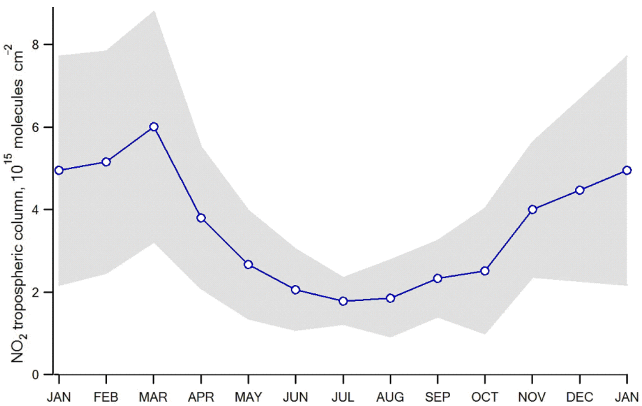

Figure 4. Seasonal component of tropospheric NO_2 variation, obtained from the satellite measurements over St. Petersburg in 1996–2009. The standard deviation of monthly averaged is shown by grey line.

Then, to decide if the trend estimate is statistically significant a commonly used rule is applied, considering that a trend B is real with 95% confidence level if $|B/\sigma_B| > 2$ [*Weatherhead et al.*, 1998].

To compute seasonal component μ_t, each of the 12 months was averaged over the full data record for the years 1996–2009. Resulting time series of μ_t obtained from the satellite measurements over St. Petersburg, along with the standard deviation for each month, is presented on Figure 4. According to that data, high NO_2 columns are observed in winter and early spring – up to $\sim 6 \times 10^{15}$ molecules cm^{-2} in March; the minimum values are found in summer – about $\sim 2 \times 10^{15}$ molecules cm^{-2}. Such typical seasonal cycle is caused by the longer lifetime of NO_x during the cold period, increased local NO_x emissions from the municipal heating system and frequent episodes of calm and temperature inversion, facilitating the accumulation of pollutants in the wintertime surface air.

Figure 5 shows a measured time series of monthly average tropospheric NO_2 column over St. Petersburg and the fitted function (model approximation). The solid green line is the model fit and the remainder between model and measurements is denoted by the squares. Resulting trend estimate provides a linear growth of $4.3 \pm 1.0\%$ per year (growth is the percentage calculated with respect to the mean NO_2 column, 3.3×10^{15} molecules cm^{-2}). The fitted trend value is statistically significant ($|B/\sigma_B| > 2$).

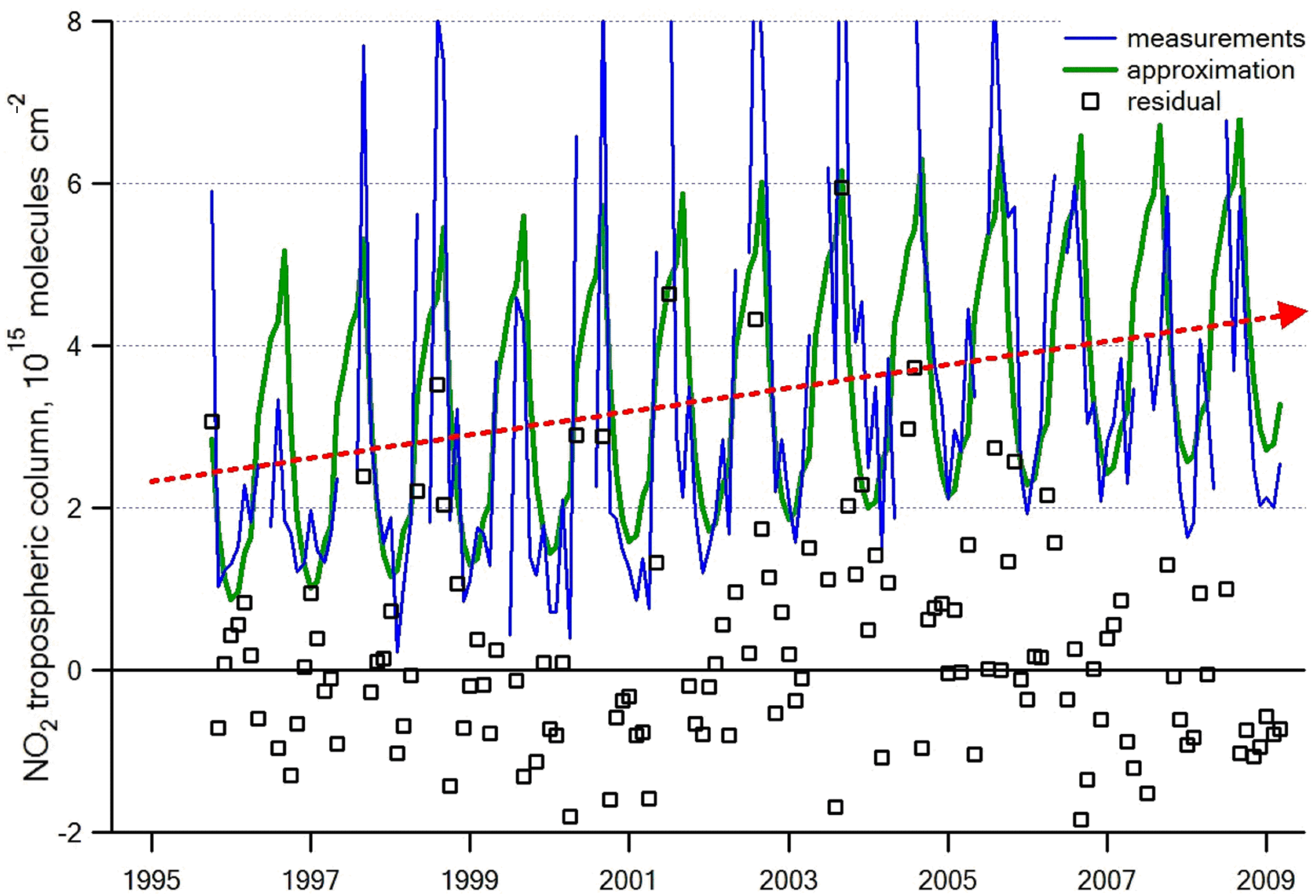

Figure 5. Time series of monthly average tropospheric NO_2 column over St. Petersburg and the fitted function (model approximation). The solid green line is the model fit and the remainder between model and measurements is denoted by the squares. The linear component is shown by red line.

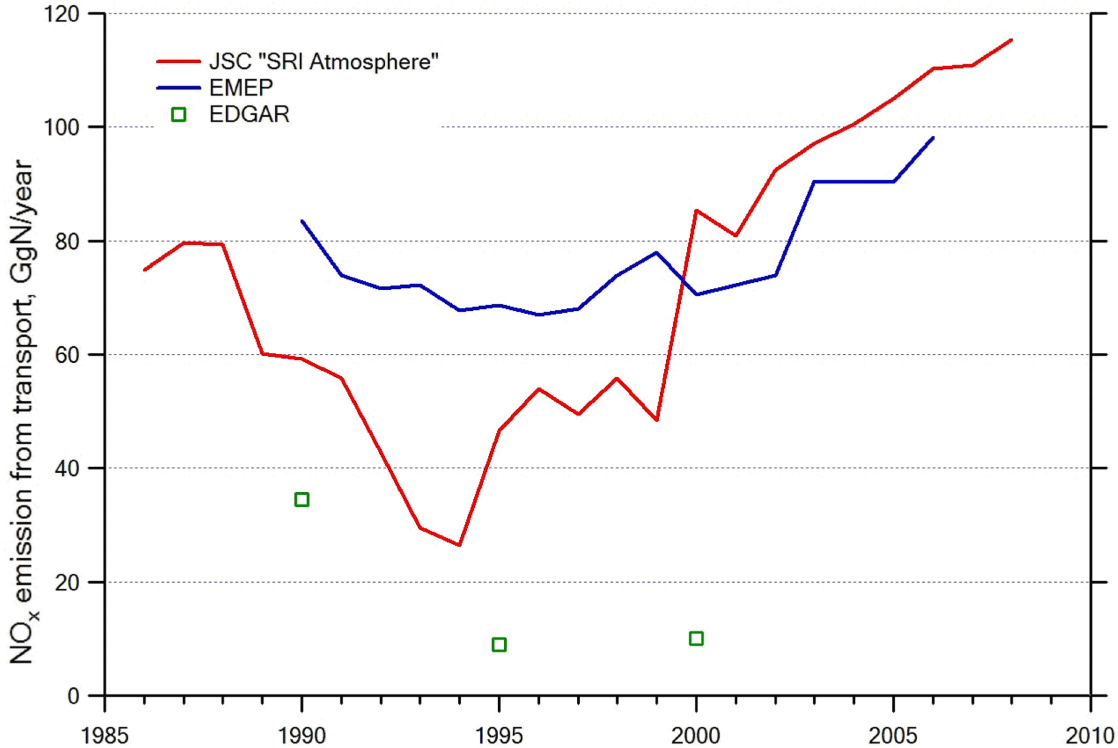

Figure 6. Inventory estimates of NO_x emission from transport of St. Petersburg as given by EMEP (http://emep.int), EDGAR (http://www.mnp.nl/edgar) and JSC "SRI Atmosphere" (http://www.nii-atmosphere.ru).

Discussion, Conclusions and Further Work

Fossil fuel combustion is known to be responsible for more than 50% of the total production of NO_x. According to the studies done at the Scientific Research Institute for Atmospheric Air Protection, JSC "SRI Atmosphere" (http://www.nii-atmosphere.ru) the share of traffic in the total emission of anthropogenic pollutants at St. Petersburg has increased from 56% in 1990 to 78% in 2005 and 92% in 2008. The number of vehicles officially registered in St. Petersburg has changed from $\sim$ 1.0 million in 2000 to $\sim$ 1.5 million in 2008 – with an average linear growth rate of $\sim$ 6% per year. Inventory estimate, reported by the Committee for Nature Use, Environmental Protection and Ecological Safety of St. Petersburg (http://infoeco.ru) for NO_x emission from transport was 115 GgN yr^{-1} in 2008. The calculations of this estimate, provided by JSC "SRI Atmosphere" show, that this value has dropped down manifold beginning from 1988, then started to grow in 1995 but dropped again in 1998, and now it is growing steadily since 2002 with a linear rate of $\sim$ 4% per year (see Figure 6). *Konovalov* [2007] combined satellite measurements of tropospheric NO_2 with a continental scale air quality model (CTM CHIMERE) over the European part of Russia in 1996–2005, to elaborate so-called "top-down" emission estimates, and also found a positive and statistically significant trend of NO_x emission during summer at St. Petersburg (3.6 ± 1.6% per year). Thereby, the trend estimate derived in the present study ($\sim$ 4% per year) is in agreement with an expected growth of anthropogenic emissions at St. Petersburg during last decade.

Looking at the time series of tropospheric NO_2 over St. Petersburg (see Figure 5), one may notice some decline, starting from 2006. A similar feature was found recently by *Sitnov* [2009], who analyzed tropospheric NO_2 measured by OMI over the region of Moscow in 2004–2009 and suggested that the observed growth has stopped in 2006, and now the tropospheric NO_2 is decreasing. Indeed, municipal inventory surveys of St. Petersburg report the growth of annual surface NO_2 concentration from 29 mkg m^{-3} in 2001 to 57 mkg m^{-3} in 2006, but then it dropped back to 36 mkg m^{-3} in 2008 [*Golybev and Sorokin*, 2009]. The duration of that decline is too short to find any reliable explanation, but it indicates the need of additional parameter (e.g. local meteorology) to be introduced into the trend analysis.

Existing emission inventories based on the so-called "bottom-up" estimates (e.g. compilation of individual sources statistics), are known to be inaccurate. For instance, two spatially gridded inventories – the European Monitoring and Evaluation Programme (EMEP, http://emep.int) and Emission Database for Global Atmospheric Research (EDGAR, http://www.mnp.nl/edgar) provide considerably different estimates of NO_x emission from the road transport source over the region of St. Petersburg (see Figure 6). A number of studies have demonstrated already the perspectives to verify and improve available emission estimates with the help of satellite data [e.g. *Konovalov et al.*, 2008; *Leue et al.*, 2001; *Martin et al.*, 2003, 2006]. A further development of current work will be to combine the data of satellite measurements with the regional chemical-transport model and improve the estimate of anthropogenic NO_x emission from the city of St. Petersburg.

Acknowledgment. This study was supported in part by the Ministry of Education and Science of the Russian Federation (project no. RNP.2.1.1.4166, RNP.2.2.1.1.3836).

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