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Validation of GOME (ERS-2) NO_2 vertical column data with ground-based measurements at Issyk-Kul (Kyrgyzstan)

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Abstract

Here we present the results of comparison between operational NO_2 vertical column data by Global Ozone Monitoring Experiment (GOME) onboard ERS-2 satellite and ground-based measurements at Issyk-Kul station in Kyrgyzstan, northern Tien Shan. The data of GOME, taken for the period of 1996–2002, was found to be reasonably close to the results of ground-based sunrise measurements. The latter were adjusted to the time of GOME overpass nearby noon, providing direct comparison between satellite and ground-based data. According to the results, there is 0.6×10^{15} mol/cm^2 ($\sim$18%) overestimation of NO_2 vertical column by GOME, compared to our ground-based data.

Keywords: Atmospheric nitrogen dioxide; Satellite data validation; Twilight observations

1. Introduction

Nitrogen dioxide (NO_2) is one of the key species in atmospheric chemistry. In the stratosphere, it is involved in catalytic ozone destruction, whereas the photolysis of tropospheric NO_2 results in O_3 formation. In addition, it is indirectly responsible for the atmosphere oxidizing capacity and contributes to radiative forcing of climate.

Stratospheric NO_2 has been measured by a number of satellite instruments, e.g., LIMS (Limb Infrared Monitor of the Stratosphere), SME (Solar Mesosphere Explorer), SAGE-II/III (Stratospheric Aerosol and Gas Experiment), ISAMS (Improved Stratospheric and Mesospheric Sounder), HALOE (Halogen Occultation Experiment), and POAM (Polar Ozone and Aerosol Measurement). Despite of the global coverage of satellite observations, these measurements are characterized by the limited time sampling and high uncertainty in the lower stratosphere. The Global Ozone Monitoring Experiment (GOME) in 1995 was the first satellite mission to provide a global picture of atmospheric NO_2 with reasonable spatial and temporal resolution. Unlike previous satellite systems, aiming on individual NO_2 vertical profile measurements, GOME is designed to map the global distribution of NO_2 vertical column. However, the true accuracy of that data is not fully understood up to now. The present study contributes to geophysical validation of operational NO_2 vertical column measurements of GOME by means of correlative ground-based observations at a remote station of Issyk-Kul in the central part of Eurasia.

2. Ground-based measurements of NO_2 vertical column

The measurements of nitrogen dioxide vertical column have been started at Issyk-Kul station in 1983 (Aref'ev et al., 1995; Kashin et al., 2000; Semenov et al., 2001). The station is located on the northern coast of Issyk-Kul lake in Tien Shan mountains (42.6°N/77.0°E, 1650 m above the sea level). This region represents one of the largest intermountain hollows of Tien Shan, located on the

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northeast of Kyrgyzstan. Being enclosed by the two large arcs of mountain circuits in the south and north, which converge in the west and east sides, this area form a closed space. The dominant breezes at the shore of the lake block the movement of cumulus clouds generated above the enclosing mountain ridges, and increase the duration of clear sky conditions over the station.

The observations of NO_2 vertical column are based on the twilight measurements of zenith scattered solar radiation in the visible range of spectra, each sunrise and sunset at solar zenith angle 85–92°. Before 2001, the retrieval technique applied a three-wavelength method (Kerr et al., 1982; Sinyakov and Spektorov, 1987), considering measured intensities at three wavelengths: 437.6, 439.8, and 442.0 nm, located at relative maximum and minimum of NO_2 absorption band. Since 2001 the instrument was upgraded and the retrieval is now similar to well-known DOAS (differential optical absorption spectroscopy) method, consisting of least squares fitting the observed differential optical thickness with the absorption cross-sections of molecular species (Elokhov and Gruzdev, 1993). Both instruments, the old and the new one, were running for 1.5 years simultaneously in 2001–2002 to ensure resulting time series be uniform. Since then, the measurements are carried out by means of grating spectrometer operating in ~435–453 nm range with a spectral resolution of ~1.0 nm. Based on the similar spectra registered at noon under clear sky conditions, when NO_2 optical absorption is expected to be negligibly small (so-called reference spectra), the column NO_2 density along the optical path (or slant column) is derived considering Beer–Lambert's law. The solution takes into account ozone and NO_2 absorption, molecular scattering, aerosol attenuation and so-called Ring effect (inelastic scattering). Slant NO_2 columns are then converted into vertical columns using so-called air mass factor (AMF), calculated with radiative transfer model.

The instrument design and retrieval technique is similar to that operating at Zvenigorod station and described in detail in (Elokhov and Gruzdev, 1993). The system use Russian manufactured scanning monochromator MDR-23 (LOMO) with 1200 grove/mm grating, providing a spectrum sampling of ~0.01 nm. In the DOAS processing chain, O_3 and NO_2 absorption cross-sections by P. Graham and H. Johnston (unpublished data) are used to retrieve slant columns. The Ring effect for this short range of spectra (435–453 nm) is considered as a constant term added to the intensity of direct solar radiation (Hofmann, 1992). As for the AMF, a specially developed model accounting for O_3 and NO_2 absorption, aerosol attenuation, molecular single scattering and refraction in 45-layer spherical atmosphere was used. The data of vertical ozone distribution was taken from (Keating and Pitts, 1987), temperature and pressure – from CIRA 86 model (Labitzke et al., 1985), aerosol scattering coefficient – from (Elterman, 1968), and NO_2 vertical profiles – from (Chu and McCormick, 1986; Roscoe et al., 1986).

The Issyk-Kul instrument was certified by the Network for the Detection of Stratospheric Change (NDSC) as a complementary station, following the results of intercomparison carried out at Zvenigorod (55.7°N/36.8°E) in 1997. According to conclusions from official NDSC intercomparison campaigns (Roscoe et al., 1999; Vandaele et al., 2005), such instruments agree for the measurements of NO_2 slant column within 7% or better, when common spectral interval and analysis parameters are imposed.

3. Satellite GOME data on total NO_2

The Global Ozone Monitoring Experiment (GOME) was launched on April 21st 1995 on board the second European Remote Sensing Satellite (ERS-2) (Burrows et al., 1999a). ERS-2 flies in a Sun-synchronous polar orbit at mean altitude of 780 km with an inclination of 98°, and a local time of 10:30 h of the equator crossing at the descending node. GOME is a nadir-viewing grating spectrometer that measures the solar irradiance and the solar radiation backscattered from both the atmosphere and the earth's surface. The instrument is operating in UV–visible range of spectra (240–790 nm) with a moderate spectral resolution of 0.2–0.4 nm sensed by four individual linear detector arrays each with 1024 detector pixels. The field of view may be varied in size from 320 km × 40 km (forward scan) to 960 km × 40 km (back scan). With the large swath, global coverage is achieved every 3 days at the equator and earlier at higher latitudes.

Providing the global picture of atmospheric ozone, GOME was also the first spaceborne instrument having the capability to measure the vertical column amount of nitrogen dioxide (NO_2). The trace gas column densities, given in the GOME products (ozone and NO_2), are derived by applying the DOAS technique. The input is the ratio of earth radiance and solar irradiance, measured by GOME. The accurate derivation of total ozone and NO_2 from GOME data presents several difficulties and is still a matter of research. Since the release in summer 1995 of its first developmental version, the GOME Data Processor (GDP) was upgraded on many occasions and the quality of both ozone and NO_2 products has improved significantly (ESA, 2002).

GOME GDP total NO_2 has been validated from pole to pole by comparison with the data of ground-based measurements from a network of SAOZ (Systeme d'Analyse par Observation Zenithales)/DOAS UV–visible spectrometers and Fourier Transform Infrared spectrometers associated with the NDSC, and with global data from the HALOE and POAM satellite sensors and from both tropospheric and stratospheric modeling tools (ESA, 2002).

According to these studies, GOME total NO_2 (GDP 3.0) is in a reasonable agreement with ground-based and other satellite measurements: within $\pm 5 \times 10^{14}$ mol/cm^2 in areas of low tropospheric NO_2 and within $\pm 8 \times 10^{14}$ mol/cm^2 in areas of very low slant column of NO_2. Although it is difficult to evaluate precisely the accuracy of this product due to various problems such as the diurnal photochemical variation of NO_2, the overall accuracy is estimated to fall within the 5–20% range. GOME total NO_2 is affected by larger errors under particular circumstances, e.g., over polluted areas and in the South Atlantic Anomaly (ESA, 2002).

The data of GOME have been also compared to ground-based NO_2 measurements at Zvenigorod (Russia), which are similar to those carried out at Issyk-Kul station (Timofeev et al., 2000). However, these observations were found to be affected with high tropospheric pollution over Moscow region, which is only 50 km to the east from Zvenigorod.

4. Correlative data set

In this study, we used the data of ground-based twilight NO_2 observations at Issyk-Kul, collected for the period from January 1996 till December 2002. The total number of data available for comparison was 1124 daily values at sunrise and 1084 at sunset. For these days the corresponding NO_2 vertical columns have been extracted from GOME level 2 data (GDP version 3.0), related to the pixels located within 1000 km from ground-based station. According to that selection, the overall number of comparisons between satellite and ground-based measurements was limited to about 500 pairs of values. Only half of that data set corresponds to the period of 1996–2000, as the ground-based observations were limited to clear-sky conditions during these years, while in 2001–2002 nearly everyday measurements became possible with the new instrument.

Within the data set, the average distance of GOME ground pixel from Issyk-Kul was 475 km; the cloud fraction was on the average 0.16. Sun zenith angle was changing from 21° in summer to 72° in winter (44° on the average). The average error of NO_2 vertical column retrieval (GDP 3.0) was 17%. The average NO_2 column, measured at Issyk-Kul, was 3.26×10^{15} mol/cm^2 at sunrise and 5.10×10^{15} mol/cm^2 at sunset; the average GOME value was 3.42×10^{15} mol/cm^2.

5. Results of comparison and analysis

The time series of correlative GOME and ground-based data at Issyk-Kul for 1996–2002 is presented in Fig. 1. There are frequent gaps in the data before 2001 due to the ground-based instrument limitation, but starting from 2001 the measurements are continuous (see Fig. 2). Anyway, the data set regularly cover the whole period, and both satellite and ground based measurements reproduce known seasonal variation of NO_2, with maximum values in summer and minimum – in winter. As it is seen from the plot, the data of GOME is much closer to ground-based sunrise measurements, than to the sunset ones. The average and rms (root-mean-square) deviations between GOME and ground-based sunrise data are -0.08×10^{15} mol/cm^2 and 0.69×10^{15} mol/cm^2, accordingly.

We have studied how does these deviations depend on different factors of data selection and observation conditions. First, the spatial criteria of satellite data search have been examined to check if it may influence the agreement with ground-based measurements. However, the difference between satellite and ground-based NO_2 was not found to depend on the distance of selected GOME pixels from the site location (<1000, ~480 km on the average). Next, the influence of Sun elevation (solar zenith angle), NO_2 vertical column, GOME cloud fraction and season, have been investigated in a similar way. The most evident among them is a seasonal dependence of deviations between satellite and ground-based data, presented in Fig. 3 (comparison of GOME with sunrise ground-based measurements). According to the plot, GOME NO_2 columns are below ground-based sunrise values in summer, but appear to exceed them in wintertime.

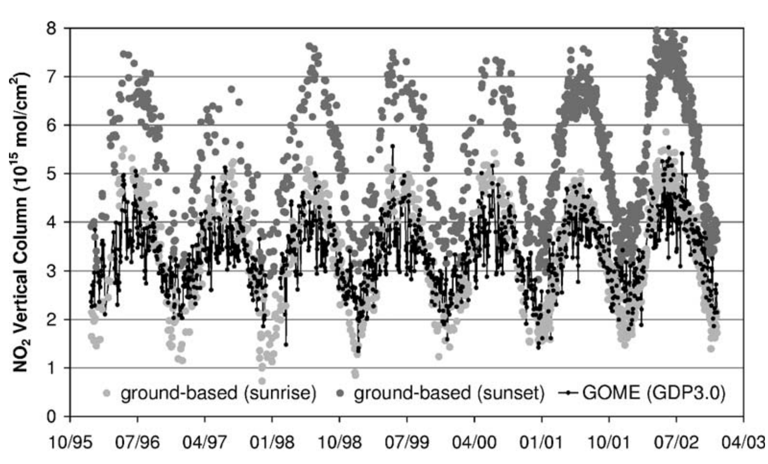

Fig. 1. Comparison of GOME (GDP 3.0) NO_2 vertical column data with correlative ground-based measurements (sunrise and sunset) at Issyk-Kul station (43°N/77°E) in 1996–2002.

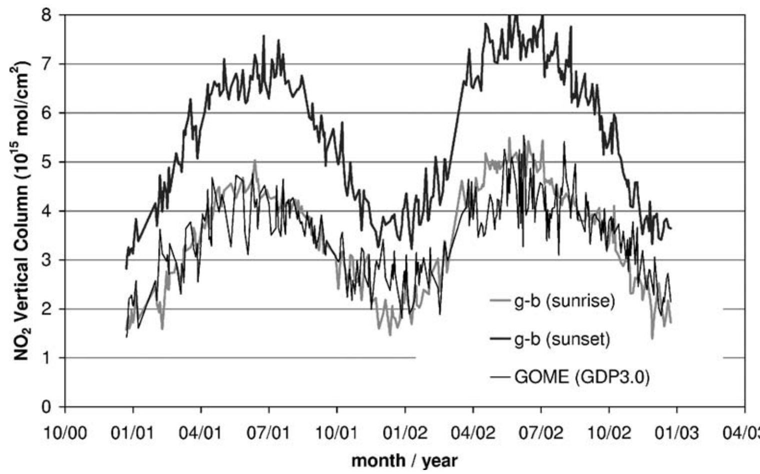

Fig. 2. Comparison of GOME NO_2 vertical column data with correlative ground-based measurements (sunrise and sunset) at Issyk-Kul station in 2001–2002.

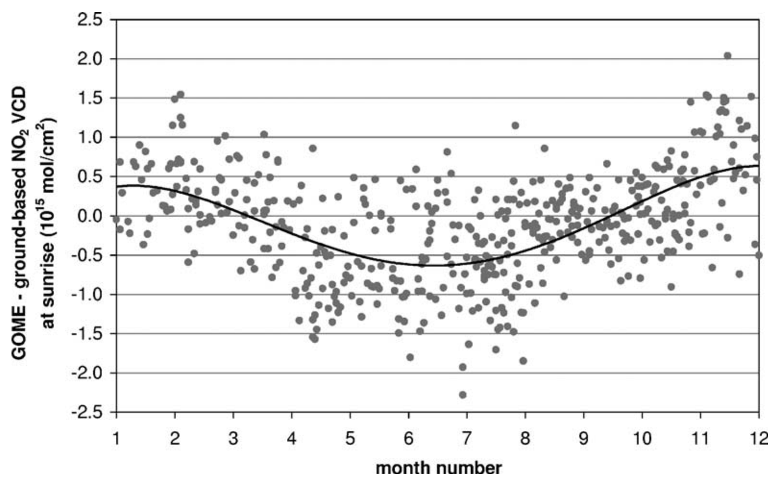

Fig. 3. Absolute difference in NO_2 vertical column density between GOME and ground-based measurements at Issyk-Kul (sunrise) plotted as a function of month.

To understand the reasons of such behavior we have investigated a number of effects that may introduce bias in the results of our ground-based DOAS observations. First, the NO_2 slant column fit is known to depend on the data of O_3 and NO_2 cross sections being used. Also, accounting for O_4 and H_2O molecular absorption (which was missing in our processing) may also matter. To analyze this in detail, we processed a number of our spectra with WINDOAS program, developed at Belgian Institute for Space Aeronomy (IASB/BIRA) (Fayt and Van Roozendael, 2001). According to these calculations, the use of low temperature cross-sections, e.g., measured at ~220 K instead of 243 K (O_3) and room temperature (NO_2) (Vandaele et al., 1998; Bogumil et al., 2000) may decrease resulting NO_2 column at 90° SZA to 14% on the average. In addition, the NO_2 slant columns retrieved at 90° SZA with room temperature NO_2 cross-sections by Graham and Johnston, were found to be ~7% higher than those, calculated using (Vandaele et al., 1998), also measured at room temperature. Finally, we have estimated an effect of molecular absorption by O_4 and H_2O, which we do not consider in our processing, to be within 3% of change in our NO_2 slant column.

In order to match GOME DOAS NO_2 as close as possible, we used O_3 and NO_2 cross sections obtained from the measurements of GOME-FM (Flight Model) recorded at 221 K (Burrows et al., 1998, 1999b) to re-calculate our ground-based data. This has shifted our results ~11% down, increasing difference with GOME to 0.3×10^{15} mol/cm^2 on the average (see Fig. 4). However, not only slant column retrieval may be a subject of bias, but also conversion to vertical column, based on a proper AMF value. For our ground-based data one AMF set was used all over the year (being equal to 16.5 at 90° SZA), which in fact exhibits some seasonal variation due to the changes in atmospheric vertical profiles of pressure, temperature and NO_2. To take this into account, we used seasonal mid-latitude AMF based on the AFGL standard atmosphere, which were calculated at IASB/BIRA with ray-tracing model for 440 nm, with multiply scattering in

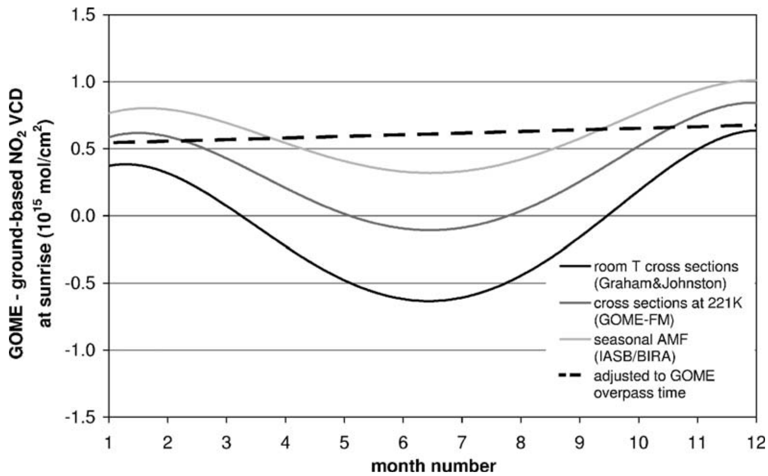

Fig. 4. Difference in NO_2 vertical column between GOME and sunrise ground-based measurements at Issyk-Kul, for different setting of processing: original data with room temperature O_3 and NO_2 cross sections (Graham and Johnston), reprocessed with O_3 and NO_2 cross sections at 221 K (GOME-FM), corrected for seasonal NO_2 AMF (IASB/BIRA), and adjusted to GOME overpass time with chemical box model.

spherical atmosphere, resulting in 18.76, 17.96, 18.17, and 18.19 at 90° SZA in March, July, September, and December, respectively (M. Van Roozendael, personal communication), that decreased our data by ~10% more, producing difference of 0.6×10^{15} mol/cm^2 with GOME, on the average (see Fig. 4).

However, there is still a clear seasonal variation of difference between ground-based sunrise and GOME NO_2 vertical column, along with some offset. To find an explanation, we have investigated diurnal cycle of NO_2 content, which should certainly effect the difference between twilight ground-based and local noon satellite observations. Atmospheric NO_2 is known to exhibit a strong photochemical cycle throughout the day due to its daytime photolysis into NO and its nighttime conversion into N_2O_5. The typical cycle of NO_2 daily run is formed by sharp decrease at sunrise, followed by a quasi-linear slow increase during daytime, then sharp increase at sunset, and finally a slow decrease during night. This standard cycle is therefore characterized by the maximum right after the sunset and a sharp drop to a minimum at sunrise, with the local noon value somewhere in between. To see it in detail, we have looked into the calculations of chemical box model derived from the SLIMCAT 3D chemical-transport model (Chipperfield, 1999), as proposed in (Denis et al., 2005). The model is stacked vertically at 17 levels with time step set to 1 min. It includes 98 chemical and 39 photochemical reactions, including heterogeneous chemistry on liquid and solid particles. Vertical NO_2 columns are obtained by integrating all layers, assuming a constant density in each layer.

An example of such calculation, carried out for the location of Issyl-Kul in January, April, July, and October, is presented in Fig. 5. As it is seen from the plot, the model simulation explains the observed closeness of GOME local noon NO_2 value (~minimum SZA) to the ground-based measurements at dawn (SZA ~90°). Furthermore, as the shape of NO_2 cycle slightly varies with season, the

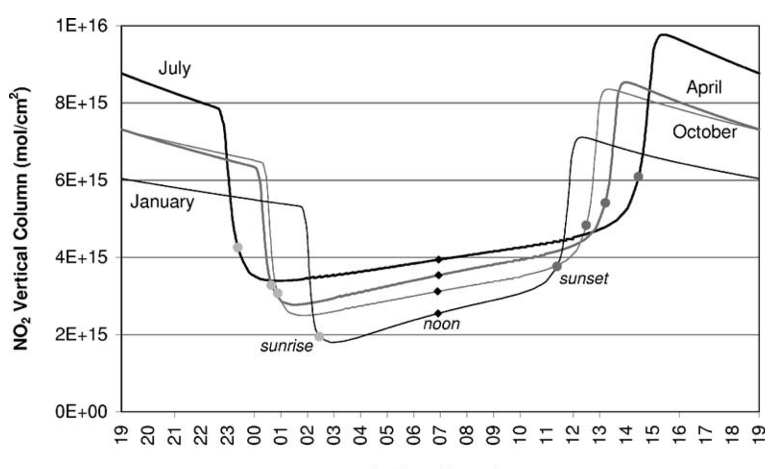

Fig. 5. Diurnal variation of NO_2 vertical column at Issyk-Kul, simulated for different months of the year – January, April, July, and October.

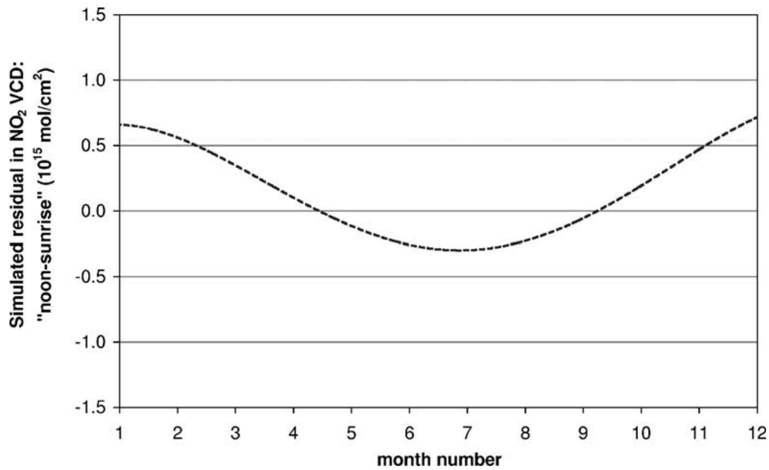

Fig. 6. Seasonal plot of absolute difference between NO_2 vertical column simulated for the moment of local noon and sunrise.

relationship of NO_2 vertical column at sunrise to the local noon also changes. Thus, while the noon NO_2 column is found to be well above sunrise value in winter, it has the same level with sunrise in the autumn and comes below it in the summer. Resulting absolute differences as a function of season are presented in Fig. 6, which reproduce similar curve to what we have seen before in Fig. 3. The model is thus predicting the observed difference between the GOME data and ground-based observations of NO_2 vertical column, and may be used to consider the effect of diurnal variation in our comparison.

Based on the polynomial approximation of chemical model calculations, we have generated a photochemical adjustment factor to bring ground-based sunrise measurements to the moment satellite overpass (~1 h before noon) for the quantitative comparison with GOME. Using that factor we had mostly removed seasonal dependence in our comparison, which could be seen on the same plot in Fig. 6. However, there is a remaining offset between GOME and ground-based data by 0.61×10^{15} mol/cm^2 (see Fig. 4).

6. Conclusions

We have compared the data of operational measurements of NO_2 vertical column with correlative ground-based observations at Issyk-Kul station in Kyrgyzstan for the period of 1996–2002. The Issyk-Kul is a remote site in the central part of Eurasia continent, which makes our study representative for validation of GOME operational NO_2 product in the pollution-free area. According to the results of comparison, the data of GOME is reasonably close to corresponding value of ground-based sunrise twilight observations, presenting some seasonal variation of absolute difference. A number of effects were considered to explain and remove that discrepancy, including the use of similar molecular absorption cross sections and seasonal NO_2 AMF in the processing our ground-based data. Also, simulated NO_x photochemistry has been used to adjust sunrise ground-based measurements to the time of GOME overpass nearby noon. Finally, the data of GOME was found to produce ~18% more NO_2 in vertical column, compared to twilight ground-based measurements, adjusted to noon. This may be an indication to some additional source of disagreement, e.g., incorrect treatment for tropospheric NO_2 content for the region of ground-based measurements. Anyway, presented results are consistent with related GOME validation by recognized NDSC and SAOZ ground-based networks (ESA, 2002), which prove our measurements being valuable for further validation of GOME and similar satellite missions – ENVISAT SCIAMACHY, AURA OMI, and METOP GOME-2.

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