COMPARISON OF THE GOME NO2 TOTAL CONTENT DATA WITH THE RUSSIAN GROUND-BASED MEASUREMENTS

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ABSTRACT

Information on a method, measurement instrumentation, interpretation algorithms used by the Institute of Atmospheric Physics RAS (Zvenigorod Scientific Station) for deriving the column abundance and vertical distribution of NO_2 from the measurements of diffused solar zenith radiation during morning and evening twilight is given. Comparison of the NO_2 ground-based and GOME measurements revealed the large deviations between the satellite and ground-based data. Possible causes of these mismatches (NO_2 daily run, town influence, etc.) are analyzed.

Three-dimensional Atmospheric Chemical Transport Model (ACTM) of the atmosphere has been used for computing the NO_2 column abundance fields. Some examples of comparing the GOME measurements with the model prediction are given.

1. INTRODUCTION

Understanding of NO_2 and NOx global distribution is very important due to well known very active participation of these radicals in sources and sinks of the ozone and other climatically active photochemical gases. In particular the information on NO_2 content and horizontal distribution in a layer involving the tropopause is crucial for estimation of NOx emissions from the current fast growing world fleet of subsonic air transport aircrafts. These emissions affect the ozone content in the above layer, which produces the maximal greenhouse effect. NOx emissions in the stratosphere by future supersonic massive transports have also to be monitored.

For the most part the NO_2 content measurements from satellites use the slant path geometry. The SAGE-2 and POEM-2 instruments have measured the NO_2 atmospheric profiles by the solar radiation absorption in visible spectral range at sunrise and sunset [Refs. 1, 2]. Similar measurements in IR spectral range were carried out in a number of satellite experiments with the Spacelab grill spectrometer [Ref. 3] and the ATMOS interferometer [Refs. 4, 5]. Studies of spatial - temporal variations of the NO_2 vertical structure were also conducted by measurements of thermal radiation of the Earth horizon [Ref. 6, 7]. Beginning in 1995, nadir measurements of the NO_2 total content have been carried out by the GOME spectrometer (ERS-2 satellite) through interpreting the scattered and reflected solar radiation in visible spectral range [Ref. 8].

Ground-based, aircraft and balloon measurements collocated in time and space with the satellite ones are of great importance in estimating the real accuracy of satellite data (e.g. [Ref. 9]). In this study, the results of comparing the GOME data on NO_2 total content with ground-based measurements at Zvenigorod Scientific Station (1996, 1998) and with the prediction of three-dimensional ACTM are given.

2. GROUND-BASED THE NO_2 MEASUREMENTS

Zvenigorod Scientific Station (ZSS) of the Oboukhov Institute of Atmospheric Physics (56°N, 38°E,) is located at a distance of 50 km to the west from Moscow. However, because of the westerly winds prevail in this region through out the year the influence of the polluted air from Moscow on the observations is not too important. In the region of station there are no sources of pollution of the environment. The highways and main roads are far from the station.

The regular measurements of column abundances and vertical profiles of NO_2 at ZSS have been performed since March 1990. The station is a member of the International Network for Detection of Stratospheric Change (NDSC) as a station for complimentary measurements of column NO_2. Vertical NO_2 profiles are retrieved out of the framework of the NDSC activity. First retrievals of NO_2 vertical distributions from ground-based measurements were undertaken by McKenzie et al. [Ref. 10]. In the papers [Ref. 11,12], the NO_2 total content measurements at ZSS were analyzed.

Column amount NO_2 contents are obtained from zenith-scattered solar radiation measured by the grating spectrometer MDR-23 operating in the 435-450 nm wavelength range with spectral resolution 0.7 nm and time of scanning 40 s. A

photomultiplier optimized in the visual wavelength range is used as a detector. The measurements are taken at twilight in mornings and evenings at solar zenith angles 84°-96°, and during daytime , if necessary, to control NO_x pollution of the boundary layer. Detected spectrum is proposed to be the sum of the solar spectrum exponentially attenuated by NO_2 and O_3 absorption and by Rayleigh and aerosol scattering, and a constant. The constant is believed to include effects of detector dark current, spectrometer stray light leakage, and the Ring effect. The slant NO_2 contents are derived from the observed spectra with the use of differential NO_2 absorption.

To retrieve columnar NO_2 contents and NO_2 vertical distributions from the slant NO_2 contents, air mass factors for NO_2 are needed. They are computed using a spherical scattering model for solar radiation and a non-stationary one-dimensional photochemical model including the O_x and NO_x photochemistry. The spherical scattering model takes into account ozone and NO_2 absorption, single molecular and aerosol scattering, refraction and refraction divergence. Parameters of the models: ozone, temperature and air density vertical distributions are seasonally dependent and taken from simultaneous measurements, if available, or from empirical models [Refs. 13-15] specified for the latitude of observations.

The photochemical model provides the altitude-dependent diurnal variation of NO_2, which is the input parameter in the scattering model. Taking into account photochemical processes is very important as NO and NO_2 undergo rapid changes at sunrise and sunset.

Given calculated air mass factors for NO_2, the NO_2 contents in 5-km thick layers and in the thin near-surface layer are then obtained as a solution of the inverse mathematical problem, with the use of modified method similar to that used by McKenzie et al. [Ref. 10] (Chahine method). The modification is concerned with introducing into consideration the thin near-surface layer where NO_2 concentration can be large during pollution episodes. The NO_2 content in the 0-5 km layer does not include NO_2 in the near-surface layer. Derived quantities are (1) NO_2 contents within 5 km thick layers in the stratosphere and the troposphere (0-50 km), (2) NO_2 content in the thin atmospheric surface layer, and (3) columnar NO_2 contents in the troposphere (0-10 km) and the stratosphere (10-50 km) as integrals over appropriate layers. Comparison of the NO_2 profiles retrieved from ground-based and SAGE-2 measurements demonstrated a good agreement of the data.

The precision of determination of NO_2 slant abundances using the only spectrum is about 3 % at the solar zenith angle 90^o, the precision of determination of NO_2 slant abundances with a step 0.5^o is better than 1 %.

3. COMPARISON OF THE GOME AND GROUND-BASED NO_2 MEASUREMENTS

The results of comparing all the ground-based (GS) and satellite (GOME) total NO_2 measurements in pairs are given in Fig. 1. The sets of compared data consisted from 331 GOME measurements with 280 sunrise and 286 sunset ground-based observations for 1996. The simplest approximation was used to consider the diurnal variation of total NO_2 - the GOME measurements were compared with a half-sum of corresponding sunrise and sunset ground-based observations. The reason for such approach is that the GOME measurements occur near the local noon time, as the ERS-2 satellite has a so-called sun-synchronized orbit.

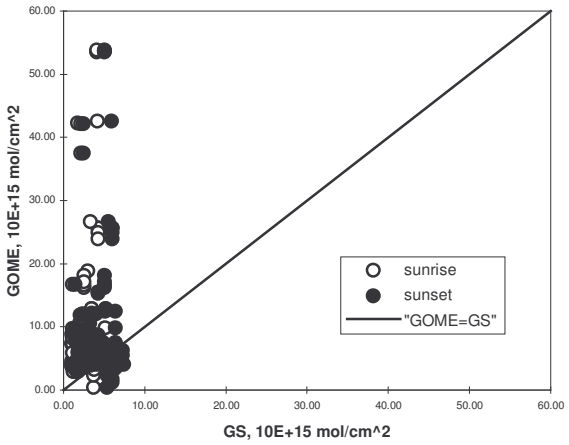

Figure 1. Comparison of satellite (GOME) and ground-based (GS) total NO_2 measurements over ZSS in 1996 (Solid line - the equality "GOME = GS")

Results of comparison demonstrate a very poor agreement between satellite and ground-based measurement systems (it should be taken into account that even negative values were found in the satellite data set, and those were excluded from the comparison). Statistical characteristics of observed discrepancies between GOME (s) and ground-based (g) total NO_2 measurements also prove that conclusion. The mean deviation Δ_{s-g} between the GOME measurements and a half-sum of ground-based observations is 176 % ("sunrise" and "sunset" observations separately deviate from satellite ones by 260 % and 114 %, respectively); standard deviation σ_{s-g} is 549 % (727 % and 394 % for comparison of GOME with "sunrise" and "sunset" observations, respectively); the coefficient of correlation R is - 0.04±0.05 (when comparing GOME measurements with "sunrise" and "sunset" observations those are + 0.01±0.06 and - 0.05±0.06, respectively). It is clear that the GOME significantly (several times!) overestimates the value of total NO_2 in comparison with ground-based observations.

Fig. 2 presents the similar comparison for the limited data set of satellite and ground-based observations - only relatively accurate the GOME measurements (error < 25 %) at the high sun (sun zenith angle SZA < 55°) were included. Results of that comparison demonstrate much better agreement between satellite and ground-based data. The mean deviation Δ_{s-g} for the comparison of GOME measurements with a half-sum of ground-based observations is - 0.1 %, with standard deviation σ_{s-g} = 23.7 %. Although the absolute values of satellite and ground-based measurements agree rather well, correlation of these data sets is still very poor - $R = 0.13 \pm 0.14$. 49 satellite measurements were compared with 39 "sunrise" and 43 "sunset" observations, covering time period from 26 of March to the 5 of September 1996. The corresponding temporal variation of the GOME NO_2 total content data together with a half-sum of "sunrise" and "sunset" observations is presented in Fig. 3. It is clearly seen, that GOME measurements hardly

reproduce variation of daily total NO_2 over the time, if compared with ground-based observations.

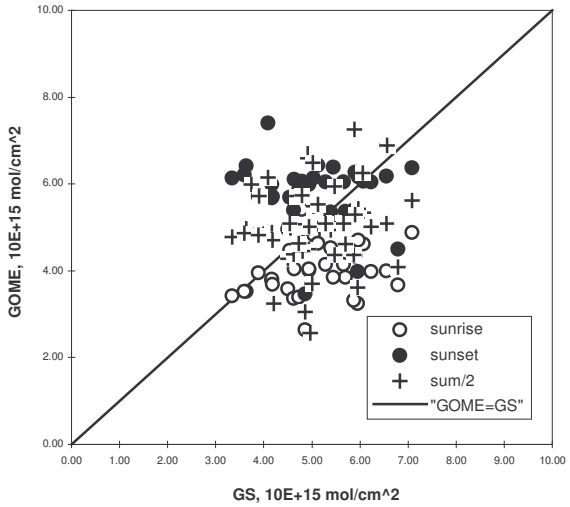

Figure 2. Comparison of satellite (GOME) and ground-based (GS) total NO_2 measurements over ZSS in 1996 (limited dataset).

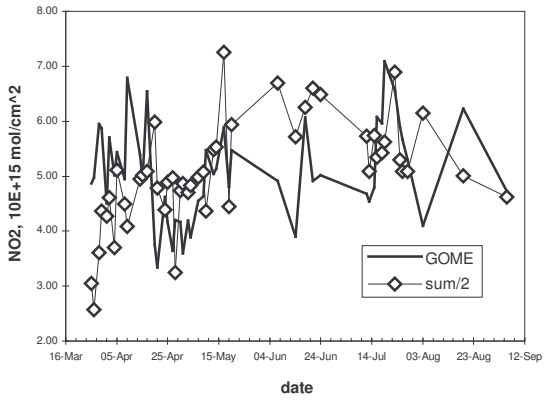

Figure 3. Temporal variation of NO_2 total content from satellite (GOME) and ground-based measurements over Zvenigorod (limited dataset)

Additional comparison was performed for the GOME NO_2 total content data (s) with ground-based observations (g) over Zvenigorod station in March 1998 ("sunset" measurements only). The temporal variations of total NO_2 satellite and ground-based observations are presented in Fig. 4. Satellite measurements give the total NO_2 values exceeding, on the average, by 30.5 % the ground-based ones, with corresponding standard deviation σ_{s-g} = 60.0 %. The number of measurements in the comparison (only 10 pairs were available) is insufficient to make any strong conclusion, but it is possible to note that the agreement between two data sets is much better than it was in 1996 (the average discrepancies were Δ_{s-g} = 114 %, σ_{s-g} = 394 %). GOME measurements performed on March 26 and 29 are nearly 2 times higher the ground-based ones; an elimination of these data from the comparison reduces the average discrepancy between satellite and ground-based systems to 8.9 %, with corresponding standard deviation σ_{s-g} = 20.2 % relative to 30.1 % in 1996. So, there is some reason to conclude that the GOME data processed by new improved code are better agree with ground-based measurements.

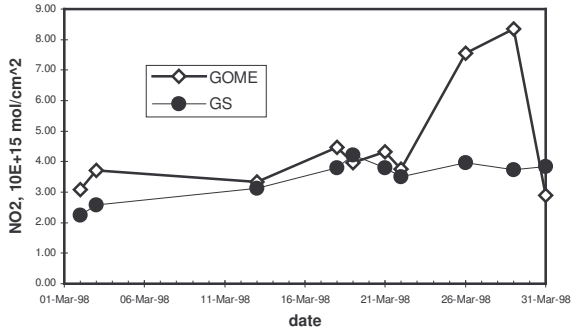

Figure 4. Comparison of satellite (GOME) and ground-based "sunset" (GS) total NO_2 measurements over ZSS in March 1998.

Diurnal cycle of total NO_2 is well expressed. Total NO_2 value changes several times, with a rapid decrease at sunrise, growing slowly during daylight until sunset, when it quickly reaches the maximum and starts to reduce gradually until the next dawn. An example of one-day total NO_2 variation, calculated with a use of special photochemical routine of the UIUC 3-D ACTM (see sec. 4) is presented in Fig. 5. Simulation was performed for the first day of each month, giving diurnal cycle of total NO_2 over location of Zvenigorod station. It is clearly seen from this Figure, that the "noon" value is found between "sunrise" and "sunset" values. According to ACTM simulations, the relative variation of total NO_2 over Zvenigorod during the daylight is minimal in May - less than 7 %, and maximal in August - up to 15 %.

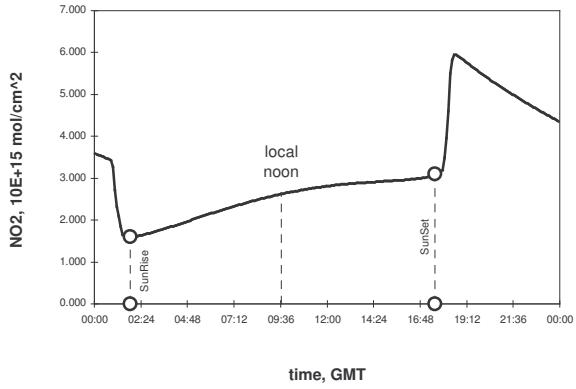

Figure 5. Diurnal cycle of total NO_2 over Zvenigorod (August 1998) (dashed lines indicate the time of sunrise, sunset and local noon)

4. NUMERICAL SIMULATION OF NO_2 GLOBAL FIELDS

In modeling, the 3-Dimensional Atmospheric Chemical Transport Model, developed at University of Illinois at Urbana-Champaign , USA (UIUC 3-D ACTM). The UIUC tropospheric/stratospheric 24-layer 3-D ACTM [Ref 16] have been used. It consists of 3 main modules: (1) a Hybrid advective transport routine, which includes Prather scheme for vertical transport and Semi-Lagrangian scheme for horizontal

transport, (2) a photochemical routine that includes the principal gas-phase and heterogeneous reactions and uses a pure implicit iterative Newton-Raphson routine for solving the set of continuity equation for gas species, (3) a module with prescribed temperature, tropospheric humidity and circulation fields. The developed ACTM is global with a horizontal resolution of 4^0 latitude and 5^0 longitude. In the vertical direction the model extends from the earth's surface to 1 hPa. The sources of NO_X and CO as well as near surface mixing ratio of other source gases (CFC's, CH_4 and N_2O) are prescribed for 1995 conditions. The circulation and temperature fields are acquired from 24-L UIUC AGCM as well as from assimilated UKMO (U. K. Meteorological Office) dataset.

Evaluations of 3-D ACTMs have been performed using observed climatological data or observed species distributions for particular locations or time periods. These comparisons showed that 3-D models can reproduce many features of the observed species distributions [e.g. Ref. 17]. However, substantial disagreements between model-simulated and observed data have also been reported in these studies. On the other hand, an extensive validation of the model results (that reflect the current level of theoretical knowledge) with observation data and especially the evaluation of any kind of substantial disagreement between measured and simulated trace gas distributions can be very useful for further progress of the science and may also help to improve the quality and accuracy of the measurements.

4.1. Comparison of the GOME total NO_2 global distribution with model prediction

Comparing the GOME total NO_2 global measurements with simulated spatial distributions may help to understand the reasons of the observed discrepancies between satellite and ground-based systems. Such investigation was started in 1998 year. ??

As an example, 3-days averaged total NO_2 GOME data (25-27 July 1996) was compared with corresponding local noon ACTM simulation. Satellite measurements were averaged and interpolated onto the ACTM spatial grid. Fig. 5 presents a map of relative difference of the observed total NO_2 from simulated values, in %. The GOME NO_2 total content data significantly deviate from the simulation - up to 130%. The best agreement between modeled and GOME data is found in the low latitudes of northern hemisphere, and in some parts of southern hemisphere - the relative discrepancy is less then ± 30 %. In most part of northern hemisphere the GOME total NO_2 data exceed the simulated ones- discrepancy is more than 30 %. On the south, results are opposite - the GOME total NO_2 values are less the simulation by 30-90 %. It is necessary to note, that both data sets (satellite and modeled) produce similar zone distribution of total NO_2 over the globe - it is maximal in the high northern latitudes and decrease to the south. So, it is possible to conclude, that GOME overestimate (in comparison with ACTM) the high NO_2 total contents and underestimate the low ones. Of course, these results are just preliminary; such investigations are at the initial stage and will be continued.

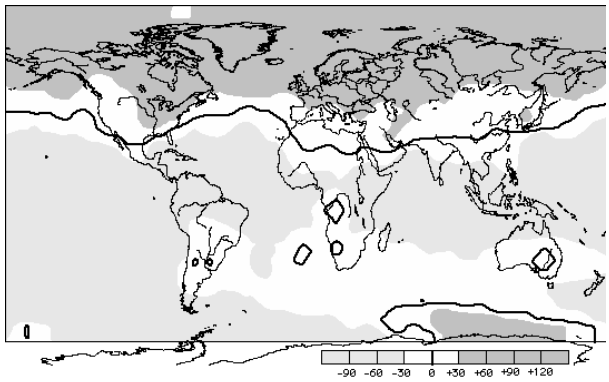

Figure 5. Relative difference of the GOME total NO_2 from simulated (ACTM) values, in % (25-27 July 1996).

4.2. Comparison of the ground-based total NO_2 measurements with model prediction

The results of an 8-year steady-state model run have been reprocessed and the total vertical NO_2 column compared with the data collected at Zvenigorod in 1996 an 1997 years. In this case the model NO_2 values were sampled every 6 minutes for the first day of every month to describe better the diurnal NO_2 variations. Results of comparison for sunrise conditions show that the model mimics rather good the observed seasonal cycle which is characterized by the maximum of vertical column NO_2 in June and minimum vertical column NO_2 during the winter season. However, the difference between simulated and observed data could be rather substantial. The model underestimates vertical column NO_2 from April to August up to 20-40 %. During January and February simulated vertical column NO_2 exceeds measured values by almost 100%. More or less reasonable agreement (within uncertainty of the measurements) occurs in March and from August to November. The causes of such disagreements can be connected with not accurate time coincidence of the measured and simulated data. The other possible sources of the disagreement are rather rough horizontal resolution of the model and the using of GCM generated wind fields to drive ACTM.

It is evident that the agreement between NO_2 ground-based and simulated data is much better then that between the GOME and modeled values.

5. SUMMARY

1. Results of comparison between GOME and ground based NO_2 column amount measurements in 1996 (ZSS, Russia; 56°N, 38°E) (331 GOME measurements, 280 sunrise and 286 sunset ground-based observations) demonstrate a very poor agreement between satellite and ground-based measurement systems. The mean deviation Δ_{s-g} between the GOME measurements and a half-sum of ground-based observations is 176 %, standard deviation σ_{s-g} is 549 %, the coefficient of correlation R is - 0.04±0.05. It is clear that GOME significantly overestimates the value of total NO_2 in comparison with ground-based observations.
2. The similar comparison for the limited data set of satellite and ground-based observations in 1996 - only relatively accurate the GOME measurements (error < 25 %) -

demonstrates much better agreement between satellite and ground-based data. The mean deviation Δ_{s-g} for the comparison of GOME measurements with a half-sum of ground-based observations is - 0.1 %, with standard deviation σ_{s-g} = 23.7 %. Although the absolute values of satellite and ground-based measurements agree rather well, correlation of these data sets is still very poor - $R = 0.13 \pm 0.14$.

3. Comparison of the GOME NO_2 total content data and ground-based observations over Zvenigorod station in March 1998 shows that satellite measurements give, on the average, 30.5 % higher the total NO_2 values than ground-based system, with corresponding standard deviation σ_{s-g} = 60.0 %. GOME measurements performed on March 26 and 29 are nearly 2 times higher than the ground-based ones. Elimination of these data from comparison reduces the average discrepancy between satellite and ground-based systems to 8.9 %, with corresponding standard deviation σ_{s-g} = 20.2 %. Our preliminary conclusion is that the GOME data of March 1998 has the better quality than data of 1996.
4. Comparative analysis of total ozone measured by GOME and modeled by three-dimensional Atmospheric Chemical Transport Model for 25 - 27.07.96 shows some differences both in tropical and in polar zones. Model overestimates the total NO_2 in polar regions and underestimates it in tropics relative to GOME results, but their globally averaged values are close to each other. The more smoothed latitudinal distribution of the modeled total ozone relative to GOME data is partly due to using the climatic July wind field in calculations having no actual wind field data for the considered period.

6. REFERENCES

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