ABSTRACT

SAOZ (Système d’Analyse par Observations Zenithales) is a ground-based UV-Visible zenith-sky spectrometer installed between 1988 and 1995 at a number of NDSC stations at various latitudes on the globe. The instrument is providing ozone and NO$_2$ vertical columns at sunrise and sunset using the Differential Optical Absorption Spectroscopy (DOAS) technique in the visible spectral range. The ERS-2 GOME 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. It was then followed by SCanning ImAging spectroMeter for Atmospheric ChartographY (SCIAMACHY) onboard ENVISAT in 2002, and Ozone Monitoring Instrument (OMI) onboard EOS-AURA in 2004, with a similar capacity to monitor total NO$_2$. All these instruments are nadir viewing mapping spectrometers, applying the DOAS technique in the visible for deriving the NO$_2$ total column. Here we present the results of NO$_2$ long-term comparisons between GOME and SAOZ for the whole period of GOME operation since 1995 at all latitudes – tropics, mid-latitudes and polar regions - in both hemispheres. Comparisons are also shown with the most recently available SCIAMACHY and OMI data in 2004-2005. Overall, the daytime satellite measurements (around noon) are found consistent with sunrise ground-based data, with an average smaller difference at the tropics and mid-latitudes than in the polar areas in the summer. The agreement is even improved after correcting for the NO$_2$ photochemical change between sunrise and the satellite overpass using a box model. However, some seasonal dependence of the difference between ground-based and satellite total NO$_2$ still remains, related to the accuracy of photochemical simulations and the set of NO$_2$ air mass factors used in the retrievals of both systems.

1 SAOZ AND SATELLITE TOTAL NO$_2$ MEASUREMENTS

1.1 SAOZ

SAOZ is a network of ground-based UV-Visible spectrometers for zenith-scattered sunlight observations, installed at a number of NDSC stations over the globe [1]. The NO$_2$ vertical column is retrieved using the DOAS technique applied to zenith-sky measurements in the visible range (410-530 nm) at twilight. The measurements are averaged in the range of 86°-91° sun zenith angle, providing an accurate estimate of NO$_2$ vertical column at sunrise and sunset. Here we present the comparison of satellite data with the measurements of 8 SAOZ instruments: Scoresby Sund, Sodankyla, Salekhard, OHP, Reunion Island, Bauru, Kerguelen and Dumont d’Urville (Fig. 1).
1.2 Satellite NO$_2$ measurements – ERS-2 GOME, ENVISAT-SCIAMACHY and AURA-OMI

GOME was launched on April 21st 1995 on board the second European Remote Sensing Satellite (ERS-2) [2]. ERS-2 is on a sun-synchronous polar orbit crossing the equator at 10:30 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 the UV-visible spectral range (240-790 nm) with a moderate spectral resolution of 0.2 to 0.4 nm. The field of view may be varied in size from 40x320 km$^2$ (forward scan) to 40x960 km$^2$ (back scan). With the large swath, global coverage is achieved every three days at the equator and more frequently at higher latitudes. Providing the global picture of atmospheric ozone, GOME was also the first spaceborne instrument having the capability of measuring the total column amount of nitrogen dioxide (NO$_2$). The trace gas column densities, given in the GOME products (ozone and NO$_2$), are derived by the DOAS technique.

SCIAMACHY is a UV/visible/NIR grating spectrometer covering 220-2400 nm range with 0.2-1.5 nm spectral resolution depending on wavelength [3]. It was launched on ENVISAT on March 1st 2002 into a sun-synchronous orbit with a 10:00 equator crossing local time (descending node). The UV-visible nadir measurements of SCIAMACHY are very similar to those of GOME, but with better spatial resolution - 30×30 to 30×240 km$^2$. Global coverage at the equator is achieved in 6 days (due to alternative nadir and limb measurements along the orbit) and more frequently at higher latitudes.

OMI is a nadir viewing, wide swath, UV-visible imaging spectrometer [4]. It combines the advantages of GOME, SCIAMACHY and TOMS, measuring the complete spectrum in the UV-visible wavelength range (270-500 nm) with a very high spatial resolution (13×24 km$^2$) and achieves daily global coverage of all products (2600 km wide swath). It is using two-dimensional detectors for simultaneous spatial and spectral registration (0.4-0.6 nm resolution). OMI was launched on July 15th 2004 on board NASA’s AURA platform into sunsynchronous near polar orbit, with 13:45 equator crossing local time.

2 COMPARISON OF SAOZ NO$_2$ WITH SATELLITE DATA

2.1 Comparison of SAOZ sunrise and sunset with satellite total NO$_2$ data

Time series of SAOZ NO$_2$ vertical column (sunrise, sunset) are compared in Fig. 2 to with GOME (operational GDP4) and OMI (provisional overpass total column) data in 2004-2005. The GOME data are those of the closest pixel from the SAOZ location within a distance of 500 km. For OMI of higher spatial resolution the pixel selected is the closest from SAOZ, within a distance of one pixel size (20-30 km). Unfortunately, there is no GOME data to compare with SAOZ in the Southern Hemisphere in 2004-2005 due to failure of the on-board recorder since June 2003. The satellite data are very close to the SAOZ sunrise measurements. Best agreement could be seen at mid-latitudes (OHP). At high latitudes - Scoresby Sund, Sodankyla, Salekhard, during summer, satellites show less NO$_2$ compared to SAOZ, and fluctuates due to the strong NO$_2$ diurnal variation along the polar day (discussion later). Provisional OMI total NO$_2$ data also shown on the plots, display high day-to-day variations compared to SAOZ, and extremely high values during the polar day. This overestimation comes from high tropospheric NO$_2$ detected by OMI, but not observed in SAOZ, the zenith sky observation mode at twilight being little sensitive to tropospheric NO$_2$. For improving the comparison with SAOZ, only the stratospheric part of OMI NO$_2$ will be used in the following obtained by taking the difference between total and tropospheric NO$_2$ provisional products. For SCIAMACHY, the stratospheric NO$_2$ column provided by the University of Bremen is used [5]. The pixel selected is the closest within 200 km from SAOZ.

2.2 Correction of satellite data for NO$_2$ diurnal variation

Atmospheric NO$_2$ is known to exhibit a strong photochemical cycle throughout the day due to its daytime photolysis into NO and nighttime conversion into N$_2$O$_5$. The NO$_2$ daily cycle starts with a fast drop a little after sunrise followed by a quasi-linear slow increase during the day, a fast increase at sunset, and finally a slow decrease during night. The diurnal cycle has been simulated with a box photochemical model derived from the SLIMCAT 3D chemical-transport model [6]. It includes 98 chemical and 39 photochemical reactions, including heterogeneous chemistry on liquid and solid particles. Calculations are made at 17 altitude levels with a time step of 1 minute. The NO$_2$ total column is obtained by integrating the profile assuming a constant density in each layer.
Figure 3 shows the results of simulations at two SAOZ stations, OHP at mid-latitude and Scoresby Sund in the Arctic for summer and winter. Excepted in the Arctic summer, the noontime column is close to its sunrise value. However, the ratio between these two changes with the season. The noontime column is larger than that of sunrise in the winter and smaller in the summer, especially during polar day. In addition, a difference also appears between satellites measurements at 10:00 (SCIAMACHY), 10:30 (GOME) and 13:45 (OMI) which needs to be taken into account particularly at mid-latitude. For doing this, the daily variation of the ratio NO₂(sunrise)/ NO₂ as been calculated for each of the 12 months at each SAOZ location. As SAOZ is an average of measurements between 86 and 91 SZA the NO₂ column at 88.5° SZA is considered is used as sunrise reference. All satellite measurements have been normalized to sunrise values using these ratios. This approach proved to be useful for the comparison of satellite NO₂ data with ground-based SAOZ [7]. An example of this correction applied to the measurements over the Arctic Scoresby Sund SAOZ station is displayed in Fig. 4. After correction for the photochemical change the agreement between satellite and SAOZ sunrise measurements improves showing also less scatter during summertime.
2.3 Comparison of SAOZ and satellite columns adjusted to sunrise

All GOME, SCIAMACHY and OMI data selected for comparison with SAOZ have been normalized to sunrise using this method. Figure 5 displays the resulting NO$_2$ column time series in 2004-2005 over 8 SAOZ stations. The agreement is excellent almost everywhere, but at Salekhard where the SAOZ column is underestimated. The reason for that is being investigated. However, there is clear systematic difference between OMI and SAOZ at OHP in France and Bauru in Brazil. This is very likely due to a small contribution of tropospheric pollution frequent in these regions still present in the SAOZ data but removed from the OMI column. The average difference, RMS and correlation between each satellite and SAOZ measurements at each station is given in Table 1. Best agreement with SAOZ is found for the SCIAMACHY stratospheric column (average difference less than 0.2·10$^{15}$ mol/cm$^2$ at 4 out of 8 SAOZ stations, and RMS between 0.3 to 0.7·10$^{15}$ mol/cm$^2$. The OMI stratospheric column retrieved agree with that of SAOZ within ±0.6·10$^{15}$ mol/cm$^2$, but systematically lower – by 0.4·10$^{15}$ mol/cm$^2$. In contrast, the GOME total column is larger than that of SAOZ by 0.2±0.7·10$^{15}$ mol/cm$^2$ on average. The absolute differences between SAOZ and satellite data, at polar, mid-latitude and tropical regions as a function of season, are summarized in Fig. 6 (Salekhard excluded). The SCIAMACHY stratospheric column agree with SAOZ within ±0.6·10$^{15}$ mol/cm$^2$, with an average difference ∼0 at polar and mid-latitudes, and -0.1·10$^{15}$ mol/cm$^2$ lower in the tropics. The GOME column is larger than that of SAOZ by 0.2 to 0.3·10$^{15}$ mol/cm$^2$ at all latitudes. The OMI stratospheric column calculated from the difference between OMI total and tropospheric columns is lower than that of SAOZ by 0.2 to 0.6·10$^{15}$ mol/cm$^2$ at all latitudes suggesting an overcorrection of the data for the tropospheric column. A systematic seasonal variation of the difference between all satellites and SAOZ could be observed due to the use of a constant AMF throughout the year in the SAOZ retrievals though the altitude of the maximum ozone concentration increases in the summer.

3 SUMMARY

GOME, SCIAMACHY and OMI NO$_2$ columns, and stratospheric columns in the case of SCIAMACHY and OMI have been compared with correlative ground-based SAOZ measurements in polar mid-latitudes and tropical regions. For comparison purpose all satellite data have been compensated for NO$_2$ diurnal change and normalized to sunrise values using a photochemical box model. Best agreement with SAOZ is found for SCIAMACHY stratospheric NO$_2$ column within 0.5·10$^{15}$ mol/cm$^2$. The operational GOME total column is larger than that of SAOZ by 0.2·10$^{15}$ mol/cm$^2$ on average, and the OMI stratospheric column smaller by 0.4·10$^{15}$ mol/cm$^2$. A systematic seasonal variation with a summer maximum is observed in the difference between SAOZ and all satellite satellite data, attributed to the use of a constant AMF instead of a seasonal dependent AMF in the SAOZ retrievals which will need further correction.
Fig. 5. Comparison of ground-based SAOZ satellite data, adjusted to sunrise – GOME total NO$_2$, SCIAMACHY and OMI stratospheric NO$_2$ (2004-2005)

4 ACKNOWLEDGEMENT

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Netherlands (NIVR/KNMI), Finland (FMI) and the United States of America (NASA) in the Earth Observing System (EOS) Aura mission. This work was partly supported by ESA within the TASTE project.

Table 1. Absolute average and r.m.s. difference (Δ, σ), and correlation (R) between ground-based SAOZ and satellite data, adjusted to sunrise – GOME total NO$_2$ (1995-2005), SCIAMACHY stratospheric (2004-2005), and OMI total and stratospheric NO$_2$ (2004-2005)

<table>
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<tr>
<th>STATION</th>
<th>GOME GDP4</th>
<th>SCIAMACHY IUPB, strato</th>
<th>OMI OMNO2, total</th>
<th>OMI OMNO2, strato</th>
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<td></td>
<td>Δ</td>
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Fig. 6. Absolute difference between SAOZ and satellite data adjusted to sunrise – GOME total NO$_2$ (1995-2005), SCIAMACHY and OMI stratospheric NO$_2$ (2004-2005), as a function of season (at polar, mid-latitudes and tropics)

5 REFERENCES