Comparison of the GOME ozone and NO_2 total amounts at mid-latitude with ground-based zenith-sky measurements

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Abstract. This paper reports on the details of the comparison method used to validate the GOME vertical column amounts of ozone and nitrogen dioxide by means of ground-based zenith-sky observations, with emphasis on the SAOZ data from the International Scientific Station at the Jungfraujoch (46.6°N, 8°E, Switzerland). For validation purpose, the accuracy of the SAOZ measurement is analysed, a special care being given to the retrieval of total ozone. Possible SAOZ contributions to the origins of discrepancy between GOME and the ground-based observations are identified and quantified. A methodology of comparison is defined, taking into account the optimisation of the co-location of the air masses probed by the satellite and the ground-based instruments. The capabilities and the limitations of the method are investigated with special care. The diurnal variation of the NO_2 total column has also been considered.

Introduction

The GOME instrument (Global Ozone Monitoring Experiment), on-board the second ESA Earth Remote Sensing platform (ERS-2), was launched on the 21st of April 1995 onto an heliosynchronous polar orbit. Its main scientific objective is the study of trace constituents in the lower and the middle atmosphere. GOME is the first spaceborne instrument using the Differential Optical Absorption Spectroscopy technique (DOAS) to detect atmospheric trace species such as ozone, nitrogen dioxide, OClO and BrO. A GOME Geophysical Validation Campaign started on 20 July 1995, requiring the collection of ground-based, balloon and spaceborne correlative observations. Within the scope of the validation of the GOME total vertical columns of ozone and nitrogen dioxide, the Belgian Institute for Space Aeronomy has co-ordinated the activities based on the NDSC (Network for the Detection of Stratospheric Changes) Alpine and complementary stations, and has also contributed to the world-wide validation project relying on the SAOZ network. First validation results of the previous version of the GOME ozone and nitrogen dioxide measurements were reported in January 1996 (Lambert et al., 1996[a, b]). The detection of BrO and OClO signatures in the GOME spectra in Antarctica from July to September 1995 was reported as well (Eisinger et al., 1996). Since, the GOME algorithms have been revisited and the agreement with correlative measurements has been improved. GOME total vertical columns of ozone and nitrogen dioxide are now routinely measured.

The purpose of this paper is to define the methodology of comparison used for the validation of the GOME vertical column amounts of ozone and nitrogen dioxide by means of ground-based zenith-sky DOAS observations, with emphasis on the SAOZ data from the International Scientific Station at the Jungfraujoch (ISSJ). The capabilities and the limitations of the method have been investigated with special care. Possible SAOZ contributions to the origins of the discrepancy between GOME and the ground-based observations have been identified and quantified. First validation results of the current GOME version 2.0 data obtained with the world-wide SAOZ network are reported by Lambert et al. in this issue.

Description of the instruments

The Global Ozone Monitoring Experiment

GOME is a combination of four grating spectrometers observing the solar radiation scattered from the atmosphere or from the Earth's surface, covering the spectral range 240-790 nm. The instrument is operated in the nadir-viewing geometry with a 960 km swath width. Narrow absorption features due to ozone, nitrogen dioxide and other key constituents such as OClO and BrO are detected in the UV and the visible by means of the DOAS technique, based on the fit of the calculated differential optical thickness with the observed one. The observed column densities along the line of sight, or slant columns, are converted into total vertical columns by using a geometrical enhancement factor, or air mass factor (AMF), which is calculated by a radiative transfer model assuming given vertical distributions of the atmospheric constituents controlling the penetration of the solar radiation in the atmosphere.

The ground-based instruments

The SAOZ instrument (Système d'Analyse par Observation Zénithale) is a grating spectrometer looking at the sunlight scattered at the zenith (Pommereau and Goutail, 1988). The UV-visible part of the zenith-sky spectrum is recorded during twilight periods for SZA ranging from 86° up to 91°. The DOAS technique is used here to detect absorption features due to ozone, nitrogen dioxide, O_4, H_2O, OClO and BrO. In particular, ozone slant amounts are derived from the absorption in the Chappuis bands, between 470 and 540 nm, while the 406-526 nm window is used for nitrogen dioxide. The slant columns are converted into total vertical columns by using a standard AMF, which is calculated by a validated radiative transfer model (Sarkissian et al., 1995).

For the present study, the nearly co-located total ozone measurements performed by the Dobson and Brewer spectrophotometers at Arosa (46.5°N, 9.4°E, Switzerland) and Hohenpeißenberg (47.8°N, 11°E, Germany) and the Brewer-Mast ozone soundings at Payerne (46.5°N, 6.6°E, Switzerland)

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have provided useful ancillary information. These instruments, in addition to the SAOZ at the ISSJ, are part of the NDSC/Alpine and complementary stations.

Accuracy of the ISSJ SAOZ data

In order to detect possible SAOZ contributions to the origins of discrepancy between GOME and the ground-based observations, the accuracy budget of the SAOZ measurement is analysed with respect to various critical parameters. Possible GOME contributions are highlighted as well.

Absorption cross-sections

In the Chappuis bands, the uncertainty on the laboratory absorption cross-sections used in the fitting procedure introduces a systematic error of about 3% in the total ozone. For NO_2, the systematic error introduced by the uncertainty on the absorption cross-sections can reach 15%. The spectral analysis itself generates a pseudo-random noise lower than 1%. The temperature dependence of the NO_2 absorption cross-sections introduces an additional uncertainty of about 3% (Johnston and McKenzie, 1989), rising from fluctuations in the atmospheric temperatures. The weak temperature dependence of the ozone differential absorption cross-sections in the Chappuis bands has no significant effect on the DOAS ozone retrieval (Burkholder and Talukdar, 1994). On the other hand, the ozone measurement of GOME is based on the absorption of ozone in the Huggins bands, where the ozone absorption cross-sections are known to be temperature dependent. Hence, the SAOZ can be used to assess the uncertainty of the GOME total ozone associated with atmospheric temperature fluctuations.

SAOZ Air Mass Factor

The AMF, used to convert the observed ozone or NO_2 line-of-sight amounts into vertical total amounts, depends on the scattering geometry and is sensitive to fluctuations in pressure, temperature and ozone vertical distributions. SAOZ data obtained with the standard SAOZ AMF were compared to those retrieved with a zenith-sky AMF calculated with ozone vertical profiles measured at Payerne by means of Brewer-Mast ozonesondes (Lambert et al., 1996[a]). The comparison showed that the daily fluctuations in the scattering geometry might account for ± 1% of scatter in the SAOZ total ozone while the seasonal variation introduces a seasonal systematic bias of about 3% at mid-latitude. Following previous works (Solomon et al., 1987, Perliski and Solomon, 1993), the uncertainty on the NO_2 zenith-sky AMF is estimated to be about 10% at 90°SZA.

The SAOZ data retrieved with the two different AMF were compared to the total ozone measured with the Dobson spectrophotometers operated at Arosa and at Hohenpeißenberg, as displayed in Figure 1 for the summer-autumn 1995. The figure demonstrates the better agreement reached with the SAOZ total ozone retrieved with a corrected AMF. The mean difference between the SAOZ and the Dobson total ozone is cut down from 1.6±2.8% to 0.6±2.1% at Arosa and from 3.9±3% to 2.7±2.5% at Hohenpeißenberg. The superior agreement with Arosa can be partly explained by the closest location of the stations. Figure 1 still shows that, even after the AMF correction, a seasonal dependence remains between SAOZ and Dobson, more pronounced with Hohenpeißenberg.

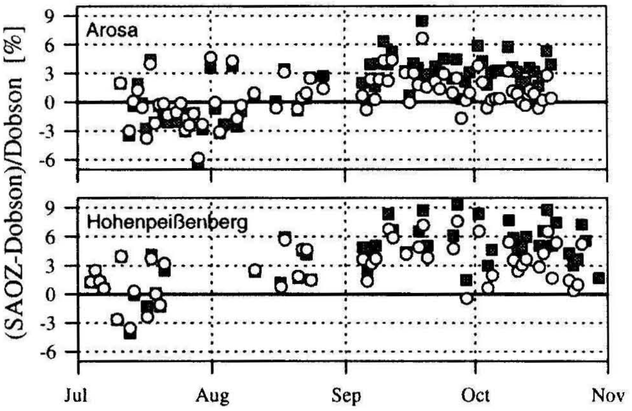

Figure 1 Relative differences (in percentage) between the total ozone measured in summer-autumn 1995 by the SAOZ at the ISSJ and by the Dobson at Arosa and Hohenpeißenberg. SAOZ data are obtained with: (1) standard AMF (dark squares); (2) AMF calculated with the Payerne ozone soundings (open circles).

Tropospheric perturbations

At twilight, the tropospheric contribution to the total absorption seen by a zenith-sky instrument is lower than 4%. However, the tropospheric contribution could occasionally increase due to pollution events. Moreover, tropospheric multiple scattering, generated by fog, thick clouds or snow showers, not taken into account in the SAOZ AMF calculation, could also increase the tropospheric contribution by enhancing the light path. In addition, absorption by O_4 and H_2O, which interfere with ozone, are enhanced by tropospheric multiple scattering as well, producing a bias in the retrieved ozone amounts (Van Roozendael et al., 1994). The long-term comparison of the total ozone measured by the SAOZ at the ISSJ and the Dobson at Arosa shows that this random error should not exceed 1% on average.

Residual amount in the reference spectrum

The observed optical thickness consists in the logarithm of the ratio between the observed and a reference spectrum recorded at low SZA. The uncertainty on the residual amount of absorber contained in the reference spectrum introduces a constant offset in the retrieved total column, depending on the method used to estimate the residual amount (Vaughan et al., 1996). With the classical methods using a zenith-sky reference spectrum, the uncertainty on the residual ozone, mainly attributed to the error on the zenith-sky AMF at low SZA, leads to a systematic offset in the total ozone of about 3%. The use of a direct Sun reference spectrum reduces significantly this uncertainty and hence the offset.

Long-term quality control

Calibration changes should not exist for SAOZ instruments since they are self-calibrated in wavelength with the solar Fraunhofer lines. SAOZ and other UV-visible DOAS zenith-sky spectrometers, as well as the instruments operated at the NDSC/Alpine stations, regularly participate to intercomparison campaigns in order to control their quality, to assess their accuracy and to examine their consistency with other types of instruments. For example, at the intercomparison campaign held in September 1994 at Camborne (UK) for UV-visible DOAS zenith-sky spectrometers, the agreement between four SAOZ and two other DOAS spectrometers was within 5% for total NO_2 and 3% for ozone, as well as for the co-located Dobson measurements and ECC ozone soundings (Vaughan et al., 1996).

In summary, the following improvements have been implemented in the retrieval of the SAOZ data at the ISSJ, especially in the frame of the GOME validation exercise: (i) the calculation of a daily AMF by means of ozone profiles measured at Payerne; (ii) the most accurate method for the ozone residual amount estimation in the reference spectrum by using a direct Sun spectrum as reference; (iii) the rejection of erroneous SAOZ data after detection of tropospheric multiple scattering events by using the measured slant amounts of O_4 and H_2O. The error budget analysis on a ground-based zenith-sky measurement leads then to an accuracy of about 3.5% for ozone and 10-15% for NO_2.

Methodology of comparison

This section describes the methodology defined for comparing the GOME and the zenith-sky ground-based total ozone and nitrogen dioxide. Some potential sources of discrepancies between the different instruments, due to their own observing mode, are highlighted.

Coincidence of the Probed Air Masses

For a reliable comparison, the air masses probed by GOME and by the ground-based instruments should be as similar as possible. While the most accurate zenith-sky measurements are obtained at twilight, ERS-2 overpasses the mid-latitudes around local noon. Therefore, a significant difference in time exists between the air masses sampled by GOME and by the SAOZ, varying from 5 hours in winter up to 8 hours in summer for a mid-latitude site. This time difference makes the comparison of single measurements irrelevant when the ozone field exhibits a high temporal variability. During a spell of low temporal variability, the comparison can be reliable if the variability is lower than the measurement accuracy, what can be verified by examining dawn and dusk SAOZ total ozone and the evolution of nearly co-located Dobson or Brewer observations through the day. The effect of the measurement time difference on the statistical results was assessed by comparing time series of GOME, Dobson and Brewer total ozone at Arosa, with varying the time span between the satellite and the ground-based observations from 2 hours up to the whole day. The comparison showed that expanding the time span increases the scatter by a few per cent but does not bias the average agreement. Therefore comparison results obtained with twilight measurements during both high and low variability periods can be used to carry out statistical investigations such as the study of trends or of seasonal dependences.

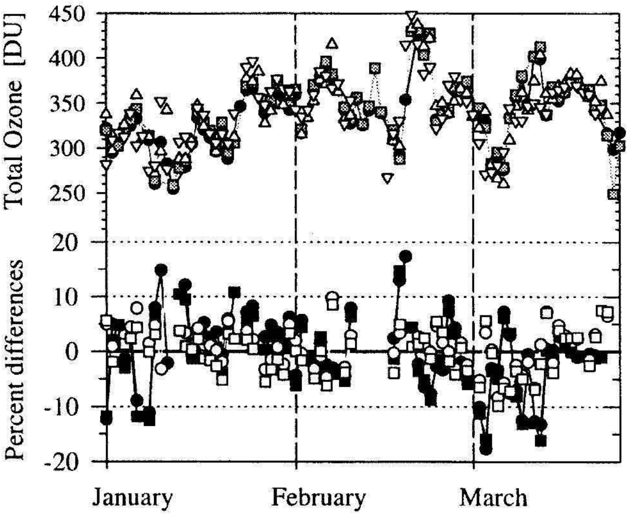

Figure 2 Upper part: total ozone (in Dobson units) measured in winter 1996 at Arosa (Dobson in dark circles and Brewer in grey squares) and by the SAOZ at the Jungfraujoch (open triangles, upward at dawn and downward at dusk). Lower part: relative differences between the SAOZ and Dobson total ozone (circles) and between the SAOZ and Brewer total ozone (squares). Open symbols stand for the AM and black symbols for the PM SAOZ data. The better coincidence of the air masses reached by comparing only AM SAOZ data does not alter significantly the mean agreement, but reduces the scatter in the comparison: -0.9±4.6% against -0.2±7.1% for the Dobson, and -0.1±4.1% against -0.9±6.5% for the Brewer.

The differences in the geometry of observation of the instruments can also introduce some scatter in their comparison. The comparison of the ISSJ SAOZ total ozone with Dobson and Brewer data at Arosa illustrated in Figure 2 demonstrates that the better spatial coincidence between the air masses reached by comparing only AM SAOZ data (at dawn, the SAOZ measures eastward, in the direction of Arosa), cuts down the scatter of about 2.5% for the 1996 winter. To estimate the geolocation of the air masses effectively probed, the absorption light path related to the nadir and the zenith-sky observation modes has been investigated by means of a radiative transfer model. The effective geolocation of the SAOZ twilight measurement extends up to several hundred kilometres from the ground-based site (from 100 up to 350 km at 87° SZA and from 150 up to 550 km at 91° SZA) within an azimuth range varying with the season, while the GOME measurement is calculated to extend from the ground pixel up to 30 km in summer and up to 100 km in winter, depending on the SZA. Such a geometrical extension of the different measurements introduces some scatter in the comparison, due to the spatial variability of the field of the measured species. As shown in Figure 3, this scatter can be dramatically enhanced by the classical comparison methods, which consist in comparing the ground-based data with all the pixels found up to a given distance from the ground-based site.

For the GOME validation with the SAOZ network, the viewing geometry of the various instrumentations has been taken into account. The GOME pixels are selected when their absorption light path - and not their footprint on the ground - offers a sufficient intersection with the absorption light path of the correlative ground-based measurement, as illustrated in Figure 3. The GOME ozone maps themselves can give some qualitative information on the homogeneity of the ozone field in the line-of-sight of the instruments.

Diurnal Variation of the NO_2 Vertical Column

Since the NO_2 vertical column exhibits a diurnal increase between sunrise and sunset, the comparison of GOME measurements performed around noon with twilight SAOZ data is not straightforward. A first option would be to interpolate linearly the dawn and dusk SAOZ measurements

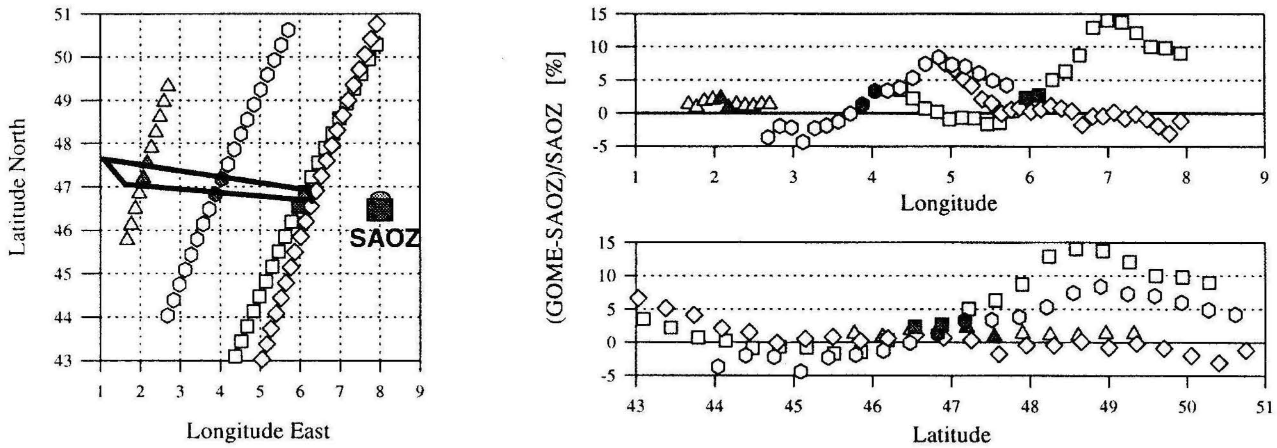

Figure 3 Illustration of the methodology of comparison between the GOME and SAOZ total ozone at the ISSJ on the 4 November 1995 (GOME version 1.20). During this day, the ozone field presented a steep gradient above the Alps. In the left-hand part, the horizontal projection of the GOME pixel centres up to 500 km from the ground-based station makes appear four tracks (triangles, hexagons, squares and diamonds) out of two different orbits. The location of the SAOZ and the horizontal projection of the air mass probed by the SAOZ are displayed in this figure as well, and the dark symbols stand for the GOME pixel centres intersecting with this projection. The right-hand part shows the relative differences (in percentage) between the GOME and SAOZ total ozone measurements, as a function of the longitude (upper part) and of the latitude (lower part). This figure clearly demonstrates that, for the 4 November 1995, the ±10% scatter introduced in the comparison by the steep ozone field gradient can be avoided if only coincident geolocations are compared.

at the local time of the ERS-2 overpass. Since the diurnal change of NO$_2$ is not linear but fast in the morning and slower in the afternoon after the complete photolysis of N$_2$O$_5$, an alternative approach would be to validate GOME with the evening SAOZ data. If needed, a small correction might be added in the future, based on a photochemical model simulation.

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