

POLE-TO-POLE VALIDATION OF THE ERS-2 GOME LEVEL 2 PRODUCTS WITH THE SAOZ GROUND-BASED NETWORK

J.-C. Lambert¹, M. Van Roozendael¹, J.-F. Müller¹, P.C. Simon¹, M. De Mazière¹, J.-P. Pommereau², F. Goutail², A. Sarkissian², L. Denis², V. Dorokhov³, P. Eriksen⁴, E. Kyro⁵, J. Leveau⁶, H.K. Roscoe⁷, W. Arlander⁸, C.W. Tellefsen⁸ and G. Vaughan⁹

1. Belgian Institute for Space Aeronomy, BIRA-IASB, Avenue Circulaire 3, B-1180 Brussels, Belgium
phone: +32-2-373.04.68; fax: +32-2-374.84.23; lambert@oma.be
2. Service d'Aéronomie du CNRS, BP 3, F-91371 Verrières-le-Buisson Cedex, France
phone: +33-1-64.47.42.88; fax: +33-1-69.20.29.99; pommereau@aerov.jussieu.fr
3. Central Aerological Institute, CAO, Dolgoprudny, Moscow, Russia
4. Danish Meteorological Institute, DMI, Copenhagen, Denmark
5. Finnish Meteorological Institute, FMI, Sodankylä, Finland
6. Faculté des Sciences, Université de la Réunion, France
7. British Antarctic Survey, BAS, Cambridge, U.K.
8. Norwegian Institute for Air Research, NILU, Kjeller, Norway
9. University College of Wales, Aberystwyth, U.K.

ABSTRACT

The comparison results between GOME level 2 products (mainly total ozone and nitrogen dioxide) from July through December 1996 and correlative ground-based observations from the SAOZ UV-visible network are presented over a wide range of latitudes from the Arctic to the Antarctic, including 1996 ozone hole conditions in Antarctica. Possible reasons for the observed discrepancies have been identified and quantified. On average the GOME version 2.0 and the SAOZ total ozone are in close agreement. However the comparison still shows a clear dependence on the solar zenith angle of the GOME measurement as well as a difference in sensitivity. When combined together, the SZA dependence and the reduced sensitivity result in a high ozone column underestimation which amplifies at large SZA. During ozone hole conditions in Antarctica, GOME and the ground-based instruments observe the high day-to-day variability similarly, although GOME overestimates the lowest ozone columns. Agreement between GOME and ground-based total NO₂ is found to depend strongly on the NO₂ profiles used as input in the AMF calculation, and particularly on the tropospheric content.

1. INTRODUCTION

The Global Ozone Monitoring Experiment (GOME) was launched on 21 April 1995 onto a polar heliosynchronous orbit, on-board the ESA ERS-2 environmental platform. The instrument is a nadir-viewing UV-visible grating spectrometer observing between 240 and 790 nm the solar radiation scattered from the atmosphere or the Earth's surface. By application of the differential optical absorption spectroscopy (DOAS), vertical columns of total ozone,

NO₂ and BrO are inferred as well as other trace species such as OCIO and SO₂ during particular events.

Since the late 1980's, ozone and NO₂ are monitored permanently from the Arctic to Antarctica by several SAOZ ground-based instruments (Pommereau and Goutail, 1988). Based on the same DOAS technique as GOME, the SAOZ (Système d'Analyse par Observation Zénithale) is a UV-visible spectrometer that detects atmospheric trace constituents by looking at the sunlight scattered at zenith by the atmosphere. Total ozone is retrieved in the 450-580 nm spectral region and nitrogen dioxide in the 400-500 nm range, twice daily during twilight periods for solar zenith angle (SZA) ranging from 86° up to 91°. After a real time spectral analysis at the station, column densities along the line of sight, or slant columns, are converted into vertical columns by using a standard air mass factor (AMF) calculated at 60°N with a radiative transfer model. These preliminary total columns are transmitted by satellite data collection to the laboratory. Several SAOZ spectrometers are currently operated at a variety of sites in the world, over an extended latitude range. With two other UV-visible DOAS spectrometers designed respectively at the Belgian Institute for Space Aeronomy (IASB, see Van Roozendael et al., 1995) and at the Norwegian Institute for Air Research (NILU), these instruments listed in Table 1 form the SAOZ/UV-visible network.

Part of the GOME Geophysical Validation Campaign started on 20 July 1995, the data of this network have been collected by IASB and by the Service d'Aéronomie du CNRS for investigating the pole-to-pole performances of the GOME. Based on 45 days of data from July to December 1995, a first comparison with the GOME v1.20 and v1.21 total ozone retrieved

Table 1. The SAOZ/UV-visible network

Location	Lat.	Long.	Institute
Ny-Ålesund	79N	12E	NILU
Longyearbyen	78N	16E	NILU
Thulé	77N	69W	DMI
Scoresbysund	70N	22W	CNRS/DMI
Sodankylä	67N	27E	CNRS/FMI
Zhigansk	67N	123E	CNRS/CAO
Harestua	60N	9E	BIRA-IASB
Aberystwyth	52N	4W	U. of Wales
Jungfraujoch	47N	8E	BIRA-IASB
O. Haute Provence	44N	6E	CNRS
Tarawa	01N	172E	CNRS/NIWA
Reunion Island	21S	55E	U. Réunion
Bauru	22S	48W	CNRS/UNESP
Kerguelen Islands	49S	70W	CNRS
Dumont d'Urville	66S	140E	CNRS
Rothera	68S	68W	BAS

in the ultraviolet Huggins bands was conducted at the NDSC/Alpine station using a variety of instruments, and was extended to all latitudes using the SAOZ network (Lambert et al., 1996^{a,b}). Both exercises concluded to i) a total ozone underestimation by the GOME; ii) a significant SZA dependence at all latitude when compared to SAOZ: 5% underestimation on average at 45° SZA, 10% at 60° SZA and even much more beyond where multiple scattering was not considered; iii) a dependence of the relative difference between the GOME and the SAOZ total ozone, on the amplitude of total ozone amount, that is a difference in sensitivity; iv) an overestimation of the ozone column by 10-20% at high latitude in summer, as well as in ozone hole conditions in Antarctica at spring. Since improvements in the preliminary GOME NO₂ retrieval were still in progress, the quantification of the discrepancies between GOME and ground-based measurements of total NO₂ was irrelevant.

Following the recommendations and conclusions drawn from this first validation exercise, the GOME retrieval of ozone in the Huggins bands has been revisited and improvements have led to the current version 2.0 of the GOME Data Processor (GDP), including the calculation of the AMF at the lowest weight wavelength (325 nm), the correction for multiple scattering up to 92° SZA, and a better treatment of the cloud cover. A first comparison between GOME v2.0 and ground-based total ozone obtained in July-August 1996 was conducted at northern latitudes with Dobson, Brewer and GUV instruments by Van Roozendaal et al. (1997^a) and from pole to pole by Lambert et al. (1997^a). These works demonstrated a general better agreement between the satellite and the ground-based measurements. Here we report on an extension of the pole-to-pole validation of the GOME v2.0 total ozone over the period from 28 June through 15 December 1996. The agreement of the GOME with correlative SAOZ observations is

analysed at low, middle and high latitudes in both hemispheres with respect to parameters such as the SZA and the column amount. A separate section presents the ground-based validation of GOME measurements during the 1996 ozone hole in Antarctica. Last section describes first comparison results of total NO₂ obtained within the same period.

2. VALIDATION METHODOLOGY

An adequate methodology to compare GOME total ozone and NO₂ with ground-based zenith-sky measurements has already been described by Lambert et al. (1996^a, 1997^b). In summary, this methodology takes into account the spatial and temporal variability of the ozone and NO₂ fields as well as the uncertainty on the SAOZ measurement. To account for large horizontal gradients in the vertical columns, GOME pixels are selected where intersecting the effective geolocation of the twilight zenith-sky measurement. This effective geolocation, calculated by a radiative transfer model, is located in the direction of the Sun between 100 and 350 km from the instrument at 87° SZA and between 150 and 550 km at 91° SZA.

The SAOZ spectral analysis program is based on a least squares iterative procedure to retrieve in the visible range slant columns of O₃, NO₂, (O₂)₂, O₂ and H₂O. This least squares procedure uses high resolution absorption cross-sections published in the literature and convolved with the SAOZ slit function. When the uncertainty on the high resolution ozone absorption cross-sections and the one sigma confidence level of the least squares fit calculated for each spectrum (0.5% for ozone and 1.5% for NO₂) are taken into account, the overall accuracy of the SAOZ ozone slant total amounts is better than 2%. For NO₂, the uncertainty of the absorption cross-sections (Merienne et al., 1994) is of the order of 5%, but a rather large temperature dependence was shown by Harwood and Jones (1994) and Coquart et al. (1995), and is not taken into account in the present analysis. If corrected for this temperature dependence, the NO₂ stratospheric columns would have to be reduced by about 15%. For ozone, a systematic seasonal cycle exists in the SAOZ AMF of about 5-6% amplitude at 67°N, 3-4% at 44°N and negligible in the tropics, originating in the change in the shape of the ozone profile (Hoiskar et al., 1995; Van Roozendaal et al., 1997^b; Sarkissian et al., 1997; Denis et al., 1995). The use of the standard SAOZ AMF also introduces an average latitudinal dependence of -3% at 67°N to +2.8% at the tropics (Denis et al., 1995). Since this seasonal/latitudinal variation is not included in the real time SAOZ data, it must be kept in mind in the interpretation of comparison results with GOME.

The SAOZ instruments participate regularly to field intercomparison campaigns with other DOAS UV-visible spectrometers, SAOZ, Dobson, Brewer and ozone soundings. At Camborne (UK) in September 1994, the results from four SAOZ, the UV-visible spectrometer of BIRA-IASB and other DOAS

instruments were consistent within 3% (10 DU) for ozone and 5% for NO₂ and consistent also with Dobson measurements and ozone soundings within 3% (Vaughan et al., 1997).

3. TOTAL OZONE

In this section, GOME v2.0 total ozone obtained from June 28 through December 15, 1996, is analysed with respect to pole-to-pole SAOZ observations. Nevertheless, results from the Antarctic Polar Circle are incorporated in the present section only when they present a reasonable consistency with other latitudes despite ozone hole conditions. Validation results during the 1996 ozone hole in Antarctica will be presented in the following section.

3.1 Overall consistency and SZA dependence

The relative differences between the GOME and SAOZ total ozone obtained in July-November 1996 at nine stations are depicted in Figure 3-a as a function of the SZA of the GOME measurement. At first glance, results are remarkably consistent among the various latitude belts. The average agreement between GOME and SAOZ depends clearly on the SZA. Below 70°, the mean difference between the two data sets does not exceed $\pm 4\%$, that is within the known uncertainty of the SAOZ real time measurement obtained with the standard 60°N AMF. At the Jungfrauoch, after taking properly into account the 3% seasonal cycle of the SAOZ AMF and the altitude of the station (3580 m a.s.l.), GOME and SAOZ total ozone are on average consistent within 2%. Dobson and Brewer observations at the NDSC/Alpine sites also show a good agreement and confirm these results. Beyond 70° SZA, GOME underestimates SAOZ total ozone by 8% on average. The seasonal/latitudinal dependence of the standard SAOZ AMF can contribute to the SZA dependence, but can certainly not account for this 8% underestimation. Moreover, this underestimation generates in summer polar regions a systematic bias between GOME data obtained in the descending and

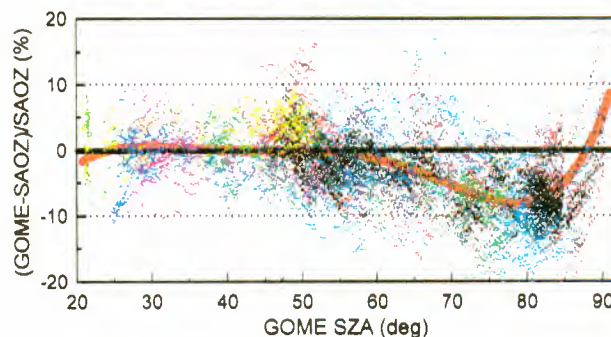


Figure 3-a. Percentage relative difference between GOME and SAOZ total ozone versus GOME SZA, along with a 5th order polynomial fit. Legend is provided with Figure 3-b.

ascending part of the orbit, that is between measurements at noon and in midnight Sun. This bias is observed in the Arctic as well as in Antarctica. The deviation from SAOZ becomes rapidly dispersed after 85° SZA and statistics become irrelevant, despite the fact that the spatial and temporal coincidence of the air masses probed by the satellite and the ground-based instruments is optimal.

3.2 Sensitivity

For eight stations distributed from the Arctic up to 50°S, the correlation between the GOME and SAOZ total ozone is shown in Figure 3-b at SZA below and beyond 70°. For ozone total amounts from 250 through 400 DU, the slopes of the linear regressions (between 0.66 and 0.85) indicates some systematic lower sensitivity of GOME compared to SAOZ. Large columns are systematically underestimated. This difference in sensitivity amplifies with the SZA: below 70° SZA, $GOME = 55.2 + 0.81 \times SAOZ$ ($r^2 = 0.79$); beyond 70° SZA, $GOME = 66.1 + 0.70 \times SAOZ$ ($r^2 = 0.79$). Regressions of higher order show that this difference in sensitivity also increases for large ozone total amounts. This sensitivity difference amplifying at large SZA could originate: i) in the wavelength dependence of the GOME AMF; ii) in any small wavelength calibration

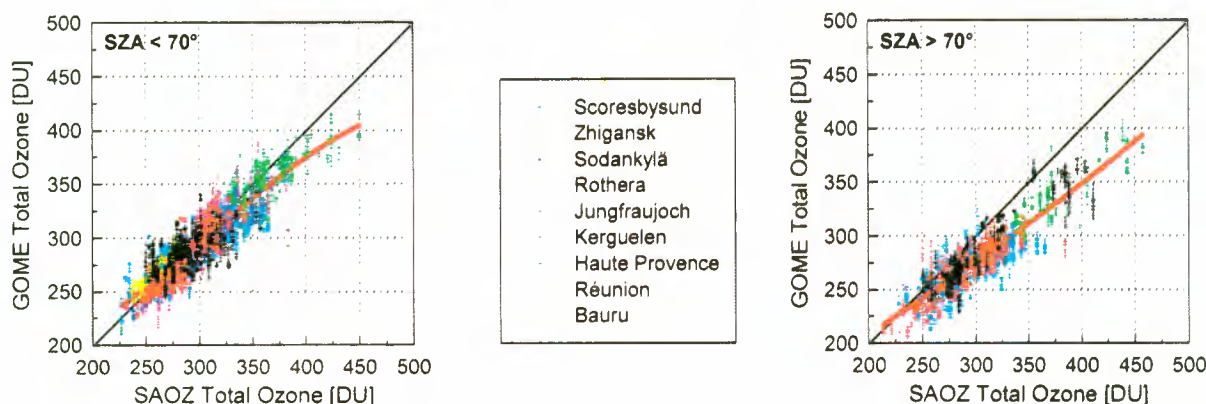


Figure 3-b. Correlation between the GOME and SAOZ total ozone for eight stations distributed from the Arctic to southern mid-latitude (the same stations as in Figure 3-a excepted Rothera), along with a 3rd order polynomial fit. Left-hand panel shows results below 70° SZA and right-hand panel beyond 70°. The difference in sensitivity combines with the SZA dependence to result in a high ozone column underestimation that amplifies at large SZA.

shift between the GOME spectra and the laboratory cross-sections; and iii) in imperfect ozone cross-section convolution in the GDP. The same comparison exercise was carried out for the same time period between total ozone observations from the SAOZ network and from the Total Ozone Mapping Spectrometer (TOMS) launched on-board the NASA's Earth Probe platform (EP) on July 2, 1996. This exercise concluded to a difference in sensitivity too, but with no clear dependence on the total column. Since TOMS uses only a set of discrete wavelengths, this difference between GOME and TOMS-EP backs up the aforementioned possible sources of SZA dependent difference in sensitivity, but it could also arise from basic differences between the algorithms, GDP v2.0 assuming a sole given state of the atmosphere while TOMS v7.0 uses, for latitudes between 15° and 75°, a combination of several ozone density and temperature profiles and hence partly accounts for scattering geometry variation.

3.3 Dispersion

The scatter increases on average from $\pm 4\%$ at the tropics and mid-latitudes, up to $\pm 10\%$ at high latitude and for SZA larger than 65°. Possible sources for this dispersion are: i) the total ozone variation between the twilight SAOZ and local noon GOME observations; ii) the difference in location between noon nadir and twilight zenith viewings combined to the presence of horizontal gradients; iii) the use of a constant AMF in the SAOZ retrieval instead of the real AMF corresponding to the actual ozone profile, which can account for 1% scatter; iv) a similar contribution from the constant climatic GOME AMFs; v) the dispersion generated in the SAOZ measurements by tropospheric multiple scattering in presence of dense clouds or haze, combined with local ozone changes (Van Roozendaal et al., 1994); and vi) the contribution of clouds to the GOME measurement which mask the tropospheric contribution. The dispersion varies from one station to another depending on its location with respect to sources of tropospheric ozone and cloudiness. The smallest dispersion is observed in absence of clouds

during the dry season at Reunion Island, in the tropics, where total stratospheric ozone is the most stable. It is already significantly larger at Bauru in Brazil during the season of biomass burning where scattered high altitude clouds are also frequent. Clouds may also contribute to significant systematic deviations like at Kerguelen, which is almost permanently overcast in the winter season.

4. ANTARCTICA: 1996 OZONE HOLE

The total ozone comparison at Dumont d'Urville points out a constant 10% overestimation of SAOZ by GOME during July and August 1996. SAOZ data at Rothera were too scarce during this period, but Dobson observations at Vernadsky/Faraday (65°S, 64°W) provided by the British Antarctic Survey (BAS) confirm this bias that vanished after August to lead to a good average agreement. The comparison at two extreme locations on the Antarctic polar circle show that GOME and SAOZ reproduce similarly the high day-to-day variability observed during ozone hole conditions near the edge of the polar vortex, as illustrated in Figure 4-a. The large dispersion of the comparison results is significantly cut down after removal of aberrant points originating in: i) a strong diurnal variation of the ozone vertical column; ii) strong gradients identified by means of GOME ozone maps; and iii) a dense cloud cover confirmed by local meteorological monitoring and synoptic observations.

From one day to another, ozone total columns can move from values as low as 130 DU to values of about 400 DU. It appears that GOME overestimates systematically the lowest ozone amounts and generally underestimates the largest columns, as shown in Figure 4-b for July-October at Rothera and in Figure 4-a. Again, these findings are confirmed by measurements from the Dobson spectrophotometers operated by BAS at Vernadsky/Faraday and at Halley (76°S, 27°W). In particular, from August through October, GOME total ozone overestimated Dobson data at Halley by 11% on average. Taking into account the observed difference in sensitivity, this offset can be explained easily by

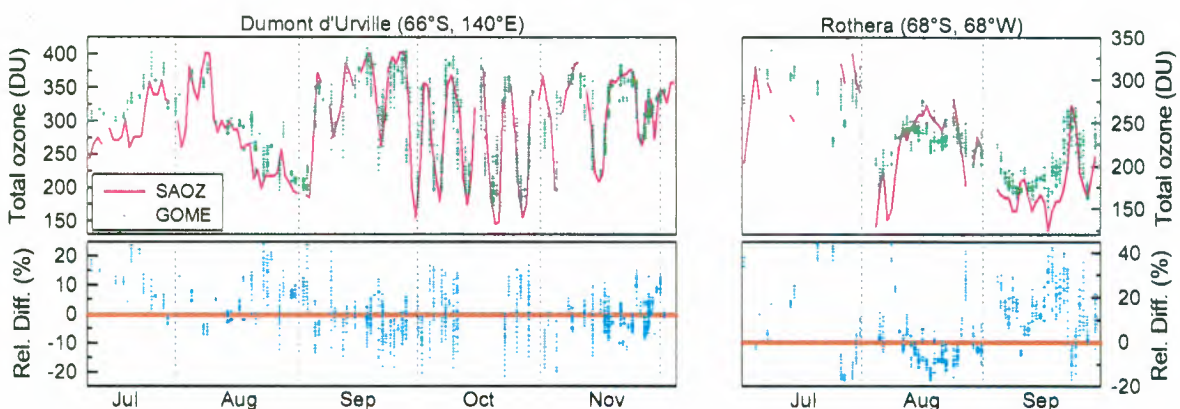


Figure 4-a. GOME and SAOZ total ozone and percentage relative difference ($(\text{GOME}-\text{SAOZ})/\text{SAOZ}$) at Dumont d'Urville and at Rothera during the 1996 ozone hole.

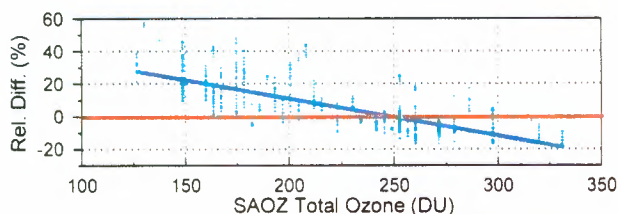


Figure 4-b. Relative difference between GOME and SAOZ at Rothera (July-October) as a function of the ozone total amount, along with a first order regression.

permanent very low total ozone values ranging from 100 DU up to a maximum of 210 DU, due to the very high latitude of the station and hence its location inside the vortex during the whole time period. The strong difference in sensitivity and the July-August offset mask any SZA dependence, but the comparison between ascending and descending orbit data leads to the same 8% bias as observed in the Arctic under similar conditions. The same comparison exercise was carried out for the same time period between TOMS-EP and ground-based total ozone and concluded to a similar overestimation of the lowest ozone columns by TOMS-EP, but also to a different seasonal behaviour of the agreement with ground-based observations, in particular a better agreement before September 1996 but a worst after. In addition to those mentioned in the previous section, possible contributions to the difference in sensitivity could be the use of a climatic atmospheric profile in the GOME retrieval which cannot match the true depleted profile during low ozone events and also a possible artefact in the GOME as well as the SAOZ data, due to dense (type II) PSCs which form at extremely low temperature in the vortex.

5. NITROGEN DIOXIDE

Since total NO_2 exhibits a diurnal increase between sunrise and sunset, the accurate comparison of GOME measurements performed around noon with dawn and dusk SAOZ data is not straightforward. However, 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_2O_5 . Therefore, in a first step, the comparison could be carried out with evening SAOZ data rather than with dawn and dusk SAOZ measurements linearly interpolated at the local time of the ERS-2 overpass. If needed, a small correction might be added in the future, based on a photochemical model simulation. In addition to the vertical column amounts, seasonal variation, day-to-day fluctuations and scatter between adjacent ground pixels must be surveyed.

5.1 Comparison of NO_2 vertical columns

For this work, GOME total NO_2 was available from June 28 through December 15, 1996, except between July 29 and October 15 when a strong wavelength calibration shift in GOME channel 3 made the retrieval of NO_2 irrelevant. The agreement between GOME and SAOZ observations depends clearly on the latitude and

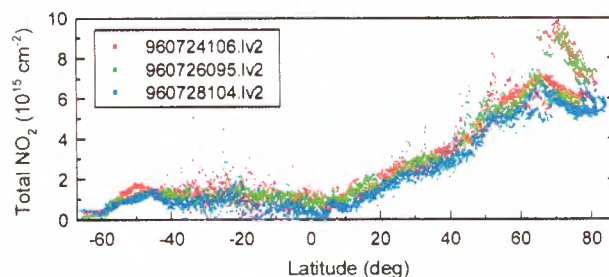


Figure 5-a. GOME NO_2 vertical column amounts measured during three orbits overpassing Scandinavia and western Europe on July 24, 26, and 28, 1996. The names of the three orbit data files are given in the legend. In the ascending part of the orbits, that is in the Arctic under midnight Sun conditions, GOME total NO_2 increases unreasonably and overestimates SAOZ data by $2\text{-}4 \times 10^{15}$ molecule. cm^{-2} .

on the season. Tropic of Capricorn is the sole region where the GOME and SAOZ vertical columns are comparable before and after the calibration shift period. Outside the Tropics, the quantification of the discrepancies between both sets of measurements is often irrelevant. After the calibration shift, GOME total NO_2 reaches values twice as large as SAOZ values in the northern hemisphere, and twice as small in the southern hemisphere. A systematic bias of about 2 up to 4×10^{15} molec. cm^{-2} occurs between noon and midnight Sun measurements in the Arctic and in Antarctica. Figure 5-a depicts NO_2 vertical column amounts measured by GOME during three orbits overpassing Scandinavia and western Europe on July 24, 26, and 28. This figure illustrates the unreasonable increase of GOME total NO_2 in the ascending part of the orbit. Moreover, in the northern hemisphere, GOME total NO_2 exhibits an aberrant seasonal variation. Nevertheless, for all latitudes, slant columns measured by GOME and by SAOZ present at similar SZA a comparable level. In general, day-to-day fluctuations of total NO_2 averaged within 350 km from the ground-based station appear also reasonable for all latitudes.

The scatter over ten adjacent ground pixels depends on the latitude and the season. It is significantly reduced when considering only satellite data under nearly clear sky conditions (cloud cover fraction lower than 40 %). In this case, the scatter within 700 km along track ($\pm 3^\circ$ latitude around the ground-based site) is still always larger than the diurnal variation observed between correlative AM and PM SAOZ data and it ranges from 0.5 up to 3×10^{15} molec. cm^{-2} . This scatter is likely to originate in the high sensitivity of GOME to tropospheric NO_2 , a quantity which is known to exhibit a strenuous temporal and spatial variability. Lowest values of scatter are observed in winter polar regions as well as at remote sites located several hundred kilometres away from any tropospheric pollution source such as Greenland, Siberia, Tarawa (middle of Pacific Ocean), Kerguelen (middle of Indian Ocean) and Antarctica stations. Highest values are observed over European polar region in summer, and outside the Arctic in all seasons at locations where GOME measurements are corrupted by notable tropospheric

pollution such as mid-latitude European sites, Brazil, and also Reunion Island when wind carries air masses influenced by African biomass burning.

5.2 AMF and NO₂ density climatologies

The standard SAOZ AMF uses as input NO₂ density profile from the 1976 US Standard Atmosphere while GOME AMFs are based on the results of a two-dimensional chemical transport model of the atmosphere developed at the Max Planck Institute (Crutzen and Gidel, 1983; Bruehl, 1991), hereafter referred to as MPI-2D profiles. As shown in parts (a) and (b) of Figure 5-b, a fundamental difference between these profiles rests in their tropospheric content: in the first 5 kilometres, less than 8×10^8 molecules.cm⁻³ in Standard Atmosphere against 10^9 up to 10^{10} for MPI-2D. It is worth noting that stratospheric densities from both types of profiles are one order of magnitude as small as lower troposphere densities given by MPI-2D. Nadir AMFs were computed by means of the IASB's single scattering radiative transfer model (Sarkissian et al., 1995) with the MPI-2D and the Standard Atmosphere profiles as input. The comparison, illustrated at 45° and 65° north in parts (c) and (d) of Figure 5-b, points out the high sensitivity of the AMF on the input NO₂ vertical distribution. When compared to the Standard Atmosphere AMF, MPI-based AMFs can be twice or more as small, that is leading to vertical columns twice or more as large as the Standard Atmosphere vertical columns. The impact of this essential difference is determining, not only for the interpretation of

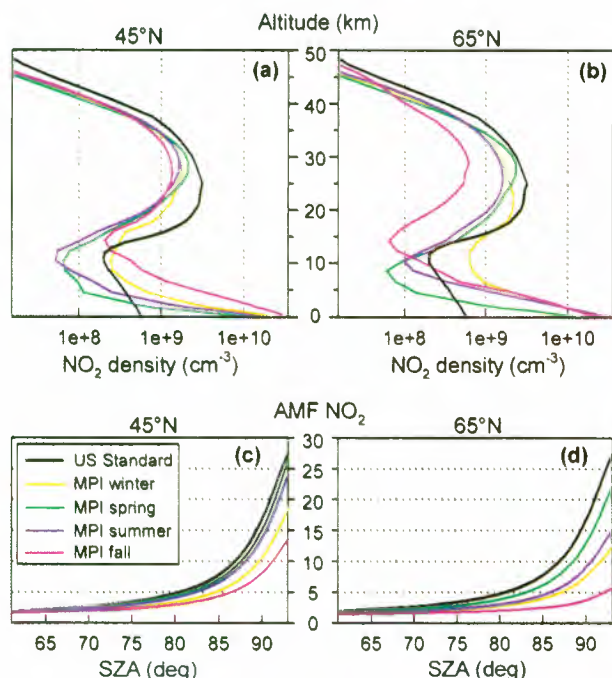


Figure 5-b. (a) and (b): NO₂ density vertical distributions at 45°N and 65°N from the 1976 US Standard Atmosphere (black curve) and from the MPI-2D model; (c) and (d): NO₂ AMFs based on these different vertical distributions and calculated with the IASB's single scattering model.

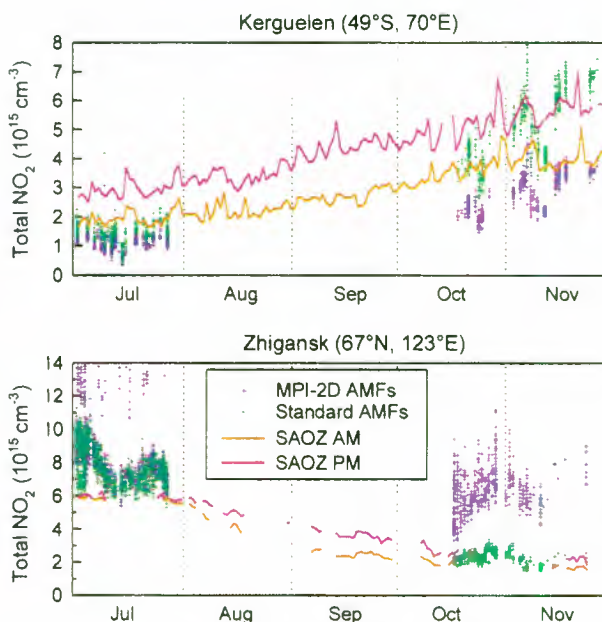


Figure 5-c. NO₂ vertical column amounts measured from July through November 1996 at Kerguelen (Indian Ocean) and at Zhigansk (Eastern Siberia) by GOME and by SAOZ. GOME vertical columns under clear sky conditions (cloud cover fraction < 40 %) are inferred from observed slant columns by means of AMFs calculated i) with MPI-2D profiles (in red), and ii) with 1976 US Standard Atmosphere profiles (in green).

comparison results between GOME and ground-based NO₂ vertical columns but also for the interpretation of the GOME vertical columns themselves. At small and moderate SZA, slant column amounts observed by a nadir viewing instrument are very sensitive to the tropospheric content. In comparison, the main contribution to slant column amounts observed by a SAOZ at zenith during twilight comes from the stratosphere. Moreover, the relative contribution of a given atmospheric layer is constrained in the AMF calculation by the input profile. Therefore, the enhanced tropospheric content in MPI-2D increases significantly the weight of lower troposphere.

A new data set of GOME NO₂ vertical columns was built up from GOME slant columns obtained under nearly clear sky conditions (again a cloud cover fraction lower than 40 %), by dividing them by the calculated Standard Atmosphere AMFs. The purpose of this exercise was to get rid of the influence of input NO₂ profiles in the comparison and to reduce the weight of tropospheric content. The comparison between GOME and SAOZ observations over the whole SAOZ/UV-visible network demonstrates that the agreement is significantly ameliorated if Standard Atmosphere AMFs are used. In the Arctic, the scatter of GOME data is cut down by a factor two. In the fall northern hemisphere, the aberrant seasonal variation disappears. At the Equator and at the Tropic of Capricorn, the 1-2 10^{15} molec.cm⁻³ underestimation of both dawn and dusk SAOZ data is attenuated. In

Antarctica, GOME total NO_2 in November and December values reaches regular values and follow a normal seasonal variation. Figure 5-c illustrates the comparison at Kerguelen Islands in the Indian Ocean and at the Siberian station of Zhigansk, several hundred kilometres away from any source of tropospheric pollution. For both stations, the improvement is clear.

5.3 Preliminary comparison with 3D model IMAGES

A three-dimensional chemical transport model of the troposphere, named Intermediate Model of Global Evolution of Species (IMAGES, Müller and Brasseur, 1995), has been developed jointly at IASB and at the National Center for Atmospheric Research (NCAR) to study the global distributions, budgets and trends of 41 chemical compounds, including ozone and nitrogen oxides. IMAGES distributions of nitrogen oxides and of other species are found to be generally in good agreement with correlative airborne in situ measurements. IMAGES has been run to provide monthly vertical distributions of NO_2 onto a $5^\circ \times 5^\circ$ grid. Modelled global distribution of NO_2 , depicted in Figure 5-d at surface level in April, demonstrates that, for an instrument such as the GOME, strong zonal gradients in the NO_2 field - that can vary from 100 pptv up to 50 ppbv over less than 500 km - make any 2D model inadequate to reach a reasonable level of accuracy. Modelled meridional gradients also suggest that a 10° longitude resolution (approximately 1110 km at sea level) would be too coarse. Moreover, the enhanced variability of tropospheric NO_2 in fall and winter makes a set of input profiles based on monthly means preferable to a seasonal set. At 45° and 65° north, the comparison of MPI-2D with IMAGES profiles confirms the larger NO_2 density values of MPI-2D in the low and middle troposphere, particularly in fall. This overestimation in the most dense layers might be the main source of the aberrant seasonal variation observed in GOME total NO_2 after the calibration shift period. In the upper troposphere, IMAGES is systematically lower than MPI-2D at 45°N .

A preliminary comparison (not shown here) was also carried out in the upper troposphere and in the stratosphere with 25 NO_2 density vertical distributions measured with the SAOZ-balloon experiment (Pommereau and Piquard, 1994) at mid and high northern latitude in various seasons. At mid-latitude, MPI-2D and SAOZ-balloon number densities present a reasonable agreement in the middle stratosphere, but MPI-2D underestimates balloon measurements by a factor two in the lower stratosphere and upper troposphere. A similar systematic underestimation in this altitude range is observed in the Arctic under midnight Sun conditions, while modelled and measured profiles are at first glance mutually consistent in winter. In the upper troposphere, the comparison between IMAGES and SAOZ-balloon profiles at 45°N and 65°N shows a general reasonable agreement whatever the season.

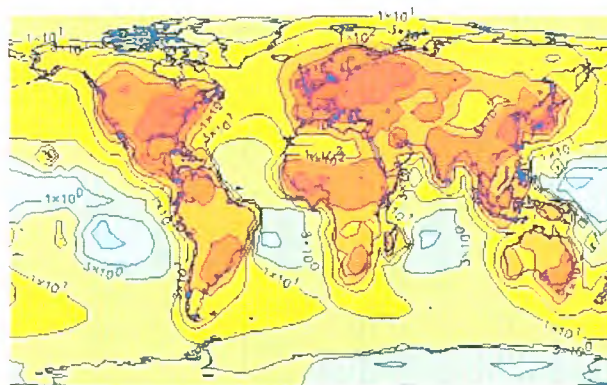


Figure 5-d. World-wide distribution of surface NO_2 mixing ratio in April (logarithmic contour steps, expressed in pptv), calculated by the 3D model IMAGES (IASB-NCAR). Although attenuated at tropopause, the same morphology is observed at least in the 5 first kilometres.

5.4 Conclusions and recommendations

A first qualitative analysis of the July-December 1996 GOME total NO_2 by means of correlative SAOZ observations shows that GOME vertical columns exhibit in many case an abnormal behaviour but that slant columns look in general reasonable. The main problem rests obviously in the AMF used to convert slant into vertical columns, and particularly in the input NO_2 vertical distributions used for AMF calculation. It is vigorously recommended to revisit these input profiles. In the troposphere, test cases with the 3D tropospheric model IMAGES demonstrate that a 2D model is inadequate to reach the required level of accuracy, due to strong gradients in the NO_2 field. Moreover, a set of input profiles based on monthly means is preferable to a seasonal set, especially in fall and winter. Tropospheric NO_2 from model should be validated by surface, balloon and airborne measurements. In the stratosphere, balloon-borne experiments such as SAOZ-balloon and FTIR could combine advantageously with global information from satellite experiments SAGE-2 or UARS (HALOE, CLAES, LIMS).

6. REFERENCES

- Bruehl, C., and P. Crutzen, The MPI Two-dimensional Atmospheric Model - Trace Gas Profiles, MPI Mainz, Private communications, 1991
- Coquart, B., A. Jenouvrier and M. F. Merienne, The NO_2 Absorption Spectrum II. Absorption Cross Sections at Low Temperature in the 400-500 nm Region, *J. Atm. Chem.*, 21, 251, 1995.
- Crutzen, P.J., and L.T. Gidel, A two-dimensional model of the atmosphere, 2: The tropospheric budgets of the anthropogenic chlorocarbons CO , CH_4 , CH_3Cl , and the effects of various NO_x sources on tropospheric ozone, *J. Geophys. Res.*, 88, 6641-6661, 1983
- Denis, L., J. P. Pommereau, F. Goutail, T. Portafaix, A. Sarkissian, M. Bessafi, S. Baldy, J. Leveau, P.V. Johnston and A. Matthews, SAOZ Total O_3 and NO_2 at

- the Southern Tropics and Equator, *Proc. 3rd Europ. Symp. on Polar Stratospheric Ozone*, EC Ed., 458-462, 1995.
- Harwood, M. H., and R. L. Jones, Temperature Dependent Ultraviolet Cross-sections of NO₂ and N₂O₄: Low Temperature Measurements of the Equilibrium Constant 2NO₂ <--> N₂O₄, *J. Geophys. Res.*, 9, 922, 1994.
- Hoiskar, B.A.K., A. Dahlbak, G. Vaughan, G.O. Braathen, F. Goutail, J.P. Pommereau and R. Kivi, Seasonal Variations in Air Mass Factors for Ozone Computations Based on Climatology Data, *Proc. 3rd Europ. Symp. on Polar Stratosph. Ozone*, EC Ed., 557-562, 1995.
- Lambert, J.-C., M. Van Roozendael, P. Peeters, P.C. Simon, M.-F. Merienne, A. Barbe, H. Claude, J. de La Noë and J. Staehelin, GOME Ozone Total Amounts Validation by Ground-based Observations Performed at the NDSC/Alpine Stations, *Proceedings of the GOME Geophysical Validation Final Results Workshop, 24-26 January 1996, ESRIN, Frascati, Italy*, ESA WPP-108, pp.115-121, May 1996^a.
- Lambert, J.-C., M. Van Roozendael, P.C. Simon, M. De Mazière, F. Goutail, A. Sarkissian, J.-P. Pommereau, L. Denis, V. Dorhokov, P. Eriksen, E. Kyro, J. Leveau, H.K. Roscoe, G. Vaughan and C. Wahlstrom, Validation of the ERS-2 GOME Products with the SAOZ Network, *Proceedings of the GOME Geophysical Validation Final Results Workshop, 24-26 January 1996, ESRIN, Frascati, Italy*, ESA WPP-108, pp.123-131, May 1996^b.
- Lambert, J.-C., M. Van Roozendael, M. De Mazière, J. Granville, P.C. Simon, F. Goutail, A. Sarkissian, J.-P. Pommereau, L. Denis, V. Dorhokov, P. Eriksen, E. Kyro, J. Leveau, H.K. Roscoe, G. Vaughan and C. Wahlstrom, Validation of the ERS-2 GOME Total Ozone Measurements with the SAOZ Ground-based Network during the Period: 28 June-17 August 1996, *Proc. of Quad. Ozone Symposium at L'Aquila, Italy, 12-21 September 1996*, in press, 1997^a.
- Lambert, J.-C., M. Van Roozendael, J. Granville, P. Peeters, P.C. Simon, H. Claude and J. Staehelin, Comparison of the GOME ozone and NO₂ total amounts at mid-latitude with ground-based zenith-sky measurements, *Proc. of Quad. Ozone Symposium at L'Aquila, Italy, 12-21 September 1996*, in press, 1997^b.
- Merienne, M.F., A. Jenouvrier and B. Coquart, The NO₂ Absorption Spectrum I: Absorption Cross-sections at Ambient Temperature in the 300-500 nm Region, *J. Atm. Chem.*, 20, 281, 1994.
- Müller, J.-F., and G.P. Brasseur, IMAGES: A three-dimensional chemical transport model of the global troposphere, *J. Geophys. Res.*, 100, 16,445-16,490, 1995.
- Sarkissian, A., G. Vaughan, H.K. Roscoe, L.M. Bartlett, F.M. O'Connor, D.G. Drew, P.A. Hughes and D.M. Moore, Accuracy of Measurements of Total Ozone by a SAOZ Ground-based Zenith Sky Spectrometer, *J. Geophys. Res.*, 102, 1997.
- Pommereau, J.-P., and F. Goutail, Ground-based Measurements by Visible Spectrometry during Arctic Winter and Spring 1988, *Geophys. Res. Lett.*, 891, 1988.
- Pommereau, J.-P., and J. Piquard, Ozone and nitrogen dioxide vertical distributions by UV-visible solar occultation from balloons, *Geophys. Res. Lett.*, 1227, 1994.
- Sarkissian, A., H.K. Roscoe, D.J. Fish, M. Van Roozendael, M. Gil, H.B. Chen, P. Wang, J.-P. Pommereau and J. Lenoble, Ozone and NO₂ air-mass factors for zenith-sky spectrometers: Intercomparison of calculations with different radiative transfer models, *Geophys. Res. Lett.*, 22, 1113-1116, 1995.
- Van Roozendael, M., M. De Mazière and P.C. Simon, Ground-based Visible Measurements at the Jungfraujoch Station since 1990, *J. Quant. Spectrosc. Radiat. Transfer*, 52, 231-240, 1994.
- Van Roozendael, M., C. Hermans, Y. Kabbadj, J.-C. Lambert, A.-C. Vandaele, P.C. Simon, M. Carleer, J.-M. Guilmot, R. Colin, Ground-Based Measurements of Stratospheric OClO, NO₂ and O₃ at Harestua, Norway (60°N, 10°E) during SESAME, *Proceedings of the 12th ESA Symposium on European Rocket and Balloon Programmes & Related Research, Lillehammer, Norway, 29 May - 1 June 1995*, ESA SP-370, 305-310, 1995.
- Van Roozendael, M., J.-C. Lambert, P.C. Simon, G. Hansen, A. Dahlback, D. De Muer, E. Schoubs, R. Koopman, H. Vanderwoerd, A. PETERS, A. Barbe, H. Claude, J. de La Noë, M.-F. Merienne and J. Staehelin, Ground-based validation of GOME total ozone measurements by means of Dobson, Brewer and GUV instruments, *Proc. of Quad. Ozone Symposium at L'Aquila, Italy, 12-21 September 1996*, in press, 1997^a.
- Van Roozendael, M., P. Peeters, H.K. Roscoe, H. De Backer, A. Jones, G. Vaughan, F. Goutail, J.-P. Pommereau, E. Kyro, C. Wahlstrom, G. Braathen, and P.C. Simon, Validation of Ground-based Visible Measurements of Total Ozone by Comparison with Dobson and Brewer Spectrophotometers, submitted to *J. Atm. Chem.*, 1997^b.
- Vaughan, G., H.K. Roscoe, L.M. Bartlett, F. O'Connor, A. Sarkissian, M. Van Roozendael, J.-C. Lambert, P.C. Simon, K. Karlsen, B.A. Kaestad Hoiskar, D.J. Fish, R.L. Jones, R. Freshwater, J.-P. Pommereau, F. Goutail, S.B. Andersen, D.G. Drew, P.A. Hughes, D. Moore, J. Mellqvist, E. Hegels, T. Klupfel, F. Erle, K. Pfeilsticker, U. Platt, An intercomparison of ground-based UV-visible sensors of ozone and NO₂, *J. Geophys. Res.*, 102, 1411-1422, 1997.

7. ACKNOWLEDGEMENTS

The authors would like to thank the staff of the SAOZ stations and particularly P. Gerard, J. Granville and J. Hottier for their computational and logistical support. They also thank J. Shanklin for providing BAS Dobson data from Antarctica. This work was supported by the PRODEX A.O. ERS-2 Project 1 and the OSTC contract GC/35/002 in Belgium, the Programme National de Chimie de l'Atmosphère in France, and the Environmental Programme of the European Commission in the frame of SCUVS-II, contract EV5V-CT93 0334.