A PSEUDO-GLOBAL CORRELATIVE STUDY OF ERS-2 GOME NO_2 DATA WITH GROUND-, BALLOON-, AND SPACE-BASED OBSERVATIONS

J.-C. Lambert, J. Granville, M. Van Roozendael, and J.-F. Müller

Belgian Institute for Space Aeronomy (IASB-BIRA), Avenue Circulaire 3, B-1180 Brussels, Belgium
phone: +32-2-373 04 68; fax: +32-2-374 84 23; lambert@bira-iasb.oma.be

J.-P. Pommereau, F. Goutail, and A. Sarkissian

Service d'Aéronomie du CNRS (CNRS/SA), BP3, 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

ABSTRACT

After nearly four years of successful operation aboard ERS-2, the current total NO_2 data record inferred routinely from GOME measurements allows a year-round evaluation of both versions 2.0 and 2.3 of the operational GOME Data Processor (GDP). Their respective performance is investigated by means of high-quality correlative observations performed by a pseudo-global ground-based network of UV-visible spectrometers, under a variety of relevant geophysical conditions. The study concludes that the total NO_2 data product derived with GDP 2.0 is unreliable, mainly due to the use of a partially inadequate NO_2 database in the evaluation of optical enhancement factors. Improved with a more acceptable database, GDP 2.3 provides a more consistent data product. However, latitudinal and seasonal changes in stratospheric NO_2 as well as tropospheric emissions of NO_x distort the shape of the actual NO_2 vertical distribution and hence affect the optical enhancement factor. To assess the resulting impact on the retrieved vertical column amount, a suitable NO_2 reference atmosphere is built up, combining long-term space-, balloon-, and ground-based measurements and modelling results. The investigation highlights the need to improve the GDP with a NO_2 database including relevant stratospheric features and a consistent three-dimensional tropospheric background.

1. INTRODUCTION

Operating since July 1995 aboard the ESA's Earth observation heliosynchronous polar satellite ERS-2, the Global Ozone Monitoring Experiment (GOME, ESA 1995) observes at nadir, between 240 nm and 790 nm, with a resolution of 0.2 to 0.4 nm, the solar radiation backscattered by the atmosphere. The atmospheric abundance of several trace species, including nitrogen dioxide (NO_2), is derived from GOME spectra using the Differential Optical Absorption Spectroscopy (DOAS). Line-of-sight column amounts, or apparent slant columns, are retrieved with an iterative least-squares procedure, fitting the observed differential optical thickness with differential absorption cross-sections derived from laboratory measurements. Slant columns are converted into vertical columns using an optical enhancement factor, or air mass factor (AMF). The AMF is evaluated with a radiative transfer model assuming vertical distributions of the target absorber and of the atmospheric constituents controlling the path of the solar radiation through the atmosphere. For several decades, the DOAS has also been widely used for the interpretation of ground-based observations of the UV-visible sunlight scattered at zenith by the atmosphere. Deployed from the Arctic to the Antarctic within the framework of the Network for the Detection of Stratospheric Change (NDSC), about 30 zenith-sky UV-visible spectrometers monitor total NO_2 twice daily at twilight (Lambert *et al.*, 1999, and references therein).

The performance of the GOME total NO_2 data product has been investigated at every step of its maturation by means of ground-based observations associated with the NDSC. After evaluation of the successive developmental versions 1.x of the GOME Data Processor (GDP), a preliminary ground-based analysis of the first operational version GDP 2.0 was carried out with a limited set of GOME data from July through November 1996 (Lambert *et al.*, 1997). The study highlighted the high sensitivity of nadir-viewing AMFs to the shape of the NO_2 vertical distribution. The NO_2 profile database used in GDP 2.0 was also shown to be partially inadequate for the evaluation of GOME AMFs. According to recommendations drawn from this preliminary study, a more acceptable NO_2 database was implemented in GDP 2.3, operational since January 1998. A first verification exercise concluded to the better general consistency of GDP 2.3 total NO_2 data (Lambert and Simon, 1998). However, it also stressed the need to investigate the impact of seasonal and latitudinal changes in stratospheric NO_2 and of tropospheric emissions of NO_x, on the GOME AMF due to alteration of the NO_2 profile shape.

Described in section 2, the data record available at the time of the present study allows the pseudo-global, year-round evaluation of both versions 2.0 and 2.3 of the GOME total NO_2 product reported in section 3. Seasonal and latitudinal biases of stratospheric origin in both the GDP 2.3 and ground-based vertical columns are estimated in section 5 by means of a composite climatology built up from ground-, balloon-, and space-based observations and modelling results, described in section 4. The impact of tropospheric NO_x emissions on the GOME AMFs is assessed in section 6. The paper concludes with recommendations to improve the quantitative derivation of the NO_2 vertical column amount from GOME data.

2. DATA SETS

2.1 GOME level-2 data record

The GOME total NO_2 data record available at the time of the present study was obtained with two versions of the operational level-1b-to-2 retrieval algorithm: GDP 2.0 for 1996-1997, and GDP 2.3 since January 1998 and also for 1995.

Between the two versions, most relevant improvements to total NO_2 processing are connected with the evaluation of AMFs: (a) substitution of the NO_2 profile database; (b) use of multiple scattering look-up table of AMFs computed with an upgraded radiative transfer model; (c) new weighting and time/latitude interpolation schemes; and (d) correction of a known error in the detection of clouds. Two time periods identified by dashed boxes in the time-series of Figures 1 and 2 are affected by irrelevant retrieval due to wavelength registration problems in the spectral channel 3 of the instrument. The atmospheric profile database used in GDP 2.0 is based on results of a two-dimensional chemical transport model of the atmosphere developed at the Max Planck Institute (hereafter MPI-2D, Crutzen and Gidel, 1983). MPI-2D profiles represent seasonal mean estimates in 10° latitude belts. GDP 2.3 AMFs are based on the daytime estimate for United States (US Standard) included in the AFGL Reference Atmosphere (Anderson *et al.*, 1986).

2.2 Ground-based vertical column amounts

Measurements of the NO_2 vertical column amount at twilight have been collected from 24 zenith-sky UV-visible DOAS spectrometers operating at the stations listed in Table 1: (a) 5 instruments developed by NIWA since the late 1970s (McKenzie and Johnston, 1982); (b) 15 SAOZ instruments (Système d'Analyse par Observation Zénithale) developed by CNRS and performing automated network operation since the late 1980s (Pommereau and Goutail, 1988); and 4 spectrometers of a similar design developed at (c) IASB (Van Roozendael *et al.*, 1995), (d) IFE (Richter *et al.*, 1998), and (e) NILU (Arlander *et al.*, 1998), respectively. Using high-resolution Fourier transform infrared (FTIR) solar spectrometry, the NO_2 column amount throughout the day has also been measured at the Jungfraujoch station by the University of Liège, as part of its monitoring activities initiated in the 1950s (Delbouille and Roland, 1995). Most of the contributing UV-visible sensors have been certified for the NDSC after fruitful participation to major intercomparison campaigns organised through the NDSC or the EC Environment Programme. During such campaigns, the agreement between the various instruments generally falls within 5-10% (e.g., Vaughan *et al.*, 1997; Roscoe *et al.*, 1999). The figure is consistent with an estimated 5-10% accuracy of the retrieved slant column amount taking into account the 5% uncertainty of the NO_2 absorption cross-sections (Merienne *et al.*, 1995), their temperature dependence (Harwood and Jones, 1994; Coquart *et al.*, 1995), and the average 1.5% one sigma confidence level of the least-squares spectral fit. The largest uncertainty in the vertical column remains the AMF, which varies by large factors with the time of the day, the latitude and the season, and has been difficult to characterise until recently due to the near absence of profile measurements. Most of the contributing total NO_2 data records are based on single profiles measured in 1983 during the MAP/GLOBUS balloon campaign (Pommereau *et al.*, 1987), or on the AFGL Reference Atmosphere. Usually, ground-based data are obtained with two different levels of data processing: (i) a version 0 (NIWA) or real-time (SAOZ) spectral analysis, generally performed at the station and transmitted via the ARGOS satellite collection system or the Internet, aiming at a quick evaluation and dissemination of preliminary data; and (ii) a state-of-the-art analysis providing high accuracy data. A characteristic of real-time data consists of their retrieval with absorption cross-sections at room temperature. Consequently, slant columns must be reduced by about 15% due to the large temperature dependence of the NO_2 cross-sections. Data acquired with instruments equipped with uncooled detectors and operating in a severely changing environment must also be corrected for the temporal variation of the instrument slit function arising from instrument temperature changes. State-of-the-art data are all retrieved with low temperature cross-sections and, when relevant, with a dynamic slit function. They have been used here for quantitative investigation of the GOME NO_2 data product, while real-time data have provided valuable support for the pseudo-global extension of the investigation and for the regular monitoring of the GOME performance since the beginning of its operation in July 1995.

Table 1. Contributing ground-based stations

Location	Lat.	Long.	Institute
Ny-Ålesund	79°N	12°E	NILU, IFE
Longyearbyen	78°N	16°E	NILU
Thulé	77°N	69°W	DMI
Scoresbysund	70°N	22°W	CNRS/DMI
Sodankylä	67°N	27°E	CNRS/FMI
Salekhard	67°N	67°E	CNRS/CAO
Zhigansk	67°N	123°E	CNRS/CAO
Harestua	60°N	10°E	IASB
Bremen	53°N	9°E	IFE
Aberystwyth	52°N	4°W	U. Wales
Jungfraujoch	47°N	8°E	IASB, U. Liège
Haute Provence	44°N	6°E	CNRS
Mauna Loa	20°N	156°W	NIWA
Tarawa	1°N	172°E	CNRS, NIWA
Saint Denis	21°S	55°E	U. Réunion
Bauru	22°S	48°W	CNRS/UNESP
Lauder	45°S	170°E	NIWA
Kerguelen	49°S	70°E	CNRS
Macquarie	55°S	159°E	NIWA
Faraday	65°S	64°W	BAS
Dumont d'Urville	67°S	140°E	CNRS
Rothera	68°S	68°W	BAS
Arrival Heights	78°S	167°E	NIWA

3. PSEUDO-GLOBAL GROUND-BASED COMPARISONS

3.1 GOME Data Processor version 2.0

Comparisons of the GOME and ground-based total NO_2 are illustrated in Figure 1 at six typical stations from north to south. Except at Kerguelen, ground-based data displayed in the figure are all retrieved with state-of-the-art algorithms, including NO_2 absorption cross-sections at stratospheric temperatures and dynamic instrument slit function. Conclusions drawn from the first pole-to-pole study (Lambert *et al.*, 1997) remain valid throughout the time period of operational processing with GDP 2.0. The agreement is found to vary significantly with the season and the latitude. A striking feature is the difference between latitudes below 30°N and beyond. Although GOME reports lower values than the ground-based instruments, the general agreement can be considered as reasonable at southern latitudes and up to 30°N, that is, within 5% to 20%. On the opposite, the agreement at higher northern latitudes is mediocre and its quantification often is irrelevant. GOME total NO_2 data beyond 30°N are affected by 3-months shifts of significant amplitude, leading to

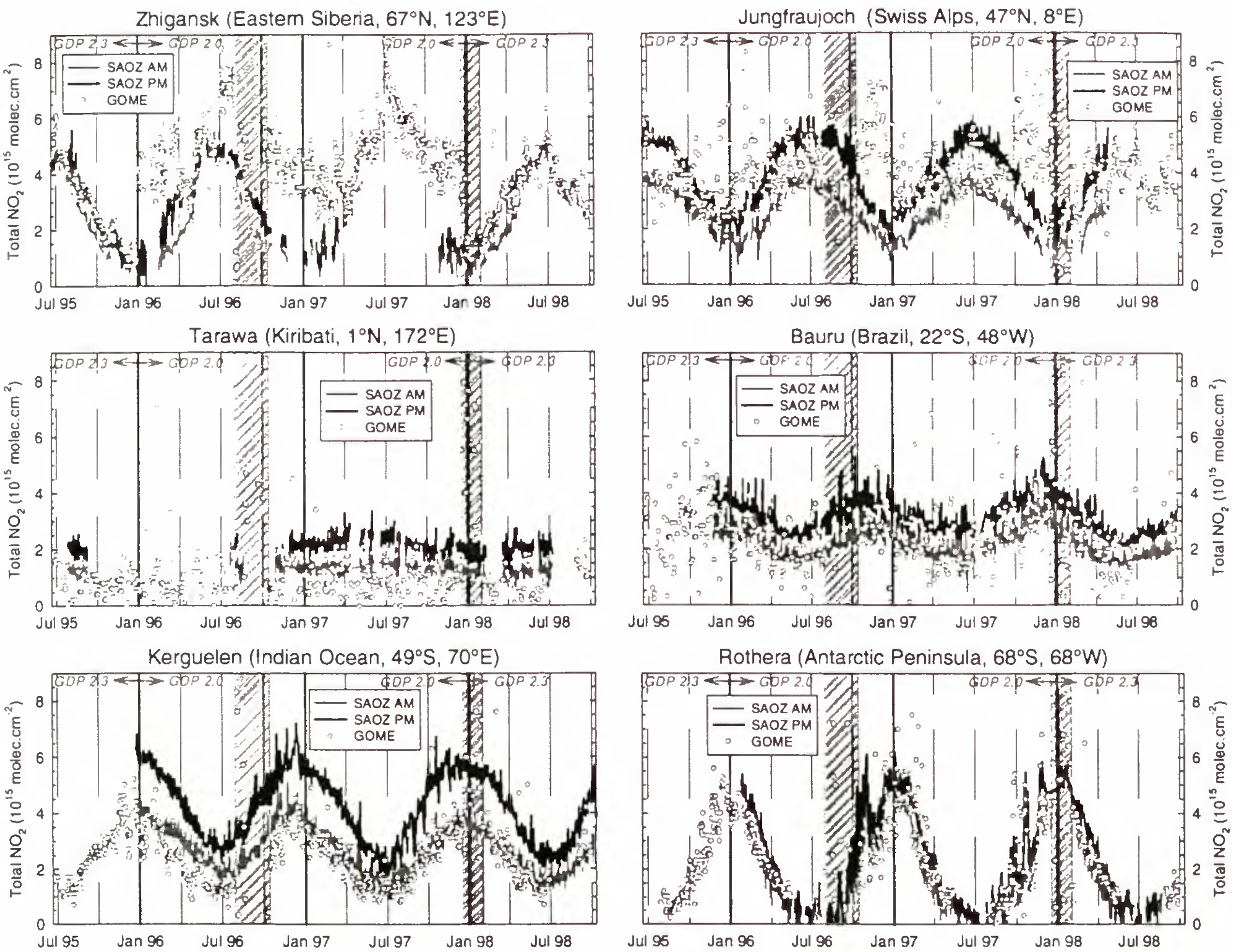

Figure 1. Time-series of the NO_2 vertical column amount derived from ERS-2 GOME and from ground-based observations at six typical stations from north to south. Dashed areas indicate time periods when NO_2 retrieval is dramatically affected by known problems in the spectral channel 3 of the GOME instrument.

a seasonal variation inconsistent with ground-based observations. Enhanced in most of the cases, the day-to-day variability of GOME data is also found to vary with the season and the latitude. The lowest dispersion is observed at middle and high southern latitudes, except during overpass of the station by the border of the polar vortex. It is already larger in the subtropics, where the measured differential optical depth and the signal-to-noise ratio are weaker. Finally, the day-to-day dispersion increases to unrealistic values at northern latitudes, especially in fall and winter, exceeding by far both the day-to-day and dusk-to-dawn differences observed from the ground. The qualitative analysis of global NO_2 maps inferred from GOME level-2 data highlights aberrant spatial structures in the Northern Hemisphere. Anomalies are also detected along orbits, such as the high dispersion of total NO_2 values from pixel to pixel along track, or an unphysical increase of the NO_2 column over summer polar regions in midnight sun conditions. The study also confirms the frequent occurrence of aberrant individual values in the vertical column amounts: negative, or too high by one and sometimes two orders of magnitude, associated with unacceptable error values on the DOAS fitting. There is also a major concern as to the reliability of GDP 2.0 slant column amounts from July 1996 through June 1997: from north to south, they are found to be systematically quantified, as illustrated in Figure 2 over the Alpine region. The apparent dispersion of total column values during this period arises from the division of the slant columns by an AMF influenced by pseudo-random parameters, such as the cloud cover in the line-of-sight, the surface albedo, or the effective line-of-sight angles.

Except aforementioned problems specific to the retrieved slant column amounts which suggest possible problems in the spectral fitting segment of the processing chain, major geophysical inconsistencies of the GDP 2.0 vertical column amount can be explained partly by looking at the corresponding AMF time-series. The three-month shifts observed in Figure 1 in the agreement with ground-based observations at the Jungfraujoch correlate with three-month shifts in the GDP AMF appearing in Figure 2. The principal cause of the problem at latitudes higher than 30°N has already been pointed out in the study reported by Lambert *et al.* (1997). It originates from the use of partially inadequate NO_2 vertical distributions in the calculation of the GOME AMF combined with the enhanced sensitivity of this latter to the actual atmospheric profile shape. The weak consistency of the GDP 2.0 total NO_2 product arises from unreasonable tropospheric burden and shape of the MPI-2D profiles at those latitudes, which decrease drastically the AMF and in addition increase the sensitivity of the AMF to the troposphere. Regions with a relatively clean troposphere – e.g.,

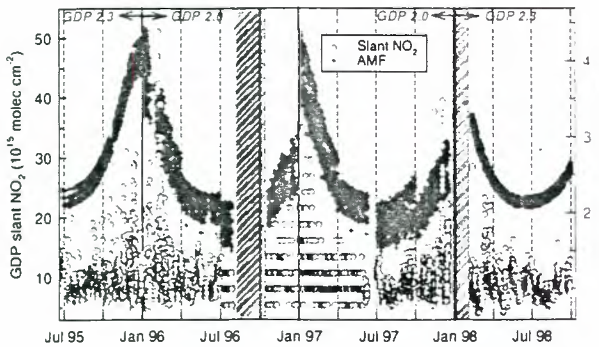

Figure 2. ERS-2 GOME NO_2 slant column amount and down-to-ground AMF over the Alpine region, extracted from level-2 data files generated with GDP 2.0 and 2.3.

the Eastern Siberian station of Zhigansk in Figure 1 - are particularly affected since AMFs are calculated with zonal mean profiles, strongly biased towards the extreme values characteristic of heavily polluted regions. Three-month shifts originate in the strong seasonal difference of the tropospheric burden of the MPI-2D profiles. The effect vanishes towards south where tropospheric amounts become insignificant.

3.2 GOME Data Processor version 2.3

Typical comparisons illustrated in Figure 1 show that the geophysical consistency of the GOME total NO_2 data at northern latitudes has improved significantly with GDP 2.3, especially in fall and winter. The implementation of the US Standard profile in the AMF calculation has produced the expected effect. Three-month shifts seem to have disappeared in both the vertical columns (e.g., Figure 1) and the AMFs (e.g., Figure 2). The seasonal variation of the vertical column is in better agreement with that observed from the ground. From the northern Tropic through the Southern Hemisphere where MPI-2D tropospheric burdens were already more realistic than beyond 30°N, the mean agreement remains reasonable but GOME persists in reporting generally lower values than the ground-based instruments. As demonstrated in section 5, both GOME and NDSC time-series are affected by cyclic biases due to changes in stratospheric NO_2. However, after taking those cyclic biases into account, differences persist, or even increase by a few percent. E.g., GOME total NO_2 at the Equator remains lower than ground-based data by a factor of two. At all latitudes, the quantitative comparison of GDP 2.3 data acquired in 1995 and 1998 does not reveal any significant long-term drift. The occurrence of aberrant individual values of the vertical column amount is less frequent, however, negative or highly scattered values are detected in both the slant and vertical columns and error values on the DOAS fitting remain high, suggesting that the spectral analysis in GDP still needs to be refined.

4. COMPOSITE NO_2 REFERENCE ATMOSPHERE

The evaluation of the optical enhancement of scattered light requires an accurate knowledge of the atmospheric parameters affecting the radiative transfer. An improved database of NO_2 atmospheric profiles has been built up, combining long-term observations from ground-, balloon- and space-based sensors, and modelling results. The concept is illustrated in Figure 3

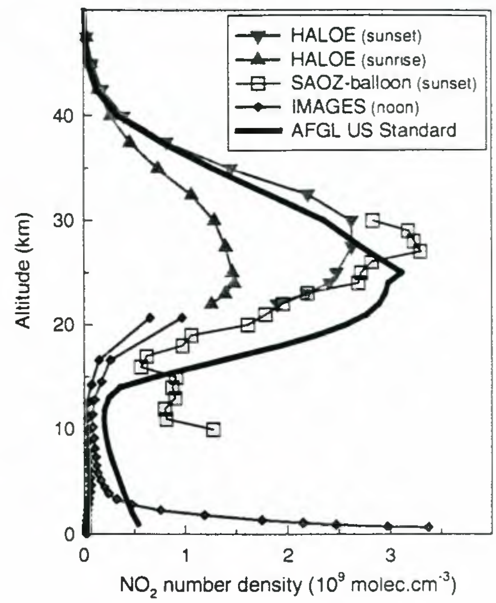

Figure 3. Vertical distribution of NO_2 over Bauru (Brazil) at the end of November, derived from: (a) climatological study of HALOE sunrise and sunset zonal means; (b) SAOZ-balloon measurement at Bauru on 29 November 1996; (c) IMAGES modelling results typical of a clean (Pacific ocean) and polluted (Brazil) troposphere; and (d) AFGL US Standard.

where the different sources for a Brazilian station are compared to the US Standard profile. The information in the middle and upper stratosphere relies on twilight profiles measured for more than 7 years by the Halogen Occultation Experiment (HALOE, Russell *et al.*, 1993) aboard the US UARS platform. HALOE NO_2 data cover altitudes spanning from above the stratopause down to 20 km, however, the accuracy of version 18 used here degrades at altitudes below 25 km, mainly because of Mie scattering by stratospheric aerosols. A three-dimensional chemical transport model of the global troposphere, named Intermediate Model of Global Evolution of Species (IMAGES, Müller and Brasseur, 1995), has been developed jointly at IASB-BIRA and at the National Center for Atmospheric Research (NCAR) to study the global distributions, budgets and trends of 41 chemical compounds, including nitrogen oxides. Modelling results are found to be generally in good agreement with correlative airborne *in situ* measurements. IMAGES has been run to provide monthly means of the vertical distribution of tropospheric NO_2 at local noon, onto a 5° x 5° grid. A database of NO_2 profiles measured during more than 80 flights of the SAOZ-balloon experiment (Pommereau and Piquard, 1994) at middle and high northern latitudes in various seasons and in Brazil, fill in the altitude gap between the HALOE and IMAGES data sets. Covering altitudes from 8 km up to 30 km, the long-term SAOZ-balloon data record gives also a unique opportunity to evaluate the quality of the HALOE and IMAGES information at altitudes where their accuracy might degrade. Climatological information on temperature, pressure and geopotential height has been derived from statistical analyses of European Centre for Medium Range Weather Forecasts data (Trenberth, 1992) and from the COSPAR International Reference Atmosphere (Fleming *et al.*, 1990). Taking into account the high sensitivity of the AMF to the profile shape of the NO_2 vertical distribution, a special care has been given to each step of the

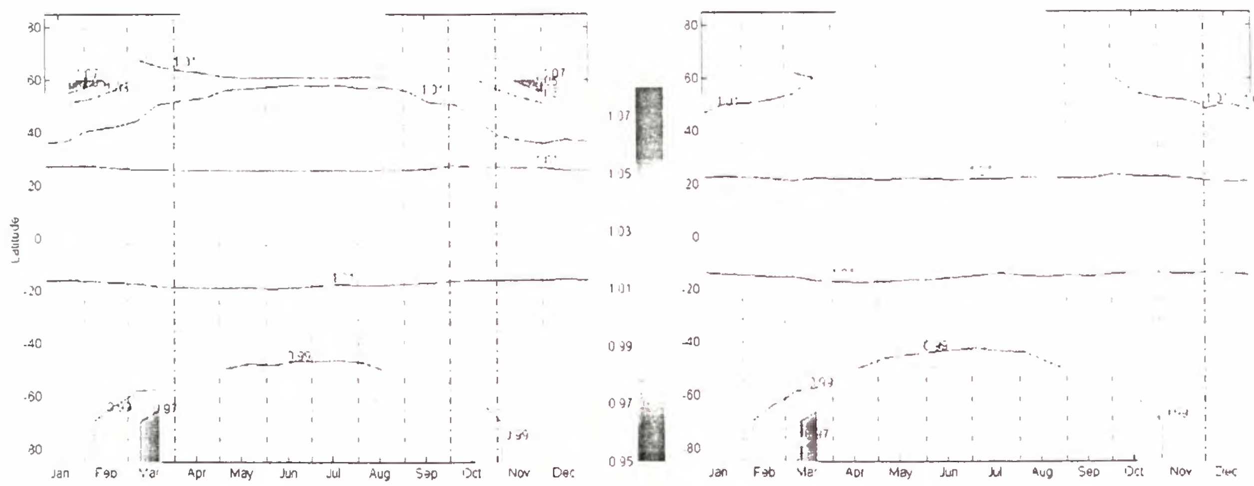

Figure 4. Ratio of nadir-viewing optical enhancement factors for NO_2 calculated with the US Standard and climatological NO_2 atmospheric profiles, as a function of latitude and time, for a sunrise (left panel) and sunset (right panel) stratosphere.

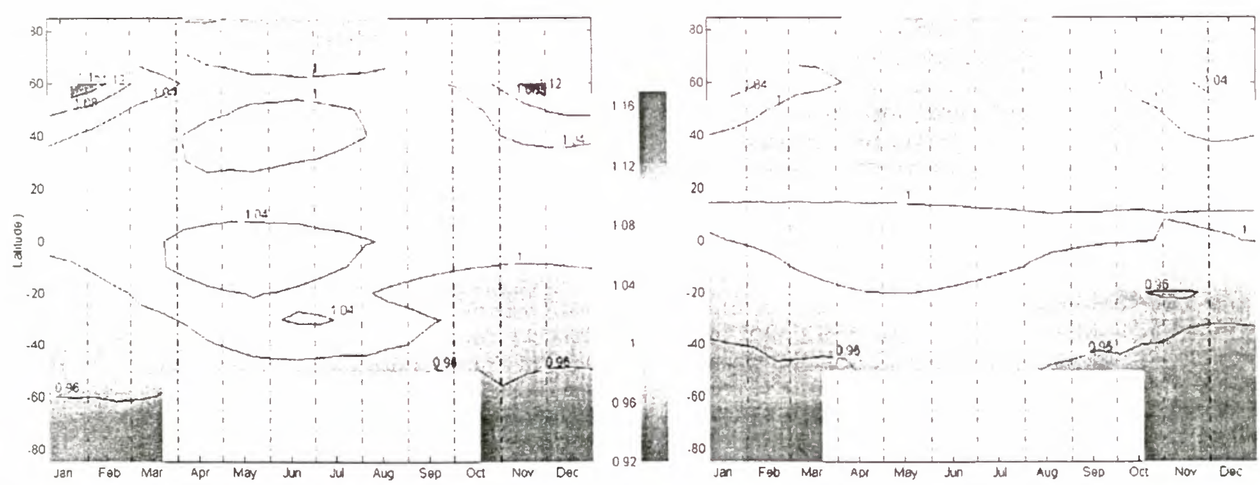

Figure 5. Same as Figure 4, but at 90° SZA in the zenith-sky observation geometry, at sunrise (left panel) and sunset (right panel).

database compilation. The geophysical consistency and the accuracy of the final composite database have been verified from pole to pole, among others by comparison with long-term ground-based observations of stratospheric NO_2. Finally, the database has been implemented in an AMF processor based on both an in-house single scattering radiative transfer model (Sarkissian *et al.*, 1995) and the UVSPEC package (Kylling, 1995) using a pseudo-spherical adaptation of the multiple scattering Discrete Ordinate Radiative Transfer model (DISORT, Dahlback and Stamnes, 1991).

5. CYCLIC SIGNATURES OF STRATOSPHERIC ORIGIN

Seasonal and latitudinal changes in solar illumination, temperature, and distribution of N_2O, the main stratospheric source of NO_2, cause the NO_2 stratospheric profile shape to vary significantly, resulting in a seasonal and latitudinal variation of the AMF. The single profile used by GDP 2.3 or by the ground-based retrieval algorithms cannot take the effect into account, and its use generates in the resulting total columns, fictitious cyclic signatures superimposed on the real total NO_2 variations observed by the instrument. To assess the NO_2 stratospheric profile shape effect and the resulting bias in both the GDP 2.3 and NDSC NO_2 vertical columns, US Standard AMFs have been compared with climatological AMFs. Seasonal and latitudinal components of stratospheric origin have been estimated from the climatological study of the aforementioned HALOE and SAOZ-balloon empirical database. Stratospheric profiles have been completed with IMAGES tropospheric profiles representative of regions free from surface emissions for each latitude belt. From pole to pole, the geophysical consistency of the resulting semi-empirical model has been verified in the long term by comparison with ground-based observations of stratospheric NO_2. Nadir-viewing AMFs have been estimated at the actual mid-morning GOME SZA. Other relevant parameters are: surface albedo of 5%; aerosols: background; line of sight: 0°; wavelength: 437 nm.

5.1 ERS-2 GOME

Figure 4 depicts seasonal and latitudinal variations of the ratio between AMFs calculated in the nadir viewing geometry with the US Standard profile and the stratospheric climatology. The

AMF ratio is a first approximation of the factor by which GDP vertical column amounts should be multiplied to take into account the seasonal/latitudinal bias of the US Standard AMF. The ratio can show more pronounced amplitude with a sunrise stratosphere. This is likely due to the reduced amount of stratospheric NO_2 before the daytime photolysis of its nighttime reservoir N_2O_5, combined to other effects affecting the profile shape, such as the possible relative importance of the tropospheric NO_2 burden and, in winter, the higher sensitivity of the AMF at large SZA to stratospheric variations. During midnight sun and polar night when the diurnal variation of NO_2 is significantly attenuated, and in regions with negligible tropospheric background, the AMF ratio exhibits no clear sunrise/sunset difference. Although sometimes different in amplitude, results with a sunrise and sunset stratosphere are qualitatively consistent, and the ratio at mid-morning should fall between sunrise and sunset values. In the subtropics, the US Standard AMF would yield underestimated vertical column amounts by 1% to 3%. At middle and high latitudes, a clear seasonal variation appears. In summertime, the overall agreement generally falls within 1%. At polar latitudes in fall and spring, the drastic reduction of stratospheric NO_2 distorts the profile shape in such a way that the US Standard AMF would lead to vertical columns too high by 2% in the Arctic to 5% in Antarctica. Denoxification inside the polar vortex would increase the effect. At northern middle latitudes, GDP vertical columns in fall and winter would be underestimated by 1% to 8%. The obvious difference between southern and northern middle latitudes might reflect the difference in the contribution of the troposphere to the net profile shape effect.

5.2 NDSC/UV-visible spectrometers

Figure 5 depicts seasonal and latitudinal variations of the ratio between US Standard and climatological AMFs calculated in the zenith-sky viewing geometry at 90° SZA. Similar results are reached when the US Standard is replaced with other profiles of the AFGL Reference Atmosphere or from the MAP/Globus balloon campaign. Qualitatively, the seasonal and latitudinal variations of the AMF ratio in the zenith-sky geometry display similar patterns compared to those of the AMF ratio at nadir. Quantitatively, variations of the zenith-sky ratio are more pronounced especially at 60°N when they would reach at sunrise a maximum of 15% between summer and winter, against 8% at nadir. Another illustration is the increase of the sunrise underestimation in May at the southern subtropics, from 3% at nadir up to 6% at zenith. Again, the amplitude of the AMF ratio and of its variations at sunrise exceeds sunset values.

6. IMPACT OF TROPOSPHERIC NO_2

Due to its viewing geometry at nadir, GOME is particularly well suited to detect absorbers located in the troposphere. On global maps of NO_2 derived from GOME observations (not shown here), massive emissions of NO_x associated with biomass burning or urban pollution are clearly identifiable by the striking enhancement of the NO_2 slant column amount. As shown in Figure 3, tropospheric emissions bend spectacularly the NO_2 profile shape in the first five kilometres. Consequently, they are expected to modify dramatically the optical enhancement factor. Moreover, it is pointed out in the previous section that the tropospheric background, although moderate, can affect AMF variations of stratospheric origin at northern latitudes. To assess the impact of the tropospheric NO_2 field on the global scale, nadir-viewing AMFs have been evaluated at the actual mid-morning GOME SZA for a variety of representative geographical distributions of tropospheric emissions. The clean zonal tropospheric background used in section 5 has been replaced by three-dimensional IMAGES modelling results, completed by associated climatological stratospheric profiles.

The ratio of AMFs calculated with a clean and a loaded troposphere in March and July is displayed in Figures 6 and 7 with two different colour scales. The dilated colour scale of Figure 6 shows the striking decrease of the AMF in the vicinity of massive NO_x emissions. Tropical biomass burning is associated with an AMF reduction reaching 5% to 20%, while urban and industrial pollution over extended areas can cause the AMF to decrease by 30% in extreme cases. In this exercise, the surface albedo has been fixed to a value of 5%, representative of oceanic conditions. At higher albedo, the reduction of the AMF is less pronounced, and this latter effect is more significant for a loaded than for a clean troposphere. Consequently, the estimated ratio clean/loaded AMF would be reduced over vegetation and ice. The impact of tropospheric emissions depends not only on their strength but also on their relative importance compared to the stratospheric column. Figure 6 shows the significant reduction of the AMF ratio over Europe between March and July, that is, for two very different stratospheric profiles. This latter finding demonstrates the importance of a reliable stratospheric climatology. The reduced colour scale of Figure 7 highlights the influence of the long-range transport of species. AMFs decrease systematically by several percent over the continents, except in a few desert regions. Changes in the AMF ratio over the oceans clearly correlate with changes in the global atmospheric circulation depicted in Figure 8. In particular, the decrease of the loaded AMF over the northern Pacific and Atlantic in March arises from the contamination of the tropospheric background by polluted air masses transported from Asia through the Pacific and from northern America through the Atlantic, respectively.

7. CONCLUSIONS AND RECOMMENDATIONS

The geophysical consistency of the GOME NO_2 data record available at present time has been investigated from pole to pole by means of high-quality ground-based observations associated with the NDSC. The reliability of the data record processed with the version 2.0 of the GDP - operational in 1996 and 1997 - is questionable due to serious problems in both the retrieved slant column amounts and the AMFs. Problems in GDP 2.0 AMFs arise clearly from the use of a partially inadequate NO_2 atmospheric profile database in the AMF evaluation, and the implementation in GDP 2.3 of the AFGL/US Standard NO_2 profile constitutes a first step towards a geophysically consistent total NO_2 product from GOME. However, due to significant variations of the actual profile shape in both the stratosphere and the troposphere, the use of the single US Standard profile generates in the retrieved vertical columns fictitious signatures superimposed on the real total NO_2 variations observed by GOME. Those signatures have been assessed by means of a composite, semi-empirical NO_2 reference atmosphere combining space-, balloon- and ground-based observations and modelling results. Seasonal and latitudinal changes in the stratospheric profile shape result in relative cyclic biases of a few percent, however, the net effect on the AMF is controlled by the tropospheric profile shape. Tropospheric NO_2 reduces the AMF considerably in the vicinity of tropospheric NO_x emissions. Although to a less

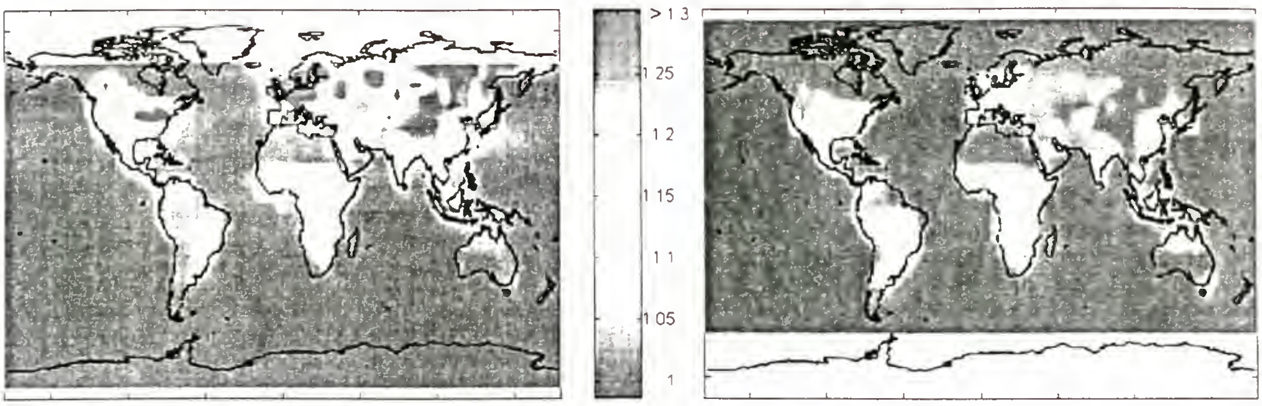

Figure 6. Ratio of nadir-viewing optical enhancement factors for NO_2 calculated with a clean and a loaded noon troposphere. Left panel: March; right panel: July.

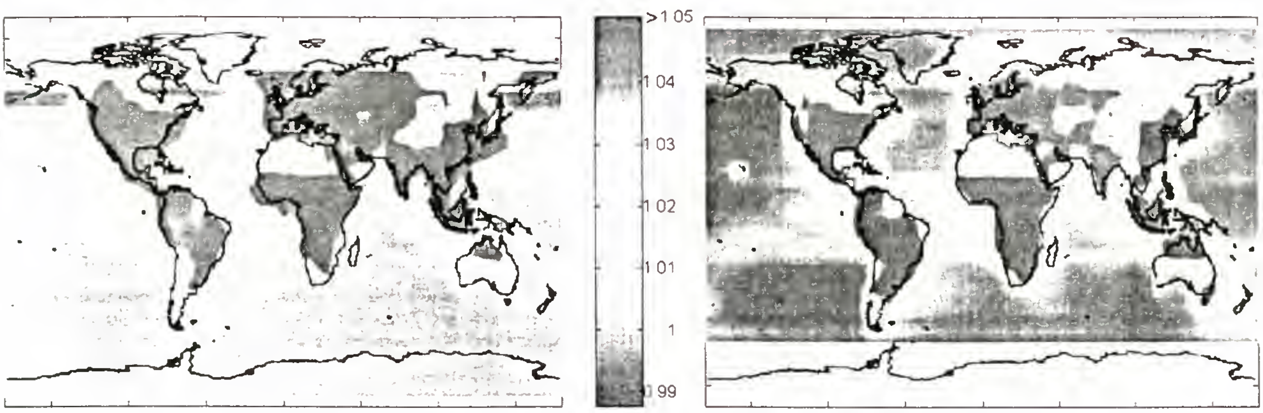

Figure 7. Same as Figure 6, but with colour scale saturated at 1.05

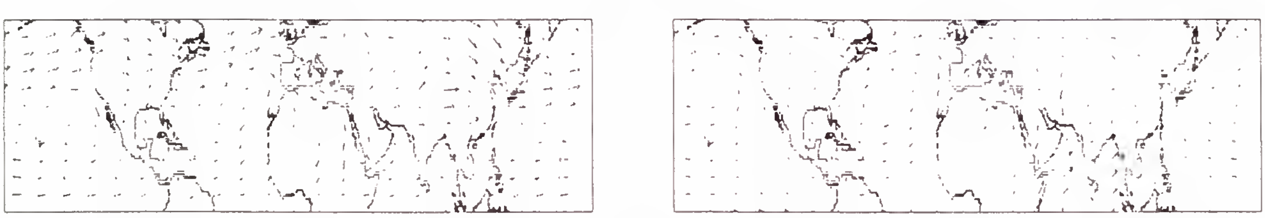

Figure 8. March (left panel) and July (right panel) monthly means of the atmospheric circulation (direction and relative intensity) over the northern Pacific and Atlantic oceans at altitudes between 500 and 850 hPa (derived from Trenberth *et al.*, 1992).

extent, the effect is also observed in remote geographical areas free of tropospheric emission sources, due to the long-range transport of polluted air masses. It is timely to emphasise that the coupled stratospheric and tropospheric profile shape effects will affect the scientific exploitation of nadir total NO_2 data from the progammed GOME series as well as tropospheric information derived from the interleaved limb/nadir mode of the SCIAMACHY to be flown aboard ENVISAT-1.

According to the conclusions drawn from the present study, it is vigorously recommended to improve the operational GDP with an adequate NO_2 reference atmosphere inspired from that developed here, including relevant stratospheric features as well as a consistent 3-dimensional tropospheric background. Over regions affected by intense NO_x emissions, the extreme spatial and temporal variability of the NO_2 field makes the use of a fixed tropospheric database hazardous. Moreover, the enhanced sensitivity of the retrieved vertical column amount to the profile shape is inherent to the static retrieval approach adopted up to now in GDP. Therefore, it might be preferred to adopt for polluted conditions an iterative approach, e.g., consisting of the selection of an adequate reference troposphere after first retrieval of information on the tropospheric burden from GOME data themselves. Finally, the observed bias between the GOME and ground-based total NO_2 time-series persists or increases after deduction of the stratospheric profile shape effect on both the GOME and ground-based data records. This latter finding suggests that other issues related to the retrieval of the slant column amount remain to be addressed, among others, the quality of the DOAS fitting procedure, or the impact of the temperature dependence of the NO_2 absorption cross-sections.

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