TROPOSPHERIC OZONE INFORMATION IN GOME LONG-TERM DATA RECORD

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ABSTRACT

Ozone Profile Retrieval Algorithm (OPERA) version R2 has generated a long-term, global record of atmospheric ozone profile data from backscattered ultraviolet spectra measured since 1996 by ESA's ERS-2 Global Ozone Monitoring Experiment (GOME). The GOME/OPERA data record contains tropospheric ozone information that has been characterised by an ensemble of complementary diagnostic techniques. In addition to classical ground-based validations based on comparisons of the retrieved ozone data with correlative ozonesonde measurements, the tropospheric information content has been explored via an extensive analysis of the averaging kernels associated with the retrieval and an assessment of the vertical smoothing error. Results highlight geographical, seasonal and longer-term variations of the tropospheric information contributed by the GOME measurement and of its agreement with ozonesonde network observations. The impact of these findings on the geophysical usability of GOME OPERA R2 tropospheric ozone data is discussed for applications in the fields of tropospheric chemistry, air pollution monitoring and atmospheric dynamics.

1. INTRODUCTION

As a result of the rapid variation of the ozone absorption cross-section in the 250-340nm spectral window, ultraviolet spectra measured at the nadir of an Earth-orbiting satellite contain height-resolved information on atmospheric ozone. Different methods have been developed to retrieve the ozone profile from backscattered ultraviolet spectra, most of them relying on detailed simulations of the radiative transfer of sunlight up to the instrument and of the instrument response to the incoming radiation. The retrieval consists in minimizing the difference between the simulated and measured radiance spectra. Since the retrieval must be performed at higher vertical sampling than the actual amount of independent pieces of profile information contained in the measurement, the retrieval is in general under-constrained and consequently unstable. Retrieval schemes include therefore additional constraints in the form of *a priori* information on the profile, its shape, and its allowed covariance. The resulting solution is a mix of information contributed by the measurement and of *a priori* constraints. The contribution of *a priori* constraints can be significant where the measurement contains little or no information on the real atmospheric profile, e.g. about fine scale structures or below optically thick tropospheric clouds. Additionally, instruments operating in the harsh space environment exhibit an expected but nevertheless severe degradation and other wavelength dependent effects that impact the signal-to-noise ratio (SNR), which in turn might affect the tropospheric ozone information content.

Operating on board of ESA's ERS-2 satellite since July 1995, Global Ozone Monitoring Experiment (GOME) [1] has measured more than 10 years of earthshine and sunshine ultraviolet spectra. In the framework of ESA-funded CHEOPS-GOME project (Climatology of Height-resolved Earth Ozone and Profiling Systems for GOME) built up on the heritage of the GOME-1 Ozone Profile Retrieval Working Group [2], Ozone Profile Retrieval Algorithm (OPERA) developed at KNMI [3], based on the aforementioned retrieval principle, has been refined in order to generate a global, long-term data record of GOME ozone profiles suitable for scientific applications, including tropospheric and stratospheric ozone research and trend assessments. The objectives of the present paper are to investigate tropospheric ozone capabilities of this data record and to pave the way for similar evaluation of existing and upcoming backscattered ultraviolet ozone profilers like Envisat SCIAMACHY, EOS-Aura OMI and the EPS/MetOp GOME-2 series in Europe, the American SBUV/2 and NPOESS/OMPS systems, and the Chinese FY-3 SBUS/TOU series. Combining information content studies (Section 4) with classical ground-based comparisons (Section 5), our work points out space and time variations of the tropospheric information contributed by the GOME measurement, the signature of level-1 data issues, and particularly the effect of long-term degradation on the global and vertical distribution of tropospheric information. Section 6 concludes with a discussion of the usability of the GOME OPERA R2 ozone profile data record in the fields of tropospheric chemistry, air quality and atmospheric dynamics.

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2. GOME/OPERA R2 DATA

GOME is a grating spectrometer measuring from 240 nm to 790nm the solar irradiance and Earth radiance backscattered at its nadir. It flies aboard ERS-2 on a quasi-polar sun-synchronous mid-morning orbit. A scanning mirror rotating perpendicularly to the flight direction feeds the spectrometer with spectra over three consecutive footprints – or pixels – from West to East, the so-called forward mode, at 320x40km² resolution, plus one spectrum at 960x40km² resolution recorded during the backward motion of the mirror. OPERA retrieves ozone from GOME level-1 data (radiance and irradiance spectra) in the 265-330 nm part of the Huggins band. To get sufficient signal-to-noise ratio, eight forward pixels are co-added, yielding a total footprint of 960x120km². The retrieval of the vertical distribution of ozone number density relies on the optimal estimation method [4]. The radiative transfer is modelled explicitly by the LIDORTA code [10]. The retrieval deduces the ozone profile by minimizing the difference between simulated and measured radiance spectra. It includes an initial *a priori* knowledge of the atmospheric state – namely, an ozone profile climatology and an estimate of the ozone profile covariance – in order to constrain and stabilise the retrieval. Cloud information is retrieved from GOME measurements of the oxygen A-band using FRESCO [5]. More details of OPERA are given in [3]. Its standard output contains ozone partial columns given within 40 layers from the ground up to 0.1hPa and the corresponding averaging kernels (see next section).

Within CHEOPS-GOME, several improvements have been brought to GOME level-1 radiometric data (including improved calibration, better treatment of polarisation features and updated correction for instrumental degradation [6]) and to the OPERA algorithm [3]. The study presented here addresses the OPERA data set version R2, dated June 2006. Note that further improvements of level-1 data and OPERA were implemented more recently. A new reprocessed data set is anticipated, which likely will yield improved results.

3. ANALYSIS METHODOLOGY

The remote sensing nature of GOME observations limits the retrievable ozone profile information to a limited amount of independent vertical layers. Moreover, as already mentioned, a profile retrieved with the optimal estimation method is not a simple measurement of the true atmospheric state but rather a mix of information contributed by the measurement but also *a priori* knowledge of the atmosphere. The contribution of a-priori information can be significant where the measurement contains little or no information, e.g. about fine scale structures of the profile or below optically thick clouds.

With the optimal estimation method [4], if we neglect measurement errors, the retrieved ozone profile can be characterised at first order by the following equation:

$$\hat{x} = x_a + A(x - x_a) \quad (1)$$

This equation describes the linearised relationship between the true (x), *a priori* (x_a) and retrieved ($\hat{x}$) profiles. Each row of the averaging kernel matrix (A) shows, at a given altitude, how the system either smoothes or amplifies departures of the true profile from the *a priori*. A study of algebraic properties of the averaging kernel matrix can help understanding how the system captures actual atmospheric signals. Such a study is described in Section 4, where several diagnostic parameters derived directly from the averaging kernels associated with every OPERA retrieval are used to characterise time and space variations of the tropospheric information content of the GOME ozone data record. Comparisons with ozonesonde measurements are performed in Section 5 to complement the information content analysis and to discuss in Section 6 the potential use of the GOME/OPERA tropospheric ozone data record for several scientific domains of current interest.

4. INFORMATION CONTENT STUDIES

Several mathematically deduced diagnostic parameters have been studied within CHEOPS-GOME. Here we focus on parameters relevant to the analysis of the tropospheric information content: the degree of freedom of the signal, the eigenvalues and eigenvectors of the averaging kernel matrix, and the height-resolved sensitivity to the measurement.

4.1. Degrees of freedom of the signal

The Degree of Freedom of the Signal (DFS) represents the number of independent pieces – that is, independent atmospheric layers – of information that have been retrieved by OPERA. The DFS can be calculated as the trace of the averaging kernel matrix:

$$DFS = tr(A) \quad (2)$$

DFS time series of OPERA ozone profile data over nearly ten years are shown in Figure 1. Data have been separated within six 30°-wide latitude bands.

OPERA provides five up to six independent pieces of information at the beginning of GOME operations. A small seasonal variation exists with about 0.5 DFS of amplitude. High and middle latitude profiles have about one piece of information more than sub-tropical profiles. The DFS presents a long-term decrease at all latitudes and about one unit of DFS is lost between the first three years of measurement and the end of the data record. The observed drift correlates with instrumental degradation and can be understood as a consequence of the reduction of the signal-to-noise ratio. Although an updated correction for instrumental degradation has been implemented in the algorithm, the slow SNR loss with time can not be totally compensated for, resulting in a loss of information content in the retrieved profiles.

4.2. Eigenvalues and eigenvectors

The DFS study gives access to the total information content of the profile. In order to better understand the vertical distribution of the different pieces of information, and in particular to better identify the tropospheric information, we will look at the eigenvectors of the averaging kernel matrix. Consider the eigenvector matrix (U) and the diagonal eigenvalue matrix (Λ) of the averaging kernel matrix, as:

$$AU = \Lambda U \tag{3}$$

Rewriting Rodger's equation (1) in the eigenvector base, it can be demonstrated that the state vector decomposes into patterns, some corresponding to eigenvalues close to one that will be well captured by the measurement, and some with eigenvalues close to zero that will mainly come from the a-priori state [4]. Eigenvectors corresponding to eigenvalues close to one show the different layers in the atmosphere that can be well reproduced by the measurement system.

Figure 2 shows an example of the first six eigenvector associated with the six largest eigenvalues, for a GOME profile measured at Northern middle latitude in June 2000. The eigenvectors illustrate the shape and altitude of the independent layers from which information can be retrieved. The first four eigenvectors correspond to pieces of information located mainly in the stratosphere, while the last two include also tropospheric information. The first fourth are associated with eigenvalues close to one: the retrieved profiles will contain information contributed mainly by the measurement. The fifth pattern comes from the measurement and *a priori* information. The last pattern comes mainly from *a priori*. It corresponds to the fifth degree of freedom of the signal observed at that latitude and time period.

Figure 3 shows time series of the six largest eigenvalues (each one with an offset) for all GOME ozone profile data retrieved between 30°N and 60°N. The decrease in the number of degree of freedom observed at section 4.1 results here in the progressive decrease of the fifth and sixth eigenvalues. As the fifth and sixth eigenvectors contain tropospheric patterns, the effect can be interpreted as a loss of tropospheric information due to instrumental degradation. Similar conclusions can be drawn at other latitudes.

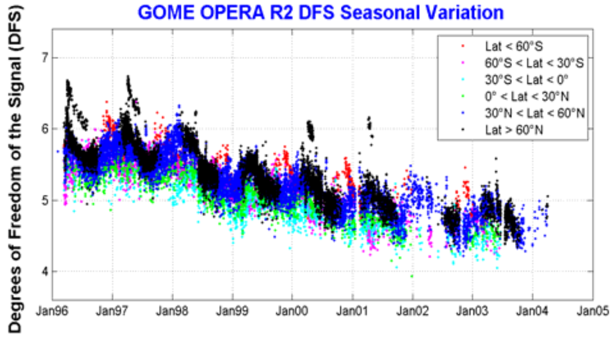

Figure 1 – Time series of the number of degree of freedom of the GOME/OPERA ozone profile signal. Data have been separated into six latitude bands.

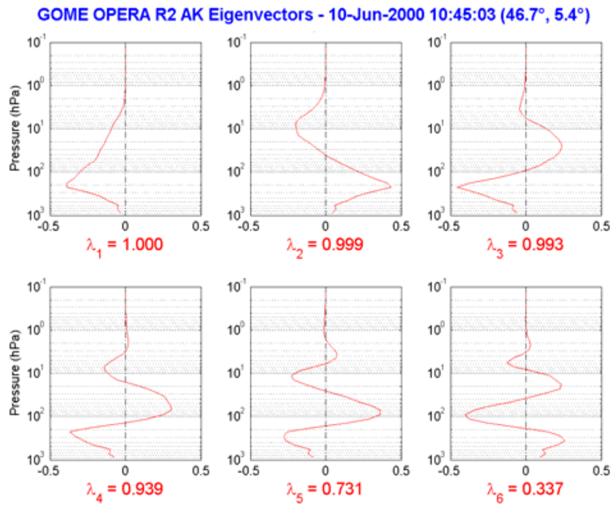

Figure 2 – Eigenvectors of OPERA averaging kernel matrix associated with the largest six eigenvalues.

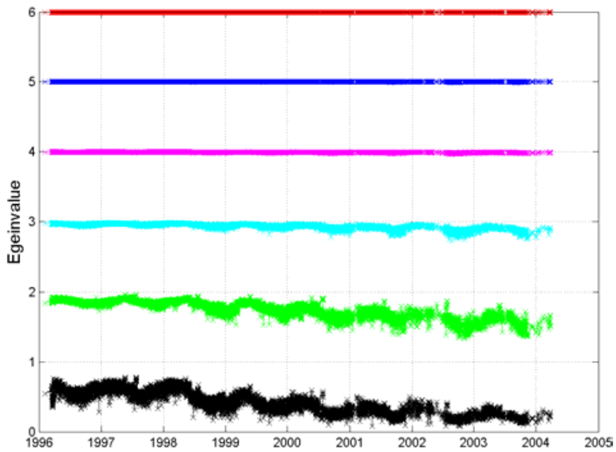

Figure 3 – Time evolution of six largest GOME/OPERA eigenvalues (with offset) for all ozone profile retrievals between 30°N and 60°N.

4.3. Height-resolved sensibility to the measurement

The sensitivity of the retrieval to the measurement can be estimated by another diagnostic parameter calculated from the averaging kernel matrix. The area of the averaging kernel function – the rows of the averaging kernel matrix – is close to one at levels where the retrieval is accurate and can be seen as a measure of the fraction of the retrieved value that comes from the measurement rather than from the *a priori* profile. A vector of areas is calculated from the averaging kernel matrix by multiplication with a unit vector (u):

$$areas = Au \tag{4}$$

It can also be interpreted as the response of the retrieval to a unit perturbation in all elements of the state vector.

Figure 4 and Figure 5 show the time variation of the vector of areas of GOME/OPERA averaging kernels at Northern middle latitudes and Southern low latitudes. The figures reveal bands of positive and negative sensitivity that alternate from the ground up to the stratopause. They correspond to the independent pieces of information in the retrieved ozone profile. During the first three years of GOME operation, we observe six bands of sensitivity at middle and high latitudes and five bands in the tropics. There is one piece of tropospheric information in the tropics and a maximum of about 1.5 at higher latitudes. The instrumental degradation of GOME impacts clearly the tropospheric information content of the retrieval: the sensitivity to the low troposphere decreases rapidly with time after mid 1998 and even vanishes after 2002 in the tropics. The instrumental degradation also impacts the altitude of the bands of sensitivity, the latter being redistributed in altitude as a function of time.

Figure 4 and Figure 5 also reveal a seasonal variation of the altitude of maximum sensitivity, in particular during the first three years of measurement, when the total information content was optimal. From the troposphere up to the middle stratosphere the information comes from upper layers in winter and from lower layers in summer. This behaviour correlates with a similar seasonal variation exhibited by the DFS, confirming that the total information content is about 0.5 larger in winter than in summer. This seasonal variation decreases with latitude. A detailed study has shown that the seasonal variation correlates with the ozone column along the GOME optical path, or slant column amount. At first order it is a function of the total ozone column and the solar zenith angle. Figure 6 shows the height-resolved sensitivity of GOME profiles versus ozone slant column amount. As the ozone slant column increases, the sensitivity to the layers in the stratosphere move upward, and the sensitivity to the lower troposphere disappears.

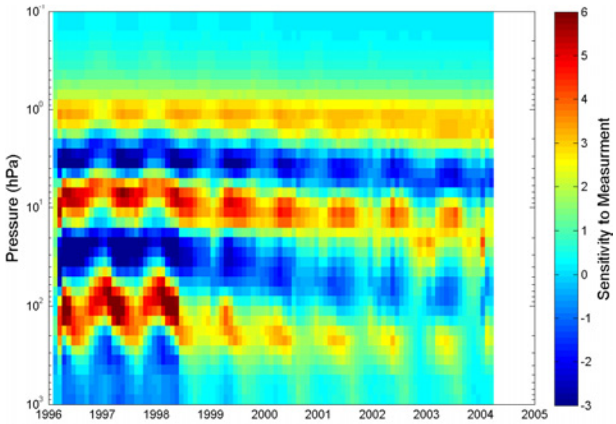

Figure 4 – Time series of areas of GOME/OPERA R2 averaging kernels (versus atmospheric pressure), as an estimate of the sensitivity to the measurement. Monthly mean data from 30°N to 60°N.

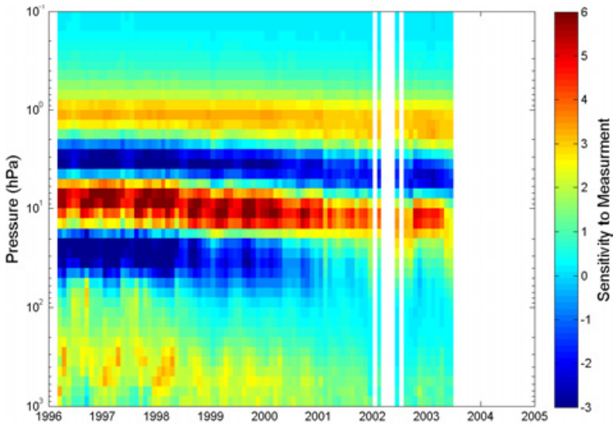

Figure 5 – Same as Figure 4, but monthly mean data from the equator to 30°S.

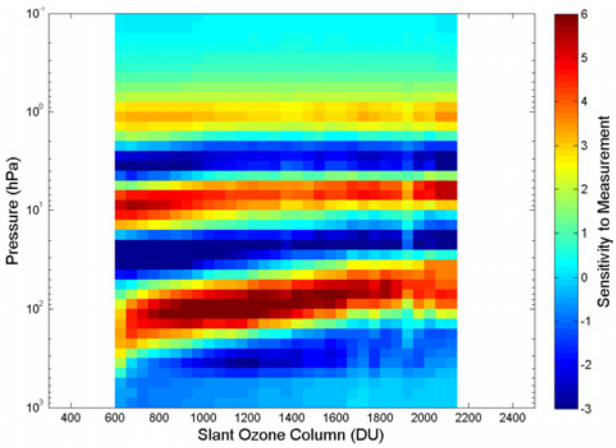

Figure 6 – Same as Figure 4, but areas of averaging kernels plotted as a function of the ozone slant column amount, in the Arctic, averaged over 1997.

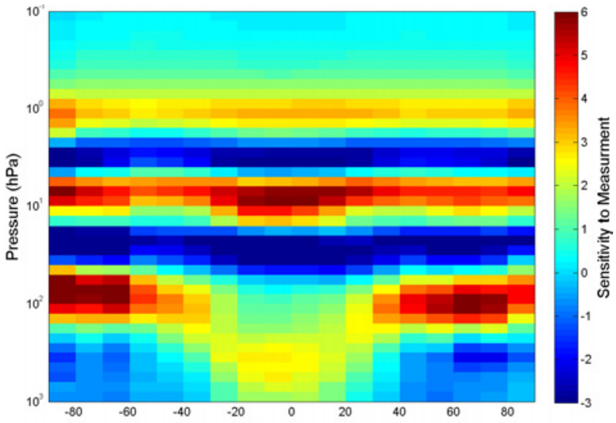

Figure 7 – Same as Figure 4, but areas of averaging kernels plotted as a function of latitude, for year 1997.

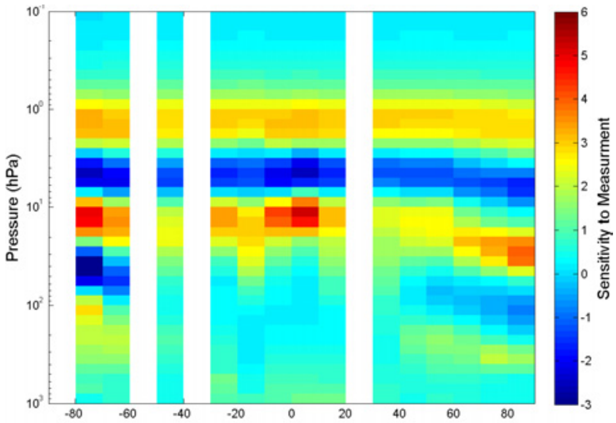

Figure 8 – Same as Figure 7, but for year 2002.

This behaviour might be related to the increasing optical thickness of the atmosphere when the ozone amount a-long the optical path increases. More absorption by ozone increases the GOME measurement SNR, hence, the retrievable information content. Consequently, the information should move slowly to the altitude of the ozone maximum, where most of the slant column increase occurs. Moreover, as large amounts of ozone increase the optical thickness of the atmosphere, the solar flux reaching the lower troposphere decreases, this atmospheric layer being occulted by the thicker and thicker stratospheric ozone layer, which, in addition, is passed through two times by the sunlight backscattered to the satellite. Consequently, the tropospheric information measured from the satellite vanishes.

Figure 7 depicts the meridian variation of the GOME/OPERA sensitivity averaged over 1997. The most striking feature is the merging of the two lower bands of sensitivity when going into the tropics. There is only one piece of information in the subtropical troposphere. Its altitude differs strongly from the sensitivity altitudes outside of the extra-tropical barrier. Figure 8 shows results for 2002. Measured tropospheric information has clearly vanished at subtropical latitudes while some limited sensitivity persists in the upper troposphere of the high and middle latitudes.

5. COMPARISON STUDIES

5.1. Correlative ozonesonde measurements

In situ measurements acquired by lightweight balloon-borne electrochemical ozonesonde provide accurate and independent ozone profile data from the ground up to typically 30km [7]. The typical vertical resolution of the ozone profile is 100-150 m with a typical accuracy of 5% to 7% [8]. Ozonesondes are launched at a variety of stations from pole to pole under the auspices of WMO's Global Atmosphere Watch programme (GAW) [11] and contributing networks like the Network for the Detection of Atmospheric Composition Change (NDACC) [12] and the Southern Hemisphere ADditional OZonesonde programme (SHADOZ) [13]. GOME and ozonesonde network data have been processed to select a robust set of collocated profiles within a maximum spatial distance of 500km and time difference of 6h. The vertical resolution of the ozonesonde profile is much higher than that of GOME/OPERA retrievals. We will see later that the way high resolutions are degraded can impact significantly the interpretation of comparisons.

5.2. Vertical smoothing error of GOME/OPERA

Ideally, the averaging kernel matrix should be equal to the identity matrix. If that was the case, according to Eq. 1, the retrieved profile $\hat{x}$ should be equal to the true profile x, and the *a priori* profile x_a should have no influence on the retrieved profile. Practically, the averaging kernel matrix contains non-diagonal elements, which reflect the vertical spread of the ozone information contributing to the retrieved value. This spread of the information generates the so-called vertical smoothing error [9,4]. For an individual retrieved profile $\hat{x}$, knowing the associated averaging kernel matrix A and *a priori* profile x_a, the vertical smoothing error can be estimated using the convolution/offset function described by Eq. 1, assuming that the true profile is the one measured by the ozonesonde. The error is then the difference between the result of Eq. 1 and the ozonesonde profile, yielding Eq. 5:

$$\Delta x = x_a + A(x_{os} - x_a) - x_{os} \quad (5)$$

The exercise has been performed at several stations offering adequate ozonesonde time series. Figure 9 illustrates smoothing error estimates for tropospheric column obtained by integrating GOME ozone profiles from the ground up to the tropopause at three stations: Ny-Ålesund in the Arctic (79°N/12°E, Spitzbergen), Uccle

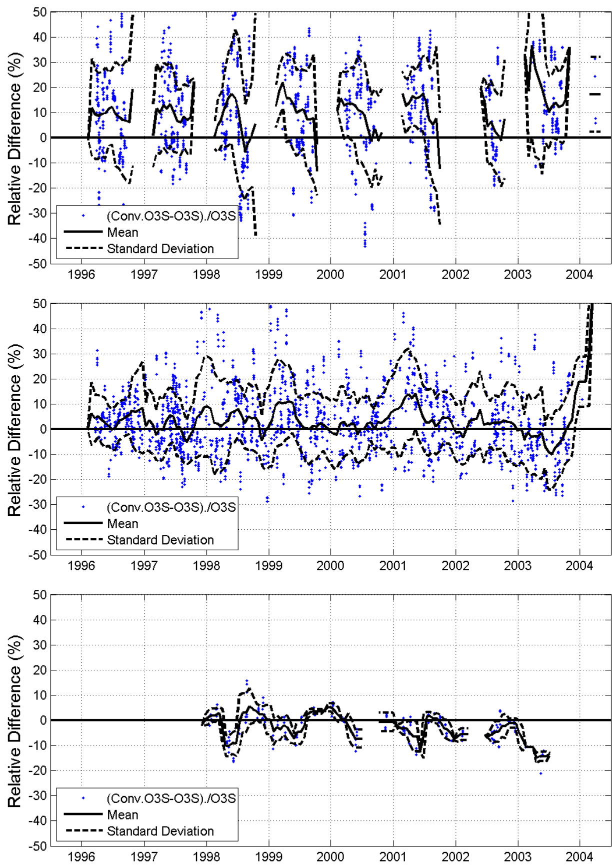

Figure 9 – Percent relative difference between degraded and actual ozone tropospheric columns at the ground stations of, from top to bottom, Ny-Ålesund, Uccle, and Ascension Island.

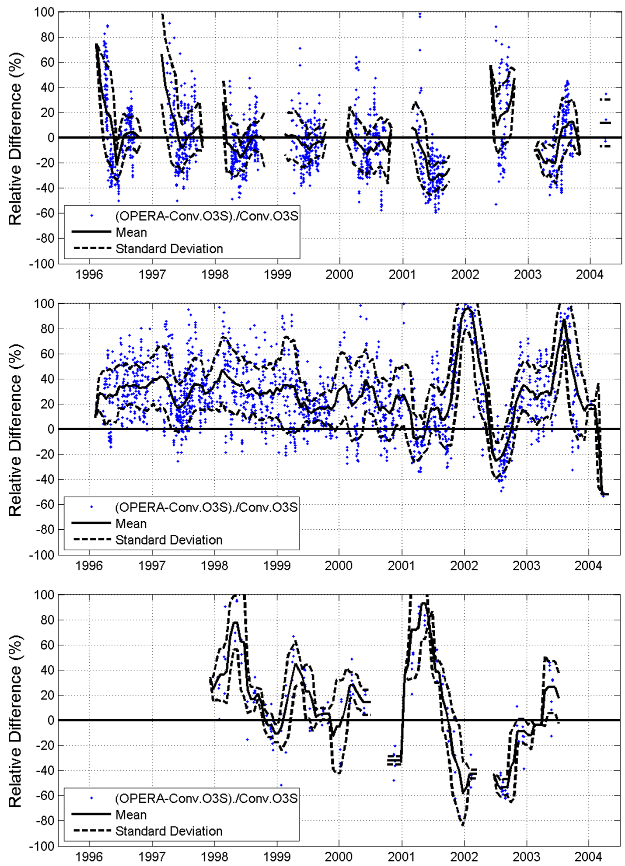

Figure 10 – Percent relative difference between GOME/OPERA R2 and ozonesonde tropospheric columns at the ground stations of, from top to bottom, Ny-Ålesund, Uccle, and Ascension Island.

in mid-latitude Europe (51°N/4°E, Belgium), and Ascension Island in the inter-tropical zone of the Atlantic Ocean (8°S/14°W). As expected from information content studies reported in Section 4, the vertical smoothing error varies with latitude, and even differs from one geolocation to another. At Ny-Ålesund, vertical spread of the measured ozone profile information confers to GOME retrievals a high bias of about 15% ± 20% (1σ standard deviation) with seasonal and inter-annual variations of 5 to 15%. Individual high biases of up to 50% can be observed especially in winter and early spring. At Uccle, the high bias reaches only a few percent on an average, but its standard deviation is as large as that observed in the Arctic. It also shows a significant seasonal variation. At Ascension Island, the standard deviation is much lower and it makes appear more clearly a seasonal cycle of 15% from peak to peak. It is important to realise that the vertical smoothing error is inherent to the nature of both the backscatter ultraviolet measurement and the Optimal Estimation retrieval method. It will affect any direct use or interpretation of the GOME ozone profile data and should always be taken into account somehow.

5.3. Comparison of tropospheric columns

The study reported in this subsection consists of comparisons of ozone tropospheric columns derived from GOME and ozonesonde data by integration of their profiles up to the tropopause. Prior to this calculation, ozonesonde data are degraded to the GOME/OPERA perception again through Eq. 1, in which the true profile is substituted by the ozonesonde observation, and A and x_a are the averaging kernels and *a priori* profile corresponding to the considered retrieval. Biased and smoothed this way, ozonesonde data lose their independence with respect to GOME data. The advantage is that GOME/OPERA vertical smoothing errors quantified in Subsection 5.2 are removed from the comparison error budget [4]. Figure 10 shows for each pair of collocated GOME and ozonesonde measurements the percent relative difference between respective tropospheric columns at the same three stations as those considered in Figure 9. Running mean and standard deviation are also indicated. At Ny-Ålesund, a reasonable agreement within ± 20% is found in summer and fall until 2001. Larger differences are observed in 1996 and 1997 near the winter terminator and in early spring. After 2001, the mean agreement deteriorates and varies from year to year. In Uccle, a mean positive bias of +30% is observed between GOME and correlative ozonesonde data over the entire data record. Similar results are obtained at a number of other European and North American stations. The bias exhibits only a small, noisy seasonal cycle and does not vary significantly from 1996 to 2001, when dramatic oscillations appear suddenly, ranging from +100% down to –50%. At Ascension Island, large differences and oscillations are observed already since 1998. Oscillations consist mainly of a clear seasonal cycle with highs in the first part of the year and lows in the second part. There is also a trend between 1998, with a mean positive bias of about +35%, and 1999-2000, with a mean positive bias of only +10%. Again, 2001 seems to be the trigger of enhanced differences ranging from +100% down to –55%.

5.4. Comparison of tropospheric profiles

Since the physics behind ozone retrieval from backscattered UV measurements limits the true vertical resolution of tropospheric ozone data to 10 km at best, direct comparison with ozonesonde profiles at the vertical sampling of the GOME retrieval (≈1 km) does not make sense if the objective is to assess the absolute accuracy of GOME ozone profile data. Nevertheless, comparisons at high vertical resolution complement the algebraic study of averaging kernels reported in Section 4, with the advantage to be independent from the retrieval system since based on external data.

Figure 11 reports height-resolved differences between GOME and ozonesonde data at Uccle. In the upper part, ozonesonde profile data have been integrated in each of the OPERA vertical layers. The GOME/OPERA vertical smoothing error contributes to the observed differences. In the lower part, ozonesonde profile data have been first convolved via Eq. 1 using corresponding OPERA *a priori* profile and averaging kernels. In the latter case, the vertical smoothing error is supposed to be removed. Figure 11 confirms the existence of a positive bias in the troposphere, already noted in the comparison of tropospheric columns (Subsection 5.3). The vertical structure of the differences is interesting: between the ground and burst point of the balloons, five seasonally varying layers of positive and negative biases alternate with altitude. Apparently, the retrieval seems to fetch information from the upper troposphere and lower stratosphere and push it down into the troposphere. Similar results are obtained at other sounding stations. As expected from estimates of the GOME smoothing error as illustrated in Figure 9, the same vertical oscillation appears in both parts of Figure 11, that is, with and without degrading ozonesonde data with OPERA averaging kernels via Eq. 1. This finding suggests some residual instability of the retrieval, calling

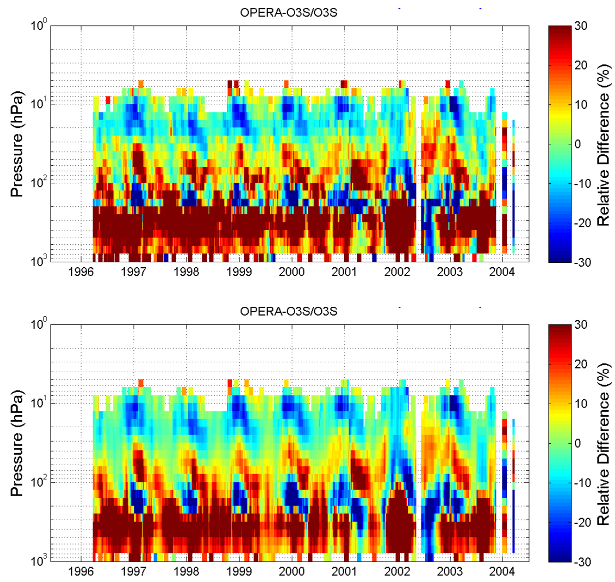

Figure 11 – Percent relative difference between GOME OPERA R2 and ozonesonde profile data at Uccle. Top: ozonesonde profiles smoothed with square box. Bottom: ozonesonde profiles smoothed with averaging kernels.

for an improvement of the retrieval constraints. After June 2001, the vertical oscillation and its seasonal variation are more pronounced, generating the large oscillations observed in the comparison of tropospheric columns (Subsection 5.3). Negative biases appear, while only positive biases were noted previously.

6. DISCUSSION AND CONCLUSION

6.1. General discussion

The present diagnostic study of the GOME ozone profile data record generated by the OPERA R2 system combines classical comparisons with correlative ozonesonde measurements, with a characterisation of the tropospheric information contributed by the GOME measurement. The latter study is based on the analysis of OPERA averaging kernels, which indicate how the system smoothes departures of the true profile from the *a priori* profile. A main finding is that the tropospheric information contributed by the GOME measurement, as opposed to the information contributed by *a priori* constraints of the retrieval, varies with altitude, latitude and time. The vertical distribution of the sensitivity to the measurement is a relatively smooth function of several parameters controlling the radiative transfer through the atmosphere. Among others, in the absence of optically thick clouds masking the lowest layers, the sensitivity to the lower troposphere depends directly on the slant column amount of ozone and the solar zenith angle. It can be moderate at middle latitudes but weak in the inter-tropical zone.

The main feature of the latitude variation is a sudden reduction and vertical redistribution of the measured tropospheric information where moving from the high and middle latitudes – with about 1.5 layers of independent information through the entire free troposphere – to the low latitudes – with roughly one layer of independent information in the troposphere above 1-2 km of altitude. The time variation has a more complex behaviour: it blends short term components, seasonal components attributed partly to the seasonally varying atmospheric optical thickness, and a longer-term component reflecting GOME level-1 data quality issues. Three main periods can be distinguished, which correspond each to a different phase of the instrumental degradation: (1) 1996 to 1998, when the retrieved information content on tropospheric ozone is at its maximum; (2) 1999 to mid-2001, when the tropospheric information content degrades progressively and is redistributed slightly with respect to altitude; and (3) after mid-2001, when the tropospheric information content in the inter-tropical zone vanishes.

Reflecting the vertical spread of the measured information, the vertical smoothing error of GOME/OPERA tropospheric ozone data consists of biases and seasonal and meridian signals that might alter the interpretation of the data and the comparisons if not taken into account properly. Biases and variations depend on the latitude, sometimes the geolocation, and maybe the orography. Surprisingly, long-term effects associated with the evolution of level-1 data quality do not seem to be significant.

Where and when information content studies conclude to a sufficient contribution of the measurement to the retrieved ozone profile, and taking into account properly the vertical smoothing error, ground-based comparisons with ozonesonde network data lead to mixed results, ranging from excellent agreements of about ±10% at some stations to, at other stations, permanent positive biases of up to +35% and time-dependent oscillations of even larger amplitude. They point out vertical oscillations suggesting residual instabilities in the retrieval. They make also appear the three distinct time periods mimicking the long-term evolution of the GOME level-1 data quality. While the first two periods seem to offer comparable data quality, GOME tropospheric ozone data after mid-2001 should be handled with the greatest circumspection.

It is timely to remind that present results relate to the GOME/OPERA data set version R2, dated June 2006, which was the best data set available at the time of the present study. Further improvements of both the level-1 data and the OPERA software were implemented more recently. Consequently, a new reprocessed data set is anticipated, which likely will yield improved results.

6.2. Tropospheric chemistry and air quality

Low and middle latitudes present different tropospheric capabilities. At low latitudes, during the first years of operation, the unique piece of independent information on tropospheric ozone spreads over about fifteen kilometres and its height varies with the season. The maximum sensitivity is located several kilometres above the surface. Consequently, the measurement of anthropogenic tropospheric ozone is expected to depend on the vertical transport of air masses rich in ozone and in ozone precursors. Although occurring during the dry season rather than the convective season, tropical biomass burning concerns large geographical areas and can last several days. Massive amounts of ozone and precursors generally have the time to reach altitudes to which GOME is the most sensitive. The actual sensitivity to fire-related ozone will depend on factors controlling vertical transport, like the surface temperature, but also the backscattered ultraviolet measurement, like absorbing and scattering aerosols. The detection of ozone related to industrial pollution, which is usually associated with dynamically stable meteorological systems with weak vertical transport, is more uncertain as ozone and its precursors remains in the vicinity of the ground. Where and when information content studies conclude to a sufficient contribution of the measurement to the retrieved ozone profile, comparisons between GOME and ozonesonde measurements show a fair agreement and usually conclude to positive deviations superior to 20% in the inter-tropical low troposphere, with sometimes negative deviations. Both information content studies and ground-based comparisons suggest that GOME/OPERA observations of fire-related ozone are feasible within the first three years of operation, become less accurate after mid 1998, and are highly questionable after 2001.

At middle latitudes, about 1.5 independent layers offer tropospheric information in 1996-1998, with a seasonal variation of the altitude of maximum sensitivity and a correlation with the ozone slant column amount. At higher latitudes, 1.5 pieces of information are also accessible to GOME/OPERA, however, under large slant columns a large part of the troposphere is masked by the optical thickness of the overlaying stratospheric ozone. Compared to the low latitudes, sensitivity at middle latitudes goes more deeply into the lower troposphere, allowing in principle a better detection of ozone related to precursors emitted by industrial and urban activities. Although satisfactory from the perspective of information content, ground-based comparisons reveal at middle latitudes a systematic overestimation by about 20% of the tropospheric ozone column. This bias cannot be explained by the vertical smoothing error of the GOME/OPERA data. At high latitudes, at moderate solar zenith angles, a better agreement within ±20% is found.

6.3. Tropospheric ozone as a dynamical tracer

From the strict point of view of information content, GOME/OPERA R2 tropospheric ozone data over 1996-1998 might be suitable for use as tracer for studies of atmospheric dynamics and in meteorological data assimilation, although limited to phenomena at low vertical resolution. The smooth meridian structure of the tropospheric information content and of the altitude of maximum sensitivity might allow tracer use of GOME tropospheric ozone across the high and middle latitudes. However, it must be kept in mind that comparisons with ozonesonde measurements at several European and North American stations have highlighted a bias of which the amplitude depends on the geolocation. This geographical dependence of the GOME bias might introduce fictitious patterns in tropospheric ozone that should not be attributed too rapidly to dynamical phenomena. The sudden change of tropospheric information content near the tropics poses a problem for studies of dynamical exchanges across the extra-tropical barrier. From 1999 to mid 2001, some GOME/OPERA information content on tropospheric ozone remains, but it degrades progressively, at all latitudes. In counterpart, the fraction of *a priori* information, thus not contributed by the GOME measurement itself, increases in the retrieved product. The altitude range covered by the available tropospheric information also decreases progressively. From 2001 onwards, the added value of the GOME measurement is questionable.

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