RETRIEVAL OF BrO COLUMNS FROM SCIAMACHY AND THEIR VALIDATION USING GROUND-BASED DOAS MEASUREMENTS

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ABSTRACT

Building on the experience acquired with ERS-2/GOME, a scientific algorithm has been developed at BIRA-IASB for the retrieval of total BrO columns from SCIAMACHY nadir measurements. In order to overcome problems related to anomalies in the polarization response of the SCIAMACHY instrument, specific BrO retrieval settings have been introduced, different from those used for the retrieval of GOME data.
In the present study, we focus first on an assessment of the accuracy of the SCIAMACHY slant column retrieval, through various sensitivity tests and comparisons with coincident measurements from GOME. Second, SCIAMACHY nadir BrO results are compared with independent correlative measurements of BrO columns resolved into their stratospheric and tropospheric contributions, and photochemically matched to satellite observations. Correlative data are derived from DOAS observations performed by IASB-BIRA at three locations (Harestua 60°N, OHP 44°N, and Reunion Island 22°S). From the combined analysis of GOME, SCIAMACHY and ground-based and balloon-borne DOAS observations the overall consistency of the remote-sensing BrO observing system is evaluated and discussed in light of recent findings published in the literature. In particular our results strengthen the existence of a worldwide background of BrO located in the free-troposphere with mixing-ratios in the range from 1-2 pptv.

1 INTRODUCTION

Bromine compounds, which have both anthropogenic and natural sources, cause about half of the chemical loss that results in the Antarctic ozone hole in the stratosphere. Low levels of ozone in the atmosphere's lowermost layer, the troposphere, during the polar spring result from bromine released from melting sea ice and 'frost flowers'[1]. But it is also becoming evident that substantial amount of bromine could be present in the free-troposphere at the global scale [2,3]. Much research is currently being devoted to understanding the sources and sinks of these organic bromine compounds, and their effects in the atmosphere [4].

Bromine monoxide (BrO) is the most abundant bromine bearing inorganic trace gas during daylight. BrO columns have been monitored globally since 1996 by the GOME instrument onboard the ESA ERS-2 platform allowing to document the extent and the time evolution of inorganic bromine in both the troposphere and the stratosphere. Due to problems with tape storage on ERS-2, GOME lost its ability to provide global coverage. In order to ensure long-term monitoring of the atmospheric inorganic bromine content, it is of great importance to link GOME records to measurements from new satellites. SCIAMACHY, the successor of GOME, has been launched in 2002 on the ESA ENVISAT platform. In the context of the ESA DUP II TEMIS project, a scientific BrO vertical column product has been developed at BIRA-IASB, based on the expertise previously developed with GOME processing.

This paper aims to validate this algorithm based on the use of GOME data and correlative ground-based measurements, as initiated in [5].

2 SCIAMACHY BrO RETRIEVAL

The technique used to derive BrO amounts from satellite measurements is the so-called Differential Optical Absorption Spectroscopy (DOAS) [6]. GOME BrO slant columns are derived in the wavelength interval from 345 to 359 nm [7], making use of the characteristic absorption structures of BrO in this region. Unfortunately the simple transfer of the GOME BrO settings to SCIAMACHY spectra has been found unsuccessful. The main reason for this is the presence of a strong Wood's anomaly centered at 350 nm in the middle of channel 2b of SCIAMACHY, which introduces spurious polarization dependent features that interfere with the BrO absorption bands. To overcome this problem, a new UV-shifted fitting interval (336-347 nm) has been proposed for SCIAMACHY and tested for stability and consistency with GOME (more details in [8,9]). By analogy with GOME settings and due to the unavailability of solar spectra in the first releases of the SCIAMACHY Level 1b data product, our slant column retrievals use as control spectrum an earthshine radiance selected daily around equator. Further processing then follows as in [2], a mean slant column value is calculated around the equator (0°±5°) for each day and then subtracted to attenuate the noise due to SCIAMACHY radiances. Afterwards an offset correction is applied, assuming a constant equatorial BrO column of 7.5 x 10^{13} molec/cm². This value has been found to be consistent with test BrO retrievals made around the equator using daily sun reference spectra (calibrated and uncalibrated ASM references).

In the current stage of the SCIAMACHY algorithm, BrO slant columns (SCD) are converted to vertical columns (VCD) using AMFs calculated under the assumption that the tropospheric BrO content is negligible (stratospheric profile).

The GOME and SCIAMACHY BrO products (Level 2 & 3) are available on the TEMIS website (http://www.temis.nl).

3 COMPARISON WITH GOME

In Fig.1., monthly averages of BrO vertical columns derived from both instruments in March over the Arctic from 1996 until 2004 are displayed. Although color plots show obvious differences related to the different sampling and spatial resolutions of GOME and SCIAMACHY, both instruments also show excellent consistency as to the way they capture the general features of polar spring BrO emissions.

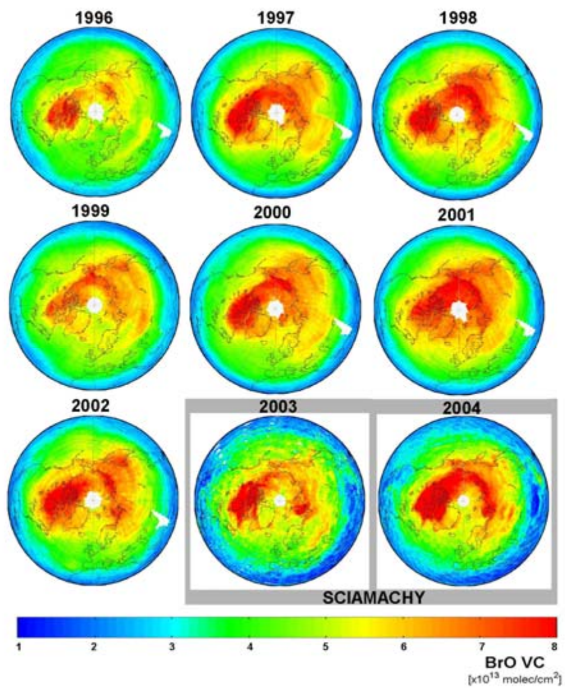

Fig. 1. Monthly averaged BrO vertical columns in March over the Northern Hemisphere, derived from GOME (1996-2002) and SCIAMACHY (2003-2004).

The consistency between GOME and SCIAMACHY BrO products has been investigated using the data in the period from July 2002 until June 2003 for which both instruments were operated simultaneously. A more refined comparison between SCIAMACHY and GOME BrO vertical columns retrievals is presented in Fig. 2. Here, the relative differences (in %) between SCIAMACHY and GOME columns (averaged on the overall data sets from January to June 2003) are displayed at the global scale. Since both instruments have different spatial resolutions, comparisons have been made between GOME VCD and the average of the VCD values of the SCIAMACHY pixels falling into the corresponding GOME pixel.

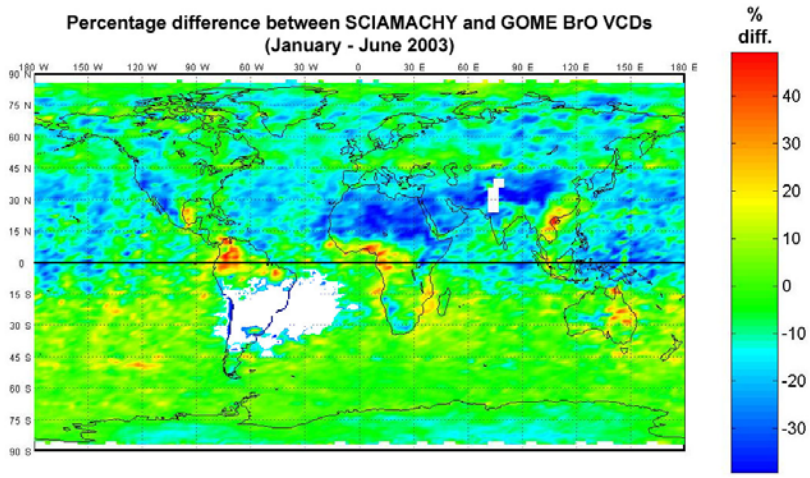

Fig. 2. Relative difference (in %) between SCIAMACHY and GOME BrO total vertical columns, averaged on the January to June 2003 measurement period.

Fig.2. reveals interesting features in the differences between SCIAMACHY and GOME BrO vertical column evaluations:

- SCIAMACHY retrievals tend to overestimate BrO columns over regions of high formaldehyde emissions. This artifact is due to a spectral interference between BrO and HCHO differential absorption features in the (non-optimal) wavelength range selected for fitting
- SCIAMACHY BrO columns are underestimated over the Sahara desert and over most of the chain of mountains (e.g. Himalaya)
- There seems to be an asymmetry between both hemispheres

The origin of the latter two problems is currently not fully understood. The positive biases observed in polluted regions like eastern China or regions affected by biomass burning like Central Africa are clearly related to a problem of spectral interference involving formaldehyde absorption. In contrast the origin of the negative bias systematically observed above desertic regions is still largely unresolved. More work is needed to better assess the overall consistency between GOME and SCIAMACHY BrO columns.

4 COMPARISON WITH GROUND-BASED MEASUREMENTS

UV-visible BrO observations have been conducted by BIRA-IASB at three ground-based stations (see Table 1), belonging to the international Network for the Detection of Stratospheric Change (NDSC). From the analysis of the measured spectra using the DOAS technique, the slant column of BrO can be retrieved during day time. In these observations, the sensitivity to stratospheric BrO is largest at twilight around 90° of solar zenith angle (SZA), while for moderate SZA, ground-based BrO slant columns are sensitive to both tropospheric and stratospheric BrO columns. In contrast SCIAMACHY nadir measurements are made around noon and are representative of total BrO columns.

Table 1. Instrumental sites

Name	Location	Viewing geometry
Reunion Island	21°S 55°E	Zenith-sky + 3°, 6°,10°,18° elev.
Observatoire de Haute-Provence	44°N 6°E	Zenith-sky + 3°, 6°,10°,18° elev.
Harestua	60°N 11°E	Zenith-sky

Exploiting the change in sensitivity of the zenith-sky measurements to the BrO vertical distribution in between noon and twilight, methods of inversion of stratospheric and tropospheric BrO vertical columns from ground-based (GB) observations have been developed.

The first retrieval technique introduced here, has been designed for multi-axis DOAS (MAX-DOAS) observations performed at the sites of Reunion Island and Observatoire de Haute-Provence (OHP). The retrieval technique [10] is basically a "2-layer" inversion method where the measured slant columns at different elevation angles (3°, 6°, 10°, 18° and zenith) are fitted to model simulations of the BrO slant column, accounting for the different sensitivity to the troposphere of the various elevation angles measurements. The radiative transfer model used for this purpose is the UVspec/DISORT package [11] which has been extended at BIRA-IASB to treat the effect of 2-dimensional inhomogeneities in the BrO concentration field due to rapid photochemical changes at twilight. The BrO inversion is indeed complicated by the fact that the retrieved parameters (stratospheric and tropospheric BrO columns) vary with solar zenith angle, since atmospheric BrO is characterized by a strong diurnal cycle. For this reason, a stacked box photochemical model (PSCBOX)[12,13] is required to reproduce the effect of the rapid variation of the BrO concentration. For Reunion Island, the diurnal variation of the stratospheric BrO columns is directly constrained by the measurements, by fitting 5 stratospheric columns (at 45°, 80°, 85°, 87.5° and 92.5° of SZA) in addition to a single tropospheric BrO column. At OHP, instead of retrieving the stratospheric BrO diurnal variation, we impose that the stratospheric BrO column varies in a way consistent with the diurnal variation given by the photochemical model. With this simplified method, the stratospheric BrO column can only be derived at a single reference SZA (fixed here at 80°). Due to the absence of measurements of the BrO diurnal variation in the troposphere, we assume that the tropospheric BrO diurnal variation can be approximated by the one provided by the photochemical model in the lowest stratospheric layer. For both stations, the measured BrO SCDs can be best reproduced by the simulations when assuming a tropospheric profile peaking around 6 km altitude, suggesting a substantial contribution from the free-troposphere to the tropospheric BrO vertical column.

For Harestua, a more advanced retrieval algorithm has been developed with the aim to provide vertical distributions of BrO. Since profiling is fundamentally an under-constrained problem, an Optimal Estimation Method (OEM)[14] has been used. The retrieval technique [15] is based on the dependence of the mean scattering height with SZA. During twilight, the mean scattering altitude scans the stratosphere rapidly, yielding height-resolved information on the absorption by stratospheric BrO. Zenith-sky observations are also sensitive to the troposphere, so GB observations enable us to extract some information about tropospheric BrO. The forward model describing the physics of the measurement is identical to the one described above. The total vertical column is calculated simply by integrating the height-resolved BrO profile.

Fig. 3, 4 & 5 show the comparisons between the calculated GB total BrO vertical columns and the coincident SCIAMACHY BrO total VCDs, respectively for Reunion Island, OHP and Harestua. The tropospheric BrO columns retrieved from ground-based observations are also displayed.

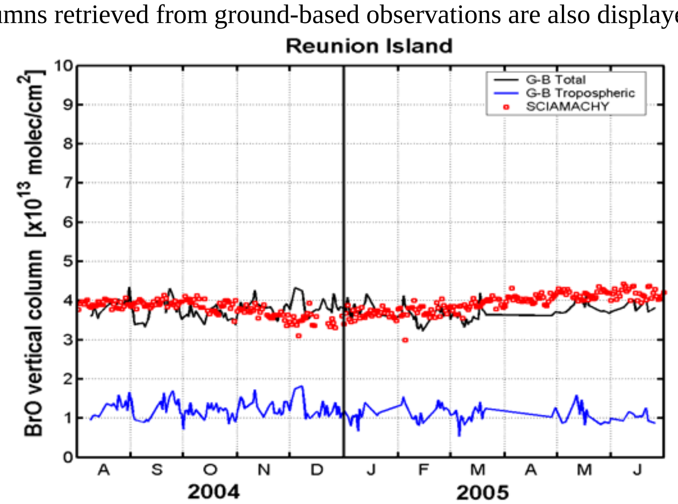

Fig.3. Total and tropospheric BrO vertical columns retrieved from ground-based DOAS observations at Reunion-Island. SCIAMACHY vertical columns are daily zonal averages (10° latitudinal band around the latitude of the station).

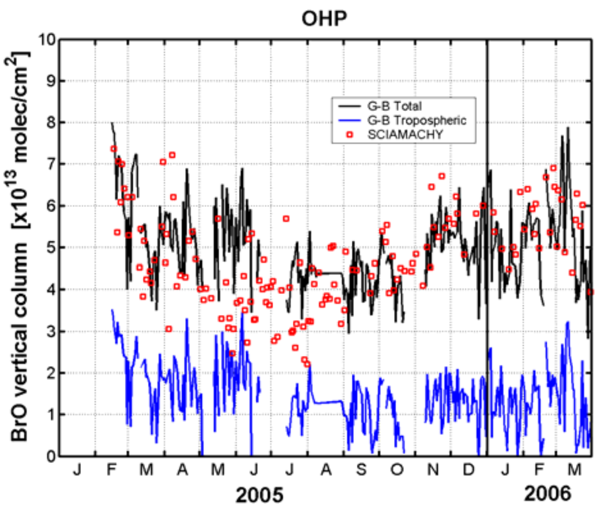

Fig.4. Total and tropospheric BrO vertical columns retrieved from ground-based DOAS observations at Observatoire de Haute-Provence (Preliminary results). SCIAMACHY vertical columns are daily averages of all pixels falling within a radius of 200 km around the station.

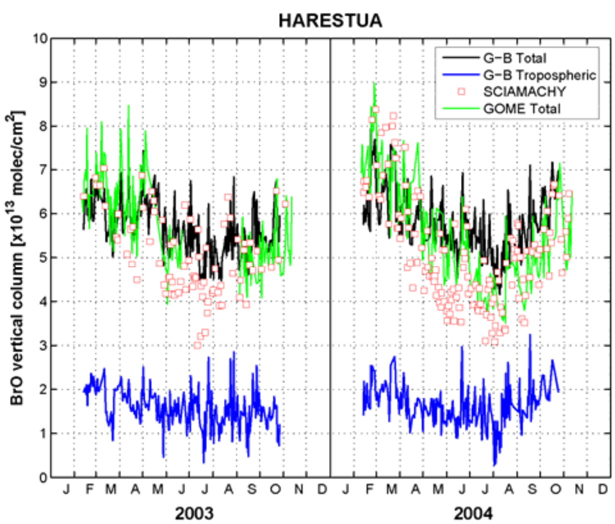

Fig.5. Total and tropospheric BrO vertical columns retrieved from ground-based DOAS observations at Harestua. SCIAMACHY and GOME vertical columns are daily averages of all pixels falling within a radius of 200 km around the station.

Since satellite nadir total BrO columns are made at high sun, the GB retrieved total columns must be evaluated adequately at the SZA of the satellite overpasses. A photochemical correction is thus applied to the GB vertical columns. For validation purposes (and in contrast to the more basic product delivered on the TEMIS web-site), satellite VCDs have been estimated by applying total AMFs (which account for a tropospheric BrO contribution derived from the ground-based measurements) to the SCIAMACHY SCDs.

In general, SCIAMACHY BrO vertical columns are found to be in good agreement with the total BrO VCDs inferred from the ground-based observations for the three stations. At Reunion Island, no noticeable seasonal variation is found but the mean level of total BrO VCDs from SCIAMACHY and GB evaluations are consistent. At OHP and Harestua, the seasonal variations (and even short-term variations) of the total BrO VCDs are captured in a similar way by SCIAMACHY and ground-based instruments. It has to be noticed however that systematic discrepancies tend to appear in summer conditions when both satellite and GB retrievals have their largest uncertainties. More work is needed to better characterize the origin of these summer discrepancies.

Besides validation results, Figs. 3 to 5 also indicate that both satellites and ground-based observations are consistent with the presence of a tropospheric BrO layer of approximately 1-3 $x10^{13}$ molec/cm^2 in column (corresponding to mixing ratios in the range from 1 to 3 pptv is well mixed in the free troposphere). This finding is in agreement with estimates from recent studies published in the literature [16,17]. In Harestua, large and highly variable tropospheric BrO VCDs are observed in late winter-early spring, suggesting possible transport from polar sources.

5 CONCLUSIONS

An algorithm to retrieve total BrO columns from SCIAMACHY nadir data has been developed. SCIAMACHY and GOME BrO total VCDs were found to be in good agreement, despite some systematic features in SCIAMACHY data, in particular a strong interference with formaldehyde. SCIAMACHY BrO total VCDs are also in good agreement with ground-based measurements performed at three stations sampling tropical, mid- and high-latitudes. However, larger differences seem to be observed during summer time when both satellite and GB retrievals have largest uncertainties. Satellite and ground-based data are consistent with the presence of a tropospheric BrO column of 1-3 $x10^{13}$ $molec/cm^2$ at all latitudes.

6 REFERENCES

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