BrO PROFILING FROM GROUND-BASED DOAS OBSERVATIONS: NEW TOOL FOR THE ENVISAT/SCIAMACHY VALIDATION

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ABSTRACT

Time-series of BrO vertical profiles and corresponding total columns retrieved from ground-based zenith-sky DOAS observations at Harestua (60°N, 10°E) are compared to ENVISAT/SCIAMACHY limb and nadir and GOME scientific products. Retrieval algorithms from the following groups are involved in the comparison exercise: IUP-Bremen, IUP-Heidelberg, and Harvard Smithsonian Center for Astrophysics for SCIAMACHY limb BrO profiles, BIRA-IASB and IUP-Bremen for SCIAMACHY nadir BrO total columns, and BIRA-IASB for GOME BrO total columns. Comparisons between ground-based UV-visible and SCIAMACHY and GOME data cover the 2002-2004 period and are performed in the same photochemical conditions since the BIRA-IASB profiling algorithm includes a stacked box photochemical model, enabling the retrieval of BrO profiles at any solar zenith angle.

1. INTRODUCTION

The profiling technique applied to ground-based DOAS (Differential Optical Absorption Spectroscopy) measurements offers new perspectives in the use of ground-based UV-visible networks such as the NDACC (Network for the Detection of Atmospheric Composition Change, formerly NDSC). With this technique, based on the dependence of mean scattering height with solar zenith angle (SZA), not only columns but also low resolution vertical profiles are made available for the purpose of satellite data validation. So far, profiling from ground-based UV-visible observations has been mainly used for the retrieval of stratospheric NO_2 profiles (e.g., [1,2,3]). Very recently, [4] have applied this technique to combined zenith-sky and direct sun UV-visible observations in order to infer time-series of tropospheric and stratospheric BrO vertical column densities (VCDs) from the retrieved profiles.

At BIRA-IASB, a profiling algorithm has been developed in order to retrieve stratospheric but also tropospheric BrO profiles from ground-based zenith-sky UV-visible observations. BrO profiling, especially the ability through this technique to differentiate stratospheric and tropospheric contributions, is important for a least two reasons: (1) There are still open questions on bromine compounds in the atmosphere, mainly concerning the bromine loading in the stratosphere and the presence of BrO in the free troposphere; (2) There are few correlative BrO observations, especially profiles, available for the validation of satellite experiments. So far, the BIRA-IASB algorithm has been applied to ground-based DOAS observations of BrO performed at the NDACC station of Harestua in Southern Norway (60°N, 10°E) during the 2002-2004 period. In the present study, the time-series of retrieved profiles and corresponding columns are compared to the following ENVISAT/SCIAMACHY limb and nadir and GOME scientific products: SCIAMACHY limb BrO profiles from IUP-Bremen, IUP-Heidelberg, and Harvard Smithsonian Center for Astrophysics (SAO), SCIAMACHY nadir BrO total columns from BIRA-IASB and IUP-Bremen, and GOME BrO total columns from BIRA-IASB.

2. GROUND-BASED UV-VISIBLE OBSERVATIONS

Ground-based zenith-sky UV-visible observations have been continuously performed by BIRA-IASB at the Harestua station since 1998. The instrument and the data analysis have been validated through several NDACC campaigns (Lauder 1992, OHP 1996, and Andøya 2003). In case of BrO, the DOAS fitting window used is 345-359 nm and the BrO cross-sections set is from [5]. The DOAS analysis is performed using a noon spectrum in summer condition as reference and the residual amount in the reference spectrum is calculated using a modified Langley-plot analysis taking into account the diurnal variation of stratospheric BrO. The application of the profiling algorithm to the resulting absolute slant column densities (SCDs) makes the retrieval sensitive to the troposphere.

3. BIRA-IASB BRO PROLIFING ALGORITHM

The profiling algorithm used in the present study is the BIRA-IASB NO_2 profiling algorithm [3] adapted to BrO. It is based on the Rodgers Optimal Estimation method (OEM; [6]) and the forward model consists in the radiative transfer model UVspec/DISORT [7] coupled to the BIRA-IASB stacked box photochemical model PSCBOX. Both models have been validated through intercomparison exercises ([8, 9]). The inclusion of a photochemical model in the retrieval algorithm, reproducing the effect of the rapid variation of the BrO or NO_2 concentration, makes possible profile retrieval at any SZA. An important step in the OEM method is the choice of the a priori profile. In the stratosphere, the a priori BrO profile is the output of the photochemical model PSCBOX daily initialized with SLIMCAT 3D-CTM chemical and meteorological fields [10]. In the troposphere, a constant BrO concentration of about $1.0x10^7$ molec/cm^3 is used. The diurnal variation of the tropospheric profile is similar to the one given by the photochemical model in the lower stratosphere.

The averaging kernels (AVKs) matrix **A** is the key parameter in the characterization of the retrieval information content. The shape of the AVKs can give a rough estimate of the vertical resolution and the trace of the AVKs matrix **A** gives the number of independent pieces of information contained in the measurements. A typical example of AVKs, obtained for the sunrise 12/07/2004 retrieval, is shown in Fig. 1. From the examination of these AVKs, it is found that the vertical resolution is 10 km at best and a significant sensitivity to the troposphere is obtained. The value of the trace of **A** is 3.5, so there are about 3 independent pieces of information in the measurements.

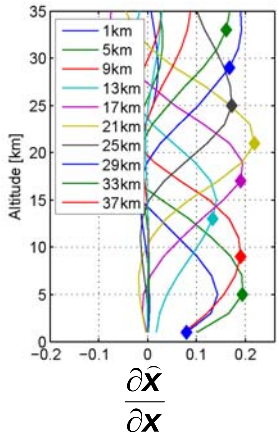

Fig. 1: Typical example of ground-based BrO averaging kernels. They are calculated for the sunrise Harestua 12/07/2004 retrieval. Plain diamonds indicate the altitude at which each averaging kernel should peak in an ideal case.

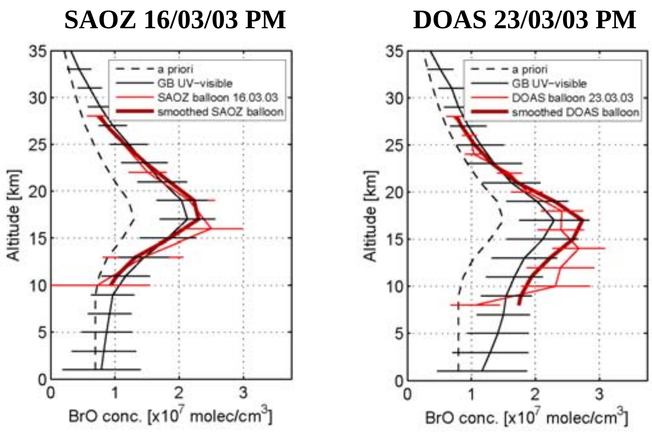

Fig. 2: Comparisons between ground-based UV-visible and SAOZ and DOAS ascent balloon BrO profiles. Both balloon flights originated in Kiruna (68°N, 21°E). Ground-based profiles have been retrieved at about 85° (SAOZ comparison) and 80° (DOAS comparison) SZA. Due to the higher vertical resolution of the balloon BrO profiles, they have been smoothed by convolution with ground-based UV-visible AVKs to allow direct comparison with ground-based BrO profiles. These comparisons also show also the sensitivity of the ground-based retrievals to the troposphere (see [11] for time-series of tropospheric BrO VCDs at Harestua).

It should be noted that in case of NO_2, the BIRA-IASB profiling algorithm has been extensively validated through comparisons with correlative balloon (SAOZ and DOAS) and satellite data (HALOE and POAM III) [3]. In case of BrO, preliminary comparisons with SAOZ and DOAS balloon profiles show a good overall agreement as it can be seen in Fig. 2 (see [12] for a description of the SAOZ and DOAS balloon BrO profiles).

4. COMPARISON WITH SCIAMACHY LIMB BrO PROFILES

These comparisons are part of the BOOST (Bromine Oxide in the lOwer STratosphere) project [13] of which the objectives are to investigate possible reasons for the disagreement between the vertical distributions of BrO retrieved from the SCIAMACHY limb measurements and subsequently improve existing retrieval algorithms. For this purpose, several comparison tests between SCIAMACHY limb BrO profiles and correlative balloon and ground-based data have been started. The three involved SCIAMACHY limb retrieval groups are IUP-Bremen, SAO, and IUP-Heidelberg. The characteristics of each algorithm can be found on the BOOST web site [13]. In a preliminary step, four comparison cases between ground-based UV-visible BrO profiles at Harestua and coincident SCIAMACHY limb profiles have been selected. These are: 23/08/2002, 16/04/2004, 03/06/2004, and 12/07/2004. Due to the latitude of Harestua (60°N), the vortex conditions corresponding to ground-based and SCIAMACHY measurements on 16/04/2004 had to be checked. Fig. 3 shows that no strong PV gradient was present around the Harestua station on this day and that both ground-based and SCIAMACHY measurements have been performed in the same PV conditions.

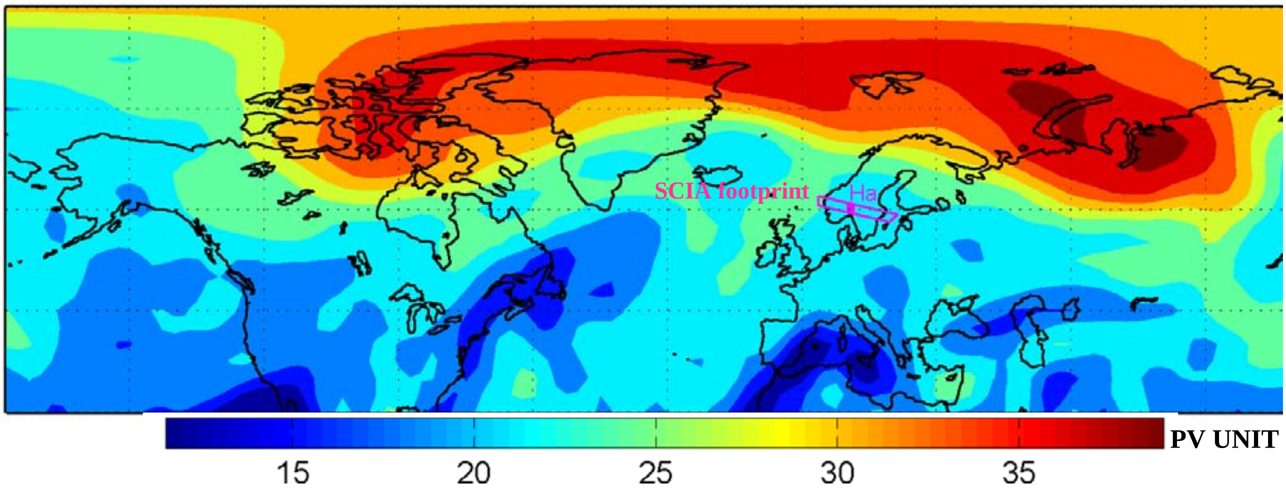

Fig. 3: Potential Vorticity (PV) map at 475 K for 16/04/2004.

Comparison results for the four selected cases are shown in Fig. 4. It should be noted that the ground-based UV-visible BrO profiles have been retrieved at the SZA of the SCIAMACHY tangent point. Concerning the IUP-Bremen retrievals, except for the 03/06/2004 case, good agreement with ground-based profiles is found between 13 and 23 km of altitude whereas SCIAMACHY gives generally lower BrO concentration values below and above this altitude range. SAO profiles are systematically higher than the ground-based profiles below 21-23 km of altitude and lower above this altitude. However, below 21-23 km, ground-based BrO profiles are generally within the SAO SCIAMACHY error bars. In case of IUP-Heidelberg retrievals, a good overall agreement is obtained with ground-based profiles. IUP-Heidelberg SCIAMACHY algorithm also gives larger BrO concentration above 25 km of altitude than the two other algorithms, in better agreement with ground-based retrievals. However, comparisons with balloon profiles made by the University of Bremen [13] show that IUP-Bremen and SAO retrievals agree quite well with balloon data above 25 km, suggesting therefore a possible overestimation of BrO by ground-based UV-visible and IUP-Heidelberg retrievals in this altitude range. Possible origins for this behaviour (e.g., uncertainty on the photochemical correction applied to ground-based BrO profiles in order to match with SCIAMACHY tangent point SZA, which is ranging between 40 and 55° for the four selected coincidences) are currently being investigated. These results have also to be confirmed through a larger number of coincident events between ground-based UV-visible and SCIAMACHY limb measurements.

5. COMPARISON WITH SCIAMACHY NADIR BrO TOTAL COLUMNS

Three different SCIAMACHY nadir BrO VCD evaluations were compared to ground-based BrO VCDs at Harestua calculated by integrating the BrO profiles retrieved at the SCIAMACHY overpass time. These are the following: BIRA-IASB SCIAMACHY evaluation using a daily earthshine radiance (equatorial off-set: 7.5×10^{13} molec/cm^2) and IUP-Bremen [14] and BIRA-IASB evaluations using daily uncalibrated ASM solar spectra and similar settings for the DOAS analysis (e.g., same wavelength range (336-347 nm) and BrO cross sections set from [5]). BIRA-IASB GOME retrieval [15] using a daily earthshine radiance (equatorial off-set: 7.5×10^{13} molec/cm^2) was also included in the comparison. It should be noted that in both SCIAMACHY and GOME DOAS analyses, the equatorial off-set acts as a free parameter and the value of 7.5×10^{13} molec/cm^2 was estimated from ground-based DOAS observations performed in the tropics ([11]). Concerning the nadir AMFs, they were calculated using a tropospheric contribution consistent with ground-based profiling estimation.

Fig. 5 shows ground-based and all SCIAMACHY and GOME time-series of BrO VCDs at Harestua. Quite good agreement is obtained between BIRA-IASB and IUP-Bremen SCIAMACHY evaluations using daily uncalibrated ASM solar spectra, the IUP-Bremen columns being somewhat lower. Both evaluations generally give smaller BrO VCD values than ground-based observations, the largest discrepancies being observed in summer and fall with mean relative difference values between –12 and –24% (see Fig. 6).

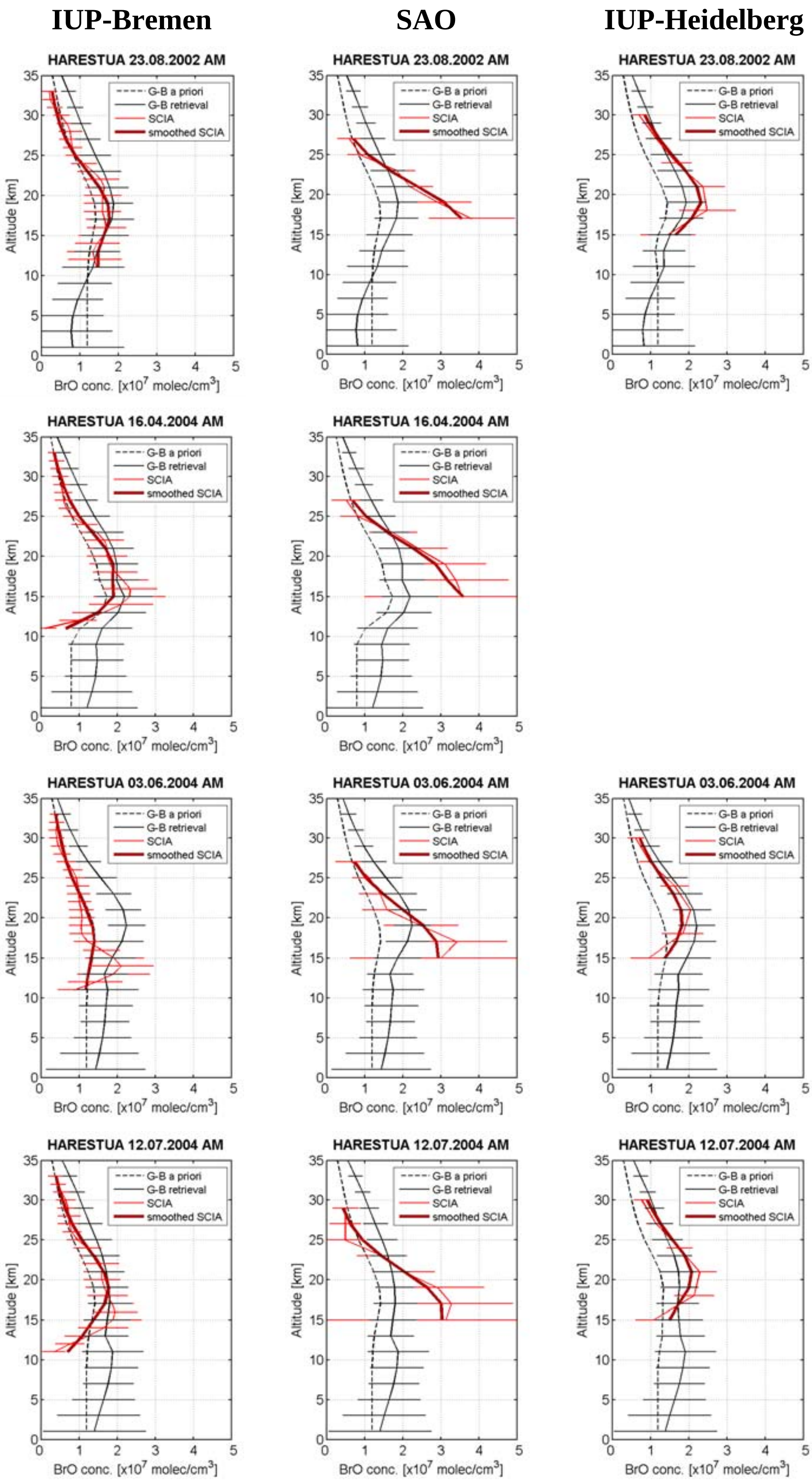

Fig. 4: Comparison between ground-based UV-visible and SCIAMACHY limb BrO profiles at Harestua for four selected coincidences. The involved SCIAMACHY retrieval algorithms are: IUP-Bremen, SAO, and IUP-Heidelberg. Due to the higher vertical resolution of the SCIAMACHY BrO profiles, they have been smoothed by convolution with ground-based UV-visible AVKs to allow direct comparison with ground-based profiles.

SCIAMACHY BrO VCDs evaluated using a daily earthshine radiance show a good agreement with ground-based data in spring and fall (mean relative difference smaller than 11% in absolute value) but SCIAMACHY underestimates ground-based BrO VCDs in summer (mean relative difference values between –16 and –23%). In case of comparison with GOME, good agreement is found with ground-based BrO VCDs, with relative difference smaller than 12% in absolute value (see Fig. 6). It should be noted that as for all SCIAMACHY evaluations, GOME displays smaller BrO VCD values than ground-based observations in summer. Investigations are currently under progress in order to determine whether the origin of this systematic behaviour is to be found in the satellite or ground-based data sets. For this purpose but also to study the long-term evolution of the consistency between ground-based, SCIAMACHY, and GOME data sets, the time-series of ground-based BrO VCDs will be extend to years before 2002 and after 2004.

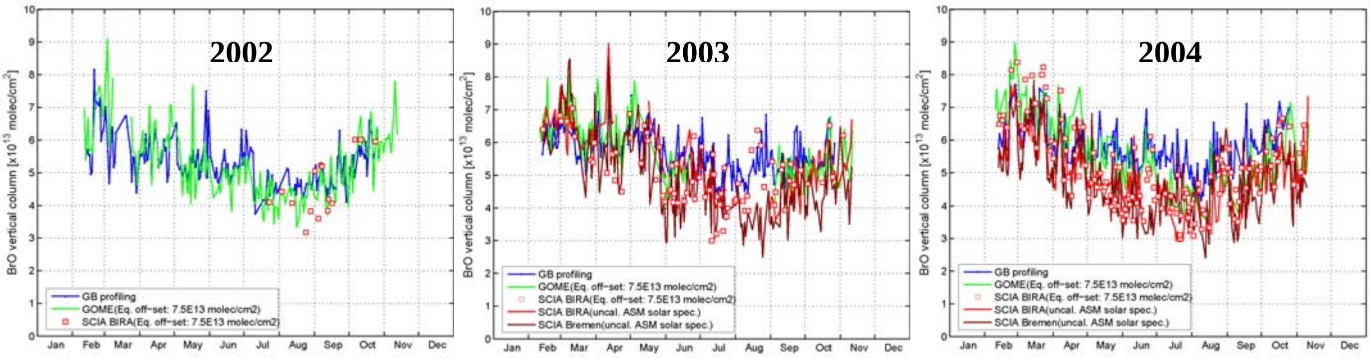

Fig. 5: ground-based UV-visible, SCIAMACHY, and GOME BrO VCD time-series at Harestua for the 2002-2004 period.

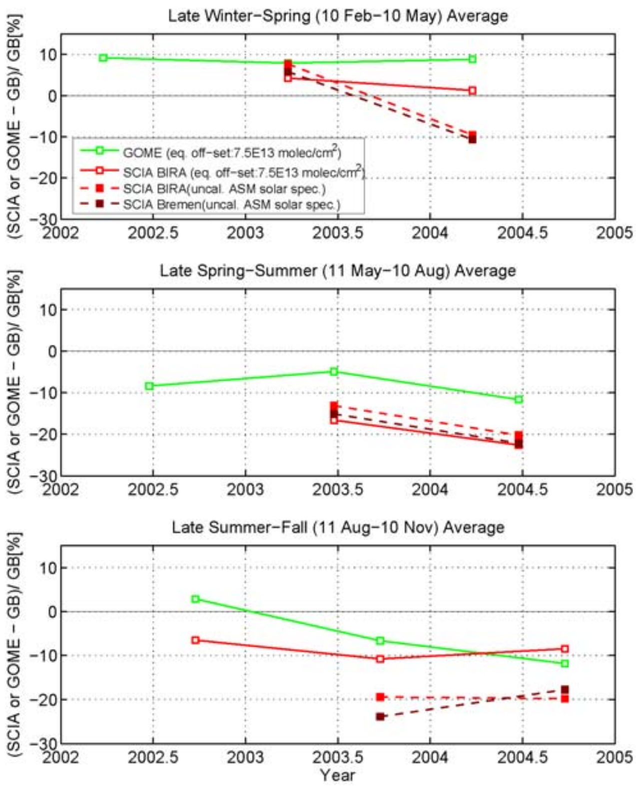

Fig. 6: Differences between SCIAMACHY or GOME and ground-based BrO VCDs (reference) averaged for three periods: late winter-early spring (10 February-10 May), spring-summer (11 May-10 August), and late summer-fall (11 August-10 November).

6. CONCLUSIONS

BrO vertical profiles have been retrieved from ground-based zenith-sky UV-visible observations at Harestua (60°N, 10°E) for the 2002-2004 period. These profiles and their corresponding integrated total columns have been compared to SCIAMACHY limb and nadir and GOME BrO scientific products from IUP-Bremen, BIRA-IASB, IUP-Heidelberg, and SAO. Concerning the comparison with SCIAMACHY limb BrO profiles, a good overall agreement is generally obtained between IUP-Bremen and IUP-Heidelberg retrievals and ground-based UV-visible profiles between 13 and 23 km. Above this altitude, good agreement persists with IUP-Heidelberg whereas IUP-Bremen profiles are lower than ground-based ones. Concerning the SAO retrievals, they give systematically larger BrO concentration values than the ground-based profiles below 21-23km of altitude. However, the ground-based BrO profiles are generally within the SAO SCIAMACHY errors bars. Above 25 km, SAO SCIAMACHY profiles are lower than ground-based retrievals. Since a rather good agreement is found between IUP-Bremen and SAO retrievals and balloon BrO profiles above 25 km, these comparison results suggest a possible overestimation by ground-based UV-visible and IUP-Heidelberg retrievals in this altitude range. This behaviour is currently under investigation. In case of comparison with SCIAMACHY nadir and GOME BrO total columns, quite good agreement is found between BIRA-IASB and IUP-Bremen SCIAMACHY evaluations using uncalibrated ASM solar spectra. However, best agreement with ground-based BrO columns is generally obtained with SCIAMACHY but also GOME evaluations using a daily earthshine radiance with an equatorial off-set of $7.5x10^{13}$ molec/cm^2. In summer, GOME and all SCIAMACHY evaluations give systematically lower BrO total column values than ground-based observations, requiring further investigation. In the near future, the number of comparison cases between ground-based and SCIAMACHY limb BrO profiles will be increased and the ground-based BrO column time-series will be extended to years before 2002 and after 2004 in order to study the long-term evolution of the consistency between ground-based, SCIAMACHY, and GOME data sets.

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