OMI total bromine monoxide (OMBRO) data product: Algorithm, retrieval and measurement comparisons

Raid M. Suleiman¹, Kelly Chance¹, Xiong Liu¹, Gonzalo González Abad¹, Thomas P. Kurosu², Francois Hendrick³, and Nicolas Theys³

¹Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, USA
²Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, USA
³Royal Belgian Institute for Space Aeronomy, Brussels, Belgium

Correspondence to: Raid M. Suleiman (rsuleiman@cfa.harvard.edu)

Abstract. This paper presents the retrieval algorithm for the operational Ozone Monitoring Instrument (OMI) total bromine monoxide (BrO) data product (OMBRO) developed at the Smithsonian Astrophysical Observatory (SAO), and shows comparisons with correlative measurements and retrieval results. The algorithm is based on direct nonlinear least squares fitting of radiances from the spectral range 319.0-347.5 nm. Radiances are modeled from the solar irradiance, attenuated by contributions from BrO and interfering gases, and including rotational Raman scattering, additive and multiplicative closure polynomials, correction for Nyquist undersampling, and the average fitting residual spectrum. The retrieval uses albedo and wavelength-dependent air mass factors (AMFs), which have been pre-computed using a single mostly stratospheric BrO profile. The BrO cross sections are multiplied by the wavelength-dependent AMFs before fitting so that the vertical column densities (VCDs) are retrieved directly. The fitting uncertainties of BrO VCDs typically vary between 4 and 7×10¹² molecules cm⁻² (~10-20% of the measured BrO VCDs). The retrievals agree well with GOME-2 observations at simultaneous nadir overpasses and ground-based zenith-sky measurements at Harestua, Norway, with mean biases less than -0.216±1.13×10¹³ molecules cm⁻² and 0.12±0.76×10¹³ molecules cm⁻², respectively. Global distribution and seasonal variation of OMI BrO are generally consistent with previous satellite observations. Global distribution of BrO from OMBRO shows spatial and temporal patterns similar to GOME-2 retrievals. The OMBRO retrievals show enhancement of BrO over U.S. Great Salt Lake. It also shows significant BrO enhancement from the eruption of the Eyjafjallajökull volcano, although the BrO retrievals are affected under high SO₂ loading conditions by the sub-optimum choice of SO₂ cross sections.
1 Introduction

Bromine monoxide (BrO) is a halogen oxide, predominantly located in the stratosphere and upper troposphere where, like chlorine monoxide (ClO), it is a catalytic element in the destruction of stratospheric ozone (von Glasow et al., 2004; Salawitch et al., 2005), but with higher efficiency per molecule. Sources of tropospheric BrO include bromine release (“explosions”) during the Polar Spring (Hausmann and Platt, 1994; Hollwedel et al., 2004; Simpson et al., 2007; Begoin et al., 2010; Salawitch et al., 2010; Abbatt, et al., 2012; Blechschmidt et al., 2016), volcanic eruptions (Bobrowski et al., 2003; Chance, 2006; Theys et al., 2009), salt lakes (Hebestreit, et al., 1999; Hörmann et al., 2016), and stratospheric transport (Salawitch et al., 2010). Global BrO measurements from space were first proposed for the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) instrument (Chance et al., 1991) and were first demonstrated with Global Ozone Monitoring Experiment (GOME-1) measurements (Chance, 1998; Platt and Wagner, 1998; Richter et al., 1998), and since with SCIAMACHY nadir (Kühl et al., 2008) and Global Ozone Monitoring Experiment 2 (GOME-2) measurements (Theys et al., 2011; Toyota et al., 2011). Initial observations of BrO by OMI were first reported by Kurosu et al. (2004). Polar Spring BrO enhancements are known to be associated with boundary layer ozone depletion (Hausmann and Platt, 1994; von Glasow et al., 2004; Salawitch et al., 2005; Simpson et al., 2007; Salawitch et al., 2010; Abbatt, et al., 2012). OMI measurements of BrO have been used together with chemical and dynamical modeling to investigate stratospheric versus tropospheric enhancements of atmospheric BrO at high northern latitudes (Salawitch et al., 2010). OMI BrO retrieval using the Differential Optical Absorption Spectroscopy (DOAS) method has been used to study the seasonal variations of tropospheric bromine monoxide over the Rann of Kutch salt marsh (Hörmann et al., 2016). The Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) campaign (Choi et al., 2012) found consistency between BrO column densities calculated from Chemical Ionization Mass Spectrometer (CIMS) measurements with the tropospheric BrO columns derived from OMI using our operational retrieval algorithm. BrO has been observed from the ground in Harestua, Norway (Hendrick et al., 2007), Lauder, New Zealand (Schofield et al., 2004a, 2004b), Antarctica (Schofield et al., 2006), Utqiagvik (Barrow), Alaska (Liao et al., 2011; Frieß et al., 2011; Liao et al., 2012; Sihler et al., 2012; Peterson et al., 2016), Eureka, Canada (Zhao et al., 2015), Summit, Greenland (Stutz et al., 2011) and the Arctic Ocean (Burd et al., 2017).
Enhancement of BrO in the vicinity of salt lakes like the Dead Sea and the Great Salt Lake have been observed from ground-based measurements (Hebestreit et al., 1999; Matveev et al., 2001; Stutz et al., 2002; Tas et al., 2005; Holla et al., 2015). The active bromine compound release is due to the reaction between atmospheric oxidants with salt reservoirs. Satellite observation of salt lake BrO was first reported over the Great Salt Lake and the Dead Sea from OMI (Chance, 2006; Hörmann et al. 2016). Seasonal variations of tropospheric BrO over the Rann of Kutch salt marsh have been observed using OMI from an independent research BrO product (Hörmann et al. 2016). Bobrowski et al. (2003) made the first ground-based observations of BrO and SO$_2$ abundances in the plume of the Soufrière Hills volcano (Montserrat) by multi-axis DOAS (MAX-DOAS). BrO and SO$_2$ abundances as functions of the distance from the source were measured by MAX-DOAS in the volcanic plumes of Mt. Etna in Sicily, Italy and Villarica in Chile (Bobrowski et al., 2007). The BrO/SO$_2$ ratio in the plume of Nyiragongo and Etna was also studied (Bobrowski et al., 2015). The first volcanic BrO measured from space was from the Ambrym volcano, measured by OMI (Chance, 2006). Theys et al. (2009) reported on GOME-2 detection of volcanic BrO emission after the Kasatochi eruption. Hörmann et al. (2013) examined GOME-2 observations of BrO slant column densities (SCDs) in the vicinity of volcanic plumes; it showed clear enhancements of BrO in ~1/4 of the volcanos, and revealed large spatial differences in BrO/SO$_2$ ratios.

The purpose of this paper is to describe the OMI BrO operational algorithm and the data product, compare it with ground-based and other satellite measurements and analyze its spatiotemporal characteristics. This paper is organized as follows: Section 2 describes the OMI instrument and the data product. Section 3 gives a detailed description of the operational algorithm including algorithm and product history, spectral fitting, AMF calculations, destriping, and fitting uncertainties. Section 4 presents results and discussion including comparison with GOME-2 and ground-based zenith-sky measurements at Harestua, Norway, global distribution, seasonality, enhanced BrO from the U.S. Great Salt Lake and Iceland’s Eyjafjallajökull volcano. Section 5 concludes this study.
2 OMI instrument and OMBRO data product

2.1 OMI instrument

OMI instrument and OMBRO data product

OMI was launched on the NASA Earth Observing System (EOS) Aura satellite into a sun-synchronous orbit on 15 July 2004. It is a push-broom imaging spectrometer that observes solar backscattered radiation in the visible and ultraviolet from 270-500 nm in three channels (UV1: 270-310 nm, UV2: 310-365 nm, visible: 350-500 nm) at spectral resolution of 0.42-0.63 nm and spatial resolution in the normal (global sampling) mode ranging from 13×24 km$^2$ at direct nadir to about 28×150 km$^2$ at the swath edges. The global mode (GM) has 60 ground pixels with a total cross-track swath of 2600 km. There are also spatial and spectral zoom modes with twice finer across-track spatial resolution at nadir. The spatial zoom mode (SZM) is employed every 32 days (Levitt et al., 2006): data from this mode are spatially rebinned to global-mode sampling sizes, known as the rebinned spatial zoom mode. The SZM, like the global mode (GM), has 60 cross-track pixels. These are re-binned to 30 pixels, to form “the rebinned spatial zoom mode” (RSZM) which is equivalent in pixel size to the GM data, but with reduced spatial coverage.

Since June 2007, certain cross-track positions of OMI data have been affected by the row anomaly (http://projects.knmi.nl/omi/research/product/rowanomaly-background.php): some loose thermal insulating material likely appeared in front of the instrument’s entrance slit, which can block and scatter the light thus causing errors in level 1b data and subsequently the level 2 retrievals (Kroon et al., 2011). Initially, the row anomaly only affected a few positions and the effect was small. But since January 2009, the anomaly has become more serious, spreading to ~1/3 of the positions and retrievals at those positions are not recommended for scientific use. A flagging field has been introduced in the OMI level 1b data to indicate whether an OMI pixel is affected by this instrument anomaly.

OMI measures ozone and other trace gases, aerosols, clouds, and surface properties. Products developed at the SAO include operational BrO, chlorine dioxide (OClO), and formaldehyde (H$_2$CO; González Abad et al., 2015) that are archived at NASA Goddard Earth Sciences (GES) Data and Information Services Center (DISC), and offline (“pre-operational”) ozone profile and tropospheric ozone (O$_3$) (Liu et al., 2010; Huang et al., 2017, 2018), glyoxal (C$_2$H$_2$O$_2$) (Chan Miller et al., 2014, 2016) and water vapor (H$_2$O) (Wang et al., 2014, 2016) that are available at
the Aura validation data center (AVDC). All the products except for the ozone profile product are produced using nonlinear least-squares (NLLS) fitting methods based on those previously developed at the SAO for the analysis of measurements from the GOME (now GOME-1) (Chance, 1998; Chance, et al., 2000) and SCIAMACHY instruments (Burrows and Chance, 1991; Chance et al., 1991; Martin et al., 2006).

2.2 OMBRO data product

The current operational BrO product, OMBRO version 3, contains BrO vertical column densities (VCDs), slant column densities (SCDs), effective air mass factors (AMFs) and ancillary information retrieved from calibrated radiance and irradiance spectra in OMI GM and RSZM level 1b data product. Each BrO product file contains a single orbit of data, from pole to pole, for the sunlit portion of the orbit. The data product from 26 August 2004 through the present is available at GES DISC. Data used in this study cover the period from 1 January 2005 to 31 December 2014.

3 Retrieval algorithm

3.1 Algorithm and product history

OMBRO Version 1.0 was released on 1 February 2007, based on a spectral fitting window of 338–357 nm. Version 2.0 was released on 13 April 2008. It included major adjustments for Collection 3 Level 1b data, improved de-striping measures, change of the fitting window to 340–357.5 nm, improvements to radiance wavelength calibration, and several improvements for processing near-real-time data. In both Versions 1 and 2, total BrO VCDs were retrieved in two steps: first performing spectral fitting using the basic optical absorption spectroscopy (BOAS) method to derive SCDs from OMI radiance spectra, and then converting from SCDs to VCDs by dividing AMFs. This is similar to current SAO H₂CO, H₂O and C₂H₂O₂ as mentioned previously. The latest Version 3.0.5, released on 28 April 2011, includes major algorithm changes: the fitting window was moved to 319.0–347.5 nm, and BrO cross sections are multiplied by wavelength-dependent AMFs, which are a function of albedo, before fitting, for a direct retrieval of BrO VCDs. SCDs are similarly retrieved in a separate step by fitting BrO cross sections that have not been multiplied with wavelength-dependent AMFs, and an effective AMF = SCD/VCD is computed. Diagnostic
cloud information from the OMCLDO2 product (Acarreta et al., 2004) was added, and the row-anomaly indicating flags were carried over from the level 1b product.

The current algorithm is described in detail in the rest of this section, with spectral fitting in Section 3.2, AMF calculation prior to spectral fitting in Section 3.3, post-processing de-stripping to remove cross-track dependent biases in Section 3.4, and fitting uncertainties and error estimates in Section 3.5.

3.2 Spectral fitting

Most aspects of the algorithm physics for the direct fitting of radiances by the BOAS method were developed previously at SAO for analysis of GOME and SCIAMACHY satellite spectra (Chance, 1998, Chance et al., 2000, OMI, 2002; Martin et al., 2006) and in the various algorithm descriptions of other SAO OMI products (Wang et al., 2014; Chan Miller et al., 2014; Gonzalez Abad et al., 2015). Unlike the often-used DOAS fitting method (Platt, 1994), radiances are not ratioed to irradiances, logarithms are not taken, and no high-pass filtering is applied.

The spectral fitting in the SAO OMI BrO retrieval is based on a Gauss-Newton NLLS fitting procedure, the CERN ELSUNC procedure (Lindström and Wedin, 1987), which provides for bounded NLLS fitting. Processing begins with wavelength calibration for both irradiance and radiance. In each case the wavelength registration for the selected fitting window is determined independently for each cross-track position by cross-correlation of OMI spectra with a high spectral resolution solar irradiance (Caspar and Chance, 1997; Chance, 1998; Chance and Kurucz, 2010) using the preflight instrument slit functions (Dirksen et al., 2006). To improve cross-track stripe correction (Section 3.4) and reduce the noise in the solar irradiance data, the OMI irradiance spectra are composites derived from a principal component analysis of three years of individual OMI irradiance measurements (2005-2007). Radiance wavelength calibration is performed for a representative scan line of radiance measurements (usually in the middle of the orbit) to determine a common wavelength grid for reference spectra.

Following wavelength correction, an undersampling correction spectrum is computed to partially correct for spectral undersampling (lack of Nyquist sampling: Chance, 1998; Slijkhuis et al., 1999;
Chance et al., 2005). The calculation of the corrections for the undersampling is accomplished by convolving the preflight slit functions with the high-resolution solar spectrum and differencing its fully-sampled and undersampled representations (Chance et al., 2005).

Fitting is then performed for all scan lines in the OMI swath granule. In each stage, the fitting is performed individually for the 60 cross-track pixels of a block of 100 OMI across-track swath lines along the flight direction (30 cross-track pixels for the RSZM) according to Eq. (1):

\[
I = (aI_0 + \sum \alpha_i A_i)e^{-\sum \beta_j B_j + \sum \gamma_k C_k}Poly_{scale} + Poly_{baseline},
\]

where \(I_0\) is the solar irradiance (used in our operational BrO retrieval) or radiance reference measurement, \(I\) is the Earthshine radiance (detected at satellite), \(a\) is albedo, \(\alpha\), \(\beta\), \(\gamma\), are the coefficients to the reference spectra of \(A_i\), \(B_j\), \(C_k\), (for example, trace gas cross sections, Ring effect, vibrational Raman, undersampling correction, common mode, etc.) of model constituents. The reference spectra are derived separately for each cross-track position from original high-resolution cross sections convolved with the corresponding OMI slit functions after correcting for the solar \(I_0\) effect (Aliwell et al., 2002). Fig. 1 shows the trace gas cross sections and Ring spectra used in the current operational algorithm. The black lines are the original high-resolution reference spectra, and the color lines show the corresponding spectra convolved with OMI slit function, which are used in the fitting.

For improved numerical stability, radiances and irradiances are divided by their respective averages over the fitting window, renormalizing them to values of ~1. BrO is fitted in the spectral window 319.0–347.5 nm, within the UV-2 channel of the OMI instrument. The switch from the previous fitting window of 340–357.5 nm to this shorter and wider fitting window is based on extensive sensitivity analysis following the method described by Vogel et al., 2013. This new fitting window aims at reducing the fitting uncertainty by including more BrO spectral structures as shown in Fig. 1 and reducing retrieval noise while preserving the stability of the algorithm. An analysis of the retrieval sensitivity to different windows is included in section 3.5.
The rotational Raman scattering (Chance and Spurr, 1997; Chance and Kurucz, 2010) and undersampling correction spectra, $A_i$, are first added to the albedo-adjusted solar irradiance $a I_0$, with coefficients $\alpha$ as shown in Eq. 1. Radiance $I$ are then modeled as the this quantity attenuated by absorption from BrO, O3, NO2, H2CO, and SO2 with coefficients $\beta_j$ fitted to the reference spectra $B_j$ as shown in Eq. 1. A common mode spectrum $C_k$, computed on line, is added by fitting coefficient $\gamma$ after the Beer-Lambert law contribution terms. An initial fit of several hundred pixels per cross-track position determines the common mode spectra (one spectrum per cross-track position, between $30^\circ$N and $30^\circ$S) as the average of the fitting residuals. The common mode spectra include any instrument effects that are uncorrelated to molecular scattering and absorption. This is done to reduce the fitting root-mean-square (RMS) residuals, and the overall uncertainties. These are then applied as reference spectra in fitting of the entire orbit. The fitting additionally contains additive ($Poly_{baseline}$) and multiplicative closure polynomials ($Poly_{scale}$), parameters for spectral shift and, potentially, squeeze (not normally used). The operational parameters and the cross sections used are provided in Table 1.

As part of the development of the OMBRO retrieval algorithm, a significant amount of effort was dedicated to algorithm “tuning”, i.e., the optimization of elements in the retrieval process, including interfering absorbers like O4. The spectral region of 343 nm, where O4 has a small absorption feature, essentially is impossible to avoid in BrO retrievals: the fitting window would have to either terminate at shorter wavelengths or start past this feature, and both approaches yield to unacceptable low information content for the BrO retrievals to succeed. During the tuning process, we investigated the effects of, among many other things, including or excluding O4, the use of different spectroscopic data sets (Greenblatt et al., 1990 and Hermans et al., 1999 cross-sections), shorter or longer wavelength windows for the retrieval, and even extending the retrieval window beyond the O4 absorption feature but excluding the approximate wavelength slice of the feature itself. The only approach that provided quantitatively satisfactory results - i.e., stability of the retrieval under a wide range of conditions, minimized correlation with clouds, low fitting uncertainties, consistency of OMI global total column BrO with published results, and low noise in pixel-to-pixel retrievals - was to exclude O4 from the fit. It is impossible to quantify O4 atmospheric content from the absorption feature around 343 nm alone, and its correlation with
absorption bands of BrO and CH₂O leads to spectral correlations in the course of the non-linear least squares minimization process that are detrimental to the OMI BrO retrievals.

5 3.3 Air mass factors

Due to significant variation in ozone absorption and Rayleigh scattering in the fitting window AMFs vary with wavelength by 10-15% as shown in Fig. 2. At large solar and viewing zenith angles it is difficult to identify a single representative AMF ad hoc. The wavelength dependent AMFs are introduced to take into account for such strong variation within the BrO fitting window. They are applied pre-fit to the BrO cross sections, and the spectral fit retrieves VCDs directly. This direct fitting approach is a major departure from the commonly employed 2-step fitting procedure (OMI, 2002). It was first developed for retrievals of trace gases from SCIMACHY radiances in the shortwave infrared (Buchwitz et al., 2000) and has been demonstrated for total O₃ and SO₂ retrievals from GOME/SCIAMACHY measurements in the ultraviolet (Bracher et al., 2005; Coldewey-Egbers et al., 2005; Weber et al., 2005; Lee et al., 2008).

The albedo- and wavelength-dependent AMFs were pre-computed with the Linearized Discrete Ordinate Radiative Transfer code (LIDORT, Spurr, 2006) using a single mostly stratospheric BrO profile (Fig. 3, left panel). The BrO profile, based on the model of Yung et al. (1980), has ~30% BrO below 15 km, ~10% BrO below 10 km, and ~2% BrO below 5 km. For conditions with enhanced BrO in the lower troposphere, using this profile will overestimate the AMFs and therefore underestimate the BrO VCDs. The AMF profile used can lead to errors up to 50% with variations of albedos and viewing geometry. The mean absolute difference between AMF calculations using the stratospheric profile and the stratospheric-tropospheric profile (Fig. 3, right panel) is 41%. Surface albedos are based on a geographically varying monthly mean climatology derived from OMI (Kleipool et al., 2008). Although AMFs based on this BrO profile only slightly depend on surface albedo, albedo effects can be significant over highly reflective snow/ice surfaces, reducing VCDs by 5-10%.
In order to provide the AMF in the data product for consistency with previous versions based on a two-step approach, a second fitting of all OMI spectra is performed with unmodified BrO cross sections, which yields SCDs. An effective AMF can then be computed as AMF = SCD/VCD.

The green line in the top right panel of Fig. 1 shows the modified BrO cross section after multiplication with the wavelength-dependent AMF (albedo = 0.05, SZA (Solar Zenith Angle) = 5.0°, and VZA (Viewing Zenith Angle) = 2.5°). The wavelength-dependence in AMF is visible from the varying differences near BrO absorption peaks and the right wings at different wavelengths. The correlation of the unmodified BrO cross sections with the rest of the fitted molecules is small (typically less than 0.12), except with H₂CO (0.43). However, it is safe to assume that in most polar regions with enhanced BrO there are no high concentrations of formaldehyde. It will be worthwhile for future studies to assess the interference of H₂CO under high H₂CO and background BrO conditions similar to De Smedt et al., 2015. In addition, the AMF wavelength dependence increases with the increase of solar and viewing zenith angles and surface albedo, which increases the correlation between modified BrO cross sections and O₃ cross sections. However, the correlation with O₃ becomes noticeable (~0.10) only at SZAs above ~80°.

3.4 Destriping

OMI L1b data exhibit small differences with cross-track position, due to differences in the dead/bad pixel masks (cross-track positions are mapped to physically separate areas on the CCD), dark current correction, and radiometric calibration, which lead to cross-track stripes in Level 2 product (Veihelmann and Kleipool, 2006). Our destriping algorithm employs several methods to reduce cross-track striping of the BrO columns. First, we screen outliers in the fitting residuals. This method, originally developed to mitigate the effect of the South Atlantic Anomaly in SAO OMI BrO, H₂CO, and OClO data products, is now also being employed for GOME-2 (Richter et al., 2011). Screening outliers is done through computing the median, \( r_{\text{med}} \), and the standard deviation \( \sigma \) of residual spectra \( r(\lambda) \) and in subsequent refitting excluding any spectral points for which \( r(\lambda) \geq |r_{\text{med}} \pm 3\sigma| \). This can be done repeatedly for every ground pixel, which makes the processing slow. However, we do it once for a reference scan line, recording the positions of the bad pixels, and excluding them by default in each subsequent fit. Second, after the completion of
the spectral fitting process for all ground pixels in the granule, a post-processing cross-track bias correction is performed: an average cross-track pattern is calculated from the along-track averages of all BrO VCDs for each cross-track position within a ±30° latitude band around the equator, to which a low-order polynomial is fitted. The differences between the cross-track pattern and the fitted polynomial is then applied as a cross-track VCD correction (or “smoothing”) factor. The smoothed VCDs are provided in a separate data field, ColumnAmountDestriped. Smoothed SCDs are derived in an analogous fashion and are also included in the data product.

3.5 BrO VCD Error Analysis

Estimated fitting uncertainties are given as $\sigma_i = \sqrt{c_{ii}}$ where $C$ is the covariance matrix of the standard errors. This definition is strictly true only when the errors are normally distributed. In the case where the level 1 data product uncertainties are not reliable estimates of the actual uncertainties, spectral data are given unity weight over the fitting window, and the $1\sigma$ fitting error in parameter $i$ is determined as

$$\sigma_i = \epsilon_{rms} \sqrt{\frac{c_{ii} \times npoints}{npoints - nvaried}}$$

where $\epsilon_{rms}$ is the root mean square of the fitting residuals, $npoints$ is the number of points in the fitting window, and $nvaried$ is the number of parameters varied during the fitting.

The fitting uncertainties for single measurements of the BrO VCDs typically vary between $4 \times 10^{12}$ and $7 \times 10^{12}$ molecules cm$^{-2}$, consistently throughout the data record. The uncertainties vary with cross-track positions, from $\sim7 \times 10^{12}$ at nadir positions to $\sim4 \times 10^{12}$ at edge positions due to the increase of photon path length through the stratosphere. Relatively, the VCD uncertainties typically range between 10-20% of individual BrO VCDs, but could be as low as 5% over BrO hotspots. This is roughly 2-3 times worse that what was achieved from GOME-1 data.

The BrO VCD retrieval uncertainties listed in the data product only include random spectral fitting errors. Error sources from AMFs (i.e., BrO climatology), atmospheric composition and state (pressure/temperature vertical profiles, total ozone column, etc.) and other sources of VCD uncertainty are not included. We provide here error estimates for these additional error sources.
Uncertainties in the AMFs, used to convert slant to vertical columns, are estimated to be 10% or less except when there is substantially enhanced tropospheric BrO. Hence the total uncertainties of the BrO vertical columns typically range within 15-30%. To estimate the AMF error associated with enhanced tropospheric concentrations we have studied the difference between AMFs calculated using a stratospheric only BrO profile and a stratospheric-tropospheric profile. In this case, we find that the mean absolute difference is 41%. Fig. 4 shows the dependency of the AMFs relative error with respect to wavelength (bottom panel), albedo (middle panel) and VZA (top panel) as a function of the SZA.

We have investigated the sensitivity of OMI BrO VCD with respect to the retrieval window. We studied four wavelength windows including the current operational window (319.0-347.5 nm) version 2 window (323.0-353.5 nm), version 1 (340.0-357.5 nm) and two extra windows exploring the impact of extending the window to shorter wavelengths (310.0-357.5 nm) and reducing it by limiting its extension to wavelengths above 325 nm (325.0-357.5). Table 2 summarizes the results of these studies. The current window results in the most stable retrievals with the smallest number of pixels with negative VCD values. The difference in the mean of the VCDs retrieved using the different fitting windows are always smaller than %50.

Additional sensitivity studies also shown in Table 2 include excluding from the fitting interfering molecules (O₄ and CH₂O), using pre-flight measurements of the slit function or calculating them for each orbit, not including the mean residual (common mode) in the spectral fitting, and changing the order of the closure polynomials. In these experiments, everything else is kept the same as in the operational retrieval. In Table 2, we list the median VCDs and the median uncertainties for BrO for 13 July 2009 orbit number 26564 for each one of these retrieval configurations except for the test of the different orders of the closure polynomials. The results of the polynomial sensitivity test are summarized in Table 3. To study the impact of the slit function variation we have performed the retrieval using both an online slit function modelled as a Gaussian and the preflight instrument function. The mean difference between these two retrievals is 14% (see Table 3) for orbit number 26564.
To study the impact of the radiative transfer effects of the ozone absorption in our retrieval we have adopted the correction method described by Pukite et al., 2010. We find that between 60 degrees south and 60 degrees north the average difference is smaller than 10% with values around 2% near the equator. However, as we move near the poles with solar zenith angles above 60 degrees the differences start to be bigger arriving to mean values around 30%.

4 Results and discussions

Comparisons of the OMI OMBRO product with GOME-2 satellite retrievals and remote sensing ground based measurements over Harestua, Norway as well as monthly mean averages illustrate the quality of the retrieval on a global scale. On a local scale, recent scientific studies looking at BrO enhancements in volcanic plumes and over salt lakes are pushing the limits of the current OMBRO setups. In the following sections, we provide details of these comparisons (section 4.1) and discuss OMI OMBRO global distribution (section 4.2) and local enhancements over salt lakes and volcanic plumes observations (section 4.3), and their applicability and strategies to correctly use the publicly available OMBRO product.

4.1 Comparisons with GOME-2 and ground-based observations

To assess the quality of the OMBRO product, we first compared OMI BrO VCDs with BIRA/GOME-2 BrO observations (Theys et al., 2011). GOME-2 and OMI have different orbits: descending orbit with a local equator crossing time (ECT) of 9:30 am for GOME-2 and afternoon ascending orbit with an ECT of 1:45 pm for OMI. To minimize the effects of diurnal variation especially under high SZAs (e.g., McLinden et al., 2006; Sioris et al., 2006) on the comparison, we conduct the comparison using simultaneous nadir overpasses (SNOs) within 2 minutes between GOME-2 and OMI predicted by NOAA National Calibration Center’s SNO prediction tool (https://ncc.nesdis.noaa.gov/SNOPredictions). Due to different orbits, all these SNOs occur at high latitudes around 75°S/N. Fig. 5 shows the time series of comparison of individual OMI/GOME-2 BrO retrievals from February 2007 through November 2008. The temporal variation of BrO at the SNO locations is captured similarly by OMI and GOME-2 BrO. The scatter
plot in Fig. 6 quantifies the comparison between OMI and GOME-2 BrO. OMI BrO shows excellent agreement with GOME-2 BrO with a correlation of 0.74, and a mean bias of -0.216 ± 1.13×10^{13} molecules cm^{-2} (mean relative bias of -2.6 ± 22.1%). Considering very different retrieval algorithms including different cross sections and BrO profiles, such a good agreement is remarkable. GOME-2 retrievals use the BrO cross sections of Fleischmann et al. (2004) while our BrO retrievals use the BrO cross sections of Wilmouth et al. (1999). According to the sensitivity studies by Hendrick et al. (2009), using the Fleischmann cross section increases BrO by ~10%. So, accounting for different cross sections, OMI BrO underestimates the GOME-2 BrO by ~10%. In addition, the GOME-2 algorithm uses a residual technique to estimate tropospheric BrO from measured BrO SCDs by subtracting a dynamic estimate of stratospheric BrO climatology driven by O_3 and NO_2 concentrations and by using two different tropospheric BrO profiles depending on surface albedo conditions. This is very different from the approach of using a single BrO profile in the OMI BrO algorithm, and can contribute to some of the BrO differences. Furthermore, additional algorithm uncertainties in both algorithms and different spatial sampling can also cause some differences. Fig. 7 shows the VCDs monthly averages of GOME2 data (green) to OMBRO (black) from February 2007 to December 2009 where the seasonal variations are clearly seen. GOME2 VCDs show an upward trend that is not seen in OMI retrieved VCDs. Our study shows the mean difference for the whole period is 12%, 10%, 17%, and 10% for Alaska, Southern Pacific, Hudson Bay, and Greenland respectively.

We also used ground-based zenith-sky measurements of total column BrO at Harestua, Norway (Hendrick et al., 2007) to estimate the quality of the OMI BrO. We compared daily mean total BrO at Harestua with the mean OMI BrO from individual footprints that contain the location of Harestua site. Fig. 8 shows the time series of the comparison between OMI total BrO and Harestua total BrO from February 2005 through August 2011 with the scatter plot shown in Fig. 9. Ground-based BrO shows an obvious seasonality with high values in the winter/spring and low values in the summer/fall. Such seasonality is well captured by OMI BrO. OMI BrO shows a reasonable good agreement with Harestua BrO with a correlation of 0.46 and a mean bias of 0.12±0.76×10^{13} molecules cm^{-2} (mean relative bias of 3.18±16.30%, with respect to individual Harestua BrO). Sihler et al. (2012) compared GOME-2 BrO to ground-based observations at Utqiagvik (Barrow) finding the correlation to be weaker (r = 0.3), likely due to both elevated and shallow surface layers.
of BrO. However, their correlation between GOME-2 BrO and ground-based measurements at Amundsen, U.S. (r = 0.4) is closer to our correlation here. From the Harestua data, tropospheric BrO typically consists of 15-30% of the total BrO, larger than what we have assumed in the troposphere. The use of a single BrO profile in the OMI BrO algorithm will likely underestimate the actual BrO. Accounting for the uncertainty due to profile shape, OMI BrO will have a larger positive bias relative to Harestua measurements, which can be caused by other algorithm uncertainties and the spatiotemporal differences between OMI and Harestua BrO.

4.2 Global distribution of BrO VCDs

Table 2 shows the median VCD, median uncertainties and the number of negative pixels for the current operational version (V3), V2, and V1. Fig. 10 shows the monthly mean averages for V3 and V3 for the months of February and May of 2008. The differences on the VCD are about 30% in V3 comparing to V2. In comparison with V2 retrieval, the new retrieval (V9) does not show a large increase in the VCD concentrations, especially at the north polar region. The BrO background concentrations over the Pacific Ocean remain the same between the two versions, however, there are more retrieved VCDs.

Fig. 11 presents the global distribution of monthly mean BrO VCDs for selected months (March, June, September, December) showing BrO seasonality for three different years (2006, 2007 and 2012). BrO typically increases with latitude, with minimal values in the tropics (~2×10^{13} molecules cm^{-2}) and maximum values (~10^{14} molecules cm^{-2}) around polar regions especially in the northern hemisphere winter/spring. In the tropics, BrO shows little seasonality but at higher latitudes in polar regions, BrO displays evident seasonality. The seasonality is different between northern and southern hemispheres. In the northern hemisphere, BrO values are larger in the winter/spring and smaller in the summer/fall, and the enhancement is more widespread during the spring. In the southern hemisphere, BrO values are larger in southern hemispheric spring and summer (i.e., September and January) and smaller in the winter. Such global distribution and seasonal variation are generally consistent with previous satellite measurements (cf. Chance, 1998; http://bro.aeronomie.be/level3_monthly.php?cmd=map). BrO in the tropics shows consistent zonal distributions with lower values over land and in the intertropical convergence zone. This
might be related to the impacts of clouds on the retrievals (e.g. BrO below thick clouds cannot be measured, there are uncertainties in the AMF calculation under cloudy conditions) and will be investigated in detail in future studies. The global distribution and seasonal variation are consistent from year to year, but the distributions from different years disclose some interannual variation. For example, BrO values in 2007 are smaller in January but are larger in March compared to those in 2006. Although OMI data since 2009 have been seriously affected by the row anomaly at certain cross-track positions, the monthly mean data derived from good cross-track positions are hardly affected by the row anomaly as shown from the very similar global distribution and seasonality in 2012.

4.3 Salt lakes and volcanic plumes enhancements of BrO

Following recent work by Hörmann et al. (2016) over the Rann of Kutch we have explored the capability of OMBRO to observe similar enhancements in other salt lakes. Fig. 12 shows monthly averaged OMI BrO over the Great Salt Lake for 06/2006, the corresponding surface albedo used in the retrieval, cloud cover (assuming a cloud filter of 40%) as well as the cloud pressure. Over the Great Salt Lake, BrO enhancement occurs predominantly over the lake bed with enhancements of ~5-10×10^{12} molecules cm^{-2} over background values (3-4×10^{13} molecules cm^{-2}). Despite observing these enhancements, the users of OMBRO for these kinds of studies should be aware of three limitations of the current retrieval algorithm. First, the BrO columns assume a mostly stratospheric BrO profile (Figure 3) for the AMF calculation. Second, the OMI derived albedo climatology (Kleipool et al. 2008) used in OMBRO has a resolution of 0.5 degrees. At this resolution OMBRO retrievals can have biases given the size of OMI pixels and the inherent sub-pixel albedo variability. Finally, high albedos inherent to salt lakes surface yield abnormally high cloud fractions and low cloud pressures over the salt lakes (Hörmann et al., 2016). All these factors should be considered in studies addressing the spatiotemporal distribution of BrO over salt lakes using OMBRO. We have done a preliminary analysis of salt water bodies including the Rann of Kutch. Although this work is not fully complete and will be a separate paper, however, we see maximum BrO VCDs appearing during March–May every year from 2004 – 2015 similar to what was reported by Hörmann et al., 2016. The BrO VCDs we see are around 4.5 x 10^{13} molecules cm^{-2}.
During our analysis of volcanic eruption scenarios, it was discovered that the currently implemented SO$_2$ molecular absorption cross sections (Vandaele et al., 1994) are a sub-optimum choice (see Fig. 13). Compared to more recent laboratory measurements (Hermans et al., 2009; Vandaele et al., 2009), the original SO$_2$ cross sections implemented in OMBRO do not extend over the full BrO fitting window and exhibit the wrong behavior longward of 324 nm, overestimating the most recent measurement by up to a factor of 3. As the correlation between BrO and both SO$_2$ cross sections are very small (-0.03 for the current SO$_2$ and 0.11 for the latest SO$_2$ cross sections) over the spectral range of SO$_2$ cross sections, interference by SO$_2$ in BrO retrievals is usually not an issue at average atmospheric SO$_2$ concentrations, but strong volcanic eruptions will render even small SO$_2$ absorption features past 333 nm significant. Around 334 nm, the Vandaele et al. (2009) data show an SO$_2$ feature that correlates with BrO absorption when SO$_2$ concentrations are significantly enhanced. As a consequence of this spectral correlation, SO$_2$ may be partially aliased as BrO, since the implemented SO$_2$ cross sections cannot account for it. Fig. 13 presents an example from the 2010 Eyjafjallajökull eruption to show that the BrO retrieval can be affected by the choice of SO$_2$ cross sections. The next version of the OMBRO public release will be produced using the updated SO$_2$ absorption cross sections. Until then, caution is advised when using the OMI BrO product during elevated SO$_2$ conditions. We recommend to use OMBRO product together with the operational OMI SO$_2$ product (Li et al., 2013) to flag abnormally high BrO retrievals.

The top panels of Fig. 14 show daily average operational BrO VCDs from the eruption of the Eyjafjallajökull volcano on May 5 and 17, 2010, respectively. Enhanced BrO values in excess of 8.0×10$^{13}$ are detected in the vicinity of this volcano (e.g., plume extending southeast ward from the volcano on May 5 and, high BrO over Iceland on May 17). Some of these enhanced BrO values correspond to the locations of enhanced SO$_2$ as shown from the NASA global SO$_2$ monitoring website (https://so2.gsfc.nasa.gov/). This enhancement of BrO is not related to the seasonal variation of BrO as no such BrO enhancement is detected over Eyjafjallajökull during May 5-17, 2011 (a year after the eruption), with BrO values of only up to ~5.3×10$^{13}$ molecules cm$^{-2}$ (not shown). The bottom panels of Fig. 14 show the same BrO retrievals using SO$_2$ cross sections by
Vandaele et al. (2009). Using the improved SO$_2$ cross sections increase the BrO over a broader area on both days, supporting that the choice of SO$_2$ cross sections can affect the BrO retrievals. However, BrO enhancement around the volcano can still clearly be seen with the improved SO$_2$ cross sections. This suggests that this BrO enhancement is not totally due to aliasing of SO$_2$ as BrO, but real BrO from the volcanic eruption.

5 Conclusions

This paper describes the current operational OMI BrO retrieval algorithm developed at SAO and the corresponding V3 OMI total BrO (OMBRO) product in detail. The OMI BrO retrieval algorithm is based on nonlinear least-squares direct fitting of radiance spectra in the spectral range 319.0-347.5 nm to obtain vertical column densities (VCDs) directly in one step. Compared to previous versions of two-step algorithms, the fitting window was moved to shorter wavelengths and the spectral range was increased to reduce the fitting uncertainty. Because air mass factors (AMFs) vary significantly with wavelengths as a result of significant variation of ozone absorption, the wavelength and surface albedo dependent AMF, which is precomputed with the Linearized Discrete Ordinate Radiative Transfer (LIDORT) code using a single mostly stratospheric BrO profile, is applied pre-fit to BrO cross sections for direct fitting of VCDs. Prior to the spectral fitting of BrO, wavelength calibration is performed for both irradiance and radiance at each cross-track position and reference spectra are properly prepared at the radiance wavelength grid. Then radiances are modeled from the measured solar irradiance, accounting for rotational Raman scattering, undersampling, attenuation from BrO and interfering gases, and including additive and multiplicative closure polynomials, and the average fitting residual spectrum. To maintain consistency with previous versions, a second fitting of all OMI spectra is performed with unmodified BrO cross sections to derive SCDs and the effective AMFs. Then a destriping step is employed to reduce the cross-track dependent stripes.

The uncertainties of BrO VCDs included in the data product include only spectral fitting uncertainties, which typically vary between 4 and 7×10$^{12}$ molecules cm$^{-2}$ (10-20% of BrO VCDs, could be as low as 5% over BrO hotspots), consistent throughout the data record. The uncertainties vary with cross-track positions, from ~7×10$^{12}$ at nadir positions to ~4×10$^{12}$ at edge positions. The
use of single stratospheric BrO profile is another source of uncertainty, overestimating AMFs and therefore underestimating BrO VCDs for conditions with enhanced BrO in the lower troposphere. In addition, the used SO$_2$ cross sections are a sub-optimum choice and can cause errors in the retrievals under high SO$_2$ concentrations.

We compared OMI BrO VCDs with BIRA/GOME-2 BrO observations at locations of simultaneous nadir overpasses. OMI BrO shows excellent agreement with GOME-2 BrO with a correlation of 0.74, and a mean bias of -0.216±1.13×10$^{13}$ molecules cm$^{-2}$ (mean relative bias of -2.6 ± 22.1%). We also compared OMI BrO with ground-based zenith-sky measurements of total BrO at Harestua, Norway. This BrO seasonality in Harestua total BrO is well captured by the OMI BrO and OMI BrO shows a reasonable good agreement with a moderate correlation of 0.46 and a small mean bias of 0.12±0.76×10$^{13}$ molecules cm$^{-2}$ (mean relative bias of 3.18±16.30%). The global distribution and seasonal variation of OMI BrO are generally consistent with previous satellite measurements. There are small values in the tropics with no much seasonality, and large values at high latitudes with distinct seasonality. And the seasonality is different between the northern and southern hemisphere, with larger values in the hemispheric winter/spring (spring/summer) and smaller values in summer/fall (winter) for the northern (southern) hemisphere. This spatiotemporal variation is generally consistent from year to year and is hardly affected by the row anomaly, but does show some interannual variation. The retrievals show enhanced BrO of 5-10×10$^{12}$ molecules cm$^{-2}$ over the U.S. Great Salt Lake and also significant enhancement from the eruption of Eyjafjallajökull volcano despite BrO retrievals under high SO$_2$ conditions can be affected by the current use of a sub-optimal choice of SO$_2$ cross sections.

Acknowledgements

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is managed by NIVR and KNMI in the Netherlands. We acknowledge the OMI International Science Team for providing the SAO OMBRO data product used in this study.
References


Table 1. Fitting window and parameters used to derive BrO vertical column densities

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Description/value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fitting window</td>
<td>319.0 - 347.5 nm</td>
</tr>
<tr>
<td>Baseline polynomial</td>
<td>4th order</td>
</tr>
<tr>
<td>Scaling polynomial</td>
<td>4th order</td>
</tr>
<tr>
<td>Instrument slit function</td>
<td>Hyper-parameterization of pre-flight measurements, Dirksen et al., 2006</td>
</tr>
<tr>
<td>Wavelength calibration</td>
<td>Spectral shift (no squeeze)</td>
</tr>
<tr>
<td>Solar reference spectrum</td>
<td>Chance and Kurucz, 2010</td>
</tr>
<tr>
<td>BrO cross sections</td>
<td>Wilmouth et al., 1999, 228K</td>
</tr>
<tr>
<td>H$_2$CO cross sections</td>
<td>Chance and Orphal, 2011, 300K</td>
</tr>
<tr>
<td>O$_3$ cross sections</td>
<td>Malicet et al., 1995, 218K, 295K</td>
</tr>
<tr>
<td>NO$_2$ cross sections</td>
<td>Vandaele et al., 1998, 220K</td>
</tr>
<tr>
<td>SO$_2$ cross sections</td>
<td>Vandaele et al., 1994, 295K$^1$ Hermans et al., 2009; Vandaele et al., 2009, 295K$^2$</td>
</tr>
<tr>
<td>OCIO cross sections</td>
<td>Kromminga et al., 2003, 213K</td>
</tr>
<tr>
<td>Molecular Ring cross sections</td>
<td>Chance and Spurr, 1997</td>
</tr>
<tr>
<td>Undersampling correction</td>
<td>Computed on-line, Chance et al., 2005</td>
</tr>
<tr>
<td>Residual (common mode) spectrum</td>
<td>Computed on-line between 30°N and 30°S</td>
</tr>
</tbody>
</table>

1. Used in the current operational algorithm.

2. Used for testing sensitivity to SO$_2$ cross sections and will be used in the next version.
Table 2. Error analysis studies.

<table>
<thead>
<tr>
<th>Description</th>
<th>Median VCD (Molec. cm$^{-2}$)</th>
<th>Median uncertainty (Molec. cm$^{-2}$)</th>
<th>Number of negatives</th>
</tr>
</thead>
<tbody>
<tr>
<td>Operational (V3)</td>
<td>$3.89 \times 10^{13}$</td>
<td>$7.85 \times 10^{12}$</td>
<td>1222</td>
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<tr>
<td>323.0 - 353.5 nm (V2)</td>
<td>$2.69 \times 10^{13}$</td>
<td>$1.01 \times 10^{13}$</td>
<td>4393</td>
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<tr>
<td>340.0 – 357.5 nm (V1)</td>
<td>$2.48 \times 10^{13}$</td>
<td>$1.29 \times 10^{13}$</td>
<td>9390</td>
</tr>
<tr>
<td>310.0 - 357.5 nm</td>
<td>$1.91 \times 10^{13}$</td>
<td>$6.83 \times 10^{12}$</td>
<td>7372</td>
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<tr>
<td>325.0 - 357.5 nm</td>
<td>$3.10 \times 10^{13}$</td>
<td>$8.75 \times 10^{12}$</td>
<td>3107</td>
</tr>
<tr>
<td>With O$_2$-O$_2$</td>
<td>$3.57 \times 10^{13}$</td>
<td>$8.65 \times 10^{12}$</td>
<td>1265</td>
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<tr>
<td>Online slit function</td>
<td>$5.00 \times 10^{13}$</td>
<td>$7.92 \times 10^{12}$</td>
<td>1003</td>
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<tr>
<td>Without common mode</td>
<td>$3.72 \times 10^{13}$</td>
<td>$1.11 \times 10^{13}$</td>
<td>2093</td>
</tr>
<tr>
<td>Without HCHO</td>
<td>$2.53 \times 10^{13}$</td>
<td>$6.93 \times 10^{12}$</td>
<td>1703</td>
</tr>
</tbody>
</table>
Table 3. Summary of different errors sources in the BrO vertical column.

<table>
<thead>
<tr>
<th>Error source</th>
<th>Type</th>
<th>Parameter uncertainty</th>
<th>Averaged uncertainty on BrO VCD</th>
<th>Evaluation method</th>
<th>reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measurement noise</td>
<td>Random</td>
<td>S/N 500 - 1000</td>
<td>4-7x10^{12} molec. cm^2</td>
<td>Error propagation;</td>
<td></td>
</tr>
<tr>
<td>HCHO</td>
<td></td>
<td></td>
<td>5%</td>
<td>Chance and Orphal,</td>
<td>2011, 300K</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Malicet et al., 1995, 218K, 295K</td>
<td></td>
</tr>
<tr>
<td>O₃</td>
<td></td>
<td></td>
<td>2%</td>
<td>Wilmouth et al., 1999, 228K</td>
<td></td>
</tr>
<tr>
<td>BrO</td>
<td>Systematic</td>
<td>Based on literature reported error estimates</td>
<td>8%</td>
<td></td>
<td></td>
</tr>
<tr>
<td>NO₂</td>
<td></td>
<td></td>
<td>3%</td>
<td>Vandaele et al., 1998, 220K</td>
<td></td>
</tr>
<tr>
<td>SO₂</td>
<td></td>
<td></td>
<td>5%</td>
<td>Vandaele et al., 1994, 295K</td>
<td></td>
</tr>
<tr>
<td>OCIO</td>
<td></td>
<td></td>
<td>5%</td>
<td>Kromminga et al., 2003, 213K</td>
<td></td>
</tr>
<tr>
<td>Ring</td>
<td></td>
<td></td>
<td>5%</td>
<td>Chance and Spurr, 1997</td>
<td></td>
</tr>
<tr>
<td>Offset order</td>
<td>Systematic</td>
<td>Vary polynomial order</td>
<td>10%</td>
<td></td>
<td></td>
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<tr>
<td>Polynomial order</td>
<td>Systematic</td>
<td>Vary polynomial order</td>
<td>10%</td>
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<tr>
<td>Instrumental slit function and wavelength calibration</td>
<td>Systematic</td>
<td>Preflight and online slit function</td>
<td>28%</td>
<td>Sensitivity analysis</td>
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<tr>
<td>Wavelength interval</td>
<td>Systematic</td>
<td>Varying fitting window</td>
<td>50%</td>
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</table>
Figure 1. Cross sections used in the current operational BrO algorithm except for the SO\textsubscript{2} cross section at 298 K which is to be used in the next version. The black lines are the original cross sections, the color lines show the cross sections convolved with OMI slit function (which is assumed to be a Gaussian with 0.42nm full width at half maximum. The BrO cross section after multiplication with the wavelength-dependent AMFs used these parameters for the AMF calculation: albedo = 0.05, SZA= 5.0°, and VZA = 2.5°).
Figure 2. Wavelength- and albedo-dependent air mass factors calculated using a mostly stratospheric fixed BrO profile. The blue box shows the fitting window used in our previous versions, and the red box shows the fitting window used in the current operational algorithm.

Figure 3. A mostly stratospheric vertical BrO profile used for air mass factors (left panel). Total BrO, BrO < 15 km, BrO < 10 km, and BrO < 5 km are $1.55 \times 10^{13}$, $5.06 \times 10^{12}$, 1.55 $\times 10^{12}$, and 2.87 $\times 10^{11}$, respectively. A stratospheric tropospheric vertical BrO profile for air mass factors (right panel) can reduce errors by 50%.
Figure 4. AMFs relative errors as a function of the SZA and the wavelength (bottom panel), albedo (middle panel) and VZA (top panel).

Figure 5. Time series comparison of SAO OMI (red) BrO and BIRA GOME-2 (blue) BrO VCDs from February 2007 to November 2008 using simultaneous nadir overpasses within 2 minutes between OMI and GOME-2 observations.
Figure 6. Correlation and orthogonal regression of OMI and GOME-2 BrO for the data shown in Fig. 5. The legends show the mean bias and standard deviation of the differences, correlation, and the orthogonal regression.
Figure 7. VCD of GOME2 (green) comparison to OMI (black) over four regions from February 2007 to December 2009 for four regions.

Figure 8. Time series comparison of ground-based zenith-sky total BrO (black) at Harestua, Norway and coincident SAO OMI BrO (red) from February 2005 through August 2011.
Figure 9. Correlation and orthogonal regression of OMI and Harestua BrO for the data in Fig. 8. The legends show the mean biases and standard deviations of the differences, correlation, and the orthogonal regression.
Figure 10. Monthly averages for February and May 2008 for version 3 and version 2.
Figure 11. Global distributions of monthly mean BrO VCDs in March, June, September and December (in different rows) of 2006, 2007, and 2012 (different columns). Bromine release “explosions” during the Polar Spring months can be seen clearly.
Figure 12. Mean June 2006 BrO VCD over the Great Salt Lake area. Averages have been calculated on a 0.2 x 0.2 degree grid including only pixels with cloud fractions smaller than 0.4.
Figure 13. Comparison of BrO absorption (red) and SO$_2$ absorptions under volcanic scenarios based on cross sections used in the operational algorithm (Vandaele et al., 1994) as shown in black and the recent laboratory cross sections (Vandaele et al., 2009) as shown in purple. For BrO, a SCD of 1.0×10$^{14}$ molecules cm$^{-2}$ is assumed; for SO$_2$, a SCD of 15 Dobson Units (i.e., 4.03×10$^{17}$ molecules cm$^{-2}$) is assumed. Cross sections have been convolved with OMI slit function (which is assumed to be a Gaussian with 0.42nm full width at half maximum).
Figure 14. Daily average BrO VCDs from Eyjafjallajökull on May 5 (a) and 17 (b), 2010 produced using the operational SO$_2$ cross sections and for the same days (c) and (d) using the Vandaele et al. (2009) SO$_2$ cross sections.