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Atmospheric Measurement Techniques Discussions

Interactive comment on "OMI total bromine monoxide (OMBRO) data product: Algorithm, retrieval and measurement comparisons" *by* Raid M. Suleiman et al.

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We thank referee's helpful and constructive comments and review. Our responses are in **bold** starting with **"Response:"**
Figures have been moved around:
Old manuscript Updated manuscript
Figure 1 still Figure 1
Figure 2 still Figure 2
Figure 3 still Figure 3
Figure 4 moved to become Figure 10

Figure 5 moved to become Figure 4
Figure 6 moved to become Figure 5
Figure 7 moved to become Figure 6
Figure 8 moved to become Figure 7
Figure 9 moved to become Figure 8
Figure 10 moved to become Figure 9
Figure 11 still Figure 11

General Comment

In the paper "OMI total bromine monoxide (OMBRO) data product: Algorithm, retrieval and measurement comparisons" Raid M. Suleiman and co-authors present the operational retrieval algorithm for bromine monoxide (BrO) columns from measurements by the Ozone monitoring instrument (OMI). Since BrO is a trace-gas with significant impact on atmospheric chemistry, the paper fits to the scope of AMT. In my opinion, however, the scientific quality would need more than major revisions because the presentation of the retrieval method in its current form is far from being scientifically publishable. Therefore, I suggest to reject the current manuscript but I would like to encourage resubmission after the following issues have been addressed. The decision to suggest the rejection of a manuscript is never easy. In this particular case, however, especial care must be given to scientific quality because operational products are potentially applied by fellow scientists, which may not be trained enough to assess the quality and reliability of the product by themselves. The manuscript, however, rather obfuscates potential quality issues instead of presenting a transparent analysis of the algorithm performance.

Specific Comments

1) The most critical aspect of the presented algorithm for the retrieval of BrO from OMI measurements lays in the choice of the wavelength range. The presented algorithm applies a fitting window between 319 to 347.5 nm. Not being an expert for the retrieval of BrO myself, I found the arguments of Vogel et al., 2013 concerning the fit interval of OMBRO particularly alarming. Vogel et al. state in the caption of their Fig. 11 that

"Wavelength evaluation ranges with a lower limit <325 are dominated by O3 and SO2 features". In my opinion, this is really alarming since BrO, O3, and SO2 chemistry are highly correlated. The manuscript itself even contains proof that the applied wavelength range might be an issue:
a) Figure 2 shows that the applied AMFs ("OMI current") are structured by O3 absorption indicating that interferences with O3 are close to inevitable.
b) Section 3.6 and Figure 11 reveal interferences with SO2.
c) If scaled properly, Figure 2 would reveal many absorbers to have much larger structures than BrO.
d) Figure 4 is clipped below 330nm. Why? Please show the whole story
In the current manuscript, however, the choice of the new fitting window is justified in (p.7, l. 2) by the simple statement: "to reduce fitting uncertainty by including more BrO spectral structures". This does not convince me and I really would like to urge the authors to present significant arguments to justify the applied wavelength range, which is far-off compared to the wavelength ranges used by other groups (cf. Table 1 in Vogel et al., 2013). The least the authors could have done would be to include a plot showing results using the "Traditional" and "OMI current" wavelength ranges. I would like to propose some questions that may lead the authors to find profound arguments: How does the residual change when changing the fitting window? What about systematic structures in the residual? How large are biases by other absorbers depending on the fit range? These questions may be tested using real and synthetic measurements as well using the methods by Chan Miller et al., which was co-authored by many coauthors of this paper. Hence, I wonder why methods for reliably comparing different wavelength ranges were not applied here even though they are existing at SAO. Studies building on this data skate on thin ice if the above issues are not addressed appropriately.
Furthermore (p. 1, l. 18), the authors detail that also "the average fitting residual spectrum" is included in the fit. This approach may obfuscate potential systematic interferences and is, in my opinion, only appropriate if its influence is thoroughly

studied. Please provide more information. Please investigate the cross-correlations with other absorbers and include it in a revised Figure 1.

Response:

We first proposed that BrO could be measured globally from satellites (Chance, K.V., J.P. Burrows, and W. Schneider, Retrieval and molecule sensitivity studies for the Global Ozone Monitoring Experiment and the SCanning Imaging Absorption spectroMeter for Atmospheric CHartographY, Proc. SPIE, Remote Sensing of Atmospheric Chemistry, 1491, 151-165, 1991; http://www.cfa.harvard.edu/atmosphere/publications.html). We were then the first to fit BrO from GOME-1 and have fitted BrO from SCIAMACHY, OMI, and OMPS. The update of the BrO fitting window from V2 to V3 in 2011 is not arbitrary, but is based on substantial quantitative analysis by checking the quality of BrO retrievals and the correlation with other trace gases while systematically varying the lower and upper limits of fitting windows, similar to the studies of Chan Miller et al. (2014). It is obvious that O3 absorption and SO2 absorption are much stronger than BrO absorption below 325 nm, but it does not mean BrO cannot be retrieved using part of this wavelength range as we are simultaneously fitting O3 and SO2. For example, operational SO2 measurements from UV are almost always retrieved from a window that is entirely dominated by the O3 absorption for typical SO2 abundance. We state in this current paper that "The correlation of the unmodified BrO cross sections with the rest of the molecules fitted 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."

The caption to Fig. 11 in Vogel et al., (2013) states that "wavelength evaluation ranges with a lower limit <325 are dominated by O3 and SO2 features, whereas the other wavelength ranges may be influenced mainly by NO2 and HCHO." This is a comparison of where different molecules might interfere based

on the absorption not an absolute determination of the best range. Our careful evaluation of correlation stands. We have been very systematic, as in Chan Miller et al., 2014, in the selection of our wavelength range. If one puts a point on Fig. 11 of Vogel et al. (2013) corresponding to our fitting window, one would see that the correlation is not significant for our measurements. In fact, Vogel et al. (2013) stated in their Appendix B conclusion (B3) that "If the retrieval wavelength interval is sufficiently wide and includes strong BrO absorption lines, the results are not strongly affected by changes in retrieval wavelength intervals (lower wavelength limits 320–337.5 nm and upper wavelength limit > 342 nm)."

"a) Figure 2 shows that the applied AMFs ("OMI current") are structured by O3 absorption indicating that interferences with O3 are close to inevitable."

Yes. AMFs should be affected by the dominant O3 absorption. We agree that interferences with O3 is close to inevitable if O3 is not fitted. We simultaneously fit O3 and account for this wavelength-dependent AMF to minimize the O3 interference so the correlations between BrO and O3 (218 K) and O3 (298 K) are typically very small, less than 0.03

"b) Section 3.6 and Figure 11 reveal interferences with SO2."

This is fully explained in Section 4.3 second paragraph (starting on line 15 of the updated manuscript). Correlation with volcanic SO2 will be reduced when improved cross sections are used. Correlation with normal SO2 is negligible given the low levels of SO2 present in the atmosphere. As we point out in the volcano detection section, when high levels of SO2 are present OMI BrO should be utilized with extra care.

"c) If scaled properly, Figure 2 would reveal many absorbers to have much larger structures than BrO."

Looks like you meant Figure 1. This is why spectra must be fitted very carefully and correlations evaluated. Otherwise, nobody could fit atmospheric BrO or most other trace gases in the UV/visible. We have changed the approach to Figure 1 by showing four different panels with 4 different optical density orders of magnitude.

"d) Figure 4 is clipped below 330nm. Why? Please show the whole story."

We are making no attempts to hide information. On the contrary, we use this wavelength range to highlight the region where the new and old SO2 cross sections differ the most. We have modified the figure to show the whole fitting window.

In summary, we urge the referee to discount Comment 1 arguments for rejection and accept our paper. We consider that the goal of the paper, to provide a scientific description of SAO OMI BrO retrievals and some particular examples, is achieved. We appreciate the comments that helped us to improve the clarity and transparency of the manuscript. However, we want to clearly reject the idea, as hinted throughout the comments, that we are making an intentional effort to keep the paper obscure.

2) It is not clear to me to what extend the presented paper is dedicated to validation of the retrieval results. The title suggests "measurement comparison" and the abstract details that the paper "shows some validation", which is confusing. Please be more specific on the purpose and the results of the validation exercises. Example: The measurements at Harestua are not ideal for evaluating BrO close to the surface due to

a lack of tropospheric BrO events. Therefore, I suggest to state that the sensitivity of OMBRO towards near-surface BrO may not be evaluated using those measurements. If the authors aim at near-surface BrO with their product, which they do because they claim to have detected BrO over the Great Salt Lake, I suggest to use a data set featuring a significant measurement sensitivity for BrO columns at the ground, for example Frieb et al., 2012.

Response:
The purpose of the paper is not to do a dedicated validation exercise but to describe the algorithm and show some comparisons, consistent with the title. We show some comparisons of total BrO with GOME-2 and ground-based observations, and show examples of BrO enhancement from volcanic eruptions and salt lakes. We are not trying to evaluate BrO close to the surface or in the troposphere as the retrieval currently assumes a mostly stratospheric BrO profile. But, this does not prevent us from showing enhanced BrO due to sources near the surface as these sources will contribute to the total BrO although total BrO will be underestimated due to the assumed BrO profile. In the abstract, we replaced "shows some validation" with "comparisons."

3) I would like to suggest to review the selection of references in the introduction. There are many citations of papers (co)authored by the coauthors of this paper while papers from other groups seem to be often ignored. E.g. for the sources of tropospheric BrO mostly satellite papers are cited even though there are many observations by groups using ground-based methods. This way of introducing the different findings may be misleading for the readers. Even more, informed fellow scientist readers may be offended if their contribution is not appropriately acknowledged. I suggest to be a bit more generous here. Simpson et al., 2015 may provide a start for a comprehensive list of publications.

Specifically, I miss the following references in an up-to-date BrO satellite paper:

- Please add (Hörmann et al., 2013), which is one of the most comprehensive surveys of volcanic BrO sources using satellites.

Response:
We added this reference in the revision with the text: "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/SO2 ratios."

- Please add (Liao et al., 2011) and (Frieß et al., 2012) to the references for Barrow, Alaska since both papers present significant BrO observations of near-surface BrO.

Response:
The recommended two references were added. Please note that Liao, et al., 2011 was finally published in 2012 and Frieß was published in 2011 not 2012.
Text added to manuscript: Frieß et al., 2011; Liao et al., 2012a,b;

- Further BrO satellite papers well worth citing: Begoin et al., 2010, Toyota et al., 2011, Sihler et al., 2012, and Blechschmidt et al., 2016

Response:
Begoin et al. (2010) was added in the Introduction.
Toyota et al. (2011) is a modeling paper and it has no observations or comparison to any satellite or ground-based measurements, so it is not added.
Sihler et al. (2012) and Blechschmidt et al. (2016) were added to the Introduction.

4) The investigation of the BrO over the Great Salt lake is insufficient and I am missing a rationale for including this issue in the paper at all. However, the results may be due to systematic effects caused by a variety of geophysical parameters (see investigation by Hörmann et al., 2016). Without an appropriate discussion of these influences I would not accept the authors claim that the signal is really due to emissions from the Great Salt Lake.

Response:
We have added several sentences at the beginning of section 4 to show the rationale "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."

The paragraph has been significantly updated. In addition to BrO enhancement over the Great Salt Lake, we added BrO enhancement also over the Dead Sea Valley. The impacts of geophysical parameters were discussed. This is the updated text:

Following recent work by Hörmann et al. (2016) we have checked the capability of OMBRO to observe similar enhancements in other salt lakes. Fig. 9 shows monthly averaged OMI BrO over the Great Salt Lake for 02/2013.

and the Dead Sea for 07/2009. Over the Great Salt Lake, BrO enhancement occurs predominantly over the lake bed with enhancements of 5-10$\times$1012 molecules cm-2 over background values (4-4.7$\times$1013 molecules cm-2). Over the Dead Sea, the BrO enhancement of 5-8 $\times$1012 molecules cm-12 occurs to the South-West, where BrO accumulates at a small hill due to the prevailing north-easterly winds. Despite observing these enhancements, the users of OMBRO for these kinds of studies should be aware of two limitations of the current retrieval. First, the actual BrO enhancement is actually underestimated since we are assuming a mostly stratospheric BrO profile for the AMF. 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 sub-pixel albedo variability not represented in the albedo climatology. We also raise attention to the fact that abnormally high cloud fractions are reported over the salt lakes due to enhanced albedos. All these considerations are important for future studies studying spatiotemporal distribution of BrO over salt lakes.

5) In my opinion, the treatment of the interferences with SO2 is not appropriate. If the issue is known, why not solve it right-away and then publish an improved version? What is the purpose of an OMBRO product featuring this imperfection? I suggest to solve this issue together with choosing an appropriate fitting window before resubmission.

Response:
All operational algorithms of all gases contain known issues. There are usually suggestions as to how future improvements can be made and how to use the data properly. Interference with SO2 is only an issue when SO2 concentrations become significant. Keeping that in mind the main purpose of discussing the impact of the SO2 cross sections in the retrieval is to educate possible users

about the limitations of the current set up in specific scenarios (i.e. volcanic plumes and smelters). We discuss the effect of the new cross sections (Vandaele et al., 2009) since we plan to include them in future updates to the operational processing. We also look forward to laboratory measurements over an extended temperature range, and have encouraged them. However, given the limited impact in the retrieval results (from a geographic and occurrence perspective), the complications and limitations arising from operational processing and the computing cost of full mission reprocessing we prefer to apply several updates in each new version. With plans to update other aspects of the retrieval we favor the option of saving the SO2 cross section update for the future while we still think is important to single out the limitations of the current setup so that all known limitations of the OMI BrO are available to the science community in order to make educated decisions on which kind of science investigations are supported by the data product, and which are not. This is completely standard in the development of operational products.

Specifically:
a) Fig. 11 is really hard to interpret. I suggest to show a comparison plot based on single OMI measurements instead of gridded maps.

Response:
A plot of a single OMI measurement does not add much. Due to significant orbit overlapping at this location. The results need to be averaged to show entire structures clearly. For reference, two separate orbits are shown below (Figures 1-4).

b) In the introduction (p.2, l.31): Please rephrase "a known issue" for something more specific. In my opinion, the interference with SO2 is not just an issue but a significant flaw.

Response:
As explained above, it is not a significant flaw, nor another reason to reject the paper. We have changed "a known issue" to SO2 interference.

c) Fig. 4: The x-axis must contain the entire fitting window at least. I find the figure in its current form rather disturbing.

Response:
"Rather disturbing" is not appropriate language for an unbiased scientific review. We use this wavelength range to highlight the region where the new and old SO2 cross sections differ the most. Figure 4 (in the updated manuscript moved to be Figure 10) is updated to include the entire fitting window. Additionally, we plotted cross sections that have been convolved with OMI slit function (which is assumed to be a Gaussian with 0.42nm full width at half maximum).

6) The following plots need to be improved:

a) Fig. 1: I strongly suggest to refrain from scaling arbitrarily to allow an open discussion of the results. For example, the amplitude of O3 and SO2 cross sections and the Ring spectra seem to be strongly manipulated in order to downplay their potential impact. I suggest to apply an y-axis in optical density space and scale the cross-sections according to a typical fit.

Response:
It was not intentional to downplay any contribution from any cross sections through arbitrary scaling. Our intention was to provide a qualitative image of the species involved in the fitting while keeping the figure simple. We have changed the approach to Figure 1 by showing four different panels with 4 different optical

density orders of magnitude.

b) Fig. 6: Please use orthogonal regression for the comparison. Linear regression is not appropriate for independent data sets.

Response:
We corrected old Figure 6 (now Figure 5) using orthogonal distance regression.

Changed caption of old Figure 6:
Correlation and orthogonal regression of OMI and GOME-2 BrO for the data in Figure 4 when both are available. The legends show the mean bias and standard deviations of the differences, correlation, and the orthogonal regression.

c) Fig. 7: The frequency of the time series is too high to allow a one-to-one comparison. I recommend to also show a zoomed plot of two months or so.

Response:
Old Figure 7 (now Figure 6) have been updated to include only the time series from Harestua and OMI total BrO only since we are not discussing in the manuscript the Harestua stratospheric/tropospheric BrO. We also changed the x-axis to be Time (years).
We are including in this discussion a plot showing only few months (Figure 5), however, we believe it does not add value to the manuscript.

d) Fig. 8: see 6c)

Response:âĂĺ
You mean see comment 6b. We updated old Figure 8 (now Figure 7) using orthogonal regression instead of linear regression.

Changed the caption of Figure 8:
Correlation and orthogonal regression of OMI and Harestua BrO for the data in Figure 6. The legends show the mean biases and standard deviations of the differences, correlation, and the orthogonal regression.

e) Fig. 10: This plot does not allow an independent judgment whether this is a significant signal or not. Suggested improvements:
- Increase area significantly
- Use full colorscale
- Thicker coast lines
- Align with other geospatial properties: cloud statistics, albedo, precipitation etc.

Response:
Old Figure 10 (now Figure 9) has been updated: We increased the covered area, used full-color scale, and added the lake line. We also added another panel to show BrO enhancement over the Dead Sea Valley for September 2007.

Further Comments
(p. 5, l. 16) "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." I wonder whether this is an advantage or disadvantage of the described method. What is the intention behind this statement? My suggestion would be not to confuse the reader and just remove it from the manuscript.

Response:âĂĺ
We do not see anything wrong with the above sentence. We are trying to describe the algorithm and how it differs from other approaches (c.f., Platt, U.,

"Differential optical absorption spectroscopy (DOAS)", Chem. Anal. Series, 127, 27 - 83, 1994). Describing the algorithm in detail should not confuse the readers. In our long experience with analysis of satellite spectra, the added DOAS steps do not improve the result and thus our approach has an advantage.âĂİ

Response:âĂİ
We do not see anything wrong with the above sentence. We are trying to describe the algorithm and how it differs from other approaches (c.f., Platt, U., "Differential optical absorption spectroscopy (DOAS)", Chem. Anal. Series, 127, 27 - 83, 1994). Describing the algorithm in detail should not confuse the readers. In our long experience with analysis of satellite spectra, the added DOAS steps do not improve the result and thus our approach has an advantage.âĂİ

(p. 11, l.17) Why are OMI and GOME-2 data treated differently with respect to spatial averaging? Without discussion, the reader may assume that OMI data are more noisy and needed some smoothing. Please be more specific.

Response:âĂİ
We have changed to use individual OMI BrO measurements without averaging. We generated new Figure 4 (in the updated manuscript) and Figure 5 (in the updated manuscript) corresponding to the use of OMI BrO data on individual pixels.
We see excellent agreement between OMI BrO and GOME-2 BrO with a correlation of 0.74, and a mean bias of -0.216 $\pm$ 1.13$\times$1013 molecules cm-2 (mean relative bias of -2.6 $\pm$ 22.1$\%$).

(p. 13, l.21 -> l. 25) -> move to introduction

Response:âĂİ
The text was moved to the Introduction and was changed to "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). 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)."

(p. 14, l. 2 -> l. 99 -> move to introduction

Response:
âĂİPage 14 Line 2 to Line 9 was moved to the Introduction and was changed to "Bobrowski et al. (2003) made the first ground-based observations of BrO and SO2 abundances in the plume of the Soufrière Hills volcano (Montserrat) by multi-axis DOAS (MAX-DOAS). BrO and SO2 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/SO2 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/SO2 ratios."

(p.2, l. 29): "briefly analyze" characterizes an approach not suitable for a scientific article. An analysis is either profound or not scientific. In my opinion, an AMT paper should only contain profound content.

Response:âĂĺ
The statements “An analysis is either profound or not scientific. In my opinion, an AMT paper should only contain profound content” are truly unnecessary. The standard of our research is as high as that of anyone in the field. Our analysis is not brief and we should not have used that word. We removed it from the manuscript.

(p.3, l. 22): Please add reference documenting the OMI row anomaly:
http://projects.knmi.nl/omi/research/product/rowanomaly-background.php

Response:âĂĺ
The link “(http://projects.knmi.nl/omi/research/product/rowanomaly-background.php)” has been added after OMI row anomaly:

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2018-1, 2018.

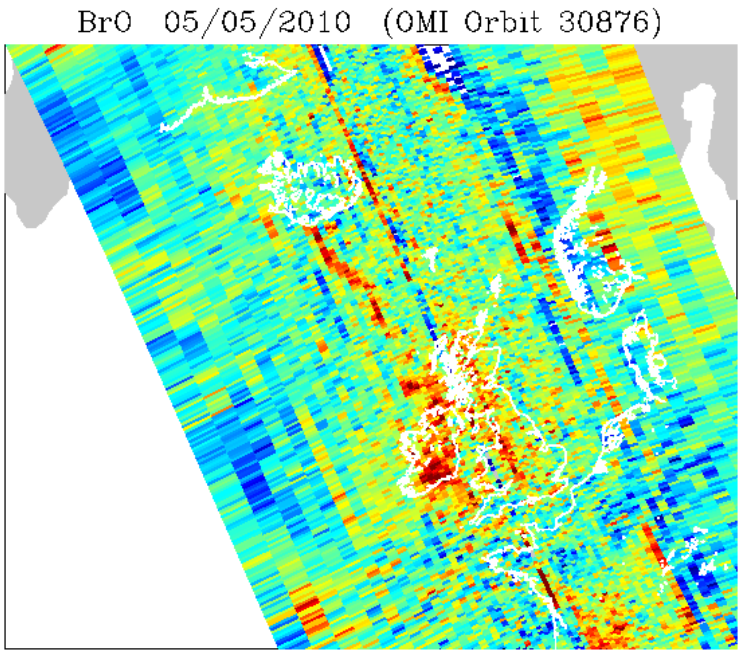

Fig. 1. Retrievals with new SO2 cross section (Vandaele et al. (2009)âĂĺ. OMI orbit 30876

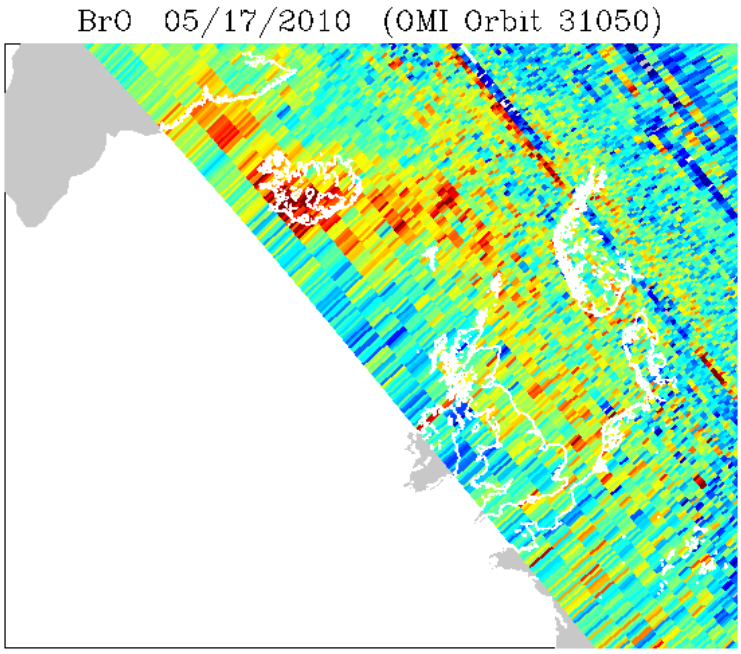

Fig. 2. Retrievals with new SO2 cross section (Vandaele et al. (2009)âĂĺ. OMI orbit 31050

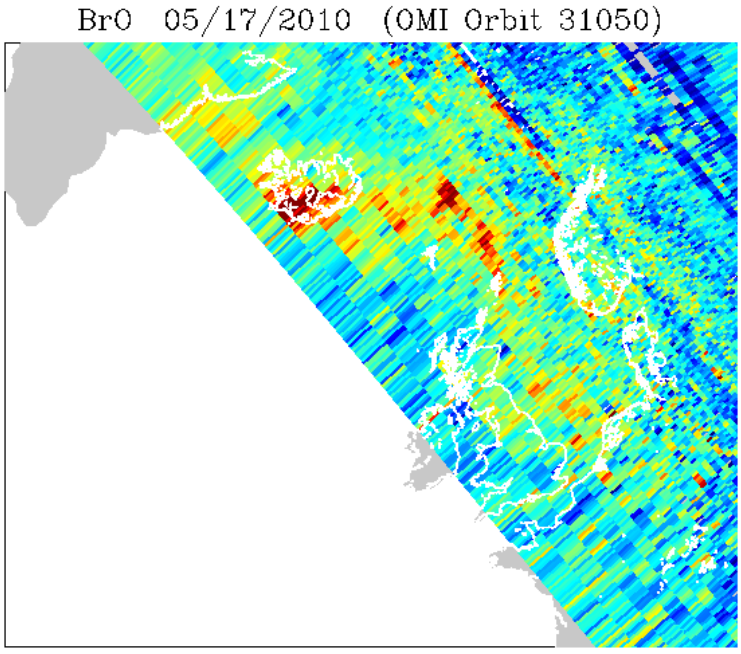

Fig. 3. Retrievals with operational SO2 cross section (Vandaele et al. (1994) OMI orbit 31050

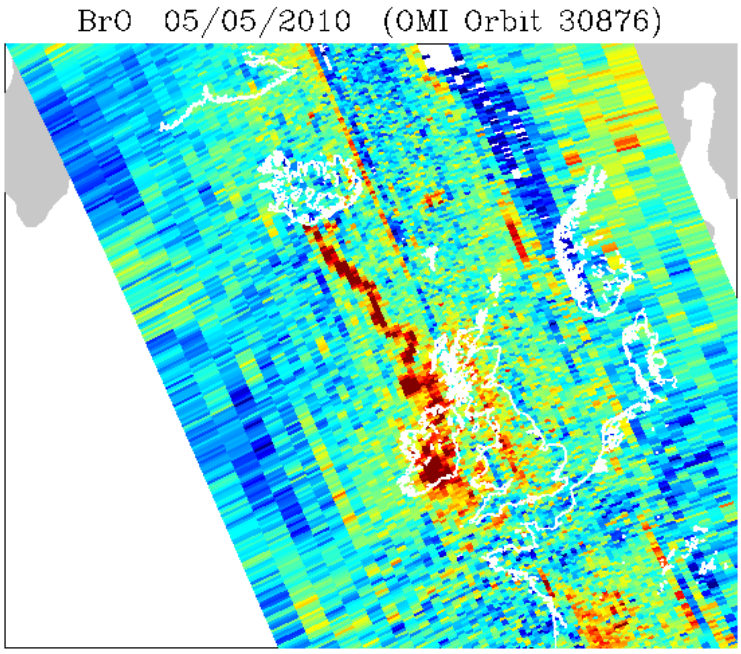

Fig. 4. Retrievals with operational SO2 cross section (Vandaele et al. (1994) OMI orbit 30876

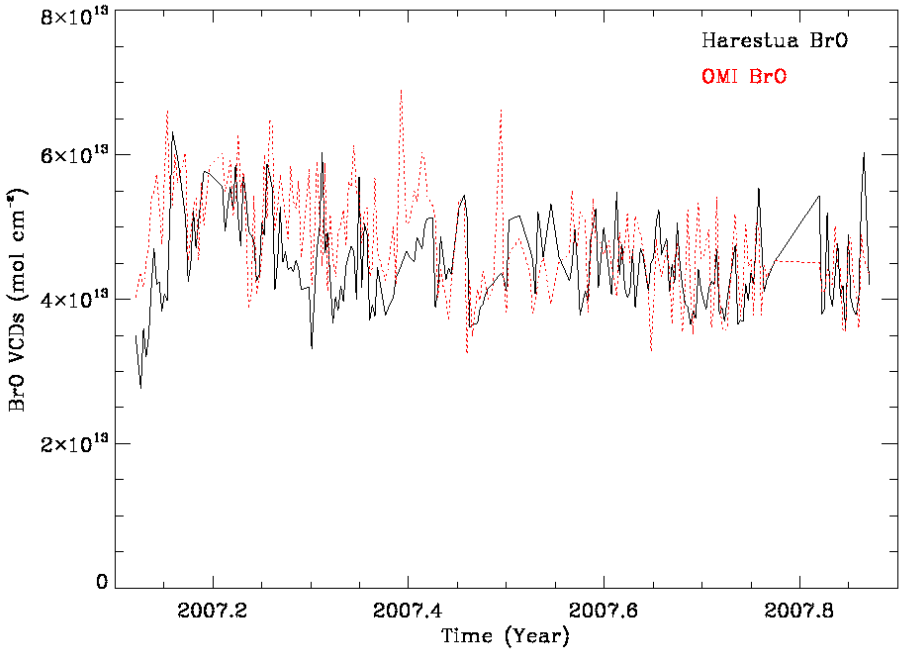

Fig. 5. Time series comparison of ground-based zenith-sky total BrO (black) at Harestua, Norway and coincident SAO OMI BrO (red) from February 2007 through August 2007.