Author comment on

Sihler, H., Platt, U., Beirle, S., Marbach, T., Kühl, S., Dörner, S., Verschaeve, J., Frieß, U., Pöhler, D., Vogel, L., Sander, R., and Wagner, T.: Tropospheric BrO column densities in the Arctic from satellite: retrieval and comparison to ground-based measurements, Atmos. Meas. Tech. Discuss., 5, 3199-3270, doi:10.5194/amtd-5-3199-2012, 2012.

We would like to thank the anonymous Referee 2 for the positive review of our paper and the constructive comments which, in our opinion, helped to significantly improve the quality of our manuscript.

The answers to the comments by Referee 2 are compiled in this document. The list also contains the references to changes in the manuscript. Comments of Referee 2 are denoted "R2C", typed in normal face, and coloured blue. Author comments are denoted "AC" and an *italic font* is used. For any text in the either old or new manuscript, **bold face** is used in the author comments.

Furthermore, minor corrections (typos), which have been corrected in the new manuscript, are summarised in the answers for Referee 1.

R2C: In this paper, the authors report on a new approach to derive tropospheric BrO columns from nadir observations of UV/visible satellite instruments. The novel aspect of the retrieval is to use a parametrisation of the dependence of the stratospheric BrO column on SZA, O3, and NO2 based only on the measurements themselves. In addition, a sensitivity filter is developed to identify those measurements having substantial sensitivity to the lower troposphere using O4 columns and the reflectivity of the measurements. Based on a large number of radiative transfer calculations, the same observations are also used to estimate the most appropriate airmass factor for the tropospheric column. The data are compared to an AVHRR scene and to measurements from the CALIPSO lidar to verify the cloud treatment. The resulting tropospheric BrO columns are validated against ground-based observations and very good agreement is found in several cases.
The topic of the paper is very relevant, and in spite of a significant number of publications on the retrieval and application of tropospheric BrO columns from satellite observations, there still is an on-going debate on the applicability and the reliability of these retrievals. The methods presented here are novel and have the potential to address several of the problems present in existing algorithms. The study fits well into the scope of AMT, is relevant for developers and users of polar tropospheric BrO products alike. The paper is clearly structured and generally well written in spite of some repetitions and a few difficult to follow sections. I therefore recommend the paper for publication in AMT once the comments below have been addressed.

AC: We thank Referee 2 for this summary and the positive review.

Major comments

R2C: 1. The proposed retrieval approach is impressive and has the beauty of relying solely on the measurements themselves. However, it is also complex and relies on a multitude of more or less arbitrarily selected thresholds, parametrisations, and choices made on scenarios. While most of these seem reasonable to me (although it is often hard to judge), my overall impression is that the method is much more complex than necessary. I had this impression for the algorithm determining the stratospheric mode and the selection of the partitions (does the difficulty of the problem really justify the complexity of an approach which even after reading the text twice carefully I have not yet fully understood?) but is even more obvious in the filtering approach illustrated in Fig. 8. In my opinion, a simple O4 AMF threshold of 3.75 would do as good a job as the proposed 6 parameter model using h, g0, g1, g2, R, and O4 AMF. This becomes obvious when looking at the real data in Fig. 8b. While I cannot judge if there are other scenarios where things are less simple thus justifying a complex approach, I'm convinced that it is beneficial for an algorithm to be simple and transparent to the user. I think the algorithm should be simplified or the need for the complexity be shown.

AC: We agree with the Referee that a retrieval should be as simple and as transparent as possible. On the one hand, the authors are aware that the presented algorithms for the column separation as well as the surface sensitivity filter are complex and the description of both contain many technical details. On the other hand, however, we still believe that this complexity is required to adequately address the problem. The extensive description is included in the manuscript in order to make the retrieval as transparent as possible. The technical details may be skipped by the less technically interested reader who could turn directly to the section on validation, which has been even furthermore extended following the suggestions by the Referee 2 (see below). A simplified retrieval would not be able to extract the tropospheric BrO VCD from the data as reliable as the one presented. A less detailed description would reduce the transparency of the retrieval.

With this in mind, we would like to focus on the specific Referee's comment about the filtering approach illustrated in Fig. 8. We agree with the Referee that the results presented in Fig. 8b of the manuscript may not be sufficient to justify an approach using both the radiance R and the O4 AMF as the dependence of g on R is rather small. In fact and as suspected by the Referee, this dependence may become stronger for other conditions. In particular, it depends on the observation geometry. In order to emphasise the R dependence of g in the new manuscript, we replaced Figs 8a and 8b by two plots using modelled values and data for 76° SZA and AMF_{500}^{min}=1 instead of 66° SZA and AMF_{500}^{min}=2 as in the submitted manuscript, respectively.

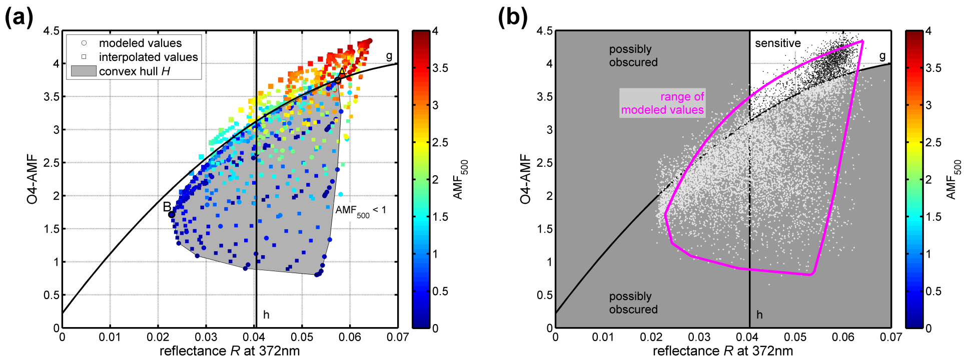

Fig. XX. (a) Modelled and interpolated (R, A_0, A_{500})-triplets for a nadir geometry at SZA=76°. The convex hull H (shaded area) including all A_{500}<1=AMF_{500}^{min} is parameterised in order to provide a threshold for the surface sensitivity filter. (b) Classification of all GOME-2 nadir observations of 2008 at the same SZA based on measured R and A_0 with a threshold of AMF_{500}^{min}=1. The described filter distinguishes between measurements sensitive to the lowest 500m of the atmosphere (black dots) and those that are possibly obscured by clouds and/or too low albedo (grey area, bright dots). The convex hull (magenta) of modelled values contains approx. 88% of the measurements.

The new plots reveal a much stronger R dependence of g. Hence, we still think that using two parameters as proxies to determine the sensitivity to the surface is justified because there are at least some measurements which would be filtered in a one-dimensional algorithm as proposed by the Referee and would be unnecessarily lost. From numerical radiative transfer simulations we may furthermore conclude that the measurements which would be lost are *potentially located at the sea-ice edge. These measurements are particularly precious for the investigation of bromine activation in the Arctic as relevant abundances of BrO are often found at the sea ice edge, in connection with open or freshly frozen leads. We do not believe that the advantage of a simpler approach would outweight losing these data. A further, and more technical, problem of the approach suggested by the Referee would be the determination of the O_4 AMF threshold as the apex of g in the new figure is outside the reasonable range of R.*

The following paragraph is added to the new manuscript in order to motivate the rather complex approach implemented. It is inserted after line 16 on page 3219:

"Figure XX furthermore illustrates the advantages of using the two parameters R and A_O instead of using just a single A_O threshold. There is a significant number of measurements located in the sensitive range featuring an O_4 AMF below point A but also at a lower radiance. These measurements would be lost if only one threshold criterion based on S_{O4} was applied. Furthermore, the measurements gained from using the two-parameter approach are particularly precious for the investigation of bromine activation in the Arctic. These measurements are more likely located at the sea-ice edge, because, at a given radiance R, A_O is maximal for clear-sky scenarios over pixels partially covered by sea-ice."

Moreover, compared to using only one parameter, the approach based on two parameters offers the advantage that AMF_{500} may be interpolated more accurately using a bilinear model (Eq. (20) in the paper). This issue is further discussed in our answer to a further comment below.

R2C: 2. Not in all cases, the choice of parametrisations is clear. For example, I'm not convinced that for a daily image of stratospheric BrO columns, the NO2 column is an important parameter. While in Fig. 6 (left), some dependence on the NO2 column is apparent, I suspect that this is really a latitude dependence and could just as well (and simpler) be parametrised by latitude. I agree that reaction with NO2 is a relevant sink for BrO but the NO2 columns measured from satellite are dominated by NO2 altitudes much above the BrO layer making a direct link between NO2 and BrO less obvious. I'm also surprised by the choice of region for determination of the relationship – by including latitudes down to 30N, polar, midlatitude and sub-tropical regimes are mixed, complicating parametrisation without clear benefit. Please comment on why these parameters and this geographical region were chosen.

AC: This comment contains two issues which need to answered separately:

1) The choice of parameters may be understood from their cross-correlation:
a) The latitudinal dependence, which is mentioned by the Referee, is already indirectly accounted for by using the SZA. When the satellite observations are limited to a narrow LOS and time interval, the SZA to latitude dependence is continuous.
b) We agree with the Referee, that the latitude range down to 30°N includes different regimes of stratospheric chemistries. However, these regimes are not intentionally mixed by the retrieval as the SZA criterion preselects these regions through its direct latitude dependence (see above). The intention for including this comparatively large latitude range is that the interpolation performed in Figs 6a and 6b becomes smoother towards the edges of the domain.

2) NO_2 VCD as a parameter for the BrO to O_3-ratio.
a) If the NO_2 dependence shown in Fig. 6a was due to the latitude, there would be no variations along the y axis, because the latitude directly depends on the SZA as explained above.
b) NO_2 is an important additional parameter for the retrieval of the BrO/O_3-ratio. On the one hand, as, for example, shown in Fig. 6(a), there are regions in the SZA/NO_2-plane where the BrO/O_3-ratio is increased by up to 15% compared to the baseline of 4.8×10^{-6}. Hence, including this dependence in the retrieval significantly improves its accuracy. On the other hand, an almost linear relation between BrO/Br_y and NO_2 was found for the stratospheric climatology presented by Theys et al. (2009) (Fig. 12 therein) also suggesting to include NO_2 in the parameterisation of stratospheric BrO.

In conclusion, the authors still believe that the parameterisation chosen is well justified and, therefore, left unchanged. No changes have been applied to the manuscript.

R2C: 3. On a similar note, most of the scenarios in Fig. 8 will never be used as they have too little sensitivity to the surface layer. The remaining scenarios above g are small in number, most of them coming from interpolating values which is a complex step in the procedure and interestingly not well reflected in the real data shown in Fig. 8b.

AC: The replacement figure for Fig. 8 (see above) illustrates the situation at 76° SZA and for a lower threshold AMF_{500}^{min}=1. It shows a larger number of simulated and interpolated scenarios above g and right of h. Furthermore, a significant number of real data now falls into this range. These measurements are particularly important for the investigation of bromine activation in the Arctic as discussed above.

R2C (continuation of 3.): From these few values, a relation is formed linking the AMF 500 to R and A0 using a linear model and "selected scenarios", but at least in Fig. 8, I cannot see a clear pattern that would justify such a model. I'm therefore again not convinced that this complex parametrisation of AMF 500 is adding accuracy to the AMF relative to just using the mean or median value. I think it would be good to show in a scatter plot how the parametrisation of the AMF and the values from the model runs and interpolations compare for the example shown in Fig. 8.

AC: The authors understand the concerns of the Referee and believe that the replacement of Fig. 8a shown above better demonstrates that the implemented algorithm can be justified. The new plot shows a clear pattern of the modelled/interpolated values for AMF_{500} in the upper right of the plot (above g, right of h): The highest values can be found in the upper right decreasing towards smaller R and O4-AMF. The simplest approximation to assign an AMF_{500} to a measurement would be to take the mean (or median) of these values as suggested by the Referee. However, this would introduce rather large systematic errors because the modelled/interpolated values vary approx. between 1 and 3.9. Therefore, the simple bilinear model is chosen in order to improve the mapping between retrieved and modelled/interpolated values.

As suggested by the Referee, the following plot shows a scatter plot on how the parameterisation of the AMF and the modelled/interpolated values compare for the above example (SZA=76°, nadir, AMF_{500}^{min}=1).

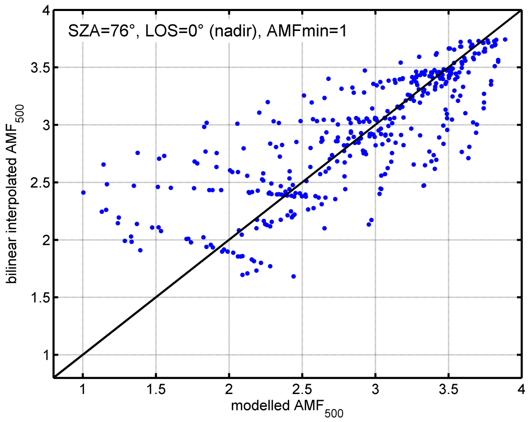

Fig. XX. Comparison between modelled/interpolated values for A_{500} and values from the parametrisation at a fixed nadir geometry of SZA=76° and a threshold of AMF_{500}^{min}=1 (same as in Fig. 8).

This plot has been added to the manuscript. It shows that the modelled and parameterised AMF_{500} are correlated and therefore justify the presented bilinear interpolation approach. This plot also reveals, that a single value (mean or median) as proposed by the Referee would add a significant systematic error to the retrieved AMF_{500} compared to the real AMF_{500}. We therefore conclude that using the two proxies (R and AMF O_4) for the determination of AMF_{500} offers the opportunity to even quantify AMF_{500} to some degree instead of using a constant value.

The following discussion of the scatter plot has been included in the new manuscript. The text is appended to paragraph (iv) at the end of line 8 on page 3220.

"Fig. XX compares A_{500} resulting from the bilinear model to the modelled and interpolated input values of the fit for one example geometry (SZA=76°, same as in Fig. 8). This plot reveals that a single value (mean or median) would add a significant systematic error to the retrieved A_{500} compared to the real A_{500}. It is therefore concluded that using the two proxies (R and A_O) for the determination of A_{500} offers the opportunity to even quantify A_{500} to some degree instead of e.g. using a constant value."

R2C: 4. The section on validation is not very convincing in its current form although it shows that the algorithm is performing as expected.

AC: The authors are convinced that demonstrating the performance of the algorithm before its application is at least as important as detailing the underlying principles. There may be some improvements possible, but this part should nevertheless be kept in the manuscript. However, we thank Referee 2 for the particular suggestions which have been applied to the new manuscript and therefore, in our opinion, improved the section on validation significantly. We certainly believe that it is now more convincing for the reader.

R2C: • Testing the column separation algorithm with a purely statistical data set with normal distributed values is in my opinion just showing that the algorithm is properly implemented. The real test would have to be done on BrO, O3, and NO2 slant columns as modelled by a radiative transfer model based on CTM profiles with some occasional BrO events added in the lower 500 m of the troposphere. This would test the algorithm on a (more) realistic ensemble of data which might not follow the idealised assumptions in the test data.

AC: We would like to thank the Referee for suggesting this useful test. We followed the suggestions and the results are described below. The authors, however, still believe that demonstrating a test of the presented algorithm using purely statistical data is valuable because it shows that its implementation was successful. We therefore intend to keep section 3.1 in the manuscript and add the results from the suggested study as an additional subsection between sections 3.1 and 3.2 in the new manuscript.

The following plots show the results for the retrieved stratospheric BrO SCD and the retrieved corresponding SCD error. It has also been included in the new manuscript.

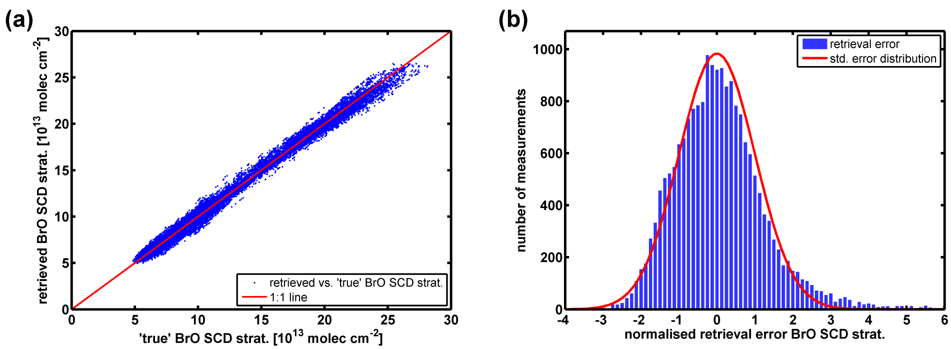

Fig. XX. Benchmark results of the column separation algorithm

using an ensemble (n=20000) of concentration profiles of BrO, O_3, and NO_2 simulated by the EMAC model. (a) Retrieved stratospheric BrO SCDs against 'true' input BrO SCD. (b) Distribution of the difference between retrieved and input BrO SCD normalised by the BrO standard deviation σ_{strat} as provided by the retrieval (see text).

Within numerical errors, the proposed algorithm is able to retrieve the partial stratospheric BrO SCD column over a wide range of SCDs (a). Furthermore, the distribution of deviations from input SCD and retrieved SCD almost perfectly match the distribution given by the retrieved standard deviation of a single measurement as shown in (b). The complete description and discussion of this study, which has been included in the manuscript, reads:

"3.2 Proof of concept of column separation algorithm using profiles simulated by EMAC

In addition to simulated measurements, it is also possible to benchmark the proposed column separation algorithm applying concentration profiles of BrO, O_3, and NO_2 simulated by a chemistry climate model (CCM). SCDs of BrO and O_3 as well as VCDs of NO_2 are computed from an ensemble of profile triplets provided by the CCM and using radiative transfer calculations. Then, the algorithm presented in Sect. 2.2 is applied on the computed SCDs and VCDs in order to retrieve again the stratospheric BrO SCDs. These BrO SCDs are compared to the original BrO SCDs and, hence, benchmarked. This approach is presented here and has two distinct advantages compared to the study in Sect. 3.1: It incorporates radiative transfer effects which may lead to deviations due to differences in the concentration profiles, and the ensemble of computed values should be more realistic.

The data basis for this study are concentration profiles of BrO, O_3 and NO_2 which were computed by the ECHAM5/MESSy Atmospheric Chemistry (EMAC) model described by Jöckel et al. (2010). This model, of which the results of a "nudged" (towards ECMWF analysis data) simulation in T42L90MA resolution are used, incorporates the Modular Earth Submodel System (MESSy) in the ECHAM5 global climate model (GCM) developed by the MPI for Meteorology, Hamburg. One distinct feature of the EMAC output is provided by the SORBIT submodel, which saves the result at the overpass times and locations of sun-synchronous satellite instruments like GOME-2 (Jöckel et al., 2010). Therefore, compared to the application of typical model output (global snapshots), a higher correlation between model and satellite measurement can be expected. It is noted that the output of EMAC used here features only a resolution of 128 times 64 grid cells in longitudinal and latitudinal direction, respectively. Therefore, model data of seven consecutive days between 22 and 28 March 2007 are used in order to increase the total number of different concentration profiles. The model profiles are filtered applying the same selection criteria as to the measurements (Sect. 2.2.2).

An ensemble of n=20000 simulated satellite measurements of BrO, O_3 and NO_2 is generated from the EMAC profiles. n is similar to the typical number of measurements in one ψ-range. Hence, the choice of n is reasonable because only nadir measurements are considered here for the sake of simplicity. A random concentration between 10 and 40 ppt is added to the lowest 500m of 50% of the randomized BrO profiles in order to simulate events of enhanced near-surface BrO. From these profiles, the total SCDs of BrO and O_3 are computed using again the McArtim model applying a purely Rayleigh atmosphere, a random surface albedo between 3% and 96%, and the respective SZA of the profile. The computation of the NO_2 VCD is trivial. Finally, the column separation algorithm is applied on the simulated column measurements in order to retrieve a stratospheric BrO SCD S_{strat} and its standard deviation σ_{strat} according to Eqs. (15) and (16), respectively.

Figure 9a correlates the retrieved S_{strat} to the 'true' stratospheric BrO SCD S^*_{strat} without the random tropospheric BrO enhancement. An almost perfect correlation

(r^2=0.99) is found between S_{strat} and S^*_{strat}. The deviation of the slope (not shown) from the 1 to 1 line is of the order of the numerical error. Hence, it can be concluded that the proposed algorithm succeeded in retrieving the correct stratospheric BrO SCD with negligible systematic bias. This finding is particularly important because it indicates that the requirement of the column separation algorithm for sufficiently similar vertical profiles of BrO and O_3 is probably also fulfilled in reality. In reality, however, additional interferences due to clouds and more complex structures of the surface albedo may arise potentially decreasing the correlation.

Finally in this study, the difference between the retrieved and original BrO SCD $\Delta_{strat}=S^*_{strat}-S_{strat}$ are compared to the σ_{strat} as provided by the retrieval. Figure 9b shows the distribution of the Δ_{strat} divided by the retrieved σ_{strat}. The red line is the normal probability density function with a standard deviation of unity. The agreement between the retrieved distribution and the model assumptions for normally distributed data is remarkable. Despite the small asymmetry, this figure demonstrates that the error computed by the proposed retrieval is a realistic estimate for the real measurement error of the separated stratospheric BrO SCD."

Furthermore, the insertion of this additional subsection required several changes elsewhere in the text:

1 Changed word order and added sentence part (p. 3222, l.6)
"[..] using data from instruments other than GOME-2: [..]"
becomes
"[..] using either simulated data or measurements from instruments other than GOME-2: [..]"

2) Another item has been added to the enumeration following the previously changed sentence (p. 3222, l. 8):
"[..] is tested on simulated measurements (Sect 3.1) and (2) using concentration profiles of BrO, O_3 and NO_2 provided by atmospheric chemistry model simulations (Sect. 3.2)."
The following numbers (2), (3), and (4) are accordingly increased by one.

3) The following sentence is included in the acknowledgements (p. 3238, l. 10) after **"Hamburg."***:*
"Patrick Jöckel (DLR) is acknowledged for providing data from EMAC model simulations."

4) The following reference has been added to the references section:

Jöckel, P., Kerkweg, A., Pozzer, A., Sander, R., Tost, H., Riede, H., Baumgaertner, A., Gromov, S., and Kern, B.: Development cycle 2 of the Modular Earth Submodel System (MESSy2), Geosci. Model Dev., 3, 717–752, doi:10.5194/gmd-3-717-2010, 2010.

R2C: • The comparison with the AVHRR data is again not adding much – reflectivity at 630 nm should have similar information as reflectivity at 337 nm as used in the GOME-2 data and the discussion of the clouds is qualitative at best.

AC: It is intentional that the comparison with the AVHRR data is qualitative. It illustrates the performance of the sensitivity filter in a realistic scenario. The authors believe that Fig. 10 of the old manuscript is an interesting visualisation contributing to the GOME-2 special issue on AMT and should therefore be kept in the final version of the manuscript. No changes have been made to the manuscript.

R2C: • The comparison with CALIPSO data is very interesting but I'm confused by the quantities shown. Why are there AMFs of 3.5 for both COT of 0 and 1.5? Why is that proving that the algorithm works? What I would like to see in such a comparison is the dependence of

BRO AMF on COT and CLT and which of the points were classified as sensitive. If the majority of the points at high COT are classified as possibly obscured, then the algorithm has performed well in detecting cloud shielding over ice as suggested in the abstract.

AC: We agree with the Referee that the presentation of the results in Sect.3.3 can be improved. Furthermore, we thank the Referee for the comment on algorithm performance, which has been added to the manuscript almost literally. Following the suggestions by the Referee, this section has been altered using a revised figure with a more detailed statistics also requiring a more extensive description and discussion accordingly. Figures 11a and 11b are now replaced by a composite of 6 subfigures illustrating the dependence of the sensitivity filter on cloud optical thickness (COT) and layer top altitude (LTA), respectively. Furthermore, the dependence of AMF500 (for a moderate threshold of AMFmin=1) on COT and LTA are shown in the new subfigures 11e and 11f, respectively.

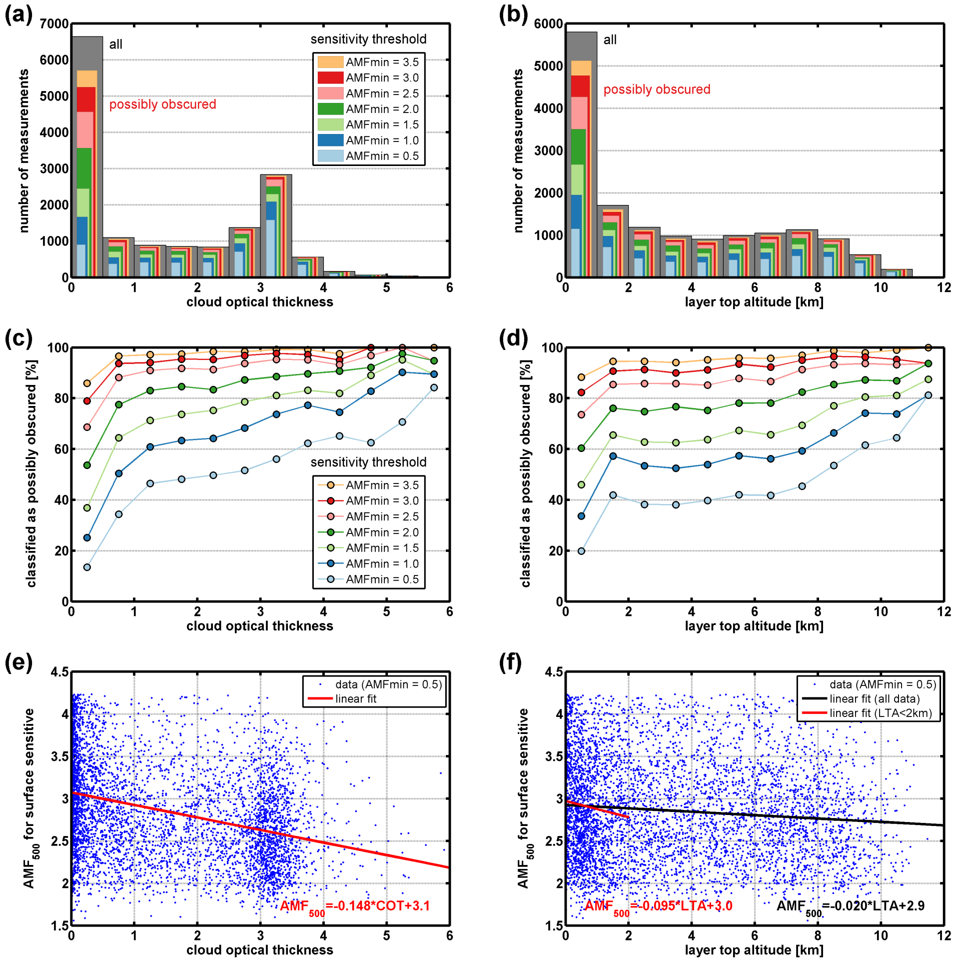

Fig. XX. Comparison between results of the surface sensitivity filter and collocated CALIPSO measurements over sea-ice: CALIPSO cloud optical thickness (COT, left column) and CALIPSO layer top altitude (LTA, right column). (a) and (b) histograms of the unfiltered measurements compared to the histograms of measurements identified as possibly obscured at different sensitivity thresholds AMF_{500}^{min}; (c) and (d) ratio of filtered measurements depending on COT and LTA, respectively; (e) and (f) respective dependence of A_{500} for AMF_{500}^{min}=0.5.

During the preparation of these new plots, the data was reanalysed resulting in an updated value for number of measurements: in the revised manuscript **"15379"** *is inserted on page 3226 in line 6 instead of the* **"17000"** *stated in the old version.*
Furthermore, the results shown in the replacement for Fig. 11 requires a revised discussion

which is printed below. It is meant as a replacement of the text starting in the second third of line 11 on page 3226 (after **"[...] collocated with."***) and ending in line 24 on the same page.*

"Figure 11 shows the comparison between the surface sensitivity filter, retrieved A_{500} and collocated CALIPSO measurements. The comparison to COT (Fig. 11, left column) is discussed first and followed by the comparison to LTA (Fig. 11, right column).

The histogram in Fig. 11a shows the distribution of all collocated measurements compared to measurements classified as possibly obscured by the sensitivity filter for different thresholds AMF_{500}^{min}=0.5, ...,3.5. There are two accumulation points, one for COT<1 and another between 3 and 3.5 COT. The first accumulation point is due to essentially cloud-free pixels and the second one is probably caused by clouds which are optically thicker than can be resolved by CALIOP leading to a systematic underestimation for these clouds. For increasing AMF_{500}^{min}, however, an increasing percentage of measurements are flagged as possibly obscured which is also shown in Fig. 11c. Figure 11c furthermore illustrates that the percentage of flagged measurements increases with increasing COT and the choice of AMF_{500}^{min} as expected. Hence, it may be concluded that the proposed surface sensitivity filter is COT selective over sea-ice and able to classify the majority of pixels with high COT as possibly obscured. The dependence of A_{500} on COT plotted in Fig. 11e approves that a larger COT on average leads to a smaller surface sensitivity.

The right column in Fig. 11 shows the respective plots for the LTA revealing a similar but weaker dependence of the sensitivity filter on LTA than COT. This is not surprising since there is presumably some cross-correlation between COT and LTA because clouds with a larger top altitude are potentially optically thicker. The histogram in Fig. 11a shows one dominating accumulation point for LTA<1km caused by cloud-free CALIOP measurements which are set to LTA=0. Therefore, the dependence of the number of measurements classified as possibly obscured (Fig. 11d) shows the strongest gradient between 0 and 2km LTA. The dependence on LTA is vanishing between 2 and 8km but increases again for high clouds (LTA>8km). Finally, Fig. 11f shows the dependence of A_{500} on LTA. The linear fit to all data shows a relatively slowly decreasing slope (black line). The slope becomes steeper, however, if only measurements below 2km LTA are taken into account (red line). Hence, A_{500} depends stronger on the presence of low clouds which results from the concentration profile of O_4 whose slope decreases with altitude.

Within the limitations of the CALIOP data set (relatively low maximum cloud optical depth, which can be measured) and of a comparison of different data-sets, it can be concluded that the algorithm is capable to identify the shielding effect of clouds over sea-ice. GOME-2 pixels with a higher average COT and LTA are more likely classified as possibly obscured. A higher threshold AMF_{500}^{min} increases the sensitivity of the filter towards filtering thinner and higher clouds. The LTA, however, was expected not to play such an important role because A_{500} is almost constant for clouds higher than 500 m.

The dependence on LTA illustrates the limits of the presented filter approach based on the utilisation of O_4 as a tracer for near-surface air. O_4 is also abundant above 500 m altitude implying the following limitations. Firstly, the shielding effect of very low and optically thick clouds may be underestimated because, in this case, S_{O4} is almost not affected. Secondly, a pixel may also be filtered although the measurement is sensitive to the BrO present in that pixel. Therefore, filtered measurements are flagged as only 'possibly' obscured. Example scenarios which appear as obscured but are in fact sensitive could be either a layer of BrO over a relatively dark surface elevated high enough to be detected anyway or near-surface BrO residing below high, optically thin clouds over a rather bright surface which may reduce S_{O4} stronger than the real A_{500}. The strength of the presented filter algorithm, however, is that measurements flagged sensitive are very likely to be actually sensitive to near-surface BrO as the first limitation can be assumed less frequent in reality than the

second."

Furthermore, section 3.4 (Discussion of validation results) has been removed from the new manuscript following the suggestion by Referee 1. The according changes are summarised in the answers for Referee 1.

Minor Comments

R2C:• page 3201, "polar regions reach about full coverage. . ." => "at polar latitudes, full coverage is reached. . ."

AC: The bracketed text now reads **"at polar latitudes, full coverage is reached once per day"** *(p. 3201, l. 18f)*

R2C:• page 3207, one point that should be briefly discussed is the fact, that the method implicitly relies on similar vertical profiles of BrO and O3 (when taking the ratios of the Scs).

AC: We agree with the Referee and included the following text in the manuscript:
"This approach implicitly relies on similar vertical profiles of BrO and O_3 which is further discussed in Sect. 3.2." *(p. 3207, l. 17)*

R2C:• page 3210, normalisation of BrO columns – why is this step needed at all?

AC: The authors agree that this step should be motivated more clearly in the paper. The normalisation is needed for several reasons:
1) The SCDs of weak absorbers measured by satellite instruments potentially contain an unknown offset due to spectral structures introduced by the diffuser used to measure direct sunlight. The issue is discussed by Richter et al. (2002) who also proposed a normalisation scheme equivalent to the one applied here.
2) Furthermore, detector degradation of the instrument may introduce further problems which is especially the case for the GOME-2 sensor as discussed by Dikty et al. (2011). The reported degradation does not only increase the statistical error of the retrieved SCDs but also introduces spectral features resulting in a trend. Dikty et al. (2011) therefore propose to normalise BrO data applying the scheme already implemented in this study.
3) The proposed retrieval algorithm for tropospheric BrO VCDs is intended to be applicable also on satellite sensors other than the GOME-2 on MetOp-A. The normalisation step introduces the possibility to homogenise the BrO data gained from the measurements of different satellite instruments.
Therefore, the normalisation of BrO columns is included in the retrieval presented in this paper.

Referee 1 furthermore suggested to move the paragraph describing the normalisation of the BrO columns to section 2.1. The authors follow this suggestion and, therefore, the following changes have been applied to the manuscript:

1) Erase paragraph enumerated ***"i"*** *(p. 3210, l. 12ff).*

2) The text below describing the normalisation of BrO columns and the definition of A_{geom} replaces the text starting with **"The fit result furnish us with total SCDs of BrO S."** *(p. 3205, l. 23) and ending with* **"over the Arctic are plotted in Fig. 1a."** *(p. 3206, l. 6):*

"The fit result provides total SCDs of BrO S, which need to be subsequently normalised for several reasons: (i) the SCDs of weak absorbers potentially contain an unknown offset due to spectral structures varying over time as discussed by Richter et al. (2002). (ii) The GOME-2 instrument suffers from sensor degradation

leading to increased statistical and, more problematic, systematic errors of the BrO SCDs as revealed by Dikty et al. (2011). (iii) The proposed retrieval algorithm for tropospheric BrO VCDs is intended to be applicable also on satellite sensors other than the GOME-2 on MetOp-A. The normalisation step introduces the possibility to homogenise the BrO data gained from the measurements of different satellite instruments.

Measured BrO SCDs are normalised to a VCD of $V_{norm} = 3.5\times10^{13}$ molec cm^{-2} within a reference sector over the Pacific Ocean as suggested by Richter et al. (2002). This normalisation is performed for each pixel-number of one scan separately (GOME-2: 32 pixels per scan, pixel-numbers correspond to discrete LOS angles). The boundaries of the reference sector are ±10 latitude and 150°E to 100°W longitude. Pixels with a footprint significantly different from the nominal ≈80×40 km^2 (narrow-mode and backscan pixels) are excluded from counting as reference measurements. The normalised SCDs S are calculated by subtracting the median difference between SCDs in the reference sector and the normalised SCD $S_{norm} = V_{norm} \cdot A_{geom}$ from the measured SCDs applying the geometrical AMF A_{geom}. While the AMF is defined as the ratio of SCD and VCD in general, A_{geom} displays an adequate approximation for stratospheric absorbers for SZA<80°. A_{geom} is defined as

$$A_{geom} = 1/\cos\theta + 1/\cos\psi \qquad (1)$$

where θ denotes the SZA, and ψ denotes the line-of-sight (LOS) angle.

Total VCDs V of BrO can be approximated from S using

$$V = S/A_{geom} \qquad (2)$$

again applying A_{geom}. As an example, total VCDs of BrO measured on 25 March over the Arctic are plotted in Fig. 1a."

3) Erase **"(i) normalization of the BrO SCDs,"** *(p. 3209, l. 26).*

4) Decrease roman enumeration **"(ii)"**, **"(iii)"**, *and* **"(iv)"** *in lines 1 to 4 on page 3210 by one.*

5) Decrease roman enumeration in (p. 3210, l. 22), (p. 3211, l. 14), and (p. 3213, l. 18) by one.

6) The following reference has been added to the list of references:

Dikty, S., Richter, A., Weber, M., Noël, S., Bovensmann, H., Wittrock, F., and Burrows, J. P.: *GOME-2 on* MetOp-A Support for Analysis of GOME-2 In-Orbit Degradation and Impacts on Level 2 Data Products – Final Report, Tech. rep., Inst. of Environ. Phys., Bremen, Germany, available at: www.eumetsat.int, doc. ITT 09/10000262 (last access: 25 September 2012), 2011.

R2C:• page 3211, "depends only slowly" => "depends only weakly"

AC: **"slowly"** *is replaced by* **"weakly"** *in line 28 on page 3211 and in line 1 on page 3212 as well in order to be consistent*

R2C:• page 3213, "still a reasonable small residual" => "still a reasonably small residual"

AC: **"reasonable"** *is replaced by* **"reasonably"** *(p. 3213, l. 8f)*

R2C:• page 3216, if O4 is integrated from sea level to top of atmosphere for the VC, what about elevated regions such as Greenland?

AC: The O4 VCD integrated from sea level to the top of the atmosphere is used as a constant factor in the calculation of both the measured and the simulated O4 AMF alike. Hence, the reduction of O4 VCD over elevated regions such as Greenland cancels out in the proposed algorithm. However, Fig. 7b depicts the measured O4 AMF calculated by using the reduced O4

VCD. This may be confusing, and, therefore, page 3216 of the manuscript is altered between line 3 and 9 as follows:
"[..] A_0 is calculated from the measured O_4 SCD, S_{O4}, using
$A_0=S_{O4}/V_{O4}*0.8$ (19)
where $V_{O4}=1.33\times10^{43}$ molec^{2}cm^{-5} is the O_4 VCD integrated from sea level to the top of the atmosphere. Equation (19) furthermore applies an empirical correction factor of 0.8 which has already been suggested by Wagner et al. (2009b) and Clémer et al. (2010) and was confirmed by sensitivity studies conducted for this work. The same definition is used for the computation of A_0, and, hence, the reduction of the real O_4 VCD over an elevated surface cancels out in the comparison between measurement and model. However, the illustration in Fig. 7b depicts A_0 measured on 25 March 2009 depending on the surface elevation."

R2C:• page 3217, "derivation the" => "derivation of the"

AC: **"of"** *is inserted (p. 3217, l. 7)*

R2C:• page 3219, "SZA = 60" – in the figure caption, it says 66

AC: The value of **"60"** *in the main body is a typo indeed and should read* **"66"***. However, as Figs. 8a and 8b are replaced as described above, the value is now changed to* **"SZA=76"** *instead.*

R2C:• page 3221, discussion of errors – this comes as a bit of a surprise here as it is not discussed elsewhere in the paper

AC: We agree with the Referee that this particular feature of the presented algorithm could be more emphasised in the paper. The estimation of the measurement errors, however, is already mentioned at several places in the manuscript (e.g. (p. 3207, l. 6), (p. 3208, l. 6), (p. 3212, l. 23), (p. 3213, l. 20ff), Fig. 6b (p. 3256)), but may be potentially overread. Therefore, the following changes have been applied to the manuscript in order to make this issue more clear:

1) Added one sentence to the Abstract (p. 3200, l. 11):
"[..] measured properties. The presented algorithm furthermore allows to estimate a realistic measurement error of the tropospheric BrO column. The sensitivity [..]"

2) Appended additional information to one sentence in the introduction of Sect. 2 (p. 3204, l. 17). The sentence now reads:
"Separation into a stratospheric and residual tropospheric BrO SCD and error estimation (Sect. 2.2)."

R2C:• page 3227, reference to Heue et al, 2011 – as the data sets used are different, this cannot really be seen as validation of this product which is fine tuned for observations of surface BrO in the Arctic

AC: It is correct that the data set used in the study by Heue et al., 2011 is different from the one presented in this paper. The first step, calculation of the BrO SCDs is, however, identical and we found the correlation shown by Heue et al., 2011 worth mentioning. The differences between the data sets are limited to the application of a different AMF, the application of the sensitivity filter and an alternative approach for extracting the BrO enhancements within the volcanic plume from the total BrO VCDs. These differences are justified by the alternative application of detecting enhanced BrO concentrations in an elevated layer within a volcanic plume.

Following the suggestion by Referee 1 to delete section 3.4 of the old manuscript anyway, also the reference to Heue et al., 2011 on page 3227 is discarded. However, the cross-validation presented by Heue et al., 2011 remains mentioned in the introduction for section 3 on page 3222. The number **"(5)"** *(p. 3222, l. 12) is deleted in the revised manuscript in order to signalise the reader that there have been previous publications using the same data which are* *however not applied in this section any more.*