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 1 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 1 are compiled in this document. The list also contains the references to changes in the manuscript. Comments of Referee 1 are denoted "R1C", 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.

After answering the comments by Referee 1, this document summarises minor corrections (typos) which have been corrected in the new manuscript.

R1C: This paper reports on a new method to retrieve tropospheric BrO columns from satellite observations in the Arctic. It appears to be a very good paper and a carefully done piece of work. The measurements are of high quality and the methodology is rigorous. Although quite technical, a lot of creative solutions are presented. In particular, the sensitivity filter is an elegant concept that nicely complements other recent studies on tropospheric BrO retrieval (mostly concerned by correcting for the stratospheric absorption). A very strong point of this paper is also the validation part, which shows unique comparisons of satellite and ground-based BL BrO columns in Arctic spring (during bromine explosion events). I believe this paper should be published, after addressing minor points (see below).

AC: We thank Referee 1 for this summary and the positive review.

Specific comments

R1C: - The paper could be improved for a better readability (especially section 2). Sometimes two consecutive paragraphs are not well linked, so it is difficult to have a smooth read. I also found the description part of the paper a bit long and a lot of mathematical formulas and symbols are not necessary. It would be better to spend few lines here and there to give more intuitive/physical explanations.

AC: We thank the Referee for this comment. We tried to optimise the paper at several places. Furthermore, we would like to add that the changes made in the manuscript based on the comments provided by both Referees also improved the readability significantly. After applying the changes listed below, we are confident that the new manuscript allows a smoother read.

1) In order to avoid confusion, all **"AMF_{500}"** *are converted to* **"A_{500}"**.
Replaced appearances of **"AMF_{500}"**:
a) in the main body of the paper: (p. 3215, l. 11), (p. 3215, l. 23), (p. 3216, l. 14), (p. 3216, l. 24), (p. 3217, l. 7), (p. 3217, l. 11), (p. 3217, l. 17), (p. 3217, l. 21), (p. 3219, l. 12), (p. 3219, l. 17), (p. 3219, l. 18), (p. 3219, l. 21), (p. 3220, l. 14), (p. 3224, l. 18)
b) in caption of Fig. 10 (three times)
c) in text changed or added as described in this document as well as the answers for Referee 2

2) replace **"and a geometric AMF"** *(p. 3206, l. 12f) with* **"using A_{geom}"**

3) erase **"on different concentration profiles of BrO and O3 and the corresponding stratospheric AMFs, but"** *(p. 3208, l. 10f)*

4) insert **"which is parameterised by the VCD of NO_2 and the SZA"** *after* **"stratospheric NO_2 chemistry"** *(p. 3208, l. 12)*

5) erase **"of BrO SCD to O_3 SCD"** *(p. 3209,l. 1)*

6) erase **"Although the stratospheric AMFs of BrO and O_3 are similar, Eq. (10) can, in addition, also compensate differences of both AMFs since z^-_0 depends also on θ and ψ."** *(p. 3209, l. 9f)*

7) replace **"thus"** *(p. 3209, l . 15) with* **"so that"**

8) replace **"a mean background stratospheric BrO/O3-SCD ratio z^-_0"** *(p. 3210, l . 2f) with* **"z^-_0 and σ_0"**

9) replace **"ratios"** *(p. 3210, l . 4) with* **"z'"**

10) replace **"the stratospheric BrO SCD"** *(p. 3210, l. 5) with* **"S_{strat}"**

11) replace **"and can be justified since the stratospheric chemistry usually only changes slightly within one week"** *(p. 3210, l. 10f) with* **"and relies on a stratospheric chemistry changing only slightly within one week"**

12) insert **"background"** *(p. 3211, l. 14) after* **"BrO/O_3"**

13) replace **"the present discussion,"** *(p. 3211, l. 27) with* **"this paper"**

14) replace **"points at which z is given"** *(p. 3212, l. 2) with* **"measured z"**

15) erase **", Eq. (11),"** *(p. 3212, l. 12)*

16) insert **","** *after* **"Now"** *(p. 3213, l. 10)*

17) erase **"represented by AMF500"** *(p. 3215, l. 11)*

18) erase **"directly"** *(p. 3215, l. 22)*

19) insert **", A_O"** *after* **"O_4 AMF"** *(p. 3215, l. 24)*

20) replace **"The O_4 AMF, A_0,"** *(p. 3216, l. 3) with* **"A_O"**

21) replace **"Finally, the AMF for the boundary layer A^{meas}_{500} is derived from the same modelled values depending on R and A_O."** *(p. 3216, l. 20f) with* **"Finally, the AMF for the boundary layer A^{meas}_{500} is depending on R and A_O."**

22) replace **"each set of viewing geometries"** *(p. 3217, l. 2) with* **"each viewing geometry"**

23) delete **"through interpolation"** *at (p. 3217, l. 26) and it insert after* **"cloud cover"** *(p.3217, l. 27)*

24) replace **"all triples"** *(p. 3220, l. 7) with* **"all modelled and interpolated triples"**

25) replace **"Additionally,"** *(p. 3222, l. 12) with* **"It is noted that"**

R1C: - Section 2.2. It would be good to say why 'O3 is chosen as a tracer for the stratospheric partial column' or give a reference.

AC: We agree with the Referee that this is slightly unclear at this point. We therefore replaced **"O3 is chosen as a tracer for the stratospheric partial column."** *(p. 3207, l. 12) in the old manuscript with*
"Two substances, O_3 and NO_2, are used to parametrise S_{strat} similar to the approach initially proposed by Theys et al. (2009) but without utilising any model output. O_3 is chosen as a parameter for tropopause dynamics whereas NO_2 is used as a parameter for variations in the stratospheric chemistry."
This description repeats some of the information already given in lines 19 through 21 on page 3202 of the old manuscript. We think that repeating this information may help the reader to better understand the steps described in Sect. 2.2.1.

R1C: - Section 2.3.1. 'It is noted, that the exact value of the BrO mixed layer height may differ in reality, but radiative transfer simulations showed its choice is not critical for the presented considerations' please specify that this is because of the high albedo conditions in Arctic

AC: We agree with the reviewer that the weak dependence on mixed layer height is due to the high surface albedo. The sentence cited by the Referee (p. 3215, l. 7ff) has therefore been changed to:
"It is noted, that the exact value of the BrO mixed layer-height may differ in reality. Radiative transfer simulations, however, showed that its choice is not critical for the presented considerations because the sensitivity of nadir measurements is only slightly depending on altitude above surfaces with high albedo which are typical for polar regions."

R1C: - Section 2.3.1: I found the explanation of the sensitivity filter a bit hard to understand. It would be good to explain (intuitively and in simple words) how the two measured parameters (Reflectance and O4) help to determine the AMFs.

AC: **"On the one hand, R is a well suited measure to discriminate either clouds/ice (bright) and ocean/land (dark). A_o, on the other hand, helps to discriminate between ice and clouds and furthermore provides information about the height and optical thickness of potential clouds."** *These two sentences have been included in the manuscript after* **"The two proxies used in the proposed algorithm are the reflectance *R* and the O4 AMF."** *(p. 3215, l. 24)*

R1C: - Table 3. It is hard to understand why these values have been used. Why these settings and not others? Are they representative enough?

AC: We agree with the Referee that the choice of these settings may appear a bit arbitrary. Through extensive radiative transfer simulations before calculating the entire look-up tables, the presented settings were found to be largely representative for the scenarios. They are capable of being differentiated by the sensitivity filter relying on the radiance and the O_4 AMF. Two settings (0-1km, OD 20 and 3-4km, OD 50) were included after the entire radiative transfer calculation was performed. The results of both additional settings were found critical for the subsequent automated interpretation of the results that were performed for all geometries (Table 2). Computing the entire radiative transfer again was not possible due to the extensive run-time of our code. The resulting settings, however, were found to provide sufficient information to parameterise the sensitivity filter look-up tables.

In order to clarify the chosen approach, the following paragraph has been included in the manuscript on page 3217 after 22.

"Before the entire look-up tables were calculated, the scenarios summarised in Table 3 were found to be largely representative for the presented sensitivity filter through extensive radiative transfer simulations. However, two scenarios (0-1km, OD 20 and 3-4km, OD 50) were added at a later stage in order to further improve the accuracy of the algorithm. It is noted, that future studies may benefit from using even more selected scenarios yet increasing the computational cost of the algorithm."

R1C: - Section 2.3.2. Specify the wavelength used for the AMF500 calculation.

AC: The following sentence has been inserted after the first sentence of (i) in line 18 on page 3217 of the manuscript. It compiles the used wavelengths:
"[..] (Deutschmann et al., 2002). Two different wavelengths are used in the radiative transfer calculations: R is derived from radiative transfer simulations at 372nm, whereas A_O and A_{500} are simulated at 360nm. For each LUT [..]"

R1C: Fig.11. I find the message very qualitative. It only shows that the sensitivity filter is not failing and does not give any limits of its applicability (subjective to the value of AMF threshold). I feel it could be expanded a bit.

AC: This issue has also been raised by Referee 2 and is discussed in detail in our answers for Referee 2. Figure 11 has been altered according to the comments by Referee 2. The changes in the corresponding section 3.3 are summarised in the accompanying document. The new text also discusses the limitations of the proposed filter algorithm (last paragraph of the new section 3.4: Comparison to CALIPSO cloud data).

Minor comments

R1C: - Some acronyms are not defined (e.g. LP-DOAS, CIMS,..). Please check the entire manuscript.

AC: The following changes have been applied to the new manuscript in order to meet the Referee's recommendation:

1) Added definitions for LP-DOAS, MAX-DOAS and CIMS. The sentence in lines 14 to 17 on page 3201 now begins with
"Compared to ground-based measurement techniques like long-path DOAS (LP-DOAS) ([..]), multi-axis DOAS (MAX-DOAS) ([..]), or chemical ionization mass spectrometry (CIMS) (Liao et al., 2011), observations [..]"
Accordingly, the obsolete definitions of LP-DOAS and MAX-DOAS have been deleted in line 2 on page 3228 and the sentence starting in line 1 now reads
"The data-set from the Amundsen includes LP-DOAS and yet unpublished MAX-DOAS measurements."

2) Added **"the first"** *for the discrimination between GOME and GOME-2 (p. 3205, l. 10) and the definition of SCIAMACHY (p. 3205, l. 11). The sentence now reads starting in line 10:*
" [..] combines the standard wavelength ranges used for the first GOME ([..]) and the Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) ([..]) instruments and encompasses [..]."

3) Changed **"OMI"** *into* **"the Ozone Monitoring Instrument (OMI)"** *in the conclusions (p. 3235, l. 16).*

4) Definition for the FRESCO acronym also FRESCO (p.3203, l. 10):
"[..] The ice-mode of FRESCO+ (Fast Retrieval Scheme for Clouds from the Oxygen A band, Koelemeijer et al., 2001, Wang et al., 2008) derives [..]"

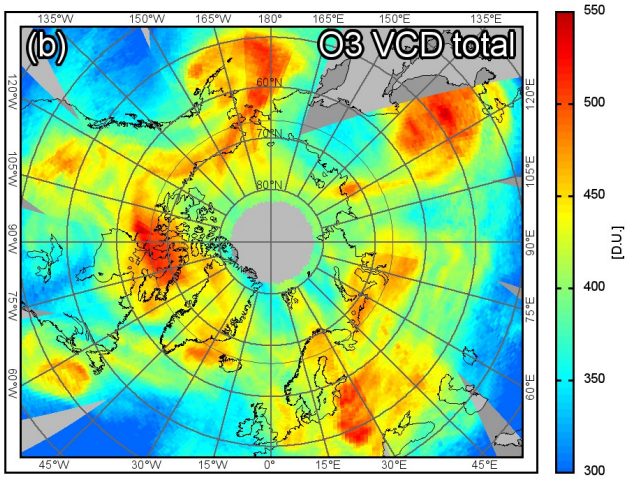
Accordingly, the following reference has been included in the references section:

Koelemeijer, R. B. A., Stammes, P., Hovenier, J. W., and de Haan, J. F.: A fast method for retrieval of cloud parameters using oxygen A band measurements from Global Ozone Monitoring Experiment, J. Geophys. Res., 106(D4), 3475-3490, doi:10.1029/2000JD900657, 2001.

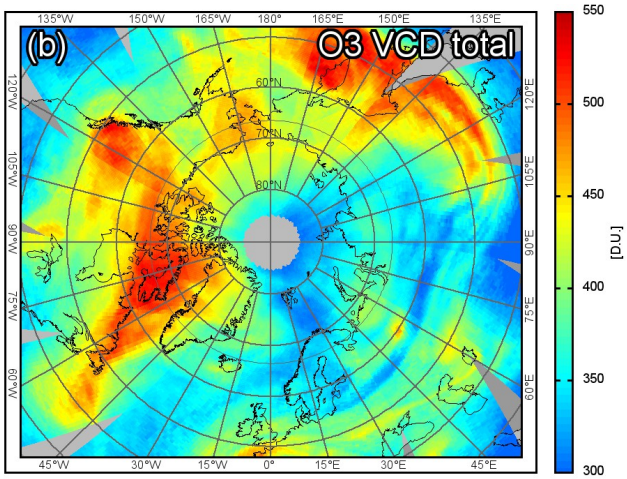
R1C: - Fig1b. It would be better to express the O3 VC in Dobson units (the most used unit).

AC: The authors would like to follow this suggestion and replaced Figs. 1b and 2b with the following plots expressed in dobson unit (D.U.), respectively.

new Fig. 1b



new Fig. 2b



It needs to be noted that the retrieval presented in this paper has not been optimised for the quantitative retrieval of the O3 column and, hence, the above figures are potentially biased, especially because no adequate AMF from RTM is used. This bias, however, will cancel in the retrieval as long as the O_3 SCD retrieved from analysing the GOME-2 spectra are linearly correlated to the real O_3 SCD.

As the BrO/O3-ratio is furthermore expressed dimensionless, the following sentence needs to be appended to Eq. (4) in the new manuscript:

"[..] (4) [line break] where $S_{strat,O3}$ is expressed in molec cm^-2 using the definition of the Dobson unit (1DU=2.69x10^16 molec cm^-2)." *(p. 3207, l. 15)*

R1C: - Section 2.2.1: 'It turns out that the BrO/Bry concentration ratio, which is typically of the order of 0.6, is primarily depending on the stratospheric NO2 concentration'. -> 'It turns out that the BrO/Bry concentration ratio, which is typically of the order of 0.6 during daytime, is primarily depending on the stratospheric NO2 concentration'.

AC: Following the Referee's suggestion, **"during daytime"** *is inserted and the sentence now reads in the revised manuscript:*
"It turns out that the BrO/Br_y concentration ratio, which is typically of the order of 0.6 during daytime, is primarily depending on the stratospheric NO_2 concentration" *(p. 3208, l. 16f)*

R1C: - Section 2.2.2. The description of the normalization of the BrO SCDs would be better placed in Section 2.1.

AC: In order to comply with the Referee's recommendation, the description of the normalisation is now placed in Section 2.1. Furthermore, the paragraph now contains a

rationale for the normalisation as required by the Referee 2. The changes applied to the manuscript are therefore summarised in the answers for Referee 2.

R1C: I would skip Fig.3.

AC: We agree with the Referee and Figure 3 has therefore been deleted in the revised manuscript. Furthermore, the sentence **"The geographical area encompassing *T* is illustrated in Fig. 3."** *(p. 3211, l. 12f) is removed.*

R1C: - In my opinion, Section3.4 is not necessary. It only repeats what has already been explained in the previous sections.

AC: Following the suggestion by Referee 1, section 3.4 has been removed from the revised manuscript. Subsequent minor changes are detailed in the answer for Referee 2.

Additional minor corrections applied to the new manuscript:

1) Appearances of **"normalized"** *have been replaced by* **"normalised"***:*
(p. 3205, l. 19), as well as in the altered paragraph describing the BrO SCD normalisation (see answers for Referee 2).

2) **"parametrized"** *has been replaced by* **"parametrised"***(p. 3215, l. 23)*

3) replace **"its"** *(p. 3209, l. 23) with* **"is"**