We thank Ross Salawitch for his review, and we appreciate his non-anonymous comments and suggestions.

Before addressing each of the reviewer's comments, we would like to remind the readers that our paper is submitted to AMT in the "Tropospheric profiling: integration of needs, technologies and applications" special issue. Within the scope of that issue, the paper describes a new algorithm for the retrieval of trace gas vertical profiles from aircraft-borne UV/vis limb measurements. The method is particularly applied to the retrieval of BrO vertical profiles and, within that framework results are compared to BrO columns retrieved from satellite measurements (Table 1). We understand that the satellite retrieval of tropospheric BrO is a matter of current discussion (e.g., Salawitch et al. 2010, Theys et al. 2011) and it seems to be the main focus of this review. However, the goal of our paper is not to address particular issues concerning satellite retrievals which are emphasized in other publications e.g. Rozanov et al. (2010), the reviewer's work (Salawitch at al., 2010) and the recently published work of Theys et al. (2011).

Salawitch, R. J., Canty, T., Kurosu, T., Chance, K., Liang, Q., da Silva, A., Pawson, S., Nielsen, J. E., Rodriguez, J. M., Bhartia, P. K., Liu, X., Huey, L. G., Liao, J., Stickel, R. E., Tanner, 10 D.J., Dibb, J .E., Simpson, W. R., Donohoue, D., Weinheimer, A., Flocke, F., Knapp, D., Montzka, D., Neuman, J. A., Nowak, J. B., Ryerson, T. B., Oltmans, S., Blake, D. R., Atlas, E. L., Kinnison, D. E., Tilmes, S., Pan, L.L., Hendrick, F., Van Roozendael, M., Kreher, K., Johnston, P. V., Gao, R. S., Johnson, B., Bui, T. P., Chen, G., Pierce, R. B., Crawford, J. H., and Jacob, D. J: A new interpretation of total column BrO during Arctic Spring, Geophys. Res. Lett., 37, L21805, doi:10.1029/2010GL043798, 2010.

Rozanov, A., Kühl, S., Doicu, A., McLinden, C., Puķīte, J., Bovensmann, H., Burrows, J.P., Deutschmann, T., Dorf, M., Goutail, F., Grunow, K., Hendrick, F., von Hobe, M., Hrechanyy, S., Lichtenberg, G., Pfeilsticker, K., Pommereau, J. P., Van Roozendael, M., Stroh, F., and Wagner, T.: BrO vertical distributions from SCIAMACHY limb measurements: comparison of algorithms and retrieval results, Atmos. Meas. Tech. Discuss., 3, 5079-5178, doi:10.5194/amtd-3-5079-2010, 2010.

*Theys, N., Van Roozendael, M., Hendrick, F., Yang, X., De Smedt, I., Richter, A., Begoin, M., Errera, Q., Johnsto*n, P. V., Kreher, K., and De Maziere, M.: Global observations of tropospheric BrO columns using GOME-2 satellite data, Atmos. Chem. Phys., 11, 1791-1811, doi:10.5194/acp-11-1791-2011, 2011.

Nevertheless, we agree that providing more details concerning the applied satellite retrievals may enhanced the clarity of the paper. Thus, in the revised manuscript more information concerning the satellite retrievals is given which we hope is suited for improving the manuscript according to the reviewer's request.

All comments are addressed below.

RC: Overall evaluation:

This paper provides a new approach for retrieving profiles of the atmospheric abundance of the trace species bromine monoxide (BrO): first, the measured radiance isused to estimate the light path through the atmosphere, considering both Rayleigh and and Mie scattering. Once the scattering profile has been determined, the profile of BrO is obtained, using an approach (regularization) that does not involve specification of an a priori. Finally, important scientific questions are addressed using retrieved profiles of BrO.

I found the material in Sections 2 and 3 to be well written, interesting, and quite important. Although this is not my specific area of expertise, the authors seem to have advanced the state of the art for remote sensing of an important atmospheric species.

AC: We are grateful to this comment.

RC: In Section 4, entitled "Results and discussions", the authors use four profiles of BrO, retrieved on two days, to address scientific problems of extreme interest to a subset of the atmospheric sciences community: whether satellite observations of total column BrO are consistent with profiles obtained by sub-orbital techniques and whether the satellite is sensitive to BrO in the boundary layer (BL). Section 4 leads to statements such as:

"These findings are well in agreement (sic) with satellite and balloon-borne soundings of total and partial BrO atmospheric column densities" (abstract)

and

"overall, worth mentioning is also that compared to airborne values, the satellite retrieval does not systematically underestimate BrO, a behavior one would expect if the satellite detection of near surface BrO would be systematically obscured in the Arctic, e.g., by scattering due to aerosol and cloud particles" (page 3951).

Neither of these statements are well supported by material in the paper. For instance, there is no meaningful quantitative analysis of the agreement, or lack thereof, between the estimates of VCDtotal given in Table 1 by the airborne and satellite platforms.

AC: For our reactions to the reviewer's overall comment see below.

RC: The paper gives very terse treatment to how several important components of Table 1 were found: i.e., one sentence is devoted to VCDstrat. Important details such as latitude/longitude, SZA, etc of the measurements are completely lacking.

AC: In the revised manuscript, the Table 1 now includes the geo-location (latitude and longitude), time (UT) and solar zenith angle of the flights. For the comment related to VCDstrat. please see bellow.

RC: In many ways, this paper reads like a novel where many chapters are used to develop a well nuanced, complicated plot. Then, the story concludes in a short chapter, in which several critical new details are abruptly introduced. I have an unsettled feeling upon reading a novel written in such a manner.

Similarly, for the paper under review, I am unsettled. If the paper was published as submitted, the strong statements resulting from section 4, which have not been adequately demonstrated, will likely be quoted in many subsequent papers, either by this team or by others. This would be a disservice to the atmospheric sciences community.

Conclusions such as:

a) consistency between the satellite and sub-orbital measurements of the BrO

b) satellite retrievals of column BrO are not obscured by clouds should be suitably demonstrated, including a treatment of uncertainties and a description of the context of the observational setting, or else Section 4 (and the attendant conclusions) should be dropped. Perhaps Atmospheric Measurement Techniques is not the venue for the type of science discussed in Section 4. If so, perhaps this material should be saved for a subsequent paper. Otherwise, Section 4 must be expanded considerably. Below, I will address some of the elements lacking in Section 4, which the author team is welcome to consider for either a revision to AMT or for submission to an alternate journal. I believe this paper requires substantial revisions before it will be suitable for publication in AMT.

AC: After describing the trace gas retrieval algorithm in Section 2, and testing it in Section 3, Section 4 focuses on the application of the method to inferring BrO volume mixing ratios from aircraft-borne UV/vis measurements performed in limb viewing geometry. Hence we feel that Section 4 may not only improve the quality of the paper, but it may also complete the study in describing the algorithm, its application and validation. In that respect and for a tight comparison with the satellite data, we selected only low-cloud scenarios (i.e., 1 April 11:25UT and 8 April 14:30UT) based on the visual inspection of a video recorded with a camera on board the Falcon (Table 1). Please note that the present paper does neither intend to detail the satellite retrieval, nor the issues arising from it (e.g., possible cloud interference). For those matters, as well as for error estimations regarding satellite retrieval, please refer to specific satellite papers (e.g., Rozanov et al., 2010; Theys et al., 2011).

In our study, we found a reasonable agreement (i.e., within error bars) in the two (cloud-free) passages selected for airborne and satellite VCD comparison (i.e., 1 April 11:25UT, and 8 April 14:30UT), see also below.

RC: Please note Sections 2 and 3 are EXCELLENT. This material, by itself, constitutes a highly appropriate contribution for AMT.

AC: We appreciate the reviewer's opinion.

RC: But this paper is the tale of two stories: a novel retrieval (Sections 2 and 3) and science related to this retrieval (Section 4), and it is the science related to the retrieval that, I believe, either needs to be expanded or perhaps omitted.

Major Comments:

1. Key details must be added to Section 4

Table 1 of Section 4 compares total column BrO retrieved from GOME to the sum of the BrO column in the troposphere inferred from the scanning mini-DOAS instrument plus the BrO column in the stratosphere inferred from prior balloon campaigns.

There is so much about the discussion of Table 1 that is lacking that it is hard to know where to begin. Nonetheless:

The description of VCDstrat (the BrO column in the stratosphere) is way too terse. The bottom of page 3950 states "In addition, estimates of stratospheric BrO columns, inferred from balloon measurements (Dorf et al., 2006) are provided after adapting them for similar tropopause height". Table 1 of Dorf et al. (2006) lists 14 profiles. Which were used? Was the sensitivity of BrO to O3 and NO2 considered? If so, how? Was the BrO profile "slided" or "stretched" to match the tropopause height at the specific locations?

AC: The mentioned sentence has been changed to "In addition, estimates of stratospheric BrO columns inferred from balloon measurements are provided. Those measurements were performed in Kiruna (67.9° N, 21.1°E) on 23 March 2003 and on 24 March 2004. Details of those balloon flights, characterized by a tropopause altitude of 8 km and 8.9 km (resp.), are given in the work of Dorf et al., 2006".

Regarding corrections for the estimation of the airborne VCDstra values, see below.

RC: Given the nature of atmospheric transport, the sensitive dependence of Bry (and hence BrO) on past photolytic history and possible contributions from VSLs (very short lived substances), neither sliding or stretching a BrO profile is particularly appealing, especially as a co-author of the paper, N. Theys, has developed a climatology of stratospheric BrO that seems to be a better choice for specifying VCDstrat. Upon revision, a detailed description of VCDstrat should be provided. If the balloon profiles are used as baseline, sensitivities to O3, NO2, SZA, and non-linear transport effects in the lowermost stratosphere (i.e., whether a profile in March from Kiruna should be both "slid" and "stretched" to simulate conditions in April near Spitsbergen) should be discussed. If the BrO climatology of Theys et al. is used, the sensitivity of the resulting BrO to Bry from VCDs should be discussed.

AC: The climatology of Theys et al. (2009) is indeed used to estimate the stratospheric VCD of BrO. However it is not used for estimating the airborne values, but the satellite (BIRA) VCDstrat. Note that some of the BrO profile measurements used to validate Theys et al. (2009) climatology are actually those presented by Dorf et al. (2006).

The estimates of VCDstrat for the aircraft-borne observations are based on the SZA (i.e., balloon data are photochemically corrected to 80°, see Dorf et al. 2006 and Butz et al. 2006), and the altitude of the thermal tropopause.

Data provided in Table 1 of the manuscript are based on the following tropopause heights as inferred from on-board measurements (in the case of the aircraft and the balloon), and from climatology (in the case of satellite-BIRA):

Platform	Date	Tropopause altitude (km)
Aircraft	1 April 2007	7
	8 April 2007	8.5
Balloon	(23 March 2003) 1 April 2007	8
	(24 March 2004) 8 April 2007	8.9
Satellite (BIRA)	1 April 2007	7.5
	8 April 2007	8.7

Note that the MPIC satellite retrieval does not include tropopause height per se (see below).

Considering the tropopause altitudes detailed above, the values inferred from the 23 March 2003 balloon flight are given as VCDstra for 1 April 2007 in Table 1, and the BrO column inferred from the 24 March 2004 balloon flight, is used as VCDstra for 8 April 2007 (now also mentioned in the caption of Table 1).

Note that, since we lack further information regarding dynamics, the airborne VCDstrat given in the manuscript (Table 1) do not include any tropopause altitude correction. Note that the airborne VCDstra provided for the 1 April 2007 should be taken with caution due to the difference in the tropopause height (i.e. 7 km inferred from the aircraft, 8 km inferred from the balloon). That difference in the thermal tropopause altitude is estimated to affect the airborne BrO VCD_{TOTAL} values provided in less than $0.5 \cdot 10^{13}$ molec/cm^2.

RC: Table 1 gives four estimates of VCDtrop from airborne sampling: one profile on 1 April 2007 and three profiles on 8 April 2007. Perhaps I missed it, but the paper does not seem to describe the flight of 1 April in any manner. Where was the profile acquired? At what latitude, longitude, and SZA (UTs are given in Fig 9 . . . would be nice for this to be converted to SZA for Table 1)? Was the sampling for clear sky conditions?

AC: The geo-location (latitude and longitude), time (UT) and solar zenith angle for the flight of 1 and 8 April are now included in Table 1. For further description of those aircraft deployments, the reader is kindly referred to thesis of Prados-Roman (2010) in Table 1 and Figure 9 of the revised manuscript.

Prados-Roman, C.(2010): Aircraft-borne spectroscopic limb measurements of trace gases absorbing in the UV-A spectral range. Investigations of bromine monoxide in the Arctic troposphere, Ph.D. thesis, Heidelberg University, http://archiv.ub.uni-heidelberg.de/volltextserver/velltexte/2011/11451.

In particular, the 11:25UT (1 April 2007) aircraft sortie took place over sea ice and during clear-sky conditions. Hence it is chosen for the satellite comparison exercise shown in Table 1. This is now clearly stated on Page 3950, line 29: “In the cases of the cloud-free passages flown over the sea ice of 1 April 2007 (11:25 UT) and 8 April 2007 (14:30 UT), our airborne data (IUP-HD) are compared to the satellite columns (MPIC and BIRA).”.

RC: How does the comparison of IUP-HD epsilon M versus in situ extinction profiles look for this flight?

AC: As mentioned in Section 3.2, the comparison of the aerosol extinction coefficient retrieved from remote sensing measurements, with that inferred from in situ measurements involves some limitations (Page 3942-3943). In the study, this comparison aims at investigating the quality of our radiative transfer calculations and it is applied to the case study (14:30 UT, 8 April 2007). Results shown in the section 3.2 address the relevance of maintaining a consistent approach toward the RT calculations. Thus, the aerosol extinction coefficients inferred from the in-situ measurements are not used further on.

RC:

c) Estimates of VCDtotal from GOME-2 are given for two groups in Table 1. These estimates barely agree within the respective uncertainties. On page 3950, no references are given for the satellite retrievals of VCDtotal (the Theys studies are, to my knowledge, modeling studies and not retrieval studies).

The notion that the satellite radiances alone can be used to separate the stratospheric and tropospheric contributions to BrO is new. The paper must, upon revision, provide a lot more detail or else appropriate citations. One particularly important aspect, the use of "a linear relationship between measured O3 and stratospheric BrO slant columns" to arrive at a stratospheric correction appears to have been first described by a paper in circulation at the time of submission:

http://www.agu.org/journals/gl/gl1021/2010GL043798/

yet neither this paper, or any others, are cited for this important detail.

AC: Table 1 aims at the comparison of airborne BrO VCD, with VCD inferred from satellite measurements and not at comparing satellite retrievals (done by e.g. Rozanov et al. 2010). The fact that two different satellite groups are included in the inter-comparison exercise may help to indicate that, in the two selected cloud-free scenarios, the airborne and the satellite BrO VCD fall within the error margins, regardless the retrieval method applied to the satellite measurements. This is now mentioned in Page 3950, line 15:"Note that two different satellite retrievals are considered for this inter-comparison exercise in order to investigate the consistency of the airborne and the satellite BrO VCD regardless the retrieval method applied to the satellite measurements.".

Concerning the different satellite approach to distinguish between tropospheric and stratospheric BrO columns, the BIRA group uses the climatology presented by Theys et al. (2009). More details are given in the study of Theys et al. (2011). Note that in the corrected manuscript the reference to the dissertation work of Theys (2010), referred to as Theys (2010a) in the old manuscript, is erased. Instead, the work of Theys et al. (2011)- recently published- is cited.

The MPIC team uses a slightly different method. The stratospheric contribution to the measured slant column is estimated using a filter algorithm based on statistical ensembles rather than stratospheric BrO inferred from a climatology. The method is based on the following assumptions:

(1) There is a linear correlation between stratospheric O_3 and BrO slant column density (Salawitch et al, 2010). This implies identical radiative transfer, which is an approximation best fulfilled when the information about the stratospheric column is most needed, i.e. the tropopause is low and the stratospheric contribution to the total column is large.

(2) Similarly to the parameterization of the BASCOE climatology (Theys et al. 2009, Theys et al. 2011), the ratio of the slant columns of BrO and O_3 is also depending on both the BrO/Bry chemistry altered by the stratospheric concentration of NO_2, and the solar zenith angle as a proxy for the photochemical situation of the probed air mass.

(3) Apart from stratospheric photo-chemistry, deviations towards a higher BrO/O_3 indicate enhanced BrO below the tropopause.

However, a retrieval based on these three basic assumptions will not be able to clearly distinguish between any background BrO in the troposphere and the stratosphere (i.e., the VCDstrat may contain free tropospheric BrO). Therefore this approach merely allows us to identify and study observations that have an 'above normal' BrO column density (indicated as * in Table 1).

Applying the assumptions (1)-(3), the MPIC retrieval includes the following steps. Ratios of all slant column densities of BrO and O_3 acquired during 5 days centered around the evaluated day are computed and binned according to the corresponding solar zenith angle and the vertical column of NO_2. The partitioning begins with both domains divided into 10 intervals and then iterating the boundaries in order to get a similar number of measurements in each bin. This procedure approximately leads to 1000 measurements per bin and bin sizes of about 5 degrees and 10^{14} molec/cm^2, respectively. Only measurements with a significant tropospheric contribution to the NO_2 column are masked out. Then, if the distribution of the O_3/BrO ratios within one bin is significantly skewed towards high BrO/O_3 ratios, a filter is applied to successively remove these ‘outliers’ until the skewness is close to zero. The mean of the remaining distribution is interpreted as the ratio between the purely stratospheric of BrO per O_3 slant column density for the considered selection with respect to SZA and NO_2 column. Finally, the mean values of all bins are interpolated on the measurements using the O_3 slant column, the NO_2 vertical column, and the solar zenith angle as an input in order to derive a stratospheric slant column of BrO. The difference between the measured total SCD and the interpolated stratospheric SCD of BrO yields a tropospheric SCD. The vertical column is then derived by applying an tropospheric air mass factor calculated assuming a tropospheric box profile.

Although the MPIC satellite retrieval is under current development, the BrO columns obtained through this approach have already been compared to ground measurements. In the poster presented in AGU 2008 meeting by H. Sihler (see attachment), results show good correlation between satellite and LP-DOAS measurements performed from aboard the Amudsen ice-breaker (Pöhler et al., 2010). This inter-comparison gives us confidence on the potential of the MPIC retrieval. Also a manuscript describing details of the retrieval is in preparation.

In the corrected manuscript the MPIC retrieval method is now briefly described. Lines 9-15 (Page 3950) are now changed to: “The MPIC team uses a slightly different method under current development. In this case the stratospheric contribution to the measured slant column is estimated using a filter algorithm based on statistical ensembles. The method relies on the following assumptions: (1) There is a linear correlation between stratospheric O_3 and BrO slant column densities (Salawitch et al, 2010). (2) Similarly to the parameterization used by the BIRA-IASB team (Theys et al. 2009, Theys et al. 2011), the ratio of the slant columns of BrO and O_3 depends on the BrO/Bry chemistry altered by the stratospheric concentration of NO_2, and on the solar zenith angle. (3) Apart from stratospheric photo-chemistry, deviations towards a higher BrO/O_3 indicate enhanced BrO below the tropopause. However, a retrieval based on these three basic assumptions is not able to clearly distinguish between any background BrO in the troposphere and the stratosphere (i.e., the VCDstrat may contain free tropospheric BrO). Therefore the MPIC approach merely allows us to identify and study observations that have an 'above normal' BrO column density (indicated as * in Table 1).”

RC: The abstract states “these findings are well in agreement (sic) with satellite and balloon-borne soundings of total and partial BrO atmospheric column densities” which follows a statement on page 3951 that “within the limits of experimental errors, the integrated BrO column amounts using the airborne and the satellite approaches compare reasonably well”. For 1 April 2007, the sub-orbital column (6.9 +/- 1.2) x 10ˆ13 molec/cm2 is in much better agreement with the MPIC satellite retrieval (6.7 +/- 1.9) than the BIRA estimate (7.9 +/- 2.3). I understand that, within error bars, all elements agree. However, on 8 April, the two airborne profiles that do not represent lower limits, with columns of 9.1 +/- 1.8 and 11.0 +/- 2.1, agree much better with the BIRA value (9.0 +/- 2.3) than the MPIC estimate (7.0 +/- 2.0). Indeed, 11.0 +/- 2.1 and 7.0 +/- 2.0 do not agree, strictly speaking.

AC: The authors believe there is a mis-understanding. As stated in the submitted manuscript “no satellite data are given for the 13:00 and 15:20 UT profiles on 8 April 2007, due to the small number of satellite pixels meeting our selection criterion” (Page 3951, line 1-3).

The satellite VCDs shown in Table 1 for 8 April 2007 are inferred only from measurements performed around 14:30 UT. Results as given in the table are 7.0 +/- 2.0 x 10ˆ13 molec/cm2 (MPIC), and 9.0 +/-

2.3 x 10ˆ13 molec/cm2 (BIRA). These satellite values are meant to be compared with the aircraft data, corresponding to the 14:30 UT passage (9.1 +/- 1.8 x 10ˆ13 molec/cm2). Accordingly the phrase "these findings are well in agreement (sic) with satellite and balloon-borne soundings of total and partial BrO atmospheric column densities" reads now as "These findings agree reasonably well with satellite and balloon-borne soundings of total and partial BrO atmospheric column densities".

RC: I do not mean to split hairs but rather point out that the notion of "reasonably well" agreement is subject to much interpretation given the way the material has been presented. I am also particularly concerned about the statement, on page 3950, that "only the satellite pixels displaying the highest sensitivity to surface BrO have been kept for the comparison". Much more detail is needed about how this selection was carried out, and how such selection may effect the high level conclusions.

AC: As shown in Figure 4 of Theys et al. (2011), in the case of a surface with an albedo of 0.5 the Box Air Mass Factors become almost constant for the whole troposphere.

In our work, the selection of the satellite pixels displaying the highest sensitivity to surface BrO is based on an O_4 Air Mass Factor proxy. Due to the selection of pixels with a measured O_4-AMF exceeding a threshold of 3.7, a significant sensitivity for the lower troposphere is assured. This value has been determined by comparing GOME-2 measurements with radiative transfer simulations.

In addition we want to point out that already in the thesis work of e.g. E. Lehrer (1999) the anti-correlation between satellite BrO VCD and surface O_3 mixing ratio was analyzed (see attachment).

Lehrer, E.: Polar tropospheric ozone loss, PhD thesis, Institut für Umweltphysik, Universität Heidelberg, 1999.

Page 3950, line 23 now reads as: "Finally, based on measured O_4 airmass factors, only the satellite pixels displaying the highest sensitivity to surface BrO have been kept for the comparison."

RC: Also, the statement "background BrO in the troposphere is implicitly accounted for in the stratospheric columns and not in the tropospheric estimates" is unclear and requires further explanation.

AC: See above.

RC: Finally and most importantly, some connection between the BIRA and MPIC estimates of column BrO given in Table 1 and values of GOME-2 BrO in the literature is needed, so that the reader can relate the results to prior scientific studies.

RC: Page 3949 now includes "(e.g.,Wagner and Platt, 1998; Richter et al., 1998)".
RC:

e) Page 3938, lines 23 to 25: the statement that the particular ascent was selected because it has the simplest RT scenario of the flight raises several questions: i) how much more complicated is the RT for the other portions of the 8 April flight and for the 1 April flight? ii) how does the comparison shown in Figure 5 look for these other portions; iii) since BrO profiles are retrieved for three other profiles, besides the one with the simplest RT scenario, how is BrO affected by uncertainties in the light path for these other, more complicated scenes?

AC: The selection of the simplest RT scenarios (i.e., 1 April 11:25 UT, 8 April 14:30 UT) was based on (a) the visual inspection of the video recorded with the camera on board the Falcon aircraft, (b) on the in situ measured clouds and aerosols, and (c) on the signal-to-noise of the mini-DOAS measurements. The uncertainties of the BrO profiles retrieved from measurements performed during more complicated cases (i.e., 8 April 13:00 UT and 15:20 UT) are estimated based on forward modeling and sensitivity runs (e.g., Page 3949, line 12).

RC:

f) Page 3941, lines 17 and 18: why ammonium sulfate? What does aged mean? (no reference are given!). I though soot was common in the Arctic due to Siberian fires. How does the different absorption and scattering properties of soot, compared to ammonium sulfate, affect the results? I can not criticize the team for use of spherical particles, but if the actual particles were fresh soot, they probably would not be spherical. Some discussion of this possibility, and the impact on the results, would be appreciated. Also, there is no mention of the phrase Angstrom coefficient, which represents the wavelength dependence of aerosol scattering. Is this not important, due to the tight proximity of the various spectral regions. If so, this should at least be stated.

AC: In the case of our observations, we have no particular evidence for the chemical composition of particles, neither from the wavelength dependence of Mie scattering for our optical measurements, nor from the in-situ particle detection instrumentation operated on the Falcon aircraft. Also, as stated by the reviewer, due to the tight proximity of the various spectral regions assumptions regarding the single scattering albedo of the particles are not likely to be very important for the inferred trace gas profiles.

Being more specific: It is well known that the retrieval of size information from aerosol spectrometer probes, which work on the principle of detecting part of the light scattered from a single particle passing through a laser beam, depends in detail on knowledge of some properties of the particle (refractive index, shape, mixing state). We have given the Weinzierl et al. (2009) reference in this context (Page 3941, line 16). Therefore, if such information on the aerosol type is not known from the measurements, a priori guesses and assumptions e.g. on the particle refractive index and shape of the particles need to be made.

In our case, the PCASP is known to be rather insensitive to particle refractive indices (see also the Weinzierl et al., 2009). For the FSSP-300, the presence of absorbing material does affect the inversion of particle size significantly, but this is relevant only for the coarse mode particles in the super-micron size range.

Further, the aerosol measurements performed by the DLR group indicate that for the particular case study (and actually for the whole ASTAR measurement period), rather clean conditions dominated (i.e., soot did not dominate the aerosol number concentration or composition). Only thin pollution layers appeared sometimes embedded in the boundary layer or the free troposphere (Currently, a paper on this matter is only in planning phase).

In addition, we know that the Arctic is largely free of local aerosol sources and that transport analysis points to very long transport times of aerosol sources at lower latitudes. For some discussion on these aspects see for instance Stohl et al. (2006) and Quinn et al. (2009).

Stohl, A.: Characteristics of atmospheric transport into the Arctic troposphere, J. Geophys. Res., 111, D11306, doi:10.1029/2005JD006888, 2006.

Quinn, P. K., Bates, T. S., Schulz, K., and Shaw, G. E.: Decadal trends in aerosol chemical composition at Barrow, Alaska: 1976–2008, Atmos. Chem. Phys., 9, 8883-8888, doi:10.5194/acp-9-8883-2009, 2009.

Therefore, due to a good reasoning we general assumed particles of refractive index of 1.54 + 0.0i, which represent an ammonium sulfate-type aerosol. In fact, in the context of the paper, the extinction values derived from the size distribution measured in situ by the aircraft, serve as a very valid first guess, in spite of the uncertainty in the assumption of the refractive index.

RC:

g) Figure 8 suggests the retrieval of BrO has been constrained such that it can be not negative at any altitude? Is this the case? Regardless, this needs to be clarified upon revision and the figure should be re-drawn so that the full extent of the negative error bars can be seen. Also, it is unclear what the dashed vertical line represents.

AC: In fact, our trace gas profile retrieval is not constrained to positive values. Figure 8 has been modified in order to include the complete error bars, as suggested by the reviewer. The dashed vertical line in the right panel of Fig. 8 and Fig. 9 indicates the BrO detection limit of 1.5 pptv (averaged in altitude). This is now included in both figures: "The dashed vertical line indicates the BrO detection limit". In the corrected manuscript (end of Sect. 2.1), the reader is kindly referred to Prados-Roman (2010) for further instrumental details such as signal-to-noise ratio and BrO detection limit.

RC:

2. The large scale context of the observations considered in Section 4 needs to be developed. Page 3932 states "During the ASTAR 2007 campaign one sortie, performed on 8 April 2007, was specially (sic) devoted to probe the Arctic atmosphere for halogen activation (e.g., BrO detection) and the development of ODEs over sea ice regions". The references to the work of Simpson et al. (pages 3929 and 3953) as well as the discussion of ODEs in the abstract and conclusion section leads on to believe the analysis is focused on what has become to be known as satellite hotspots of BrO related to the bromine explosion.

AC: Among many other Falcon flights during the ASTAR 2007 campaign, only two (April 1 and April 8) were devoted to measure BrO, and to probe ODEs. The flight was not guided by "satellite hot spots". The flight trajectory was designed considering the location of first year sea ice (i.e., more probability of enhanced BrO), and the endurance of the Falcon aircraft.

RC: However, my examination of measurements of total column BrO on 1 April 2007 and 8 April 2007, provided by examination of OMI radiances, reveals values of VCDtotal BrO near Spitsbergen on these dates were no where close to the values commonly associated with satellite hotspots of BrO related to the bromine explosion. If the 30ppt of BrO found on the 8 April 2007 descent in the BL (blue curve, Figure 9) is associated with a satellite VCD total of either 7.0 +/- 2.0 (MPIC) or 9.0 +/- 2.3 (BIRA) x 10ˆ13 molec/cm2,

AC: As explained above, the satellite data for the 8 April 2007 are only considered for comparison with the low-cloud scenario at 14:30UT, i.e. the red part of the aircraft trajectory in Figure 9.

then how much BrO in the BL would be needed to explain the values of BrO VCDtotal that existed over Hudson Bay on 1 April or over the Alaskan sea on 8 April? This issue is ignored in the paper because the global distribution of BrO VCDtotal is never shown.

AC: The examination of the GOME-2 total BrO column maps show higher BrO values over Hudson Bay (April 1st) and Alaskan sea (April 8th) than at Spitsbergen, in agreement with the referee's statement (see attachment). However, it does not mean that Spitsbergen is not affected by boundary layer BrO (although probably to a lesser extent). Further we feel that the argument of the referee "were no where close to the values (sic) commonly associated with satellite hotspots of BrO related to the bromine explosion" is not completely true because:

(1) no values of total BrO VCDs 'commonly' associated to satellite hotspots related to bromine explosions. One should always consider possible localized or rather weak boundary layer BrO emissions.

(2) for the 1st and 8th April our measurements above Spitsbergen suggests that the total BrO VCDs measured by the satellite are mostly due to BrO within troposphere. Indeed the total BrO columns can not be explained by a stratospheric origin as the total ozone columns above Spitsbergen were typically below 375 DU (Salawitch et al., 2010). In fact, dynamical effects of a varying tropopause height are well accounted for in the stratospheric BrO correction for both satellite retrievals (as well as the effect of the photochemistry), see Theys et al., 2009 and 2011.

We also feel unsettled by the comparison made by the referee between the satellite total BrO VCDs observed at Spitsbergen and over Hudson Bay or Alaska sea, since for the three locations the stratosphere is not necessarily in the same dynamical and photochemical state. Moreover, associating BL BrO mixing ratios to columns values is particularly dangerous as information on the vertical extent of the boundary layer needs to be considered, and evidently this is a function of the meteorology, geographical location and many other parameters.

RC: Upon revision, the paper would be of much greater utility to the community of interested colleagues if polar projections of BrO VCDtotal were shown for both dates.

AC: Polar projections of BrO VCDtotal, VCDstrat, and VCDtrop as inferred from the BIRA-IASB group are now provided in Figure 10 (and referenced to in Page 3950, line 1). Together with the VCD projections inferred by the MPIC retrieval, these plots are also provided in the attachment. Note that VCD values provided by both approaches are very similar. Hence, and since the reader can easily refer to Theys et al., (2011) for any concern regarding the satellite retrieval, we decide to show only the projections retrieved by the BIRA team to avoid redundancy, because comparisons of different retrievals or inter-satellite comparisons are not within the scope of the present study.

RC: Also, the statement (page 3951) suggesting that aerosols and clouds do not obscure BrO

AC: Page 3951, lines 19-22 now read as "Overall, worth mentioning is also that in the selected passages and compared to airborne values, the satellite retrievals do not systematically underestimate BrO."

from the view of the satellite requires:

a) placing the observations in context of commensurate measurements of particles on 1 April and the entire portion of 8 April (the paper discusses aerosols and clouds for only one of the four profiles that appear in Table 1);

b) placing the observations in context of commensurate satellite measurements of cloud cover, which are routinely available for the Arctic. If the perturbation to BrO due to the bromine explosion is confined to altitudes below 1 km, as suggested by Figure 9, then is flies in the face of common sense that satellite measurements will not often be obscured by clouds, because clouds extending to altitudes above 1 km are frequently present during Arctic spring. Perhaps for the chosen profiles the sky was clear and GOME-2 was able to see to the surface. The paper, as written, does not provide enough detail to evaluate this possibility. Regardless, the authors have chosen to address the effect of clouds on the satellite retrieval of column BrO.

AC: Regarding how the clear-sky passages and the satellite pixels were selected, see above.

RC: As written, the paper states clouds do not obscure BrO. The paper must make clear whether this result is driven by the nature of the observations chosen for analysis (this comes back to the statement on page 3950 that “only the satellite pixels displaying the highest sensitivity to surface BrO have been kept for the comparison”) and the robustness of this conclusion for the totality of the satellite fields.

AC: See above.

RC:

Minor comments:

1. Page 3927, line 27: phrase “are well in agreement” is awkward and, as noted above, some quantification is very much needed.

   AC: The phrase “are well in agreement” changed to “agree reasonably well”.

2. Page 3928, line 20: Suggest starting a new paragraph at “As solutions largely depend . . .”. As written, this paragraph is very long; not a good way to start a paper.

   AC: The sentence “As solutions largely depend on the individual kind of observations, different strategies have been developed to solve these ill-posed inversion problems (e.g., Rodgers, 2000).” is shortened to “Different strategies have been developed to solve these ill-posed inversion problems (e.g., Rodgers, 2000).”

3. Page 3930, lines 1 and 2: I think the team associated with GRL paper 2010GL043798 would dispute the notion that “the horizontal extent of the BrO associated with young sea ice is fairly well captured by total satellite measurements”. Is it really? Would be nice to provide a reference or two, perhaps also point out the recent questions that have been raised and, as noted above, place the particular analyses of total and partial columns in the context of the much higher values of total column BrO observed at locations other than those sampled on 1 and 8 April 2007.

   AC: See response to Schofield’s review.

4. Page 3930, lines 7 and 12: Section is abbreviated on line 7 but not on section 12.

   AC: Following the “Textual and visual conventions” from the AMT website (Manuscript preparation), “The abbreviations "Sect." and "Fig." should be used when they appear in running text followed by a number unless they come at the beginning of a sentence , e.g.: "The results are depicted in Fig. 5. Figure 9 reveals that..."”.

5. Page 3931, line 23: Was “air-tight” intended rather than “air-tide” ?

AC: That typographical error was already reported as “Authors Comment” or “AC” during the reviewing process. Accordingly it is corrected in the new manuscript.

6. Page 3932, line 1: text describes a broad spectral region, including two spectrometers, then states “data referred to in this work are exclusively related to the measurements collected by the UV channel”. Does this mean only data from QE65000 was used, and not USB2000?

AC: Yes. As the sentence indicates, data shown and used in this work only refer to the UV channel (QE65000 spectrometer).

Regardless, this should be clarified. Would be good, in the sentence in question (top of page 3932), to quantify the UV region considered (give lower and upper limits of region).

AC: On top of that page “(320-402 nm)” is now included as the spectral range of the QE65000.

7. Pages 3932 and 3933: the 1 April 2007 flight should be described.

AC: The geo-location (Lat, Long, SZA, UT) of that flight is now included in Table 1. Additionally in the new manuscript the sky and ground conditions are also mentioned (Page 3950, line 29): “In the case of the cloud-free passages flown over the sea ice of 1 April 2007 (11:25 UT) and 8 April 2007 (14:30 UT),...”. For further description of the aircraft deployments, in Table 1 and Figure 9 of the new manuscript, the reader is kindly referred to thesis work of Prados-Roman (2010).

8. Page 3934, line 19: not sure the word “artificial” is appropriate. Perhaps “simplified”?

AC: Changed to “simplified”.

9. Page 3935, lines 3 and 4: the notion of “no trace gas absorption” should be quantified. Of course, there had to be some. Perhaps a plot showing optical depth due to O_3, O_4, and BrO vs wavelength can be considered, so that the reader could judge how clear the window at 353 nm really is. It is difficult, given what is presented in the paper, to know if the assumption of no trace gas absorption is potentially problematic.

AC: “no trace gas absorption” is changed to “no major trace gas absorption, i.e., optical density smaller than several 0.001”.

10. Page 3937, line 11. would be helpful to include a simple statement regarding whether the Jacobian was found numerically or analytically (I would guess numerically).

AC: The Jacobians are found numerically. Details concerning the radiative transfer model are given in the work of Deuschmann et al., (2011). The reference to that work is now also included in the mentioned line.

Deutschmann, T., Beirle, S., Friess, U., Grzegorski, M., Kern, C., Kritten, L., Platt, U., Prados-Roman, C., Pukite, J., Wagner, T., Werner, B., and Pfeilsticker, K.: The Monte Carlo atmospheric radiative transfer model McArtim: introduction and validation of Jacobians and 3-D features, J. Quant. Spectrosc. Ra., 112, 6, 1119-1137,,ISSN 0022-4073, doi:10.1016/j.jqsrt.2010.12.009, 2011.

11. Page 3939, line 14 and page 3940, line 13: phrase “in situ measured” is awkward.

This combination of words is unusual.

AC: Changed to “were measured by the in situ UV absorption photometer” and “aerosol number densities measured by the in situ instruments”

12. Page 3940, line 2: the 20% uncertainty for albedo, while perhaps reasonable, comes out of thin air. Some better justification for this number is appropriate. Can albedo be as high as 99% over snow and ice? Can it be as low as 59%? What is the role of mid-level clouds along some of the flight portions on this value?

AC: As stated in the last paragraph of Page 3939 “in this work the ground albedo is inferred with the assistance of an albedometer measurement platform, and of a digital camera installed on the Falcon cabin looking in the direction of the flight. The albedometer was aboard the AWI Dornier-228 Polar 2 aircraft that was also deployed during the ASTAR 2007 campaign, and performed measurements of the albedo of sea ice, snow and open water (Ehrlich 2009). Measurements from the albedometer reported a sea ice albedo of 79% in the UV-A spectral range.”

Indeed, the ground albedo values reported during the ASTAR 2007 campaign ranged between 10% (open water) and 100% (snow covered glacier) for the UV-A spectral range (e.g., Ehrlich 2009).

As mentioned in the same paragraph,
„During the 30 min of the aircraft ascent, the Falcon aircraft flew over closed sea ice, some leads covered by thin ice, and snow covered glacier“. Hence, a 20% deviation to the sea ice albedo (i.e., values between 59-99%) is considered to cover the mixed ground scenarios (recorded by the camera on board the Falcon).

In the new manuscript the reference to Ehrlich et al., (2008) cited in Page 3941 (line 13) is corrected:

Ehrlich, A., Bierwirth, E., Wendisch, M., Gayet, J.-F., Mioche, G., Lampert, A., and Heintzenberg, J.: Cloud phase identification of Arctic boundary-layer clouds from airborne spectral reflection measurements: test of three approaches, Atmos. Chem. Phys., 8, 7493-7505, doi:10.5194/acp-8-7493-2008, 2008.

Additionally, the public link to Ehrlich (2009) is now included in the bibliography:
http://ubm.opus.hbz-nrw.de/volltexte/2009/2001/pdf/diss.pdf

13. Page 3940, line 11: should read “the most challenging parameter”

AC: Changed.

14. Page 3941, line 17: when the phrase “selected spectral range” is used, would be good to again note what this range is, even if it has been given before (i.e., in response to comment 6 just above).

AC: (349–360.8 nm) is now included in the line.

15. Page 3943, line 25: better to repeat the integration time, 10 s, here. A small amount

of redundancy can be very helpful.

AC: “(~10 s)” is now included.

16. Page 3949, line 4: “planning aiming at flying” is quite awkward.

AC: "planned aiming at flying" is now changed to "planned with the goal of flying".

17. Page 3954, line 7: why does "3932" appear at the end of this citation; indeed, why

do integers appear at the end of every citation ?!?

AC: They do appear due to the AMTD editing process indicating the pages where the given reference was cited. They are indeed not included by the authors.

18. Page 3954, line 20: "o" missing in Hartmut's last name.

AC: Corrected.

19. Table 1: the meaning of the asterisk should be explained in the Table, whether or not it is repeated in the text: i.e., please include explanation as a brief footnote.

AC: The header of the table now reads as "Note that VCDstrat retrieved by the MPIC team may contain free tropospheric BrO (indicated as *)…". Accordingly, the * has been erased from the VCD_{BL} values provided by the MPIC team in Table 1.

20. Figure 1: would be useful to show SZA somewhere or else state the range of SZA in the caption. Also, rationale for identifying and ODE should be stated either in the caption or in the text.

AC: "(72-80° SZA)" is now included in the caption of Fig. 1. In addition, "Here, based on the ozone measurements performed by in situ instrumentation, the threshold of an ODE situation is 45 ppbv" is now included in Page 3932 (line 6).

21. Figures 4 and 5: can similar figures be shown for 1 April? If so, they would be quite helpful. If not, please explain why this is the case.

AC: See above.

22. Figure 7: neither the text or caption explains how Xtrue is known. I assume that the true value of this quantity is found from pressure. Regardless, this should be spelled out either in the text or caption.

AC: It is actually included in the text (Page 3944, line 10): "the true O_4 state (***x***) is given by Eq. (1)".

For consistency with Eq. (1), in the corrected manuscript "extinction coefficient (ϵ_{O4})" is now included in Page 3944, line 3. Accordingly, "O_4" is changed to "ϵ_{O4}" in Page 3944 (lines 5, 10, 14, 16, 17) and Page 3952 (line 16). Also, the caption of Fig. 7 now reads as "Retrieval of the vertical distribution of the O_4 extinction coefficient (ϵ_{O4})….", and the symbol ϵ_{O4} is in the same way included in the caption.

Figure 9: It would be nice to be able to see the O3 values in the BL for the four profiles. Please consider either an insert showing detail for this region or another panel. The essence of the figure is lost for many interested readers in the present form, because the actual values of O3 in the BL are obscured.

AC: As mentioned in Page 3939, lines 21-25: "a more detailed discussion of observations with respect to the sources of reactive bromine, its atmospheric transport and photochemical transformation is not within the scope of the present study"

We believe that the boundary layer ozone depletion and bromine activation is fairly captured by Fig. 1 and Fig. 9. We understand that an additional figure detailing the chemistry in the boundary will deviate the aim of the paper (i.e., describing a retrieval algorithm). Indeed, and as also mentioned in the manuscript “Such an approach is the objective of a forthcoming study”.

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RC: Again, the new retrievals seem to be EXCELLENT. The description of these retrievals in Sections 2 and 3 is overall outstanding. But the paper, as submitted, does not, in my opinion, provide a comprehensive enough discussion of the attendant science as it should. I hope this is addressed upon revision, regardless of the editorial decision on this paper.

AC: We want to take the chance to thank the reviewer for his work. Overall, we believe that the reviewer’s criticism is partly based on a misunderstanding of the intention of the study (i.e., to present a new algorithm for inferring trace gas vertical profiles from aircraft limb observations).