

We thank both reviewers for extremely helpful, positive comments that have led to improvements in the manuscript. Our responses are given below in boldface. We have also boldfaced some words of the referee's comments, to highlight certain points. As part of the reply to reviewer comments, we have made significant changes to the abstract of the paper, which now includes more details of what was found in the study. We have added two new figures to the Supplemental Materials section, to address detailed points raised by the reviews. Finally, we have added numbers to the comments from Referee #2, to facilitate any possible future iteration.

Anonymous Referee #1

Received and published: 21 September 2011

a) This paper describes an analysis and comparison of aircraft vertical profiles of BrO (and other bromine species) with tropospheric retrievals of column BrO from two satellite instruments, GOME2 and OMI, for a number of cases in 2008. The objective is to address the question as to whether or not we understand the physical and mechanistic origin of relatively intense BrO column events, and if they are consistent with the surface level phenomenon of ozone depletion in the Arctic springtime, or if they may be more a reflection of changing tropopause heights and/or stratosphere-troposphere exchange events, involving bromine. The methods involved a model of the stratospheric component of the observed satellite column measurement, and which assumed 7 ppt of "very-short-lived" organobromine compounds in the stratosphere. The approach used for the stratospheric simulation seems to be the current state of the art; while the 7 ppt is near the high end of the likely range, it was reasonably well justified in the paper (and in Salawitch et al., 2010). What makes this paper **unique** and **important** is the fact that there is a good combination of aircraft-based column measurements (for the troposphere) plus a number of cases where one can argue that relatively reliable (reflective surface and few obscuring clouds) satellite-based tropospheric column BrO retrievals were obtained. This paper has some very important conclusions/findings, and I believe it is a paper that will have some significant impact. It is well-conceived and well written, and therefore I believe that it should be published.

> Thank you very much for your positive comments and noting the uniqueness and importance of our manuscript.

b) The big impact conclusion of the paper, in my judgment is stated in one sentence, the penultimate sentence of Section 4: "Our results are consistent with a surface origin of elevated BrO as well as transport of BrO enriched air parcels away from the source of origin". This should be the focus, and should be highlighted and strengthened in the paper.

> We have added two new sentences to the abstract to draw attention to these points:

The aircraft profiles reveal that tropospheric BrO, when present during April 2008, was distributed over a broad range of altitudes rather than being confined to the planetary boundary layer (PBL).

The rapid activation of BrO due to surface processes (the bromine explosion) is apparent in both the OMI and GOME-2 based tropospheric columns.

c) However, the paper is 35 pages of text, with 6 tables, and 18 figures, each of which has multiple panels, in some cases many panels. Reading this paper is a serious piece of homework, and the paper makes the reader really work and dig, going back and forth between text and figures. To me, it is worth it, but there are a very few such specialists keenly motivated to learn about the latest development in this area. For most readers then, the paper does itself a disservice; the impact of the paper could be much greater if it were rewritten with a serious attempt to focus on the supporting evidence for the main conclusions of the paper.

> We have chosen to maintain the general outline and structure of the paper, but shorten in places (see below), in part because referee #2 wrote “I believe it is high quality work... rather long but well structured and easy to read.”

d) Besides the conclusion stated above, the other important points were that satellite retrievals (making use of a good stratospheric model) for tropospheric BrO columns can be reliable if a few conditions are met, specifically that the surface albedo is large, the solar zenith angle is not too large, and that there are not optically thick clouds. The observation of some BrO events above the BL top is interesting, and perhaps they may result from convection from leads?

> We agree that BrO events above the top of the BL may indeed be associated with convection from leads. We had mentioned this on line 6, page 32 of the submitted paper. In the revised paper, we retain this sentence but now also mention the affect on BrO of convection over leads in the conclusion section.

e) In any case, I believe the paper should be shortened with an eye toward asking whether inclusion of material addresses the main conclusions of the paper. I have some suggestions, and there are some minor concerns and errors in the paper which should be addressed.

> We have shortened the paper in a few places (see below) and have responded to all of the minor comments, in most cases by making changes to the text and/or figures.

These are listed below in the order they arose in the paper.

1. Lines 6-16 on page 9 are not needed, an outline of a paper just gets in the way of reading the paper!

> We have deleted this paragraph.

2. Top of page 11 - the surprisingly low BrO values shown in Figure 2 should perhaps warrant more comment; while the aircraft vertical profiles show medians at the surface of perhaps ~1-4 ppt, as an example, the median surface measurements reported recently for Barrow in April are more like 8-9 ppt, and much higher in partially depleted conditions. Was 2008, or the conditions during the flight, anomalous in some way?

> We have added the following new text, to top of page 11, to address this comment:

The low values of BrO observed during ARCTAS and ARCPAC could be due to preferential sampling of O₃ depleted air by the two aircraft (which would titrate inorganic bromine from BrO to Br) or the fact the aircraft generally sampled to an altitude of 100 m above the surface (Neuman et al., 2010). While it is tempting to argue the highest mixing ratios of BrO are confined to the lowest region of the marine boundary layer (a region inaccessible to the two aircraft), ground-based MAX-DOAS observations from Barrow, Alaska obtained during April 2008 suggest that when BrO was elevated, the perturbation extended to ~1 km altitude (Salawitch et al., 2010).

The ARCTAS and ARCPAC in situ profiles of BrO show, consistent with MAX-DOAS data from April 2008, that the highest mixing ratios of BrO tended to be observed above the boundary layer.

3. Page 21, line 9 - or that the modeled stratospheric column BrO is too large?

> We have re-examined our description of BrO over the Barents Sea based on this comment, as well as a comment from Reviewer #2 (see below). We have entirely re-written a key paragraph, which now reads:

OMI total column BrO in Figure 5, within the region of the Barents Sea (black box), shows significant gradient. The region of high total BrO towards the upper left hand corner of the black box is associated with enhanced stratospheric burden (Fig. 5b, orange). Another region of large total BrO (lower portion of black box) is associated, by our analysis, with an enhanced tropospheric burden (Fig 5c, red). This region of enhanced tropospheric BrO occurs over a bright portion of the Barents Sea (Fig 5d, crimson, indicating snow or ice). Nearly zero tropospheric BrO column amounts are obtained over low surface albedo areas (OMI reflectivity < 0.5) of the Barents Sea (Fig. 5d, blue). Here, the retrieved total columns are generally less than retrieved columns over adjacent areas with higher reflectivity, leading to low tropospheric column BrO over parts of the Barent Sea. However, satellite-derived tropospheric BrO may not be reliable when the surface albedo is low. While it is possible tropospheric column BrO was truly low on 22 April over this region of the Barents Sea due to the lack of snow or ice leads that may be needed for bromine activation, it is also possible that our inference of low tropospheric BrO could either be a result of limited sensitivity over dark surfaces or an over-estimation of the stratospheric burden. The complicated sensitivity of satellite-derived tropospheric BrO to surface reflectivity and stratospheric burden requires concerted future study.

4. Page 24 - line 11 - yes, indeed, this variability is important to stress, and the impact of using things like zonally fixed column BrO can be problematic.

> We agree an accurate estimate of the stratospheric contribution is essential to derive accurate tropospheric BrO column amounts, as stratospheric variability is significant. In the submitted paper, we had stressed this point in the last two sentences of the second paragraph of the conclusions. In the revised paper, we retain these sentences and we have added a new sentence to the second paragraph of Sect. 4.1.1 (page 24, line 16 in the submitted paper), that reads:

Essentially all derivations of tropospheric column BrO prior to Salawitch et al. (2010) and Theys et al. (2011) relied on the use zonally fixed stratospheric BrO, which, as first pointed out by Theys et al. (2009), will lead to large errors.

We have also added the following new sentences to the conclusions section to emphasize this point:

Many prior studies have used a zonally symmetric representation of stratospheric BrO to derive tropospheric BrO from satellite observations.

As put forth by Theys et al. (2009), Salawitch et al. (2010), and Theys et al. (2011) and re-inforced here, this is a deeply flawed assumption.

5. Page 25, bottom paragraph - why discuss profile #13 if you are not showing the figure?

> Good point. In the revised paper, we now show in Figure 2 the profile of BrO that was obtained at locations #13 and #14. We refer to Figures 2 and 6 for this paragraph.

6. Page 26, line 9 - Hudson Bay is barely visible in the figure. Perhaps this discussion can be dropped?

> We thank the referee for noting our improper focus on the BrO anomaly just over Hudson Bay. In the revised paper, for this Section 4.1.2, we have replaced the first use of "Hudson Bay" with "vast regions of eastern Canada including Hudson Bay" and the two other uses of "Hudson bay" with "Eastern Canada". We have rewrite two sentences including "Hudson Bay" to avoid this.

7. Page 27, line 13 - well, GOME-2 trop is in fact shown.

> Thank you. We have deleted "(not shown)".

8. Top page 28 and Figure 9b - this information is useful and interesting to see! I worry that the top panel of Figure 9b will be even harder to read in the published version. The same applies to Figure 6b, top. The top panels are so important, but are hard to read. Perhaps the other panels in these figures could be in supplemental?

> Yes. Indeed, these time series plots (Fig. 6b and 9b) are important plots and we agree with your concern that these important figures may not be very readable in a final published version. To make the top panel more readable, we have enlarged the vertical span of top panel of each time series plot. We will request these figures appear as full column. Also, the legend that had appeared on the top right has been placed below the top panel, so that, if ACP does print these figures full column, the data portion of the plot will span the entire journal page.

We feel that it is important to show the altitude of the aircraft, as well as mixing ratios of ozone, BrO and active bromine from the aircraft measurements to provide the readers with the context of the measurements. So, we have decided to retain this information in the main body of the paper. We feel that the change to the aspect ratio of the top panel and the legend placement adequately addresses this concern.

9. Page 28, line 18 - the term “tropospheric perturbation” has no meaning, equivalent to saying “we don’t know”.

> **We have rewritten this sentence to read:**

Our analysis indicates that the large enhancement in total column BrO seen by OMI and GOME-2 over the Chukchi Sea on April 19 was tropospheric in origin, rather than stratospheric.

10. Page 29, for example - throughout the paper there is reference to aircraft column #s. Without reference to a specific figure in the **context** of these numbers, it is often cumbersome and difficult to follow.

> **We understand based on the length of the paper, the large number of figures, and the fact we did not refer to Figure 1 in Section 4, that it may have been easy to overlook Figure 1. However, in the submitted paper, Figure 1 (unchanged in the revised paper) is designed to provide context to the aircraft column numbers.**

In the revised paper, we now refer to Fig. 1, where appropriate, throughout Sect. 4.

11. Page 29, line 6 “not significant”, but please note that they are as much as 3 times smaller!

> **We thank the referee for pointing this out. We have re-written the text to read:**

Time series plots indicate that WP-3D in-situ columns #25, #26, and #27 are lower than the OMI tropospheric column, but the differences are not significant given the uncertainties in OMI and in-situ tropospheric columns (i.e., error bars overlap). However, the absolute magnitude of the OMI tropospheric column is about a factor of 3 larger than the in-situ BrO column.

12. Page 29, Line 18 - here and in other parts of the paper there is reference to transport (vertically in this case, on others horizontally (page 32, line 22), even using the term “long-range”, page 35, line 6) of BrO. BrO is too short-lived (perhaps ~30s, from just photolysis alone), and does not undergo significant transport itself. I feel sure that the lifetime of BrO is very short compared to the timescale for transport across the tropopause! What is transported is an airmass/parcel that retains the ability to continue generating/recycling bromine radicals. This is indeed an important distinction. All references to transport of BrO itself must be removed, and the discussion technically improved/corrected.

> This is correct! We have replaced BrO with inorganic bromine (Br_y), here and in other places, when we talk about transport. BrO of course goes away at night. The inorganic bromine family, however, has a long lifetime and is subject to redistribution by transport.

One factor not considered in our analysis is irreversible, cross-tropopause exchange of air parcels with elevated levels of Br_y from the stratosphere to the troposphere (STE).

13. Page 31, line 28 - on the other hand, the chain chemistry is quadratic in BrO, so, a deeper BL could slow down the chemistry as BrO (its precursors) is diluted. (“On the other hand, O₃ may not become completely depleted in a deeper boundary layer, allowing concentrations of BrO to remain high.”)

> Thank you for the suggestion. We have adopted your sentence in the revised paper, which now reads:

On the other hand, the ozone loss rate is quadratic in BrO, so a deeper boundary layer could slow down the chemical removal of O₃ as BrO is diluted.

14. The bottom map in Figure 6a is not necessary.

> We feel the bottom map is necessary because it indicates the location of places referred to in the text. Some of readers may not know the location of the Chukchi Sea, for instance. This map was motivated by Figure 10 of Theys et al. (2011).

15. As an example, Figure 10b is a very important and interesting case where the agreement is not good.

> We have modified the discussion regarding Fig. 10b, as noted in our reply to comment #11 above. We will also request that Fig. 10b be a full column figure, which will allow the reader to clearly see this case where OMI and in-situ tropospheric BrO are not in good agreement.

16. The two little panels on the right side of Figure 13 need different colors for T and BrO. Why are there only two rows of these latter figures?

> The original colors were too close to be easily distinguished. In the revised paper, easily distinguished colors are now used for the side panels of Figure 13.

The first row does not have a side panel showing profiles of BrO, active Br and temperature because there are no aircraft measurements corresponding to the time and the location of the depicted bromine event. We have revised the figure caption to clarify this point.

Anonymous Referee #2

Received and published: 26 September 2011

1) Choi et al. reports on satellite observations of tropospheric BrO using OMI and GOME- 2 and on comparisons with in-situ observations of bromine species during aircraft campaigns. I believe it is **high quality work**, with a lot of nice interpretations of the results and I found the manuscript **rather long** but **well structured** and **easy to read**. I recommend publications in ACP, provided that the following points are addressed.

> Thanks for these very positive comments.

2) -The author uses as stratospheric correction the model-based data introduced by Salawitch et al. (2010) that assumes a very strong supply of Bry from VLSL. There has been considerable debate on these VLSL settings (assuming 9 pptv of Bry in total: 7pptv from PGI +2pptv from SGI) and I'm still not convinced that the resulting stratospheric correction is appropriate. Indeed the stratospheric vertical columns were not compared to any independent stratospheric BrO observations and it has not been demonstrated that this model-based stratospheric correction could actually correct for the BrO absorption in the stratosphere. Moreover, the author of the present manuscript reported negative tropospheric BrO column in the maps they show which I believe is a sign of an overestimation of the stratospheric correction. However, in the authors' defense, I believe that this aspect is probably not crucial for the comparisons between satellite and in-situ data displayed here.

> The reviewer is correct that, in the Choi et al. paper, we do not compare to independent stratospheric observations of BrO. However, in Salawitch et al. GRL 2005, a similar model study is compared to independent aircraft (Figure 1 of that paper) and balloon-borne (Figure 2 of that paper) observations of BrO. We have added two new sentences to the revised paper in Section 1 stating:

Although we do not compare to independent observations of BrO here, such comparisons to aircraft and balloon-borne observations of BrO in Salawitch et al.(2005) support the use of these values for the delivery of stratospheric bromine by PGI and SGI. Other observations of BrO, such as some of those discussed in Section 2.5.2 of WMO (2007) support smaller values for these parameters.

The reviewer is correct that we do, on occasion, have negative values of tropospheric BrO column. We have added two new sentences to Section 4.1.2 of the revised paper to address this detail:

There are segments where inferred OMI tropospheric BrO is negative, such as the early time period of Fig. 8b. These values of negative BrO could be indicative of an over-correction for the stratospheric column, measurement noise for total column BrO , or some other error in calculation. Overall, we do not find extended regions of negative tropospheric BrO (either spatially or temporally).

3) Indeed, they discussed the results **honestly and rigorously**, using a **quite detailed error analysis**. The **strength of this paper** is clearly the **link between the aircraft and the satellite data** as it brings additional information on the processes involved (e.g., bromine activation after blowing-snow events).

> **Thank you for these very positive comments!**

4) Also carefully discussed by the author is the limitation in the use of the aircraft data as in numerous cases the aircraft does not fly low enough to pick up the important contribution of the tropospheric BrO column located close to the surface. Indeed, Friess et al., 2011 showed - using MAXDOAS observations at Barrow - that most of the PBL BrO was actually located very close to the ground, few hundred meters.

> **We now cite the Friess et al. paper. We have added, to Section 2.1, a description of Friess et al. 2011 that reads:**

In contrast, MAX-DOAS near Barrow obtained during April 2009 by another group suggest the enhancement to BrO usually occurs in the lowest 300 m, with one case where elevated BrO extended to 500 m altitude (Friess et al., 2011).

The two aircraft are not able to sample BrO between the lowest altitude on descent and the ground. However, the contribution of this partial column to our estimate of in situ tropospheric BrO column would be small, because the column of air below the aircraft is a tiny fraction of the overall tropospheric column, unless mixing ratios of BrO in this sliver of air reach 40 ppt. We have added a new figure, Figure S2, which shows a probability distribution function of surface BrO, to the paper. This figure shows that $\frac{2}{3}$ of the time, surface BrO as measured by a CIMS instrument at Barrow, Alaska in Spring 2009 was below 8 pptv. We present a calculation in the caption of Figure S2 documenting that 8 pptv of BrO distributed between the surface and 75 m altitude would contribute 0.18×10^{13} to the column.

We have also added a new paragraph, to the main body of the paper (second to last paragraph of Section 2.1) that addresses this issue.

The lowest altitudes sampled by the DC-8 and WP-3D during their descents over BrO-enhanced regions were 75 and 77 m, respectively. In our analysis we use the composite DC-8 or WP-3D profile to extrapolate between the lowest sampled altitude and the surface, for each formulation of in-situ column BrO shown in Tables 3, 4, and 4. At times, surface BrO can reach mixing ratios as high as 40 pptv (Liao et al., 2011a). We have assessed the impact of elevated surface BrO on our analysis of aircraft, satellite, and modeled stratospheric columns by conducting a probability distribution function for daytime surface BrO, observed at Barrow, Alaska (71°N, 156°W). Two thirds of the time, surface BrO is below 8 pptv (Figure S2). A uniform distribution of 8 pptv of BrO between the surface and 75 m altitude would contribute $0.18 \times 10^{13} \text{ cm}^{-2}$ to the column, an amount much smaller than the in situ and satellite-based columns discussed throughout the paper (see caption, Figure S2). Levels of BrO reaching 40 pptv below the aircraft would contribute $1 \times 10^{13} \text{ cm}^{-2}$ to the column if the BrO were present, at this amount, uniformly between the surface and 75 m altitude. The PDF analysis shows that while surface BrO did reach 40 ppt in spring 2009, such occurrences were rare. While layers of highly elevated BrO below the aircraft could on occasion compromise our comparisons, our overall conclusions are robust because surface measurements indicate only on rare occasion are BrO enhancements large enough to significantly perturb the column.

5) In its introduction, the author write that Salawitch et al could reconcile the total columns from OMI with the aircraft tropospheric columns and the model-based stratospheric columns using 9 pptv of Bry. How does this finding changes knowing the limitation of the aircraft data (read above) and the fact that the tropospheric AMFs were not treated?

> The reviewer raises two points here.

I. The analysis of Salawitch et al. (2010) indeed did not treat tropospheric AMFs in a rigorous manner. So, their so-called reconciliation of the budget needs to be treated with caution. We have added the phrase “and thus must be treated with

caution” to the place, in the Introduction, where the budget reconciliation of Salawitch et al. is discussed.

II. The reviewer wrote “knowing the limitation of the aircraft data (read above)”. We think the reviewer is referring to the fact the aircraft profiles sample only down to altitudes of ~75 to 387 m (Table 1). Again, as noted above, there was a sharp gradient in BrO below the lowest altitude commonly sampled by the aircraft and the surface and if the BrO mixing ratio in this region reached 40 pptv, then the assumption we have used would be flawed. But the occurrence of BrO levels this high, at the surface, is exceedingly rare (see our new Figure S2). Therefore, we do not believe the altitude region sampled by the aircraft poses a limitation to our findings. We do appreciate that this issue has been raised in the review because we think Figure S2 is an important addition to the paper.

6) -In section 3.2: I’m not convinced that the algorithm gives useful results for SZA as high as 85°. At these SZA, the photochemistry of BrO becomes important and this can lead to photochemical gradients along the slant stratospheric photon path. More importantly, the tropospheric column accuracy is controlled by the AMFstrato/ AMFtropo ratio. Any error on the stratospheric correction will be amplified by this factor. In this respect, the SZA threshold of 85° given is too optimistic. E.g., it is difficult to believe that the enhanced tropospheric BrO column values North of Greenland (Fig5) and on other figures (8,9,10) are real (no enhanced values in the corresponding total columns). If one calculates a realistic error on the tropospheric columns, one would certainly find huge values there.

> We have taken a close look at our SZA threshold and, based on this re-examination as well as this comment, we have revised the SZA threshold to 80 degrees for all of the figures, except Figs 5. Fig 5 shows the effect of SZA, so we feel it is reasonable to extend to 85 (this constitutes a tiny area of the data). We also believe we can accurately estimate the stratospheric correction out to SZA of 80 deg because the model that has been used has been compared, extensively, to gases that vary with SZA (see, for example, Salawitch et al., GRL, 2002).

7) Personally I find deceiving not to see a plot displaying WFs for thin cloud and high albedo conditions and showing that even for cloudy conditions we have good sensitivity to PBL BrO (provided of course that the cloud optical depth is not too high). It would be convincing and more useful than the results of Figs 3 & 4 that are very well known by people retrieving tropospheric constituents in the UV.

> In the submitted paper, we had provided a long paragraph describing a paper (Vasilkov et al., 2010) in which simulations of thin clouds over high surface albedo were performed. Vasilkov et al. (2010) showed that the sensitivity of UV satellite measurements to trace gas absorption near the surface in clear skies

with moderately high surface albedo (70%) is approximately the same as for substantially cloudy conditions (optical thicknesses up to about 30) over a higher albedo surface (90%); the cloud shielding effect is much reduced for high albedo surfaces. In addition, enhanced absorption takes place above a cloud of moderate to high optical thickness. We did not feel it was necessary to repeat these calculations.

For the revised paper, to place less emphasis on the optically thick cloudy case (which was already covered by Theys et al., 2011) and more emphasis on the thin cloud study, we replaced the paragraph describing the thick cloud case with a single sentence that reads:

However, as shown by Theys et al. (2011) and in Fig 3b, optically thick clouds can shield satellite measurements from absorbers including BrO.

We then added more detail in the description of Vasilkov et al. (2010) with the following two sentences:

Vasilkov et al. (2010) showed that the sensitivity of UV satellite measurements to trace gas absorption near the surface in clear skies with moderately high surface albedo (70%) is approximately the same as for substantially cloudy conditions (optical thicknesses up to about 30) over a higher albedo surface (90%); the cloud shielding effect is much reduced for high albedo surfaces. In addition, enhanced absorption takes place above a cloud of moderate to high optical thickness.

We like Figs. 3 and 4 because they expand on the simulations of Theys et al. (2011), showing results at higher SZAs over high albedo surfaces. Figs 3 and 4 are important for our study, so they have been retained.

Minor comments:

Introduction:

8) -I found the list of past studies on BrO remote-sensing nice. The author might want to complete it by referring to recent works of Prados-Roman et al., AMT, 2011 and Friess et., JGR, 2011 (in press).

> Thank you for bringing to our attention these studies. We cite both in the revised paper. We have described, in reply to comment #4, how Friess et al. was cited. We now cite Prados-Roman et al. (2011) in Section 1 with the sentence:

A retrieval of the BrO profile using radiances measured in the Arctic during April 2007 by an airborne limb scanning mini-DOAS instrument also suggest elevated BrO is confined to the boundary layer (Prados-Roman et al., 2011).

We also now cite both studies in the fourth paragraph of Section 1, where we present the list of past studies.

9) -P8, l 6-7: the author should be more precise as for the “best case” of Salawitch et al. and for (same page, l8) “quite a bit of geographical variability”. It is rather vague. (“The stratospheric BrO burden found by Salawitch et al. (2010) for their “best case” simulation of 7 ppt from PGI and 2 ppt from SGI is considerably larger (□27% overall difference, with quite a bit of geographic variability) than the stratospheric BrO burden used by Theys et al. (2011).”)

> In the revised paper, we now refer to Figure S7 of Salawitch et al. (2010), which they used to define their “best case”. Also, we have included in the Supplemental Material section of the revised paper a new figure, Figure S1, that shows the geographic variability of the ratio of stratospheric BrO found by Salawitch et al. (2010) to that found by Theys et al. (2011). We have embedded relevant discussion of this issue in the caption of Figure S1 and refer to this figure in the main body of the paper.

Data description:

10) -Binning the profiles on 500 m thick layers and then taking the median mixing ratio as representative at the altitude can potentially leads to a problem: the median as a tendency to be less affected by outliers than the mean calculation. This could be an issue for very high near-surface BrO mixing ratio values (bromine explosions, the ‘outlier’ in the profile), leading to a likely underestimation of the tropospheric BrO in-situ columns for these conditions (precisely what you want to avoid). What is the original vertical sampling (m) of these BrO profiles?

> The in situ airborne observations of BrO obtained during ARCTAS and ARCPAC show little evidence for highly elevated mixing ratios of BrO, in the boundary layer (BL), due to the bromine explosion. This is discussed at length by Neuman et al. (ACP, 2010): they postulate this surprising aspect of the data is due to the tendency of the aircraft to sample ozone depleted air while flying in the BL.

The sampling of the DC-8 and WP-3D observations of BrO used here and our relation of these observations to satellite data was designed, after careful consideration of alternatives, to represent the essential nature of the “signature” of ARCTAS and ARCPAC BrO. This “signature”, depicted in our figures as well as Neuman et al. (2010) and Salawitch et al. (2010), is the tendency for BrO enhancements to occur in relatively thick altitude segments well above the top of the BL.

We have examined various other methods to derive in situ BrO columns, such as using mean BrO mixing ratio within the layer. Use of the mean, rather than the median, has no impact on our results. Also, use of the maximum BrO mixing ratio observed in the lowest vertical bin does not substantially change the magnitudes of the aircraft-based tropospheric columns. The aircraft very often sampled within 200m above the surface and we believe the amount of BrO could possibly present in the air masses below aircraft, when converted to column abundances is too small to appreciably affect the scientific conclusion. Tables 1, 2a and 2b provide detailed information on the vertical sampling of BrO. Also, Figure 4 of Salawitch et al. shows the original vertical sampling of a BrO plume on 16 April. There is strong variability in BrO very close to the surface but clearly, as shown by this figure, the tropospheric column is dominated by contributions from above the top of the convective BL.

So, in summary, we feel Neuman et al. (2010) and Salawitch et al. (2010) have described, in considerable detail, the original vertical sampling of BrO and here, we use a strategy that was designed to properly represent the signature of these aircraft measurements.

We have added the following two new sentences to Section 2.1 of the revised paper:

This sampling strategy was chosen to properly represent the signature of the ARCTAS and ARCPAC BrO profiles, which show that the tropospheric column is dominated by contributions from above the top of the convective boundary layer (i.e., the region of constant potential temperature; see Neuman et al. (2010) and Salawitch et al. (2010)) and, when BrO enhancements occur, they are present in relatively thick altitude segments (i.e., extent larger than 500 m). Our results are insensitive to use of mean BrO within the layers rather than median BrO.

11) -The time (UTC) in the Tables often exceed 24:00. It is weird.

> The aircraft community denotes flight dates based on the calendar dates of aircraft take-off, at the location of the take-off. We have chosen here to use this protocol. Often the calendar date changes in Greenwich, England when the aircraft is still in the air; when this happens our UTC will exceed 24:00. We have added the following sentence in the third paragraph of Section 4.1.1 to make this clear:

We indicate time in hours relative to the starting date of the flight; therefore times greater 24:00 refer to the following day.

12) -P12: The author refers to Tables 3, 4a and 4b, but the satellite data are not introduced yet. It would be better to use section 2.2 for Tables 3 and 4.

> We have added the phrase:

(see Section 2.2 and 3.2 for a description of the satellite data and related parameters in these tables).

to the end of the sentence that introduces Tables 3, 4a, and 4b. We would like the introduction of these tables to stay in Section 2.1, because the tables are indexed with respect to aircraft profiles.

13) -Fig 7 & 8: the 5th and 8th April DC-8 flights were not mentioned in the section 2.1. They should be added.

> We have added to Section 2.1 the sentence:

We also examine measurements of other quantities for DC-8 flights of 5 and 8 April 2008, for which BrO was below the instrument detection limit.

14) -P13: The author probably forgot to mention that O₃ is also an absorber considered in the OMI BrO retrieval.

> Thank you for pointing out this omission! We have added O₃ to the sentence in question.

15) Section 3.1: I don't understand why the AMF_{strato} are not calculated using equation 8 applied to the simulated stratospheric BrO profiles. This would have avoided an unnecessary source of error on the tropospheric column (not discussed by the way). Error on stratospheric AMFs due to this could be as large as 10%

> The stratospheric AMF used in this study is a part of the current official OMI BrO product, which is based on profiles of BrO from a model climatology provided by Chris McLinden (using a model provided to him by Michael Prather). In general, the stratospheric AMF is not sensitive to details of the BrO profile, unless there is a significant contribution to total absorption from molecules close to the surface (well below the tropopause). We believe, based on sensitivity studies we have conducted over the past several years, that our overall scientific conclusions would not change if we were to use our simulated stratospheric BrO profiles to guide the stratospheric AMF. Use of our simulated strat BrO profiles

would require an enormous effort. We can easily tolerate errors of ~10% in stratospheric AMF, because this is a lot smaller than the uncertainty due to VSL bromocarbons on the simulated stratospheric BrO burden.

16) Section 3.2: Figure 5: the results over the Barents Sea: I feel unsettled by the results because it is dangerous to say anything about the albedo effect here. Indeed the fact that you see high/low BrO values over high/low albedo scenes can always be perturbed by the fact that this could be real (emissions over sea-ice regions).

> We understand your concern. However, we feel that the discussion of the albedo effect is important because whether or not BrO was present, it will be difficult to detect from space owing to low sensitivity over dark surfaces.

We believe Fig. 5, which shows OMI tropospheric BrO and many other geophysical parameters, is central to our paper because one region of elevated total column BrO is associated with high stratospheric burden whereas another region of elevated total BrO is not associated with enhanced strat BrO. Both of these examples occur over highly reflective surfaces.

We have re-examined our description of BrO over the Barents Sea based on this comment, as well as a comment from Reviewer #1. We have entirely re-written a key paragraph, which now reads:

OMI total column BrO in Figure 5, within the region of the Barents Sea (black box), shows significant gradient. The region of high total BrO towards the upper left hand corner of the black box is associated with enhanced stratospheric burden (Fig. 5b, orange). Another region of large total BrO (lower portion of black box) is associated, by our analysis, with an enhanced tropospheric burden (Fig 5c, red). This region of enhanced tropospheric BrO occurs over a bright portion of the Barents Sea (Fig 5d, crimson, indicating snow or ice). Nearly zero tropospheric BrO column amounts are obtained over low surface albedo areas (OMI reflectivity < 0.5) of the Barents Sea (Fig. 5d, blue). Here, the retrieved total columns are generally less than retrieved columns over adjacent areas with higher reflectivity, leading to low tropospheric column BrO over parts of the Barent Sea. However, satellite-derived tropospheric BrO may not be reliable when the surface albedo is low. While it is possible tropospheric column BrO was truly low on 22 April over this region of the Barents Sea due to the lack of snow or ice leads that may be needed for bromine activation, it is also possible that our inference of low tropospheric

BrO could either be a result of limited sensitivity over dark surfaces or an over-estimation of the stratospheric burden. The complicated sensitivity of satellite-derived tropospheric BrO to surface reflectivity and stratospheric burden requires concerted future study.

We feel this re-write addresses the concerns of both reviewers.

Section 3.3:

17) -Why does the author not consider any systematic error on the slant columns?

> We have assumed all of the error in SCD_{Total} is random (precision) and, as noted by the reviewer, the measurement is un-biased (no systemic error). The primary reason for this assumption is we lack a basis for estimating the systematic component of the error in the slant column.

During April 2011, a ground-based campaign led by George Mount was conducted at Fairbanks, Alaska, devoted to validation of OMI BrO SCD_{Total} and VCD_{Total} . This campaign has revealed that there may indeed be a systematic error in SCD_{Total} due to choice of fitting window. The submitted paper did include a statement in reference to this campaign, the sentence in Section 2.2 that reads:

There are potentially large uncertainties associated with the derivation of both slant and vertical absolute column BrO related to the choice of spectral fitting windows and polynomial parameters used in retrievals as well as assumed BrO vertical profiles (G. Mount, private communication, 2011; DLR, 2009).

The Fairbanks April 2011 results are still undergoing analysis and can not yet be used, or cited, in our paper.

In our paper, we have used a large estimate of the random error (18%) to compensate, in part, for lack of consideration of a systematic error in SCD_{Total} .

We revised text in Sect. 2.2 to address systematic errors in slant columns:

All of the analyses conducted in this study are based on the assumption that the magnitude of satellite-derived slant column is correct. A detailed analysis of the errors involved in deriving satellite-based slant columns of BrO is beyond the scope of this paper. There are potentially large uncertainties associated with the derivation of both slant and vertical absolute column BrO related to the choice of spectral fitting windows and

polynomial parameters used in retrievals as well as assumed BrO vertical profiles (G. Mount, private communication, 2011; DLR, 2009).

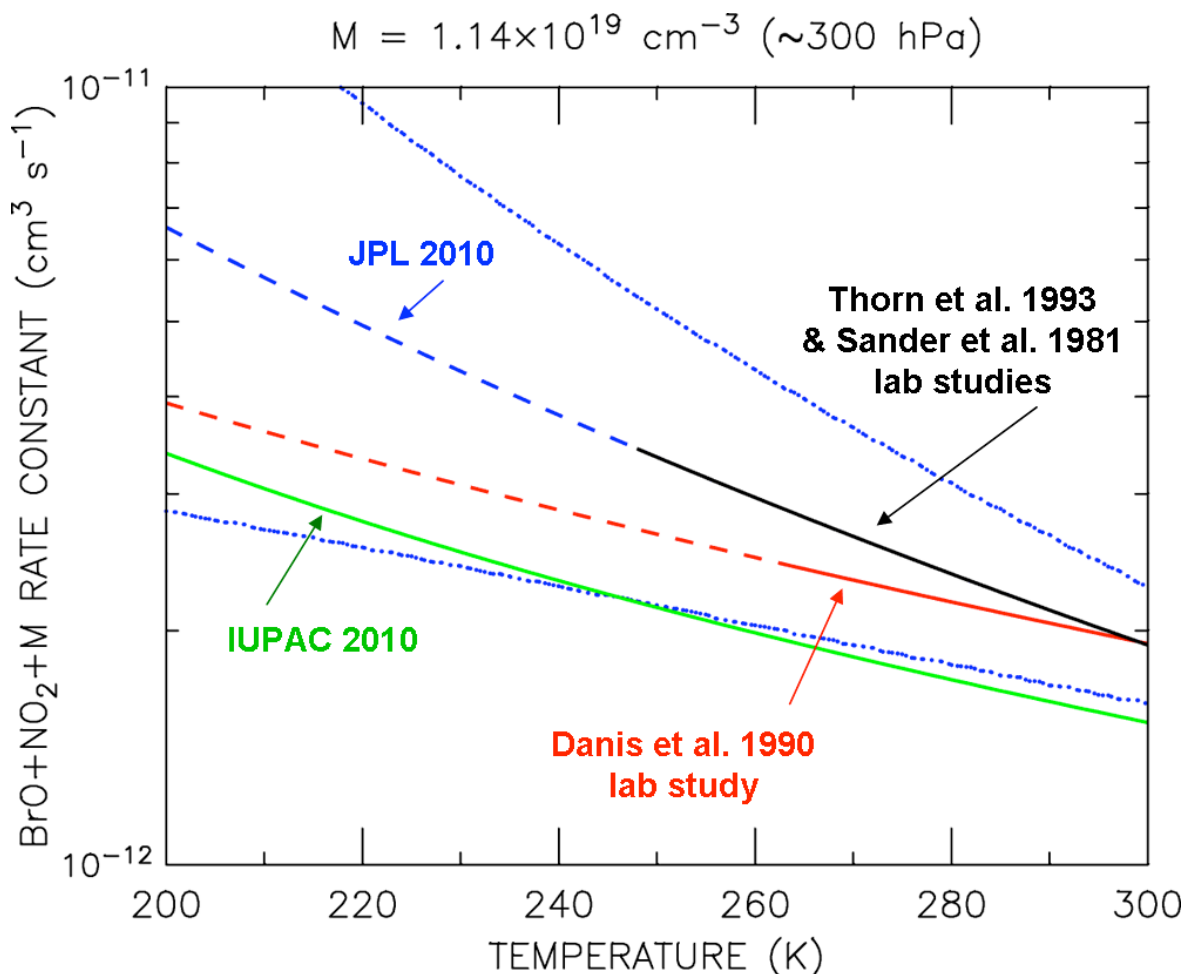
We have also added the following new text to Section 3.3:

Here, we do not consider systematic errors in SCD_{Total} or VCD_{Total} . Systematic error will generally result in either a geographically uniform over-estimate or under-estimate of total column BrO (VCD_{Total}). There is synergy between systematic error in VCD_{Total} and our prescription of the contribution of VSL species to stratospheric Br_y . If subsequent analysis shows the estimates of VCD_{Total} BrO used here are biased high by a considerable margin, then clearly we must use a smaller contribution to stratospheric Br_y to derive similar overall magnitude of tropospheric BrO. However, the geographic distribution of tropospheric BrO will not be strongly altered due to this synergy. An exploration of the systematic error in VCD_{Total} and the implication for tropospheric BrO will occur following analysis of a ground-based, OMI BrO validation campaign conducted in Fairbanks, Alaska during April 2011.

which we feel addresses this important point.

18) -P22, l 22: the author states the BrO+NO₂ rate constant is uncertain by a factor of 2 without giving any reference. Sinnhuber et al. (2002) gives a much smaller uncertainty (~25%).

> The uncertainty of this rate constant for the temperature of the lower stratosphere really is a factor of 2, as indicated in the figure below:



In the submitted version of the paper, we had referred to Sander et al. (2006) in the sentence prior to the discussion of BrO+NO₂ rate constant on page 22. The uncertainty for this rate constant is the same in Sander et al. (2010). We would like to keep the Sander et al. (2006) reference, and not use Sander et al. (2010), because Sander et al. (2010) was not available at the time we conducted the calculations in this study. We believe the use of Sander et al. (2006) in the sentence immediately prior to the one in question constitutes our “giving a reference”.

The uncertainty has a strong T dependence. Sinnhuber et al. (2002) perhaps looked at much warmer temperature, or perhaps earlier compendiums had smaller uncertainties. Regardless, we are certain our statement is correct and it is properly referenced to the NASA/JPL evaluation.

19) -The uncertainty on the stratospheric column due to VLS uncertainty is likely much higher than 27% especially for low tropopause conditions.

> We have added a new figure to a new Auxiliary Material section showing the geographic variability of the difference between the Salawitch et al. (2010) and

Theys et al. (2011) estimate of stratospheric BrO for 17 April 2008. We have spent a lot of time trying to understand the nature of this difference. While the low altitude tropopause plays a role, it is not the sole factor. Regardless, inclusion of this new figure is an important step forward, as it documents the differences between these two commonly used representations of the stratospheric burden.

Section 4.1.1

20) -Fig 6a, P24, l4-8: The patterns in the maps are very dependent on the color bars. The one used (same for total and tropospheric columns) is a bit misleading.

> We prefer to use the current scale because it successfully shows spatial structure of both total and tropospheric column BrO and, at the same time, also shows the difference in the magnitudes of these two columns. We experimented quite a bit with the color scale before settling on the one we have used.

21) -Fig 6b, P25, l1: There are indeed low values of derived tropospheric columns over thick clouds but it seems that there are also high values (although it is not easy to see from Figure 6). Maybe the author should investigate this a bit more.

> We now state, in the revised paper, that high values of satellite-derived tropospheric column BrO associated with thick clouds can be a result of BrO above or inside of thick clouds. For example, profile #12 (Figs 2 and 6b) shows a plume of enhanced BrO at about 3 km altitude that could have been above the cloud deck.

We have added a new sentence to the revised paper, Section 4.1.1, stating:

On the other hand, high values of satellite-derived tropospheric column are found along the DC-8 flight path both before the descent in altitude at ~24:15 UTC and after the plane has ascended. Data from both OMI and MODIS indicate optically thick clouds were present during and after the ascent. The high value of satellite-derived tropospheric column during and after ascent could be due to the presence of BrO above or within these clouds. Profile #12 (see Fig. 2) shows a plume of enhanced BrO at about 3 km altitude, which could have been above the cloud deck.

Further investigation is beyond the scope of the paper. Our work, along with that of Theys et al. (2011), are the first to quantify the impact of clouds on satellite-derived tropospheric BrO. Indeed, should we so desire, we could criticize many prior studies for not going as far, in this regard, as our study and that of Theys.

22) Section 4.1.3

-P27, L14: It seems that the GOME -2 are shown on Fig 9a. Is there a mistake in the sentence? "(not shown)"

> Thank you. We have deleted “(not shown)”

23) -Fig 9b, P28, L 13: It is maybe good to recall that the minimum height of the aircraft was 151 m (according to Table 2b) and that is likely that the aircraft miss an important part of the BrO profile.

> Thank you for the suggestion. We have indicated this possibility in a newsentence:

The aircraft may have missed an important part of the BrO profile as the minimum sampled height was 151 m above the surface.

Conclusions:

24) -P33, l 13: The author gives thresholds for the albedo (>0.8) and SZA ($<80^\circ$) that are different from the values given in the manuscript.

> Good catch! We have changed the paper and now use a consistent SZA thresholds of <80 deg throughout. Also, “albedo (>0.8)” has been changed to “albedo (>0.7)” to be consistent with the rest of the paper. Thanks for pointing out this inconsistency (we’ve read the paper too many times to spot items such as this).

25) -P33, l27: The author should be cautious in the conclusions. Although it is true that the variability of stratospheric BrO is such that it can modify our interpretation of total columns maps, I think that at high SZA ($>75^\circ$) the error bars are large and the regions of enhanced tropospheric BrO not apparent in maps of total columns should be treated carefully.

> We have added the following new sentence to the revised paper to address this concern:

Caution should be applied when interpreting satellite-derived tropospheric BrO for SZA greater than ~ 80 degrees because, under these conditions satellite radiances have decreased sensitivity to absorption by tropospheric BrO (Fig 4).

We prefer to use 80 deg, rather than 75 deg, for this new sentence to be consistent with our SZA threshold (see above comment).

26) Typos: -P10, l14: “miunte” → “minute” -P12,l24: what is ‘UV-2 pixel size’? do you mean OMI pixel size?

> Thank you for your careful reading! We have fixed the typo in the word “minute”.

UV-2 refers to the OMI channel used to retrieve BrO. We have clarified our use of UV-2 by changing the sentence to read:

The spectral resolution in the OMI UV-2 channel used to retrieve BrO columns is approximately 0.5 nm. ... The pixel size of OMI UV-2 channel is approximately 13 x 24 km² at the swath center and significantly larger at the swath edges.