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Atmospheric Measurement Techniques Discussions

Interactive comment on "Airborne DOAS limb measurements of tropospheric trace gas profiles: case study on the profile retrieval of O_4 and BrO" *by* C. Prados-Roman et al.

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Review of doi:10.5194/amtd-3-3925-2010, Airborne DOAS limb measurements of tropospheric trace gas profiles: Case study on the profile retrieval of O4 and BrO by Prados-Ramos et al., for publication in Atmospheric Measurement Techniques.

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 is used 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.

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. 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.

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.

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

a) 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 O_3 and NO_2 considered? If so, how? Was the BrO profile "slided" or "stretched" to match the tropopause height at the specific locations? 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 O_3, NO_2, 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.

b) 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? How does the comparison of IUP-HD epsilonM versus in situ extinction profiles look for this flight?

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.

d) 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 no agree, strictly speaking. 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. 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. 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.

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?

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.

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.

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.

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 $\sim$30 ppt 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, 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.

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. Also, the

statement (page 3951) suggesting that aerosols and clouds do not obscure 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. 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.

Minor comments:

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

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.

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 asso-

ciated 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.

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

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

6. Page 3932, line 1: text describes a broad spectral region, including two spectrometer, 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? 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).

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

8. Page 3934, line 19: not sure the word "artificial" is appropriate. Perhaps "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 O3, O4, 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.

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).

11. Page 3939, line 14 and page 3940, line 13: phrase "in situ measured" is awkward. This combination of words is unusual.

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?

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

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).

15. Page 3943, line 25: better to repeat the integration time, 10 s, here. A small amount of redundancy can be very helpful.

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

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 ?!?

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

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.

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.

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.

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.

23. Figure 9: It would be nice to be able to see the O_3 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 O_3 in the BL are obscured.

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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.

END OF REVIEW.

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