

Interactive comment on “Rapid methods for inversion of MAXDOAS elevation profiles to surface-associated box concentrations, visibility, and heights: application to analysis of Arctic BrO events” by D. Donohue et al.

Anonymous Referee #2

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Donohue et al. present measurements of BrO by MAX-DOAS at Barrow, Al. MAX-DOAS is a maturing technique that relies on the well established DOAS technique, and has been applied multiple times to measure BrO in the Arctic. The technique requires radiative transfer in order to convert the measured slant column densities into photon path independent measures for BrO abundance, e.g., surface associated concentrations, or vertical column densities. Three approaches are discussed and compared for internal consistency. The paper is interesting and potentially meritorious of publication

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in AMT. However, the paper would benefit from major revisions prior to publication.

General comments:

The actual process of measurements, data classification and filtering procedures etc are not made explicit in the paper. Fundamental quantities such as the elevation angles used for measurements are not mentioned in the text, but only implicitly given in form of captions and legends of Figures. As a result the paper is somewhat difficult to follow, and frankly not transparent. What sequence was used for scanning the angles? Which direction was the telescope pointing? How are the four cloud scenarios defined, and how did each scenario apply to the measurement period discussed? The dSCD measurement accuracy appears to be equated with the '1-sigma fit error' of the program; however, it is well known that the fit error underestimates the actual accuracy of the DOAS retrievals by factors as large as 6 (Stutz and Platt, 1996) yet this fact is not discussed. How does uncertainty propagate?

Further, much of what is presented in the paper in form of the three analysis approaches is not actually 'new'. Rather, the discussion of the three approaches as it stands is rather disconnected from existing DOAS literature that deals at a more fundamental level, and/or in more detail with each and any one of the three approaches. I agree with the community comment by Volkamer and Sinreich that this literature should be discussed. The possibly to transfer approaches to other environments remains essentially undefined partly due to limited discussion of the existing literature. Similarly, the authors miss an opportunity to discuss recent literature on BrO in the Arctic, which would benefit the paper but is currently missing.

The authors do not give a definition for 'visibility' in the text. From the Caption to Figure 5 it later becomes apparent that 'visibility' is taken as the $1/e$ extinction length due to aerosols alone, ignoring Rayleigh scattering at 360nm. This is incorrect. First, the term 'visibility' is reserved for wavelengths where the human eye is sensitive. It has no meaning at the UV wavelengths measured by the authors. Second, the physics of

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‘visibility’ depends on factors like ‘color of objects’, ‘detectable contrast ratios’ and it is proportional to the inverse of the ‘combined extinction length from aerosol and Rayleigh scattering’ (ignored here). The actual ‘visibility’ is several times larger than the ‘aerosol extinction length’ calculated by the authors. Hence, the title is somewhat misleading, and ‘visibility’ should be removed. The authors should also substitute ‘visibility’ for more accurate language that relates more closely to their measurements throughout the manuscript.

Specific comments:

- 1)P4647, I2: the list of gases does not match the citations, nor do the references quote the original work (for OVOC). The authors should revisit the literature to correct this.
- 2)P4647, I14: What is the difference between a surface associated VCD and a VCD? The vertical column density (VCD) is defined from ground to the top of the atmosphere. If found the use of another than the usual notation confusing.
- 3)P4651, I15: What is the reason that LZ-MAXDOAS should not be useful to provide information on daily or seasonal variations in the trace gas abundances? Such a general claim appears to be unsupported.
- 4)P4651, I22: The retrieval of BrO is known to be sensitive to the wavelength window fitted. What sensitivity tests were performed to establish the robustness of the BrO retrieval? What is the absolute accuracy claim of the BrO retrievals?
- 5)P4651, I28: A description of the actual procedure used for the spectra wavelength correction is missing.
- 6)P4652: A description of the measurement sequence, elevation angles, time resolution is missing. The paper would benefit if the retrievals were better described. How is detection sensitivity defined? What is the absolute accuracy of the retrievals?
- 7)Fig. 1: the RMS for panel (A) is not listed.

8)Fig. 2: The error bars appear to be very small, given the RMS noise level listed in Fig. 1. Notably, the RMS noise levels in Fig. 1 do not appear to be free of systematic structure. Under those conditions the absolute accuracy of DOAS fits can be up to 6 times higher than the 1-sigma fit error (Stutz and Platt, 1996). Indeed, if the RMS value from Fig. 1b is typical (the spectra shown may be best cases – are they?) then the absolute accuracy in the BrO dSCD could be as high as 1×10^{14} molec cm⁻² for the LZ-MAX-DOAS evaluation procedure, and higher for the DZ-MAX-DOAS procedure. This is about five times larger than the error bars listed in Fig. 2 (consistent with the 1-sigma fit error being plotted?). What is the information content of error bars in Fig. 2? How were they calculated, and what do they mean to indicate?

9)Throughout the manuscript (each Figure + text) discussion of uncertainty is currently largely missing. How is the error from the dSCD retrievals propagated in the three retrieval methods?

10)P. 4653, I18: Which ranges of SZA and Azimuth apply? Which ranges were explored?

11)P. 4654, I10-14: Reference to ample MAXDOAS literature that discusses these effects is missing.

12)P. 4655, I3ff: The authors appear to imply that there is ambiguity in the solutions of MAXDOAS retrievals of VCDs. What is the purpose of averaging? Why is this needed? Is the 'range of SA-VCDs' exploited further by the approaches? Does the average of a 'range of SA-VCDs' increase or decrease the signal to noise? Can the need for averaging be understood from an argument that has radiative transfer at its core? The paragraph would benefit from addressing these questions, and might need some reorganization of information.

13)P.4655, I16: The following two statements appear to be in contradiction: 'the O₄ concentration is independent of the height of the boundary layer' and it 'exponentially decreases with height'.

14)P. 4657, l6: Notably, clouds do not necessarily limit the RMS of solar straylight retrievals. On the contrary, the more frequent scenario is that the higher photon count associated with ground based DOAS observations reduces photon shot noise and thus improves the RMS. How do the authors define 'quality of MAXDOAS analysis'? What are the reasons for choosing clouds as the basis for classifying the data in four categories? What is the mechanics of how this was done?

15)L. 4657, l15ff: The 1-sigma fit error appears to be equated with the dSCD error. See general comments, and comment #8 for why this is a lower limit measure of uncertainty. The bias between programs points to a systematic error, in addition to the statistical uncertainty. How was the overall error of data products from the three approaches calculated? And how large is the error? Notably, the telescope elevation angle uncertainty will give rise to a different error depending on which elevation angle the telescope is pointing at. This is not discussed.

16)P. 4657, l26ff: several typos, 'km' should probably be 'm'.

17)P. 4658, l2: How are the cloud types defined? The authors should provide some justification for the given errors.

18)P. 4658, l13: The elevated viewing method is well established for geometric AMF calculations. A reference is missing. Also, some discussion of geometric AMFs would be useful in context of the aerosol profiles observed by the authors. The benefit of the approach of Sect. 2.6 over conventional approaches is currently neither transparent, nor demonstrated. How does the approach from Sect. 2.6 compare to the geometric approximation? Is an elevation angle of 10 degrees still suitable for the elevated viewing method, or does the uncertainty in the radiative transfer cancel out the benefit of higher sensitivity to the boundary layer? What is the improvement of the authors method over the geometric approximation? What measure do the authors apply to decide that there is indeed a benefit of their method over the traditional geometric approach?

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19)P. 4658, I15: What defines the threshold at 2km? In principle MAXDOAS remains sensitive also to gases in the free troposphere if the measurement precision is sufficient. This appears too generic a statement.

20)P. 4658, I25: The horizon viewing method is not entirely new. I agree with the community comment that a reference is missing. Also, the approach as described here seems to be an oversimplification that – though interesting – is difficult to generalize based on the results discussed.

21)P. 4660, I11: There is no information given about the frequency of the observed aerosol profiles. Such information could possibly be published as supplementary information.

22)P. 4660, I21: The authors should give the full equations. What is the offset? Is it significant? The full equation (incl. errors) would be more informative.

23)P. 4660, I3: How can the VCD (units of molec cm⁻²) be determined by dividing the SA-VCD (same units) by the layer height (units: cm)? Something is wrong here.

24)P. 4660, I12: See comment #22. The full equation would be more informative. What is the offset? Is any offset significant?

25)There is considerable need for more information in the text, yet the conclusion section can probably be synergized by a bulleted list of findings and research needs. The conclusion should not repeat information from the abstract. Rather it might list the assumptions that underlie the individual approaches, which assumptions are likely to be the most sensitive? Where are the boundaries of the applicability of the approaches? The 'demonstration of success of the method' would require more discussion on sensitivity studies with respect to when the assumptions are not fulfilled (to be done in the text, else: what is demonstrated in the paper?). The ninety profiles are calculated for a single SZA, Azimuth and surface albedo value. The boundaries of the approaches (e.g., if aerosol and gas were not collocated, what is the role of clouds, Azimuth, SZA

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effects, measurement geometries and conditions) are reduced in the conclusion section to the effect of 'visibility', yet it remains difficult to judge the possible role of other factors from the given information. It might not be the intention of the authors to imply a general claim, but the paper creates the impression that simple and largely linear approaches – while appealing – might be a new 'gold standard' to 'check that a more advanced analysis. . . is giving reasonable results' (quote from concluding sentence of the paper). This notion should either be avoided, or needs to be further justified.

Interactive comment on Atmos. Meas. Tech. Discuss., 3, 4645, 2010.

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