

Interactive comment on “A New Differential Optical Absorption Spectroscopy Instrument to Study Atmospheric Chemistry from a High-Altitude Unmanned Aircraft” by Jochen Stutz et al.

Anonymous Referee #2

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Referee Comment on amt-2016-251 by J. Stutz et al., 2016

The manuscript describes a new DOAS (Differential Optical Absorption Spectroscopy) instrument which is deployed on NASA's Global Hawk UAS (unmanned aerial system) and measures scattered sunlight in the UV and vis wavelength regions in limb scanning geometry. Analyses of atmospheric trace gases are performed at altitudes between the upper troposphere and the lower stratosphere.

The setup of the instrument, its viewing geometry, as well as the purpose and benefits

C1

of the specific methods are described and presented. In addition, first measurement results of several trace gases are presented and discussed. Derivation of mixing ratios from DOAS observations is a challenge especially at high altitudes. The authors use and describe a new method to derive BrO and NO₂ mixing ratios at high altitudes from the observed slant columns by using in-situ ozone observations for scaling purposes. Many aspects including the error influences of several model assumptions are analysed in detail. Deployed on the GH on a frequent basis, the instrument may yield valuable insights into and better understanding of the lower stratospheric abundances of O₃, BrO and NO₂. Reference to a companion paper is given in a suitable way. The content and significance of the manuscript are well within the scope of AMT.

Most aspects are well explained and the quality of the measurements and analysis is good. However, the text would benefit in some parts if the authors put some more carefulness in the writing, i.e. the phrasing and correct grammar. There are quite a number of mistakes/oversights such as missing words, as well as a few passages which are not so well described. This makes the reading somewhat cumbersome in the respective places, while the content itself is mostly fine. Please check the text once more in addition to the comments given here. With some additional effort on the text and some clarifications, the manuscript is certainly suitable for publication in AMT.

I. Main Comments

Comment 1. Section 4.2, I. 1059

Please describe and/or cite what is used as "typical BrO concentration profile". It is also unclear how the choice of this influences the uncertainty determination.

C2

Comment 2. Section 4.3, I. 1202-1208

Please give some more clear explanation on how (strong) the model assumptions and especially the finally applied values of SCD_{ref} and the vertical trace gas profiles from the TOMCAT/SLIMCAT model, which constrain the RT simulations, influence the mixing ratio results received from the O_3 -scaling method. This influence is also mentioned further below (Section 4.3, I. 1242). It becomes clear that certain effects and assumptions cancel in the applied method but others don't. It is also clear from the detailed descriptions and equations that this input information is needed. However, it is necessary to understand in how far the results and resulting interpretations from the announced new scaling method are independent from the initial assumptions, especially SCD_{ref} and the vertical profiles.

The Supplement gives a wide range of valuable information on influences from the model assumptions on the final results, as summarized in Table 5. However, SCD_{ref} is not considered and the vertical profiles are only scaled (either for the tropospheric or the stratospheric part) but not changed in their actual shape. If this (i.e. absolute changes as opposed to the shape in the two separate altitude ranges) is representative for the entire profile influence on the final results this should be stated more clearly. One could imagine that especially relative changes in the profile shapes between ozone and NO_2 or BrO are relevant for the final mixing ratio results. Please add some explanations as clarification.

Comment 3. Section 4.3, II. 1269-1303, Figures 12 and 13

In part this comment is connected to Comment 2. It is not clear how the shifting procedure is performed. At first reading the procedure seems easy. However, comparing with Figures 12 and 13 some questions arise. First of all, the caption of Figure 13 is really misleading. It states that the measurement results are forced to match the model results, which according to text and figures should be vice versa. The measurement curves are the same as in Fig. 12, and the model results change from Fig. 12 to Fig. 13. As stated in the text, the model is forced to match the measurements, correct?

C3

The change in ozone between pre and post shifting in some parts of the flight, however, is quite large up to a factor of two it seems (e.g. between 18:00 and 20:00 UTC), and maybe around 10-20% in other parts of the flight. The text, however, states that the vertical shift needed for matching the curves is usually smaller than 1km. Is the ozone changing that strongly between one model layer and the next? Maybe there is some misunderstanding. Maybe it would be also useful to give some more information on the used vertical profiles in general, e.g. in the Supplement, see also Comment 1. In addition: Why is a vertical shift sufficient? Is this simply because of monotonically increasing concentrations? Please briefly clarify.

Comment 4. Section 6

As outlook and for a better understanding of how the measurements performed by the new DOAS instrument on the UAS will yield further information on the UTLS, it would be very helpful if a statement - e.g. at the end of the conclusions - would be given on the future plans, especially on how often and for how long etc. the instrument can - and will be - used for the type of measurements presented here, and for which further research the results will be helpful.

II. Minor Comments

1. Lines 19-20: It is somewhat strange to concentrate on relative errors here. In order to judge the performance of the instrument, absolute errors are in addition helpful.
2. Line 57: Please state and cite actual values (current best knowledge) of total amounts of Br_y^{org} and Br_y so the given numbers can be directly set into relation.
3. Line 87: Is 3ppt the current uncertainty or the goal for the improved uncertainty of Br_y ? Please state the respective other quantity. (The newly achieved uncertainty would be interesting, however, according to the grammar of the sentence the given

C4

value is the current uncertainty. Please clarify.)

4. Line 97: Earliest measurements of? Does this refer to BrO?
5. Lines 305-307: Please give a short reason (too dark due to lack of scattered sunlight?).
6. Lines 388-390: This does not really explain how the opening angles are actually determined. The scanning capability obviously helps in the calibration. Please shortly state how the FOV was measured.
7. Lines 425-428: Although this range of the telescope stability is within the uncertainty of the elevation calibration, it obviously adds to the overall uncertainty of the determined elevation. Is this included in the uncertainty considerations? It doesn't sound as if it is. Or maybe the phrasing is misleading?
8. Lines 457-461: What about the roll angle? Probably it is just missing in the list.
9. Lines 549-551: A common shift would not be an error of the trace gas reference spectra which are all from individual data, but a (very small in the presented case) uncertainty from the instrument calibration which seems to be fine.
10. Line 556: Please refer to Table 3.
11. Lines 567-570: Reading this remark, the reader asks for an explanation, which is only given much later. Either shortly state or refer to Section 4.
12. Lines 575-578: The excluded range lies within the fitting windows for O₃ and BrO also, but is not excluded there, right? A short comment of reasoning would be helpful. (Why does the feature lead to a problem only in the O₄ fit?)
13. Line 608: Please give the ozone window also in the text as for the other trace gases, not only in the table.
14. Line 644: New paragraph?
15. Lines 698-705: Please check this passage, there is some inconsistency. If the reduction in O₃ and NO₂ is "not as pronounced" it would mean that at EA 1° the DSCD is similar as in the lower EA, so the relative error for O₃ and NO₂ would be even more problematic if using EA 1° as reference instead of direct sun. Please clarify.
16. Lines 749 and 763: Is it true that one instrument/method gives relative er-

C5

rors and the other one absolute errors only? Otherwise please use comparable data/information.

17. Line 792: Is an albedo of 0.2 realistic for the VIS? This seems rather large.
18. Lines 845-849: Please briefly state which observation leads to the interpretation (transition from lower stratosphere to UTLS region).
19. Lines 1006-1007: This information should be given earlier in the section, i.e. at first mentioning.
20. Lines 1389-1391: As the sentence can be read in a somewhat misleading way, a recommendation would be a rephrasing of the following kind: "As further analysis goes beyond the scope of this manuscript, we only briefly discuss the results (...)."
21. Lines 1485-1486: (...) i.e. low mixing ratios in the troposphere (...). This is, however, not generally true for NO₂ at all. This statement requires some clarification.
22. Lines 1216-1219 and Figure 10: Does the calculation depend on certain experimental flight conditions? Just trying to understand why there are kinks in the curve, e.g. at 700s. This is not clear. Please give a short explanation.

III. Technical and text/grammar corrections

1. Line 5-9: (...) developed for (...) GH (...) _and tested/applied_ during (...) ATTREX.
2. Line 26: (RT) not needed in abstract, abbreviation should be given in main text (Line 199?).
3. Line 151: (...) interpretation _of_ the data.
4. Line 159: Sounds as if the name of the aircraft is missing.
5. Line 181: New paragraph?
6. Line 193: Please add abbreviation DSCD at the end of the sentence as definition.
7. (...) the side facing _of_ the diffusers (...)
8. Line 541: The term "dispersion" seems to be the wrong choice. Do you rather mean

C6

"convolution"?

9. Line 673: (...) and in _the/a_ companion paper (...)
10. Lines 770-773: Sentence misses a verb. E.g. (...) we also employ a 3D atmospheric chemistry model to _determine_ vertical trace gas concentration profiles.
11. Lines 813-816: Although (probably) not wrong, the sentence structure makes it really hard to understand the meaning, especially the subject of the sentence "TOMCAT/SLIMCAT model meteorology".
12. Line 832: Remove the space at the end of the sentence.
13. Lines 860-861: (...) at the time and location _of_ the measurement (...).
14. Lines 987-989: Missing word "and": (...) _and_ a non-linear retrieval (...) is employed.
15. Line 996: The solution of this iteration is _a_ (instead of "an") vertical (...) profile (...).
16. Line 1052: (...) the general error of _a_ (instead of "an") non-linear extinction (...)
17. Lines 1077-1082: Devide into two sentences please.
18. Line 1081: "than" instead of "that"
19. Line 1124: typo in "predomina_n_tely"
20. Line 1125: (...) the ratio of the two parameters _is_ (instead of "are") a measure (...).
21. Line 1140-1142: Please correct this sentence.
22. Line 1144: Due _to_ (...)
23. Line 1171: "B" should be in italic format.
24. Lines 1269-1270: (...) requires addition_al_ scrutiny (...)
25. Line 1457: Please delete the extra "the".
26. Line 1460-1461: Apparently this sentence belongs to a different paragraph. It does not fit to the content here at all, but probably should rather be shifted to line 1490.
27. Lines 1470-1471: The verb "are" needs to be repeated here at least once: (...) and _are_ optically more powerful (...).
28. Line 1481: (...) needed to be develop_ed_ (...).

C7

29. Table 2: The wavelength range for the VIS instrument really seems to be wrong. The given range from 410-425 nm would not cover the retrieval windows given in Table 4.
30. Table 4: Please correct the title of the last column. e.g. Average DSCD error (molec/cm², for O₃: molec²/cm⁵) or similar.
31. Figure 2 and 3: O₃ (please use subscript).
32. Caption of Figure 2: delete second "the" in second of last line.

Missing plural "s", e.g., at the following locations:

Line 153: Few research aircraft_s_
 Line 158: (...) a number of aircraft_s_
 Line 214: Theoretical consideration_s_
 Line 1046: (...) these result_s_
 Line 1290: (...) the modeled O₃ profile_s_ are (...)
 Line 1326: (...) spectral retrieval error_s_ (...) which are (...)
 Line 1471: (...) instrument_s_

Corrections w.r.t. references and citations:

1. Line 40 (and many other locations): The format of references / citations has some issues. Please remove the extra parentheses around the year of publication.
2. Line 163: Parentheses around the two references are missing.
3. Please correct typo in name Baidar in Baidar et al., 2013.

Corrections concerning the Supplement:

Text correction in the figure captions: (...) parameters given _in_ Table 5.

C8