

Interactive comment on “Probing the subtropical lowermost stratosphere, tropical upper troposphere, and tropopause layer for inorganic bromine” by B. Werner et al.

Anonymous Referee #2

Received and published: 13 November 2016

This study by Werner et al. presents DOAS measurements of bromine monoxide in the tropical and subtropical upper troposphere, tropopause region and lower stratosphere from the Global Hawk. These are important observations in a key region of the atmosphere. While the interpretation of the DOAS observations, and in particular the applied O₃ scaling technique, has to rely on a number of assumptions, this may be the best technique available to measure bromine monoxide in this important atmospheric region. However, I suggest that more details on this method and the uncertainty due to the assumptions made are given here, rather than referring to the companion paper by Stutz et al.

The paper would clearly benefit from some rearrangement of the presented material,

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as further explained below. At parts also more detail is needed, as given in my specific comments below. With these modifications and after consideration of the other comments I recommend publication in Atmos. Chem. Phys.

Specific comments:

Abstract: The abstract is too long and should focus on the main findings.

p1, I1: why does this list here starts with CH₄, O₃ and NO₂ ? I suggest using a similar statement as on page 4, where the list starts with BrO, as this is really the focus of this study.

p2, I2: split: "...LS. In the TTL ..."

p2, I9: how do these numbers relate to the reported range of BrO in the TTL?

p2, I12: top of TTL defined to be 425K in line 3

p2, I14: "chemical depletion": not clear what this means. 1/3 of observed global ozone trends (and if yes: over which period, which altitude region,...) or 1/3 of the chemical loss?

p2, I16: what does "mostly by natural and anthropogenic" mean? Are there other sources than natural and antropogenic? Or do you mean mostly by natural, but also some antropogenic sources?

p2, I31: Maybe useful to include a sentence or two on observations of BrONO₂ in the stratosphere (e.g., Höpfner et al.)

p5, I1: a few lines below, the phrase "a large number of species, including O₃, NO₂ and BrO" is used, which may be appropriate here as well. I suggest to give this list of possible species only once (and consistent) and refer to it.

p5, I1: maybe better move long list of references into section 2.1

p7, I29: "The received limb radiances ...": need a few more words that this refers to the

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mini-DOAS measurements. However, I suggest merging section 2.6 with 2.1, as this is an essential part of the mini-DOAS data analysis.

p8, l4: Strange sentence: "Demonstrates that Earth sphericity, ...are relevant". I suggest to rather say, "...are relevant and taken here into account."

p8, l23: "which together contain 1 ppt of bromine atoms" just repeats the first part of the sentence, or I don't understand what is meant here.

p8, l25: include explicitly that no sea salt aerosol source is assumed, in contrast to some other recent studies (e.g., Saiz-Lopez et al.)

p8, l28: "growth rate": why is the growth rate relevant? Because the CH₄ content varies with age-of-air?

p9, l1: How? By changing the BrONO₂ photolysis, by changing the rate of BrO + NO₂, or both?

p9, l18: include "as well as" after the reference to Jensen et al.

p10, l1: closing bracketed has to be after "tropical"

p10, l6+l7: "optical" -> "optimal"

p10, l32: I couldn't easily find information on the integration time. Please organise the description in a way that all relevant information for the DOAS measurements can be easily found at one place of the manuscript. Currently this is distributed over Secs. 2.1, 2.6 and 3

p11, l1: Is really a higher spatial resolution required, or a finer temporal (or SZA) resolution?

Fig 7: Maybe it would make sense to indicate in the caption of Fig. 7 that DOAS data quality for SF₄ is reduced?

Fig. 10: I assume for Fig. 10, model data are used as they are, i.e. not altitude

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adjusted?

p12, l4: The agreement between measured and modeled NO₂ is indeed strong evidence for the validity of the approach, but be careful with the reasoning: It is not possible to validate both measurements and model at the same time from this comparison.

p12, l7: better say "surface air mixing ratios"

p12, l9: "data is " -> "data are"

p12, l13: Not sure if you can draw this conclusion from this comparison. Does this not simply show that there is some spatial variability in CH₂Br₂ while the model assumes a constant mixing ratio at the surface? See next sentence.

p12, l16: The sentence should finish before "... to be implemented in the model". Whether or not this should be implemented in a model is a totally different issue.

p12, l27: I assume Wang et al and Volkamer et al use the same measurements, so better say "the TORERO measurements reported by Wang et al. and Volkamer et al."

p13, l3: I suggest to discuss similarities and differences to the results of Wang et al and Volkamer et al., but limit the speculations about possible discrepancies. My impression is that too much weight is given on explaining possible differences to the TORERO results, while other studies are not mentioned.

p13, l3: spell out the name of the aircraft

p13, l6: rephrase sentence, avoid the double use of "but"

p13, l6/7: This sentence does not contain any solid information and could be removed: It is trivial that any two measurements that are not performed at the same place at the same time could differ just by chance.

p13, l8-12: I find this statement problematic: What do you want to imply?

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p13, l33: remove "Again"

p14, l3: remove "and others" - already contained in "e.g."

p14, l4: What does this mean: "with these features in mind"?

p14, l5: Probably misleading formulation: if you really know there is a bias of 2ppt you could correct for it.

p14, l6: "bottom to" -> "bottom of"

p14, l10: Not sure what you mean here. Increasing CH₂Br₂ in the model would be easy, and does not require a detailed back trajectory study. But would this "remove flight to flight scatter"?

p14, l14: "well be" -> "will be"

p14, l15: "gap in" or "gap between" ?

p16, l26: "climate is most sensitive": maybe better say more carefully "where ozone changes have the largest impact on radiative forcing"

p16, l33: "oxidizing capacity due to expected increase in VSLs emissions" is probably not what you mean. There are actually three possible processes at work: (1) changes in atmospheric transport, (2) changes in OH, affecting VSLs lifetimes and (3) changes in VSLs emissions due to aquaculture.

p17, l4: "some" -> "important" (?)

p17, l12: what kind of "adjustments" are performed here? Please give more details and justify!

p18, l3: Hossaini et al., 2016 is now published

p18, l9: remove extra "to"

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-656, 2016.