

BrO and inferred Br_y profiles over the Western Pacific: Relevance of Inorganic Bromine Sources and a Br_y Minimum in the Aged Tropical Tropopause Layer

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Response to Reviewer 2; 02 December 2017

Black: Referee's comments

Blue: Author's reply

Green: sentence added/modified in the manuscript

We greatly appreciate Reviewer 2's comments on the manuscript. Particularly, they have identified where specific statistical or methodological details were and missing and have helped sharpen the precision of language.

In this manuscript, several airborne DOAS BrO profile measurements over the Western Pacific are reported which were taken during the CONTRAST field campaign. Using a box model constrained by the DOAS BrO data and additional measurements taken on board the aircraft, Br_y profiles and bromine partitioning are derived. The Br_y profiles are then discussed with respect to altitude and characteristics of the respective atmospheric layer, compared to 3d model output and to previous results. The manuscript is overall well written although in some places hard to read. The topic of the study is of atmospheric relevance and fits well into the scope of ACP. The measurements presented as well as the discussion and interpretation provided are interesting and thought provoking and I therefore recommend this manuscript for publication in ACP. I do however have some concerns and suggestions which the authors should address before the paper can be accepted for final publication.

General comments:

- An important limitation of the paper (which the authors acknowledge) is that out of the Br_y budget, only BrO was measured. None of the other gas-phase bromine species were measured, nor any aerosol phase bromine. Aerosols are only constrained via aerosol surface area but the authors don't seem to trust these measurements. Washout is another unknown in this system. As a result, the inferred Br_y profiles must have considerable uncertainty which the authors address by two sensitivity runs of their box model. However, in the discussion (and also title, abstract and summary), the Br_y profile often is treated as if it had been measured. In my opinion, the authors should

– consider changing the title into something like “BrO profiles over the Western Pacific: Implications for Br_y and . . .”

We have changed the title to “BrO and inferred Br_y profiles over the Western Pacific: Relevance of Inorganic Bromine Sources and a Br_y Minimum in the Aged Tropical Tropopause Layer”

– make clear throughout the discussion that the Br_y is inferred, not measured

We have made a number of changes to further clarify that Br_y is “inferred” or “estimated” rather than “measured” or “observed”. Specifically:

P02,L06: “observed” changed to “inferred”

P13,L32: “inferred” inserted before “Br_y”

P16,L12: “inferred” inserted before “Br_y”

P16,L21: “inferred” inserted before “Br_y”

P16,L33: “observed” changed to “inferred”

P17,L20: Change: “There is a Br_y minimum” to “There is an inferred Br_y minimum”

P18,L21: Change “better reproduces Br_y” to “better reproduces box model inferred Br_y”

P20,L33: “measured” changed to “inferred”

P20,L34: “measured” changed to “inferred”

– add a discussion of the uncertainties in the Br_y profiles (I’m not sure what the error bars in Fig. 2 represent but they seem rather small to me considering the uncertainty of the BrO measurement and allowing for some uncertainty in the partitioning)

Thank you for this comment. Indeed, a definition of error bars was missing. The following text has been added in Sect. 2.3.1:

The primary box model output is the ratio BrO/Br_y computed with 30% error (Dix et al., 2013; Wang et al., 2015). This is added in quadrature with the BrO errors outlined in Sect. 2.1.2 above when computing the error in Br_y.

We have additionally added the following text in a revised Sect. 3.1:

The additional error from modeling is not immediately apparent in the rightmost panels of Fig. 2 because the 30% error contribution (Sect. 2.3.1) is relatively small compared to the error in the BrO measurements (mean proportional error of 54%).

... and in Sect. 3.2:

Again, as in Fig. 2, the error from modeling is not always apparent in the lower panels of Figs. 4 and S7 (see Sect 2.3.1).

– explain how the confidence limits quoted in the manuscript were derived

The statistical treatment is now outlined in the first paragraph of the conclusions:

We identify a gas phase Br_y minimum in the aged TTL which is robust to assumptions regarding heterogeneous chemistry. Considering all box model sensitivity studies which differ significantly (excluding Case 3 in the troposphere, and Case 2 in the stratosphere as they are not significantly different) the mean Br_y observed is 3.6 ppt (2.9-4.4 ppt) in the upper FT and TTL, 6.3 ppt (5.6-7.0 ppt) in the stratospheric middleworld, and 6.9 ppt (6.5-7.3 ppt) in the stratospheric overworld; where the values in brackets reflect 95% confidence intervals of the mean. This contrasts with 2.7 ppt (2.3-3.1 ppt) in the aged TTL. Even taking into account the uncertainty spanned by the different box model cases utilizing single tailed Wilcoxon-Mann-Whitney rank test (Mann and Whitney, 1947) (refer to supplement for details) we find that Br_y in the aged TTL is: 1) less than in the lower stratosphere (>99% confidence), 2) less than in the convective TTL and upper FT (>96% confidence), and 3) that it is a local minimum (>95% confidence). To our knowledge such a Br_y minimum has not previously been observed or hypothesized.

The statistical tests are further described in more detail in the supplement:

Statistical tests of the aged TTL minimum

We test whether there is a local minimum in the aged TTL in three steps, first we assess the likelihood that Br_y in the aged TTL is less than Br_y in the convective TTL and upper FT ($B_{\text{ry}}^{\text{aged TTL}}$

$< Br_{y\text{ uFT,cTTL}}$), second the probability that Br_y in the aged TTL is less than Br_y in the lower stratosphere ($Br_{y\text{ aTTL}} < Br_{y\text{ LS}}$), and finally the joint probability that these are simultaneously true.

Comparison of inferred Br_y in the upper FT and convective TTL with the aged TTL presents a challenge, the distribution of inferred Br_y values in the upper FT and convective TTL is skew and not normally distributed; to tackle this a Wilcoxon-Mann-Whitney rank test is used (Mann and Whitney, 1947). In brief, this method sums for elements from one distribution the number of elements from the other distribution which they are greater than, termed the U -statistic. The U -statistic is assessed against a null hypothesis; because the distributions (e.g. variance and skew) of Br_y in the upper FT and convective TTL, and in the aged TTL are dissimilar a one-tailed null hypothesis is stated in terms of stochastic ordering. Specifically, we test against the null hypothesis, $H_0: Br_{y\text{ a}} \geq_{st} Br_{y\text{ b}}$, i.e. that for any and all concentrations of Br_y the likelihood of a random inferred Br_y in region a is greater than that concentration is greater than the likelihood that a random inferred Br_y in region b is greater than the same concentration. A statistic related to U , ρ , assesses the overlap of the data sets; a value of 0.5 indicates a statistical tie, while values of 0 and 1 indicate a sweep for one set or the other.

Applying Wilcoxon-Mann-Whitney rank tests we find that Br_y in the aged TTL is lower than in the convective TTL and in the upper FT ($U = 2781$, $n_{\text{uFT,cTTL}}=111$, $n_{\text{aTTL}}=60$, $\rho=0.4176$, $p<0.04$, reject $H_0: Br_{y\text{ aTTL}} \geq_{st} Br_{y\text{ uFT,cTTL}}$), and that it is lower than in the lower stratosphere (LS) ($U = 4$, $n_{\text{uFT,cTTL}}=51$, $n_{\text{aTTL}}=60$, $\rho=0.0013$, $p<<0.01$, reject $H_0: Br_{y\text{ aTTL}} \geq_{st} Br_{y\text{ LS}}$). The p value for the second test is vanishingly small; applying the Vysochanskij-Petunin modification of the Chebyshev inequality obtains a more conservative value of $p \approx 0.02$. Taking the more conservative significance and using the Bonferroni-Holm (Holm, 1979) method we can reject both null hypotheses with $p<0.05$. With 95% confidence there is a Br_y minimum in the aged TTL.

As reflected above, we have further revised confidence intervals lowering the statistical leverage by a factor of 3 to reflect that the Br_y inferred for different box model cases is not independent.

These revised confidence intervals are reflected in the abstract.

– acknowledge that to some degree the comparison of the Br_y from the 3d-models with the Br_y profiles from their work is a model-model intercomparison, not a validation with measurements
itemize

We have added the following sentence early in Sect. 4.3.

Br_y from the experimentally constrained box model estimates is compared with chemistry/climate models, this is fundamentally a model-model comparison. However, significant information can be gleaned by leveraging the constraints on the box model and the representation of transport processes in the global models.

- One of the main results of the manuscript is detection of a BrO / Br_y minimum in the “aged TTL”. This is nicely visible in Fig. 7 and also Fig. 6 but not in the BrO profiles in Fig. 2, which give very little indication of a maximum between 12.5 and 13.5 km. I assume that this differences results from inclusion of RF15 and RF06. This raises the question of how

representative the profiles in Fig. 6 or Fig. 2 are. This is of particular relevance considering that the CIMS BrO measurement do not appear to support the C-shape finding. Please comment.

We have modified Figs. 6 and 7 to better illustrate the C-shape. The previous Fig. 6 had whiskers calculated on data from case 1 only, and included data from the aged TTL which contributed to a masking of the C-shape due to the complex structure in Br_y in the TTL. We have further revised Fig. 7 to show where profiling and jet crossing case studies can be separated and added the following text to reinforce that the minimum and TTL structure does not rely solely on the jet crossing case studies. The dashed line in Fig. 7 now makes transparent which data are from the profiling case studies, and which data are from the horizontal lags at altitude. This helps address concerns whether the profile case studies are representative. Indeed, the data from profiling and jet crossing flights connect quite naturally.

We have added the following text to Sect. 3.2. to clarify:

Observations from the profiling case studies are all in the convective TTL by these criteria. This is to be expected, given that the profiles are in the Western Pacific warm pool, where convection to 15 km and above is persistent. The GV can only probe the TTL closer to the tropopause through latitudinal transects where the relevant region is at lower altitude.

We have also inserted the following sentences in later in Sect 3.2. to reinforce the point.

The aged TTL was probed during RF06 and RF15.

The convective TTL was probed briefly in RF03 and subsequently in RF06, RF07, and RF15.

We have added a statistical treatment of the inferred Br_y data in Sect. 4.5 to further support the C-shape in inferred Br_y for convective air masses. Data in the aged TTL has been removed in the revised Fig. 6 for both BrO and Br_y. For BrO, including the aged TTL data leads to a more pronounced C-shape, but this change is due to changes in BrO_x partitioning due to elevated O₃ in the aged TTL (not shown in the manuscript).

Regarding the CIMS measurements, the MU CIMS does observe BrO in the MBL, but measurements are limited to altitudes below 9 km. The GT CIMS had limited sensitivity, and no data were reported in the MBL. We note that BrO peaks of up to 30+ pptv in the GT-CIMS data were measured in the LS. These data were considered non-physical, and showed clear correlations with O₃ peaks; these data have been filtered during QA in Chen et al. 2017. The agreement between the filtered CIMS data and DOAS is reasonable in the LS, as is discussed in more detail in another paper (Wales et al., 2017).

Figure S10 shows the comparison of tropospheric data, AMAX-DOAS data are generally in agreement with the range of CIMS measurements in the FT, where measurements disagree is in the MBL and TTL. We have added the following language to further clarify regarding the MBL clear.

Below 3.5 km AMAX-DOAS observes more BrO than the MU CIMS (Fig. S10a), though there is overlap in the distribution of observations. The MU CIMS does observe instances where BrO

is high near the surface (Le Breton et al., 2017) corroborating the variability observed by the AMAX-DOAS.

- Another important finding is the relevance of a sea salt aerosol bromine source to reproduce the observed BrO profiles. While I agree with this conclusion for the lowest altitudes, I do not see “strong evidence” from the measurements or the comparison to the models for such a link in the upper FT. I therefore suggest to remove this statement or at least to make it less bold.

We have modified the conclusion text as follows:

Our data provide evidence that a SSA derived bromine source is necessary to reproduce the observed profiles of BrO and Br_y near the surface and suggest the influence of SSA is relevant even in the upper FT.

- More details need to be given on which measurements exactly were used to constrain the box model. In Table 1, several quantities listed are measured by multiple instruments. Is IO from the DOAS instrument really used to constrain the model, and if so, which values did you observe? Which measurements are included in the GV data called “water”?

IO was measured, but it was not used to constrain the model and has been removed from the table accordingly.

Details of how measurements were used to constrain the box model are provided in the supplemental material under the heading “**Gas Phase Measurements Used to Constrain the Box Model**” this includes descriptions of how measurements were used when available from multiple instruments. Further details pertaining to the use of data from a specific instrument are discussed along with instrument descriptions in subsequent portions of the supplement.

The water measurement aboard the GV was made using the Vertical Cavity Surface Emitting Laser (VCSEL) hygrometer also described in the supplement. VCSEL is described in Zondlo et al. (2010).

- The authors define several abbreviations and use them extensively throughout the text which in places makes for difficult reading. I’d suggest using only standard abbreviations, in particular in the summary.

We have avoided abbreviations in the abstract summary, and the conclusion section.

Abbreviations are introduced at first use in the main text, and summarized in a Table as part of the SI text.

- I think that it would be helpful for the readers to have a brief outline of what is presented in the manuscript and why at the beginning of the manuscript, for example at the end of the introduction

Following on the introduction of methods and the CONTRAST campaign, we have added the following further paragraph outlining the content of the remainder of the paper.

The measurements and models are presented in Sect. 2. Section 3 describes the DOAS measurements aboard the GV, as well as measurements used to constrain a chemical box model to infer total gas-phase inorganic bromine (Br_y); this includes sensitivity studies and a discussion of different chemical regimes for heterogeneous chemistry that recycles Br_y . Section 4 discusses the results by comparing with two global models (GEOS-Chem and CAM-Chem), and places them in context with the existing literature. The atmospheric implications of tropospheric halogens for atmospheric composition and our understanding of bromine sources are summarized and discussed. Section 5 presents the conclusions and provides an outlook.

Minor comments:

- P2,12: profiles is found => profiles are found

Corrected

- P2,16: Cl not defined

Defined on first instance

- P2,17: TTL not defined

Defined on first instance

- P3,126: suggest to rephrase: “microwave radiometry has . . .”

Rephrased as suggested

- P4,116: whcih => which

Corrected

- P4, 123: “and is actively” – rephrase (grammar)

“and is” dropped, sentence now reads:

Scattered sunlight enters the telescope along a well-defined field of view (vertical dispersion of 0.17°), actively motion stabilized with a pointing accuracy better than 0.2° in real time

- P4,125: “above geopotential horizontal” – is geopotential horizontal something different than just horizontal?

“geopotential” is specified here because angles are defined against the plane perpendicular to the local gravitational field, i.e. the horizontal plane is assessed against the geopotential field. This is important for accurate modeling of the radiative transfer; gravitationally downward is toward the center of the Earth at relevant precision. This is not always the case if the horizontal were defined against constant buoyancy approximately an isopycnic surface nor for isobars which an aircraft will maintain at long range as pressure altitudes are used to define flight level.

- P5, 11: “Low altitude . . .” duplication

Eliminated the clause “Low altitude zenith spectra were used as references” appended the other clause “with the nearest qualifying spectrum in time being used.” to the previous sentence. Now reads:

Zenith spectra with at least two minute integration times collected at low altitude under relatively clear skies were used as reference spectra, with the nearest qualifying spectrum in time being used.

- P5, 15: “in one of two ways optimal...” Something missing here (colon?)

Colon inserted, “parameterized” changed to “parameterization” to match part and case of “optimal estimation”.

- P5,124: What is the advantage of orthogonalising some of the cross-sections to the first coefficients of the polynomial?

Fitting out broadband absorption as a polynomial is a defining feature of DOAS. A complicating effect is that some absorbers such as NO₂ and O₃ have broadband absorption in addition to narrowband differential absorption. If the cross-sections are not orthogonalized this broadband absorption can have spectral crosstalk with the polynomial. Even if this does not impact fitting of the target gas BrO, modified fits of NO₂ and O₃ in the BrO spectral window cannot be compared to optimized fits for the gases which can serve as an important check.

- P5,127: mention that non-linearity is from wavelength alignment only

The relevant passage has been clarified as follows:

Absorption by relevant species is fitted simultaneously using the non-linear Marquardt-Levenberg algorithm with full non-linear treatment reserved for shift, stretch, and intensity offset (Danckaert et al., 2012). This is done in finite wavelength windows targeting specific trace gases.

Note that while the most terms are fitted linearly following each step through M-L, the results of this fitting then seed the following M-L step impacting the non-linear minimization. The overall algorithm could be described as Marquardt-Levenberg with a QR decomposition subroutine for linear parameters, except for the fact that the steps are run sequentially for a given iteration.

- P8,128: “An additional . . .” – what is the benefit of mentioning an additional sensitivity study without reporting any results?

There should be a further clause making clear that the results are not shown because they are nowhere significantly different from Case 1.

An additional sensitivity study (not shown) explored the potentially large surface area contribution from super-micron particles (not measured by UHSAS) that are visible in the CDP data at lower altitudes and found no significant difference from Case 1.

- P9, 130: organic compound => organic compounds

Corrected

- P10,18: emissions six => emissions of six

Corrected

- P11, 13: at last => at least

Corrected

- P12, 14: with altitudes => with altitude

Corrected

- P13, 115: “Relative to the aged TTL is characterized. . .” – something missing?

The sentence is meant to refer to the convective TTL, the subject of the previous sentence. Relevant passage corrected as follows:

As the name suggests, the convective TTL is influenced by recent convection, and includes high altitude air which is essentially tropospheric. Relative to the aged TTL, it is characterized by high H₂O and corroborated by elevated CO, low O₃, and low θ .

- P14, 125: “The impacts . . . is” => The impacts . . . are

Following the use of “effect” as singular just prior changed to:

The impact . . . is

- P14,128 and 130: What do you mean by “convoluted”?

Replaced with “indistinguishable”. Since reaction rates operate on the product of SA and γ since there are no terms to be assessed dependent on one and not the other. Similarly, a zero dimensional model cannot separate washout from heterogeneous chemistry since this would require knowledge of transport which the model lacks.

Assumptions about reaction probabilities on surfaces, γ also play a role; SA and γ are indistinguishable and the product of SA \times γ determines many heterogeneous reaction rates. The box model does not represent washout, and this too is indistinguishable from SA and γ

- P16, 15: “. . . is most sensitive in the . . .” – sensitive to what?

“to chemical assumptions”

- P18,128: “. . . ae more consistent” => is more consistent

Corrected

- P19,110: during same period => during the same period +enditemize

Corrected

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