

We thank the referees for their valuable time and careful reading and constructive comments on our research and paper. Below we provide specific responses to the referee comments and outline the corresponding revisions made to our paper. Note that the revisions to the manuscript do not change the reported data or the conclusions from this work.

Referee #1

Referee Comment: Page 32964, line 18. Is it possible to specify sources of bromoform into the atmosphere?

Response: We can include some additional general information in the introduction to provide the reader with a better perspective of CHBr_3 sources.

Action: The sentence has been revised as follows: “ CHBr_3 (bromoform) is a short-lived atmospheric trace gas primarily emitted from natural sources such as marine phytoplankton and coastal macrophytes (seaweeds), and represents a source of reactive bromine (Br_y ; $\text{Br} + \text{BrO}$) in the troposphere as well as the stratosphere.”

Referee Comment: Page 32969, line 14. What is meant by “had pathlengths”

Response: Here, we are referring to the single pass absorption cells of different lengths used in the course of our experiments.

Action: The sentence has been revised as follows to improve the clarity: “The Pyrex absorption cells used had optical pathlengths of 10, 50, and 100 cm depending on the range of CHBr_3 concentrations being used, which was between $(0.2\text{--}150) \times 10^{15}$ molecule cm^{-3} .”

Referee Comment: Page 32972, line 16. Correct : the 532 nm measurements (not: the 532 measurements)

Response: Agree

Action: Text has been revised to: “...the 532 nm measurements established an upper-limit to the possible contribution of a Br_2 impurity to our UV measurements.”.

Referee Comment: Page 32972, line 26. Equation 3: It would be useful to give the wavelength and temperature range for the use of Eq. 3 for the calculation of absorption cross sections.

Response: This is an important consideration in the application of our data to calculate atmospheric photolysis rates. The range is described in the heading of Table 2, which contains the actual results of the parameterization. We feel that this is the most appropriate place to give this range as this is where modelers will get the parameterization numbers, i.e., they may not catch this if given in the body of the paper and it is not necessary to repeat it.

Action: No changes made to manuscript.

Referee Comment: Page 32974, line 14 In equation 4, the altitude symbol should be capital Z

Response: Agree

Action: Equation revised.

Referee #2

Referee Comment: Indicate in figures (at least one) the regions where results are extrapolated and therefore less reliable (lower temperature regions). It would be useful for these regions be readily identifiable in at least some of the figures even if those regions aren't precisely definable. I'd like to be able to easily understand the altitude in the different panels of Figure 3 above which the mean temperature $<260\text{K}$, and whether or not the uncertainties shown in this figure include any additional magnitude associated with extrapolation.

Response: We agree that including a visual indication of the regions where the cross section parameterization was extrapolated in our calculations would be useful. The loss rate uncertainty calculations shown in Figure 3 include the estimated uncertainty in the extrapolated cross sections, i.e., the uncertainty estimate is included in the high/low loss rate calculations. Detailed modeling calculations would be needed to provide more specific attribution of the errors than is already given in our paper and was not included in this study.

Action: A four panel figure of the temperature climatology used in our loss rate calculations, using the same format used in the paper, has been added to the supplementary material. This should enable the reader to compare the various atmospheric regions in the calculation without overly complicating the figures within the paper.

Referee Comment: Make the Abstract more informative. Other than the ODP estimates, the abstract provides little concrete information regarding the new results. I'd suggest you mention how the new results differ from earlier ones (even in general terms), and highlight one or more of the four points discussed in the figures with respect to changes in our understanding of atmospheric loss of CHBr_3 (e.g., lines 20-22 on p. 32976, or lines 3-5 on p. 32977, or lines 3-5 on p. 32978).

Response: We can provide some additional general statements within the abstract for better reader perspective.

Action: The abstract text has been revised as: "A parameterization of the CHBr_3 UV spectrum for use in atmospheric models is developed and illustrative photolysis rate calculations are presented to highlight the impact of the revised $\sigma(\lambda, T)$ values on its calculated local lifetimes, e.g. the photolysis rate in the tropical region obtained with the present spectral data is 10–15% lower (longer lifetime) than obtained using currently recommended cross section values."

Referee Comment: Be clearer about conclusions related to quantum yields of CHBr_3 (p. 32974, lines 3-8). Is there a possibility that the dissociative quantum yield of CHBr_3 is much less than 1.0 from 324 to 436 nm, making discussions of lifetime related to photolysis in this region inappropriate? Quantum yield of Br at 300-324 is indicated as being "less than unity" but with the large error bars can one say if it is likely to be different from unity? Line 21, p. 32966, don't you mean: at wavelengths between 300 and 436 nm?

Response: We have reviewed the available literature and current state-of-knowledge regarding the quantum yields in CHBr_3 photolysis. It seems that the quantum yield is indeed unity, but this is not something addressed in our study other than indirectly in our photolysis rate calculations. We do, however, need to clarify the wavelength range over which knowing the quantum yield is most important, that being <350 nm.

Action: The text has been revised as "...; the Br atom quantum yield was expected to be unity at the wavelengths most critical to atmospheric photolysis, >300 <350 nm (Peterson and Francisco, 2002).

Referee Comment: Improve wording. p. 32965, the WMO Assessment did not list a "global lifetime" for CHBr₃ from all loss processes or from OH. It provided a local lifetime estimated for specific conditions. Different wording is required here. Also, 22 ppt total Br discussed in WMO(2011) was for 2008, not present-day, is it the same number now?

Response: Several minor changes to the text are needed to more accurately describe the CHBr₃ lifetime and stratospheric Br abundance.

Action:

Revisions to address lifetime issues:

"Loss rates (lifetimes) of CHBr₃ occur on timescales shorter, or comparable, to the time scales for atmospheric circulation and are, therefore, dependent on the location and season of emission as well as the local conditions (e.g. UV flux, OH radical abundance, and temperature). UV photolysis is thought to be the predominant atmospheric loss process for CHBr₃ with a globally averaged photolysis lifetimes on the order of 10s of ~36 days in the Tropics. The global lifetime with respect to the OH reaction is ~75 days (WMO, 2011) yielding a total global lifetime of ~24 days. A recent kinetic study has reported an OH + CHBr₃ reaction rate coefficient that is greater than used in previous modeling estimates of the CHBr₃ lifetime, implying a shorter lifetime with respect to the OH reaction (Orkin et al., 2013), and shorter overall lifetime.

Revision to address Br abundance: "Current best estimates are that very short-lived substances (VSLs) contribute 3 to 8 ppt to the stratospheric bromine budget; the ~~present day~~ total stratospheric bromine abundance in 2008 was is estimated to be ~22 ppt (WMO, 2011)."

Referee Comment: p. 32977, line 4, do you mean a shift in the spatial "distribution" or "relative contribution" of the different loss processes?

Response: We mean relative not spatial.

Action: Revised text slightly as: "...and a shift in the relative distribution of the loss processes ..."

Referee Comment: p. 32977, line 21, perhaps "most efficient transport" rather than "greatest transport". Those authors didn't argue that Asia is a large source.

Response: Agree, we need to revise text slightly.

Action: Revised text as: "... which have found that the Asian sub-continent ~~emissions~~ in the Summer leads to the greatest transport of VSLs to the stratosphere."

Referee Comment: p. 32978, line 20, why "potent"? What defines potent from not? You've chosen to give ODPs for emissions from the region with the largest ODPs. Are emissions from other regions also potent? More precise wording seems warranted here.

Response: The magnitude of the ODP values themselves imply the potency of CHBr₃.

Action: The text has been revised slightly for further clarification of the results from the Brioude et al. study: " The mass transfer for emissions from Europe, mid-latitude North America, and East Asia regions were also considered in the Brioude et al. study and were found to be considerably less effective (about one order of magnitude) in transport of VSLs to the stratosphere and, therefore, are not included in the present discussion (see Brioude et al., 2010)."

Referee Comment: Supplement Figures S1-S11, describe what is different about the isothermal results indicated with different symbols.

Response: The different symbols represent independent experiments performed under different conditions. This is addressed in the Experimental Section of the paper as variations in the experimental conditions.

Action: The following text has been added to the appropriate figure captions in the supplement: “The different symbols are results from independent experiments including variations in experimental conditions as described in the text.”