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Interactive Comment

Interactive comment on "New Aura Microwave Limb Sounder observations of BrO and implications for Br_y" *by* L. Millán et al.

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We thank the reviewer #3 for his thoughtful comments and suggestions that have helped to improve the paper. In addition to enacting changes he suggested, as detailed below, we have also made some additions and updates to the paper to improve clarity and underscore areas where our product represents a significant improvement on earlier versions.

In the course of making our modifications, we recognized that the averaging kernel plot (figure 3) was incorrect in the submitted draft. The earlier version indicated the new product had more sensitivity to the lower stratosphere than is in fact the case. In the light of this we have revised our estimate of the valid vertical range of the product to 10-4.6 hPa. We note that our estimate of total Bry is unaffected by this update.

However, the vertical range over which the new MLS observations usefully overlap with the other sensors is narrower, restricting the range of interest for the analysis shown in the comparison with other datasets.

Reviewer comments:

This paper reports new retrievals of BrO profiles from the Aura MLS instrument and interprets these observations in terms of how much bromine is supplied to the stratosphere by very short lived substances.

I believe the paper could be acceptable for AMT after major revisions. The article falls short of adding much to our knowledge of atmospheric bromine in its present form. According to the AMT website, the main subject areas of this journal are the development, intercomparison and validation of measurement instruments and techniques of data processing and information retrieval for gases, aerosols, and clouds. The submitted paper falls particularly short in the areas of intercomparison and validation.

Most of the paper is devoted to a description of the new BrO retrieval. This is overall solid but still needs work before it can be considered up to the norm for AMT. It looks like the grey line for retrieval is clipped at 1 hPa for the lower left hand panel of Figure 5. Most importantly it is stated that "throughout most of the profile, the main source of systematic bias arises from retrieval numerics. While unsatisfactory this is expected due to overlapping O3 signals in contrast to the small BrO signature : : :" (page 331). In other words, I think, the authors are stating that interference from O3 is a major limiting factor. But Figure 5 contains a line for Contaminant species errors, and the uncertainty for shown by this line is small. So, I am honestly quite confused as to whether, or not, uncertainty in overlapping O3 is driving the large bias shown by the grey line in Figure 5. If so, the decision to use average T, O3, HNO3 (page 329) from the standard product, and apply to the averaged radiances, needs a much more thorough consideration. Upon revision, need a more thorough description of the grey line in Figure 5 and, if overlapping gases are an issue, should calculate how uncertainties in each overlapping species impacts the BrO retrieval. If it is not overlapping species, then need to explain what exactly is meant by retrieval numerics.

It is explained that the gray line is the difference between the unperturbed run and the model atmosphere, estimating the errors due to the retrieval.

The contaminant species errors do not quantify the effect of the overlapping species per se but rather the uncertainty in their mixing ratio (this will be explained in the new draft).

The real weakness of the paper is "intercomparison" and "validation". Essentially there is no validation despite the fact that over the time span of Aura there have been a number of balloon-borne measurements of stratospheric BrO (i.e., see for instance http://www.sciamachy.org/products/index.phpspecies=BrO&subspec=BrOp&institute=IUP).

The "intercomparison" in the paper is presented entirely in Figure 7, which is qualitative at best. Upon revision, would like to see some accounting for differences in local solar time of observations. This is straightforward to accomplish; see for example Appendix A of: http://www.agu.org/pubs/crossref/2006/2005JD006479.shtml) Once this is done, a scatter plot including correlation coefficients, estimates of mean offsets, etc. is needed.

Through the years there have been a number of ballon-borne measurements of stratospheric BrO, unfortunately, only one of the publicly available flights coincide with the latitudinal and vertical range of the MLS OL2 measurements. A comparison was made against it to address the "intercomparion" issue in the study.

To address the local time issue a tabulated photochemical model was used to map the balloon, SCIAMACHY and OSIRIS datasets to the MLS local time. The section 5.1 of the new version of the paper will explained the details.

The main science result of the paper, an estimate of Bry from VSLS, is at best a "fuzzy message" because: 1) between about 20 and 50 hPa, the Bry values inferred from

MLS agree quite well with the lower limit for Bry, represented by the blue WACCM curve (WACCM neglected VSLS Bry) whereas at altitudes above 10 hPa, the MLS value of Bry agrees quite well with the green SLIMCAT curve (model that includes significant Bry from VSLS). This "jumping" of MLS Bry from one curve to the other is not discussed. Of course, it is hard to interpret physically, but this "jumping" would lead many readers, including me, to question whether the new retrieval is adequate to use for quantification of VSLS Bry. Or perhaps the MLS team believes they have pushed forward our understanding of the shape of the BrO profile. Regardless, the shape of inferred Bry should be addressed. Had balloon-borne BrO profiles been part of the analysis, we could possible assess whether the shape of BrO profile reported by MLS is realistic.

Due to the revised vertical range of the product, the 20 and 50hPa agreement is not recommended for scientific use.

2) the value for VSLS Bry is leveraged to an estimate for the tropospheric burden of bromine from Montzka et al., 2003. The difficulty in using a tropospheric CH3Br time series is that, due to its short tropospheric lifetime, CH3Br at the tropopause is almost certainly lower than CH3Br at the surface. This 10 to 15% difference between surface and tropopause level CH3Br, which is well known to the aircraft measurement community, is neglected here (as well as many other studies). But it is likely important for the accounting that is being attempted.

This was taken into account in the Montzka et al (2003), study, and it will be mentioned in the new draft.

3) uncertainties in the inference of Bry from BrO are similarly neglected. Nearly all other papers on this subject examine the uncertainties of chemical kinetics and J values, including (but my no means limited to) the aforementioned Sioris et al. Paper.

An 18% percent error was considered for the kinetics used to compute the scaling factor, as derived by Siriors et al (2006) and Hendrick et al. (2008).

Interactive comment on Atmos. Meas. Tech. Discuss., 5, 325, 2012.