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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Air-sea fluxes of biogenic bromine from the tropical and North Atlantic Ocean" *by* L. J. Carpenter et al.

L. J. Carpenter et al.

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We thank both referees for their careful reading and considered review of this manuscript which we believe has led to significant improvements. We would also like to point out we have slightly revised some of the numbers in this manuscript. Firstly, we reanalysed our permeation tube weighings for 2007 and based upon the new calibration factors, have increased the CH2Br2 concentrations for the RHaMBLE campaign by 12%. Secondly, we came across an error in the calculation of kw for the RHaMBLE cruise. Correction of this error has led to about 13% decrease in the fluxes of CHBr3 and CH2Br2 for this campaign (i.e. the CH2Br2 fluxes remain more or less as they were). This does not change the conclusions of this manuscript.

Referee #1 Minor mistakes (points 1-7). These have been addressed. Points 3) and 6) were in the ACPD conversion; i.e. they do not appear in the original word document.

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Interactive Discussion
Discussion Paper

We have left the units in ppt since these are more commonly reported than pmol mol-1.

8) We have removed the actual % contribution to global fluxes in the Abstract and elsewhere and included (particularly in section 3.3) more discussion of other coastal data (see also points 17-22 and 26 below).

9) It is clear in the text that we were referring to the analytical uncertainty, not the total uncertainty. Clearly, errors in kw play a significant role; these have been well-documented. However, we do agree that the errors on the air measurements have been downplayed, and have therefore changed; "The analytical uncertainty for a sea-air flux calculation (Wanninkhof, 1992), is equivalent to the uncertainty for a water measurement for compounds which are supersaturated in seawater"; to "is dominated by the uncertainty in the water measurement....."; . Note, for the conditions of the RHaMBLe cruise, an increase in atmospheric CHBr3 concentration of 50% resulted, on average, in a 6% decrease in the calculated flux.

10) Agreed. This statement has been removed.

11) The statements regarding correlation with Chl a have been removed from the discussion of the individual campaigns and instead the following has been added to the 2nd paragraph of section 3.3: "Outside of coastal regions, Table 1 shows that the average seawater CHBr3 and CH2Br2 concentrations for each region broadly increase with increasing Chl a. However, unaveraged data points show only a weak correlation (e.g. r2 of 0.39 for the CHBr3-Chl a correlation of MAP, excluding coastal data). "

12) The statement has been changed to clarify the point that the airmasses had no terrestrial or coastal input "We attribute this to the fact that during the RHaMBLe cruise, the air masses were transported from the NE over the upwelling without touching the African continent or coast (Lee et al., this issue)." (We refer the reader to the Lee et al. paper which is in preparation for this special issue and shows reverse domain filling/trajectory plots which back up this statement). Previous work (Carpenter et al., GRL 2007) shows that the increase in [CHBr3] in this region is associated with increased

ACPD

8, S10449–S10453, 2009

Interactive Comment

[CO] and [particles], indicating a coastal or terrestrial source.

13) Further analysis of the NACW and SACW water masses show that there is little difference between the Chla, CHBr3 or CH2Br2 observed, within the spread of the data (see below). On balance, we have decided to remove this discussion (and therefore Fig 4). SACW SACW SACW NACW NACW NACW CHBr3 (w) pmol/L CH2Br2 (w) pmol/L chl a mg/m3 CHBr3 (w) pmol/L CH2Br2 (w) pmol/L chl a mg/m3 mean 9.82 3.42 2.23 8.14 2.35 1.98 stdev 5.064242 1.349869 1.943605 6.293028 1.377709 3.210043

14) We thank the reviewer for bringing this discussion in Quack et al. 2007 JGR to our attention, and have included discussion of these possible other effects for apparent surface depletion of CHBr3 relative to CH2Br2 as referred to by these authors.

15) The referee is correct to state that to estimate the global emission ratios accurately, one would need to do this on the basis of saturation anomalies. However, there is insufficient data reported in the literature on co-measurements of CHBr3 and CH2Br2 in air to make this viable, hence we used the more crude approach of using water concentrations. The text has been slightly modified to explain the approach in more detail. We also note that our data shows that the approach is valid, yielding flux ratios only ˜2% lower than those calculated using both air and water data.

In changing this section, we realised that in the original discussion we had used the values of the slopes of Figure 8 (now Figure 7) (i.e. neglected the offset) rather than the actual ratios. This has been corrected, with the result that there is an even greater difference between the CH2Br2/CHBr3 emission ratios of coastal and open ocean water.

16) We mean macroalgal-rich regions - this has been clarified.

17) The literature section has been significantly expanded to include other references. We also removed "and comparison with the literature" from the section heading, since this is not intended to be a full review.

ACPD

8, S10449–S10453, 2009

18) Done

19) Range is now included.

20) The method for the sea-air flux calculation is now included (apologies, this was an oversight). We have also included further discussion of how our coastal data compares with the literature and the possible range of flux values (see also additions to section 3.3).

21) Table 2 has been expanded to include the ranges of data. A short discussion of the possible flaws in this approach has been included.

22) We have removed the sentence regarding revision of the Butler et al. coastal emission values and simply stated the different coastal emission estimates.

23) Sentence has been reworded to "Refinement of these estimates requires further coastal seawater data, particularly in tropical regions which appear to produce large quantities of bromocarbons (Yokouchi, 2005)"

24) We have added the statements "Along with previous studies, we find that macroalgae-rich coastlines are a very significant source of bromocarbons. Surface water bromocarbon concentrations in these regions can be related to ocean depth, i.e. they decrease as the waters are diluted away from the coastline."

We also removed the final sentence "However, our data, along with others, show that coastal regions are a highly significant and possibly dominant source of bromocarbons, although the exact contribution to the global budget remains unresolved. " to avoid repetition

25) Changed to "We suggest that upwelling systems are regional hot spots of bromocarbons, but globally, supply at most a few percent of total emissions"

26) We have decided to remove estimated % contributions of different regions, instead have just highlighted the fact that our coastal emission estimates are close to current

ACPD

8, S10449–S10453, 2009

Interactive Comment

estimates of the total open ocean source (see also Abstract).

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 18409, 2008.

ACPD

8, S10449–S10453, 2009

Interactive Comment