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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Bromine partitioning in the tropical tropopause layer: implications for stratospheric injection" *by* R. P. Fernandez et al.

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Fernandez et al. use a global model, together with sensitivity calculations from a box model, to investigate the processes that control the transport of bromine from very short-lived substances through the tropical tropopause layer and into the stratosphere. Two key findings of this study are that a "tropical ring of atomic bromine" is predicted for the TTL and that heterogeneous reactions in the TTL and in the troposphere are important for the VSLS product gas injection. The manuscript is generally well written and significantly adds important aspects to previous studies by investigating the role of heterogenous reactions and the role of atomic bromine in the TTL. I recommend publication in Atmos. Chem. Phys. after consideration of the following comments.

General comments

One of the key messages of this study is the important role of heterogenous recycling for inorganic bromine. However, I am slightly confused on which types of surfaces heterogenous reactions are considered in the troposphere. In particular, is recycling on tropospheric non-sea-salt aerosol included? Fig. 7a shows SA_LIG throughout the troposphere from the surface to the upper troposphere. Is this all sea-salt aerosol?? Section 2.1.3 list only reactions on stratospheric sulfate aerosol, NAT, ice particles and sea-salt aerosol. Table S1 list reactions on stratospheric sulfate aerosol, NAT, "water-ice aerosol" and sea-salt aerosol.

In addition to a recycling of HBr and HOBr to active bromine, sea-salt aerosols provide non-stoichiometric reactions that act as an additional source of bromine. It would be interesting if numbers could be given of how large this additional flux of bromine is in this model simulation. How are sea-salt aerosols treated in the model? By providing a flux of sea-salt aerosols and subsequent loss due to deposition? Or by prescribing a sea-salt aerosol surface area? How detailed is the treatment of bromine depletion or enrichment in sea-salt aerosols (e.g., Sander et al., Inorganic bromine in the marine boundary layer: a critical review, Atmos. Chem. Phys., 3, 1301-1336, 2003) in the cam_Full_Br-simulation? Given that the role of sea-salt aerosols is one of the key points of the present study, I feel that more details should be provided here.

I'm a bit worried about exclusion of ice uptake for HOBr and HBr (p.17865). Why does the implemented scheme give unrealistic results? Are there any mistakes or unrealistic assumptions that have been identified? How do you judge that the results are unrealistic, given that basically no measurements exist of these species in the TTL?

In this study, "rapid transport of Bry-rich air masses from the MBL to the lower TTL reduces the wet-deposition of PG_VSLS species" (p.17880). This appears to be in contrast to the recent paper of Liang et al. (Atmos. Chem. Phys., 14, 5781-5792, 2014), who found that stronger convection leads to more wet-deposition of Bry in their

model. So details of the scavenging and wet-deposition in the models seem to be crucial? In this light, giving more details on the treatment of wet-deposition in the current model simulations and a discussion of its effects could be helpful.

Table 4 gives for the cam_Full_Br-simulation more total bromine from VSLS in the Western Pacific upper TTL (7.8ppt) than in the lower TTL (7.6ppt). What does that mean? Is this transported into the Western Pacific from other regions? Does this apparent increase in VSLS-bromine with altitude provide an estimate for the uncertainties associated with these numbers?

Specific comments

p.17859, l.28: "increases lifetime of the more hydrophilic inorganic portion": not fully clear what that means. I guess you mean the lifetime of inorganic bromine against washout is increased by shifting the balance more to the hydrophilic portion? The lifetime of the hydrophilic portion itself should not change, right?

p.17861, l.23: Would be good to specify for how long the 5 sensitivity calculations have been run: 15 years as well, or only for the last 3 years?

p.17861, l.26: What is "dynamical transport" in contrast to just "transport"?

p.17863, l.26: I suppose you specify surface mixing ratios not surface concentrations?

p.17864, ll.15: Again, it is not clear if recycling on tropospheric non-sea-salt aerosols is included, or not.

p.17865, l.27: Define "NEU". Does this refer to Neu and Prather (2012)?

p.17867, l.2: the word "recent measurements" and the reference to the now more than 10-year old study by Marcy et al. (2004) sound contradicting.

p.17867, l.5: I don't think there is anything wrong with the assumptions for SA_ICE here, but I don't understand the rationale of assuming H2O=12.5ppm (saturation mixing ratio for the assumed conditions) and then calculating SA_ICE by condensing 12.5ppm

of water.

p.17870, l.13: why limited to O3 between 50 and 500ppb?? Fig. 8 shows that the Br/Bry ratio even increases further for O3 lower than 50ppb (as expected).

p.17871, l.8: now a range of 50ppb to 25ppb is given. Why the lower bound of 25ppb?

p.17874, ll24: why are the sensitivity calculations shown in Figs. 8 and 9 done for Run_0, when you say a few lines above that "conditions of Run_0 are unrealistic"?

p.18879, l.1: this will lead to an increase in the Br/BrO ratio in the WP, but the statement that this "corresponds to a stronger formation of the tropical ring of atomic bromine" is a bit misleading, as this does not enhance the ring of atomic bromine around the tropics outside of the WP. Or do you suggest that this also leads to higher Br/BrO outside of the WP?

Fig. 7: Something is wrong here with the notation of the runs. E.g., panel (c) states that Run_1b is SA_ice & Cly=0, while the caption states that Run_1b is SA_sulf & Cly=50ppt. According to Table 2, the caption to Fig. 7 is wrong.

Fig. 10b: I guess the scale for Cly has to be "pptv", not "ppmv"

In panel (a) you give also the SA for liquid droplets. Would it make sense to show also HOBr + HBr for liquid in panel (c)?

Technical corrections

p.17859, l.10: double bracket

Supplement, Table S1: What is the meaning of the "§"-sign after water-ice aerosol reactions?

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 17857, 2014.