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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Vertical transport rates and concentrations of OH and Cl radicals in the Tropical Tropopause Layer from Observations of CO_2 and halocarbons: implications for distributions of long- and short-lived chemical species" *by* S. Park et al.

Anonymous Referee #2

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Review of Park et al., ACPD, 10, 6059, 2010

This is a nice analysis. Measurements of a suite of tracers are used to infer concentrations of [OH] and [Cl] in the TTL. A simple model is then used to estimate lifetimes for removal of short lived bromocarbons. The authors conclude that "VSL source gases could reach the stratosphere even before chemical breakdown to product gases and make contributions to total stratospheric halogen" levels.

Major comments:

1. While this is a sound analysis and the paper is worthy of publication in ACP, I am struck that the authors do not mention published evidence showing that CH2Br2 crosses the tropopause (e.g., Wamsley et al., JGR, 1998 must be cited and discussed, and Schuaffler et al., JGR, 1999 which is cited should also be discussed). Notably lacking as well is reference to recent work of Dorf et al. ACP 2008 and Laube et al. ACP 2008, which examine the same topic. Data shown in Laube et al. quantify the impact of VSL source species due to Product Gas Injection on stratospheric Bry and Cly. Most importantly, stratospheric air masses were sampled by the WB-57 during the TC4! There certainly are observations present in the archive for O3 > 120 ppb; much of the mission was focused on stratospheric dehydration. So, the data themselves can be used to assess injection of VSL bromine and chlorine source species into the stratosphere! My own analysis of these data, for bromine, shows that CH2Br2 crosses the tropopause but that the total bromine content of all other VSL bromine bearing organics is quite small for air in the true stratosphere. Thus, the primary uncertainty for the stratospheric bromine budget remains the fate of the product gases formed upon decomposition of the organics.

The paper has great value for the estimates of [OH], [Cl], and vertical ascent rates. But the overall science impact is a lacking, since the data themselves tell us the "punch line".

I suggest, upon revision, examination of the bromine and chlorine content of VSL organics, above and below the tropopause, and commenting on the ability of the VSL species to impact the stratospheric budget of Bry and Cly from the gradient in these source gas observations relative to the tropopause. I suspect the authors will conclude that, for bromine at least, the key uncertainty remains the fate of the gases produced during decomposition of most of the VSL bromocarbons. Not sure about chlorine ... but the data are present for this to be examined!

2. There is great value in the comparison of modeled and empirically inferred [OH], yet little discussion is given to the reason for the discrepancy. Did the GMI and GEOS-Chem models allow for production of HOx from non-methane hydrocarbons? How much acetone was present in these models? Some discussion over the $\sim$factor of 2 difference between the GMI and GEOS-Chem values of OH, and the empirical values shown in Figure 6 is needed.

Minor comments:

a. The definition of the tropopause, page 6065 line 4, should be stated (i.e., is this the WMO definition? Some other?)

b. Page 6071, line 28: text reads “linked together including together”

c. Page 6075, line 14: Are the results shown from SPADE? It was STRAT that sampled in the tropics :)

d. Page 6077, bottom: numerical results for lifetimes would convey better if placed in a table.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 6059, 2010.