Conceptual investigation of the interaction of water vapor and Br_y transport across the tropical transition layer

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Introduction

The process of water vapor transport across the tropical transition layer involves both microphysical and convective processes. The bromine budget of the stratosphere can not be reconciled with ground-based bromine measurements using long-lived bromine containing substances alone. In this study we look at the interaction of the water vapor transport processes in the context of very short lived (VSL) bromine species. Lagrangian back trajectories initiated in the stratosphere are generated using ECMWF reanalysis data. A simplified microphysical and bromine chemical box model representation are then calculated along the trajectories. We present the transport of the VSL bromine in a conceptualized form and investigate sensitivities to the general chemical, microphysical and convective processes that result in VSL bromine contributing to the stratospheric bromine budget.

Conceptual Model

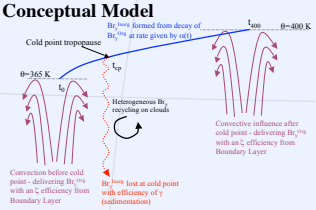


Figure 1: Schematic model of the conceptualized model

Trajectories

A detailed description of the trajectory calculations is given by Wohltmann and Rex, 2008. The trajectories are calculated using vertical winds derived from the radiative transfer equation and have been shown to be more physically realistic than those using the continuity equation. As a result however, the residence times for example between 365 - 375 K is increased to ~19 days relative to the ~ 10 days found when using the continuity equation (Wohltmann and Rex, 2008).

Three month back-trajectories from 400 K are calculated starting on of 28/2 (DJF), 31/5 (MAM), 31/8 (JJA) and 30/11 (SON) for 2000.

Convection - ζ

Deep convection over tropical coastal regions has the potential to rapidly deliver elevated quantities of brominated species from marine phytoplankton sources to the upper troposphere. To investigate this we introduced for the tropical coastal regions (defined as 2° x 2° boxes containing both land and ocean) elevated concentrations for the species known to have significant marine biogenic sources - see α determination. This process is complicated also by convection occurring under conditions where wet deposition of inorganic products also occurs - therefore we introduce a term ζ to account for the emission to convection outflow washout/loss.

Convection to before the cold point

The two extremes of no convectional addition ζ=0 (WMO values for UTLS Br_y are taken at 365K) and convectional addition from oceanic and coastal regions ζ=1 to the UT on the resulting Br_y crossing at 400 K is shown in figure 2.

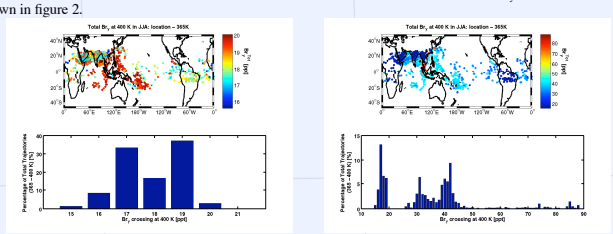


Figure 2: left - ζ=0 (no convective influence). Right - ζ=1 convective influence from every coastal and oceanic location to 365 K.

Convection after cold point crossed

Using the water vapor fields of ECMWF at 370-410 K a parameterization for daily convection using the water vapor enhancements over the median value of 1 ppmv or more, was developed. Figure 3 displays the enhanced water vapor at 400 K associated with storm systems on the 15th of August 2000, and the resulting convective strength - this being calculated with 1ppm enhancements assigned as 0 and >= 4ppm enhancements assigned as 1 and linear in between. Taking also into account whether the convection was land, oceanic or coastal, and the BL to outflow parameter ζ, the delivered Br_y is determined. The age of the air is adjusted when convection occurs (necessary as α is time dependant), thus a convective event late in the trajectory ascent results in a 'Br_y age' considerably less than the no convection case. Figure 4 displays the role of ζ and convection after the cold point on Br_y concentrations reaching 400K. Even ζ of 5% delivers substantial Br_y to the lower stratosphere.

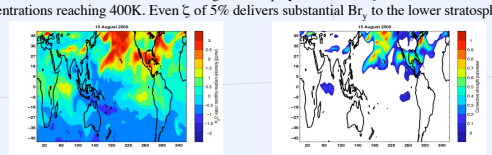


Figure 3: Difference from August median water vapor value (left) and the resulting convective strength parameter for θ=400 K (right).

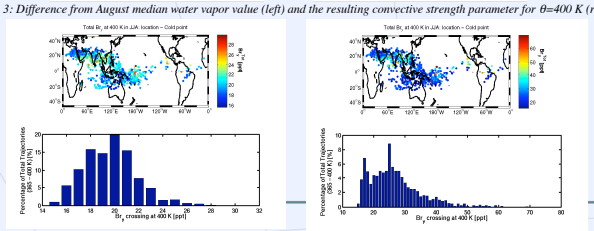


Figure 4: Convection after cold point left: ζ=0.01 and right ζ=0.05 - compare with left panel figure 2.

References

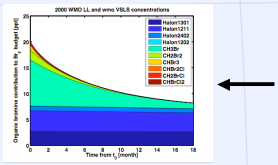
Fernandez, M.A., R.A. Cox, and R.G. Hynes, J. Phys. Chem. A., 109(44), 9986-9996, 2005
Von Glasow, R. R. von Kuhlmann, M.G. Lawerence, U. Platt, and P.J. Crutzen, Atmos. Chem. Phys., 4, 2481-2497, 2004
Wohltman and Rex, Atmos. Chem. Phys., 8, 265-272, 2008
WMO - Global Ozone Research and Monitoring Project - Report No. 50 Scientific Assessment of Ozone Depletion: 2006, 2006

Br_y Budget - α

α - is the effective lifetime controlling the rate of Br_y^{Inorg} formation and is calculated from a cumulative lifetime for all the bromine species that have lifetimes shorter than one year (VSLS +CH_3Br) - these are termed organic short-lived species Br_y^{OrgSL}. The degradation of the Br_y^{OrgSL} is given by the following simple integration:

$$Br_y^{OrgSL}(t) = Br_y^{OrgSL}(t-1) - \frac{Br_y^{OrgSL}(t-1)}{\alpha(t)}\Delta t$$

In figure 5 the expected contribution of each of the species to the stratospheric Br_y budget (Br_y^{OrgSL} + Br_y^{Halons}) as a function of time from the initial time of t_0 is displayed. The concentrations for the upper tropospheric organic bromine containing species and their lifetimes were obtained from the WMO report (2006). t_0 is treated as the UTLS entry time. Figure 4 shows that while the time spent in the UTLS is essentially irrelevant for the halons, this is not true for CH_3Br or the VSL bromine substances. Therefore, α is derived only for the VSLS +CH_3Br - the organic short-lived species Br_{OrgSL}. Note that CH_3Br is usually treated as a long lived species within CTMs and this could lead to erroneous results, as there is significant loss to inorganic bromine after 2 months with implications for lower stratospheric Br_y



Species	Upper Troposphere Conc [ppt]	Lifetime [yr (days)]	Number of Br atoms
Halon1301	2.71	65	1
Halon1211	4.01	16	1
Halon2402	0.41	20	2
Halon1202	0.05	2.9	2
CH_3Br	8.90	0.7 (256)	1
CH_2Br_2	1.29	0.33 (121)	2
$CHBr_3$	0.68	0.07 (26)	3
$CH2Br_2Cl$	0.13	0.19 (69)	2
CH_2BrCl	0.13	0.41 (150)	1
$CHBrCl_2$	0.02	0.21 (77)	1

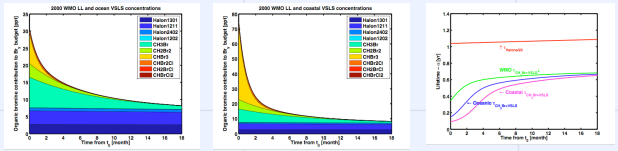


Figure 5: Upper- table and plot of bromine containing substances contributing to the stratospheric Br_y budget from t_0 (entrance time in the upper troposphere). Also shown are the plots for oceanic and coastal SLS concentrations. Far right panel displays the cumulative lifetimes of the Halons and the VSLS + CH_3Br (this being α(t)). Note as 5 km lifetimes are used these are just approximate.

Residence times

The time spent ascending through the TTL is crucial in determining how much Br_y^{Inorg} is formed (then lost) and also how much time is available for convective influence after reaching the cold point. Residence times are displayed in the following figure.

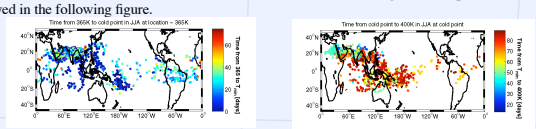


Figure 6: Time from 365K to the cold point (left) and time from cold point to 400K (right).

Microphysics - γ

As air ascends through the tropical tropopause almost all of the water condenses and is removed. Wet deposition of inorganic Br_y is the largest removal mechanism for bromine containing substances. The question of how the soluble inorganic Br_y interacts with the ice particles is a topic of current research (Fernandez, 2005). The heterogeneous recycling of inorganic Br_y into insoluble reactive forms has been demonstrated to increase the aerosol/cloud washout times from 6-9 days to 9-15 days at altitudes above 500 hPa (von Glasow, 2004). To investigate the relative importance of the microphysics we introduce a term γ that varies between 0 and 1 to represent the fraction of soluble Br_y^{Inorg} that is removed. Thereby, γ captures both the uptake efficiency of the aerosol/cloud particles and the heterogeneous recycling to insoluble bromine substances. When γ=0 then there is no removal at the cold point and the Br_y (Inorg + Org) that reaches 400 K is just the value at t_0 above in figure 5. Figures 2 and 4 assume complete washout of Br_y^{Inorg} at the cold point (γ=1), figure 7 below displays γ for 0.5 and 0.75 so washout of 50% and 75% of Br_y^{Inorg} respectively.

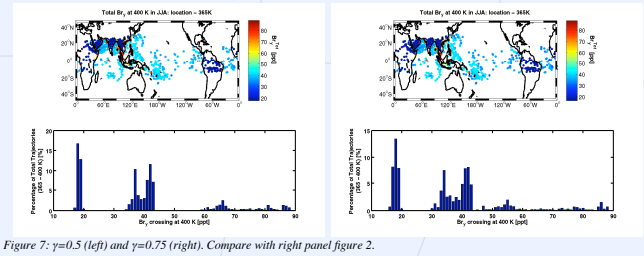


Figure 7: γ=0.5 (left) and γ=0.75 (right). Compare with right panel figure 2.

Conclusions

With this conceptual study, while a number of areas can be improved upon, we have been able to explore the sensitivity of Br_y arriving at 400 K due to convection, microphysics and emission source regions. We conclude:

- Washout at the cold point γ of 50% differs in distribution somewhat - minor sensitivity
- Convection before cold point - minor role (but caution as only the last convective event is tested)
- Convection after cold point - major role, especially if convection due to storm systems lead to sustained delivery of oceanic/coastal Br_y to the lower stratosphere - even 5% delivery efficiency influences Br_y at 400 K significantly.