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Convective transport of very-short-lived bromocarbons to the stratosphere

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Abstract

We use the NASA GEOS Chemistry Climate Model (GEOSCCM) to quantify the contribution of two most important brominated very short-lived substances (VSLS), bromoform (CHBr_3) and dibromomethane (CH_2Br_2), to stratospheric bromine and its sensitivity to convection strength. Model simulations suggest that the most active transport of VSLS from the marine boundary layer through the tropopause occurs over the tropical Indian Ocean, the Western Pacific warm pool, and off the Pacific coast of Mexico. Together, convective lofting of CHBr_3 and CH_2Br_2 and their degradation products supplies ~ 8 ppt total bromine to the base of the Tropical Tropopause Layer (TTL, ~ 150 hPa), similar to the amount of VSLS organic bromine available in the marine boundary layer (~ 7.8 – 8.4 ppt) in the above active convective lofting regions. Of the total ~ 8 ppt VSLS-originated bromine that enters the base of TTL at ~ 150 hPa, half is in the form of source gas injection (SGI) and half as product gas injection (PGI). Only a small portion ($< 10\%$) the VSLS-originated bromine is removed via wet scavenging in the TTL before reaching the lower stratosphere. On global and annual average, CHBr_3 and CH_2Br_2 , together, contribute ~ 7.7 pptv to the present-day inorganic bromine in the stratosphere. However, varying model deep convection strength between maximum and minimum convection conditions can introduce a ~ 2.6 pptv uncertainty in the contribution of VSLS to inorganic bromine in the stratosphere ($\text{Br}_y^{\text{VSLS}}$). Contrary to the conventional wisdom, minimum convection condition leads to a larger $\text{Br}_y^{\text{VSLS}}$ as the reduced scavenging in soluble product gases, thus a significant increase in PGI (2–3 ppt), greatly exceeds the relative minor decrease in SGI (a few 10ths ppt).

1 Introduction

Very short-lived (VSL) bromocarbons originate mostly from ocean biogenic sources, and when transported into the stratosphere, they exert a significant impact on the bromine budget and stratospheric ozone depletion (Kurylo and Rodriguez, 1999;

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Sturges et al., 2000). Recent years have seen significant progress in modeling efforts to quantify the contribution of brominated VSL substances (VSLs) to stratospheric inorganic bromine (Br_y^{VSL}) (e.g. Warwick et al., 2006; Liang et al., 2010; Aschmann et al., 2011; Aschmann and Sinnhuber, 2013; Hossaini et al., 2012a, b, 2013). These modeling studies suggest that brominated VSLs contributes 4.8–7 ppt to reactive stratospheric bromine, in good agreement with the 1–8 ppt estimate from satellite and balloon-borne observations (WMO 2011; Sinnhuber et al., 2005; Sioris et al., 2006; Dorf et al., 2006a, 2008; Salawitch et al., 2010).

The most important VSL pathway to the stratosphere is via convective lofting through the tropical tropopause layer (TTL). Atmospheric chemistry and transport of brominated VSLs involves the coupling of various complex processes, e.g. highly ununiform ocean emissions, convective transport of source gases and product gases, as well as wet scavenging of soluble product gases. An accurate modeling representation of these processes remains a challenging task. Modeling of these processes in general requires some extent of simplified assumptions and/or parameterizations, which may differ greatly from one model study to another. These differences have led to significant differences in the modeled Br_y^{VSL} estimates. For example, a recent model study by Hossaini et al. (2013) found that modeled Br_y^{VSL} could vary by a factor of 2 when using four different recently published emission inventories (Liang et al., 2010; Pyle et al., 2011; Ordóñez et al., 2012; Ziska et al., 2013). In general, model studies agree relatively well on source gas injection (SGI), suggesting about ~ 50 % of bromoform (CHBr_3) and ~ 90 % of dibromomethane (CH_2Br_2) can reach the stratosphere through SGI (e.g. Dvortsov et al., 1999; Nielsen and Douglass, 2001; Aschmann et al., 2011; Hossaini et al., 2010, 2012a; Brinckmann et al., 2012). However, modeled contribution on product gas injection (PGI) is highly uncertain depending on how wet scavenging is implemented. Many early model studies assume a uniform washout lifetime against wet scavenging (e.g. Dvortsov et al., 1999; Nielsen and Douglass, 2001; Hossaini et al., 2010; Aschmann et al., 2009), which was inadequate and lead to an underestimate in modeled Br_y^{VSL} (Hossaini et al., 2012a; Aschmann et al., 2011). A recent study by

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Aschmann and Sinnhuber (2013) shows that treating $\text{Br}_y^{\text{VSLs}}$ as a single soluble tracer (e.g. Liang et al., 2010) can also lead to an underestimate. They found that $\text{Br}_y^{\text{VSLs}}$ increased from 3.4 ppt to 5 ppt when switching from an idealized setup with a single soluble inorganic bromine tracer to a full chemistry scheme. Thus, an accurate modeling of transport and wet scavenging of PGI seems to be the key in narrowing the uncertainty of model estimate of $\text{Br}_y^{\text{VSLs}}$.

Understanding how the contribution of VSLs to stratospheric bromine varies with convection strength has significant climate implications. A recent modeling analysis by Hossaini et al. (2012b) suggests that VSLs source gas injection increases from ~ 1.7 ppt in 2000 to ~ 2.0 – 2.7 ppt in 2100 using the Intergovernmental Panel on Climate Change (IPCC) representative concentration pathways (RCPs) scenarios, as the future simulations feature stronger tropical deep convection transport to the lower stratosphere. On the other hand, the overall response of $\text{Br}_y^{\text{VSLs}}$ to convection strength is somewhat murky. Earlier studies deploying a uniform washout lifetime found different washout rates result a significant range in the contribution of CHBr_3 to Br_y , 0.5–3 ppt in Sinnhuber and Folkins (2006) and 1.6–3 ppt in Aschmann et al. (2011). Liang et al. (2010) which deploys explicit wet scavenging in convective updrafts found that convective scavenging only accounts for ~ 0.2 ppt difference in modeled $\text{Br}_y^{\text{VSLs}}$. Similar results were reported in Aschmann et al. (2011), who showed while SGI is highly correlated with convective activity, the impact on total stratospheric bromine in a full chemistry scheme is nearly insensitive to dehydration, likely due to convection dilution and increased scavenging.

In this study, we use a 3-dimensional Chemistry Climate Model with fully interactive CHBr_3 and CH_2Br_2 , the two most important brominated VSLs, to better understand how VSLs and their degradation products enter stratosphere. We will also test the uncertainty in modeled $\text{Br}_y^{\text{VSLs}}$ due to varying strength in convection and scavenging.

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2 Model

We conduct model simulations using the NASA GEOS Chemistry Climate Model (GEOSCCM) Version 2, which couples the GEOS-5 GCM (Reinecker et al., 2008) with a stratospheric chemistry module. Model simulations have a horizontal resolution of $2.5^\circ \times 2^\circ$ (longitude by latitude) with 72 vertical layers from surface to 0.01 hPa. The GEOSCCM model uses a flux-form semi-Lagrangian dynamical core (Lin, 2004) and the Relaxed Arakawa Schubert (RAS) parameterization for convection (Moorthi and Suarez, 1992). The GEOS-5 moist processes are represented using a convective parameterization and prognostic cloud scheme. All chemical kinetics and photolysis rates are calculated following JPL 2010 (Sander et al., 2011).

The GEOSCCM V2 stratospheric chemistry module includes all important gas phase stratospheric reactions as described in Douglass and Kawa (1999). We modify the chemistry scheme to include CHBr_3 and CH_2Br_2 . The two organic source gases are released at the ocean surface following the geographically resolved emission distribution described in Liang et al. (2010) and are destroyed in the atmosphere via photolysis and reaction with the hydroxyl radical (OH). OH above the tropopause is calculated online in the stratospheric chemistry module. OH in the troposphere is relaxed to a monthly mean climatological fields documented in Spivakovsky et al. (2000). Global annual mean OH from Spivakovsky et al. (2000) is $1.16 \times 10^6 \text{ molecules cm}^{-3}$, yielding an atmospheric methyl chloroform (CH_3CCl_3) lifetime of ~ 5.5 yr. Below tropopause, only simple bromine chemistry is considered which partition 80 % of inorganic bromine products into HBr and the remaining 20 % as HOBr, ratios adopted from Yang et al. (2005). Above the tropopause, bromine from degradation of CHBr_3 and CH_2Br_2 is released as Br and interacts fully with stratospheric chemistry. We assume high solubility for all three inorganic bromine reservoir species, HBr, HOBr, and BrONO_2 , and compute their wet scavenging. The wet removal processes are detailed in Liang et al. (2010), which include scavenging in rainout (in-cloud precipitation) and washout (below-cloud precipitation) in both large-scale precipitation (Giorgi and Chameides, 1986) and deep

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convective updrafts (Balkanski et al., 1993). We also consider re-evaporation back to the atmosphere. No heterogeneous chemistry for VSLS is included in our simulations, but results from Aschmann and Sinnhuber (2013) suggest that the impact of heterogeneous chemistry is minor and the inclusion of heterogeneous activation prevents loss
5 in scavenging and can increase $\text{Br}_y^{\text{VSLS}}$ by 10 %.

In this study, we present results from two 51 yr simulations between 1960–2010, one with brominated VSLS chemistry (R_{VSLS}) and one without (R_{BASE}), to examine troposphere to stratosphere transport (TST) of VSLS and their contribution to stratospheric bromine. It takes a considerable long time (~ 15 yr) for the full impact of VSLS to reach
10 the upper stratosphere, thus the first 16 yr are considered as spin-up. A full model evaluation and discussion of the impact of VSLS on stratospheric ozone are presented in a separate paper, currently in preparation. For this study, we focus on model results from the last 10 yr of the two simulations.

To quantify the uncertainty in $\text{Br}_y^{\text{VSLS}}$ due to changes in convection strength, we conduct two sensitivity simulations from 1980 to 2010 to represent minimum convection
15 (R_{MINCNV}) and maximum convection (R_{MAXCNV}) conditions by varying five convective parameters in RAS that impact the strength of deep convection, clouds, convective condensate and re-evaporation (Ott et al., 2009, 2011). These five convective parameters are identified as the most strongly influencing out of the total 16 parameters examined
20 using a large number of “Monte Carlo” type simulations and ensemble simulations in both a Single-Column Model as well as the GEOS-5 GCM (Ott et al., 2011). We aim to use these two extreme conditions to the range of uncertainty that can be introduced due to variations in the strength of convection and wet scavenging.

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3 Results

3.1 The contribution of VSLS to stratospheric bromine

The simulated CHBr_3 and CH_2Br_2 have been evaluated extensively in Liang et al. (2010) and compares well with aircraft and surface observations in their atmospheric distribution, geographically and vertically, as well as the associated seasonality. Here we present a summary comparison of observed and modeled CHBr_3 in the troposphere for 30–60° S, 30° S–30° N, 30–60° N, and 60–90° N latitude bands (Fig. 1). The observed profiles are compiled using Whole Air Sampler canister measurements (Schauffler et al., 1999; Blake et al., 2003) from eight NASA aircraft missions, as detailed in Liang et al. (2010), and averaged within each latitude band at 1 km vertical interval. The comparison clearly shows that the model captures well the source gas concentrations and vertical gradients in the tropics, mid and high latitudes in both hemispheres.

Figure 2 shows a comparison of our simulated BrO with balloon measurements from the LPMA (Limb Profile Monitor of the Atmosphere)/DOAS (Differential Optical Absorption Spectroscopy) at Teresina (5.1° S), Aire sur l'Adour (43.7° N), and Kiruna (67.9° N) collected between 2003–2005 (Dorf et al., 2006a, b, 2008; Rozanov et al., 2011). The lower levels of BrO from R_{BASE} in comparison to the measurements indicate the essential role of VSLS in completing the stratospheric bromine budget. With VSLS, the GEOSCCM model simulates well the observed BrO at all sampled locations in the tropics, mid and high latitudes. Since the model is running freely by specifying only surface source gas emissions, the fact that the model simulates well the observed concentrations of both the source gases and BrO suggests that the model presents a credible representation of stratospheric bromine chemistry.

Comparing results from the R_{VSLS} and R_{BASE} runs (Fig. 3), we find that the inclusion of CHBr_3 and CH_2Br_2 adds a uniform ~ 7.7 ppt Br_y throughout most of the stratosphere. The current $\text{Br}_y^{\text{VSLS}}$ estimate is 55 % higher than our previous estimate of

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~ 5 ppt (Liang et al., 2010) and ~ 6 ppt estimate in Hossaini et al. (2013), though the latter two are driven with the same VSLs emissions. Compared to the idealized case in Liang et al. (2010) that tracks $\text{Br}_y^{\text{VSLs}}$ in a single highly soluble tracer, $\text{Br}_y^{\text{VSLs}}$ is present in both soluble forms (HBr, HOBr, and BrONO_2) and insoluble forms (Br, BrO, BrCl) in the fully interactive chemistry scheme. The ratio of insoluble and soluble Br_y varies with time of the day as well as altitude and location. At daytime, when most convective lofting occurs, the majority of Br_y exists as insoluble Br and BrO, therefore greatly increases the amount of $\text{Br}_y^{\text{VSLs}}$ that survives wet scavenging during TST and ultimately reaches the stratosphere. The increase from ~ 5 ppt to ~ 7.7 ppt when switching from an idealized case to fully interactive chemistry is consistent with results from Aschmann and Sinnhuber (2013), who found that $\text{Br}_y^{\text{VSLs}}$ increased from 3.4 ppt to 5 ppt when switching from an idealized setup with a single soluble inorganic bromine tracer to a full chemistry scheme. The different estimates between Hossaini et al. (2013) and this work is likely due to how the ratio of soluble and insoluble inorganic bromine is determined inside the two models. Compared to this work which partitions inorganic bromine in its various forms based on chemical reaction rates, Hossaini et al. (2013) used a mean altitude-dependent HBr : Br_y ratio. This likely leads to an excessive washout of $\text{Br}_y^{\text{VSLs}}$ as during daytime when most of the convective lofting occurs, the majority of the inorganic bromine exists as insoluble Br and BrO. This suggests that a close approximate of soluble and insoluble inorganic product gases and the associated diurnal variation are critical to accurately quantify the contribution of VSLs to stratospheric halogen in VSLs modeling.

3.2 Troposphere-to-stratosphere transport of VSLs

We use the simulated monthly mean CHBr_3 distribution on the 355 K potential temperature surface (just below TTL) to show important tropical regions where active TST initiates (Fig. 4). Theoretically, trace gas distribution on the 365 K surface should be more indicative of TST as 365 K marks the zero radiative heating and air mass ele-

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vated above this level can enter the lower stratosphere through slow radiative ascend (e.g. Gettelman and Forster, 2002; Fueglistaler et al., 2009). However, it is difficult to identify active TST regions on the 365 K map (not shown) as a significant portion of CHBr_3 is converted to product gases. This is not surprising for a short-lived compound with lifetime ~ 26 days (WMO, 2011) while on average it takes about 10 days for air to transport by ± 10 K (Fueglistaler et al., 2004; Levine et al., 2007). Compared to CHBr_3 , it is much more difficult to identify active TST regions on a CH_2Br_2 map, as CH_2Br_2 distribution appears more zonal with a smaller meridional gradient due to a longer lifetime (~ 120 days, WMO, 2011) and thus more mixing with the surrounding background air (not shown). The 355 K CHBr_3 map shows three active regions that can efficiently deliver VLSL to the base of TTL: (1) the tropical Indian ocean, (2) the Western Pacific warm pool, and (3) off the Pacific coast of Mexico (Fig. 4). There is significant seasonality associated with each entry region. The Indian Ocean appears as the most active region for TST of CHBr_3 and occurs all year long with a maximum in boreal winter (DJF). Lofting in the Western Pacific reaches its maximum in boreal summer (JJA), while TST off the Mexico coast occurs mostly in boreal summer (JJA) and fall (SON). The importance of convective lofting in the Western Pacific warm pool in TST has been noted in many previous studies (e.g. Hatsushika and Yamazaki, 2003; Fueglistaler et al., 2004; Aschmann et al., 2009; Hossaini et al., 2012a; Aschfold et al., 2012) with several others suggesting that the Indian Ocean is also an important region in TST of VLSL (Levine et al., 2007, 2008; Brioude et al., 2010; Hoyle et al., 2011).

We show in Fig. 5 the vertical profiles of organic source gases ($\text{CH}_2\text{Br}_2 \times 2$, $\text{CHBr}_3 \times 3$) and inorganic product gases ($\text{Br}_y^{\text{VLSL}} = \text{Br} + \text{BrO} + \text{BrCl} + \text{HOBr} + \text{HBr} + \text{BrONO}_2$) in the three critical convective lofting regions to illustrate the transport and washout of the brominated VLSL during TST and the relative importance of SGI and PGI. Over the tropical Indian Ocean, on annual average, the mixing ratio of total bromine from VLSL (organic + inorganic) at 150 hPa (~ 355 K) is ~ 8.5 ppt, the same as its surface abundance (~ 8.5 ppt). This implies convective lofting in this region is so efficient that the amount of VSL bromine that enters the base of TTL is set by the concentration at the

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marine boundary layer. As air ascends to higher altitudes, a small portion of $\text{Br}_y^{\text{VLSL}}$ (~ 0.8 ppt, 10 %) is gradually removed from the atmosphere via wet scavenging until reaching a constant 7.7 ppt above 10 hPa. The somewhat wavy structure in the total bromine profile in the lower stratosphere implies that not all wet scavenging happen locally and the dip in total bromine concentration between 100–50 hPa is likely associated with recirculation of air back from the mid-latitude lower stratosphere where additional scavenging occurs via in-cloud rainout. Results from the Western Pacific and off the coast of Mexico are similar, with a smaller surface abundance of organic bromine (7.8 \sim 8.1 ppt) and slightly less washout around 100 hPa. The global averaged total bromine from VLSL (Br^{VLSL}) profile shows a maximum value of ~ 8 ppt at ~ 100 hPa. To supply this amount of Br^{VLSL} to the LS, transport from the MBL has to initiate from tropical regions where active convective lofting is co-located with high surface concentrations, where the collective CHBr_3 and CH_2Br_2 abundance exceeds 8 ppt Br.

Mapping streamlines on top of the VLSL organic bromine distribution in the tropics clearly illustrates the importance of co-location of deep convection with high surface concentration regions (Fig. 6). Among the three ascending branches of the Walker circulation (Webster, 1983), while ascending in the Western Pacific penetrates deepest into the TTL, ascending in the Indian Ocean is capable in delivering more VLSL bromine to the base of TTL due to higher surface concentrations. The surface abundance of VLSL bromine is largely dependent on the emission distribution used. Note Liang et al. (2010) assume uniform zonal emission strength across all longitudes with a few surface observation constraints from the Western Pacific and none from the tropical Indian Ocean. Hossaini et al. (2013) conducted a recent model study comparing four independent brominated VLSL emission estimates. Of all four emission inventories examined (Liang et al., 2010; Pyle et al., 2011; Ordóñez et al., 2012; Ziska et al., 2013), modeled CHBr_3 and CH_2Br_2 concentrations using the emission estimates from Liang et al. (2010) compare very well against tropical observations from multiple years of National Oceanic and Atmospheric Administration/Earth System Research Laboratory (NOAA/ESRL) surface flask measurements and the HIAPER Pole-to-Pole Observa-

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tions (HIPPO) measurements between 2009–2011 supported by the National Science Foundation (NSF). When compared against the aircraft measurements collected in the Western Pacific during the Stratospheric Ozone: Halogen Impacts in a Varying Atmosphere (SHIVA) campaign, Liang et al. (2010) yields the best CH_2Br_2 (mean bias of +0.2 ppt compared with observations), compared to the other three emissions, but the simulated CHBr_3 is $\sim +0.88$ ppt too high on average. Note the high bias in modeled CHBr_3 in the Western Pacific will likely lead to a high bias in the model $\text{Br}_y^{\text{VLSL}}$, although the lofting in Indian Ocean seems to be more critical in determining the total amount of VLSL bromine in the lower stratosphere. The model simulated high concentration of brominated VLSL over the Indian Ocean is yet to be evaluated when more surface observations become available and the importance of the Indian Ocean in delivering higher amounts of VLSL bromine into TTL needs to be assessed. The simulated high surface concentrations of CHBr_3 and CH_2Br_2 near tropical Central America were validated with the NASA TC⁴ and INTEX-B measurements (Liang et al., 2010). This region, compared to the above two, is much less efficient in delivering VLSL bromine into the TTL due to the relatively weaker ascending motion. However, this ascending branch can be important in particular seasons and in individual years as the Walker circulation moves in the east-west direction between different phases of El Niño Southern Oscillation (ENSO). For example, using trajectory calculations, Levine et al. (2008) found a clear shift in the TTL origin of air parcels from the Western Pacific and Indonesia to those from the Eastern Pacific and South America in El Niño years.

Of the total VLSL bromine that enter the base of TTL, about half (~ 4 ppt) is in the form of inorganic product gases (Fig. 5) despite our simple assumption that all VLSL degradation products are released in the troposphere as highly soluble HBr and HOBr. A commonly adopted approach in many previous modeling studies is to prescribe the VLSL concentration at the base of TTL with observed organic source gas mixing ratios in the upper troposphere and tracks the subsequent chemistry and transport (e.g. Sinnhuber and Folkins, 2006; Aschmann et al., 2011; Aschmann and Sinnhuber, 2013). Our result implies that $\text{Br}_y^{\text{VLSL}}$ estimate from such approach is not complete as it misses

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an important component associated with PGI, which is as large as SGI at the base of TTL in our model simulation. A second implication of this result is that if we were to use measurements from the TTL region to quantify the contribution of VSLS to stratospheric bromine, it is necessary to make measurements of both organic and inorganic forms to fully account for the impact of VSLS on the atmospheric bromine budget.

3.3 The impact of convection strength

We examine the difference between the two convection sensitivity simulations, R_{MINCNV} and R_{MAXRUN} , to illustrate the impact of deep convection on $\text{Br}_y^{\text{VSLS}}$ (Fig. 7). Difference between these two simulations ($R_{\text{MINCNV}} - R_{\text{MAXRUN}}$) suggests that in regions that mattered most (the three tropical convection centers), surprisingly, the minimum convection condition is more favorable for TST of VSLS. While weaker convection slightly decreases SGI (a few tenths ppt), PGI increases significantly (2–3 ppt) due to less scavenging under minimum convection condition. TST of the longer-lived CH_2Br_2 is less sensitive to convection strength compared to that of CHBr_3 because of its longer lifetime. The significant increase in CHBr_3 and CH_2Br_2 in the mid-latitude bands under minimum convection mainly reflects the compensating responses in large-scale descent in a general circulation model as a result of changing convection strength (Ott et al., 2011). The decrease of the descent does not impact TST of VSLS.

Globally, the differences in convection strength and wet scavenging introduce a ~ 2.6 ppt uncertainty in $\text{Br}_y^{\text{VSLS}}$ (~ 6.5 ppt in R_{MAXRUN} and ~ 9.1 ppt in R_{MINRUN}) (Fig. 3). Although these two simulations represent extreme parameterization conditions, BrO from both simulations still falls within the ± 2.5 ppt uncertainty range of the DOAS balloon observations (Fig. 2, Dorf et al., 2006b).

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4 Conclusions

We use the NASA Goddard 3-D Chemistry Climate Model, GEOSCCM, to quantify the contribution of brominated very short-lived substances (VSLS) to reactive stratospheric bromine and to test its sensitivity to the strength of deep convection and the associated scavenging.

The inclusion of CHBr_3 and CH_2Br_2 in a fully interactive stratospheric chemistry module adds ~ 7.7 pptv to present-day stratospheric inorganic bromine. The most active transport of VSLS from the marine boundary layer (MBL) through the tropical tropopause layer (TTL) occurs where high surface concentrations of VSLS co-locate with deep convection centers: (1) the tropical Indian Ocean, (2) the Western Pacific warm pool, and (3) off the Pacific coast of Mexico. On annual average, almost all VSLS-originated bromine available in the MBL (~ 7.8 – 8.4 ppt) at these active convective lofting regions enters the TTL, half in the form of source gas injection (SGI) and half in the form of product gas injection (PGI). After lofting above 150 hPa (~ 355 K), the majority of the VSLS-originated bromine survives TST and reaches the lower stratosphere, with only a small portion (~ 0.8 ppt, 10 %) removed by wet scavenging. Our model results point to a clear need for more surface measurements in the active VSLS lofting regions, in particular the tropical Indian Ocean and the Western Pacific, for improved emission estimate as well as a better quantification of how much VSLS bromine is available before ultimately enters the stratosphere through TTL.

Our current estimate of the contribution of CHBr_3 and CH_2Br_2 to stratospheric bromine ($\text{Br}_y^{\text{VSLS}}$) is higher than previous modeling estimates, mainly reflecting the differences in how transport and wet scavenging of product gases are treated in different models (Hossaini et al., 2012a; Aschmann and Sinnhuber, 2013). Interestingly, even though the models are driven with same emission estimates (Liang et al., 2010; Hossaini et al., 2013) and/or with similar source gas abundance at the base of TTL (Aschmann et al., 2011; Aschmann and Sinnhuber, 2013), this study still yields higher $\text{Br}_y^{\text{VSLS}}$ for two main reasons. Modeling VSLS and their degradation products in full

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chemistry scheme leads to a better representation of the partition between soluble and insoluble product gases and the associated diurnal variation, both are critical in an accurate Br_y^{VLS} estimate. Secondly, tracking PGI from the free troposphere to the base of TTL, which is as large as SGI at the base of TTL in our model simulation, is also important. This was not included in many earlier model studies (e.g. Aschmann et al., 2011; Aschmann and Sinnhuber, 2013), thus implying a missing component in these earlier model VLS contribution estimates.

Differences in model deep convection strength can introduce $\sim 30\%$ (6.6–9.2 ppt) uncertainty in simulated Br_y^{VLS} within the same convection scheme. It is important to point out that this uncertainty is likely larger among different models or a same model but different versions when different convective parameterizations are used. Contrary to conventional wisdom that the VLS impact on stratospheric bromine is larger under more intense deep convection, our simulations suggest that minimum convection condition is favorable for TST of VLS due to reduced scavenging in soluble product gases under weaker convection condition. The impact of convection strength on PGI greatly outweighs the impact on SGI with the change in PGI \sim ten times larger than that of SGI.

Acknowledgements. Funding for this research is from the NNX11AN71G project supported by the NASA ACPMAP program. Funding for the DOAS team are from the German Ministry of Economy (BMW) (50EE0840), the European Space Agency (ESA-ESRIN: no. RFQ/3-12092/07/1-OL) and the Deutsche Forschungsgemeinschaft, DFG (grants PF-384/5-1 and 384/5-1 and PF384/9-1/2), as well as Additional funding from the EU projects Reconcile (FP7-ENV-2008-1-226365) and SHIVA (FP7-ENV-2007-1-226224).

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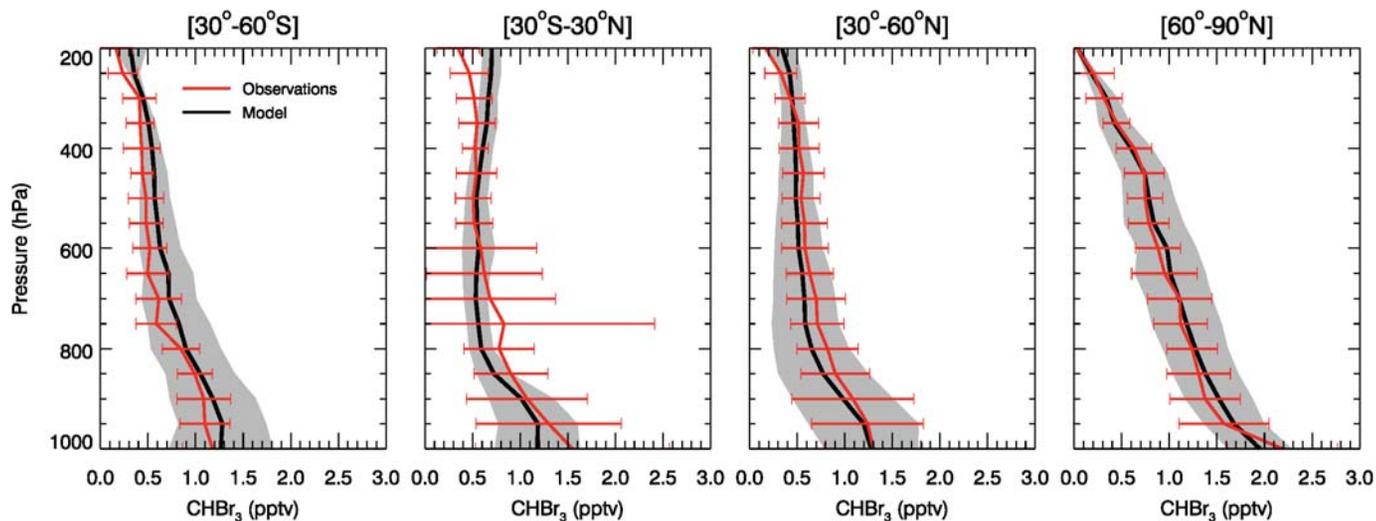


Fig. 1. Comparison between the observed (red lines with horizontal bars indicating one standard deviation) and simulated (black lines with gray shading showing one sigma variability) vertical profiles of CHBr_3 in the troposphere. Observations are compiled using Whole Air Sampler canister measurements from eight NASA aircraft missions are averaged for 30–60° S, 30° S–30° N, 30–60° N, and 60–90° N latitude bands at 1 km vertical interval. Model is sampled at the same location as the observations in the corresponding month in 2010.

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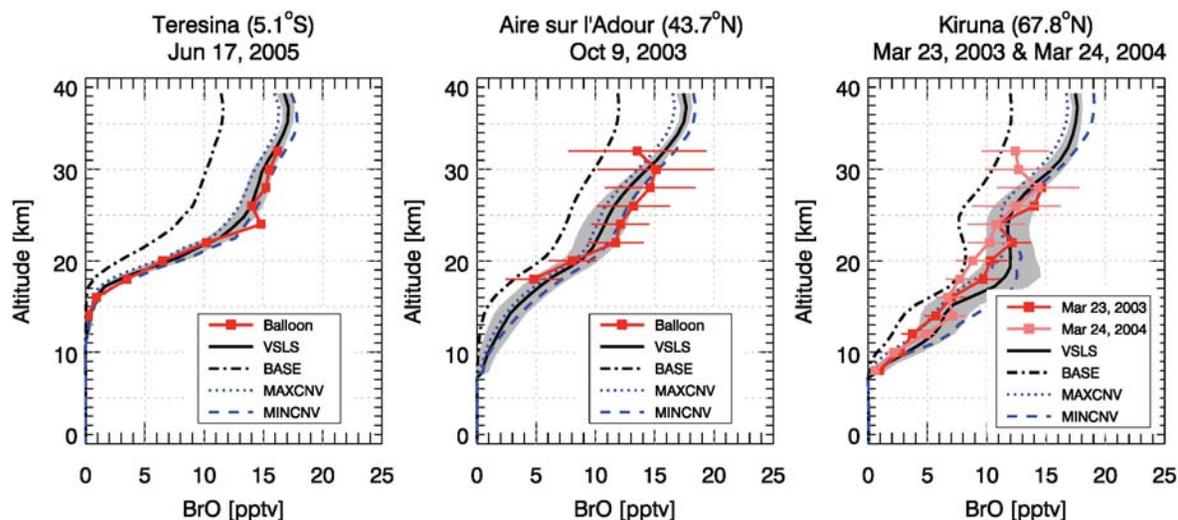


Fig. 2. Comparison of simulated BrO from the GEOSCCM run R_{VLS} (black solid line with 1-sigma variance in gray shading) with balloon measurements (red line with 1-sigma uncertainty in horizontal bars) from the LPMA/DOAS Spectrometers in the stratosphere. The black dash-dotted line shows simulated BrO from the run without VLS (R_{BASE}). Simulated BrO from the two convective sensitivity simulations are also shown (dotted line for maximum convection condition and dashed line for minimum convection condition). The model is sampled at the same month and latitude as the balloon measurements but only daytime profiles from all available longitude grid points are collected to calculate the 1σ variance of simulated BrO.

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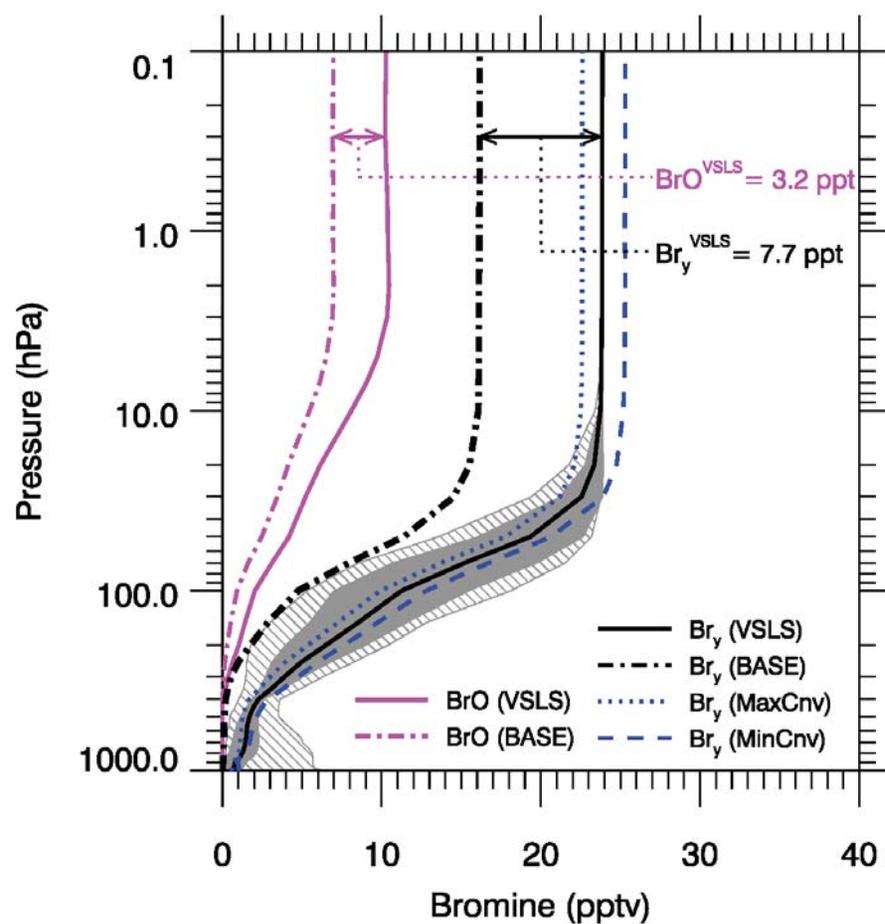


Fig. 3. Model simulated global and annual mean BrO and Br_y profiles from the simulation with VLSL (R_{VLSL} , solid lines) and the base simulation without VLSL (R_{BASE} , dash-dotted lines) for Year 2010. The gray shadings indicate the spread of annual mean Br_y in the R_{VLSL} run with the hatched areas indicate the minimum to maximum range and the filled shadings indicate 1σ variance. Br_y from the two convective sensitivity simulations are also shown (blue dashed and dotted line for minimum and maximum convection conditions, respectively).

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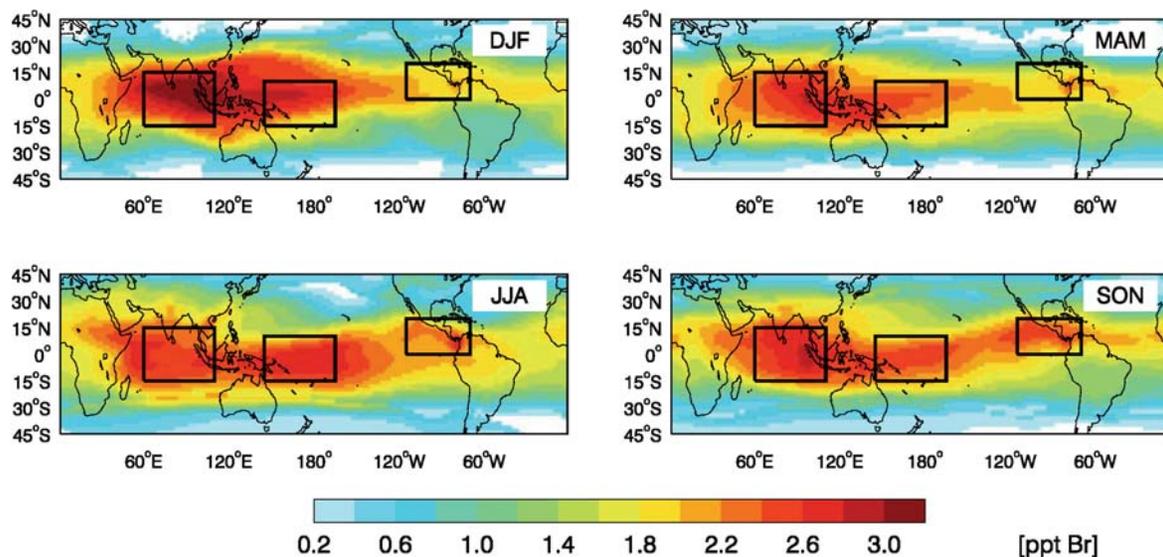


Fig. 4. 10 yr averaged (2001–2010) seasonal mean distribution of simulated CHBr_3 (unit ppt Br) at the 355 K potential temperature layer (just below tropopause) in DJF, MAM, JJA and SON. The black boxes outline the three regions where most active troposphere-to-stratosphere transport occurs.

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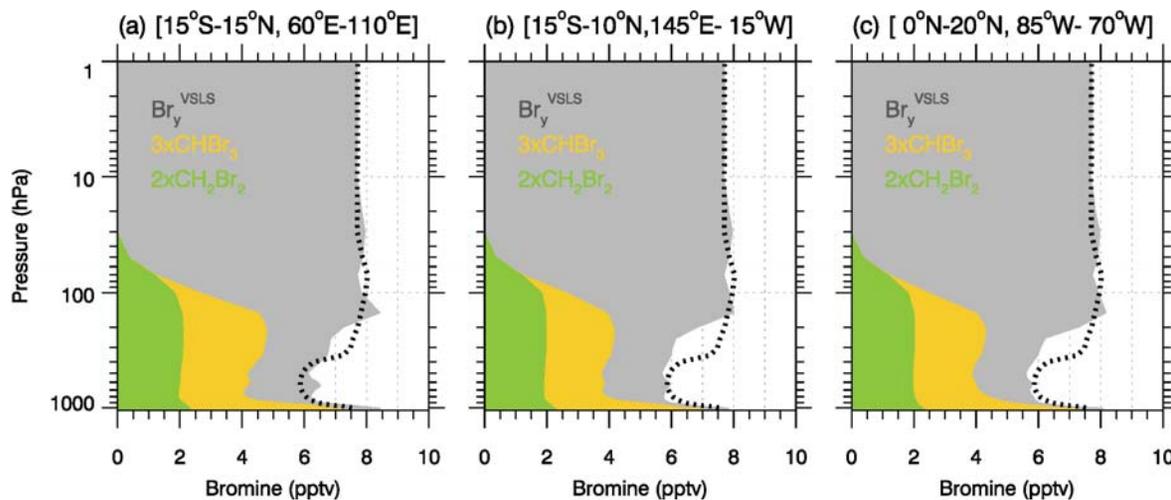


Fig. 5. The contribution of organic bromine ($\text{CH}_2\text{Br}_2 \times 2$, $\text{CHBr}_3 \times 3$) and inorganic bromine (Br_y^{VLS}) from CHBr_3 and CH_2Br_2 degradation to atmospheric bromine in three active TST regions **(a)** the tropical Indian Ocean (left), **(b)** the Western Pacific warm pool (middle), and **(c)** the Pacific Coast of Mexico (right). Global mean Br_y^{VLS} profile is also shown (thick black dotted line). The model results are 10 yr annual averages from 2001–2010.

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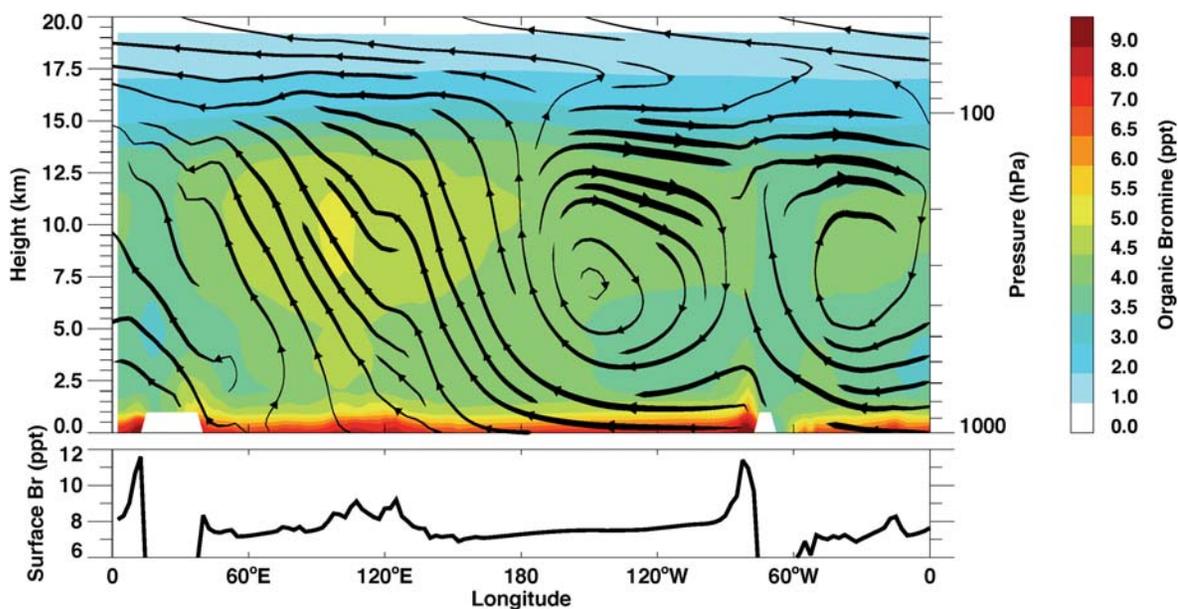


Fig. 6. Longitude-height cross-section of modeled total VSL organic bromine ($\text{CHBr}_3 \times 3 + \text{CH}_2\text{Br}_2 \times 2$) (color contours) and streamlines (black lines) in the deep tropics. The organic bromine and wind streams are 10 yr annual averages (2001–2010) between 10°S – 10°N . Surface organic bromine abundance is shown as the black solid line at the bottom panel.

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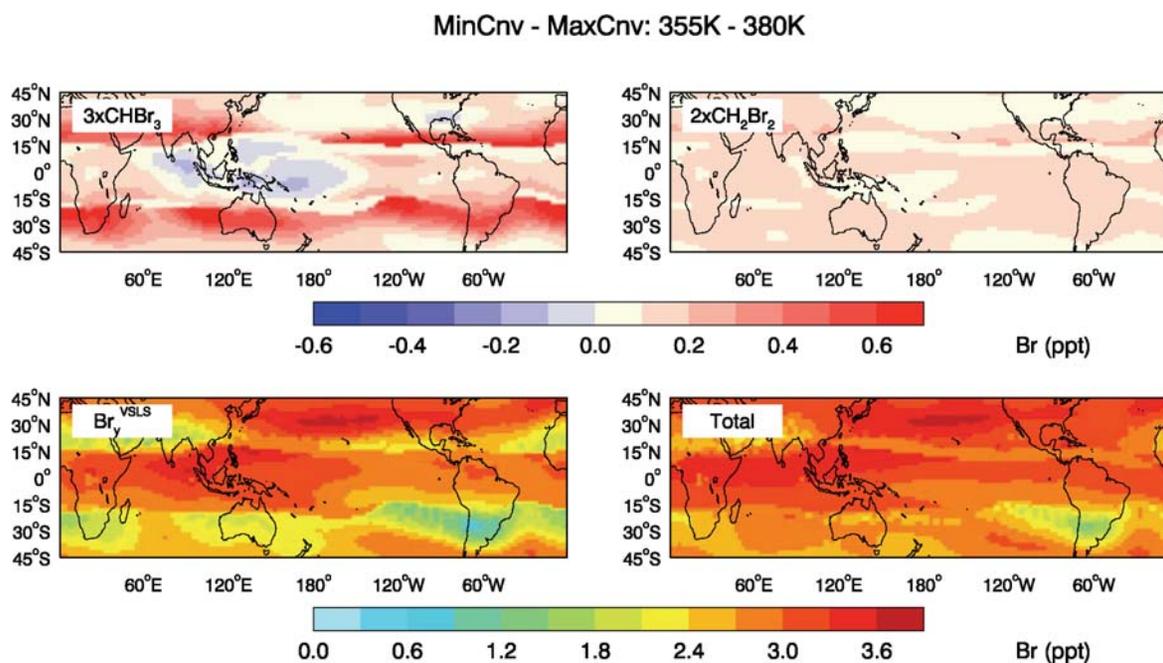


Fig. 7. The simulated annual mean difference between the R_{MINCNV} and R_{MAXRUN} runs in organic bromine ($3 \times \text{CHBr}_3$, $2 \times \text{CH}_2\text{Br}_2$), inorganic bromine and total bromine from VLSL between 355–380 K. The model results are 10 yr annual averages from 2001–2010.

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