We thank Dr. Sinnhuber for great and constructive comments. To address both reviewers' comments and concerns, we have revised our manuscript accordingly. In particular, we have revised the model description section significantly to elaborate in details the chemistry scheme, convection and wet scavenging scheme, the convection sensitivity studies. Below is our one-to-one correspondence to the comments (original comments in italic).

1. A main conclusion of this study is the finding, that product gas injection (PGI) could be much larger than deduced from previous studies. As is stated in the manuscript, an explicit treatment of bromine chemistry and the processes leading to its washout are critical to nailing down the estimate of stratospheric Bry from VSLS. However, on p.655, l.22 it is stated that bromine chemistry below the tropopause is not explicitly calculated here, but prescribed, with 80% of Bry being in the form of HBr and 20% as HOBr. What about the TTL? Is chemistry calculated in the TTL or only above the tropical cold point tropopause? If chemistry is indeed only calculated above the cold point, I'm having difficulties in understanding the differences in PGI between the present study and that of Liang et al. (2010).

The full stratospheric chemistry scheme, including interactive $CHBr_3$ and CH_2Br_2, are carried out above the 27th eta layer (approximately 350 hPa). Therefore, this setup should be adequate to address the chemistry, transport and scavenging of VSLS and its degradation products across the TTL into the stratosphere. We apologize about the unclear and misleading description in the previous manuscript. This is now clarified in the revised manuscript (section 2). We have also added in section 2 a brief description on the differences in model setup between Liang et al. (2010) and this study.

2. The only information on PGI from atmospheric measurements comes from BrO observations. Recently, Kreycy et al. (2013) suggested, using balloon borne DOAS BrO observations, that the ratio of J(BrONO2)/k(BrO+NO2) should be much larger (factor~1.7) than calculated from current recommendations, which would have a significant effect on the calculated BrO/Bry ratio. I assume this was not considered here? A comment how this would affect the comparison with BrO here would be useful. On a different, but related point: The present study considers only the two most abundant VSLS CH2Br2 and CHBr3. Observations indicate that the minor VSLS such as CH2BrCl, CHBr2Cl and CHBrCl2 together contribute another ~1ppt, which would need to be taken into account when comparing with BrO observations.

We are aware of the result from Kreycy et al. (2013), but philosophically speaking, we are somewhat conserved about altering model photolysis and thermal kinetic rates that are not yet included in the JPL recommendations. However, it is still possible to discuss the likely impact of this proposed change in the $J(BrONO_2)/k(BrO+NO_2)$ to Br_y^{VSLS} estimate without actually running an additional model simulation. Currently, $BrONO_2$ only accounts for ~0.5 ppt of the total Br_y^{VSLS} in the TTL during daytime. In addition, convective scavenging of the inorganic bromine in the TTL is not an efficient removal process (section 3.2). Increasing the ratio of $J(BrONO_2)/k(BrO+NO_2)$ in the model will shift Br_y^{VSLS} partition from $BrONO_2$ to BrO, which in turn will result an increase in

Br_y^{VSLS} due to less scavenging of $BrONO_2$. However, this increase will be small (at most a few tenths ppt). We have included this in the manuscript. The impact of the rate change on the mid-latitude BrO comparison can be a big tricky without actually conducting a model run. An important point on the Krecey et al. (2013) reported rate calculation is that it is based on balloon measurements from 68N, where the bromine photochemistry and partition ratios are very different from those in the TTL (where it matters for TST of VSLS). As shown in the figures below (Figure A for the tropics and Figure B for the high latitudes), the UT/LS high latitudes show much smaller day vs. night contrast in $BrONO_2$ levels compare to that in the tropics. This suggests that $BrONO_2$ in the high latitude UT/LS is much longer lived, therefore transport from other latitudes/altitudes plays an important role in determining the level of $BrONO_2$ (subsequently the $BrONO_2$/BrO ratio) in addition to just photochemical balance. Kreycy et al. (2013) reported a maximum likely change of 1.4 ppt in Br_y with their suggested new J($BrONO_2$) and k(BrO+NO_2) rates. This implies the impact of this rate change on the BrO/Br_y ratio is at maximum ~1.4/22 (assuming approximately 22 ppt total Br_y in 2005), ~6%. Using our current model estimate of 24 ppt Br_y and the BrO mixing ratio of 5 ppt at 10 km to 15 ppt at 30km, such a change will lead to an increase in modeled BrO from 0.3 ppt at 10 km to 0.9 ppt at 30km, which still yields a reasonable comparison with the DOAS measurements. On the comment of the contribution of CH_2BrCl, $CHBr_2Cl$ and $CHBrCl_2$, since all three are rather long-lived (lifetimes of 70-150 days), it is reasonable to adopt the SGI ratio for CH_2Br_2 and assume 90% of the 1 ppt tropospheric abundance makes to the stratosphere and adds ~0.9 ppt to stratospheric Br_y. Use the current GEOSCCM BrO/Br_y ratio, such an increase will result a stratospheric BrO increase up to 0.6 ppt (Figure C below). This increase yields a better agreement with the DOAS BrO measurements in the tropics and mid-latitudes and reasonable agreement at Kiruna with the standard model run, considering the large spatial variability in BrO in the high latitudes and rather localized balloon measurements. An additional increase of BrO by 6% on top of the above 0.9 ppt Br_y increase to account for the changes due to the newly proposed J($BrONO_2$) and k(BrO+NO_2) rates shows similar results. We have included in the revised manuscript the discussion on the inclusion of CH_2BrCl, $CHBr_2Cl$ and $CHBrCl_2$ for BrO comparison. However, we decide to leave out the impact of the new rates on the BrO comparison, as a more accurate quantification of this impact should be addressed in a future study, rather than a simple and crude approximation done here.

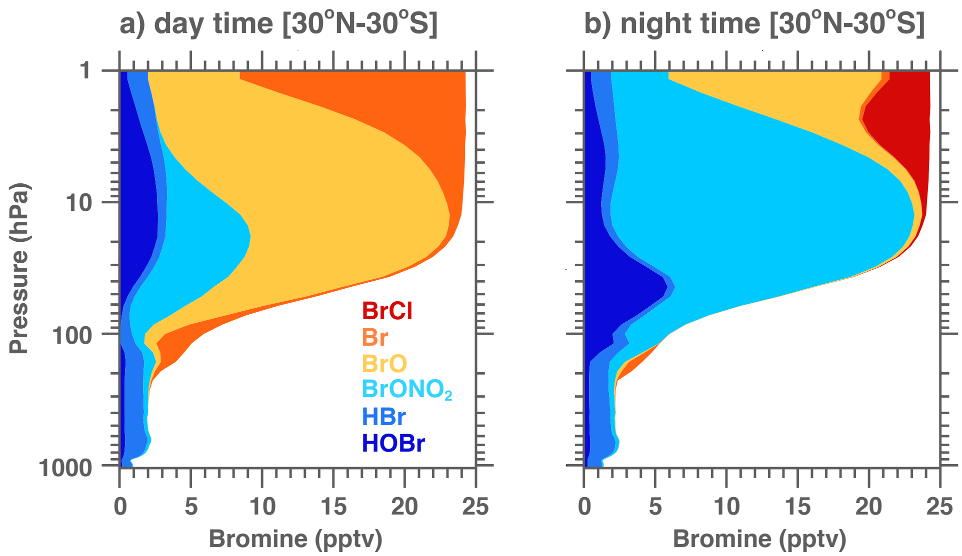

Figure A (Figure 4 in revised manuscript). Annual mean model inorganic bromine (BrCl, Br, BrO, $BrONO_2$, HBr, HOBr) tropical vertical profiles for daytime (left panel) and nighttime (right panel), averaged between 30ºN-30ºS.

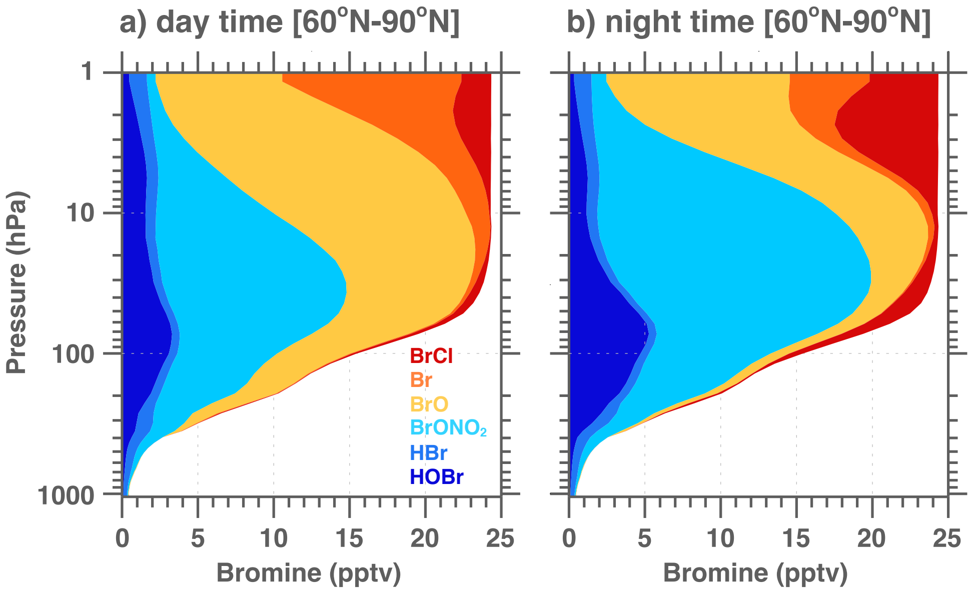

Figure B. Same as Figure A, but for the high-latitudes (averaged between 60ºN-90ºN).

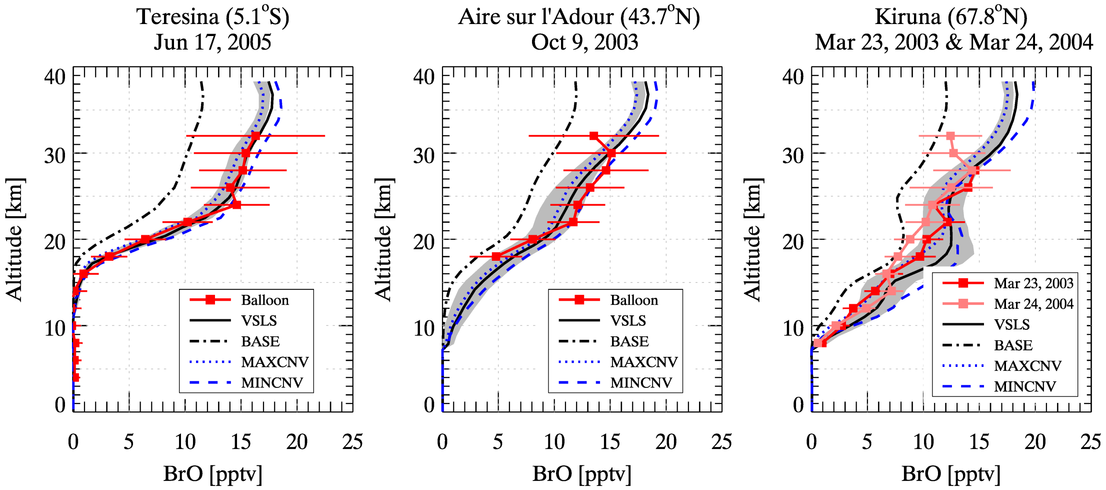

Figure C. Same as Figure 2 in the manuscript but with BrO increased proportionally assuming a total of 25 ppt Br_y instead of the model calculated 24 ppt, to account for the contribution of CH_2BrCl, $CHBr_2Cl$ and $CHBrCl_2$.

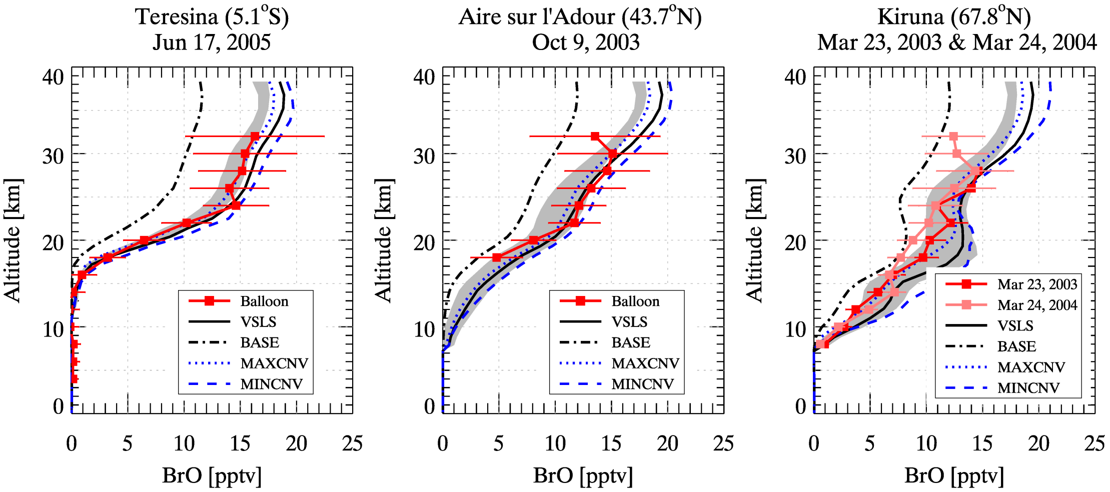

Figure D. Same as Figure D above but with BrO increased by 6% to approximate the change due to newly proposed J(BrONO2) and k(BrO+NO2) rates.

Specific comments:

p.655, l.9: Is the RAS convection parameterization different from the convective parametrization in the underlying GEOS-5 model? Would be good to provide a bit more detail.

We apologize for the somewhat misleading wording in the previous manuscript. The large-scale dynamic transport (semi-Lagrangian) and convective transport (RAS) are done inside the GCM side of the GEOSCCM model – the GEOS-5 GCM. The coupled chemistry model communicates the chemical fields with the underlying GCM at every time step to carry out transport, but it does not have a separate convection scheme. We have corrected this in the revised manuscript.

p.658, l.5: Again, is bromine chemistry modelled below the tropical tropopause (cold point) or specified?

See response to Major comment #1. For clarity, we have changed this to "fully interactive chemistry scheme in the TTL and the stratosphere".

p.660, l.7: Is there active washout above the tropopause in the model? I'm surprised that there is "in-cloud rainout" in the mid-latitude lower stratosphere - or did I misunderstand this statement? Please provide more details on the process.

There is active washout above the tropopause and the Giorgi and Chameides (1986) parameterization includes a rather simple yet adequate parameterization of in-cloud rainout and below-cloud washout. We have included in the revised manuscript a detailed description of the wet scavenging scheme as suggested by the reviewer.

p.653, l.6: should we really call this a "good agreement" given the large range of values (1-8ppt)?

We changed this to "within the 1-8 ppt estimate range".

p.653, l.24, somewhat misleading, as Brinckmann et al. is not a modelling study

We apologize for this mistake. Yes, Brinckman et al. does not belong to this list of references and therefore is deleted.

p.655, l.22: As noted above, this simple treatment is certainly not ideal when you want to investigate the fate of product gases. At least a caveat should be included and a more detailed specification how the tropopause is defined in the tropics. If chemistry is interactively calculated throughout the TTL this would be less critical than if chemistry is calculated only above the cold point.

See response to Major comment #1.

p.656, l.4: Aschmann et al. (2011) showed that heterogenous reactions in the TTL have a large impact on bromine partitioning.

While heterogeneous chemistry can shift Br_y partition and increase HBr up to 4 times between 12-18 km in the tropics (Aschamnn et al, 2011), the absence of heterogenous chemistry in GEOSCCM is likely to have only a small impact on Br_y^{VSLS} since the model HBr is present at very low abundance during day time when the majority of convective scavenging take place (see section 3.1 and Figure 4) and the overall convective scavenging in the TTL is rather inefficient (see section 3.2). We have added this in the revised manuscript.

p.658, l.7: Where (altitude, latitude) is the majority of Bry in the form of Br and BrO?

In the tropical near-tropopause region and lower stratosphere, the majority of the Br_y exist as Br and BrO during daytime. To illustrate the contrast in bromine speciation between day and night as well as to address some other comments, we have added a new figure (Figure 4) showing the tropical vertical profile of Br, BrO, BrCl, $BrONO_2$, HBr and HOBr for daytime and nighttime.

p.659, l.9: In the TTL the local lifetime of CH2Br2 is even much longer than 120 day (Hossaini et al., 2012)

We have changed this to "a longer lifetime, ~ 120 days (WMO 2011) and up to ~ 450 days locally in the TTL (Hossaini et al., 2010)". We used Hossaini et al. (2010) instead of the suggested Hossaini et al. (2012) as the TTL local lifetime for CH2Br2 is listed in Hossaini et al. (2010) instead of (2012).

p.659, l.12: Note the different definitions of "Western Pacific" in this study and other studies, which may explain some of the differences in seasonality between the studies.

We thank the reviewer for pointing this out. We have changed "Western Pacific" to "tropical western Pacific" throughout the text to more accurately reflect the region of interest.

p.660, l.19: Why are surface concentrations higher in the Indian Ocean in this model simulation? The following paragraph remains somewhat unspecific.

The rate of bromocarbon emissions for the coastal regions (per unit area) are much higher than the open oceans. In the tropics, the prevailing surface easterly trade winds tend to bring recent emissions of $CHBr_3$ from the adjacent Indonesian coastal regions while the tropical western Pacific Ocean sees recent emissions of $CHBr_3$ from the open ocean. Therefore, the surface concentrations are higher in the Indian Ocean than the tropical western Pacific. We have also added a more specific and quantitative discussion on model vs. observation comparison in the following paragraph (see below).

p.661, l.6: 0.88 ppt of CHBr3 (which would be a lot) or 0.88pt of bromine due to CHBr3?

The model vs. observation difference for the SHIVA campaign is ~ +0.76 ppt of $CHBr_3$ (value from the final published Hossaini et al., 2013). I agree with the reviewer that this difference does seem to be a lot. However, it is hard to conclude whether this large difference during SHIVA indicates a systematic alarmingly high model bias. The comparison of the model $CHBr_3$ with the long-term NOAA GMD measurements only shows $\Delta_{\text{model-obs}}$ = +0.12 ppt in the tropics and the comparison with the HIPPO measurements between 2009-2011 yields $\Delta_{\text{model-obs}}$ = +0.30 ppt in the Pacific tropics. It will be interesting to see how the model results compare with the newly available measurements in the tropical western Pacific obtained during the Jan-Feb 2014 NSF CONTRAST/British CAST field missions. We have included a more detailed and quantitative bias discussion in the revised manuscript.

p.662, l.5: I fully agree with these statements. However, two aspects remain puzzling: How can Bry be transported into the upper troposphere when assumed to be present in the form of the two highly soluble species HBr and HOBr? And secondly, observations suggest that there could be a significant amount of BrO in the troposphere, which would offer a pathway for TST PGI, but is not accounted for in this study.

To answer the reviewer's question and to better explain this in the text, we have modified Figure 5 (now Figure 6) to show all inorganic bromine from $CHBr_3$ and CH_2Br_2 degradation, instead of the sum of Br_y. Although tropospheric Br_y are assumed highly soluble, only a fraction of the grid boxes and a fractional area of the precipitating grid boxes actually experience precipitation/scavenging. On average, of all inorganic bromine (~4 ppt) produced from $CHBr_3$ and CH_2Br_2 in the tropical troposphere, about 50% (~2 ppt) is removed below 500hPa, mainly by large-scale precipitation (Liang et al., 2010), with an additional few 10ths ppt scavenged in the upper troposphere. Together, about 1.5 ppt of HBr and HOBr survive large-scale and convective scavenging and remain in the tropical upper troposphere. The current chemistry scheme does allow Br, BrO, and $BrONO_2$ production above ~350hPa. A previous detailed tropospheric chemistry model using the Harvard GEOS-Chem model and VSLS emissions from Liang et al (2010) shows about 0.1-0.2 ppt BrO below 10km in the tropics (Parrella et al., 2012). However, the impact of the absence of this 0.1-0.2 ppt tropospheric BrO on stratospheric bromine is small.

p.662, l.18: Just as a side comment, could the relation between convective strength in the tropics and large-scale descent in mid-latitudes be used to better constrain convective uplift? I guess it should be possible to validate large-scale descent from temperature and trace-gas observations better than the convective processes.

This is an interesting thought. I am not a dynamist, and I don't have enough background knowledge to comment on the feasibility of this approach. From my limited understanding, I can think of a few processes that regulates the large-scale descent in the mid-latitudes, e.g. synoptic-scale wave driven exchange between the lower stratosphere and upper troposphere, diabatic descent associated with radiative cooling. In a GCM, it might be very difficult to separate the compensating large-scale descent that needed to balance the tropical convective lofting from the above processes. Therefore, using the large-scale descent in mid-latitudes to constrain tropical convection might be in practice a hard approach.

p.662, l.18: You may cite Aschmann et al. (2011) here, who found a qualitatively *similar behavior for changes of SGI and PGI under El Nino versus La Nina conditions: While SGI slightly increased under enhanced deep convection, the sum of SGI and PGI decreased.*

Added as suggested.

p.664, l.3: Again, it is critical that you specify clearly if full chemistry was used here also in the TTL, or only above the cold point.

See response to Major comment #1. For clarity, we have changed this to “fully interactive chemistry scheme in the TTL and the stratosphere”.

Technical corrections:

p.652, l.14: of the VSLS

Corrected.

p.653, l.5: VSLS contribute

Corrected.

p.654, l.24: enter the stratosphere

Corrected.

p.660, l.11: please define LS and MBL when first used.

Since LS and MBL only used in a few number of places in the manuscript, we have spelled out LS as lower stratosphere and MBL as marine boundary layer throughout the manuscript.

p.661, l.22: that enter

Corrected as “that enters”

p.664, l.14: “scavenging in” or “scavenging of”?

Corrected.