Response to Referee #1 and #2

We thank both reviewers for their reviews and minor suggestions to improve the manuscript. We agree with all the suggestions and will change the text accordingly, as summarized below.

General Comment:

The authors use the WACCM CCM and artificially increase stratospheric inorganic bromine (Bry) with an assumed contribution from VSLS (Bry^{VSLS}) of 0 ppt, 6 ppt and up to 10 ppt, in a sulphur injected geo-engineered atmosphere. Similarly, assumptions are made regarding the contribution to stratospheric inorganic chlorine from VSLS (Cly^{VSLS}) . In my view, a stratospheric Bry^{VSLS} contribution of 10 ppt is somewhat large and I wonder what WACCM would show if VSLS were treated explicitly (i.e. VSLS emissions, tropospheric degradation etc.).

We are currently working on implementing tropospheric emissions of organic halogens along with emissions from sea salt aerosol, and polar emissions on snow and sea ice. However at this time we have not made enough progress to assess the stratospheric Br_y and Cl_y increase from inclusion of these processes. Therefore, in this study, we consider values for the contribution of VSLS to the stratospheric Br_y and Cl_y burden that are based on estimates derived from observations, as discussed in the text. The Br_y^{VSLS} contribution of 10 pptv is an upper limit based on a number of studies, as summarized for instance in Table 1-14 of the WMO/UNEP Scientific Assessment of Ozone Depletion: 2010 report.

Specific Comments:

*The introduction is lacking adequate background on VSLS. Given they are clearly central to the focus of the paper, I would expect more than just the citation to WMO (2003) on page 21926 (line 14). In fact, a citation to the most recent WMO assessment is probably more appropriate in order to be up-to-date on VSLS literature. At the very least it should be mentioned that bromine-containing VSLS, such as bromoform (CHBr**3**) and dibromomethane (CH**2**Br**2**), are predominately of natural biogenic/oceanic origin (e.g. Quack and Wallace, 2003). This is mentioned in the supplementary material but should also be included in the main text. What are the chlorine-containing VSLS? Do they have a biogenic source also? A few more sentences should be included.*

*Page 21926 (line 15): Feng et al. (2007), using a 3-D model, also showed the impact of VSLS on stratospheric O**3** to be larger during periods of high aerosol loading. A reference to this study should also be included.*

We agree with the reviewer that more details should be given to VSLS in the introduction. Therefore we will change the following paragraph in the manuscript:

'The additional supply from very short-lived (VSL) bromocarbons – with largest contributions from dibromomethane (CH2Br2) and bromoform (CHBr3) (Chapter 2 of WMO, 2003) –has been suggested to have a significant impact in a volcanic-like stratospheric aerosol layer (Salawitch et al., 2005).'

To:

'The additional supply from very short-lived (VSL) bromocarbons has been suggested to have a significant impact in a volcanic-like stratospheric aerosol layer (Salawitch et al., 2005, Feng et al., 2007). The largest contribution to Br_y from VSL bromocarbons is likely due to dibromomethane (CH_2Br_2) and bromoform ($CHBr_3$). These species are produced by biogenic processes in the ocean (e.g., Quack and Wallace, 2003). The contribution of VSLS to the total stratospheric inorganic bromine (Br_y^{VLS}) is estimated to range between 1 and 8 ppt (WMO 2010, Table 1-14) with an upper limit of 10 ppt. The need for a significant contribution to stratospheric bromine from VSL bromocarbons is also supported by the recent study of Choi et al. (2012).

Chlorine-containing VSL source gases have both anthropogenic and natural sources (WMO 2010, Chapter 1, Section 1.3.1.1). Anthropogenic sources likely have largest contributions from dichloromethane (CH_2Cl_2) and tetrachloroethene (CCl_2CCl_2) from a variety of applications. Anthropogenic sources constitute about two thirds of the total stratospheric inorganic chlorine source from VSL species (Cl_y^{VSL}) for present day conditions. The natural source is dominated by bromochloromethane compounds produced by oceanic, biogenic processes. The value for Cl_y^{VSL} is estimated to range between 25 and 170 ppt (WMO 2010, Table 1-9).

Page 21927 (line 1): The authors correctly note that halogen loading from VSLS will gain (relative) importance as the contribution from long-lived gases declines in coming years (under the terms of the Montreal Protocol). The authors should also note here that the contribution from VSLS to stratospheric halogen loading may increase in absolute terms. The recent CCM study of Hossaini et al. (2012) shows enhanced CHBr3 in the lower stratosphere, relative to present day, under future climate projections. A reference to this new work is relevant here and should be included.

We thank the reviewer for the reference and have changed the following paragraph:

'Furthermore, the role of VSL halogen species is expected to gain importance in the coming years, as stratospheric Bry and Cly from long-lived sources decline due to the Montreal Protocol.'

To:

'Furthermore, the role of VSL halogen species is expected to gain importance in the coming years, as stratospheric Bry and Cly from long-lived sources decline due to

the Montreal Protocol. The transport of halogen containing VSL gases to the stratosphere might also rise in the future, due to increases in tropical deep convection and alteration of the oxidation capacity of the troposphere driven by climate change (Hossaini et al., 2012)'

Page 21927 (experimental design): WACCM is well known but a few more simple details of the runs would be appropriate here (e.g. resolution, vertical domain and levels).

'The Whole Atmosphere Community Climate Model (WACCM) Version 3548 (Garcia et al., 2007) is used to investigate the impact of geo-engineering on ozone.'

We changed this paragraph to:
'WACCM is a global model with 66 vertical levels from the ground to $4.5x10^{-6}$ hPa (approximately 140 km geometric height). The vertical resolution is variable: 3.5 km above 65 km, 1.75 km around the stratopause (50 km), 1.1-1.4 km in the lower stratosphere (below 30 km), and 1.1 km in the troposphere (except near the ground where much higher vertical resolution is used in the planetary boundary layer). The horizontal resolution is 1.9° × 2.5° (latitude × longitude). The chemical module is based upon the 3–D chemical transport Model of OZone and Related Tracers (MOZART), Version 3 [*Kinnison et al*., 2007]. WACCM includes a detailed representation of the chemical and physical processes in the troposphere through the lower thermosphere [*Garcia et al.,* 2007].'

Technical Comments

Page 21927 (line 16): Has UTLS been defined?

We now define Upper Troposphere Lower Stratosphere (UTLS) in the manuscript

Page 21927 (line 25): Sulfur dioxide (SO2).

We have added sulfur dioxide, as suggested

Page 21927 (line 26): tropics, state latitude range.

We changed:

'The sulfate distribution was taken from Rasch et al. (2008b), who considered an injection of 2 $TgSyr^{-1}$ of volcanic-sized aerosols into the tropics.'

To:

'The sulfate distribution was taken from Rasch et al. (2008b), who considered an injection of 2 $TgSyr^{-1}$ of volcanic-sized aerosols into a 2km thick layer at 25km

altitude in the tropics, between 10N and 10S.'

Page 21944 (Figure 3 caption): First sentence is too long. Please split or include punctuation.

The sentence has been changed to:

'Relative changes of UV radiation weighted for human erythema (skin-reddening) due to geo-engineering, for projected chemical conditions in year 2040. Changes are the result of differences in column ozone between the geo-engineering and the baseline simulation.'