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## ***Interactive comment on “Stratospheric SO<sub>2</sub> and sulphate aerosol, model simulations and satellite observations” by C. Brühl et al.***

### **Anonymous Referee #2**

Received and published: 21 June 2013

Review of Bruhl et al. (2013)

The stratospheric aerosol layer exerts one of the most important natural radiative forcing on the climate system. Improving our understanding of the sources and physical processes related to its formation and evolution is fundamental to assess how changes in stratospheric aerosol may affect climate and how climate change may in turn affect the stratospheric aerosol burden. The manuscript by Bruhl et al. (2013) combines model simulations and satellite observations of stratospheric aerosol and SO<sub>2</sub> to evaluate the principal sources (volcanoes, COS and H<sub>2</sub>SO<sub>4</sub>) and mechanisms which influence the stratospheric aerosol layer. This paper also explores the effects of stratospheric aerosol changes on radiative heating, stratospheric temperature and water vapor content of the stratosphere. Overall, the paper lacks of clear objectives

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and tend to address too many points, which are only superficially explored. I would think that the paper should have been divided into two parts : - One part dealing with the Mt Pinatubo eruption effects on stratospheric aerosol burden radiative heating, dynamics, chemistry and climate. - A second on the mid-size volcanic eruptions of the last decade, including SO<sub>2</sub> and aerosol observations by satellites.

Before publication, the authors need to fully address a number of science questions, which are not insufficiently explored.

Major Comments:

1) COS appears to have a very small contribution to the total SO<sub>2</sub> burden of the middle stratospheric and is likely a minor contributor to the total stratospheric aerosol burden. The tropical reservoir is mainly supply by the upward branch of the Brewer-Dobson circulation by which even small tropical volcanic plumes reaching 19-20 km can be transported. Overall, the MIPAS SO<sub>2</sub> observations support the idea that COS has only a very small influence on the stratospheric SO<sub>2</sub> budget. It is not clear to me if the maintenance of the Junge layer near 25-30 km (see fig.1 from Vernier et al., 2011,GRL) is the result of the fact that aerosols have a longer lifetime in this region or because of the source from COS. I think the importance of COS is overestimated and the abstract and conclusion should be modulated.

2) Anthropogenic emissions of SO<sub>2</sub> : this paper does not provide a set of simulations allowing to verify the importance of anthropogenic emissions of SO<sub>2</sub> on the stratospheric aerosol budget during the past decade. While the conclusion states that this study confirm previous findings related to the minor influence of anthropogenic emission of SO<sub>2</sub> from Asia, I think this is not completely explored in this paper. For example, Fig.12 of Hopfner et al. (2013) using also MIPAS SO observations seems to indicate an increase of the SO<sub>2</sub> total burden in the 15-23 km layer over the past decade. To be discussed deeply in the paper.

3) While model simulations performed for this paper suggest an increase of strato-

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spheric H<sub>2</sub>O followed by Mt Pinatubo eruption, due to the less effective warmer cold trap tropopause, allowing more H<sub>2</sub>O to enter the stratosphere, there are no observations of TTL temperature as well as TTL water vapor, to my knowledge, which support these findings. This part of the paper needs to be fully explored.

4) The authors suggest that organic aerosols are important sources of particles in the tropical upper troposphere. However, the SAGE II observations, which are used to support these findings, can be potentially contaminated by ice clouds. The authors should include the version number of the SAGE II dataset used in this paper well as discuss the possible contamination by cirrus clouds, which would affect the retrieval of aerosol extinction at 1020nm.

Additional comments:

Abstract :

11396-2 : “at high altitude resolution”. A more appropriate term would be “high vertical resolution”. This sentence should be reformulated. It sounds like if the aerosol module GMXe is vertically resolved, but it is actually the meteorological and aerosol fields which have X number of vertical levels. 11396-6. It is not clear here what “ aerosol modes: means. Could you clarify ? 11396-9. When you speak about SAGE, do you mean SAGE II or SAGE III ? 11396-12/13. This sentence should be reformulated. I would suggest “ The model...the radiative impacts of stratospheric and tropospheric aerosol and its associated dynamical response”.

Introduction :

The introduction is very brief and does not reflect/summarize enough past research activities on stratospheric aerosol and SO<sub>2</sub>. In addition, the plan announcement of the paper is vague and lack of clear objectives.

2. Model Set up :

11397-20/22 : The description of the model is not completely clear. What T42 means ?

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how many bins of the aerosol size distribution ? what kind of aerosols ? Please provide a more complete description of the model. 11397-22 : “ The vertical grid structure. . . ” This could be reformulated by “The 90 vertical levels of the model allow to. . . ” 11397-26. The initialization of the model with volcanic SO<sub>2</sub> is relatively obscured. Do you use satellite data of SO<sub>2</sub> or aerosol to initialize the model ? 11398-12 : The choice of the boundary between coarse and accumulation mode sounds rather arbitrary. Could you provide more explanation?

### 3. Stratospheric aerosol :

11398-25. The description of the SAGE data is incomplete ? Correct SAGE by SAGE II. What SAGE II version do you use, V6.2 ? Have you been careful of using the feature flag to remove clouds in the upper troposphere ? Thomason and Vernier. (2013) argue that some clouds in the Upper Troposphere could have been missed by the previous cloud-clearing method know as the “Kent method”. I guess that you must be careful in interpreting the SAGE II data in the tropical upper troposphere. The reasons for the two simulations are not clear. To fill the gaps during the Mt Pinatubo period, the SAGE team has used lidar and aircraft observations during the year following the eruption. You could use this product (published in SPARC, 2006, ASAP) to initialize your model. Where does the 14 and 20.6 Mt values come from ? This part needs to be clarified. I also suggest the following paper : Aquila et al., 2011, JGR, <http://www.agu.org/pubs/crossref/pip/2011JD016968.shtml>

11398-24 : please correct “was injected at “ by “was injected on” 11399-25-26. One/two sentences about SAGE II would be useful. How does it work ? what does it measure ? I would suggest to be more clear on the reasons why the SAGE II extinction profiles are incomplete after Mt Pinatubo eruption. Something like : “Due to opacity of the Mt Pinatubo volcanic plume, no extinction could be retrieved below a certain altitude (usually near 22-23 km) “. 11399-4 : “some extrapolation into the region without data “, what do you mean by some ? What don’t you use the filling procedure proposed by the SAGE team ? 11399-11 : ATMOS, MLS are acronyms and need to be spelled. Which

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version of the data ? a few sentences about these measurements technique would be useful. 11399-16. You should describe fig.1 and fig.2 before discussing/interpreting your results. In general figures are poorly described in the manuscript. 11400-3/5 : You quote SPARC, 2000 to somehow prove that the positive anomaly of water vapor raising into the stratosphere is consistent with observations. Please be more accurate. What kind of measurements do you refer to ? Satellites, aircrafts? I'm not aware about reliable satellite measurements of water vapor during the years following Mt Pinatubo eruption which would support your findings. 11400-14/15 : "Note filled with extrapolations", You could probably use the recent SAGE II data which are actually filled up during this period. 11400-14 : Change "SAGE" by "SAGE II" 11400-26 : What do you mean by outdated ? which data are you speaking about ? Please be clearer. 11400-29/30 : " This decrease..." Please clarify this sentence. The volcanic aerosol forcings published Solomon et al. (2011) are not the results of complex radiative transfer calculations, but use the simple conversion factor to derive the forcing from the stratospheric aerosol optical depth (-23W/m<sup>2</sup>/unit of AOD).

### 3.2 Background and Medium volcanoes

11401-6/7 : I do not think that SO<sub>2</sub> injected by Nyiragongo reached the lower stratosphere. Vernier et al. (2009,2011) do not show evidence of this plume in the CALIPSO dataset at this level. The eruption was likely in the upper troposphere only. 11401-9/10 : "Inferred from TOMS derived masses.." I do not understand this sentence. TOMS does not provide information on the vertical distribution of SO<sub>2</sub>. Do you mean that you have used satellite data (SAGE II, CALIPSO) to redistribute the SO<sub>2</sub> mass from TOMS on the vertical levels ? Please clarify. 11401-9/10 : "These assumptions agree with MIPAS observations". Actually, the paper lacks of coherence at this point. I would think that MIPAS SO<sub>2</sub> of these volcanic plumes should have been used to initialize this simulation and that the resulting stratospheric aerosol have been validated with SAGE II and eventually CALIPSO after 2005. Please justify your choice [Major comment]. 11401-25/28 : Fig.8 : I do not really see a reason to show the sulfate mixing ratio

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derived from SAGE II instead of Extinction at 1 $\mu$ m.

11402-5/10 : Fig.12 : while trying to isolate the temperature response of the TTL from the moderate volcanic eruptions of the past decade, you must indicate if the results are significant (and at which level ?). Indeed, the signature of the volcanic plumes on the temperature seems to be embedded in the noise. You may need to use an ensemble of simulations to extract such information. Fig.12 does not provide strong evidences that these volcanic plumes had a significant impact on the temperature of the TTL.

#### 4. Stratospheric SO<sub>2</sub>

11402-15/20 Fig.13 : The tropical evolution of SO<sub>2</sub> mixing ratio profiles from MIPAS does not really support model simulations especially between 15-32 km. While increasing the photolysis rate of H<sub>2</sub>SO<sub>4</sub> allows SO<sub>2</sub> above 35 km to reach levels observed by MIPAS, the effect of the conversion of COS can barely been seen on the MIPAS profiles near 28-32 km. In addition, because of the gaps in MIPAS observations between 2004 and 2006, the SO<sub>2</sub> inputs from Manam and Soufriere Hills volcano are extremely faint. 11402-20/25 Fig.14 : While fig.14 provides better information on the volcanic sources of SO<sub>2</sub>, the signal from MIPAS is less pronounced and more confined in the tropics. In addition, the first volcanic plume in oct 2002 appears earlier in the MIPAS data than in the simulation.

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 11395, 2013.

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