Atmos. Chem. Phys. Discuss., 13, C5425–C5437, 2013
www.atmos-chem-phys-discuss.net/13/C5425/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Stratospheric SO_2 and sulphate aerosol, model simulations and satellite observations" *by* C. Brühl et al.

C. Brühl et al.

christoph.bruehl@mpic.de

Received and published: 1 August 2013

We thank the referee for the detailed suggestions for improvements and corrections.

1 General comments:

The text will be expanded and some additional material will be supplied in an electronic supplement.

Ignoring the volcanoes just leads to background conditions because almost no tropospheric SO_2 penetrates the tropopause in the monsoon region or other regions with

strong convection because of wet removal. Neely et al. (2013) could only distinguish an effect because they scaled the surface emissions by a factor of 10.

A comparison of our results with the findings of English et al. (2013, which was not available at submission) and Aquila et al. (2012) concerning Pinatubo is an interesting suggestion, which will be addressed in section 3.1. Our results concerning total optical depth are closer to the observations presented there because in contrast to these studies we compute the QBO (see attached Figure 1 for comparison with English et al. (2013)). The reviewer is right that Pinatubo is included in the manuscript as part of validation as suggested in Brühl et al. (2012) and indicated at the beginning of the section.

2 Specific comments:

Abstract: 1: Other SO_2 sources will be mentioned too. We will also replace "middle atmosphere" by "from the tropopause to the mesosphere". In line 16 the following will be included: "The MIPAS data and the model results show that anthropogenic SO_2 from China can only make a minor contribution to stratospheric aerosol."

11396.12: More information on simulated tropospheric aerosol and comparison with observations will be provided in the text and an electronic supplement.

12: The word "realistically" will be replaced.

12: For the medium size tropical volcanoes the effect of radiative heating on dynamics is not significant in the shown example simulation. Here we agree with the reviewer that the text might be misleading. For the big Pinatubo eruption it is clearly significant, however. The text will be expanded here for clarification.

13: OMI/TOMS SO_2 and the model will be mentioned here.

16: "only..." will be replaced by "only when additional processes are included in the model. A working solution is to include a sulphur sink onto the meteoric dust as proposed decades ago and to increase photolysis of gaseous H_2SO_4." The improvements are shown in Figs. 15 and 16. To include relative deviations to the observations is only useful if the model dynamics is nudged to observations. For a free running CCM only patterns can be compared. The text that explains the figures will be expanded.

Introduction: 21: Removal of SO_2 by rainout and the different solubility of COS and SO_2 will be mentioned here, too, for clarification.

23-26: This is supported by observations of ACE-FTS and other satellites, and references will be included.

11397.1,4: Words will be corrected

4: "upward" will be included here. This transport would hold also for SO_2 from other sources if it can reach the lower stratosphere without prior removal in clouds and precipitation. Note that the SO_2 that might survive rainout and reach the TTL is transported into the stratosphere rather slowly (months) during which it is oxidized into sulfuric acid and to a large degree scavenged by sedimenting ice particles, also leading to the drying of air that passes the tropical tropopause.

10: Will be replaced by "from surface to mesosphere"

13-14: To be corrected

Model setup: 24: Text will be expanded. There was no nudging applied except for the initial conditions. The agreement is related to the initialization and subsequently by chance. The agreement deteriorates in 2007.

24: We will include more information on surface emissions and refer with some sentences to the given references. Anthropogenic SO_2 emissions are taken from EDGAR, tropospheric volcanic emissions are included too as climatological average values. The volcanic emissions are from continuously degassing volcanoes only, there is no erup-

tive emission into the troposphere in this dataset. 26: Tests with local injections using the same mass of SO_2 have also been performed with similar results in case of tropical injections. Also a simulation with a local injection of 17Mt SO_2 at July 1 of Pinatubo and 1.3Mt from Cerro Hudson in late August has been carried out (see below). Zonal averages were used to be comparable with the simulations of CCMI and CCMVal of SPARC where zonal average perturbations were used. The text will be expanded.

11398.7: This means different aerosol models or climatologies and/or an optional separation between stratospheric and tropospheric aerosol. The text will be expanded.

8: To replaced by " chemical and dynamical effects of the aerosol"

8: "i.e. an ozone climatology (Fortuin and Kelder, 1998) instead of the online ozone is used in the radiation module."

12: "from 1.0 to"

12: ".. particles (a too large fraction in coarse mode)"

14: A figure will be provided in the supplement.

Section 3.1: 20: Repetition? 21: The reference Guo et al. (2004) will be included.

21: This is not in contradiction to Arfeuille et al. (2013) if the detailed development directly after the eruption is not simulated but instead SAGE data after lofting are used. At 22 km we have a secondary peak.

24: We use recent SAGE data of Thomason (personal communication 2011 and 2013) close to the ASAP dataset (v6.2) but with further corrections for the "high estimate" and sensitivity studies. In the so-called lower estimate old data with aerosol at too large altitudes (used as estimate for SO_2 injection heights) were taken. To avoid confusion we will remove these simulation results from the manuscript.

11399.1: This is used mostly to get an estimate for the region where SO_2 is injected without extrapolations for data gaps due to saturation. As shown in Table 1, the estimate causes a minor overestimate of total sulphur injections compared to other estimates (3%–12%). A sensitivity study describing the early phase in more detail is performed even though the simulation of Pinatubo is not the main objective of this study. The results will be provided in Figure 2, Figure 6 and the supplement.

4: Low estimate will be removed, it was based on old data.

5: This part will be modified including results of the above sensitivity study for improved quantification.

7: The information can be found in the figure but text will be expanded for clarification.

9: The enhanced tropical upwelling in case of dynamical coupling causes higher SO_2 in the upper stratosphere in better agreement to the ATMOS-observations. The sentence will be improved for clarification. The sentence in parentheses will be removed and "both" added in the caption.

14: An e-folding time will be included too.

18: We will explicitly mention "enhanced tropical upwelling" instead of"Brewer-Dobson circulation" and include a figure on the tracer N_2O and the H_2O tape recorder in the supplement. The issue is controversial in the literature, indeed.

18: The reference will be added, thanks.

19: ? results of longer run ? (residence time)

22: Percentage will be included, figure 2 (lower part) will be replaced as attached. An additional figure including the compilation of lidar data by Heckendorn et al. (2009) and results of the detailed sensitivity will be in the supplement.

25: There are few data on this but we will be more careful. Text will be modified.

11400.1: A short summary of Arfeuille's findings will be included.

2: Altitude region to be included.

3: "6-9 mo in the model and 8-11 mo in real life (or the sensitivity study in the supplement)"

6: A slight long wave cooling of 0.2 K/d above the aerosol plume occurs in the first 2 month, but not later. The information can be extracted from the output of the radiation module, there is no additional simulation needed.

6: There have been several sensitivity studies with similar findings. Here we want to point more to the qualitative result.

10: The prognostic quantity in the model is mixing ratio but it is not essential to show this figure in the main text. It will be moved to the supplement. The conversion of SAGE data via SAD was used in several earlier studies, not only by us. (the inversion is not unique).

14: "8 months (6 months in figure)"

15: Reference to Arfeuille et al. (2013) and to Thomason (personal comm.) will be included.

21: A figure will be included in the supplement. Meant is "typical median wet effective radius"; this will be corrected.

26: Thanks, this will be corrected. The reference to Toohey et al. (2011) will be included. The background of the Pinatubo signal seen by ERBE shown there has a bias of more than 1 W/m^2 so that the simulated 10 to 11 W/m^2 is well within the uncertainty range.

Section 3.2: 7: Table 1 will be referenced earlier.

11401.10: to be done.

12: Maybe, percentage of monthly averages (see supplement).

15: Will be expanded.

17: The inclusion of organic carbon helps in the lower stratosphere. Figure 8b will be replaced because of a mistake causing a slight overestimate of the contribution of OC (see attached Figure 3). Further investigation needed?

18: The integration over the latitude range is done for simulations and observations, i.e. the sample bias cannot be negative here. The range is selected because there are many months with full coverage and it covers 87% of the earth's surface. Text will be slightly modified.

21: A figure will be added to the supplement.

24: km-range?

11402.2: In contrast to Pinatubo the differences are small and not significant due to meteorological variability. An example is shown later. The text will be expanded here.

6-12: The text will be clarified. Shown is the difference between a coupled simulation and a simulation with an aerosol climatology. A significant difference occurs only shortly after a perturbation.

Section 4: 14, 16: The dates are given in section 3.1 in connection with Figure 1. Text will be improved for clarity and will include also El Chichon.

20: This is possible but we doubt that percentage difference to the noisy data helps much. A figure with monthly averages will be in the supplement for assessment of the smoothing effect. In line 21 a "probably" will be inserted because there are also other error sources.

23: A paper by Höpfner et al. (2013b) on detailed analysis of the temporally high resolved SO_2 plume structure on the basis of MIPAS single scan retrievals is in preparation which will be cited. We have the figures. See also interactive discussion (paper C5089) to Höpfner et al. (2013a).

25: This refers to Figs. 13 and 14. SO_2 was injected around 20 and 18 km, depending

on the volcano. In Fig. 13 it is clearly visible that the volcanic SO_2 plumes propagate upward. At 22 km no SO_2 was injected. This information would be mostly lost if just monthly averages are shown.

11403.2: This refers again to the paper in preparation.

21: This is estimated but will be checked in our laboratory.

11404.5: In Figure 15 the improvement is visible especially in high southern latitudes.

Conclusions: 10: No, the statement is supported by multi-instrument satellite observations of COS (see also reply to other reviewer). The wording will be slightly changed.

12: In the text more on this will be included. Concerning MIPAS, the statement was based on a paper in preparation (Höpfner et al., 2013b) to be cited.

14: Wording will be changed.

17: There were several simulations (see above).

22: Concerning photolysis of H_2SO_4 several sensitivity studies have been performed. The best estimate was selected for the manuscript. Concerning meteoric dust the settings are based on available literature. The value for the sticking coefficient is confirmed in the laboratory, see also the short comment in ACPD.

11405.2: A figure will be included in the supplement and referenced in line 11403.8. "higher up" will be included after "aerosol layer" for clarification.

3 References

Aquila, V., L. D. Oman, R. S. Stolarski, P. R. Colarco, and P. A. Newman: Dispersion of the volcanic sulfate cloud from a Mount Pinatubo –like eruption, J. Geophys. Res., 117, D06216, doi:10.1029/2011JD016968, 2012.

Arfeuille, F., Luo, B. P., Heckendorn, P., Weisenstein, D., Sheng, J. X., Rozanov, E., Schraner, M., Brönnimann, S., Thomason, L. W., and Peter, T.: Uncertainties in modelling the stratospheric warming following Mt. Pinatubo eruption, Atmos. Chem. Phys. Discuss., 13, 4601–4635, doi:10.5194/acpd-13-4601-2013, 2013.

Brühl, C., Lelieveld, J., Crutzen, P. J., and Tost, H.: The role of carbonyl sulphide as a source of stratospheric sulphate aerosol and its impact on climate, Atmos. Chem. Phys., 12, 1239–1253, doi:10.5194/acp-12-1239-2012, 2012.

English, J.M., O. B. Toon, and M. J. Mills: Microphysical simulations of large volcanic eruptions: Pinatubo and Toba. J. Geophys. Res., 118, 1880–1895, doi:10.1002/jgrd.50196, 2013.

Fortuin, J. P., and H. Kelder, A ozone climatology based on ozonesonde and satellite measurements, J. Geophys. Res., 103, 31,709–31,734, 1998.

Guo, S., G. J. S. Bluth, W. I. Rose, I. M. Watson, and A. J. Prata: Re-evaluation of SO2 release of the 15 June 1991 Pinatubo eruption using ultraviolet and infrared satellite sensors. Geochemistry Geophysics Geosystems, 5, No.4, doi:10.1029/2003GC000654, 2004.

Heckendorn, P., D. Weisenstein, S. Fueglistaler, B. P. Luo, E. Rozanov, M. Schraner, L. W. Thomason and T. Peter: The impact of geoengineering aerosols on stratospheric temperature and ozone. Environ. Res. Lett. 4, 045108, 2009.

Höpfner, M., Glatthor, N., Grabowski, U., Kellmann, S., Kiefer, M., Linden, A., Orphal, J., Stiller, G., von Clarmann, T., and B. Funke: Sulphur dioxide (SO_2) as observed by MIPAS/Envisat: temporal development and spatial distribution at 15-45 km altitude. Atmos.Chem.Phys.Disc. 13, 12389-12436, 2013a.

Höpfner, M. et al.: Sulfur dioxide as observed by MIPAS/Envisat: the upper troposphere and lower stratosphere. In preparation 2013b.

Neely III, R.R., O. B. Toon, S. Solomon, J.-P. Vernier, C. Alvarez, J. M. English, K.

H. Rosenlof, M. J. Mills, C. G. Bardeen, J. S. Daniel, and J. P. Thayer: Recent anthropogenic increases in SO_2 from Asia have minimal impact on stratospheric aerosol. Geophys. Res. Lett., 40, 999–1004, doi:10.1002/grl.50263, 2013.

Toohey, M., K. Krüger, U. Niemeier, and C. Timmreck: The influence of eruption season on the global aerosol evolution and radiative impact of tropical volcanic eruptions. Atmos. Chem. Phys., 11, 12351–12367, doi:10.5194/acp-11-12351-2011, 2011.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C5425/2013/acpd-13-C5425-2013-supplement.pdf

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

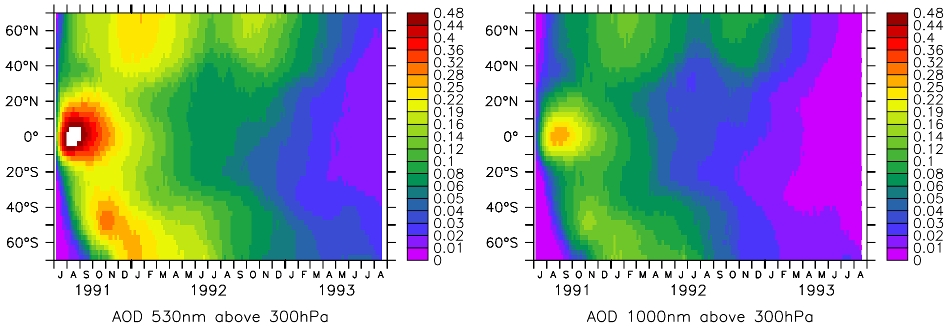

Fig. 1. Aerosol optical depth above 300hPa at 2 wavelengths as simulated by EMAC, corresponding to Fig. 3 in English et al. (2013).

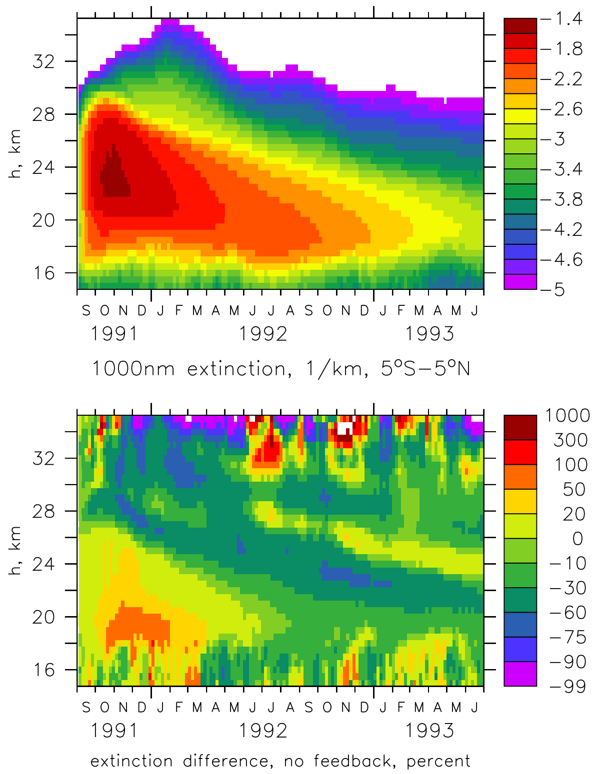

Fig. 2. 1000nm extinction (with radiative coupling to dynamics, decadal logarithm) and percentage change in the simulation without radiative coupling.

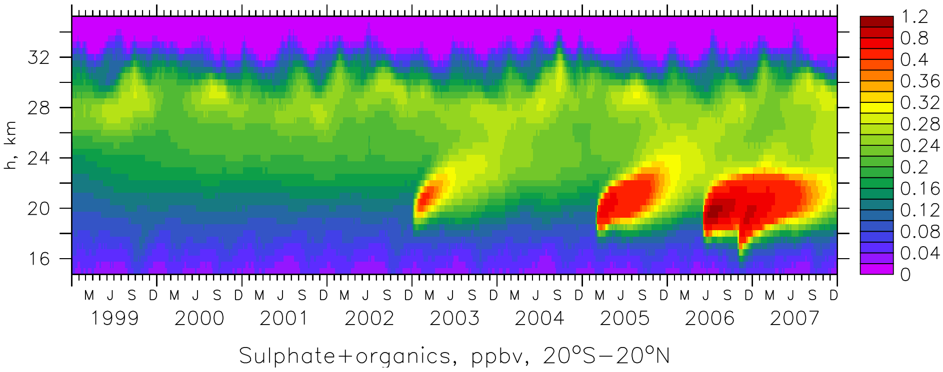

Fig. 3. Improved Figure 8b, sulphate and organics.