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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #1

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Summary

This study presents results from the coupled aerosol-chemistry-climate model EMAC, and comparisons with satellite observations, with a focus on stratospheric SO2 and sulfate aerosols. Most attention is focused on large perturbations to SO2 and sulfate aerosols due to volcanic eruptions: that of Pinatubo in 1991, and a number of smaller magnitude eruptions in the 2000's.

General comments:

The manuscript is written so as to address the current "debate" on the origin of the recent trend in the stratospheric sulfate aerosol layer. However, it is not clear that this study can address this issue with the set up of the presented model runs. There is

not, for example, a clear experiment to dissect the influences of volcanic eruptions vs. surface SO2 emissions as in Neely et al. (2013). Furthermore, the comparisons of model SO2 with observations from MIPAS seem to clearly show that the model underestimates lower stratospheric SO2 mixing ratios in non-volcanic periods. Therefore, I do not find that the results presented in the study support the main conclusion of the manuscript, that (to paraphrase) non-volcanic SO2 is a negligible source of stratospheric sulfate aerosol.

The manuscript has <6 pages of text in describing the results, while it contains 16 figures. This works out to an average of about 10 lines of text per figure, which is not nearly enough to adequately explain the figures, or the implications of what the figures show.

Many interpretations of the presented results simply are overstated. For example, the authors conclude that "radiative heating anomalies ... significantly influence the Brewer-Dobson circulation", however there is no presentation of typically used diagnostics of the BDC (the transformed Eularian mean tropical upwelling, mass streamfunction, wave breaking, etc). Furthermore, the statistical significance of any of the differences shown (e.g., between dynamically coupled or uncoupled simulations) is not addressed at all, so it is impossible for the authors to call any of the results "significant". More examples are included in the specific comments below.

What is most obviously missing from the manuscript is a presentation and discussion of the simulated aerosol size in the different simulations, and comparison with observations (e.g., English et al., 2013). Aerosol size has important implications for sedimentation (i.e., lifetime) and heating rates, and model bias in aerosol radius is likely to be the reason for modeled biases in lifetime and heating.

Given the stated aim of the paper, namely the issue of the stratospheric aerosol trends, the motivation for including a great number of results shown by the authors is not clear. For example, what is the importance of tropical heating rates after Pinatubo (Fig 3)

to the issue of aerosol trends? Much of these results are likely presented as model validation. If so, the manuscript needs better organization, and may include a section on model validation before addressing a scientific issue.

The manuscript suffers greatly from its lack of section describing the observations used within, which should describe which satellite instruments are used (e.g., SAGE II or SAGE III?), the retrieval version(s), the data sources, the time period(s) covered by the observations, the uncertainties in the observations, how the data are averaged (monthly zonal means?), from where the data is available, etc etc.

A number of different simulations are presented, without any overall explanation for the experimental design, or a clear description of the different simulations and their unique configurations. The "Model setup" section should include a list and description of simulations used in the study, ideally with names for those simulations which can be used for the rest of the paper.

Specific comments

(Line numbers are given here as pg.line, i.e., 11396.1 refers to pg 11396, line 1)

11396.1: A major portion of this first sentence (that referring to COS) refers not to work in this study, but to the previous work by Bruehl et al. (2012). As such, this sentence misrepresents what is included in this study. Secondly, since the study does not present a focused experiment to assess the relative roles of volcanic SO2 injection vs. transport of surface-emitted SO2, I don't agree that the study necessarily "demonstrates that the sulfur gases COS and SO2, the latter from low-latitude volcanic eruptions, predominantly control the formation of stratospheric aerosol", with the implication being that other sources are negligible.

11396.12: There is no analysis of tropospheric aerosols in the paper, thus "tropospheric" should be removed from this sentence.

11396.12: "The model realistically..." this is overstated. Model results show some

level of agreement with some observed radiative quantities, but this doesn't necessarily mean the model simulation is realistic.

11396.12: "including the effects on model dynamics". Again, overstated. One example is shown (with more comments below) which hints at the impact of coupled dynamics showing a better agreement between modeled and observed vertical distribution of sulfate. Concerns about this result aside, one cannot use one example to make general comments about the realisticness of the models simulation of dynamics.

11396.13: It has to be clearly stated that these radiative forcing estimates are model-based estimates, themselves completely dependent on SO2 injection estimates from satellite studies.

11396.16: This sentence is also overstated: most likely there are a number of different ways to improve simulation of upper stratospheric SO2. In any case, formulating the sentence with "only" goes too far when no other possibilities have been explored. A more accurate formulation would be something like "Simulated upper stratospheric SO2 showed improved agreement with observations when X and Y were adjusted in the model". However, for this type of formulation, it must be actually shown that the model changes led to quantifiable improvements in the model/observation comparisons.

11396.21: The "$\sim$30%" is presumably the percent of anthropogenic COS compared to total COS emission, although also relevant to the statement is the ratio of COS emission to total S emission. This sentence could be clarified.

11396.23-26: These statements are a model result, or are they validated by observations?

11397.1: proposed

11397.4 volcanic eruptions

11397.4: It's not clear why "transport by the Brewer-Dobson circulation" is a necessary

part of the explanation here – the BDC should impact all chemical species in the lower stratosphere, irrespective of whether they represent the results of volcanic eruptions or convective transport of surface emissions.

11397.10: "lower-middle atmospheric" implies it pertains to the lower part of the middle atmosphere, which is not the intended meaning.

11397.13-14: "caused transport" → injected, "vent" → injection

11397.24: It's not clear quite is meant here with respect to the QBO. Is the QBO realistic in its frequency and amplitude? Or is it actually comparable to the real QBO in terms of its timeseries through the period of time simulated here. And if the latter, how was this achieved? Was it just chance? Or was there some nudging performed in an initialization period?

11397.24: More details are required concerning the emissions, especially in regards to the issue of Asian SO2 emissions. Are the emissions time varying? What other sources of sulfur emission are included (e.g., volcanic silent degassing)?

11397.26: This is an exceptionally strange way to initiate the volcanic SO2. Why not just inject the SO2 at the location of the volcano? One gets the feeling that this method is used to alleviate model deficiencies of some sort (perhaps using this method fixed some problems with too-rapid aerosol growth?). In any case, such a unrealistic way of injecting the volcanic SO2 needs some explanation/justification.

11398.7: "for different aerosol options" in not clear what this means.

11398.8: Is it "aerosol, chemical and dynamical effects", or "aerosol-chemical and dynamical effects"

11398.8: In other words, O3 is fixed, or is it more complicated?

11398.12: from what to 1.6 um?

11398.12: At least a sentence or two should be included here to describe how shifting

the boundary between coarse mode and accumulation mode affects sedimentation velocities.

11398.14: This is quite subjective: some quantitative indication of the level of agreement between the Pringle et al. (2010) tropospheric aerosol burdens and those with the adjusted model set up should be provided.

11398.20: The volcanic eruption injected...

11398.21: An updated estimate of SO2 injection by Pinatubo by Guo (2004) puts the inection at ∼18+/- 4. This uncertainty is good to include.

11398.21: The quoted altitude of maximum aerosol formation seems quite high. The newer SAGE II retrievals shown by Arfeuille et al. (2013) place the initial peak at around 22 km.

11398.24: Again, the SAGE data version is quite important, as Arfeuille et al. (2013) show that newer SAGE retrievals have a very different vertical profile than older retrievals. In fact, this may have quite important implications for the present study, since older retrievals places a significant amount of aerosol below the tropopause, which may explain some degree of the too-fast sedimentation seen in the model simulations which use the old SAGE retrievals to initialize SO2.

11398.24: In reality, the sulfur burden of Sep 1 is probably quite different from the initial injection, since there is already 2.5 months of loss before Sep 1. This should be discussed.

11399.1: The "high" estimate seems almost guaranteed to produce an estimate which is larger than the real injection, since it can easily "double count". Imagine (as a thought experiment) the aerosol plume moving poleward as a relatively intact, zonal "parcel". By the method described, one would find the maximum burden at any latitude at different times. Summing up all these maximum values would lead to a "total" much larger than the actual initial injection.

11399.4: Unclear, does the “low” estimate use a different version of SAGE data?

11399.5: both estimates are within the uncertainty range quoted by Guo (2004). Furthermore, one would expect that the Sep 1 burden would be smaller than the initial (June 15) injection since there is undoubtedly some loss of stratospheric sulfur between June 15 and Sep 1. Therefore, this justification for the focus on the “high” estimate of SO2 injection is not strong.

11399.7: More precise figure descriptions are needed, e.g., Figure 1 shows the temporal development of the vertical profile of tropical SO2.

11399.9: By saying that the simulation with dynamical coupling shows agreement with ATMOS observations, the authors imply that the dynamical coupling simulation shows better agreement than the simulation without dynamical coupling. However, there is not a clear difference between the two simulation results shown in Fig 1.

11399.14: It is hard to objectively state at which point volcanic aerosols are completely removed from the stratosphere, as the aerosol layer decays in an exponential manner towards its background state, which may be different in the model compared to reality. A better measure of the removal rate is the e-folding lifetime, which should be calculated for the modeled sulfate burden and compared to that of the SAGE observations.

11399.18: An increase in aerosol lofting in the coupled dynamics run suggests a possible influence on tropical upwelling. However, it is also hypothetically possible that aerosol heating creates convection-like circulation cells in the tropical stratosphere, where aerosol-lofting upwards motion would be balanced by downwards motion. In such a case, the presence of aerosols may have a zero-mean net effect on tropical upwelling, even while leading to increased lofting of those aerosols. In order to actually say that the aerosols affect tropical upwelling, one would need to actually calculate the transformed Eularian mean vertical upwelling (“w bar star”) within the tropics. Furthermore, the Brewer-Dobson circulation (BDC) is characterized by upwelling in the tropics, poleward motion in the midlatitudes and downwelling in the high latitudes. In

order to say that aerosol heating affects the BDC, one would need to investigate the full structure of changes in stratospheric circulation. This is related to the fact that while tropical upwelling is often used to characterize the magnitude of the BDC, the BDC by definition is driven by the breaking of planetary waves in the mid-latitudes, not by heating in the tropical stratosphere. Therefore, it should not be taken as necessarily granted that aerosol heating in the tropical stratosphere should affect the BDC.

11399.18: The role dynamical coupling to aerosol heating has on the transport of aerosols has been studies in detail in previous studies,. Any discussion here should include references to related prior work (e.g., Aquila et al., 2012 and references therein).

11399.19: “further lofting” is apparent in Figure 2., but “longer residence time” is not.

11399.19: “better agreement with observations” is not shown.

11399.22: Please quantify “considerably”.

11399.25: McCormick et al. (1995) clearly state that temperature anomalies at $\sim$24 km were of the order of 3.5 K, not 7 K ! To say the modeled temperature response is “consistent with observations” is a gross overstatement.

11400.1: “consistent with the finding of Arfeuille (2013)” needs some further explanation.

11400.2: “tropical tropopause region” needs to be defined, i.e., what altitude is referred to.

11400.3: 9 moths after the modeled injection, or the real-life injection?

11400.6: The cooling above the aerosol layer could also be due to a relative decrease in upwelling LR radiation, since more has been absorbed below (compared to a control simulation). How can the authors be sure that the cooling is rather a dynamical effect?

11400.6: Figure 4 deserves more than just passing reference. The impact of aerosols upon the QBO is an interesting issue, however, the results shown here have question-

able significance. The difference shown of dynamically coupled vs uncoupled aerosols uses only one ensemble member for each simulation. It could be argued that inclusion or non-inclusion of aerosol heating serves to add a random dynamical perturbation between the simulations, and that the difference shown could be then the result of this random perturbation. In order to convincingly show the influence volcanic aerosols may have on the QBO, an ensemble of simulations would be required, and the difference compared to natural variability in order to estimate the significance of any difference between the coupled and uncoupled runs.

11400.10: More details on the SAGE data should be included in a Data section. It is, for example, not clear why SADs are converted to mixing ratios and not the measured extinctions (since the SADs are presumably themselves based on the measured extinctions).

11400.14: Because of the mismatch between the modeled SO2 injection and reality, statements like "6 months after the eruption" need a actual date connected (Dec 1991 in this case?) so the reader can assess exactly what is meant.

11400.15: "not filled with extrapolations here" needs more discussion.

11400.21: "typical" means median? Effective radius? And it should be stated whether these radii are realistic or not. And if too large, what implications does this have for the heating rates and temperatures shown earlier?

11400.26: The 8 W/m2 value reported by McCormick et al. (1995) is based on measurements from the Earth Radiation experiment (ERBE). Arfeuille et al. (2013) discuss updates to the SAGE II retrieval version, therefore its not clear why the authors question the accuracy of the McCormick (1995) reported value. Also, ERBE data is publicly available and the model results can be directly compared (e.g., Toohey et al., 2011).

11401.7: A table of the eruptions and their eruption/injection dates would be useful.

11401.10: "corresponding" → "resulting"

11401.12: A difference plot would be needed to visualize such differences.

11401.15: Its not clear from Fig 8 how the QBO "modulates the sulfate distribution".

11401.17: Actually, the gradient in sulfate at $\sim$ 22 km is more realistic in the plots without organic carbon. Statements like this need to be quantified: again, a difference plot would go a long way to being able to better see the impact of including organic carbon or not.

11401.18: Burdens form 60S to 60N are not "total stratospheric burdens".

11401.18: Actually, I would be surprised if there are any months in which the SAGE II latitudinal coverage covers the full 60S-60N area. And for months in which the coverage is incomplete, why can't the SAGE sampled mean be larger than the real full 60S-60N mean? (In other words, is there a reason that the sample bias must be negative?)

11401.21: While in general, more information about aerosol size is welcome, this statement is lost in this location.

11401.24: Define "middle atmosphere".

11402.2: Why is the uncoupled simulation shown here, when previous results showed the coupled runs are more realistic?

11402.6-12: More discussion would be appreciated here. Description of the model (in Sec 2) implied that the radiation module can be used to directly calculate the aerosol heating rates, while the temperature anomalies shown in Fig 12 must be differences of the simulated temperatures from a climatology (not stated). So, the conclusion from Fig 12 would be that the heating rates resulting from these eruptions lead to insignificant temperature changes compared to the natural variability of lower stratospheric temperatures?

11402.14: Please give years of ATMOS measurements.

11402.16: Unexplained reference to the model results in Fig 1 when discussing ATMOS

observations is misleading.

11402.20: Model results should be averaged in the same manner as the observations in order to properly compare them. Computing monthly mean of the model fields should be trivial.

11402.23: “zonal averages ... are well above 1ppbv” - but the maximum values shown in Figure are around 0.15. This is probably the monthly mean vs. daily mean problem, which should be fixed.

11402.25: “The peaks in the tropics...” does this sentence refer actually to Fig 13, because it is impossible to get information about the vertical transport from Figure 14? Also, in contrast to the authors text here, there is no clear signal of upward motion of the MIPAS-observed volcanic aerosol signals. Also, in the model simulations there is some indication of upward motion, but no clear signal that the volcanic injections affect the aerosol layer centered at 28 km.

11403.2: “From analysis...” Without more exposition, this statement seems suspicious, given the rather constant nature of the tropical SO2 measured by MIPAS, and the statements that follow discussing the possible sources of EMAC’s low bias in lower stratospheric SO2.

11403.21: Are these assumptions just wild guesses, or based on evidence?

11404.5: The difference between the two model setups seems negligible at 31 km (Fig 15), therefore this sentence needs some fine-tuning.

Conclusions

11404.10: “transport of COS” is not directly dealt with in this paper, therefore its presence in the main conclusions of the paper does not fit. Moreover, based on Fig 14, the reader sees that the model actually underestimates SO2 in the lower stratosphere during volcanic quiescent periods, implying that the model underestimates SO2 as a source of stratospheric aerosol. This would imply that agreement between the model

and observed stratospheric sulfate aerosol could be a consequence of the model overestimating the contribution of COS!

11404.12: There was little to no discussion of the anthrogogenic SO2 emissions used in the different model simulations, so this statement is not justified.

11404.14: As discussed above, the impact of volcanic aerosols on the BDC is not adequately proven by the analysis presented here. Moreover, no convincing quantification of any changes in aerosol lifetime in the simulations was shown.

11404.17: The results concerning the QBO are also unconvincing, given that only two simulations are compared and no account of possible random changes in dynamics between the simulations was given.

11404.22: As the authors have not performed a sensitivity study comparing the relative impact of different uncertainties in upper stratospheric sulfur chemistry, they overstate their confidence in the importance of the photolysis of H2SO4 mechanism.

11405.2: “Inclusion of the oxidation of DMS...” No results concerning this were shown in the paper, as such its presence in the Conclusions is not justified. Moreover, the statements here seem inconsistent with those of 11403.8.

References

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