



Interactive
Comment

***Interactive comment on* “The global
middle-atmosphere aerosol model
MAECHAM5-SAM2: comparison with satellite and
in-situ observations” by R. Hommel et al.**

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Reply to Anonymous Referee #2 , referring to interactive comments on “The global middle-atmosphere aerosol model MAECHAM5-SAM2: comparison with satellite and in-situ observations” by R. Hommel et al. From 04 February 2011

We thank the referee for the review of our manuscript and the constructive comments. We hope the revised manuscript will be much improved. Below, each comment (in quotation marks) is followed by an individual response.

“Manuscript evaluation criteria Scientific Significance: Does the manuscript represent a substantial contribution to modelling science within the scope of Geoscientific Model

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Development (substantial new concepts, ideas, or methods)? 2 - The paper describes an improved version of a model in a good amount of detail, and this paper will likely be the citation source for those wishing to perform simulations with this model. Scientific Quality: Are the scientific approach and applied methods valid? Are the results discussed in an appropriate and balanced way (consideration of related work, including appropriate references)? Do the models, technical advances and/or experiments described have the potential to perform calculations leading to significant scientific results? 2 - The results of the experiments are discussed well, and this model will undoubtedly be useful in generating future scientific results. Scientific Reproducibility: To what extent is the modelling science reproducible? Is the description sufficiently complete and precise to allow reproduction of the science by fellow scientists (traceability of results)? 2 - The description is good, and with a few possible details, this paper serves as a great description of the model's capabilities, with a specific focus on stratospheric sulfate aerosols. However, there is the issue of reproducibility which could be improved with a more detailed discussion of the specific processes involved that are new to this version. Presentation Quality: Are the methods, results and conclusions presented in a clear, concise, and well-structured way (number and quality of figures/tables, appropriate use of English language)? 2 - The paper is nicely done. Some of the language/word choice could be improved, but I'm far from a competent copy editor, so I'll refrain from commenting further on this.

Detailed comments

The journal suggests the model code and user manual be made available in a publicly accessible way. There should be a mention of this somewhere in the paper. Page 1362, Line 1 - This sentence is a very blanket statement that needs to be qualified. I can think of any number of papers that discuss the climate response to stratospheric aerosols, so you really need to say what you mean here."

This statement is explained in detail in subsequent paragraphs. We will rewrite respective paragraphs.

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“Page 1363, line 6 - "distribution median radius $<0.2\mu\text{m}$ ": I've seen quite a few volcanic eruptions that fit this criterion. It's right to say that background aerosols are generally different, but you should be a bit more careful here.”

We will find another formulation of the context. We agree, there is no common rule to distinguish stratospheric aerosols from their sources by simply investigating the particles size. Several works, in particular those analysing aerosols from the powerful eruption of Mt. Pinatubo, showed, that aerosol size distributions of the volcanically perturbed stratosphere have significantly larger geometric radii than those in a “clean” background stratospheric environment.

“Page 1363, line 7 - "distribution median radius $>0.4\mu\text{m}$ ": This is for large (mostly tropical) eruptions, so you may want to say that.”

This is right. Will be considered.

“Page 1363, line 15 - "measurements": Measurements of what, specifically? Radius? AOD? Everything?”

This phrase refers to the relative error in retrieving the size distribution of particles from Sun and sky radiance measurements. Will be revised.

“Page 1363, line 19 - "formation and global dispersion": Do these also characterize the background aerosol layer? Up until this point, you've only talked about radius, so some discussion of this (if relevant) would be useful.”

Yes, indeed, also the background aerosol formation is dependent on these processes. Up to this point we talked about the importance of stratospheric aerosol, and deficits in the scientific understanding of stratospheric aerosol-climate interactions. We noticed that the latter in principle can be divided into two groups: climate effects of the volcanically perturbed stratosphere and aerosol-climate interactions of the volcanically quiescent stratosphere. In the subsequent paragraph, the paragraph your comment refers to, we address how modelling helps to understand those issues. And the most prominent

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aerosol dynamical processes affecting the Earth climate, the processes which have to be modelled as accurately as possible, are aerosol formation and the subsequent transport of those particles (including all additional sink and source terms).

“Page 1363, line 21 - "soluble aerosol above the tropopause": Do you mean the tropical tropopause? Or is this true for mid and high latitudes as well? Maybe some of the mechanisms could be briefly described.”

It is not constrained to the tropics. We think, however, going too much into detail and explaining the life cycle of stratospheric particles will be beyond the scope of the paper. At this point, let us refer the reader to a respective and well written article in BamS: “The life cycle of stratospheric aerosol particles.” by Hamill et al. (1997).

“Page 1363, line 28-29 - "systematically affects model predictions of aerosol and precursor transport and mixing": How much is the effect?”

It is not easy to answer this question. In principle, an effect can be derived qualitatively from the works we cited at this point, and later when we discussed our model's results (Section 3.1 – 3.4). The magnitude of the effect differs between the quantities of interest. In our understanding, the effect will be relatively small when global aerosol dispersion is analysed in terms of integrated global quantities (burden, aerosol optical depth or equivalent quantities) and apparently large when analysed in terms of aerosol size quantities or aerosol concentration. Because the total scale of both of the latter is much larger than for the first category quantities (integrated quantities are spanning normally not more than 2 orders of magnitude whereas e.g. concentrations easily are spanning several, let us say 5 or 6 orders of magnitude). We will include a sentence explaining this.

“Page 1364, line 1 - "aerosol direct and indirect radiative forcing": I've seen plenty of treatments using bulk forcing where they did the direct forcing pretty well. Are you referring to anything in particular?”

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No, nothing in particular. Also aerosol forcing is not subject of this work, because during the time of our studies the microphysics code was not interactively coupled to the general circulation model's radiation code. As we mentioned before, the effect can be small for global integrated quantities. We agree with the referee and make this point clearer in the revised version.

“Page 1364, lines 2-4 - I'd be very careful with this statement, especially when using a reference as old as this one. IPCC (2007) says clouds are the greatest source of uncertainty. Please clarify what you mean here.”

We did not say “the largest contributor to general model uncertainty”, we said aerosol are “one of the largest contributors”. We will mention the big uncertainties due to clouds “... next to clouds aerosols are one of the largest contributors to uncertainty.”

“Section 1 (general comment): At some point, you might want to mention a shortcoming of the approach you've chosen. GCMs use the bulk treatment because it allows them to include other climate-relevant processes (including a dynamic ocean) which these more specialized models can't do due to the large computational time required. So what I'd like to see mentioned in this paper is what questions you plan to answer with your model and what the boundaries are for what questions you can ask of it.”

In recent years, much effort was made to improve aerosol microphysics codes for atmospheric general circulation models (GCM's). The list is long, some of the works and respective models were mentioned in Section 1 of our manuscript. In many of those works aiming at significantly improved complexity of aerosol processes of the models. More specific is the situation of atmospheric GCM's which explicitly consider the circulation of the stratosphere. Their computational effort is much larger than that of tropospheric models. Our model set up belongs to the middle-atmosphere group of GCM's. In this work we attempt to significantly improve the description of the stratospheric aerosol layer and their underlying processes. We believe, we succeeded and built a model showing a nice performance. Also our model has not many competitors

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so far. The largest shortcoming of our system is, that the aerosol composition is restricted to the binary system sulphate-water. But this still is a suitable assumption for the stratosphere, where other aerosol compounds play a minor role (details are given in WMO/SPARC, 2006). In the troposphere, however, it is clear that this yields an incomplete picture of the aerosol composition and affects associated climate process interactions. In Section 3.1 and 3.3 we have addressed this issue when we compared parts of our results to model systems which were developed for tropospheric conditions, model systems which are widely used by the community and in the meantime known for its quality and performance. However, we had to carefully choose which data are comparable. Naturally we focused on modelled sulphate quantities, and had to make sure that such quantities from other works do not relate to mixed aerosol compounds. In the revised manuscript we consider the referees comment and add a paragraph exactly categorising our model's set up.

“Section 2.2 (general comment): In this paper, you only talk about sulfur. If SAM2 is restricted to sulfur parameterizations/calculations only, you might want to mention that.”

See comment above. We will state that more clearly.

“Page 1367, line 9 - "time scales of interest": What are the scales of interest?”

We are referring to the time integration interval typically applied in GCM's. We will find another formulation.

“Page 1367, lines 24-25: Doing the simulations this way sounds very expensive. It would be interesting to give an estimate of the increase in required computer time.”

Indeed, the computational expense is much larger than in models where the aerosol prognostic quantity is transported as a bulk, as e.g. in Timmreck (2001). On the other hand, this treatment is one of the advances of our model and is the only way to predict the size spectra of an aerosol population. Previous models, in particular middle-atmosphere models, did not make use of this great effort due to computational

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limitations. For the Echam5 model family, any additional passive tracer increase the computational expense by approximately 4 percent (Hauke Schmidt, personal communication). However, simultaneously solving the aerosol dynamics equation, and in particular the coagulation equation, is much more expensive than tracer advection.

“Page 1368, first full paragraph: Excellent discussion of methods.” – “Page 1369, lines 4-6: This needs a bit more description, such as actual values of stratospheric injections you include. Do you also include quiescent emission rates?”

We will improve the description of volcanic emissions in the revised manuscript. As all other sulphur emissions in our model, the treatment of volcanic emissions follows the AEROCOM protocol, scenario B (present day scenario, representing year 2000; <http://dataipsl.ipsl.jussieu.fr/AEROCOM/>). Non-eruptive SO₂ emissions are taken from Andres and Kasgnoc (1998) and are distributed to model levels between volcano height and one third below. Eruptive SO₂ emissions were taken from Halmer et al. (2002) and are distributed to model levels 500 m to 1500 m above the volcano height. Our study focuses on the stratospheric background period after 1995, so that any stratospheric injections are not considered. We will provide the details in the revised manuscript.

“Page 1370, lines 3-5: This will certainly affect your results. Do you have estimates of how much/what kind of error is introduced into your calculations by doing this offline?”

Errors cannot be estimated easily without an appropriate experiment applying a fully coupled chemistry module, including a formulation for photolysis. Respective data useful for a direct comparison of the values we diagnosed from the scheme are not published in peer reviewed journals as far as we know. We will not speculate about quantitative effects. Qualitatively, however, the results are more than reasonable.

“Page 1371, line 1 - "data from km": Is there a number missing here?”

Type setting error. Must be “...considers data from 2 km above the tropopause...”

“Page 1373, line 23 - "last year of integration": Is this stable, or is there weather noise?”

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Would it not be better to take averages of the last few years?”

All the years considered in the analysis are “stable” since the model was spun up for 6 years. Hommel (2008) showed that this time was sufficiently long to reach a well balanced state of the stratospheric aerosol layer. Since we used climatological mean boundary conditions (sea surface temperatures and sea ice concentrations), and this model set up is not able to reproduce the quasi biennial oscillation of the stratosphere (details will be given in a companion paper), the stratospheric aerosol layer is predominately showing a distinct annual cycle with very small interannual variations. Therefore, we can make the analysis in any year we want. In particular global integrated quantities as shown in the budget analysis do not differ at pre-decimal positions in between the years. That’s why the budget of the model climatological mean, in our case 11 years from 1996 to 2006, or any other mean period, is not significantly different from that of a specific year. The situation will be different when the quasi biennial oscillation of the stratosphere is considered – this will be shown in a companion paper.

“Page 1374, lines 9-10 - "where aerosol processes are more or less constrained to the troposphere": Does this imply stratospheric aerosol processes do not impact the global aerosol burden?”

No. Several works, such as ours, are showing that in the absence of very large volcanic eruptions, stratospheric aerosols contribute 15 – 20 % to the global annual mean sulphate aerosol burden. We said this clearly, underlined by numbers from other studies. If our burden, which includes the stratospheric contribution, is more or less the same as diagnosed from tropospheric models, then in the latter some processes are of different strength than in our model. This is due to different mechanisms within different models or process parametrisations. Even if identical boundary conditions and emission scenarios are applied in respective models, such discrepancies might occur.

“Page 1375, line 14: Whose treatment would you say is in better agreement with observations?”

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As we showed in Section 3.2, the SO₂ abundance in the stratosphere is a big unknown. With respect to SO₂ conversion in the stratosphere, we believe we did a better job than the study of Weisenstein et al. (1997), because we applied latest JPL recommendations and also show a more consistent picture of the life cycle of stratospheric aerosols and their precursors.

“Page 1376, line 4 - “benchmark model”: This benchmark model needs a lot more description. How does it work? References? Description? Why is this the benchmark? Is it more accurate than your current model, in which case, why are you using your current one? Also, why are you using a benchmark _model_? Aren’t observations the benchmark?”

The benchmark model is described in some detail in the work of Kokkola et al. (2009). Also, in Kokkola et al. (2009) the performance of the benchmark model compared to other aerosol modules (HAM, SAM2, SALSA) is investigated in detail. We believe it does not make sense to go to much into detail here. However, in the revised version of the manuscript we will provide name and reference of the benchmark model. Interested readers may be referred to Kokkola et al. (2009). Observations have very large error bars, so a combination of observations and very high resolution models (offline!) is the best testbed we can have.

“Page 1376, lines 4-9: If you have to reserve some of the sulfuric acid vapor for nucleation, doesn’t this suggest there’s something wrong with the rate of condensation?”

No. Our knowledge of the nucleation process is much more insufficient than that of vapour condensation, so that the big unknown in the study we are referring to this point is the nucleation process. However, so far the global aerosol modelling community does not have an alternative to the binary homogeneous nucleation parametrisation of Vehkamäki et al. (2002) for upper troposphere/lower stratosphere (UTLS) conditions. The work of Hommel and Graf (2010) simply introduces an empirical parameter, which role it is to avoid model shortcomings which occur under large H₂SO₄ vapour

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saturations (shown in Kokkola et al., 2009). A similar parameter also exists in other models, e.g. the modal aerosol module HAM for the GCM Echam5 (Stier et al., 2005). However, due to the modal concept of HAM, resulting relationships might be different from what Hommel and Graf (2010) showed for a sectional model. In Hommel and Graf (2010) it was said, that the condensation sink is overestimated when the vapour saturation is high, and SAM2 fails to reproduce observations of integrated aerosol quantities (in that case the aerosol surface area). The empirical factor corrects this in a simple, non-iterative way, and makes the module available for conditions as found in a volcanic plume.

“Page 1378, line 9: I’m not sure what this line means.”

We will find a different formulation. What we mean is: When the H₂SO₄ vapour pressure increases with altitude, the condensation sink of aerosols decreases.

“Page 1380, lines 10-11: Is this due to conversion into aerosols? Or is it due to transport?”

This refers to modelled SO₂ mixing ratios compared to the model of Takigawa et al. (2002). It is likely that the Tagikawa model under-represents SO₂ oxidation in the stratosphere. They estimate a one order of magnitude lower flux from SO₂ into aerosols in the stratosphere. The chemistry scheme of Takigawa et al. (2002) is not as comprehensive as in our set up, this was stated in Section 3.1. In contrast transport, mainly through the tropical tropopause layer (TTL), seems to play a minor role here, since both models are fully 3-D and transport issues are not believed to account for an order of magnitude difference when emission scenarios are more or less of similar strength.

“Page 1380, line 11 - "missing links": Please say a bit more about these.”

We will revise this formulation and give details as far as the description of the Takigawa model (see above) allows.

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“Page 1380, line 13: I’m not sure what this sentence means. Does the H₂SO₄ spend less time in the gas phase?”

If you express it that way, yes. It seems that in Takigawa et al. (2002) H₂SO₄ vapour is converted into aerosol quickly, regardless of the exact method. It’s not easy to interpret which process accounts for that, nucleation or H₂SO₄ condensation.

“Section 3.3 (general comment): In talking about total atmospheric sulfate or tropospheric sulfate, there is the implication that the tropospheric treatment in MAECHAM5-SAM2 is different from ECHAM5. This is counterintuitive, because MAECHAM5 is a middle atmosphere model, so presumably most of the changes were made there. A bit of discussion on this would be helpful.”

Echam5 does not have an explicit aerosol component. Echam5-HAM (Stier et al., 2005) is an aerosol module, which in the meantime is used widely by the community. Echam5-HAM describes an aerosol population by means of 7 log-normal distributions, which are called modes. It also considers other aerosol components than sulphate and allows predictions of the aerosol’s mixing state. Technically the sulphate component is treated identical in the modules HAM and SAM2. Only minor adoptions are made in SAM2 to describe an aerosol population by means of fixed size sections (the so called sectional method). That means that the tropospheric treatment is not different. Apart from the aerosol composition, the only difference to Echam5-HAM is that the stratosphere is fully resolved and thermodynamic equations and parametrisations are valid for stratospheric conditions (which is not the case in HAM). We will make this clear in the revised manuscript.

“Page 1384, line 19 - "reproduced": Is that because these specific mechanisms were added to the model?”

No. It refers to the fact that in our study this prominent feature of aerosols in the stratosphere is seen in the results. The feature was pointed out first by model studies of Mills et al. (1999). Older work did not pay much attention to that. We will make that

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clear in new text.

“Page 1385, line 28: Is there any particular reason you chose this range?”

The numbers refer to contour lines in Figure 8b which mark the particular region where the meridional gradient in the condensation transfer concentration is weak. We will make it clear in a revised manuscript.

“Page 1386, line 23: This assumes SAGE II is accurate. Do you have another source of data or at least some estimates on the error involved in SAGE II measurements?”

Your comment refers to a qualitative statement. If the error is larger in one or another SAGE II quantity, then such a statement is relative. We will check this and correct the phrase if necessary.

“Page 1387, lines 8-27: Very nice discussion.” – “Page 1390, lines 6-7: Does this grid cell include the nearby Rockies and thus have the possible influence of orography? If so, it might be better to shift your grid cell downwind, since that’s where the balloon will travel anyway.”

In the respective references one does not find any information, how far the balloon travel during an ascent. At first instance, a single model grid cell, covering a horizontal area of $2.8 \times 2.8^\circ$, seems to be large enough for a comparison with in-situ balloon measurements and has been a common practise by global modellers. Orographic effects cannot be ruled out and are subject for further analysis of the model results.

“Page 1390, line 8: You have to be careful with using the years 1998-2006, because the background aerosol layer has increased pretty dramatically over this time period (Hoffmann et al., 2009, GRL). It might be better to pick a few years that match your other simulation parameters (1999-2000, for example) rather than the whole range.”

By analysing Lidar data from Mauna Loa and Boulder, for the volcanically quiescent period after 2000, Hofman et al. (2009) infer an annual increase in the stratospheric aerosol backscatter of 4 – 7%, potentially from increased coal burning in China. So far

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this is the only work showing an increase in the stratospheric aerosol mass (which is proportional to the backscatter). By contrast the Assessment of Stratospheric Aerosol Properties (WMO/SPARC, 2006) did not reveal a clear trend in the stratospheric aerosol content after the decay of the volcanic material from Mt. Pinatubo. Nevertheless we find the works of Hofmann et al. (2009) very interesting and remarkable. With the help of our model, future studies might confirm an increase in the stratospheric aerosol mass from intensified sulphur emissions in East Asia. We will mention this in revised text.

“Page 1391, line 4 - "relative large uncertainties": If $\pm 30\%$ is a lot, you need to say why.”

We understand and will find a better formulation in the revised manuscript.

“Page 1391, lines 9-10: Is this an indication of a systematic bias in the model or the observations?”

This feature is caused by two reasons. One relates to a characteristic of the modelled growth process, the other to the method how integrated aerosol size quantities are inferred from optical particle counter (OPC) number concentration measurements. It is explained in the following full paragraph of Section 3.5.

“Page 1391, line 15: Well, sure, but consider the mass or radiative effectiveness of the aerosols in the two modes. In that context, it's effectively unimodal, which is more likely what the "typical" discussion has been.”

We understand the referees view. From the aerosol dynamics modellers view however, “typical” aerosol size spectra are bimodal. However, the smallest mode does not contribute significantly to the radiative effectiveness of the aerosol, so that finally one might think of it as a unimodal distribution.

“Page 1391, lines 21-26: These particles are probably way too small to have a significant impact on radiation, surface area, and volume density. So you need to be a bit

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more clear about what you mean here. Also, back some of this up with calculations. For example, calculate surface area, volume, and radiative forcing with and without including this small mode.”

The fine mode particles do not scatter effectively, so they do not contribute much to the radiative forcing. The paragraph, however, explains why we see a systematic bias between the modelled effective radius and that inferred from OPC measurements in Figure 13. As mentioned above, one reason relates to a model characteristic, the other to the method how integrated size quantities are inferred from OPC measurements of aerosol concentrations. Your comment refers to the part which explains the model characteristics. Let us explain the situation in other words once more: If, in a certain region of the lower stratosphere, many ineffectively scattering fine mode aerosols are available and the reservoir of condensable vapour is large, the fine mode aerosols grow quickly and effectively to (with optical instruments) “detectable sizes”. An aerosol population’s volume distribution is shifted towards the larger end of an arbitrary aerosol size spectrum compared to the aerosol population’s surface distribution. Therefore, the integrated aerosol surface area increases faster than the integrated volume density, when the above mentioned fine mode particles are growing into the range of “detectable sizes”. Thus, their quotient, which is the effective radius (multiplied by 3), decreases when fine mode particles are growing into this range of “detectable sizes”. In our model this ratio, and thus the ratio of the strength of the modelled aerosol dynamical processes, is fixed and not variable over wide range of conditions and regions. We assume that this results from the single moment approach for aerosol dynamics predicting only the aerosol mass. We will revise the paragraph and will state the relationships more clearly.

“Table 2 would be improved if there were a line or two of observations.”

Unfortunately we did not find consistent values from observations for both of the parameters shown in Table 2. Therefore we concentrated on comparable model results.

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"Figure 2 - "CSO" should be "OCS" for consistency with the rest of the paper."

Will be changed.

"Figure 8 - Parts of this graph don't make a lot of sense. How can a concentration be negative? Perhaps some more description is needed."

Yes, more description is needed. Figure 8c shows the mass transfer away from the particles. This transfer will be negative when mass transfer onto the aerosols (Figure 8b) is defined positive.

"Figure 13, last two lines: Should $\leq$ be $\geq$ instead?"

Another typo. Thank you.

References, additional

Hofmann, D., J. Barnes, M. O'Neill, M. Trudeau, and R. Neely (2009), Increase in background stratospheric aerosol observed with lidar at Mauna Loa Observatory and Boulder, Colorado , Geophys. Res. Lett. , 36 , L15808, doi:10.1029/2009GL039008.

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