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Atmospheric Chemistry and Physics
Discussions

Interactive comment on “Stratospheric aerosol radiative forcing simulated by the chemistry climate model EMAC using aerosol CCI satellite data” *by* Christoph Brühl et al.

Christoph Brühl et al.

christoph.bruehl@mpic.de

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1 Introduction

The reviewers are right that some sections are too short because the manuscript was written under time pressure. We also understand that the text might be more precise and that several clarifications are needed, and we thank the Referees for their effort to specify what is odd or missing and what causes misunderstandings in our formulation. In the revised version we would follow the suggestions of Referee 2, and also replace preliminary results in the figures by the full simulation noted as ’ongoing’ in the

published version criticized be reviewer 1.

Concerning Referee 1’s suggestion to submit this manuscript to another journal, our opinion is that GMD is not an adequate one for this study because it addresses both the satellite and modelling community and its focus is on presenting model results and observations, not on model development. This will be addressed more clearly in the revised abstract, introduction, results section and conclusions. Therefore, we believe that the choice of ACP is the right one to reach the concerned communities. One important aspect here is that the model is able to calculate observed quantities at the original wavelengths of the instruments, providing consistent information, also for radiative forcing.

The suggestion for revised abstract and conclusions is:

"**Abstract**. This paper presents decadal simulations of stratospheric and tropospheric aerosol and its radiative effects by the chemistry general circulation model EMAC constrained with satellite observations in the framework of the ESA-Aerosol-CCI project such as GOMOS (Global Ozone Monitoring by Occultation of Stars) and (A)ATSR ((Advanced) Along Track Scanning Radiometer) on the ENVISAT (European Environmental Satellite), IASI (Infrared Atmospheric Sounding Interferometer) on MetOp (Meteorological Operational Satellite), and, additionally, OSIRIS (Optical Spectrograph and InfraRed Imaging System). In contrast to most other studies, the extinctions and optical depths from the model are compared to the observations at the original wavelengths of the satellite instruments covering the range from UV (ultraviolet) to terrestrial IR (infrared). This avoids conversion artifacts and provides additional constraints for model aerosol and interpretation of the observations.

MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) SO_2 limb measurements are used to identify plumes of more than 200 volcanic eruptions. These three-dimensional SO_2 plumes are added to the model SO_2 at the eruption times. The interannual variability of aerosol extinction in the lower stratosphere, and of stratospheric aerosol radiative forcing at the tropopause is dominated by the volcanoes. To explain the seasonal cycle of the GOMOS and OSIRIS observations, desert dust simulated by a new approach and transported to the lowermost stratosphere by the Asian summer monsoon and tropical convection turns out to be essential. This applies also to the radiative heating by aerosol in the lowermost stratosphere. The existence of wet dust aerosol in the lowermost stratosphere is indicated by the patterns of the wavelength dependence of extinction in observations and simulations. Additional comparison with (A)ATSR total aerosol optical depth at different wavelengths and IASI dust optical depth demonstrates that the model is able to represent stratospheric as well as tropospheric aerosol consistently."

"**Conclusions**. Satellite data are not only important to constrain model parameters; they are very important for model improvement. Comparing satellite data with model results at different wavelengths simultaneously provides additional information and is also valuable for the satellite community to check internal consistency, as in our case for GOMOS and OSIRIS.

Sophisticated modelling of dust and organic aerosol as well as a detailed volcano dataset are necessary to reproduce the seasonal cycle and interannual variability of extinction in the lowermost stratosphere observed by GOMOS at different wavelengths. From the wavelength dependence in observations and simulations regions in the UTLS with enhanced particle size due to water uptake can be identified as aged dust in the Asian monsoon region. Convective transport of dust into the UTLS is resolution dependent because of differences in convection top height and overshooting convection, A resolution of T63L90 (1.88^o in longitude and latitude, 90 vertical layers) fits best to the observations. For the low resolution T42L90 (2.75^o) dust SAOD (and mixing ratio) has to be downscaled by a factor of about 0.33, while for higher resolutions (e.g. T106L90) upscaling is required. The resolution dependent differences in convection modify also the residence time of sulfur species in the lowermost stratosphere, especially at low latitudes, in resolution T42L90 it appears to be too short.

The total AOD in the visible spectral range is very sensitive to aerosol water and the composition of sea salt. In the modal model, the bulk fraction has to be increased compared to ions to reduce artifacts of too much water uptake by sea salt. The satellite data helped to identify a preferred parameter set for the sea salt emission composition.

Our simulated dust total aerosol optical depth agrees with satellite data in the visible (ATSR SU) and the infrared (IASI ULB, version 8). The combined comparison at visible and infrared wavelengths provides strong constraints on the modelled particle size distribution. The direct comparison of observations and model reveals different structures in the extinction patterns at both spectral ranges. From this, we conclude that simply assuming a spatially constant factor of (about) 2 for conversion of DAOD from 10 μm to 550 nm, as commonly applied in the AEROCOM/AEROSAT community, is too crude.

Satellite datasets identifying volcanic SO_2, including its vertical distribution or enhanced extinction by aged dust enable the model to get closer to observationally based estimates for radiative forcing, showing the interest of a close interaction between modelling and observation research teams."

2 Reviewer 1, questions and answers

2.1 General comments

The introduction of the paper doesn't state an aim, and doesn't formulate a scientific question. It provides some background information in particular on stratospheric aerosols, satellite data, and on earlier studies with EMAC. Furthermore, it makes some general statements, e.g. concerning the usefulness of specific satellite data "to validate and optimize assumptions . . . in the model", of which it is unclear if they are a conclusion of some earlier work or just the opinion of the authors. No knowledge gaps are mentioned, no strategy how to tackle gaps, and how this work makes progress in *comparison to the rich body of literature on the topic. Even the reference to an earlier paper with involvement of several of the authors remains vague and it is unclear what the present study may add ("Some aspects . . . of this study have been addressed in Bingen et al. (2017)").* The introduction is expanded and clarified, examples see below in the reply to reviewer 2. Now the third sentence in the introduction is: "The aim of the present paper is to use jointly model simulations and satellite observations, taking into account the multiple spectral channels of the instruments to better understand of the spatio-temporal evolution of the stratospheric aerosol burden and the contribution of the different aerosol types to the observed dynamical aerosol patterns at the different altitudes. Most earlier studies..."

In section 5 (and also in the abstract) some "conclusions" are actually drawn, but in several cases they do not seem to be backed up thoroughly by the main body of the manuscript. In addition, there are very few statements that could be understood as knowledge gained on the atmosphere. And if they can (like "The total AOD in the visible . . . is very sensitive to aerosol water and the composition of sea salt."), they tend towards being very general and again it is unclear how potential discoveries in this study relate to earlier works. Text modified for clarity. An important conclusion is that from multiple wavelength observations the enhanced size of wet dust particles simulated by the model in the monsoon region can be detected. Please see also the revised version of abstract and conclusions in the introduction of this comment.

Most of the concluding statements relate to model tuning ("simply assuming a factor of 2 for conversion . . . is too crude"), it is unclear how general or model specific they are, and like in this case they are not well developed in the rest of the paper. The factor of 2 refered to a relation of observations at different wavelength often used. The other factor of 0.33 is for correction of model output if a too coarse resolution is selected.

Because I understand the manuscript as mostly related to model evaluation and tuning I would suggest the authors consider resubmitting it to a more model development related journal like GMD or JAMES. We don't agree, see second paragraph of introduction above. Text modified to avoid misunderstandings.

2.2 Specific comments

- Abstract: The abstract contains some statements that can be considered as conclusions, e.g. "sulfate particles from . . . volcanic eruptions dominate the interannual variability of aerosol extinction . . .". But if this is considered important enough to make it to the abstract: why doesn't it appear in the "conclusions"? And is it a new discovery? Abstract and conclusions rewritten for clarity, see above in introduction of this comment.

- P3L3: "The development work of these CDRs showed . . ." The formulation is odd. How did the work show this importance? And why does it come to this non-linearity in the averaging process? Wouldn't this depend on the way averages are built? Any reference for it? We revised the formulation of the discussion taking into account the questions posed by the Referee, and rearranged somewhat the text. We are not sure about what the Referee means by "nonlinearity", since all is mainly about how the observed patterns are visible in small bins or lost when the averaging occurs in a large volume compared to the size of the pattern (e.g.: a volcanic plume), which is just a linear problem, but we hope that our revision clarifies the text and removes all kind of misunderstanding. We also added a reference to the paper describing the whole study, in order to avoid duplication with this previous paper (Bingen et al., 2017).

- P3L25: "size and optical thickness on a 10 km grid". Shouldn't size be provided with some vertical resolution? Both optical thickness and size are retrieved as vertical column values. Size is not resolved vertically, but is represented by fraction of fine and coarse mode aerosol in the total.

- Section 3: Model Setup. It is not sufficiently clear from this section how observations are used in the simulations. Vague formulations are used in many places, like "use of

data . . . for input an" validation", "aerosol module parameters . . . were optimized on the basis of satellite data". Which aerosol parameters are actually prognostic quantities, which data are prescribed how (e.g. as boundary or initial conditions?) in the simulation process, which are used how to tune which model parameters. It is important to be very specific here, also to understand how dependent or independent the simulation results are from the data used for evaluation. (see also below) The text is improved and expanded a lot, providing more details and references, examples see below.

- P4L6: "we used different model resolution to improve the dust simulations". Sounds odd. I guess one can assume that higher resolution might improve simulations, but this is not what is said, here. The text is modified here. This points to a problem that is often ignored by modellers. New text: "In contrast to Jöckel et al. (2016), who use stratospheric aerosol extinction climatologies derived from observations, in our model setup aerosol and its optical properties are calculated from precursor gases and emissions. As dust reaching the upper troposphere/lower stratosphere region (UTLS) turned out to be sensitive to model resolution, we used different model resolutions: the T42 resolution (spectral, 2.75^o in latitude and longitude) of the previous studies, T63 resolution (1.88^o), the standard resolution for the stratosphere used in this study and T106 resolution (1.1^o) for a one year sensitivity test. The vertical grid has 90 layers from the surface up to 0.01 hPa (80 km altitude, short L90) with finest resolution in the boundary layer and near the tropopause. For T106 only simulations with the low top model version with 31 levels up to 30 km altitude (L31), the setup used by Klingmüller et al. (2018) which is well tested regarding the representation of tropospheric aerosol are discussed here in detail."

- P4L16: ". . . particle radius of 1.6 um to avoid too fast sedimentation . . ." Again an odd formulation. Would any other parameter lead to too fast sedimentation? And would this parameter be chosen differently based on observed particle size distributions? Text clarified: "The boundary between accumulation mode and coarse mode, a model

parameter, is set at a dry particle radius of 1.6 μm to avoid too fast sedimentation of a too large coarse mode fraction in case of major volcanic eruptions. For dust sensitivity studies in T106 which focus on the troposphere, also a boundary of 1.0 μm is used. The mode parameters are used for every aerosol type and listed for convenience in Table S1 of the supplement. "

- P4L25-35: ". . . superimposed to the simulated SO2 . . ."; "boundary conditions are taken from . . ."; "Marine DMS . . . is also included . . ." Again, these formulations are not specific enough. What does that mean? Are simulated fields simply updated at eruption times? For which boundaries are the observations used? How is marine DMS used? In terms of emissions? Is this important when SO2 concentrations are anyhow newly "superimposed" after every eruption? And what does all this use of observations mean for the evaluation of the simulations? This paragraph is expanded and corrected to be unique. "superimposed" is replaced by "added as volume mixing ratio". We provide references for the data and mention uncertainties from the model resolution. SO_2 from DMS is important in volcanicly quiet periods, i.e. when gaps between the eruptions exceeds about a month. For DMS we add in the text: "... model, using a module for exchange fluxes between seawater and atmosphere by Pozzer et al. (2006) and the Lana et al (2011) climatology".

- P5L5 "due to transport" Which transport is meant, here? From the troposphere to the stratosphere? Section expanded, "convective" and "gradual uplift to UTLS" added.

- Fig. 1 shows comparisons of extinction profiles for different wavelengths. However, this is not discussed in the text. What do we learn from the different comparisons? This part is expanded, providing more details on GOMOS.

- P5L16 "account for dust in a proper way, also with respect to particle size". What would be a proper way to account for the particle size of dust? And if you say also, what else? This section is rewritten, including new frames for Fig.2 which demonstrate the connection between bigger wet dust particles in the monsoon region and a relatively

larger extinction in near IR compared to the visible range. Figure 2 contains now also the contribution of wet dust and wet sulfate at 2 wavelengths individually, together with the observations and the calculated median wet radius in the accumulation mode.

- P5L17 ff: The way the "downscaling" is described here is very misleading. Only much later in the manuscript one learns that the authors have just multiplied the extinction with some factor to obtain a better comparison to observations. It is also not clear where this sensitivity to model resolution comes from. Why does the convection lead to very different transport for a relatively modest change of horizontal resolution? What are the tuning parameters? And how does the tuning of the convection parameterization for fitting sulfur transport influence other important quantities like the radiative balance? This is addressed in sections 3, 4.2, conclusions and the supplement now. A reference to the used Tiedke scheme is given. We don't tune the convection scheme, we just modify the output of the routine providing SAOD in case of the coarse resolution.

- P5L22, reference to Bingen et al. (2017). It is nice to know that other things are shown elsewhere, but it would be more important to get to know what has been learned elsewhere and what additional knowledge is provided by the figures in this paper. This was misplaced here. There is a reference to earlier work in section 4.1 now. The novel results are presented in the new Figure 2 where the effect of big wet dust particles can be identified in observed and simulated extinction in the monsoon area.

- P7L3, "our findings that desert dust is also important for the UTLS". A very vague finding. One could try to quantify it. A first step could be to look at difference plots of Fig. S1 (center and bottom panels). See above.

- P7L10, "global radiative forcing" I guess this is probably instantaneous forcing from a double radiation call? It would be important to spell this out. This is mentioned in section 3 now.

- P7L13, "Green lines and symbols show . . . observations like . . ." There are

no symbols in the figure. And what means "observations like"? Be specific. This is supposed to be a scientific paper. We have increased the size of the symbols for the annual averages. The curves and symbols for observations are taken from the references provided in the text ("like" removed).

- P7L18, "This is clearly seen in Fig. 4 . . ." How do I see in Fig. 4 which part of the AOD is transport-related? And how do I see the monsoon effect if only 20S-20N and 45N-70N are shown, not the monsoon region? Figure 4 is expanded, thanks for this important advice. To achieve this we had to process the OSIRIS data directly (and to include a coauthor) in addition to GOMOS which has not enough coverage for narrow latitude bands. With the new data the monsoon effect is clearly visible, including the problems concerning the overestimated dust with low T42 resolution of the older studies.

- Fig. 3 and 4: Legends would be nice. We improved the color scheme and the description. Unfortunately our graphics package cannot create legends in a convenient way, but if the reviewer or the editor insists on it, we have a postprocessing tool.

- Fig. 4: How is "stratospheric AOD" defined? Lower boundaries of integrals now provided in the text, dependent on latitude (sorry, somehow this was lost in the manuscript). Upper boundary is at the top of the Junge layer, i.e. about 30km.

- Caption of Fig. 4: All other caption don't include interpretation. And what means "differ mostly"? This is obsolete now.

- P9L6: "a clear signal from biomass burning organic aerosol". It's not clear to me from just looking at the figure. Please explain why this is clear. In the figure are spikes in the lower stratosphere in September, correlated with maxima in the mixing ratios of absorbing organic aerosol. A figure on that will be in the supplement.

- P11L10ff: There is no motivation provided for the change to resolution T106. How would the problem of tuning convection mentioned for T42 vs. T63 affect T106? We

don't tune convection. Resolution T106L31 was the standard setup for tropospheric aerosol modelling in earlier studies (indicated now in section 3). We like to show here that this setup and the stratospheric setup T63L90 are consistent concerning the comparison with IASI and ATSR.

- P13L8, "additional (ongoing) simulations". What do I make of the "ongoing"? Either the simulations are ready to be used for scientific interpretation or not. Removed. The results in the figures are replaced by the ones of the completed "ongoing" simulation, see also introduction of this comment.

3 Reviewer 2, questions and answers

3.1 General comments

- The introduction is too short and does not put this study in the context of existing literature. Similar papers on the subject shall be cited and related to the work presented in the manuscript. The introduction is expanded and misleading words (e.g. in line 6, page 2) were replaced. In line 23, page 1 two more references are cited, in line 26 the following is inserted: "Most earlier studies focus on the effects of major volcanic eruptions like Pinatubo (e.g. Aquila et al., 2014, English et al., 2013). For the post Pinatubo-period with only medium size eruptions Mills et al (2016, 2017) present simulations with the chemistry climate model WACCM (Whole Atmosphere Community Model) with interactive aerosol using estimates for volcanic injections mostly from nadir sounders. That and the present ....".

- It is also not immediately clear what are the novelty aspect of this study and what is its added value to the current knowledge in the field. It should be stated that the paper focuses on the evaluation of the EMAC model in different configurations, specifically on the aspects controlling the optical and radiative properties of stratospheric aerosol.

Please see revised abstract and conclusions in the introduction of this comment. Model evaluation is only one aspect. Novel is e.g. the clear indication of the presence of dust in the LS from multiwavelength observations and model simulations (abstract and conclusions).

- The description of model setup is also not very exhaustive: it is not clear for example how the SO2 plumes from the data are used in the model (last paragraph of Sect. 3). It is also not mentioned which time period is covered by the simulations (although this can be found out later in the figures) and whether a nudging technique has been used. Concerning nudging, we include in line 10 of section 3: "In all simulations, except the T42L90 one of the previous studies, the meteorology below about the 100 hPa level is nudged to the reanalysis ERA-Interim." The part on SO_2 injections is considerably expanded providing references (see reply to reviewer 1).

- As far as I could understand, the setup has been derived from one of the simulations in Jöckel et al. (2016). If this is the case, I would recommend to write that more explicitely at the beginning of Sect. 3. The rest of the section could then discuss just the differences and the additional features considered for the present study. The choice of particular configuration settings shall also be motivated in view of the analysis which is performed. Summarizing all the performed experiments and the relevant parameters in a table would be helpful. The most important difference to Jöckel et al. (2016) is now in second sentence of section 3: "In contrast to Jöckel et al. (2016), who use stratospheric aerosol extinction climatologies derived from observations, in our model setup aerosol and its optical properties are calculated from precursor gases and emissions." The definitions and names of the simulations are clarified.

- The results section is confusing: I miss the connection between Sect 4.1-4.2 (which are quite short) and the rest of the section, which is much more clear and goes into the details of the comparison for the different model configurations and possible reasons for deviations. I would suggest to revise Sect. 4, trying to set a common thread through the whole section. Sections 4.1 and 4.2 are expanded and related to section 4.3 with

the most important results. Section 4.3 contains now additional satellite data to support the findings that dust transported by the monsoon can be important.

- The downscaling of dust in EMAC is mentioned in the result section and in the conclusion, but it is not discussed in detail. It seems to be an important issue and shall be discussed in Sect. 3. The resolution dependent scaling is mentioned in detail for the volcanic SO_2 sink, an additional problem we detected after submission of the first version of the manuscript, in section 3. In section 4.2 and the supplement details scaling of stratospheric dust extinction for low resolution are given.

- Another interesting point which is mentioned in the conclusions but not discussed in sufficient detail is sea salt composition in the aerosol model and how it can be "tuned" using satellite data. We now provide text and references in section 3 and a table in the supplement.

3.2 Specific comments

There are several sentences which are hard to interpret and more precise statements are sometimes desirable. See detailed suggestions in the following:

P1.L20: this sentence is unclear: a consistent representation of tropospheric and stratospheric aerosol in the model and the good agreement with observations are two different things. You could have a model which represents both domains consistently but compares badly with the observations, and the other way round. I would suggest rephrasing this, stressing that you have a consistent model in terms of aerosol representation (this is a plus) AND that the results reproduce satellite observations well (this is another plus). Sentence changed, see revised abstract in introduction of this comment.

P1.L24: you should also explain why radiation is reduced (scattering processes?). Done.

P1.L26: please summarize what are the scope and the goals of this initiative and add a reference, if available. Timmreck et al. (2018) and Kremser et al. (2016) references given.

P1.L31: I would rephrase this as: "like the EMAC model (Brühl et al., 2015)". Done.

P2.L17: please provide the exact wavelength range. Done.

P2.L33: do you mean that the extinction in cloud-free fraction is attributed to sulfate aerosol? If yes, please rephrase and make it more explicit. We thank the referee for this pertinent question. We clarify earlier in the text the way to retrieve the extinction and changed the expression "total aerosol extinction" in the more adequate expression "total extinction from non-gaseous species". We also explain now how the type of particulate matter is inferred, which makes possible the derivation of separate CDRs for the total extinction, the aerosol fraction, and the polar stratospheric cloud fraction. It should be noted that the extinction by particulate matter is retrieved using a parameterization that doesn't require any knowledge of the aerosol type. So, the statement concerning the sulfate aerosols simply refers to the well-known fact that sulfate aerosols is the most common aerosol type observed in the stratosphere, but we do not claim bringing any new information on the aerosol type. Hence the use of the qualification "in good approximation": at this stage, we cannot infer more precisely the nature of the aerosol particles. A warning is also added about the limitations of the criteria used for the cloud detection, and the risk for cloud contamination in the aerosol CDR. Please note that all these aspects are detailed in the different references cited in the text, so that we don't think that more detail is needed on the way the retrieval algorithm is implemented.

P3.L30: I would mention that AERONET is recognized as the reference dataset for validating satellite products and cite Holben et al. (1998) Done.

P4.L8: please identify the vertical resolutions with a number (L90, L31), that you can refer to in the rest of the paper. Done.

P4.L16: is this the wet or the dry radius? "dry" added here in text.

P4.L18: how are the optical properties calculated? Please provide more details. Done, added in text: "...calculated using Mie-theory-based lookup tables consistent with the selected size distribution widths of the modes. The resulting optical depths, single scattering albedos and asymmetry-factors are used in radiative transfer calculations...."

P4.L20: given their relevant role in this study, more details on the dust emission parametrization should be given here. We have expanded the text to "The mineral dust emissions are calculated online using the emission scheme of Astitha et al. (2012) which builds on previous studies by Pérez et al. (2006), Spyrou et al. (2010), Laurent et al. (2008, 2010), Marticorena et al. (1997), Zender et al. (2003) and Tegen (2002). The emission scheme parameterizes saltation bombardment and aggregate disintegration by sand blasting combining the surface friction velocity with descriptions of land cover type, clay fraction of the soil and vegetation cover. For an improved representation of dust at higher resolution, we adopted the updates presented by Klingmüller et al. (2018) in the T106 L31 simulation."

P4.L30: does this generate any inconsistency/discontinuity in the emissions at 200 hPa? Please clarify. This effect is secondary and in most cases smeared out by transport. I have added "monthly" in the text. The Diehl climatology is mostly for volcanoes outgassing over long periods, for explosive events it introduces local artifacs in the upper troposphere which have negligible effects in the LS.

P4.L31: please provide references for the various emission inventories mentioned in this paragraph. The detailed description can be found in Jöckel et al 2016. This covers several pages including references and is not the main focus of this study. "CCMI" is added in the text.

P5.L18: it is not clear what has been downscaled here and why. This section is rewritten using the results of the T63L90 simulation. Results for T42L90, the resolution of earlier studies, are shown in the supplement and mentioned at the end of the section.

P5.L22: please add in which Figure of Bingen et al. this is shown. Done at end of section 4.1. In section 4.2 this is skipped.

P12.L16: which dust size distributions parameter are adopted in the model? The dust size distribution is calculated online resulting from the physical and chemical processes acting on the aerosol modes. Fixed parameters are the widths of the log-normal modes and the dry radii separating the modes. The size distribution can be adjusted by modifying these parameters, but also by modifying parameters of relevant processes such as emission, deposition, coagulation and hygroscopic growth. For clarification, we have added the following text to line 18 on page 12: "This could involve modifying the parameters of the log-normal modes, i.e., their widths and boundaries, but also reassessing the parametrisation of relevant processes such as emission, deposition, coagulation and hygroscopic growth, or even adding an extra mode for extremely coarse particles which can be relevant close to dust sources." Furthermore, we refer to a table in the supplement with the mode parameters in section 3.

P13.L8: horizontal or vertical resolution? What is the expected outcome of these simulations? Are further publications planned? Please elaborate more on this sentence. This sentence was premature. I had estimated the typical results for the whole timeseries from an ongoing incomplete simulation.

P13.L16: where does this conversion factor come from? It looks like an important issue, but it is mentioned for the first time in the conclusions. This issue is now addressed also in section 4.4. The factor is often used in the AEROCOM community but not physically based.

3.3 Technical corrections:

P1.L9: "EMAC" acronym is not defined at the first occurrence. In the abstract this should be considered as a name because the full expression of this acronym of acronyms given in section 3 is too long (3 lines).

P1.L10: "such as" instead of "like" (you intend inclusion, not comparison). Done.

P1.L13: "the observations". Rewritten.

P2.L6: add "The present paper is organized as follows:" or similar. Done, and misleading words replaced.

Fig.4: red and purple are very hard to distinguish, please consider a different color (or dash pattern). Color scheme now as in Fig. 3.

P12.L24: I would simply write "at T106L31 resolution" and use this notation consistently through the paper. Done.

4 Additional references

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Stratospheric aerosol radiative forcing simulated by the chemistry climate model EMAC using aerosol CCI satellite data

Christoph Brühl[1], Jennifer Schallock[1], Klaus Klingmüller[1], Charles Robert[2], Christine Bingen[2], Lieven Clarisse[3], Andreas Heckel[4], Peter North[4], Landon Rieger[5]

[1]Max Planck Institute for Chemistry, Mainz, Germany; [2]BIRA Brussels, Belgium; [3]ULB Brussels, Belgium; [4]Swansea University, UK; [5] University of Saskatchewan, Canada

Correspondence to: Christoph Brühl (christoph.bruehl@mpic.de)

Abstract. This paper presents decadal simulations of stratospheric and tropospheric aerosol and its radiative effects by the chemistry general circulation model EMAC constrained with satellite observations in the framework of the ESA-Aerosol-CCI project ~~like~~such as GOMOS (Global Ozone Monitoring by Occultation of Stars) and (A)ATSR ((Advanced) Along Track Scanning Radiometer) on the ENVISAT (European Environmental Satellite), ~~and~~ IASI (Infrared Atmospheric Sounding Interferometer) on ~~Metop~~MetOp (Meteorological Operational Satellite~~).~~), and, additionally, OSIRIS (Optical Spectrograph and InfraRed Imaging System). In contrast to most other studies, the extinctions and optical depths from the model are compared to the observations at the original wavelengths of the satellite instruments covering the range from UV (ultraviolet) to terrestrial IR (infrared). This avoids conversion artifacts and provides additional constraints for model aerosol and interpretation of the observations.

~~The EMAC simulations with modal interactive aerosol and observations by GOMOS show that sulfate particles from about 230 volcanic eruptions identified mostly from~~ MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) SO_2 limb measurements ~~dominate the~~are used to identify plumes of more than 200 volcanic eruptions. These three-dimensional SO_2 plumes are added to the model SO_2 at the eruption times. The interannual variability of aerosol extinction in the lower stratosphere, and of stratospheric aerosol radiative forcing at the tropopause~~.~~ is dominated by the volcanoes. To explain the seasonal cycle of the GOMOS and OSIRIS observations, desert dust ~~and organic and black carbon,~~simulated by a new approach and transported to the lowermost stratosphere by the Asian summer monsoon and tropical convection~~, are~~ turns out to be essential. This applies also ~~important. This holds also for~~to the radiative heating by aerosol in the lowermost stratosphere. ~~Comparison~~The existence of wet dust aerosol in the lowermost stratosphere is indicated by the patterns of the wavelength dependence of extinction in observations and simulations. Additional comparison with (A)ATSR total aerosol optical depth at different wavelengths and IASI dust optical depth ~~shows~~demonstrates that the model is able to represent stratospheric ~~and~~as well as tropospheric aerosol ~~in a consistent way.~~consistently.

1. Introduction

Climate effects of stratospheric aerosols can be important, as analyzed for example by Solomon et al. (2011~~).~~), Santer et al. (2014) and Ridley et al. (2014). Stratospheric aerosol exerts a negative radiative forcing on the troposphere because ~~it~~enhanced scattering by the particles reduces solar radiation reaching the surface and the lower atmosphere. In addition, changes in diffuse light fraction have shown their potential to enhance photosynthesis (Gu et al., 2003). ~~The present study contributes~~The aim of the present paper is to use jointly model simulations and satellite observations, taking into account the multiple spectral channels of the instruments to better understand the spatio-temporal evolution of the stratospheric aerosol burden and the contribution of the different aerosol types to the observed dynamical aerosol patterns at the different altitudes. Most earlier studies focus on the effects of major volcanic eruptions like Pinatubo (e.g. Aquila et al., 2014, English et al., 2013). For the post Pinatubo-period with only medium size eruptions Mills et al. (2016, 2017) present simulations with the chemistry climate model WACCM (Whole Atmosphere Community Model) with interactive aerosol, using estimates for volcanic injections mostly from nadir sounders. That and the present study contribute to the SPARC/SSIRC initiative (Stratosphere-troposphere Processes And their Role in Climate/ Stratospheric Sulfur and Its Role in Climate~~). The~~, see for example Timmreck et al. 2018), aiming at a better understanding of the composition, microphysical and radiative properties characteristics of stratospheric aerosols and their impact on climate (Kremser et al., 2016). In this work, we rely on the multiple instrument satellite dataset provided in the Climate Change Initiative (CCI) of the European Space Agency (ESA) (Popp et al., ~~2016) is very valuable~~2016), which was developed as tool for evaluation and improvement of the treatment of stratospheric and tropospheric aerosols in global chemistry climate models, like ~~for example the one used by Brühl et al. (2015),~~the EMAC (ECHAM5/MESSy Atmospheric Chemistry) model (Brühl et al., 2015). The datasets providing extinctions or total optical depth at wavelengths from ultraviolet (UV) to terrestrial infra-red (IR) are very useful to validate and optimize assumptions on the size distribution and on the composition of aerosol in the model, but also on aerosol sources. Some aspects of the stratospheric part of this study ~~have been addressed in~~follow up Bingen et al. (2017). The ATSR and IASI datasets provide additional constraints on tropospheric aerosol, especially desert dust~~, which~~. We find in the present study that this latter aerosol compound can penetrate the tropopause via the Asian Summer Monsoon system and, to a smaller extent, via tropical convection.

The present paper is organized as follows: In Section 2, we briefly present the satellite datasets used to ~~feed~~evaluate the model, and to check for consistency of observations at different wavelengths: GOMOS, IASI~~ and~~, (A)ATSR~~. Further, iin~~ and OSIRIS. In Section 3 we ~~briefly~~ describe the EMAC model and the various versions and resolutions used in our work~~.~~, including the use of MIPAS SO_2 for input. In Section 4, we ~~analyze~~study the impact of the main aerosol sources on the upper tropospheric and lower stratospheric aerosol burden. The influence of volcanic sources derived from satellite data, but also of dust and organic aerosols is ~~described~~analyzed. We present examples on the constraints by satellite observations in different spectral regions on different aerosol types with respect to particle size and composition. ~~Finally, we~~We discuss the evolution of the optical depth and radiative forcing by stratospheric aerosols, including uncertainties introduced be horizontal model resolution.

Finally, we show that the findings concerning the importance of dust for the lower stratosphere are consistent with observations and simulations of tropospheric aerosol. Conclusions are drawn in Section 5.

2. Satellite data products from Aerosol_cci II

2.1. GOMOS (Global Ozone Monitoring by Occultation of Stars)

GOMOS is an instrument based on the stellar occultation technique (Bertaux et al., 2010) and provides atmospheric measurements in the UV-Visible~~ range. ~~IR range (248-690 nm, 755-774 nm, and 926-954 nm). The use of stellar occultation results in a high rate of occultation measurements, and consequently, a very good spatial coverage compared to solar occultation. As a drawback, the signal-to-noise ratio of each measurement is much lower than in the solar case, and varies with the star characteristics (especially its magnitude and its temperature). The operational retrieval, IPF, provides density profiles for trace gases such as ~~O_3 (~~ozone~~), NO_2 (~~ (O_3), nitrogen dioxide (NO_2), and ~~NO_3 (~~nitrogen trioxide (NO_3) (Kyrola et al., 2010), as well as aerosol extinction. However, the extinction shows a poor quality out of the reference wavelength at 500 nm. For this reason an alternative inverse algorithm called AerGOM was developed specifically to optimize the aerosol retrieval (Vanhellemont et al., 2016; Robert et al., 2016). AerGOM provides ~~as primary aerosol quantity~~vertical profiles of the same gas species, and the total extinction coefficient for the non-gaseous species and its spectral dependence, currently over the range 250-750~~-~~ nm. The nature of the total extinction fraction for non-gaseous species is then inferred using simple criteria based on the geolocation, associated temperature value and extinction value, and each point of the vertical extinction profile is attributed to aerosols, cirrus clouds, polar stratospheric clouds or meteoritic dust.

From the AerGOM extinction, climate data records (CDRs) were developed in the framework of the ESA Aerosol CCI project for different quantities including the aerosol extinction and the related aerosol optical depth at several wavelengths (355, 440, 470, 550 and 750 nm, Bingen et al., 2017). A particular attention was paid to the grid choice, which should optimally render the information contained in the GOMOS measurement set. The most important conclusions of this optimization were that grid resolution should be chosen to ensure a reasonable statistical sampling in most of the grid cells, and that it should optimally reflect the typical transport of volcanic plumes after an eruption reaching the upper troposphere or the lower stratosphere (UTLS). Therefore, the grid should represent in a coherent way the longitudinal and latitudinal air mass transport, and the time needed for this transport. Also, the temporal resolution should be short enough to enable the detection of volcanic signatures, taking into account the typical lifetime of the plume. In this respect, we could verify that time intervals of about 5 days are able to represent the signature of most of the eruptions injecting sulfuric gases in the UTLS, while such signature is often diluted, underestimated, or even disappears in the case of coarser grid cells. This is the case, for instance, for monthly zonal means, even though this representation is very commonly used in the field. The ability of the grid to reproduce the signature of volcanic plume in a satisfactory way is of particularly great importance when the CDRs are used to constrain climate models. More detail about the investigations of the optimal grid choice and all other aspects of the implementation of the CDRs can be found in (Bingen et al, 2017).

In their current version (version 3.0), these CDRs are defined on a grid with a resolution of 5° in latitude, 60° in longitude, 1 km in altitude, and 5-day time period. The records cover the whole ENVISAT period (March 2002 - April 2012) and include the total ~~aerosol~~ extinction of non-gaseous species, but also the polar stratospheric cloud (PSC) fraction and the ~~non-~~cloud-free aerosol[SJ1] fraction which ~~can be attributed, in good approximation, to~~ is dominated by sulfate aerosols below an altitude of ~~35~~32 km. It is important to mention that cloud detection is not yet optimal, and that cloud contamination of the aerosol fraction is possible in the UTLS region. This issue is still under investigation.

~~The grid resolution was chosen to ensure a reasonable statistical sampling in most of the grid cells, and to optimally reflect the typical transport of volcanic plumes after an eruption reaching the upper troposphere or the stratosphere, by providing a good coherence between longitudinal and latitudinal air mass transports, and a temporal resolution enabling the detection of volcanic signatures. The development work of these CDRs showed the importance of such a high temporal resolution to derive a realistic estimate of the impact of all volcanic eruptions on the stratospheric aerosol burden: when large temporal intervals are used (such as in the very common case of monthly means), the aerosol signature is diluted in the averaging process of the gridded extinction value or even gets lost, and the contribution of volcanic aerosols to the gridded extinction is underestimated. This effect has a significant impact when the CDRs are used to constrain climate models.~~

2.2. IASI (Infrared Atmospheric Sounding Interferometer)

The IASI dust dataset of the Université Libre de Bruxelles (ULB) was generated in the context of ESA CCI's project (Popp et al., 2016). It is based on a statistical regression technique and the use of a neural network trained on synthetic IASI data. A similar scheme has already been applied for the retrieval of NH_3 (ammonia) (Whitburn et al., 2016 and Van Damme et al., 2017). As input variables it uses the IASI L2 pressure, humidity and temperature information, spectral information and a CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) derived dust altitude climatology. The main output variables are dust optical depth at 10 and 11~~-~~ µm (and 550 nm). Initial results and validation performance are provided in (Popp et al., 2016).

2.3. (A)ATSR ((Advanced) Along Track Scanning Radiometer)

The ATSR (SU) algorithm has been developed at Swansea University for estimation of atmospheric aerosol and surface reflectance for the ATSR-2, AATSR sensors, and SLSTR (Sea and Land Surface Temperature Radiometer) on Sentinel-3. Over land, the algorithm employs a parameterized model of the surface angular anisotropy (North, 2002), and uses the dual-view capability of the instrument to allow aerosol property estimation without a priori assumptions on surface spectral reflectance. Over ocean, the algorithm uses a simple a priori model of ocean surface reflectance at both nadir and along-track view angles. A climatology (Kinne et al., 2006) is used to constrain chemical composition of the aerosol components at 1° x 1° latitude-longitude grid, while the method retrieves aerosol size and optical thickness on a 10 km grid. Both optical thickness and size are retrieved as vertical column values. Size is not resolved vertically, but is represented by fraction of fine and coarse mode aerosol in the total. The algorithm has been developed from initial prototype (Bevan et al., 2012) under the Aerosol CCI program, and results and validation performance for version 4.21 are provided in Popp et al. (2016). The version used here (V4.3) differs from that summarized in Popp et al. (2016) by improvements in retrieval of coarse/fine mode fraction, and improved cloud screening over ocean in the region of dense plumes, resulting in approximately 10% greater coverage, with small improvement in correlation against AERONET (AErosol RObotic NETwork) values. AERONET is recognized as reference dataset for validation of satellite data products (Holben et al., 1998).

2.4. OSIRIS (Optical Spectrograph and InfraRed Imager), external

OSIRIS was launched on board the Odin satellite, and has provided vertical profiles of limb scattered radiance between 280 and 810 nm since 2001 (Llewellyn et al., 2004). The radiance profiles are inverted to provide aerosol extinction measurements at 750 nm at altitudes between 10 and 35 km with a vertical resolution of approximately 2 km (Bourassa et al., 2012). This technique provides high sampling rates with hundreds of measurements per day over the sunlit portion of the globe, enabling excellent spatial and temporal sampling of short-lived events. OSIRIS aerosol extinction retrievals agree well with coincident occultation measurements from Stratospheric Aerosol and Gas Experiments II and III during background periods but have known low biases above approximately 25 km, and will have some cloud contamination near and below the tropopause (Bourassa et al., 2012; Rieger et al., 2015). Additionally, seasonal biases are possible due to the orbital geometry and changes in aerosol optical properties such as after volcanic eruptions may also bias the retrievals. These effects are described in more detail by Rieger et al., (2014, 2018). This work uses the OSIRIS version 5.10 aerosol retrieval (Bourassa et al., 2018) averaged into daily, 5° latitude by 30° longitude bins for comparisons.

3. Model Setup

For the simulations of the radiative and chemical effects of stratospheric aerosol, the ECHAM5 (5th generation of European Centre Hamburg general circulation model) general circulation model coupled to the Modular Earth Submodel System Atmospheric Chemistry (EMAC) was used (Brühl et al., 2015, updated to the version of Jöckel et al., 2010). In contrast to Jöckel et al. (2016), who use stratospheric aerosol extinction climatologies derived from observations, in our model setup aerosol and its optical properties are calculated from precursor gases and emissions. As dust reaching the upper troposphere/lower stratosphere region (UTLS) ~~is~~turned out to be sensitive to model resolution, we used different model resolutions~~ to improve the dust simulations: the standard ~~: the T42 resolution (spectral, 2.75° in latitude and longitude~~),~~) of the previous studies, T63 resolution (1.88~~°)~~°), the standard resolution for the stratosphere used in this study and T106 resolution (1.1~~°).~~°) for a one year sensitivity test. The vertical grid has 90 layers from the surface up to 0.01 hPa (~80 km altitude, short L90) with finest resolution in the boundary layer and near the tropopause. For T106 only ~~test ~~simulations with the low top model version with 31 levels up to 30 km altitude (L31), the setup used by Klingmüller et al. (2018), which is well tested regarding the representation of tropospheric aerosol, are discussed here~~.~~ in detail. In all simulations, except the T42L90 one of the previous studies, the meteorology below about the 100 hPa level is nudged to the reanalysis ERA-Interim (Jöckel et al, 2006). The simulations were performed for the ENVISAT time period from July 2002 to March 2012 to allow for the use of data from MIPAS~~, GOMOS~~ for input, and GOMOS and ATSR for ~~input and~~ validation. The period from 1997 to 2002 using SAGE~~-~~ II (Stratospheric Aerosol and Gas Experiment) was simulated first to get consistent initial conditions.

The applied aerosol module GMXE (Pringle et al., 2010) accounts for seven modes using lognormal size distributions (nucleation mode, soluble and insoluble Aitken, accumulation and coarse modes). The boundary between accumulation mode and coarse mode, a model parameter, is set at a dry particle radius of 1.6 µm to avoid too fast sedimentation of a too large coarse mode fraction in case of major volcanic eruptions. For dust sensitivity studies in T106 which focus on the troposphere, also a boundary of 1.0 µm is used. The mode parameters are used for every aerosol type and listed for convenience in Table S1 of the supplement. Optical properties for the types sulfate, dust, organic and black carbon (OC and BC), sea salt, and aerosol water are calculated ~~and~~using Mie-theory-based lookup tables consistent with the selected size distribution widths of the modes. The resulting optical depths, single scattering albedos and asymmetry-factors are used in radiative transfer calculations which (except for the T106 low top sensitivity studies) feedback to atmospheric dynamics. ~~Desert dust simulations are based on the emission schemes by Astitha et al. (2012) and Klingmüller et al. (2018).~~ The contribution of stratospheric aerosol to (instantaneous) radiative forcing and heating is calculated online via multiple calls of the radiation module.

The mineral dust emissions are calculated online using the emission scheme of Astitha et al. (2012) which builds on previous studies by Pérez et al. (2006), Spyrou et al. (2010), Laurent et al. (2008, 2010), Marticorena et al. (1997), Zender et al. (2003) and Tegen (2002). The emission scheme parameterizes saltation bombardment and aggregate disintegration by sand blasting, combining the surface friction velocity with descriptions of land cover type, clay fraction of the soil and vegetation cover. For an improved representation of dust at higher resolution, we adopted the updates presented by Klingmüller et al. (2018) in the T106L31 simulation.

Aerosol module parameters, like for example, the composition of sea salt, were optimized on the basis of the satellite data**.** We apply the chemical speciation of the sea salt emission flux used by Abdelkader et al. (2015) as listed in Table S2 of the supplement. The sea salt composition affects the hygroscopic growth and thereby the AOD. The setting of Jöckel et al. (2016), dominated by Na and Cl ions, which we initially applied in our simulations produced very high AOD levels over the North Pacific which are not consistent with the satellite observations.

SO_2 plumes (sulfur dioxide) from about 230 explosive volcanic eruptions into the stratosphere were derived from 3-dimensional data fields of MIPAS (Höpfner et al., 2015) and, in case of data gaps, of GOMOS on ENVISAT with a temporal resolution of 5 days, and ~~superimposed~~added as volume mixing ratio to the simulated SO_2 at the time of the eruption. Each identified volcanic eruption (with names from the Smithsonian volcanic database, www.volcano.si.edu) is listed in an emission inventory published recently (Bingen et al., 2017), which provides an estimate of the altitude and the amount of SO_2 injected into the atmosphere. The table and the 3-D-fields of volcanic SO_2 are available at https://doi.org/10.1594/WDCC/SSIRC_1. These data were derived from MIPAS within the uncertainty range but more near the upper end for best results with the model resolution T42L90 and free running mode, which has some artifacts from the convection scheme and a dry bias at the tropical tropopause. For the nudged T63L90 simulation the volcanic SO_2 data of the inventory have to be downscaled by about a factor of 0.7 which is actually closer to the most likely MIPAS measurements. The actual values for each injection, which depend on the time span between the eruptions and on corrections for data gaps, are given in the supplement (Table S3). Boundary conditions for background concentrations of SO_2 from outgassing volcanoes into the troposphere are taken from the monthly climatology of Diehl et al. (2012) truncated at 200~~-~~ hPa to avoid double counting~~.~~ in the stratosphere. The sulfur source gas OCS (carbonyl sulfide) is constrained by observed monthly zonal average surface volume mixing ratios (update of the data by Montzka et al., 2007). Marine DMS (dimethyl sulfide) as natural sulfur source is also included in the model~~.~~, using a module for exchange fluxes between seawater and atmosphere by Pozzer et al. (2006) and the Lana et al. (2011) climatology. For anthropogenic emissions of CO (carbon monoxide), NOx (nitrogen oxides), sulfur, OC, and BC the DLR-MACCity emission inventory is used. Biomass burning is based on ACCMIP-MACCity and GFEDv2, OC-SOA (secondary organic aerosol) on AEROCOM_UMZ1. For details on ~~the~~these emission inventories selected for the Chemistry Climate Model Initiative (CCMI) see Jöckel et al. (2016).

4. Stratospheric Aerosol and its radiative effect

4.1. Volcanic eruptions

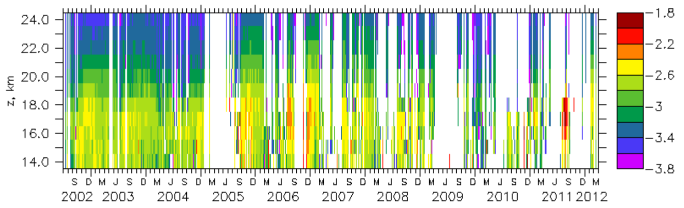
Volcanic emissions have a large impact on the stratospheric aerosol burden. Even small and moderate eruptions contribute to the stratospheric aerosol load due to ~~transport and~~convective transport of SO_2 and its gradual uplift to the upper troposphere and the lower stratosphere, and resulting accumulation of sulfate aerosol. Volcanic SO_2 injections explain most of the interannual variability of stratospheric aerosol extinction (decadal logarithm) observed by GOMOS~~ at 3 wavelengths, as depicted in Fig. 1 for the altitude dependence in the tropics. Comparisons for the latitude dependence in the lower stratosphere are shown in Bingen et al. (2017) for 550 nm (b), the wavelength, where the GOMOS data quality is best.~~, as depicted in Fig. 1 at three wavelengths. For each wavelength (350 nm on Fig. 1a, 550 nm on Fig. 1b, and 750 nm on Fig. 1c, respectively), the GOMOS time series (upper panels) showing the altitude dependence in the tropics, is compared with the EMAC simulation in resolution T63L90 including the dust contribution (lower panel; see Section 4.2 for more detail). Fig. 1 shows, at all three wavelengths, an enhancement of the extinction value is observed around 16-18 km, corresponding to the aerosol load resulting from a succession of volcanic eruptions during the whole period 2002-2012. The eruptions of Nabro in June 2011 and the successive eruptions of Soufriere Hills and Rabaul in 2006 have the largest effects on extinction in the lower stratosphere in the observations and the simulation. The best agreement between GOMOS and EMAC is found in the case of the extinction at 550 nm (Fig. 1b), where the quality of the GOMOS retrieval is the best. At 750 nm (Fig. 1c) also, GOMOS measurements agree well with EMAC for the aerosol layer (16-22 km) where measured extinction values exceed ~2 10^{-4} km^{-1}. At lower altitudes (14-16 km), rather unstructured patterns of enhanced extinction are found by GOMOS, probably corresponding to cloud contamination. At 350 nm, where a decrease of the GOMOS quality is expected due to a loss in signal-to-noise ratio obtained in the UV spectral region while using cold stars, still the volcanic events stick out. More details over these aspects can be found in references (Robert et al., 2016; Bingen et al., 2017). Bingen et al. (2017) present also the latitude dependence of 550 nm-aerosol extinction at 17 km altitude as observed by GOMOS and simulated by EMAC in the coarse resolution T42L90 in their figure 10.

4.2. Dust and organics from the troposphere in the upper troposphere/lower stratosphere (UTLS)

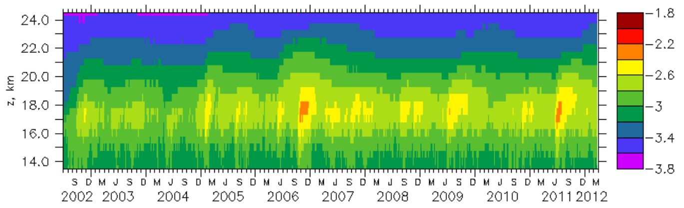
Extinction in the lowermost stratosphere and upper troposphere is ~~mostly~~to a large fraction due to desert dust and organic carbon aerosol. These contributions were strongly underestimated in Brühl et al. (2015) due to a crude parameterization in the used model version based on Jöckel et al. ~~(2006). To reproduce the GOMOS observations of extinction at the 3 different wavelengths it is essential to account for dust in a proper way, also with respect to particle size.~~(2006), but overestimated in Bingen et al. (2017). Both simulations were performed in the relatively coarse resolution T42L90. Dust reaching the UTLS is sensitive to model resolution, mostly via the convection parameterization~~.~~ (Tiedke, 1989). In Fig. 1 the simulated extinction ~~with the dust contribution~~ in ~~T42 downscaled to the values in T63~~ resolution ~~(i.e. with a scaling factor of 1/3)~~T63L90 fits ~~best~~well to the GOMOS observations~~. Fig.~~ which appear to have a seasonal contribution from the Asian summer monsoon. For more detailed analysis Fig. 2 shows observed and simulated extinction in the Asian sector at 17 km~~.~~ in the visible and the near IR. The largest extinction values are found indeed at the location and time of the Asian summer monsoon~~.~~ at the altitude of outflow. This feature is clearest in years not perturbed by medium strength volcanic eruptions, like for example 2010. ~~The corresponding zonal average extinction is illustrated in Bingen et al. (2017). Results with the full, not downscaled dust (overestimate) and without dust at all and without organic carbon (underestimate) are shown in the supplement.~~For a clear separation the contributions of wet dust and wet sulfate to extinction are displayed separately (Fig. 2cd). The wet dust particles in the monsoon region have a larger median wet radius than the volcanic sulfate particles (e.g. from Sarychev in 2009, Fig. 2e) which is consistent with a relatively larger extinction in the infrared compared to the visible in the monsoon region in observations and simulations. Fig. 2ab demonstrates that dust is essential to reproduce the observations. Total extinction without wet dust in T63L90 is shown in the supplement. Comparing Fig. S1b with Fig. 2c (lower frame) shows a small contribution of organics from biomass burning in northern spring (for volume mixing ratios see Fig. S2). Fig. S1 contains also results from the T42L90 simulation of Bingen et al. (2017), showing that there the contribution of wet dust to extinction has to be downscaled (i.e. devided) by a factor of 2 to get agreement (Fig. S1d, factor of 3 if only dry dust is considered).

Observations by IASI and ATSR indicate a maximum in dust aerosol optical depth (DAOD) in early northern hemispheric summer over the Asian deserts located in the inflow regions of the monsoon (see section 4.4). A similar feature is found in the simulations by EMAC. This supports our findings that desert dust is also important for the UTLS.

~~a)~~

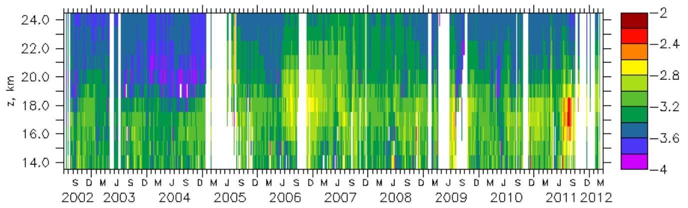


1/km, 350nm, 20°S−20°N, GOMOS 3.00

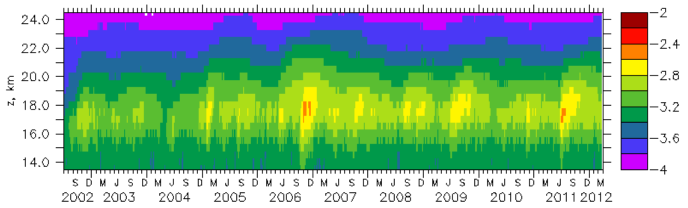


1/km, 350nm, 20°S−20°N, EMAC,V2,0.33DU

~~b)~~

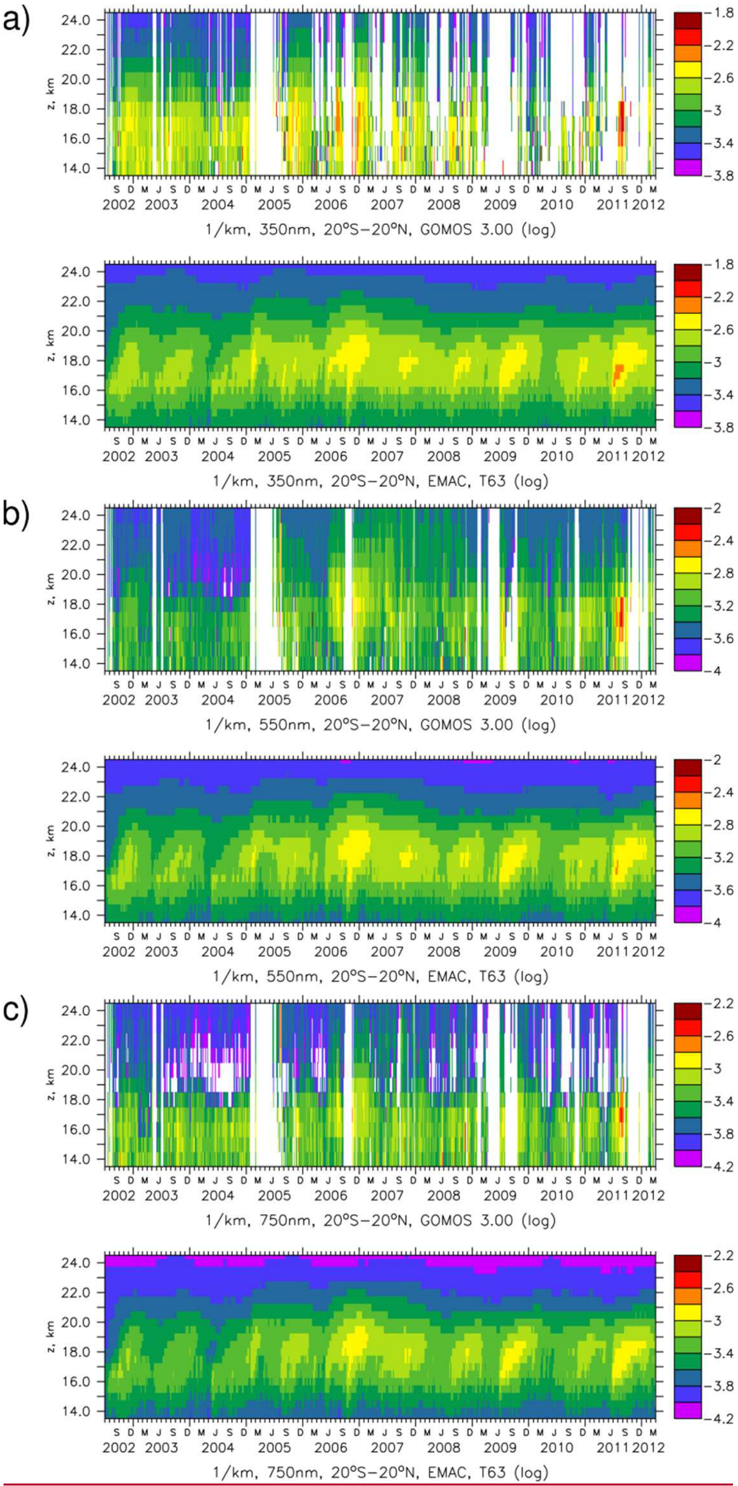


1/km, 550nm, 20°S−20°N, GOMOS 3.00



1/km, 550nm, 20°S−20°N, EMAC,V2,0.33DU

~~c)~~


a)
b)
c)
z, km
24.0
22.0
20.0
18.0
16.0
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S D M J S D M J S D M J S D M J S D M J S D M J S D M J S D M J S D M J S D M J S D M
2002 2003 2004 2005 2006 2007 2008 2009 2010 2011 2012
1/km, 350nm, 20°S−20°N, GOMOS 3.00 (log)
1/km, 350nm, 20°S−20°N, EMAC, T63 (log)
−1.8 −2.2 −2.6 −3 −3.4 −3.8
1/km, 550nm, 20°S−20°N, GOMOS 3.00 (log)
1/km, 550nm, 20°S−20°N, EMAC, T63 (log)
−2 −2.4 −2.8 −3.2 −3.6 −4
1/km, 750nm, 20°S−20°N, GOMOS 3.00 (log)
1/km, 750nm, 20°S−20°N, EMAC, T63 (log)
−2.2 −2.6 −3 −3.4 −3.8 −4.2

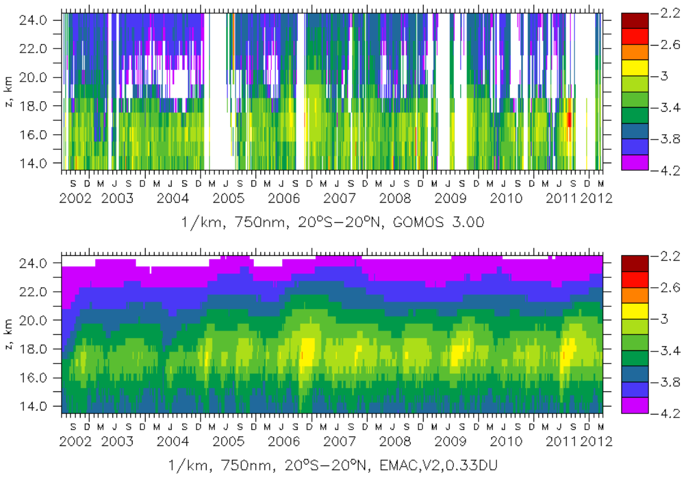
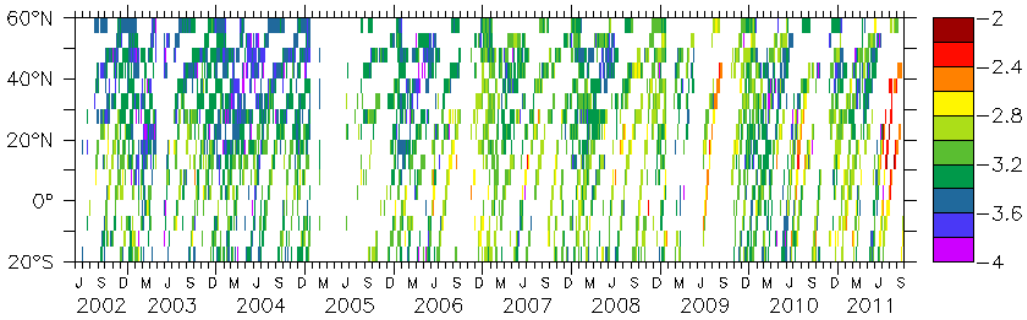
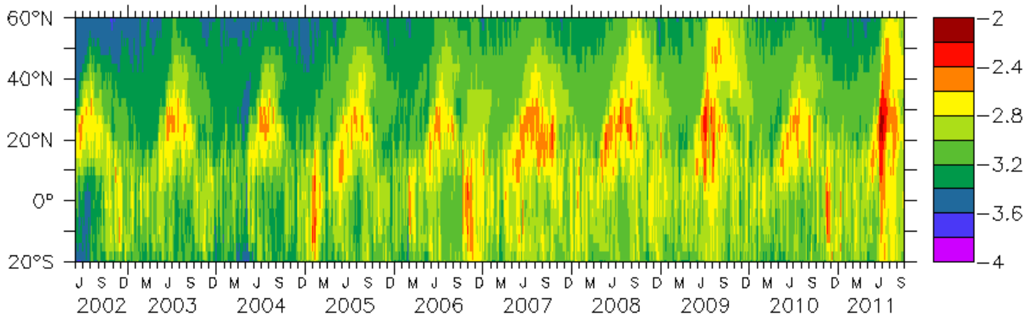


Figure 1: GOMOS and EMAC extinctions (log) in the tropics as function of altitude for different wavelengths: (a) UV 350 nm, (b) visible 550 nm and (c) near infrared 750 nm; ~~dust in EMAC~~ resolution ~~T42 downscaled to results in resolution T63~~T63L90.



1/km, 550nm, GOMOS 3.00, 17km, 60−120E



1/km, 17km, EMAC, 60−120E, 0.33*DU (log)

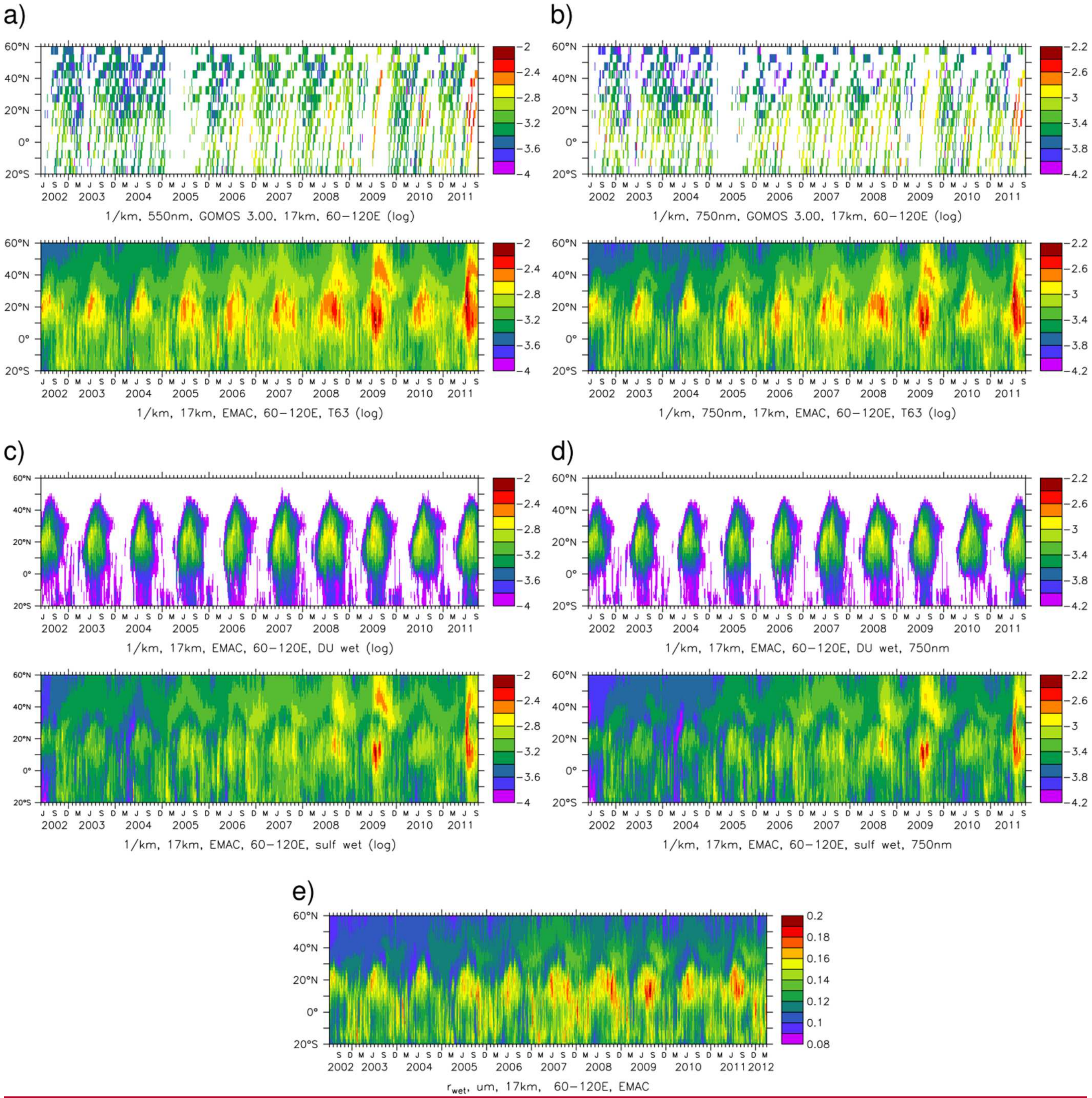

a)
1/km, 550nm, GOMOS 3.00, 17km, 60−120E (log)
1/km, 17km, EMAC, 60−120E, T63 (log)
b)
1/km, 750nm, GOMOS 3.00, 17km, 60−120E (log)
1/km, 750nm, 17km, EMAC, 60−120E, T63 (log)
c)
1/km, 17km, EMAC, 60−120E, DU wet (log)
1/km, 17km, EMAC, 60−120E, sulf wet (log)
d)
1/km, 17km, EMAC, 60−120E, DU wet, 750nm
1/km, 17km, EMAC, 60−120E, sulf wet, 750nm
e)
r_wet, um, 17km, 60−120E, EMAC
60°N
40°N
20°N
0°
20°S
2002 2003 2004 2005 2006 2007 2008 2009 2010 2011 2012
−2 −2.4 −2.8 −3.2 −3.6 −4
−2.2 −2.6 −3 −3.4 −3.8 −4.2
0.2 0.18 0.16 0.14 0.12 0.1 0.08

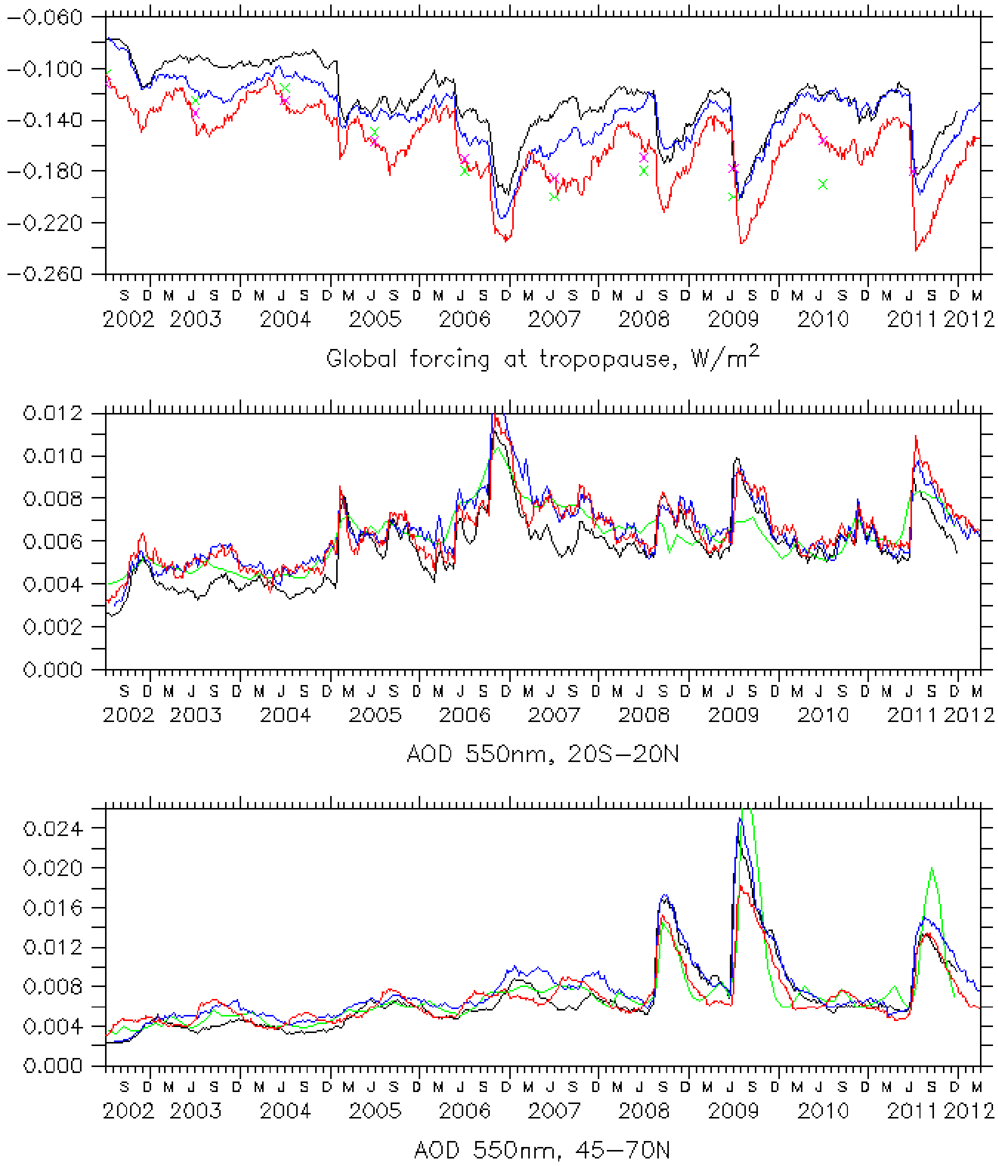
Figure 2: a) + b) Observed (~~top~~upper) and simulated (~~bottom~~lower, EMAC T63L90) extinction in the Asian sector (60°E-120°E, 20°S-60°N) for 550 nm and 750 nm. c) + d) Contribution of wet dust (upper) and wet sulfate (lower) to extinction for 550 nm and 750 nm. e) median wet radius in accumulation mode (for effective radius multiply by 1.4).

4.3. Stratospheric aerosol radiative forcing~~ and total optical depth of~~ , stratospheric aerosol optical depth and radiative heating

Desert dust transported to the UTLS mostly via the Asian summer monsoon contributes significantly to the seasonal cycle of total stratospheric aerosol optical depth (SAOD) in satellite observations and the EMAC simulations shown in Fig. ~~3~~3b for the tropics (middle panel, vertical integral of extinction above about 16 km) and for midlatitudes (bottom panel~~).~~, above about 14 km). Global radiative forcing at the tropopause is depicted in the upper panel a. The figure contains in black results from the ~~old model version with less volcanoes (Brühl et al., 2015~~T42L90 simulation of Bingen et al. (2017) and in blue the ~~old model version~~T63L90 simulation with ~~about~~ the ~~same~~high volcanic sulfur input ~~as in~~ derived for the coarse resolution.~~Bingen et al. (2017).~~ Green lines and symbols show estimates derived from satellite observations ~~like~~ (SAGE II, OSIRIS ~~(Optical Spectrograph and InfraRed Imaging System) and~~ and CALIPSO~~ (~~, Solomon et al., 2011; Santer et al., 2014; Bourassa et al., 2012~~).~~; Glantz et al., 2014). Red shows results of the current model version in T63L90 with the Astitha et al. (2012) dust scheme~~.~~ and corrected SO_2 input (see section 3 and supplement). Concerning global radiative forcing, the volcanoes are the dominating effect with up to 0.13 W/m^2 for Rabaul and Nabro compared to the volcanicly quiet period in 2002. Here the use of the SO_2 inventory for T42L90 in the T63L90 simulation (blue) causes an overestimate of up to 50% in 2006 and 2007 due to accumulation effects of eruptions following in short sequence. This is visible in the overestimate of tropical SAOD depicted by the blue curve in Fig. 3b (upper).

Especially in Northern midlatitude summer ~~these aerosol optical depths (AOD) appear~~SAOD in T42L90 appears to be high because in ~~standard horizontal~~that resolution ~~T42~~ the convective transport of dust to the UTLS in the Asian monsoon region is overestimated~~.~~ (Fig. 3b, lower). This is clearly seen in Fig. 4 which shows in black the T42L90 simulation, in green the observations of 550 and 750 nm SAOD by GOMOS~~.~~ and in light blue (Fig. 4b only) by OSIRIS in different latitude bands, including the monsoon region. For the narrow latitude bands in Fig. 4b inclusion of OSIRIS data is important because here GOMOS has often too low coverage. Nevertheless, for a lot of features the two satellite datasets agree well. Using the higher resolution ~~T63~~T63L90, for which the convection parameterization was developed, the agreement to the satellite observations is much better (~~Fig.~~Figs. 3 and 4,

~~purple),~~red) than T42L90, especially at midlatitudes~~.~~ and in subtropics. In the subtropics (Fig. 4b, lower) the simulation with low resolution (black) always overestimates the monsoon peaks in August compared to the ones seen in the observations. Comparing the model results with OSIRIS in the northern tropics (Fig. 4b, upper) indicates that some volcanic events are still missing in the inventory, for example in spring 2007 and 2010. This would also explain the differences in radiative forcing (indicated by crosses in Fig. 3a) in these years.



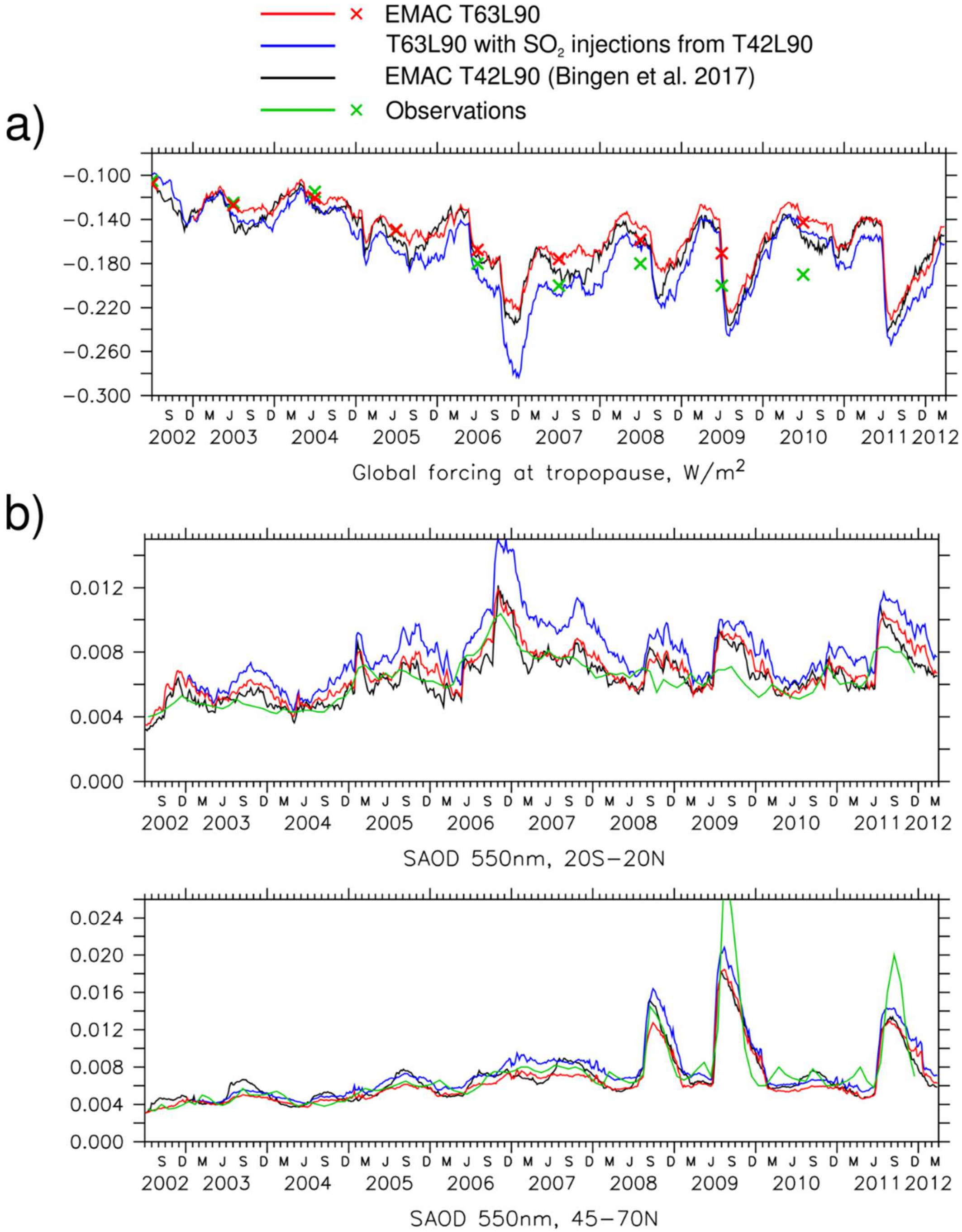
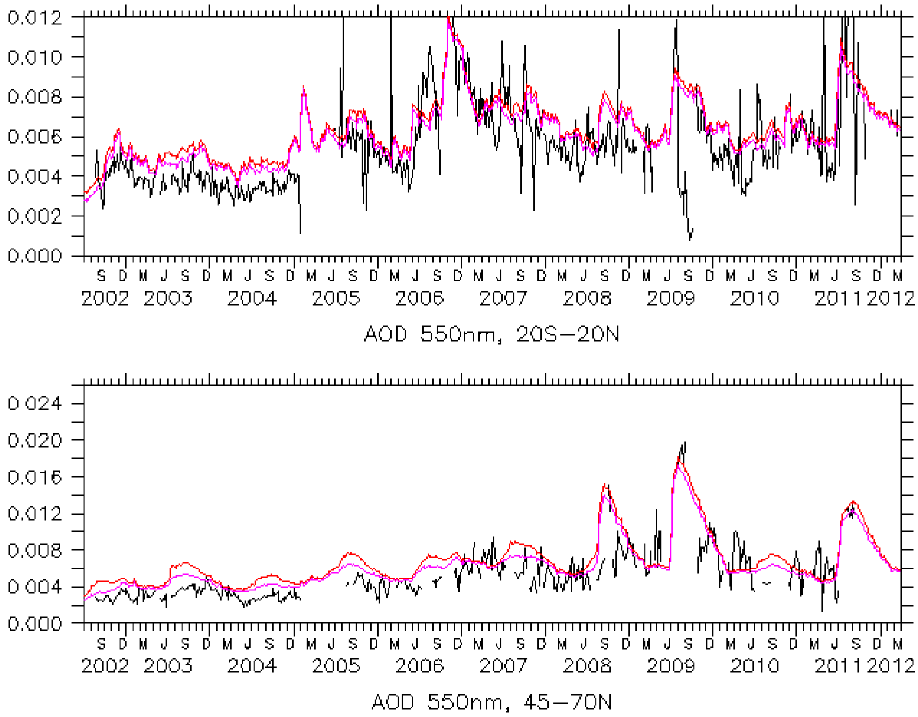


Figure 3: a) Stratospheric aerosol radiative forcing and b) stratospheric AOD. Red lines and crosses: EMAC,~~42,~~, resolution T63L90, current version; black: EMAC T42L90 (Bingen et al., 2017); ~~black: EMAC, version used in Brühl~~

~~et al. (2015);~~ blue: ~~As black, but with more volcanoes~~T63L90 without downscaling the SO_2 injections for T42L90. green: From observations (crosses annual mean for forcing (Solomon et al., 2011), SAGE II, CALIPSO, OSIRIS).



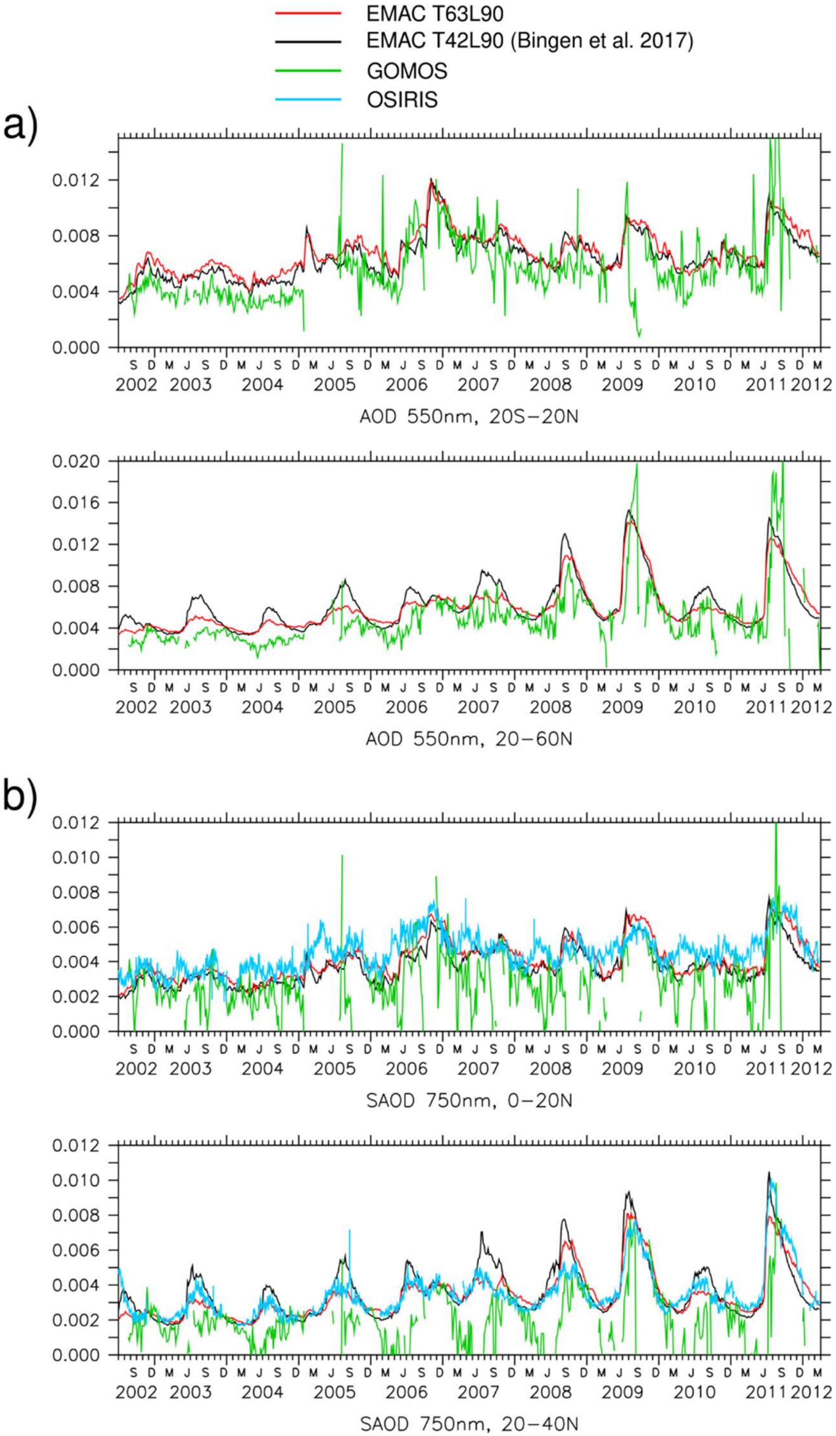

EMAC T63L90
EMAC T42L90 (Bingen et al. 2017)
GOMOS
OSIRIS
a)
0.000
0.004
0.008
0.012
S D M J S D M J S D M J S D M J S D M J S D M J S D M J S D M J S D M J S D M J S D M
2002 2003 2004 2005 2006 2007 2008 2009 2010 2011 2012
AOD 550nm, 20S−20N
0.000
0.004
0.008
0.012
0.016
0.020
AOD 550nm, 20−60N
b)
0.000
0.002
0.004
0.006
0.008
0.010
0.012
SAOD 750nm, 0−20N
SAOD 750nm, 20−40N

Figure 4: a) Stratospheric AOD at 550 nm observed by GOMOS (~~black~~green) and simulated by EMAC in resolutions ~~T42~~T42L90 (black) and T63L90 (red~~) and T63 (purple). The simulations differ mostly by dust transported to the UTLS.~~). b) Stratospheric AOD at 750 nm tropics and subtropics (SAOD above 15 km), additionally with OSIRIS observations (light blue).

The simulated aerosol radiative heating~~ shown in Fig. 5~~, derived from radiation calls with and without aerosol, reflects the medium volcanic eruptions with the largest effects near 18 km~~.~~ (Fig. 5). There, desert dust causes additional heating at the time of the Asian summer monsoon. In the UTLS below every year in September a clear signal from biomass burning organic aerosol, its volume mixing ratio is shown in Fig. S2 of the supplement, is visible. Above, around 22 km, the dust below in northern hemispheric summer causes a reduction of absorption of terrestrial radiation by ozone. ~~This effect is smaller in the simulation with higher horizontal resolution (T63) but still visible.~~

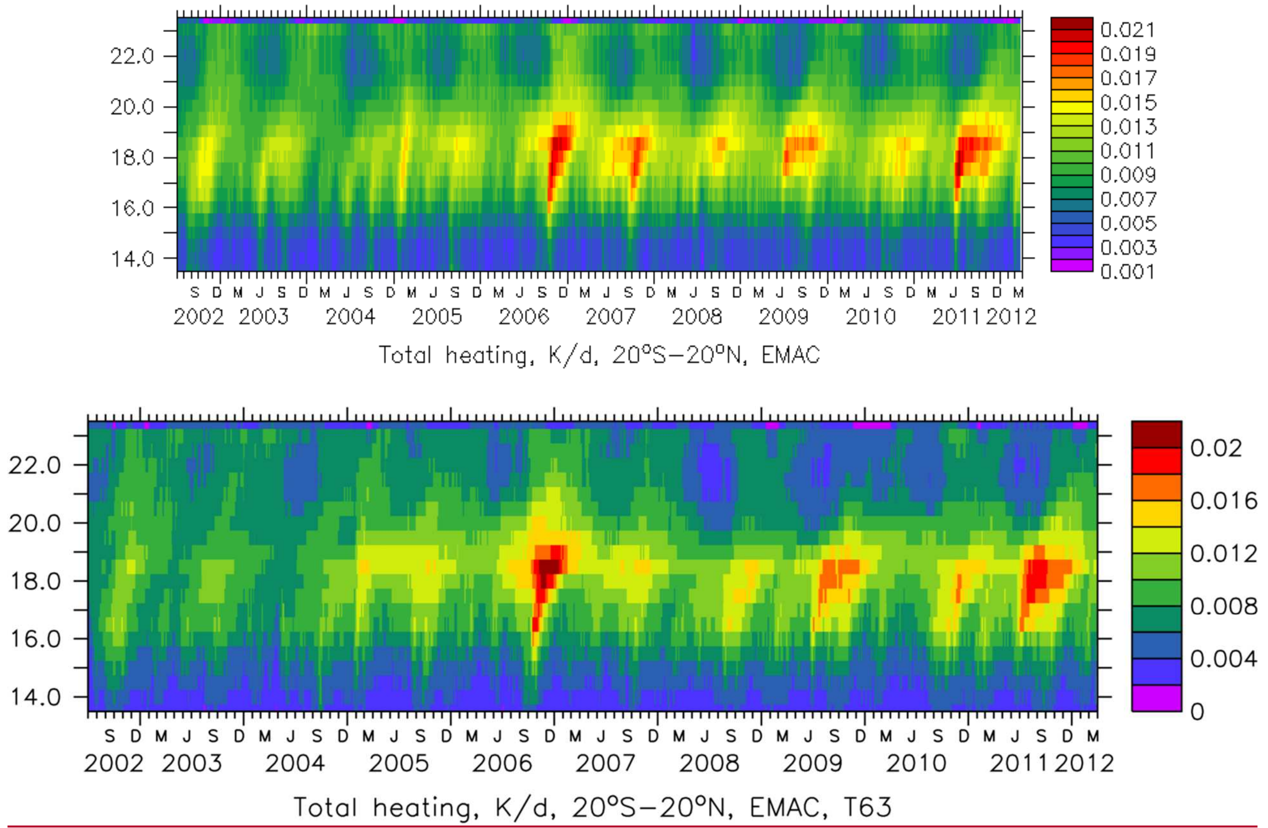
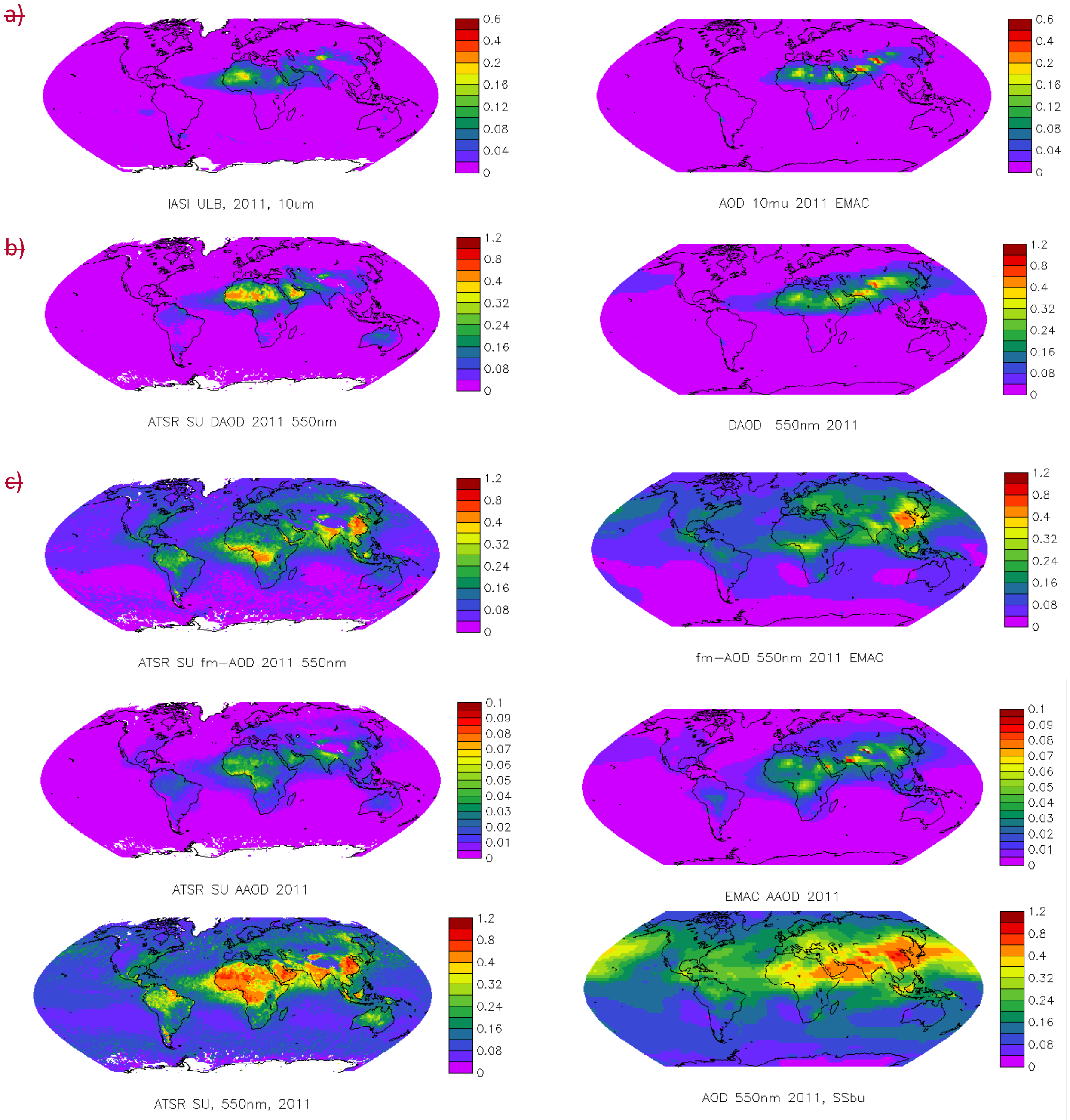


Figure 5: Simulated aerosol radiative heating in tropics (solar + infrared, ~~T42, full dust).~~T63L90).

4.4. Constraints from total aerosol optical depth in different spectral regions and for different aerosol subsets

The first comparisons are carried out for EMAC in ~~T42L90~~T63L90, the standard resolution used in the previous sections. ~~The AOD~~Here AOD refers to troposphere and stratosphere. The DAOD (dust AOD) in terrestrial infrared is most sensitive to the coarse mode of tropospheric dust. Fig. ~~6 (a)~~6a shows that the model reproduces most of the IASI features~~ but overestimates the AOD in the Himalaya region. This feature appears also in~~. DAOD in the visible spectral region (~~b),~~Fig. 6b) is too high over central Asia, pointing ~~also~~ to an overestimate of dust in the accumulation mode near the Taklamakan desert. The patterns in the IR and visible spectral range are different despite considering the factor 2 often applied by AEROCOM/AEROSAT (Aerosol Comparison between Observations and Models) community for conversion in the color scales of Fig 6a,b. This holds for model and observations. The fine mode AOD fraction~~ is ~~, which is dominated by the accumulation mode, is slightly overestimated over Europe ~~(c~~and underestimated in the biomass burning regions in Africa (Fig. 6c). In the model this is sensitive to the way how the extinction of aerosol water is attributed to the soluble aerosol species, especially sea salt. Absorbing AOD, i.e. AOD times (1-~~ω)~~ -ω) with ~~ω~~ω single scattering albedo, agrees surprisingly well (~~d~~Fig. 6d). In the total AOD (~~e~~Fig. 6e) there appears to be too much sea salt in the model, or still not optimum parameters for the sea salt composition which controls water uptake~~.~~ (see section 3).



~~d)~~

~~e)~~

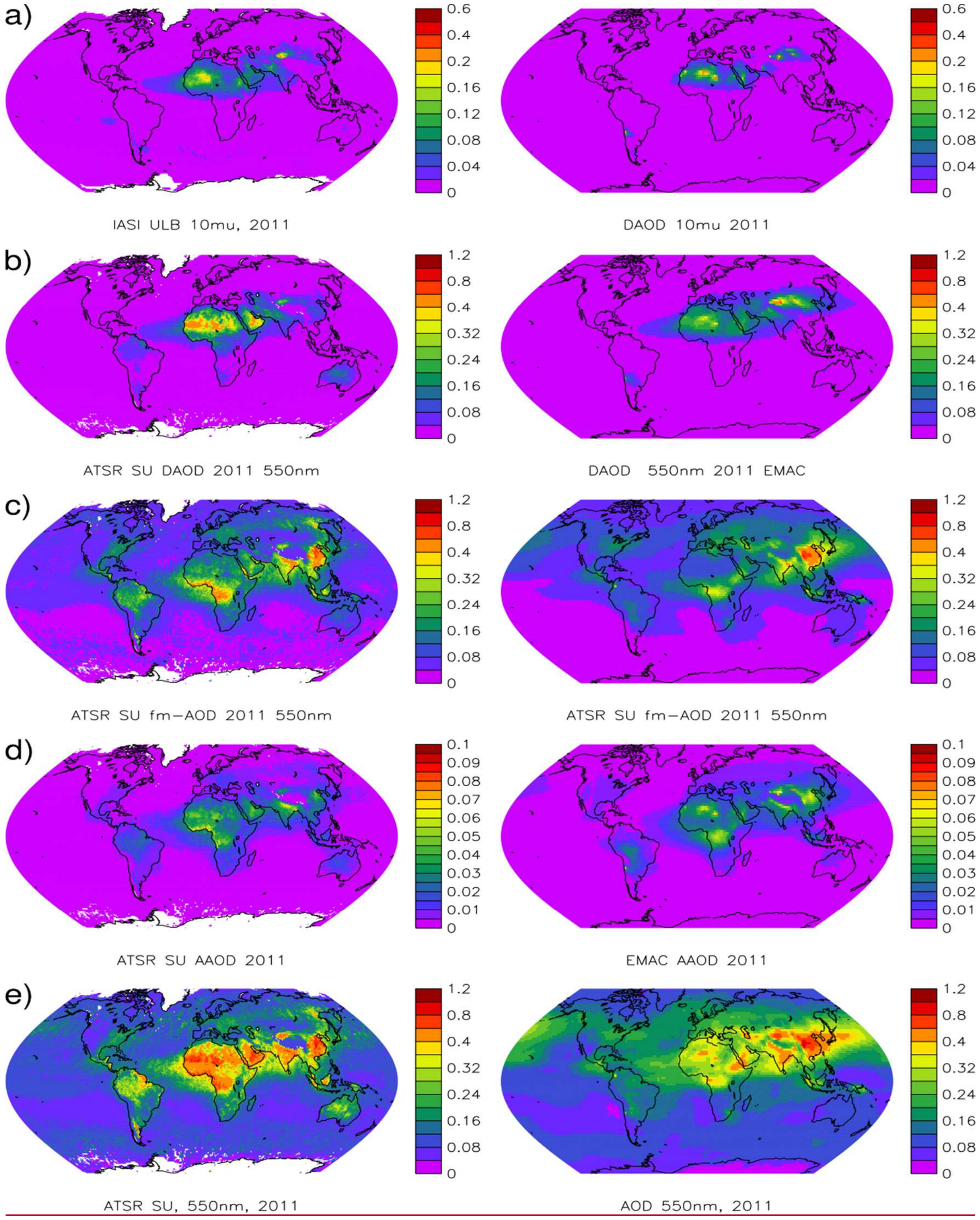
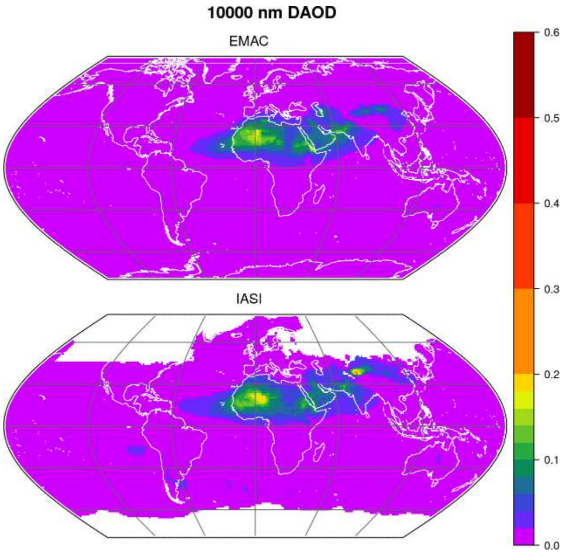

a)
0.6
0.4
0.2
0.16
0.12
0.08
0.04
0
IASI ULB 10mu, 2011
DAOD 10mu 2011
b)
1.2
0.8
0.4
0.32
0.24
0.16
0.08
0
ATSR SU DAOD 2011 550nm
DAOD 550nm 2011 EMAC
c)
1.2
0.8
0.4
0.32
0.24
0.16
0.08
0
ATSR SU fm−AOD 2011 550nm
ATSR SU fm−AOD 2011 550nm
d)
0.1
0.09
0.08
0.07
0.06
0.05
0.04
0.03
0.02
0.01
0
ATSR SU AAOD 2011
EMAC AAOD 2011
e)
1.2
0.8
0.4
0.32
0.24
0.16
0.08
0
ATSR SU, 550nm, 2011
AOD 550nm, 2011

Figure 6: Observed (left) and simulated (right) (a): 10 µm dust AOD (DAOD) for IASI and EMAC; ~~(b):~~ and (b): 0.55 µm ~~dust aerosol optical depth (DAOD);~~DAOD; (c): fine mode AOD; (d): absorbing AOD (AAOD) and (e): total AOD for ATSR (SU) and EMAC in ~~T42L90~~T63L90 resolution, annual mean 2011.

Figure 7 compares the annual average for 2011 of the 10 µm DAOD observed by IASI and simulated by EMAC in the low top version with high horizontal resolution (~~T106~~T106L31, about 1.1°). The satellite retrievals are taken from version 8 of the ULB dataset. The simulation utilizes the dust emission scheme of Klingmüller et al. (2018) which calculates the emissions online considering the meteorological conditions. To extract the DAOD from the total EMAC AOD at 10 µm, we apply a filter nullifying sea salt dominated AOD values. To identify the latter, we compare the AOD weighted with the volume of sea salt and dust.



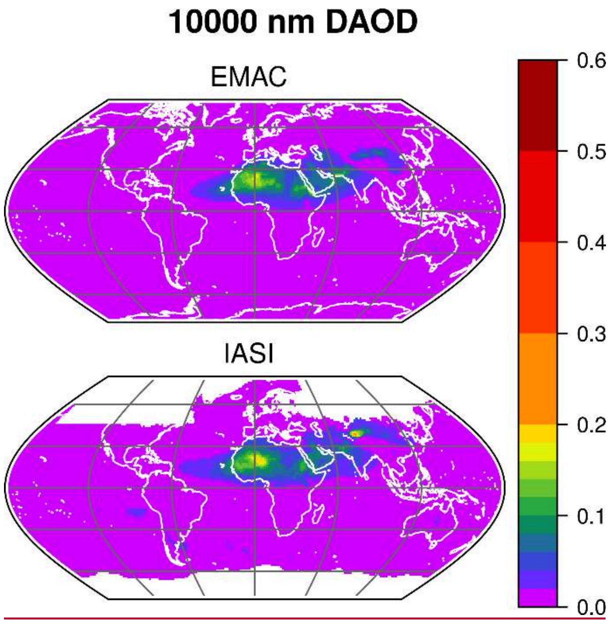
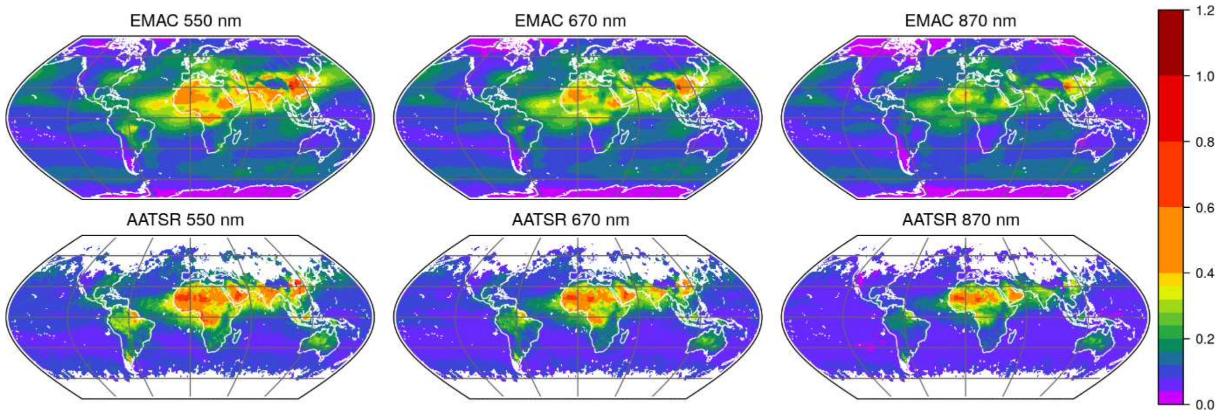


Figure 7: Annual mean for 2011 of the DAOD at 10 µm wavelength observed by IASI (bottom, IASI ULB dataset version 8) and simulated by EMAC (top) at ~~T106 horizontal~~T106L31 resolution~~ (about 1.1°) using 31 vertical levels~~.

The observed and modelled global DAOD distributions shown in Fig. 7 agree remarkably well. The pixel values of each map are strongly correlated with a correlation coefficient of 0.91. The overall AOD level is consistent as well, so that a similar variance of the pixel values is obtained for the observed (0.00038) and the modelled (0.00041) DAOD distribution. Interestingly, the DAOD from the older version 7 of the ULB dataset yields a pixel by pixel correlation coefficient of only 0.89 and a pixel value variance of only 0.00029. We conclude that the agreement of EMAC and IASI has improved with the update from version 7 to version 8 of the IASI ULB dataset.

The main disagreement of the two maps in Fig. 7 is the less pronounced maximum over the Taklamakan Desert in Central Asia in the model result. This underestimation is related to the model surface friction velocity in mountainous regions like the surroundings of the Taklamakan Desert, which tends to be lower in simulations at higher horizontal resolution (e.g. T106) than at lower resolution (e.g. ~~T42~~T63), possibly resulting in an underestimation of the dust emissions.

Figure 8 compares results from the T106L31 EMAC simulation for the annual average of the total AOD at visible and near-infrared wavelengths with AASTR retrievals using the ATSR (SU) algorithm version 4.3. Generally good agreement is obtained at 550 nm which is consistent with the good agreement between the 550 nm MODIS (Moderate-resolution Imaging Spectroradiometer) AOD and model results based on the same EMAC version (Klingmüller et al., 2018). As for the ~~T42L90~~T63L90 simulation, the model yields higher sea salt related AOD levels over the oceans. In contrast, the model AOD over the Sahara is lower than the satellite retrieved values. This becomes even more evident at larger wavelengths (Fig. 8, right): the model AOD over the Sahara, in contrast to most other regions, has a stronger wavelength dependence than the observed AOD, corresponding to a larger Ångström exponent. This discrepancy might be resolved by adjusting the dust particle size distribution in the model under the constraint of not sacrificing the good agreement of model and observed AOD at 550 nm and at 10~~ µm.~~ µm. This could involve modifying the parameters of the log-normal modes, i.e., their widths and boundaries, but also reassessing the parameterization of relevant processes such as emission, deposition, coagulation and hygroscopic growth, or even adding an extra mode for extremely coarse particles which can be relevant close to dust sources. Over South America, the biomass burning regions of Africa, India and China the wavelength dependence of model and observed AOD is largely consistent.



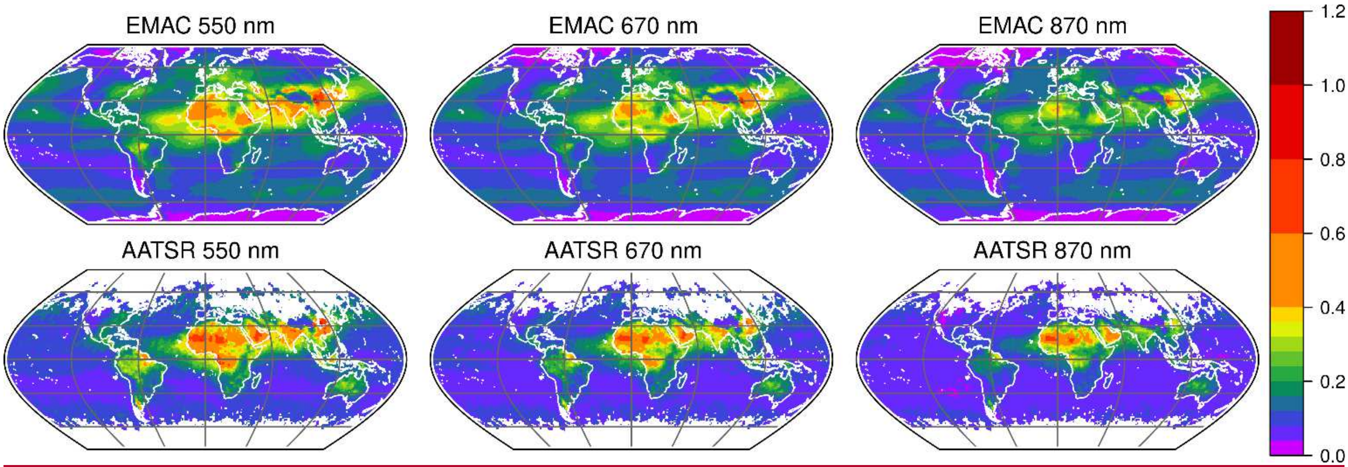


Figure 8*:* Annual mean for 2011 of the AOD at (from left to right) 550 nm, 670 nm and 870 nm wavelength observed by AATSR (bottom, SU-ATSR algorithm version 4.3) and simulated by EMAC (top) at ~~T106 horizontal~~T106L31 resolution. ~~(about 1.1°) using 31 vertical levels~~

5 Conclusions

Satellite data are not only important to constrain model parameters; they are very important for model ~~development~~improvement. Comparing satellite data with model results at different wavelengths simultaneously provides additional information and is also valuable for the satellite community to check internal consistency, as in our case for GOMOS and OSIRIS.

Sophisticated modelling of dust and organic aerosol as well as a detailed volcano dataset are necessary to reproduce the seasonal cycle and the interannual variability of extinction in the lowermost stratosphere observed by GOMOS at different wavelengths~~ (Bingen et al., 2017). The scaling of EMAC dust AOD~~. From the wavelength dependence in ~~the stratosphere by 0.33 was done to correct for an artifact of convective transport in the resolution T42 which is not apparent in the higher resolution T63. The additional (ongoing)~~observations and simulations regions in the UTLS with ~~higher~~enhanced particle size due to water uptake can be identified as aged dust in the Asian monsoon region. Convective transport of dust into the UTLS is resolution dependent because of differences in convection top height and overshooting convection. A resolution of T63L90 (1.88° in longitude and latitude, 90 vertical layers) fits best to the observations. For the low resolution ~~were stimulated by the satellite observations~~T42L90 (2.75°) dust SAOD (and stratospheric mixing ratio) has to be downscaled by a factor of about 0.33, while for higher resolutions (e.g. T106L90) upscaling is required. The resolution dependent differences in convection modify also the residence time of sulfur species in the lowermost stratosphere, especially at low latitudes, in resolution T42L90 it appears to be too short.

The total AOD in the visible spectral range is very sensitive to aerosol water and the composition of sea salt. In the modal model, the bulk fraction has to be increased compared to ions to reduce artifacts of too much water uptake by sea salt. The satellite data ~~help~~helped to ~~find~~identify a preferred parameter set for the ~~best choice of parameters~~sea salt emission composition.

~~Simulated~~Our simulated dust total aerosol optical depth agrees with satellite data in the visible (ATSR SU) and the infrared (IASI ULB, version 8). The combined comparison at visible and infrared wavelengths provides strong constraints on the modelled particle size distribution. The direct comparison of observations and model reveals different structures in the extinction patterns at both spectral ranges. From this, we conclude that simply assuming a spatially constant factor of (about) 2 for conversion of ~~AOD~~DAOD from 10 µm to 550 nm, as commonly applied in the AEROCOM/AEROSAT community, is too crude.

~~Dust and volcanic eruptions based on satellite datasets help~~Satellite datasets identifying volcanic SO_2, including its vertical distribution or enhanced extinction by aged dust enable the model to get closer to observationally based estimates for radiative forcing, showing the interest of a close interaction between modelling and observation research teams.

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