Author's Response

Dear editor:

For the major changes in the revised manuscript, we included our response to the reviewers' comments. This shows all the major changes made in the revised manuscript. However, we additionally included the revised manuscript with track changes, which includes:

1. The same changes presented in our response to the reviewers' comments
2. Affiliation change for Y.H. Lee
3. Acknowledgements for the two reviewers
4. Minor changes, mostly in the Tables and Figures captions.

Response to Referee #1

Response) We thank the reviewer for their constructive comments and suggestions and have made several changes to the paper to address the issues raised. Reviewers' comments are shown in italics with our response shown after each.

The paper is an evaluation of the aerosol distributions simulated by the TOMAS aerosol scheme used in the GISS modelE2 global climate model. The evaluation is similar to most papers of its kind, but covers a larger number of observational datasets to address more aspects of the aerosol model (for example including size distributions). The paper is most interesting and useful to other aerosol model developers when it clearly explains the reasons behind the choices of parameters (e.g. size distribution of emissions, parameterisation of DMS emission rates, fraction of primary sulphate emissions, differences with bulk mass scheme, etc.) and behind model skill, or lack thereof, when compared to observations. This good level of explanation is not always present, however, and most of my comments below suggest improvements in that direction.
The paper could have taken a different – and arguably more original – direction, looking at how improvements in the host model, from II-prime to ModelE2, have affected the skill of the aerosol scheme at reproducing observed aerosol distributions with fidelity.
This is an important aspect of global aerosol modelling, as yet almost unexplored in the literature. The paper is long, with a large number of Tables and Figures. However, considering the breadth of the model evaluation, it is difficult to recommend shortening the discussion or removing figures (with the possible exception of Figure 25). The conclusion (section 6) is a good summary of the findings. I recommend publication after the following comments, aimed at improving the discussion, are addressed by the author.

1 Main comments
• *Section 2: Since the main motivation for developing ModelE2-TOMAS is to be able to use TOMAS in a better model than II-prime (page 5835, lines 8), one would have expected a more complete discussion of ModelE2 compared to IIprime, especially on those aspects that are relevant to the life cycle of aerosols and their radiative effects. So section 2 should be extended with a discussion of changes in cloud, precipitation, and transport schemes, summarising the improvements in those and how they are expected to impact on the quality of the aerosol simulation. A lot of a model's skill at simulating aerosols does not depend on the aerosol scheme itself, but on the host model – and this dependence has been little investigated in the literature so far. I would strongly encourage the authors to look into that aspect.*
Response) I agree with the reviewer that it would be very interesting to explore how aerosol model predictions depend on a host model configuration. It is disappointing to mention that because GISS-TOMAS has been unofficially retired and is not available to run at this time, it is not possible to compare two TOMAS models. Also, especially due to the lack of documents for the old II-prime model, it is difficult to describe how non-aerosol GCM components are different between the two GCM versions.

• *Section 5.2: The AeroCom 1 simulations are a decade old now, and comparing against those*

does not mean much – aerosol modelling has progressed since then, and comparing to older – presumably poorer – models does not really demonstrate skill. The comparison should therefore be restricted to AeroCom 2 models (comparing total carbonaceous if a more detailed split is not possible) and ACCMIP. Also, since emissions vary between the different studies, comparing absolute values for burdens and mass fluxes is not really useful: the focus should only be on lifetimes and relative contributions to deposition rates, with burdens given on Table 5 for information only.

Response) We have deleted the comparisons to AeroCom Phase 1 budgets except for the lifetime and removal rate coefficients comparison, as the reviewer suggested. We only use the ACCMIP multi-model budgets to compare the burden, source, and deposition rates to compare sulfate and elemental carbon. We can't use the AeroCom Phase 2 budgets from Myhre et al. (2013), mainly because their budgets are anthropogenic portion, which is the differences between the present-day run and the preindustrial run. As the changes in the revised manuscript are large, please see the revised manuscript for the changes.

2 Other comments

• *Page 5831: The title is misleading, because the paper is really about evaluation. Development details are delegated to previously-published papers. I would therefore drop the word "development" from the title.*

Response) We have modified the title to

"Evaluation of the global aerosol microphysical ModelE2-TOMAS model against satellite and ground-based observations"

• *Page 5833, line 16: Strictly speaking, the pre-industrial atmosphere matters only for the radiative forcing, not for the radiative effect.*

Response) Thanks for catching that. It's now corrected in the revised version.

• *Page 5834, line 11: "very accurately": I'm not sure what the point is here. Are the authors suggesting that single-moment representations are inherently less accurate in what they can simulate?*

Response) Yes – at least when compared for the same number of size sections. Single-moment sectional model assumes an average mass of particles in each bin to be constant and it is not changed over time, leading a substantial numerical error in a number prediction. In reality and in the two-moment scheme, the average particle mass in a bin is varied with time, and this flexibility leads to greater accuracy. A two-moment model does not assume the average mass in a bin to be constant and overcomes the problem stated above. Harrington and Kreidenweis et al. (1998) shows much more variables are need for a single-moment sectional model to predict accurate simulation. Also, Feingold et al. (1988) shows the superior of a two-moment method compared to a single-moment. We have included these references in the revised manuscript.

Debra Y. Harrington and Sonia M. Kreidenweis, "Simulations of Sulfate Aerosol Dynamics Part II: Model Intercomparison," Atmospheric Environment 32, no. 10 (May 1, 1998): 1701–9,

doi:10.1016/S1352-2310(97)00453-6.

G Feingold, S Tzivion, and Z Levin, "Evolution of Raindrop Spectra. Part I: Solution to the Stochastic Collection/Breakup Equation Using the Method of Moments," Journal of the Atmospheric Sciences 45, no. 22 (1988): 3387–99, doi:10.1175/1520-0469(1988)045<3387:eorspi>2.0.co;2.

• *Page 5837, section 2.1: What is the mixing assumption in the bulk model: external? Same question for TOMAS: how are aerosols mixed within each size bin (Page 5838, line 11)?*
Response) The bulk aerosol model (i.e., ModelE2-OMA in the revised manuscript) assumes external mixing, and the TOMAS microphysical processes assume internal-mixing state in a size bin. We included the following sentence in the revised manuscript.

"In general, TOMAS treats all aerosols as internally mixed during microphysics such as calculating condensation and coagulation rates. However, a portion of EC is treated as externally mixed for purposes of wet deposition."

• *Page 5838, line 17: Ammonium has just been discussed, so this statement seems redundant.*
Response) The bulk tracer NH_4 has been removed.

• *Page 5838, line 23: How is hygroscopic growth represented then?*
Response) For sulfate and sea-salt, we use a polynomial fit based on ISORROPIA, a thermodynamic equilibrium model for inorganic aerosols (Nenes et al., 1998). For organic carbon, it is based on the observations of Dick et al. (2000). We have added the following statements in Section 3.

"Water uptake by sulphate and sea salt is based on a polynomial fit based on ISORROPIA, a thermodynamic equilibrium model for inorganic aerosols (Nenes et al., 1998). For water uptake by hydrophilic OM, it is based on the observations of Dick et al. (2000)."

• *Page 5839, line 5: Why are the other kinds of nucleation unused? Ok, it is mentioned in section 4.2: it may be useful to point out here that the reason for not using a given parameterisation will be discussed later in the paper.*
Response) We have moved the following sentences from Section 4.2 to Section 3 (model description).

"The boundary-layer nucleation is off in all simulations because it tends to overpredict aerosol number concentrations in our model. Also we do not show any run with the ternary nucleation (Napari et al., 2002) because it overpredicts aerosol number concentration severely (not shown)."

• *Page 5839, lines 6–19: For the benefit of other model developers, it would be interesting to say how the size bin/cutoff configurations are selected. I assume it is a compromise between*

computation cost and fidelity of the model, but how is that latter quantified?
Response) The computation costs and numerical errors of the Fast TOMAS models are presented in Lee et al. (2013b). In Lee et al. (2013b), we compared the different versions of TOMAS models to an analytical solution of condensation and coagulation in box-scale and also compared the fast versions of TOMAS models to the original TOMAS model, which was our benchmark. We have added the following in Section 3.

"As discussed in Lee and Adams (2012), the Fast TOMAS reduces the computational burden by 2-3 times while generally predicts CCN concentrations within a few percent of the original TOMAS."

• *Page 5840, lines 1–3: Is the fraction of precipitating cloud water computed in each model layer? Does the wet deposition flux account for re-evaporation of precipitation?*
Response) Yes. The fraction of precipitating clouds water is computed for each model layer. Wet deposition accounts for the changes of tracers in precipitation-water by its re-evaporation. We have modified the following sentence.

"The fraction of activated aerosols removed by wet deposition is proportional to the fraction of cloud water that precipitates, **which is computed in each model layer. Wet deposition accounts of re-evaporation of precipitation.**"

• *Page 5882, caption of Figure 1: The caption could be improved to make clear that the Table lists aerosol and precursor emissions, and how nucleation is accounted for.*
Response) I think the comment is for Table 1. The Table 1 caption is now revised as below.

"Table 1. Aerosol and precursor gas emissions used in TOMAS and the bulk aerosol models and the nucleation scheme used in the TOMAS simulations"

• *Page 5841, lines 15–18: I understand the need for pragmatic choices like this one, but it would be useful to offer an explanation as to why the Nightingale et al. (2000) leads to an overprediction in the Southern Hemisphere in both aerosol schemes. Is it because of other aspects of the model?*
Response) Boucher et al (2003) simulates DMS using the Nightingale et al. (2000) scheme (used in ModelE2-OMA) using a global climate model, LMD-ZT. The overprediciton of DMS is not shown in Boucher et al. (2000). Their DMS mixing ratios at the same observation sites (Amterdam Island and Cape Grim) are about 50% of the ModelE2 DMS values and is more comparable to ModelE2-TOMAS DMS, which uses the Liss and Merlivat (1986) scheme. This seems to suggest that ModelE2 has a stronger surface wind speed than the LMD-ZT model used in Boucher et al. (2003), but we need to investigate this further in the future to confirm.

While doing this, we have updated the DMS measurements at Amsterdam Island and Cape Grim to cover a longer time period. The most noticeable change from this update is the observed DMS mixing ratios during the winter at Amsterdam Island, which are about two

times higher than our original DMS observation. This is because DMS has a large year-to-year variability (Sciare et al., 2000). With this revised data, the DMS in ModelE2-OMA is overpredicted by about 30% at Amsterdam Island (much less than before) and still more than a factor of two at Cape Grim. We have included the following in the revised text.

"The DMS concentrations seem to agree well against the observations when using the sea-air transfer function of Liss and Merlivat (1986), i.e, the case for ModelE2-TOMAS, but this run underpredicts during the winter season at the Amsterdam Island site and all season at Dumont site. Earlier, we mentioned that the global DMS budgets from Boucher et al. (2003) agree well with those from ModelE2 when using the same DMS emission parameterization. However, Boucher et al. (2003) shows better agreement to the same DMS measurements when using the sea-air transfer function of Nightingale (2000) at Amsterdam Island and Cape Grim (i.e., the case for ModeleE2-OMA), because their DMS mixing ratios from that simulations are actually closer to ModelE2-TOMAS. This may suggest that, at least over SH high latitude regions, the surface wind speed in ModelE2 is much stronger than that in LMD-ZT, resulting in higher DMS emissions and burden. We need to investigate further to find out a source for the difference though."

J. Sciare, N. Mihalopoulos, and F. J. Dentener, "Interannual Variability of Atmospheric Dimethylsulfide in the Southern Indian Ocean," Journal of Geophysical Research: Atmospheres 105, no. D21 (November 16, 2000): 26369–77, doi:10.1029/2000JD900236.

O Boucher et al., "DMS Atmospheric Concentrations and Sulphate Aerosol Indirect Radiative Forcing: A Sensitivity Study to the DMS Source Representation and Oxidation," Atmos. Chem. Phys. 3, no. 1 (2003): 49–65, doi:10.5194/acp-3-49-2003.

• *Page 5883, Table 2: Need to define GMD and GSD in the caption.*
Response) Added.

• *Pages 5884 and 5885: It would make sense to merge Tables 3 and 4, since they are analysed together.*
Response) We have merged the two tables.

• *Page 5846, lines 24–25: It would be useful to remind the reader that anthropogenic emissions are supposed to be representative of the year 2000, which justifies the choice of period for observations.*
Response) We would like to note that, as mentioned in Section 4.1, the year 2000 emissions for SO_2 include the anthropogenic emissions as well as the natural emissions such as volcanic eruptions and DMS oxidation. The year 2000 intends to represent the "present-day". The choice of observational period is not to evaluate only anthropogenic emissions but rather the "present-day" conditions.

• *Figure 2: The red and blue signs are difficult to distinguish. Perhaps use filled circles, or*

another, thicker, symbol? Also, the model overestimates concentrations by more than 10 times for 5 sites of the EMEP network. Do those 5 sites have some common characteristics that could be the signature of a specific weakness of the model?
Response) The modified Figure 2 uses a different symbol to make it more distinguishable. For the EMEP sites showing significant overpredictions, we could not find any common characteristics except that their observed concentrations are on the low side. This requires more detailed investigation, but, for now, we guess that those locations are influenced by overpredicted SO_2 emission nearby.

• *Page 5848, lines 4–5: This is interesting, but also worth an explanation. Why would DMS concentrations increase aloft? Lack of oxidants?*
Response) We suspect that the model has too strong vertical transport by clouds. In fact, we provided the explanation the below paragraph (Pg. 5848, L: 10-15). We have decided to delete the sentence and modified the following part to avoid any confusion.

(Old) "Considering the small DMS peak at 8 km and the elevated SO_2 in the upper/free troposphere"
(New) "Considering the small DMS peak at 8 km and the elevated SO_2 in the upper/free troposphere in the model (see Figs. 4 and 5)"
(Deleted) Interestingly, the model DMS seems to show an increase above ~8 km quite consistently throughout the sites.

• *Page 5848, lines 16–17: It looks like volcanic emissions help the model do the right thing in Mar-Apr, but not Aug-Oct. Is the agreement in Mar-Apr coincidental, or should one apply a seasonality to volcanic emissions? Also what about the peaks in observed SO2 visible above 4 km over Tahiti and Easter Island: that looks like transported aerosol layers. Are those mentioned in the papers on the PEM-Tropic campaigns? Where do they come from?*
Response) Please read below for our response. The following is now included in the revised manuscript.

"A large peak in the mid-troposphere at Hawaii in the models results from volcanic SO_2 emissions, while the observations show a similar peak only during March-April 1999, which is heavily influenced by volcanic emissions (Thornton et al., 1999). During August-October 1996, the observations at Tahiti and Easter Island show transport of volcanic SO_2 emissions in the middle and upper troposphere (Thornton et al., 1999), which the model does not capture. Since our model includes only continuous volcanic emission with a yearly resolution, our model fails to simulate variability in volcanic SO2 emissions at higher time resolution."

D. C. Thornton et al., "Sulfur Dioxide Distribution over the Pacific Ocean 1991–1996," Journal of Geophysical Research: Atmospheres 104, no. D5 (March 20, 1999): 5845–54, doi:10.1029/1998JD100048.

• *Page 5849, lines 5–6: TOMAS sulphate lifetime is one third longer than the AeroCom mean. I would not qualify that as "slightly longer".*
Response) Changed from "slightly longer" to "longer".

• *Page 5849, line 7: Are we to understand that weak dry deposition rates explain the longer lifetime of sulphate in TOMAS?*
Response) The longer SO_4 lifetime in TOMAS should be influenced by both weak wet deposition and dry deposition. Both deposition coefficients are small compared to the AEROCOM mean values (0.04 less for wet deposition and 0.027 less for dry deposition). When increasing the dry deposition coefficient to the AEROCOM mean alone, the overall SO_4 lifetime is decreased from 5.6 days to 4.8 days. Doing the same for wet deposition with no change in the dry deposition, the overall SO_4 lifetime decreases from 5.6 days to 4.5 days. We have added the following right after this.

"However, the longer overall sulphate lifetime is contributed by both dry and wet deposition, rather than dry deposition. When increasing the dry deposition coefficient to the AEROCOM Phase 1 mean alone, the overall lifetime is decreased from 5.6 days to 4.8 days. Doing the same for wet deposition with no change in the dry deposition, the overall lifetime decreases from 5.6 days to 4.5 days."

• *Page 5849, line 9: Is the remainder of wet deposition caused by large- scale precipitation? Also, I guess "moist convective clouds" are in fact simply convective clouds.*
Response) Yes, the remainder (73%) is by large-scale precipitation and the moist convective clouds are convective clouds. In case it could be confusing, we have dropped "moist" from moist convective clouds throughout the text. Also, we have included the following in Section 2 to make sure that the model clouds are distinguished into convective and large-scale stratiform clouds.

"In the model, clouds are distinguished into convective and large-scale stratiform clouds."

• *Page 5849, line 13: Sulphate lifetime is given at 5.6 days on line 6 and in Table 5.*
Response) It was typo. It is now corrected to 5.6 days.

• *Page 5850, lines 1–14: Comparing lifetimes of coarse mode aerosols is only meaningful if the models cover the same size ranges. Is that the case here?*
Response) We agree with the reviewer. We have deleted the comparison of our model sea-salt and dust particles lifetime to the AEROCOM Phase 1.

• *Page 5851, line 6: Readers are left to draw their own conclusion, here, so the paper should be more affirmative: is the dry deposition parameterisation in TOMAS better – or equivalently, are dry deposition rates in other models likely overestimated?*

Response) First of all, we'd like to make it clear that this is to explain why dry deposition rate and lifetime differ dramatically between the two models. Unfortunately it would be difficult to judge which parameterization is superior. One might think the TOMAS dry deposition is better, because it is more physically based parameterization (i.e. accounting for a size-dependent resistance and gravitational-settling velocity) than the one used in the bulk aerosol model. However, the dry deposition velocity assumed in the bulk aerosol model could be more comparable to some of the observations, as it is based on the observations from Wesely et al. (1985). More information is required to draw a reasonable conclusion.

About the second question, we can't say whether other models overpredict their dry deposition rates, not only because we don't have any measurement of dry deposition rates to judge and the topic is further complicated because a dry deposition rate is influenced by the dry deposition parameterization as well as many other factors (e.g., boundary layer scheme, and model surface definitions/properties).

• *Page 5896, caption of Figure 6: Are OC surface concentrations given in terms of [C] or [OM]? There is a factor 1.4 between the two in TOMAS.*
Response) The unit is in term of [OM]. The model simulates OM concentrations rather than OC concentrations, as it matters for particle size.

• *Page 5851, line 24: OM aerosol concentrations are high over North Hemisphere continents, including in regions I would not particularly associate with industry or biomass-burning, especially on an annual average (midwest US, central Siberia). Are those biogenic sources?*
Response) Yes. We have included the following sentence in the revised text.

"Due to the SOA formation, the OM concentrations over Midwest US and Central Siberia are also noticeably high."

• *Page 5897, caption of Figure 7: I presume that units are the same as in Figure 6?*
• *Page 5898, caption of Figure 8: Sulphate in ug[S] m-3 as before?*
Response) Thanks for pointing this inconsistency. The unit used in Figure 7 and 8 was $\mu g\ m^{-3}$. The Figure 6 is now also shown in $\mu g\ m^{-3}$ instead of $\mu g\ [S]\ m^{-3}$.

• *Page 5852, line 6: Figure 7 really shows zonal cross-sections – but the vertical aspect of the Figures is not discussed.*
Response) The following sentences are added to describe the vertical distributions.

"The two small spikes shown the EC and OM concentrations between 10° S and 10° N are a result of the injection height used in the biomass burning emissions. Since a significant amount of sulfate and OM are also formed through chemical reactions in the atmosphere, their vertical gradients are relatively small."

"A strong dust plume is shown at around 0°-30° N due to the large Northern African and

Middle East dust emissions, and a small dust plume at around 30° S due to Australian emissions. In the model, the PM10 concentrations in upper troposphere are dominated by sulfate and dust particles."

• *Page 5853, lines 18–20: Isn't that statement in contradiction with the statement on lines 11–14, where TOMAS overestimated sulphate?*
Response) We understand that they may sound as if we are contradicting each other. The referee #2 also raised a question for the line 11-14, and we have modified the manuscript. Please see our response to the referee #2's comments.

• *Page 5854, lines 1–13: The sea-salt evaluation lacks discussion. The overestimations inland are probably due to the model resolution, which would produce an abnormally large transport from the ocean. The low bias in the Tropics is more interesting: perhaps a consequence of poor simulation of near-surface wind speeds in the host model?*
Response) We have added the following discussions in the text.

"The overprediction of sea salt in continents may suggest that ModelE2 has a stronger transport from ocean to in-land, as there is no obvious overprediction over adjacent oceanic sites. The ModelE2-OMA model exhibits a particularly large overprediction over most SH sites. Both models tend to be biased significantly low at some of the sites near the tropics where the observed sea salt concentrations are high. Similar underprediction is also shown in mineral dust (see Fig. 13). This might be due to fast wet scavenging due to overpredicted precipitation in that area (see Fig. 9 in Schmidt et al., 2014)."

• *Page 5854, from line 14: Again, lack of discussion, this time for mineral dust aerosols. It is interesting that TOMAS seems to be doing better than the bulk aerosol model here. Could that better performance be explained by specific differences between the two models?*
Response) We have explained why ModelE2-TOMAS performs better over continents than ModelE2-OMA. The followings are included in the revised manuscript.

"This is due to the emission size assumptions (resulting in more clay emissions than ModelE2-OMA) and the differences in the deposition parameterizations (resulting in slightly longer lifetime for clay particles; ~ 9 days in ModelE2-TOMAS and 6.5 days in ModelE2-OMA)."

• *Page 5855, lines 24-26: Why not show the comparison against IMPROVE where the OC:OM ratio is consistent with the choice made in the model? The other comparison is of little interest and can be removed. Again, the paragraph could discuss the results of the comparison of EC and OC – the comparison is not too bad in fact. Emissions will probably be the main source of error for the networks used here, which are located relatively close to the source.*
Response) We now use OM/OC ration of 1.4 for IMPROVE sites, and Figure 15 has been updated. The following is either newly added or modified in the revised text.

"The simulated EC and OM in both models agree very well at the IMPROVE sites (for OM, LMNB=-0.17 to -0.08; for EC, LMNB=-0.08 to 0.05). Note that we applied an OM/OC ratio of 1.4 to the IMPROVE network to make it consistent with our model assumption, but the IMPROVE OM data provided to us was based on the ratio of 1.8. Over Europe, the model predictions are still reasonable (within a factor of 2-3), but the agreement is slightly worse than the IMPROVE sites. Since these sites are mostly adjacent to the source/emissions, the good agreement suggests that the emission inventory (used in this study) is well represented for these regions."

• *Page 5856, line 7: It does not mean that they don't contribute to PM2.5 – so seasalt and mineral dust aerosols may be partly to blame for the under-prediction.*
Response) Thanks for pointing this out. Doing more investigation, we found that the model PM2.5 in Oceania is little influenced by sulfate or dust and we also found a small error in PM2.5 calculation for the bulk aerosol model. Please see below for the modified text.

"The PM2.5 overprediction in Oceania is mainly due to too much fine mode sea-salt particles (the overall agreement in Oceana is little influenced by sulfate or dust particles). Note that the sea salt comparison to the Miami dataset (in Figures 11) shows severe underpredictions in several sites in Oceania because their concentrations are likely dominantly by coarse mode sea salt particles."

• *Page 5856, lines 16–19: Please give the full details of the satellite products used here: collection for MODIS, version for MISR, and whether level 3 (monthly, gridded distributions) were used.*
Response) We have included the following statement.

"Specifically, we use Terra MODIS Level 3 (MOD08_M3.051), Aqua MODIS Level 3 (MYD08_M3.051), and Terra MISR Level 3 (MIL3MAE4), which are monthly products with 1x1 degree resolution"

• *Page 5856, lines 25–26: This statement is unclear: does that mean "in the cloudfree fraction of gridboxes" or "in cloud-free gridboxes"?*
Response) It is cloud-free gridboxes. The following is now added.

"where clouds are not present." ➔ ""where clouds are not present (i.e. cloud-free grid-box only)."

• *Page 5857, first paragraph: It should be said that the satellite products do not seem to support the bands of large sea-salt AOD at high latitudes. In that respect, TOMAS does better than the bulk model. Also, the quality of the comparison will depend on the host model simulating clear skies in the right regions and seasons.*
Response) First of all, we want to clarify that both models and satellites show an enhanced AOD band in SH high latitude (compared to the neighboring areas). However, the magnitude

of the satellite AOD seems to be in between the TOMAS AOD and the OMA AOD. We agree with the reviewer that the AOD evaluation would be more meaningful when the host model simulates clear skies in right time. Our current model outputs have monthly resolution, but we think sub-daily model outputs using a satellite simulator seems to be more appropriate to evaluate model clear sky predictions with a satellite production.

• *Page 5859, line 15: Number concentrations from the bulk aerosol model could be derived from the simulated mass and prescribed size distributions, but the comparison would probably not be useful.*
Response) It is possible to compute number concentrations from the bulk-aerosol model. However, the number concentrations in bulk model are not useful, as they are quite inaccurate.

• *Page 5861, lines 2–4: Why is that surprising?*
Response) It was surprising because our previous study using GISS-TOMAS (Lee et al., 2009) did not show a large contribution of dust particles to CCN-sized particles. Instead, when including mineral dust, the model CCN(0.2%) is decreased by 10-20% over dust regions due to microphysical feedback by dust particles. The direct source of CCN by dust emissions was less significant. However, ModelE2-TOMAS shows a significant CCN concentration over dust regions. We added the followings in the revised text.

"This is opposite to the results from GISS-TOMAS (Lee et al., 2009), which shows 10-20% reduction in CCN(0.2%) when introducing mineral dust emissions. Despite the direct source of CCN-sized particles from dust emissions, CCN and ultrafine particles that grow to become CCN are scavenged via coagulation with coarse dust particles, and dust particles compete for condensable sulfuric acid, leading to a slower growth rate of ultrafine particles)."

Y H Lee, K Chen, and P J Adams, "Development of a Global Model of Mineral Dust Aerosol Microphysics," Atmospheric Chemistry and Physics 9, no. 7 (2009): 2441–58.

• *Page 5861, line 16: How was the 30% number obtained? By reducing the altitude where simulated aerosol numbers are taken until the agreement with observations is satisfactory? If so, isn't that being too easy on the model?*
Response) "30% reduction of the altitude" was chosen based on the improvement in the model CN seasonality. It indicates a problem in model orographic transport issue in free tropospheric sites, but this might be an inherent problem by the coarse grid resolution in GCMs.

• *Page 5861, line 25 and Figure 21: It is difficult to tell which of the three simulations does best. Is it BASE? If not, wouldn't that be a reason to redefine the scientific configuration of the BASE simulation, at least as far as nucleation is concerned? Figure 24 strongly suggests that LowNUC would make a good BASE.*

Response) We concluded that LowNUC is the better configuration than BASE in terms of predicting number concentration related to nucleation, but we prefer to use "the least perturbed case" as our BASE scenario.

• *Page 5862, line 6: It is difficult to reconcile Figures 21 and 23. The model seems to a have a high bias in aerosol number over Europe - something that Figure 21 doesn't really show. Should we conclude that this is specific to that region?*
Response) The measured size distribution data are obtained from Putaud et al. (2003), while the measured CN concentration used in Fig. 21 are obtained from Spracklen et al. (2010). The temporal coverage between two datasets are not necessarily same. We found out that except for Jungfraujoch, most sites used in Figure 23 have comparable CN concentrations to the ones used in Figure 21. For Jungfraujoch, the total CN from the size distribution data (from June 1997 to May 1998) is about a factor of two lower than the ones used in Figure 21 (from 1995 to 1999, 2003-2007). This seems indicate a strong year-to-year variability in CN at that site. We have included the following statement in the revised text to warn the difference.

"This data is obtained from Putaud et al. (2003) and, for the same sites shown in Figure 21, the temporal coverage used in Putaud et al. (2003) is not necessarily matched with them. Most sites are reasonably close to the dataset used in Figure 21. However, for Jungfraujoch, the total CN concentrations summed from the size distribution data, which covers from June 1997 to May 1998, is about a factor of two lower than the CN shown in Figure 21, which covers from 1995 to 1999 and 2003-2007."

• *Page 5863 and Figures 25 and 26: Both Figures show similar things, but Figure 26 does it better. With Figure 25, I guess the authors wanted to show the extend of observational variability, but that backfires since the observations are shown to provide little actual constraint. So perhaps Heintzenberg et al. (2000) is not suited for this kind of comparison.*
Response) We do not agree with the reviewer's point. Given that the observations compiled by Heintzenberg et al. (2000) are the collection of some 30 years of marine aerosol observations, the large observational variability (shown in the error bar) is somewhat expected. Their data could help to reveal some climatology of marine aerosols, and thus it is useful for GCM evaluation.

• *Page 5863, lines 22–24: It should be easy to check that convective clouds are indeed more frequent in ModelE2.*
Response) This should be easy to check. However, unfortunately, we don't have an access to GISS GCM II' output (GISS-TOMAS has been retired), so the exact comparison is not possible. Based on other relevant parameters (and some personal communications), we can confirm that the current version of model has more frequent moist convective clouds.

• *Page 5864, line 6: Does the "rest" cover non-marine environments, non- boundary layer locations, or both?*

Response) All CCN data compilation from Spracklen et al. (2011) is in the boundary layer. The rest refers to non-marine boundary layer. However, we have decided to use continental boundary layer (CBL) instead. We have modified the following in Section 5.6.

"In Fig. 27, the CCN data is divided into two groups: CCN in the MBL (marine boundary layer) and CCN in the CBL (continental boundary layer). Note that all CCN measurements used here are in the boundary layer.

• *Page 5864, lines 19–21: Again, the authors rely on the fact that the observational dataset does not provide a strong constraint. This is a bit unfortunate – could a better use of the dataset be made?*
Response) Unlike CTM models, the observations with short duration can't be a strong constraint for GCM. Since climatological CCN measurements are very rare, we performed the evaluations to the short-term CCN measurements, as this is still very useful for a quick check.

• *Page 5866, line 25–28: Saying that aerosol modelling produces large differences is not uninteresting, but if host model impacts had also been discussed in details in the paper, it would have been possible to tell which of the two is the dominant factor. Also, it is possible that the two aerosol schemes are affected by the host model in different ways. Host model effects would then be misattributed to aerosol modelling differences.*
Response) We agree with the reviewer that the impact of host model on aerosol model is an interesting subject, but this is not a scope of this paper. Our main goal of this paper is to evaluate ModelE2-TOMAS.

3 Technical comments
Page 5840, line 22: Typo: "InitiAtive".
Page 5842, line 12: Typo: Stier et al., 2005.
Page 5844, line 5: Delete "that".
Page 5886, caption of Table 5, typo: "standard" deviation.
Page 5853, line 1: The sentence does not read well. I suggest: "For details of the GBD PM2.5 dataset, the reader is referred to..."
Page 5859, line 4: Typo: "underprediction"
Response) Thank you for catching those mistakes. All Technical comments are now corrected as suggested.

Interactive comment on **"ModelE2-TOMAS development and evaluation using aerosol optical depths, mass and number concentrations"** by **Y. H. Lee et al.**
Anonymous Referee #2

Response) We thank the reviewer for their constructive comments and suggestions and have made several changes to the paper to address the issues raised. Reviewers' comments are shown in italics with our response shown after each.

This manuscript describes the implementation of a new aerosol microphysics module (TOMAS) into the ModelE2 general circulation model, contrasting in detail various aspects of the new scheme with those for the existing mass-based aerosol scheme. The paper then presents an evaluation of the ModelE2-TOMAS simulated aerosol properties against an impressive number of observational datasets covering aerosol optical properties, mass concentrations and number concentrations. The paper is indeed appropriate for publication in GMD, and will provide a very useful reference for users of the model to understand the details of the new scheme and its expected skill against these benchmark observational datasets. However, although the Figures and results sections are well presented, the Abstract needs some attention and some aspects of the Introduction section require some correction which I have identified in my comments below.
As per the interactive comment from the Executive Editor, the manuscript also requires the addition at the end of the paper of a "Code availability" section giving the information of how the code for the model can be made available on request. http://www.geoscientific-model-development.net/submission/manuscript_types.html. Overall however the paper is a worthy addition to GMD and the authors are to be commended on a very comprehensive assessment of the aerosol properties simulated by the model. I therefore recommend the paper be published once these minor revisions have been made.

1) Title – I would suggest to replace "using" with "of simulated" which better describes the evaluation carried out in the paper
Response) Title has been changed to

"Evaluation of the global aerosol microphysical ModelE2-TOMAS model against satellite and ground-based observations"

2) Authors – I was surprised to see that there were only 3 co-authors on this paper, which seemed to bring together the aerosol microphysics module, the general circulation model, use a range of emissions datasets and compare to a large number of observations. Are there any model developers or observational PIs who should also be offered co-authorship to recognise their contribution here? Does the GISS model have any "Publication Policy" to provide guidance on how best to recognise such contributions? Do the observational datasets used have any data policy about offering co-authorship? I am aware that some monitoring networks require that co-authorship be offered for publications using their observational datasets.

Response) First of all, we provided the acknowledgements to the observational PIs after communicating with them individually and two GISS personnel who have provided technical support during the model development. Y.H. Lee performs the ModelE2-TOMAS development and evaluations. Y.H.Lee and P.J. Adams at Carnegie Mellon University developed the Fast TOMAS microphysics modules. D.T. Shindell at NASS GISS supports the development and evaluations of ModelE2-TOMAS. ModelE2-TOMAS is linked with online gas chemistry model and uses some existing aerosol module that D.T. Shindell have developed/involved as a main developer. Additional GISS personal have of course contributed to other portions of the model, but it is not customary at GISS to include co-authors who have not contributed more directly to the particular investigation of a given paper. Similarly, observations are cited whenever used, acknowledging those who made the measurements.

3) Model naming - Is there a recognised acronym for the GISS ModelE bulk aerosol scheme that could be used? In many parts of the text there is the phrase "bulk aerosol model" or "bulk aerosol scheme" which could usefully be abbreviated to BAM or BAS for example. Also the abstract explains that the TOMAS scheme presented is the computationally efficient 15-bin version of TOMAS. It would be useful if this was reflected in the acronym for the aerosol scheme. Is there also an existing acronym for this "fast" version of TOMAS that could be used e.g. TOMASf or TOMAS15? A related comment is also that many of the features of the global aerosol distribution are affected by parameters within the other parts of the general circulation model, and consequently I would recommend to refer to ModelE2-BAM/ModelE2-BAS in the text describing the results and evaluation. This also goes for the microphysical scheme which I would recommend to refer to as ModelE2-TOMASf or ModelE2-TOMAS15. It is correct to just refer to BAM or TOMAS when describing the aerosol scheme itself but when presenting aerosol properties simulated in the GCM then one could use ModelE2-BAM or ModelE2-TOMAS.

Response) We now use ModelE2-OMA for the bulk aerosol model in the revised manuscript and ModelE2-TOMAS when presenting the results. Although we think the reviewer's suggestion for ModelE2-TOMAS15 might be a good idea, we did not further distinguish ModelE2-TOMAS15 from ModelE2-TOMAS because a) using ModelE2-TOMAS15 in the results section may bring potential confusions for readers; and b) TOMAS15 becomes a default configuration in ModelE2-TOMAS. We have added the following in the Section 3.

"In this paper, we used TOMAS with 15 bins covering 3 nm to 10 μm (TOMAS15; see Table S1 in the supplementary materials): 3 bins cover from 3nm to 10 nm, 10 bins from 10 nm to 1 μm and the last two bins from 1 μm to 10 μm. The TOMAS15 version becomes a default model configuration for ModelE2-TOMAS, so we will be continuously refereed to as ModelE2-TOMAS throughout the paper."

4) 1st section of the Abstract need attentions and some quantitative statements required. The first 3 sentences of the abstract contain too much specifics and seem out of place here. The authors need to re-draft this first part of the abstract to give the overview of the aims of the

paper rather than this level of detail about the new aerosol scheme. I would suggest to move the 4th sentence to instead be at the start of the abstract. Perhaps the existing first 3 sentences could even be removed – or else condensed into a single sentence giving brief general explanation of the microphysical scheme. The sentence beginning "The TOMAS model successfully captures observed aerosol number...." and other statements would be much improved with some measures of skill against the observations. The Figures have a good set of bias and correlation measures presented and I suggest to cite some of these in the Abstract to give some quantitative metrics to back up the statements made about the model skill. The sentence "With TOMAS, ModelE2 has three...." seems out of place in the Abstract – suggest to remove it.

Response) We have modified the Abstract as below.

"The TwO-Moment Aerosol Sectional microphysics model (TOMAS) has been integrated into the state-of-the-art general circulation model, GISS ModelE2. This paper provides a detailed description of the ModelE2-TOMAS model and evaluates the model against various observations including aerosol precursor gas concentrations, aerosol mass and number concentrations, and aerosol optical depths. Additionally, global budgets in ModelE2-TOMAS are compared with those of other global aerosol models, and the ModelE2-TOMAS model is compared to the default aerosol model in ModelE2, which is a One-Moment Aerosol (OMA) model (i.e., no aerosol microphysics). Overall, the ModelE2-TOMAS predictions are within the range of other global aerosol model predictions, and the model has a reasonable agreement (mostly within a factor of two) with observations of sulphur species and other aerosol components as well as aerosol optical depth. However, ModelE2-TOMAS (as well as ModelE2-OMA) cannot capture the observed vertical distribution of sulphur dioxide over the Pacific Ocean possibly due to overly strong convective transport and overpredicted precipitation. The ModelE2-TOMAS model simulates observed aerosol number concentrations and cloud condensation nuclei concentrations roughly within a factor of two. Anthropogenic aerosol burdens in ModelE2-OMA differ from ModelE2-TOMAS by a few percent to a factor of 2 regionally, mainly due to differences in aerosol processes including deposition, cloud processing, and emission parameterizations. We observed larger differences for naturally emitted aerosols such as sea salt and mineral dust, as those emission rates are quite different due to different upper size cutoff assumptions."

5) Merging Tables 3 and 4 together into one table for DMS, SO2 burden & budget

I suggest to merge Tables 3 and 4 together so that the reader can easily compare the DMS and SO2 burdens and budgets between the ModelE2 runs with the bulk and microphysical schemes.

Response) We have merged the two tables.

6) 2nd paragraph of Introduction needs quite some revision.

This para mentions 3 categories of aerosol microphysics model – moment, modal and sectional. But I'm a bit puzzled by what is meant by "moment". It is said that "moment-based methods track lower-order (radial) moments of a size distribution." My understanding of aerosol microphysics models is that they can be categorized as either modal and sectional. And that the radial moment tracked by the scheme then describes which variables are treated

prognostically by the model. Both modal and sectional schemes can be either single-moment or double-moment. TOMAS is a double-moment sectional scheme for example. I'd recommend the authors re-write this paragraph with this classification. I would remove the text "moment, " from the 1st sentence and replace "In general, moment-based methods track lower-order (radial) moments of a size distribution, and modal-based methods.." with "Modal methods...". Suggest then to replace "represent a mode" with "represent a subset" later in that sentence. In the 3rd sentence suggest to replace "predicting the amount of" with "representing" and re-write the last 2 sentences explaining that one can have single-moment, double moment or triple-moment schemes with reference to existing models which have these approaches.
Response) We have modified this paragraph as below.

"Aerosol microphysics models can be broadly categorized into modal and sectional methods, depending on how they represent the aerosol size distribution. In general, modal-based methods use an analytical function (e.g. a lognormal distribution) to represent a subset of the particle population. Sectional methods represent a size distribution by predicting aerosols in several size sections or "bins". Additionally, sectional and modal methods may differ from each other in numerous ways, including the number of moments of the size distribution that are tracked in each section or mode."

7) Introduction 3rd para – 1st sentence – further to my recommended changes above here I suggest to replace the existing text "(i.e. zeroth moment)" with "(i.e. zeroth radial moment)" and replace the existing text "mass (i.e. 1st mass moment)" with "mass (i.e. third radial moment)". The current text is confusing because the sentence could confuse the reader with the use of mass moments. My suggested revised text just refers to radial moments as they are the usual one referred to in terms of size distributions.
Response) We do not think "mass moment" is particularly confusing, but we have included "3rd radial moment" in order to help some readers.

"… both aerosol number (i.e. 0th moment) and mass (i.e. 1st mass moment or 3rd radial moment) in each size section"

8) Introduction 3rd para – 3rd sentence – as per my comment 6) I suggest here to not consider moment methods separately from modal and sectional methods. Suggest to replace "The modal and the moment-based approaches are..." with "Modal approaches are...."
Response) Changed as suggested.

9) Introduction 4th para – I'd suggest to reword the sentence beginning "Despite the accuracy..." – perhaps shorten that sentence to instead say: "Despite the accuracy in predicting aerosol microphysical processes, the original version of TOMAS has a heavy computational burden." Then in the sentence after that I'd suggest to replace "more computationally efficient" with "less computationally expensive configurations..".
Response) Changed as suggested.

10) Introduction 5th para – the 1st sentence beginning "Since uncertainties..." seemed out of place here. I'd suggest to start that para with the current 2nd sentence changing the start of it from "Therefore, here we..." to "Here, we... I think the current 1st sentence would fit well at the end of the paragraph changing the start of it from "Since uncertainties in..." with "We also note however that uncertainties in...". I would also reword from the current "...come from not only aerosol modelling itself but..." with "come not only from aerosol modelling but..." and finish the sentence after the text boundary layer, and advection)" – delete the text "it is important to include the improvements in both aerosol modelling and the other parts of GCM." as that's implied already in the rest of that sentence.
Response) Changed as suggested. However, we modified the last sentence to the following.

"We also note that it was important to implement the TOMAS aerosol model into the ModelE2 host model because uncertainties in the estimates of aerosol forcing come not only from aerosol modelling itself but also other parts of the host GCM (e.g., cloud physics, planetary boundary layer, and advection)."

11) Introduction 6th para – suggest to replace "that has a goal of understanding" with "which aims to understand"
Response) Changed as suggested.

12) Introduction 6th para – suggest to shorten substantially the sentence beginning "The model description...." to instead simply say something like "Here we give a detailed description of ModelE2-TOMASf and evaluate simulated aerosol mass, number and optical depth against those from ModelE2-BAM (Schmidt et al., 2014) and observations." The sentence afterwards should have a citation for the expected paper if it is already well advanced in its preparation. If not then the sentence should be removed. With that re-worded sentence the next sentence beginning "In this paper, as a comparison with TOMAS, we include..." can be deleted.
Response) Changed as suggested.

13) Introduction 6th para – Be clear when you're referring just to a description of the aerosol scheme and where it's describing the full model ModelE2-TOMAS. For example in the sentence beginning "Section 2...." when you say "including the bulk aerosol model" I suggest you say here the bulk aerosol scheme".
Response) Changed as suggested.

14) Introduction 6th para – Insert "the" between "design of" and "simulations".
Response) Changed as suggested.

15) Introduction 6th para – sentence beginning "Section 5.." can be made shorter and easier to read by deleting "the" between "presents" and "global budgets" and replacing "and the evaluation of the TOMAS and bulk aerosol scheme model..." with "and evaluates ModelE2-TOMAS and ModelE2-BAM..."
Response) Changed as suggested.

16) Section 2 – 1st para – suggest not to begin a sentence with "The newest version of" as this will rapidly become not the case as time passes... Also this sentence is clumsily worded and makes this whole para difficult to read. Suggest to re-write that sentence to instead be something like "In this section we briefly describe ModelE2 (Schmidt et al., 2014), the GISS climate model used to perform simulations for the Coupled Model Intercomparison Phase 5 (CMIP5)". Suggest to refer here to Taylor et al. (2012) when CMIP5 is mentioned. With this re-wording the later sentence "A brief description of ModelE2 is given here." can be deleted.
Response) Changed as suggested.

17) Section 2 1st para – the Prather (1986) paper is missing in the References – please add.
Response) Added.

18) Section 2 1st para – replace "hydroscopic" with "hygroscopic".
Response) Thanks for catching that. It has been corrected.

19) Section 2.1 title – this para is not really describing the bulk aerosol scheme but rather the way it is implemented within ModelE2. Suggest to change the title to "Implementation of the bulk aerosol scheme in ModelE2" or "ModelE2-BAS description" or similar.
Response) The title has been changed to "ModelE2-OMA description".

20) Section 2.2 last sentence – reference is missing for "(2002)" – also the authors need to add sentence giving brief explanation of this – cloud droplet number concentration as a function of aerosol number or mass?
Response) It is now corrected as follows.

"Aerosol indirect effects are based on an empirical parameterization that compute cloud droplet number concentrations as a function of aerosol mass (Menon et al, 2002; 2008). "

21) Section 3 title – again this section is describing the overall model not just the TOMAS aerosol microphysics scheme. As in my comment 19) I suggest to have this section as "Implementation of TOMASf aerosol microphysics scheme into ModelE2"
Response) We have changed the title to "ModelE2-TOMAS description".

22) Section 3 1st para – 1st sentence – see my comment 7) above I suggest to refer to radial moments only throughout to avoid confusion. Suggest to replace the existing text "(i.e. 0th moment)" with "(i.e. zeroth radial moment)" and replace the existing text "mass (i.e. 1st mass moment)" with "mass (i.e. third radial moment)".
Response) Please see our response to comment 7.

23) Section 3 1st para – 3rd sentence – suggest to replace "the TOMAS model tracks ten quantities for each size bin..." with "ten quantities are tracked for each size bin..."

Response) Changed as suggested.

24) Section 3 1st para – that 3rd sentence is very long and needs to be re-written as at least 2 sentences. Also please clarify what is meant by "the ammonium mass is diagnosed in each size bin based on sulphate mass..." Later in that sentence you mention that the scheme tracks aerosol ammonium so is it transported or diagnosed?
Please take care with the wording here when revising the manuscript.
Response) The sentence has been broken into two sentences. For the sentence starting with "the ammonium mass is diagnosed in each size bin~", we meant that size-resolved ammonium is determined by sulfate mass in each bin, as it is assumed to be fully neutralized with sulphate. We have modified as follows.

"In TOMAS, all ammonia becomes aerosol ammonium until sulfate is neutralized to form ammonium sulfate; the excess ammonia after neutralization remains as free gas-phase ammonia. The aerosol ammonium is partitioned into each size bin in proportion to the sulfate mass. However, ammonium is not size-resolved (i.e., bulk tracer) for purposes of model processes outside of TOMAS such as advection and deposition.

25) Section 3 1st para – you say "TOMAS uses a moving sectional approach to treat water uptake" – Please can you clarify this – I assume this moving sectional approach deals with the aerosol dynamics. Isn't that moving sectional approach based on dry size – what is meant here? Please re-word to clarify.
Response) Unintentionally it was neglected to provide the TOMAS size boundary definition in the original manuscript. We have been clarified the part as below.

"The size section boundary is defined by dry particle mass, such that addition or removal of aerosol water mass does not move particles between sections."

26) Page 5839 line 1 – insert "alterative" between "Several" and "nucleation schemes".
Response) Added.

27) Page 5839 line 6 – you have already introducted the faster configuration of TOMAS on page 5835 so you don't need this wording here – please reduce this sentence. I have also suggested to give it a name such as "TOMASf" or "TOMAS15". So please replace "With the development of computationally efficient TOMAS models (i.e. Fast TOMAS), the TOMAS microphysics module became more flexible...." with something like "As well as being computationally faster, the development of TOMASf (see section 1) also made the scheme more flexible...."
Response) It has been shortened to "With the development of computationally efficient TOMAS models (i.e. Fast TOMAS)," ➔ "With Fast TOMAS models,"

28) Page 5839 lines 10 to 12 – this sentence says "TOMAS" much too many times. Suggest to delete the "compared to the original TOMAS" at the end as that's implicit in the wording

already – then can delete "in TOMAS" after "lower size cutoff" – again it's clear already you're referring to TOMAS.
Response) Changed as suggested.

"For the size range of 10 nm to 10 μm, the original TOMAS uses 30 bins, and the Fast TOMAS uses 15 bins or 12 bins, which reduces the computational burden by 2-3 times."

"The lower size cutoff can also vary from 10 nm to 3 nm or from 10 nm to 1 nm (Lee et al., 2013b)."

29) Page 5839 lines 12-19 – these sentences would be much better illustrated in a Figure showing the different size bin configurations across the size spectrum. Perhaps they don't even need to be shown at all? Is this already described elsewhere in another paper?
Response) Previous TOMAS publications present the model configurations in a Figure and Table. However, none of them present the exact model configuration used in GISS ModelE2. So we include a table that describes the model configuration in Supplemental material.

30) Page 5839 line 21 to 23 – this para needs some rewording – the current text says "condenses" but the sentence describes aqueous sulphate production so condensation is not the right term. Suggest to change "First, the TOMAS model condenses the sulphuric acid formed from aqueous oxidation by hydrogen peroxided (H2O2) directly onto sulphate aerosols in ambient air..."
with "First, ModelE2-TOMAS adds sulphate mass produced in the aqueous phase directly to the bin-resolved sulphate mass in ambient air...."
Response) The sentence is now modified with the reviewer' suggestion.

"First, ModelE2-TOMAS adds sulphate mass produced in the aqueous phase directly to the bin-resolved sulphate mass in ambient air rather than maintaining a separate tracer for dissolved sulphate"

31) Page 5839 line 25 – replace "...sulphate formed from aqueous oxidation should release to the air only when the cloud water evaporates" with "...sulphate formed in the aqueous phase will only be released as interstitial aerosol when the cloud water evaporates".
Response) Changed as suggested.

32) Page 5839, line 28 – replace "For in-cloud scaenging, modified Kohler theory is used..." with "Modified Kohler theory is used..." – this sentence is referring to activation not in-cloud scavenging.
34) Page 5840, line 1 – suggest to replace "activate and are subject to" with "activate (i.e. contribute to cloud droplet number) and which are subject to..."
35) Page 5840, lines 3-7 – this sentence needs to be clarified – is this referring to scavenging or activation or both? Also replace "hopple" with "Hoppel".
Response to the comment 32, 34, and 35)

First of all, nucleation scavenging implies activation. Activation described in this section is only for in-cloud scavenging, not determining cloud droplet number concentration (i.e. aerosol-cloud interaction). Although activation (=nucleation scavenging) is a part of in-cloud scavenging and thus some of the reviewer's suggestions are not necessary, we have replaced "activation" to nucleation scavenging to avoid confusion. Also, we have included the following description of activation used in ModelE2-TOMAS.

"To compute the cloud microphysics properties as a function of aerosols (i.e., the aerosol-cloud interactions), ModelE2-TOMAS uses a physical-based activation parameterization from Nenes and Seinfeld (2002). A critical supersaturation is computed in the parameterization using a model updraft velocity that is computed based on a large-scale vertical velocity and sub-grid velocity."

33) Page 5839, line 29 – "for activation of each size section..." somewhere in this para the kappa values used for each component need to be given.
Response) We have provided the kappa values for each soluble aerosols in the revised manuscript.

"To determine activation, we assume kappa values of 0.6 for sulfate, 1.28 for sea-salt, and 0.15 for hydrophilic OM."

36) Page 5840, within the description of ModelE2-TOMAS there is no mention of how aerosol-radiation interactions (i.e. aerosol direct radiative effects) are represented. Do the size-resolved aerosol information feed into aerosol scattering and absorption in the ModelE2 radiative transfer model? There needs to be at least a sentence or two describing what is done here.
Response) We have included the followings at the end of Section 3.

"In ModelE2-TOMAS, Mie theory is used to compute size-resolved AOD. For each grid cell, particle compositions (including aerosol-water) in each individual size bin are used to compute the volume-averaged refractive index and optical properties based on Mie theory. The optical properties are used to compute aerosol optical depth taking into account the aerosol concentration."

37) Page 5840, section 4 title – suggest to replace "Simulation setup" with "Description of the simulations"
Response) Changed as suggested.

38) Page 5840 line 18 – replace "2000" with "year-2000" and replace "CMIP5" with "ACCMIP".
Response) Changed as suggested. But we stick with CMIP5 instead of ACCMIP because this emission inventory has been originally provided for CMIP5.

39) Page 5840 lines 21-22 – give the original reference for continuous volcanic emissions from

GEIA – is it the Andrea & Kasgnoc (1998) dataset that you mean here?
Response) Yes. The reference has been added.

40) Page 5842 – title for section 4.2 – suggest to change to "The ModelE2-TOMAS run setup"
Response) Changed as suggested.

41) Page 5842 – line 2 – please give reference for the MERRA re-analysis fields.
Response) The reference has been added.

42) Page 5842 – lines 14-18 – reword this sentence to make it easier to read. Suggest to replace "Note that an emission size distribution used for the biofuel emissions is generally the same" with "Note that although the emissions size distribution for biofuel emissions are generally assumed to be the same.." and delete "as their burning materials are the same". Then replace ", but our model assumes the.." with ", in the ModelE2-TOMASf run we assume the ..." and replace "follow the fossil fuel because the CMIP5 emissions does not..." with "follow the finer fossil fuel size settings because the ACCMIP emissions do not..."
Response) We followed the suggestions from the reviewer, but we kept "CMIP5 emissions" instead of "ACCMIP emissions".

43) Page 5842 – line 22 – replace "sulphate and carbonaceous aerosols" with "primary sulphate and carbonaceous emissions" so it is clear that you mean the assumed size for the emissions.
Response) Changed as suggested.

44) Page 5842 – line 23 – here you give the Lee et al. (2013) reference for GISSTOMAS but earlier in the manuscript you cite Lee and Adams (2010) for GISS-TOMAS – which is the best one to refer to – presumably the same?
Response) We have several references available for GISS-TOMAS, and we have cited the most relevant ones for the topic discussing there. For example, when Fast TOMAS microphysics scheme is being discussed, Lee and Adams (2010) is the most appropriate reference. For the emission size assumption here, Lee et al. (2013b) is the appropriate one.

45) Page 5843 – lines 1-2 – delete the sentence beginning "Whereas the GISS-TOMAS does not...." – you've already said that in point 1).
Response) Deleted as suggested.

46) Page 5843 – line 2 – suggest to replace "Additionally the" with "Note also that the" – that reads better in my opinion.
Response) Changed as suggested.

47) Page 5843 – lines 6-8 – Presumably this info is for SO2 emissions here right? If so please put this information into Table 3 rather than writing it as a sentence.
Response) Table 3 is modified to present the information, and the sentence is deleted.

48) Page 5843 – lines 12-13 – replace "and thus they are excluded" with "and is therefore not received by any of the TOMAS size bins."
Response) Changed as suggested.

49) Page 5843 section 4.3 title – suggest to change to "The ModelE2-BAS run setup" or similar.
Response) Changed as suggested.

50) Page 5843-5844 section 4.3 1st sentence – the 1st half of this sentence can be deleted as you've already explained earlier in the article and so I suggest to start this as "To compare to the ModelE2-TOMAS run, we also ran the ModelE2-BAS model nudged to the same MERRA reanalysis meteorology with 3 years spin-up."
Response) Changed as suggested.

51) Page 5844 section 4.3 2nd sentence – suggest to replace "The natural emissions and emissions-relevant setup are not necessarily the same between the bulk and TOMAS models. This is because we maintain..." with "However, the natural emissions and associated settings are not always the same between the two models because we chose to maintain...."
Response) Changed as suggested.

52) Page 5844, line 5 – Suggest to replace "Here, we note that the differences..." with "To assist the interpretation of the results, we briefly summarize the differences...."
Response) Replaced.

53) Page 5844, line 14 – insert "whereas ModelE2-TOMAS assumes only 1%" after "(Dentener et al., 2006)".
Response) Added.

54) Page 5845, lines 13-14 – suggest to replace "In case of the bulk aerosol model in ModelE2.." with "For ModelE2-BAS.." or similar acronym.
Response) Replaced with "For ModelE2-OMA".

55) Page 5845, line 16 – delete "newer".
Response) Deleted.

56) Page 5845, line 20 – replace "in both models." with "in both simulations."
Response) Replaced.

57) Page 5846, line 7 – why are the H2SO4 and SOA precursor gas budgets in the text rather than in a Table. It would be much better to tabulate them alongside the DMS and SO2 in Table 3.
Response) Because H_2SO_4 and SOA do not have a budget for most processes shown in Table 3, we do not present their budgets in Table 3.

58) Page 5846, lines 10-11 – is this the same for the bulk aerosol scheme? How is SOA handled? State if this is the same or not in the text.
Response) We have included the SOA description in the bulk aerosol model (ModelE2-OMA) in Section 2.1, and their SOA budget in Section 5.1.

In Section 2.1,
"The secondary organic aerosol formation is computed using a two-product model with isoprene, monoterpenes, and sesquiterpenes as SOA precursors (described in Tsigaridis and Kanakidou, 2007)."

In Section 5.1,
"For ModelE2-OMA, the total production rate of SOA is 14.6 Tg yr^{-1}. This is quite comparable to ModelE2-TOMAS, which treats SOA much more simply and has a production rate of 17.1 Tg yr^{-1}. Global burden of SOA in ModelE2-OMA is 0.6 Tg yr^{-1}."

59) Page 5846, lines 13-14 – "and OH and NO3 concentrations" – state in brackets whether these oxidants are interactive or prescribed.
Response) Changed to " and **interactive** OH and NO3 concentrations".

60) Page 5847, line 2 – But what about the chemical sinks. That authors should comment here. Do the ModelE2-TOMAS and ModelE2-BAS have the same approach for oxidants and hence the same chemical sinks? This should be stated here as it could make a big difference.
Response) Thank for pointing out this. We have included the following information in Section 3. Note that we did explain the difference in SO_2 oxidations in Section 5.1 (from Page 5845; Line 23 to Page 5846 line 3).

"ModelE2-TOMAS is coupled to the same gas chemistry model (Shindell et al., 2013) as ModelE2-OMA. So the oxidation fields used for sulphate formation are from the chemistry model. However, unlike ModelE2-OMA, the photolysis rates are not affected by aerosols."

61) Page 5848, line 10 – replace "that the model does not capture" with "that neither of the simulations is able to capture".
Response) This part has been further modified in the revised manuscript.

"Except at Hawaii, our model do not capture the enhanced SO_2 concentrations in the boundary layer shown in the observation, even though the model DMS is quite well captured."

62) Page 5849, line 8 – add "in ModelE2-TOMAS" after "98% of the total deposition"
Response) Done.

63) Page 5849, line 13-17 – rewrite this sentence to shorten it. How about Note that the GISS-

E2-R-TOMAS simulation used for ACCMIP is almost identical model except for the ...."
Response) The sentence has been shorten to below.

OLD : "Note that GISS-E2-R-TOMAS included in Shindell et al. (2013) is a basically identical model to the ModelE2-TOMAS, but the sulphate budget in the two TOMAS models is different because the sulphate and DMS emissions assumptions used in GISS-E2-R-TOMAS are similar to those used in the bulk aerosol model in this paper."

New: "Note that the GISS-E2-R-TOMAS model used for ACCMIP is almost identical to the ModelE2-TOMAS evaluated here except for the sulfate modeling. The sulphate and DMS emissions used in GISS-E2-R-TOMAS are identical to those used in ModelE2-OMA in this paper."

64) Page 5850, lines 11-12 – "has a significantly faster removal rate and increases the mean value" – is this for dust or for sea-salt – or for both – please clarify in that sentence.
65) Page 5850, line 14 – Another issue is that, for components in the coarse part of the particle size range, some of the variation between the models for burden and lifetime can be explained by differences in the upper size cut-off used in the models. Please add a sentence at the end of this para noting this in the interpretation.
66) Page 5850, line 19 – insert "and much lower than the AeroCom median value" after "than in the TOMAS model."
67) Page 5850, line 22 – insert "and a factor of two lower than the AeroCom mean after "compared to TOMAS".
Response to 64~67) We no longer compare our model sea-salt and dust particles lifetime to the AEROCOM Phase 1 in the revised manuscript. So those sentences have been deleted.

68) Throughout results sections replace TOMAS with ModelE2-TOMAS.
Response) We have updated the text as suggested.

69) Page 5853, lines 11 to 14 – I don't understand why there is such a big difference between the surface SO4 in the 2 model runs in these SH marine regions. Is this related to the differences in the treatment of aqueous sulphate production and wet removal between the ModelE2-BAS and ModelE2-TOMAS runs?
Response) We intended to state that TOMAS overprediction is quite pronounced in a few SH marine region compared to other sites, but there is no particularly big difference between the two models in the SH marine regions. TOMAS generally predicts a higher SO4 concentration than the bulk aerosol model in most regions. We decided to delete this sentence, as it can be misleading.

70) Page 5854, line 18 – insert "for both models" after "severe underprediction"
Response) Inserted.

71) Page 5855, lines 1-4 – the observations at Heimaey, Iceland show a big peak in June or so

that is not seen in other months – what is the cause of the higher dust emissions here? Are there papers that have attributed this to a spike in emissions from certain sources? Add reference to these.
Response) We have added the followings in the revised manuscript.

"The observed peak concentration at Heimaey Iceland is the second highest after Sal island. Our models underpredict this site severely probably because our dust emission parameterization is not designed to simulate a dust event in humid areas such as Iceland. Prospero et al. (2012) points out that dust emissions at high latitudes (e.g., Alaska and Iceland) are mostly due to individual dust events or single seasons and link large dust events at Heimaey Iceland during 1997 to 2002 with glacial outburst floods."

Joseph M. Prospero, Joanna E. Bullard, and Richard Hodgkins, "High-Latitude Dust Over the North Atlantic: Inputs from Icelandic Proglacial Dust Storms," Science 335, no. 6072 (March 2, 2012): 1078–82, doi:10.1126/science.1217447.

72) Page 5855 line 10 – you state this may be showing the dust emission are too low – but could it alternatively (or as well as) be that there is too rapid removal in the model? If so insert "or the removal timescale is too fast" after "are too low".
Response) We do not think this is due to too rapid removal. If there is rapid removal, the total deposition fluxes disagree more severely in downwind. Figure 14 does not seem to support that.

73) Page 5857 lines 28-29 – "indicating a possibility of aerosol emissions being underestimated in these regions". That's a bit speculative. Couldn't it also be that something in the model that could be causing the bias? You need to give a bit more to back up your statement here. Are there references which have also shown this similar bias in other models?
Response) The ACCMIP models are also shown the underprediciton in these regions (Shindell et al, 2013). We have modified the following sentence (bold for the newly added part).

"Both models show lower AOD over China, India, and biomass burning regions **and a similar underprediction is shown by the ACCMIP models (Shindell et al., 2013)**, indicating a possibility of aerosol emissions being underestimated in these regions."

74) Page 5858 line 28 – replace "In contrary" with "By contrast"
Response) Replaced.

75) Page 5859 line 4 – "undeprediction" –> "underprediction".
Response) corrected.

76) Page 5859 line 18 – "particles with diameters" –> "particles with dry diameters" in both CN3 and CN10 definitions. That's certainly how its measured – please can you confirm whether

your model values are based on dry or wet diameter.
Response) It is based on dry diameter. We have changed the definition as below.

"CN3 (particles with dry diameters larger than 3 nm), CN10 (particles with dry diameters larger than 10 nm), and CN100 (particles with dry diameters larger than 100 nm)"

77) Page 5859 line 22 – I would delete the word "obviously" – it is worth stating this – it may not be obvious to some readers.
Response) Deleted.

78) Page 5859 lines 26-28 – you can see this in the ratio of CN3 to CN100. For the base case, CN3 is about a factor of 40 higher than CN100 and only about a factor 6 higher in the surface layer. In the LowNUC and NoNUC these ratios shift substantially. Worth stating this in the text.
Response) Thanks for the suggestion. CN70 and J3 are better parameters than CN70 and CN3, so we have included the following.

"We can see this using the number budgets in Table 8. The increase in CN70 with the BASE case nucleation (i.e., BASE CN70 – NONUC CN70) is 51 cm^{-3} for the nucleation rate (J3) of 0.131 cm^{-3} s^{-1} and that with the LowNUC case nucleation, 37 cm^{-3} for J3 of 0.013 cm^{-3} s^{-1}. In the BASE run, J3 is 10 times higher but the CN70 increase by nucleation is only ~1.4 times higher than those in the LowNUC run."

79) Table 9 – suggest to delete the 1st column "Emission rate". You don't refer to these values in the text and it is not obvious why the values are given here.
Response) We do not delete those value, as it is important to show the contribution of primary emissions to total number source.

80) Page 5860 – line 1 – you have "Aerosol number burdens" but the values are given in particles per cm3 which suggests they are concentrations not burdens. Burden implies it's a column-integrated property which would have be given per unit area rather than per unit volume. Please give a different term.
Response) We corrected the term to "aerosol number burden normalized by tropospheric volume". This was done because an actual number burden is too high.

81) Page 5860 – lines 17-19 – Change "Obviously when turning nucleation off, CN3 is very close to CN10" to "When nucleation is switched off CN3 is very close to CN10 near to the surface (Figure 19 g and h)." Again this is not necessarily obvious to the reader – it is worth stating.
Response) We changed to the following.

"When nucleation is switched off, CN3 is very close to CN10 near to the surface (Figs 19 g and h) because nucleation contributes most CN between 3 nm to 10 nm"

82) Please add labels a), b), c) … to all Figures with more than 1 panel so that it is possible to refer to them in the text. Figures 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 15, 16, 17, 18, 19, 20, 22, 23, 24, 25, 26 and 28.
Response) We have modified all figures.

83) Page 5860, line 19 – insert "primary" between "anthropogenic" and "emissions" – I'm assuming that's what was intended here.
Response) Changed as suggested.

84) Page 5861, lines 2-3 – You say "Rather suprisingly, dust particles in our model contribute to CN100 quite significantly" – why is that surprising? It is not obvious to the reader why that is surprising. You should explain why it is surprising or else delete the sentence.
Response) The same comment is also from the reviewer #. Please see our response to the reviewer #1.

85) Page 5861, line 22-24 – the use of whisker lines in Figure 21 is confusing. The reader will assume that the circle in the centre of the whisker is the one to refer to. I would suggest that the circle should be showing the BASE case – you could then have one vertical whisker down from that with two horizontal whiskers indicating the 2 sensitivity runs. Please change Figure 21 accordingly.
Response) Instead of circle symbol, all model runs are shown in horizontal whiskers.

86) Page 5861 line 25-29 – the BASE run seems to high according to the Figure with the LowNuc in much better agreement at some sites. Please can you comment on this in the text.
Response) First of all, we agree that LowNuc shows the best agreements among the simulations. In that sentence, we meant that all three simulations are quite well comparable to the observation, as the overall errors and biases for all simulations are small. However, we added the new phrase (bolded for the new part) right after the sentence.

"On average, the annual-mean CN concentrations in the model agree with the observations well for the all three categories (LMNB= -0.26 to 0.16; LMNE=0.13 to 0.22), **although the LowNUC simulation shows the best agreement to observation."**

References:
Andres, R. and Kasgnoc, A.: A time-averaged inventory of subaerial volcanic sulfur emissions, J. Geophys. Res.-Atmos., 103, 25251–25261, 1998.

Taylor, K. E., Stouffer, R. J., and Meehl, G. A.: An overview of CMIP5 and the experiment design,_B. Am. Meteorol. Soc., 93, 485–498, 2012.

Evaluation of the global aerosol microphysical ModelE2-TOMAS model against satellite and ground-based observations

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Abstract

The TwO-Moment Aerosol Sectional microphysics model (TOMAS) has been integrated into the state-of-the-art general circulation model, GISS ModelE2. This paper provides a detailed description of the ModelE2-TOMAS model and evaluates the model against various observations including aerosol precursor gas concentrations, aerosol mass and number concentrations, and aerosol optical depths. Additionally, global budgets in ModelE2-TOMAS are compared with those of other global aerosol models, and the ModelE2-TOMAS model is compared to the default aerosol model in ModelE2, which is a One-Moment Aerosol (OMA) model (i.e., no aerosol microphysics). Overall, the ModelE2-TOMAS predictions are within the range of other global aerosol model predictions, and the model has a reasonable agreement (mostly within a factor of two) with observations of sulphur species and other aerosol components as well as aerosol optical depth. However, ModelE2-TOMAS (as well as ModelE2-OMA) cannot capture the observed vertical distribution of sulphur dioxide over the Pacific Ocean possibly due to overly strong convective transport and overpredicted precipitation. The ModelE2-TOMAS model simulates observed aerosol number concentrations and cloud condensation nuclei concentrations roughly within a factor of two. Anthropogenic aerosol burdens in ModelE2-OMA differ from ModelE2-TOMAS by a few percent to a factor of 2 regionally, mainly due to differences in aerosol processes including deposition, cloud processing, and emission parameterizations. We observed larger differences for naturally emitted aerosols such as sea salt and mineral dust, as those emission rates are quite different due to different upper size cutoff assumptions.

1. Introduction

Aerosols perturb the energy balance of the Earth-atmosphere system by scattering and absorbing solar and terrestrial radiation, known as the aerosol direct effect, and by modifying cloud properties such as via acting as cloud condensation nuclei (CCN),

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known as aerosol indirect effects (e.g. Lohmann and Feichter, 2005; Forster and Ramaswamy, 2007). The recently published IPCC AR5 (Intergovernmental Panel on Climate Change Fifth Assessment Report) refers to these as aerosol-radiation interactions and aerosol-cloud interactions, respectively (Boucher et al., 2013). For light-absorbing aerosols such as black carbon and mineral dust, the ambient air can be heated as a result of their direct effect, affecting relative humidity and atmospheric stability, which is known as the semi-direct effect. The largest uncertainty in estimating anthropogenic climate forcing is from the aerosol indirect effects (Myhre et al., 2013b). Since it is not easily estimated from observations due to natural variability in cloud properties and the lack of observations of the pre-industrial atmosphere, estimates of aerosol indirect forcing have been mainly based on general circulation models (GCMs). Thus, there have been growing efforts to develop and improve aerosol microphysics models for a more physically based representation of atmospheric aerosol number and CCN concentrations (e.g. Adams and Seinfeld, 2002; Easter et al., 2004; Vignati et al., 2004; Lauer et al., 2005; Liu et al., 2005; Spracklen et al., 2005; Stier et al., 2005; Bauer et al., 2008; Trivitayanurak et al., 2008; Yu and Luo, 2009; Mann et al., 2010; Lee and Adams, 2012).

Aerosol microphysics models can be broadly categorized into modal and sectional methods, depending on how they represent the aerosol size distribution. In general, modal-based methods use an analytical function (e.g. a lognormal distribution) to represent a subset of the particle population. Sectional methods represent a size distribution by predicting aerosols in several size sections or "bins". Additionally, sectional and modal methods may differ from each other in numerous ways, including the number of moments of the size distributions that are tracked in each section or mode.

Sectional methods can be divided into single-moment sectional methods that typically track either aerosol number or mass in each bin and two-moment sectional methods that explicitly track both aerosol number (i.e. 0^{th} moment) and mass (i.e. 1^{st} mass moment or 3^{rd} radial moment) in each size section. Unlike single-moment sectional approaches, two-moment sectional methods can conserve both number and mass very accurately (Tzivion et al., 1987; Feingold et al., 1988; Harrington and Kreidenweis, 1998; Tzivion et al., 2001; Adams and Seinfeld, 2002; Jung et al., 2006) but have a high computational burden. The modal approaches are generally more computationally

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efficient but may not represent abrupt transitions in a size distribution well, which can occur during cloud processing (Zhang et al., 1999).

The TOMAS aerosol microphysics model (Adams and Seinfeld, 2002; Tzivion et al., 1987;Tzivion et al., 1989) has been developed to study tropospheric aerosol microphysics and predict cloud condensation nuclei (CCN) concentrations. The TOMAS model has been previously implemented into the climate model of Goddard Institute for Space Studies General Circulation Model II-prime (GISS GCM II-prime), referred to as "GISS-TOMAS" (Lee and Adams, 2010). It has also been incorporated into GEOS-CHEM (Trivitayanurak et al. 2008), the regional model PMCAMx-UF (Jung et al., 2010), and the Large-Eddy Simulation model (Stevens et al., 2012;Singh et al., 2014). The GISS GCM II-prime has horizontal grid dimensions of 4˚ latitude and 5˚ longitude, with nine vertical sigma layers between the surface to the 10 hPa level (Hansen et al., 1983). Modules for each of the major aerosol species have been developed for the GISS GCM II-prime, and the GISS-TOMAS model has been evaluated with ground-level measurements such as number and mass concentrations, deposition fluxes, and remote sensing observations (Adams and Seinfeld, 2002; Pierce and Adams, 2006; Pierce et al., 2007; Lee et al., 2009; Lee and Adams, 2010). Despite the accuracy in predicting aerosol microphysical process in TOMAS, the original version of TOMAS has a heavy computational burden. Lee and Adams (2012) developed less computationally expensive configurations of the TOMAS model (Fast TOMAS), which are 2-3 times faster than the original TOMAS model with only a few percent increases in microphysical errors. However, a remaining weakness for the GISS-TOMAS model is the outdated host model, the GISS GCM II-prime.

Here, we incorporate the TOMAS model into the new version of GISS GCM (i.e., ModelE2), referred to as "ModelE2-TOMAS". ModelE2 now has three different aerosol models available: TOMAS, the One-Moment Aerosol model (hereafter, referred to as OMA) (e.g. Koch et al., 2006) that has no microphysics, and the modal-based aerosol microphysics model, MATRIX (Multiconfiguration Aerosol TRacker of mIXing state) (Bauer et al., 2008). The combination of several aerosol models into the same host GCM allows ModelE2 to explore the uncertainties in predicting aerosol characteristics and their climate effects that are associated with aerosol modelling (e.g. different numerical

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approaches) in the same host model. We also note that it was important to implement the TOMAS aerosol model into the ModelE2 host model because uncertainties in the estimates of aerosol forcing come not only from aerosol modelling itself but also other parts of the host GCM (e.g., cloud physics, planetary boundary layer, and advection).

ModelE2-TOMAS has been used in several recent studies under the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) which aims to understand composition changes and the associated radiative forcing between 1850 and 2100 (Bowman et al., 2013; Lamarque et al., 2013a; Lamarque et al., 2013b; Lee et al., 2013a; Nabat et al., 2013; Naik et al., 2013; Shindell et al., 2013; Stevenson et al., 2013; Young et al., 2013). Here we give a detailed description of ModelE2-TOMAS and evaluate against ModelE2-OMA (e.g., Schmidt et al., 2014) and observations of aerosol mass and number as well as aerosol optical depth. Section 2 provides the description of ModelE2-OMA, and Section 3, the description of the TOMAS aerosol microphysics model. Section 4 explains the emissions and design of the simulations. Section 5 presents global budgets of the simulated aerosols and the evaluation of the ModelE2-TOMAS and ModelE2-OMA against observations of aerosol mass concentrations and aerosol optical depth and the evaluation of the TOMAS number predictions against observations. Conclusions follow in Section 6. We note that aerosol direct and indirect forcings using ModelE2-TOMAS will be discussed in a separate paper.

2. GISS GCM ModelE2

In this section, we briefly describe ModelE2 (Schmidt et al. (2014)), the GISS climate model used to perform Coupled Model Intercomparison Phase 5 (CMIP5; Taylor et al, 2012). The model physics are mostly similar to GISS ModelE (CMIP3 version: Schmidt et al., 2006). The model has 2° latitude by 2.5° longitude resolution, with 40 vertical hybrid sigma layers from the surface to 0.1 hPa (80 km). Tracers, heat, and humidity are advected using the highly nondiffusive Quadratic Upstream Scheme (Prather, 1986). The radiation scheme accounts for size-dependent scattering properties of clouds and aerosols based on Mie scattering (Hansen et al., 1983) and non-spherical light scattering of cirrus and dust particles based on T-matrix theory (Mishchenko et al., 1996). It also includes the impact of water uptake by hygroscopic species on their radiative properties. In the model,

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clouds are distinguished into convective and large-scale stratiform clouds. The clouds parameterizations are similar to Del Genio (1993) and Del Genio et al. (1996) but have been improved in several respects (see details in Schmidt et al., 2006; Schmidt et al., 2014). The physics timestep is 30 minutes, and the radiation is calculated every 2.5 hours.

2.1 ModelE2-OMA description

ModelE2 includes a default aerosol module, OMA (One-Moment Aerosol), which has no microphysics. ModelE2-OMA has sulphate (Koch et al., 2006; Koch et al., 2007; Koch et al., 2011), carbonaceous aerosols (Koch et al., 2007), secondary organic aerosols (Tsigaridis and Kanakidou, 2007), sea salt (Koch et al., 2006; Tsigaridis et al., 2013), dust (Miller et al., 2006), and nitrate (Bauer et al., 2007). Along with sulphate, the model also predicts sulphur dioxide, dimethyl sulfide (DMS) and methanesulfonic acid (MSA) (Koch et al., 2006). The secondary organic aerosol formation is computed using a two-product model with isoprene, monoterpenes, and sesquiterpenes as SOA precursors (described in Tsigaridis and Kanakidou, 2007). Sea salt particles have two size classes with a fine mode (0.1 μm to 1 μm in dry radii) and a coarse mode (1 μm to 4 μm in dry radii). Dust particles have four size classes with radii between 0.1-1 μm (clay), 1-2 μm (silt1), 2-4 μm (silt2), and 4-8 μm (silt3). The model accounts for heterogeneous chemistry on mineral dust particle surfaces to form nitrate and sulphate (Bauer and Koch, 2005).

In ModelE2, the surface boundary conditions are defined using dry deposition and interactive surface sources (Koch et al., 2006). The dry deposition scheme is tightly coupled to the model boundary layer process and is based on a resistance-in-series scheme derived from the Harvard GISS-CTM, which is applied between the surface layer (10 m) and the ground (Koch et al., 2006). Wet deposition is determined by several processes including rainout within clouds, washout below precipitating regions, scavenging within and below cloud updrafts, evaporation of falling precipitation, transport along with convective plumes, and detrainment and evaporation from convective plumes (Koch et al., 2006; Shindell et al., 2006). ModelE2 includes a dissolved species budget scheme for stratiform clouds, which has an impact on sulphate

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formation via aqueous oxidations, since some sulphate formed in clouds undergoes wet scavenging instead of being added back to the sulphate in air (Koch et al., 2006).

Tropospheric/stratospheric chemistry in ModelE2 includes 156 chemical reactions among 51 gas species (Shindell et al., 2013). In ModelE2, chemistry and aerosols are fully interactive, so that the oxidation fields used for sulphate formation are from the chemistry model (not prescribed) and the photolysis rates are affected by light attenuation by aerosols (Shindell et al., 2013). Photolysis rates are computed using the Fast-J2 scheme (Bian and Prather, 2002). Aerosol indirect effects are based on an empirical parameterization that compute cloud droplet number concentrations as a function of aerosol mass (Menon et al., 2002; Menon et al., 2008).

3. ModelE2-TOMAS description

The TOMAS aerosol microphysics model uses a sectional approach that represents the aerosol size distribution by predicting the amount of aerosol in several size categories or "bins". TOMAS tracks two moments of the aerosol size distribution in each size bin: total aerosol number (i.e., 0^{th} moment) and mass (i.e., 1^{st} mass moment). Total mass is decomposed into several aerosol species, allowing prediction of the size-resolved aerosol composition. In total, ten quantities are tracked for each size bin: sulphate mass, sea-salt mass, mass of pure (hydrophobic) elemental carbon (EC), mass of mixed (aged) EC, mass of hydrophobic organic matter (OM), mass of hydrophilic OM, mass of mineral dust, mass of ammonium, mass of water and the number of aerosol particles in that bin. In TOMAS, all ammonia becomes aerosol ammonium until sulfate is neutralized to form ammonium sulfate; the excess ammonia after neutralization remains as free gas-phase ammonia. The aerosol ammonium is partitioned into each size bin in proportion to the sulfate mass. However, ammonium is not size-resolved (i.e., bulk tracer) for purposes of model processes outside of TOMAS such as advection and deposition. In addition, the model tracks four bulk gas-phase species: sulphur dioxide (SO_2), dimethylsulfide (DMS), sulphuric acid (H_2SO_4), and a lumped gas-phase tracer that represents oxidized organic vapors forming secondary organic aerosol (SOA). The gas-phase H_2SO_4 is assumed to be in pseudo-steady state equilibrium between its chemical production and condensational/nucleation losses (Pierce and Adams, 2009a). Water uptake by sulphate

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and sea salt is based on a polynomial fit based on ISORROPIA, a thermodynamic equilibrium model for inorganic aerosols (Nenes et al., 1998). For water uptake by hydrophilic OM, it is based on the observations of Dick et al. (2000). The size section boundary is defined by dry particle mass, such that addition or removal of aerosol water mass does not move particles between sections. In general, TOMAS treats all aerosols as internally mixed during microphysics such as calculating condensation and coagulation rates. However, a portion of EC is treated as externally mixed for purpose of wet deposition. Detailed descriptions of the TOMAS microphysics scheme can be found in Adams and Seinfeld (2002), Lee and Adams (2012), and Lee et al. (2013b).

Several alternative nucleation schemes are available in TOMAS, including binary nucleation (Vehkamaki et al., 2002), ternary nucleation (Napari et al., 2002), ion-induced nucleation (Modgil et al., 2005), and activation nucleation with an A factor of 2×10^{-6} s^{-1} (Sihto et al., 2006) for the boundary layer (~up to 900 mbar). For the simulations used in this paper, only binary nucleation is used. The boundary-layer nucleation is off in all simulations because it tends to overpredict aerosol number concentrations in our model. Also we do not show any run with the ternary nucleation (Napari et al., 2002) because it overpredicts aerosol number concentration severely (not shown).

With Fast TOMAS models, the TOMAS microphysics module became more flexible in term of varying particle size resolution, i.e. the number of size bins (Lee and Adams, 2012). For the size range of 10 nm to 10 μm, the original TOMAS uses 30 bins (size boundary is defined with mass doubling), and the Fast TOMAS uses 15 bins or 12 bins (size boundary is defined with mass quadrupling). As discussed in Lee and Adams (2012), the Fast TOMAS reduces the computational burden by 2-3 times while generally predicts CCN concentrations within a few percent of the original TOMAS. The lower size cutoff in TOMAS can also vary from 10 nm to 3 nm or from 10 nm to 1 nm (Lee et al., 2013b). Among several possible configurations, ModelE2-TOMAS currently uses either 12 bins covering 10 nm to 10 μm or 15 bins covering 3 nm to 10 μm, which is the most computationally efficient version of TOMAS for the given size range. In this paper, we used TOMAS with 15 bins covering 3 nm to 10 μm (TOMAS15; see Table S1 in the supplementary materials): 3 bins cover from 3nm to 10 nm, 10 bins from 10 nm to 1 μm and the last two bins from 1 μm to 10 μm. The TOMAS15 version becomes a default

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model configuration for ModelE2-TOMAS, so we will be continuously refereed to as ModelE2-TOMAS throughout this paper. More configurations will be available in the near future.

The wet deposition scheme in ModelE2-TOMAS is identical to the one used in ModelE2-OMA except for the following. First, ModelE2-TOMAS adds sulphate mass produced in the aqueous phase directly to the bin-resolved sulphate mass in ambient air rather than maintaining a separate tracer for dissolved sulphate. Compared to ModelE2-OMA, this is a simplification because the sulphate formed in the aqueous phase will be only released as interstitial aerosol when the cloud water evaporates. It is adopted here for simplicity but will be improved in the future. The other difference is that the wet/dry deposition in ModelE2-TOMAS accounts for particle size dependence. For in-cloud scavenging, modified Köhler theory is used to obtain the critical supersaturation for activation of each size section and to determine which particles activate and are subject to in-cloud (nucleation) scavenging (Pierce et al., 2007). To determine activation, we assume kappa values of 0.7 for sulfate, 1.3 for sea-salt, and 0.15 for hydrophilic OM. The fraction of activated aerosols removed by wet deposition is proportional to the fraction of cloud water that precipitates, which is computed in each model layer. Wet deposition accounts for re-evaporation of precipitation. For in-cloud scavenging, the large-scale and convective clouds in the model are assumed to have a supersaturation of 0.2%; unlike GISS-TOMAS that used a supersaturation of 1.0% for convective clouds, a supersaturation of 0.2% is assumed in ModelE2-TOMAS in order to capture the observed Hoppel gap (~100 nm) in the marine boundary layer. Note that the activation described here to determine in-cloud scavenging is not used for computing cloud droplet number concentrations (see below). For below-cloud scavenging, a first-order removal scheme implemented for bulk aerosols by Koch et al. (1999) is modified for size-resolved aerosols (Adams and Seinfeld, 2002). Dry deposition is identical to the existing resistance-in-series scheme in ModelE2, but ModelE2-TOMAS treats a size-dependent gravitational settling of particles and a size-dependent resistance in the quasi-laminar sublayer (Seinfeld and Pandis, 1998; Adams and Seinfeld, 2002).

To compute the cloud microphysics properties as a function of aerosols (i.e., the aerosol-cloud interactions), ModelE2-TOMAS uses a physical-based activation

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parameterization from Nenes and Seinfeld (2002). A critical supersaturation is computed in the parameterization using a model updraft velocity that is computed based on a large-scale vertical velocity and sub-grid velocity.

In ModelE2-TOMAS, Mie theory is used to compute size-resolved AOD. For each grid cell, particle compositions (including aerosol-water) in each individual size bin are used to compute the volume-averaged refractive index and optical properties based on Mie theory. The optical properties are used to compute aerosol optical depth taking into account the aerosol concentration.

ModelE2-TOMAS is coupled to the same gas chemistry model (Shindell et al., 2013) as ModelE2-OMA. So the oxidation fields used for sulphate formation are from the chemistry model. However, unlike ModelE2-OMA, the photolysis rates are not affected by aerosols.

4. Description of the simulations

4.1 Emissions

The emissions used in this study are summarized in Table 1. The simulations used year-2000 emissions from the anthropogenic emissions inventory created for CMIP5 (Lamarque et al., 2012) and climatologically averaged biomass burning emissions from GFED3 for 1997 to 2009 (van der Werf et al., 2010). For SO_2, in addition to the anthropogenic emissions, continuous volcanic emissions from GEIA (Global Emissions InitiAtive; Andres and Kasgnoc, 1998) are used but increased by a factor of 1.5 as in the AEROCOM intercomparison emissions in Dentener et al. (2006). Sea-salt emission is based on Gong (2003), which extends the lower size limit of the Monahan et al. (1986) emission from 0.4 μm to 0.02 μm. Dust emission is based on the source distribution from Ginoux et al. (2001) and is proportional to the third power of the wind speed (at 10 m in height) above a threshold that is a function of soil moisture. Subgrid-variation of the wind speed in a GCM grid box, which is created by boundary-layer turbulence and dry/wet convection, is accounted for the modeled dust emissions (Cakmur et al., 2006; Miller et al., 2006). DMS emission is based on the seawater DMS concentrations of Kettle et al.

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(1999). For the sea-to-air transfer function used in the DMS emissions, The ModelE2-TOMAS runs are based on Liss and Merlivat (1986), and the ModelE2-OMA model run is based on Nightingale et al. (2000).

Nightingale et al. (2000) provides a revised parameterization based on observations of the sea-to-air transfer rate scatter between two classical parameterizations (i.e., Liss and Merlivat, 1986;Wanninkhof, 1992), and it has been more favoured in many global DMS models than the two classical parameterizations. However, DMS emission is quite uncertain. Estimates of the global DMS emissions range from 16 to 54 Tg S yr^{-1} (Kettle and Andreae, 2000), depending on the choice of DMS sea surface climatology, sea-to-air transfer rate parameterization, and wind speed data. DMS emission rates from ModelE2-TOMAS and ModelE2-OMA are within the range. The Liss and Merlivat (1986) parameterization is used in ModelE2-TOMAS, because a ModelE2-TOMAS run based on Nightingale et al. (2000) overpredicts the SO_2 concentrations over remote oceanic regions especially in the southern hemisphere. Koch et al. (2006) showed that the sea-to-air transfer function from Nightingale et al. (2000) increased annual DMS emissions by roughly a factor of two compared to the emission based on Liss and Merlivat (1986). This was desirable in ModelE2-OMA because of the underprediction of sulphate in remote oceanic regions in that model, although the model DMS and MSA (oxidized from DMS) tended to be excessive in SH oceanic regions especially near Antarctica. However, despite the higher DMS emissions, it turned out that the sulphate was still underpredicted because sulphate formed by aqueous oxidation was subject to wet scavenging before releasing to the ambient air as a result of the updated dissolved species budget scheme (Koch et al., 2006).

4.2 ModelE2-TOMAS run setup

We performed the simulations nudged with winds from the MERRA (Modern Era Retrospective-analysis for Research and Applications; Rienecker et al., 2011) reanalysis meteorological fields from 2000 to 2003 with 3 years spin-up (i.e., 1997-1999). Primary emission of particulate sulphate is assumed to be 1.0 percent of total sulphur emissions. Emissions size distributions assumed for ModelE2-TOMAS are summarized in Table 2. Primary sulphate emission is assumed to have a bi-modal lognormal distribution that

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assigns 5% of the primary sulphate emissions as a nucleation mode with a geometric number mean diameter (GMD) of 10 nm and a geometric standard deviation (GSD) of 1.6 and the rest as an Aitken mode with GMD of 70 nm and GSD of 2. For fossil fuel and biofuel emissions, the size of primary carbonaceous aerosol emissions is assumed to fit a lognormal size distribution with a GMD of 60 nm and a GSD of 1.59 for both EC and OM (Stier et al., 2005). For carbonaceous aerosols of biomass burning emissions, a lognormal size distribution is assumed to have a GMD of 150 nm and a GSD of 1.59. Note that although the emission size distribution for biofuel emissions is generally assumed to be the same as that for biomass burning emissions (e.g. Dentener et al., 2006), in ModelE2-TOMAS run we assumes the biofuel emission size distributions follow the finer fossil fuel size settings because the CMIP5 emissions does not provide a separate category for biofuel emissions (e.g. biofuel used for cooking and heating are assigned as the residential sector, which also includes fossil fuel usage). The OC (organic carbon): OM (organic matter) ratio is assumed to be 1:1.4.

ModelE2-TOMAS assumes larger particles for primary sulphate and carbonaceous aerosols than GISS-TOMAS (e.g., Lee et al., 2013b) to capture the observed aerosol number concentrations better. This is very likely due to the following: 1) GISS-TOMAS attributed the primary sulphate emissions only to anthropogenic sulphur emissions (excluding biomass burning emissions), while ModelE2-TOMAS attributes these to all sulphur emissions; 2) GISS-TOMAS applied the biomass-burning emission size distributions of carbonaceous aerosols to the biofuel emissions, which is coarser than the fossil fuel emission size distribution. Note also that the emission size distributions used for biomass burning and volcanic emissions are finer than the AEROCOM recommendations in Dentener et al. (2006). However, the model number concentrations and size distributions are changed little when applying the AEROCOM recommended emission distributions (not shown). Note that the biomass burning and volcanic emissions for sulphur are 1.4 Tg S $yr^{-1}$and 12.5 Tg S yr^{-1}, respectively.

Following the soil size assumptions used in GISS-TOMAS (Lee et al., 2009), the clay distribution is assumed to have a GMD of 0.14 μm and a GSD of 2, and the silt distribution, a GMD of 1.15 μm and a GSD of 2. Using this distribution, fifteen percent

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of the silt emissions flux falls out of the upper size cutoff (i.e., 10 μm), and is therefore not received by any of the TOMAS size bins.

Compared to the run setup described above (hereafter, referred to as the "BASE" run), we additionally ran two other sensitivity runs with the ModelE2-TOMAS model by perturbing the nucleation process to evaluate changes in number concentrations (Table 1). The first sensitivity run is called "NoNUC", in which we turned off nucleation to estimate the contribution of primary emissions to aerosol number concentrations. The other run is called "LowNUC", in which we reduced the nucleation rate by using 5 times lower sulphuric acid concentrations to compute nucleation rates. Note that sulphuric acid concentrations are not perturbed in other processes, and the model sulphuric acid budget is little influenced by this treatment.

4.3 ModelE2-OMA run setup

To compare to the ModelE2-TOMAS run, we also ran the ModelE2-OMA model nudged to the same MERRA reanalysis meteorology with 3 years spin-up. However, the natural emissions and associated settings are not always the same between the two models because we chose to maintain the natural emissions/setup used in ModelE2-OMA, which has been chosen carefully in previous studies. To assist the interpretation of the results, we briefly summarize the differences between ModelE2-OMA and ModelE2-TOMAS. First, as mentioned in Section 4.1, the ModelE2-OMA model uses the sea-to-air transfer function of Nightingale et al. (2000) instead of Liss and Merlivat (1986), because Koch et al. (2006) prefers the newer DMS emissions to improve a sulphate prediction in the remote marine locations. Second, the same sea salt and dust emission schemes are applied in both aerosol models, but different assumptions for the upper limit of particle size are used: 8 μm in diameter for sea salt and 16 μm in diameter for dust in the ModelE2-OMA model; 10 μm in ModelE2-TOMAS. Third, the ModelE2-OMA model assumes 2.5% of the total sulphur as primary sulphate as followed by the AEROCOM study (Dentener et al., 2006) whereas ModelE2-TOMAS assumes only 1%. Aerosol number predictions are sensitive to the primary sulphate assumption, but sulphate mass concentrations are not. When using the 2.5% assumption in ModelE2-TOMAS, we found that the simulated aerosol number concentrations were biased high,

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and the model size distribution predictions wer also poor. Note that Pierce and Adams (2009b) shows that GISS-TOMAS also overpredicts aerosol number concentration with the 2.5% assumptions.

5. Model results and evaluations

In this section, we present global-annual budgets, spatial distributions, and evaluations of the model aerosol precursor gases (in Sect. 5.1), aerosol mass (in Sect. 5.2 and 5.3), aerosol optical depths (AODs; in Sect. 5.4) and aerosol number (in Sect. 5.5 and 5.6). The observations used for model evaluations are from surface-based, aircraft-based and remote-sensing measurements. More details of the observations are provided in each subsection. To compare with the ModelE2-TOMAS results, we included the ModelE2-OMA results in global-annual budgets and model evaluations. Only the BASE run results are used in Section 5.1 to 5.4 because the predicted aerosol precursor gases concentrations, aerosol mass concentrations, and AODs from the nucleation sensitivity runs are quite similar to the BASE run.

Model skill is quantified in terms of log-mean normalized bias (LMNB) and log-mean normalized error (LMNE) when evaluating with annual-mean concentrations measurements and normalized mean bias (NMB) and correlation coefficient (R) when evaluating with an observed annual cycle and aerosol optical depth (both monthly and annually averaged AODs).

5.1 Aerosol precursor gases

Global budgets of DMS and SO_2 in ModelE2-TOMAS are presented in Table 3 with a range obtained from several global models including Wang et al. (2011), Liu et al. (2005), and those listed in Liu et al. (2005). The DMS and SO_2 budgets in ModelE2-TOMAS are within the ranges of the other global models. In case of ModelE2-OMA (in Table 3), the global burden of DMS is about a factor of two higher than the ModelE2-TOMAS model because the DMS emission rate is ~78% higher by using the sea-to-air transfer functions by Nightingale et al. (2000). Despite the different DMS emissions and SO_2 emissions (due to the primary sulphate emission assumption, 1% versus 2.5%), the

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global burden of SO_2 is quite similar to that in the ModelE2-TOMAS model. The dominant SO_2 removal processes are aqueous oxidation and dry deposition in both simulations. Boucher et al. (2003) simulates atmospheric DMS in the LMD-ZT model using the same DMS emission schemes as ModelE2-OMA (i.e., EXP1 in their study) and ModelE2-TOMAS (i.e., EXP4 in their study). The global DMS budgets from ModelE2-OMA and ModelE2-TOMAS agree quite well with the EXP1 (within 25 %) and EXP4 (within 15%).

When using the same DMS emissions in ModelE2-TOMAS as in ModelE2-OMA, the DMS global budgets are almost identical, but the SO_2 budgets vary substantially due to the differences in SO_2 modelling, i.e., heterogeneous SO_2 oxidation and photolysis (see Section 2). The heterogeneous sulphur dioxide oxidation on dust aerosol surfaces, which is only included in ModelE2-OMA, accounts for 25% of the total gas-phase oxidation loss. Based on Bauer and Koch (2005), including the heterogeneous chemistry, global SO_2 burden can decrease 32%, and global sulphate burden can increase 3%. The simulated photolysis rates in ModelE2-OMA are affected by aerosol optical depth, affecting hydroxide (OH) and other gas tracer concentrations - Naik et al. (2013) show a higher OH concentration in ModelE2-TOMAS than ModelE2-OMA. Overall, using the same DMS emissions in ModelE2-TOMAS results in a higher SO_2 burden and worse agreement for SO_2 and sulphate concentrations over remote oceanic regions (not shown).

The global budgets of H_2SO_4 and SOA precursor gas in the ModelE2-TOMAS model are not included in Table 3 but are summarized here. The simulated H_2SO_4 has a total production rate of 12.3 Tg S yr^{-1}, matching the SO_2 gas-phase oxidation, and is used in aerosol microphysics (i.e. 12 Tg yr^{-1} for condensation and 0.3 Tg yr^{-1} for nucleation). The model SOA precursor gas has a total production rate of 17.1 Tg yr^{-1}, assumed to be 10% of the terpene emission, and is condensed as hydrophilic OM. For ModelE2-OMA, the total production rate of SOA is 14.6 Tg yr^{-1}. This is quite comparable to ModelE2-TOMAS, which treats SOA much more simply and has a production rate of 17.1 Tg yr^{-1}. Global burden of SOA in ModelE2-OMA is 0.6 Tg yr^{-1}.

Figure 1 shows global maps of annual-mean DMS and SO_2 column mass concentrations. The spatial distribution of DMS concentrations shown in Fig. 1a is driven by its emission and interactive OH and NO_3 concentrations, which oxidize DMS to form

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MSA and SO_2. The model DMS concentrations are most pronounced in the Southern Ocean and the Northern Atlantic oceans due to high seawater DMS concentrations during summer. The simulated SO_2 concentration shown in Fig. 1b is very high over industrial regions due to the anthropogenic emissions and is also high over the Southern Ocean due to DMS oxidation. Several local hotspots of SO_2 shown in Fig. 1b are due to volcanic emissions.

Annually averaged surface-layer SO_2 concentrations from both ModelE2-TOMAS and ModelE2-OMA are evaluated against observations from the EMEP (European Monitoring and Evaluation Programme, http://www.emep.int) and CASTNET (Clean Air Status and Trends Network, http://epa.gov/castnet/javaweb/index.html) networks (see Fig. 2). We used 2000-2004 mean SO_2 measurements for the EMEP network and 1995-2005 mean SO_2 data for the CASTNET network. Performance of ModelE2-TOMAS and ModelE2-OMA for predicted SO_2 concentrations in these locations is almost the same (i.e. LMNB=0.25-0.26 and LMNE=0.34 for the EMEP network; LMNB=0.09 and LMNE=0.29 for the CASTNET network), and the continental SO_2 predictions agree with the observation on average roughly within a factor of two. The two aerosol models are almost the same because the anthropogenic emissions, which are identical in both models, are dominant at these locations.

Figure 3 compares surface-layer SO_2 and DMS mixing ratios from the two aerosol models against observations at three Southern Hemisphere remote sites: Amsterdam Island (DMS from Sciare et al., 2000; SO_2 from Nguyen et al., 1992), Cape Grim (Ayers et al., 1995), and Dumont (Jourdain and Legrand, 2001). Note that Dumont has only DMS measurements. For DMS, both models capture the observed seasonal cycle (i.e. R>0.8) generally well but are less successful over Dumont. The DMS concentrations seem to agree well against the observations when using the sea-air transfer function of Liss and Merlivat (1986), i.e., the case for ModelE2-TOMAS, but this run underpredicts during winter season at the Amsterdam Island site (in Fig. 3a) and during all season at Dumont site (in Fig. 3c). Earlier, we mentioned that the global DMS budgets from Boucher et al. (2003) agree well with those from ModelE2 when using the same DMS emission parameterization. However, Boucher et al. (2003) shows the better agreement to the same DMS measurements when using the sea-air transfer function of Nightingale et

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al. (2000) at Amsterdam Island and Cape Grim (i.e., the case for ModeleE2-OMA), because their DMS mixing ratios from that simulations are actually more close to ModelE2-TOMAS. This may suggest that, at least over SH high latitude regions, the surface wind speed in ModelE2 is much stronger than that in LMD-ZT, resulting in higher DMS emissions and burden. We need to investigate further to find out a source for the difference though.

The ModelE2-TOMAS SO_2 shows very good agreement at Amsterdam Island but a high bias at Cape Grim (see Figs. 3d and e). We considered whether the overpredicted SO_2 in the model might be influenced by the emissions in the adjacent grids, which is mentioned in Mann et al. (2010), because the measured SO_2 at Cape Grim is filtered to include the marine sector only (Ayers et al., 1991). Sampling the model SO_2 from adjacent grids toward marine areas, the overprediction is reduced significantly (LMB is reduced from 10 to 3) but is still severe. The most plausible reason for the overprediction of SO_2 at Cape Grim might be the lack of SO_2 oxidation by ozone on sea salt particles, which is missing in our model. Korhonen et al. (2008) shows a reduction of SO_2 concentrations by a factor of 5 in January and a factor of 20 in July at Cape Grim when including SO_2 oxidation on sea spray particles, although their treatment of the reactions might overestimate the SO_2 oxidation rates.

Simulated DMS and SO_2 vertical profiles over the Pacific Ocean are compared against two sets of aircraft observations in Figures 4 and 5: PEM-Tropics-A performed during August-October, 1996 (Hoell et al., 1999) and PEM-Tropics-B during March-April, 1999 (Raper et al., 2001). Note that PEM-Tropics-A DC8 aircraft data is used and most of them were during September 1996, but model evaluation is little changed by comparing with model outputs in September or August-October average. The observed vertical profile data used here are binned into altitude ranges (Emmons et al., 2000). Model outputs are averaged over the observational time period and domain. Simulated DMS vertical profiles are very similar between the two aerosol models, although the surface DMS is different as their emissions are not the same. Both models show good agreement with the observations (mostly within 25th and 75th percentile of observed values), especially capturing a strong concentration decrease from the surface to the free troposphere.

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In the case of SO_2, even though the agreement is not as good as that seen for DMS, both aerosol models seem to capture the observed magnitude approximately within a factor of two (see Fig. 5). The overall vertical patterns shown in the models are frequently not in agreement with the observations. Except at Hawaii, our model do not capture the enhanced SO_2 concentrations in the boundary layer shown in the observation, even though the model DMS is quite well captured. The poor prediction of SO_2 vertical profile might be due to 1) too much precipitation near tropics in ModelE2 (see Fig. 9 in Schmidt et al., 2014) and 2) too strong vertical transport (e.g., via deep convection over the tropical Pacific Ocean) in the model. The latter can be supported by that the small DMS peak at 8 km and the elevated SO_2 in the upper/free troposphere in the models (see Figs. 4 and 5). Although the elevated SO_2 mixing ratios might be due to too weak wet scavenging (including aqueous chemistry) of SO_2, we did not see any noticeable improvement when increasing SO_2 Henry's law constant by a factor of two in the model (not shown). A large peak in the mid-troposphere at Hawaii in the models results from volcanic SO_2 emissions, while the observations show a similar peak only during March-April 1999, which is heavily influenced by volcanic emissions (Thornton et al., 1999). During August-October 1996, the observations at Tahiti and Easter Island show transport of volcanic SO_2 emissions in the middle and upper troposphere (Thorton et al., 1999), which does not capture in the model. Since our model includes only continuous volcanic emission with a yearly resolution, our model fails to simulate variability in volcanic SO_2 emissions at higher time resolution.

5.2 Aerosol mass budgets and distributions

Globally and annually averaged budgets of aerosols in the ModelE2-TOMAS model are shown in Table 4. For the sulphate and EC budgets, we use the ACCMIP multi-model mean from Shindell et al. (2013) and Lee et al. (2013a), which is based on 8 ACCMIP models using the same AR5 emission scenario. Note that the biomass burning emission in this study is GFEDv3 inventory averaged from 1997 to 2009, while the ACCMIP models use GFEDv2 inventory for 2000. We do not compare with the AEROCOM phase 2 multi-model mean presented in Myhre et al. (2013a) because the aerosol budgets in Myhre et al. (2013) are for anthropogenic aerosols, which is defined as the difference

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between the present-day run and pre-industrial run. For the lifetime and deposition rate coefficient budgets, we compare with the AEROCOM Phase 1 multi-model mean presented in Textor et al. (2006) –hereafter, referred to as AEROCOM Phase 1.

For sulphate, the ModelE2-TOMAS total source rate is lower than the ACCMIP mean (43.7 Tg yr^{-1} vs. 51.7 Tg S yr^{-1}), and the global burden is the same as the ACCMIP mean burden (0.67 Tg S) due to the slightly longer lifetime in the ModelE2-TOMAS model (5.7 days vs. 5.0 days). Note that the GISS-E2-R-TOMAS model used for ACCMIP is almost identical to ModelE2-TOMAS evaluated here except for the sulfate modeling. The sulphate and DMS emissions used in GISS-E2-R-TOMAS are identical to those used in ModelE2-OMA in this paper. For dry deposition coefficient (the inverse of the lifetime), ModelE2-TOMAS has a particularly small value. However, the longer overall sulphate lifetime is contributed by both dry and wet deposition, rather than dry deposition. When increasing the dry deposition coefficient to the AEROCOM Phase 1 mean alone, the overall lifetime is decreased from 5.6 days to 4.8 days. Doing the same for wet deposition with no change in the dry deposition, the overall lifetime decreases from 5.6 days to 4.5 days.. Wet deposition accounts for 98% of the total deposition in ModelE2-TOMAS, which is much higher than AEROCOM Phase 1, and convective clouds contribute 27% of the wet deposition.

Global-annual EC in ModelE2-TOMAS is 0.19 Tg, which is very comparable to the ACCMIP mean (0.16 Tg). Similar to sulphate, wet deposition contributes >95% of total deposition of EC, which is higher than other ACCMIP models (see Table 3 in Lee et al., 2013a), and 24% of wet deposition is by convective clouds. For EC and OM, their lifetimes are longer than the ACCMIP mean as well as AEROCOM Phase 1 means but still within the standard deviation. Despite smaller dry deposition coefficients for EC and OM compared to the AEROCOM mean, their wet deposition coefficients are quite comparable to the AEROCOM mean.

The global annual burden of sea salt and dust in ModelE2-TOMAS are 3.6 Tg and 9.1 Tg, respectively. For sea salt and dust, dry deposition is as important as wet deposition due to their large particle sizes, accounting for 68% and 52% of total deposition, respectively. Since the size coverage of sea salt and dust in our model do not necessarily match with those in the AEROCOM Phase 1 models, we do not compare the lifetime and

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removal rate coefficients, which is strongly influenced by the upper size cutoff used in their emissions.

Despite the same host model and the same anthropogenic emission scenarios as the ModelE2-TOMAS model, the ModelE2-OMA model shows significantly different aerosol mass budgets (in Table 5), which must arise from using different deposition assumptions and other aerosol modelling treatments (see Section 2 for the details). Sulphate burden and lifetime in ModelE2-OMA is roughly half of that in ModelE2-TOMAS. The total source rate of SO_4 is about 20% higher than ModelE2-TOMAS and is close to the ACCMIP mean value. The ModelE2-OMA model has a shorter lifetime for EC, leading to ~40% lower burden compared to ModelE2-TOMAS. The OM burden is quite similar between two models. SOA formation rate is slightly different (14.6 Tg yr^{-1} for ModelE2-OMA and 17.1 Tg yr^{-1} for ModelE2-TOMAS), but the difference is only a small portion (about 3-4%) of the total OM source rate. The sea-salt emission rate is lower than that in ModelE2-TOMAS due to the maximum size cutoff of 8 μm assumed in sea-salt emission, but its burden is more than a factor of two higher. For dust particles, the emission rate is higher than that in ModelE2-TOMAS due to the coarser size cutoff in their emissions (i.e. up to 16 μm), and the burden is higher. For sea salt, the contribution of wet deposition to total deposition is more than 2 times higher than that in ModelE2-TOMAS. Unlike ModelE2-TOMAS, ModelE2-OMA has nitrate aerosol, which has a global burden of 1.6 Tg with a lifetime of 6.4 days.

The removal rate coefficient of dry deposition is about 50-60 times higher for sulphate, EC and OM in ModelE2-OMA, making it more comparable to the AEROCOM mean values. The lower dry deposition rates with the ModelE2-TOMAS model is likely due to the size-dependent dry deposition parameterization. The dry deposition velocity is not saved in the ModelE2 output currently, so alternatively we refer to the Figure 1 in Adams and Seinfeld (2001) that presents the global and annual-average of size-resolved dry deposition velocities in GISS-TOMAS (sulphate alone) compared to the size-independent one. Although the dry deposition velocities from ModelE2-TOMAS might not be exactly the same as the ones in the model version used in Adams and Seinfeld (2001) due to the updates made in ModelE2 (e.g. the boundary layer module), this point should be valid because the dry deposition parameterizations in both models have been

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little changed. Despite the large differences in dry deposition rates for accumulation mode particles, dry deposition is a fairly minor removal pathway in both models.

Figure 6 shows simulated global distributions of annual-mean concentrations of ModelE2-TOMAS sulphate, EC, OM, sea salt and dust in the lowermost layer. The sulphate concentrations are high over industrial regions, driven by the SO_2 emissions and OH/H_2O_2 oxidant concentrations. Simulated EC and OM concentrations are high over the biomass burning regions and the industrial regions, especially East Asia and South Asia, but the OM concentrations are particularly pronounced over biomass burning regions due to their higher emissions. Due to the SOA formation, the OM concentrations over Midwest US and Central Siberia are also noticeably high. The sea salt concentrations are distributed fairly uniformly over the oceans but are higher over the Southern Ocean and lower over the oceans near the tropics as expected due to wind speed variations. The dust concentrations are pronounced over the source regions such as Northern Africa, Arabia, Northern/Northwestern China, and Australia.

Figure 7 shows zonal distributions of annual-mean aerosol concentrations in ModelE2-TOMAS. The sulphate concentrations are highest between 0° N and 50° N due to the high anthropogenic emissions in the NH. Over the SH, the sulphate concentrations are mostly a result of DMS oxidation. The zonal-mean EC and OM concentrations are high from the tropics to ~50° N. Similar to Fig. 6, the high EC and OM concentrations are around 30° N to 50° N due to fossil fuel emissions, but the OM concentrations are also large around the tropics due to biomass burning emissions. The two small spikes shown the EC and OM concentrations between 10° S and 10° N are a result of the injection height used in the biomass burning emissions. Since a significant amount of sulfate and OM are also formed through chemical reactions in the atmosphere, their vertical gradients are relatively small. The sea salt concentrations are high from 60° S to 50° N with a peak around 30° S to 60° S due to the large open ocean in the SH. A strong dust plume is shown at around 0°-30° N due to the large Northern African and Middle East dust emissions, and a small dust plume at around 30° S due to Australian emissions. In the model, the PM10 concentrations in upper troposphere are dominated by sulfate and dust particles.

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5.3 Aerosol mass concentrations evaluation

The simulated surface-layer mass concentrations of aerosols are evaluated against various observations: 1) sulphate/sea salt/dust concentrations at 23 long-term observation sites operated by University of Miami (e.g. Prospero and Bonatti, 1969; Savoie and Prospero, 1989; Arimoto et al., 1990); 2) speciated PM2.5 concentrations from the Interagency Monitoring of Protected Visual Environment (IMPROVE) sites in the United States that are annual-averages from 2000 to 2008 (Debell et al., 2006; Hand et al., 2011); 3) speciated PM2.5 concentrations from various European observations (Putaud et al., 2010) (hereafter, referred to as European sites); 4) a large set of PM2.5 observations assembled in support of the Global Burden of Disease Study (GBD Study 2010, http://www.globalburden.org); 5) deposition flux measurements obtained from Ginoux et al. (2001), Tegen et al. (2002), and Mahowald et al. (2009). The PM2.5 dataset from the GBD Study consists of a worldwide set of annual-average PM2.5 largely drawn from official monitoring networks for 2005 (in some cases from 2004-2006). The GBD PM2.5 dataset includes the IMPROVE network and the European site measurements, so we only present the PM2.5 evaluation with the GBD dataset. For details of the GBD PM2.5 dataset, the reader is referred to the description in the supplementary material in Shindell et al. (2011).

Figure 8 compares the model annual-mean surface-layer sulphate mass concentrations to the observations from the IMPROVE network, the European sites, and the University of Miami network. Simulated sulphate agrees well with observations, mostly within a factor of two. Compared to ModelE2-OMA, the ModelE2-TOMAS model shows better agreement in the European sites (i.e. LMNB=-0.06 and LMNE=0.13 in Fig. 8b) but worse in the IMPROVE sites (i.e. LMNB=0.06 and LMNE=0.16 in Fig. 8a) and the remote oceanic sites (i.e. LMNB=0.04 and LMNE=0.22 in Fig. 8c). Over the US, both models overpredict systematically at lower observed concentrations (i.e. below 1 μg m^{-3} of measured SO_4 concentrations), which are mostly located over the western US.

Monthly mean surface-layer sulphate concentrations are evaluated using observations from the University of Miami in Figure 9. The simulated sulphate mass concentrations from both models usually falls within the standard deviation of the observed values. The

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sulphate predictions in both ModelE2-OMA and ModelE2-TOMAS become quite similar over the SH because about a factor of two lower DMS emissions are used in ModelE2-TOMAS. Using the same DMS emissions as ModelE2-OMA, ModelE2-TOMAS tends to overpredict sulphate concentrations noticeably over the SH (not shown), and the higher SO_4 concentration with ModelE2-TOMAS could be explained by 1) a longer lifetime due to different deposition parameterizations, 2) letting all SO_4 formed from aqueous oxidation to evaporate without accounting for cloud evaporation, 3) a stronger oxidation state resulted from un-degraded photolysis rates by aerosol optical depth (more SO_4 can be formed from DMS oxidation). Despite the fact that ModelE2-OMA accounts for the heterogeneous SO_4 formation on dust particles, SO_4 concentrations in near dust source regions are still higher in the ModelE2-TOMAS model due to the shorter lifetime in the ModelE2-OMA model.

Simulated annual-mean surface-layer sea salt concentrations are evaluated with the IMPROVE network, the European sites, and the University of Miami network (Fig. 10). Both aerosol models are biased strongly high over the US and Europe but biased low near the tropics. Unlike ModelE2-OMA, ModelE2-TOMAS underpredicts sea salt concentrations at several remote sites (see Fig. 10c). Compared to other aerosol components, the agreement between model sea salt and the observations is worse over the United States and Europe. For the evaluation of monthly mean surface-layer sea salt concentrations against the University of Miami dataset in Figure 11, the model predictions fall within the observed standard deviation at about a half of the 26 sites, but these sites are not necessarily the same between the two aerosol models. The overprediction of sea salt in continents may suggest that ModelE2 has a stronger transport from ocean to in-land, as there is no obvious overprediction over adjacent oceanic sites. The ModelE2-OMA model exhibits a particularly large overprediction over most SH sites. Both models tend to be biased significantly low at some of the sites near the tropics where the observed sea salt concentrations are high. Similar underprediction is also shown in mineral dust (see Fig. 13). This might be due to fast wet scavenging due to overpredicted precipitation in that area (see Fig. 9 in Schmidt et al., 2014).

For ModelE2-TOMAS, simulated annual-mean surface-layer dust concentrations are mostly within a factor of two of the measurements (in Fig. 12): 6 sites are excluded in

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Fig. 12 due to an incomplete annual cycle. ModelE2-TOMAS shows good agreement at the IMPROVE and European sites, while ModelE2-OMA tends to underpredict. This is due to the emission size assumptions (resulting in more clay emissions than ModelE2-OMA) and the differences in the deposition parameterizations (resulting in slightly longer lifetime for clay particles; ~ 9 days in ModelE2-TOMAS and 6.5 days in ModelE2-OMA). Both models are biased low compared to the University of Miami dataset, and only 7-8 sites among 20 sites fall within a factor of two agreement. A few sites show a severe underprediction for both models, mostly located near the tropics and in SH high latitudes.

Monthly mean surface-layer dust concentrations are evaluated using the observations from the University of Miami in Figure 13: no measurement data is available at 3 sites (Reunion Island, Invercargill, and Marion Island), but we still include them to compare the two aerosol model predictions. The model captures the observed annual cycle of dust very well at most NH sites but not in the SH sites except Norfolk and Mawson. The model captures the observed magnitude well at sites located relatively near the source regions (e.g., Sal Island, Barbardos, Bermuda for African dust; Jeju Island and Hedo for Asian dust; Cape Grim and Norfolk Island for Australian dust). Both models underpredict dust concentrations in the NH/SH high latitude (45°) regions except Mace Head, but the simulated dust seems to be within the observed standard deviation as the observations have a large standard deviation for dust. The observed peak concentration at Heimaey Iceland is the second highest after Sal island. Our models underpredict this site severely probably because our dust emission parameterization is not designed to simulate a dust event in humid areas such as Iceland. Prospero et al. (2012) points out that dust emissions at high latitudes (e.g., Alaska and Iceland) are mostly due to individual dust events or single seasons and link large dust events at Heimaey Iceland during 1997 to 2002 with glacial outburst floods.

Figure 14 compares simulated annual-mean dust deposition fluxes against observations obtained from Ginoux et al. (2001), Tegen et al. (2002), and Mahowald et al. (2009). Data is classified by the influencing source region, presented in different colours in Fig. 14. Except for the minor source category, the model dust deposition fluxes tend to be underestimated at most locations and agree with observations only within a

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factor of 5-8 on average. This may indicate that dust emissions are too low in ModelE2, but deposition fluxes measurements could contain large particles especially close to source regions (e.g., Duce, 1995) or local emissions (Uematsu et al., 1985) that are not simulated in the model. For ModelE2-TOMAS, the upper size limit being 10 μm, which is too small for the dust particles near the sources, can explain some of the severe underprediction, and Lee et al. (2009) also shows similar disagreement using the GISS-TOMAS model.

Figure 15 shows simulated annual-mean surface-layer EC and OM concentrations compared against the observations from the IMPROVE network and the European sites from Putaud et al. (2010). The simulated EC and OM in both models agree very well at the IMPROVE sites (for EC, LMNB=-0.08 to 0.05; for OM, LMNB=-0.17 to -0.08). Note that we applied an OM/OC ratio of 1.4 to the IMPROVE network to make it consistent with our model assumption, but the IMPROVE OM data provided to us was based on the ratio of 1.8. Over Europe, the model predictions are still reasonable (within a factor of 2-3), but the agreement is slightly worse than the IMPROVE sites. Since these sites are mostly adjacent to the source/emissions, the good agreement suggests that the emission inventory (used in this study) is well represented for these regions.

Figure 16 compares simulated annual-mean PM2.5 concentrations against the GBD dataset. Note that ModelE2-OMA includes nitrate mass into its PM2.5, and the nitrate contributes to PM2.5 rather significantly. The GBD data is classified/presented by a region listed in Figs. 16a and b. The aerosol models capture the observation quite well in most locations (overall LMNB is -0.08 to -0.03 and LMNE is 0.2 to 0.25), but both models show the worst agreement for Oceania regions (LMNB/LMNE=0.4-0.58) and Latin America (LMNB/LMNE =-0.33 to -0.48). The PM2.5 overprediction in Oceania is mainly due to too much fine mode sea-salt particles (the overall agreement in Oceana is little influenced by sulfate or dust particles). Note that the sea salt comparison to the Miami dataset (in Figures 11) shows severe underpredictions in several sites in Oceania because their concentrations are likely dominantly by coarse mode sea salt particles. The underprediction of PM2.5 in Latin America might be related to the biomass burning emissions. It is consistent with the model AOD being biased low over biomass burning source regions that is shown in Section 5.4. Model evaluation with the observed PM10

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concentrations using the IMPROVE and European sites was also performed and is similar to the PM2.5 evaluation results (not shown): LMNB and LMNE are -0.01 and 0.17 for ModelE2-TOMAS and 0.0 and 0.29 for ModelE2-OMA, respectively.

5.4 Aerosol optical depth evaluation

Simulated annual-mean clear-sky aerosol optical depths (AODs) at 550 nm are compared with observations from the Terra MODIS (MODerate resolution Image Spectroradiometer; e.g. Abdou et al., 2005; Remer et al., 2008) and MISR (Multiangle Image SpectroRadiometer; e.g. Diner et al., 1998; Kahn et al., 2005) satellite instruments averaged over 2004-2006 (Fig. 17). Specifically, we use Terra MODIS Level 3 (MOD08_M3.051), Aqua MODIS Level 3 (MYD08_M3.051), and Terra MISR Level 3 (MIL3MAE4), which are monthly products with 1x1 degree resolution. We also use the Deep Blue AOD (e.g., Hsu et al., 2006) from Terra MODIS to increase its spatial coverage, and all the satellite data was obtained from http://disc.sci.gsfc.nasa.gov/giovanni. When the Deep Blue AOD and the "regular" AOD from Terra MODIS are both available, the former is used. However, we notice that the Deep Blue AOD shows some local hotspots (e.g. over South America) that are not shown in the "regular" MODIS AOD as well as the MISR AOD. Our model calculates clear-sky AOD by including only AOD values calculated in model locations where clouds are not present (i.e., cloud-free grid-box only). The ModelE2-TOMAS and ModelE2-OMA models capture the broad spatial features seen in the satellite measurements: 1) very high AODs over desert regions in and near Northern Africa and the Arabian Peninsula associated with mineral dust, 2) the band of locally enhanced AOD over the Southern Ocean associated with sea salt, 3) high AODs over East Asia and India due to high anthropogenic emissions. However, the models underestimate the AOD over East Asia and the Indo-Gangetic plain, especially compared to MODIS. Including nitrate in ModelE2-OMA, the AOD predictions increase particularly over Europe and East Asia. Without nitrate, they become quite similar to the ModelE2-TOMAS AOD values in the regions where the model nitrate predictions are significant. The models show an enhanced AOD over the biomass burning regions such as tropical South America, Africa and Indonesia but it is clearly underestimated. The simulated AOD in North America and

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high latitude regions appears also to be lower than the satellite observations. Comparing with ModelE2-OMA AOD, ModelE2-TOMAS shows a stronger AOD over Africa due to its higher dust burden and a lower AOD over the marine areas especially the Southern Ocean associated with sea salt.

We present spatial correlations and biases between the models and the satellite data (Table 6). For these analyses, the annual-mean satellite AOD fields are regridded to 2°x2.5° horizontal resolution, and the models are sampled only where the satellite AOD is available. Correlation coefficients between the model and satellite AODs are around 0.6-0.7 for ModelE2-TOMAS and around 0.4-0.5 for ModelE2-OMA. Given that the correlation coefficient between MODIS and MISR is 0.79, the ModelE2-TOMAS model shows a good correlation with these satellite data. Both models show better correlations with MISR AOD. Compared to ModelE2-OMA, ModelE2-TOMAS shows a strong negative bias (about -29 to -34%) for both satellite datasets because of noticeably low AODs over the oceanic regions (where sea salt is dominant) shown in Fig. 16 and possibly because of missing component such as nitrate aerosols, which contributes to AOD significantly over Europe and China in the ModelE2-OMA model. Over Europe, the ModelE2-OMA model overpredicts AOD due to nitrate though. Without nitrate in ModelE2-OMA, the normalized mean bias (NMB) falls from 8-16% to -16 to -21%. Both models show lower AOD over China, India, and biomass burning regions and a similar underprediction is shown in by the ACCMIP models (Shindell et al., 2013), indicating a possibility of aerosol emissions being underestimated in these regions.

Figure 18 presents simulated monthly mean AODs compared against AERONET (AErosol RObotic NETwork; Holben et al., 1998; Holben et al., 2001) measurements at 28 sites that represent the following characteristic regions: polluted continental, marine, biomass-burning, and dusty regions (see Table 7 for individual site information). Both ModelE2-TOMAS and ModelE2-OMAunderestimate the maximum AOD during summer by a factor of 2-3 in the biomass burning sites (1 to 6) but capture the observed annual cycle quite well (R>~0.9). Unlike other biomass burning sites, simulated AODs at Ilorin and Banizoumbou are comparable to the observations possibly due to the influence of mineral dust. The agreement between the models and the AERONET AOD is generally good in the dusty regions (9 to 16). The ModelE2-TOMAS model shows a

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slight overprediction of AOD during spring at Capo Verde and Bidi Bahn which are located near the African dust sources, although it shows a good agreement at Barbados where is also influenced by African dust.

For ModelE2-TOMAS, all polluted continental sites (17 to 24) show large underpredictions, while the model tends to capture the observed annual cycle well. However, the model surface aerosol mass concentrations agree well with the observations from IMPROVE network and several European sites (see Figs. 8, 10, 12, 15, and 16). This might be due to the fact that column AOD depends on many additional factors (e.g., optical properties and vertical distribution of aerosols) and provides a measure of total radiatively active aerosols in the atmosphere. Misrepresentation of these factors or missing a chemical component could introduce a bias in the model AODs. For ModelE2-OMA, the model AOD without nitrate is quite similar to the ModelE2-TOMAS AOD. Including nitrate in ModelE2-OMA, it simulates the annual-mean AERONET AOD relatively well but the observed annual cycle worse. Although the inclusion of nitrate is helpful for the underpredicted AOD in polluted regions, the overprediction of wintertime AOD suggests that the model nitrate is too large.

With the ModelE2-TOMAS model, the oceanic sites (25 to 30) are generally underpredicted roughly by a factor of two except for Bermuda (25) and Lanai (26). By contrast, the ModelE2-OMA model captures the observed magnitude relatively well but overpredicts at Bermuda and Lanai. Note that Bermuda (23) and Rottnest Island (27) are influenced by long-range transported mineral dust. The ModelE2-OMA model predicts AOD that is more comparable to observations in these oceanic sites than ModelE2-TOMAS. Compared to ModelE2-OMA, the underprediction of sea salt concentrations at the remote sites (shown in Figs. 10 and 11 in Sect. 5.3) and the underpredictions of AODs in the remote oceanic sites in ModelE2-TOMAS may be due to a faster sea salt removal rate (see global budgets in Table 4 for the details). It is worth mentioning that the inclusion of marine organic aerosols, which are not included in this paper, may not increase AOD noticeably in remote oceanic sites. Using the same host model, Tsigaridis et al. (2013) shows that Southern Ocean AOD is quite insensitive to the inclusion of marine organic particles but is strongly sensitive to the sea-salt emissions parameterization (see Figure 9 of Tsigaridis et al. (2013).

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5.5 Aerosol number budgets and its distributions

This section includes only ModelE2-TOMAS results, as ModelE2-OMA does not predict aerosol number concentrations. Global mean number budgets for all three simulations, including two sensitivity runs for nucleation rates, are presented in Table 8 (see Section 4.2 and Table 1 for the details of run descriptions). Compared to the BASE run, global mean CN3 (particles with dry diameters larger than 3 nm), CN10 (particles with dry diameters larger than 10 nm), and CN100 (particles with dry diameters larger than 100 nm) concentrations in the LowNUC run are decreased by 74%, 33%, and 7% in the troposphere and 29%, 17%, and 3% in the lowermost layer, respectively. The smaller impact on CN in the lowermost layer is due to the presence of the primary emissions near the surface. It is also because the binary nucleation parameterization used in this model produces few particles in the boundary layer; however, nucleated particles in the upper/free troposphere may be carried down to the surface and influence CN there. Since the LowNUC run produces fewer nucleated particles than the BASE run, each nucleated particle is more likely to grow more efficiently to form CCN-sized particles, as there is less competition for sulphuric acid and condensable organics. We can see this using the number budgets in Table 8. The increase in CN70 with the BASE case nucleation (i.e., BASE CN70 – NONUC CN70) is 51 cm^{-3} for the nucleation rate (J3) of 0.131 cm^{-3} s^{-1} and that with the LowNUC case nucleation, 37 cm^{-3} for J3 of 0.013 cm^{-3} s^{-1}. In the BASE run, J3 is 10 times higher but the CN70 increase by nucleation is only ~1.4 times higher than those in the LowNUC run.

Aerosol number burdens normalized by tropospheric volume are dramatically reduced when nucleation is turned off in the NoNUC run. For example, global mean CN3 and CN10 are decreased by 95% and 76% in the troposphere and by 42% and 31% in the surface layer. We found that 24% of tropospheric CCN-sized particles (i.e., CN100) and ~10% of surface-layer CCN-sized particles result from binary nucleation in our model; the contribution of the nucleated particles to the CCN concentrations is larger as the cutoff size (e.g. 100 nm in CN100) gets lower.

Figure 19 shows global distributions of annually averaged CN3, CN10, and CN100 in the lowermost layer for the BASE, LowNUC, and NoNUC runs, and Fig. 20 presents their zonal distributions. For the CN3 distributions, the BASE run shows a high

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concentration (over 5000 cm^{-3}) in the upper/free troposphere and the entire Antarctica troposphere due to nucleation and over polluted continental regions due to the primary emissions (see Figs. 19a and 20a). Note that the CN3 near the surface in Antarctica comes from nucleated particles from the upper/free troposphere, as the binary parameterization does not predict nucleation in the boundary layer. CN3 in the LowNUC run (Figs. 19d and 20d) is decreased significantly in the free troposphere and in the surface layer over Eurasia and western Antarctica (fewer nucleated particles formed above subside to the surface). When nucleation is switched off, CN3 is very close to CN10 near to the surface (Figs. 19g and h) because nucleation contributes most CN between 3 nm to 10nm. For CN10, its spatial pattern over the continents is quite similar among the runs as it is mostly driven by primary emissions except for some locations heavily influenced by the nucleated particles formed in the upper/free troposphere (Figs. 19b, e and h and Figs. 20b, e, h). A pronounced difference in CN10 is shown over oceans, indicating a larger contribution of nucleated particles to CN10 in these regions. This is consistent with Merikanto et al. (2009), which shows higher contribution of nucleation to CN over oceanic regions. CN100 differs little among runs except for SH high latitudes where binary nucleation plays an important role (Figs. 19c, f, and i). This is consistent with Pierce and Adams (2009) showing that the exhibited high SH latitude region for the most positive changes in CCN(0.2%) by turning on binary nucleation and, again, with Merikanto et al. (2009) showing 65% of CCN(0.2%) in Antarctica resulted from upper tropospheric nucleation. CN100 shows a maximum at 10° N to 40° N because most anthropogenic primary emissions are located in mid-NH latitudes. Rather surprisingly, dust particles in our model contribute to CN100 quite significantly over the source regions. This is opposite to the results from GISS-TOMAS (Lee et al., 2009), which shows a 10-20% reduction in CCN(0.2%) when introducing mineral dust emissions. Despite the direct source of CCN-sized particles from dust emissions, CCN and ultrafine particles that grow to become CCN are scavenged via coagulation with coarse dust particles, and dust particles compete for condensable sulfuric acid, leading to a slower growth rate of ultrafine particles.

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5.6 Aerosol number evaluation

The CN measurement dataset compiled by Spracklen et al. (2010) is used to evaluate simulated annual-mean CN concentrations (in Fig. 21) and monthly mean CN concentrations (in Fig. 22). Details of the measurement procedures for each site, including the instrument type and minimum cutoff diameter (varying from 3nm to 14 nm), can be found in Table 2 of Spracklen et al. (2010). Due to an incomplete annual cycle, we excluded Mt. Waliguan, Finokalia, Listvyanka, and Weybourne. The measurement sites are classified into three categories: FT (free troposphere; 1 to 8 in Fig. 22), MBL (marine boundary layer; 9 to 15 in in Fig. 22), and CBL (continental boundary layer; 16 to 32 in Fig. 22). We sampled the model values to match the altitude of each measurement site except for the free troposphere sites that use an altitude 30% lower because it improved the annual cycles prediction significantly. Previous studies pointed out that free tropospheric sites can be influenced by upslope winds that carry the planetary boundary layer air, so it cannot be assumed to be in the free troposphere all the time (e.g., Baltensperger, 1997; Collaud Coen et al., 2011). Our model does not seem to simulate this well, so sampling the model predictions at lower altitude (i.e., 30% lower) helps to increase the influence of PBL air. In Fig. 21, a whisker plot is used to present the three run results; the maximum of the whisker line for the BASE run; the circle symbol in the middle of the whisker line for the LowNUC run; the minimum for the NoNUC run.

On average, the annual-mean CN concentrations in the model agree with the observations well for the all three categories (LMNB= -0.26 to 0.16; LMNE=0.13 to 0.22; see Fig. 21), although the LowNUC simulation shows the best agreement to observation. Simulated annual cycles at individual sites also show that the model captures the measured magnitude reasonably but overpredicts the CN during winter (November to March) that is worse at the CBL sites and results in poor seasonality (especially in the BASE run). The poor seasonality in ModelE2-TOMAS in all three runs suggests that other factors may play a role such as missing seasonal variation in primary emissions or scavenging that causes the poor seasonality rather than nucleation. The evaluation of CN at South Pole shows that the model predicts too strong nucleation throughout the year.

Figure 23 compares the observed size distributions at six European sites during winter (DJF: December to February; Figs. 23a to f) and summer (JJA: June to August; Figs. 23d to l) to the model. The observed size distributions are averaged during morning,

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afternoon, and night, while the model results are not broken into the three periods. This data is obtained from Putaud et al. (2003) and, for the same sites shown in Figure 21, the temporal coverage used in Putaud et al. (2003) is not necessarily matched with them. Most sites are reasonably close to the dataset used in Figure 21. However, for Jungfraujoch, the total CN concentrations summed from the size distribution data, which covers from June 1997 to May 1998, is about a factor of two lower than the CN shown in Figure 21, which covers from 1995 to 1999 and 2003-2007. As expected, the simulated size distributions of the model simulations differ according to what nucleation scheme is used. The impact of nucleation is more notable for particles smaller than ~50 nm, as expected, and also during winter. In general, the higher nucleation rates tend to overpredict Aitken model particles at most sites. For Harwell (Figs. 23c and i), particles below 50 nm are overpredicted even without nucleation. For the summer season, the model shows less sensitivity to nucleation rates and has better skill at capturing the observed size distributions.

The observed CN5, CN15, and CN120 concentrations from the LACE campaign (Petzold et al., 2002) are compared with three model runs (Figs. 24a to c). Below 700-800 mbar, all three model runs predict concentrations roughly within the observed CN5, CN15, and CN120 ranges. Above 700-800 mbar, the BASE simulation overpredicts CN5 and CN15 by approximately an order of magnitude and 2-3 times, respectively. For the NoNUC simulation, it captures the lower side of the observed CN15 but fails to capture the increasing CN5 concentrations with height above 600 mbar. All model runs basically show almost identical CN120 concentrations and fall on the lower edge of the observed range.

We compare CN3 vertical profile measurements averaged into the 3 latitude bands over the Pacific Ocean (Clarke and Kapustin, 2002) with the model (Figs. 24 d to f). The simulated CN3 profiles in LowNUC agree well with the observation, capturing the increasing CN3 with height. Although the BASE run shows the increasing pattern correctly, it overpredicts CN3 severely above approximately 6 km. The NoNUC run disagrees with the observations for all latitudes and altitutes and clearly fails to reproduce the high number concentrations in the upper troposphere that result from nucleation.

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The observed Aitken mode and accumulation mode concentrations and size distributions in the marine boundary layer (Heintzenberg et al., 2000) are compared with the model in Figs. 25 and 26. The measurements are aggregated into 15-degree latitude ranges. For the Aitken mode (Fig. 25a), the LowNUC run shows the closest agreement to observations. However, whereas the observations show higher concentrations in the SH than in the NH, all model simulations show the opposite tendency. Similarly, other global models with binary nucleation show underpredicted CN concentrations in the SH and either well-simulated or overpredicted CN in the NH (e.g. Easter et al., 2004; Spracklen et al., 2005; Pierce and Adams, 2006; Pierce et al., 2007; Trivitayanurak et al., 2008; Mann et al., 2010; Wang et al., 2011). For the accumulation mode (Fig. 25b), the three model runs are quite similar to each other and are within the observed range.

In Fig. 26, ModelE2-TOMAS captures the bimodal size distribution shown in the observations reasonably, which is mainly determined by the activation diameter assumed in wet deposition (and cloud processing): ~80 nm as an activation diameter (supersaturation of 0.2%) for both large-scale clouds and convective clouds in the model. Note that ModelE2-TOMAS cannot capture the observed bimodal distribution when a supersaturation of 1.0% is assumed for convective clouds, unlike the other TOMAS models (e.g. Pierce et al., 2007;Trivitayanurak et al., 2008): a peak at around 20-30 nm appears when the supersaturation of 1.0% is assumed for convective clouds. This suggests that, compared to GISS GCM II', convective clouds are more frequent in ModelE2. Although the model captures the observations successfully using fixed supersaturation assumptions, future work is needed to link the in-cloud supersaturation to cloud and aerosol properties.

Simulated CCN concentrations are compared against a dataset of CCN measurements compiled by Spracklen et al. (2011) in Figs. 27 and 28: see Table 1 in Spracklen et al. (2011) for the details regarding each site. The CCN dataset includes a total of 277 measurements at 80 locations using various instruments from 1971 to 2009. Approximately 70% of the observations were taken after 1990. Most have sampling periods of days to weeks except the observations at Cape Grim and Mace Head. For Cape Grim and Mace Head, an annual cycle is available, so we present them separately in Fig. 28. In Fig. 27, the CCN data is divided into two groups: CCN in the MBL (marine

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boundary layer) and CCN in the CBL (continental boundary layer). Note that all CCN measurements used here are in the boundary layer. For CCN in the CBL, the model CCN shows good agreement with the observation in all three simulations (LMNB=0.11-0.2 and LMNE=0.31-0.34). For CCN in the MBL, the model predictions are, on average, within a factor of 1.5-2 of the observations for all three runs but, relative to several measurements of CCN concentration between ~100 and ~300 cm^{-3}, are biased high, by roughly a factor of two.

For the annual cycle of CCN concentrations at Cape Grim and Mace Head (Fig. 28), the model overpredicts in all months even without nucleation (NMB>1.0). The CCN overpredictions at Cape Grim might be influenced by overpredicted SO_2 (shown in Fig. 3), which could lead to overpredicted condensational growth of Aitken mode particles. As with the SO_2 evaluation in Fig. 3, the CCN overprediction decreases by sampling adjacent grids toward the ocean (not shown), but the model CCN is still higher than the measurements. Given the fact that most CCN observations have very short duration (days to weeks) in a single year and, according to Spracklen et al. (2011), the relative uncertainties in the measurement data range from about 5-40%, mostly in 10-20% very roughly, the overall model-to-observation agreement is satisfying.

6. Conclusions

We have implemented the TwO-Moment Aerosol Sectional (TOMAS) microphysics model into the new version of the GISS GCM (i.e. ModelE2), called "ModelE2-TOMAS". This paper has compared the global budgets of ModelE2-TOMAS to other global aerosol models and evaluates the model with various observations such as aerosol precursor gas concentrations, aerosol mass and number concentrations, and aerosol optical depth.

Global budgets of aerosols and aerosol precursor gases in ModelE2-TOMAS are similar to those in other global aerosol models, and the ModelE2-TOMAS model agrees reasonably (mostly within a factor of two) with long-term observed aerosol precursor gas and aerosol mass concentrations. The model captures the broad spatial features shown in the MODIS and MISR annual-mean AOD distributions as well as the observed seasonal

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trends of AOD at several AERONET sites. The model predicts the observed annual-mean CN (the minimum cutoff varying from 3nm to 14 nm) concentrations very well and the observed vertical profiles of aerosol number over Germany (i.e., LACE campaign) and in the marine boundary layer. For CCN, the model shows good skill in capturing the observations. We conclude that the model is realistic enough to be useful for many types of scientific study.

However, the evaluations have also highlighted some weaknesses in ModelE2-TOMAS to be revisited in the future. First, ModelE2-TOMAS predicts too much SO_2 lifted into the upper/free troposphere over the Pacific Ocean possibly due to overly strong convective transport. This is also seen in ModelE2-OMA (and in gaseous tracers), suggesting this might be a host model problem. Second, the model AOD is underpredicted over polluted continents, even though mass concentrations of each aerosol component at the surface are well simulated (or at least not underpredicted) in the model. Missing nitrate in ModelE2-TOMAS may not be the main contributor, as the inclusion of nitrate in ModelE2-OMA decreases its ability to capture the observed seasonality in AERONET polluted continents sites. Third, the model tends to underpredict aerosol loading (and AOD) over biomass burning emission regions. This is a common issue in global aerosol models, and this might be due to underestimation of biomass burning emissions. Fourth, the ModelE2-TOMAS AOD prediction is biased low over the SH high-latitude oceans, which suggests underpredicted sea salt burden in this area. Fifth, the simulated CN seasonality is poor at some CBL sites due to overpredicted CN during the wintertime. Similarly, ModelE2-TOMAS predicts the observed number size distributions over European sites during the summer season reasonably but not for the winter season. The model overpredicts Aitken mode particles during the winter season, which happens even without nucleation, possibly due to a problem in primary emissions representation or a bias in the model scavenging that causes the poor seasonality. Finally, ModelE2-TOMAS seems to predict faster nucleation rates using binary nucleation (Vehkamaki et al., 2002) than other global microphysics models using the same binary nucleation (including GISS-TOMAS). For instance, the observed CN vertical profiles are captured the best when nucleation rates are reduced (our LowNUC run), while Lauer et al. (2005) presents significantly underpredicted CN profiles with the same binary nucleation scheme

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using ECHAM/MADE. To investigate this issue further, we need to perform a model evaluation against observation-derived nucleation-relevant metrics, which has been done in Westervelt et al. (2013). Because of the fine size assumptions for the primary emissions (see Section 4.2 for the details) and/or the fast nucleation rates using binary nucleation in ModelE2-TOMAS, we do not include boundary nucleation as it overpredicted CN near the surface. We conclude that further work is necessary to improve the realism of some aspects of ModelE2-TOMAS and to better understand the size-resolved physical processes (e.g. microphysics, emissions, and depositions).

Comparing the ModelE2-TOMAS model with the ModelE2-OMA model in ModelE2, some aerosol species burdens and concentrations differ significantly, even with using the same host model and identical anthropogenic emissions, mainly because the deposition parameterizations and some of the emission-related assumptions are different. In the case of sea salt and dust, the size ranges assumed by each model are different, resulting in different emission rates and burdens.

Analysis of multiple aerosol model results help to identify where a model bias might originate from: aerosol modelling or the host GCM or elsewhere such as emissions. We found that some of the large differences in aerosol predictions between the two aerosol models are due to aerosol modelling. This is valuable information, as this is not easy to constrain using observations. Having more than one aerosol physics representation in the NASA GISS ModelE2 will serve as a useful tool to study the uncertainty in aerosol modelling and to guide our efforts to improve the models.

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Acknowledgements

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Table 1. Aerosol and precursor gas emissions used in ModelE2-TOMAS and ModelE2-OMA and the nucleation scheme used in the ModelE2-TOMAS simulations

Emission/Process	TOMAS model	Bulk model
Anthropogenic emissions	CMIP5 2000 emissions (Lamarque et al., 2012)	CMIP5 2000 emissions (Lamarque et al., 2012)
Biomass burning emissions	Climatological-average GFED3 emissions from 1997 to 2009 (van der Werf et al., 2010)	Climatological-average GFED3 emissions from 1997 to 2009 (van der Werf et al., 2010)
Primary sulfate emission assumption	1.0% of total sulfur emissions	2.5% of total sulfur emissions
DMS emission	Seawater DMS concentrations from Kettle et al. (1999) Sea-to-air transfer function from Liss and Merlivat (1986)	Seawater DMS concentrations from Kettle et al. (1999) Sea-to-air transfer function from Nightingale et al. (2000)
Sea-salt emission	Gong et al. (2002); the upper diameter limit of 10 um	Gong et al. (2002); the upper diameter limit of 8 um
Dust emission	See text for the details; the upper size diameter of 10 um	See text for the details; the upper size diameter of 16 um
Nucleation	Three nucleation cases 1. BASE - Binary nucleation 2. LowNUC - Binary nucleation with 5 times lower sulfuric acid concentrations 3. NoNUC – no nucleation	N/A

Table 2. Emission size distributions assumed in ModelE2-TOMAS. *This is the soil size assumption used in ModelE2-TOMAS, and the dust emission size distribution is additionally influenced by meteorological variables. GMD stands for geometric mean diameter, and GSD for geometric standard deviation.

Species	Emissions	Size assumptions
Sulfate	All emissions	Bimodal distribution GMD=10nm, GSD=1.6 (5% of total mass) GMD=70 nm, GSD=2.0 (95% of total mass)
EC and OC	Fossil fuel and Biofuel Biomass burning	GMD=60 nm, GSD=1.59 GMD=150 nm, GSD=1.59
Dust*	Clay Silt	GMD=140 nm, GSD=2.0 GMD=1.15 μm, GSD=2.0

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Table 3. Global budgets for DMS and SO_2 from the BASE run in ModelE2-TOMAS and ModelE2-OMA. The ModelE2-TOMAS values are presented before slash and the ModelE2-OMA values are after slash. Values in parentheses are ranges from other global models including Wang et al. (2011), Liu et al. (2005), and those listed in Liu et al. (2005).

	DMS	SO2
Burden [Tg S]	0.05 / 0.11 (0.02-0.15)	0.36 / 0.38 (0.2-0.69)
Total source [Tg S yr^{-1}]	16.1 / 28.7	80 / 90
Emission	16.1/ 28.7 (10.7-23.7)	65.6 / 64.7 (61.2-92.0)
Chemistry	-	14.4 / 25.3
Sink [Tg S yr^{-1}]	16.1/ 28.7	80 / 89
Gas-phase oxidation	16.1/ 28.7	12.3 / 14.6 (6.1-22.0)
Aqueous-phase oxidation	-	30.8 / 35.8 (24.5-57.8)
Wet deposition	-	0.36 / 0.4 (0-19.9)
Dry deposition	-	37 / 38.8 (15.78-55)
Lifetime [days]	1.2 / 1.5 (0.5-3.0)	1.9 / 1.5 (0.6-2.6)

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Table 4. Global aerosol budgets of the BASE run in ModelE2-TOMAS. Values in the parentheses are the mean and normalized standard deviations obtained from Table 10 in Textor et al. (2006). The mass budgets for sulfate are presented as Tg S.

	Sulfate	Elemental carbon (EC)	Organic matter (OM)	Sea-salt	Dust
Burden [Tg]	0.67	0.19	1.2	3.6	9.1
Total source [Tg yr^{-1}]	43.7	7.4	60.8	3231.9	705.8
Emission	0.66	7.4	43.7	3231.9	705.8
Wet deposition [Tg yr^{-1}]	42.9	7.1	59.1	1046.9	336.8
By convective clouds [%]	27	24	24	54	29
Dry deposition [Tg yr^{-1}]	0.8	0.3	1.6	2184.9	369.9
Lifetime [days]	5.6 (4.1, 18%)	9.6 (7.1, 33%)	7.2 (6.5, 27%)	0.4 (0.5, 58%)	4.7 (4.1, 43%)
Removal rate coefficient [day^{-1}]	0.18 (0.25, 18%)	0.1 (0.15, 21%)	0.14 (0.16, 24%)	2.4 (5.1, 188%)	0.21 (0.31, 62%)
Wet deposition	0.18 (0.22, 22%)	0.1 (0.12, 31%)	0.14 (0.14, 32%)	0.79 (0.79, 77%)	0.1 (0.08, 42%)
Dry deposition	0.0032 (0.03, 55%)	0.004 (0.03, 55%)	0.0037 (0.03, 49%)	1.6 (4.3, 219%)	0.11 (0.23, 84%)
Wet/(Wet+Dry) [%]	98 (89, 8%)	96 (79, 17%)	97 (80, 16%)	32 (31, 65%)	48 (33, 54%)

Table 5. Global aerosol budgets in ModelE2-OMA. Note that the sulfate and nitrate budgets are presented as Tg S and Tg N, respectively.

	Sulfate	EC	OM	Sea-salt	Dust	Nit
Burden [Tg]	0.38	0.12	1.1	9.3	11.4	0
Total source [Tg yr-1]	52.0	7.4	58.5	2866.7	1071.8	21.0
Emission	1.7	7.4	43.7	2866.7	1071.8	
Wet deposition [Tg yr-1]	46.6	5.4	44.9	2059.1	407.7	17.7
By convective clouds [%]	21	37	32	39	52	29
Dry deposition [Tg yr-1]	5.4	2.0	13.7	806.9	664.1	3.3
Lifetime [days]	2.6	5.8	7.1	1.2	3.9	6.4
Removal rate coefficient [day-1]	0.39	0.17	0.14	0.84	0.26	0.16
Wet deposition	0.35	0.13	0.11	0.61	0.10	0.13
Dry deposition	0.04	0.046	0.033	0.24	0.16	0.025
Wet/(Wet+Dry) [%]	90	73	77	72	38	84

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Table 6. Statistical measures of model predictions compared to satellites. For ModelE2-OMA, the model prediction without nitrate aerosols is also presented in parentheses.

		Model vs. MODIS	Model vs. MISR	MODIS vs. MISR
ModelE2-TOMAS	Correlation	0.63	0.73	0.79
	NMB [%]	-29%	-34%	8%
ModelE2-OMA	Correlation	0.45 (0.45)	0.52 (0.55)	0.79
	NMB [%]	16% (-16%)	8% (-21%)	8%

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Table 7. Locations of AERONET sites and corresponding measurement time periods.

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	Sites	Longitude	Latitude	Years
1	Alta Floresta	56.0° W	9.9° S	1999-2005
2	Abracos Hill	62.0° W	11° S	1999-2005
3	Cuiaba-Miranda	56.0° W	15.7° S	2001-2005
4	Mongu	23.2° E	15.2° S	1995-2005
5	Ilorin	4.3° E	8.3° N	1998-2005
6	Banizombou	2.0° E	13.0° N	1995-2005
7	Capo Verde	22.9° W	16.7° N	1994-2004
8	Bidi Bahn	2.5° W	14.1° N	1996-1997
9	Barbados	59.5° W	13.2° N	1996-2000
10	Sede Boker	34.8° E	30.9° N	1998-2005
11	Bahrain	50.6° E	26.2° N	2004-2005
12	Solar Village	46.4° E	24.9° N	1999-2005
13	Dalanzadgad	104.4° E	43.6° N	1997-2005
14	Yulin	109.7° E	38.3° N	2001-2002
15	Sevilleta	106.9° W	34.4° N	1994-2005
16	Cart site	97.5° W	36.6° N	1996-2005
17	Bondville	88.4° W	40.1° N	1996-2005
18	GSFC	76.8° W	39.0° N	1995-2005
19	Mexico city	99.2° W	19.3° N	1999-2005
20	Ispra	8.6° E	45.8° N	2001-2005
21	Kanpur	80.3° E	26.5° N	2001-2005
22	Shirahama	135.4° E	33.7° N	2000-2005
23	Bermuda	64.7° W	32.4° N	1996-2005
24	Lanai	156.9° W	20.7° N	1996-2004
25	Dry Tortugas	82.9° W	24.6° N	1996-2003
26	Tahiti	149.6° W	17.6° S	1999-2005
27	Rottnest Island	115.5° E	32.0° N	2001-2004
28	Nauru	166.9° E	0.5° S	1999-2005

Table 8. Summary of global annual-average of tropospheric and surface-layer aerosol number budgets including J3 (new particle formation rates at 3 nm), CN3 (number concentration of particles with Dp ≥ 3 nm), CN10(Dp ≥ 10 nm), CN100(Dp ≥ 100 nm) in ModelE2-TOMAS. Values normalized by tropospheric volume at 273 K and 1 atm.

	Emission rate [cm^{-3} s^{-1}]	Tropospheric J3 [cm^{-3} s^{-1}]	Tropospheric CN3 [cm^{-3}]	Tropospheric CN10 [cm^{-3}]	Tropospheric CN100 [cm^{-3}]	Surface-layer CN3 [cm^{-3}]	Surface-layer CN10 [cm^{-3}]	Surface-layer CN100 [cm^{-3}]
Base	5.47×10^{-4}	0.131	4852	939	211	1622	1331	416
LowNUC	5.47×10^{-4}	0.013	1277	628	197	1152	1111	405
NoNUC	5.47×10^{-4}	0.000	222	221	159	935	919	374

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Figure 1. Annual-average column mass concentrations of (a) DMS and (b) SO_2 in the ModelE2-TOMAS BASE run. Units are μg S m^{-2}.

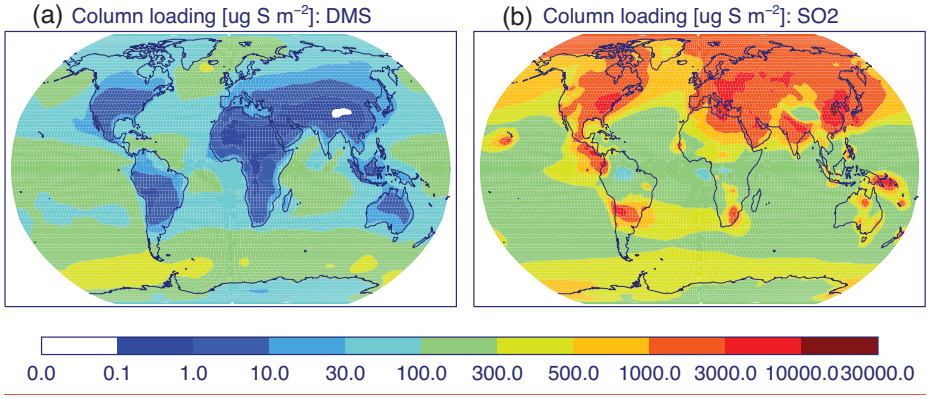

Figure 2. Scatter plot of annual-mean surface SO_2 concentrations [μg m^{-3}] for the model (red for ModelE2-TOMAS and blue for ModelE2-QMA) compared to the observations in the EMEP (European Monitoring and Evaluation Programme, Loevblad et al., 2004; a) and CASTNET (Clean Air Status and Trends Network, Malm et al., 2002; b) networks. Log-mean normalized bias (LMNB) and log-mean normalized error (LMNE) are given.

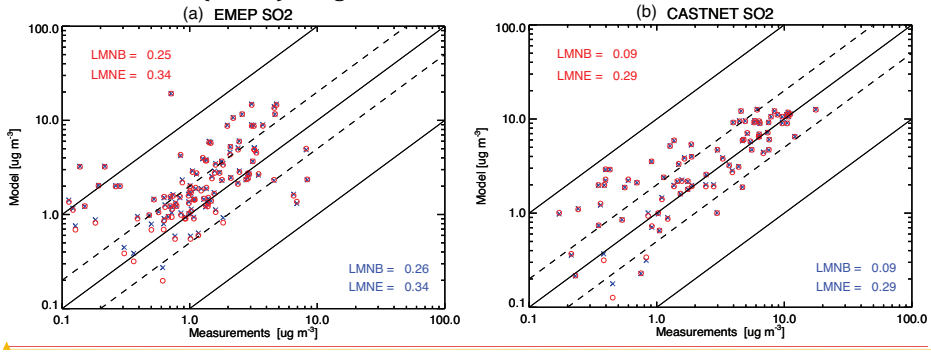

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Figure 3. Comparisons of monthly averaged surface DMS (a to c) and SO_2 (d to e) mixing ratios [pptv] simulated (red for ModelE2-TOMAS and blue for ModelE2-OMA) and measured (black) at Amsterdam Island [Nguyen et al., 1992] and Cape Grim [Ayers et al., 1991]. Only DMS at Dumont D'Urville [Jourdain and Legrand, 2001]. Correlation (R) and normalized mean bias (NMB) are given.

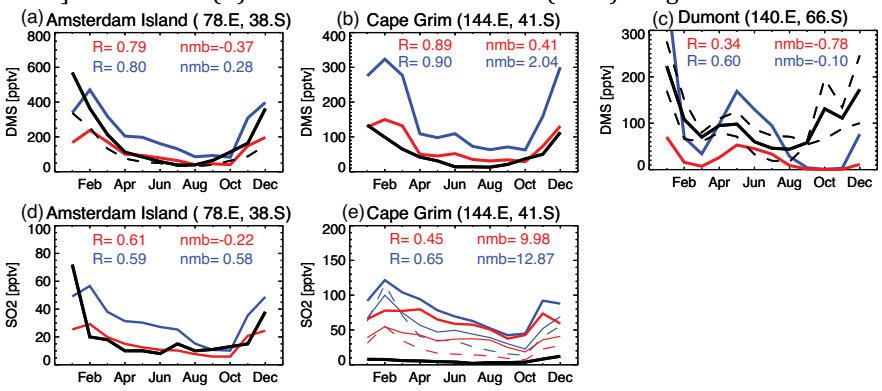

Figure 4. Comparison of DMS vertical profiles over the Pacific Ocean simulated (red for ModelE2-TOMAS and blue for ModelE2-OMA) and observed (black solid line). Observations are from PEM-Tropic-A (August-October 1996 in the tropical Pacific; Christmas Island in a; Hawaii in c; Tahiti in e; Easter Island in g; Fiji in h) and PEM-Tropic-B (March-April 1999 in the tropical Pacific; Christmas Island in b; Hawaii in d; Tahiti in f). The dashed lines represent 25th and 75th percentiles of the observed values.

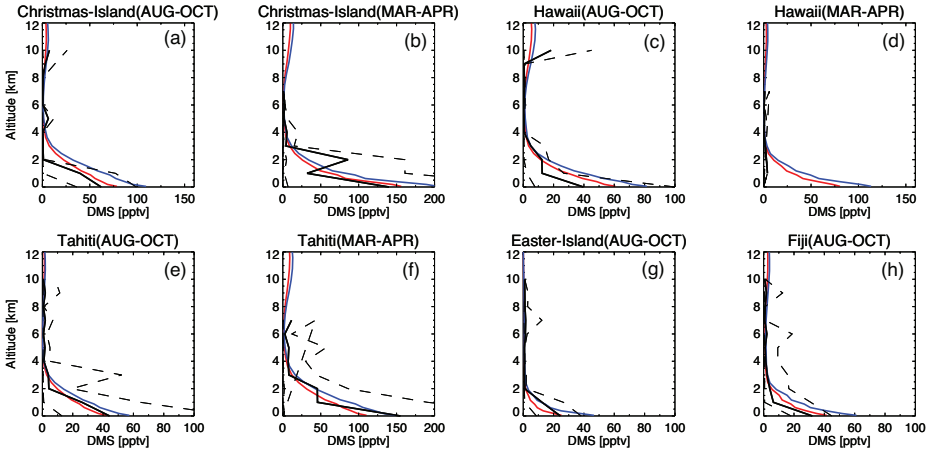

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Figure 5. The same as Fig. 4 but for SO_2 vertical profiles.

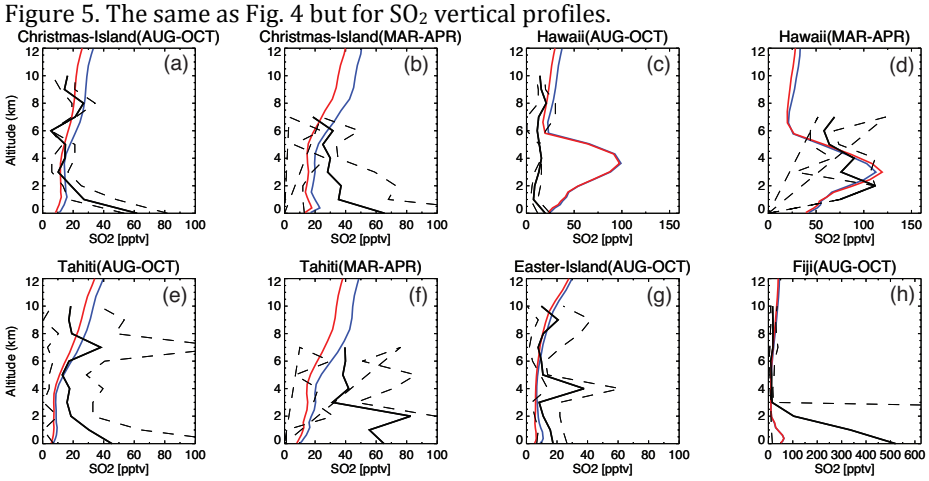

Figure 6. Annual-mean concentrations of (a) sulfate, (b) EC, (c) OM, (d) sea-salt, and (e) dust in the lowermost layer in the ModelE2-TOMAS BASE run. Units are μg m^{-3}.

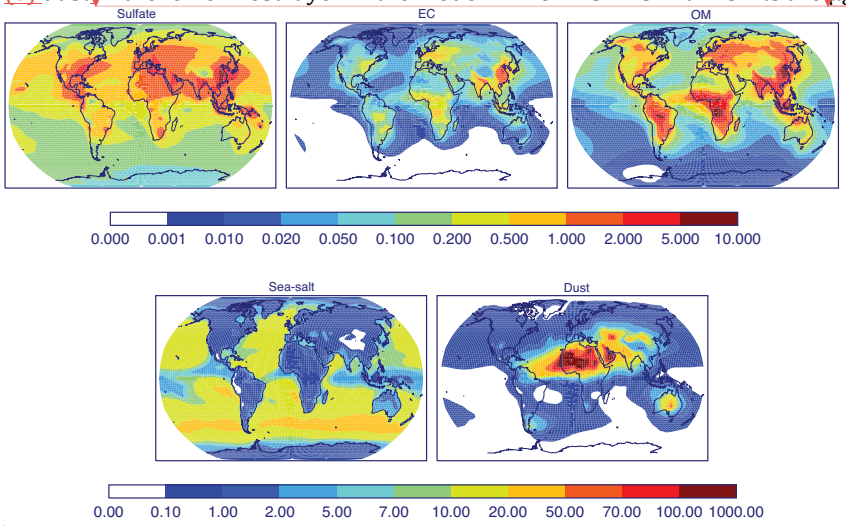

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Figure 7. Annually and zonally averaged concentrations of (a) sulfate, (b) EC, (c) OM, (d) sea-salt, and (e) dust in the ModelE2-TOMAS BASE run. Units are μg m^{-3}.

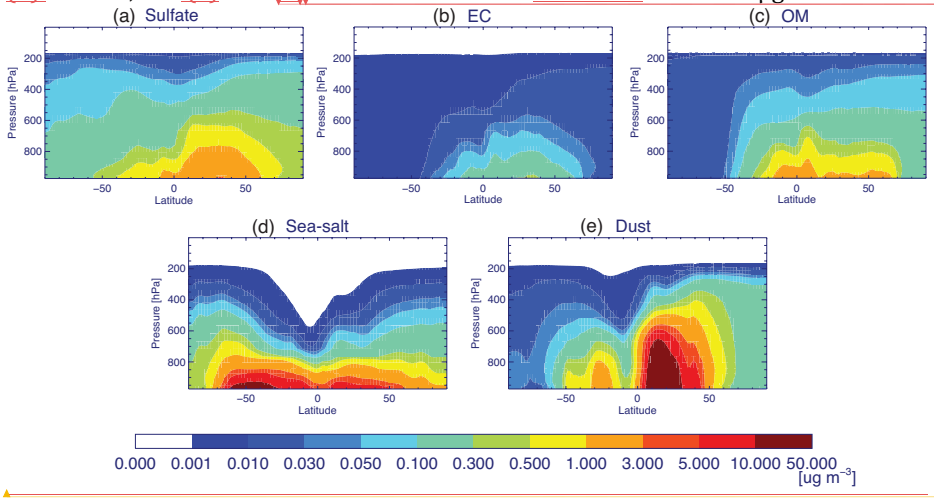

Figure 8. Scatter plot of annually averaged surface sulfate mass concentrations [μg m^{-3}] in the model (red for ModelE2-TOMAS and blue for ModelE2-OMA) and observation from (a) the IMPROVE network, (b) the European sites from Putaud et al. (2010), and (c) University of Miami. Note that the sulfate in the model is simply converted to ammonium sulfate at the IMPROVE sites.

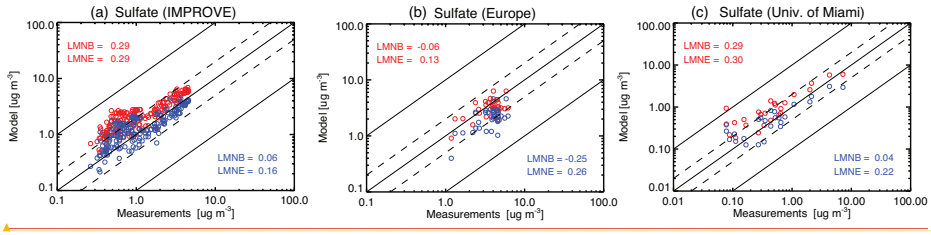

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Figure 9. Comparisons of monthly averaged surface sulfate mass concentrations [μg m^{-3}] simulated (red for ModelE2-TOMAS and blue for ModelE2-OMA) and measured (black solid line with an error bar showing a standard deviation) by University of Miami. Correlation (R) and normalized mean bias (NMB) are provided only when the observation is available for 12 months.

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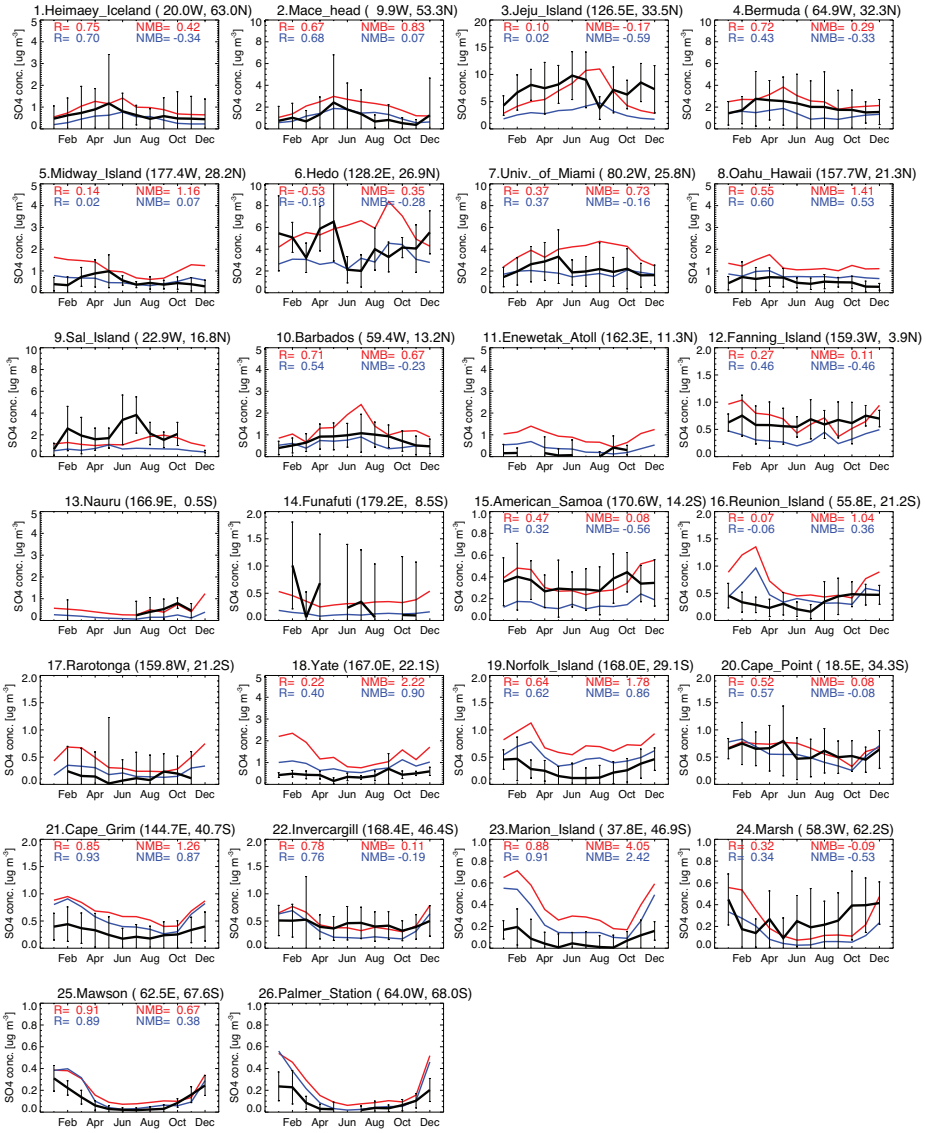

Figure 10. Same as Figure 8 but for sea-salt mass concentrations

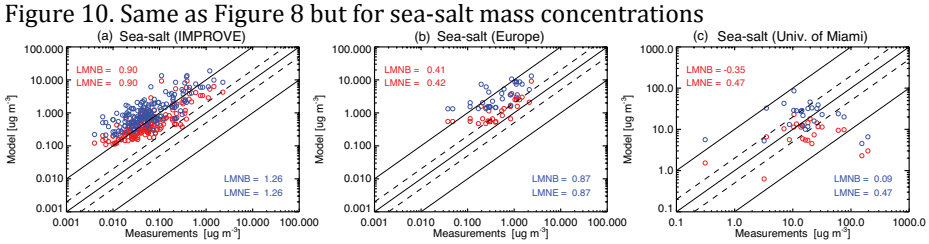

Figure 11. Same as Figure 9 but for sea-salt mass concentrations

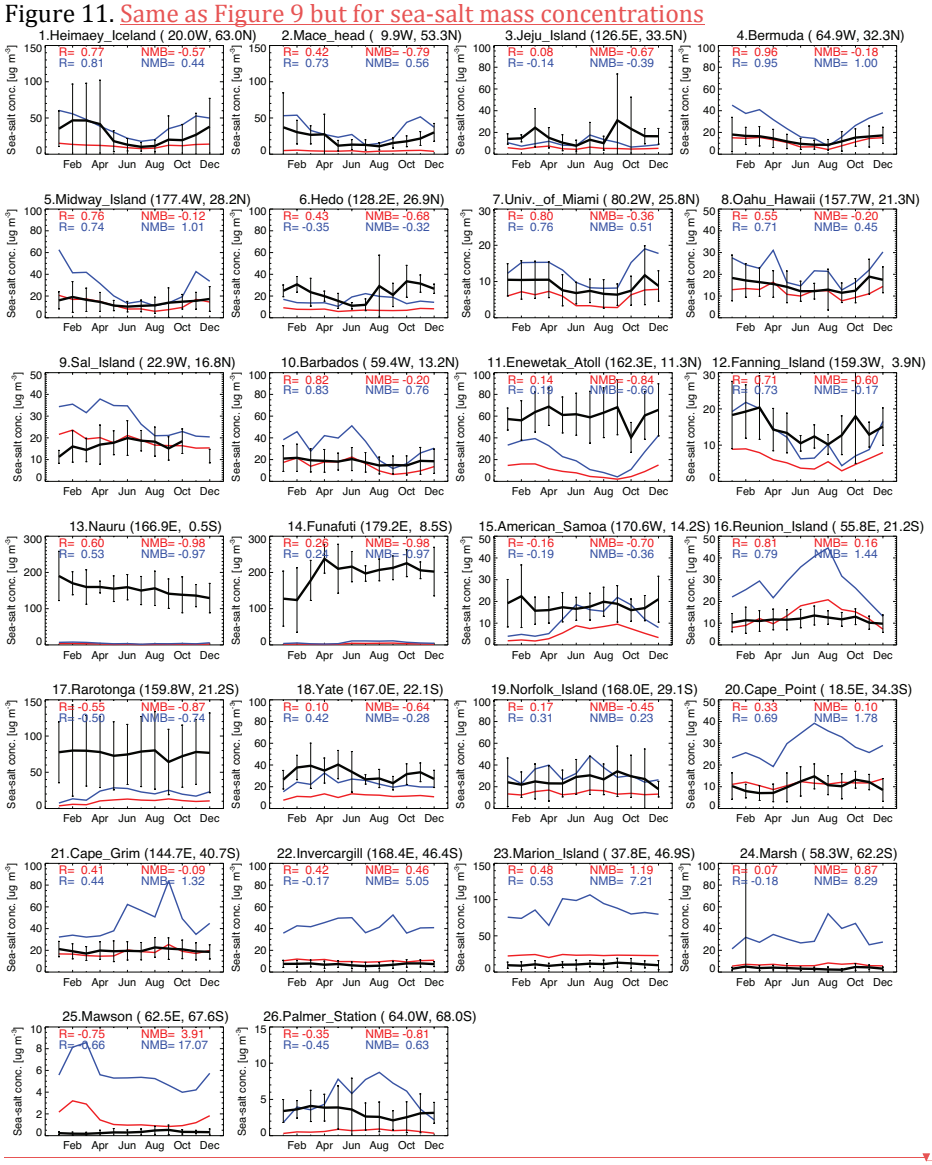

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Figure 12. Same as Figure 8 but for dust mass concentrations

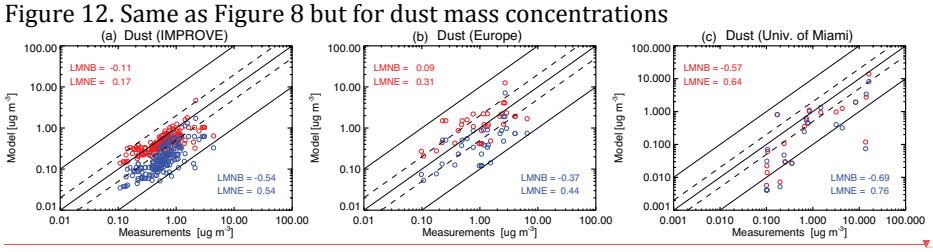

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Figure 13. Same as Figure 9 but for dust mass concentrations.

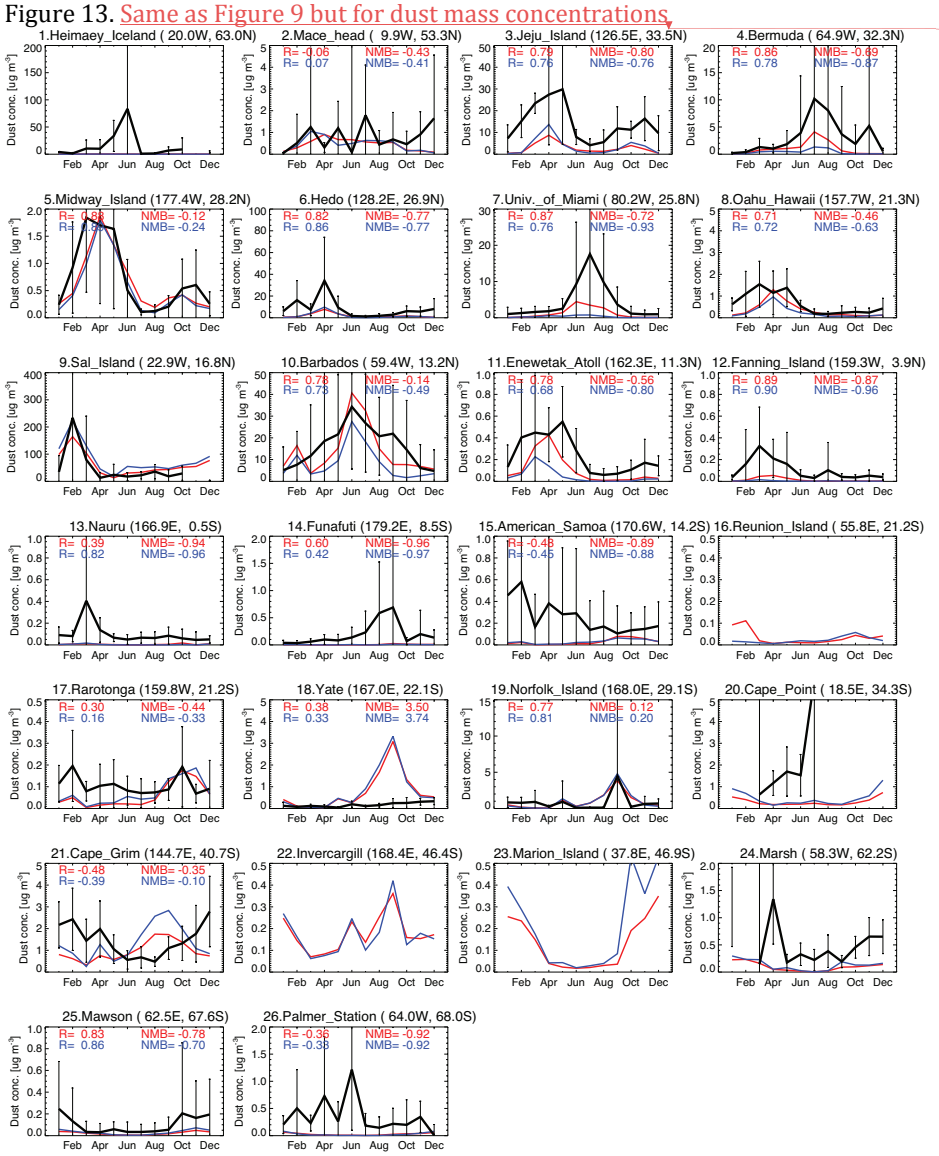

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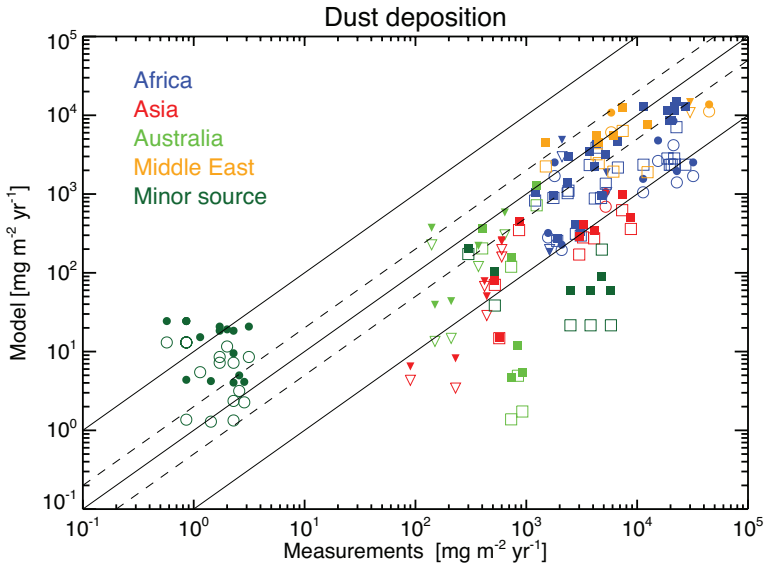
Figure 14. Scatter plot of annual-average dust deposition fluxes [mg m^{-2} yr^{-1}] simulated and observations obtained from Ginoux et al. (2001), Tegen et al. (2002) and Tables S2 in Mahowald et al. (2009). Open symbols are for ModelE2-TOMAS and the filled symbol for ModelE2-OMA. LMNB and LMNE are presented below the plot: the first values are for ModelE2-TOMAS.



Mahowald et al.[2009] - o	Ginoux et al.[2001] - v	Tegen et al.[2002] - sq
LMNB = -0.04	LMNB = -0.77	LMNB = -0.90
LMNE = 0.68	LMNE = 0.81	LMNE = 0.91
LMNB = 0.24	LMNB = -0.54	LMNB = -0.61
LMNE = 0.82	LMNE = 0.64	LMNE = 0.65

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Figure 15. Scatter plots of annually averaged surface EC and OM mass concentrations [μg m^{-3}] simulated (red for ModelE2-TOMAS and blue for ModelE2-OMA) and observations from the IMPROVE network (a and b) and the European sites from Putaud et al. (2010) (c and d).

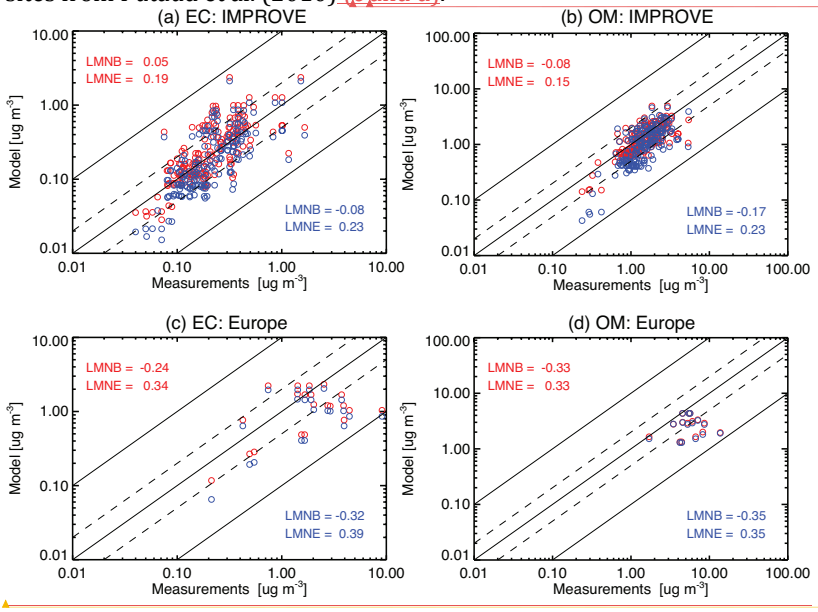

Figure 16. Scatter plots of annually averaged surface PM2.5 concentration [μg m^{-3}] simulated and observations compiled by the Global Burden of Disease (GBD) Study. ModelE2-TOMAS is shown in (a) and ModelE2-OMA in (b).

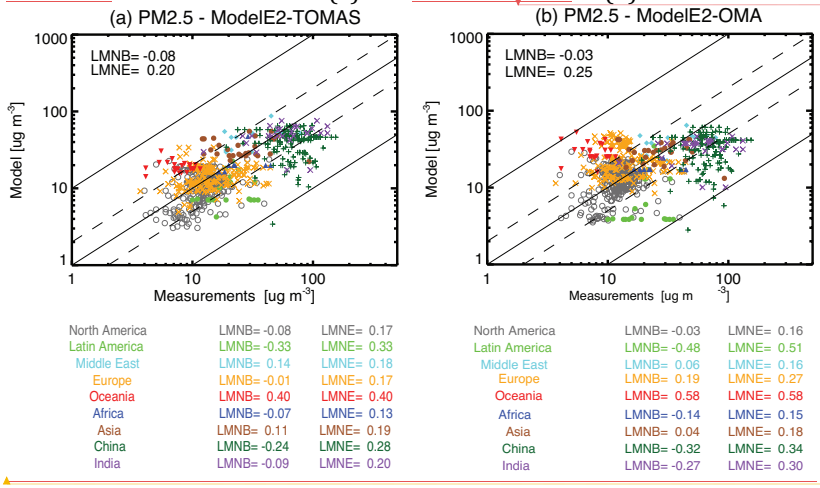

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Figure 17. Global distributions of the annual-mean AOD from (a) ModelE2-TOMAS, (b) ModelE2-OMA, (c) MISR, and (d) MODIS. See Section 5.4 for the details of the MISR and MODIS AOD information.

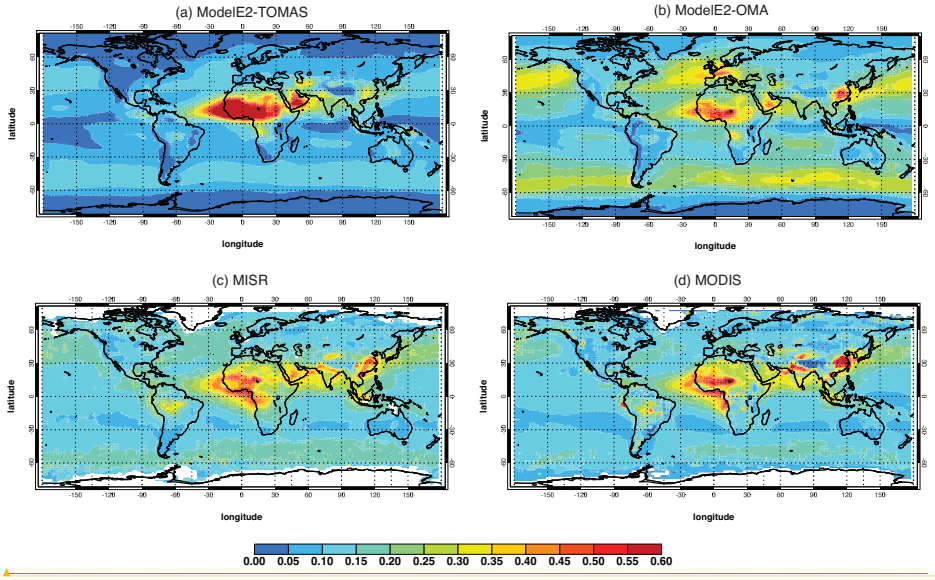

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Figure 18. Comparisons of monthly averaged model AOD (red for ModelE2-TOMAS and blue for ModelE2-OMA) and AERONET AOD (black solid line). Correlation (R) and normalized mean bias (nmb) are provided only when the observation is available for 12 months.

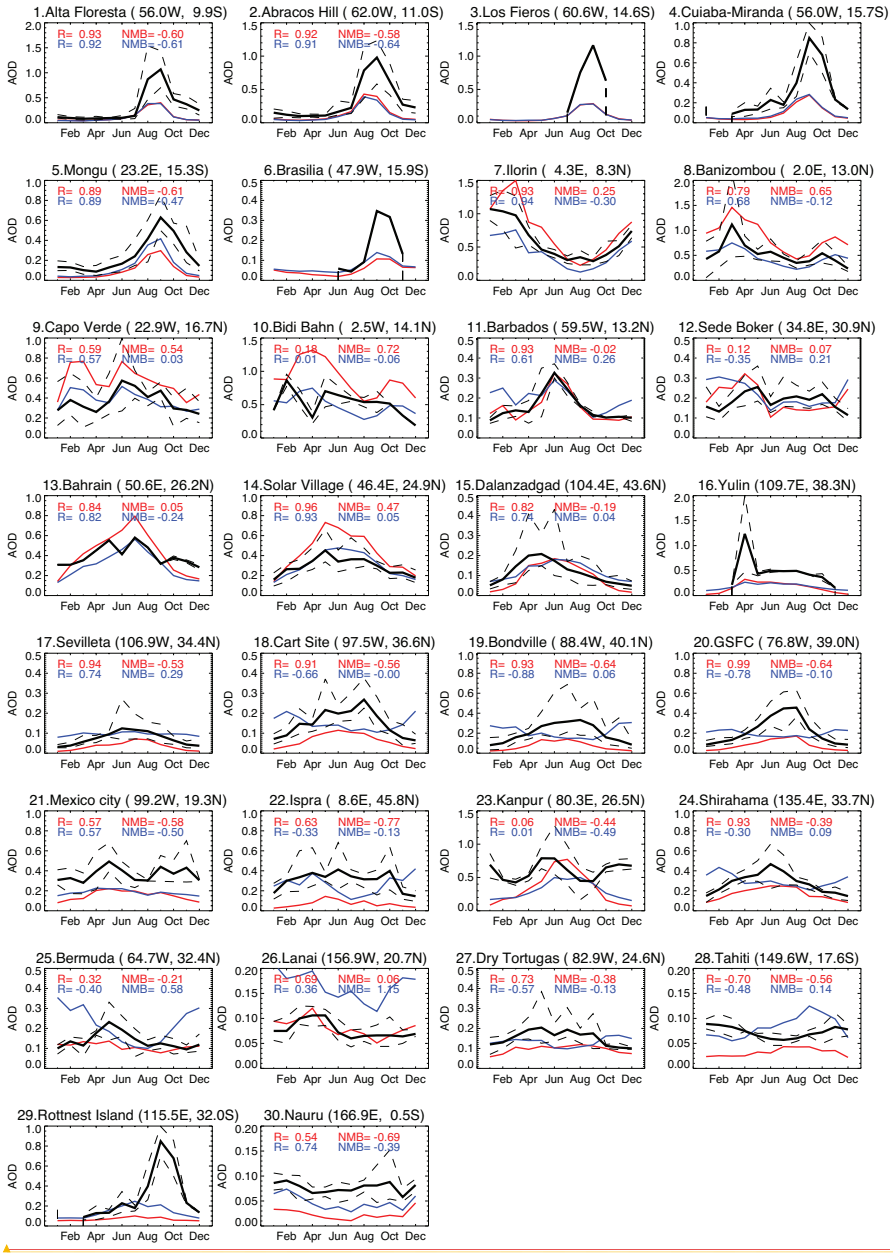

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Figure 19. Annually averaged CN3 ($D_p \geq 3$ nm), CN10 ($D_p \geq 10$ nm), CN100 ($D_p \geq 100$ nm) concentrations in the lowermost layer for the BASE run (a to c), the LowNUC run (d to f), and the NoNUC run (g to i) of ModelE2-TOMAS. Units are cm^{-3}.

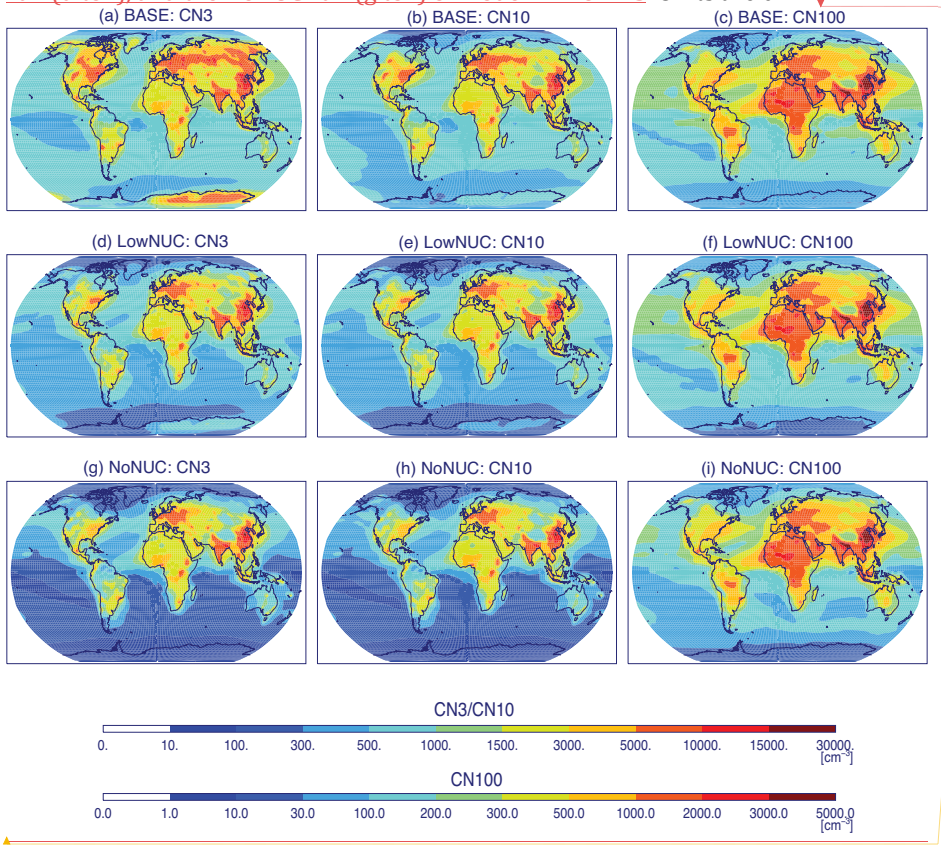

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Figure 20. Annually and zonally averaged CN3 (Dp ≥ 3 nm), CN10(Dp ≥ 10 nm), and CN100(Dp ≥ 100 nm) concentrations for the BASE run (a to c), the LowNUC run (d to f), and the NoNUC run (g to i) of ModelE2-TOMAS. Units are cm^{-3}.

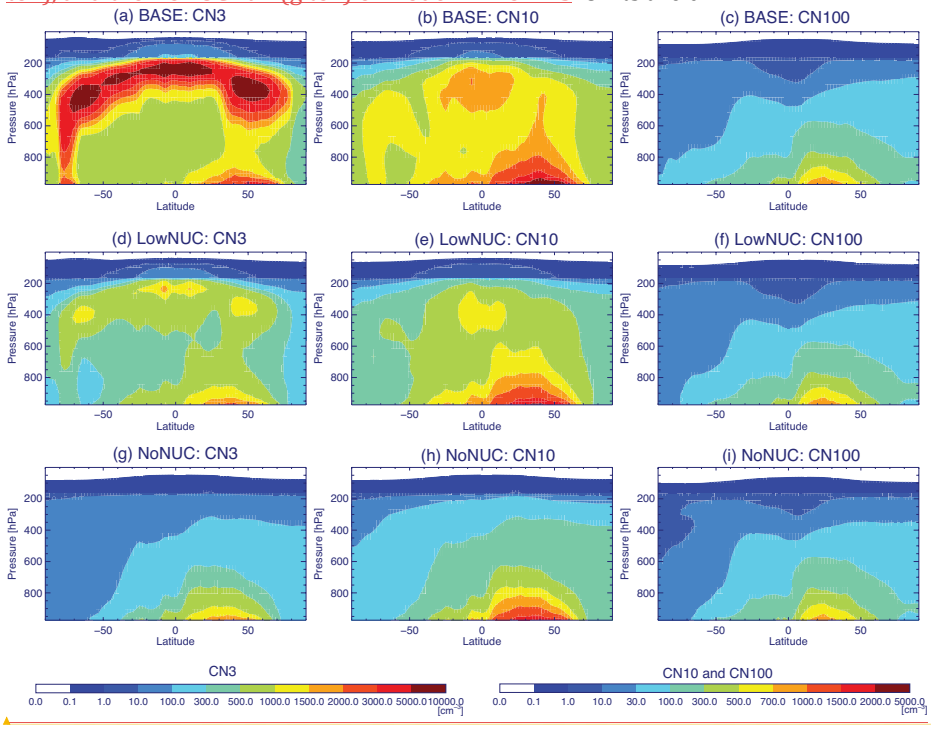

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Figure 21. Scatter plot of simulated annual-mean aerosol number concentrations in comparison with a dataset of surface number concentrations measurements at 36 sites around the world compiled by Spracklen et al. (2010). The top horizontal bar represents the BASE results, and the middle bar the LowNUC results and the lower bar the NoNUC results. Red color is for free troposphere (FT); blue for marine boundary layer (MBL); green for continental boundary layer (CBL).

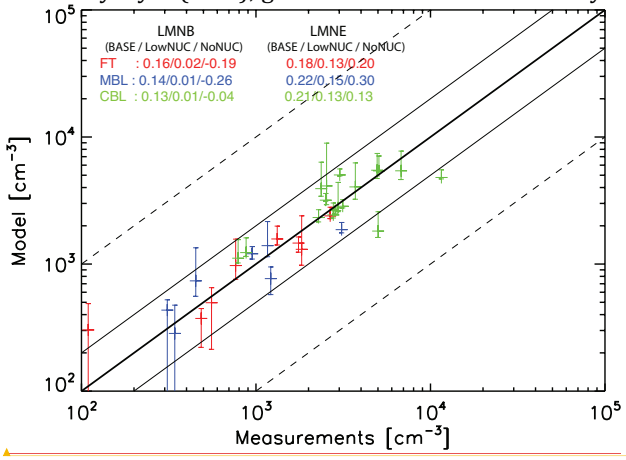

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Figure 22. Annual cycle of aerosol number concentrations [cm^{-3} at ambient conditions] at 32 sites. The observations are shown in black, and three model results are presented: red for BASE, blue for LowNUC, and green for NoNUC. The free tropospheric (FT) sites are from 1 to 8; the marine boundary layer (MBL) sites, 9 to 15; the continental boundary layer (CBL) sites 16 to 32. Correlation (R) and normalized mean bias (NMB) are provided only when the observation is available for 12 months.

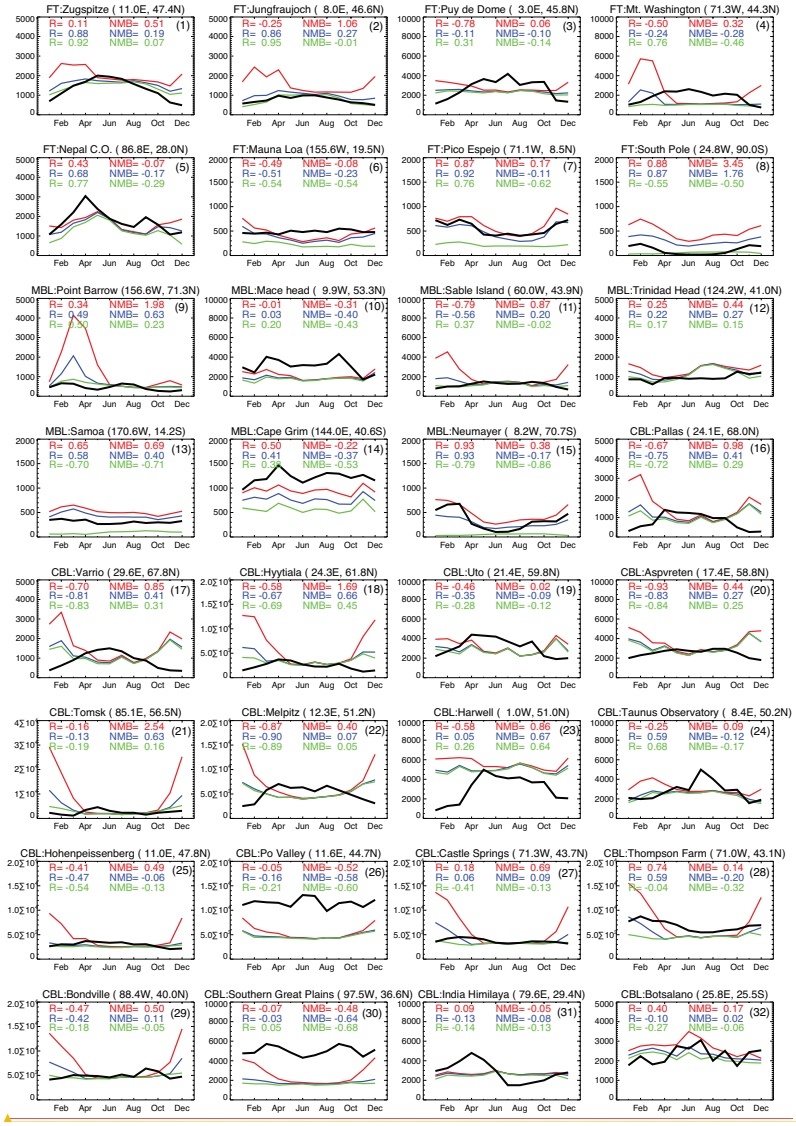

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Figure 23. Number size distributions from European sites during winter (DJF; a to f) and summer (JJA; g to l) that are obtained from log-normal 3-mode fits during morning (black solid), afternoon (black dotted), and night (black dashed). The model results are the seasonal mean, shown in red lines for the BASE run, blue lines for the LowNUC run, and green lines for the NoNUC run.

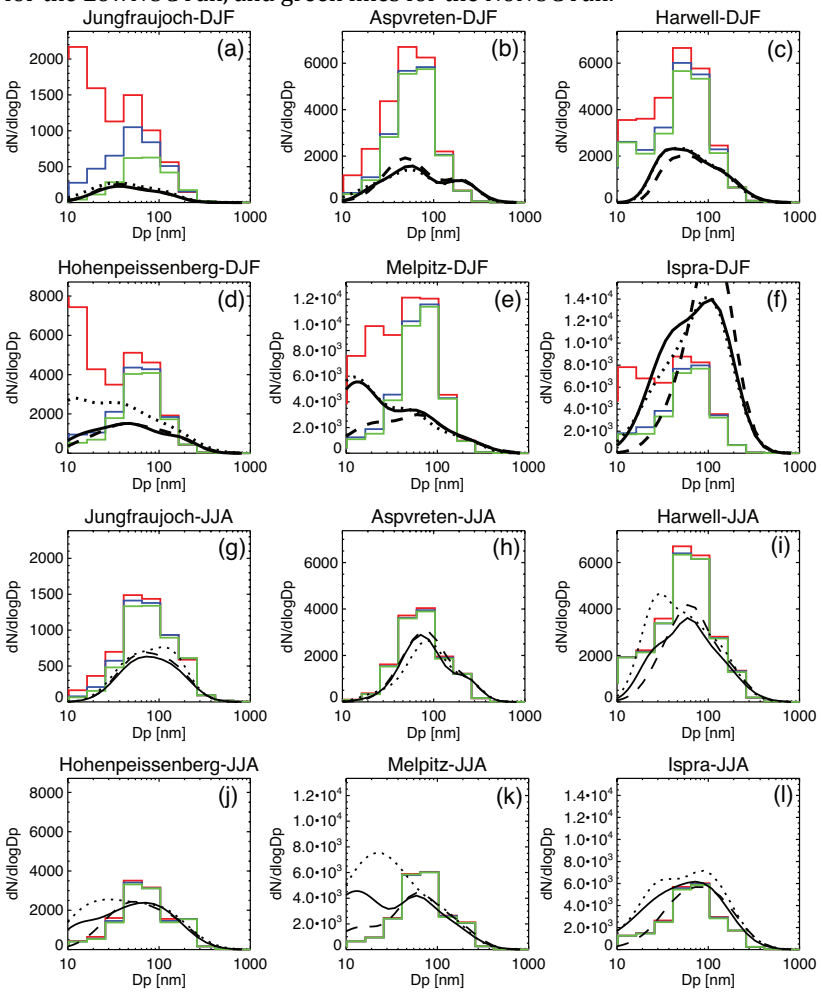

Figure 24. Vertical profiles of aerosol number concentrations from the observations (black lines) and ModelE2-TOMAS (red lines for the BASE run, blue lines for the LowNUC run, and green lines for the NoNUC run). The CN5, CN15, CN120 concentrations (ambient conditions) from LACE campaign (Petzold et al., 2002, in north-east Germany) are shown in a to c, respectively. The observed CN3 concentrations (STP conditions: 1atm, 273 K) over the Pacific Ocean, which are averaged into the 3 latitude bands (70 S to 20S, 20S to 20N, and 20N to 70N; Clarke and Kapustin, 2002), are shown in d to f. The dashed lines show the standard deviation for the observations and the min/max monthly mean for the model (only in d to f).

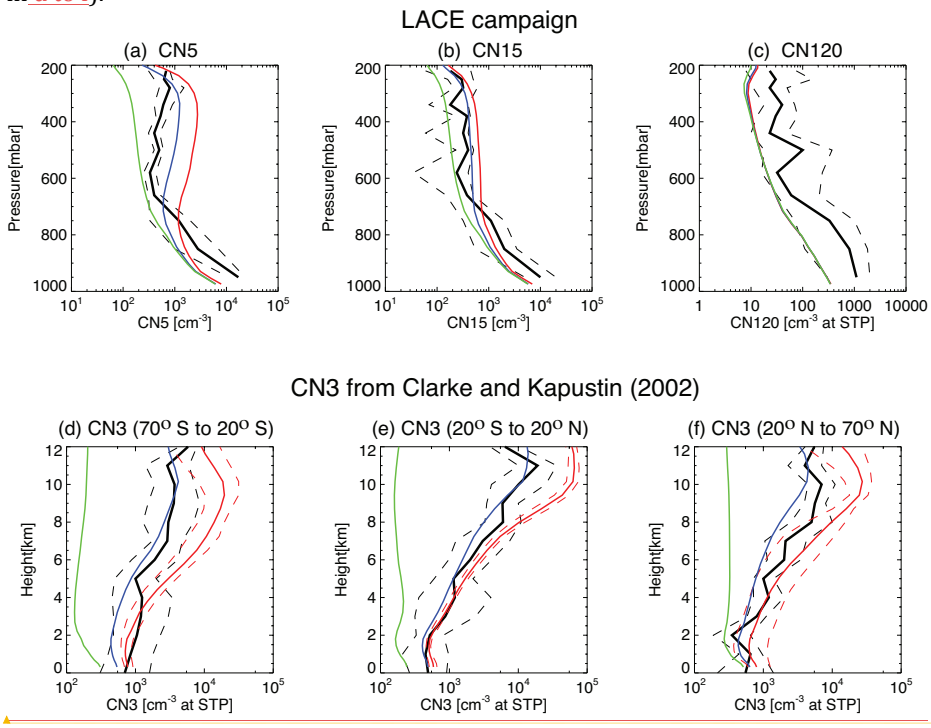

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Figure 25. Simulated number concentrations in (a) Aitken mode (10 nm ≤ Dp < 100 nm) and (b) accumulation mode (100 nm ≤ Dp < 1 μm) compared to the observations ("x" symbol with error bar representing minimum and maximum observed concentrations) that were compiled and aggregated into 15-degree latitude ranges (Heintzenberg et al. 2002). The model is also averaged to the 15-degree grid and is shown in red lines for the BASE run, blue lines for the LowNUC run, and green lines for the NoNUC run.

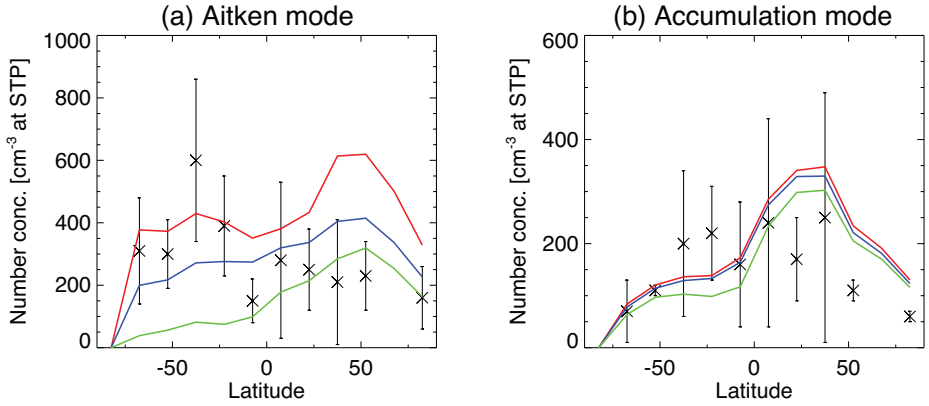

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Figure 26. Annually and zonally averaged aerosol size distributions in the marine boundary layer. Observations are from Heintzenberg et al. (2000) and were compiled and aggregated into a 15° x 15° grid. The model is also averaged to the 15-degree grid and is shown as red lines for the BASE run, blue lines for the LowNUC run, and green lines for the NoNUC run.

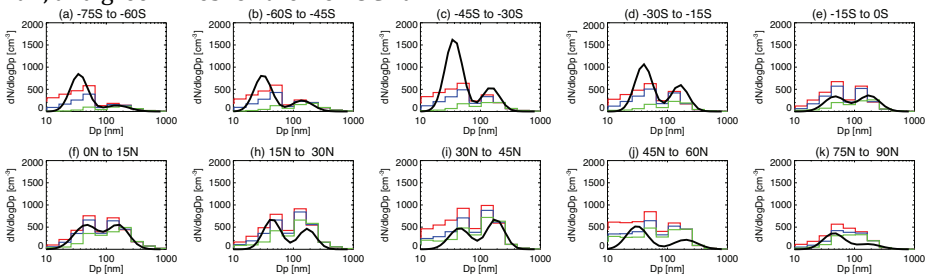


Figure 27. Scatter plot of simulated CCN concentrations in comparison with a dataset of CCN measurements compiled by Spracklen et al. (2011). Data is classified into two categories: CCN in the MBL (in black) and CCN in the CBL (in red). The upper error bar is for the BASE run, the middle symbol for the LowNUC run, and the lower error bar for the NoNUC run. Large and filled symbols are for measurement duration longer than 10 days, and small and open symbols for less than 10 days. Circle symbols are for supersaturations (*s*) less than 0.2%; upward triangles for *s* greater than 0.2% and less than 0.8%; downward triangle for *s* greater than 0.8%.

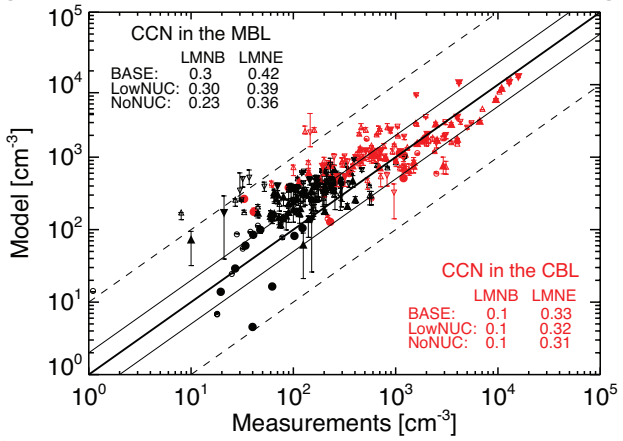
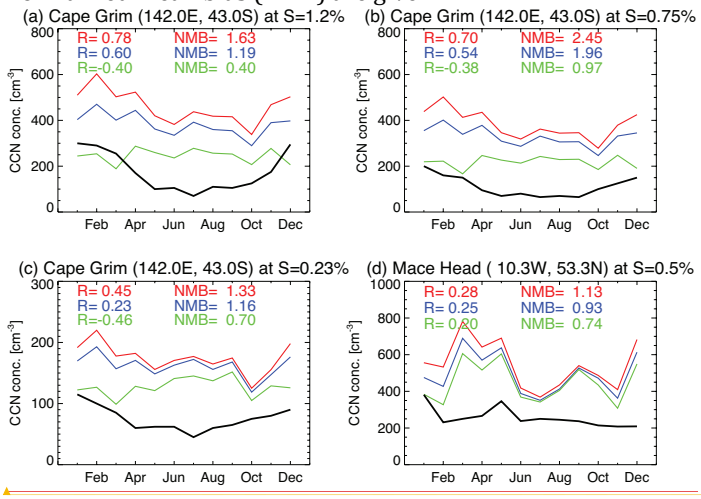


Figure 28. Annual cycle of CCN concentrations at Cape Grim (supersaturations (*s*) of 1.2% in a, 0.75% in b, and 0.23% in c) and Mace Head (*s* of 0.5% in d). The measured CCN concentrations are shown in black, and the simulated CCN in red for the BASE run, blue for the LowNUC run, and green for the NoNUC run. Correlation (R) and normalized mean bias (NMB) are given.



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