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.