Author's response to interactive comments by anonymous referee #2:

The authors would like to thank the referee for very constructive review comments. Each of the review comments are repeated below in bold font, followed by our replies.

This manuscript describes the latest version of the OsloAero module used for modelling atmospheric aerosol in the aerosol-climate model CAM5.3-Oslo and earth system model NorESM1.2. OsloAero has a quite different formulation to most aerosol schemes, using a set of "background aerosol" tracers, whose prescribed size distributions and composition are then modified according to a second set of tracers which are tagged according to aerosol production processes. The resulting variations in size and composition, which are not restricted to e.g. a log-normal form, are then looked up in tables computed offline to determine the relevant optical and other properties of these mixtures. This is a novel approach to aerosol modelling which the authors have developed over a number of previous papers, adding distinct capabilities compared to the more widespread modal and sectional schemes. The present manuscript describes new developments for nucleation and secondary organic aerosol, as well as new online emission schemes for sea-salt, mineral dust and oceanic DMS and organics. There has also been an effort to ensure better consistency of parameters between the different components of the scheme, which is always welcome from a physical point of view. These are important additions, both in terms of updating this modelling framework with the latest understanding of physical processes, and for tighter coupling in the earth system context. A good basic set of evaluation plots and metrics are included, though the overall results compared to the previous CAM4-Oslo are somewhat mixed, with some biases and errors improved but others becoming larger; nevertheless this is a well-presented paper documenting a significant advance in the physical and chemical capabilities of the model and I would recommend it for publication in GMD subject to the minor comments below:

p.3, line 28 – p.4, line 10 This section is quite programmatic in terms of discussing the evolution of the projects to which OsloAero is related; I would consider whether all of this is relevant to a model description paper.

The first part of this text, p.3, line 28 – p.4, line 2, explains how CAM5.3-Oslo (or NorESM1.2) relates to CAM5.3 (CESM1.2), as well as to the online and offline parts of the aerosol schemes, OsloAero5.3 and AeroTab5.3, which we think is an important part of the description of the model, and being of high relevance also for potential users of the model. The remaining part links the present model version, already being used as a pre-CMIP6 version (in the CRESCENDO project, not mentioned in the text since that at least may be considered too project centered) of the final model versions planned to be used in CMIP6. We think this is of relevance for the reader, especially when seen together with the subsequent lines (p.4, linea 9 - 11) which in a similar way mention the participation of NorESM1 in CMIP5. We advocate for retaining this text as it is, for the sake of completeness. We will instead remove the last sentence of this paragraph (on lines 11-13) about NorESM1 participation in PDRMIP, since this is not imortant for the history or description of CAM5.3-Oslo. Two papers in the reference list will therefore also be removed: Samset et al. (2016) and Myhre et al. (2017).

p.4, line 27 "same method of aerosol activation as Liu et al. (2012)": is this the primary reference for the parameterisation in use, or does it describe a particular implementation of a well-known parameterisation (e.g. Abdul-Razzak and Ghan, 2000) which should also be cited here.

This was orginally meant to be written in more general terms, not only to concern the activation treatment, which indeed follows Abdul-Razzak and Ghan (2000) (referred to later, on the last line of p. 12). We suggest to rewrite the sentence starting at line 27 as follows:

«OsloAero5.3, as it is implemented in CAM5.3, applies the same method of aerosol activation (Abdul-Razzak and Ghan, 2000) and transport of and transition between aerosols in interstitial and cloud-phase as in Liu et al. (2012), with the simplifications proposed by Ghan and Easter (2006) that cloud-borne aerosols are not advected, except by vertical turbulent mixing.»

p.6, line 4 It's not clear either why a portion of the modifying tracer is redistributed to the background ones, or the basis on which the amount redistributed is calculated; some explanation here is needed.

The confusion seems to be due to a misunderstanding. What we are trying to explain is that the mass from the modifying tracers are distributed onto the modes (or mixtures) which initially only are constituted by the background aerosol tracers. Hence, no redistribution from modifying to background tracers is being performed at any point. While the tracers operate in «chemistry space», the mixtures (containg mass from various tracers) are treated separately in «physics space», if we can put it that way. How this is done quantitatively is outlined in the subsequent paragraph. Aiming to clarify this point, we suggest to rewrite this sentence (from line 4) to:

«OsloAero calculates how much of each "modifying" tracer should be distributed onto each of the background modes (thus forming mixtures of mass from the various tracers) within a time-step.».

p.6, lines 12–13 In what way are these sized "augmented to take into account atmospheric growth"? This is a rather vague description.

The sizes are increased (for mixtures 1-4 in Table 2) to typical sizes in the lower troposphere, which are rough estimates (prescribed values) originally from OsloAero4.0 (Kirkevåg et al., 2013), but with minor adjustments based on table look-ups of median radii (for the log-normal fits to modified size-distributions) from earlier CAM5.3-Oslo model simulations. To make this more clear, we suggest to add a second row with these modified sizes in Table 2, and rewrite the sentence on lines 11-16 (moving some of the detailed size info on lines 15 and 16 to the Table instead) to:

«First, during atmospheric transport the background tracers are assigned typical tropospheric dry sizes (i.e., the sizes at the point of emission, augmented to take into account atmospheric growth for the finest particles, mixtures no. 1 - 4 in Table 2). The size-modifying tracers are also assigned prescribed sizes (Table 2). Calculation of sizes (for dry deposition) after hygroscopic growth is as in OsloAero4.0 (K13).»

The «augmented» dry sizes (as NMR) for mixture 1, 2 and 4, to be listed in Table 2, are 0.025, 0.025 and 0.06 μm , respectively.

In addition to the above changes in the main text, we modify the fourth sentence in the caption of Table 2 slightly to

«The initial number median dry radius (NMR) and standard deviation (SIGMA) of each background mode are listed in the second and third column.», and follow up with:

«Also listed (with numbers in brackets) are the prescribed dry NMR values assumed during transport (including atmospheric growth), for the finest particle mixtures (no. 1, 2 and 4). For other mixtures, the dry sizes of transported tracers are assumed to be identical to the initial sizes.»

Similarly, we add at the end of the table caption: «Assumed dry size parameters for the size-modifying tracers during transport are NMR = 0.04 μ m and SIGMA = 1.8 for SO4_A1, and NMR = 0.1 μ m and SIGMA = 1.59 for SO4_A2, SO4_AC, OM_AC, BC_AC and SOA_A1."

p.6, lines 13–16 A little more explanation of what is meant by lumping the size-modifying tracers would be good, as well as the meaning of the modal size parameters of these modifying tracers, if these are not particles in their own right, but act to adjust the sizes of the background modes. (This may be explained further in earlier papers e.g. that labelled K13, but should ideally be self-explanatory in this manuscript.)

The «lumping of size-modifying tracers (with respect to transport and deposition)» alludes to the fact that, e.g., sulfate condensate has only one tracer even though it is used in (distributed onto) several background modes to form mixtures, as shown in Table 2. But the different size-modifying tracers are here not lumped together to even fewer tracers, so this expression is a bit misleading. We have therefore chosen to change the text to that above, in the reply to the previous remark.

The modal size parameters were already defined in the same sentence, as «number median radius (NMR) and standard deviation (SIGMA)», but this has now been moved to the caption of Table 2 and rephrased (to size parameters). These tracers, as transported tracers in the model, behave as if they were «particles in their own right». Only in the microphysics part of the model (through use of look-up tables) do they adjust the sizes of the backround modes. To make this more clear in the text, we suggest to add the following sentence at line 15, just before the sentence starting with «Secondly»:

«In the parts of OsloAero5.3 which deal with aerosol chemistry, transport and dry deposition (the aerosol life-cycle scheme), both the background tracers and size-modifying tracers are treated as if they were particles.»

p.27–29 I assume "lost from the model" means that monoterpene and isoprene do not exist as tracers in the model, but only as "transient" species near the surface within a given model time step?

Both Isoprene and Monoterpene exist as gas tracers in the model, see e.g. Fig. 1. What we mean here on p. 7, line 27-29, is that the fractions of these gases which do not form SOA gas (85% or 95% in R1 – R5), are no longer tracked and taken into account, as they are assumed not to produce SOA aerosols. This (that they are not just transient species) can also be seen from Karset et al. (2018), Table 4., where their lifetimes for present-day conditions are estimated to 3.2 and 2.6 days, respectively. To clarify this in the text, we suggest to rewrite this sentence to

«The fractions of monoterpene and isoprene which do not react to form SOA gas in (R1) - (R6) are not taken into account, assuming that they form other gas or aerosol products which we do not track in the model.»

To make the list of aerosol and gas tracers complete and easy to find in the manuscript, we will also list of the 8 transported gas tracers at the end of the caption of Table 1:

«The aerosol precursor and oxidant gas tracers transported by the model are: SO_2 , H_2SO_4 , DMS, isoprene, monoterpene, SOAG_LV, SOAG_SV, and H_2O_2 .»

p.11, line 19 This should be "... scheme is different from that..." or "... schemes are different from those..." for singular/plural consistency.

Thank you, we will correct this to «...scheme is different from that...»

p.14, line 8 It would be good to recap the emission sources here, even if they haven't changed from the previous paper.

The exact meaning of this comment is a bit unclear, but we suggest to modify and add to this paragraph (on lines 6-9) as follows:

«For aerosol and precursors not mentioned above, as in K13, the emissions are taken from the IPCC AR5/CMIP6 (Lamarque et al., 2010) for the years 2000 (for simplicity called present day, PD), and 1850 (preindustrial, PI) conditions. The emissions and their vertical distribution are essentially the same as those used by Liu et al. (2012): the IPCC AR5 emission data set includes anthropogenic emissions for primary aerosol species OC and BC, as well as the precursor gas SO₂. We assume that 2.5% of the sulfur emissions are emitted directly as primary sulfate aerosols, and the rest as SO₂. Anthropogenic emissions are defined as originating from industrial, energy, transportation, domestic and agriculture activity sectors.»

p.15, line 8 What is the height of the model top with 30 levels?

We may here add the following extra nformation (as a reply also to referee#1):

«In hybrid sigma pressure coordinates, the uppermost eta level mid (or top of the level) value is 3.64 (2.26), and for the lowermost level it is 992.56 (985.11). The number of layers below approximately 1km and 2km height a.s.l. are 5 and 8, respectively.»

p.15, line 9 Clarify that "microphysical schemes" here refers to cloud and precipitation rather than aerosol, if that is the case. Also, a brief description of the nature of these schemes (single/double-moment, what is prognostic etc.) would be welcome to provide context for aerosol-cloud interactions in this model.

Yes, thank you, we here mean cloud and precipitation microphysical schemes. To add a brief description of the two schemes and the main difference between them, we suggest to rewrite and add to the sentence on lines 9-11 as follows:

«CAM5.3, and therefore also CAM5.3-Oslo, has two choices for stratiform microphysical cloud schemes: MG1.0 (Morrison and Gettelman, 2008) and MG1.5 (Gettelman and Morrison, 2015). Both are double-moment (i.e., mass and number predicting) bulk cloud microphysics schemes with prognostic cloud droplet and cloud ice mass mixing ratios and number concentrations. MG1.5 is an update of the original formulation MG1, where the location for updating prognostic droplet number mixing ratios with the tendency from droplet activation has been moved to the beginning of the scheme. We have in this study used MG1.5.»

p.15, line 24 A reference for the nudging technique would be welcome (e.g. Jeuken et al., 1996, 10.1029/96JD01218 or equivalents in other models).

The references for the nudging technique applied in CAM5 are Koopermann et al. (2012) and Zhang et al. (2014). These papers are already mentioned a bit later in Sect. 3, but we may add the references also at the end of the sentence at lines 24-26, p. 15:

«... using a relaxation time scale of six hours (Koopermann et al., 2012; Zhang et al., 2014).»

p.16, line 18 Is there a reason why CAM4-Oslo couldn't be tested at the higher resolution, or CAM5.3-Oslo at the lower one, in order to assess the impact of the resolution change separately from the actual model updates?

This was not originally done with the present model version and in this study, but two unnudged AMIP simulations (PI and PD) with 2° resolution have later been run to help answer this review comment. The main focus of this study is to document the changes in aerosol treatment from CAM4-Oslo to CAM5.3-Oslo, together with the comparison of their performance, while the effort to understand the differences in simulated results are more of a spin-off from the main study (not mentioned in the abstract). We will therefore not include the results of the 2° simulations (hereafter referred to as AMIP2degPD and AMIP2degPI) in the tables and figures of the manuscript, which would mean an immense amount of extra work, especially when it comes to the AeroCom intercomparison with other models and with maesurements (which are regularly updated on the aerocom.met.no server). We instead add comments on the effect of differences due to resolution some places in the manuscript where differences between the two model versions are being discussed (the basis diagnostics for this may be found here, together with diagnostics from the other simulations: http://ns2345k.web.sigma2.no/nudged_NorESM_c12/7310AMIP20002nAMIP_PI_wPDoxi_vs _AMIP2deg_PDnPI/ModIvsModII.htm):

At the end of the paragraph on p. 17, line 2:

«An additional test simulation with CAM5.3-Oslo with AMIP PD set-up and 2° resolution show that the effect of increased resolution (to 1°) on DMS emissions and lifetime alone is only about 5% and 0.2%, respectively (not shown).»

At the end of the paragraph on p. 17, line 21:

«The additional 2° test simulation with CAM5.3-Oslo reveals that the effect of increased resolution on vertical profiles is very small compared to the differences between the two model versions, for all species (not shown).»

At the end of p. 17, line 32:

«The additional 2° test simulation reveals that the effect of increased resolution on the lifetime of BC is only about 0.3% (not shown).»

At the end of the paragraph on p. 18 line 11:

«The additional 2° test simulation (note that this by default set-up has a slightly different cloud tuning) reveals that the effect of increased resolution on LWP and on total (low) cloud cover is small compared to the differences between the two model versions, only about 1% and -1% (-3%), respectively (not shown).»

At the end of the paragraph on p. 18, line 17:

«The effect of increased resolution from 2° to 1° is here found to be 11% for the emissions (due to stronger winds), 9% for the burden, and only -2% for the lifetime (not shown).»

At the end of the paragraph on p. 18, line 24:

«The additional 2° test simulation reveals that the effect of increased resolution on the OM lifetime is only about 1% (not shown).»

At the end of p. 18, line 32: «The additional 2° test simulation reveals that the effect of increased resolution on the SO₂ lifetime is only about -0.4% (not shown).»

At the end of the paragraph on p. 19, line 12: «The additional 2° test simulation reveals that the effect of increased resolution on the mineral dust lifetime is only about 2% (not shown).»

Since the above differeces due to increased resolution alone are so small, the impacts on effective radiative forcing are also small, compared to other changes from CAM4-Oslo to CAM5.3-Oslo. E.g., the SW cloud radiative forcing at TOA from AMIP2degPD minus AMIP2degPI is estimated to be -1.47 W/m2, compared to -1.45 W/m2 in AMIP_PD – AMIP_PI (Fig. 10). We therefore believe that the above additions to the text are sufficient, especially given that the weight on the parts of the mansucript where understanding differences in model results between the two models to some degree are discussed, is small (not mentioned in the abstract).

p.16, line 25 If the DMS burden is doubled compared to the earlier version, are there some observations which could be cited here to indicate which is more realistic, or is the uncertainty even larger than this?

The text should here have read «emissions», not «column burdens», and the background for these changes (with references) are discussed directly below. The text will here be corrected to:

«The result of the change in DMS emission parameterization described in Sect. 2.4 is an almost doubled DMS emission (34-35 Tg S yr⁻¹) compared to the 18.1 Tg S yr⁻¹ found in K13,...».

The increase in column burden (for which we do not have any obersvations) is much smaller (see Table 1), going from 0.12 Tg S in CAM4-Oslo to 0.14 Tg S in CAM5.3-Oslo. For completeness, we add at the end of the paragraph, on line 3, p. 17 (after the additional text concerning model resolution), as well as including the new reference under the «References» section:

«Note also that the increase in column burden from CAM4-Oslo to CAM5.3-Oslo is much smaller than the increase in emissions (see Table 1), going from 0.12 to 0.14 Tg S. These both lie well within the range of estimates (0.015 - 0.17 Tg S) from other model studies reported by Liu et al. (2007), see their Table 1.»

p.17, line 29–30 The attribution of upper-tropospheric excess to the treatment of convective processes based on comparisons to HIPPO was also made by Kipling et al. (2013, 10.5194/acp-13-59690-2013 in the context of another model.

Thank you. We here suggest to change the text as follows, and include the new reference in «References»:

«This is probably related to the way aerosols are transported and scavenged in deep convective clouds in the model (see e.g. Kipling et al., 2013; 2016).»

The latter reference is already used elsewhere in the text.

p.27, lines 8–12 The rationale for using the grid-box-mean RH and weighting by clearsky fraction, rather than the more common approach is not clear.

Different models make use of different assumptions about what can be considered representative for optical properties from observations or remotely retrievals under clear-sky conditions (such as those from AERONET), and it is not obvious to us which ones are the most accurate. We would argue that the assumed RH for hygroscopic growth is not the only relevant parameter, since aerosol optical properties also depend on the general abundance of aerosol, its size distribution and chemical composition, which again have links to the cloud cover (and precipitation). Although using the clear-sky RH fraction and not taking into account the actual cloud cover is the most common approach, it is not the only approach applied. The question of how AeroCom modellers calculate clear-sky optical properties was taken up by the AeroCom community during phase II of the project, which resulted in an attempt of documentation via a wiki page: https://wiki.met.no/aerocom/optical properties. Among the 10 AeroCom model versions (in addition to our own) which are both appearing in the wiki and in Tables 6-8 in the present study (GISS-MATRIX, GISS-modelE, GOCART-v4Ed, HADGEM2-ES, INCA, MPIHAM_V2_KZ, OsloCTM-v2, SPRINTARS-v384, and TM5.V3), only one estimates clear-sky in a similar way as we do, i.e. by using weights based on total 2D cloud cover but all-sky RH for hygroscopic growth of the aerosol: SPRINTARS_v384 averages only AOD for time steps and grid points with CLDTOT<0.2, using all-sky RH in these almost cloudfree cases. GISS-MATRIX, GMI and GOCART_v4E average all-sky AOD (no cloud cover weighting), also based on growth factors from all-sky RH. GISS-MATRIX also provide clear-sky optics, but this has not been used at aerocom.met.no. Although not very clear from the wiki, the rest of the models seem to estimate AOD (tacitly meant to be a clear-sky parameter) for all cloud conditions, but based on hygroscopic particle growth using the clear-sky RH value. As a short summary of this (also as reply to a related comment by referee #1), we suggest to rewrite parts of the caption of Table 6 (lines 8-11) from

«Optics diagnostics listed for the AP2 and AP3 models are mostly clear-sky values, in the sense that the clear-sky humidity of the grid cell is used for calculating the hygroscopic swelling. CAM4-Oslo and CAM5.3-Oslo compute all-sky optical properties using the average humidity of the grid cell. Clear-sky (CS) properties are represented by a cloud fraction weighted average of the all-sky properties.»

to

«Optics diagnostics listed for most of the AP2 and AP3 models (exact number is not available) are clear-sky values, in the sense that the clear-sky humidity of the grid cell is used for calculating hygroscopic swelling of the aerosol. Information about this for 11 of the AP2 models included here, plus some others, may be found at

https://wiki.met.no/aerocom/optical_properties. CAM4-Oslo and CAM5.3-Oslo compute allsky optical properties using the average humidity (RH) of the grid cell. Clear-sky (CS) properties are instead represented by a 2D cloud-free fraction weighted average of the allsky properties. Only a few other AeroCom models follow a similar clear-sky optics definition, and the optics data submitted to AeroCom for a few of the models are all-sky values both in terms of cloud conditions and RH for hygroscopic growth.»

p.37, line 30 NUDGE_PI appears twice; one should be NUDGE_PD.

Thank you, this will be corrected.

p.38, line 9 Who should be contacted to obtain a user agreement for access to the code and data?

The contact person has traditionally been Øyvind Seland, but we have a more general mailing list which should be used: <u>noresm-ncc@met.no</u>. Based also on another interactive comment, we suggest to rewrite the second line of the Code and data availability paragraph to:

«The source code for CAM5.3-Oslo is part of a restricted NorESM2 pre-release and stored within the private github NorESM repository

(<u>https://github.com/metno/noresm/tree/NorESM1.2-v1.0.0</u>). Access to the code and simulation output data produced in this study can be obtained upon reasonable request to <u>noresm-ncc@met.no</u> and requires entering a NorESM Climate modeling Consortium (NCC) user agreement.»

Tables Many of the tables contain a large number of numerical values, either mass budget terms or statistics. While the actual numbers may be useful for reference, summarising these in charts would probably be much easier for the reader to understand at a glance.

Since the tabulated values are still needed for reference in the main text and are valuable for inter-comparison with past and future studies, such charts would cause a drastic increase in the number of figures and therefore the total length of the mansucript, which is already quite long. Each table lists quite many model variables (and observed variables for some tables), meaning that each of the tables would require several fugures/charts in order to avoid overbusy plots with multiple axes. We therefore strongly suggest to keep the tables as they are, but with one small change to make it easier to spot the largest biases at a glance in Tables 5 and 6: Using bold fonts for NMB numbers with absolute values larger than (e.g.) 50%.

Reference:

Liu, X., Penner, J. E., Das, B., Bergmann, D., Rodriguez, J. M., Strahan. S., Wang, M., and Feng, Y.: Uncertainties in global aerosol simulations: Assessment using three meteorological data sets, J. Geophys. Res., 112, D11212, doi:10.1029/2006JD008216, 2007.