

**Reply to anonymous referee #2**, interactive comment (C829–C832) on “Aerosol-climate interactions in the Norwegian Earth System Model – NorESM” by A. Kirkevåg et al:

Thank you for this very positive and constructive review, which has helped to improve the manuscript markedly. I hope you will find the following replies to your minor comments and questions (repeated below) satisfactory. The full references of the publications referred to but not found below are found in the manuscript.

**1. Page 2600, line 23: Explain which forcing is meant here (total aerosol, anthropogenic aerosol, : :?)**

We will change this sentence to: “The global anthropogenic aerosol direct radiative forcing (DRF) at the top of the atmosphere has changed from a small positive value to  $-0.08\text{W m}^{-2}$  in CAM4-Oslo.”

**2. The authors discuss that their model developments result in forcings which are closer to the AeroCom median or IPCC AR4 best estimates than the results of previous model versions. I think this could be misunderstood as a hard quality criterion. It is probably not really clear whether the AeroCom or AR4 results could be regarded as a reference since individual models could be superior to ensemble means when compared to measurements. I would recommend mentioning this in the text to prevent from misinterpretation. If the authors feel that, in some cases, the AeroCom or AR4 results could be regarded as benchmark, this should be justified in the paper.**

We do not feel that the median or mean AeroCom or IPCC AR4 results should be regarded as benchmarks, but we chose to compare our results with these because (compared to most other models) the forcing estimates in the previous model versions, CAM-Oslo, were on the high side (positive) for DRF and on the low side (large negative) for IndRF. We stated in the abstract (p. 2600, line 26) that “*Although it has not been a goal in this study*, the new DRF estimate is closer both to the median model estimate from the AeroCom inter-comparison and the best estimate in IPCC AR4.” Furthermore (on p. 2601, line 8), “The IndRF of  $-1.2\text{Wm}^{-2}$ , which is closer to the IPCC AR4 estimates than the previous estimate of  $-1.9\text{Wm}^{-2}$ , has thus been obtained without imposing unrealistic artificial lower bounds on cloud droplet number concentrations.” In the latter statement our point is that weaker IndRF values can be obtained without unphysical tuning of CDNC, which has been used in some climate models to constrain the total aerosol forcing. Nevertheless, we agree that these messages still might be misunderstood, and propose to further clarify these points in the summary and conclusion on p. 2653, by rephrasing the sentence starting on line 14 (new text is underlined for clarity):

“Although this is not to be regarded as an objective quality criterion, the new value,  $-1.2\text{Wm}^{-2}$ , is closer to the IPCC AR4 estimates constrained by the observed climate response (IPCC AR4, e.g. Fig. 2.20, Sect. 2.9).

Compared to Seland et al. (2008), the global DRF at TOA has changed from a small positive value to  $-0.08\text{Wm}^{-2}$ , which incidentally is also closer to the best estimate by IPCC AR4 (Forster et al., 2007), as well as to the AeroCom median model estimates (Schulz et al., 2006; Myhre et al., 2012).

**3. Page 2603, lines 6-8: The authors mention that they do not discuss the semi-direct effect in this manuscript. It would be interesting for the reader why this discussion has been omitted.**

It is perhaps more accurate to say that we have not *studied* the semi-direct effect, which would require extra sets of simulations where the effect of aerosol absorption on atmospheric heating and subsequent cloud development has been switched off. We propose to change this sentence to:

“We have not specifically studied the semi-direct effect in the present paper, although it is included in the model experiments which couple the aerosols and their radiative forcing online with the atmospheric thermodynamics (see Sect. 4.4). The semi-direct effect on the net radiative budget at the top of the atmosphere may be positive or negative, and its potential magnitude is characterized as small in the IPCC AR4 (Denman et al., 2007). The level of scientific understanding is furthermore characterized as very low.”

**4. Page 2608, line 9: The meaning of ‘thus’ is unclear to me. The fact that modes are changed according to processes does not imply to use size-segregated schemes.**

This sentence will be rewritten so that “thus” may be skipped, see also our reply to referee #1:

“In the separate scheme for size resolved aerosol physics these modes are changed in accordance with the processes to which the aerosol mass concentrations are tagged in the life-cycle scheme, and are described without assuming log-normality. As described in detail by Kirkevåg and Iversen (2002), the size distributions of number and mass concentrations used in the look-up tables (see below) are estimated by solving the discrete form of the respective continuity equations, using 44 size-bins with radii ( $r$ ) ranging from 0.001 to 20  $\mu\text{m}$ . The size bins are equally wide ( $=0.1$ ) along a  $\log_{10}(r)$  axis, so that the resolution, in terms of linear radius  $r$ , is finest for the smallest particle sizes.”

**5. Section 2.1, introduction on pages 2607-2609: It should be explained in more detail how the processing and transformation of the different aerosol species is realized in the model.**

Here the text will be updated, partly also based on the comments from referee #1. We propose to modify the part starting on line 24 on p. 2607 to:

“Our approach differs from the often applied modal method such as e.g. M7 (Stier et al., 2005) and MAM3 (Liu et al., 2012). The details of the approach are described by Seland et al. (2008), although the principles probably are more easily understood from the description of the simpler aerosol life cycle scheme in Iversen and Seland (2002, 2003) and the corresponding scheme for size resolved aerosol physics in Kirkevåg and Iversen (2002). As in Seland et al. (2008), the aerosol life-cycle scheme calculates mass-concentrations of aerosol species. These mass-concentrations are tagged according to production mechanisms in clear and cloudy air. There are up to four size modes for each of these tagged mass concentrations (nucleation, Aitken, accumulation, and coarse modes). The processes are gas phase and aqueous phase chemical production, gas to particle nucleation, condensation on pre-existing aerosol surfaces, and coagulation of smaller particles onto pre-existing Aitken, accumulation and coarse mode particles. The chemical components are sulphate ( $\text{SO}_4$ ), black carbon (BC), organic matter (OM), sea-salt (SS), and mineral dust (DU). In addition comes water which is mixed into the particles based on their hygroscopicity and ambient relative humidity.

The aerosol mass concentrations calculated in the life-cycle scheme (and transported in the model) are 11 components for externally mixed particles emitted or produced in air. These are the 10 modes listed in Table 1, where one of the modes, OM(a)/BC(a), contains two components. In addition there are 9 components which are tagged to production mechanisms in air or cloud droplets, so that the size resolved transformations into internal mixtures by interactions with the above 11 compounds can be estimated a posteriori by use of look-up tables, see below. These 9 components are:  $\text{SO}_4(\text{cond})$ , the

part of the sulphate mass produced in gas phase by oxidation of  $\text{SO}_2$  by OH ( $\text{SO}_4(\text{gas})$  in Fig. 1) which is estimated to condense on existing particles (note that the remaining part is assumed to produce nucleation mode sulphate,  $\text{SO}_4(\text{n})$ );  $\text{SO}_4(\text{a})$  is the part of the nucleation-mode sulphate ( $\text{SO}_4(\text{n})$ ) mass which is subject to condensation of gaseous sulphate produced in clear air and thus produces externally mixed sulphate in the Aitken mode;  $\text{SO}_4(\text{coag})$  is the mass of sulphate originating from  $\text{SO}_4(\text{n})$ ,  $\text{SO}_4(\text{a})$  and  $\text{SO}_4(\text{cond})$  which coagulates with accumulation and coarse mode particles in clear air and ends up as accumulation and coarse mode particles (note that the transfer rate for  $\text{SO}_4(\text{cond})$  is assumed to be the same as that calculated for  $\text{SO}_4(\text{a})$ );  $\text{SO}_4(\text{in water})$  is the mass of sulphate oxidized from  $\text{SO}_2$  in cloud droplets and the part of  $\text{SO}_4(\text{n})$  and  $\text{SO}_4(\text{a})$  which is collected by cloud droplets and ends up in accumulation and coarse mode particles after evaporation;  $\text{BC}(\text{cond}, \text{n})$  is the mass of Aitken mode BC originating from  $\text{BC}(\text{n})$  after condensation of  $\text{SO}_4(\text{cond})$ ;  $\text{BC}(\text{cond}, \text{a})$  is the mass of accumulation mode BC originating from  $\text{OM}/\text{BC}(\text{a})$  after condensation of  $\text{SO}_4(\text{cond})$ ;  $\text{BC}(\text{coag})$  is the mass of BC originating from  $\text{BC}(\text{n})$ ,  $\text{BC}(\text{a})$ ,  $\text{OM}/\text{BC}(\text{a})$ , or  $\text{BC}(\text{ac})$  that coagulates in clear air or in cloud droplets which subsequently evaporate and end up as accumulation and coarse mode particles;  $\text{OM}(\text{cond}, \text{a})$  is the mass of OM originating from  $\text{OM}/\text{BC}(\text{a})$  after condensation of  $\text{SO}_4(\text{cond})$ ;  $\text{OM}(\text{coag})$  is the mass of OM originating from  $\text{OM}/\text{BC}(\text{a})$  that coagulate in clear air or in cloud droplets, which subsequently evaporate and end up as accumulation and coarse mode particles.

This adds up to 20 aerosol components in addition to two gaseous precursors ( $\text{SO}_2$  and dimethyl sulphide, DMS). Fig. 1 gives an updated schematic representation of the aerosol processes in CAM4-Oslo, which facilitates comparison with the corresponding schematic for CAM-Oslo in Fig. 1 of Seland et al. (2008). It should be noted that the externally mixed OM mode from fossil fuel combustion, labeled  $\text{OM}(\text{a})$  in Seland et al. (2008), is removed in this work. The rationale for this is that the recommended size distribution for organic matter from fossil fuel is the same as for biomass burning particles (Dentener et al., 2006), and that the relative fraction of fossil fuel OM is small compared to OM emitted from biomass burning. This was in an early version of CAM4-Oslo shown to give very small changes in concentrations and life-time of OM, and small changes in the estimated aerosol properties in general.

As in Seland et al. (2008), the internally mixed mass from the processes described above is only added to and distributed onto the primary particles when calculating aerosol size distributions and optical properties for use in the cloud droplet activation code and in the radiative transfer code (cf. Kirkevåg and Iversen, 2002). ”

Immediately after this follows (see also our reply to editorial comment 2 and the new text proposed under question 4 above):

“The particle numbers and sizes are here estimated based on assumptions about the primary particles that are emitted or produced in air, of which there are 10 modes with log-normal size distributions as detailed in Table 1. In the separate scheme for size resolved aerosol physics these modes are changed in accordance with the processes to which the aerosol mass concentrations are tagged in the life-cycle scheme, and are described without assuming log-normality. As described in detail by Kirkevåg and Iversen (2002), the size distributions of number and mass concentrations used in the look-up tables (see below) are estimated by solving the discrete form of the respective continuity equations, using 44 size-bins with radii ( $r$ ) ranging from 0.001 to 20  $\mu\text{m}$ . The size bins are equally wide ( $=0.1$ ) along a  $\log_{10}(r)$  axis, so that the resolution, in terms of linear radius  $r$ , is finest for the smallest particle sizes.”

After the text originally ending on p. 2608, line 25, we further propose to add:

“When apportioning condensate and coagulated material between the various primary particle modes, condensed SO<sub>4</sub> is lumped together with coagulated SO<sub>4</sub> as input to the look-up tables for size distributions and optical parameters for sea-salt, mineral dust and SO<sub>4</sub>(ac) particles. This is done in order to keep the number of dimensions (for interpolation) of the look-up tables for each internally mixed mode down to five (Kirkevåg et al., 2005).”

Kirkevåg, A., Iversen, T., Seland, Ø., and Kristjánsson, J. E.: Revised schemes for optical parameters and cloud condensation nuclei in CCM-Oslo, Institute Report Series. **No.128**, Section for Meteorology and Oceanography, Department of Geosciences, Oslo, Norway, 2005.  
([http://folk.uio.no/kirkevag/instrep128\\_Kirkevag-etal.pdf](http://folk.uio.no/kirkevag/instrep128_Kirkevag-etal.pdf)).

**5.Section 2.1, introduction on pages 2607-2609 (cont'd): It should also be motivated why the described approach has been chosen and what the advantages and disadvantages are when compared to other methods as the log-normal approach.**

We propose to add on p. 2608, line 26:

“The main advantage of this approach, described above, is that the degree of internal vs. external mixing can be estimated based on physicochemical processes instead of explicit assumptions, and that the CPU costs are low compared to the full sectional approach. A disadvantage is that there is no explicit information about the size and mixing state of the aerosol masses (after growth) in the life-cycle scheme. A further disadvantage of this method is its rigidity. The need for complex and huge look-up tables makes it cumbersome to introduce changes to the basic physical properties of the aerosol, such as the assumed size parameters at time of emission of primary particles.”

**5.Section 2.1, introduction on pages 2607-2609 (cont'd): It is not clear to me why the aerosol components are classified according to single processes while being subject to a number of different kinds of processes. As an example, if SO<sub>4</sub>/OM/BC(coag) is formed by coagulation, it is not clear why it should not be subject to condensation of SO<sub>4</sub>. Or can aerosol mass be present in different components at the same time in the model? It would be helpful to the reader to find some brief information in the manuscript to avoid consulting previous literature.**

This is addressed in the first reply to this long comment, above.

**5.Section 2.1, introduction on pages 2607-2609 (cont'd): It should also be mentioned how gas phase precursor chemistry is realized (a short description with additional reference to section 2.1.6 would be sufficient).**

To answer this, we propose to add on p. 2608, line 26:

Apart from some exceptions described in Sect. 2.1.6, the sulphur chemistry is as described by Seland et al. (2008). Prescribed oxidant fields are still used, but now with an updated replenishment rate of H<sub>2</sub>O<sub>2</sub> in clouds (see Sect. 2.1.6). The DMS fraction converted into MSA is calculated explicitly by use of reaction rates given by Seinfeld and Pandis (1998). SO<sub>2</sub> is oxidized to sulphuric acid gas [SO<sub>4</sub>(gas)] in clear air by OH, and to particulate sulphate in aqueous phase cloud droplets [SO<sub>4</sub>(in water)] with an efficiency which is determined by the availability of H<sub>2</sub>O<sub>2</sub>, ozone, liquid water, and the rate of dynamic replenishment of cloudy air.

**6.Page 2616, line 1: the compensating effects should be described in more detail.**

We have not studied these compensating effects (which are accounted for in the prognostic CDNC scheme and not the diagnostic scheme) in any detail, but some of these are: i) In the prognostic CDNC scheme, the cloud droplet number tends to be lower than in the diagnostic scheme because the droplet number can be reduced after activation by collision-coalescence processes, evaporation and freezing over the course of more than one time-step. These sinks were all ignored in the diagnostic CDNC scheme used in the CAM-Oslo version of e.g. Seland et al. (2008). CDNC was there simply estimated as the activated concentration of CCN(S), where S was a prescribed supersaturation which only varied with cloud type (stratiform or convective); ii) Calculated super-saturations are here also smaller for PD than PI conditions due to competition effects (for water vapour), while the supersaturation was the same for PD and PI in the diagnostic approach.

We propose to rewrite the sentence starting on p. 2615, line 25:

“A preliminary sensitivity test involving prognostic calculation of both CDNC and LWC, with activation of CCN following Abdul-Razzak and Ghan (2000), indicated a reduction of the first indirect forcing (the radius effect) by 36% due to compensating effects not accounted for in the diagnostic scheme. One such compensating effect is the competition for available water vapour, which leads to smaller realized supersaturations for polluted present day conditions than the more pristine preindustrial conditions. Another effect is the loss of cloud droplets due to collision-coalescence processes, evaporation and freezing, which were not considered in the diagnostic scheme of e.g. Seland et al. (2008), therefore giving larger CDNC in general in that study.”

**7.Section 4.1, Table 3: The reasons for differences between ‘total emissions’ and ‘total sources’ in the case of primary particles should be discussed. I would expect the two quantities to be identical.**

What we name total sources is in reality total deposition. Averaged over multi-year simulations these two should ideally be equal, but they differ due to numerical inaccuracies and finite simulation lengths. Total sources include secondary production in addition to the emissions. For the primary particles SS and DUST we see that the total deposition in CAM4-Oslo is within 1% of the total sources/emissions within less than 1% accuracy. To clarify this point we propose to add on p. 2620, line 7:

“Total source numbers are here simply assumed to equal the total deposition, since secondary produced aerosols are not standard output in the model. Due to numerical inaccuracies and finite simulation lengths this assumption is seen (from primary Sea-salt and mineral dust in Table 3) to be accurate to within about 1%.”

**8.Section 4.2.2: To learn more about the vertical transport of aerosol in the model it could be useful to consider also observed vertical profiles of primary particles, which are not subject to in-situ generation from precursors and therefore largely constrained by transport and deposition. Data on primary aerosol vertical distribution from aircraftbased field campaigns is available for black carbon (see e.g. Schwarz et al., 2008, JGR, D03203; Schwarz et al., 2006, JGR, D16207) or dust (see e.g. Weinzierl et al., 2009, Tellus B, 61, 96–117).**

This is an interesting suggestion for future work, but we consider it to be a bit outside the scope of this already quite comprehensive study, and too late to include at this stage. Furthermore, a problematic aspect of comparing modeled data with the suggested measurements is that they are not climatological averages, but are referring to specific situations. The already included comparison with vertical extinction profiles therefore probably provides a more suitable evaluation of the vertical aerosol

distribution as such. However, there is one somewhat related study with NorESM/CAM4-Oslo (lead by Risto Makkonen) in progress, focusing on improvements and validation of the aerosol nucleation scheme in CAM4-Oslo. This is already mentioned very briefly on p. 2654, line 4. Near surface values and height profiles of total aerosol *number* concentrations are here being compared with measurements. Scheduled submission for this paper is early next year.

### **Editorial changes:**

#### **1. Figure 1, caption, line 7: ‘assumed transformed’ makes no sense. Q(xx) should be explained.**

We propose to change the figure caption text as follows (new text is underlined for clarity):

“Schematic for aerosol particle processing in CAM4-Oslo. The source terms on the left side, labeled Q(X)y, where X is the constituent name and y is the source type, may come from primary emission or secondary production. The source labels bb, ff and bio respectively indicate biomass burning, fossil fuel combustion, and biogenic sources. Primary particles are emitted (dashed-dotted arrows) as accumulation-mode sulphate (SO<sub>4</sub>(ac)), nucleation and accumulation mode black carbon (BC(n), BC(ac)), Aitken mode BC (BC(a)), internally mixed Aitken mode organic matter and BC (OM/BC(a)), Aitken, accumulation, and coarse mode sea-salt (SS(a), SS(ac), SS(c)), accumulation and coarse mode mineral dust (DU(ac), DU(c)). Model calculated gas-phase components are DMS and SO<sub>2</sub>. Gaseous sulphate (SO<sub>4</sub>(gas)) produced in air is assumed to be transformed to nucleation-mode sulphate (SO<sub>4</sub>(n), dotted arrow) if insufficient particle surface area is available for condensation (solid arrows). Sulphate produced in cloud water droplets (SO<sub>4</sub>(in water), short-dashed arrow) is partly added to SO<sub>4</sub>(ac) but mainly to a broad internal mixture (dashed rectangles) of accumulation and coarse mode particles (of which there are two types with respect to complexity). Long-dashed arrows represent coagulation which contributes to the latter particle types.

#### **2. Page 2608, line 6: replace ‘is’ by ‘are’.**

I think “is” is the correct form to use here, but we may rewrite this sentence to: “The particle numbers and sizes are here estimated based on assumptions about the primary particles that are emitted or produced in air, of which there are 10 modes with log-normal size distributions as detailed in Table 1.”

#### **3. Page 2630, line 23: replace ‘data prepared by Brigitte Koffi for year 2007’ by ‘data for 2007 (B. Koffi, personal communication, : : .)’ , or similar.**

“data prepared by Brigitte Koffi for year 2007” will be replaced with “data for year 2007 (Brigitte Koffi, personal communication, 2012)”.

#### **4. Section 4.3: the notation of units (W/m<sup>2</sup>) is partly erroneous.**

Thank you, this will be corrected.

#### **5. Page 2651, line 14: Rewrite ‘other changes have proven important’.**

We propose to rewrite this sentence to

“Although the properties and effects of increased natural aerosols are emphasized, other changes have also been shown to be important, such as the omitted mixing between updrafts and downdrafts in deep convective clouds, and the shift in basic years for present day and preindustrial emissions.”