<u>Reply to anonymous referee #1</u>, interactive comment (C762-C765) 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.

• Page 2608, line 1: Is each aerosol species present in each mode (5 species x 4modes = 20)? In figure 1 it appears to be more than 20 components.

Not all species are present in all the size classes: As seen from Fig. 1, there are 2 species (SO4 and BC) in the nucleation mode, 4 (SO4, BC, OM and SS) in the Aitken mode, and 5 (SO4, BC, OM, SS and DU) in the accumulation and coarse modes, adding up to only 16 size-modes. To explain what is meant more explicitly, we propose to change the text starting on line 2 (see also reply to referee #2):

"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. This is the 10 modes listed in Table 1, where the OM(a)/BC(a) mode contain 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."

This will then be followed by a list and a short description of the 9 components. For the full text, see our reply to question 5 from referee #2.

• Page 2608, line 10: What does "size-bins with equal width with respect to the logarithm of the particle radius" mean?

This sentence (lines 8-10) was perhaps not formulated very clearly. What we mean, mathematically, is that $log_{10}(r_i) = d^*(i-1)-3$, where d=0.1. We propose to add a reference and rewrite the sentence to:

"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 μ m. The size bins are equally wide (=0.1) along a log₁₀(r) axis, so that the resolution, in terms of linear radius r, is finest for the smallest particle sizes."

• Page 2608, line 21: Do you mean that the Köhler theory (line 13) is used for calculating the lookup tables?

If you mean the sentence ending on line 21, the answer is no. For clarity we rewrite this sentence to: "Output from one set of tables are dry aerosol modal radii and standard deviations based on lognormal fits to the size distributions, which are used as input to the CCN-activation calculations (Hoose et al., 2009; see also Abdul-Razzak and Ghan, 2000)."

The Köhler theory is here embedded in the activation code, but not used for calculating the *dry* aerosol size distribution. If you instead meant the sentence starting on line 21, the answer is yes, since the optical properties of the *wet* aerosol also depend on hygroscopic swelling. As stated in the following line, however, this is described in more detail in Seland et al. (2008) and Kirkevåg and Iversen (2002).

• Page 2608, line 28 and everywhere in the manuscript: I am not sure whether it is correct to use a period before i.e.

We will rewrite the 3 sentences (on p. 2608, l. 28; p. 2619, l. 15; and p. 2639, l. 27) which started with "I.e.," to instead start with "This means that".

• page 2610, line 1: there is a typo in "exercise".

Thank you, this will be corrected.

• page 2612, line 1: I am confused by the notation: do the authors want to say that the ratio of MSA to S is 3 to 1? If so, I think writing "MSA to S (3:1)" is clearer.

We agree, and we will change the text accordingly.

• page 2612, line 15: Is W the white cap fraction? I think it should be directly stated.

This was stated a bit further down, on line 19, but to avoid unnecessary confusion we will follow up Eq. 1 with: ", where W is the white cap fraction:", and then move Eq. 2 up accordingly.

• page 2614, line 9: I understand that CAM4-Oslo does not actually have an oxidant chemistry. The oxidant chemistry is calculated with Oslo-CTM2, and then read in by CAM4-Oslo. If so, the title is confusing. I would rather call this section "Oxidant fields" or anything that does not imply the presence of a chemistry module.

We fully agree, and we will rename section 2.1.6 to "Oxidant fields" in the final manuscript.

• page 2614, line 15: the new H_2O_2 values are smaller than Seland et al. (2008) in the UT and in the stratosphere; is this a problem? Is CAM4-Oslo suitable for studies in the UTLS and stratosphere?

The model does not describe chemical processes specific for the stratosphere, and thus the answer to both questions is no.

• page 2615, line 4: Is dust emitted only offline, or is it possible to have it emitted online depending on the simulated winds?

A parameterization for prognostic/online dust emissions is unfortunately not available in the present model version.

• page 2615, line 12: is there a reference for the 0:25 value of the scavenging coefficient, or was it assessed through sensitivity tests?

The value 0.25 can be said to be a result of sensitivity testing, although not very rigorous as only a very small number of values have been tested. As mentioned in the text, a value of 1 was used by Seland et al. (2008), 0.1 was used by Hoose et al. (2010). Based on those papers, 0.25 was chosen in this study.

• page 2620, line 19: the sea salt lifetime is slightly longer in this work than in Seland et al. (2008), in spite of the fact that the wet and dry deposition are higher. Is it consistent?

It is correct that the wet deposition is higher in this work, but the dry deposition is lower than in Seland et al. (2008), both seen as percentages and as Tg/yr (see Table 3). Aerosol lifetimes also

depend on how emission and deposition fields vary in time and space, which are different in the two model versions. This is tentatively explained as follows in the following text (lines 19-23):

"In spite of the enhanced importance of gravitational settling, the fraction deposited by precipitation scavenging is considerably higher in this work. This is probably a consequence of the wind (and SST) driven emissions in CAM4-Oslo. Strong winds over oceans are often co-located with precipitation. The prescribed emissions in Seland et al. (2008) would more often, and erroneously, not be associated with the actual storms predicted in the atmospheric model, leaving a higher preference for dry rather than wet deposition."

• page 2622, line 20: could it also be a sign of too high deposition, which limits long range transport?

The negative bias is not rigorously shown to be a result of missing sources, and we agree that the possibility of too high deposition still cannot be ruled out, even though it is known that non-desert dust emissions are likely under-estimated. We therefore propose to add, from line 22:

"Although the treatment of deposition has been updated since Seland et al. (2008) and Hoose et al. (2009), the possibility that the deposition of mineral dust is still over-estimated cannot be ruled out, e.g. close to sources where many dust-compounds are hydrophobic."

• page 2628, line 15: the simulated AOD is much smaller than observed over India. Do you know why that happens?

We have presented two possible explanations to this on p 2627, lines 21-26. We propose to modify the last of these two sentences slightly:

"As in the previous model version by Seland et al. (2008), too much and too frequent precipitation over the Indian Ocean and parts of the continent of southern Asia (not shown) lead to exaggerated wet deposition estimates, which probably explains some of the negative biases for that region. In the densely populated South Asia region we also anticipate that we miss important contributions from secondary aerosol formation of organics, and possibly also mineral dust from dry soils."

• page 2631, line 5: "possible" or "possibly"?

Thank you, "possibly" is the correct word to use here.

• page 2631, line 26: "it is overestimated...". What is "it"?

By "it" we here meant the extinction. We will rephrase this sentence to "The aerosol extinction is overestimated by up to about 100% between 1.5 and 3km height a.s.l."

• page 2633, line 26 and ff: in this section the units of RF should be W m-2.

Thank you, this will be corrected.

• page 2634, line 18: How does exactly internal mixing affect AOD? Couldn't also increase it? For instance, the presence of a coating increases the absorption by BC (Bond and Bergstrom, Aerosol Sci Tech, 2006, vol 4(1) pp27-67).

In CAM4-Oslo coating is only assumed to affect CCN activation, while the optical properties are estimated by assuming homogeneous internal mixing (which is radius dependent within a size mode)

for particles that are subject to processes leading to internal mixing. What we compare in this section is not the effect of external vs. internal mixing on optics per se, but rather the effect on AOD or ABS for internal mixing of all aerosol components (at the same time) compared to the sum of each of the separate contributions which are also internally mixed. These separate contributions are found from the difference between total AOD or ABS and AOD or ABS from all components except the one in question. This follows from the special set-up of these experiments, as described in section 3.1 and by Myhre et al. (2012). Without running further specially constructed sensitivity experiments, we cannot explain in detail why the very small non-linearity we found is of this particular nature. We therefore propose to modify the sentence starting on p. 2634, line 18 to:

"Due to non-linear effects, probably related to small differences in size and mixing state of the internally mixed aerosol in these experiments, the sum of each contribution is (only) about 1.5% larger than the total AOD increment for PD-PI of 0.0527."

• page 2635, line 8: when you say "internally mixed OM/BC", do you mean that OM and BC are internally mixed with soluble material, or that they are internally mixed together? How hygroscopic do you consider them?

As indicated in Fig. 1, this mode is assumed to be an internal mixture of only BC and OM at the point of emission, but it may be further mixed with sulphate (and H_2O) condensate, as well as with larger sea-salt and dust particles through coagulation.

BC is assumed to be practically hydrophobic, while the hygroscopicity of OM is 0.25 times that of ammonium sulphate. The hygroscopicity of all internally mixed particles is calculated based on the volume fraction of each of the internally mixed components, as explained on page 2608 (with some changes, see my reply to the interactive comment by K. Zhang for more details).

• page 2643, line 1: yields

Thank you, this will be corrected.

• page 2647, line 12: the presence or absence of a hydrophilic coating should affect the scavenging, too. Is this feedback not included in CAM4-Oslo? If it is, it should affect the life cycle.

In the present model version the hydrophilic coating is still only assumed to affect CCN activation, not deposition and the aerosol life cycle, nor the optical properties. So no such feedback is yet included in CAM4-Oslo. This is described in more detail in Sect. 5.9.

• page 2650, line 13: fluffy BC particles?

We agree that "fluffy" is not an accurate description of these particles. We will remove this word and change the sentence starting on line 12 to:

"The relative amount of fossil fuel BC from rapid combustion which in the model is directly emitted as fractally shaped (Ström et al., 1992) accumulation mode particles of low mass density, BC(ac) in Table 1 and Fig. 1 (agglomerates of BC(n) monomer particles, see Kirkevåg et al., 2002; Kirkevåg et al., 2005; Seland et al., 2008), is in *Ctrl* assumed to be 10 %, as in Seland et al. (2008).

Ström, J. S., Okada, K. and Heintzenberg, J. 1992: On the state of mixing of particles due to Brownian coagulation, J. Aerosol Sci. 23, 467–480.

• page 2669, table 3: I am not sure about the difference between emission and sources. Is "sources" the production of secondary particles? If so, why SS and dust have different numbers for emission and sources? SS and dust should be only primary particles.

What we name total sources is in reality total deposition from the model output. Averaged over multiyear simulations these two should ideally be equal. However, they may 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. 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%."

• page 2675 and ff: The fonts in all figures is much too small, hardly readable.

This will be corrected in the final manuscript version.