Response to review of "Sensitivity of black carbon concentrations and climate impact to aging and scavenging" by Marianne T. Lund, Terje K. Berntsen and Bjørn H. Samset.

We thank the anonymous referee for the carefully and thorough reviews of our paper and the useful suggestions. Responses to individual comments are given below.

Anonymous Referee #2

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The paper evaluated simulated black carbon in OsloCTM2-M7 against various observations and performed several sensitivity simulations by varying BC aging and scavenging parameters. The paper particularly focused on improving BC predictions over the high latitude, which is a particularly interesting topic as potentially important role of BC on climate changes occurring in high latitude such as Arctic.

Despite this importance, I have major concerns with this paper. I agree with all the concerns addressed by the referee #1. I particularly agree that this paper does not provide new findings. Here is the list of my major comments. Please consider them to improve this manuscript.

Major comments:

1) Regarding BC modeling in OsloCTM2-M7, please explain any difference/update in BC modeling used in this study compared to the ones used in previous studies. Without such information, this paper appears to be very redundant to previous studies with OsloCTM2. This is particularly because OsloCTM2 has participated several multi-model inter-comparison studies (e.g., AEROCOM) focused on black carbon evaluations against observation. Also, there were previous studies using OsloCTM2 (maybe with bulk aerosol model) improved BC prediction by adjusting aging/deposition parameterization and shortening BC lifetime (e.g., Skeie et al. 2011; Hodnebrog et al, 2014). The authors should make it clear how the model different from previous studies with OsloCTM2, and how BC predictions in this model are improved from previous OsloCTM2 evaluation. Specifically, Lund and Berntsen (2012) evaluated OsloCTM2-M7 BC predictions against the same observation. Please explain how the BC modeling and evaluation results in this paper differ from that previous paper.

Lund and Berntsen (2012) performed the first analysis of BC simulated by the M7 in the OsloCTM2 and compared the M7 with the standard bulk parameterization (OsloCTM2-BULK). A basic evaluation against selected measurements was performed, showing that the M7 improved the representation of Arctic surface concentrations compared with the bulk scheme, but that considerable overestimation of high altitude concentrations remained a problem. Because the M7 significantly increases the required computing time, the simulations of Lund and Berntsen (2012) were used to generate a latitudinally and seasonally varying look-up table of aging rates for use in the bulk scheme as documented in Skeie et al. (2011). In this way, at least some of the spatial and temporal variability in aging could be accounted for in the bulk scheme. However, this approach can only capture variations under present-day conditions, e.g., changes in variability with changing emissions not captures. Furthermore, despite existing issues, using an aerosol microphysical module like M7 provides a physically more realistic parameterization, which we believe is needed in order to improve the modeling of aerosols. Building on the findings in Lund and Berntsen (2012), we here perform a much more thorough documentation of the model (which is also needed due to recent important updates to the emission inventories) and explore possible reasons for the high-altitude discrepancies identified previously. Moreover, using a microphysical module allows us to investigate parameters beyond those examined by Hodnebrog et al. (2014) who used the bulk aerosol module. We also provide additional information by focusing simultaneously on model capabilities at high latitudes and remote regions over the Pacific; whereas other sensitivity studies often focus on one or the other. See also response to comment 1) by referee #1.

To clarify, we have modified the introduction section:

"Here we examine the sensitivity of modeled BC concentrations to factors controlling aerosol lifetime in the OsloCTM2 (Sovde et al., 2008) coupled with the aerosol microphysical parameterization M7 (Vignati et al., 2004) (hereafter OsloCTM2-M7). The chemical transport model OsloCTM2 has been documented and used in several multi-model aerosol studies (Balkanski et al., 2010; Myhre et al., 2013; Schulz et al., 2006; Shindell et al., 2013; Textor et al., 2007). These studies used a simplified bulk aerosol scheme. Lund and Berntsen (2012) (hereafter LB12) performed the first analysis of BC simulated by the M7 in the OsloCTM2 and compared results with those from the bulk parameterization. A basic evaluation against selected measurements was performed, showing that using M7 improved the representation of Arctic surface concentrations compared with the bulk scheme, but exacerbated the overestimation of high-altitude BC.

Building on the findings in LB12, we perform a range of sensitivity experiments varying key assumptions in the treatment of aging and scavenging in OsloCTM2-M7 and investigate the resulting range in vertical BC profiles, as well as high-latitude surface concentrations. Using updated emission inventories, three years of model results and observations from surface stations, flight campaigns, and snow samples, we also perform a more thorough documentation of the current model performance. Our experiments include a first step towards accounting for BC aging by gas-phase nitric acid condensation. Measurements have shown that nitrate is frequently present in internal aerosol mixtures (Pratt & Prather, 2010; Shiraiwa et al., 2007). Aging through interaction with nitrate may also become more important in the future following strong projected decreases in SO_2 emissions and increasing NOx and greenhouse gas emissions (Bauer et al., 2007; Bellouin et al., 2011; Makkonen et al., 2012), but has so far not been accounted for in the model. We also take the analysis one step further and estimate the range in global RF and surface temperature resulting from the changes in the model parameters.."

2) The BC sensitivity results do not seem so informative. Large portion of the paper results are focused on BC evaluations, not the sensitivity results - This paper actually fit better as OsloCTM2-M7 BC evaluation paper, rather than BC sensitivity study. If the authors wish to stay focused on BC sensitivity study, I strongly recommend examining details comparison (i.e., spatial and temporal distributions of concentrations and radiative forcing) among the sensitivity simulations to find any interesting spatial and temporal differences. This may be helpful to understand the climate impact.

This is a good point. While we would argue that a careful documentation of the model performance in its original setup is needed before examining the range of results in the sensitivity tests and attempting to identify potential improvements, we agree with the referee that the paper should increase the focus on the sensitivity experiments. We have kept most of text describing the model evaluation, but shortened it where possible. We have also moved the figures showing the model-measurement comparison of other species than BC to a supplementary material. We also analyze the difference in spatial distribution of BC concentrations in the sensitivity experiments in more detail, including two new figures. Finally, the global vertical profiles of forcing and temperature response are examined in more detail.

Minor comments:

Abstract section

1) Please re-write Abstract. I got an impression that the current abstract is just a short version of the conclusion section. I found some identical sentences or phrases between abstract and conclusions. Also, the abstract seems too long and needs to improve readability. Here are some examples:

P1 L12 : please modify "microphysical aerosol to "aerosol microphysical"

P1 L14 : Please clarify "Arctic surface concentrations". Is this BC ambient concentrations or BC in Arctic snow or both?

P1 L14: please modify "remote region BC vertical profiles" to "BC vertical profiles at remote region".

P1 L17 : please modify "annual averaged" to "annually averaged" or "annual average"

P2 L22: Please re-write this sentence: "Several processes can achieve this".

The abstract has been rewritten and minor comments below addressed where still applicable*:*

"Accurate representation of black carbon (BC) concentrations in climate models is a key prerequisite for understanding its net climate impact. BC aging scavenging are treated very differently in present models. Here, we examine the sensitivity of 3-dimensional, temporally resolved BC concentrations to perturbations to individual model processes in the chemistry-transport model OsloCTM2-M7. The main goals are to identify processes related to aerosol aging and scavenging where additional observational constraints may most effectively improve model performance, in particular for BC vertical profiles, and to give an indication of how model uncertainties in the BC life cycle propagate into uncertainties in climate impacts. Coupling OsloCTM2 with the microphysical aerosol module M7 allows us to investigate aging processes in more detail than possible with a simpler bulk parameterization. Here we include, for the first time in this model, a treatment of condensation of nitric acid on BC. Using radiative kernels, we also estimate the range of radiative forcing and global surface temperature responses that may result from perturbations to key tunable parameters in the model. We find that BC concentrations in OsloCTM2-M7 are particularly sensitive to convective scavenging and the inclusion of condensation by nitric acid. The largest changes are found at higher altitudes around the Equator and at low altitudes over the Arctic. Convective scavenging of hydrophobic BC, and the amount of sulfate required for BC aging, are found to be key parameters, potentially reducing bias against HIPPO flight-based measurements by 60 to 90 percent. Even for extensive tuning, however, the total impact on global mean surface temperature is estimated to less than 0.04K. Similar results are found when nitric acid is allowed to condense on the BC aerosols. We conclude, in line with previous studies, that a shorter atmospheric BC lifetime broadly improves the comparison with measurements over the Pacific. However, we also find that the model-measurement discrepancies can not be uniquely attributed to uncertainties in a single process or parameter. Model development therefore needs to be focused on improvements to individual processes, supported by a broad range of observational and experimental data, rather than tuning of individual, effective parameters such as the global BC lifetime."

Section 2.3

1) Regarding BC aging by HNO3 condensation, please explain why HNO3 produced in the aq. Chemistry has to be excluded. Is this to estimate gasphase production?

Yes, this is to estimate gas-phase production of HNO3. We realize that since it's in fact the gas-phase nitric acid we're after here, showing results also from the case including the aqueous phase production is confusing and not very informative. We have therefore removed the latter experiment in order to focus on the correct one.

2) It looks like the required ML is different for sulfate and nitrate. In reality, these hydrophilic aerosols will condense on BC surface and change BC properties. Isn't it more realistic to set the required ML combined for sulfate and nitrate? Am I missing something?

We realize this description is very unclear (this was also pointed out by referee #1). There is only one variable giving the number of MLs required for moving a BC aerosol from the insoluble to the soluble mode and the number of particles than can be moved, i.e., has sufficient associated soluble material, is determined from the total sulfate and nitric acid condensation. However, when adding nitric acid, we perform additional simulations with 5 and 10 MLs, reflecting the range of values used in previous studies. The text has been clarified:

"The number of MLs used as the criterion for aging ranges in existing literature. In its original setup M7 assumes 1 ML, based on the best agreement with a sectional model found by Vignati et al. (2004), but this considers sulfate as the only condensable species. Other studies have used a 5 (Pringle et al., 2010) and 10 (Mann et al., 2010) monolayer scheme. Reflecting this range and examining the subsequent impact on concentrations, we here perform three runs assuming 1, 5 and 10 ML are required for aging."

Section 2.4

1) I can't follow the first paragraph describing the method (L226-L238) to distribute BC burden to CESM-CAM4 model. Can you please re-write this method more clearly? Did you have to re-gridding BC burden?

We have expanded and clarified this method description and the section now reads:

"To estimate implications of the concentration changes in our experiments for the global BC climate impact, we use precalculated radiative forcing (RF) and surface temperature (TS) kernels derived with the CESM-CAM4 (Samset & Myhre, 2015). These 3-dimensional, temporally varying kernels were constructed by systematically applying a uniform BC burden to one model layer at a time, and calculating the resulting responses. Effective radiative forcing (ERF) was extracted from simulations with prescribed sea-surface temperatures, while temperature responses were taken from simulations with a slab ocean setup. As shown in (Samset & Myhre, 2015), it is possible to take a perturbation to the 3D concentration, multiply it with the kernels, and get an estimate for the resulting ERF and temperature change. However, because the BC perturbations were applied uniformly throughout a single model layer, the temperature response at each grid point will be due to both BC forcing exerted locally and to forcing in surrounding gridboxes. In the present analysis, we therefore focus on global mean vertical profiles. For each experiment, the globally averaged vertical BC profile from the OsloCTM2-M7 is multiplied with the globally averaged vertical forcing and temperature change kernels, respectively. The kernels are interpolated to the OsloCTM2-M7 resolution before use."

Section 3.1.1

1) L314-L315 : Please provide a citation.

Text slightly modified and citation added:

"Studies have found that models often struggle to capture the seasonal cycle and magnitude of measured high-latitude BC surface concentrations (e.g., Eckhardt et al. 2015; Shindell et al. 2008)."

2) L323-340 : This study applies seasonality in agricultural waste burning and domestic BC emissions. What about other emission sources? What is the impact of missing seasonality of other emission sources?

The ECLIPSEv4 emission inventory used in this study does not include seasonality of other sectors than domestic and agricultural waste burning (AWB). However, monthly data is provided in a more recently released version, ECLIPSEv5. Aside from AWB and domestic emissions, the seasonal variation in this inventory is negligible, both globally and north of 30 degree north. We have added the following text in the methods section 2.2:

"Seasonality of emissions in other sectors is not included in ECLIPSEv4. In the more recently released ECLIPSEv5 inventory (MLhttp://eclipse.nilu.no/, the monthly variability in emissions from other sectors is minor or negligible."

3) L335-336 : Is this for certain year? 2008?

The emissions in the Wiedinmyer et al. (2014) study is based on year 2010 population and waste generation data. This has been clarified in the manuscript.

4) L 348-349 : Please present the CO evaluation for SH region.

We have included the SH evaluation in the supplementary material (see response to comment major comment #2 above).

Section 3.1.2

1) L361 : Please provide a citation.

Citations have been added and the text modified as follows for clarification:

"During April 2008, when these campaigns were undertaken, there was unusually strong fire activity in Siberia and air masses were heavily influenced by biomass burning emissions (Brock et al., 2011; Jacob et al., 2010; Warneke et al., 2009). During several flights, the biomass burning plumes were specifically targeted. Possible reasons the strong discrepancies could be underestimation of the fire emissions during these extreme fire events or inaccurate representation of the plumes in the model."

Jacob, D. J., et al.: The Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) mission: design, execution, and first results, Atmos. Chem. Phys., 10, 5191-5212, doi:10.5194/acp-10-5191-2010, 2010

Brock, C. A., et al.: Characteristics, sources, and transport of aerosols measured in spring 2008 during the aerosol, radiation, and cloud processes affecting Arctic Climate (ARCPAC) Project, Atmos. Chem. Phys., 11, 2423-2453, doi:10.5194/acp-11-2423-2011, 2011.

Warneke, C., *et al.* (2009), Biomass burning in Siberia and Kazakhstan as an important source for haze over the Alaskan Arctic in April 2008, Geophys. Res. Lett., 36, *L02813*

2) L364-367: This doesn't apply to ARCTAS summer. Please explain why.

The majority of flights during the summer campaign of the ARCTAS took place over Canada, further south than the spring campaign. During summer 2008, there was considerable fire activity in Northern California and Siberia, but the fire activity over Canada was generally low. One possible reason for the better agreement with the measurements from ARCTAS summer is that the model was better able to reproduce the plume transport in this region and/or time of year. Our evaluation against monthly surface concentrations of BC also suggest a generally better agreement at high latitudes during summer than spring. Of course, these differences also underline that flight campaigns only provide a snap shot comparison and that one should be careful not to generalize results. The following paragraph has been added:
"The majority of flights during the ARCTAS summer campaign took place over Canada, where the fire activity was generally low that year. Moreover, our evaluation against monthly surface concentrations of BC also suggest a generally better agreement at high latitudes during summer than spring (Sect. 3.1.1)."

3) L385-386 : I am not sure what this mean. Please explain why it is less important for aerosol distribution.
We agree that this is unclear. The sentence in question has been deleted and the following sentence modified to:
"In contrast, the HIPPO campaigns sampled older air masses, where loss processes have had more time to influence the distribution."

Conclusion section
1) Please see the comments for Abstract section above, which are also applied to this section as well.
We have rewritten the conclusion section to the comments above; please see revised manuscript.
2) L561: put comma between "aging" and "and".
Corrected
3) L581: please specify how much MNB is changed
Specification has been added and text slightly modified accordingly:
"Allowing for convective scavenging of hydrophobic BC and reducing the amount of soluble material required for aging results in a 60 to 90 percent lower MNB in the comparison with vertical profiles from HIPPO, relative to the baseline."
4) L584: It looks like this part has a grammatical error: "… available for removal, a parameter with large ".
Corrected
5) L584 : "uncertaines" typo
Corrected
6) L587: "fligh" typo
Corrected
7) L589: please specify how big is the overestimation.
No longer applicable after this section has been rewritten.
8) L607- L609 : This sentence should be rewritten. It doesn't read well.
We agree and have modified the sentence:
"In the experiments resulting the most pronounced BC concentration changes relative to the baseline, we calculate changes in global RFari (i.e., direct RF) on the order of 10-30% of the total pre-industrial to present BC direct forcing."

9) L610: please change “is” to ”are”.
Corrected
10) L617 : please fix this part: “dependen on”
Corrected
11) L614-618: This is very long sentence and it is not well read. Please re-write this.
We agree and have rephrased:
“The existing model-measurement discrepancies in the OsloCTM2-M7 can not be uniquely attributed to uncertainties in a single process or parameter.”
12) L618-619: Please explain more what you mean by “tradeoffs … between different regions”.
Modified:
“Furthermore, improvements compared to measurements in one geographical region, can be accompanied by a poorer model performance in other.”
13) L621 –L622: If possible, please specify what kind of observation data that would be especially useful to improve BC modeling?
Examples have been added: *“(…) e.g., of BC IN efficiency, aging rate and mixing state (…)”*