**Response to review of "Sensitivity of black carbon concentrations and climate impact to aging and scavenging in OsloCTM2-M7" by Marianne T. Lund, Terje K. Berntsen and Bjørn H. Samset.**

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

**Anonymous referee#3**

The paper represents a useful addition to the growing body of literature concerning the sensitivity of BC concentrations to parameterization uncertainties. The paper would benefit from a more detailed description of the modeling approach and discussion of key results. In particular, the meaning of results from sensitivity tests with modified scavenging of hydrophobic BC (ConvBC) and condensation of nitrate (NitCond) is not sufficiently clear.

Major comments:

How much does convective aerosol scavenging contribute to total deposition? How is convective scavenging and transport represented in the model? Is a mass flux scheme used and how are aerosols treated in that scheme? Are interactions between entrainment, detrainment, and scavenging processes in convective aerosol mass budgets accounted for?
We have added a description of the parameterization of large-scale and convective cloud systems, to compliment the description of assumptions related to BC solubility and removal. The following paragraph has been included:

*"Wet deposition is calculated based on ECMWF data for convective activity, cloud fraction and rain fall, and on the solubility of individual species. Removal in large-scale cloud systems follow the scheme by Berge [1993]. The parameterization of deep convection is based on the Tiedtke mass flux scheme [Tiedtke, 1989], with mass redistributed in the vertical by a so-called "elevator" principle, i.e., surplus or deficit of mass in the column [Berglen et al., 2004]."*

Why do BC concentrations respond strongly to changes in the scavenging of hydrophobic BC in the convection? Various studies have shown that externally mixed BC particles are very rare in the remote atmosphere which implies that concentrations of hydrophobic BC should be quite low. This would likely imply a low sensitivity of model results to hydrophobic aerosol concentrations in model sensitivity studies. Please quantify the amount of hydrophobic BC in the model and further information about aging time scales etc.

This is an important observation. In fact, in the OsloCTM2-M7 baseline setup, the insoluble Aitken model BC particles (BCi) make up the largest fraction of total BC at both high latitudes and around the tropics. The high-latitude BCi concentrations were also noted by Lund and Berntsen (2012), but we agree that a discussion of this feature is needed in the present study as well. We have added a figure showing the zonal mean concentration of total BC in the baseline simulation and the relative contribution of BCi to the total in the supplementary material. The output required to

calculate aging time scales for all sensitivity experiments (i.e., separate information of the wet removal of BCi) is not available, but we quantify the aging time in the baseline case. The following has been added to the discussion:

*"The strong sensitivity of concentrations at high northern latitudes and around the tropics to changes in the convective scavenging of hydrophobic BC and inclusion of aging by nitric acid may seem surprising given that measurement suggest that significant the majority of freshly emitted hydrophobic BC particles quickly acquire coatings and become internally mixed (Gong et al., 2016; McMeeking et al., 2011; Moteki et al., 2007). Consequently, externally mixed BC particles are likely rare in the remote atmosphere. However, there is still little information about the aerosol mixing state in aged air masses, especially at high latitudes (Raatikainen et al., 2015). In the baseline OsloCTM2-M7, a considerable fraction of total BC is in the insoluble Aitken mode BC (BCi) (45%, or 60 Gg, of the global BC burden with an aging time scale of 2.7 days). In particular, BCi constitutes the dominating fraction of total annual mean BC concentration north and south of 60° (Fig. SI4 b). However, there are important seasonal variations and high-latitude BCi concentration is highest during winter, when the aging is slower due to less efficient production of sulfate. Convective scavenging can be an important loss mechanism during Arctic winter, resulting in a considerable reduction in BC concentrations in the ConvBCi experiment. BCi also dominates above 300 hPa around the tropics."*

What sources and sinks of nitrate are accounted for in the model? Do simulated aerosol size distribution respond to changes in nitrate mass and how is this response parameterized? Is ammonia accounted for in the calculation of the thermodynamic nitrate/HNO3 equilibrium and how is ammonia represented in the model?

The description of EQSAM in Sect. 2.1 has been expanded to include the role and source of ammonia. In response to comment #1 above, a general description of wet and dry deposition in the model is included. As already mentioned, the source of nitric acid is photochemistry. We have modified slightly to clarify and included a reference for further details. A more detailed description of EQSAM is beyond the scope of the study and we refer to further details in the cited literature.

*"Based on the ammonium to sulfate ratio, EQSAM first calculates the preferred state of sulfate. Excess ammonia is available to partition to the aerosol phase, together with gaseous nitric acid, as described in Myhre et al. (2006). Emissions of ammonia are described in Sect. 2.2, while nitric acid is produced through photochemistry as described in Berntsen and Isaksen (1997)."*

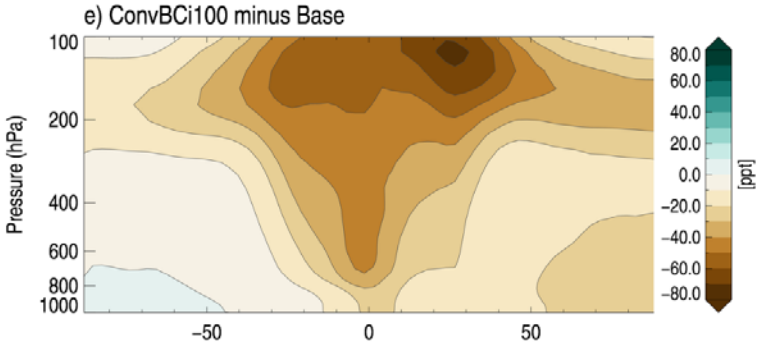
We have also clarified that ammonium-nitrate aerosols are not included in M7 and that nitric acid condensation only contributes to transfer of BC from the insoluble to soluble Aitken mode. The following is added in Sect. 2.3:

*"In this experiment, nitric acid contributes only to the transfer of BC from insoluble to soluble Aitken mode, with no further impact on aerosol size distribution."*

Results from the study seem to differ from results of at least 2 other models in the Arctic (Mahmood et al., 2016). In these models, convective scavenging of BC mainly affects BC in the upper troposphere and lower stratosphere, which seems opposite to responses shown in Fig. 3 and 4. Why are concentrations near the surface sensitive to convective scavenging, which presumably occurs outside the Arctic? This does not seem plausible considering that transport of BC to the Arctic is mainly isentropic. Similar, aging processes are more likely to affect mid and upper tropospheric concentrations of BC in the Arctic than near-surface concentrations. It would be beneficial to change units to ppb in the graphs in order to illustrate impacts of scavenging processes relative to transport processes in the simulations.

As described in response to comment #2 above, the model has a high fraction BC mass residing in the insoluble Aitken mode, in particular in the lower Arctic atmosphere during winter, when convective precipitation connected to cold fronts can be an important loss mechanism. Including convective scavenging of hydrophobic BC therefore gives a large impact on the concentrations in this region.

We agree that changing the units to ppb would give a better picture of the influence of transport and scavenging processes. When doing this, there is a general agreement with the Mahmood et al. (2016) with the largest change occurring at around 200 hPa, thereby illustrating the importance of convective scavenging at higher altitudes. We show this here for our ConvBC experiment.




There is, however, little BC at these altitudes. On the other hand, the changes in the BC mass concentration resulting from the changes to the model's parameterizations are important for the consequent impact on climate. For instance, the climate impact is stronger for near-surface BC, especially in the Arctic. To better be able to illustrate the potential climate impact of changes in our experiments as discussed in later sections of the paper, we therefore consider it beneficial to keep the current unit.

A mean normalized bias (MNB) is introduced but is only applied to a few select simulation results, which limits the usefulness of this metric. In addition, the root mean square error, or similar metric, should be considered, too.

We have added MNB for vertical profiles of CO and replaced the correlation coefficient in Fig. S1 with the RMSE.

The main purpose of including a metric for model performance is to quantify potential improvements in model-measurement comparison in the experiments. We feel that the MNB serves this purpose.

Further comments:

P 2 L 62: Allen and Landuyt (2014) seems relevant in this context, too.

Yes. The reference has been added.

P 4 L 112: Is below-cloud scavenging accounted for?

Below cloud scavenging of BC is not included. This has been specified.

P 5 L 159: How is soluble material formed in the model? Through condensation?

As described in the preceding paragraphs of Section 2.1, the sulfate that contributes to aging of insoluble particles is produced through gas-phase oxidation of $SO_2$ by OH. To clarify we have added:

*"(…) (i.e., sulfate from gas-phase oxidation of $SO_2$) (…)"*

P 7 L 224: 3D kernels are apparently available but global mean concentration profiles are used to calculate radiative forcings. Why? A lack of regional information limits the analysis for the Arctic in the second half of the paper.
In the model simulations used to construct the forcing and temperature kernels, the BC perturbations were applied uniformly throughout a single model layer. Consequently, the temperature response at each grid point will be due to BC forcing exerted both locally and in surrounding grid boxes. At a given grid point, the temperature response cannot be uniquely attributed to the concentration change or forcing in that grid box and this kernel-based approach is less useful for a detailed grid point analysis. In addition, due to the significant altitudinal dependence of forcing and temperature response on the BC perturbation, we are mainly interested in the impact of changes in the vertical here.

For radiative forcing, using the full 3D kernels is justified; here the spatially averaged profiles are used simply for consistency with the temperature response calculations. We have confirmed that the prior averaging of the profiles has a <10% impact on the net RF results presented, except in two cases where differences are approx. 20%. For a fuller discussion on this issue, we refer to Stjern et al. (ACP, 2016).

However, note that we do not only use globally averaged profiles. In additional to quantifying the global mean RF and temperature response to global mean concentration changes, the Arctic changes are estimated using Arctic averaged vertical concentrations. We think the description of the methodology is perhaps a bit unclear here and have tried to clarify further. The description in Sect. 2.4 now reads:

*"Combined with the strong altitudinal dependence of forcing and temperature response on the BC perturbation, we therefore focus on horizontally averaged vertical profiles and not on changes at the grid point level in the present analysis. 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 prior averaging of the profiles has a small impact on the net RF estimates (<10%, except in two cases where the difference compared to using 3D kernels is 20%) (see also Stjern et al. (2016) for a fuller discussion on this issue). We also estimate the Arctic average forcing and response to Arctic BC concentrations changes and briefly investigate the potential uncertainties in Arctic TS using this kernel-based approach due to influence from forcing exerted outside the region. The latter is done by using a kernel for the temperature response caused only by the local Arctic BC perturbation from Flanner (2013) (see Sect. 3.3)."*

P 10 L 307: AMAP models are shown to produce dramatically different seasonal variations in BC burdens in the Arctic (Mahmood et al., 2016). Currently available observational data sets are insufficient to validate seasonal variations in atmospheric BC burdens in the Arctic. Does this statement refer to near-surface concentrations?

The reviewer raises an important point. We have clarified that this statement, as well as the improvements found in several modeling studies, concerns near-surface concentrations, and that considerable inter-model variability remain. We have also included a reference to the Mahmood et al. (2016) paper, which is a very relevant study we missed in the original submission. The revised version reads:

*"While there has been considerable progress and several current models capture the seasonality in high-latitude surface concentrations better than previous generations [Breider et al., 2014; Browse et al., 2012; Liu et al., 2011; Sharma et al., 2013], discrepancies remain and there is considerable inter-model variability in simulated Arctic atmospheric BC burdens [Mahmood et al., 2016]."*

P 11 L 332: Please quantify. What does "somewhat high" mean?

We have added the following based on visual inspection of modeled concentrations in the studies in question:

*"In the present analysis, we find higher modeled concentrations than LB12, on the order of 5-15 ng $g^{-1}$ over large areas north of 70°N, likely owing to the updated emission inventory and shorter model time step for precipitation."*

Fig 3: Include a profile from the control simulation for comparison with concentration differences.
Added to supplementary as part of the response to major comment #2 above.

P 15 L 451: "...in order for models to reproduce the HIPPO data...". Clearly, these profiles cannot be "reproduced" in models but perhaps the general agreement with observations has been improved? Also, this depends on the metric that is used. Typically, BC concentrations near the surface are systematically too low in models and this bias tends to become more severe at reduced lifetimes.

We agree that the wording could be improved here and have rephrased:

*"This is in line with other recent studies showing that modeled BC vertical profiles agree better with HIPPO data, in particular at higher altitudes, when the global BC lifetime is reduced."*

P 18 L 544: Please provide RFari due to BC from all sources from the model. How does it compare to the IPCC estimate? How do the individual values of RFari in Fig. 6 compare to this value?

For comparison with the magnitude of absolute changes in RFari in the sensitivity experiments (Fig.6), we have added the RFari total BC estimated from the baseline simulation using the kernel method:

*"These changes are on the order of 25-50% of the RFari in the baseline simulation, estimated to be 0.35 W $m^{-2}$."*

However, this estimate does not account for the absorption enhancement due to BC aging and is relative to a case with no BC, not relative to pre-industrial, and should therefore not be compared to the IPCC value. We have therefore also added the following paragraph:

*"We emphasize that the RFari from the baseline simulation in the present study should not be compared with estimates of pre-industrial to present forcing by BC as it does not include the absorption enhancement due to BC aging and is given relative to a no-BC situation."*

P 20 L 606: For comparison, please provide the global mean surface temperature response that corresponds to RFari to BC from all sources. How do the temperature responses in Fig. 6 compare to this value?

Presumably, the reviewer means temperature response to ERFari here, corresponding to Fig.6? Given that we use a kernel-based approach to provide estimates of temperature response, and that this does not account for absorption enhancement, we have reservations about presenting a temperature response from the baseline simulation here in the conclusion section. However, to place the absolute changes in Fig.6 in context, we have estimated the baseline TS and added the following to the Section 3.3:

*"(…), resulting in a decrease of -14 to -25 mK compared to the baseline (13-22% of the TS in the baseline simulation estimated using the kernel-based approach)."*

References:

Allen, R. J., and W. Landuyt (2014), The vertical distribution of black carbon in CMIP5 models: Comparison to observations and the importance of convective transport, J. Geophys. Res. Atmos., 119, 4808-4835, doi:10.1002/2014JD021595.

Mahmood, R., K. von Salzen, M. Flanner, M. Sand, J. Langner, H. Wang, and L. Huang (2016), Seasonality of global and Arctic black carbon processes in the Arctic Monitoring and Assessment Programme models, J. Geophys. Res. Atmos., 121, 7100-7116, doi:10.1002/2016JD024849.