

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 review of our paper and the useful suggestions. Responses to individual comments are given below.

Anonymous Referee #1

Received and published: 19 October 2016

In this paper, the authors investigated the sensitivity of black carbon (BC) concentrations in the chemistry-transport model OsloCTM2-M7 to parameters controlling aerosol and scavenging. They especially focused on surface concentrations over the Arctic and vertical profiles over remote regions. Many sensitivity simulations were conducted considering the uncertainties in the coating thickness of sulfate, scavenging by convective and ice precipitation, nitrate formation, and emissions, and the authors showed the importance of the BC ice nucleating efficiency and the change in hygroscopicity with aging.

It is very important to understand the sources of uncertainties in simulating BC concentrations especially over remote regions. So, the theme of this paper is interesting and important. However, I feel there are some fundamental problems in the method (the model representation of aging processes) and the description of this paper, as shown in the major comments below. I suggest the authors to consider these comments carefully because they may be important for the results of this study. The modifications of the model and/or additional sensitivity simulations will be useful to consider these comments.

Major comments:

(1) New findings in this study

What are new findings in this paper scientifically? The authors show the results of many sensitivity simulations, but I think most conclusions obtained from the simulations are already shown by previous studies. For example, previous studies (listed on the references in this paper) showed the uncertainties of BC scavenging by convective precipitation, the poor agreement of BC concentrations over the Pacific (HIPPO) and Arctic (ARCTAS, ARCPAC), the overestimation of BC concentrations at higher altitudes, and sensitivity simulations focused on the aging timescale of BC. Some global aerosol models already consider nitrate formation. Considering these points, I suggest the authors to highlight the important conclusions (new scientific findings) obtained in this study.

The objective of our study is to explore the range of results under varying assumptions in a specific model, how these influence existing model-measurement discrepancies and identify potential improvements that can be implemented before further applications of this model. This is crucial in order to advance BC modelling, e.g. as several recent studies have documented that the current model ensembles do not accurately reproduce measured BC vertical profiles. In the years to come, several new aircraft campaigns are planned. It is of imperative that the modelling groups carefully document the current performance of the global models, before further comparison against new measurements. Furthermore, information about the sensitivity of BC to key processes and parameters may contribute insight to where efforts could be focused in upcoming campaigns in order to provide the best possible data for further constraining global models. Since the global

models differ considerably in their treatment of aerosols aging and scavenging, it is important to examine a broad range of processes in a several models.

However, we also go beyond testing of model performance, to ensure that our results contribute to the growing body of literature on BC modeling. E.g., we focus simultaneously on model capabilities at high latitudes and remote regions over the Pacific, whereas previous studies often focus on one or the other. Additionally, using a microphysical module allows us to investigate parameters beyond those examined in studies using bulk modules (e.g., Hodnebrog et al. (2014)), thereby providing additional information about the importance of underlying processes. Finally, as input to the discussion surrounding the role of BC in the climate system, we also move beyond differences in concentrations and examine the consequent impact on global BC radiative forcing and temperature response.

To better reflect our main objective and the points above, we have changed the title and modified the abstract, introduction and conclusions sections.

(2) BC aging by organic aerosol formation

BC aging processes by organic aerosol (OA) formation will be important because OA mass concentrations are high and are roughly similar to sulfate mass concentrations on global average (at the surface). Considering the concentrations in the atmosphere, OA formation is probably more important than nitrate formation in terms of BC aging by condensation. However, I could not find any description about the BC aging by OA formation. If the OsloCTM2-M7 model does not consider the BC aging by OA formation, the model is insufficient to represent BC aging processes. It is better to improve the model to consider OA formation and the BC aging by OA formation. If it is difficult for the authors to modify the model in a short time, I suggest the authors to add some sensitivity simulations to show the potential uncertainties due to the BC aging by OA formation processes by using the current model.

The M7 accounts for interaction with organic carbon through coagulation, but is so far only limited to primary organic carbon and does not include condensation by secondary organics. While the OsloCTM2-M7 includes a treatments for the gas-aerosol partitioning of secondary organics, a coupling of this module with the M7 require resources and time beyond that is available for this study. Furthermore, the objective of the current study is not to develop a new aerosol parameterization, but to test the range of concentrations and vertical profiles to changes in selected parameters. However, we agree that the potential limitations of not accounting for secondary organics should be made clear and have added the following paragraph:

“In addition to nitrate, condensation of organic aerosols could play an important role in the aging of BC. For instance, He et al. (2016) recently found that a microphysics-based BC aging scheme including condensation of both nitric acid and secondary organics resulted in improved representation of BC in GEOS-Chem compared with HIPPO measurements. This process is currently not included in the OsloCTM2-M7, but should be addressed in future work.”

(3) BC aging by nitrate

Please clarify the treatment of nitrate evaporation. As for sulfate, it is relatively easy because it is enough to consider the conversion from hydrophobic BC to hydrophilic BC. However, as for nitrate, the conversion of both directions will be important. The evaporation of nitrate is especially important over remote regions, and it may be possible to change from hydrophilic BC to hydrophobic BC over the regions through evaporation of nitrate. If the model already considers the effect of nitrate evaporation, please describe about it and show its importance (e.g., as a

sensitivity simulation). If not, please add the effect to the model or please show some results that the effect is not important.

The referee raises an important point. Changes in BC hydrophilicity due to evaporation of nitric acid is not something we have considered in our simulations. In this way, our sensitivity test likely represent an upper estimate of the efficiency of nitric acid in the BC aging process. Furthermore, we find very little literature on the parameterization of this process or its impact. To highlight the uncertainty and limitation in our study, we have added the following paragraph:

“Another important caveat is that we do not account for changes in hydrophilicity resulting from evaporation of nitric acid already condensed on the aerosols. This may result in an overestimation of the contribution from nitric acid to the aging, at least in certain regions.”

Other comments:

(1) Section 3.1.3

I suggest the authors to add a figure showing the results of this section.

Based on the comment by referee #2 regarding the balance of the paper, we have chosen to focus more on the sensitivity studies and somewhat less on the evaluation. We have therefore chosen to include a shortened version of the BC in snow documentation in the section describing the surface concentrations, rather than as a separate section. In light of this, we also do not include additional figures of model evaluation.

(2) Figures 6 and 7

It is hard to see the lines in Figures 6 and 7. Please revise these figures to make them easy to understand.

The figures have been revised with changes to the colors and line thickness.

(3) The number of ML (lines 491-492)

Please describe why the authors use different MLs for sulfate and nitrate.

We realize this description is very unclear. 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 that 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 consider 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.”

(4) Typos

There are some typos in the text. Please correct them.

Line 212: onHoose et al.

Line 581: amont

Line 617: dependen

Typos have been corrected.