

## 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 two anonymous referees for their carefully and thorough reviews of our paper and the useful comments. 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 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 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.

## **Anonymous Referee #2**

Received and published: 2 November 2016

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<sub>2</sub> emissions and increasing NO<sub>x</sub> 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 HNO<sub>3</sub> condensation, please explain why HNO<sub>3</sub> produced in the aq. Chemistry has to be excluded. Is this to estimate gasphase production?

Yes, this is to estimate gas-phase production of HNO<sub>3</sub>. 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 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 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/](http://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 (...)”*

# Sensitivity of black carbon concentrations and climate impact to aging and scavenging in OsloCTM2-M7

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## Abstract

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.

~~Despite recent improvements, significant uncertainties in global modeling of black carbon (BC) aerosols persist, posing important challenges for the design and evaluation of effective climate mitigation strategies targeted at BC emission reductions. Here we investigate the sensitivity of BC concentrations in the chemistry transport model OsloCTM2 with the microphysical aerosol parameterization M7 (OsloCTM2-M7) to parameters controlling aerosol aging and scavenging. We focus on Arctic surface concentrations and remote region BC vertical profiles, and introduce a novel treatment of condensation of nitric acid on BC.~~

~~The OsloCTM2-M7 underestimates annual averaged BC surface concentrations, with a mean normalized bias of -0.55. The seasonal cycle and magnitude of Arctic BC surface concentrations is improved compared to previous OsloCTM2 studies, but model-measurement discrepancies during spring remain. High altitude BC over the Pacific is overestimated compared with measurements from the HIPPO campaigns. We find that a shorter global BC lifetime improves the agreement with HIPPO, in line with other recent studies. Several processes can achieve this, including allowing for convective scavenging of hydrophobic BC and reducing the amount of soluble material required for aging. Simultaneously, the concentrations in the Arctic are reduced, resulting in poorer agreement with measurements in part of the region.~~

~~A first step towards inclusion of aging by nitrate in OsloCTM2-M7 is made by allowing for condensation of nitric acid on BC. This results in a faster aging and reduced lifetime, and in turn to a better agreement with the HIPPO measurements. On the other hand, model-measurement discrepancies in the Arctic are exacerbated. Work to further improve this parameterization is needed.~~

~~The impact on global mean radiative forcing (RF) and surface temperature response (TS) in our experiments is estimated. Compared to the baseline, decreases in global mean direct RF on the order of 10–30% of the total pre-industrial to present BC direct RF is estimated for the experiments that result in the largest changes in BC concentrations.~~

~~We show that globally tuning parameters related to BC aging and scavenging can improve the representation of BC vertical profiles in the OsloCTM2-M7 compared with observations. Our results also show that such improvements can result from changes in several processes and often depend on assumptions about uncertain parameters such as the BC ice nucleating efficiency and the change in hygroscopicity with aging. It is also important to be aware of potential tradeoffs in model performance between different regions. Other important sources of uncertainty, particularly for Arctic BC, such as model resolution has not been investigated here. Our results underline the importance of more observations and experimental data to improve process understanding and thus further constrain models.~~

## 1 Introduction

Black carbon (BC) aerosols play an important role in the climate system through several mechanisms including direct absorption of solar radiation (Bond et al., 2013; Myhre et al., 2013), changing surface albedo (Flanner et al., 2009; Warren & Wiscombe, 1980), modification of cloud properties and thermal stability (Koch & Del Genio, 2010; Lohmann & Feichter, 2005), and influence on precipitation and circulation (Bollasina et al., 2014; Wang et al., 2015). The potentially strong climate warming, combined with short atmospheric residence time and harmful health impacts (Anenberg et al., 2012; Aunan et al., 2006; Shindell et al., 2011), has made BC reductions an attractive mitigation measure (AMAP, 2015; Bowerman et al., 2013; EPA, 2012; Grieshop et al., 2009; Kopp & Mauzerall, 2010; UNEP/WMO, 2011).

~~However, accurately representing the distribution~~Modeling atmospheric concentrations of BC concentrations in global atmospheric models remains challenging and considerable inter-model variability and model-measurement discrepancies exist. In particular, it has been well documented

that the current model ensembles do not accurately reproduce measured BC vertical profiles (e.g., (Koch et al., 2009b; Lee et al., 2013; Samset et al., 2014; Schwarz et al., 2013; Wang et al., 2014)). Additionally, global models often underestimate~~ion of Arctic BC surface concentrations and fail to capture the the magnitude and difficulty capturing the seasonal cycle of Arctic BC surface concentrations~~ (e.g., Eckhardt et al. (2015); Shindell et al. (2008)) ~~and an overestimation of high altitude BC concentrations over remote regions (e.g., (Koch et al., 2009b; Lee et al., 2013; Samset et al., 2014; Schwarz et al., 2013; Wang et al., 2014)).~~ (Bourgeois & Bey, 2011; Browse et al., 2012; Fan et al., 2012; Kipling et al., 2016) Because the radiative forcing (RF) and temperature response to a perturbation in BC impact of the aerosols on radiation and temperature depends strongly on altitude, such discrepancies propagate to lead to uncertainties in the net BC -climate impact ~~of BC~~. For instance ~~While,~~ overestimating high ~~—~~altitude BC concentrations can result in an overestimation of the subsequent BC RF radiative forcing (Samset & Myhre, 2011), while too low surface concentrations may lead to an underestimation of the temperature response due to the reduced efficacy of BC forcing with altitude (Ban-Weiss et al., 2011; Flanner, 2013; Samset & Myhre, 2015). This in turn poses significant challenges for the design and evaluation of effective BC mitigation strategies. Studies have shown that both modeled global vertical BC profiles and the transport of the aerosols to the Arctic is strongly influenced by the parameterization of scavenging and aging (Bourgeois & Bey, 2011; Browse et al., 2012; Fan et al., 2012; Kipling et al., 2016). However, these parameterizations differ considerably between current models. Increasing the understanding of factors controlling the distribution of BC in different global atmospheric and climate models is therefore essential.

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 in the BC aging parameterization. Measurements have shown that nitrate is frequently present in internal aerosol mixtures and contribute to the aging of BC (Pratt & Prather, 2010; Shiraiwa et al., 2007), a process currently excluded in many models. Aging through interaction with nitrate This process may also become more important in the future following strong projected decreases in SO<sub>2</sub> emissions and increasing NO<sub>x</sub> 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 (Bourgeois & Bey, 2011; Browse et al., 2012; Fan et al., 2012); Hødnebrog et al. (2014); Kipling et al. (2016) 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. The model setup and experiments are described in Sect. 2, results presented and discussed in Sect. 3 and conclusions given in Sect. 4

~~Several studies have explored how scavenging processes and uncertainties in emissions contribute to the inter-model and model-measurement discrepancies. Some have investigated how these processes influence transport of BC to the Arctic (Bourgeois & Bey, 2011; Browse et al., 2012; Liu et al., 2011), others have focused on the vertical BC distribution in remote regions (Fan et al., 2012; Hødnebrog et al., 2014; Kipling et al., 2016; Kipling et al., 2013).~~

~~In this study we use the chemical transport model OsloCTM2 (Sovde et al., 2008) with the microphysical aerosol parameterization M7 (Vignati et al., 2004) (hereafter OsloCTM2-M7) to investigate the sensitivity of modeled BC concentrations to changes in a range of parameters related to aging and scavenging processes and how these influence the model-measurement~~

discrepancies, focusing simultaneously on Arctic surface concentrations and remote region vertical distributions of BC.

The OsloCTM2 has been used in several multi-model studies of aerosol impacts (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 parameterization of carbonaceous aerosols. Lund and Berntsen (2012) compared the M7 to this bulk parameterization and found improved representation of modeled BC seasonal cycle and magnitude at high latitudes. However, a comparison against BC vertical profiles measured during one aircraft campaign suggested that M7 exacerbated high-altitude overestimation of concentrations.

Here we use updated inventories of anthropogenic and biomass burning emissions and three years of model results, and further evaluate the OsloCTM2-M7 against a range of observations from surface stations, flight campaigns, and snow samples. Next, we perform sensitivity experiments to quantify the impact of changes in a range of physical and microphysical parameters on the BC distribution. Our sensitivity experiments include a first step towards accounting for gas phase nitric acid condensation in the BC aging parameterization. Measurements have shown that nitrate is frequently present in internal aerosol mixtures and contribute to the aging of BC (Pratt & Prather, 2010; Shiraiwa et al., 2007), a process currently excluded in many models. This process may also become more important in the future following strong projected decreases in SO<sub>2</sub> emissions and increasing NO<sub>x</sub> and greenhouse gas emissions (Bauer et al., 2007; Bellouin et al., 2011; Makkonen et al., 2012). Finally, we estimate the subsequent impact of BC concentrations changes on global radiative forcing and surface temperature response.

Section 2 describes the model setup and experiments, Sect. 3 presents and discusses results and Sect. 4 gives the conclusions.

## 2 Methodology

### 2.1 The OsloCTM2-M7

The OsloCTM2 is a global off-line 3-dimensional chemistry transport model with transport driven by meteorological data generated by the Integrated Forecast System (IFS) model at the European Center for Medium Range Weather Forecast (ECMWF) (Sovde et al., 2008). The model is run for

2008-2010 with a T42 resolution (approximately  $2.8^\circ \times 2.8^\circ$ ) and 60 vertical layers from the surface to 0.1 hPa.

The microphysical aerosol module M7 (Vignati et al., 2004) includes ~~the main aerosol species~~ sea salt, mineral dust, sulfate and organic carbon, in addition to BC. Aerosols are represented by seven modes with size distribution given by a lognormal distribution function. BC ~~aerosols are separated into soluble (mixed) and insoluble particles and can exist~~ in the Aitken soluble (mixed) and insoluble mode, soluble accumulation and soluble coarse modes. All BC aerosols are emitted assumed to be 100% hydrophobic and in the insoluble Aitken mode upon emission. Aging and growth subsequently then occurs due to condensation of sulfuric acid produced in the gas-phase reaction  $\text{OH} + \text{SO}_2 \rightarrow \text{H}_2\text{SO}_4$  ~~and/or~~ coagulation with soluble sulfate particles. See LB12 for additional details. ~~M7 is coupled to the sulfur/oxidant chemistry in the OsloCTM2, i.e., the production of sulfate is explicitly calculated and is dependent on the  $\text{SO}_2$  emissions and oxidant levels and thus variable in time and space.~~

Particles in the soluble modes are assumed to be hygroscopic and are removed according to the fraction of the liquid plus ice water content of a cloud that is removed by precipitation (Berntsen et al., 2006), assuming 100% scavenging efficiency by both water and ice in both large-scale and convective precipitation in the baseline setup. Since LB12 the temporal frequency of wet scavenging in OsloCTM2-M7 has been reduced from three to one hour. ~~The dry deposition velocities for all aerosols depends on particle size and density, turbulence close to surface and the resistance of the laminar sub layer (Grini, 2007).~~ The OsloCTM2-M7 also keeps track of the BC deposition and concentration in snow; see ~~Snow depth and snowfall data from ECMWF is used to build snow layers in the model and BC is dry and wet deposited in these.~~ For detailed description see Appendix A of Skeie et al. (2011) for description.

The sulfate and nitrate modules are described in detail in Berglen et al. (2004) and Myhre et al. (2006), and we only give brief summaries here.

The sulfur cycle chemistry scheme includes dimethyl sulfide (DMS),  $\text{SO}_2$ , sulfate,  $\text{H}_2\text{S}$  and methane sulfonic acid (MSA) and the concentrations of sulfur is calculated interactively with the oxidant chemistry. Sulfate is formed by Ggas-phase and aqueous-phase oxidation of  $\text{SO}_2$  by  $\text{OH}$ , ~~forms sulfate and  $\text{SO}_2$  is also oxidized to aqueous-phase sulfate by  $\text{H}_2\text{O}_2$ ,  $\text{HO}_2\text{NO}_2$  and  $\text{O}_3$ .~~ When

M7 is used, the gas-phase sulfate is saved in a separate tracer and allowed to condense on the insoluble aerosol modes. The aqueous phase sulfate is distributed to the ~~transferred to the~~ accumulation and coarse mode sulfate tracers in M7 according to a prescribed fraction. The treatment of sulfate aerosols then follows M7.

The chemical equilibrium of semi-volatile inorganic species is treated with the Equilibrium Simplified Aerosol model (EQSAM) (Metzger et al., 2002a; Metzger et al., 2002b). EQSAM considers the  $\text{NH}_4^+/\text{Na}^+/\text{SO}_4^{2-}/\text{NO}_3^-/\text{Cl}^-/\text{H}_2\text{O}$  system and calculates the gas/aerosol partitioning of ammonium nitrate under the assumption that aerosols are internally mixed and obey thermodynamic gas/aerosol equilibrium. Nitrate aerosol is represented by two modes; a fine mode comprised of sulfate and a coarse mode comprised of sea salt. After  $\text{H}_2\text{SO}_4$  and nitric acid $\text{HNO}_3$  have been generated by the photochemistry, the thermodynamic equilibrium is solved using EQSAM.

## 2.2 Emissions

Anthropogenic emissions for 2008-2010 are from the ECLIPSEv4 inventory developed with the GAINS model (Amman et al. 2011) as part of the activities under the ECLIPSE project funded by the European Commission 7<sup>th</sup> Framework (Amann et al., 2011; Klimont et al., 2009; Klimont et al., 2016) (available upon request from <http://eclipse.nilu.no/>). Emissions from international shipping and aviation are from the Representative Concentration Pathway (RCP) 6.0 (Fujino et al., 2006; Hijioka et al., 2008). Biomass burning emissions are from the Global Fire Emission Database version 3 (GFEDv3) (van der Werf et al., 2010). Seasonal variability in domestic emissions is accounted for by using monthly weights (2000-2006 average) for each grid based on spatially distributed temperature data from the Climate Research Unit (CRU) following the methodology described in Streets et al. (2003), while seasonality in agricultural waste burning is obtained from GFEDv3. Seasonality of emissions in other sectors is not included in ECLIPSEv4. In the more recently released ECLIPSEv5 inventory (<http://eclipse.nilu.no/>), the monthly variability in emissions from other sectors is small or negligible. Total BC emissions in 2010 are 5866 Gg from fossil fuel plus biofuel sources and 2273 Gg from biomass burning.

## 2.3 Experiments

We first perform a three-year base simulation with meteorological data and emissions for 2008-2010, which forms the basis for the model evaluation. Next, we perform a range of sensitivity experiments described in the following paragraphs and summarized in Table 1.

Several sensitivity experiments are related to the aging of BC. First, we explore the impact of varying the amount of soluble material required to transfer the BC aerosols to the soluble mode. The M7 uses the concept of monolayers (ML); when sufficient soluble material is associated with a hydrophobic particle to form “n” monomolecular layers around the particle, the particle is assumed ~~to be hygroscopic~~ hygroscopic and is transferred to the mixed mode. ~~Currently, n=1 is used based on the best agreement with a sectional model found by Vignati et al. (2004). However,~~ €The amount of soluble material required for a particle to become hygroscopic is an important source of uncertainty (Vignati et al., 2010). Popovicheva et al. (2011) used a laboratory approach to quantify the water uptake by particles with varying amounts of sulfates in order to simulate the aging of combustion particles. Based on a quantification measure for separating hygroscopic and non-hygroscopic soots (Popovicheva et al., 2008), the laboratory results suggest that the transformation of soot particles from hydrophobic to hydrophilic requires an H<sub>2</sub>SO<sub>4</sub> surface coverage of 0.5-1.4 ML, while 1.4-2.3 ML were required for transformation to hygroscopic mode. Based on these results we perform three model simulations where the ML requirement is changed from 1 in the baseline to assuming 0.5, 1.4 and 2.3, respectively ML required for BC aging. Next, we perform a test where 50% of BC from biomass burning sources is emitted directly in the accumulation mode instead of in the insoluble Aitken mode. This is based on observational evidence suggesting that biomass burning BC tends to be larger and more aged, with thicker coatings than BC from urban source (Schwarz et al., 2008). Finally, we test the impact of allowing for BC aging by condensation of gas-phase nitric acid (HNO<sub>3</sub>), first including total HNO<sub>3</sub> and then excluding HNO<sub>3</sub> produced in the aqueous-phase reaction with N<sub>2</sub>O<sub>5</sub>. We extend M7, in a simplified manner, ~~M7~~ to also account for condensation by nitric acid HNO<sub>3</sub> on insoluble particles after gas/aerosol partitioning with ammonium-nitrate is calculated in EQSAM. We follow the same treatment of condensation as for sulfate in M7 (Vignati et al., 2004) and adopt an accommodation coefficient for HNO<sub>3</sub>-nitric acid of 0.1 (Pringle et al., 2010). -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 different runs assuming 1, 5 and 10 are performed where the number of required ML are required for aging, assumed to be one (as for sulfate in the standard M7 case), five or ten. Results presented in Sect. 3 uses ML=5.~~

The second set of sensitivity ~~experimentstests~~ is related to ~~emissions and~~ wet scavenging, the main loss mechanism of BC and ~~thus hence~~ a key parameter for the lifetime and distribution. Hydrophilic BC is originally assumed to be 100% removed by both liquid and ice in large-scale mixed-phase clouds in the OsloCTM2-M7. However, this high efficiency of BC removal by ice-phase precipitation is uncertain. Koch et al. (2009a) found that assuming 12% ice removal of BC gave optimal agreement with observations. This fraction was also supported by observations in Cozic et al. (2007) and has been adopted in studies with the OsloCTM2 bulk aerosol parameterization (e.g., Skeie et al. (2011)). Here we compare results with 100% and 12% removal efficiency for large-scale ice-phase clouds. The removal scheme in OsloCTM2-M7 also assumes no wet scavenging of hydrophobic particles. However, hydrophobic BC aerosol may still be subject to removal by impact scavenging or act as ice nuclei (IN) in convective and mixed-phase clouds (Ekman et al., 2006; Kajino et al., 2012; Park et al., 2005). The BC IN activity is not well known. ~~In order to~~ represent ~~at least~~ some of this uncertainty, we perform two sensitivity ~~experimentstests~~ assuming either 100% or 20% removal efficiency of hydrophobic BC by convective precipitation, with the latter loosely based on Hoose et al. (2010). We also perform a combination ~~experimenttest~~ assuming 12% removal efficiency of hydrophilic BC by large-scale ice-phase clouds and 20% removal of hydrophobic BC by convective precipitation.

Finally, we perform two additional tests to investigate the impact of seasonality in ~~domestic and agricultural waste burning~~ emissions and ~~an increase in higher~~ emissions in Russia following a recent study by Huang et al. (2015). ~~In the first, w~~We alternately remove the seasonal variation in domestic and agricultural waste burning emissions ~~and , while in the second the replace~~ ECLIPSEv4 emissions in Russia ~~are replaced bywith the~~ Huang et al. (2015) inventory. These ~~experiments~~ have limited impact on the global BC distributions, but their influence on the seasonal cycle of Arctic BC concentrations is discussed in Sect. 3.1.1.

## TABLE 1

### *2.4 Radiative forcing and temperature response calculations*

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 radiative forcing (RF) and surface temperature response (TS) kernels— were derived from simulations with the CESM-CAM4, constructed by systematically applying where 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 and 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. was systematically added to each model layer to investigate the climate response to a BC perturbation at a given altitude (Samset & Myhre, 2015). However, Bbecause the BC perturbations were applied uniformly throughout a single model layer, the temperature response at each grid point will be due to is caused partly by the both BC forcing exerted locally and partly by to forcing in surrounding gridboxes. For each experimentIn the present analysis, we therefore focus on global mean vertical profiles. For each experiment, multiply 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. Both direct and semi-direct effects due to aerosol-radiation interactions are included in the kernel response. In line with the nomenclature of the IPCC Fifth Assessment Report, we hereafter refer to the net effect as effective radiative forcing (ERFari) and the direct effect only as RFar<sub>i</sub>.

As discussed by Samset and Myhre (2015), CAM4 does not account for the absorption enhancement due to BC aging, resulting in a lower direct-RFar<sub>i</sub> per BC burden than earlier studies, especially at higher altitudes (e.g., Samset and Myhre (2011)). The consequent temperature response per unit BC may also be underestimated. However, here we focus on the changes from the baseline in each sensitivity experiment rather than absolute climate impacts.

## 2.5 Observations

Modeled concentrations are evaluated against measurements from surface stations, flight campaigns and snow-pack samples.

Measured surface concentrations of BC, sulfate, nitrate, sulfur dioxide and nitric acid across North America are from the IMPROVE and CASTNET networks, while measurements across Europe and the rest of the world are from the EBAS and NOAA GMD databases. We also ~~use~~ compare against measurements in China from Zhang et al. (2012) and ~~from~~ Aerosol Mass Spectrometer (AMS) campaigns summarized in Zhang et al. (2007).

To evaluate the model performance we calculate the correlation coefficient and the mean normalized bias (MNB). The MNB for each species is given by Equation 1:

$$MNB = \frac{1}{N} \sum \left( \frac{C_{mod} - C_{obs}}{C_{obs}} \right) \quad (1)$$

where  $C_{mod}$  and  $C_{obs}$  is modeled and observed concentration and  $N$  is the total number of observations.

Following the recommendations by Petzold et al. (2013) observational data ~~is~~ referred to as equivalent BC (EBC), refractory BC (rBC) or elemental carbon (EC) depending on whether measurements are derived from optical absorption methods, incandescence methods or methods that specify the carbon content in carbonaceous matter. To convert to BC concentrations we adopt a mass-absorption cross-section (MAC) of  $9.7 \text{ m}^2/\text{g}$  (Bond & Bergstrom, 2006), ~~except for all stations except for~~ Alert and Zeppelin, where we use the station-specific MAC reported given in by Lee et al. (2013).

BC in snow is compared to snow sample measurements across the Arctic in 2008/2009 (Doherty et al., 2010) and across Northern China in 2010 and 2012 (Wang et al., 2013; Ye et al., 2012). In the latter case, model results for 2010 are used.

Vertical profiles of modeled BC is compared with measurements from several flight campaigns, including ARCPAC (Aerosol, Radiation, and Cloud Processes affecting Arctic Climate), ARCTAS (Arctic Research of the Composition of the Troposphere from Aircraft and Satellites), HIAPER Pole-to-Pole Observations (HIPPO) and A-FORCE (Aerosol Radiative Forcing in East

Asia). During ARCPAC and ARCTAS, flights were made across Alaska and Canadian Arctic in spring and summer of 2008 (Brock et al., 2011; Jacob et al., 2010), while HIPPO measured atmospheric constituents along transects from approximately pole-to-pole over the Pacific Ocean during different seasons from 2009 to 2011 (Wofsy et al., 2011). The A-FORCE campaign sampled air masses around Japan in March-April 2009 (Oshima et al., 2012). Data from ARCPAC, ARCTAS and HIPPO is available online from [www.esrl.noaa.gov/csd/projects/arcpac/](http://www.esrl.noaa.gov/csd/projects/arcpac/), [www.air.larc.nasa.gov/missions/arctas/arctas.html](http://www.air.larc.nasa.gov/missions/arctas/arctas.html) and [hippo.ornl.gov/](http://hippo.ornl.gov/). Data from A-FORCE was provided by Professor Yutaka Kondo, University of Tokyo (personal communication). Model data is also compared with CO concentrations measured during the campaigns.

Model data is interpolated in time and space and extracted along the flight track. An average profile for each campaign and latitude band is calculated by averaging observations and model results in 100 hPa altitude bins (25 hPa for HIPPO data between 400 and 200 hPa). The HIPPO data is also separated into five latitude bands. To evaluate the model performance in each experiment, we calculate the MNB for each campaign following Eq. 1, where N is determined by the number of altitude and latitude bins.

### 3 Results and discussion

#### 3.1 Model evaluation

~~Before examining the impact of our sensitivity experiments on BC distribution, the baseline We first evaluate the general performance of the OsloCTM2-M7 must be documented. (Lund & Berntsen, 2012)~~ While the main focus of this paper is BC, the evaluation is extended to include species relevant for the BC aging process, including sulfate and sulfur dioxide. We also look at the modeled CO distribution. CO is another product of incomplete combustion and therefore has many of the same emission sources as BC. However, due to the longer lifetime of CO a comparison with observations, in particular in the more remote regions mainly influenced by long-range transport, can give an indication of how well the model represents the atmospheric transport.

##### 3.1.1 Surface concentrations

~~Figure 1 shows annual mean (year 2008) modeled and measured surface concentrations of BC, sulfate, nitrate, sulfur dioxide and nitric acid.~~

#### **FIGURE 1**

~~On annual mean (year 2008), the OsloCTM2-M7 underestimates BC and sulfate surface concentrations in Europe, North America and China with, with an overall MNB of -0.55 (Fig. S1), and -0.45, respectively. The underestimation is largest for measurements in China.~~ **FIGURE 1**

~~The model also underestimates annual mean surface concentrations of sulfate (MNB -0.45), while Nitrate concentrations are in better agreement with measurements, with MNB of 0.08. The model overestimations of sulfur dioxide surface concentrations of sulfur dioxide, especially in Europe, with (MNB of 0.70). This may be due to too inefficient conversion to sulfate, which is supported by the underestimation of sulfate aerosols, and/or an overestimation of emissions. Also nitric acid concentrations in Europe and North America are overestimated (MNB 0.75).~~

~~Figure 1 shows We also investigate the seasonal cycle of BC. Figure 2 shows monthly mean modeled BC and measured EBC surface concentrations averaged over 2008-2010. The model reproduce captures the magnitude relatively well at Mace Head, Cape Point, Trinidad Head, Barrow and Pallas, but fail to capture some of features of in the seasonal variation. Concentrations are also underestimated at Lulin, Hohenpeissenberg and Jungfraujoch during winter and spring.~~

#### **FIGURE 12**

~~Studies have found that Many models often typically struggle to capture the seasonal cycle and magnitude of measured high-latitude BC surface concentrations e.g., (Eckhardt et al., 2015) (Eckhardt et al., 2015; Shindell et al., 2008). While there has been considerable progress and current models capture high-latitude seasonality better than previous generations (Breider et al., 2014; Browse et al., 2012; Liu et al., 2011; Sharma et al., 2013), problems remain. This is also the case for the OsloCTM2-M7. —showed LB12 showed that inclusion of aerosol microphysics significantly improved both magnitude and seasonality of Arctic BC. Here we find further This is further improvements by the use of updated emissions in the current study, partly due to the inclusion of emissions from flaring, which is an important local Arctic source of BC (Stohl et al., 2013). However, the model still underestimates concentrations during spring. The seasonal~~

variability in emissions is an important factor. Accounting for seasonality in domestic BC emissions in the ECLIPSEv4 inventory increases the burden of total fossil fuel plus biofuel BC north of 65°N by approximately 15% during winter and by 2% on annual average compared to assuming constant monthly emissions. Over the same region, including seasonality in agricultural waste burning results in a 2-3% higher total BC burden during spring. This is a relatively small increase, but agricultural waste burning contributes only around 6% to total BC emissions north of 40°N on an annual basis. Another potentially important factor is missing or underestimated emission sources. A recent study by Huang et al. (2015) estimate total anthropogenic BC emissions in Russia of 224 Gg, about 40% higher than in the ECLIPSEv4 inventory. Replacing the ~~Russian BC emissions in the ECLIPSEv4 inventory~~ with those from Huang et al. (2015) increases the modeled BC burden north of 65N by about 16% during fall, winter and early spring and 2-10% during summer. Another possibly underestimated emission source is open waste burning. Wiedinmyer et al. (2014) use year 2010 population data and estimate that 631 Gg BC is emitted globally from open waste burning, nearly a factor 7 more than in the ECLIPSEv4 inventory. Moreover, they suggest that open waste burning may contribute 30-50% to total anthropogenic PM<sub>10</sub> emissions in Russia, from where the near-surface transport of BC to the Arctic is effective (Stohl, 2006). Underestimation of this emission source may thus contribute to the too low modeled Arctic BC concentrations.

The OsloCTM2-M7 underestimates BC concentrations in snow compared to measurements, in particular in Russia, Svalbard and the Canadian Arctic. Here we find somewhat higher modeled concentrations than in previous studies (Lund & Berntsen, 2012; Skeie et al., 2011) owing to the updated emission inventory and shorter model time step for precipitation. However, this increase is ~~not~~ insufficient to ~~fully~~ compensate for the existing underestimation. The model and measurements agree better for many of the snow samples taken in China.

Eckhardt et al. (2015) show that models, including the OsloCTM2, have similar difficulties capturing the ~~sulfate~~ seasonality in ~~the~~ Arctic sulfate concentrations as they have for BC. For instance, At Zeppelin, the OsloCTM2-M7 underestimates ~~also~~ sulfur dioxide during spring at Zeppeling, but overestimates concentrations during summer. Through the aging process, such problems add to the uncertainties in modeled BC.

~~The model captures measured CO concentrations in the Northern Hemisphere during summer (Fig. S2). Figure 3 shows the seasonal cycle of CO for the same stations as in Fig. 2. In the Northern Hemisphere, the model captures the measured concentrations during summer, but underestimates the magnitude during winter/spring, a feature that has been shown also for other models in previous studies (Emmons et al., 2015; Monks et al., 2015). We also compare results at additional Southern Hemisphere locations (not shown here). In the Southern Hemisphere, the model generally reproduces the magnitude better, with a slight overestimation during winter/spring at several stations. The ability of the model to reproduce the seasonal cycle and magnitude of CO, in particular at remote Southern Hemispheric stations that are mainly influenced by long-range transport, suggests that the model represents atmospheric transport reasonably well and points to other processes as the dominant source of uncertainty in the model.~~

### FIGURE 3

#### 3.1.2 Vertical profiles

~~Figures 42 and 5 shows modeled vertical BC and CO profiles against measurements from six aircraft campaigns. Compared to measurements from ARCPAC and ARCTAS spring the OsloCTM2-M7 underestimates the magnitude of BC concentrations throughout the atmosphere (Fig. 24 (p),(r); MNB -0.8). However, the shape of the profile is reproduced reasonably well. During April 2008, when both these campaigns were undertaken, there was unusually strong fire activity in Siberia and air masses were heavily influenced by biomass burning emissions biomass burning plumes, which are often not captured by global models (Brock et al., 2011; Jacob et al., 2010; Warneke et al., 2009). During several flights, the biomass burning plumes were specifically targeted. Possible explanations for the strong discrepancies could therefore be underestimation of the fire emissions or inaccurate representation of the plumes in the model. The same springtime discrepancy was also seen in the surface concentrations. However, the shape of the profile is reproduced reasonably well. The model also underestimates the magnitude of CO concentrations during these two campaigns (Fig. S3 (p,r)), but again captures the profile shape reasonably well, providing further indication that too low emissions could be an important reason for the discrepancy. The agreement is better for ARCTAS summer (Fig. 24 (q); MNB 0.05). 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 suggest a generally better agreement at high latitudes during summer than spring (Sect. 3.1.1)., but the model underestimates near surface concentrations. The model also underestimates the magnitude of CO concentrations during these two campaigns (Fig. 5 (p-r)), but again captures the profile shape reasonably well, providing further indication of too low emissions as an important source of the discrepancy.~~

Measurements from HIPPO are separated into five latitude bands (Fig. 24 (a-o), Fig. 5 (a-o)). For most latitude bands and flights, there is reasonable agreement close to the surface. In the 60-80N latitude band, the model overestimates concentrations close to the surface during HIPPO1 and 2, but underestimates concentrations during HIPPO3. HIPPO3 was undertaken during spring and a similar underestimation was also seen in the modeled surface measurements at Barrow during this time of year (Fig. 12). The model typically fail to reproduce the layered structure of the measured vertical profiles. In particular, the high-altitude concentrations in tropics and the southern mid-latitudes are overestimated. It should be noted that there are substantial differences between the three HIPPO campaigns although they all cover the Pacific. A better model-measurement agreement is found for HIPPO3 than for HIPPO1 and 2 (MNB 1.1, 3.3 and 2.8, respectively). In contrast to BC, both the magnitude and shape of most vertical CO profiles compare well across all latitude bands (Fig. S3 a-o).

There is quite good agreement between measured and modeled BC ~~and CO~~ during the A-FORCE campaign (Fig. 24 (s), Fig. 5 (s);, MNB -0.1), with model results falling within one standard deviation of the measured profile. The A-FORCE campaign was carried out downstream of nearby emission sources and the good agreement with observations suggests reasonable representation in the model of both emission magnitude in the region and the mixing with the free troposphere on timescales of a few days. ~~On these temporal and spatial scales, the loss processes are of less importance for the aerosol distribution.~~ In contrast, the HIPPO campaigns sampled older air masses, ~~where and~~ loss processes have had more time to influence the distribution ~~are more important.~~

Our overall findings are in line with other recent studies. The tendency to overestimate high altitude BC concentrations over the Pacific has been noted for several other model (Kipling et al., 2013; Samset et al., 2014; Schwarz et al., 2013; Wang et al., 2014). The vertical profiles from

OsloCTM2-M7 also fall roughly within the range of the AeroCom Phase II models (Samset et al., 2014).

**FIGURE 24**

**FIGURE 5**

### *3.1.3 BC in snow*

~~The OsloCTM2-M7 underestimates BC concentrations in snow compared to measurements, in particular in Russia, Svalbard and the Canadian Arctic. Here we find somewhat higher modeled concentrations than in previous studies (Lund & Berntsen, 2012; Skeie et al., 2011) owing to the updated emission inventory and shorter model time step for precipitation. However, this increase is not sufficient to fully compensate for the existing underestimation. The model and measurements agree better for many of the snow samples taken in China.~~

~~Towards late spring, the modeled concentrations are occasionally very high compared to the measurement, especially in Tromsø and the Arctic Ocean. This feature was also shown by . During melting, the model assumes that all BC accumulates at the surface. Observational evidence suggest this assumption may lead to an overestimation. For instance, scavenging fractions of 10-30% due to percolation of meltwater were found by from measurements made in Alaska, Greenland and Norway during melt season.~~

### *3.2 Sensitivity of BC concentrations to changes in aging and scavenging*

This section discusses the changes in sensitivity of modeled BC concentrations to the changes in aging and scavenging processes in our experiments, and examines the resulting range in vertical profiles and Arctic surface concentrations.

Table 2 summarizes the global BC burden and lifetime in each experiment. The global mean burden (lifetime) is 133 Gg (6 days) in the base simulation, while there is considerable range from 81 Gg (3.6 days) to about 185 Gg (8 days) across the experiments. The largest changes result from increasing the number of MLs required for aging (CoatThick2.3), allowing convective scavenging

of hydrophobic BC (ConvBCi) and including aging by nitric acid (NitCond). Theis range of BC lifetimes still falls within that of estimates BC lifetimes from different global models (e.g., Samset et al. (2014)).

## **TABLE 2**

## **FIGURE 3**

To examine the impacts in more detail, we calculate the differences in zonal mean concentration (Fig. 3) and burden (Fig. S4) between each experiment and the baseline. The changes follow a similar spatial distribution in most all experiments, with largest changes in the lower model layers north of 60°N and around the equatorial Atlantic. The changes in concentrations north of 60°N are largely determined by changes in the potential for long-range transport of BC from the northern hemisphere source regions with decreasing or increasing lifetime. The pronounced maxima over the equatorial Atlantic coincides with the outflow region with frequent precipitation from areas with high biomass burning activity, where changes in aging rate or lifetime strongly influences the amount that is removed by wet scavenging. In contrast to the other experiments, assuming that more biomass burning BC is emitted directly in the accumulation mode only has a very small effect on concentrations (Fig. 3 d). Asia is also an important BC source region; however, the absolute changes are smaller here than around the Equator. In the coating thickness experiments (Fig. 3 a-c), one possible explanation contributing to the differences could be sulfate levels, with higher concentrations available for coating of the BC particles, and thus lower sensitivity to changes in the aging requirement, in Asia.

Not surprisingly, allowing for convective scavenging of hydrophobic BC results in considerable high-altitude changes, in particular over the tropics where convective activity is strong (Fig. 3 e,f). The maximum change in concentrations is shifted to northern mid-latitudes in the experiments when the large-scale ice scavenging efficiency of hydrophilic BC is reduced (Fig. 3 g,h), with the largest absolute changes over East Asia (Fig. S4 g,h). An increase in this region could reduce the general underestimation of surface concentrations found in the base simulation (Fig. S1). These experiments illustrate that the modeled concentrations are sensitive to the fractions of BC available for scavenging. For convective scavenging of hydrophobic aerosols, we assume either 20% or 100%

to represent a wide range. However, this parameter, and the BC IN efficiency, are uncertain parameters poorly constrained by observations.

The largest changes in concentration results from inclusion of condensation of nitric acid (Fig. 3 i, Fig. S4 i). The number of MLs assumed as the criterion for aging is a key parameter. Different values have been assumed in studies with different aerosol schemes. Here we test the range of results under varying MLs in the same model. The resulting global burden and lifetime is approximately 40% higher in the simulation assuming 10 MLs than with 1 ML. The largest differences are found over South and East Asia (Fig. S5). With no firm agreement on the most correct number to use, we focus on results from the simulation with the middle value of 5 MLs in the following paragraphs.

~~The largest changes in BC concentrations in the sensitivity experiments occur in remote regions and we find only small differences in the model-measurement comparison at the more urban/rural stations in Fig. 2. Next, we examine the impact of the changes in concentrations on the existing model-measurement discrepancies. In the following w~~We therefore focus on the Arctic stations (Alert, Barrow, Pallas and Zeppelin), as well as the vertical profiles from the ~~six~~ aircraft campaigns discussed in Sect. 3.1.2. Figure ~~46~~ shows seasonal Arctic surface concentrations compared to the measurements (left column) and the absolute difference from the base in each experiment (right column). Figure ~~57~~ shows the vertical BC profiles for each campaign and experiment, compared to the baseline and measurements.

**FIGURE ~~46~~**

**FIGURE ~~57~~**

A shorter atmospheric BC lifetime reduces the high-altitude overestimation at mid- and tropical latitudes over the Pacific. This is in line with other recent studies, which have suggested that the lifetime of BC in global models must be reduced in order for the models to reproduce the HIPPO data (Hodnebrog et al., 2014; Samset et al., 2014; Wang et al., 2014). ~~Both allowing for convective scavenging of hydrophobic BC (ConvBCi) and reducing the amount of soluble material required for aging (CoatThick0.5) substantially reduces~~ The MNB for the HIPPO campaigns is substantially reduced in the ConvBCi and CoatThick0.5 experiments compared to the baseline (from approx. 3 to -0.3 and 1, respectively). ~~{Kipling, 2016 #234}~~In most latitude bands, the

~~reduction in MNB is largest for the former of the two. Given that the largest concentration changes in most of the experiments in the present analysis are found over the equatorial Atlantic (Fig. S4), a future comparison of our results against vertical profiles from the ongoing ATom campaign (espo.nasa.gov/home/atom/content/ATom) will be a useful exercise. Our results are supported by {Kipling, 2016 #234}, who also found convective scavenging to be an important parameter for the global vertical BC profile in the HadGEM3-UKCA. For vertical profiles in most latitude bands, the former experiment results in the lowest MNB of the two. However, the model is very sensitive to the fraction of hydrophobic BC assumed to be available for removal (here 100% or 20%), which is an uncertain parameter.~~ Surface concentrations at Alert, Zeppelin, and Pallas are also reproduced reasonably well in these experiments, although the springtime underestimation discussed above remains. In other parts of the Arctic however, the model performance is exacerbated. More specifically, the MNB for the ARCTAS and ARCPAC campaigns increases, and the underestimation of surface concentration at Barrow is larger compared to the baseline. Similar effects are also found in the 60°-70°S region (Fig. 57 (e), (j)). ~~In addition to aging and scavenging,~~ Several other factors ~~not considered here could also likely~~ contribute to the too low modeled Arctic concentrations, including uncertainties in emissions and model resolution. For instance, Aa recently published study point to the importance of model resolution as a source of uncertainty, suggesting that a kilometer-order resolution is required for more accurate representation of BC concentrations in the Arctic (Sato et al., 2016).

Conversely, increasing the amount of soluble material required for aging increases the BC lifetime. This in turn results in an increased potential for long-range transport and increase in Arctic surface concentrations. However, with the exception of Barrow during spring, increasing the number of required ML (CoatThick1.4, CoatThick2.3) does not result in marked improvements in modeled Arctic surface concentrations compared to measurements. The modeled seasonal cycle in Arctic concentrations changes very little in all experiments. The longer aging time in ~~these~~ CoatThick1.4 and CoatThick2.3 experiments also results in a poorer agreement with the HIPPO measurements, both close to the surface and at high altitudes. Moreover, even with the longer lifetime and consequent increases in Arctic BC concentrations, the model does not reproduce the vertical profiles from ARCTAS and ARCPAC. ~~These~~ experiments also result in reduced concentrations of BC in snow in our model, since. ~~In these cases, the aging time is longer and~~ more BC ~~hence~~ resides

in the insoluble mode, unavailable for wet scavenging. Hence, in the OsloCTM2-M7 a slower BC aging alone does not ~~result in significant improvements in any~~ model-measurement discrepancies.

Reducing the scavenging of BC by large-scale ice clouds and increasing the fraction of biomass burning emissions initially in the accumulation mode, have only a minor influence on the comparison with both Arctic surface concentrations and HIPPO modeled vertical profiles compared to the baseline. The smaller impact in the former experiment contrasts the results by {Fan, 2012 #232}, who found a good agreement with HIPPO data when reducing the removal efficiency of hydrophilic BC by snow in the AM3 model. However, Fan et al. (2012) used a more detailed treatment of large-scale ice precipitation and adopted even lower scavenging coefficients than in our analysis. This is also the case for the combined reduction in scavenging by large-scale ice clouds and increased convective scavenging of hydrophobic aerosols.

~~In terms of BC concentrations in snow, smaller improvements are found, but none of the experiments improve the model-measurement comparison of BC in snow simultaneously in all regions.~~

Measurements at mid-latitudes have shown that nitrate is frequently present in internal aerosol mixtures and contribute to the aging of BC (Pratt & Prather, 2010; Shiraiwa et al., 2007). The addition of BC aging by nitric acid in the microphysical BC parameterization is a new development treatment in the OsloCTM2-M7 and results from these experiments are discussed separately here. Allowing for nitric acid to condense on the aerosols results in a faster aging as more soluble material is available ~~than when only sulfate is allowed to contribute~~ and hence reduces the global BC lifetime. This in turn reduces high-altitude BC concentrations and the discrepancies in the HIPPO comparison leads to a better agreement with the HIPPO measurements (MNB between 0.4 and 0.7 for HIPPO1 and HIPPO2, respectively in NitCondv2). Furthermore, BC snow concentrations across all regions except Greenland increase in this experiment, although not enough to fully account for the existing underestimation compared to measurements. However, the Arctic atmospheric BC concentrations are significantly reduced, resulting in a poorer model performance compared to both measured vertical profiles and surface concentrations in this region.

~~The work in this study is a first step and tests the potential importance of accounting for, we have taken a first step towards inclusion of nitrate in the aerosol microphysical aerosol parameterization. There are, however, important limitations. This should however be studied further in future work.~~ For instance, the current setup only treats the condensation by nitric acid, not coagulation with nitrate aerosols. ~~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. Furthermore, it is uncertain how effectively nitric acid increases the hygroscopicity of BC. Here we have assumed 5 ML. In two additional sensitivity tests we also investigate the impact of 1 and 10 ML and find substantial impacts on modeled BC concentrations. Existing model measurement discrepancies in nitrate and sulfate concentrations also contribute to uncertainties.~~ 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.

~~Our analysis in this work, we have~~ does not considered combinations of or regionally differing sensitivity experiments, for instance increased coating thickness required at high-latitudes in combination with more efficient removal by convective precipitation in low and mid-latitudes. Moreover, there are important details that are not captured in the OsloCTM2-M7. One example is related to the particle hydrophilicity/hygroscopicity. The OsloCTM2-M7 assumes that particles can automatically act as cloud condensation nuclei once they are transferred from the hydrophobic to hydrophilic mode. However, the cloud condensating activity of hydrophilic and hygroscopic particles also depends on the atmospheric supersaturation (Koehler et al., 2009; Petters & Kreidenweis, 2007). Furthermore, particles may not merely be hydrophilic or not, as assumed by models, but rather can exhibit a whole range of degrees of hydrophilicity. ~~The ice nucleating efficiency of BC is also relatively poorly known.~~ Our results underline the importance of more observations, in particular of the mixing state and scavenging of BC, as well as experimental data, to improve process understanding.

### 3.3 Climate impacts

As input to the discussion around the role of BC as a climate forcer, the impact of the changes in model parameters on radiative forcing (RF) and surface temperature (TS) is estimated using the kernel-based approach described in Sect. 2.4. The changes in BC concentrations in our experiments can in turn affect the climate impact, especially when changes occur at altitudes where the efficacy of BC forcing and temperature response is strong. Changes in global radiative forcing (RF) and surface temperature (TS) from the baseline in each experiment are estimated using a kernel approach based on results from (see Sect. 2.4) and presented in Table 2.

Figure 6 shows the change in BC ERFari (i.e., net of direct and semi-direct aerosol-radiation interactions), RFari (i.e., direct aerosol effect only) and TS in each experiment. Relative to the baseline, decreases in global-mean BC RFari up to  $-180 \text{ mW m}^{-2}$  are estimated for the two experiments that lead to the most marked improvements (i.e., strongest reduction in MNB) in vertical profiles compared to measurements over the Pacific (ConvBCi and NitCond) (Fig. 6). A notable decrease in RFari of  $-90 \text{ mW m}^{-2}$  is also estimated for the CoatThick0.5 experiment. The Fifth IPCC Assessment Report reports a best estimate of RFari due to BC from all sources of  $0.6 \text{ W m}^{-2}$  (Boucher et al., 2013), while Bond et al. (2013) give a slightly higher estimate of  $0.71 \text{ W m}^{-2}$ . Hence, depending on experiment, the changes in global mean RFari estimated here are on the order of 10 to 30% of the total BC RFari relative to pre-industrial.

Including the semi-direct aerosol impacts partly offsets the RFari. The a decrease in global-mean BC ERFari (i.e., net of direct and semi-direct aerosol-radiation interactions) is of  $-49$ ,  $-45$  and  $-26 \text{ mW m}^{-2}$  in the ConvBCi, NitCond and CoatThick0.5 experiments.  $-mW/m^2$  is estimated for the two experiments that lead to the most marked improvements (i.e., strongest reduction in MNB) in vertical profiles compared to measurements over the Pacific (ConvBCi and NitCondv2). Reducing the amount of sulfate required for BC aging also gives a notable decrease in ERFari of  $-26 \text{ mW/m}^2$ . Changes in ERFari of similar magnitudes but opposite sign are estimated for the CoatThick1.4 and CoatThick2.3 experiments. The change in TS is also largest for three former experiments, resulting in a decrease of  $-14$  to  $-25 \text{ mK}$  compared to the baseline.

Both forcing and temperature response is sensitive to the altitude of BC concentration change. Figure 7 examines the vertical variability behind results in Fig. 6. The globally averaged ERFari (Fig. 7b) peaks below 900 hPa and around 200 hPa, driven by contributions from both the semi-direct and direct radiative effects. The direct radiative effect per BC burden increases with altitude (see also Fig.1 of Samset and Myhre (2015)), resulting in a larger change in RFar<sub>i</sub> at higher altitudes in the present analysis, especially in the ConvBCi and NitCond experiments (Fig. 7a). In contrast, the semi-direct effect per BC burden is positive below 900hPa, but negative and increasing in strength at higher altitudes. Between 800 and 400 hPa the ERFari is smaller due to cancelling direct and semi-direct effects. The net changes in ERFari in Fig. 6 are thus largely determined by an RFar<sub>i</sub> contribution due high altitude concentration changes in our experiments and a low altitude contribution from the semi-direct effect. The TS change is largest in the lower models layers (Fig. 7c), in agreement with the decreasing efficacy of BC forcing with altitude.

~~{Samset, 2015 #218}. In these two experiments, allowing for convective removal of hydrophobic BC and adding condensation by gas phase nitric acid reduces BC concentrations at high altitudes where the forcing efficacy is strong. Reducing the amount of sulfate required for BC aging also gives a notable decrease in ERFari of  $-26 \text{ mW/m}^2$ . Changes in ERFari of similar magnitudes but opposite sign are estimated for the CoatThick1.4 and CoatThick2.3 experiments. The change in surface temperature response is also largest for three former experiments, resulting in a decrease of  $-25 \text{ mK}$  compared to the baseline.~~

~~Since our study also focuses on Arctic BC, we also estimate the changes in Arctic -ERFari and TS caused by the changes in the BC profiles over this region. These are generally larger than the global mean changes. In the CoatThick and NitCond experimentsThe resulting, we estimate 30-50% higher change in ERFari due to Arctic BC concentration changes changes are generally larger compared to than the global mean change in the global-mean case. For all except two cases the Arctic TS changes are also larger than the global-mean changes. This partly reflects that e large the concentration changes are larger in BC concentration the lower model layers at high northern latitudes in these experiments (Fig. 3), combined with a stronger direct radiative efficiency over this region and changes in this region in our experiments and partly a relatively smaller smaller contribution of the semi-direct effect to the ERFari, which acts to offsets less of the RFar<sub>i</sub> than on globally averaged. Surface temperature response per BC burden is also larger for low altitude~~

~~perturbations at high latitudes than globally averaged, and even becomes slightly negative, i.e., a cooling, above 400 hPa north of 70°N. The low altitude concentration changes in The Arctic therefore also results in larger surface temperature response TS changes (by a factor 2-4) compared to the global mean change. to BC forcing exerted in the lower atmosphere, where a substantial impact on BC concentrations is seen in several of the experiments, is also stronger than in lower latitudes.~~

There is, however, an important caveat when using the temperature kernel from Samset and Myhre (2015) to estimate Arctic ~~changes impacts~~. Because ~~the globally uniform~~ BC perturbations ~~were imposed in each at each altitude were applied uniformly in that~~ model layer, the impact on temperature in a specific gridbox may be due both to forcing exerted locally and to remote forcing through large-scale circulation impacts. To exclude any ~~possible~~ influence of BC forcing exerted outside the Arctic region, we also use results from Flanner (2013) to estimate the ~~change in Arctic TS changes. Using the same model as,~~ Flanner (2013) imposed BC perturbations at five different altitudes over the Arctic only, ~~using the same model as Samset and Myhre (2015),~~ hence calculating the Arctic TS ~~caused only to by a local perturbation only local effects~~. The resulting temperature kernel has previously been used to assess the impact of regional on-road diesel BC emissions (Lund et al., 2014). When used here to estimate the impact of our experiments, we find similar changes in Arctic TS to those estimated using results from Samset and Myhre (2015), with ~~some small differences one notable exception~~. In ~~the LSice12 and CombPert three of the experiments, the (EmisTest, LSice12 and CombPert) the two different approaches produce changes in net Arctic TS estimated using {Flanner, 2013 #5} are of opposite sign from results using the kernel from {Samset, 2015 #218}~~. This is caused by ~~slightly~~ different efficacies ~~in the two temperature kernels~~ above 500 hPa ~~altitude~~, where these experiments ~~give the have their largest change effect in on Arctic~~ BC concentrations. ~~For most of the remaining experiments, using the temperature kernel from result in slightly stronger changes in net Arctic TS, reflecting the higher efficacy below 850 hPa compared to the kernel.~~

~~To place the impact of our experiments in context, we calculate the change in direct forcing only (i.e., RFari) and compare with existing best estimates of the total pre-industrial to present BC RFari. The Fifth IPCC Assessment Report reports a best estimate of RFari due to BC from all sources of~~

0.6 W/m<sup>2</sup> (Boucher et al., 2013), while Bond et al. (2013) give a slightly higher estimate of 0.71 W/m<sup>2</sup>. Depending on experiment, the changes estimated here are on the order of 10 to 30% of the total BC R<sub>F</sub> relative to pre-industrial.

#### 4 Summary and conclusions

We have performed a range of experiments to investigate the sensitivity of BC concentrations modeled by the OsloCTM2-M7 to parameters controlling the aerosol scavenging and aging, including, for the first time in the model, a treatment of condensation of nitric acid on BC particles. The impact of changes in these and how these processes influence on the existing model-measurement discrepancies in Arctic surface concentrations and high-altitude concentrations over remote regions of the Pacific is investigated, in order to identify potential improvements to be included in future work. Our analysis is further extended to include an assessment of the effect of the concentration changes on subsequent radiative forcing and surface temperature response.

We find changes of up to 40% in global BC burden and lifetime compared to the baseline, with the largest decreases resulting from inclusion of convective scavenging of hydrophobic BC and aging by nitric acid condensation. In most experiments, the largest changes in concentrations are found in lower model layers north of 60°N and at higher altitudes around the equatorial Atlantic. In the experiments resulting the most pronounced BC concentration changes relative to the baseline, we calculate changes in global-mean R<sub>F</sub> (i.e., direct RF) on the order of 10-30% of the total pre-industrial to present BC direct forcing. However, even with the considerable changes in concentrations, the total impact on global mean surface temperature is estimated to less than 0.04K.

A shorter atmospheric BC lifetime in the OsloCTM2-M7 reduces the high-altitude overestimation at mid- and tropical latitudes over the Pacific and improves the comparison with HIPPO measurement data, providing further support to findings from recent studies (Hodnebrog et al., 2014; Samset et al., 2014; Wang et al., 2014). However, this required shorter lifetime can be achieved through changes in several different model parameters, focusing simultaneously on Arctic surface concentrations and remote vertical distributions of BC. The experiments include a

~~novel treatment of condensation of nitric acid on BC. Furthermore, the subsequent impact of concentration changes on radiative forcing and surface temperature response is estimated.~~

~~The OsloCTM2-M7 underestimates annual averaged BC surface concentrations, with a mean normalized bias (MNB) of -0.55. The model is better able to reproduce the observed seasonal variation and magnitude of Arctic BC surface concentrations compared to previous OsloCTM2 studies, but model-measurement discrepancies remain, particularly during spring. The OsloCTM2-M7 overestimates high altitude BC concentrations over the Pacific compared to measurements from the HIPPO flight campaign, as has been found also for several other global models.~~

~~We find that a shorter atmospheric BC lifetime in the model reduces the high altitude overestimation at mid- and tropical latitudes over the Pacific. This is in line with other recent studies which have suggested that the lifetime of BC in global models must be reduced in order for the models to reproduce the HIPPO data (Hodnebrog et al., 2014; Samset et al., 2014; Wang et al., 2014). Both a Both inclusion of ~~flowing for~~ convective scavenging of hydrophobic BC and reduction in ~~the amount of~~ soluble material required for BC aging results in a 60 to 90 percent lower MNB in the comparison with vertical profiles from HIPPO, relative to the baseline ~~significantly improves (i.e., reduces the MNB) the comparison with vertical profiles from HIPPO compared to the baseline.~~ In the case of convective scavenging, the model is sensitive to the fraction of hydrophobic BC assumed to be available for removal, which is a poorly constrained parameter ~~with large associated uncertainties.~~ The OsloCTM2-M7 is better able to reproduce the observed seasonal variation and magnitude of Arctic BC surface concentrations compared to previous OsloCTM2 studies, although model-measurement discrepancies remain, particularly during spring. ~~While the s~~Surface concentrations at ~~the Arctic stations of~~ Alert, Zeppelin and Pallas remain in reasonable agreement with observations in the two abovementioned ~~former~~ experiments, but the agreement comparison ~~with measurements at Barrow becomes poorer and the ARCTAS and ARCPAC flight campaigns becomes poorer.~~~~

We also find similar improvements in the comparison with HIPPO measurements when including BC aging by condensation of nitric acid. ~~However, the Arctic atmospheric BC concentrations are substantially reduced, resulting in a poorer model performance compared to both measured vertical~~

profiles and surface concentrations in the region. The treatment of BC aging by nitric acid included here is a first step. Further work to resolve uncertainties and incorporate missing processes, such as coagulation with nitrate aerosols and secondary organics, is needed for the development of a more comprehensive aerosol microphysical parameterization in the OsloCTM2-M7.

The existing model-measurement discrepancies in the OsloCTM2-M7 can not be uniquely attributed to uncertainties in a single process or parameter. Furthermore, improvements compared to measurements in one geographical region, can be accompanied by a poorer model performance in other. This underlines the need for better process understanding supported by observational and experimental data, e.g., of BC IN efficiency, aging rate and mixing state, rather than tuning of individual, effective parameters such as global BC lifetime, to further improve models and constrain estimates of BC climate impact. Sensitivity studies may therefore provide important insight ahead of upcoming measurement campaigns regarding where experimental efforts could be focused in order to provide the best possible data for further constraining global models.

~~Conversely, changes in processes that lead to a longer BC lifetime exacerbates the high-altitude overestimation over the Pacific and result in an overestimation of Arctic surface concentrations during winter. Moreover, despite increases compared to the baseline, the BC concentration in snow and during flight campaigns in the Arctic is still underestimated.~~

~~Measurements at mid latitudes have shown that nitrate is frequently present in internal aerosol mixtures and contribute to the aging of BC . In this study, we have taken a first step towards including this process in the OsloCTM2-M7 by allowing for aging of BC by condensation of nitric acid. This results in a faster aging and hence a reduced global lifetime, which in turn reduces high-altitude BC concentrations and leads to a better agreement with the HIPPO measurements. Furthermore, BC snow concentrations across all regions except Greenland increase in this experiment, although not enough to eliminate the underestimation compared to measurements. However, the Arctic atmospheric BC concentrations are substantially reduced, resulting in a poorer model performance compared to both measured vertical profiles and surface concentrations in this region. A number of uncertainties remain, including how effectively nitric acid increases the~~

~~hygroscopicity of BC and how coagulation with nitrate aerosols influence aging, and should be studies further.~~

~~Our experiments result in a non-negligible impact on radiative forcing (RF) and surface temperature (TS). Compared to the baseline, decreases in the global RF<sub>dir</sub> (i.e., direct RF) on the order of 10-30% of the total pre-industrial to present BC direct forcing is estimated for the experiments that result in the largest changes in BC concentrations. Notable decreases in both ERF<sub>dir</sub> (i.e., direct plus semi-direct RF) and TS is also estimated for the experiments which leads to the most marked improvements (i.e., strongest reduction in MNB) in vertical BC profiles compared to measurements over the Pacific.~~

~~While we find that globally tuning parameters related to aging and scavenging can improve the representation of BC in the OsloCTM2-M7 compared to measurements in specific regions, our results also show that such improvements can result from changes in several processes and depend on assumptions about uncertain parameters such as the ice nucleating efficacy of BC and the change in hygroscopicity with aging. Our results underline the importance of more observations and experimental data to improve process understanding and thus further constrain models.~~

~~It is also important to be aware of potential tradeoffs in model performance between different regions. Other important sources of uncertainty, particularly for Arctic BC, such as model resolution has not been investigated here. Our results underline the importance of more observations and experimental data to improve process understanding and thus further constrain models.~~

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## TABLES

Table 1: Summary and description OsloCTM2-M7 experiments performed in this study.

Experiment	Description
Baseline	Standard M7 OsloCTM2 simulation
CoatThick0.5	Required coating thickness reduced to 0.5ML
CoatThick1.4	Required coating thickness increased to 1.4ML
CoatThick2.3	Required coating thickness increased to 2.3ML
EmisTest	50% of biomass burning BC emitted directly in soluble accumulation mode
ConvBCi100	Hydrophobic BC removed by convective precipitation, 100% efficiency
ConvBCi20	Hydrophobic BC removed by convective precipitation, 20% efficiency
LSice12	Scavenging by ice in large-scale precipitation reduced from 100% to 20%
CombPert	LCice12 + ConvBCi20
NitCond	Aging by <u>gas-phase HNO<sub>3</sub>nitric acid</u> condensation included
<del>NitConcV2</del>	<del>As above, but excluding HNO<sub>3</sub> produced by aqueous phase N<sub>2</sub>O<sub>5</sub> reaction</del>
<del>EmisSeasonality</del>	<del>Seasonality in domestic or agricultural waste burning BC emissions removed</del>
<del>EmisBCRUS</del>	<del>BC emissions in Russia replaced by Huang et al. (2015) inventory</del>

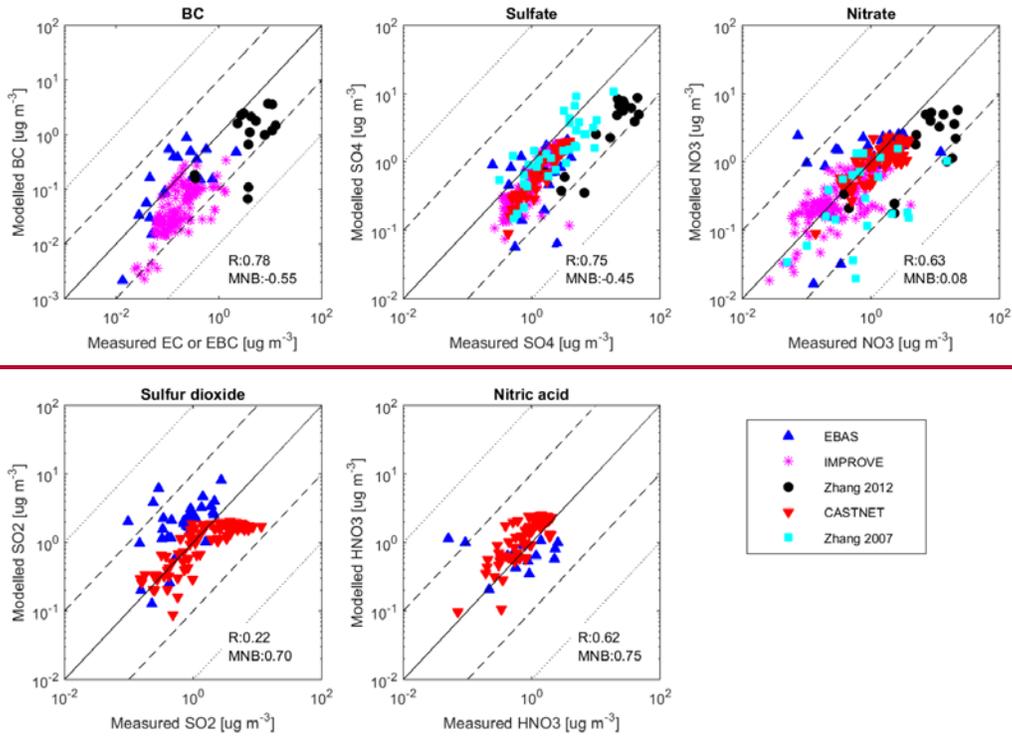
Table 2: *Global BC lifetime and burden, and the change in global mean RF, DRF and surface temperature response from the baseline in each experiment.*

	<b>Lifetime</b> [days]	<b>Burden</b> [Gg]
<b>Base</b>	6.0	133
<b>CoatThick0.5</b>	4.8	106
<b>CoatThick1.4</b>	6.7	150
<b>CoatThick2.3</b>	8.3	185
<b>EmisTest</b>	5.9	131
<b>ConvBCi100</b>	3.6	81
<b>ConvBCi20</b>	4.8	107
<b>LSice12</b>	6.6	147
<b>Combpert</b>	6.6	148
<b>NitCond</b>	3.9	87

**Global**

	<b>Lifetime</b> [days]	<b>Burden</b> [Gg]	<b><math>\Delta</math>ERFari</b> [mW/m <sup>2</sup> ]	<b><math>\Delta</math>RFari</b> [mW/m <sup>2</sup> ]	<b><math>\Delta</math>TS</b> [mK]
<b>Base</b>	6.0	133	-	-	-
<b>CoatThick0.5</b>	4.8	106	-26	-88	-14
<b>CoatThick1.4</b>	6.7	150	18	55	11
<b>CoatThick2.3</b>	8.3	185	52	166	32
<b>EmisTest</b>	5.9	131	-1	-7	-0.3
<b>ConvBCi100</b>	3.6	81	-49	-181	-25
<b>ConvBCi20</b>	4.8	107	-24	-91	-11
<b>LSice12</b>	6.6	147	15	46	8
<b>Combpert</b>	6.6	148	15	49	8
<b>NitCond</b>	4.9	109	-24	-84	-11
<b>NitCondv2</b>	3.9	87	-45	-157	-23

## FIGURES



*Figure 1: Annual mean measured versus modelled BC, sulfate, nitrate, sulfur dioxide and nitric acid surface concentrations across Europe, North America and Asia.*

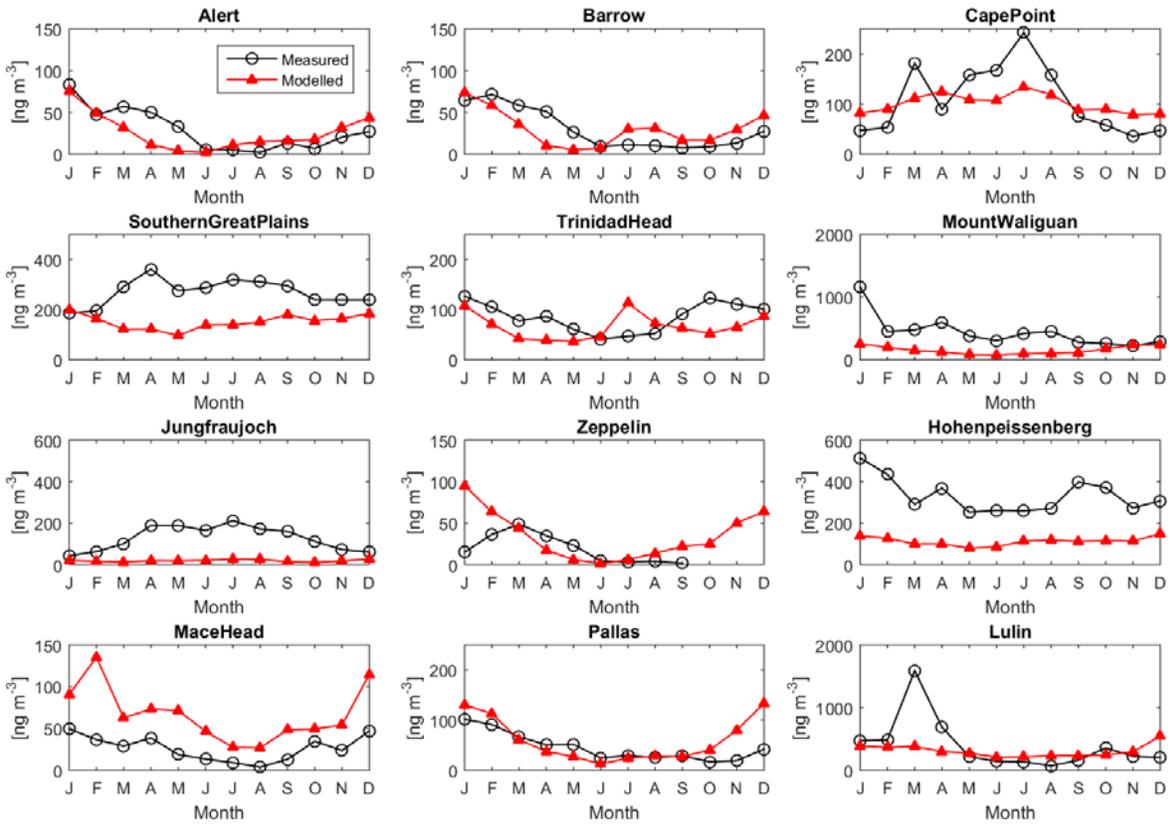
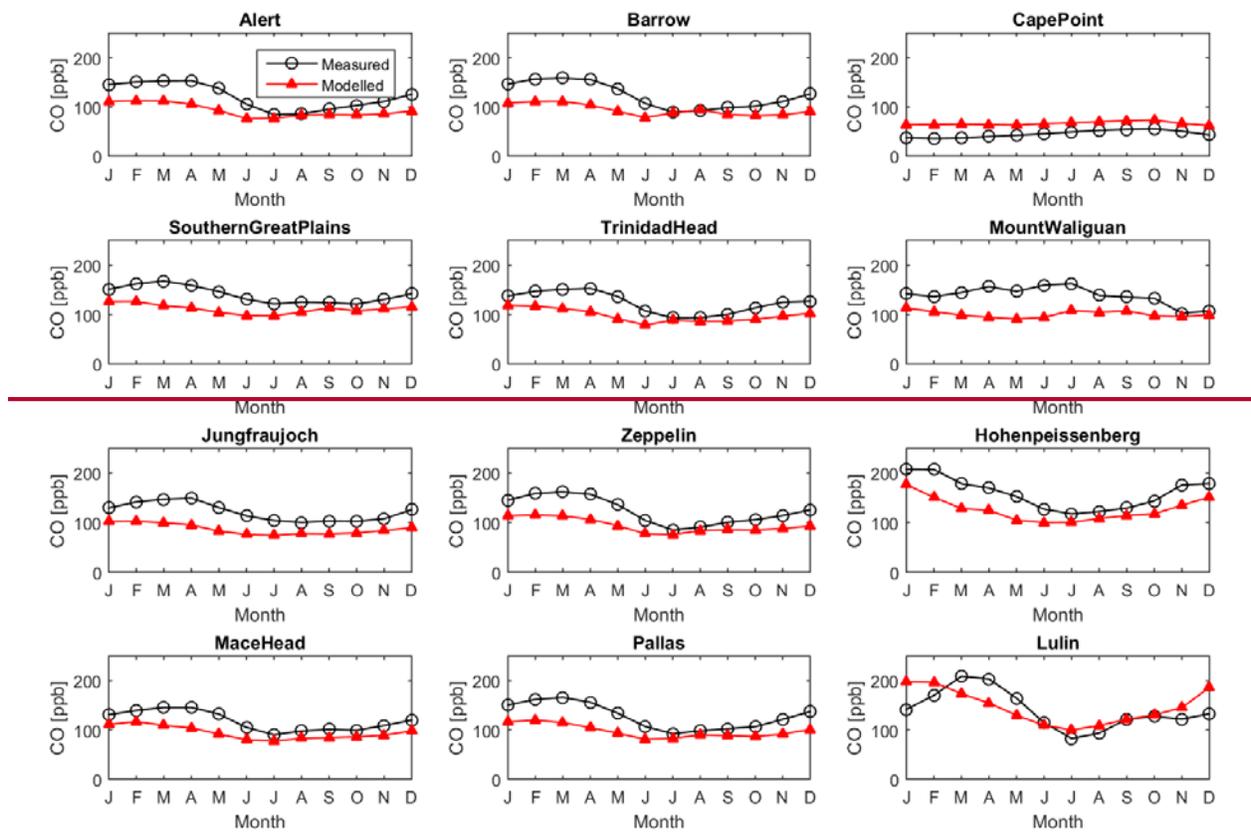
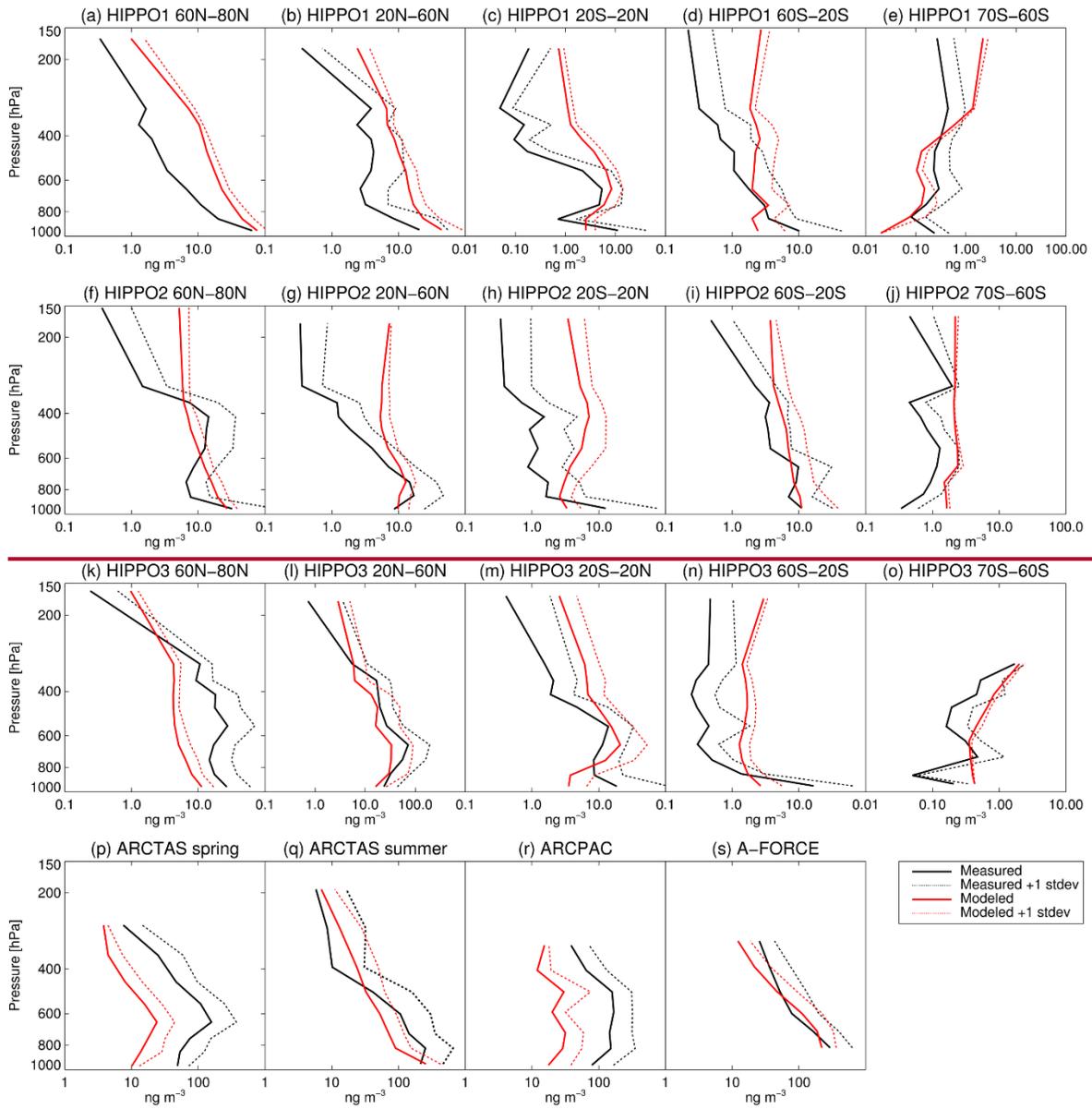


Figure 12: Monthly mean measured EBC versus modelled BC surface concentrations [ $\text{ng/m}^3$ ] averaged over 2008-2010 (data at Lulin only available for 2009-2010).



*Figure 3: Monthly measured and modelled surface CO concentration [ppb] averaged over 2008-2010.*



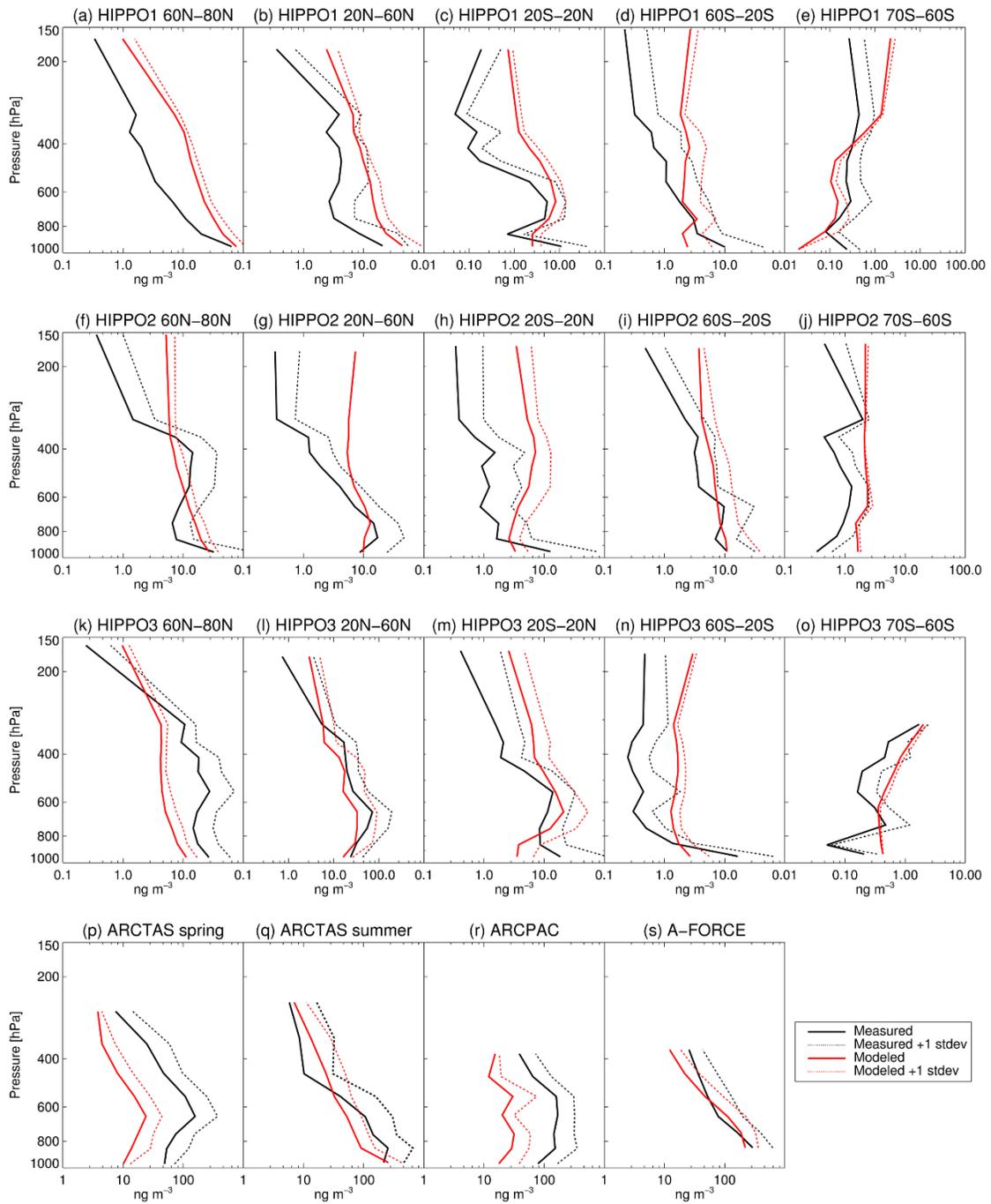


Figure 24: Comparison of modeled vertical profiles of BC with measured rBC from six flight campaigns: (a)-(o) HIPPO 1-3, averaged over five latitude bands, (p)-(q) ARCTAS, spring and summer, (r) ARCPAC and (s) A-FORCE. Solid lines show the average of observations and

model results binned in 100 hPa intervals (25 hPa for HIPPO data between 400 and 200 hPa), while dashed lines show the standard deviation in each interval.

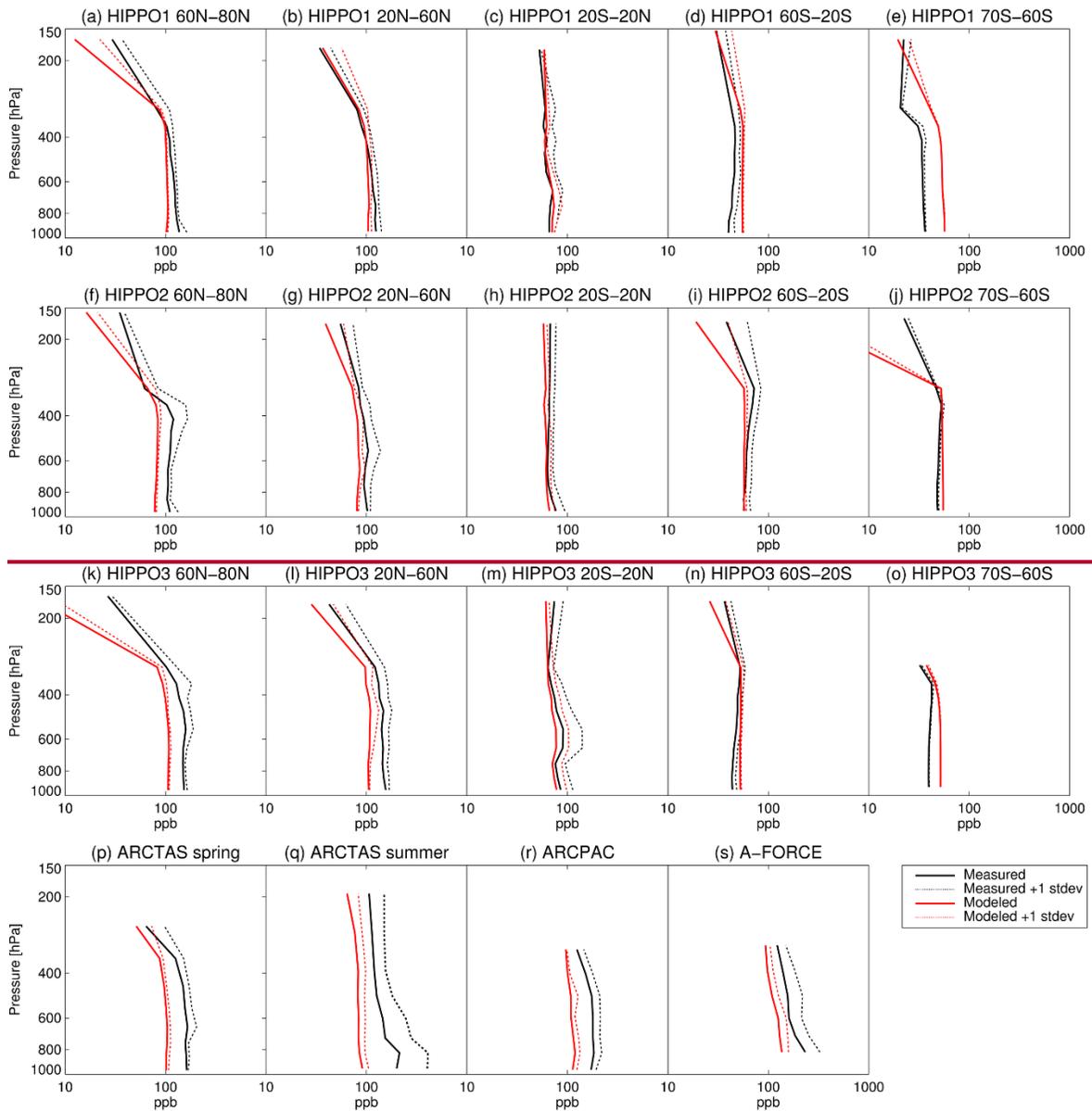


Figure 5: same as Fig. 4, but for CO.

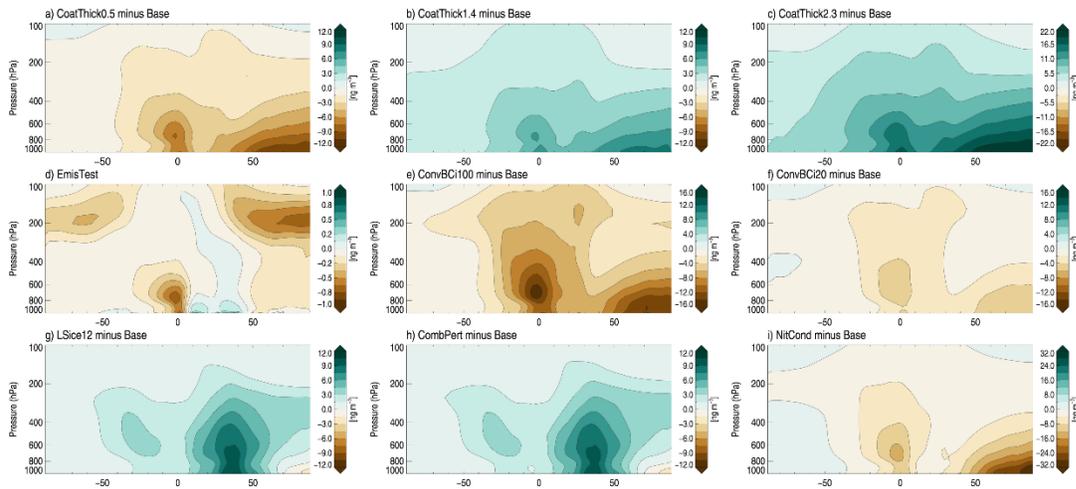
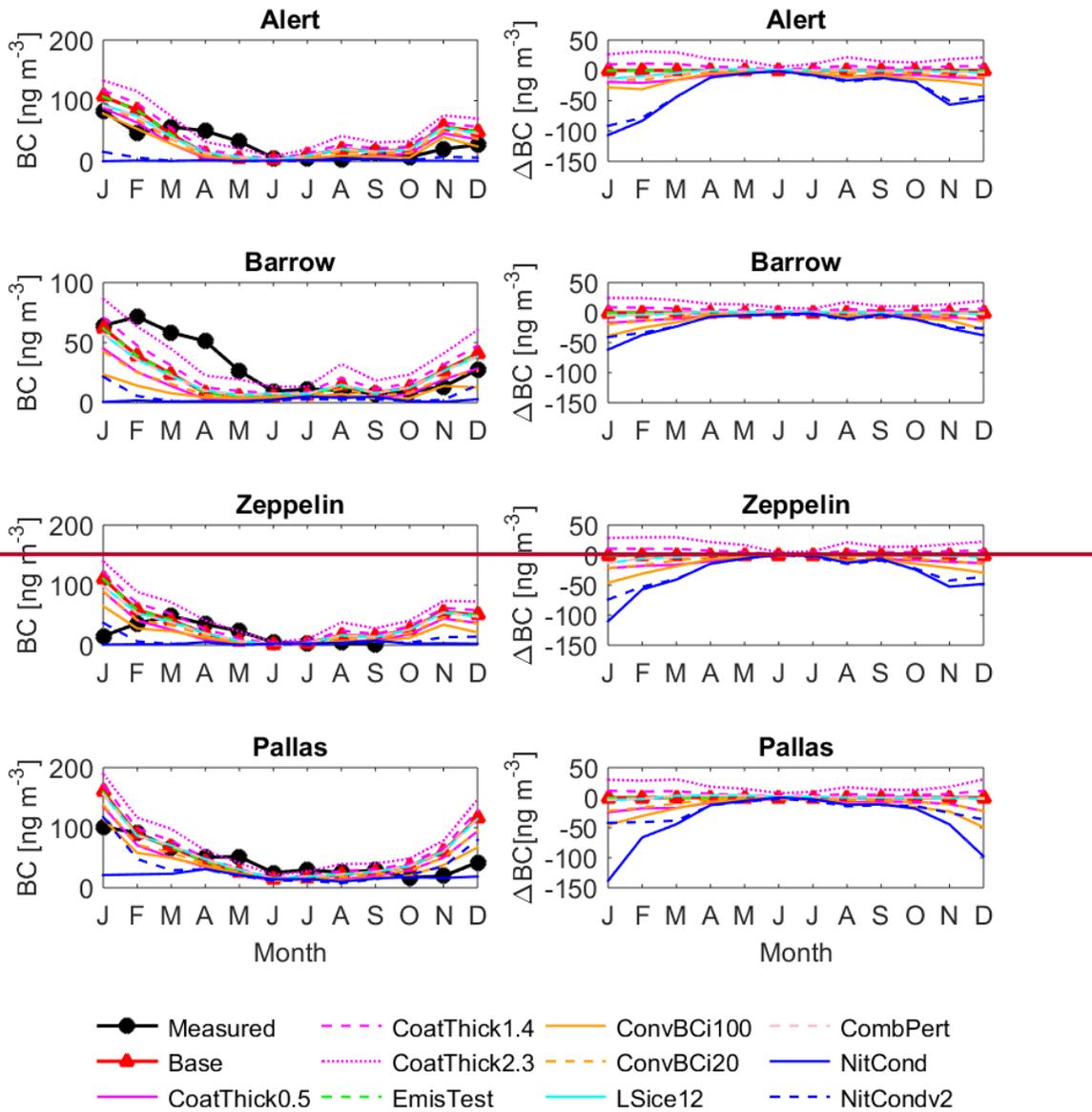


Figure 3: Difference in zonal, annual mean burden [ $\text{ng m}^{-3}$ ] between each sensitivity experiment and the baseline simulation.



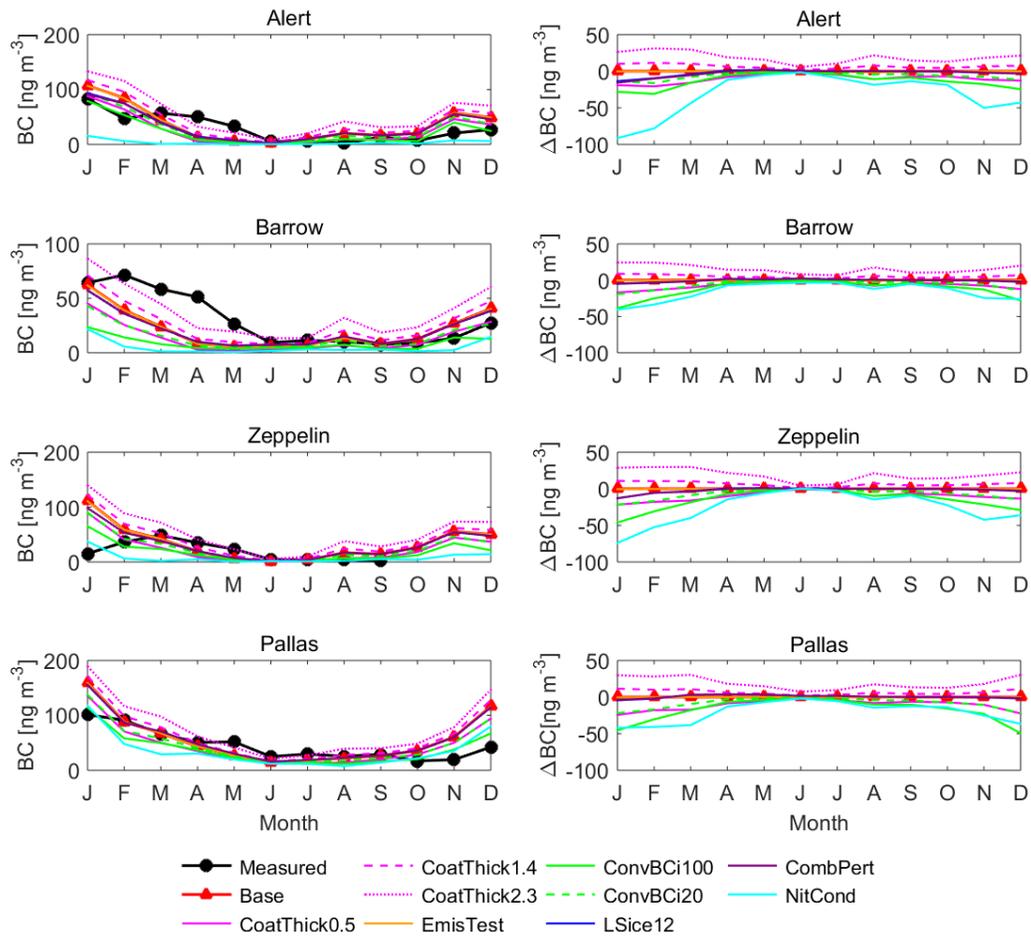
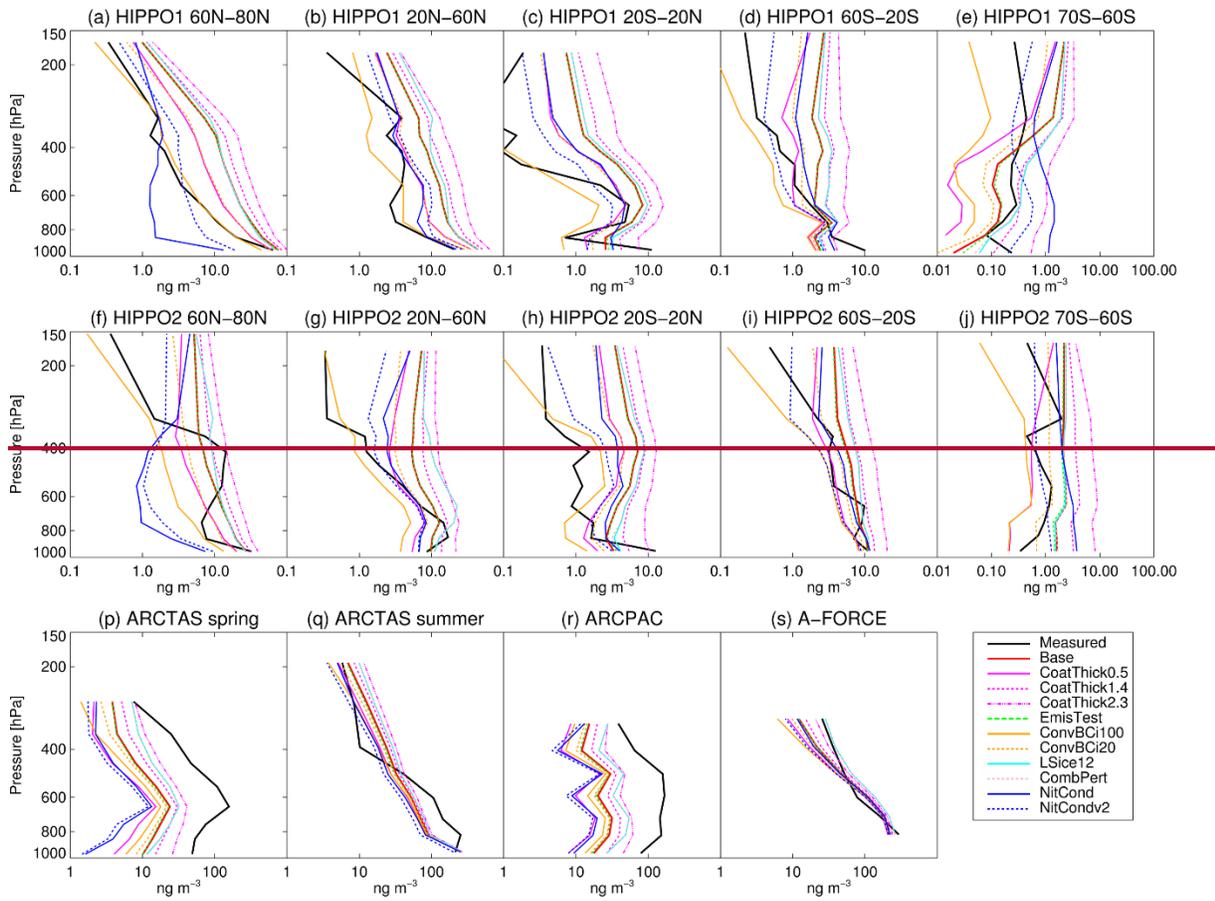


Figure 46: Monthly surface concentrations of BC at Arctic stations in 2008: measurements versus baseline and sensitivity simulations (right column) and difference between each sensitivity simulation and the baseline (left).



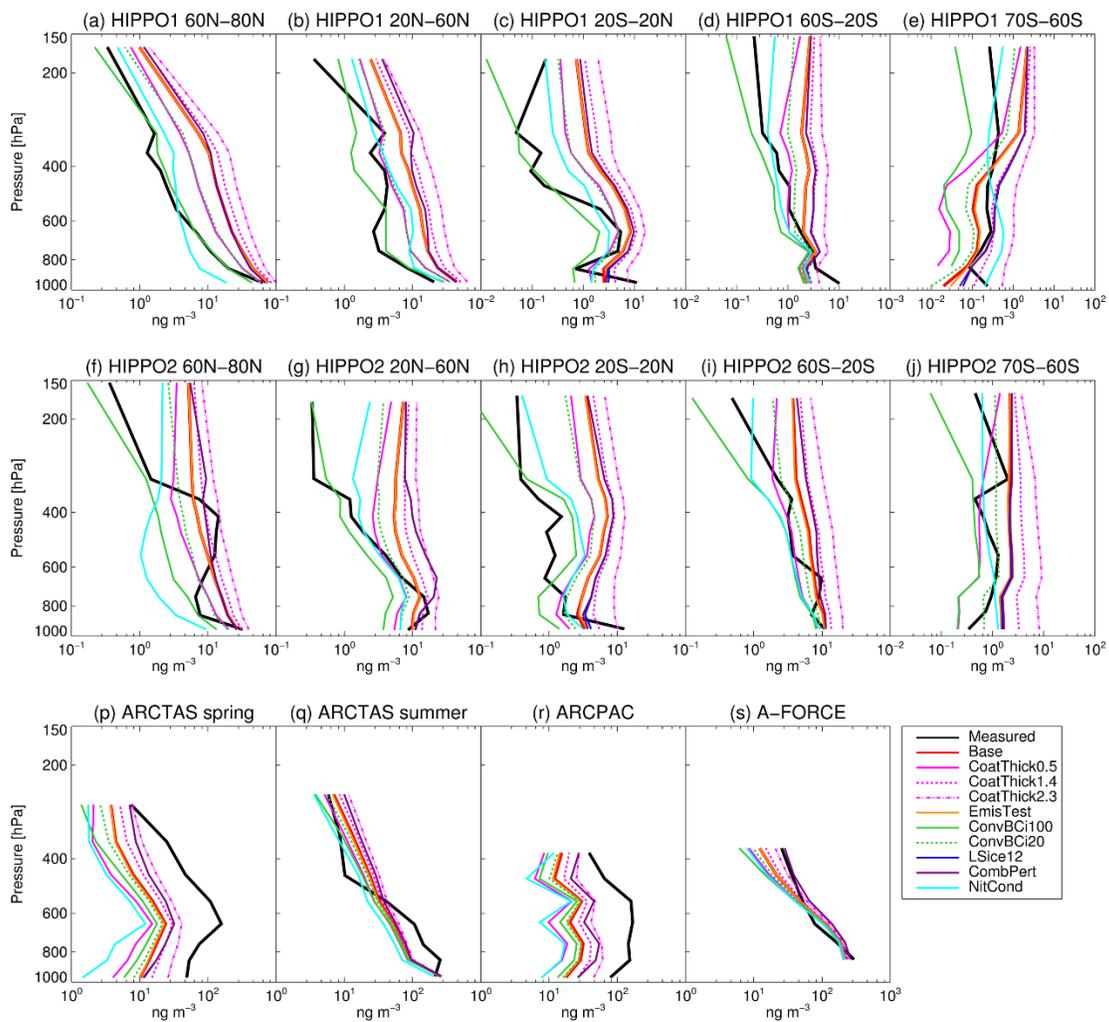


Figure 57: Vertical profiles of BC in the control and sensitivity runs compared to flight campaigns.

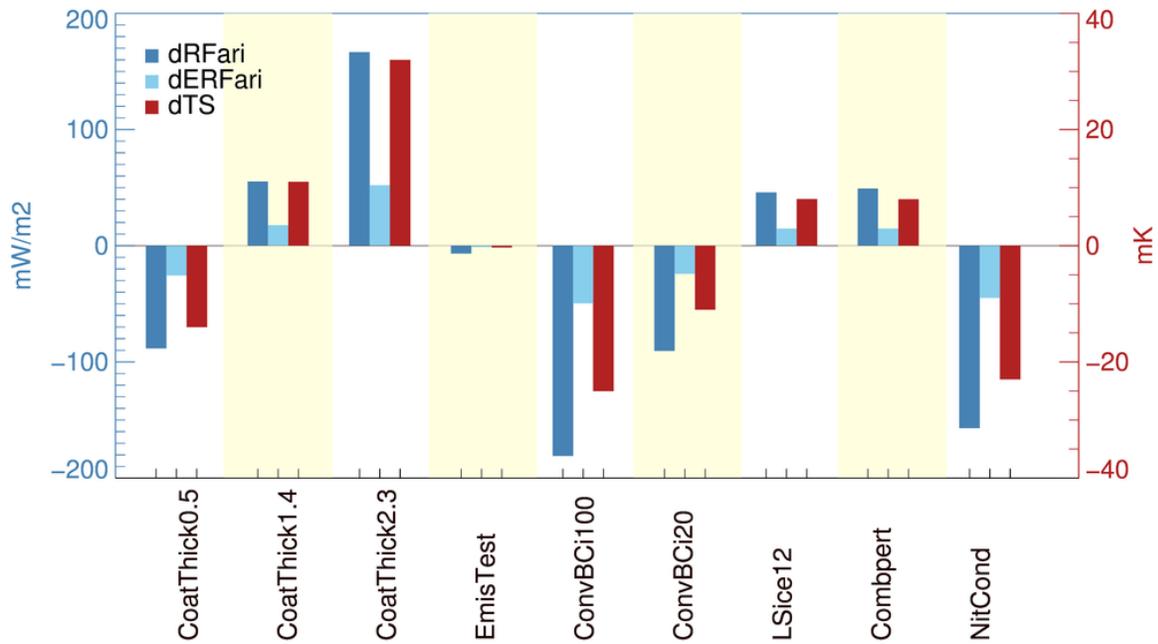


Figure 6: Net change in RFar, ERFar and TS between each sensitivity experiment and the base simulation, estimated using the kernels from {Samset, 2015 #218}

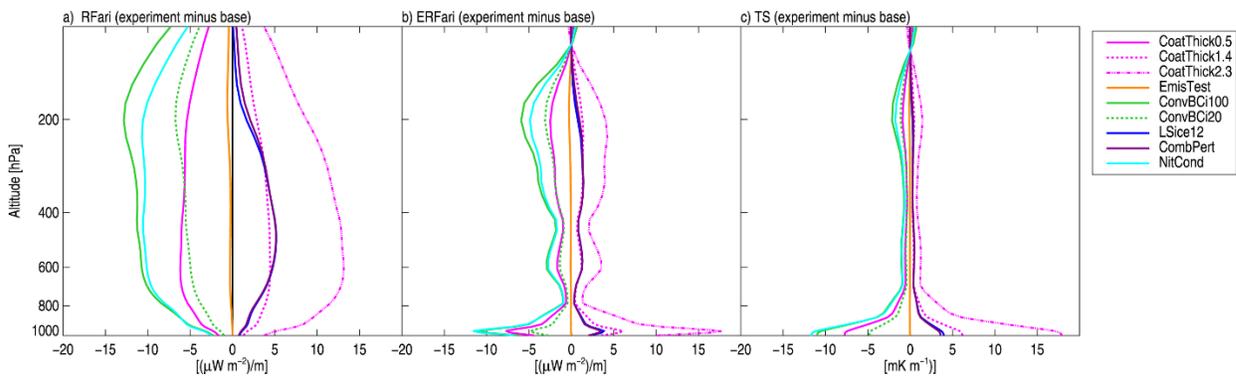


Figure 7: Change in a) RFar, b) ERFar and c) TS between each sensitivity experiment and the base simulation as a function of altitude.