We thank the referees for thorough and interesting suggestions which we feel have helped to improve the paper. Answers to individual comments are given below. This has been a first study looking at M7 in the CTM2 and we have focused on the current version of the module and contrasted it to the original BC parameterization. There are however many aspects of M7 that should be further investigated and possible improvements to be made. We have already started this second step and are planning new simulations with M7 to look more closely at the microphysical processes and scavenging and to compare with more measurements. While we were not able to perform new simulations which could fully answer all referee questions within the framework of this study and for this review, the referees' comments have given us good ideas which we will follow up in the next phase.

Reply to comments by Referee #1:

1. A curiosity of the results presented is that the use of the more sophisticated parameterization (M7) improves the representation of near-surface atmospheric concentrations of BC in the Arctic but it worsens a high bias in modeled concentrations aloft. The finding that modeled values of surface concentrations are too low but concentrations aloft are too high is not new to this paper, but I am wondering if the model studies with BULK and M7 tell us anything about what processes are leading to this result; e.g. does this indicate anything about the relative roles in transport versus deposition in these biases? The paper could do better than to just present the numerical results of the comparison.
   This is an interesting point. The overestimation of high altitude concentrations in the Arctic as well as the underestimation of BC concentration in snow/ice suggest that one reason for the discrepancy could be too inefficient wet removal. The high latitude concentration of BC consists mainly of particles in the insoluble Aitken mode (particularly during winter), which are not removed by precipitation. It is possible however that insoluble BC particles can act as ice nuclei and thus should partly be removed by snow. This is discussed in Section 3.2.1. It is also possible that the high altitude transport of BC from emission sources in e.g. China is too high.
   The same high altitude discrepancy is also seen at lower latitudes; in this case it is possible that the removal by convective precipitation is too inefficient. The following has been added in Section 3.1.1:
   "The high-altitude overestimation compared to measurements may be caused by too inefficient wet removal. There is no removal of particles in the insoluble mode in the CTM2. However, it might be that also insoluble particles can be scavenged in regions of convective precipitation. This will be tested in future simulations. "
   Based on our current results it is not possible to say something quantitative about this. We are however already working on new simulations which among other things will test wet removal of BC in the model.

2. It is noted that the conversion from hydrophobic to hydrophilic in M7 is via condensation of sulfate onto the B C, and it's pointed out a couple of times that in reality measurements have found that organics (usually co-emitted with the BC) may also mix with BC and convert it to hydrophilic. On pg. 32509, lines 5-9 the slow conversion from hydrophobic to hydrophilic in the Arctic winter is noted to be due to the lack of sunlight and therefore slow production of sulfate. I would like to see some discussion here of how accounting for internal mixing with organics might change this, and therefore how it might change the calculated wintertime atmospheric and surface snow concentrations of BC. The implications of accounting for organics could also be expanded upon on page 32513, lines 15-19.
   We have added a discussion of accounting for coating by nitrate and secondary organic aerosol at the end of Section 3.1.2.

3. Section 3.1.2 discussion + Conclusions, pg 32517, line 21 to end of paragraph + Figure 5: The contributions of fossil fuel (FF) and biofuel (BF) emissions from different regions is presented and discussed. It has been observed (e.g. during POLARCAT) that biomass burning may account for a large fraction of Arctic BC, and a lot of the biomass burning emissions may be anthropogenic. I think it is important therefore that in addition to showing the FF and BF contributions to the Arctic from different regions they also to show the regional BB emissions. BB emissions are of course very specific to a given year, but it can be specified whether the year(s) shown are particularly low, high or typical burn years within a given region.
   We have focused on fossil and biofuel emissions in this study and do not have regional contributions from biomass burning available. We agree that it would be interesting to see however, and will do this in future studies. A recent study (Skeie et al. 2011) with the same model and using results from simulations with M7 to improve the aging in the BULK scheme has also looked at the regional contributions from biomass burning. We have added a reference to and description of results from this study. We expect similar change between BULK and M7 for biomass burning emissions. However, since co-emissions of SO_2 *are lower, the changes are expected to be smaller. We have added the following section:*
   "Here we have focused on BC from fossil fuel sources. There are also contributions from biomass burning emissions, of which significant fractions can be of anthropogenic origin. Because the amount of co-emitted SO_2 *is lower from biomass burning than from fossil fuel sources, the changes between BULK and M7 are likely smaller than for fossil fuel BC. The biomass burning emissions also occur mainly during spring and summer, while differences between the parameterizations are most pronounced during fall and winter. Skeie et al. (2011) explore also the contributions from biomass burning BC using the BULK scheme of the CTM2, finding that biomass burning in Russia contributes most to higher-altitude concentrations during spring, while biomass burning in North America is important during summer. The contributions from Europe are mainly of fossil and biofuel origin."*

4. Section 2.2.2, page 32504, line 22: "RF" should be spelled out.
   This has been corrected.

5. Section 3.1.1, page 32508, lines 5-9 and Figure 2c-f: Are the model data averaged over the same grid boxes as the location of the aircraft measurements, or are the model averages over all data in given latitude bands? Please give more detail here on how the model averages were calculated for the comparison.
   Model averages are over all data in the longitude range spanned by the flight track in each latitude band, i.e. for latitude 20S to 20N averages are over all data in longitudes -172.9 to -149.61. This has been specified in the text.

6. Section 3.1.2, page 32512 line 26 to page 32513 line 2: "The contributions from Europe and Russia increase strongly below 5km in January (in M7) compared to BULK. Annual average percentage contributions change only by a few percentage points, to 18% for Europe AND 13% FOR RUSSIA. There is no change in the contribution from China. The contribution FROM RUSSIA INCREASES TO 13% WITH M7." a. It's not clear if you are still talking about column burdens (versus e.g. only below 5km), so it would be good to be explicit. b. If I'm reading this correctly, you are repeating the statement that Russia increases to 13% with M7; i.e. the two bits of text in all caps above.
   a. Clarifications have been added.
   b. This should say "… to 18% for Europe and 7% for North America" and has been corrected.

7. pg. 3253, line 23: “averaged...over the top three modeled snow layers”. Please specify the depth of the top three snow layers (cm?).
   When checking this we discovered that the three top snow layers can be quite thin, so we decided to instead plot the snow concentration in the uppermost 5 cm of the snow. This also makes it more consistent across all regions. We have changed the text correspondingly.

8. Section 4 Conclusions, pg 32516, line 25: “: : :and hence does not lead to significant improvements.” This wording is a bit misleading/odd: Using M7 doesn’t lead to significant or insignificant improvements; it leads to worse agreement between the modeled and measured values. Please reword.
   This has been rephrased to “and hence exacerbates the overestimation.”

Reply to comments by Referee #2:

(1) As described by the authors, aging is an important process determining the rate of soluble materials coating on BC particles. However, the simulated concentration and deposition of BC also depend on how the removal processes (i.e., wet + dry) are parameterized. In section 2, there are some descriptions on BC aging, dry deposition and wet deposition of the bulk method. For the microphysical method, however, only the aging process is described. Details on the parameterizations of dry and wet deposition of BC and other aerosols (e.g. sulfate) in the microphysical method, as well as brief comments on their difference to the bulk scheme are needed.
There is no difference in the representation of wet removal processes between the two parameterizations of aerosols; this has now been specified in the text. A brief description of dry deposition in M7 was already included in the text and we have expanded this.

(2) On Page 32505 “Aging then occurs due to condensation of sulfuric acid produced in the gas-phase reaction $OH+SO_2 \rightarrow H_2SO_4$ or coagulation with sulfate particles : : :” I’d like to know which process (condensation or coagulation) is more important for BC aging (usually both BC and sulfate concentrations are enhanced over the Arctic during the haze period).
This is a very interesting point. With our current simulations it is not possible to obtain this information. However, we are planning a new study to further explore and improve the microphysical module and will certainly try to include such information in the output from these new simulations.

(3) Is OH simulated online or offline?
OH is simulated online using an updated version of the chemistry scheme in the CTM2 (Berntsen & Isaksen 1997). The chemical scheme is solved using the quasi steady state approximation, with iterations to improve the accuracy. A reference to the chemistry scheme has been added.

(4) The authors should state how aqueous oxidation of SO_2 is treated in the bulk and M7 schemes. Some previous studies found sulfate aerosols are mainly oxidized in the liquid phase. It would be nice to have a discussion on the effects of aqueous-phase chemistry on BC aging and then wet deposition.
In simulations with the bulk aerosol parameterization, the CTM2 is run without the sulfate module and the aging of black (and organic) carbon aerosols is calculated using a fixed time constant. To clarify we have added the following sentence:

"[…] Hence the model is run with only the BC/OC aerosol application […]".

In the sulfur module of the CTM2, aqueous phase sulfate is produced from oxidation of SO_2 by H_2O_2, HO_2NO_2 and O_3. A heterogeneous scheme by Jonson&Isaksen (1993) and Jonson et al. (2000) is included to simulate the aqueous chemistry. For application of M7, a new tracer SO4gas is introduced to keep track of sulfate in gas phase. This is the only sulfate used as input to M7. We have added the following to the description of M7:
"The chemistry scheme used in the CTM2 when M7 is applied calculates gas and aqueous phase sulfate. Only the gas-phase sulfate is input to the M7. Nucleation and condensation then proceeds as described in Vignati et al. (2004)."

(5) The aging rate of BC in M7 depends on both SO2 and OH concentrations, which are variable by locations. It would be nice to add a figure which shows the distribution of BC aging time based on the M7 parameterization.
This is an interesting suggestion. However, we do not have the required transfer rates from insoluble to soluble mode as output from the current simulations. For the calculations of regional aging times we use the burden of insoluble Aitken mode particles and total BC emissions. This approach is correct on the global scale and can be used as an approximation on the regional scale, but not for individual grid points due to the transport of species.

(6) In the bulk simulation, dry deposition velocity is set to 0.025cm/sec, which seems to be too small according to both measurements and previous modeling studies. As a result, drydep will contribute little to BC deposition and wetdep will be the dominant term. This could indirectly overstate the role of aging. The authors may want to provide the reference of these values. In addition, in the microphysical method, drydep is based on Grini 2007. I wonder how this would be different to the bulk method.
A reference to Cooke et al. (1999) for dry deposition velocities used in BULK has been added.
This is an interesting point and it would indeed be interesting to look at the actual numerical differences between the constant dry deposition velocities used in BULK and the velocities calculated in M7 (which depend on particle size and density, turbulence and resistance of the laminar sublayer). The currently available simulations do not provide this information. However, we are already preparing for new simulations where we will investigate possible improvements in the wet removal parameterizations and in the M7. As a part of this we will also look more closely at the contributions of dry and wet deposition and differences between BULK and M7.

(7) As shown in Figure 4, the observed BC concentrations at Alert, Barrow and Zeppelin are highest during February to April (i.e., the haze period), which seems different to the seasonality of oxidation of SO2, a process determined by the availability of oxidants which are lowest in DJF and highest in JJA in the northern hemisphere. This may partly explain why the simulated BC with improved aging is usually highest in winter. The authors may want to have a discussion on this and differentiate the role of aging and treatment of other processes on the Arctic BC concentrations.
We have rewritten and added: "The oxidation of SO_2 depends mainly on availability of oxidants, which is lowest during winter. This leads to the largest differences between BULK and M7 during winter. During spring, measured concentrations are likely to be influenced by emissions from biomass burning, which might not be properly captured

by the model emission inventory. For instance, agricultural burning in Eastern Europe caused record high air pollution episodes in the European Arctic in spring 2006 (Stohl et al., 2007) as can be seen in from the Zeppelin measurements in Fig. 4."

(8) A number of previous studies have reported the global budget of deposition flux (i.e., drydep and wetdep). This study has quantified the atmospheric burden and lifetime of BC aerosols in Section 3.1.1. It would be also useful to quantify the global budget of dry and wet deposition, and compare them to the previous work.
This is a good and interesting point. The deposition fluxes are not provided by the simulations forming the basis for this paper. However, we are already planning new simulations where wet deposition of BC will be explored in more detail (see also reply to comment (6)).

(9) There are always some difficulties to evaluate model results with aircraft measurements, which need to match the exact conditions of the sampling period, including emissions (particularly the biomass burning emissions) and meteorology. The simulation period of this study is 2006 and the measurements were made (as a few snapshots) in 2009. The authors should be careful about those differences.
We have added:
"There are important differences between measurements and model results in terms of e.g. meteorological conditions, emissions (such as episodic biomass burning not captured by the model), averaging area and temporal resolution. These should be kept in mind as they make direct comparison difficult."

(10) It would be good to evaluate the model with some surface measurements in the Southern Hemisphere.
Because little information on aerosol absorption coefficient measurements were available in the NOAA/ESRL/GCM or NILU EBAS data bases we have written to the responsible people at several stations. In the time available for this review we have received data for Amsterdam Island, Neumayer and Cape Point and have added a comparison of model results to measurements at these stations.

(11) In Fig. 6a, are these high spots over the high-latitude regions the observed values?
No, there are no observations in this figure.