

We appreciate the useful comments by both reviewers. The original reviewer's comments are in italics.

Referee # 1

The paper discusses the historical evolution of black carbon (BC) aerosols and their associated direct and snow-albedo forcings. This is an interesting study, with section 3.5 on regional contributions to Arctic BC and the discussion in section 4 being particularly informative. I suggest some minor revisions below in order to make some points clearer and the wording more accurate in places.

1 Main comments

- Latitudinal and seasonal ageing coefficients are taken from an M7 version of the CTM. Is there any reason why that model version was not used for the study and the authors chose to parameterise ageing instead? Also, as discussed in section 4, it is likely that ageing coefficients have varied historically. I would be interesting to discuss how results can be affected by that variation: if pre-industrial lifetimes were longer, would that mean that BC radiative forcing were stronger than modelled here?

Response: The M7 version of the Oslo CTM2 is much more computationally expensive since full tropospheric chemistry calculations are needed. It takes approximately 10 days for one time slice simulations with the M7, compared to a day for the model version used in this paper. We have included a sentence on page 7474 line 28: "Due to computing time limitations we were not able to do all the simulations with the coupled model system."

We wrote in section 4: "Stier et al. (2006) found that the atmospheric lifetime of BC was reduced from 4.3 days in the pre-industrial atmosphere to 3.8 days for 2000 conditions". Not only the RF in year 2000 versus pre-industrial are interesting, but also the time development with largest SO₂ emissions in Europe and North America around 1980 (Smith et al., 2011). It is not clear how the geographical changes in the SO₂ emissions will affect the RF time series, and modelling studies with microphysics is necessary to quantify this effect, as we suggest in the conclusion section page 7500 line 18.

- What is the residence time of BC in snow, from deposition to removal when snow melts?

Response: In the model there is no pre defined residence time of BC in snow. The BC is deposited to the snow and contained in the snow until the whole snow deck has melted. The residence time can be from 3 hours to the whole snow season depending on the lifetime of the snowpack.

- Figures 1 and 9 provide important informations but a timeseries of zonal means is a lot to put on one figure. It could be clearer to present timeseries of global averages on one panel, with the corresponding zonal means for selected years on another panel. Selected years could be limited to 1750, 1970, and 2000 since those years incorporate the more important changes in magnitude and location.

Response: We agree that it is too much information in the panels. We have kept the years 1750, 1850, 1910, 1950 and every 10th year until 2000. We have added two more panels in Fig. 1 and 9 with global averages. For best comparability, we want to have the same number of years for the burden plot and the emission plot. From the discussion in the text it is important to include the years 1960, 1990 and 2000. We think including every 10th year since 1950 is useful for presenting the southward shift in the global burden. We think the figures are clear by doing these changes.

- Figures 12: I understand that the inter-annual variability shown in 12b is used to obtain the error bars in 12a. I also understand (although it is not clear from the text) that since 2006 is associated with the largest values (12b), then all model simulations done with meteorology for the year 2006 (12a) are assumed to be high estimates, with error bars applied downward. It is a strange assumption: using meteorology for 1977 could have lead to even higher values, for example. So you can't assume that using meteorology for 2006 will always lead to the highest values. Your measure of year-to year variability (error bars) should be centered on 2006 values. The same comment is valid for figure 15b.

Response: We do not have meteorological input data for the Oslo CTM2 going longer back in time. Therefore it is problematic to state from our modelling more about the interannual variability, than the information we have from the 2001 to 2008 simulations. We agree that we should state clearer in the text our assumptions for the error bar in Fig 12a. However, we disagree that the error bars should be centred on the 2006 values, since 2006 was a year with high concentrations. We have now added an error bar based on 2 times the standard deviation around the mean of the modelled concentrations for the year 2001-2008, and scaled it to the historical concentrations. On page 7488 line 29 we have rephrased:

“Uncertainty bars based on the 2001-2008 variability are added to Fig. 13a, however there might have been long term changes in the circulations (e.g. connected to the Atlantic Multidecadal Oscillation) that could systematically affect the modelled concentrations.”

This is discussed in section 4. In the figure legend we have included:

“Error bars indicate two standard deviation around the mean of the modelled concentrations for the year 2001-2008 scaled to the historical concentrations assuming that the relative variability is constant in time.”

Regarding the snow albedo effect in Fig. 15b, we also do not have information about interannual variability for a longer time period than 2001-2008. We have included a short discussion regarding changes in snow cover over the historical time period, e.g. larger snow cover historically will give the possibility of a larger snow albedo forcing on page 7494 line 2: “It should be noted that any changes in the historical snow cover, e.g. larger snow cover and a longer snow season historically compared to 2005-2006, may have reduced the RF of BC in snow during more recent years.”

2 Other comments

Page 7470, line 23: Forest fires can be anthropogenic as well - in fact, in some countries wildfires are mostly man-made. I suggest giving "lightning-induced forest fires" as an example of purely natural biomass burning.

Response: We have replaced "forest fires" with "lightning-induced forest fires" on page 7470 line 23.

Page 7470, line 25: Black carbon is known for its large absorption, but around 40% of its extinction remains due to scattering. The direct effect is due to both scattering and absorption.

Response: The direct aerosol effect of BC is clearly dominated by the absorption. The critical single scattering albedo for warming of aerosols is around 0.85 in global calculations and BC has a single scattering albedo of around 0.25 (see discussions in Bond and Bergstrom (2006) and Bond et al. (2006)).

Page 7471, line 25: Be more specific than "recently". Page 7473, line 28, and page 7487, line 18: "real time" has a different meaning than used here, typically referring to operational systems where data is processed shortly after being produced. I suggest "meteorological data for the years simulated".

Response: We have replaced "recently" on page 7471, line 25 with: "over the three last decades".

We have replaced "real time meteorological data" with "meteorological data for the years simulated" as suggested on page 7473 line 28 and we have rephrased on page 7487 line 18: "using the results from the 2001-2008 simulations"

Page 7474, line 21: I would think that other species, such as volatile organic compounds, also play a role in BC coating.

Response: Yes, we have rephrased the sentence:

Page 7474 line 21: "This is a simplification since the aging depends on coagulation with soluble aerosols and condensation of organics and inorganic species."

Page 7477, line 14: Lamarque et al. [2010] provide both biomass and fossil-fuel emissions of BC and OC, yet only their biomass emissions are used here, with FF emissions coming from another dataset. What is the reason behind that choice?

Response: At the time of our simulations were made, the Lamarque et al. paper was not published, but data were available, and we chose to use the data published in Bond et al. (2007) for the fossil fuel and biofuel emissions.

Page 7477, line 25: Different spatial distributions? What are the main differences? Could those differences be due to the industrial revolution that occurred between 1750 and 1850? If so, the scaling applied by the authors is not justified: spatial distributions have changed.

Response: We chose to scale down the 1850 emissions, to get a consistent gridding of the emissions as the available gridded data for 1750 was on a different grid. This will indeed give a too large emission over early industrialized regions as the UK. However, the total anthropogenic BC emissions in 1750 were quite low (0.4 Tg yr^{-1}) so that even if the spatial distribution put somewhat too large BC emissions in the early industrialized regions the errors are small. At this stage it will be a large effort to do a new pre-industrial simulation and thereafter to redo many of the figures. We will delete the sentence: "These emissions have a different spatial distribution than the historical dataset used in the rest of this work" and include "This will put somewhat too large BC emissions in the early industrialized regions, however the total anthropogenic BC emissions in 1750 were quite low (0.4 Tg yr^{-1}) so that the errors are small." on page 7477 line 28.

Page 7478, line 21: Comparing to the ranges of observed BC concentrations given in the text, Figure 2a seems to show that modelled concentrations over China, Europe, and the Arctic are on the low side.

Response: We have rephrased the sentence: "The modelled annual surface pattern (Fig. 2a) is in general good agreement with the observations listed above, however the modelled surface concentrations seems to be too low compared with the observations. A more detailed comparison with observations is given below."

Page 7480, line 21: A reference is needed for issues with PSAP measurements at high relative humidity.

Response: The anomalous behaviour is increased noise in the absorption coefficient as the humidity increases. We've reproduced this effect in the lab. However, if we look at hourly-averaged data, we don't see a dependence of the average on the standard deviation. In other words, even if the instrument gets noisy at high RH, the magnitude of the absorption doesn't change. We have therefore deleted the sentence from the text.

Page 7486, line 2: The authors should discuss why fossil-fuel BC dominates in the model, while biomass-burning BC dominates in the observations. Is it due to different regions of the Arctic being dominated by different source types? Is there a large uncertainty in the source attribution of observed BC?

Response: This is common problem with all models using current emission inventories that the BC in snow is dominated by the fossil and biofuel sources. It may be that the positive matrix factorization method used by Hegg et al. have interpreted some of the biofuel emissions as coming from open biomass burning as they have similar source signatures. A recent study found that EC measured at

Nordic sites was dominated by fossil fuel sources using carbon isotope analysis. We have added the following to the text Page 7486, line 5:

“Their analysis indicates that most of the BC in the snow samples had a biomass source. It may be that part of the discrepancy is because large contributions from residential wood burning (included in our FFBC source) were interpreted as open biomass burning as they have similar source signatures. A recent study by Yttri et al. (2011) investigated the source of elemental carbon in the atmosphere measured at Nordic rural sites in late summer using $^{14}\text{C}/^{12}\text{C}$ ratio. They found however that fossil fuel totally dominated the ambient elemental carbon loadings.”

Page 7496, line 20: The strength of the direct effect is not only related to the amount of sunlight. The solar zenith angle is also important, with the direct effect being stronger at intermediate values. Optical depth and single-scattering albedo being fixed, the direct effect is stronger at mid- latitudes, although the large absorption of BC will complicate things. To be more convincing on the issue of changes in radiative forcing efficiency, I would show the distribution of annual-averaged RF exerted by a constant BC optical depth.

Response: We have added the following sentence on page 7496, line 28: “The normalized RF with respect to AOT shows similar development as the normalized RF with respect to burden both close to source regions and in the Arctic”. As a note Haywood and Shine (1997) show that RF increases almost linearly with solar zenith angle for BC.

Page 7497, line 2: Any physical explanation for this counter-intuitive result?

Response: This indeed counter-intuitive result is explained by Shindell and Faluvegi as being a result of reduced temperature gradient between the Arctic and the mid-latitude atmosphere causing a reduction in the meridional heat transport by transient eddies. A sentence referring to the explanation given by Shindell and Faluvegi is included in the text: “since it leads to a reduced temperature gradient between the Arctic and mid-latitudes which caused a reduction in the northward heat transport by transient eddies.” on page 7497 line 2. While this is a plausible explanation, we believe that further detailed sensitivity studies with more than one GCM are needed to confirm this result.

Page 7501, line 8: This statement is speculative since the paper has not demonstrated such a strong link between BC forcing and global-averaged temperature.

Response: We have added to this sentence: “and should be investigated further in attribution studies”.

Page 7502, line 26: Which physical process is represented by including wet-deposited BC into the two top snow layers instead of the topmost layer only?

Response: This is included to represent dry deposition that only affects BC in the uppermost layer (until the next snowfall). For wet deposition, the snow layers are generated and the amount of BC deposited is stored. After a snow fall a top layer of 1 cm is separated from the rest of the new snow layer, both with the same BC concentrations. The 1 cm top layer is kept separated due to possible dry deposition. We have rephrased to make this clearer in the Appendix.

3 Technical comments

Page 7471, line 9: "reduce" should read "reducing".

Page 7471, line 16: "climate forcers" is a bit awkward. I suggest "climate forcing agents".

Page 7475, line 8: Opening bracket in citation is missing.

Page 7484, line 17: "relative" should read "relatively".

Page 7485, line 22: "in the western Russian" should read "in western Russia".

Page 7492, line 24: "but only less than 20% higher than the RF in 1910" could be better written as "but remains within 20% of the RF in 1910".

Page 7494, line 13: "range from 1-30m² g¹" should read "are in the range 1-30 m²g⁻¹".

Page 7494, line 23: "there are a relatively good consistency" should read "there is".

Page 7495, line 16: Period is missing at the end of the sentence.

Response: All the suggestions under technical comments have been taken into account.

Referee # 2

This paper discusses the simulation of black carbon deposition onto snow surfaces since preindustrial times. It also examines the radiative forcing and aging time of black carbon during this period. It contains some novel information. I would recommend revisions as suggested below. If these comments are addressed with modifications in the text, I would recommend publication.

P. 7472. The authors state, “Most of these models do not include aerosol microphysics that lead to conversion of BC to a hydrophilic state where it can be scavenged by precipitation.” However, some models do account for aging by treating microphysical processes. Please identify such models.

Response: We have included a sentence on page 7472 line 28: “The aging times used in this study is based on simulations using the M7 module (Vignati et al., 2004). Other models including aerosol microphysics are e.g. Bauer et al. (2010) and Jacobson (2001).”

P. 7473. The authors state they use a CTM. However, the authors do not acknowledge the disadvantage of using a CTM versus an interactive climate model-CTM– namely, the lack of feedback of the transported pollutants in the CTM back to weather and climate, which could change results. The authors should acknowledge this limitation.

Response: We have included in the model and setup section page 7473 line 27: “The changes in BC concentrations do not feed back on the meteorological data”. In the discussion section, the use of constant meteorology over the historical time period is discussed in general. For calculations of radiative forcing, where meteorological conditions should be kept fixed, it is an advantage to use a CTM.

P. 7474. The authors claim that the advection scheme they use is “low-diffusive.” However, this is not demonstrated here. Most global models are significantly diffusive, particularly in the vertical transport of trace species, as demonstrated by the model comparisons with vertical BC profile measurements over the Pacific ocean in Schwarz et al. Geophys. Res. Lett., 37, L18812, doi:10.1029/2010GL044372, 2010. The nearly uniform vertical profile of the modeled concentrations up to high altitude compared with the sloped profile of the data in that comparison suggests numerical diffusion. The authors have compared with some different vertical profile data here, and the results are ambiguous with respect to both numerical diffusion and accuracy. It would be helpful to compare their vertical profiles of BC with the Schwarz et al. data as well since then their results can be compared also with those from other models along with the data and a better determination of the numerical diffusivity could be obtained.

Response: The advection scheme used is shown to be “low-diffusive” in Prather (1986). But there are other effects as well, as vertical transport by convection and wet removal that determines the vertical profile.

The reason why we did not include a comparison with Schwarz et al. (2010) was that the observations were done in January 2009, and we did not simulate BC concentrations for January

2009. We have now included a new figure comparing the vertical profile from the latitude band in Schwarz et al. (2010) with the modelled profiles from the flight track for the years 2001-2008.

We have included a figure and a discussion on page 7483 line 13:

“Figure 7 compares measurements of BC from a flight campaign from 67S to 80°N in the Pacific in January 2009 (Schwarz et al., 2010) with profiles corresponding to the flight track from the modelled monthly mean BC for the years 2001 to 2008. The model overestimates the mean observed BC at all latitudes and altitudes, except close to the surface at high northern latitudes. As in Fig. 6, the model over predicts in the upper troposphere, especially in the tropics (Fig.7c) where the large gradient between 600 and 400 hPa is not captured by the model. Compared to the AeroCom model results Schwarz et al. (2010) found that the models greatly overestimate in this region as well, and related this to insufficient wet removal in the models and of minor importance errors in advection processes. Koch et al. (2009) also related the discrepancies between models and aircraft measurements to removal processes being to low and also vertical mixing may be too excessive. We should note that in Fig. 7 the averaged January mean concentration is compared with an observed snap shot of BC. “

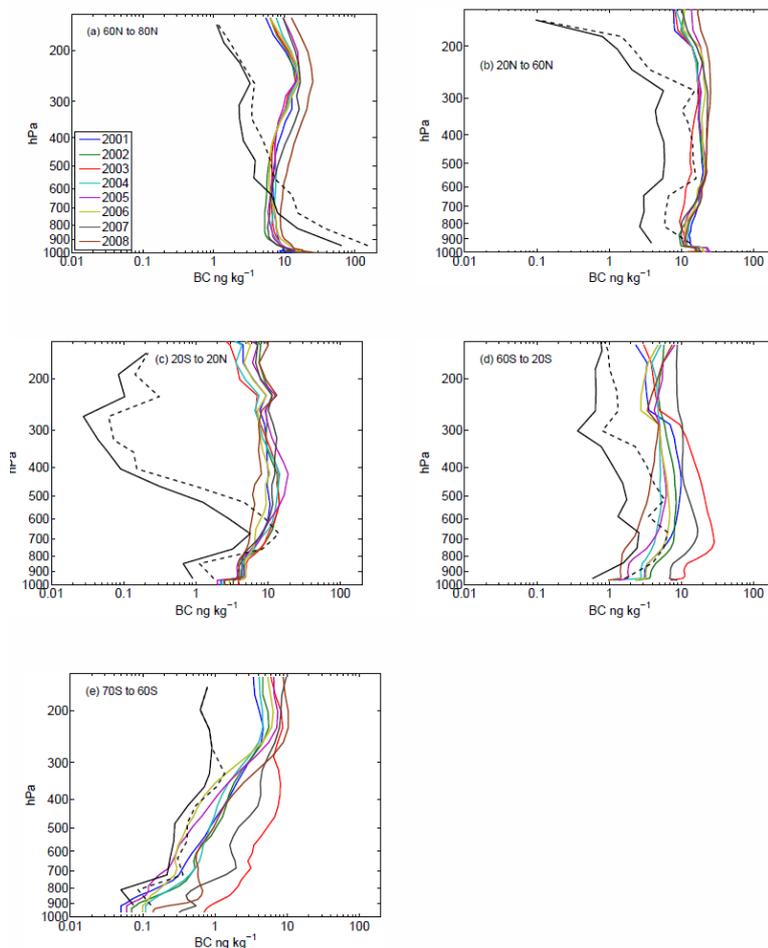


Figure 1 The average vertical profile of BC observed in each latitude zone from Schwarz et al. (2010) (black line) and dashed line indicating the standard deviation of the observations. The coloured lines show the corresponding vertical profiles from the modelled monthly mean corresponding to the flight track, one line for each of the simulated years 2001 to 2008.

P. 7474. Aging depends not only on coagulation of BC with sulfate but also on condensation of sulfuric acid and other vapor onto BC-containing particles and heterogeneous reaction on BC-containing particle surfaces. These processes do not seem to be accounted for. The authors should state explicitly that they are excluding these additional processes. Also, the aging process varies as a function of particle size, but the authors seem to treat only bulk BC. The authors should explain how this simplification might affect results.

Response: The M7 version includes condensation of sulphuric acid and coagulation of BC with sulphate. To make this clearer we wrote on:

Page 7474 line 21 regarding the use of a constant exponential lifetime: "This is a simplification since the aging depends on coagulation with soluble aerosols and condensation of organics and inorganic species."

and on Page 7474 line 29: "This model version allows for condensation of sulphuric acid and coagulation of sulphate on BC aerosols which determines the aging time, but excludes other effects. The M7 version may therefore overestimate the aging time."

P. 7476. The authors assume hydrophobic BC does not obtain a coating for radiative calculations. However, sulfuric acid condenses on soot particles containing BC regardless of whether soot is hydrophobic or hydrophilic, and water will then hydrate to the sulfuric acid. The authors should discuss the potential error from not treating this process.

Response: We have added the following sentences on page 7476 line 7: "We note that this is a simplification of the aging process of BC and the coating of BC with no enhanced absorption for the intermediate between hydrophilic and hydrophobic BC. However, given the uncertainties in the absorption efficiency of BC the approach suggested by Bond et al. (2006) is useful."

P. 7478. With respect to the calculated global mean BC concentration, please distinguish between land and ocean concentrations.

Response: We have included: ", 0.25 $\mu\text{g m}^{-3}$ over land and 0.04 $\mu\text{g m}^{-3}$ over ocean" on page 7478 line 11.

P. 7478. It is not clear how Fig. 2a shows that the modeled surface concentrations are in "good agreement with observations" as no observations are shown in that figure.

Response: We have rephrased the sentence: " The modelled annual surface pattern (Fig. 2a) is in general good agreement with the observations listed above, however the modelled concentrations

seems to be too low compared with the observations. A more detailed comparison with observations is given below.”

P. 7479. Figure 3 does not provide useful information. The authors should instead show a time series comparison of the model with observations at each location instead of a scatterplot. The scatterplot does not pair model with data in time so serves no real benefit.

Response: In Figure 3 monthly mean concentrations from observations are plotted against model results for the corresponding month and year, and thus indeed pair model with data in time. Different symbols and colours indicate different months. We have added to the first sentence in the figure legend: “for the same month and year” to make this clearer. We have also rephrased on page 7479 line 18 to make this clearer: “In Fig. 3 observed monthly concentrations of BC at several sites (Table 3) are compared with the corresponding model results for the same month and year for the period 2001 to 2008, when observations are available.”

Conclusions and abstract. The authors provide a single number rather than a range for their radiative forcing estimate of BC in air and snow. The authors should provide a range around the mean number.

Response: Providing an uncertainty range is challenging in a study using only one model. Schulz et al. (2006) stated that “an estimate from one model is not sufficient but a combination of several model estimates is necessary to provide a mean and to explore the uncertainty.” In the conclusions we have clearly presented our results in light of the RF ranges provided by IPCC AR4. The results from our model will be an input to future assessments which will explore the uncertainty in RF. In the abstract we have added “in this study” to the sentence: “The calculated radiative forcing in 2000 for the direct aerosol effect is 0.35 W m^{-2} and for the snow-albedo effect 0.016 W m^{-2} ”.

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