

Anonymous Referee #1

This paper discusses an intercomparison among six global models of the indirect plus semidirect effects of biofuel soot and of fossil-fuel soot.

Major points

1. The benefit of the paper appears to be primarily that it gives information about the variation among a few models in the field in certain behavior, namely how the modeled cloud fields respond to different levels of black carbon. However, the paper lacks context in that the authors do not discuss whether nor demonstrate that the models represent clouds physically or not. Very little is mentioned about what physical treatments are included and what treatments are missing in each model. This is one of the most important points that the paper can make and is relevant since one conclusion could be that none of the models tested is adequate for simulating the effects of black carbon on clouds thus climate. This cannot be determined unless a proper identification of processes treated and missing (e.g., cloud drop nucleation, condensation, collision-coalescence, breakup, sedimentation, precipitation, evaporation, scavenging of aerosol particles, how BC interacts with cloud drops and how radiation interacts with cloud drops and BC together, and the associated numerical methods in all cases) in the models is given. It is suggested that a table be provide of both important cloud processes treated and missing in all models. Also details of resolution should be included (e.g., number of cloud drop sizes (or bulk treatment), number of clouds formed per column at a given time, and types of clouds that can form).

Response: One major benefit of this paper is that it provides a multi-model comparison of identical experiments in order to see a range of responses. We have added this to the beginning of the Conclusion: “We have provided a multi-model investigation of how soot may affect liquid-clouds, by comparing results from three soot-reduction experiments in six global models.”

This paper is intended to be a follow-up to the previous AeroCom study Quaas et al. (2009) which provided more detail on the cloud microphysics and radiative treatment. We have added text to clarify this: “This study is largely a follow-up to the earlier AeroCom study of Quaas et al. (2009) that considered the liquid cloud indirect effect response to all aerosols in ten global models, and compared these responses to satellite retrievals. The study indicated a positive relation between cloud droplet number concentration (CDNC) and aerosol optical depth (AOD) that was generally well captured by the models. The models generally overestimated a positive relation between cloud liquid water path (LWP) and AOD, suggesting possible deficiencies in their cloud water conversion to rain, or autoconversion parameterizations. On the other hand, the models generally underestimated the positive relation between cloud cover (CC) and AOD. The modeled global mean cloudy sky forcing due to aerosols, scaled to the satellite CDNC-AOD regression slopes, was $-1.2 \pm 0.4 \text{ Wm}^{-2}$.” We have also added more information on model and cloud treatment. A new table, Table 2, contains information on model resolution, cloud microphysical treatments, autoconversion treatment, etc. We have also added the following text in section 2.2: “In all models, aerosols are taken into cloud droplets during cloud formation and then rained out following autoconversion; aerosols are also scavenged by falling rain below-cloud.” Also section 2.2 has more discussion of radiative treatments for BC-cloud: “The

radiative effects resulting from interstitial treatment of BC within clouds is included in most models however BC within cloud droplets is not included in these models. The latter effect was estimated to enhance BC absorption by about 5% according to Chuang et al. (2002). Jacobson (2006) found that surface warming by BC was enhanced about 10% due to BC inclusions in both cloud liquid droplets and ice particles.”

2. Based on the analysis of the first comment, the authors need to provide information as to whether some models are more suited for simulating the effects examined than others. Merely providing results from several different models without scrutinizing whether some models are better than others results in a false sense that all models are equally valid so the wide range in results is attributed to “uncertainty.” This is not a correct attribution since some models simply have no business simulating cloud effects of BC. Do models with certain characteristics (e.g., those that assume more hygroscopic organic matter or models with coarser vertical resolution) provide different results from other models? Similarly, are there other models in the field that might be more appropriate for a comparison?

Response: We have provided more detail on model cloud microphysical as well as aerosol microphysical treatments so that the reader may assess model suitability. We were going to add mean and range for the estimates of the 4 models that include aerosol microphysical treatment, but interestingly these numbers are almost identical to those that include all 6 models. So we have left the text discussion of the estimates as it is.

3. The paper presumes that everyone is in agreement that knowing indirect and semidirect effects in isolation is relevant for determining the effects of black carbon on climate. Yet, the authors provide no evidence that indirect radiative forcing is even linearly additive to direct forcings or other forcings, so they have not demonstrated that there is any point to simulating the indirect or semidirect effects in isolation. Further, they provide no evidence that the forcing calculations of BC effects on clouds are proportional to climate response. In fact, it is well known that when two effects are isolated, they give individual climate responses that sum to a different value compared with when the two effects are combined and a single response is determined. This is simply because feedbacks operate between the two effects that are ignored when they are calculated independently. The authors need to state clearly in the abstract and conclusion that their results may or may not have relevance to the overall effects of BC on climate for this reason and state explicitly that the overall effects depend on several other processes not examined, including the effects of BC on snow and sea ice albedo, on surface water evaporation, and on cloud absorption (e.g., as discussed next).

Response: These are good points, and we have changed the text to give a more balanced discussion of the value of simulating indirect and semi-direct effects independently. The abstract already explained that both indirect and semi-direct effects are included in the radiative flux perturbations. In the discussion we have modified our discussion of this (4th paragraph from the end) to: “Future experiments might isolate the indirect from the semi-direct effect, as was done in Chen et al., 2010 by switching off the aerosol-radiation interaction. However these

effects probably interact and therefore do not add linearly. Furthermore, ultimately we are interested in the net effect of soot and co-emitted species on climate, including semi-direct, direct, indirect and snow/ice-albedo effects together.”

4. All models tested ignore the treatment of cloud drop absorption by BC inclusions (both nucleated and scavenged BC), so the authors cannot say as they do that they are determining “cloud radiative responses.” The authors need to clarify in the abstract and text that they are ignoring this effect and, as such, cannot draw conclusions about the effects of BC on cloud radiative responses (thus overall effects of BC), only on the responses that they treated, and only based on the detail they treated.

Response: We have clarified in the text: “The models include radiative interactions among BC and cloud particles within a cloud, but do not account for the effects of absorption enhancement of BC within cloud droplets as described and treated in Jacobson (2002, 2006). Scattering between cloud and aerosol layers are typically included.” We do not feel this level of detail is appropriate in the Abstract.

5. None of the models appear to treat radiative transfer through discrete size-resolved cloud drops or ice crystals for each wavelength of solar and thermal-IR, so it does not appear possible for the models to account for multiple scattering of cloud light through BC particles that lie interstitially between cloud drops and below or above clouds. As such, the statement that the paper includes “semi-direct” effects appears overly optimistic.

The authors should first clarify exactly what aspect of semi-direct effects each model treats and then state clearly that these treatments are simplistic relative to what could be treated in an ideal model. Are separate spectral radiative calculations performed for cloudy and clear portions of each grid cell? How are cloud optical properties versus aerosol optical properties calculated when aerosols are present within clouds?

Response: Yes, in general the semi-direct effect included in the models is simple and we are now more specific in the beginning of section 3.2: “All models also include semi-direct effects, or the change in cloud distribution resulting from aerosol direct radiative perturbation of the atmospheric thermal structure”. And “The models include radiative interactions among BC and cloud particles within a cloud, but do not account for the effects of absorption enhancement of BC within cloud droplets as described and treated in Jacobson (2002; 2006). Scattering between cloudy and aerosol layers is also typically included.”

6. The indirect effects examined here appear to apply only to warm clouds, but not mixed phase clouds or ice-only clouds (except one model is stated to treat mixed phase clouds in an uncertain manner). Given that most clouds worldwide are mixed-phase, it is unclear what the difference in results would be if BC effects on such clouds as well as ice clouds were considered. The authors need to acknowledge clearly in the abstract and paper that their results do not apply to mixed-phase (except the one model) or ice-only clouds and they are not treating explicit cloud microphysics in any clouds in any of the models.

Response: The models simulate BC effects on liquid clouds, including effects on droplets in mixed phase clouds – the initial manuscript was incorrect regarding mixed phase clouds). In the abstract, in general, we feel it is appropriate to focus on

what the study does include, rather than what it does not. We have added the modifier “liquid” also to the abstract. Within the Introduction we have added the clarifying sentences “In this study we do not consider the effects of BC on ice clouds, but rather focus on the effects of BC on liquid droplets within liquid or mixed phase clouds only. We also note that the effects of BC on ice-phase clouds as observed in the field and laboratory are very uncertain (e.g. Kärcher et al. (2007).” Many of the models do include cloud microphysics, such as impact on droplet size and therefore on collision-coalescence. More detail on the model cloud schemes has been provided in section 2.2.

7. Although all models are run forward in time and differences are taken among multiple simulations for each model, there is no significance testing of the results. As such, it is not possible to tell whether any of the results are statistically significant relative to deterministic chaotic variations due, for example, to a random change in initial condition in each model. The authors should each provide a global plot of the regions of the world where results are statistically significant (using a t-test) relative to a set of random perturbation simulations.

Response: Unfortunately it would be impossible at this point to have each modeling group repeat their experiments and perform t-tests, since their models have evolved since performing these experiments. We do present global mean standard deviations for 3 models over their 5-year simulations, for perturbations in radiative forcing, cloud cover and cloud optical depth (end of section 3) and we discuss the large inter-annual variability in the models there and in the Discussion section.

8. It appears that simulations were run for only 1 year. If so, how do we know that results over 1 year are representative of what occurs over 2, 3, 4, 10, or 20 years? Cloud effects are climate responses, so the changes in temperature due to indirect effects and radiative heating will result in changes in feedbacks that will alter clouds and natural emission over a period much longer than one year. If the simulations are one year long, they do not appear long enough. The authors should really run their tests over at least 15-20 years if they have not done so.

Response: The models were run for six years, with analysis performed on five. As stated in the Discussion “Our simulations were performed for five years, and longer experiments would be better when considering the effects on clouds from relatively small aerosol perturbations” so we are aware that the experiment length is probably problematic. On the other hand, as now stated more clearly in section 2.2, the models used prescribed SST’s which tends to reduce the amount of cloud feedback, which is a limitation in terms of full climate response analysis, but does tend to reduce variability allowing for shorter simulation length.

A couple of these points are discussed in more detail below.

If the models tested are treating indirect and radiative effects of clouds physically, they should be able to simulate the increase in cloud fraction or optical depth with increasing aerosol optical depth followed by a decrease in cloud fraction or optical depth upon further increase in cloud optical depth in the presence of absorbing aerosols, as found by satellite correlations (e.g., Koren et al., 2004, 2006; Ten Hoeve and Jacobson, 2010). The

reduction in COD with increasing AOD is due to radiative absorption in cloud drops (cloud absorption effect) as well as interstitially between drops and below clouds (semidirect effect). The authors are including only the semi-direct effect and only part of that effect apparently, since much of the effect is due to multiple scattering within clouds, and such scattering cannot be simulated correctly when clouds are treated as bulk properties and individual wavelengths do not interact with individual cloud drops of different size and aerosol particles of different size between the cloud drops. With regard to the cloud absorption effect, this has been simulated in 1-D studies in which nucleation scavenging of BC was treated (e.g., BC was treated as a single core; e.g., Conant et al., 2002). However, to account for washout (impaction scavenging of BC), it is necessary to treat polydispersion of BC in cloud drops. The absorption due to multiple BC inclusions in cloud drops is much higher than that due to a single inclusion of the same summed volume at most visible wavelengths (e.g., Jacobson, 2006, Figure 1). The effect of such treatment appears to be a much stronger tropospheric warming (Figure 4a of that paper), which is stated by Jacobson (2010) to be found strong at the surface as well in that study.

Response: According to Koch and Del Genio (2010), models even with relatively simple aerosol-cloud radiative interactions can simulate either an increase or decrease in cloud cover, depending on cloud type, the dynamical environment, surface hydrology, etc. However it is beyond the scope of this study to discuss semi-direct effects in detail. We have added specifics regarding model treatments of semi-direct and cloud-aerosol radiative interactions as discussed in points 1, 3, 4, 5 of our response above.

Table 1. Please include the number of vertical layers in each model, the model top height, the number of layers, in the boundary layer, the number of layers in the troposphere, and the number of layers in the stratosphere (if applicable).

Response: The vertical resolution of the models and model references are provided in Tables 2 and 3. We feel this is sufficient detail for this study.

Table 1. Three models assume either 50%, 65%, or 70% of OC from fossil fuels is hygroscopic. This assumption appears unjustifiable, as almost all OC from liquid fuel combustion is insoluble lubricating oil or unburned fuel oil. The authors need to self criticize results from these models.

Response: This is an interesting point, we have added the following to section 2.2 “There is some disparity in assumption about OC hygroscopicity among the models, with emitted hygroscopicity ranging from 0 to 70%. We note that low hygroscopicity is generally appropriate for fossil fuels while higher values are appropriate for most biofuels.”

Table 2. The title of the table says “total soot emissions,” where soot is particulate matter yet the emissions seem to include “14 TgOC from natural terpene sources” which imply gas emissions. This is inconsistent. Also the ratio of OC/BC of 10:1 for fossil fuels is unsubstantiated if this is particulate matter emissions. The ratio should be 1:1 or 2:1. The authors should have a separate category for biomass burning and separate gas from particle emissions. If the organics include gas emissions, how are they converted to particles in the models? Finally, please provide references for all numbers in the table.

Response: We have changed that Table (now Table 1) to show the soot *reduced* in the experiments and the ratio of the reductions; before we had the soot *included* in the experiments and the ratio of soot included.

The natural terpene-derived OC is now removed from the table and put in a footnote with other non-soot aerosol and aerosol precursor emissions included in the experiments.

Note that our FF experiment reduced only BC and not OC, it is an idealized experiment to see maximum BC impact, as stated in many places in the text. The references for the emissions are given in the text: “The emissions are from Dentener et al. (2006), including carbonaceous aerosol pollution emissions from an updated version of Bond et al. (2004).” We have added the following to section 2.2: “Most models assume that secondary organic aerosols are emitted as particulate OC (14 Tg y⁻¹); CAM-PNNL includes secondary organic aerosol (SOA) formation from reversible SOA condensation integrated over the size distribution of each mode.”

Table 2. How is it possible for there to be more diesel emissions than total fossil-fuel emissions?

Response: See previous response.