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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global cloud condensation nuclei influenced by carbonaceous combustion aerosol" *by* D. V. Spracklen et al.

D. V. Spracklen et al.

d.spracklen@see.leeds.ac.uk

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We thank the reviewers for their comments on our paper. To guide the review process we have copied the reviewer comments in italics. Our responses are in regular font. We have responded to all the referee comments and made alterations to our paper.

J. Pierce (Referee)

This paper evaluates (1) GLOMAP model predictions of CCN against observations and (2) the model-predicted sensitivity of CDNC, the aerosol direct effect and the cloud albedo indirect effect to changes in carbonaceous aerosol. This second point is addresses the important discussion of the efficacy of BC reduction for climate control. The paper is well written and certainly of interest to the ACP readership. I recom-

mend it being published in ACPD after several minor issues are addressed. General comment:

1. In my opinion, the most important finding in the paper is the huge sensitivity of the AIE of carbonaceous aerosol due to the emitted size distribution of the carbonaceous aerosol (factor of 3!!). To be fair, the smallCCA sim had very small-sized carbonaceous emissions; however, the sensitivity would still be very large even if the emissions sizes in the smallCCA sim were made a bit larger.

I think that it is unlikely scientists will be able to give policy makers any useful estimate of how a reduction of carbonaceous emissions affects climate until this uncertainty is reduced. Also, it is likely that the emissions size distribution of carbonaceous aerosols would change if control techniques were used to reduce BC, which introduced additional uncertainties beyond those shown here.

It appears that the AIE is more sensitive to uncertainties in carbonaceous primary emissions sizes than it is to uncertainties in nucleation rates (e.g. Wang and Penner, 2010. I tried to deduce a sensitivity from Merikanto et al, 2010, but couldn't quickly.) Yet, in my opinion, the community is focused much more on nucleation. There is no doubt that nucleation is important, but your results here show just how important it is for us to better understand primary emissions size.

I would make this large uncertainty in the carbonaceous AIE due to uncertainties in the emissions size distribution a main point of the paper in both the abstract and the conclusions.

The referee is probably correct in suggesting that the AIE will be more sensitive to uncertainties in the emission size of primary particles than it is to uncertainties in the nucleation rate. However, we have not directly performed these comparisons. We have previously shown a relatively low sensitivity of simulated CCN concentration to changes in the nucleation rate (this can also be seen in Fig., 2c and 2d). The assumed emitted size distribution in experiment smallCCA is based on AEROCOM recommendations

(Dentener et al., 2006). It may well be too small, especially for coarse resolution global models. We agree that the uncertainty in the primary aerosol size distribution needs to be addressed in future studies. As suggested we emphasise this uncertainty. We add the following additional text to the conclusions (note this additional text also addresses some concerns raised by referee 2 but is included fully here for clarity):

"We have synthesised observations reported in the published literature to produce a worldwide dataset (277 mean observations from 55 published studies) of cloud condensation nuclei (CCN) concentrations. We used the dataset along with a global aerosol microphysics model to explore the contribution of carbonaceous combustion aerosol to CCN. We find that in polluted locations (defined based on the simulated black carbon mass concentrations) the model is biased low unless carbonaceous combustion aerosol is able to act as CCN. Uncertainty in the emitted size distribution of carbonaceous combustion aerosol results in a substantial uncertainty in the contribution of this aerosol source to global CCN. When we use the emission size suggested by AEROCOM (Dentener et al., 2006) carbonaceous combustion aerosol contributes 64% to simulated global CCN concentrations. Using a larger emission size (Stier et al., 2005) reduces the calculated contribution of carbonaceous combustion aerosol to global CCN to 52%.

We calculated the first (cloud albedo) aerosol indirect effect due to carbonaceous combustion aerosol. We do not calculate the second (cloud lifetime) aerosol indirect effect or semi-direct effects. Therefore our study should not be used to calculate the overall impact of carbonaceous combustion aerosol on climate. Furthermore, since different aerosol effects are unlikely to linearly combine care should be taken when comparing the forcing numbers calculated here with those from other studies. We calculate a global mean top of atmosphere first aerosol indirect effect due to carbonaceous combustion aerosol (from fossil fuel, biofuel and wildfire) of -0.34 W m-2 when we assume particles are emitted at larger sizes and -1.08 W m-2 when we assume the smaller emitted size. This large sensitivity to particle size underlines the importance

of improving emission databases to include emitted particle size and the need to understand sub-grid evolution of particle size distributions from emission to the regional scale (Pierce et al., 2009). We calculate that carbonaceous combustion aerosol from pollution sources (fossil fuel and biofuel emissions) results in a aerosol indirect effect of -0.23 W m-2 (when we assume particles are emitted at larger (Steir et al., 2005) sizes). The smaller particle size of fossil fuel carbonaceous combustion emissions means that they contribute more to CCN concentrations than biomass burning emission per unit mass of emission. We find that whilst pollution (fossil and biofuel) emission sources only account for 31% of the total emitted mass of carbonaceous combustion aerosol they account for 75% of the simulated enhancement in CDNC."

Specific comments: 1. Page 7007, line 12: Why is BLN not used in the default simulation since it was shown in earlier papers to improve GLOMAP results? Wouldn't it be important to have it in the base case as well as the cases where carbonaceous aerosols are turned off in order to better simulate the changing size distribution?

For the default simulation we choose a model set-up that is most comparable to other global aerosol models reported in the literature. Since previous studies of carbonaceous combustion aerosol did not include BL particle formation we chose not to include this in our default simulation, instead including this in sensitivity simulations. The response of the aerosol system when carbonaceous combustion aerosols are removed is a substantial uncertainty. Certainly this work (and that of Chen et al. (2010)) needs to be repeated with different empirical nucleation mechanisms to test how this changes the model response to removing carbonaceous combustion aerosol. In our paper we emphasise the need to further study the non-linear response between emissions and CCN concentrations.

2. Section 4.3.1: To estimate the aerosol direct effect, you use the forcing per mass from AeroCom averages and the global mass burdens predicted in your GLOMAP simulations. However, forcing per mass is a fairly strong function size (information that you have in GLOMAP) and location (i.e. surface reflectance/cloud cover). I understand

that it may be more work than is necessary to do a full rad x-fer calculation on your aerosol fields, but do you have an idea of how similar your aerosol size distributions are to those assumed by the AeroCom models when they calculated the radiative forcing? Do you have an idea of how much uncertainty might be associated with differences in the size distribution?

The main reason that I'm curious about this is because I wonder if uncertainties in the size distribution (e.g. smallCCA) have important effects on the direct effect calculations too. I'm fairly sure they won't be a factor of 3 change in the direct forcing like you saw with the AIE, but they still could be important.

I'm not asking you to recalculate your direct effects, but maybe add a few sentences on these uncertainties. Maybe its something we should try to quantify in the future.

The referee is correct that there are different and possibly more accurate ways of calculating the direct radiative effect. However, since this was not the main objective of our paper we chose the current simplified technique which does have the added advantage of including some of the AEROCOM model diversity into our calculated direct radiative effect. We agree that uncertainties in the size distribution of carbonaceous combustion aerosol are unlikely to introduce the same level of uncertainty in the aerosol direct effect (as that seen in the aerosol indirect effect). However, this uncertainty should be quantified in the future. We emphasise that our calculation of the aerosol direct effect is an order of magnitude estimate.

3. Page 7014, Line 24: What are the units for the effective radius in this equation? Since the "100" at the beginning of the right-hand side has no listed units, you can figure out the Re units.

The re unit is μm, as specified on page 7014, line 20. This equation corresponds to eq. (2) from Bower et al. (1994). However, we thank the reviewer for pointing this out, as the w term (density of liquid water, 1g cm^{-3}) is missing from the original Bower equation. For clarity, we now add this into our equation. The text on page 7014, lines

20-26 is now:

"For the unperturbed and perturbed runs, cloud effective drop radius re (in μm) for low and mid level water clouds was calculated from the GLOMAP CDNC (in cm^{-3}) and ISCCP derived liquid water paths (LWP, in g m^{-2}), using the Bower et al. (1994) parameterisation, namely:

r_e=100 $\times$ [LWP/(Δz) $\times$ 3/(4π $\times$ w $\times$ CDNC)]$^{1/3}$,

where Δz (in m) is the cloud thickness, which in our climatology is roughly 1400 m and 2900 m for low and middle clouds, respectively, and w (in g cm^{-3}) is the density of liquid water. Only water clouds were modified. "

4. Page 7015, Line 2: Can you elaborate more on this perturbation experiment? Did you keep that ratio of drop volumes constant?

We assumed that the liquid water content (LWC = (4/3) $\times$ π $\times$ r_e^3 $\times$ w $\times$ CDNC) is constant, meaning that a change in CDNC leads to a change in drop volume (since w remains constant). Therefore the re change was calculated as follows:

r_{e2} = r_{e1} $\times$ (CDNC1 / CDNC2)$^{1/3}$

In the alternative approach, due to the methodology used to derive LWP from the ISCCP data, we considered a constant r_{e1}=10 μm for the unperturbed experiment, while for the perturbed experiment we calculated the corresponding r_{e2} using the above formula. This approach gave very similar forcings as when using the Bower et al. (1994) parameterisation to derive independent values for r_e for the unperturbed and the perturbed experiments.

The text on page 7014, line 26 – page 7015, line 5 was modified:

Note that in its derivation of LWP, ISCCP assumes a constant effective radius of 10 μm. This creates an inconsistency between our method and the original ISCCP retrieval. To investigate this our results were compared to an alternative approach, where a

control effective radius of r_{e1}=10μm was employed in the unperturbed experiment. For the perturbed experiment, the effective radius re2 was scaled to account for the drop volume change (needed in order to maintain the same water content) caused from the GLOMAP CDNC change, i.e. $r_{e2}=r_{e1}\times(\text{CDNC1/CDNC2})^{1/3}$. Both approaches gave very similar answers, suggesting that the methodology is robust.

Anonymous Referee 2

This paper discusses modeled estimates of the indirect radiative forcing due to wildfire plus pollution carbonaceous combustion aerosols. The paper uses some observations of CCN in an effort to corroborate some of the assumptions made in the modelling exercise.

Whereas the comparison with CCN data is a unique aspect of the paper, the main issue with this paper is that it claims to calculate the effects of carbonaceous combustion aerosol on global cloud albedo (e.g., P. 7002, "To evaluate the impact of carbonaceous combustion aerosol on global cloud albedo....") when in fact neither the model nor the analysis accounts for the necessary physical processes occurring in clouds to do this. The authors make the familiar mistake of assuming that only indirect effects affect cloud thickness, fraction, and lifetime, thus cloud albedo. It then uses this limited information to suggest (abstract) "this cooling effect must be accounted for to ensure that black carbon emissions controls....have the desired net effect on climate" when in fact, several climate response modeling studies have accounted for indirect effects together with other major cloud effects not accounted for here in a physical manner and found that black carbon warms the climate.

It was not our intention to fully calculate the impact of carbonaceous combustion aerosol on climate. Calculating the full set of interactions and feedbacks between aerosols and clouds requires a high resolution cloud resolving model coupled to a calculation of aerosol microphysics. Parameterising such aerosol-cloud interactions in a global model with coarse resolution introduces additional aspects of uncertainty that

are difficult to evaluate. For this reason we quantify the first order effect of aerosol on clouds: the first aerosol indirect effect. This diagnostic is recognised by the community and used by the Intergovernmental Panel on Climate Change. We add text to our paper to emphasise that we only treat the first aerosol indirect effect and that out paper should not be seen as treating the full climate impact of carbonaceous combustion aerosol. For example we add the following text to the Introduction:

"We do not calculate the cloud lifetime (second indirect) effect or semi-direct effects and so we are unable to estimate the full climate impact of carbonaceous combustion aerosol."

We have also added the following text to the Conclusions:

"We do not calculate the second (cloud lifetime) aerosol indirect effect or semi-direct effects. Therefore our study should not be used to calculate the overall impact of carbonaceous combustion aerosol on climate. Furthermore, since different aerosol effects are unlikely to linearly combine care should be taken when comparing the forcing numbers calculated here with those from other studies."

We acknowledge that other papers have accounted for these effects by adding the following text to the Conclusions:

"Some previous studies (Jacobson, 2002, 2004, 2006, 2010) that have attempted to include a diverse range of aerosol direct, indirect and semi-direct effects have concluded that carbonaceous combustion aerosols have a net positive warming impact on climate."

Major missing processes are as follows: Aside from producing indirect effects, combustion aerosols affect clouds through the semi-direct effect (which the authors acknowledge but state that they do not account for on p. 7012) and the cloud absorption effect, which is the heating of cloud drops by absorbing inclusions within them as well as heating of clouds by absorbing particles interstitially between cloud drops (Jacobson, 2006;

2010).

We acknowledge that carbonaceous combustion aerosol affects clouds in multiple ways and that many of these processes are poorly understand and ill quantified. We modify our paper to include a more detailed discussion of the potential effects of combustion aerosol on clouds (P7001, Line 21):

"Additionally, carbonaceous combustion aerosol affect clouds through a range of mechanisms including semi-direct effects (Hansen et al., 1997), the cloud absorption effect (Jacobson 2002, 2010) and potentially through acting as ice nuclei (e.g., DeMott et al., 1999). The semi-direct effect is used to describe the range of cloud responses that occur due to changes in the temperature structure of the atmosphere driven by absorbing aerosols. Koch and Del Genio (2010) reviewed model studies of the semi-direct effect and showed that the net-negative semi-direct effect due to BC may be similar in magnitude but opposite in sign to the positive direct effect of BC. The climate response due to BC also critically depends on the vertical profile of the aerosol, with BC resulting in less warming when present at higher altitudes (e.g., Ban-Weiss et al., 2011). Jacobson (2002, 2004, 2006, 2010) account for a diverse range of aerosol-climate interactions including the aerosol direct effect, indirect effect, semi-direct effects and cloud absorption effects, and calculate a net-warming due to carbonaceous combustion aerosol."

As shown in Koren et al., 2004 and Kaufman and Koren, 2006 with respect to cloud fraction and Ten Hoeve et al. (2011) with respect to cloud optical depth, whereas carbonaceous aerosol particles increase cloud optical depth (COD) at low aerosol optical depth (AOD) (due to the first indirect effect), they burn off such clouds at higher AOD (around 0.2-0.3). In the present case, the authors account only for the increase in COD with increasing AOD in the absence of absorption in clouds, so essentially assume that COD increases linearly with increasing AOD for all AODs, whereas Ten Hoeve et al. (2011, Figure 6), shows that this is not the case based on satellite analysis. As the three effects, indirect, semi-direct, and cloud absorption, occur simultaneously, it is not possible to add them linearly, nor have the authors or any other study demonstrated

they are.

We acknowledge that we do not address all the climate effects of carbonaceous combustion aerosol, that the different climate effects are unlikely to combine linearly and that our paper should not be seen as an estimate of the full climate impact of carbonaceous combustion aerosol. We add text to the Conclusion and Introduction to emphasise these points (see responses elsewhere).

We add the following discussion on remote sensing analysis to our paper:

"Remote sensing studies have been used to explore the relationship between cloud (cloud fraction and cloud optical depth) and aerosol (aerosol optical depth) properties (Koren et al., 2004; Kaufman and Koren, 2006; Ten Hoeve et al., 2011). For the Amazonian biomass burning season Tan Hoeve et al. (2011) showed that at lower values of AOD (<0.3) cloud optical depth and AOD were positively correlated whereas at higher values of AOD they were negatively correlated. The authors postulated that positive correlations at low AOD may be due to the aerosol first indirect effect whereas negative correlations at high AOD may be due to inhibition of cloud development by absorbing aerosols or potentially due to retrieval artefacts."

After producing estimates of the indirect radiative forcing, the authors contend in the abstract, "This cooling effect must be accounted for to ensure that black carbon emissions controls that reduce the high number concentrations of small pollution particles have the desired net effect:..." In fact, cooling due to indirect effects of carbonaceous aerosols has been accounted for in several global climate response simulations over the last decade, such as in Jacobson (2002, 2004, 2006, 2010), where two of these studies are listed in the authors' paper. These studies also accounted for the semidirect effect, and the last two accounted for the cloud absorption effect and treated microphysical growth of cloud drops and their aerosol inclusions as a function of discrete size in many subgrid clouds, which is not done in the present study (the authors use a parameterization that assumes one cloud per global model grid cell and modal treat-

ment of the cloud distribtuion). All studies, though, found a strong warming due to black carbon, so the implication that the indirect effects have not been treated and might lead to a net warming, is unfounded based on the published literature of climate response simulations.

We did not intend to give the impression that indirect effects of carbonaceous combustion aerosol had not been previously treated. On the contrary we cited a number of studies of the indirect radiative forcing due to carbonaceous combustion aerosol (see for example P7001 and P7016). We appreciate the referee pointing out a number of papers that we did not previously cite (Jacobson, 2004 and Jacobson, 2006). We now include citations to these studies (see our response above) and also a more complete discussion of the range of potential aerosol-cloud interactions. We also add the following text to the Conclusions:

"Some previous studies (Jacobson, 2002, 2004, 2006, 2010) that have attempted to include a diverse range of aerosol direct, indirect and semi-direct effects have concluded that carbonaceous combustion aerosols have a net positive warming impact on climate. It is now necessary to evaluate the predictions of such models against new datasets of cloud condensation nuclei that have been synthesised here."

Along these lines, the authors further contend on P. 7001, "However, carbonaceous combustion aerosol also contains particulate organic matter (POM), which can have a cooling effect on climate because it scatters solar radiation...." "Poor understanding of these effects has forced many previous studies to account only for atmospheric BC heating when assign the global warming potential of carbonaceous combustion aerosol." However, all four of those studies listed above accounted for POM emissions as well as particle aging due to internal mixing over time. The authors should clarify that previous studies have accounted both for indirect effects and treatment of POM emitted with carbonaceous aerosols. The authors should also recognize that BC is emitted with a coating of some carbonaceous aerosols (e.g., lubricating oil and unburned fuel oil in the case of fossil-fuel combustion), and that coating has been shown to enhance

warming due to BC due to the optical focusing effect in laboratory studies.

The statement "Poor understanding of these effects..." is true: many studies only consider BC heating, see the cited references. The uncertainty of BC impacts on climate is acknowledged by recent policy reports. For example, the recent UNEP Integrated Assessment of Black Carbon and Tropospheric Ozone states that uncertainty in the climate effects of BC are large "because BC and OC can influence clouds that have multiple effects on climate that are not fully understood".

The referee is correct that previous studies have included the aerosol indirect effect and have accounted for POM emissions and we modify our paper to clarify these points (see responses elsewhere). Additionally we have included a more detailed discussion of aerosol-cloud interactions (see above).

The novelty of the present paper lies in comparisons with some CCN data, which I applaud, but not with the modeling or the idea of simulating indirect effects or POM. However, the authors overstate how the comparisons with data help to demonstrate their point. In the abstract, they state, "The net radiative effect of carbonaceous combustion aerosol is uncertain because their contribution to cloud drops has not been evaluated to date on the global scale." This statement should be rephrased. First, merely evaluating a model against some data does not reduce the uncertainty. First, any evaluation must be accurate and the accuracy must be for the right reason. In the present case, the model is missing many processes and sources of emissions so a good evaluation against data could easily be for the wrong reason. Second, comparisons need to be performed against many parameters simultaneously, and statistical significance testing is needed. Third, some other studies have evaluated the effects of carbonaceous combustion aerosols on cloud drops in different ways. The statement implies that this has not been done.

We do not know of a previous study that has evaluated the contribution of carbonaceous combustion aerosol to CCN concentrations at the global scale. We change the

statement to say "...contribution to cloud condensation nuclei..." rather than "cloud drops". We have included discussion of other techniques that have been used to constrain the impact of carbonaceous combustion aerosol on clouds (see above).

Our results qualitatively agree and provide additional confirmation for recent papers that have also demonstrated a large contribution of primary carbonaceous aerosol to CCN (e.g., Pierce et al., 2007, Chen et al., 2010). It is harder to compare our results against some of the studies cited by the referee (e.g., Jacobson, 2002, 2004, 2006, 2010) because they have not reported key diagnostics such as concentrations of cloud condensation nuclei or cloud droplet number concentrations.

With respect to missing processes, the treatment of cloud absorption and the semi direct effects are two. In addition, the authors do not treat aerosol-cloud interactions completely. In order to get the number of cloud drop number concentration (CDNC) correct, it is necessary not only to account for CCN activation (which the authors do), but also the microphysical growth and coalescence of cloud drops, drop breakup, and precipitation as a function of size. These treatments are needed to calculate scavenging by new and existing cloud and precipitation drops of interstitial aerosol particles, which the authors do not do. The simultaneous scavenging reduces the available number of particles, particularly of carbonaceous particles that the authors are interested in. As such, by not accounting for this process, the authors overestimate the number of CCN available.

We have previously compared simulated cloud droplet number concentrations against observations and shown that the model produces reasonable concentrations (Merikanto et al., 2010). We state this in the current paper (Page 7013, Line 11-13). This study uses the same model and model setup as experiments CCA (2) and bln (3) of the present study (see table 2). We have also previously shown that our model captures the observed relationship between aerosol number concentrations and CDNC (Pringle et al., 2009). We acknowledge that there are considerable uncertainties in both the measurement and modelling of CDNCs. In the absence of these unresolved

sub-grid processes, which we do not believe can be adequately treated in a global model, we prefer to restrict our study to CDN at cloud base, which is likely to be far less uncertain.

The authors argue that their results are correct because they reasonably match observations of CCN; however, not only is the error in CCN versus observations sufficiently large and the number of measurements sufficiently small to question whether the comparisons are meaningful, but even if the comparisons of CCN are close, the authors show no evidence that their aerosol particle number concentrations are correct or that their distribution of anthropogenic versus natural CCN are correct.

We believe we have gone much further in assessing the realism of our aerosol microphysics fields than the majority of the studies listed by the referee. In those papers we found very little evaluation against in situ aerosol measurements. In previous papers we have evaluated our model against a diverse range of aerosol observations including aerosol particle number concentrations and aerosol number-size distributions (e.g., Korhonen et al., 2008, Merikanto et al., 2010; Metzger et al., 2010, Spracklen et al., 2005, 2006, 2007, 2008a, b, 2010). We point the reader to the most relevant of these comparisons in the current paper (see P7005, Line 22-28). We have previously made comprehensive comparison against observed aerosol particle number concentrations. For example, in Spracklen et al. (2010) we synthesised observed aerosol particle number concentrations from around the world and made a detailed evaluation against our model. This effort involved many scientists from the measurement community and to our knowledge is one of the most comprehensive evaluations of aerosol particle number against a global model made to date. Our current paper represents the most comprehensive evaluation of simulated CCN concentrations by a global aerosol model. Therefore we believe that the evaluation of our model against aerosol observations is at least as comprehensive and in many cases more so than papers cited by the referee. For example, whilst Jacobson (2002) compare against observations of near surface BC and sulfate mass concentrations, Jacobson (2004) compare against limited obser-

vations of BC in snow, sea-ice and rainwater and Jacobson (2010) compares against observed vertical profiles of sulfate, nitrate, organic matter and ammonium made over the U.K. and northeast Atlantic, there is little evidence from these papers for a comprehensive evaluation against aerosol particle number, CCN concentrations or aerosol number size distributions.

The referee asks us to show evidence that our simulated distributions of anthropogenic versus natural CCN are correct. Since there are no observations that can distinguish between anthropogenic and natural CCN this is an impossible model diagnostic to evaluate. We have included additional analysis to show that the model is not strongly biased in clean, remote conditions (see below) which suggests that we do not greatly underpredict natural aerosol sources.

For example, we don't know if the soil particle number concentrations are correct. Also, it does not appear that the authors include several sources of natural aerosol particles, including pollen, viruses, bacteria, all of which are of sufficient size to activate cloud drops. The larger the background number of particles, the less the influence of anthropogenic particles.

The referee is correct that increases in the natural background of particles will decrease the influence of anthropogenic particles. Our treatment of natural particle sources is of similar complexity to many other global aerosol models. We do not include primary biological aerosol particles. However, both model (e.g., Heald Spracklen, 2009; Hoose et al., 2010) and observational (e.g., Elbert et al., 2007, Poschl et al., 2010) studies have suggested that they are unlikely to make a substantial contribution to particle number or CCN concentrations even over biogenically active regions such as the Amazon basin.

More important, the authors have not published a paper on the algorithms showing that their cloud-aerosol interaction algorithms reproduce data for cloud optical depth, cloud thickness, cloud lifetime, etc., or physically-expected results. Instead, the model is run and results from the aerosol-cloud interaction algorithms are assumed to be correct

without evaluation at high resolution.

Our model is an offline CTM and therefore these diagnostics are read in from observations and reanalysis. It is likely that, being based on observations, these model fields are at least as reliable as internally calculated fields used in GCMs. For example, many un-nudged GCMs have cloud coverages that are quite far from observations. Our estimations of the first aerosol indirect effect use water vapour, temperature and ozone from ECMWF reanalysis data, surface albedo and cloud liquid water path from the International Satellite Cloud Climatology Project (see P7014). These products have been extensively evaluated and we take them as reliable.

Finally, the authors do not demonstrate the statistical significance of their result. This is needed for simulations of climate effects, including indirect effects.

We include additional analysis to demonstrate the statistical significance of our results. We add the following text:

"We used the Wilcoxon signed-rank test (an alternative to the paired t-test used when the population cannot be assumed to be normally distributed) to assess whether the means of the simulated and observed populations differ. The model simulation where carbonaceous combustion aerosol does not act as CCN is significantly different from the observations ($P \ll 0.0001$). When carbonaceous combustion aerosol acts as CCN the difference between simulated and observed CCN is statistically insignificant ($P=0.38$)."

Additional comments.

P. 7001. "...particulate organic matter, which can have a cooling effect." The authors should recognize that absorbing organic carbon (e.g., brown carbon) mitigates cooling due to non-absorbing organic carbon and that coating of black carbon with organic carbon enhances the warming due to black carbon. It appears the authors do not account for this effect.

We don't understand this comment.

The model contains little evaluation of parameters aside from CCN. It is important for the authors to compare their model results with at least some of the following satellite retrieved parameters on a global scale: cloud optical depth, cloud fraction, aerosol optical depth, and precipitation, from the same simulation. It is also not sufficient to point to previous comparisons if these were done, since the model has been updated for the present case. In addition, the authors should compare with observed BC profiles, such as those from Schwarz et al. (2010), particularly as Schwarz et al. compared their data with multiple other models.

Our model is an "off-line" chemical transport model. We force our model using European Centre for Medium Range Weather Forecast (ECMWF) analyses, which provides diagnostics cloud cover and rain rates (see page 7004). Therefore GLOMAP does not prognose cloud optical depth, cloud fraction or precipitation.

Our model has been evaluated against aerosol observations in at least as much detail as other global models. In some cases we have made some of the most comprehensive evaluations. The referee appears to suggest that we should include all our previous comparisons against aerosol observations in the current paper which is clearly not appropriate. The model used here is the same model version and model setup as that used within numerous previous works [e.g., Korhonen et al., 2008a, b; Merikanto et al., 2009, 2010; Meztger et al., 2010, Spracklen et al., 2008a, b, 2010]. We alter our paper to clarify this and we include references to these papers. In ongoing work we are making comparisons against the observed BC profiles from Schwarz et al. (2010) and a manuscript on this evaluation is in preparation.

The comparison of CCN using a scatterplot in Figure 2 is not encouraging, particularly considering that the plots are on a log-log scale and many of the model results are far out of the 25th-75th percentile of the data. It is also not clear what the dashed lines are for. Results without combustion aerosols appear to have less bias than results with

combustion aerosols except at very high CCN observations. Thus, over most of the world, it appears that the author's treatment overpredicts CCN. In any case, as the authors have not evaluated particle number from natural and anthropogenic source, it is not possible to conclude that the number of CCN, when predicted reasonable, contains the right number of natural versus carbonaceous aerosol particles.

The solid line is the 1:1 relationship, the dotted lines are the 2:1 and 1:2 relationships and the dashed lines are the 10:1 and 1:10 relationships. We clarify this in the figure caption. In Fig. 2a and Fig 2b the error bars represent the observational error (not the 25th-75th percentile). We clarify this in the figure caption.

To further explore the issue of model bias we extend our analysis (see below). We find that in remote regions the model is not strongly biased. The small model bias in remote regions suggests that we do not greatly underpredict natural aerosol sources. We already acknowledge that in clean remote conditions it is harder to isolate the impact of carbonaceous combustion aerosol since particle formation, which is itself poorly constrained, makes a substantial contribution to CCN concentrations in these locations. We find that over remote ocean regions carbonaceous combustion aerosol can lead to a reduction in CDNC (see Fig 4d) through suppressing particle formation. This microphysical response means that removing carbonaceous combustion aerosol results an increase in CDNC and a warming in these remote regions that offsets a fraction of the cooling in polluted regions (Fig. 5).

To address the referee comments on our model observation comparison we include additional analysis and discussion and an additional figure (Fig. 3 in paper, Figure 1 in this comment):

"Another metric of model performance is the ratio of simulated to observed CCN concentrations. Figure 3 shows this ratio as a probability density function for the model simulation when we allow carbonaceous combustion aerosol to act as CCN and for the model simulation when we do not. We fit a Gaussian to each probability density func-

tion. We use simulated concentrations of BC aerosol to define clean (BC < 50 ng m-3) and polluted (BC > 100 ng m-3) conditions in line with previous studies (Yoon et al., 2007). In clean conditions (Fig. 3a) the simulated to observed CCN ratio is not strongly biased in either simulation (the center of the fitted Gaussian changes from a simulated to observed ratio of 0.85 when we do not let carbonaceous combustion aerosol act as CCN to 1.16 when we do let carbonaceous combustion aerosol act as CCN). This suggests that there is not a strong bias in our representation of natural CCN sources. In polluted conditions (Fig 3b) the model is biased low (center of fitted Gaussian at a model to observed ratio of 0.26) when we do not allow carbonaceous combustion aerosol act as CCN. The model bias is greatly reduced when we allow carbonaceous combustion aerosol to act as CCN (center of fitted Gaussian at model to observed ratio of 0.98)."

As stated previously there is no observation that can determine natural versus anthropogenic CCN. Therefore it is not possible to evaluate CCN number concentrations from natural and anthropogenic sources directly. However, since the model is not strongly biased under very clean conditions (Fig. 3a) it is likely that we do not greatly under represent natural sources of CCN.

Conclusions. "Our study shows that mitigation strategies need to take account of the impact on the size distribution and number concentration of emitted carbonaceous combustion aerosol and the fact that BC and POM are present in the same particle, which shifts the technological challenge considerably." This conclusion was shown previously in several climate response papers listed above, including those cited in the present manuscript. The differences are that some of those papers accounted for cloud absorption and aerosol-cloud interactions with a microphysical approach, whereas the present study does not. Those papers also showed that, despite the strong indirect effects of aerosol particles, which are stated to be treated explicitly in those papers, the multiple effects of BC on warming outweigh the cooling due to the indirect effects. They further showed that fossil fuel soot behaves different from biofuel soot due to the

different cloud activation properties of each.

We include and have added citations for these previous studies. In our paper we have synthesised observations of cloud condensation nuclei to produce a new dataset for evaluating global aerosol models. We use the dataset to evaluate the impact of carbonaceous combustion aerosol on global CCN. This has not been done before and is the focus of our paper. Where possible we have put our results in context with previous studies. Since some of the papers cited by the referee (Jacobson, 2002, 2004, 2006, 2010) do not report simulated CCN concentrations, nor do they evaluate simulated against observed CCN it is not possible for us to compare our results against these previous studies in any meaningful way. We add text to the conclusions to address the concerns raised by the referee (see our response to referee 1).

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Figure 3. Probability density functions of model to observed ratios of CCN concentration when carbonaceous combustion aerosol does not act as CCN (black line) and when carbonaceous aerosol does act as CCN (red line) for (a) clean conditions (simulated EC < 50 ng m-3), (b) polluted conditions (EC > 100 ng m-3). Dashed line indicates the model to observed ratio of 1. The dotted lines show Gaussian fits to the probability density function.

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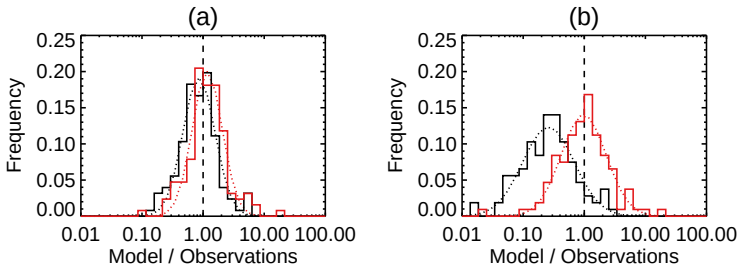

Fig. 1. See caption in text