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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.

Anonymous Referee #2

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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 modeling 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.

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).

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.

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 semi-direct 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 treatment 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.

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 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.

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.

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. 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.

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.

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

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.

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.

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.

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.

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