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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Correlation between cloud condensation nuclei concentration and aerosol optical thickness in remote and polluted regions" *by* M. O. Andreae

M. O. Andreae

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Response to Anonymous Referee #2

(Referee comments in *italics*, my response in plain font)

This manuscript presents a nice set of data that shows the correlation between observed AOT and boundary layer CCN concentrations. The combined data set and analysis are unique for the peer-reviewed literature, and as such makes a significant contribution. I have two major comments which must be addressed as a condition of acceptance.

1) There are numerous places in the abstract, Section 3, and Section 4, where the au-

thor asserts that some of the difference between remote land and remote ocean CCN concentrations must be anthropogenic. This is plausible, at best, but I am confused why it is asserted so prominently in the abstract and conclusions. The statement is not supported in a quantitative way. The only support for the statement is the observation that pollution events occur more frequently in remote land observations than in remote marine observations. This is not sufficient to quantify at all whether the land-marine CCN difference in remote regions is anthropogenic, natural, or some meaningful combination between the two. If there is a more quantitative way of demonstrating this conclusion, it needs to be shown in the paper rather explicitly. At present, the assumption is stated as a conclusion, and should be eliminated from the abstract and conclusions and softened in the text.

The arguments for this assertion have been made before in the peer-reviewed literature (Andreae, 2007a and 2007b; Andreae and Rosenfeld, 2008), and I had not thought it necessary to repeat them here. They are based on observations that aerosol number concentrations over the cleanest regions on land approach those over the cleanest oceans, and that they increase with proximity to pollution sources. In our own studies from the Amazon Basin and Siberia, we find that combustion-derived particles (soot) are present even under fairly clean conditions, that there is some sulfate that can be related to transport from pollution sources, and that there are correlations between particle number and pollution tracers (soot, CO, etc.) even under nearly pristine conditions. A further argument is that the atmospheric lifetime of aerosols is of the same order as the transport time across continents. Finally, the same conclusion comes out of several global aerosol modeling studies (all cited in the papers indicated above). I am including a brief statement and appropriate references in the revised manuscript.

2) The penultimate sentence of the paper needs to be eliminated: "...the radiative and microphysical effects of aerosols on clouds, and therefore on climate and precipitation, are correlated and cannot vary independently of one another, at least not on larger scales." Again, this is totally unsupported by the data. The correlation shown between

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AOT and CCN statistics is regional and statistical, and is hardly a general constraint. Furthermore, the implication that average CCN concentration constitutes the primary forcing parameter in aerosol-cloud-climate interactions is incredibly naive. The author seems to be unaware of black carbon effects on precipitation and clouds, the importance of IN on precipitation, dynamical feedbacks that vary in sign from one cloud type to another, etc...

This paper came out of a discussion on what would be the appropriate variables to use as drivers in global models of aerosol effects on clouds and climate, and what ranges of values would need to be considered for these variables. The first results from this effort, a conceptual analysis of the combined radiative and microphysical effects of aerosols on clouds, have just been published in Science (Rosenfeld et al., 2008). We selected AOT as a proxy for the radiative effect of aerosols on cloud formation and the production of convectively available potential energy. $CCN_{0.4}$ was chosen to represent microphysical effects. Initially, we thought that the entire AOT-CCN parameter space might be populated and would have to be included in an analysis. One of the benefits of the present study is that the observations cluster along the diagonal represented by the fit line in Figure 1, removing one degree of freedom for a first, conceptual analysis.

I don't think that I have asserted anywhere that "average CCN concentration constitutes the primary forcing parameter in aerosol-cloud-climate interactions" – I don't think I am that "incredibly naïve". I will, however, defend that boundary-layer CCN concentrations are a major forcing variable for the microphysical effects of aerosols on clouds, climate and precipitation. As the author and co-author of several recent reviews on aerosol-cloud-precipitation interactions, I am also keenly aware of all the complications in this field. I do not feel, however, that the issues the reviewer mentions, and the many problems he doesn't, are at all germane to the present paper. Here, the only point is the – surprising – empirical correlation found in the analysis. This correlation is not regional, as the reviewer states, but global, since the data set presented here covers all continents and oceans. To accommodate the reviewer's concerns, I am softening

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the offending statement, and adding some discussion to make the intended point more clearly.

References:

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 11293, 2008.