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Interactive Comment

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 #1

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

My (weak) recommendation for (eventual) acceptance is only based on the fact that the paper compiles a large data base of aerosol data that someone might find useful. My major concern is that in its current form, the work will be seriously misused. The author should carefully express the many caveats. I urge the author to look at a much more interesting aspect of the in-situ data, i.e. at the relationship between CCN and aerosol extinction (i.e., "local optical depth") measured at the same point. Such a study will be of far more use to the community, and to climate models.

I'm reassured by the reviewer's statement that "someone might find [this compilation] useful". In contrast to the reviewer, however, I have enough faith in the community's wisdom that I am not overly concerned about its misuse. In order to alleviate the reviewer's concerns, I am including some additional caveats in the revised version (more specifics below). I agree with the reviewer that investigations of local relationships are of interest. This has been done many times (often in the original papers that are cited here), and to great scientific benefit, but is not the point of this paper, which looks at large-scale, empirical relationships.

Major comments: Presumably the goal of such a study would be to connect AOT to CCN and eventually connect CCN to cloud microphysical characteristics such as drop concentration or effective radius. Although there is a good correlation between AOT and CCN over large enough spatial/temporal scales, the use of an empirical equation such as that in Figure 1 completely misses the mark because clouds are localized, phenomena, driven by convective scale processes. Column conditions, averaged over many data sets, do not drive cloud-scale processes.

Firstly, the equation in Figure 1 is simply the quantitative description of an empirical statistical relationship. As such, it cannot "miss the mark". Secondly, such an equation can indeed be used in a statistically predictive sense. I.e., for a given value of the independent variable (here AOT) there is a defined probability that the dependent variable (here CCN concentration) will be within a certain range. Thus, while the "column condition" obviously does not "drive cloud scale processes", it does define a probability density function for one variable (boundary layer CCN concentration) that does play a role at the process level.

1) The study ignores a list of processes that make application of such an equation dangerous in the context of indirect effect studies. A few of these are mentioned here:

(i) A column measurement like AOT does not represent the aerosols affecting the cloud, particularly, but not only, in conditions of mid-tropospheric transport; the troposphere is

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frequently poorly mixed and aerosols have a distinct scale height.

The analysis shows, that in spite of the obvious fact that a column property like AOT does not have to predict a surface or near-surface property, such as boundary layer CCN concentration, there is an empirical correlation in this global meta-analysis. In fact, I started out with the same concerns, and was surprised myself to find the relationship reported in this paper. It appears that, at least in a statistical sense, the variability in scale height and the layering of the troposphere are not strong enough to obscure the observed empirical relationship.

(ii) Measurement of AOT is affected by clouds because skies are rarely clear;

The AERONET AOT product, which was used for most of the AOT data reported here, is carefully screened to minimize the effects of clouds; the algorithms used for this purpose are well validated and have been published in the peer-reviewed literature. For the remaining AOT data, one must assume that the authors of the original publications have also used appropriate quality control procedures. It is not possible in a meta-analysis to evaluate all the methodological details of each original study.

(iii) RH affects AOT and introduces variability. (Kapustin's work is referred to but the lessons learned by that study are ignored.)

This is undoubtedly true, and may ultimately limit the applicability of remote-sensing detection of CCN. This variability is part of the reason for the large error bars in Figure 1, but again, does not contradict or obscure the existence of an empirical correlation.

(iv) N=CSˆk is a poor approximation to CCN spectra; activation spectra exhibit curvature on log-log plots.

Regrettably, not all authors of the many studies included here reported data at the same supersaturation, or showed complete supersaturation spectra. In these cases, using this equation was the only way I could think of to interpolate to a common supersaturation. I am open to better suggestions! On the other hand, in most of the studies included in the data set used here, these interpolations were either not needed, or had to cover only a small range of supersaturations, so that the effects of curvature or errors in the choice of k would be minor.

The danger is that those unfamiliar with these issues will use the empirical relationship and produce results that are only tenuously linked to atmospheric processes like aerosol indirect effects.

I prefer to give my colleagues enough credit to assume that they understand the meaning and proper use of statistical, empirical relationships. Caveats based on the comments received from the reviewers will be added to the revised version to protect the innocent.

2) It is claimed that empirical equations of this kind will be useful in climate modeling studies. Climate models at least have the benefit of a height-resolved aerosol and don't have to rely on column-integrated quantities. Of what use will a relationship like this be to a climate model? Why not explore the relationship between light extinction and CCN concentration? This would be analogous to the work by Hegg and Kaufman (JGR 1998) which looked at the relationship between number concentration and volume of an aerosol population. The author should explore this avenue even if the size of the data set is smaller than what is currently used.

I appreciate the reviewer encouraging me to write a paper on another topic than the one I chose here, and will take it into consideration for my future research. On the other hand, I have been assured by climate modelers that the existence of the kind of constraint expressed by the empirical relationship shown here is quite useful to them. In fact, this study came out of a discussion with modelers who requested just such an analysis. Some of the early applications have been published in Science (Rosenfeld et al., 2008).

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Other comments:

1) The font size in the Tables is so small that they are basically illegible.

I have faith in the skill of the editorial office of ACP that the font in the final version will be acceptable. I agree that the present font is a challenge to ageing eyes, including my own.

2) There is no mention of the published relationships between AOT and surface PM2.5 which is very similar in concept and has similar problems.

If the reviewer were to be kind enough to supply a reference, I would be happy to include it.

3) What is the mean and standard deviation of the distance between AOT and CCN measurements? How well synchronized are they in time?

I don't quite understand what use a mean and standard deviation would be here. Huge gradients can exist over short distances in some regions, while the aerosol properties in others may change little over hundreds of km. Rather than presenting such a metric, I have indicated the geographic coordinates of the AOT and CCN measurement sites for each case in Table 2 (with apologies for the small font). Specifics of the time periods used are also given in the Tables.

References:

Rosenfeld, D., U. Lohmann, G. B. Raga, C. D. O'Dowd, M. Kulmala, S. Fuzzi, A. Reissell, and M. O. Andreae, Flood or drought: How do aerosols affect precipitation?, *Science*, 321, 1309-1313, 2008.

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