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Interactive comment on “Global distribution of the effective aerosol hygroscopicity parameter for CCN activation” by K. J. Pringle et al.

Anonymous Referee #2

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The authors have used a global chemical transport and transformation model to compute size-resolved composition of atmospheric aerosols, both for present day emissions and for preindustrial assumptions. They then use the composition fields to compute the hygroscopicity parameter at each grid point and over an annual cycle. Reasons for the simulated hygroscopicities are discussed based on regional and seasonal variations in aerosol composition.

The model fields are not shown; they are to be presented in another paper where comparisons to observations will be made. This is somewhat of a concern because the calculations in this paper are only meaningful to the extent they reflect observed aerosol composition. However, since the models used are well known and tested, presumably the fields are reasonable. Assuming this, mostly the results shown here

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are not surprising, since they reflect current knowledge of the composition of the fine fraction of the atmospheric aerosol, which contains most of the number concentration and thus influences CCN concentrations most strongly. The authors do make a good point regarding the variations in composition with height and noting that in general, hygroscopicity decreases with height, either because an enhanced fraction of organic species or of dust is present. Since these altitudes are most relevant to cloud formation, surface CCN measurements may not provide a complete or accurate picture of how aerosols affect cloud formation.

Overall, if I have understood this presentation correctly (see below), the findings presented here really present global variations in bulk, fine mode aerosol composition, as reflected in the hygroscopicity parameter (whose calculation is somewhat uncertain, especially for the assumed single value for the organic fraction). These fields cannot be immediately translated into CCN concentrations, even given fine aerosol size distributions, unless the assumption of complete internal mixing is valid. The authors' earlier papers have explored the validity of these assumptions for some observational data sets, and the validity of this assumption on the global scale needs to be shown here – or at the very least, the authors should make more clear that CCN predictions using these recommended values are valid for only very aged, internally-mixed particles.

Most of the questions I have for the authors refer to Section 2.2, where the calculation of kappas is discussed. First, the mixing rule as presented is valid for an internally mixed aerosol, and it is stated that this rule has been applied to each of the seven aerosol models of the model. So is it assumed in the model, or perhaps only in these calculations, that each simulated mode is completely internally mixed? This is an important point that must be stated explicitly – it is not explained in section 2.1, Model Description.

Also, please note that although the kappa values assumed for each component are listed, one needs also the assumed densities in order to compute volume fractions

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from the modeled mass fractions.

Second, it is stated that “We calculated kappa for each of the seven aerosol modes of the model. [in Section 2.1, these are identified as 4 hydrophilic and 3 hydrophobic log-normal modes, but it’s not clear how these are related to the “Aitken,” “accumulation”, etc. modes?] We took the volume weighted mean value of the Aitken and accumulation modes as this size range is most relevant for atmospheric CCN concentrations in the atmosphere and thus for comparison to field observations. . . . We considered the volume fraction of each component in both the hydrophilic and hydrophobic modes, which corresponds to the bulk composition of fine particulate matter”.

So if I understand the above correctly, what was done was equivalent to a filter-based estimate of aerosol hygroscopicity: The contributions of the nucleation and coarse mode were ignored, and the rest of the aerosol mass (which essentially would show up on a PM1 filter) was assumed completely internally mixed, and then kappa was calculated?

If the approach was this simple, then the discussion needs to be revised to make this clear. It’s confusing to talk about how each mode was treated, if this information is not really used. Further, if indeed this simple approach was used, why go to all the trouble of using a size-resolved aerosol module to simulate the aerosol composition? Since the authors do have more resolved information, however, it seems like an opportunity was missed here to discuss what the model predicts as far as mixing state and chemical heterogeneity and where that matters for the estimation of CCN concentrations. Although on p. 6306 the authors cite a number of papers that they say support a bulk-composition approach as adequate for the prediction of CCN concentrations, I don’t think this has been uniformly found in prior closure studies (e.g., Cubison et al., 2008, suggested that knowledge of the mixing state was of primary importance in determining the activation properties of urban aerosols). I have difficulty understanding how the information presented here can be extrapolated to the accurate global prediction of CCN concentrations, using simulated size distributions, as implied in the manuscript.

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Specific comments:

Abstract:

1. When the mean values are presented, it should be noted that these were based on assumed internal mixtures of the fine mode aerosol (if that was indeed the case).
2. “The influence of industrialisation on aerosol hygroscopicity appears to be less pronounced than the influence on the aerosol burden”. On what basis is this statement made? The mass loadings? But that is not totally relevant to number concentrations. The sensitivity of the predicted CCN at some selected supersaturation would be a more relevant comparison.
3. Last sentence (and elsewhere): the factor change of kappa is not the most relevant metric, since for example a factor of 2 could mean 0.6-> 1.2 or 0.001->0.002, having very different magnitudes of impact.

2.1 Model description

1. T42 resolution is approximately 2.8 by 2.8 degrees, considered coarse resolution. This should be pointed out, because many fields will be “smeared” due to this factor alone.

3.1 Comparison with observations

Much of this discussion would be the same for an earlier-generation model that predicted only aerosol mass concentrations in the fine mode. The contrast between continents and oceans is expected, as are the plumes of pollution that modify aerosol composition in certain oceanic regions.

3.4 Annual cycle

1. “the chemical composition can change throughout the year, which can potentially affect kappa”: isn’t it certain that it will affect kappa?
2. I can’t accept that the seasonal cycle is not important. One has to only look at the data collected by the IMPROVE network at rural sites in the US to see that there is a

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strong seasonal cycle in composition that must certainly affect kappa.

3.5.1 Regional distributions, continental

1. p. 6312, line 18: I don't agree with this reasoning. I would guess that the coarse resolution of the model is responsible for much of the homogenization that is observed, and it is not accurate to ascribe it to an actual physical process without much more checking against observations. A similar comment applies to p. 6314, line 10.

3.5.3 Regional vs. temporal variability

1. p. 6314, line 12: the minimum in kappa in North America in June and July is quite surprising, and I would guess inaccurate for the Eastern U.S. Again, a lot of information is based on modeled chemical fields that are not shown and, until the companion paper is published, cannot be evaluated against observations.

3.6 Effect of regional mean kappa on critical supersaturation required for CCN activation

1. The study by Reutter et al assumed a single, chemically uniform lognormal mode. Thus, the findings cited here are fully relevant only to the extent that the atmospheric aerosol conforms to this picture.

2. Figure 7 reinforces the idea that the aerosol can be considered chemically homogeneous, and that this single value together with the modal size distributions can be used to predict CCN active at any supersaturation. Is this indeed what the authors are claiming? I also want to ask about the statement on p. 6316, line 13-15, that although particle size is the main factor in activation, the small differences in kappa between regions can also play a role. I find it hard to believe that these variations in the mean kappa matter more than external mixing with modal differences in kappa. Can this hierarchy of importance be demonstrated for the ranges of situations modeled here (ie the actual mixing states / sizes of various modes and compositions)? That would be a fascinating and highly useful result.

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3.7 Present vs. Pre-industrial

p. 6318, line 7-9: Doesn't one have to weight changes in number, diameter AND composition to really understand these differences? It seems like this analysis is over-simplified. Similar comments apply to the note about applicability of mass-dependent parameterizations.

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