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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Physical properties of the sub-micrometer aerosol over the Amazon rainforest during the wet-to-dry season transition – comparison of modeled and measured CCN concentrations" *by* J. Rissler et al.

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Answear to the specific comments

1. Nenes and Seinfeld (2003) suggested a new parameterization of cloud droplet formation in global climate models. Their model takes into account some of the important kinetic limitations that our static CCN parameterization does not. The Nenes and Seinfeld (2003) model requires a description of the CCN properties of the aerosol entering the cloud. This input is described in a very general way in their paper in order not to be exclusive, and should be in the form of dN/ds, that is the number of particles that activate as a function of the supersaturation in the cloud. The cumulative representation of dN/ds is simply the CCN spectrum, which we propose can be estimated from four

fundamental physical properties of each mode, namely the number concentration, the geometric mean diameter, the geometric standard deviation, and finally the (modal average) soluble volume fraction (equations 11-13). The latter quantity can be estimated from H-TDMA measurements, and refers to a model compound, for instance a certain salt.

Already now a similar parameterisation of CCN data from another campaign (SMOCC, heavy biomass burning aerosol) is used in an adiabatic cloud parcel model (Khain, A. P., Rosenfeld, D., Pokrovsky, A.: Simulating convective clouds with sustained supercooled liquid water down to -37.5°C using a spectral microphysics model, Geophys. Res. Let. 28 (2001) 3887-3896), which will be used for calibration of cloud model in a GCM (Frank J. Nober, Hans-F. Graf, Daniel Rosenfeld: Sensitivity of the global circulation to the suppression of precipitation by anthropogenic aerosols. Global and Planetary Change 767 (2003) 1-24).

In Nenes and Seinfeld (2003), the calculation of dN/ds is not the main issue. Here, we have determined the physical properties that determine dN/ds (or rather the integral CCN spectrum) in various air masses in the Amazon region. In combination with a later paper describing the dry season heavy biomass burning aerosol, we are able to characterize all three major aerosol types of the Amazon region (unperturbed rain forest background air, aged biomass burning aerosol, and heavy biomass burning aerosol). Numerous other H-TDMA and SMPS/DMPS data sets exist for a large variety of other environments world-wide, and they could be used for the same purpose.

Even though GCMs at present are not able to fully incorporate this information and the parameterization of cloud droplet formation suggested by Nenes and Seinfeld (2003), they must at some point address this issue. If not, the large uncertainty in our ability to predict future climate change due to the aerosol indirect forcing will remain.

The referee statement “What is the value of CCN measurements other than to help identify the water uptake characteristics of particles?” is actually rather provocative.

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Should we stop worrying about CCN and the aerosol indirect effect? We certainly don't think so. We quote the latest IPCC report that we also feel support our line of research (IPCC, 2001, section 5.3.3, last paragraph): "Large-scale models must be able to represent several factors related to CCN in order to better assess the indirect effect: the size distribution of the mass of water-soluble species, the degree of solubility of the represented species, and the amount of mixing of individual species within a given size fraction." We also have strong support from within Brazil. On 18 March 2004, the Brazilian Senate approved a "Resolution of Appreciation" for the LBA studies in Amazonia on aerosol and cloud interactions, of which CLAIRE-2001 is a part. But perhaps we misunderstand the question.

See more on comment 10.

2. Section 4.1.2, top of page 3178
The following was added to section 4.1:
"The air masses representing clean background and aged biomass burning smoke are well defined and can be generalized to be valid for larger regions of the Amazon.". . .
"The observations made on recent biomass burning injected into a clean background air mass should be used with more care since the source was local and rather small."

The last paragraph in section 4.1 adds to the discussion. For instance: "Furthermore, the measurement site was situated in a forested area, while most biomass burning characterization experiments have been carried in regions with ongoing rapid deforestation or already deforested regions such as the central part of Rondônia state. The particle concentrations observed during CLAIRE-2001 were therefore low compared to studies in regions with intense biomass burning."

The concentration much depends of the extent of the fire. The Balbina site was situated in the pristine forest and no heavy biomass burning takes place in the area – only small-scale fires. The fact that the concentrations were rather stable during the "recent biomass burning" indicates we were inside the plume and not on the edge.

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In section 4.1.2 we only discuss the classification of periods. We want to leave the detailed discussion regarding the number size distribution for section 4.2.1. But as suggested we rewrote and shortened down the discussion about the issue in section 4.2.1.

3. Section 4.2.1, p. 3179
During the selected clean period (no biomass burning) during CLAIRE 2001, we write that the total concentration were very similar to that of CLAIRE 1998 (also clean period), but with slightly lower concentrations in the Aitken mode. Here we clearly say that this is the case only for the selected clean period and not in general.

CLAIRE 2001 and CLAIRE 1998 were made almost at the same site (some km apart) situated in the pristine forest. No heavy biomass burning takes place in the area, even during dry season. This is the reason for the concentrations during the "recent biomass burning" for CLAIRE 2001 being lower than other studies. This is pointed out earlier in the article, but in order to make it more clear we add a sentence in the new version of chapter 4.2.1.

4. Section 4.2.1, paragraph at bottom of 3179 going onto 3180 and Figure 3. The terms "nucleation mode" and "Aitken mode" are well known and generally used terms. We are aware that they can be defined differently and could be ambiguous. Here a clear definition is given where the terms are first used (chapter 4.2.1). To avoid misunderstanding, in the new version a sentence containing the term "Aitken" used before the definition is removed (chapter 4.1.3).

5. Page 3180, bottom paragraph
We are in an area with high precipitation and since the largest particles activate more easily into CCN they are expected to be removed by wet deposition. Cloud processing of aerosol particles is more efficient than coagulation in this humid environment. This is explained later in the section.

After reading the text again, having in mind the referee comments, we rewrote and

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shortened down paragraph 4-6 in chapter 4.2.1 (see comment 2). We agree with the second comment and remove the sentence “This is more in line with the expectations”.

6. The reason for the two distributions being so similar we can unfortunately not determine. We made some speculations in the text of why the distribution looked like they did. Maybe it is just chance. We have not found any reference on aged biomass burning superimposed on a background aerosol in a region similar to the Amazon region.

The classification of air masses is not based on number size distributions but on other quite clear indicators such as trajectories, satellite images, gas data, and total number conc.

7. Page 3181, last paragraph
The dip at 100 nm is not instrumental. The HGFs do show an increase with size, but not as much as if SO_2 were responsible for all the increase in size from Aitken mode particles to accumulation mode particles. This has been observed also in other H-TDMA data sets. A possible explanation could be condensable organic compounds that are not as hygroscopic as ammonium sulfate. One should also keep in mind that the most hygroscopic particles are efficiently washed out.

8. Section 4.2.2
The nucleation mode was difficult to fit but still gives information about the presence of the nucleation mode as well as some information about size. Therefore we found it worthwhile to show the parameters.

We do not present any time series for particles in the 3-5 nm size range, simply since there are almost no particles of this size and therefore give no information. The nucleation mode is typically somewhat larger, around $\sim$10-15 nm. This as well as our conclusion is explained in chapter 4.2.2, paragraph 2.

Even though the nucleation mode was present during heavy rain events the typical diurnal pattern seen for the occurrence of the nucleation mode particles is not at all

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correlated with rain events. About the splashing of raindrops our feeling is that the particles seen are too small to be formed from such a process.

The daily pattern of the the nucleation mode particles can be seen very clearly in contour plots showing the variability of the entire size distribution over one whole day (not shown). Since we didn't want to focus on this issue we only included 2c, where as pointed out, the diurnal pattern is not shown very clear. Because the figure is small as it is we remove 2c from Fig. 2, leaving more room for 2a, b, d and e.

Since we claim to describe the "Physical properties of the sub-micrometer aerosol", we think that the nucleation mode particles have to be mentioned. They contribute noticeable to the total number concentration. The diurnal pattern is obvious and a similar pattern has been observed during a later campaign in Rondônia, Brazil. Since there are large differences compared to observations over boreal forests (Kulmala, M., et al., J. Aer. Sci., 35, 143-176, 2004), we consider it to be of interest to the reader. In the new version of the article the discussion about nucleation was made shorter.

We leave to the editor do decide if the section should be removed or not.

9. Section 5

We have tried to add some references that could be considered missing. If the referee has any suggestion for reference all contributions are gratefully received.

10. Section 6

Also the section about parameterisation was looked through and the use of an error function removed, shortening the section somewhat. We see a value in the parameterisation to make the data usable for any other application. A motivation is added in chapter 6. They could be used as input data in GCM's and other cloud models (see comment 1).

A reference to figure 9 was added in the text.

11. Section 7

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We changed the range in parenthesis from 3-850 nm to 3-30 nm.

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