Atmos. Chem. Phys. Discuss., 12, C10092–C10096, 2012
www.atmos-chem-phys-discuss.net/12/C10092/2012/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud condensation nuclei production associated with atmospheric nucleation: a synthesis based on existing literature and new results" *by* V.-M. Kerminen et al.

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Received and published: 4 December 2012

We would like to thank the reviewer for the constructive comments. Below we have added our response separately after each detailed referee comment.

This paper presents a review of field and modelling studies of CCN production from atmospheric new particle formation events. NPF is potentially a very important source of CCN, and I therefore find the paper useful and timely. However, I believe that the paper could be improved especially in the sense that comparisons between the results obtained and techniques applied in the referenced papers could be more quantitative. I therefore have a few comments that the authors should address during revision of the

paper.

Overall, I am somewhat disappointed in the literature review of section 3. I think the comparison of the observations should be made more quantitative, for example a table showing how much NPF events have been found to contribute to CCN populations in the different studies would be very useful.

Our response: We have substantially revised sections 3.1 and 3.2 based on the detailed and quite relevant comments given by the referee below. The problem with trying to be more quantitative is that the practically none of the previous investigations approached this problem in the same way, i.e. using the same data analysis methods. As a result, we consider quantitative comparison of the available field studies impossible at the moment. Therefore, we prefer discussing those results qualitatively in the text rather than summarizing the results in a table.

An important reference missing from section 3.1 is Hamed et al. (ACP 10, 1071, 2010). They considered CCN production in NPF events in Melpitz, Germany, and showed that although the new particle production (both in terms of event frequency and new particle formation rates) has decreased considerably between the two periods studied (1996-97 and 2003-06), CCN production from the events did not decrease between the study periods. This was attributed mainly to higher temperatures during the latter period, causing increased SOA production and faster particle growth rates, and thus increased survival probability to CCN size range.

Our response: This is a very good point. We added the following paragraph (now a third paragraph) into section 3.1:

"Hamed et al. (2010) investigated how past reductions in SO2 emissions might have affected secondary CCN production by comparing long-term measurements in a central European location between two time periods (1996–1997 vs. 2003–2006). They found that, consistent with substantial European-scale SO2 emission reductions over this time, both the frequency of nucleation events and the magnitude of new particle

formation during the events decreased considerably. On the contrary, CCN production associated with atmospheric nucleation was found to increase over the same time period, most likely as a result of increased nuclei growth rates caused by increasing biogenic aerosol precursor emissions. The study by Hamed et al. (2010) points out the complicated interplay between natural and anthropogenic emission and atmospheric CCN production. "

In section 3.2.3 the authors note that their approach of calculating CCN production lead to clearly different results from those of Asmi et al. (2011) who used a somewhat different way of calculation. This should be made more quantitative: How big were the differences? In fact, it would be very useful (and make the excercise of presenting new results quite a bit more valuable) if the authors calculated the CCN production from the Hyytiälä, Pallas, Botsalano and Vavhill events using all the different methods that have been presented in the papers reviewed in section 3.1.

Our response: We revised the text in section 3.2.3 in two ways. Firstly, we revised the comparison of our results to those obtained by Asmi et al (2011) to make it more quantitative, as suggested. These modifications can be found in the second paragraph of sections 3.2.3. Secondly, we analyzed one full year of measurement data from one of the stations using all the 3 methods that have recently been applied in estimating CCN production using field data. Based on this analyzes, the following paragraph was added into section 3.2.3:

"In order to consistently compare the three methods described earlier in this section, a subset of Type I nucleation events in Hyytiälä in 2002 was analyzed by using all the three methods. It quickly became apparent that such a comparison is a complicated task, since certain methods may not be appropriate for a particular event (e.g., a higher CCN concentration prior to an event, leading to a negative increase in CCN concentration), while any other method may work well for the same event. Because of this, we analyzed only those events for which all the three methods produced physically reasonable results. The comparison revealed that the method used in this paper produced

the highest increases in CCN number concentrations (Fig. 7), which is logical considering that the method utilized the maximum CCN concentration during an event. The method used by Asmi et al. (2011a) produced usually the smallest increases in CCN concentrations. When comparing the different thresholds diameters defining the lower limit of the CCN concentration, no systematic pattern in estimated CCN increases between the methods by Asmi et al. (2011a) and Laakso et al. (2012) could be identified (Fig. 7)."

Section 5.3: Another way of combining the different approaches would be to use regional models with high spatial resolution to investigate how reliable the field studies are in quantifying the CCN production from NPF events. An inherent weakness in the field studies is that they are Eulerian, the pre-existing particle concentrations can fluctuate cosiderably between the start of NPF and the time that particles reach CCN sizes, creating uncertainty in the number of CCN actually formed due to particle formation and growth. Using models, it should be possible to both quantify the CCN number formed in a regional event and to investigate how well the CCN production can be estimated from increase of CCN numbers in a single gridpoint.

Our response: We agree. The last sentence of this paragraph was modified into the following form "Simple process model simulations might bring new insight into interpreting field measurements, whereas high-resolution regional models could be used to investigate how reliable field studies are in quantifying the CCN production from new particle formation events."

Minor:

- P. 22145, regarding the chemical effects potentially affecting CCN activation, the adsorption activation mechanism (Sorjamaa and Laaksonen, ACP 7, 6175, 2007) should be mentioned. Note that in the adsorption mechanism, the slopes of S* vs D(dry) are different than in the tradiotional Köhler theory (see e.g.Kumar et al., ACP ,11, 8661, 2011).

Our response: We added the following sentence to the end of section 2.1.1: “Insoluble particles, such as fresh dust particles, can be activated into cloud droplets via water adsorption (Sorjamaa and Laaksonen, 2007; Kumar et al., 2011), in which case the relation between Sc and dc is very different from that given by Eq. 2.”

- P. 22158, it is said that "The absolute CCN increase depends mainly on particle formation and growth rates". I would argue that it depends very much also on the pre-existing aerosol that acts as coagulation sink, because the growth rate and the coagulation sink together determine the survival probablity of the particles to CCN size range.

Our response: This is true. In order to avoid misunderstandings, we replaced the sentences “The absolute CCN increase depends mainly on particle formation and growth rates, whereas the relative CCN increase is affected also by primary aerosol particle number concentrations” with the following sentence: “For example, the absolute and relative CCN increases depend in very different ways on primary aerosol particle number concentrations”.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 22139, 2012.