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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Size-dependent particle activation properties in fog during the ParisFog 2012/13 field campaign" *by* E. Hammer et al.

Anonymous Referee #2

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General comments

The work describes observations of interstitial particle and fog droplet size distributions plus measurements of the CCN properties of particles smaller than 300 nm during several fog events near Paris between October 2, 2012 and January 7, 2013. The paper provides useful details of the microphysics and hygroscopicity of particles and their growth to fog droplets in these events. The paper is well written and organized, but I do have some concerns.

First, the authors assume that everything above a threshold was activated without regard to whether a critical diameter was reached or even whether a supersaturation was actually achieved in any of these fogs. These distinctions are important if we are

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to properly understand these processes in fogs. Another questionable assumption is whether the application of a kappa value of 0.14 for all particles above about 300 nm is valid. Details are given in comment 1 below.

I believe some clarification is required about the partitioning of the light scattering between droplets and interstitial particles. See comment 2 below.

Finally, while the technical aspects of the work seem excellent, the paper lacks discussion of some potentially important and scientifically interesting consequences. For example, does the presence of the urban centre and attendant large concentrations of particles impact the fog supersaturation? Did any or all of these fogs truly supersaturate? How might chemical processing in the cloud influence the results? The addition of some discussion points could make this a very interesting paper.

Detailed comments

1. The application of the term 'activate' to those particles on which the largest fog droplets may be inappropriate. In clouds and fogs, the largest of the precursor particles are required to grow to into droplets of very large size before they can be considered 'activated' under the true definition of the term, which is the droplet exceeding its critical diameter. In some clouds, but particularly in fogs where the growth rates are overall smaller due to lower and more variable excess water vapour, the droplets formed around particles with greater hygroscopic mass never grow large enough such that the precursor particles can be classified as activated. This is quite easy to see in an adiabatic parcel model if you compare the sizes of the growing droplets with the critical diameter as a function of time. For particles with critical supersaturations below the maximum cloud supersaturation, you end up with the larger droplets formed on unactivated particles (because they don't have sufficient time for growth) and the smaller droplets formed on the smaller activated particles. This aspect of activation was first pointed out to me many years ago by Bob Charlson. It is a concept that is probably less important for clouds, but potentially important for fogs. Your statement in section 3.5 to

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the effect that minima in the number distributions were not distinct is one indicator that not only may some of the particles classified as activated not be truly activated, all the particles in droplets above the threshold may not be activated; i.e. simply large haze droplets. One literature reference to the process is Phinney et al. (J. Geophys. Res., 108, 4371-4380, 2003), but that is by no means the first reference to this; e.g. see papers by Nenes discussing kinetic limitations. As an example, the 'activation' diameters for your Nov 12 case are given as 261-451 nm (Table 2). Particles of those diameters with relatively high kappa values (e.g. sodium chloride) have equilibrium diameters at 100% RH of 3-7 um, respectively. Your kappa estimates are only directly applicable to particles smaller than your cut diameters, and you show in Fig. 5 that kappa values near the 'activation' diameters were larger, but you dismiss this as an instrument limitation. I do not argue about the increased uncertainty in the CCN measurements at low supersaturations, but it is certainly possible, and perhaps likely, that your assumption that the particles on which the fog droplets form have the same kappa values as particles smaller than 300 nm is incorrect (i.e. why could the fog droplet growth not be on particles in a mode larger than 400 nm that are more hygroscopic, as the data in Fig. 5 suggest?). There are a number of reasons why the larger particles could have larger kappas, including a couple of gas-phase sources: there was more sulphate production in the larger fog droplets, or the scavenging of acidic gas-phase nitrate. Your discussion needs to show that the particles forming the larger droplets (i.e. >2-4 um as in your table 2) are truly activating, or it must acknowledge that the separation that occurs in the 2-4 um range might also result from a modal feature in the pre-fog particle size distribution with higher kappa values. A consequence of the latter is that the effective fog supersaturations are invalid; you are only able to calculate effective supersaturations because of your assumptions. In short, I suspect that what is happening in these fogs is much more complicated than just cooling leading to a supersaturation and an activation threshold.

2. Section 4.3 – In Fig. 10, the area under the mean scattering coefficient curve for the Nov 9 case appears to be much larger above 2.4 um (wet threshold for the case)

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than below it. How then is the integrated scattering coefficient for the hydrated particles (bs,hyd in Table 2) approximately six times higher than the the integrated coefficient for the droplets (bs,drop in Table 2)?

Minor comments:

3. Page 9481, line 25 to page 9482, line 3 – Was the scaling done based on the number concentrations of the part of the total distribution that overlapped with the interstitial distributions?

4. Page 9485. Line 19 – should it be 1.4 um rather than 1um on this line?

5. Section 3.7 – Can you distinguish with your uncertainties the difference between a fog droplet distribution developed at a stable RH of 99.99% and a droplet distribution developed over some minutes at a constant supersaturation of 0.01%?

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 9475, 2014.