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Discussions

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

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We would like to thank the referee for his/her valuable and detailed comments. Our answers to the comments are given below, where the referee comments are given in italics. We also improved the grammar in some sentences without changing the scientific output. Please note that the full captions to the Figures R1-R3 are given at the end before References, while the figures are placed at the very end of the document.

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

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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 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.

We addressed the above general comments in the answers to the detailed referee comments 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.

We addressed and discussed your questions in the paper with the new sections 4.2.1 and 4.2.2, while a detailed analysis of chemical processing and aqueous phase chemistry will be the subject of a manuscript that is currently in preparation (Klemm et al.):

"4.2.1 Effect of local emissions on fog properties

The results shown in Fig. 4 show that a higher particle number concentration while wind directions from the Paris city centre were measured (sector between ~ 0 and ~ 90). However, neither the obtained SS_{peak} nor the κ values show any pattern with

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the wind direction.Laborde et al. (2013) showed, through the combined analysis of aerosol composition and hygroscopicity distribution data measured at the SIRTA site in Winter 2010, that a dominant number fraction of the particles with a diameter of 110 nm often originates from local emissions from within Paris, while the dominant fraction of the particles with a diameter of 265 nm can be attributed to the background aerosol. This size dependence of local influence can be explained by the fact that the size distribution of primary particle emissions from combustion sources peaks at rather small sizes clearly below the maximum of the background aerosol size distribution. The observations of this study showed that only particles with dry diameters larger than $\sim 300 - 500$ nm are able to act as condensation nuclei for fog droplets due to the low peak supersaturation. This finding is consistent with the small number concentration of fog droplets usually measured at the SIRTA site (Elias et al., 2014). Therefore, the local emissions comprised of small particles, don't have a substantial effect on the resulting fog droplet number concentration. The fact that the freshly emitted particles are less hygroscopic than the background aerosol (Laborde et al., 2013), which shifts the activation threshold diameter for local emissions even higher up, further amplifies the dominant influence of background aerosol particles.

Crippa et al. (2013) showed, by parallel measurements of fine particulate matter (PM1) at three measurements sites across Paris in winter 2010, that variations of aerosol mass concentration and composition are dominated by regional scale influence, while local emissions only give a minor contribution. This is consistent with the above-mentioned findings from Laborde et al. (2013), given the fact that the dominant contribution to particle mass and bulk composition comes from larger particles, which are mainly from the background aerosol. The regional scale emission may potentially have some influence on fog properties, for example by growing background aerosol mode particles across the activation threshold dry diameter through condensation of secondary aerosol matter. However, this potential effect cannot be quantified from the data set available here.

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4.2.2 Possible influences of in-fog processes on measured D_{act}

The size distributions of the total, interstitial and droplet residual particles may be altered by in-fog processes such as fog droplet sedimentation, entrainment of subsaturated air, in-cloud scavenging of interstitial particles, coalescence of fog droplets, and droplet processing, which includes condensation of semi-volatile aerosol components or aqueous phase heterogeneous formation of secondary aerosol within the droplet (e.g. Noone et al., 1992; Henning et al., 2014). These processes can potentially lead to a bias in the measured D_{act} compared with the true dry activation diameter at the time of fog formation. A detailed sensitivity analysis, provided in the supplementary information, revealed that the measured D_{act} may be larger (more likely) or smaller (less likely) compared to the true D_{act} due to the effects of in-fog processes. These artefacts are most likely smaller than $\sim 15\,\%$, which is within the specified uncertainty of the measured D_{act}.

The size distributions of the interstitial and droplet residual particles may be altered by in-fog processes such as fog droplet sedimentation, entrainment of subsaturated air, in-cloud scavenging of interstitial particles, coalescence of fog droplets, and heterogeneous chemical reactions in the aqueous phase (e.g. Noone et al., 1992; Henning et al., 2014). These processes can potentially lead to a bias in the measured D_{act} compared with the true dry activation diameter at the time of fog formation. A detailed sensitivity analysis, provided in the supplementary information, revealed that the relative bias of the measured D_{act} due to in-fog processes is most likely smaller than 16\%. This is within the specified uncertainty of the measured D_{act}."

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 un-activated 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 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 μm, 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. $> 2-4$ μm as in your table 2) are truly activating, or it must acknowledge that the separation that occurs in the 2-4 μm 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.

The above referee comment challenges our data analysis approach used to infer an estimate of the critical supersaturation with a series of questions and hypotheses. Here we provide a detailed answer to all these potential issues.

First of all, the referee puts up the hypothesis that supersaturation may never have been reached in the observed fog events and that the particles forming the observed fog droplets never got truly activated (where "activated" means that the droplets grow to sizes larger than their critical droplet size, i.e. larger than the droplet diameter corresponding to the maximal equilibrium supersaturation according to Köhler theory). - We argue that supersaturation and true activation of some CCN to droplets larger than their critical wet diameter must have occurred for the following reason: if we assume that all particles always stayed on the stable side of the Köhler curve (i.e. the RH never exceeded the critical supersaturation of those particles that can activate at the lowest SS), then there is a fixed relationship between the ambient temperature and the ambient RH, for a certain total water content (if we also assume that all particles have reached their stable equilibrium diameter; if some particles would not have reached their equilibrium diameter due to kinetic limitations, then the ambient RH would be larger than that with all particles in equilibrium). This relationship between ambient temperature and RH depends, besides the total water content, also on the number concentration, size distribution and hygroscopicity of the aerosol particles, as they have a

dampening feedback effect on the RH increase with decreasing temperature through the water that condenses on the growing particles. However, the key fact is that particles on the stable side of the Köhler curve, which have a clearly defined equilibrium diameter, can by no means buffer the RH at 100 % by uptake or release of water vapor, if the ambient temperature varies. Thus, if the ambient air cools below the temperature that corresponds to 100 % RH, then the RH will exceed 100 % and the air becomes supersaturated. The peak supersaturation will only be reached when either the cooling stops, or when a sufficient number of droplets got truly activated such that they can pull the RH back down towards 100 % (truly activated droplets can simply adjust their size to accommodate all excess water vapor until the RH drops down to their equilibrium RH which asymptotically approaches 100 % with increasing droplet diameter in the range above the critical diameter). Consequently, true supersaturation and droplet activation must occur during the formation process of developed fog, as it is absolutely not plausible that the cooling always stopped virtually exactly at the temperature corresponding to 100 % RH (at least ~ 100 % must have been reached to explain the observed droplet diameters, even with assuming very hygroscopic particles, as equilibrium droplet diameters sharply drop for RH slightly lower than 100 %).

We added the following paragraph in Sect. 4.2:

"The droplet diameters observed during the events are in the range where very large and hygroscopic particles can exist in stable equilibrium. This brings up the question whether supersaturation and true droplet activation occurred at all during fog formation. We are confident that this was the case as particles on the stable side of the Köhler curve, which have a clearly defined equilibrium diameter, cannot buffer the RH at 100 % by uptake or release of water vapor, if the ambient temperature varies. Thus, if the ambient air cools below the temperature that corresponds to 100 % RH, then the RH will exceed 100 % and the peak supersaturation will only be reached when the activated droplets have taken up enough water to reduce the RH back to 100 % RH."

The referee also asks whether it could be possible that only very few rather large and

very hygroscopic CCN with critical droplet diameters above $\sim 5\,\mu m$ – e.g. a few large externally mixed sea salt particles always buffered the RH between 100 % and their extremely low critical supersaturation, either by growing on the stable side of the Köhler curve or by being truly activated? – We do not believe that this is possible, firstly because the number of such particles is too small to accommodate all water corresponding to observed LWCs reaching up to $\sim 0.25\,g/m^3$, and second, as these few fog droplets, even if activated, would offer a very small condensation sink for the excess water vapor under continued cooling, given the fact that the driving force for diffusion of water vapor to the droplets, i.e. the supersaturation minus the equilibrium RH, would be extremely small under such a scenario. Therefore, very few CCN are not able to buffer the RH at ~ 100 % under continued cooling due to their small number and kinetic limitations. Instead, in order to achieve efficient buffering of the RH at ~ 100 %, the supersaturation will initially increase to somewhat higher values such that faster droplet growth and activation of additional CCN with somewhat higher critical supersaturation occurs, until the condensation sink becomes sufficiently efficient.

The referee correctly points out that large and very hygroscopic CCN with critical droplet diameters above $\sim 5\,\mu m$ may potentially remain in stable equilibrium even under developed fog conditions, coexisting with smaller and/or less hygroscopic CCN that got truly activated. The reason for this phenomenon are kinetic limitations: depending on the conditions, the CCN with smaller critical wet diameter require less time to grow across their critical size compared to the CCN with much larger critical diameter (this is in line with the argumentation in the previous paragraph). Now, let us compare a droplet formed on a large good CCN that has a large critical wet diameter with a smaller/poorer CCN that has a smaller critical wet diameter. The former droplet will be larger than the latter at any time and will always become activated before the latter crosses the critical wet diameter of the former. In this sense, a large good CCN will contribute at least as much to the LWC as a smaller or poorer CCN, even if the good CCN isn't yet truly activated. Here it is important to note that this phenomenon does also not invalidate our data analysis approach, which uses the minimum in the droplet

size distribution between the modes from unactivated droplets in stable equilibrium and truly activated droplets, because the abovementioned "good CCN" would simply show up in the uppermost tail of the droplet size distribution.

We added in Sect. 4.2: "...fog droplet activation, as detailed in Sect. 3.5. It is important to note that large and very hygroscopic CCN with critical droplet diameters above $\sim 5\,\mu m$ may potentially remain in stable equilibrium, even under developed fog conditions, coexisting with smaller and/or less hygroscopic CCN that got truly activated (e.g. Phinney et al., 2003). The reason for this phenomenon are kinetic limitations (Nenes et al., 2001): depending on the conditions, the CCN with smaller critical wet diameter require less time to grow across their critical size compared to the CCN with much larger critical diameter. However, this phenomenon does not invalidate data analysis approach for retrieving the D_{thres}^{wet}, which uses the minimum in the droplet size distribution between the modes from unactivated droplets in stable equilibrium and truly activated droplets, because the large CCNs would show up in the uppermost tail of the droplet size distribution."

The referee further puts up the question, whether larger κ values than those measured at diameters $< 310\,nm$ should be applied for inferring the effective peak supersaturation. It is indeed true that droplet processing, e.g. nitric acid uptake into the droplet or heterogeneous sulphate production within the droplet, can potentially add hygroscopic material to the activated particles, thereby increasing their hygroscopicity compared to the non-activated particles. Such "cloud processing" of the larger particles that acted as CCN could potentially increase their hygroscopicity compared to the smaller particles with sizes just below the activation threshold. However, the extremely sharp increase of the measured κ values between 290 nm and 310 nm dry particle diameter is unphysical (Fig. 5), unless there would have been a very sharp activation cut-off at 300 nm dry diameter in all fog events. This is not plausible and therefore much more likely caused by the increased CCNC uncertainty at very low supersaturation as already argued in the ACPD version of the manuscript. (If the activation was indeed happening

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at $\sim 300\,nm$ dry diameter, then this would be an argument for the validity of our D_{act}^{lower} ($\sim$ 261–364 nm; Table 1) and corresponding SS_{peak}^{upper} (0.08–0.05 %) values.) Despite the fact that the measured increase of κ values between 290 and 310 nm dry particle diameter is within experimental uncertainty, we do agree that one could consider using somewhat larger κ values for inferring the effective peak supersaturation, due to the above-mentioned processes that can cause a size-dependence of particle hygroscopicity. This would have virtually no effect on the resulting SS_{peak}^{lower}, as the κ value is only used to extrapolate the Köhler curves from 100 % RH to ~ 100.03 % RH (this fact is already stated on p. 9488 of the ACPD-version of this manuscript). Figure R1 below shows a set of Köhler curves with different κ values that all go through exactly the same D_{thres}^{wet} of 2.6 µm at $RH = 100$ % (indicated by the black circle in panel A), which represents the median of all observed D_{thres}^{wet}. The maxima of the different Köhler curves (see panel B), which represent the corresponding SS_{peak}^{lower}, are virtually equal for all κ values. Thus, the SS_{peak}^{lower} inferred from the observed D_{thres}^{wet} are within experimental uncertainty, completely independent of the κ value used for the extrapolation of Köhler curve. However, the corresponding D_{act}^{upper}, which is obtained by extrapolating the Köhler curve from 100 % to 0 % RH, depends on the assumed κ value. Fig. 1C shows that assuming a κ of ~ 0.34 (cyan line) provides a corresponding D_{act}^{upper} of 350 nm, which would be in close agreement with the observed D_{act}^{lower} (when operating the 2-µm-cut-off). Our other approach to infer the effective peak supersaturation works in the other direction: the activation cut-off diameter of the dry particles, D_{act}^{lower}, is measured, and the corresponding supersaturation, SS_{peak}^{upper}, is obtained by "extrapolating the Köhler curve" from 0 % to > 100 % RH using a certain κ value. If we were assuming a higher κ value for this extrapolation, e.g. κ=0.34 instead of κ=0.14, then the SS_{peak}^{upper} inferred from the 2-µm-cut-off measurements would become approximately equal to SS_{peak}^{lower}. In summary, using a somewhat larger κ value, as suggested by the referee would bring the SS_{peak}^{upper} and SS_{peak}^{lower} (as well as D_{act}^{lower} and D_{act}^{upper} to closer agreement. However, in the interest of providing a conservative estimate of the likely range of the effective peak supersaturation in the fog, we keep using the lower κ value of 0.14, as measured

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by the CCNC for particles with dry diameters < 300 nm. Note, the inferred SS_{peak}^{lower} also depend on the temperature at activation (3 % relative change per 10 C temperature change).

The statement in Sect. 3.7 has been modified slightly: "A relative uncertainty of ± 3 % in SS_{peak}^{lower} arises from a temperature uncertainty of ± 10C. SS_{peak}^{lower} is virtually independent of the kappa value used to infer it, because in this approach the κ–Köhler theory is just used to extrapolate the Köhler curve over a very small change in RH and droplet diameter. On the other hand, the inferred D_{act}^{upper} would decrease by ~ 20 % if a kappa value of 0.3 was assumed instead of 0.14."

And we added in Sect. 4.2: "...range between ~ 0.016 to 0.113 %.

As described in Sect. 3.5, a rather low κ value of 0.14 as measured at dry diameters < 300 nm is used to infer SS_{peak}^{lower} from D_{thres}^{wet} and SS_{peak}^{upper} from D_{act}^{lower}. It is possible that particles in the dry diameter range above 300 nm have somewhat larger κ values due to potential addition of hygroscopic material via fog processing. By assuming a higher κ value of e.g. 0.34 instead of 0.14, the SS_{peak}^{upper} inferred from the 2-µm-cut-off measurements would become approximately equal to SS_{peak}^{lower} (which is virtually independent of κ, see Sect. 3.7). Therefore, using a somewhat larger κ value would bring the SS_{peak}^{upper} and SS_{peak}^{lower} (as well as D_{act}^{lower} and D_{act}^{upper}) to closer agreement. However, in the interest of providing a conservative estimate of the likely range of the effective peak supersaturation in the fog, the lower κ value of 0.14 is used."

The referee also asks whether the bimodal feature of the droplet size distribution simply occurs because the aerosol is an external mixture of more and less hygroscopic particles. – It is unlikely that a broad monomodal dry size distribution would grow to a bimodal droplet size distribution at high RH, also for an external mixture with a bimodal hygroscopicity distribution, for the simple reason that the convolution of a broad size distribution with a bimodal "growth kernel" simply produces a somewhat broader and distorted size distribution, unless the "growth convolution kernel" (i.e. the bimodal

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growth factor distribution) would have a large geometric standard deviation compared to the geometric standard deviation of the dry size distribution. If the bimodal feature of the droplet size distribution was to be attributed to an external mixture, then we have to distinguish between two possible cases. The first alternative would be that the smaller mode represents less hygroscopic particles in stable equilibrium while the larger mode represents more hygroscopic particles that became activated. In this case, the "D_{thres}^{wet}–approach" would provide the correct peak supersaturation (within the uncertainty of determining the exact value of D_{thres}^{wet}). Here it is to be remembered that we simply have to find the largest droplet size (at RH$= 100$ %) that did not get activated, while the κ value of the CCN that formed these particles is irrelevant. The other potential alternative is that both modes would be in stable equilibrium. The observation of "stable" droplets at diameters as large as 8 µm (and even larger), would then imply that the peak supersaturation would have been extremely low (< 0.02 %). However, this small driving force for condensation would turn it very difficult for the droplets to grow to such large diameters, which makes this an unlikely explanation of the observations.

Summarizing the above discussion: We are confident that supersaturation and true activation of fog droplets occurred and that our two approaches provide a fair estimate of the supersaturation range into which the true fog supersaturations fall. At the same time we do acknowledge that our analyses do not allow for an exact measurement of the true supersaturation, as we state in the manuscript.

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 µm (wet threshold for the case) than below it. How then is the integrated scattering coefficient for the hydrated particles ($b_{s,hyd}$ in Table 2) approximately six times higher than the integrated coefficient for the droplets ($b_{s,drop}$ in Table 2)?

Figure 10 shows results from the fog event F9 which was on November 12. A discrepancy of a factor of 1.6 remains when comparing the original Fig. 10 with the data provided in Table 2 for event F9. This discrepancy, which slipped our attention,

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can simply be explained with different approaches to "average" the two types of time-resolved data over the whole fog event. The "median size distribution" shown in the original Fig. 10 was obtained by calculating the median separately for each size bin, whereas $b_{s,hyd}$ and $b_{s,drop}$ in Table 2 are medians calculated from the corresponding values obtained by integrating individual size distributions (having a time resolution of 12 minutes). These two different averaging approaches can, depending on the temporal variability of the shape and area of the size distribution, provide different results (e.g. event F9 shown in Fig. R2), while similar results are sometimes obtained (e.g. event F15 shown in Fig. R3).

We consider the tabulated values, representing the statistics of the time-resolved $b_{s,hyd}$ and $b_{s,drop}$ values as relevant and kept them as is. Therefore we changed the averaging approach to obtain a median size distribution for the whole fog event for graphical representation. The scattering coefficient size distribution shown in the revised Fig. 10 is obtained by calculating the bin-wise median of the individual normalized size distributions, followed by scaling with the median of the individual total scattering coefficient values ($b_{s,tot}$; obtained by integrating the individual scattering coefficient size distributions). The values of $b_{s,hyd}$ and $b_{s,drop}$ calculated from the median scattering coefficient size distributions averaged with the new approach agree much better with the tabulated values (the average relative difference is within 8 %). This is also clearly seen for the two example fog events presented in Figs. R2 and R3.

Here we have to add that the values of $b_{s,hyd}$ and $b_{s,drop}$ in Table 2 got somewhat smaller because we corrected a minor error in the original calculations. We therefore adjusted in the conclusions: "The hydrated particles were estimated to contribute on average $\sim 41 - 68$ % to total light-scattering during the developed fog periods."

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?

No scaling was done. We clarified this description: "The mean size distributions measured behind either inlet agreed well during fog-free periods as both inlets deliver the total aerosol under these conditions. However, the total number concentration differed by up to 15 % between consecutive scans due temporal variability of aerosol concentration. To minimize the effects of aerosol variability on the calculated activation curves, the total size distribution was compared with the average of the two interstitial size distributions measured immediately before and after."

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

Yes, we changed it to 1.4 μm.

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 %?

In the detailed answer to the first major comment provided above, we argued that supersaturation and true activation occurs in the developed fog events and that the activated droplets buffer the RH at ~ 100 % during further temperature variations. The approach described in Sect. 3.7 infers the peak supersaturation from the largest equilibrium droplet diameter observed in the developed fog. The RH in the developed fog is not exactly known, which adds to the uncertainty of the inferred SS_{peak}^{lower} values. This is discussed in the revised Sect. 3.7:

"The RH in the developed fog will stabilize slightly above 100 % RH, depending on the fog droplet sizes that are reached while buffering the excess water. The fact that the RH in the developed fog is not exactly known, adds some uncertainty to the inferred SS_{peak}^{lower} values. If the RH in the developed fog was assumed to be as high as 100.03 % (i.e. 0.03 % SS) instead of exactly 100.00 %, then the SS_{peak}^{lower} would increase by ~ 17 % (relatively). In the interest of providing a conservative estimate for SS_{peak}^{lower}, we assume

an RH of exactly 100 % for this approach."

Figure Captions:

Figure R1: Köhler curves for different pairs of dry diameter and κ value, chosen in such a manner that all curves cross RH= 100 % at a droplet diameter of 2.6 µm. This intersect is marked by the black circle in panel A, which shows the range between 99.9 % and 100.05 % RH. B) Zoomed in to the critical supersaturation and C) zoomed out to the whole range between 0 % and 100 % RH.

Figure R2: For the fog event F9 on 12 November 2012: Averaged merged scattering coefficient size distribution (grey line) from SMPS and WELAS. The dotted line depicts the median of the normalized scattering coefficient size distributions multiplied by the median total scattering coefficient ($b_{s,tot}$). The separation of the hydrated aerosols (yellow) from activated droplets (green) is done at the local minimum representing D_{thres}^{wet}.

Figure R3: Same as Fig. R2 for the fog event F15 on 30 November 2012.

References

Crippa, M., DeCarlo, P. F., Slowik, J. G., Mohr, C., Heringa, M. F., Chirico, R., Poulain, L., Freutel, F., Sciare, J., Cozic, J., Di Marco, C. F., Elsasser, M., Nicolas, J. B., Marchand, N., Abidi, E., Wiedensohler, A., Drewnick, F., Schneider, J., Borrmann, S., Nemitz, E., Zimmermann, R., Jaffrezo, J.-L., Prévôt, A. S. H., and Baltensperger, U.: Wintertime aerosol chemical composition and source apportionment of the organic fraction in the metropolitan area of Paris, Atmos. Chem. Phys., 13, 961-981, doi:10.5194/acp-13-961-2013, 2013.

Elias, T., Dupont, J.-C., Burnet, F., Haeffelin, M., Jolivet, D., and Hammer, E.: Aerosols affecting the radiative budget during the fog life cycle: microphysical properties and extinction coefficient, Atmos. Res., in review, 2014.

Henning, S., Dieckmann, K., Ignatius, K., Schäfer, M., Zedler, P., Harris, E., Sinha, B., van Pinxteren, D., Mertes, S., Birmili, W., Merkel, M., Wu, Z., Wiedensohler, A., Wex,

H., Herrmann, H., and Stratmann, F.: Influence of cloud processing on CCN activation behaviour in the Thuringian Forest, Germany during HCCT-2010, Atmos. Chem. Phys., 14, 7859-7868, doi:10.5194/acp-14-7859-2014, 2014.

Laborde, M., Crippa, M., Tritscher, T., Jurányi, Z., Decarlo, P. F., Temime-Roussel, B., Marchand, N., Eckhardt, S., Stohl, A., Baltensperger, U., Prévôt, A. S. H., Weingartner, E., and Gysel, M.: Black carbon physical properties and mixing state in the European megacity Paris, Atmos. Chem. Phys., 13, 5831-5856, doi:10.5194/acp-13-5831-2013, 2013.

Nenes, A., Ghan, S., Abdul–Razzak, H., Chuang, P. Y. and Seinfeld, J. H., Kinetic limitations on cloud droplet formation and impact on cloud albedo. Tellus B, 53: 133–149. doi: 10.1034/j.1600-0889.2001.d01-12.x, 2001.

Noone, K. J., Ogren, J. A., Hallberg, A., Hansson, H. C., Wiedensohler, A., and Swietlicki, E.: A statistical examination of the chemical differences between interstitial and scavenged aerosol, Tellus B, 44, 581–592, doi:10.1034/j.1600-0889.1992.t01-1-00011.x, 1992.

Phinney, L. A., U. Lohmann, and W. R. Leaitch: Limitations of using an equilibrium approximation in an aerosol activation parameterization, J. Geophys. Res., 108, 4371, doi:10.1029/2002JD002391, D12, 2013.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/14/C6042/2014/acpd-14-C6042-2014-supplement.pdf
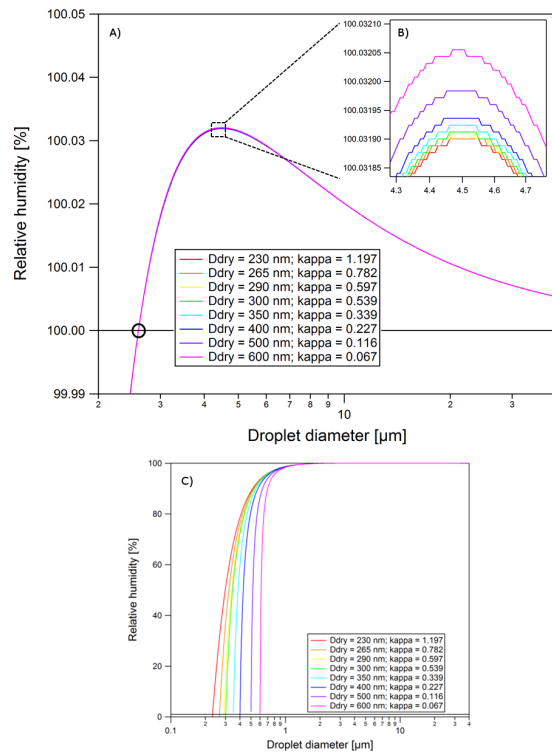
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Fig. 1. R1: K\"{o}hler curves for different pairs of dry diameter and κ value, chosen in such a manner that all curves cross RH$=100\,$\% at a droplet diameter of $2.6\,\unit{{\mu}m}$. This intersect is
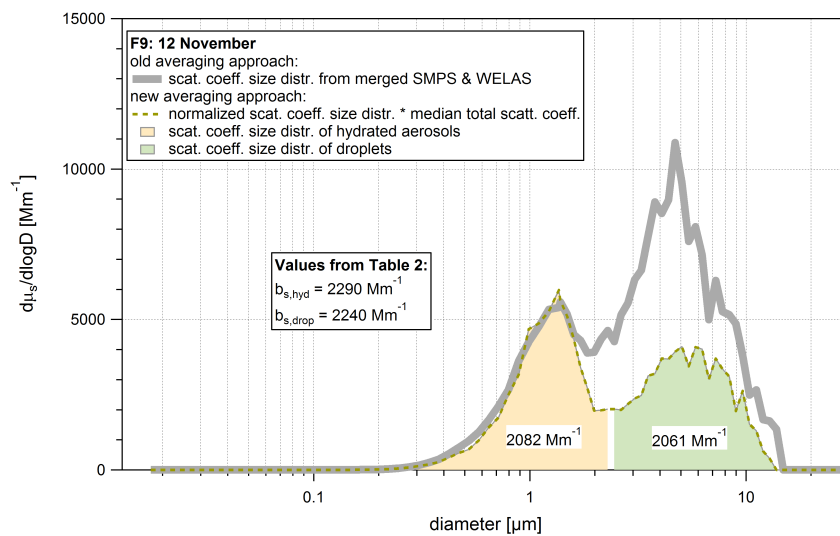
Fig. 2. R2: For the fog event F9 on 12 November 2012: Averaged merged scattering coefficient size distribution (grey line) from SMPS and WELAS. The dotted line depicts the median of the normalized scattering
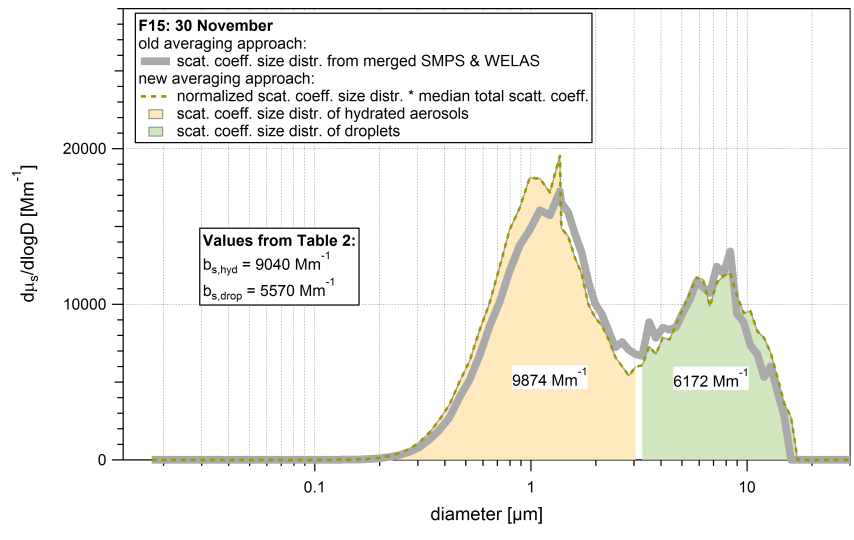
Fig. 3. R3: Same as Fig. R2 for the fog event F15 on 30 November 2012.

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