

## REVIEWER 3

*Authors thank the reviewers for their useful and very interesting comments and for the time spent to review our work. Reviewers have common concerns which we addressed to the best of our possibilities:*

- *We clarified the introduction, the method and the results sections by moving some figures to supplementary material and by re-organizing the text.*
- *The abstract has been rewritten in order to highlight the most significant results.*
- *A discussion has been added in the paper on the limitations of our methodology to derive aerosol and fog microphysical properties and on the limitation of this work due to the location of the campaign and the weak chemical variability.*
- *We also added a discussion on the relevance of our results for numerical weather prediction.*
- *Cyrielle Denjean have been added to this paper as coauthors to acknowledge their contribution to discussion related to these correction.*

**1. One of the main conclusions is that, based on the range of hygroscopicity factors  $C1(k)$  observed, aerosol composition is unimportant in determining  $N_{act}$ . However, the (large) error bars in Figure 9c must be considered only by keeping in mind that the  $k$  values for the SS smaller than 0.1 were determined only through an extrapolation.  $N_{act}$  is actually one order of magnitude smaller than the smallest concentrations measured by the CCN counter, therefore caution must be used in extrapolating the CCN data to derive hygroscopicity factors for such small subset of particles. For these reasons, I am not convinced that the scatter plot in Figure 9c provides enough evidence that hygroscopicity does not contribute to control the activation of aerosols to fog droplets.**

On figure 8 and 9 and on table 3 ones can see that the errors bars of the  $kappa$  values are not as important and won't change the interpretation. Then looking at Figure 7, moving from violet blue distribution to black ones (SMPS distribution growth with  $kappa$ -Köhler theory to WELAS distribution), only depends on  $kappa$ . The excellent connection between the SMPS and Welas size distributions (also true for episode) means that  $kappa$ 's determination is not that bad.

In addition ones can see on figure 10.a) that for a given  $S_{peak}$ , different activation fraction can correspond and they differ according to  $kappa$ . Thus  $kappa$  does have an influence on the activation fraction. But only when looking at the ratio. That means that the  $N^*$  (which divide  $N_{act}$ ) is important, and actually it can be seen as a proxy for the droplet size distribution above 200 nm.

We added in section Impact of aerosol particles on fog droplets concentration :

*“ Correlation when  $N^*$  was calculated from the range of critical diameter inferred during the campaign (150 to 400 nm). This suggest that  $N^*$  using a single critical diameter can be considered as a good proxy of the number of activable particles. This can be understood since the supersaturation occuring in fog has a narrow range of value. CCN at a supersaturation between 0.02 and 0.11 % are mostly composed of accumulation mode particles. »*

We added in the conclusion:

*“Concentration of activated particles using a single critical diameter can be considered as a good proxy of the number of activable particles.”*

**2. The Authors present a new methodology to separate activated and non-activated fog droplets (Section 3.2 and Figure 4). This is based on an iterative calculation for matching pairs of aerosol parameters ( $N_{ccn}$  and  $D_w$ , where  $N_{ccn}$  was estimated by integrating the SMPS size-distributions and using the Koehler theory and the  $k$  values from the CCN measurements to derived wet diameters at the point of activation) with pairs of fog parameters ( $N_{droplets}$  and  $D_w$  measured by WELAS+FM) and making  $N_{ccn}$  to converge to  $N_{droplets}$ . This is an interesting approach, but it is based on strong assumptions: a) it ignores that the  $D_w$  distribution in the WELAS+FM is much broader than the  $D_w$  of aerosols at the activation point because this point is reached only instantaneously and activated droplets grow to larger diameters afterwards; b) it ignores feedbacks of the activated droplets on SS so that all aerosols are assumed to activate at the same SS. In other words, the approach ignores kinetic effects, it is based on a purely equilibrium state, and neglects the variability of SS (and fog forms like inside a CCN counter where SS is externally controlled). The Authors have not acknowledged these caveats, and, in my opinion, they have not adequately presented a critical analysis of their approach. Only two examples are presented (f6 and f20 in Figure 7), where the approach seems to work, but what about other cases, such as the strange f22? For all these case, it would be very interesting to compare the present approach with alternative ones based on, e.g., mode fitting (Elias et al., 2015, etc.).**

**a) it ignores that the  $D_w$  distribution in the WELAS+FM is much broader than the  $D_w$  of aerosols at the activation point because this point is reached only instantaneously and activated droplets grow to larger diameters afterwards;**

Reviewer is right on the point that distribution of WELAS+FM must have reach larger diameter. It is the reason why we use approximatively 1h laps time after the fog beginning as explained in section “method to determine fog activation properties ». Then droplets distribution continues to evolve but that should be the point of a following paper.

**b) it ignores feedbacks of the activated droplets on SS so that all aerosols are assumed to activate at the same SS. In other words, the approach ignores kinetic effects, it is based on a purely equilibrium state, and neglects the variability of SS (and fog forms like inside a CCN counter where SS is externally controlled). The Authors have not acknowledged these caveats, and, in my opinion, they have not adequately presented a critical analysis of their approach. Only two examples are presented (f6 and f20 in Figure 7), where the approach seems to work, but what about other cases, such as the strange f22? For all these case, it would be very interesting to compare the present approach with alternative ones based on, e.g., mode fitting (Elias et al., 2015, etc.).**

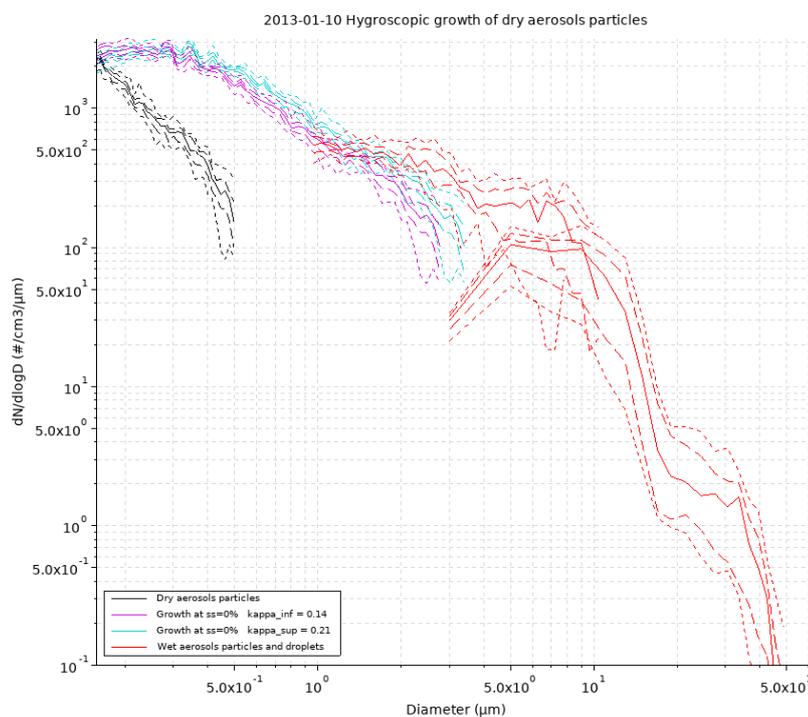
In the text we wrote:

“These values are slightly higher than values reported by Hammer et al. (2014),  $D_w$  50th = 2.6

$\mu\text{m}$ , but consistent with Elias et al. (2015) results  $D_w = 4 \pm 1.1 \mu\text{m}$  who determined it from a November 2011 dataset as the intersection between the two log-normal distributions fitting the particle volume distribution measured by the WELAS. Such a gap between hydrated and activated particles can be seen in figures 7-a) and b) and it corresponds fairly well to the average wet activation diameter derived for such cases that are equal to 4.41 and 2.62  $\mu\text{m}$ , respectively.”

Our results are consistent with the other studies and indeed Figure 7 shows that wet diameter correspond rather well to the intersection between two modes. Same behaviour is observed with other case.

For the f22 case, correspondence is rather good too considering that wet diameter is included in the range [0,98-2,65]  $\mu\text{m}$ .



Note that the supersaturation we are talking about here, is the critical supersaturation peak. That is obviously not the critical supersaturation of each individual aerosols particles but the supersaturation needed to activated the smallest aerosols particles becoming a droplet.

We add in the session Fog activation properties:

“(which refers to the supersaturation needed to activate the smallest activated aerosols particles)”  
(talking about supersaturation)

### 3. The method used to estimate $N_{act}$ from aerosol measurements relies on the SMPS

**size-distributions. However, instrumental uncertainties in sizing and counting for particles larger than 400 nm should be taken into account. In addition, the discussion that the Authors present about mixing state is oversimplified as a certain extent of external mixing is common in polluted environments. In this referee's opinion, there is a lack of adequate online aerosol instrumentation for probing concentrations, composition and mixing state in the size range which is the most critical for fog droplet activation: that of large accumulation mode particles.**

Concerning particles between 400 nm and 496 nm, agreement between the SMPS and Welas size distributions, give us confidence on the measurements.

We had actually no way to determine dry particles larger than 496 nm. However as there should be very few particles above 496 nm (considering that the concentration decrease from ~200nm), we guess the impact to be weak. According to Franck, 1998, large accumulation mode particles could be present into the fog as big hydrated particles not activated. We don't think they will have a notable impact on droplets concentration, but depending on their size, they could initiate some processes as collision-coalescence. We are also convinced that future studies on fog activation should include « dry » instrumentation above 500 nm.

We agree with the reviewer about the lack of clarity on the mixing states.

We add in the method section:

*“However atmospheric aerosols are frequently externally mixed with particles of different sizes exhibiting different chemical composition. Discussion on this hypothesis is performed in the method validation section.”*

And move the discussion on the mixing state in the method validation section.

We add in the method section:

*“In addition, number concentration of aerosol particles larger than 496 nm was assumed to be negligible in our calculation.”*