

REVIEWER 4

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 papier as coauthors to acknowledge their contribution to discussion related to these correction.*

The number of figure is high and I would suggest to move some figures to the supplementary material. For examples Fig 2 used to validate the droplet size distribution measurements can be moved in supplementary. Figure 3, as well, can be moved to supplementary, since the variability of Na and NFM is already clear in Table 2. Figure 5 and 6 can be moved to supplementary, as well, as an example to show the variability of particle size distribution and the variability of kappa.

Move to supplementary

The paper deals with fog formation considering only fog droplets formed in super saturated conditions, i.e. at RH larger than 100%. Thus, the authors analysed the aerosol and fog microphysical measurements using the k-kohler theory, derived from a parametrisation of the Kohler theory. Nevertheless, Charlson et al. 2001 claimed that “soluble gases, slightly soluble solutes, and surface tension depression by organics also influence the formation of cloud droplets in a manner unforeseen by Kohler”. They concluded that “clouds or fogs with micrometered-sized droplets may exist even though the droplets have not undergone traditional activation and even though the ambient relative humidity never exceeds 100%” It would be useful if the authors could discuss their results at the light Charlson et al. conclusions, which strongly depends on the level of aerosol and gas pollutant concentration. (Charlson et al. 2001, Reshaping the theory of cloud formation, Science, Vol. 292, Issue 5524, pp. 2025-2026, and reference therein)

Surface-active species have indeed the potential to lower surface tension of a growing droplet, thereby increasing the critical supersaturation and the CCN activation efficiency. This effect has largely been attributed to organics when the organic concentration in the aerosol population is

sufficiently large (Facchini et al., 1999; Noziere et al., 2014). The extent to which this will occur is strongly dependent upon the specific molecular properties of the organic molecules, and traditional surfactants (such as fatty acids) can actually have a negligible impact on CCN activation (Forestieri et al. 2018). Moreover to influence aerosol activation, the average properties of surface-active organic molecules must differ substantially from the long-chain fatty acids having either smaller molecular volumes or larger molecular areas. Exploring these aspects would need to combine CCN measurements and a detailed analysis of the molecular species contained in organic particles. This is out of the scope of this manuscript.

We add in the method section:

“Moreover, properties (surface tension) of atmospheric aerosols could be modified by surfactant. This effect has largely been attributed to organics when the organic concentration in the aerosol population is sufficiently large (Facchini et al., 1999; Noziere et al., 2014). The extent to which this will occur is strongly dependent upon the specific molecular properties of the organic molecules. Unfortunately exploring these aspects would need to combine CCN measurements and a detailed analysis of the molecular species contained in organic particles. This is out of the scope of this manuscript”

Fog with micrometer-sized aerosols particles not activated could exist at ambient relative humidity under 100% as also noticed by Franck, 1998. Unfortunately we don't have any measurements of particles larger than 500 nm, so we can not evaluate this effect. All we can guess if we follow the shape of the lognormal distribution for the dry aerosols distribution, is that concentration of particles larger than 500 nm may be very few numerous and should not impact the determination of the droplets concentration.

We have added in the method section:

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

The authors conclude that particles composition is less determinant than particles size for the number of activated particles. Figure 4c is used to derive such conclusion, since no correlation is observed between N_{act} and κ . Is it possible that particle hygroscopicity has a stronger influence for small activation diameter than for large activation diameters? Does κ explain the scatter of data points in fig 9b at low activation diameter (<0.35)?

Scatter in Figure 9 b) is explained by aerosols particles diameter distribution as show Fig 10.b).

On this figure, for an equal N_{act} we don't have the same D_d , meaning that the shape differs. However looking at the ratio N_{act}/N^* on Fig 10.b), correlation is rather good, that suggest that the shape differs more on intensity than on form. That would also suggest that on a given diameter range, results on dependencies between N^* and N_{act} would be the same whatever the threshold diameter. Indeed sensibilities tests made with threshold at 250 nm, 300 nm or 400 nm confirms it. Moreover, as figure 11 shows it, more aerosols particles, less high supersaturations, that suggest a 'control' of the supersaturation values by the aerosols concentration.

Sentence must be added in the text to explain that point.

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. »*

Kappa could have a larger influence for stronger supersaturation and smaller stronger diameter but range of supersaturation is limited in fog. And Fig 13 shows that at 0,1% supersaturation, Kappa get influence but is not the main factor controlling CCN.

One additional evidence to prove that “size matters more than chemistry” is the analysis reported in figure 10b. The graph shows that the ratio of N_{act} over $N_{>200}$ decreases with the activation diameter increase. This is obvious considering the log normal shape of particle size distribution and derived form the assumption that all particles larger than 200 nm can be activated, i.e. on the assumption that “size matters more than chemistry”. The result of the analysis is biased by the starting assumption.

If we consider that kappa have a stronger influence than shape and that the “biggest” shape have also the higher kappa then activation should be more important for the “biggest” shape and D_d could be smaller. Thus the ratio N_{act}/N^* would be inverted.

Authors consider only particles larger than 200 nm because activation only concerns the accumulation mode, and we wanted to avoid the pollution of the smallest particles [~ 10 nm] that will never be activated and will have a very low hygroscopic growth. We thus conclude than shape of particles above 200 nm matters more than chemistry. We added some modifications on the text about that point.

MINOR COMMENTS

The manuscript concludes that aerosol size distribution impacts fog microphysics more than chemical composition. Please discuss if the limited variability of the k parameter observed during the experiment can bias this conclusion

Kappa vary between 0.1 and 0.3. Repartition in class of kappa on this range allows to show the impact of kappa of Figure 10.a).

We agree that our conclusion is valid on the kappa range observed in our study, and our conclusion can differ for other kappa values. We add a discussion on the limitation of our results in the conclusion. However on the 23 cases observed (and selected according to the instruments limitation) during three winter, aerosol size distribution impacts fog microphysics more than chemical composition.

We agree that reservation should be issued about that.

We add in the text:

“Nevertheless, some reservation must be issued on that point. Aerosol size distribution impacts fog microphysics more than chemical composition on the measure range on the hygroscopic parameter during the three winters of observations. Higher or lower values of this parameter could bias this

conclusion. Other measurements of aerosol and fog microphysics in other environmental conditions would be needed to pursue this issue.”

Page 5: do the authors see a difference in kappa values for easterly and westerly flow conditions. Is the origin of air masses reflected in kappa variability?

We did not observe difference in kappa values for the different wind conditions. As mentioned above, a discussion on the limitation of the environmental conditions observed during the field campaign have been added in the conclusion.

We add in the conclusion:

“Other measurements of aerosol and fog microphysics in other environmental conditions would be needed to pursue this issue.”

Technical corrections

Table 1: Done

Page 5: Done

Page 6: Done

Page 7: Done

Page 10: Done

Page 10: Done