

## REVIEWER 1

*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.*

**Second parag in Discussion is not good one, people look at the fog droplet spectra and by definition droplets are above the 1 micron usually. What you did doesn't explain what is wrong with this. You also state in same parag that Nd was lower in your case..... why? Is this because you have measured drizzle but not fog droplets???? You need to show time series of Vis and summarize the results.**

Droplets are defined as hydrated particles whose diameter is above the activation diameter, it is the Köhler theory.

We add in the introduction :

*« Water droplets are also the particles whose diameter are higher than the critical threshold corresponding to that critical supersaturation. »*

We add a discussion on that point on the discussion session:

*«In a modelling purpose, distinguishing hydrated aerosols particles from droplets allows to calculate an accurate repartition of the vapour deposition on the droplet size distribution. Indeed, behaviour of hydrated particles and droplets is quite different, the second one grow as long as there are exceeding vapor while the first one stay at an equilibrium diameter. The second one may grow enough to produce drizzle and have strong interaction with radiation. A clear distinction should improve the representation of processes in numerical weather prediction model and so on the visibilities forecast.»*

A sentence have also been added in the discussion section to lighten the meaning of activated particles *« To our knowledge, no other study has retrieved experimentally concentration of activated particles (and thus droplets) ».*

Nd is rather low for a semi urban conditions, semi urban conditions being generally polluted conditions, an high loading in aerosol is expected. Experimentals studies made in China, for example Liu, 2017, reported values as high  $1000 \text{ cm}^{-3}$ . We measure concentration as high as  $150 \text{ cm}^{-3}$ , which is quite lower. Drizzle is a particle issued from collision, or it is actually considers unlikely that collection happens for diameter lower than  $40 \mu\text{m}$  (Berry et Reinhard, 1974).

However Xue et al, 2008 considers that could happens for diameter lower than 20  $\mu\text{m}$  and Niu et al, 2010, Zhao et al., 2013, Lu et al., 2013 suggest through processes studies that collision could be presents in fog. Still we do not think that the particles we measure between 2  $\mu\text{m}$  and 50  $\mu\text{m}$  are from coalescence, usually a gap in the droplet size distribution is viewable (see stratocumulus droplets size distribution).

Concerning the visibility, the threshold values used to select fog events are already presented in section 2.2

**You need to state how did you average your obs and why? Then, how did you come up with your plots if they are acceptable?**

Observation have been averaged on different time laps. This is specified in the caption of each figure.

**On pag 15; last parag., your values are much lower compared to models, what this tells you? Either model doesn't do good job or your obs are not good.**

When comparing model and observation, one can ask if errors come from model or observation. Measurement uncertainties have been developed in the method section and have been taken into account in the calculation of fog activation properties (ie. the large errors bars in our figures). The comparison of our derived parameters were close to those previously obtained in other experimental studies. This give us a relative confidence in our experimental data.

**LN11; median value  $\sim 3.8$  micron ? why it is large?**

Such a median value is large compared to the threshold of 2 micron generally used to define droplets from measurements. In the introduction and discussion sections, we suggest that it may be due to the instrumentation limitation, which generally begins at 2  $\mu\text{m}$  in diameter to measure droplets size distribution.

**In conclusions; you are saying winter time fog events???? How do I know? Why not provide T and  $U_h$  for each case?**

As stated in the abstract and in the introduction, the comprehensive field campaigns were conducted during the winters of 2010-2013. Winter fog do not mean ice fog as we don't have ice or supercooled droplets. RH is 100 % for our case and T during the fog won't bring more usefull information for our activation process study.

**$N_d/N_{act} \sim 0.25$  to  $0.67$ ; in reality,  $N_{act} \sim N_d$ , why this comes out of this work? Your figures suggest that  $N_d > N_{act}$  (see above). What is going on here?**

$N_d$  refer to  $N_{d\_cycle}$  that represents the median values for the whole field campaigns. It is why we name it  $N_{d\_cycle}$  in order to not mix it with  $N_d$  at the activation.

$N_{d\_cycle}/N_{act}$  was lower than 1, which means that concentration of droplets low down during the fog life cycle. Processes such as sedimentation, collision-coalescence, evaporation due to mixing

could occur. As mentioned in the paper lots of studies have already pointed out the key mechanism of the turbulent mixing in fog.

**Fig. 1 for 2 cases Nd about 127 and 46 cm<sup>-3</sup>, why these are selected and like to know in Table what is the fog droplet size range? Please provide min and FM100 values, additional to mean. Any drizzle formed on these cases? This can create lots of issues. I feel because of smaller Nd from fm100, for many cases, you likely have drizzle here. How did you eliminate drizzle?**

Figure 1 illustrates the discrepancies between the WELAS and the FM-100. We choose two contrasting fog event. f6 gets only one droplets mode and f20 two and less droplets.

For case f6 concentrations as measured by the FM-100 (>2μm) are, for the percentil 25,50 and 75, 103-127-142 #.cm<sup>-3</sup> respectively.

For case f20 concentrations as measured by the FM-100 (>2μm) are, for the percentil 25,50 and 75, 33-46-56 #.cm<sup>-3</sup> respectively.

Unfortunately, we did not have any measure above 50 μm, it is impossible to know if we had drizzle. Further measurements campaigns should definitely include such instruments.

However considering the size range involved for the cross section between WELAS and FM-100 ([2-10]μm), drizzle won't create any issues.

**Fig. 2 FM100 versus WE plots; these correlations are useless, no correlation in fact. You need weighted averages and then use fits. How can you do a fit if 1000 points of data at the same location but 10 points in other place? This figure can't be acceptable for fits. Same after this figure discussion**

The aim of this figure is to evaluate the use of size distributions from two different instruments to derive a single fit on the full size range. Figure 1&2 shows that no good fit can be done, a compromise must be made.

Knowing that more confidence is given in WELAS smallest classes bins and in FM-100 largest classes bins, a cross section need to be found to achieve our study. Figure 1&2, show that the best crossing diameter are [4-6] μm and [6-8] μm. WELAS underestimates for diameter larger than 8 μm and WELAS overestimates for diameter lower than 4 μm. As we want to take advantage of the WELAS measurements on the smallest size bins, we choose [6-8] μm as the best cross diameter. However, as noticed by the reviewer, figure 2 shows that depending on the case best fit could be obtained on [4-6] μm. On that point we must highlight that Figure 2 is a statistical support to our study, where all data have been used during all fog long, a perfect interpretation would need to take into account other information such as wind speed and direction. Figures 1&7 clearly show a best fit on [6-8] μm at the beginning of fog event for f6 and f20. However disparities between the two instruments, seen in figure 2, have been considered in our calculation of activation properties as added in section 2.1 : « *Nevertheless because of the large discrepancies between the two sensors on the crossing diameter, next calculation using this crossing diameter also consider a standard deviation using the min and the max size distribution over this size range.* » Considering that the true droplet size distribution should be between WELAS one and FM-100 one, taking into account

the min and the max for the standard deviation provide a reliable droplet size distribution. Nevertheless, we agree that more more investigation should be done to fit these two measure, that should be the purpose of another technical paper.

**Fig. 3; What is the sampling rate/averaging time period of these data points? Do these points represent an event averages? Then you are comparing 2 different things, you cannot do this. Also, show some fog Nd versus Na from other works here. To me no relationship exists between Nd and Na.**

These data points represent median values and 25 and 75 percentils over the fog life cycle of droplets number concentration as derived from the FM-100 [2-50] $\mu\text{m}$  function of the aerosol loading as derived from the SMPS as already written in the legend.

As expected form the first indirect effect (also know as Twomey's effect) in warm cloud, more aerosol generally lead to more droplets. That have been supported by many in situ observations for other type of cloud. Concerning fog Hudson (1980) showed that fog formed in maritim condition contains less droplets than fog formed in polluted condition.

One of the aims of our work is to determine the link between the Nd and Na for the fog in polluted conditions. The figure 3 shows that considering droplets on [2-50] $\mu\text{m}$  no relation appears, differently from other cloud types.

**Fig. 5; Show RHw here please. Also, SMPS decreases while Nd increases before 6 am, is this correct? Nccn and Nd should be correlated positively.**

As mentionned above, no correlation appears between Naero and Nd for supersaturations occurring in fog due to the low supersaturations occurring in fog leading to very selective criteria on particle diameter for droplet activation. In addition the correlation between Nccn and Nd is not straightforward because the supersaturation is much lower in fog conditions than in our CCN measurements, leading to different processes involved in fog activation.

As mentionned in the instrumentation section, SMPS measurements were performed under dry condition while Welas measurements were performed at RH close to the supersaturation values.

**Fig. 6; This is not valid for real atmosphere because we don't see  $S_w > 0.1\%$  often. Then how can we trust these kappa values applicable to real conditions? How about model predictions?**

CCNC chamber have been designed to measure CCN concentration between 0,1% and 0,5 % because usual values measured in this atmosphere are in this range. For example in stratocumulus supersaturation can be as high as 0,8%. Models actually simulate high supersaturation values (see thouron et al, 2012 for example). In our study we extrapolated CCNC measurements to lower SS values in order to derive kappa values representative to fog conditions (i.e. section 3.2 in the paper).

**Fig. 8; Again, what is the averaging length for these data points? To me no relationship can be seen.**

Though the scatter is rather large, the general trend points towards a slight decrease of  $\kappa$  values as critical supersaturation values increase, which corresponds to the expected behaviour as depicted for example in Fig. 6 from CCN data. Data are retrieved using aerosols measurement during one hour interval before the fog event. To estimate the most representative fog droplet size distribution

of the activated distribution, we average the composite wet particle size distribution derived from both WELAS and FM-100 measurements over a time interval from the beginning of the fog event during which the CDNC reaches a stable value for a sufficient long time. On average, this time period is selected from 30 min to 1 hour after the fog beginning. Calculation on the average properties take into account the two kappa and uncertainties due to use of mean time values for CDNC and aerosols distribution. This is already written in the paper page 9 line 22.

**Fig. 9; a) no relationship, b) obvious because you use same sensor for the data points, again, averaging conditions? C) no relationship.**

Fig. 9; a) shows that  $N_{act}$  values increase with SS peak. Note that even if there are quite large uncertainty intervals, they follow the same trend as the mean values. The higher is  $S_{peak}$ , the stronger is  $N_{act}$

b), same averaging conditions as Fig. 8, it's not obvious, that means that the aerosols number size distribution gets a close shape at larger diameter.

c), we said page 12 line 10 "Fig. 9-c) reveals no trend at all from the several values of  $\kappa$ ."

**Fig. 10; To me no relationship exists. Averaging scales? B) see above.**

For a same color it's undeniable that a relationship exists.

**Fig. 11; Same reasoning, see above, averaging scales?  $N_{act}$  versus  $N^*$  no relationship; why is that?**

Same answer as fig 8.

Figure 11 in contrast reveals that almost no relationship exists between  $N_{act}$  and  $N^*$ . This result demonstrates that the concentration of fog droplets is roughly independent of the aerosol number concentration in opposite to the general trend depicted by Fig. 3. The concentration of fog droplets was independent of the aerosol number concentration due to the low supersaturations occurring in fog leading to very selective criteria on particle diameter for droplet activation. This is also consistent with the very large derived dry diameter.

**Fig. 12; are your data points are from CCN chamber measurements? How  $N_{act}$  is obtained from field observations of RHw? At  $SS=0.5\%$  you have  $3-4000\text{ cm}^{-3} \sim N_d$ , did you observe this during field project? How accurate SS in the field data? At  $SS=0.05\%$ , basically no change in  $N_{act}$ ? This is your final figure, and should do a better job to explain it, presently, not enough to be understood properly.**

Grey data point are from CCN chamber measurements, black point have been retrieved by the method explained in the paper. As we are in very polluted conditions at high supersaturations a high concentrations of particle are activated. But we don't have 0,5% of supersaturation in fog, this supersaturation is "created" by the CCNC. At  $SS = 0,5\%$ , great change in  $N_{act}$ , the scale is a logarithm one.

"This is illustrated in Fig. 12 where  $N_{act}$  are plotted as a function of SS peak in log scale (black diamond, same data as Fig. 9-a)) superimposed to the statistics of CCN measurements (grey diamond)."

**Fig. 13: How come CCN can be more than  $N^*$ ? What is the uncertainty in this figure's result? Basically  $N^* \sim N_d$ , then CCN called like this because they are play a role in droplet formation. We should expect  $N_d$  getting close to CCN but not more than CCN? Please explain for your case.**

Here we plotted concentration of activated aerosols particles as measured by the CCN at 0.1 % supersaturation function of concentration of aerosols with diameter higher than 200 nm.  $N_{ccn} > N^*$ , means that at this supersaturation the dry activation diameter is lower than 200nm. The uncertainty correspond to the mean on one hour.  $N^* \sim N_{ccn}$  means that in average the dry activation diameter is close to 200nm at 0,1 % supersaturation.

**Fig. 14; is this for dry aerosols or wet aerosols? Which sensor? SMPS? If dry aerosols, then why we expect to see change in SS. To me again there is no relationship.**

The concentration of aerosols was derived from the SMPS that provides size distributions at the maximum RH of 50%. The absorption of water by continental aerosols is expected to be low at such RH. Thus hopefully dry if they are below the efflorescence point. We measure them with the SMPS, it is the only instrument we have which measure particle size distribution. It is  $SS_{peak}$  as retrieved with our method.  $SS_{peak}$  depend on the thermodynamics conditions, for example in convective situation of the vertical velocities. Here we show that aerosols could have a lowering role.

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

*« One can note in Fig. 11 that the highest values of  $N_{act}$  decrease as  $N^*$  increase. Indeed, while aerosol particles activable concentration increases ( $N^*$ ), variability of  $SS_{peak}$  decrease and tend toward low values as reported in Fig. 14, thus lower particles can be activated. »*

**Table 2; serious problems, if thick fog, then how can we see  $N_d$  less than  $50 \text{ cm}^{-3}$ ? What do you mean with thick? Like to see Vis time series to validate your results given in table.**

As explained in the legend :

*« Table 2. List of fog events analysed here. Type RAD corresponds to radiation fog and STL to stratus lowering, "thick" to fog developed on the vertical, and "thin" to fog layer with top altitude lower than 18 m.  $N_a$  is the number concentration of aerosol particles derived from SMPS data. 25th, 50th and 75th percentiles are computed from the distribution of 5 minutes samples over the last hour before the fog beginning. NFM is the droplet number concentration as measured by the FM-100 over the range [2-50]  $\mu\text{m}$ . Statistics of NFM are computed with minute average data of samples with  $LWC > 0.005 \text{ g.m}^{-3}$  collected during the whole fog event. »*