ACP-2014-750

Response to the review by H. Gerber

Summary -

This paper deals with the optical extinction due to hydrated aerosol in mist and fogs, which is a topic already dealt with previously by others in the literature. However, this paper is significant, because it presents additional evidence of this effect for a larger selection of mists/fogs in a near urban environment near Paris, France.

The scientific quality is generally good in that it deals reasonably well with two particle spectra probes that demonstrate inaccuracies over their sensitivity ranges. Comparison of extinctions integrated from the particle spectra with co-located visibility measurements shows reasonable agreement on the average supporting the extinction conclusions.

The presentation quality can use improvement because of the use of the English language. Sentence structure needs help, and word usage has issues. This leads to difficulty in understanding in places what the authors had in mind. A careful review of the paper by someone more proficient in English is recommended.

Discussion Points -

1. The calculation of pec_M sums up the contribution from the WELAS and the FM100 particle size spectrometers, with the latter only using part of the size contributions larger than 7 um, because the FM100 is thought to underestimate small drops according to the paper. There is a strong contribution of fog drops to the ratio pec_M/pec_k calculated and discussed at length in the paper, so that the FM100 measurements need to be accurate. First, Table 1 needs an uncertainty value which is left out for the FM100. Second, the use of the FM100 data in this paper should be described taking into account the thorough evaluation of this probe by Spiegel et al., 2012: Atm. Meas. Tech., 5, 2237-2260. Further, this evaluation should also be applied to the given pec ratios and their uncertainties.

We added references to Spiegel et al. (2012).

Recommendations on the channel sizes by Spiegel et al. (2012) could not be considered as the FM100 collected data at SIRTA in 2011, and manufacturer's channels were used. However, as done by Spiegel et al. (2012) we used independent measurements to evaluate the quality of the FM100 size distributions: PVM-100 which is considered as a reference instrument by Spiegel et al. (2012), but also a diffusometer:

1) We checked that LWC derived by FM100 lays within uncertainties of LWC provided independently by the PVM-100 instrument, indicating no significant impact of the particle loss. Eventually a slope of 0.80 was found between LWC by PVM and FM100, and therefore N_{FM100} can be trusted at +-10%. PVM served twice to disregard LWC computations with FM100: for ambiguous cases between mist and fog which were eventually labelled fog because LWC was larger than 10 mg/m3 according to PVM; strong over estimation of LWC and pec by FM100 during f1 and f9 fogs.

2) We checked that pecM derived from WELAS+FM100 lays within uncertainties with pecK provided independently by the diffusometer, indicating no critical impact of the size attribution by

the default FM100 procedure. We show we can reproduce pecK with a negligible bias (only 7%), and with one standard deviation of 33%. Spiegel et al. (2012) do not give values of the impact of particle loss and sizing on pec.

Moreover, DF20+ and PVM also agree together, with a slope of 0.99 and a correlation coefficient of 0.95 when comparing pec provided by both instruments (Figure R1).

The aerosol contribution to extinction was derived from WELAS and DF20+, with 40% combined uncertainty.

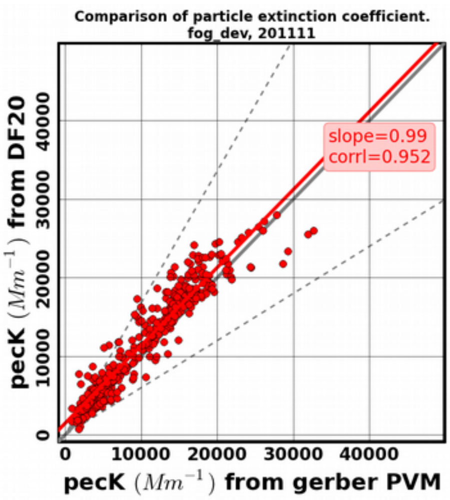

Figure R1. Comparison of particle extinction coefficient measured by PVM and by DF20+.

2. Pg. 297, lines 5-10: It is not clear to which of the two particle spectrometers this section refers to. This becomes clearer on pg. 300, lines 27-28. The clarity should already appear on pg. 297.

This section refers to WELAS, the text has been changed accordingly.

3. Pg. 300, lines 13-14: "Instrument giving largest values is assumed to provide the most reliable measurements" This assumption on the choice of WELAS vs FM100 data made by the authors is risky and should be removed unless it can be backed up with some evidence.

This assumption has been removed and we just keep that WELAS is used for smaller particles than FM100, and that the junction is made at 7 um.

For example at 03:00 15 Nov, the droplet mode would start from the 4th FM100 bin, which shows values smaller than WELAS, in number and in volume.

4. Pg. 302, lines 7-10: "The uncertainty is too high to use RH to detect aerosol activity". This is incorrect, since accurate RH measurements between 95% and 105% have been made some time ago in fog, see Gerber (1991: JAS, 48, 2569-2588). The saturation hygrometer used for those measurements had an accuracy of ~0.02% at RH=100% and response time of a few seconds. A couple of other papers by Gerber are listed in the 1991 paper describing even earlier such fog measurements and details of the hygrometer design.

Thank you, we corrected this mistake. We had to precise that measurements of RH were not precise enough AT SIRTA.

It is interesting to digress a bit and note that the closely related and published paper on Paris fogs by Hammer et al., 2014: ACP, 14, 10517-10533, in which some co-authors of the present paper also appear, does list Gerber (1991) in the reference list, but not in the text.

Perhaps the comment in the Hammer paper that "short supersaturation spikes - - - - are irrelevant" was addressed to the findings of Gerber (1991) where RAD/EVP fogs showed SS transients and droplet spectra with sizes up to somewhat larger than 10 um; even though the fog had a mean RH ~ 100%. Hammer et al also note that "...cooling of air parcels below dew point results in formation of cloud or fog". The fogs in Gerber (1991) appeared to form differently by mixing near-saturated parcels at different temperature causing supersaturations.

This raises the questions: How relevant are the values of SSpeak discussed in Hammer et al when turbulence and mixing dominates fog formation as in Gerber (1991)? See also, for example, Rodhe (1962: Tellus, 14, 49-86) for this fog-formation mechanism. Is it necessary to know the fine details of fog formation including SS, or is the use of SSpeak sufficient to produce realistic droplet spectra? A good test is to use SSpeak values and CCN spectra to calculate fog droplet spectra and compare them to accurate measured droplet spectra. Unfortunately, the latter is still appears to be somewhat of an issue for ground-based measurements. It seems more effort is needed to properly address relationships between CCN, hydrated mist particles, fog droplets, and fog (and cloud) dynamics.

Emanuel Hammer will answer these comments in a separate document.

5. Pg. 303, lines 7 -10: Unclear what is meant here.

6. Pg. 303, lines 3 -4: Unclear.

Rewritten, as "Rarely, *LWC* was observed smaller than 7 mg/m^3, while visibility was smaller than 1 km (Question 'F3'), and while LWC was larger than 7 mg/m^3 during the previous and the next time step (Question 'F4'). We can suspect particle losses (Spiegel et al., 2012) in such cases, as the PVM, in contrary to FM100, showed values of *LWC* larger than 10 mg/m^3. Such cases were then defined as fog. Figure 4 shows that this situation occurred rarely, only 5 times, which is less than 2% of the fog situations and less than 0.05% of the cases with visibility < 5 km. "

7. Pg. 304, lines 20 - 21: Unclear. Is "LWC factor" larger or lower?

These lines were rewritten. LWC was smaller in mist than in fog, by a factor of around 50, while the mist/fog ratio in visibility was around 10.

8. Pg. 304, lines 23 - 25: Rewrite. Are you trying to indicate a range from 93% to 99%?

RH was included between 93 and 100% in mist in November 2011 at SIRTA, with a month average of 99%, and an uncertainty of 2%. Such high RH confirms these low visibility events are not haze according to the definition by Quan et al. (2011).

9. Fig. 1: This figure shows a plot of measured WELAS volume size distributions and fitted lognormal curves. The fitted curves show a dip identified in the paper as the transition particle size between hydrated particles and the first fog drops. Are the transition sizes described in the paper from these volume spectra? If so do they not overestimate the actual transition sizes? Hammer et al uses a similar approach but surface area spectra for which they correct an overestimate of the transition size. Do you need to do the same for your sizes?

Hammer et al. (2014) chose to estimate the transition diameter from the surface size distributions, while we chose to estimate it from the volume size distributions (vsd). New Fig. 1 shows that the inflexion in the slope in number size distribution (nsd) occurs at the same size as the dip in the vsd. For example at 03:00 on 15 Nov, the inflexion in nsd and the dip in vsd occur at ~4 um. However Fig. 2 (previously Fig. 1) shows that the intersection of the two modes occurs at larger size than the observed slope inflexion or the dip, by around 1 um.

Anyway, we made the computation of the hydrated aerosol contribution to extinction, considering two sizes: 2.5 um, which is close to the diameter found by Hammer et al. (2014), and our varying estimate (average of 4.0 um). We commented that aerosols smaller than 2.5 um contribute for the main part, 20%, while the averaged contribution by aerosols larger than 2.5 um is 6%. Therefore the choice of the definition affects little the computation of the contribution. However, the strong advantage of method of Hammer et al. (2014) and our method lays in letting the variability of the transition diameter.

Given your comments about the shortcomings of both the WELAS and FM100 spectrometers, it would be desirable to show a comparison plot of the two probes which the paper does not have; only the WELAS spectra are shown. Such a comparison is especially pertinent for the FM100 spectra which must contribute heavily to pec_M.

One Figure is added (Figure 1) showing size distributions from both instruments on the same plot, for two fog events. They show that the junction diameter was close to 7 um, and that it did not depend on the volume or number.

The font x and y axis legends on the plots in Fig. 1 are too small.

This was corrected.

10. Fig. 2: The lettering appears too small.

Corrected.

11. Fig. 8: Looks like your data points on this plot are for RH > 99%. Is that realistic?

Observations show RH up to 100% in mist, but with uncertainty of 2%. RH was measured at several levels along the meteorological mast. Figure R2 shows comparisons between RH at 2 m a.g.l. (used in this paper) and RH at 1 m (bottom) and at 5 m a.g.l. (top), in mist (left) but also in the 5-10 km visibility range (mv) (right). Figure R2 shows satisfying agreement between the three sensors. Large differences in mist between 2 and 5 m agl (top left) are due to vertical gradient of RH in some of the mists.

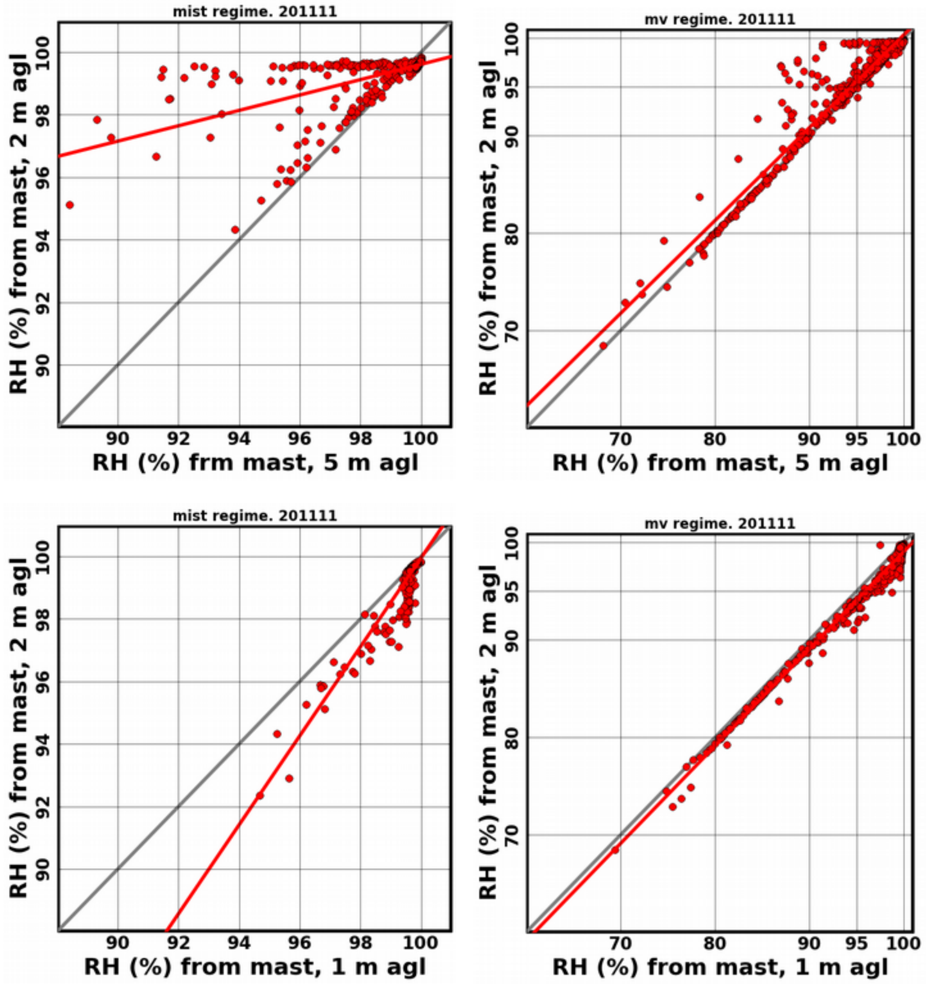

Figure R2. Comparison of RH measured at several heights of the meteorological mast, during mist and 5-10 km events.

12. Table 3: The lettering might be too small.

That may be taken care during the edition process.

13. Table 5: Include a notation that the values in this table are average values.

We mentioned that values are averaged.

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Response to the review by Reviewer 2

General Comments

The article addresses a scientific question within the scope of ACP, namely, the role that hydrated aerosols perform in light extinction in mists and fog. The paper uses an analysis of field observations made during November 2011 near Paris to make its points. The field observations are analyzed to learn about the underlying physical processes. The paper provides an interesting analysis of these observations that has not been done before, on a topic and with a field campaign already in the literature. Other studies done on the data collected during this field campaign are cited in the paper. The limitations of the instrumentation are discussed in depth and uncertainty quantification is presented. In general the structure and content of the paper is sufficient to communicate the authors' methodology, results and conclusions. Some clarifications are requested in the specific comments and presentation quality is addressed in technical comments.

Specific Comments

p. 7, lines 7-10: the sentence beginning "Consequently" is unclear, in particular the end of the sentence.

Due to the general agreement between the particle counters and the diffusometer, we eventually chose to neglect the discussion on this possible under estimation by WELAS.

p. 10, lines 1-4: provide more detail and reasoning on the contribution of the items listed to the 30% uncertainty estimate.

We refer to computations made by Elias et al. (2009) which estimated a 30% uncertainty. Discussions are provided by Wex et al. (2002) on the impact on some of the aerosol properties. Uncertainties on scattering by Mie theory were evaluated to be +-20%, and on absorption to be +-30%. Discussions are also provided by Chen et al. (2012) which estimated 34% uncertainty on extinction.

p. 10, first full paragraph: What is the instrument accuracy over the whole range? The instrument that provides the largest values is presumed to be right. Is this still likely an underestimation? Are certain ranges more likely to be accurate than others even within whichever instrument is chosen as "correct?" Are there ever cases where the instrument giving larger values is not the one with the greatest sensitivity in that region? Please clarify this paragraph and perhaps cite some more papers specifically dealing with the performance of the two instruments. There are selected bibliographies provided on the manufacturer website for the FM100 at least as a place to start:

http://www.dropletmeasurement.com/products/ground-based/FM-120

We did not keep this sentence "the instrument giving largest values is assumed ... its greatest sensitivity". As shown by Elias et al. [2009] and also suggested in new Fig. 1, WELAS alone could not reproduce extinction in fog, because of underestimation of the droplet size distribution. Given the agreement between WELAS and the diffusometer, and as suggested by the new Fig. 1, FM100 seems not reliable for smallest particles. Other details on accuracy depending on the size range are not treated here, due to the general agreements we found.

However data were screened out of situations with rain and drizzle, and of shallow fog conditions, as discussed in Sect 4.1. We also discussed the variability of the aerosol contribution to extinction by studying the impact of the main fog formation process.

Few references exist on WELAS. Advised papers on FM100 were read and more references added in the text, especially Spiegel et al. 2012.

P. 11, Figure 1 description: Are these single 5 minute sampled values at the UT listed? Or are they averaged over the 15 minute period mentioned in Section 2? Are these meant to show different characteristics of the different fog events or is this level of variability also seen within the individual fog events? Why is only one of the instruments included in the plots?

Only the WELAS size distributions are shown in Fig. 2 (old Fig. 1), because the WELAS alone is sufficient for the distinction between aerosols and droplets, covering the appropriate range from 1 to 10 um. Size distributions by both instruments are now shown in new Fig. 1.

As specified in Section 2.1, all data are averaged at 15 minutes. Even the size distributions of Fig. 2 (old Fig. 1) were averaged over the 15-minute period. Fig. 2 is meant to show the variability between fogs, while the variability in individual fog is now showed in new Fig. 1 (3-hour averages are used only in new Fig. 1).

p. 12, lines 1---3: this is unclear. Why could the 1 km convention not be applied in order to distinguish between the two types and then the events further stratified by the droplet presence? This could be used to make a conclusion about the accepted definitions of fog/mist. The reasoning becomes clearer as the section continues but there should be a clear, succinct statement here, and the phrase "could not be applied" is misleading.

First sentence was erased for clarity, and we rewrote some parts of the Section.

Literature does not show a net limit in visibility for fog definition. 1 km is usually accepted but some authors also define light fog or evolving fog at visibility > 1 km. Consistently, with the LWC threshold, and therefore droplet presence, visibility could be larger than 1 km in fog.

With the 1-km definition, droplet cases would be included in the mist regime, that WELAS alone could not reproduce properly. Consequently, the agreement between WELAS and the diffusometer in mist would be eroded.

p. 12, lines 9-10: Clarify this is because of the uncertainty in the available measurements in this field study and not because the uncertainty is too high in a general scientific sense.

We added the precision that the uncertainty is too high with measurements made AT SIRTA. Indeed

Gerber [1991] for example presents measurements with sufficient precision.

p. 13, lines 7-8: If the FM100 did not provide the correct LWC, what does that mean for its reported size distributions during these times?

It is expected that under-estimation of LWC is associated with under-estimation of pec, but with a negligible impact on the slope between optical counters and the diffusometer.

p. 18, lines 11-13: If filtering is being described here (wording "eventually agreed" makes me think that) please make it much clearer and more specific.

We now mention that filtering in Section 2.

Conclusions are clear and well stated.

Technical Comments

The article has many grammar mistakes and some unclear language and sentence structure. There should be extensive revision to correct this deficiency. A few examples are provided here but the list is not exhaustive.

Examples:

p. 2, line 5: "which are the most efficient to interact" should be "most efficiently interact"

p. 11, line 8: "varying" should be "vary"

p. 11, last line: "chart flow" should be "flow chart"

p. 13, line 28: "associated to" should be "associated with"; remove "however"

We corrected the mistakes you mention, and we revised the grammar of the paper.

Other technical comments:

Table 1: Uncertainty is missing for the FM100 and the CPC.

Values were added.

Table 5: The columns don't line up correctly so it is hard to immediately tell which method goes with which columns.

We will be careful with that matter during the editing process

Figures 1,4,5,6,7,10: Increase font sizes for axes labels and tick marks.

Font sizes were increased.

Wex, H., C. Neusüß, M. Wendisch, F. Stratmann, C. Koziar, A. Keil, A. Wiedensohler, and M. Ebert, Particle scattering, backscattering, and absorption coefficients: An in situ closure and sensitivity study, J. Geophys. Res., 107(D21), 8122, doi:10.1029/2000JD000234, 2002.