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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modes in the size distributions and neutralization extent of fog-processed ammonium salt aerosols observed at Canadian rural locations" *by* X. H. Yao and L. Zhang

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Responses To Referee #1

We greatly appreciate all of the comments, which have substantially improved the paper. Our responses are detailed below.

RC- Reviewer's Comments; AC – Authors' Comments

RC: General Comments

The authors present a list of findings on 10 aerosol samples collected during fog events in coastal and inland Canada. Fog measurements are sorely needed to better understand the role of aqueous chemistry in forming or modifying particulate matter, and these measurements provide a decent data set to address the question of particle size vs inorganic composition. However, for this paper to have any impact on the community it needs to be severely restructured and it must be edited for grammar. Some unnecessary text should be removed to focus the paper. These are the main concerns that must be addressed before publication. I have itemized some detailed comments below but have not given a complete list of grammatical changes as they are too numerous. Additional critical points are listed below and must be clarified before publication can be granted.

The structure of this paper (by conditions) results in a list of observations in each condition with little interpretation other than "this indicates fog processing." The larger structure of the sections and the internal structure of each paragraph result in the reader becoming lost in the list of what each size has, whether it is neutralized, and what the likely source is. By the fourth section it is impossible to follow the story. An alternative to this structure would be to target a specific chemical component(s) and to follow that through the various fog and temperature conditions. In this manner, one would come away from the paper with an understanding of how sulfate was distributed and how various conditions impact it's distribution and neutralization, for example.

AC: We reanalyzed our data based on all of the three reviewers' comments. In the revised paper, we chose to focus only on the supermicron modes of ammonium aerosols and generated more logical explanations behind the observed phenomena. The main findings include: (i) one or two supermicron modes were found in fog-processed ammonium aerosols, (ii) the first supermicron mode was in the size range of 1.1-1.7 μm if T>-3 C and in the size range of 2.8-3.4 μm if T< -4 C, (iii) the second supermicron mode appeared in the size ranges of 5-10 μm, but not necessarily appeared in every fog event, (iv) the first mode was mainly caused by fog-processed ammonium aerosols

and the second mode was likely the direct collection of fog droplets, (v) heterogeneous ice nucleation likely played a role in the larger size mode under T< -4 C. Based on these findings, we reconstructed our presentation of the results and discussions and rewrote the whole abstract and part of the Introduction. Unnecessary texts were removed, literatures were updated, and proof reading was done by an English native speaker.

RC: Specific Comments Title: The title of this article does not advertise the new and interesting findings of this work (neither does the abstract). As part of a reworking of this manuscript, the title should be reworded to emphasis the most important finding. What about the modes? Was a new mode found? Perhaps the title should focus on the supermicron mode found in processed particles if that is the most new and interesting result. Currently the title shares the same problem as the structure, little interpretation.

AC: Based on the new results generated from the reanalysis, we changed the title to: "Supermicron modes of ammonium salts observed in rural atmosphere - Implications of fog-processed aerosols and heterogeneous ice nucleation at warmer temperatures"

RC: Abstract: The abstract is difficult to follow as written. It would be much more informative to write what the modes mean, interpreted by the authors, with less of a list of findings. I'm already confused after reading only the abstract. What is the significance of the modes? What conclusions can be drawn from these findings? If the main conclusion is that fog forms these supermicron modes, then what? Do the authors think that by creating these supermicron particles, the fog can remove PM gravitationally the following day, even in the event the fog does remove it overnight?

AC: the abstract was completely rewritten to reflect the major analysis and findings as summarized above.

RC: Introduction: pg 5521 line 17: This is an excellent point, are the authors going to come back to this point in the discussion of their work? Did they show that in a clean environnent, fog events enhance particulate pollution? This would be great to work into

the results/abstract/title if so. If the authors are not going to comment on this question in the context of their work, it does not belong in the introduction.

AC: We removed this discussion from the Introduction in the revised paper. From our data, we just found an increase in the ammonium concentrations in supermicron particles relative to those of non-fog samples. The data does not allow us to draw any firm conclusion on fog-enhanced particulate pollution.

RC: Pg 5521 line 22: Based on the description here, it makes more sense to have one section for the role of temperature, one section for the role of acidity, and one section for pre-existing aerosol and to discuss how ammonium sulfate varies in each. For most cases, the authors seem to be making the case that fog processing is driving the observed patterns rather than primary emissions. Perhaps it would make a more focused paper to omit the discussion of other components unless they directly relate to ammonium sulfate.

AC: this suggestion gave us more ideas on how to interpret our data. In the revised paper, we separated the data based on the temperature conditions and included discussions on emission sources, pre-excising aerosols (including interstitial aerosols during fig events), and acidity conditions. We also omitted many materials that were not directly related to the focus of the paper.

RC: Pg 5523 line 10: Should "mass spectra" be "mass concentration distribution?"

AC: This part was rewritten and the phrase did not appear anywhere in the revised paper.

RC: Pg 5523 line 11: This statement is a bit confusing. Are the authors saying that 5% of samples collected during/post fog had a supermicron mode of ammonia salts while 10% of samples collected during/post fog DID NOT have this mode? It seems they are claiming the mode as a fingerprint of processing, but twice as many fog samples were lacking this mode compared to those samples with a supermicron mode. The

logic seems circular if I am following correctly. If anything, the wording "fog processed aerosols cannot be clearly identified due to the absence of the supermicron modes" must be reconciled with the wording above stating that the mode coincided with foggy days. This is a serious point that must be cleared if the main finding of this paper is supermicron ammonium salts following foggy days.

AC: We rewrote this part and provided more explanations. It was moved to Section 3.1 due to a reconstruction of the presentation. It now reads: “It should be noted that, in another 10% of samples which might have gone through foggy days, the supermicron modes of ammonium were not apparent. One explanation could be the high efficiency of fog in removing large particles from the ambient air during the sampling periods. This could happen if the sample duration was much longer than the fog events. Another possibility could be the low efficiency of MOUDI in collecting fog droplets larger than 10 μm if fog processed aerosols exceeded that size.”

RC: Pg 5525 line 25: Sulfate as a primary emission? From seasalt, yes, but in anthropogenic emissions SO2 is the primary emission while sulfate is characteristic of aqueous processing.

AC: The reviewer is right. In the revised paper, this part was removed since we focused on supermicron ammonium.

RC: Pg 5526 line 2: Is this because the supermicron modes are rarely measured? Or, because they are measured but the mode are not observed? This is a key difference.

AC: Supermicron modes were measured but not observed in most cases. Limited studies observed the supermicron modes post fog events (e.g., Moore et al., 2004). This part was rewritten.

RC: Pg 5526 paragraph starting line 6: It is difficult to follow the reasoning in this paragraph. Instead, consider providing a short sentence describing the mechanism in cases 1 and 2, then go into detail explaining why they are unlikely. In addition, it is

very difficult to understand what the authors are disputing in case 2. They are claiming hygroscopic growth was NOT part of the formation of the large mode, but then claim supersaturation was needed. How exactly do they think the larger particles formed? From what process? What exactly are they calling fog processing if not the uptake of gases and aqueous reaction of those gases into non-volatile components? This should be rewritten.

AC: In the revised paper, we focused on the supermicron modes of ammonium. This part has been removed.

RC: Pg 5527 line 15: These references are not from this study, but the statement you were reporting is that organics improve CCN efficiency in THIS particular study. Since it is not always the case that organics behave this way, you can only reference works on that study, rather than all instances when organics have done so.

AC: We removed the discussion in the revised paper.

RC: Pg 5527 line 21: The discussion of Ca as FCN is out of place here and leads to confusion. It should only be presented in the context of ammonium salt modes. It canbe reworked as part of the restructuring described above if the authors find it relevant, otherwise it should be omitted.

AC: We removed the discussion in the revised paper since we focused on the temperature effects.

RC: Pg 5528 line 6: I thought this section was about two samples? In which "particular sample" does this occur?

AC: The whole section was rewritten and the sample periods was clearly stated in the revised paper.

RC: Technical Comments

AC: All corrected.