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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

Anonymous Referee #1

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Anonymous review of "Modes in the size distributions and neutralization extent of fogprocessed ammonium salt aerosols observed at Canadian rural locations"

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

C576

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.

- 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.

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?

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 environment, 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.

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.

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

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...

Pg 5525 line 25: Sulfate as a primary emission? From seasalt, yes, but in anthro-

C578

pogenic emissions SO2 is the primary emission while sulfate is characteristic of aqueous processing.

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.

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.

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.

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 can be reworked as part of the restructuring described above if the authors find it relevant, otherwise it should be omitted.

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

-- Technical Comments

Pg 5521 line 1: replace "could" with "can"

Pg 5521 line 11: insert "A" before "high number"

Pg 5521 line 14: omit "the" before "cloud formation." Note, the authors use "the" many places it should not be and I stopped editing the paper a few pages in. This may be helped by reviewing with a native speaker.

Pg 5522 line 21: "However" is not the correct word here.

Pg 5522 line 23: Statistically significant at what level?

Pg 5526 line 2: replace "were" with "are"

Pg 5527-5528: The tense keeps changing here to past tense and back to present. Avoid past tense here.

Further technical corrections are needed but are too numerous beyond this point to itemize.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 5519, 2012.

C580