Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-373-RC2, 2020
© Author(s) 2020. This work is distributed under
the Creative Commons Attribution 4.0 License.

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of in-cloud aqueous processes on the chemical compositions and morphology of individual atmospheric aerosols" *by* Yuzhen Fu et al.

Anonymous Referee #2

Received and published: 10 July 2020

General:

This paper compares single-particle measurements of particle size, composition, and morphology of in-cloud and interstitial particles collected at a rural site in China. This paper mainly comments on the role of aqueous chemistry in forming organic shells and the observation of more branched soot particles in cloud. I have several comments about this work to be considered before publication.

Major Comments:

1. A lot of the most important details and figures are in the SI rather than in the main text. Also, a lot of the main supporting data came from SPAMS analysis which was not

described in the methods.

2. The argument regarding core-shell and liquid-liquid phase separations was a bit confusing since previous work has shown that the core-shell morphology can break down as RH is increased.

Specific Comments:

Introduction:

1. Lines 59-60: Nitric oxide is a gas, not particulate matter.

2. Line 63: not sure how "decomposed" is being used in this sentence.

3. Line 76: reword "this process might not be neglected"

4. Line 78: Also see [Moffet and Prather, 2009]

5. Lines 77-86: please also comment on the finding that organic coatings caused the collapse of soot particles from [Spencer and Prather, 2006]

Methods:

1. Lines 100-101: What is meant by "almost unaffected by local anthropogenic sources"?

2. Lines 117-120: I think that Table S1 and the air mass back trajectories should be shown in the main paper. It will help give context for what was different between the different cloud events to help interpret the results.

3. Line 125: change "folds" to "fold".

4. Line 128: change "vacuumed" to "vacuum" and define NH4NO3.

5. I couldn't follow the methodology given in section 2.4. Please add more details.

6. A lot of SPAMS data is brought in to corroborate the results. I suggest that details on the SPAMS needs to be added to the methods if the data is being used.

Results:

1. Line 153: some fresh soot particles can have sulfate, see [Moffet and Prather, 2009].

2. Lines 154-156: the methods for identifying each component should be moved from the SI to the methods section of the paper.

3. Lines 159-161: I would think it would be important to explicitly detail the mixture for your results. I found these classifications really confusing and hard to keep straight.

4. Lines 167-168: I recommend bringing Figure S1 into the main paper.

5. Lines 168-169: Not sure what is meant by “influenced by air masses”. More description of the different conditions and air mass conditions encountered for each cloud event will help the authors interpret their single particle findings.

6. Lines 171-172: What is meant by “as confirmed by SPAMS data”?

7. Line 176: what is meant by “part of”? Can this be made more quantitative?

8. Figure 3, should “coating” be “thin coating” instead to better distinguish the morphology?

9. I found the coating thickness definitions to be confusing especially because they overlap. I’m not quite sure how the coating thickness was used to robustly distinguish particles classified as “coating” vs “core-shell”

10. Lines 219-222 imply that the site is polluted, but the site was presented as a background site.

11. Line 223: reword “follow up strong interactions” to “heterogeneous and multiphase reactions”

12. Lines 225-227 seem to imply that there is more data that was not presented. Please rephrase.

13. I’m very confused as to how the O/C ratios were determined. Perhaps I missed

something, but I thought that the detector used only detected elements heavier than C and it is not clear how the background from the carbon film is accounted for.

14. One of the main conclusions of this paper is regarding oxidized coatings formed via aqueous chemistry, yet the main table showing this is in the SI. I suggest bringing Table S2 into the main paper.

15. The O/C values should be stated in the main paper.

16. Lines 242-245: If ion peak ratios from SPAMS are discussed, then SPAMS must be included in the methods section and the interpretation of the ion peak ratios needs much more interpretation to connect to the data presented in this paper.

17. Line 251: also site [Moffet and Prather, 2009]

18. Lines 254-256: I don’t follow the logic regarding non-volatile material and branching. I suggest that the authors more clearly present this argument.

19. Lines 258-260: could this just be showing the role of particle size where unaged soot is larger and more CCN active than smaller, aged particles?

20. Lines 268-270: is there a figure showing the off-center positions of the soot?

References:

Moffet, R. C., and K. A. Prather (2009), In-situ measurements of the mixing state and optical properties of soot with implications for radiative forcing estimates, Proceedings of the National Academy of Sciences of the United States of America, 106(29), 11872-11877.

Spencer, M. T., and K. A. Prather (2006), Using ATOFMS to determine OC/EC mass fractions in particles, Aerosol Science and Technology, 40(8), 585-594.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-373, 2020.