



EGUsphere, referee comment RC2  
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## **Comment on egusphere-2022-166**

Anonymous Referee #2

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Referee comment on "Measurement report: Closure analysis of aerosol–cloud composition in tropical maritime warm convection" by Ewan Crosbie et al., EGU sphere, <https://doi.org/10.5194/egusphere-2022-166-RC2>, 2022

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This manuscript represents an interesting dataset that covers a broad range of topics, and it does a nice job of demonstrating the complexities of conducting and analysing these types of studies. The breadth of the paper limits the focus that is put on any particular topic, and there is a distinct lack of discussion of the process of nucleation scavenging, despite this being fundamental to everything. Samples of water collected for chemical analysis are all classified as cloudwater (CW). A rainwater fraction (RWF) is used to identify the relative contributions from cloudwater versus rainwater, which I think is a nice approach. For a paper about clouds, I think there are too many thoughts about the discrepancies associated with the  $\text{NH}_4^+$  measurement. Overall, the authors have a very nice dataset, including the information about organic components of the cloudwater, but I would like to see this long paper more focussed on a few issues that can be addressed well. For example, since there is disagreement on  $\text{NH}_4^+$  among some of the measurements, eliminate  $\text{NH}_4^+$  from the discussion and put it in another paper after you have completed your related laboratory investigations.

1. Introduction – There seems to be a lot of discussion about why observations don't work well, but then we are told that it is clear that they are needed. The discussion fails to acknowledge observations that have worked, mostly because of the nature of the clouds and well documented instrumentation.
2. Lines 200-202 – Why were liquid drops “universally assumed”, when you have 2D instrumentation you can use to avoid an assumption?
3. Line 218 – RWF is a ratio. Unless LWC includes RWC, the fraction should be  $\text{RWF} = \text{RWC}/(\text{LWC}+\text{RWC})$ . Please clarify. Also, here it would be helpful to clearly state that all samples of water are referred to as CW with RWF used to indicate the relative proportion of rainwater in each. In Figure 6, identify samples that are RW in some way. For example, are the samples that appear to have been collected below cloud in Case II actually RW or is there some discrepancy in cloud base?
4. Section 2.5 - How were the blank samples, mentioned on page six, used? What were the detection limits?
5. Section 2.7 and Figure 1 – The implication here is that this represents one cloud. The very sharp and narrow downdraft just past 20 seconds suggests a possible outer edge of the larger cell. Does this time series represent two clouds in different stages?

6. Section 3 – For each case, it would be helpful to have a conceptual 2D cartoon showing the relative proximity of the clear-air profile to the cloud and the wind directions with heights. That is, something similar to Figure 6, but showing the proximity of the clear-air work to the sampled cloud.

7. Line 416 – “PILS SS showed a marked reduction in US”. Was that meant to be “DS”, and reduction relative to what: in the DS intervals,  $SS:nnSO_4=$  appears to be higher?

8. Lines 472-473 – The statement “A large number of air mass properties that characterize sub-micrometer aerosol” seems a bit odd. Should it be reversed: i.e., “A large number of sub-micrometer aerosol properties that help to characterize the air mass”?

9. Lines 506-507 – With an hour and a half between the stacked legs, should you expect consistency?

10. Lines 520-522 – This does not appear to be true, as increases in nitrate coincide with the spikes in calcium, which you note later in this paragraph in connection with the possible reactions of  $HNO_3$  with insoluble (or less soluble) calcium-containing soil molecules. Related - lines 525-529 – Why would insoluble components be responsible for the spikes if they’re insoluble?

11. Lines 584-586 – Why is sulphate a useful candidate for closure, since in-cloud production of sulphate is a dominant factor contributing to global sulphate? Also, if you are to undertake a closure analysis of sulphate, you must consider the fraction of sulphate scavenged by nucleation. It would seem from the many measurements you mention, including size resolved sulphate from the AMS and droplet distributions, that you have the tools to include nucleation scavenging in a reasonable manner.

12. Line 598 – Your statement about the implication of “positive curvature” is interesting, but I would like to see a little more discussion here. It seems reasonable that the wide variation of  $SO_4=$  at a near constant CO of about 0.135 ppm may be indicative of precipitation scavenging (Case I), but there can be influences affecting  $SO_4=$  as well as CO. In making your statements in this section, your discussion should consider major losses and sources of CO and  $SO_4=$  in order to rule out other explanations for the behaviour of the clear-air curves in Cases II and III.

13. Lines 709-611 – I think this statement belongs in section 2.7.

14. Lines 613-622 – It is here that you first distinguish between CW samples collected in cloud and CW samples collected in precipitation in otherwise clear air (“unsaturated precipitation” as you call it, which is might better stated as precipitation in unsaturated air.) RW is subject to some additional processes than CW. As I mention above, the distinction should be shown in Figure 6.

15. Lines 621-622 – I would appreciate a more detailed explanation of the statement that “The singular Case III data point... is perhaps indicative ... that ... a reference parcel with 100% scavenged fraction is unrealistic, even under undilute conditions, given the competition for water vapor in the highly particle-loaded smoke plume.” What is a reference parcel with 100% scavenged fraction? What does competition for water vapour have to do with the reference parcel?

16. Lines 624-635 – Are the quartiles in  $SO_4=CW/SO_4=ML$  different across  $Z/Z_{top}$ , across RWF and across  $w/\sigma_{w}$  after taking into account the uncertainties in the measurements and sampling constraints (e.g., how representative is your  $SO_4=ML$  of  $SO_4=$  at cloud base?)

17. Line 635 – By “scavenged” here, do you mean nucleation scavenged?

18. Line 641-642 – There are at least two reasons why I don’t think you can make this statement about  $nssSO_4=$ :

a. You don't appear have sufficient information to help assess the importance of sulphate production in cloud.

b. You haven't assessed nucleation scavenging at cloud base. If only 50% of the  $SO_4=$  is scavenged, some aspects of the assessments arrived at from Figures 6-8 could be quite different.

19. Line 642-643 – Four microns seems to be around the cut off for sampling coarse particles from many aircraft, but the statement about “grossly underpredicted” sea salt particles should be either toned down or better supported. You have measurements to

give you information on the profiles of coarse particles that can be used to help assess the potential contribution from larger sea salt particles to your cloudwater samples.

20. Line 649 – “as anticipated” – We expect sea salt particles to be entirely incorporated into cloud during nucleation because of their generally larger sizes. We do not expect SO<sub>4</sub>= particles to always be entirely incorporated (Jensen and Charlson, *Tellus*, 1984; Leaitch et al., *Tellus*, 1986). For that reason, it is possible that your SS/SO<sub>4</sub>= can be higher in the CW than in the PILS samples. You bring up nucleation scavenging occasionally, but you do not try to address it in anyway with your measurements, despite it being a major process impacting your discussions.

21. Line 669 – “susceptible to washout” should be referenced, e.g., Seinfeld and Pandis.

22. Lines 669-670 – “CW SS in unsaturated environments”. Are you proposing to change terminology: i.e., rain becomes cloudwater in unsaturated environments? This complicates reading.

23. Line 676 – Just the 2nd leg, not “The lower (2nd) leg”. It is not lower than 3 or 4.

24. Section 4.3.1 - Consider references to Leaitch et al. (*JGR*, 1986; <https://doi.org/10.1029/JD091iD11p11821>) and Hill et al. (*JGR*, 2007; [doi:10.1029/2006JD008002](https://doi.org/10.1029/2006JD008002)) that discuss measured enhancements in nitrate in cloudwater collected in convective cloud.

25. Line 729 – The Quinn reference appears to be missing.

26. Lines 734-747 – The aim of this paragraph seems to be a reference to justify the lab experiment some of the authors are conducting. Otherwise, it tells us nothing because there are so many uncertainties associated with the NH<sub>4</sub><sup>+</sup> measurements and so many related processes to consider. This paragraph complicates the paper and I suggest removing it.

27. Line 806 – I think it is reasonably well known that oxalic acid can be unstable, depending on temperature, but your statement that oxalate is not thermodynamically favored on sea salt particles needs a reference. Are you referring to oxalate after reaction of oxalic acid with NaCl?

28. Line 818 – Does this consider the AMS-CE and CVI collection efficiencies as well as size constraints on CW sampling?

29. Lines 831-832 – This is a good example of something that could stand to have more focus and detail. There are many indications that OA can be distributed more broadly across the size spectrum than SO<sub>4</sub>=.

30. Lines 838-841 – Interesting, but the analysis is again insufficient.

31. Lines 904-905 – about 25% of the total carbon was measured in the CW samples.