

Interactive comment on “Aircraft measurements of High Arctic springtime aerosol show evidence for vertically varying sources, transport and composition” by Megan D. Willis et al.

Anonymous Referee #4

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This paper describes a carefully analysed and well thought through interpretation of a complex aircraft data set that provides significant new insight into the transport pathways and composition of aerosol in the Arctic polar dome. The analysis sets the observations in the framework of potential temperature to best reflected the vertical distribution of layers entering and descending the lower polar troposphere in spring time. The authors use an analysis of gas phase species to discriminate different layers within the polar dome and then analyse the aerosol characteristics within those layers. This paper offers very valuable new information on aerosol in the polar springtime and contextualises the more extensive surface based measurements that extent. It is a very thorough study and one that adds important new information to aerosol characterisa-

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tion of the Arctic atmosphere. The paper is clear and very well written and despite the necessary detail conveys the information succinctly and in a way that is accessible for the non expert in most places. The figures and tables are informative and there is the right balance between information in the main text and the supplementary material. I recommend acceptance in ACP. I do have a few detailed comments that the authors should take on board:

Page 1: Abstract: “These differences in transport history were closely related to aerosol composition” It would be preferable to say that “Variations in aerosol composition were closely related to these differences in transport history” since transport history drives aerosol composition not the other way around. Page 2: line 30: It is not obvious how the work in this paper has relevance to this problem since the flights were conducted at altitudes of 3.5 km or less. I would suggest removing this as a problem that this work can shed light on. Page 3 line 18: I would start a new paragraph with “Sea salt...” Page 3: line 21-29: The text states that input to the Arctic from both Europe and Asia has been decreasing in the past decade yet most of the papers cited are either model studies or inversions constrained by surface measurements it may be worth tempering the statement to reflect the comments that follow. Page 8 lines 11-15: The technical details of AMS sampling will be lost to many non AMS users. It would be good to include references here to guide a non expert who wishes to know and include a line that summarises what data is delivered from each mode. Page 8: lines 27-28: It would be good to understand how variable these calibrations were and whether there was any trend. Were the calibrations all used and was the average taken or an interpolation performed? Page 9 lines 5-6: Given the low concentrations and the reliance of the CDCE methodology being very dependent on the ability to retrieve accurate ammonium concentrations from the fragmentation table over a wide range of water concentrations it would be good to understand the size of any potential bias in the calculation and well as the variation over the flight window. Page 9 line 21 and line 28: Figure S2 should read Figure S3 Page 9 line 32: strictly Centre not Center Page 10 line 25: demonstrate(s) Page 11 line 30: The caption and axes labels

in Figures 4 and S8 appear to be the same but the data are different. This needs clarification. Page 12 line 3: This statement agrees with Liu et al 2015 but is in contrast with the other papers. This might be made more clear by rewording. Page 14 line 5: Could these differences also be a result of the difficulties in retrieving ammonium from the MS at such low concentrations? Page 16 lines 28-30: This process would need to drive against an opposing temperature gradient driving condensation with reducing altitude though. Previous work suggests this highly aged OA is of low volatility. This might be worth commenting on. Page 18 figure 8 caption "...solid lines represent..." Page 20 figure 9: The ToF signals of organic aerosol at sizes above 500 nm are likely to have very poor counting statistics. The lack of oscillation of the ToF signal at high sizes also suggests the baseline is incorrectly accounted for. I am somewhat sceptical that the increase in OA/SO₄ above 400 nm to 600 nm can be observed above the signal to noise. I would like to see the Poisson statistics and an assessment of the baseline correction before much claim is made for this enhancement. Page 19 lines 19-22: Conclusions: is the converse true? That is do surface based observations overplay the contribution of export of sulfate to Arctic in spring compared to the aircraft measurements presented here? If so it is worth stating. Pages 20 and 21: figures 9 and 10: can you explain the differences between the sulfate size distributions in figures 9 and 10?

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