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Interactive comment on "Aerosol pH Indicator and Organosulfate Detectability from Aerosol Mass Spectrometry Measurements" by Melinda K. Schueneman et al.

Anonymous Referee #2

Received and published: 17 December 2020

This study examines the performance and validity of recently published OS estimation methods through analyzing the AMS spectral data of sulfate-related ions in ambient and lab-generated PM. This work reveals that the published OS estimation methods have major limitations and may produce erroneous results on OS concentration although could work under certain PM chemical composition regimes. In addition, this study explores the feasibility of estimating pH based on AMS spectral data and postulates the physical processes associated with sulfate fragmentation in the AMS. This exercise provides useful insights into why sulfate fragmentation changes in response to changes in aerosol chemical composition. This is a solid work that addresses an important topic related to atmospheric aerosol chemistry. This study is timely and sig-

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nificant. The manuscript is well written and fits nicely within the scope of AMT. I thus fully support the publication of this work on AMT after the following comments are addressed.

It is mentioned that the fragment pattern of sulfate ions may vary from instrument to instrument or even for the same instrument after it is tuned. What's the range of variations in the fragmentation pattern of sulfate-related ions for inorganic sulfate?

It would be helpful to give a more clear definition of organosulfate (OS) here. The paper as it reads seems to refer OS to all organic compounds that can produce SOx ions. It is useful to note that not just the ROSO3 types of compounds generate SOx ions in the AMS, compounds with sulfone, sulfoxide, and sulfonate functional groups may do so as well.

For the PM data analyzed in this study, are there measurements other than the AMS that can be used to validate the quantification of OS concentration?

Consider to increase font size in the figures to make the texts more readable.

Line 41, fraction of what mass? Please clarify.

Line 46-47, this sentence is vague, what values of measured ammonium balance or HySOx/SOx ratio that are indicative of pH < 0?

Line 102, spell out SOAS

Line 147, what "sticky" means here, in what sense or towards what substrate?

Line 221-222, please elaborate a bit more on the "alternative methods" mentioned here Line 264, remove "as".

Eq 6, stay consistent with the nomenclature, add square parenthesis to denote concentrations

Eq. 8, is it inorganic NO3 or total NO3? was the contribution of organic nitrate signals

removed?

Line 507, "in the absence of acidity" does not make sense.

Line 716, it would be interesting that the authors explain what "careful instrument calibration" involves, through analysis of pure inorganic sulfate particles?

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2020-339, 2020.