

Interactive comment on “Eddy covariance measurements with high-resolution time-of-flight aerosol mass spectrometry: a new approach to chemically-resolved aerosol fluxes” by D. K. Farmer et al.

Anonymous Referee #2

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Farmer et al. provide an interesting and useful introduction to measurement of fine particle fluxes using an HR-AMS. This work significantly extends previous efforts with respect to chemically resolved particles fluxes, including Q-AMS work, and should be of great interest to readers of AMT. The manuscript is well written and the work is thoroughly described and appears of high quality. I recommend the manuscript be accepted for publication following attention to several points:

1. The authors speak of PM₁ concentrations, fluxes, and deposition velocities. This

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is an oversimplification. Transmission efficiency curves for the standard AMS aerodynamic lens show a dropoff well below 1 micrometer. This is not clear in the manuscript. On p. 5872 the authors state that “The HR-AMS focuses particles in the 50-1000 nm size range into a narrow beam with an aerodynamic lens.” This issue needs to be clearly discussed in a revised manuscript and any implications for the authors’ conclusions clarified. For example, actual PM₁ fluxes of components may well exceed values reported here as PM₁ fluxes, if size distributions of the discussed species extend much above 500 nm. Likewise, if particle deposition velocities are size-dependent, actual PM₁ component deposition velocities may differ from those reported here.

2. P. 5872: Please elaborate on the system flow control. Since the residence time in the long sample tube is critical to properly pairing concentration and velocity fluctuations, we need to know how (and how well) the sample flow rate was maintained.

3. Have particle losses through the sample system been characterized? How do these affect reported fluxes or deposition velocities?

4. P. 5880, line 18: What do you mean by “particle flux measurements are LIMITED by particle counting statistics?”

5. P. 5884, lines 3-4: The authors state that “...a non-unity slope can be interpreted as the uncertainty in sulphate deposition velocity.” Please explain what you mean by uncertainty. Uncertainty is often used to imply precision, but the slope of this relationship really tells us little about measurement precision. It would seem to be more a measure of bias between two methods although, given that neither method is known to give a true value, it is also unclear whether the slope necessarily implies anything about accuracy. The same comment applies to lines 14-15 of p. 5885.

6. Section 5.2: Do the authors have any evidence whether ammonium oxalate is an important component of the ammonium budget at the BEARPEX site? If it is, this could affect some of the tests the authors invoke (e.g., ammonium vs. anion charge balance). Malm et al. (2005) found that summertime oxalate concentrations at another

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Sierra Nevada foothills site in Yosemite National Park were high enough to noticeably influence the fine particle charge balance.

7. P. 5889, lines 14-15: The statement about ammonia concentrations being too low to support ammonium nitrate formation at the site is too strong. Perhaps the authors mean to say that concentrations are too low to support very much ammonium nitrate formation. 1-2 ppb of ammonia can certainly result in some ammonium nitrate formation, depending on T, RH, and HNO₃ concentrations.

8. P. 5890, line 9: These are the first direct eddy covariance observations of particulate ammonium deposition over a forest.

9. Fig. 2 caption: The term nitrate equivalent mass concentration is likely to confuse readers not intimately familiar with terminology used in the AMS user community. The meaning of this set of units should be briefly explained.

References:

Malm, W. C., Day, D. E., Carrico, C., Kreidenweis, S. M., Collett, J. L., Jr., McMeeking, G., Lee, T., and Carrillo, J. (2005) Intercomparison and closure calculations using measurements of aerosol species and optical properties during the Yosemite Aerosol Characterization Study. *J. Geophys. Res.*, 110, D14302, doi:10.1029/2004JD005494.

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