

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.

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We thank the referee for his/her comments, and are submitting a revised version of the paper addressing the specific and technical comments. Our responses are in plain text below, with the referee’s comments italicized.

In this paper a novel instrument, which has been described in detail in Kimmel et al.(2010), is used to measure size- and chemically-resolved fluxes over a forest. The

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focus of the paper is on the technique of using the instrument to make flux measurements, and on the errors associated with these measurements. The measurement technique is outlined in detail, an attempt is made to estimate the measurement errors, and some diurnal flux and concentration measurements made during the BEARPEX campaign are presented. The application of this new instrument to high frequency flux measurements has great potential for improved knowledge of aerosol deposition processes. The authors do an excellent job of discussing both the limitations and the possibilities of this technique. My only complaint is that the writing is often repetitive and overly detailed in places. Specific examples and other corrections are listed below.

Pg 5869, l 28+: “EC flux measurements must be taken fast enough...” This is an oversimplification since the contribution from high frequencies decreases following a $-4/3$ power law. Similarly, the following “Measurements must be averaged over 30 min...” is not true. 30 minutes is just a convenient balance between number errors and stationarity in some (or most) circumstances. Figure 3 demonstrates that much of the contribution to the flux comes from lower frequencies. At the very least, “must” should be removed from both statements.

We have rewritten the sentences to address this point, as: “EC flux measurements over forests are typically taken at 5 to 10 Hz in order to capture the smallest eddies that contribute to the flux. Measurements are typically averaged over 30 minutes”. And “A challenge is collecting data at evenly spaced intervals to reduce errors.”

p.5870, l3-9. There is an overemphasis here on the challenge of time precision, especially given the errors associated with the time lag correction and the 20 cm distance between the inlet and the anemometer. These lines could be summarized by saying that faster, evenly spaced measurements will reduce errors. Also, the phrase “time grid” seems like a odd way of saying periodic time interval.

We have shortened these sentences as per the referee’s suggestion.

p.5873, l24. As above, this seems like too much information to say that less than 1 in

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200 data points had to be thrown out due when the processor couldn't catch up.

While this may be a large amount of information on the time grid, these numbers and the details of the data acquisition process are particularly useful for current and future users of this and similar data acquisition systems to know what typical data loss rates should be. For example, we anticipate that similar software systems will be implemented for chemical ionization mass spectrometers based on the same TOFMS technology. However, we have removed unnecessary words and phrasing to shorten this section.

p.5876-5878. Steps 1, 3, and 6 are processes, while steps 2, 4, and 5 are corrections. They should be separated. Step 3 could be removed as it doesn't contribute anything, and step 6 is a calibration or a conversion (a change in units), not a correction.

We agree with the referee that the data treatment steps outlined in this section are a combination of corrections, unit conversions and mathematical steps, and were initially written as a 'data analysis protocol', rather than a separate series of calculations and corrections. However, we have separated these two components as requested by the reviewer.

p.5876, l. 15. Rotation also corrects for surrounding slope effects as well as instrument leveling.

Re: surrounding slope effects: The reviewer is correct, and we have added this concept to the text.

p.5879-80 Fig.3 The frequency and cospectrum should both be normalized so the results can be compared with other studies. See Ahlm et al., 2009 (ACPD Vol 9), Gronholm et al., 2009 (JGR Vol 114), or Buzorius et al, 1998 (J. Aerosol Sci. Vol 29) for examples. The individual dots could be removed from the figure as they obscure the binned results, and the gray dots are buried beneath the black dots. It is also unclear if the binned results are all positive or negative, or a mixture of both.

We have altered the figure as per the referee's request to show the normalized, frequency-multiplied cospectrum as a function of dimensionless frequency, and have included only the binned points to simplify the presentation. The figure caption has been modified to reflect these changes, and includes the sentence "The data presented are binned averages of the entire cospectrum, including both positive and negative points."

p.5880-p.5881. This paragraph seems unnecessary in that errors are first calculated and discussed, after which it is stated that the calculated errors are irrelevant for this particular instrument.

We removed the paragraph.

p. 5881, l2. DL is used before it is defined at l 23.

By removing the paragraph, this is no longer an issue.

p. 5882, l11. "smaller in magnitude" might be better than "below".

Done.

p. 5882, l.27. "increasing with increasing u^ and for unstable conditions." could be "increasing with u^* and instability."*

Done.

p. 5882, l.29. Is this just the same as saying the assumption of horizontal homogeneity may not be true?

Yes, and we have rephrased to include the point that the assumption of horizontal homogeneity may not be met.

p.5884-5/Fig.4 I don't understand how the slope relates to uncertainty. Is it not possible that there could be a large amount of scatter in the measurements but the slope could randomly turn out to be 1?

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The slope of the deposition velocities of two fragments that are expected to measure the same chemical species may provide an indication of systematic error: a heavy bias in deposition velocity for one fragment over another suggests that a component is over-estimating (and the other-underestimating) - OR that one of the components is correctly representing the flux, while the other is subject to an unknown bias. In either case, the slope is indicative of the difference between the two fragments that are expected to be two estimates of the same thing, and thus it provides an estimate of the potential systematic bias in using only one of the fragments. This section has been rephrased in order to clarify the point. The effect of the uncertainty in the data points is accounted for by the weighted orthogonal regression.

p.5885, I.10-11. What is the "slope of flux"? We don't see the term "slope of the flux" around I.10-11, and assume you are referring to the 'slope of the correlation' (I.12), which refers to the slope of the correlation plot between the deposition velocities? We will clarify this text to say "slope of the regression line".

p. 5887, I.1. A better reference for temperature fluctuation dampening is Rannik et al., 1997 (JGR Vol 102, 12789-12794).

We think the reviewer means p.5877, I.1, and have added the reference at that point

p.5888 Would "Ammonium Deposition" be a better title for this section? It seems to be the only measurement result discussed here.

We have changed the title to BEARPEX-2007 Ammonium deposition" as we also discuss NR-PM1 deposition and the balance between cations and anions.

Fig.5-7. State if time is UTC or PDT.

We will specify "local time (PDT)" in the figure captions.

Technical Corrections: Eqs 1, 2 and R1 should be followed by punctuation.

Done

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Pg 5870, I 18: "Martensson" should be "Mårtensson".

Done

Pg 5871, I 22: Should "L." be italicized?

The L. refers to the surname of the scientific authority, and its usage (i.e., plain text) is consistent with standard botanical binomial nomenclature system.

Pg 5872, I 20: "were" should be "was".

Done

Pg 5873, I 12: "mode" should be "Mode" (to be consistent with I 10).

Done

Pg 5874, I 13: Should "(iii)" be "(i) and (iii)", since they are both high speed?

While example (i) is high speed, it represents the complete mass spectrum, and none of the data in the mass spectra are integrated over a defined region - and is thus not included in the list.

Pg 5874, I 16: Should this be "m/z 0.5"?

'M/z 0.5' refers to half of a m/z unit, and is thus correctly referenced in the boolean expression.

Pg 5879, I 4: "the stationarity requirement" should be in brackets or preceded by a colon.

Done

Pg 5879, I 8: "...clearly visible, though rarely occurred" is grammatically wrong, as is "..., thus typically meeting...".

Done

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Pg 5881, l 29+: If the mean errors of 248, 360, and 194discussed.

Done

Pg 5885, l 3: To avoid capitalization, “R2 for fluxes...” could be “The values of r2 for fluxes...”.

Done

Kaimal et al, and Solomon et al. are both missing from the references. Done

Interactive comment on Atmos. Meas. Tech. Discuss., 3, 5867, 2010.

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