The effect of hygroscopicity on sea-spray aerosol fluxes: a comparison of bulk and high-rate methods

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Response to Reviewer #2

We thank the reviewer for their comments. Our responses to specific points are below. Reviewer comments are highlighted in **bold**, with our responses in plaintext. Specific changes to the manuscript text are shown in *italics.*

Reviewer Comments

I was unable to convince myself I understood exactly what they did for fast processing. I read section 2.2 several times and am still somewhat puzzled. Perhaps the authors could make this section more reader-friendly.

The relevant parts of section 2.2 has been re-written to attempt to make the fast processing method clearer and easier to understand.

Figs. 10 and 12 show scatterplots comparing estimates of the deposition velocity error associated with humidity effects for bulk vs fast methods. Looking at Fig. 12, there are some clear major differences as a function of size. Channel 5 has a high correlation but a large offset. Channel 3 has two groupings of points and Channel 9 has poor correlation. I don't find any significant discussion in the paper and I can't see how 1.8 micron particles can behave so differently from 1.5 and 2.2 micron particles.

The differences seen here are not so much related to particles of different sizes behaving differently as to changes in the gradient of the aerosol spectra and spacings of the CLASP channel boundaries. The two distinct groupings visible in Channel 3 are related to a change in meteorological conditions (particularly the humidity flux) from around run 84 onwards. Before this time, the bulk method tends to overestimate the deposition velocity error and after this time the deposition velocity error is underestimated. This is also apparent, albeit to a much lesser extent and in the opposite sense, in channels 4 and 5 (see Figure RF1, below). This difference is exaggerated in Channel 3 because this channel is much larger (in a logarithmic sense) than any of the other channels – see, for example, Figure 1.

The bias apparent in Channel 5 is mainly due to a consistent underestimation of the slope of the aerosol spectra by the functional fit. It is clear from Figure 3 that the gradient of the measured spectra tend to increase significantly in gradient between Channels 4 and 5. Such a sharp increase in gradient tends to be underestimated by the functional fit to the spectra (particularly in the case of the smoothly varying quadratic form considered in Figure 12). The 'bulk' estimates are linearly dependent on the gradient of the aerosol spectra, thus this underestimation of the gradient leads to a systematic bias in the deposition velocity error.

The large scatter and poor correlation apparent in Channel 9 is most probably due to a decrease in the signal:noise ratio. At these larger particle sizes counting statistics become poor and thus eddy covariance fluxes become less meaningful. The scatter observed in Channel 9 is visible in all of the larger CLASP size channels, which we did not use in this part of the analysis for exactly this reason. Channel 9 was a borderline case considered worthy of inclusion.

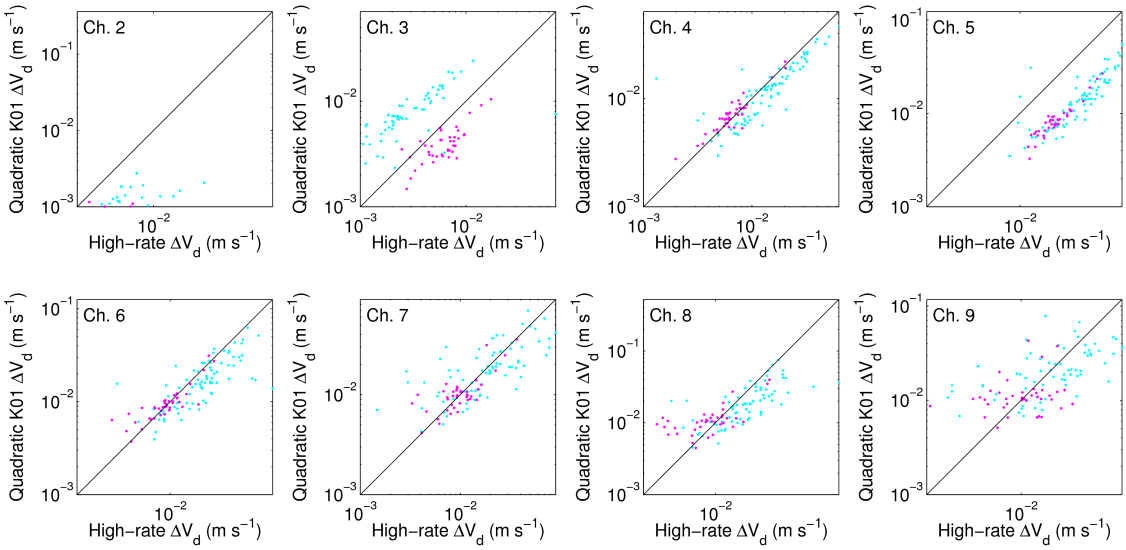

Figure RF1: As Figure 12, but points have been coloured to highlight those from runs prior to run 84 (cyan) and after (magenta).

P6288, Line 7. I don't think it is correct to characterize the covariance as the 'net aerosol flux (production-deposition)'. I believe the net flux is $\overline{w'n'} - V_g\overline{n}$ and that is equal to effective production – deposition.

This sentence has been changed to read

"Most importantly, what the eddy covariance technique actually measures is the net turbulent aerosol flux, therefore it is only an adequate approximation to the true sea-spray source function in conditions far from equilibrium"

P6291, Eq (6). A more accurate formula is

$$\overline{w'S'} = \frac{c_e}{c_d^{1/2}} u_* (1-S)\left[1 + \frac{L_e}{RT^2}(Ts - Ta)\right] \qquad \textbf{(RE1)}$$

It might be amusing to test this one.

While this provides a more accurate estimate of $\overline{w'S'}$, we do not think that including this would add anything to the paper. The two 'bulk' methods we tested, F84 and K01, use the approximation given in Equation 6 and the measured value of $\overline{w'S'}$, respectively. A new bulk method using Equation RE1 would simply provide an intermediate of these two results.

P6296. Fits to aerosol spectra are generally done in log-log space. I don't think it adds much to include the linear-linear fit.

Agreed. We have removed all references to the linear fits from the paper.

P6300, line 10. Can you not get negative Dvd if Beta is negative?

Yes, however for the standard F84 and K01 methods, a negative beta would imply a very flat aerosol spectrum for $-1 \le \beta < 0$ or a spectrum which increases in concentration with particle size for $\beta < -1$ - a rather unphysical result. We have changed this sentence to read

“There are also times when the high-rate bias is negative, which the bulk methods cannot reproduce for observed values of ß.”.

Fig. 6 mostly just shows the importance of getting the aerosol slope correct. That seems mathematically obvious. Could just state the errors associated with it and skip the figure.

Figure 6 (and by extension Figure 7) do show the importance of getting the aerosol slope correct. However, they also show exactly how the deposition velocity error from the high-rate method relates to those from the bulk methods, and provide context for later figures where different spectra estimates with non-constant slopes are used. We thus think that it is preferable to retain these figures.

Several figure panels (8,13, 14) are poorly scaled and just show spikes. This could be improved.

We have re-scaled Figure 8 and replaced Figures 13 & 14 with a figure containing log-scaled scatterplots for CLASP channels 3,5,7 and 9.