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

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Response to Reviewer #1

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

P 6288 lines 11-13: This section should be written more clearly and precisely. The authors should take care to define the net flux measured by eddy covariance, and separate it from the sea-spray flux that interests them, and other processes (deposition, etc.) that they wish to exclude. Note that these can include gas-particle conversion or coagulation which, if operative in the air below the measurement height, will also invalidate the relationship between the surface fluxes and the eddy covariance.

The discussion in this section relates to the specific cases where deposition is zero, and thus does not need separating from the eddy covariance flux, or where deposition is equal to the upward flux in which case the equilibrium method may give the best estimation of the surface flux. However, it is true that these fluxes are only representative of the surface production insofar as any processes which modify the aerosol spectrum below the measurement height can be neglected. The text has been changed to reflect this, and now reads

"In the limiting conditions of no deposition, or deposition equal to production, the eddy covariance or equilibrium method respectively give the best estimate of the surface source flux. However neither of these methods can account for processes that occur below the measurement height, such as gas-particle conversion or coagulation of droplets."

p 6290 lines 6-7: It seems to me that the number flux is not "biased", but more importantly not representative of the aerosol exchange at the surface.

When measuring size segregated aerosol fluxes in the presence of a relative humidity flux, the 'measured' aerosol flux is indeed not representative of the surface aerosol exchange, but furthermore it may not be representative of the aerosol flux at the height of the measurements. This is because, on average, downward moving air parcels will be less humid than upward moving parcels and thus, on adjustment to a reference humidity, the aerosol spectrum will be translated across the instrument size boundaries differently for upward and downward moving air parcels. This can be erroneously interpreted as an aerosol flux, the sign of which depends primarily on the sign of the humidity flux. As over the ocean the humidity flux is almost always positive (upwards), this erroneous flux will generally be of one sign, hence the term bias. However, we have changed this section to read

"..., and thus the size-segregated eddy correlation measurements of the number flux, $\overline{N'w'}$, may not be representative of the true aerosol flux, and net particle fluxes may be measured even where non is present.".

p 6292 line 1: rather than "turbulent data", day "turbulence data"

This sentence now reads *"Here, c_d is the drag coefficient which can be calculated from turbulence data or approximated through empirical relationships (Large & Pond, 1981; Yellend et al., 1998) and u* is the friction velocity."*.

p 6295 lines 15-16: Rather then saying what data are "available", scientists should specify the data that were "measured" or "used".

We have changed the word "available" to "used". The relevant text is now

"...pressure and water vapour density are used, also at 20 Hz, from a LI-COR LI-7500 open path gas analyser.".

p 6292 line 11: The main problem with the analysis is that the Junge fits have been adjusted incorrectly. This is a key aspect of the manuscript, whose purpose is to demonstrate improvement over the F84 and K01 "bulk" methods, and yet the authors appear not to have applied these methods correctly. The problem is most evident in Figure 3d, where the data display a reasonably linear behaviour, and yet the black line is clearly not a the correct least-squared fit, as is evident by simple inspection. It looks to this reviewer that the authors have failed to apply the log transformation prior to calculating least squares; put another way, the relevant values on the y-axis ticks of Figure 3 should be the exponents [2, 4, etc.], rather than [100, 10000, etc.], and the least-squares distances that are evaluated must be expressed in terms of these exponents, rather than in terms of particle counts. Such a failure to log-transform prior to least-squares minimisation would have the effect of granting extreme leverage to the smallest particles (whose absolute numbers are orders of magnitude higher) and allow them to wrench the regression line. This would explain why the "Junge fit" in Figure 3d does not come even remotely close to representing the slope that is evident in the data. Presumably, the authors have made the same mistake in all of their regression fitting and so all of the analyses, which must therefore be corrected in order to demonstrate conclusions such as those (p 6063, line 15) suggesting that the "bulk" methods can be improved upon.

The reviewer is correct that the fit in which the least-squares error is minimised in linear space (the black lines in Figures 3 and 4) are not appropriate for representing an aerosol spectrum. However, these fits have **not** been used in the later analysis; the 'correct' fits which were minimised in log-space and shown by the red lines in Figures 3 and 4 were used. The 'incorrect' (or more accurately 'inappropriate') fits are included here purely for completeness; if the aerosol spectra could be represented perfectly by a Junge power law, then performing the minimisation in log- or linear-space would be equivalent. However, as noted by Reviewer #2, including these fits adds little to the paper, and as is apparent here that their inclusion can easily lead to confusion. We have thus removed these fits from Figures 3 and 4, and any reference to them from the manuscript.

p 6297 line 23: The Junge fitting must be done via "log error minimisation", not "linear".

Please see the response to the previous point.

p 6298 line 9: rather than the blue/black, the K01/F84 lines in Fig. 5 of the PDF file is grey/black.

The blue/black referred to an old version of the figure. This has now been corrected.

p 6302 line 19: "there are times when none of the bulk methods considered here provide a suitable approximation". This is very likely related to errors in calculating the regression fits.

When referring to comparisons between the high-rate and bulk corrections, we are using the log least-squares fits as discussed above.

Figures:

Generally, the means of citing the figures in the text could be greatly improved. For example, rather than "Figure 1 shows a scatter plot of y versus x", it is preferable to say "A linear relation was found with generally very high correlation between y and x, but more noise at the low end (Figure 1)". In other words, the figure citation should explain generally what the Figure tells us, rather than give information that iss redundant in the caption or evident when glancing at the Figure (or both).

We have, where practical, improved the citation of figures throughout the manuscript.

In Figure 1-4, the ticks on the x-axis are not sufficiently labelled for the reader to understand the data. Probably, the ticks go from 0.2 to 8 microns, but this needs to be made explicit since only one tick is labelled.

Extra tick labels on the x-axis have been added to these figures.

Figure 2 (and subsequent figures) should have labelled panels (a) and (b)

Done.

I could find no reference to Fig. 7 in the text. Figure 10 is only briefly mentioned, despite containing 8 panels. Probably many of these results are not absolutely critical to the story being told, and could either be reduced or omitted entirely.

Discussion of these figures has been extended slightly; we feel that they are worth keeping in the paper as they provide an easy reference of how the various bulk corrections compare to the high-rate correction.

In Figures 7, 10, and 12 the units are missing for deposition velocity.

The units, m s^{-1}, have been added to these figures.