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

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## Abstract

The eddy covariance technique is the most direct of the methods that have been used to measure the flux of sea-spray aerosol between the ocean and atmosphere, but has been applied in only a handful of studies. However, unless the aerosol is dried before the eddy covariance measurements are made, the hygroscopic nature of sea-spray may combine with a relative humidity flux to result in a bias in the calculated aerosol flux. “Bulk” methods have been presented to account for this bias, however they rely on assumptions of the shape of the aerosol spectra which may not be valid for near-surface measurements of sea-spray.

Here we describe a method of correcting aerosol spectra for relative humidity induced size variations at the high frequency (10 Hz) measurement timescale, where counting statistics are poor and the spectral shape cannot be well represented by a simple power law. Such a correction allows the effects of hygroscopicity and relative humidity flux on the aerosol flux to be explicitly evaluated and compared to the bulk corrections, both in their original form and once reformulated to better represent the measured mean aerosol spectra. In general, the bulk corrections – particularly when reformulated for the measured mean aerosol spectra – perform relatively well, producing flux corrections of the right sign and approximate magnitude. However, there are times when the bulk methods either significantly over- or underestimate the required flux correction. We thus conclude that, where possible, relative humidity corrections should be made at the measurement frequency.

## 1 Introduction

Sea-spray aerosol, generated in the open ocean through bubble bursting in whitecaps, or as droplets are physically ripped from the crests of waves by the wind, is the second largest natural source of aerosol into the atmosphere after dust (Hoppel et al., 2002). As such, sea spray is expected to have a major impact on global climate, both through

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direct (Doss-Hammel et al., 2002) and indirect (Haywood et al., 1999) effects. An understanding of production rates and transport of sea-spray is thus a prerequisite for an understanding of the climate system as a whole. Sea-spray is also a potentially important mediator of air-sea fluxes of heat, moisture and momentum at high wind speeds ( $> 25 \text{ ms}^{-1}$ ) (Andreas, 1992), where large volumes of re-entrant sea-spray may be produced.

Calculation of a sea-spray source function requires the quantification the flux of sea-spray aerosol into the atmosphere. Traditionally this is done through one of two methods. Firstly, under the assumption that the local production and deposition are equal, the flux of sea-spray aerosol into the atmosphere may be inferred through a model describing the dry deposition (i.e. the sink) of aerosol – the “equilibrium method” (Fairall et al., 1983; Fairall and Larsen, 1984; Smith et al., 1993; Hoppel et al., 2002). The second method relies on the assumption that spray production can be inferred from the fraction of the sea surface that is covered by whitecaps. The amount of spray ejected into the atmosphere per unit area whitecap is empirically derived through studies in the laboratory (Mårtensson et al., 2003), or at field sites generally located in the surf zone (Clarke et al., 2006). The fractional area of the ocean covered by whitecaps can then – again empirically – be related to the wind speed and sea-state, allowing a wind dependent sea-spray source function to be derived (Monahan et al., 1982, 1986). Other methods used to calculate the surface source function, albeit less frequently, include the gradient method (Petelski and Piskozub, 2006); inverse modelling (Vignati et al., 2001; de Leeuw et al., 2003) and concentration increase with fetch (Reid et al., 2001).

Although the equilibrium and whitecap methods discussed above have both been widely used, they are both indirect, and source functions spanning up to six orders of magnitude have been reported (Andreas, 1998, 2002). More recent studies show source functions agreeing to within approximately one order of magnitude for particles with radii smaller than around  $1 \mu\text{m}$  (de Leeuw et al., 2011), although the equilibrium method is strictly only applicable to particles larger than about  $3 \mu\text{m}$  (Petelski and Piskozub, 2006; Andreas et al., 2010). A more direct method to measure the sea-spray



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source function is through the eddy covariance technique, whereby turbulent fluctuations in the vertical component of the wind are correlated with fluctuations in the aerosol spectrum, to produce a net vertical transport of aerosol particles (Nilsson et al., 2001; Geever et al., 2005; de Leeuw et al., 2007; Norris et al., 2008, 2012). The derivation of a sea-spray source function using the eddy covariance method still, however, relies on some assumptions. Most importantly, what the eddy covariance technique actually measures is the net aerosol flux (production–deposition), therefore it is only an adequate approximation to the true sea-spray source function when production is much greater than deposition. Note that this assumption is in direct contradiction of the conditions for which the equilibrium method is applicable, which relies on the assumption of zero net flux. In the limiting conditions of no deposition, or deposition equal to production, the eddy covariance or equilibrium methods respectively will give the correct surface source flux. In practice, however, where deposition generally is non-zero but smaller than production, both methods will result in an underestimation of the true surface source flux. It is also possible for deposition to exceed the surface source, in which case the equilibrium method will overestimate the source flux and the measured net eddy covariance flux will be of the wrong sign. In either case, eddy covariance measurement of the net flux corrected for deposition is the most direct method to determine the surface source flux.

The calculation of an eddy covariance sea-spray source function requires the collocation of high-frequency aerosol and vertical wind measurements. This requires the use of a small, weatherproof aerosol spectrometer or a long sample line from the point of measurement back to the aerosol instrument. The use of such a sample line introduces a number of issues: a lag between wind and aerosol measurements; a damping of the aerosol fluctuations at high frequencies and a loss of particles to the walls of the sample line. Corrections for these effects exist, but may rely on unproven assumptions or become significant for larger particle sizes ( $r > 1 \mu\text{m}$ ). Using a compact in-situ instrument with a short sample line, however, makes drying the aerosol prior to measurement extremely difficult, thus aerosol spectra will generally be recorded at ambient humidity.

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Sea-spray aerosol is extremely hygroscopic, and particles will rapidly grow or shrink in response to changes in the local relative humidity. In the presence of an upward relative humidity flux (which is almost ubiquitous in the marine environment), upward moving parcels of air ( $w' > 0$ ) will, on average, have a higher relative humidity than downward moving parcels. As sea-spray responds rapidly to RH fluctuations, a particle in an upward moving parcel may be larger than that in a downward moving parcel, even when these particles have equal dry radius. When measuring these aerosol in discrete size bins, this effect may cause the segregation of particles of equal dry radius into different bins, depending on the sign of  $w'$ . When using the eddy covariance technique to determine aerosol flux, this will be interpreted as a net flux of aerosol, even if none exists. Fairall (1984), hereafter F84, and Kowalski (2001), hereafter K01, have both presented “bulk” methods of correcting for this apparent flux, based on mean meteorological conditions and assumptions of the shape of aerosol spectra within the marine boundary layer. Here, we describe a method of accounting for the humidity flux at the measurement timescale (a frequency of 10 Hz) and compare these to the bulk corrections of both F84 and K01 for measurements from the SEASAW cruise in North Atlantic during March–April 2007. In the next section we discuss the theory behind the bulk correction methods and describe the high-rate method we have used to account for humidity variance in the SEASAW data, give a brief overview of the SEASAW cruise, and processing that the data has undergone. In Sect. 3 we evaluate the validity of the assumptions of spectral shape with regard to the SEASAW data, and present some alternative functional forms which better represent the aerosol spectra for these data. In Sects. 4 and 5 we compare the biases calculated through the high-rate method with those from the bulk methods, both in their original forms, and having been reformulated to use the functional forms which better represent the mean spectra recorded during SEASAW.

## 2 Theory

The hygroscopic nature of sea-spray aerosol means that the size of these particles changes rapidly in response to changes in the ambient saturation ratio,  $S$ , defined as the ratio of ambient to saturation vapour pressure. In situations where there exists a vertical flux of the saturation ratio (i.e.  $\overline{S'w'} \neq 0$ ), the number of deliquescent particles in a certain particle size interval is a non-conservative scalar, and thus size-segregated eddy correlation measurements of the number flux,  $\overline{N'w'}$ , may be biased, and net particle fluxes can be measured even where non is present.

### 2.1 Bulk corrections

Both F84 and K01 address the issue of apparent particle fluxes, both through an apparent transfer velocity,  $\Delta v_d$ , which is the transfer/deposition velocity induced through the vertical flux of saturation ratio. Both give the dependence of aerosol radius on humidity as

$$r(S) = r_0 \left[ 1 + \frac{\gamma}{1-S} \right]^{\frac{1}{3}}, \quad (1)$$

where  $r_0$  is the dry aerosol radius, and  $\gamma$  is a parameter related to the aerosol chemistry, taken as unity by F84 for clean marine air. Taking the derivative w.r.t.  $S$ , we have

$$\frac{\partial \bar{r}}{\partial \bar{S}} = \frac{\gamma \bar{r}}{3(1-\bar{S})^2 + 3\gamma(1-\bar{S})}. \quad (2)$$

Note: we have substituted  $\bar{r}_0 = \bar{r} \left[ 1 + \gamma / (1 - \bar{S}) \right]^{-1/3}$  in the above.

Under a fluctuation in saturation ratio,  $S'$ , the change in the ambient concentration at a fixed radius is a combination of the translation of the particle distribution in radial

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space and a “renormalisation” which occurs due to the change in the size intervals over which the distribution is defined. These two effects result in an induced change in the particle distribution which can be written, following K01, as

$$N' = r' \left[ \frac{\partial \bar{N}}{\partial \bar{r}} + \frac{\bar{N}}{\bar{r}} \right]. \quad (3)$$

Both K01 and F84 make the assumption that the aerosol spectrum can be approximated through a Junge power law (Junge, 1963) of the form

$$\bar{N} = \alpha \bar{r}^{-(\beta+1)},$$

where  $\beta$  is typically about 3, and thus

$$\frac{N'}{\bar{N}} = -\beta \frac{r'}{\bar{r}}. \quad (4)$$

Combining Eqs. (2) and (4) with the definition of the deposition velocity and taking  $\frac{\partial \bar{r}}{\partial \bar{S}} \equiv \frac{r'}{\bar{S}}$ , we arrive at K01’s expression for the bias velocity due to humidity fluctuations and hygroscopicity:

$$\Delta v_d = \frac{-\gamma\beta}{3(1-\bar{S})^2 + 3\gamma(1-\bar{S})} \overline{w'S'}. \quad (5)$$

K01 reformulates Eq. (5) in terms of temperature and moisture fluxes, however this is not required here. F84 makes the further approximation that, in the surface layer,

$$\overline{w'S'} = c_d^{\frac{1}{2}} u_* (1 - \bar{S}), \quad (6)$$

resulting in F84’s expression for the bias velocity:

$$\Delta v_d = \frac{\gamma\beta}{3(1+\gamma-\bar{S})} c_d^{\frac{1}{2}} u_*. \quad (7)$$

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Here,  $c_d$  is the drag coefficient which can be calculated from turbulent data or approximated through empirical relationships (Large and Pond, 1981; Yelland et al., 1998) and  $u_*$  is the friction velocity.

Both of these methods provide a bulk means of estimating the bias velocity due to hygroscopicity, but both rely on the assumption that aerosol spectra can be well represented with the use of a Junge power law, and F84's equation relies on the accuracy of the approximation of  $\overline{w'S'}$ .

## 2.2 High-rate corrections

The collocation of high frequency measurements of humidity and size-resolved aerosol spectra during SEASAW allows the effect of hygroscopicity to be explicitly calculated. In order to do this, the 10 Hz CLASP spectra must be individually corrected to a reference humidity, typically the run-mean relative humidity, following Zhang et al. (2006) and Lewis and Schwartz (2003).

Such a correction is relatively simple in the mean sense, where a long averaging period means that all size channels will have adequate counting statistics and a simple functional form (for example a Junge power law) can be fitted to the mean spectrum. Under a humidity correction, channel boundaries will change, but the number of particles,  $N$ , in each channel will be unchanged. Using the new channel boundaries,  $dN/dr$  and a new mean radius for each channel can easily be calculated. If the spectrum is required over specific channel boundaries or at a specific radius, this can easily be achieved by using the fit to the adjusted spectrum as an interpolant.

However, when adjusting spectra measured over much shorter time periods (here measurements are at 10 Hz), things are not so simple. Following an adjustment for relative humidity, each individual spectrum will be defined over a different set of channel limits. Additionally, the counting statistics for a spectrum measured over such a short period of time will be much more noisy than for a mean spectrum, and indeed may show little or no resemblance to the mean case. This makes it both difficult and inappropriate to use a functional fit to the spectra to redefine them over a consistent set

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of channel boundaries, and we must find an alternative means of evaluating these adjusted spectra over consistent limits. This is achieved as follows. For each channel  $i$  in the (adjusted) aerosol spectrum, a fit  $\mathfrak{N}_i$  through the channel of the form  $\mathfrak{N}_i = a_i r^{b_i}$  is calculated based on the values of  $\frac{dN}{dr}$  in channels  $i-1, i, i+1$ . This fit then provides an estimate of the particle distribution within each channel, and the integral  $\int_{r_i} \mathfrak{N}_i dr$  gives the total number of particles,  $\mathcal{N}_i$ , in channel  $i$ . Note that this approximation will result in a gain or loss of particles in each channel. We define this error as  $\epsilon_i = \mathcal{N}_i - N_i$ , where  $N_i$  is the CLASP measured number count in the (humidity adjusted) channel  $i$ . Then, for each (adjusted) channel  $i$  and (original) channel  $j$ , we find the interval  $[a_{i,j}, b_{i,j}]$ , if any, where channels  $i$  and  $j$  overlap. The number of particles that must be moved from channel  $i$  to channel  $j$ ,  $\Delta N_{i,j}$ , is thus

$$\Delta N_{i,j} = \left(1 + \frac{\epsilon_i}{N_i}\right) \int_{a_{i,j}}^{b_{i,j}} \mathfrak{N}_i dr, \quad (8)$$

where the factor of  $\epsilon_i/N_i$  is introduced to account for the error introduced in assuming a log-linear aerosol distribution through each CLASP channel. Note that this gives identical results to solving for  $a_i$  and  $b_i$  so that  $\int_{r_i} \mathfrak{N}_i dr = N_i$ , however it is significantly cheaper computationally.

An example of using this high-rate humidity correction method is shown in Fig. 1. The 12 000 individual aerosol spectra from a 20-min data record are shown in pink. These spectra are very variable, particularly in the larger CLASP size channels, and most have a spectral shape very different to that of the mean spectrum, shown by the solid red line. This is due to the discrete nature of aerosol measurements and the relatively small volume (5 ml) of each 10 Hz measurement. The black line shows this mean spectrum once it has been corrected from the run-mean relative humidity (60 %) to a reference humidity of 80 %. The blue line in Fig. 1 shows the mean of the 12 000 10 Hz spectra once they have been individually corrected to 80 % relative humidity via

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a 10 Hz humidity measurement collocated with the CLASP instrument. Note that after this procedure, each of these spectra is defined over a different set of channel limits, depending on the sign and magnitude of the individual relative humidity adjustment. This mean spectrum is thus defined over the mean channel limits of the high-rate aerosol spectra. While this isn't useful for any analysis, it does show that, before re-binning, correcting for humidity before time-averaging produces an almost identical spectrum to that produced by humidity correcting a run-mean aerosol spectrum. The dashed red line in Fig. 1 shows the mean of the high-rate spectra after they have been both corrected for relative humidity fluctuations and re-binned back to the original CLASP size channels at 10 Hz. This mean spectrum is almost identical to the other two adjusted mean spectra, showing that our re-binning technique works well at 10 Hz, where spectral shapes are very variable, and many spectra include isolated channels in which no aerosol particles are recorded. The only difference between this spectrum and the other two adjusted mean spectra is that the local maximum in channel 13 has been smoothed out during the re-binning, and channel 1 has a significantly smaller particle count than either of the other adjusted mean spectra. This latter effect is due to the nature of the re-binning, where individual particles are moved from one channel into another. When correcting from a low relative humidity to a higher humidity, as in this case, particles will generally be moved from smaller bins to larger (as particle size increases with increasing relative humidity), however we have no information about the aerosol spectra for particle sizes smaller than the lower limit of CLASP channel 1. This means that particles will move out of this channel into channel 2, but none will move into it. The number of particles in channel 1 after adjustment will thus be erroneously small, and we must reject this channel from any further analysis. Equally, when adjusting from a higher relative humidity to a lower, we cannot ascertain the number of large particles which would move into the largest measurement bin, thus under such an adjustment the largest measurement bin would have to be dropped. Note that adjusting relative humidity to a reference value of 80 % is a rather stringent test of the humidity correction algorithm. In practice, if the particle flux in each channel is the same sign,

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the 10 Hz spectra need only be adjusted to the the run-mean relative humidity for the purposes of evaluating the flux bias for a single record. The mean flux spectra may then later be adjusted to 80 % humidity (or any other reference value) using standard approaches. In this case, measurements will be corrected to both lower and higher relative humidities, and both the smallest and largest measurement bins must be disregarded. If the particle flux changes sign between size bins, adjustment of the flux spectrum may be inappropriate, and a high-rate correction to the reference humidity required. In this case, channels must be rejected depending on the largest RH adjustment required in this process. Two examples of our high-rate re-binning method for noisy 10 Hz aerosol spectra are shown in Fig. 2.

### 2.3 SEASAW data

The data which are used to test the high rate humidity corrections are from cruise D317 (21 March–12 April 2007) of the RRS Discovery made as part of the SEASAW project, a UK contribution to the international SOLAS programme (Brooks et al., 2009; Norris et al., 2012). Three dimensional winds and sonic temperature are available at 20 Hz from a Gill sonic anemometer, pressure and water vapour density are available, also at 20 Hz, from a LI-COR LI-7500 open path gas analyzer. Size resolved aerosol spectra are recorded at 10 Hz in 16 unequally spaced channels ranging between radii of 0.18 and 8.88  $\mu\text{m}$  by a CLASP instrument (Hill et al., 2008).

Aerosol, turbulent winds and humidity are time-matched at 10 Hz and split into “runs” of 20 min. These runs are checked to ensure that sonic temperature and momentum flux ogives and relative humidity flux ogives are suitably well behaved, with a characteristic flattening of the curve at low and high frequencies and a minimal distortion at the wave scale (Fairall et al., 1997). Any runs where this was not the case were rejected from the analysis. The run-mean 10 Hz relative humidity timeseries were also required to lie within  $\pm 10\%$  of the low-frequency relative humidity from a Vaisala HMP45 humidity probe (part of the ship’s permanent surface meteorology instrumentation), and all runs where this was the case were visually examined to ensure that the LI-COR and

low-frequency relative humidity timeseries showed a similar signal. Once this quality control had been carried out, run-mean CLASP aerosol spectra were then visually examined to remove any spectra that were clearly contaminated. This quality control left a total of 124 20-min runs with which to perform the analysis.

### 3 Functional fits to SEASAW data

Both F84 and K01 assume that, in the mean, an aerosol spectrum can be well described by a Junge relation of the form  $N = \alpha r^{-(\beta+1)}$ , with  $\beta$  typically around 3. However, it is not clear that such a relationship is appropriate for the SEASAW measured aerosol spectra, which tend to have a smaller change in  $dN/dr$  (in log space) with radius at smaller values of  $r$ , with a distinct change in gradient at around  $r = 1.5\text{--}2\ \mu\text{m}$  (between CLASP channels 4 and 5). Figure 3 shows Junge fits to the mean CLASP aerosol spectra in four randomly selected SEASAW runs, with the fit error minimised in different ways. The fits shown by the blue lines are generated by finding values of  $\alpha$

and  $\beta$  which minimise the L2 norm  $\left( \sum \left( \bar{N}_i - \alpha r_i^{-(\beta+1)} \right)^2 \right)^{\frac{1}{2}}$ . The goodness of this fit is clearly dominated by the error in the approximation to the two or three smallest CLASP channels, this results in a fit whose gradient is far too shallow, and thus the value of  $\bar{N}$  in the larger CLASP channels is overestimated by many orders of magnitude. The value of  $\beta$  for these fits is typically less than 0.2.

The green lines in Fig. 3 show Junge fits which are calculated by substituting  $N^* = \log(dN/dr)$  and  $r^* = \log(r)$  and finding a linear least-squares fit to  $N^*$  and  $r^*$ . This leaves us with  $N^* = ar^* + b$  and thus

$$\begin{aligned} \frac{dN}{dr} &= \exp(a \log(r) + b) \\ &= e^b r^a, \end{aligned} \quad (9)$$

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which is a Junge fit with  $\alpha = e^b$  and  $\beta = -(a + 1)$ . This results in a clearly improved fit over the length of the CLASP spectra, with a mean value of  $\beta$  over the SEASAW runs of 3.15. However, due to the characteristic shape of the SEASAW spectra, this may still result in an overestimation of  $\bar{N}$  by over an order of magnitude for small and large values of  $r$ , with an order of magnitude or more underestimation of  $\bar{N}$  for values of  $r$  in the middle of the spectrum. Clearly this is not an ideal situation.

Also shown in Fig. 3 are two further functional fits for  $dN/dr$  which offer an improvement over the simple log-linear Junge fits. The red line shows a quadratic fit of the form  $dN/dr = \exp\{ar^2 + br + c\}$ , while the magenta line shows a piecewise continuous Junge-type fit, with different values of  $\beta$  for channels 1–4 and 4–16. Both of these fits offer a significant improvement over a simple Junge fit. The piecewise Junge fit has the advantage that it can be used directly with the F84 and K01 adjustment methods, and will result in different bias velocities for channels 1–4 and 4–16. The log-quadratic fit has the benefit that it is continuously differentiable and so will result in a unique bias velocity for each CLASP channel, however it is strictly only valid within the radius range of the CLASP channels, and will diverge quickly from the CLASP recorded spectra outside this range. Using this functional form also complicates the expression for the K01 and F84 bias velocities due to the more complicated derivative w.r.t.  $r$ .

Figure 4 shows the mean error for each CLASP channel resulting from the four functional fits described above. The left panel shows the absolute error,  $|N_i - F_i|$ , where  $F$  is the fit to  $N$ , and the right panel shows this error normalised by the channel mean number count,  $|N_i - F_i|/\bar{N}_i$ . The Junge power law (following F84 and K01) which is fitted through linear error minimisation is clearly a very poor fit for CLASP channels greater than 3, as one would expect given the fit examples shown in Fig. 3. The Junge function fitted in log space performs much better, but still introduces a considerable error at the small and large ends of the CLASP spectrum. This Junge fit does perform better than either of the more complicated functional forms for particle sizes of around  $2\ \mu\text{m}$ , but this is simply where the function changes from underestimating to overestimating the particle count (with increasing  $r$ ). The piecewise-linear and log-quadratic fits both

perform similarly, and it is not immediately obvious from Fig. 4 which provides the best fit. We can calculate this more objectively by summing the relative errors over the CLASP channels. This gives a value of 10.9 for the piecewise-linear Junge fit and 9.2 for the log-quadratic fit, suggesting that the latter provides, on average, a superior fit to the CLASP spectra than does the piecewise-Junge fit. However, as the error metric for both of these fits are similar (especially when compared to the simple Junge fits), we will consider both of these later.

#### 4 Bulk methods for calculating bias velocity

Figure 5a shows a run-by-run comparison of the K01 (blue) and F84 (black) bias velocities,  $\Delta v_d$ , given by Eqs. (5) and (7), respectively. Both give similar, physically reasonable, values for the bias velocity, however K01's method (mean = 0.010,  $\sigma = 0.0069$ ) is generally more variable and very slightly smaller than F84's method (mean = 0.011,  $\sigma = 0.0040$ ). Given that the only difference between the K01 and F84 methods is F84's approximation of  $\overline{w'S'}$  as  $c_d^{1/2} u_* (1 - S)$ , the discrepancy between these methods suggests a significant error in the estimation of the saturation ratio flux. This can be seen more explicitly in Fig. 5b where we have directly compared the saturation ratio flux and F84's approximation to it. Where there is a significant saturation flux, F84's method tends to underestimate the saturation ratio flux by up to 50%. Where the saturation ratio is smaller, however, the F84 approximation may overestimate by a similar margin.

#### 5 Comparison of bulk and high-rate bias estimates

A comparison of the apparent deposition velocity,  $\Delta v_d$ , between the K01 and F84 methods and the high-rate method is shown in Fig. 6 for CLASP channels 2–9. Note that in the K01 and F84 methods,  $\Delta v_d$  does not depend on particle radius, so only the high-rate  $\Delta v_d$  is changing in each of these plots. In channels 2 and 3 the bias velocity

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from the bulk methods is comparable to that from the high-rate corrections, however in general they are biased significantly high. This is unsurprising given the assumption that the aerosol spectrum follows a single Junge law (with  $\beta \approx 3$ ), whereas, as we saw in Fig. 3, the SEASAW spectra generally have a much shallower gradient than this in channels 1–4, and a steeper gradient for channels 4–16. Using  $\beta = 3$  for the bulk approximations will thus inevitably lead to an overestimation of the bias velocity.

In CLASP channel 4, the bulk  $\Delta v_d$ , particularly from the K01 method, matches extremely closely with that derived from the high-rate method. Generally, it is around channel 4 where the CLASP aerosol spectrum steepens from a gradient of around 1.5 to around 5 (Table 1). Thus the mean gradient across channel 4 is only slightly larger than 3 so the bulk methods would be expected to perform well here, providing other assumptions and approximations on which these rely are met. Note that the K01 method makes fewer approximations than the F84 method does – namely the use of  $\overline{w'S'}$  rather than an estimate of it – explaining why it generally performs better. This is particularly pronounced in channel 4.

In channels 5–16 the  $\beta$ -value in the CLASP aerosol spectra increases to typically around 5, so the assumption that  $\beta \approx 3$  again fails, this time resulting in the bulk methods underestimating  $\Delta v_d$ . This is most pronounced in CLASP channel 5, although the tendency for the bulk methods to underestimate the bias flux persists through to the larger CLASP channels. As we move to the larger CLASP channels, however, the bias velocity calculated through the high rate method becomes increasingly noisy, making a direct comparison with the unchanging bulk methods less useful (this is likely due to a decreasing signal–noise ratio as fewer and fewer particles are recorded in the larger CLASP channels).

The bias velocity,  $\Delta v_d$ , is not a true deposition velocity, but an artefact of measuring size-resolved hygroscopic particle concentration in the presence of a humidity flux. However, this apparent deposition will lead to an error in the calculation of the particle flux  $\overline{w'N'}$ , by an amount equal to  $\Delta v_d \overline{N}_{RH}$ , where the RH indicates that  $\overline{N}$  has been evaluated at the run mean relative humidity. This bias flux may be comparable to, or



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larger than, the true particle flux when conditions are close to equilibrium (F84) and thus correcting for relative humidity may lead to a change of sign of the particle flux (generally from +ve to -ve when the relative humidity flux is upwards). The flux biases calculated from the high-rate and K01/F84 methods are shown in Fig. 8. In general the bias fluxes from the bulk method, particularly from the K01 method, match the high-rate method well. Where they are in error, they are generally biased high; this is particularly clear in CLASP channel 2 from around run 85 onwards, roughly corresponding to where the saturation ratio flux becomes small. There are also times when the high-rate flux bias is negative, which the bulk methods cannot reproduce (as by definition both  $\Delta v_d$  and  $\bar{N}$  are either positive or zero in the presence of an upwards humidity flux). Note that a negative flux bias may occur in a positive humidity flux as a result of the shape of the aerosol spectrum. If the spectrum is relatively flat for small values of  $r$ , then the expansion of the channel limits under increasing humidity may cause a loss of particles for certain size channels, even when adjusting to higher relative humidities. A negative bias flux may also result if the spectral shape or mean number concentration is correlated with relative humidity. This may occur as fresh plumes of sea-spray (with  $N' > 0$ ) increase the local ambient relative humidity through droplet evaporation. Such a situation would also invalidate the assumption of F84 and K01 that the dry aerosol distribution is well mixed and uncorrelated with relative humidity.

### 5.1 Bulk methods based on improved functional forms

#### 5.1.1 A piecewise linear Junge power law

Given the characteristic shape of the aerosol spectra in the SEASAW dataset, the use of a single Junge power law to approximate the spectra, as suggested by F84 and K01 is not particularly appropriate. An obvious improvement that can be made to this is to use two Junge-type fits: one between channels 1 and 4 where the CLASP spectra are generally quite shallow, and another between channels 4 and 16 where the spectra steepen, modified so these fits produce one piecewise-continuous fit. This gives us

different values for  $\beta$  for channels 1–3 and 5–16 which can then be used in the F84 and K01 expressions for the bias velocity. Due to the change of gradient at channel 4,  $\beta$  is undefined here, so we must use a mean of the two  $\beta$  values on either side. Generally this will give a value of  $\beta \approx 3$  at channel 4. Examples of this type of fit are shown over-lying CLASP spectra in Fig. 3.

### 5.1.2 Quadratic fit in $\log(dN/dr), r$

Another approach to dealing with the non-constant (in log space) gradient of the CLASP aerosol spectra is to fit a function which is continuously dependent on particle radius. For example, if we fit a quadratic in  $\log(dN/dr), r$  space, then we end up with a functional representation of the aerosol spectrum of the form

$$\frac{dN}{dr} = \exp \left\{ a\bar{r}^2 + b\bar{r} + c \right\}.$$

Again, examples of this fit are shown along with CLASP measured spectra in Fig. 3. Following K01 we may then derive a bias velocity which is continuously dependent on particle radius,  $\bar{r}$ . This gives us

$$\Delta v_d = \frac{\gamma \left( 2a\bar{r}^2 + b\bar{r} + 1 \right)}{3(1 - \bar{S})(1 - \bar{S} + \gamma)} \frac{1}{w'S'}. \quad (10)$$

### 5.2 Do these adjusted methods offer an improvement?

Timeseries and scatter-plots showing the relationship between the high-rate corrections and the piecewise-Junge F84 and K01 methods are shown in Figs. 9 and 10, respectively. Using the piecewise continuous Junge fits in place of a single Junge fit is essentially equivalent to multiplying  $\Delta v_d$  from the standard F84 and K01 methods by 0.5 in channels 1–3 (as the  $\beta$  value for a Junge fit over channels 1–4 is typically around 1.5) and by around 5/3 in channels 5–16 (where the typical  $\beta$  value is around

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5) Channel 4, where we must use the mean of the  $\beta$  values on each side, generally remains largely unchanged from the standard F84 and K01 estimations of  $\Delta v_d$ , using  $\beta \approx 3$ . This scaling has the effect of tempering the usual overestimation of  $\Delta v_d$  in channels 1–3 and the underestimation in channels 5–16. However, despite the piecewise continuous Junge fits producing a generally very good approximation of the SEASAW aerosol spectra, there are still times when there are significant differences between the bulk and high-rate methods, suggesting that variables not considered within the bulk methods may be important.

Using K01's method, modified to use a quadratic fit to the CLASP aerosol spectra (Figs. 11 and 12), offers a similar improvement to the estimation of the bias velocity over the standard K01 and F84 bulk methods, particularly at the smaller end of the spectra where the quadratic fit can represent the complete flattening of the spectra which is sometimes observed in the SEASAW aerosol data.

Although both the piecewise-Junge and quadratic forms of the K01 correction generally allow an improved bulk representation of the bias velocity (and resulting bias flux) it is clear that no one method is superior under all circumstances seen in the SEASAW data; indeed there are times when the simple Junge fit presented by K01 offers the best approximation to the “true” bias velocity calculated through our high-rate method. Furthermore there are times when none of the bulk methods considered here provide a suitable approximation of the hygroscopicity-induced bias suggested by the high-rate method. This can be seen clearly in Figs. 13 and 14, where the particle number fluxes,  $N'w'$ , are shown for CLASP channels 2–15 uncorrected for RH variations, corrected using our high-rate method, and corrected using the various bulk methods discussed here.

## 6 Conclusions

The use of the eddy covariance technique to measure the size-segregated flux of sea-spray aerosol (or other hygroscopic aerosol) in the presence of a relative humidity flux

may lead to a significant systematic bias in the recorded flux. “Bulk” methods (F84 and K01) have been presented to account for this bias using mean meteorological conditions (F84) or turbulent measurements (K01) and an assumed mean shape of the aerosol spectra, in the form of a Junge power law.

In this paper we have developed a method for correcting aerosol spectra for variations in relative humidity at the high temporal resolution required for the calculation of eddy correction fluxes, allowing the flux bias caused by the relative humidity flux to be explicitly calculated. We have also reformulated the corrections given by F84 and K01 to use a more representative shape of the mean aerosol spectra. In situations where turbulent (high-rate) measurements of humidity are not available and aerosol spectra have not been dried, the bulk correction described by F84 (1984) provides a reasonable estimation of the flux correction which must be applied to account for the effects of hygroscopicity. If turbulent measurements of humidity are available, then the bulk correction of K01 (2001) offers an improvement, particularly when modified to better model the shape of the mean aerosol spectra. However, these bulk methods are far from infallible and may, at times, significantly under or overestimate the required flux correction. In situations where collocated turbulent aerosol and humidity measurements are available, the high-rate correction method is recommended, despite the relatively high computational cost.

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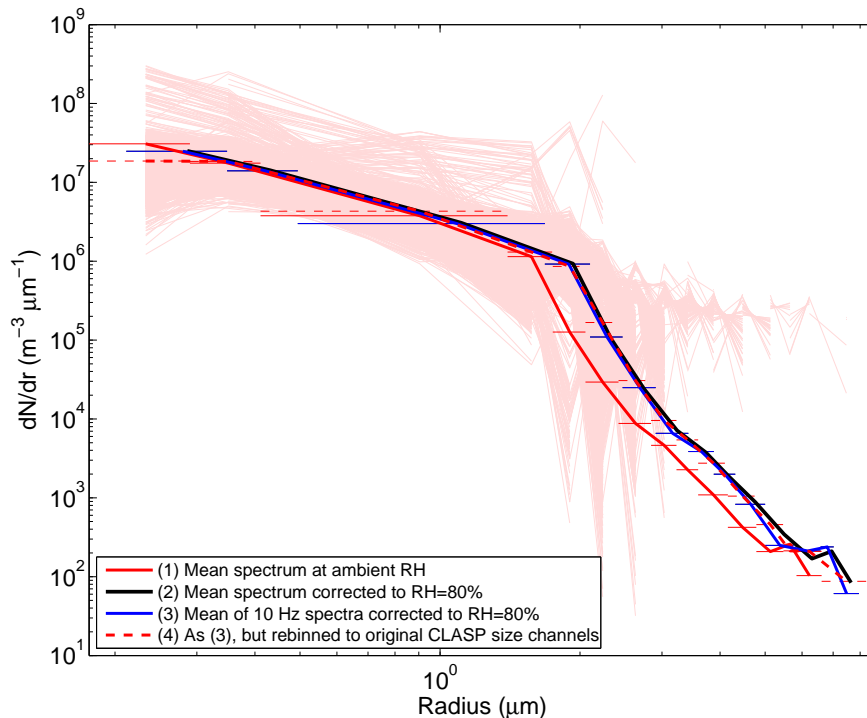
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**Table 1.** Characteristics of  $\beta$  values of different Junge-type fits to the SEASAW data.

	Mean	Min	Max	$\sigma$
Junge $\beta$ (linspace)	0.266	-0.662	5.401	0.911
Junge $\beta$ (logspace)	2.708	1.756	5.188	0.440
PW-Junge $\beta_1$	1.056	-1.698	2.730	0.709
PW-Junge $\beta_2$	5.042	3.341	6.743	0.672





**Fig. 1.** An example of using the high-rate relative humidity correction technique described here. The pale red lines show the 12 000 individual spectra which were measured at 10 Hz by the CLASP unit. The solid red line show the mean of these spectra at the mean ambient relative humidity. The black line shows the mean spectra corrected from the mean relative humidity (60 %) to a reference humidity of 80 %. The blue line shows the mean of the 12 000 individual spectra after each has been adjusted to 80 % relative humidity (note that this spectrum is then defined over the mean channel boundaries of these individual adjusted spectra). The dashed red line shows the mean of the 12 000 spectra once they have been both adjusted to 80 % and rebinned back to the original CLASP channel limits.

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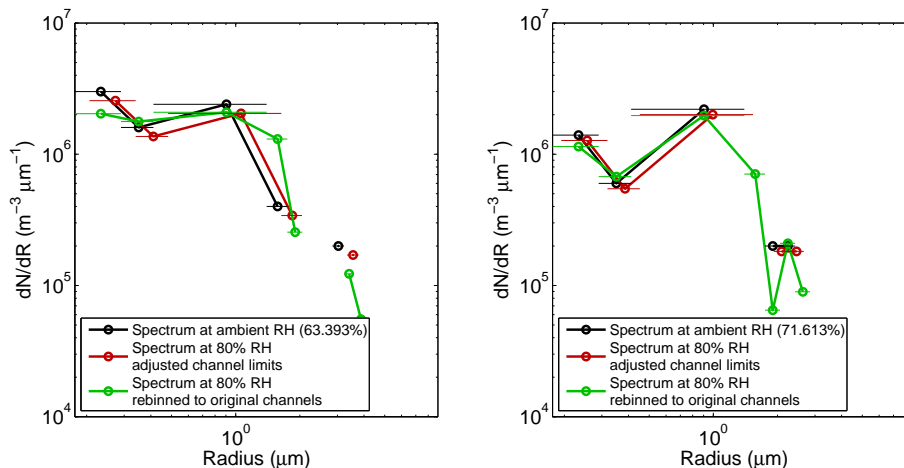
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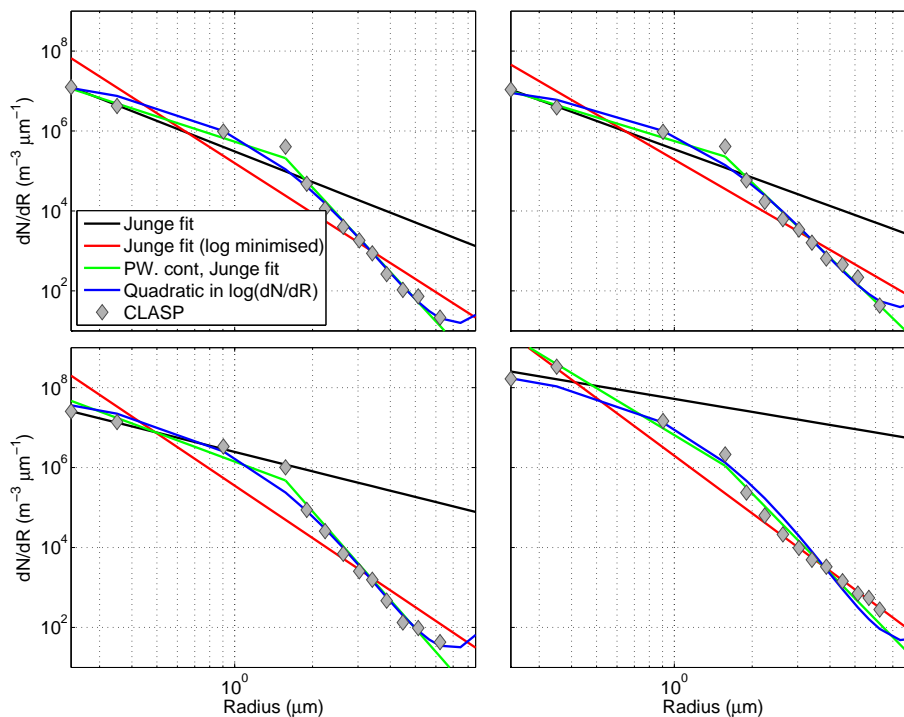


**Fig. 2.** Two examples of a relative humidity correction applied to sea-spray aerosol spectra recorded at 10 Hz. The black lines show the original spectra at ambient humidity, the red lines show these spectra corrected to 80 % relative humidity, and the green lines these corrected spectra after re-binning to the original instrument channels.

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**Fig. 3.** Four different fits encompassing three different functional forms to aerosol spectra from four randomly chosen 20-min SEASAW runs.

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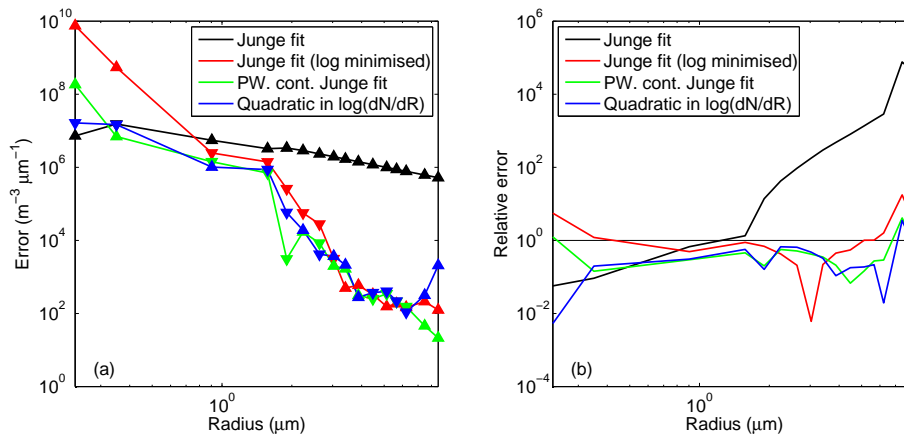
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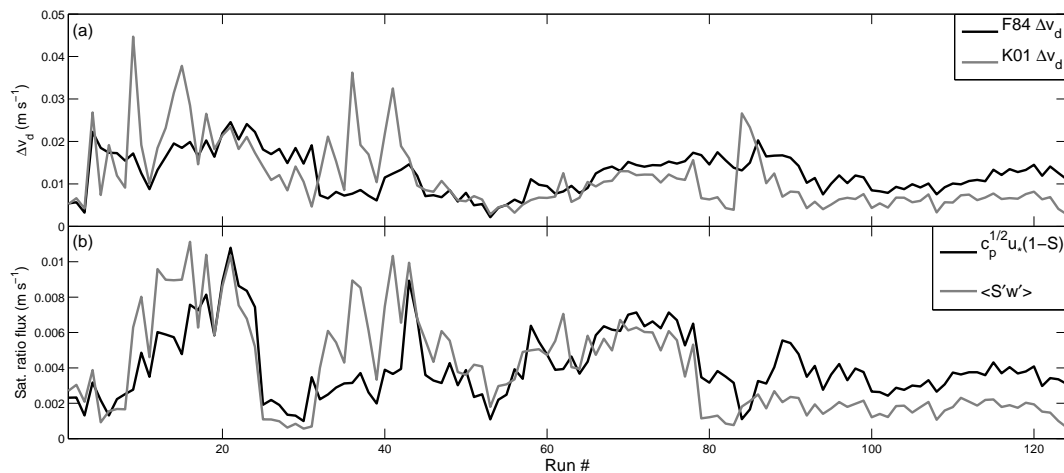
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**Fig. 4.** Mean absolute (a) and relative (b) errors for the fits shown in Fig. 1. The relative errors are simply the absolute error normalised by the mean particle count,  $\bar{N}$ . The triangles in (a) indicate whether the mean error is positive or negative.

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**Fig. 5.** (a) Run-by-run comparison of K01 and F84 methods for calculating bias velocity,  $\Delta v_d$ ; (b) run-by-run comparison of saturation ratio flux and the F84 approximation to it.

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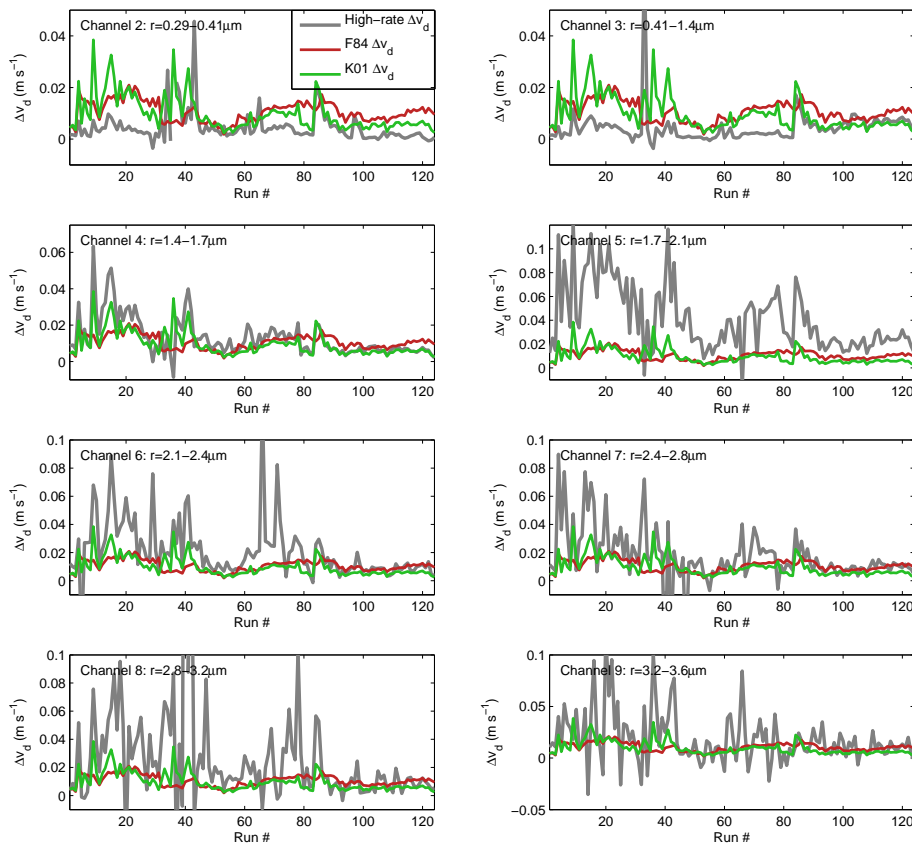
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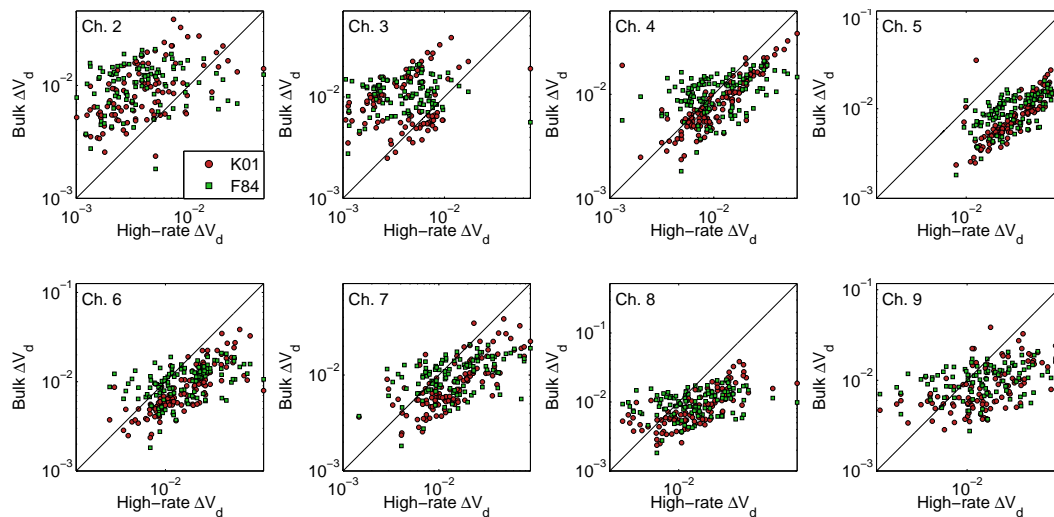
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**Fig. 6.** Apparent deposition velocity due to hygroscopicity,  $\Delta v_d$ , calculated through K01's and F84's methods, and the difference in the high-rate deposition velocity between ambient and RH-corrected (to run mean) spectra. CLASP channels 2–9 are shown.

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**Fig. 7.** Scatterplots showing the relationships between CLASP  $\Delta V_d$  and K01/F84  $\Delta V_d$  in Fig. 6.

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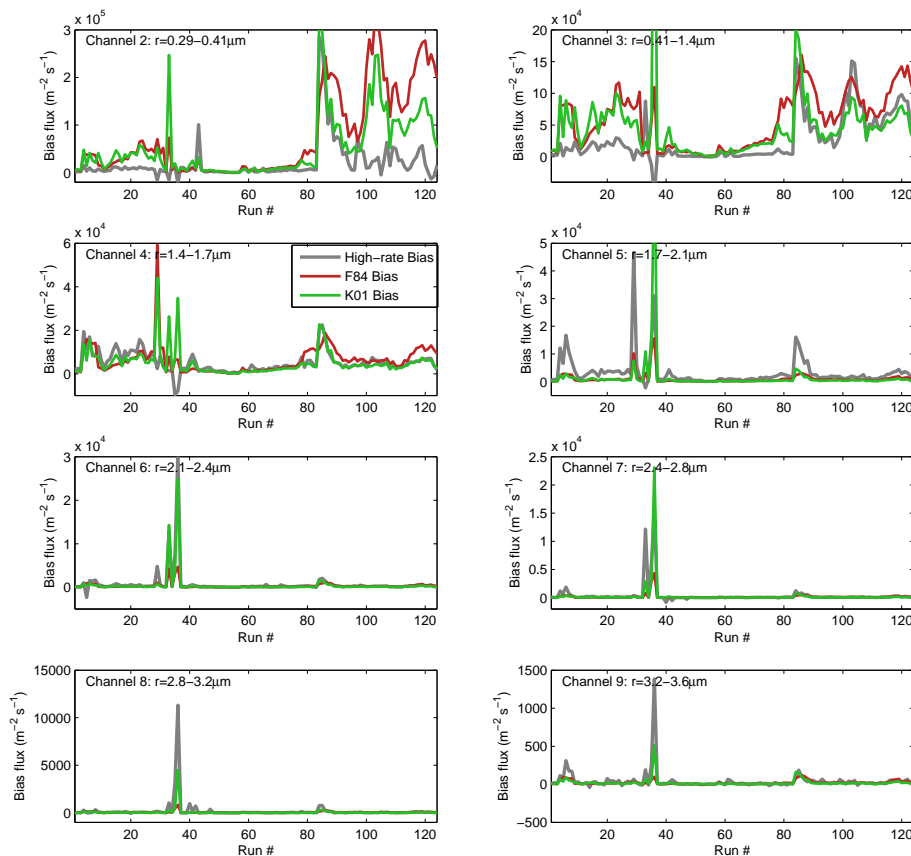
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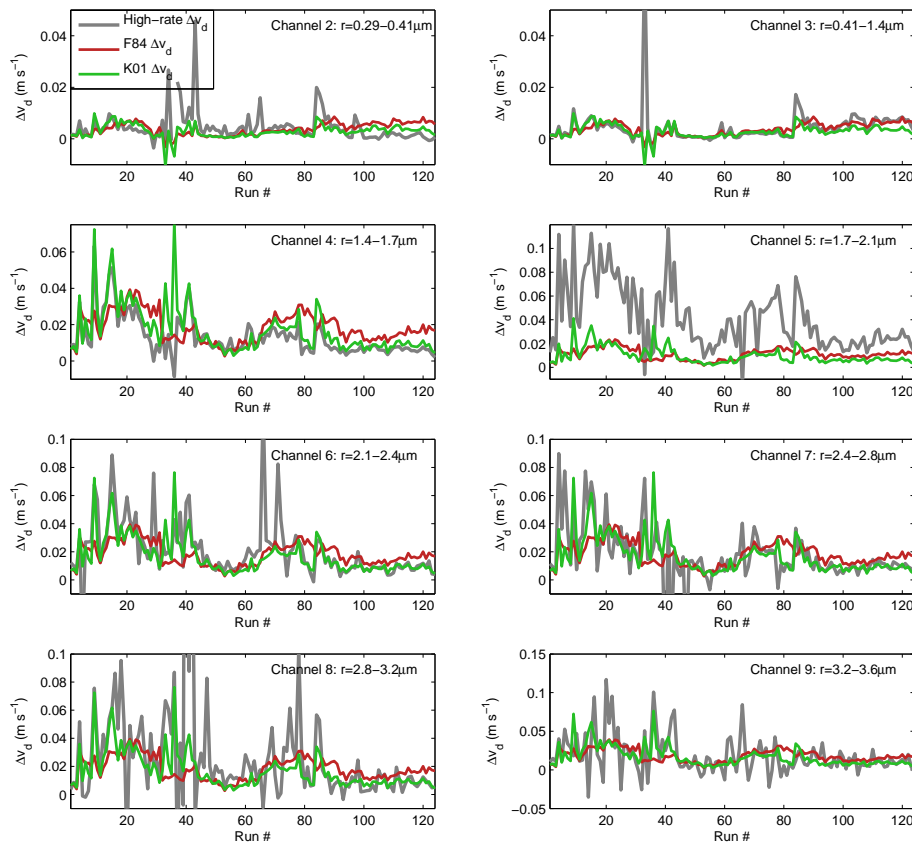


**Fig. 8.** Flux bias due to hygroscopicity,  $(= \Delta v_d \overline{N}_{RH})$ , calculated through K01's and F84's methods, and the difference in the high-rate number flux,  $\overline{N'w'}$ , between raw and RH-corrected (to run mean) spectra. CLASP channels 2–9 are shown.



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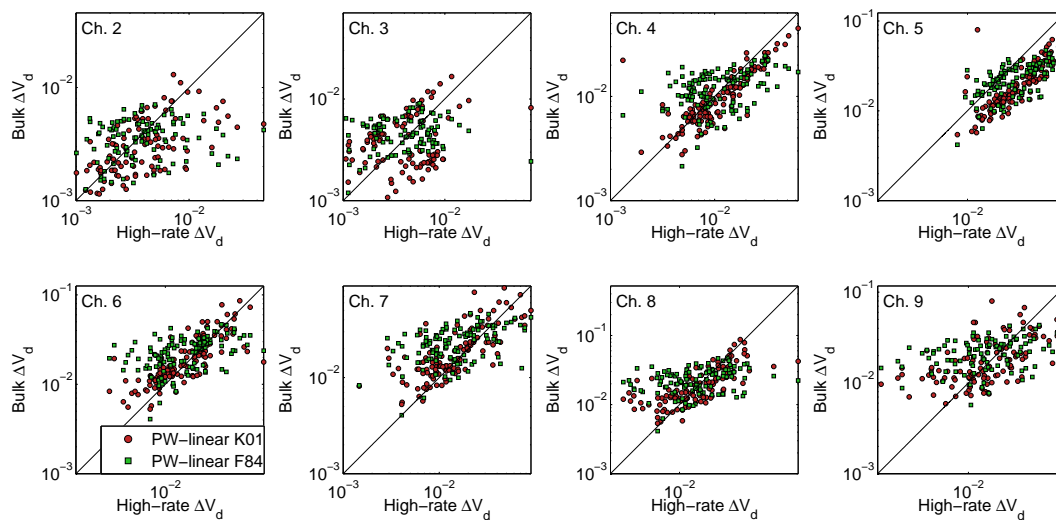


**Fig. 9.** Apparent deposition velocity due to hygroscopicity,  $\Delta v_d$ , calculated through K01's and F84's methods, modified to use a piecewise (log-)linear Junge form, and the difference in high-rate deposition velocity between raw and RH-corrected (to run mean) spectra. CLASP channels 2–9 are shown.

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**Fig. 10.** Scatterplots showing the relationships between the high-rate  $\Delta V_d$  and modified K01/F84  $\Delta V_d$  in Fig. 9.

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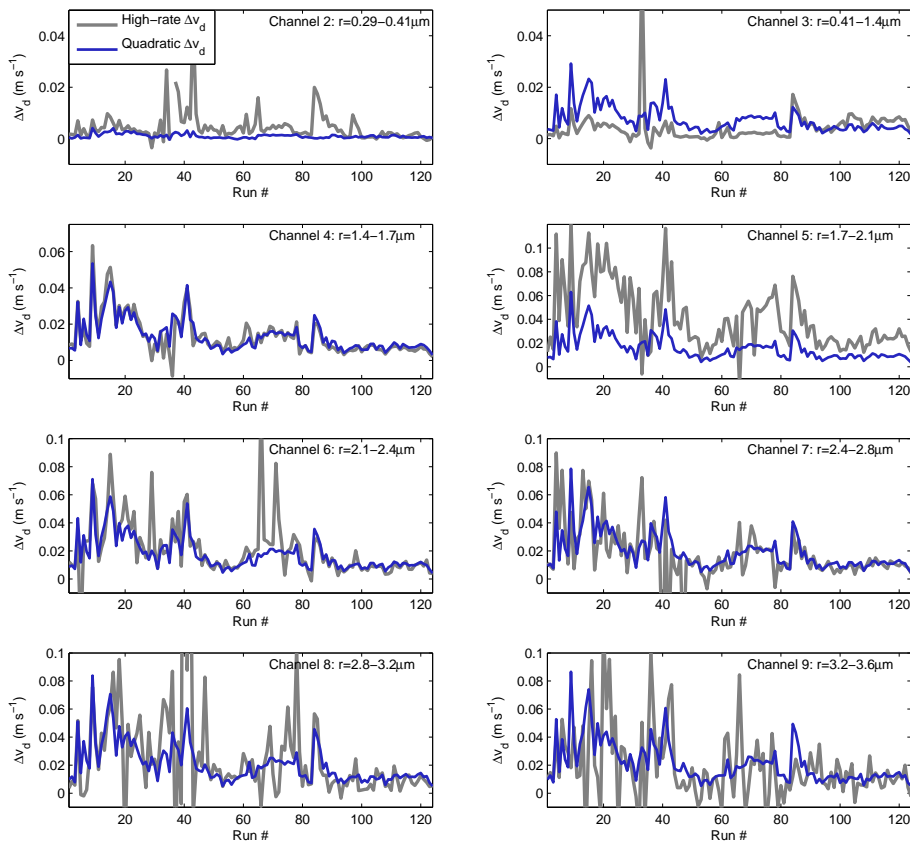
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**Fig. 11.** Apparent deposition velocity due to hygroscopicity,  $\Delta v_d$ , calculated through K01's method, modified to use a log-linear quadratic functional form, and the difference in high-rate deposition velocity between raw and RH-corrected (to run mean) spectra. CLASP channels 2–9 are shown.

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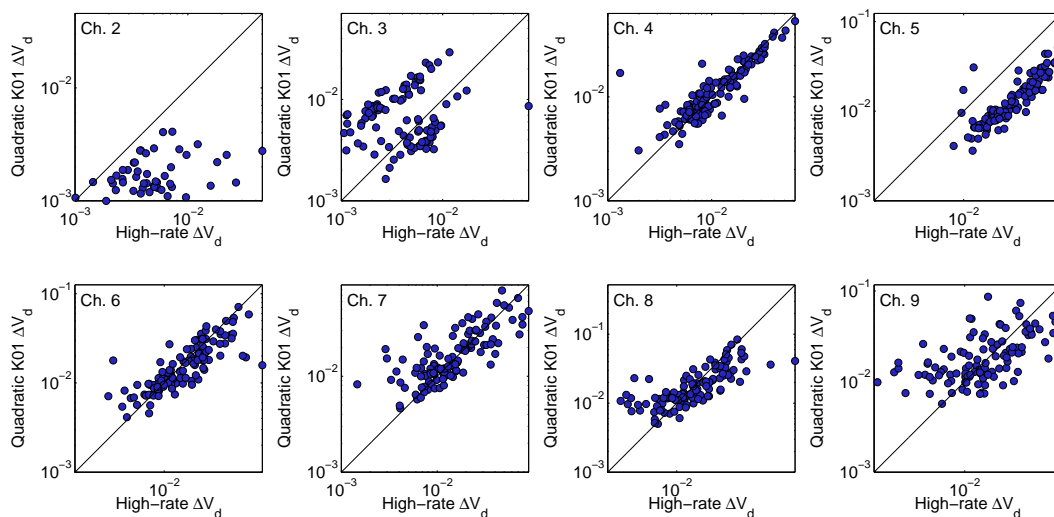
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**Fig. 12.** Scatterplots showing the relationships between high-rate  $\Delta V_d$  and K01 (modified to use a log-linear quadratic fit)  $\Delta V_d$  in Fig. 11.

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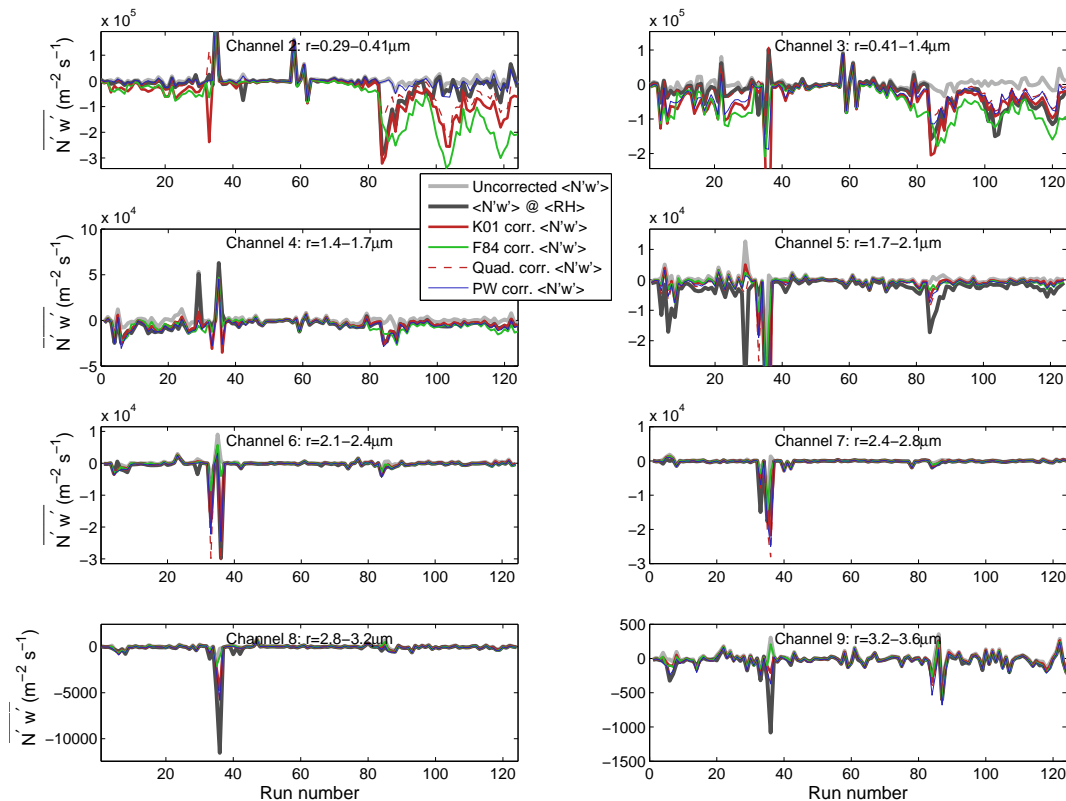
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**Fig. 13.** CLASP number flux,  $\overline{N'w'}$ , (grey line) corrected with high-rate (black), K01 (red), F84 (blue), log-linear quadratic (green), and piecewise (log)-linear (cyan) methods. CLASP channels 2–9 are shown.

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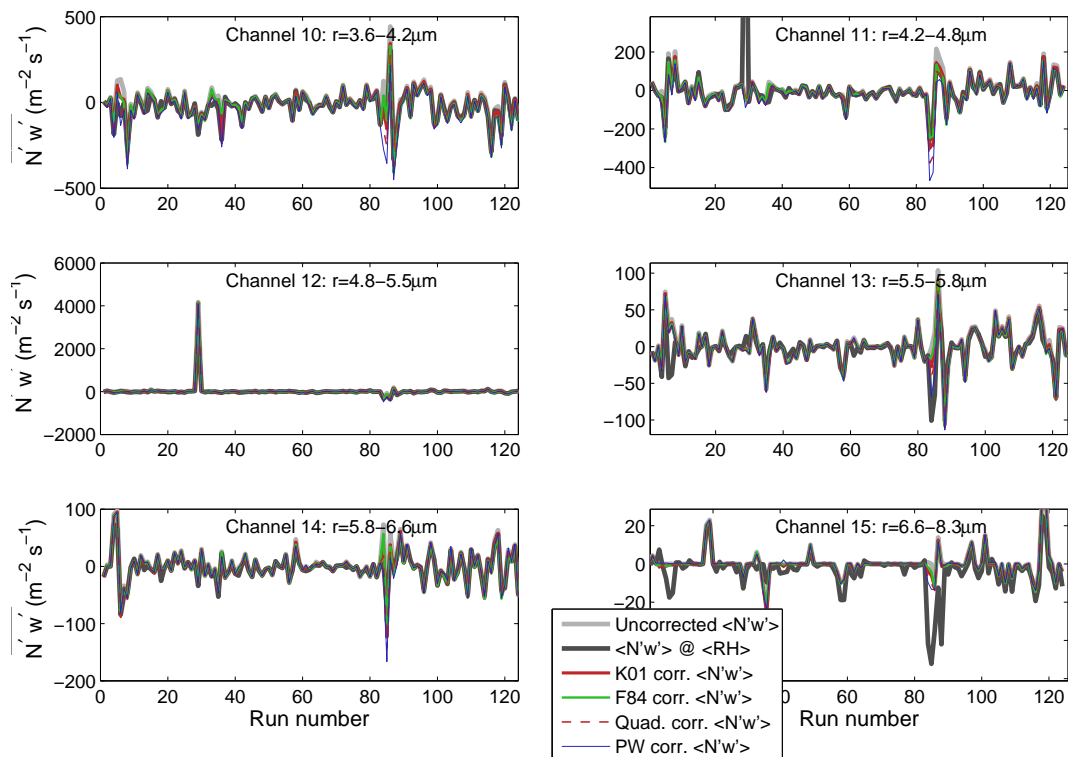
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**Fig. 14.** CLASP number flux,  $\overline{N'w'}$ , (grey line) corrected with high-rate (black), K01 (red), F84 (blue), log-linear quadratic (green), and piecewise (log-)linear (cyan) methods. CLASP channels 10–15 are shown.

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