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Atmospheric Measurement Techniques Discussions

Interactive comment on "Determining the sea-air flux of dimethylsulfide by eddy correlation using mass spectrometry" *by* B. W. Blomquist et al.

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We are grateful to the reviewer for the comments on this manuscript. We know it takes a significant investment of time and effort to undertake a critical review and agree the final result is better for it. The comments are both substantive and stylistic/typographical. We will begin by addressing the substantive comments in the order presented in the review. We will quote the comment in italics where appropriate for clarity.

1 Substantive

1.1 Frequency Response Comment

It should be pointed out in section 4.3 that the input function for the DMS frequency response estimation is not a sine wave, but a square wave. This makes a difference when looking at the amplitude response. For example, for a first-order response function with a 3 db point at 6 Hz for a sine wave, the 3 db point for a square wave is at about 7.8 Hz. That is, the sine wave response function drops off faster that the square wave response. The analysis of Lenschow and Raupach uses a sinusoidal response function. This needs some discussion.

In this case the input function generated for the lab experiment is certainly not a sine wave, but neither is it a perfect square wave due to the response characteristics of the switching valve and pressure/flow effects. At higher frequencies the waveform observed in trials with the short inlet and no dryer is a bit rounded. The difference between the synthetic rounded-square-wave and a turbulent flux signal is probably less than the comparison between a pure sine wave and perfect square wave would suggest, but we agree it is possible the true instrumental half-power frequency is a bit lower than the experimental result presented in Figure 6, and the correction applied to the cospectra may be somewhat less than required. We can discuss this in the text. This is one reason we mention the alternate or additional approach of fitting the -4/3 line to the inertial subrange as a further correction for high frequency losses (Figure 9). And there are certainly other approaches involving spectral similarity with heat flux, etc. Lenschow and Raupach generated a true turbulent flux of water vapor in a wind tunnel, not a sine wave, but this was not feasible for DMS in this case.

1.2 Comments on Eq 6-9

Equations (6) - (9) need some work. Equation (7) is in error. This should be an equation for the rate of change of mean c, not mean plus fluctuating c, and should have terms on the right involving mean advection, not mean plus fluctuating advection terms. I do not think it is necessary to go through this too abbreviated discussion of how to obtain (7) from (6), especially since it seems incorrect. I suggest just presenting (7) with an appropriate reference. (7) does not reduce to (8) under the stated assumptions. It reduces to (8) only at the surface, where turbulent diffusion is assumed to vanish, as you state in the next sentence. Integrating (8) to obtain (9) is legitimate only in the molecular sublayer. What you are doing is assuming a constant flux in the surface layer and equating the surface flux to the eddy flux within the surface layer, not integrating (8) up to some level z...

Equation 7 is indeed wrong. The proper result for decomposition of Eq. 6 into mean and fluctuating components, averaged over time (assuming ergodicity), is

$$\frac{\partial \overline{c}}{\partial t} = -\frac{\partial}{\partial x_i}\overline{u_i}\,\overline{c} - \frac{\partial}{\partial x_i}\overline{u_i'c'} + D\frac{\partial^2}{\partial x_i^2}\overline{c} - \overline{L} \tag{7a}$$

which yields the following, recognizing $\overline{w} = 0$ and assuming horizontal gradients in DMS are negligible.

$$\frac{\partial \overline{c}}{\partial t} = -\frac{\partial}{\partial z}\overline{w'c'} + D\frac{\partial^2}{\partial z^2}\overline{c} - \overline{L} \tag{7b}$$

This result reduces to Eq. 8 with the further assumptions of steady state ($\frac{\partial \overline{c}}{\partial t} = 0$) and no chemical loss. As the reviewer notes, this is not new material, and as presented in this corrected form is essentially a reiteration of the aerosol budget of Businger(1986),

adjusted slightly for DMS - i.e. excluding gravitational deposition and including a photochemical loss term. We're grateful for the correction and sorry to have missed it in earlier proof readings, but believe this short review is important to set the stage for the subsequent discussion. We are indeed equating the constant flux in the surface layer to the turbulent flux measured at height z (Eq. 9), and it is the assumptions leading to this result we wish to reinforce in the readers mind.

As the reviewer observes, and as we state in the text, we explicitly neglect advection and chemical loss in deriving Eq. 9. Within the context of most ship-based flux measurements - i.e. the determination of sea-air transfer velocity over timescales of 10-60 minutes - these are standard, though often easily overlooked, assumptions. The subsequent discussion in section 5.2 is intended to examine conditions when neglecting advection and chemical loss will lead to significant errors in the measured surface flux and derived transfer velocity. In most cases, it is sufficient to merely recognize when such conditions exist and exclude those periods from further consideration. This is in contrast to the context of developing a scalar budget for DMS in the marine boundary layer, where you would certainly not choose to neglect advection or chemical loss.

1.3 Comments on Sec 5.1

Similarly, Section 5.1 needs rewording. For DMS, $F_0 > F_i$ only increases BL DMS if the flux divergence exceeds the chemical loss. The "existence of a significant entrainment flux" is not the reason that the DMS flux is a linear function of height. Rather it is the result of assuming a constant loss term with height and a constant (or zero) gradient of DMS throughout the BL and no significant mean advection.The DMS flux can be expressed as a function of z. The boundary fluxes F_0 and F_i are prescribed.

The flux error would better be written in (11) as $\Delta F_0/F_0$, since it has been normalized by the surface flux. This is the way that it is used later, in (15) and (17).

We can certainly mention here the assumptions leading to linearity in Eq. 10, as noted by the reviewer: 1) constant loss rate with z and constant (small) gradient in DMS concentration, both of which are consistent with a well mixed MBL, and 2) no net advection, which is an assumption stated in the prior section. We can express the error as $\Delta F_0/F_0$.

1.4 Comments on Sec 5.1

The second term on the right side of Equation (12) does not follow directly from (7). Besides the fact that (7) is written for means plus fluctuations, to go from (7) to (12) you invoke the continuity equation. Also, the third term contains horizontal flux transport, which seems not present in (7).

With the corrections mentioned above, Eq. 12 should be consistent with Eq. 7a and is similar to the result in Businger (1986), with the addition of the chemical loss term.

1.5 Comments on Sec 6.1 relating to flux error

In discussions among the coauthors, we have previously determined that this section needs a major revision. Equation 15 is an approximation, and the analytical approach taken here overlooks effects of uncertainty in the derivation of this approximation. Furthermore, the way random noise was incorporated into the error formulation was incorrect. Also, we would like to examine the dependence of flux error on atmospheric stability. It is wrong to say that band-limited white noise has zero integral scale, but we will address that in the revision. We are submitting a substantial revision of Section 6.1 in a separate posting.

1.6 Comments on Sec 6.3 relating to motion correction and gradient effects

The analysis of (20) and (21) assumes that the covariance of vertical displacements with vertical changes in DMS concentration (i.e. the product of w' and the second term in the second parenthesis) is zero. There is no reason to believe a priori that this is the case.

w' is a property of the turbulent field and is uncorrelated to fluctuations in concentration represented by the motion term $\frac{\partial c}{\partial z}\left(z(t)-\overline{z}\right)$. The mean of the product of these terms will therefore be zero.

1.7 Comments on Sec 6.4 relating to frequency attenuation

The analysis of 6.4 does not take into account the phase shift that may occur between w and c. This can be larger than the amplitude attenuation, and also needs to be included.

Phase shift between w' and c' occurs for several reasons, including gas transit time through the inlet, poor clock synchronization between the w and DMS measurement, and attenuation-induced phase shift in the DMS measurement. In the computation of the covariance, correction for the total phase shift is the first step, as described in Section 3.2. The discussion in Section 6.4 assumes the two measurements have been brought into phase.

2 Stylistic

The reviewers principle stylistic objection seems to be with the placement of equations. We certainly agree an equation should be presented as close as possible to it's first point of reference. It's largely a personal preference to prefer splitting a sentence to insert the equation immediately rather than waiting a line or two. The former can lead to broken or choppy prose and the latter to a bit of searching on the part of the reader. Except for a couple cases, the equations are presented within two lines of the first reference in the text. We feel the paper reads fine as is, and a few other reviewers have indicated as much, but do not hold a strong opinion on the matter and will defer to the wishes of the editors at Copernicus if they have a preference.

3 Minor Comments

Most of the minor corrections are easily remedied. We will only comment specifically on those listed below.

p. 1982, l. 26: I suggest that it might be better to put in the derivation of (4) earlier so that the reader does not have to jump ahead to find equation (4).

We can simply place Section 2.4 after Section 2.6 to avoid the forward reference.

p. 185, l. 11: I do not understand how "rotating the wind coordinates to achieve... zero crosswind component" can correct for airflow distortion.

The correction described here is merely to achieve zero mean vertical and crosswind components, rotating the coordinates into the mean wind, streamwise, so u becomes the relative wind speed. Without this correction, the turbulent component of w will be contaminated by gustiness in horizontal wind. We can make this more clear in the text.

The rotation does not compensate for an increase in mean windspeed as the air moves over the obstruction. A further correction is often done to adjust measured wind speed to the standard 10 m reference height and neutral conditions, yielding mean wind speed over the surface of the ocean. This involves information from airflow models of the particular ship, knowledge of the ship's speed and an assessment of atmospheric stability.

While not necessary for the covariance calculation, it is essential for interpretation of the measured transfer velocity.

p. 1987, l. 14, and Fig. 5: I don't understand why you define the normalized cospectra as "transfer velocity" spectra. This seems completely misleading, as a cospectrum is in no way a spectrum.

This is a bit nonstandard (hence the quotes) and we can simply state the cospectra were normalized to seawater DMS concentration prior to averaging.

p. 2000, l. 6: This is true if there is no real contribution at low frequencies. How do you know this? You should cite a reference for this.

We are not saying flux at low frequencies will average to zero, only that the effects of variability in the flux signal, which is significant at these frequencies and leads to scatter in transfer velocity estimates, will tend to average out if you have a large number of samples, and therefore it isn't necessary to reject samples with large positive or negative flux contributions at the lowest frequencies.

If there is a significant, real flux contribution at lower frequencies, it will be apparent in a cospectrum averaged over many samples (many hours or a day, for example). Evidence of missing flux at low frequencies implies bias in the flux measurement and argues for longer integration times for the individual measurements. This is a separate issue.

p. 2001, l. 2: The quantity F_0/z by itself does not seem particularly useful. Near the surface it will be large, and higher up it will be small regardless of how close the regime is to steady-state.

In ship experiments, the flux is not measured at various heights throughout the boundary layer, but at one more or less constant distance above the surface. At that height, the quantity F_0/z can be estimated for current conditions, as outlined in Section 5.2, and can be used as a flag for situations when advection or chemical loss rate may bias the surface flux estimate. In practice, the chemical loss rate should never be a problem, but advection may lead to $\partial\hat{\bar{c}}/\partial t \approx F_0/z$ near a strong surface source.

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