

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

Anonymous Referee #1

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The paper contains lots of useful information on measuring eddy covariance flux of DMS via APIMS, but there are lots of issues to be addressed and errors to be corrected before it is acceptable for publication. Below are a list of major and minor comments to be considered.

MAJOR COMMENTS:

The usual practice when introducing equations in a written text is to write the equations immediately after introducing them. Here, the equation introductions are often followed by an equation number, and later the equation is presented. I suggest going back to the conventional way.

It should be pointed out in section 4.3 that the input function for the DMS frequency

C595

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 than the square wave response. The analysis of Lenschow and Raupach uses a sinusoidal response function. This needs some discussion.

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 . I actually don't see a reason for going through this analysis, since this is standard stuff, and there are so many errors. Just state the relevant equations, like (7), and leave out the molecular diffusion estimate of surface flux-(9).

p. 12, Eq. (7): This equation and subsequent equations are incorrect, and the derivation needs to be redone. The substitution of mean and fluctuating quantities into (7) needs to be done for all the variables. As it stands, it seems to have been done for only some of the terms in the equation, and ignored in others. Also, referring to equations presented later in the text is unconventional. This happens frequently in subsequent equations and can easily be remedied by putting in the equation when it is first referenced. An example is on l. 370. I suggest that it be reworded as follows: ...loss term, L , (7) reduces to [then write Eq. (8)]. However, the molecular diffusion term as it is written in (8) is incorrect. It is only true just above (i.e. within a few mm of) a flat surface

C596

where the flux is due solely to molecular diffusion. I agree that (9) is correct (at night and near the surface), but the path taken to obtain it is incorrect. It would be preferable to generalize this, since in daytime, L needs to also be included. I actually don't see a necessity to "derive" (9), since it's not really used, and is inconsistent with (12).

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).

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).

There is a problem with (16). The integral scale τ_i is relevant only to the actual standard deviation of DMS, not to the DMS standard deviation plus noise. The noise contribution has zero integral scale. Therefore, you cannot just substitute (16), which is signal plus noise, into (15), which applies only to the real signal. Also, you need a reference for the integral scale, and to say that this expression applies only in the surface layer. It does not apply throughout the BL. You also need a reference for the MOST scalar standard deviation.

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.

C597

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.

MINOR COMMENTS:

p. 1974, l. 19: Need to specify the measurement height to make the statement that "...bias in surface flux estimates arising from the [vertical] flux gradient [is] are not generally significant,..."

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).

p. 1983, l. 11: alternatively instead of alternately

p. 1984, l. 2 and p. 1996, l. 18, p. 2000, l. 10: principal instead of principle

p. 1984, l. 18: I'm not familiar with "heave" in this context. Need to emphasize that this is for the surface layer.

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

p. 1986, l. 2: reference to figure 7 appears before reference to earlier figures. Why not put Figure 7 closer to the reference?

p. 1986, l. 12: Counting error variance...

p. 1986, l. 17: Has Δt been defined?

p. 1986, l. 19: ...instrumental noise variance.

Figure 4 legend: ...indicating additional sources of uncorrelated random noise...

p. 1987, l. 6: Don't you mean "Cospectra" rather than covariance spectra? Covariance spectra could be interpreted as the spectrum of the covariance, which is not what you mean.

C598

- 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.
- p. 1987, l. 23: ...by moderate lengths of inlet tubing... The tube length needs to also be taken into account. Even with turbulence, if a long tube is used, significant attenuation can occur.
- p. 1990, l. 4: ...D is the molecular diffusion...
- p. 1990, Eq. (7): take out the space between u_i and c .
- p. 1990, l. 8: Separating c and $u_i = u + v + w$ into...
- p. 1991, l. 14: How about defining k_{dms} (preferably with an equation)? Also w_e needs to be defined.
- p. 1992, l. 4: You switch notation here; earlier \overline{v} is "mean" v , and here \boldsymbol{v} is "mean" v .
- p. 1992, l. 20: $\partial c / \partial t \simeq F_0/z$ is written incorrectly. The z is written as a subscript.
- p. 1993, l. 8: ...due to flux divergence and chemical loss.
- p. 1994, l. 11: How about a reference for the MOST expression for $\sigma_{c,t}$?
- p. 1994, l. 13: There seems to be a units problem with the expression for $\sigma_{c,n}$. It comes out as $pptv \text{ counts}^{-1/2}$. This carries through into subsequent equations.
- p. 1996, l. 17: instead of "motions in the vertical gradients," how about "vertical displacements in a vertically varying DMS field."
- p. 1997, l. 4: integral
- p. 1998, Eqn. (28): what is f_n ? Here you use f for frequency, and in the previous section you use ω and call it frequency when it is really angular velocity. That is,

C599

ω should be defined as $2\pi \times f$.

- p. 2000, l. 8: Figure 8 shows cross-spectra, not spectra. Also should be ...a biasing effect... on the next line. also corrections a few lines down.
- 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.
- p. 2001, l. 2: The quantity F_c/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.

—————end of review—————

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