

We thank both reviewers for their positive and constructive comments. We address their specific points below:

### **Reviewer 1: Barry Huebert**

*Lines 15-17, page 28459: The equilibrator time constant is never stated. Is the 3-4 min delay to the miniCIMS itself or (as stated) to the inlet of its equilibrator? From the experimental design I would think the former.*

The delay is to the inlet of the equilibrator. Our temperature sensor was just before the equilibrator and the delay was calculated using this data and data from the ship's thermistor, which was mounted at the entrance to the pumped seawater supply. We have previously estimated that our equilibrator/miniCIMS setup had a fast response (e-folding) time of 10 seconds (Royer et al., 2014).

*Page 28643, last 10 lines: You can't really see this point clearly in the last panel of Fig 2. Maybe put the red symbols in front? In some places it could be random scatter, just from eyeing the figure. Saying it's not is kind of absolute without some backing.*

We agree that plotting the COARE prediction over the SOAP data will help evidence the point made on Page 28463. We have made the suggested change.

### **Reviewer 2: Ian Brooks**

*P28456, line 2 – it would be useful to list the compounds for which gas transfer estimates have been made via eddy covariance here.*

Suggested change has been made.

*P28460, line 5 – the reader is referred to Bell et al. (2013) for details of the eddy covariance flux calculations. Following that reference it appears (from section 2.4, paragraph 5) that flow distortion over the ship is considered only as a source of uncertainty in the eddy covariance measurements (which is true), but no consideration is made for the effect of flow distortion on the mean wind. The mean flow over the ship is lifted and (depending on location) accelerated or decelerated relative to the free-stream flow at the same level. This means that the measured mean wind is (usually) biased, and hence U10 is biased. This can have a significant impact on the parameterization as a function of U10 and hence on the differences between estimates made from different ships. See for example Griessbaum et al, (2010).*

Suggested change has been made. This made relatively minor adjustments (< +/-10%) to the U10 data and all plots have been changed accordingly. There is no impact on the overall conclusions of the paper. A sentence has been added on Line 7 of P28460:

“Relative wind speed was adjusted according to the wind direction-dependent correction presented by Smith et al. (2011), which uses the computational fluid dynamics Gerris model (Popinet et al., 2004).”

*P28461, eqn 5 – the waterside only transfer velocity  $k_w$ , and subsequently  $k_{600}$  are derived using the measured total transfer velocity,  $K_{DMS}$  and the air-side transfer velocity,  $k_a$ , obtained from the NOAA COARE bulk flux algorithm. The results thus depend on the validity of the NOAA COARE algorithm; any bias in its  $K_a$  value will impact the value of  $k_w$ . Although  $K_w$  dominates the total transfer velocity, some discussion of the uncertainty associated with the reliance on NOAA COARE should be included. This is potentially relevant to the occasional non-random divergence of the COARE gas transfer velocity from the measured estimates noted at the bottom of p28463.*

This is a good point. Bias in  $k_a$  would indeed impact our  $k_w$  estimates. It is also inevitable that the airside resistance ( $r_a$ ) has a larger proportional contribution to total resistance ( $R_T$ ) when waterside resistance ( $r_w$ ) estimates are low. This would manifest as a greater uncertainty in  $k_w$  when our estimates of DMS gas transfer are large. As also noted by the reviewer, this impact is small because  $R_T$  is dominated by  $r_w$ .

The data in Figure R1 demonstrates that the contribution of  $r_a$  is not wind speed dependent. The scatter is evenly distributed around the COARE model prediction.

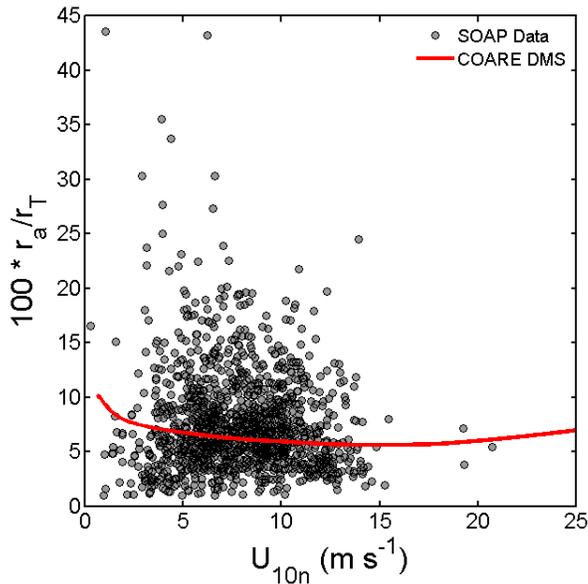


Figure R1: Percentage contribution of airside resistance ( $r_a$ ) to total resistance ( $R_T = 1/K_w$ ). Grey points =  $r_a$  (COARE estimate) /  $R_T$  (SOAP data). Red line = NOAA COARE estimates of  $r_a$  and  $R_T$ .

To give an idea of the potential bias introduced by using the NOAA COARE estimate of  $k_a$ , we have recalculated  $k_w$  using two other independent estimates of  $k_a$  (Mackay and Yeun, 1983; Duce et al., 1991). The gas transfer velocities in Figure R2 demonstrate that:

- 1) Scatter (uncertainty) due to  $k_a$  increases when high DMS gas transfer velocities are measured.
- 2) The trend in  $k_w$  is largely unaffected by the choice of  $k_a$  parameterisation.

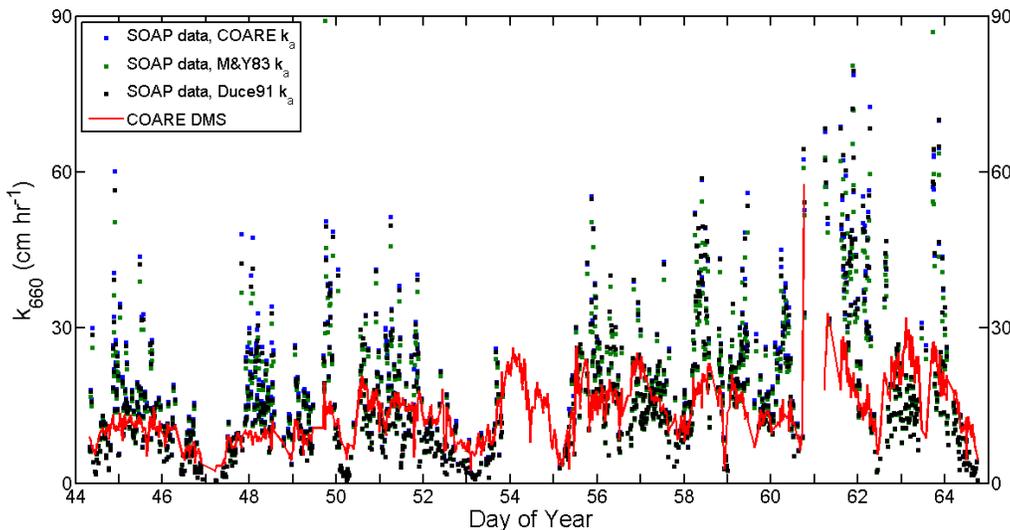


Figure R2: Time series of SOAP waterside gas transfer velocities ( $k_w$ ) normalised to Schmidt number = 660. Airside gas transfer velocity ( $k_a$ ) estimates were used to calculate  $k_w$  from measured  $K_w$  using three different models/parameterisations: COARE (Fairall et al., 2011), M&Y83 (Mackay and Yeun, 1983) and Duce91 (Duce et al., 1991). NOAA COARE model output (red line) shown for reference.

We have added Figures R1 and R2 to the Supplemental material and modified Lines 13-14 of P28461: “The relative influence of  $k_a$  upon our estimates of  $k_w$  was greater when measured  $K_{DMS}$  was high (Figure A, Supplemental material). This has little impact upon our data as the average (mean) difference between  $k_w$  and  $K_{DMS}$  was 7% and showed no wind speed dependence (Figure B, Supplemental material).”

P28464, line 3 – the line of best fit to the observed transfer velocities as a function of wind speed is discussed, but not shown on the relevant figure (fig 3a) where on the NOAA COARE function is shown. It would be useful to the reader to have the best fit show too.

Suggested change has been made.

P28465, line 7 – the authors state that for both this (SOAP) cruise and a previous cruise on the Knorr, the use of a geometric mean rather than arithmetic mean results in a ‘shallower slope’ of  $k_{600}$  with wind speed. I think this gives a slightly misleading impression. For the SOAP cruise one might make this interpretation, but for the Knorr cruise data all you can really say is that the geometric means are lower than the arithmetic means.

This is true. We have changed the sentence to now read:

“Geometric binned  $k_{600}$  data from both cruises are lower than the arithmetic binned data. The binned  $k_{600}$  SOAP data demonstrate a shallower slope using the geometric means.”

P28465, line 10-20. The reduced transfer velocity at high winds is convincing for the Knorr cruise, but rather less so for SOAP since it applies only to a single wind speed bin. In the absence of the Knorr data I would discount this point as an outlier – small data volume, and potentially suffering from the limitations inherent in evaluating data at the limit of the independent variable (over bulk of range a wind bin may include values from both increasing and decreasing winds, at the upper limit it can include values from increasing or constant wind only, by definition). Taking the Knorr data into account also, lends some support to this suppression being a real effect. The substantial difference in wind speed at which the suppression begins is then notable and worthy of discussion (or at least speculation).

We agree that the small amounts of data at high wind speeds are difficult to interpret. For this reason we included the following sentence in this section:

“In both cruises, there is limited data at wind speeds above  $10 \text{ m s}^{-1}$ , so this phenomenon should be viewed with caution.”

Line 19 – the authors suggest the suppression of transfer velocity might arise from a suppression of near-surface turbulence, but fail to specify whether that is in the water, air, or both.

We have made a change to the sentence to clarify that we are referring to near surface waterside turbulence.

P28467 – the discussion of the spatial mismatch between flux footprint and estimate of air-sea concentration difference is a really nice piece of work, and highlights the significant challenges of making such measurements. The comparison with the flux footprint model raises some questions because it fails to reproduce a spatial offset as large as that implied by the measurements. I suggest this might arise because of the slightly different physical properties being evaluated by each approach. The flux footprint model evaluates the fraction contribution to the total flux as a function of distance upstream – crucially it assumes spatially homogeneous conditions. The observational approach maximises the correlation between water-side DMS concentration and the  $U_{10}$ -normalised DMS flux\* – crucially the surface source is NOT spatially homogeneous. The location of the peak contribution to the flux is thus not (necessarily) the same as the peak in the footprint model. Where the DMS gradient increases with distance upwind, the maximum flux contribution would be expected to be further upstream than implied by the model. This is predominantly the case here, where the perturbations in DMS concentration and normalised flux used to evaluate the lag here are dominated by increase in DMS concentration upstream. It is difficult to assess confidently by eye, but my impression from Figure 7, is that the increase in flux precedes an increase in DMS concentration by more than the subsequent decrease in flux precedes the decrease in concentration. It would be interesting to partition the time series into portions that show an increase or decrease in DMS, and see if different lag intervals are produced when these are analysed separately.

This is an excellent point. We have added a paragraph to discuss this issue on P28468, Line 7:

“Despite the sensitivity of the model to the input parameters, none of these estimates are as large as the footprint derived from the lag calculation. Flux footprint models make the assumption that the surface source is spatially homogeneous. This was not true during the SOAP *B1* transect – the location of the peak contribution to the flux was not the same as the peak in the footprint model. Greater  $DMS_{sw}$  concentrations at the furthest extent of the

flux footprint will cause the flux signal to be dominated by a signal from further afield than implied by the footprint model. This is the likely explanation for the mismatch between our correlation analysis and the flux footprint model output.”

We have also added a sentence to the Conclusions, paragraph 3:

“The discrepancy between the flux footprint model output and our correlation analysis is probably because the model assumes spatial homogeneity in the  $DMS_{sw}$  concentrations within the flux footprint.”

The reviewer also suggests subdividing the SOAP *B1* transect to see if different lag intervals are produced. We attempted this but the number of data points becomes too few to draw any sensible (statistically-significant) conclusions.

*\*as an aside, surely you ought to maximise the correlation between the DMS gradient (water-air) not just water concentration, since this is what drives the flux...granted this is dominated by the water side concentration.*

We agree. We have re-analysed the lag correlation using  $\Delta C$  instead of  $DMS_{sw}$  and it did not affect the result. The text has been adjusted to reflect the fact that the correlation analysis used  $\Delta C$ .

#### *Minor issues*

*There are a few statements in the text to the effect that ‘figure N describes/plots’ etc... One could argue the ability of a bit of ink on paper (or pixels on screen) to actively do anything. Better to describe the data not the figure: ‘The gas transfer velocities are shown in figure 7’ rather than ‘Figure 7 also plots gas transfer velocities...’.*

Suggested change has been made.

*Also, it would be useful to label each panel (a), (b) etc to allow easy reference to ‘figure 7e’ rather than having to describe where in the plot the panel is.*

Suggested change has been made.

*Consider switching to an alternative colour map.*

Suggested change has been made.

#### **References:**

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