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Interactive comment on "Eddy flux measurements of sulfur dioxide deposition to the sea surface" *by* Jack G. Porter et al.

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This is a nice paper on a topic of broad relevance to air-sea gas transfer. The paper is generally well written. The introduction and method sections are complete and clear. I will make some general comments on the analysis and conclusions, followed by a list of minor edits and suggestions.

This paper deals with the deposition of a reactive gas, sulfur dioxide, to the ocean surface. Ionic equilibria and oxidation reactions in the aqueous phase (discussed in Sec 2.2) are sufficiently rapid that the equilibrium water-side concentration of SO2 is quite small, and thus we expect physical processes on the air-side will determine the deposition rate constant. But, as mentioned at the end of 2.2.1, we expect surface effects

(mass accommodation) may also be important, and this additional resistance to mass transfer is distinct from either turbulent diffusion in the bulk air or molecular diffusion in the surface microlayer. The physical diffusive mechanisms are fairly well understood. The surface effects are not. The magnitude of this barrier probably depends on surface microlayer characteristics, and for the ocean it could be significantly different from the resistance at the surface of small droplets implied by the Worsnop et al 1989 laboratory data.

Given the excellent introduction I was a bit disappointed that the discussion and conclusions did not try to address the surface resistance, but rather seem to lump it into a diffusion factor which doesn't lead to significant insights.

For gases like SO2, I think the airside resistances are better represented by

r_total = r_turbulence + r_diffusion + r_surface

It seems to me that you should be able to estimate r_surface for SO2 as the difference between r_total for SO2 and r_total for water vapor, since there is no surface resistance effect for water. You can also compare r_total for SO2 with r_a from COAREG, which is a stability-corrected estimate of the turbulent and molecular diffusion resistances but does not consider a surface resistance.

This sort of analysis could lead to improvements in the COAREG parameterization through the introduction of an additional species-dependent constant in the formulation of r_a. Even if the data available from this study are insufficient for a statistically significant determination of the surface resistance term, the exercise provides useful guidance for future investigations.

Other more minor suggestions and comments:

P2 Eq. 1: You could point out that the delta C term defined this way means a downward flux is positive, which is typical in literature dealing with deposition to the surface, but opposite the general convention for gas transfer where the upward flux is positive.

P2 Line 20: define solubility as Cw_o/Ca_o to emphasize this is the equilibrium concentration ratio? Personally, I would avoid the term ‘Ostwald coefficient’ since this is subject to several slightly different and potentially confusing definitions (see R. Battino, Fluid Phase Equilibria, 15, 231-240, 1984) and just call it the dimensionless (liq/gas) solubility.

P4 line 11: ‘This is sufficiently small…’ ?

Sec 3.2: Were blank measurements conducted and if so, how? In previous work with this method we have used a coil of HCl-washed copper tube to remove SO2 from the sample stream and determine the background signals at m/z 112 and m/z 114. Admittedly, this is not a perfect blank, because removing one reactant from the air sample perturbs the ion-molecule equilibria in the source, such that the background signal you measure in the absence of SO2 may not be exactly the same as the background when SO2 is present. If the concentration of SO2 (ambient + internal standard) is small compared to the CO2 and ozone concentrations, however, this consideration should be minor.

P7, eq 9: F is being used for both flux and flow which is confusing. Choose another variable in this equation to represent gas flow…

P7, line 30: The correct references for the COARE model are Fairall et al. 1996, Fairall et al. 2003 and Edson et al. 2013. Fairall et al. 2000 deals specifically with gas transfer.

P8 line eq 12: You might mention that, Fmom is more commonly called the Reynolds stress (tau).

Sec 5: See comments above. Also, you could mention that a linear wind speed dependence is expected for very soluble gases and has been demonstrated in other studies (i.e. little or no bubble enhancement to k from breaking waves). In comparisons with the physical model I would just use k_a from COAREG and ignore k_b which should

not be important and in any case is the more uncertain parameter.

P11 line 25: The other reference for updates to COAREG is Fairall et al., 2011.

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