Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-495-RC1, 2018

Atmospheric Chemistry and Physics Discussions

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

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Received and published: 3 July 2018

Review for "Eddy flux measurements of sulfur dioxide deposition to the sea surface"

This is a nice paper describing direct measurements of air-sea SO2 transfer, which were made at a coastal location. This is a highly challenging measurement, as evidenced by the very few previous data for comparison. The measurements reported here appear to be sound and the data processing/filtering procedure reasonable (though slightly incomplete, see detailed comments). The fact that the results are largely consistent with existent theory doesn't reduce the value of this work. As the authors mentioned, these measurements were made over a very limited range of environmental conditions. Further observations over a much wider range of conditions

have the potential to not only improve our understanding of air-sea SO2 transfer, but also the transfer of many other highly soluble gases as well as heat. In my opinion the paper can be published after some technical corrections.

Specific comments: P1, line 18. Please give references and indicate ranges. And note that anthropogenic SO2 emissions are changing globally, with for example decreasing trends in Western Europe and North America.

P2. Line 1-3. Please give references. Yang et al (PNAS 2013, ACP 2014, GTWS 2016) measured the air-sea transfer of methanol and compared its rate to those of momentum and sensible heat. A very similar method of analysis is used here.

P. 2 Line 6-7. The previous sentence just said that the Faloona et al's measurements were in the MBL. Also, it should be 2010, not 2009.

P. 5. Line 12. What's the tidal range at this site? If significant, it'd alter the measurement height above the mean sea level and so the extent of the flux footprint.

P. 7 line 12. How is the SO2 blank measurement (what I assume the authors meant by 'system blanks') made? Since Vd = -flux/[C], any error in mean [C] due to uncertain blank correction will propagate to Vd. To test, the authors can plot Vd vs. u* and color-code it by [C]. Is there any pattern?

P. 7 line 20. What's the typical tilt angle, i.e. the angle between the horizontal and the streamline?

P. 7, line 30. The authors show later that the mean Cd measured at this location is significantly greater than what the COARE model predicts. If so, U10 computed from the COARE model (assuming open ocean Cd) will be in error. In theory, to get a more accurate U10 it's probably better to use an iterative approach to estimate U10 from the measured Cd. Though the difference might not be very big in this case since the measurement height is already at ∼10 m above sea level.

p. 7 line 32. Last sentence: 'sensible heat' is left out

p. 8. Equation 11. The Licor7500 measures mass concentrations rather than the mixing ratios. Thus a 'Webb' correction for air density fluctuation is required for water vapor flux (probably not a big correction). Has this been applied?

p. 8 Equation 13. Is T here the sonic temperature or the air temperature? The sonic temperature, approximately equal to virtual temperature, is affected by humidity. Thus one needs to apply a latent heat correction to the sonic heat flux to get sensible heat flux. This probably isn't a big correction, and can be achieved by:

a) apply a high frequency (e.g. >=5 Hz, if available) humidity correction to the raw sonic temperature data, or b) use the actual latent heat flux (or bulk latent heat flux) to correct the sonic heat flux

P. 8, Equation 17. Typically a lapse rate correction is applied to the measured air temperature in the calculation of deltaT. What's the height of the mean air temperature sensor?

More generally, airside transfer is dependent on atmospheric stability. It is typical to convert the airside transfer velocities to neutral transfer velocities. This doesn't affect the authors pair-wise comparisons (e.g. kSO2 vs kSH), but does affect the kSO2 vs u* relationship, for example.

P. 9, line 1. How did the authors arrive at a cut off frequency of 1.5 Hz? What is the instrument response time?

p. 9, line 16. Have the authors estimated the flux detection limit for their SO2 system? SO2 over the open ocean (especially Southern Hemisphere) is only typically a few tens of ppt.

p. 9, line 18. Reasoning and reference for excluding data with Z/L > 0.07?

P. 9. Line 19. What were the thresholds used for excluding SO2 ship spikes?

p. 10, line 25. There is very large variability in the kmom vs. U10 relationship here.

Typically over the open ocean, the relative standard deviation of u* decreases with increasing wind speed. Could some of the variability here be due to tidal height changes or the wind direction being on the edge of the acceptable sea-sector? Also, see my comment about U10 earlier.

P. 11 line 13. At the beginning of this section, I think the authors should first compare their kSO2 vs. u* relationship to a) the only previous measurements of kSO2 (Faloona et al. J. Atmos Chem 2010), and b) kmethanol from Yang et al. GTWS 2016.

p. 11, line 18. The authors' assertion that kSO2 is more precise than kSH and kH2O appears to be backed up by their Fig. 4. With increasing wind speed, the scatter in kSO2 only increases marginally (the relative standard deviation probably decreases). In contrast, the scatter in kSH and kH2O increase substantially with wind speed.

p. 11, line 25. Technically the name COAREG started with Fairall et al. 2011 JGR (the gas transfer version), not Fairall et al. 2000.

p. 11, line 28-30. It looks like the author substituted computed Cd with the mean Cd v. U10 relationship from the measurements? Would the authors be able to explain more of the variability in the other k data if they prescribe the model with the measured Cd on a point-by-point basis?

p. 12, line 16. The authors should specify that these are airside Schmidt numbers, which are largely temperature independent. Also, how does the Sc_SO2 here compare with more contemporary predictions (e.g. from Johnson 2000 Ocean Science)?

P. 12 line 19-20. Quoted uncertainties here for the Schmidt number exponents are very large. Are they derived from a regression of rdiffH2O vs. rdiffSO2, etc? There seems to be a lot of variability in the kmom data, which isn't as apparent in the kSO2 data. Is subtracting such a noisy rturb (from kmom) the cause for the poor regression results?

p. 12 line 22. More appropriate references than Jaehne et al. 1987 are specific studies of airside transfer, including Hicks et al. 1986 as well as the earlier Liss et al. papers.

p. 23, Fig 4. Show units for slope. Also, typo in caption: ‘computed’ instead of ‘computer’

Finally, I personally find ‘higher/lower’ to be more suitable adjectives for transfer velocity than ‘bigger/smaller’.

The authors can contact me directly for further questions if they wish.

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