Atmos. Chem. Phys. Discuss., 8, S7875–S7882, 2008
www.atmos-chem-phys-discuss.net/8/S7875/2008/

Atmospheric
Chemistry
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Discussions

Interactive comment on “The effects of experimental uncertainty in parameterizing air-sea gas exchange using tracer experiment data” *by* W. E. Asher

W. E. Asher

Received and published: 3 October 2008

Responses to Comments from Rik Wanninkhof on ACPD-2008-0388

Review: *The effects of environmental variability and experimental uncertainty in parameterizing air-sea gas exchange by William Asher Asher provides a thorough analysis of the effect of experimental error on calculated gas transfer velocities and the resulting effect on gas exchange wind speed relationships. The main conclusion is that much of the observational differences in field and windwave tank studies can be explained from these uncertainties rather than the common attribution that other effects are impacting gas transfer. [As a sidebar- this does no mean that other effects are not pertinent but rather Asher suggests that much of the scatter can be explained by experimental*

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uncertainty]

The paper discusses uncertainty for the Schmidt number exponent based on wind-wave tank results using He, SF_6, and CH_4; and the uncertainty in the gas transfer velocity in this wind wave study and a compilation dual deliberate tracer field studies. The analyses is performed by a Monte-Carlo simulation spanning a 2 sigma uncertainty based on uncertainty in the measurement of concentration, and in the case of field studies uncertainty in mixed layer depth as well. The results are shown graphically. The paper is well laid out and overall quite clear. The main issue is that it does not really provide a clear sense if the situation can be improved or if we are stuck with this envelope of uncertainty. In particular, there should be some discussion on the effect of integration time, better estimates of mixed layer, and more precise concentration measurements. Also, while the study assumes steady winds, the effect of variability of wind over the measurement time of the field studies should be, at least, qualitatively assessed.

Response: Rik has raised many good points in his review and I thank him for his comments. I have modified the conclusions to include a discussion of the broader implications of this work. In terms of laboratory studies, the main point is that because measurement times are generally short, on order of several hours, uncertainties in concentrations are the most important source of variability over which an investigator has control. Therefore, particular care should be made to reducing these to as low as possible. In contrast, because integrations times are relatively long in PDTM measurements the main source of variability is the mixed layer depth. As discussed in detail below, the effect of wind speed variability on the observed scatter in measured transfer velocities is beyond the scope of this paper.

Specific comments:

Abstract: "could explain a major portion"- be more quantitative

Response: I have estimated the fraction quantitatively by first estimating the variance in the experimental data set. In the case of the wind tunnel data, for He and SF_6 I fit each data set to a straight line. The least-squares regression provides standard error of the fit. This term represents the average error of the experimental data. Then, I perform a linear regression on the simulated data and get the standard error for the simulated data. The standard error of the simulated data is somewhat less than the standard error of the simulated data, and the ratio is a very rough estimate of the fraction of the uncertainty in the experimental data explained by the experimental uncertainties. The dual-tracer data is treated in a similar fashion, except I fit the field data and the simulated data to a U2 dependence. So estimated this way, measurement uncertainty could explain roughly half of the observed variability in both the dual-tracer data and the wind-wave tunnel data. I have modified the abstract and discussion sections to describe this procedure and the results.

Introduction: First couple of comments should be referenced

Response: Agreed

Equation 2: I could not readily find how C_s *was defined.*

Response: I have added a sentence following Equation 1 explaining that the product $K_H P_A$ defines the saturation concentration, C_S.

Equation 3: provide an estimate of the magnitude of the denominator: (1-(ScHe/ScSF$_6$*)*$^{1/2}$*)*

Response: Done.

Page 5. (Or before): State somewhere what the uncertainty in the Schmidt numbers and diffusion coefficients is.

Response: The uncertainties for the diffusivities of SF_6 and He have been provided.

Page 5, bottom: Explain why despite the larger uncertainty in He concentration measurements the uncertainty in k is the same (or even slightly less) than the measurement *of k by SF_6*

Response: This occurs because the change in concentration for He is greater than the change in concentration of SF_6. Calculation of k involves taking the logarithm of ratios, and if the absolute change in the ratio is larger, the relative effect of the error goes down. In a theoretical sense this would be irrelevant since one could just let the experiment measuring SF_6 run for longer than the one for He so that the absolute change in concentration would be the same. However, I performed these calculations trying to simulate the effect that measurement error would have on the experiments as they were performed, where the experiment time is determined by how long one is willing to measure concentrations. This time is typically a few hours, which means the change in SF_6 concentration is significantly less than the change in He concentration, and the relative uncertainty in the ratio of the logarithms is larger for SF_6. If money and time were not considerations in conducting gas transfer measurements then the error for He would be greater than the error for SF_6. Realistically though, when conducting experiments at a wind-wave tunnel like the NASA/WFF-ASIRF, there is a limited time for conducting experiments, which leads to effects such as shown. I have explained this in a more succinct fashion on Page 7 of the revised manuscript.

Page 7, bottom: perhaps emphasize that with stated uncertainties n cannot be measured to better than 0.1 (?).

Response: Agreed. We have added a sentence to this effect.

Equation 8: Why should n decrease monotonically with wind speed > 2 m/s?

Response: We have added the following paragraph in explanation:

The choice of functional form for Eq. (8) was selected because it approximates the measured dependence of n as a function of U [Jähne et al., 1984; Zappa et al., 2004]. Theoretical justification for assuming n will decrease as U increases can be made through heuristic appeals to eddy structure models for air-water gas transfer [Harriott, 1962]. In these models, as the penetration depths and timescales of turbulence eddies at the water surface decrease, n decreases from a maximum of 1 to a lower limit of 189; (see Figure 3 of Harriott [1962]). Because it is known that penetration depth and timescales decrease with increasing levels of turbulence [Asher and Pankow, 1991a; b] and it is known that turbulence increases with increasing wind speed [Siddiqui et al., 2004], it is reasonable to assume that n will decrease monotonically to the lower limit of 1/2 as U increases.

Equation 8: appears to have a -; missing in the exponential function

Response: Correct. There is a typographical error in Equation 8. It will be corrected.

Page 8, middle: The field measurements were not done under steady winds. This uncertainty should be estimated.

Response: This is a rather complicated request and would require development of a much more sophisticated modeling scheme than was undertaken. My opinion is that this effort would greatly expand the paper significantly in order to treat it thoroughly. Because it is an important point, it should not be added as an afterthought. Rather than expand the paper to include the effect of variable wind speeds on dual-tracer measurements, I have added the following sentences to the end of the first paragraph of Section 4:

Rather than address the effect of averaging wind speed, which has been discussed in some detail elsewhere [Wanninkhof et al., 2004], here the effect of measurement variability under steady winds will be studied. This will overestimate the importance of measurement errors in the resulting variance of kL, but the best way to estimate the effects of wind speed variability on dual-tracer experiments is not clear and the subject of ongoing work.

As a point for further discussion, perhaps we (meaning Rik and I) could look into how one would do a similar Monte Carlo simulation for dual-tracer data where the instanta-

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neous wind speed used to model the decrease in transfer velocities was a function of time. One potential approach would be to generate a time series of wind speeds that would be used to calculate a time series of transfer velocities for each trial in the Monte Carlo simulation. These time series would then be used to numerically integrate the underlying differential equations for the decrease in gas concentrations. The measurement uncertainty would be added to these concentrations that accounted for variable wind speeds before they were used in Equation 3. Although what I have outlined above is feasible, it is not a trivial task and its exposition would make the present paper much longer. Perhaps Rik and I could collaborate on performing this study and publish the results separately?

Page 9, line 3: The line in Figure 6 does not look like a power law dependence.

Response: That is an optical illusion of a sort. Because transfer velocities for the dual-tracer simulations were calculated at only four wind speeds, what is plotted in Figure six is the four discrete data points with straight line segments connecting them. This makes it appear the data is not from a power-law relationship. However, if you take these four data points and fit them to a power relationship using a least-squares algorithm, the coefficient m for the U^{m} relationship predicted by the fit comes out to be 1.99, or essentially 2 as in Wanninkhof (1992). Rather than run more simulations using more wind speeds so that the line segments in Figure 6 are less apparent, the figure in a revised manuscript will plot the fitting power-law curve showing the underlying power law relationship as opposed to the discrete average values.

Page 9, line 6: Comment on time interval of 1 to 2 days. The time interval is critical for the concentration decrease and therefore the uncertainty

Response: Typically, the minimum time required for a significant decrease in the concentration ratio for oceanic dual-tracer measurements has been observed to be on order of one day. This has been mentioned in the text.

Page 10: While the conclusion that there is less variability in forcing mechanisms is

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an important one, some words if field and lab studies can be improved to get to these forcing functions and/or more accurate wind speed relationships would be of interest.

Response: This has been added to the conclusions. The implications of this work towards laboratory studies are that measurement error in concentrations should be minimized and that longer experimental times are to be preferred over shorter times. Application of this latter principle might be that in situations where the total time available to conduct experiments at a particular wind-wave tunnel is on the order of a few weeks and a limited number of experiments can be conducted, it might be preferable to conduct fewer experiments running but run each experiment for many hours to maximize the concentration decrease. Additionally, particular attention should be paid to reduction of measurement variability in the laboratory measurements of gas concentrations, although specific recommendations for this depend on the method used for the gas in question. Oceanic dual-tracer experiments are more problematic since main the source of variability resides in determination of the mixed-layer depth, and how it changes over the course of the DTM measurement period. It is possible that this will present an additional source of error, much as the difference between steady and variable wind speeds do. I have mentioned this in the revised discussion, but putting numbers on such an effect is beyond the scope of this paper although could be included in modeling designed to explore the effect of variable wind speeds.

Figure 1. I vaguely recall there were 4 data points for IronEx; I only see 3 plotted.

Response: Two of the IronEx data points are essentially identical, U=7.0 m s^{-1} kL=11.1 cm hr^{-1} and U=7.1 m s^{-1} kL=11.4 cm hr^{-1}. I omitted plotting one of the two. The revised manuscript now contains all four IronEx points.

Figure 3. (And 2) Comment why more SF_6 points fall outside the error envelope than for He.

Response: The variability is larger for SF_6 than for He because of the difference in the change in absolute concentrations of the two gases. This point was explained above.

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Figure 4. It is unclear why the 2-sigma error envelope broadens at low winds.

Response: The error envelope broadens for small U because the measurement error in kL increases at low winds. The measurement error increases because of the effect that measurement time has, where relatively smaller changes in concentrations amplify the effect of measurement error due to the presence of the logarithm term in Equation 2. As explained above, these simulations were conducted to mimic how data were actually collected in a wind-wave tunnel, where experiment times were restricted to at most 5-6 hours duration. For lower wind speeds, this means the change in concentration is smaller so the variance in the calculated kL increases. In an ideal world where one could run experiments at low wind speeds for much longer periods, the error envelope would be independent of wind speed.

Figure 5a: Why does error envelope broaden at low winds for CH_4/SF_6 *but not as much (for figures 5b and 5c.*

Response: The width of the error envelope is determined mainly by the ratio of the two Sc values for the gas pair. CH_4/SF_6 have the lowest ratio of the three pairs of gases so the effect of measurement error increases. This point has been explained in more detail in the text discussion Figure 5.

Figure 6. It might be worth commenting that the wind speed (as the concentration decreases faster).

Response: This point is mentioned in the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 16693, 2008.

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