

Interactive comment on “A numerical scheme to calculate temperature and salinity dependent air-water transfer velocities for any gas” by M. T. Johnson

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General Comments: In my view, there are two main components to this manuscript. Firstly, a model (and within, sub-models) of air-water gas transfer is selected. Secondly, this model is described in detail and an implementation scheme presented and made publically available. I am personally grateful for the second part and I think the community in general will share that view. However, the value of the second part does to a large extent depend on the choices in the first part. There is considerable room for improvement in discussing the background literature in general and particularly in justifying the selection. We do not all have to agree each selection (and that is not

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going to happen anytime soon), but as part of this review process I'd like to reach a point where the most relevant evidence is presented clearly, concisely and fairly. I think you have tried to do this and largely succeeded, but I do have some specific criticisms. The sub-model for liquid phase transfer velocity, k_w , is the part I am most familiar with. The “Nightingale et al” model would not be my first choice, but that is not my criticism, I am more concerned that there is a more rounded discussion of “ k_w models”. In the case of “ k_a ”, I am concerned that some of the most relevant information has not been used in the selection of the k_a sub-model. I have to admit considerably less familiarity with “ k_a ”, but even with my limited knowledge it is apparent that there are major omissions, particularly field evidence on water vapour transfer (a peculiar, but very important and well-measured soluble gas) and the formulations proposed within the NOAA COARE model (Fairall et al., 2003). The latter point raises another criticism: The NOAA COARE model is already available as a general tool applicable to gases of all solubilities and should reasonably be introduced at a much earlier stage. At present it is only mentioned with respect to GASEX-98 CO₂ data, which it can only be fitted to with unusual (I am tempted to say “unrealistic”, but that may be premature with the paucity of evidence at present) tuning constants. Since NOAA COARE can be fitted to other CO₂ and DMS results with more consistent tuning constants, the presentation of GASEX-98 alone gives a rather unrepresentative view of NOAA COARE. Note also that DMS falls into the class of gases where liquid phase transfer is more important but the gas phase transfer is significant; thus, some inclusion of recently published field studies of DMS transfer would be appropriate. I do accept the point at line 2 of page 270 that the scheme presented here is simpler than “NOAA COARE” and “MESSY AIR-SEA” but it would be better if you acknowledged their existence specifically in the Introduction, rather than only referring to “More detailed approaches [p.254, l 7]”. Elements of the numerical calculation, e.g choice of k_a and k_w models, are currently a matter of interpretation and the favoured sub-models may alter in the future. Thus, I'd suggest a “modular approach” to the code, allowing substitutions to be made easily. I haven't investigated the R code, so apologies if this has already been addressed.

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Specific Comments It may be pedantic, but I do not recognize the presented model as a “thin film model”. Indeed, the assumed Schmidt number (or molecular diffusivity) dependences of both the k_a and k_w sub-models are directly contradictory to a “thin film model” as usually defined. If transfer in each phase is controlled by molecular diffusion across a thin film, then necessarily k_a is proportional to $S_{c_a}^{-1}$ and k_w is proportional to $S_{c_w}^{-1}$. What is presented here is a more generic “two phase model”. It is certainly reasonable to refer to Liss and Slater for the model of serial transfer across layers in the gas and liquid phases, but avoid using the term “thin film model”. Similarly, Figure 1 is misleading in implying linear variation in gas concentration either side of the interface. I’d suggest borrowing the “resistance model” drawing of transfer from one of Peter Liss’s later papers (incidentally this might be useful in explaining “NOAA COARE + bubbles” where a parallel pathway (and resistance) in the liquid phase is proposed). I am not qualified to comment in detail on Section 2.1, but for what it is worth I found it interesting and convincing. Perhaps it should be clearer that this is a “fall back”, where the solubility of the gas in seawater has not been directly measured? I found Section 2.2 interesting and informative. It is clearly a matter of concern that a range in values from different parameterisations of greater than 4. What I think is missing from this section is a critical review of the experimental data that underlies the parameterization. As far as I am aware the best field experimental data available for the parameterization of k_a is measurements of the air-sea transfer of moisture (see Fairall et al. [2003] *J. Climate*, 16, 571-591, for a review). It is also important to use sensible drag coefficients and this is dealt with adequately, but a greater issue is the actual data on transfer of volatiles. I have some misgivings myself about applying results for water vapour to all other gases (there might be different behaviour), but I am not convinced that there is adequate experimental data on other soluble gases to pursue any other path. As far as I understand, water vapour measurements underpin the “NOAA COARE” parameterization of k_a and this is significantly different to the parameterizations presented in this manuscript. It is significant that Fairall et al. [2003] conclude that the bulk parameter “ C_e ” for moisture increases slightly but significantly

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with wind speed (above a minimum at low wind speeds), which would imply a weak non-linearity in k_a . I note that unlike “ K_H ” and “ k_a ” there is not a new derivation for “ k_w ” but I am concerned that k_w is treated robustly. I have read much worse summaries of k_w parameterizations than the beginning of Section 3.3, but I am compelled to quibble. There is a danger that Sweeney et al (2007) will be regularly misinterpreted and I would rather that did not start here. The bomb 14 C data considered by Sweeney et al.(2007) provides no evidence that gas transfer velocities follow a square law dependence on wind speed. Sweeney et al. do derive a very useful estimate of the global average exchange coefficient of CO₂ and chose to assume square-law dependence. It is clear that Nightingale et al (2000; “N”) is consistent with the global coefficient. Also, N. is clearly a fairly good fit to both the dual-tracer data that they originally considered and to data from subsequent dual tracer studies. Thus, I think there is a reasonable case for choosing this model. However, this model does not fit all data and particularly it is rarely consistent to micrometeorological estimates of the transfer of CO₂ and DMS. For completeness, these studies and the apparent discrepancies should be mentioned. Also, N is an empirical model and the absence of any substantial theoretical underpinning should be mentioned. The main practical importance of “the weakness of theory” is that it is not clear that N (both the wind-speed dependence and the assumed dependence on Schmidt number) can be applied to all gases. At least one interpretation is that it is not reasonable to apply it to gases much more soluble than helium; and that is backed up by the experimental evidence for DMS (see e.g., B. J. Huebert, B. W. Blomquist, M. X. Yang, S. D. Archer, P. D. Nightingale, M. J. Yelland, J. Stephens, R. W. Pascal, and B. I. Moat. 2010. *Geophys. Res. Lett.*, 37, 1, doi:10.1029/2009GL041203, 2010). Personally, I wouldn’t make a virtue of N “[sitting] roughly in the middle” –unless you consider a herd instinct beneficial – though clearly N is more acceptable for being far from extreme. At low wind speeds, rain or free convection may be important and the model may underestimate k_w . I do not see mention of whether any of the selected sub-models are inappropriate for some reactive gases (e.g k_w of SO₂, or of CO₂ at very low wind speeds). I think this should be added. As noted in the general

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comments, I am concerned that taking GASEX-98 and the tuning of NOAA COARE to that single dataset gives a rather unrepresentative view of both data and theory. I suggest looking at a few of the most recent publications of micrometeorological data (e.g. Miller et al. [2009] GEOPHYSICAL RESEARCH LETTERS, VOL. 36, L15816, doi:10.1029/2009GL038907; Huebert et al. [2010], GEOPHYSICAL RESEARCH LETTERS, VOL. 37, L01605, doi:10.1029/2009GL041203) and a recent theoretical and modelling development of NOAA COARE (Jeffery et al. [2010]; Ocean Modelling, 31(1-2), 28-35. doi: 10.1016/j.ocemod.2009.09.001) and working backwards. I am not suggesting a major revision here, but you might end up with a different "Figure 6" that gives a more rounded view of both data and theory. Technical and typographical: It would be more consistent to use small letter "a" consistently for gas phase, and "w" for liquid phase. E.g. Why capital A in the Schmidt number for air? Typo "gaess" at p.262, line 14 Is equation A1 correct? I ask because there are two linear coefficients in "S", "A" and "C"; perhaps there is a missing symbol? Typo "alond" at p.271, line 7

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