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Atmospheric
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Interactive comment on "New constraints on terrestrial and oceanic sources of atmospheric methanol" *by* D. B. Millet et al.

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This comment is concerned with a specific part of the paper of Millet et al (2008) and we do not wish to comment on the rest of the paper which appears very interesting and of significant value to the field. We are concerned by the analysis of the ocean-atmosphere exchange of methanol in the manuscript, in terms of the formulation used to calculate the transfer (piston) velocity for methanol in the model.

The authors state that their model uses the gas transfer velocity paramterisation of Nightingale et al (2000):

$k(_w) = (0.222*(U_{10})^2 + 0.333*U_{10}) * (600/Sc)^{0.5}$

where U_{10} is the windspeed (at 10m above the sea surface) and Sc is the Schmidt

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number of the gas of interest. We reproduce equation 1 from Nightingale et al (2000) directly, as it is not presented in Millet et al (2008). The transfer velocity measured by Nightingale et al (2000) is effectively a water phase transfer velocity (k_w) and is thus applicable only to those gases whose transfer across the ocean-atmosphere interface lies under water phase control i.e. those whose solubilities are low and therefore the resitance to their diffusive transfer at the interface is greatest in the water phase (Liss and Slater 1974). This is almost certainly not the case for methanol. For a compound as soluble as methanol (some 3 times more soluble than ammonia) the resistance to transfer in the water phase is likely to be tiny (due to solubility enhancement of the diffusive transfer, rendering Equation 1 innapropriate) and so the resistance to transfer on the atmospheric side of the interface is likely to dominate. The gas phase transfer velocity (k_g) can be calculated according to Duce et al (1991):

k_g = U_{10}/(770 + 45*r.m.m.$^{1/3}$)

where r.m.m. is the relative molecular mass of the gas of interest (32 in the case of methanol).

k_g calculated according to Equation 2 is 2784 cm/hr at a wind speed of 6 m/s and 5587 cm/hr at 12 m/s, compared with the inappropriately used k_w, calculated from Equation 1 to be 9.4 and 34 cm/hr for wind speeds of 6 and 12 m/s respectively. Here we have used a Schmidt number for methanol of 684, calculated according to Pozzer et al (2006). Therefore, it is possible that the authors have underestimated their ocean-atmosphere exchange fluxes by two or three orders of magnitude. Whether this is the case depends greatly on the degree of disequilibrium between atmosphere and ocean relative to the timescales of other production and loss terms in the atmosphere and ocean. For example, if in the author's model system the timestep is such that ocean and atmosphere will reach equilibrium within each timestep, then increasing the piston velocity will make little or no difference to their results. We would be interested hear the authors' thoughts on this matter and would be pleased to provide more details to aid their calculations if required.

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