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Closing the dimethyl sulfide budget in the tropical marine boundary layer during the Pacific Atmospheric Sulfur Experiment

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Abstract. Fourteen research flights were conducted with the National Center for Atmospheric Research (NCAR) C-130 near Christmas Island (2° N, 157° W) during the summer of 2007 as part of the Pacific Atmospheric Sulfur Experiment (PASE). In order to tightly constrain the scalar budget of DMS, vertical eddy fluxes were measured at various levels in the marine boundary layer (MBL) from ~30 m to the top of the mixed layer (~500 m) providing improved accuracy of the flux divergence calculation in the DMS budget. The observed mean mole fraction of DMS in the MBL exhibited the well-known diurnal cycle, ranging from 50–95 pptv in the daytime to 90–110 pptv at night. Contributions from horizontal advection are included using a multivariate regression of all DMS flight data within the MBL to estimate the mean gradients and trends. With this technique we can use the residual term in the DMS budget as an estimate of overall photochemical oxidation. Error analysis of the various terms in the DMS budget indicate that chemical losses acting on time scales of up to 110 h can be inferred with this technique. On average, photochemistry accounted for ~7.4 ppt hr $^{-1}$ loss rate for the seven daytime flights, with an estimated error of 0.6 ppt hr^{-1}. The loss rate due to expected OH oxidation is sufficient to explain the net DMS destruction without invoking the action of additional oxidants (e.g., reactive halogens.) The observed ocean flux of DMS averaged 3.1 (±1.5) μmol m^{-2} d^{-1}, and generally decreased throughout the sunlit hours. Over the entire mission, the horizontal advection contribution to the overall budget was merely -0.1 ppt hr^{-1}, indicating a mean atmospheric DMS gradient nearly perpendicular to the east-southeasterly trade winds and the chlorophyll gradient in the equatorial upwelling ocean. Nonetheless, horizontal advection was a significant term in the budget of any given flight, ranging from −1.2 to 2.5 ppt hr^{-1} , indicating a patchy and variable surface seawater DMS distribution, and thus needs to be accounted for in budget studies.

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