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Coupling between DMS emissions and the ocean-atmosphere exchange of NH_3

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A strong correlation between aerosol ammonium and non-sea salt sulfate is commonly observed in the remote marine boundary layer. It has been suggested that this relationship implies a biogeochemical linkage between the nitrogen (N) and sulfur (S) cycles at the cellular biochemical level in phytoplankton in the ocean, or a linkage in the atmosphere (see P. S. Liss and J. N. Galloway, *Interactions of C, N, P and S biogeochemical cycles and global change* (Springer, 1993), and P. K. Quinn et al. in *J. Geophys. Res. – Atmos.* 1990, 95). We argue that an oceanic linkage is unlikely and draw on mechanistic and observational evidence to make the argument that the atmospheric connection is based on simple physical chemistry. Drawing on an established analogous concept in terrestrial trace gas biogeochemistry,we propose that any emission of dimethylsulfide (DMS) from the ocean will indirectly influence the flux of NH_3 from the ocean, through the neutralisation of acidic DMS oxidation products and consequent lowering of the partial pressure of NH_3 in the atmosphere. We present a simple numerical model to investigate this hypothesised phenomenon, using a parameterisation of the rate and thermodynamics of gas-to-particle conversion of NH_X and explicitly modelled ocean–atmosphere NH_3 exchange. The model indicates that emission of acidic sulfur to the atmosphere (e.g. as a product of DMS oxidation) may enhance the marine emission of NH_3. It also suggests that the ratio of ammonium to non-sea salt sulfate in the aerosol phase is strongly dependent on seawater pH, temperature and wind speed – factors that control the ocean–atmosphere ammonia flux. Therefore, it is not necessary to invoke a stoichiometric link between production rates of DMS and ammonia in the ocean to explain a given ammonium to non-sea salt sulfate ratio in the aerosol. We speculate that this mechanism, which can provide a continuous resupply of ammonia to the atmosphere, may be involved in a series of biogeochemical-climate feedbacks.