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Thermodynamic predictions of aerosol pH in the summertime Southern Ocean marine boundary layer using high-resolution aerosol- and gas-phase observations from Cape Town to Antarctica

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Aerosol acidity is an important parameter that affects gas-particle reaction rates, heterogeneous chemistry, human and ecosystem health, global biogeochemical cycles, and climate. Aerosol acidity is difficult to measure directly and remains poorly constrained in most of the troposphere. There is a further scarcity of measurements and/or proxy-estimates of aerosol acidity in the remote marine atmosphere, a region where aerosol acidity exerts strong control on the solubility, bioavailability, and toxicity of biogeochemically-relevant species that influence the productivity of the surface ocean. Here, we measure gas-phase ammonia, and aerosol phase ($PM_{2.5}$) ammonium, sulfate, nitrate, sodium, and chloride in the summertime Southern Ocean marine boundary layer every two hours across a latitudinal gradient from Cape Town (-34.11 °S, 18.03 °E) to Antarctica (-58 °S, -0.06 °W). A thermodynamic equilibrium model, i.e., ISORROPIA-II, was run in "forward" mode to calculate aerosol pH using the measured gas + aerosol concentrations, atmospheric temperature, and relative humidity as inputs. The model was able to accurately predict the observed concentration of gas-phase ammonia across the entire dataset (R^2 > 0.9). Aerosol pH ranged from 0.96 to 4.92 with pH generally increasing with distance away from Cape Town. Observed aerosol- and gas-phase concentrations were typical for the remote marine atmosphere and will be presented in full. Temperature varied significantly across the 8 day transect from 18.42 °C near Cape Town to a minimum of -2.13 °C. Factors that control aerosol pH across the time and space scales observed will be evaluated and discussed as well as implications for improving our understanding of atmospheric chemistry and biogeochemical cycling in remote marine atmospheres.

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