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Air-sea exchange of reactive gases in the marine atmospheric boundary layer

L.L.Soerensen (1), S.C. Pryor (2,3), Sv-.E. Gryning (3)

 (1) Department of Atmospheric Environment, National Environmental Research Institute, Aarhus University, Denmark, (2) Atmospheric Science Program, Indiana University, Bloomington, IN 47405 USA, (3) Department of Wind Energy & Atmospheric Physics, Risø National Laboratory, Denmark (Contact Lotte.soerensen@risoe.dk)

Interactions between oceans and the atmosphere play a major role in climate, via exchanges of heat, gases and particles. The air-sea exchange of e.g. HNO3 and NH3 is not just important as nutrient input to the sea but the exchange processes is also affecting the particle and gas composition in the marine atmosphere. Uptake of gases on sea spray can enhance the nitrogen content in the marine aerosol, which then will affect the distribution of nutrient input (Pryor and Sørensen, 2000). The air-sea exchange of reactive species in marine coastal areas are difficult to measure due to the in-homogenous conditions in coastal waters with changing roughness, interaction between surface waves and atmosphere and sea spray influence. Thus several assumptions for applying M-O-similarity theory to calculate gas and particle fluxes are violated in the coastal zone. The assumption of a constant flux layer is not valid if the sink/source term in the continuity equation differs from zero. This is the case when sea spray is reacting with the specie of interest (Geernaert et al., 1998, Sørensen et al., 2005) causing a flux divergence.

Roughness changes due to changing wave state in the coastal zone will influence the horizontal gradient of the drag coefficient and thus the horizontal distribution of the flux and the concentrations. This will also add to the flux divergence. Here we present measurements and modeling of the interaction of nitric acid with sea spray to demonstrate their importance in dictating: (i) the spatial patterns of atmosphere-surface exchange of nitrogen (ii) vertical profiles of reactive nitrogen gases (iii) the ambient size

distribution of particle-nitrate.

Observations of nitric acid profiles at different coastal locations in Denmark and Sweden are presented. Profiles of nitric acid concentrations are measured using NaCl coated denuders placed in a mast at different heights. The profiles measured differ from the expected log profile suggesting influence by chemical reactions, stratified flows and/or horizontal gradients causing difference in foot print conditions. Simulations conducted using the ISOPART model indicate partitioning of total particle-NO3between sea spray and NH4NO3 is highly sensitive to the amount of sea spray present, and hence the initial vertical profile, sea spray source functions and the wind speed.

References Geernaert, L.L.Sørensen, Geernaert, G.L., Granby, K., and Asman, W.A.H., 1998, Fluxes of soluble gases in the marine atmospheric surface layer. Tellus 50B, pp 111-127. Pryor, S. and Sørensen L.L., 2000, Nitric Acid- Sea salt reactions: Implications for nitrogen deposition to water surfaces. Journal of Applied Meteorology. 39, 725-731. Sørensen, L.L., Pryor, S.C.; de Leeuw, G.; Schulz, M., Flux divergence of nitric acid in the marine atmospheric surface layer. J. Geophys. Res. (2005), 110, D15306, doi:10.1029/2004JD005403.