

Dry and wet deposition of reduced nitrogen to the Tampa Bay Watershed

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Ammonia and ammonium are the most abundant reduced nitrogen species in the atmosphere and can affect ocean chemistry through indirect deposition to adjacent watersheds. Ammonia (NH₃) can be transferred from the atmosphere via turbulent exchange, a process termed dry deposition. Ammonium (NH₄⁺) aerosols may be incorporated into precipitation and removed from the atmosphere by wet deposition. Dry and wet deposition of ammonia and ammonium have contributed to excess nutrient loading in coastal waters of Tampa, FL, USA [1].

In a series of experiments in 2002 and 2003, a relaxed eddy accumulation system was deployed to measure dry deposition of ammonia over natural vegetation in the Tampa Bay Watershed. Fluxes varied greatly with mean values of approximately $-0.03 \mu\text{g N m}^{-2} \text{ s}^{-1}$, which indicated a predominance of ammonia deposition to the watershed with infrequent occurrences of emission from the vegetation. However, deposition velocities were fairly consistent with means of approximately 2 cm s^{-1} [2].

Wet deposition data were retrieved from the Atmospheric Integrated Research Monitoring Network (AIRMoN), a cooperative effort of the National Atmospheric Deposition Program (NADP). Ammonium concentrations in precipitation were reported from a measurement site immediately adjacent to Tampa Bay. Concentrations of ammonium in collected precipitation samples averaged approximately 0.23 mg L^{-1} , which resulted in small ammonium fluxes during the experimental periods.

[1] Poor *et al.* (2001) *Atmos. Environ.* **35**, 3947–3955.

[2] Myles *et al.* (2007) *Enviro. Res. Lett.* **2**, 034004.

In situ characterization of fluid and melt structure in H₂O-saturated aluminosilicate systems in the deep crust and upper mantle

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The structure of H₂O-saturated haploandesitic melts and of coexisting silicate-saturated aqueous solutions, as well as that of supercritical silicate-rich aqueous liquids has been characterized *in situ* while the sample was at high temperature (to 800°C) and pressure (up to $\sim 1 \text{ MPa}$). Three glasses compositionally along the join Na₂O•4SiO₂-Na₂O•4 (NaAl)O₂-H₂O (0, 5 and 10 mol % Al₂O₃, denoted NS4, NA5, and NA10) were used as starting materials. Structural information was obtained with confocal microRaman and with FTIR microscopy. Fluids and melts were examined along pressure-temperature trajectories of isochores of H₂O at nominal densities (from PVT properties of pure H₂O) of 0.85 g/cm^3 (NA10 experiments) and 0.86 g/cm^3 (NA5 experiments) with the silicate+H₂O sample contained in an externally-heated, Ir-gasketed hydrothermal diamond anvil cell. Pressure derived from the EOS of pure H₂O was corrected by pressure-/temperature-calibrated Raman shift of synthetic ¹³C diamond.

Molecular H₂O (H₂O⁰) and OH groups forming bonds with metal cations (OH) exist in silicate-saturated fluids, water-saturated melts and in supercritical aluminosilicate liquids. The OH/H₂O⁰ ratio is positively correlated with temperature and pressure (and, therefore, fugacity of H₂O, $f_{\text{H}_2\text{O}}$) with $(\text{OH}/\text{H}_2\text{O}^0)^{\text{melt}} > (\text{OH}/\text{H}_2\text{O}^0)^{\text{fluid}}$ at all pressures and temperatures. Structural units of Q³, Q², Q¹, and Q⁰ type occur together in fluids, in melts, and, when outside the two-phase melt+fluid boundary, in single-phase liquids. The abundance of Q⁰ and Q¹ increases and Q² and Q³ decrease with $f_{\text{H}_2\text{O}}$. Therefore, the NBO/T (nonbridging oxygen per tetrahedrally coordination cations), of melt is a positive function of $f_{\text{H}_2\text{O}}$. The NBO/T of silicate in coexisting aqueous fluid, although greater than in melt, is less sensitive to $f_{\text{H}_2\text{O}}$.

The melt structural data can be used to describe relationships between activity of H₂O and melting phase relations of silicate systems at high pressure and temperature. The data were also combined with available partial molar configurational heat capacity of Qⁿ-species in melts to demonstrate how these quantities can be employed to estimate relationships between heat capacity of melts and their H₂O content.