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Seasonal and diurnal cycles of ammonia, nitrous acid and nitric acid at a forest site in Finland

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Background
In July – August 2010 a large campaign "Hyytiälä United Measurements of Photochemistry and Particles in Air - Comprehensive Organic Precursor Emission Concentration 2010 (HUMPPA – COPEC-10)", was conducted in a boreal forest at the SMEAR II station in Hyytiälä, southwestern central Finland. The general goal was to study links between gas phase oxidation chemistry and particle properties and processes. The Finnish Meteorological Institute contributed to the campaign with an on-line analyzer MARGA 2S (Ten Brink et al., 2007) for semi-continuous (1-hr time resolution) measurement of water-soluble gases and ions. Concentrations of gases (HCl, HNO_3, HNO_2, NH3, SO_2) and major ions in particles (Cl, NO_3, SO4, NH4, Na, K, Mg, Ca) were measured in two size fractions: PM2.5 and PM10.

The MARGA was kept running at SMEAR II also after the campaign. Here we discuss data collected until 30 April, 2011, and restrict the analysis to the nitrogen-containing gases. Ammonia plays a key role in neutralizing acidic atmospheric compounds and in aerosol formation. The concentration of semi-volatile aerosol species such as ammonium nitrate and ammonium chloride is strongly dependent on the gas phase precursors, NH3, HNO_3 and HCl. HONO is of atmospheric importance due to its expected significant contribution to the production of OH radicals.

Results and discussion
The median concentrations of ammonia (NH3), nitrous acid (HONO) and nitric acid (HNO_3) during whole period of 21 June 2010 – 30 April 2011 were 85, 54, and 57 ppt, respectively. The seasonal cycle was such that in summer the concentrations of all of these gases were the highest, the respective medians were 356, 70, and 81 ppt in June 21 – August 12, and lowest in winter (December – February), the respective medians were 38, 54, and 52 ppt. A very clear diurnal cycle of all these gases was observed, especially in July. In December there were no cyclic diurnal variation of these but in spring, especially in April the diurnal cycles were there again. The HNO_3 and HONO diurnal cycles were driven by the amount of solar radiation: when there was sunlight, the HNO_3 concentrations were higher and the HONO concentrations were lower than in the darkness. The concentration of ammonia was clearly positively correlated with temperature. An exponential curve was obtained from a regression: [NH3] = 77 ppt * exp(0.07 t) (r2 = 0.66) where temperature is expressed in degrees Celcius. This can be explained by agriculture-related and soil-related sources that are low when land is frozen and covered with snow

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Reference
Ten Brink H., Otjes R., Jongejan P., Slanina S. (2007) An instrument for semi-continuous monitoring of the size-distribution of nitrate, ammonium, sulphate and chloride in aerosol, Atm. Environ. 41, 2768-2779.