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Atmospheric nitrogen oxides (NO and NO_2) in ambient and firn interstitial air at Dome C: implications for modeling reactive nitrogen cycling on the East Antarctic Plateau

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The nitrogen oxides NO and NO_2 (NO_x) play a key role in determining the oxidizing capacity of the boundary layer in high latitudes. This influence is achieved via the photolysis of NO_2 – the only source for in situ production of tropospheric ozone (O_3) – and through shifting HO_x radical partitioning towards the hydroxyl radical (OH) via the reaction NO + HO_2 → OH + NO_2. Numerous field campaigns in the high latitudes demonstrated that the polar snow pack can emit significant amounts of NO_x and that one of the major driving mechanisms is UV-photolysis of nitrate (NO_3-) in snow.

Previously, we presented the first measurements of atmospheric NO_x at Dome C, East Antarctica (75.1ºS 123.3ºE, 3233 m) during austral summer 2009/2010. NO_x mixing ratios were highly perturbed, with a mean of 240 pptv (range 10-1000 pptv), but unlike at South Pole showed a strong diurnal variability. The timing of daily concentration extrema, the minimum at noon and the maximum in the evening, was shown to be largely determined by the dynamics of the local boundary layer.

Here we focus on NO_x observations in the firn interstitial air: gas phase mixing ratios at 10 cm depth were up to 10-fold those in the air above the snow and varied in phase with solar radiation, consistent with a photolytic source in the surface-near snow. Furthermore, shading experiments and firn air profiles suggest the existence of a NO_x reservoir in the upper snow pack. And finally, we estimate the total oxidant burden in the open pore space, a quantity, which is not easily measured, using NO_x flux measurements and deviations of the NO_2:NO ratio from steady state.

Observations were compared to the 1-D atmosphere-snow model MISTRA-SNOW, used previously to investigate NO_x snow-photochemistry at Summit/Greenland. Model runs constrained by wind speed, snow pack profiles of NO_3- concentrations and actinic flux are in close agreement with observations in ambient air. We examine how the parameterization of reactive nitrogen recycling at the snow-atmosphere interface can be improved and discuss future use in regional or global models.