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The Oxidant Production over Antarctic Land and its Export (OPALE) project: An overview of data collected in summer 2011-2012 at Concordia

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The need to characterize the oxidative capacity of the atmosphere of East Antarctica motivated the OPALE investigations at the top of the high plateau (Concordia) where processes are suspected to differ from those already identified at South Pole. For instance, in contrast to South Pole experiencing 24-hour sunlight, the solar irradiance at Concordia has a strong diurnal cycle. This has consequences on intensity of snow emissions as well as on the dynamic of the boundary layer. Concordia is also the inland site where the longest chemical ice core records have been extracted.

Investigations made at Concordia in December 2011-January 2012 included OH and RO_2 together with concurrent measurements of NO, NO_2, HONO, O_3, H_2O_2, HCHO, photolysis rates as well as meteorological parameters and physics of the boundary layer. HONO was investigated by deploying for the first time in Antarctica an absorption photometer (LOPAP), an analyser supposed to be free of interferences with numerous chemical species. Also investigated for the first time is the excess of 17O of ozone with a newly developed fast sampling method. The diurnal cycle of snow emissions was also documented for NO_x and HCHO that strongly influence the level of radicals.

The concentrations of OH and RO_2 radicals (median values of 3x106 and 1x108 in molecule cm-3, respectively) were found to be comparable to those observed at South Pole confirming that the elevated oxidative capacity is a common characteristic of near-surface atmospheric layer for most of the Antarctic plateau. Similar to the SP findings the major factor explaining to high radical levels at Concordia was the high levels of NO leading to fast recycling of RO_2 to OH. At the same time, in contrast to the SP where the radical levels are controlled by NO levels mostly via changing boundary layer properties, OH and RO_2 at Concordia show strong diurnal variability. The variability of NO_x at Concordia is also determined by the solar diurnal cycle via an inter-play between the NO_x snow emission rates and boundary layer dynamics.

The major radical primary sources at Concordia are represented by the photolysis of HONO, and HCHO. The main net losses of radicals are their reactions with NO_2 and cross radical reactions. It is found, however, that these results are inconsistent with radical observations leading to about 2 times overestimation of RO_2 and OH levels. At the same time, neglecting the OH production from HONO results in about 2 times underestimation of radical levels and to explain the radical observations in this case an additional OH source equivalent to about 20% of measured HONO photolysis is required. Assuming that HONO at Concordia originates from snow emissions whose the emission strength was evaluated from lab experiments done by irradiating surface collected at Concordia, model calculations suggest that HONO levels about 20% of measured are consistent with those calculated from radical measurements. We suggest that an explanation for the found overestimation of radical production could be an overestimation of measured HONO, which may originate from the interference from HO_2NO_2 affecting HONO measurements by LOPAP. Even with 5 times reduced HONO levels, the HONO photolysis represents the major primary radical source at Dome C accounting for about 40% of primary radical production.

Presented in this work analysis of the OH and RO_2 budget may be significantly biased by inconsistency of the mechanism based on the available observations with observed NO_2/NO ratios. Further studies of NO_x, HONO and radical chemistry at Antarctic Plateau are therefore strongly needed with specific efforts dedicated to increase the reliability of measurements (especially HONO) under polar conditions.