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Can laboratory data explain field observations: The fluxes of HNO₃ and HNO₄ from snow in the lab and in Antarctica

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Fluxes of trace gases between Polar snow packs and the boundary layer air are of importance because they significantly impact the composition and reactivity of the lower atmosphere and of the snow pack. They thus need to be understood to predict the oxidative capacity of the atmosphere on a regional scale and to interpret ice core data for reconstruction of past environmental conditions. A full understanding of observed fluxes is however complicated by the variety of underlying, elemental processes that may drive the fluxes synchronously: chemical reactions; diffusion in the gas, solid, or liquid phase; adsorption or desorption; or by uptake to the bulk ice.

In recent decades, some of these processes have been carefully investigated in laboratory-based experiments. Here, I will present our recent results on the partitioning of HNO4 between snow and air. These results are the first acquired at relevant low concentrations and show a significant lower tendency of HNO4 to stick to snow than previously thought. Fluxes of HNO4 in polar environments are not only of importance because HNO4 is a reservoir for HO_x and NO_x ; but also suspected to lead to interferences in HONO measurements.

A key question with this -and similar laboratory measurements- is, how well the description of physical processes derived under the idealized laboratory conditions can be applied to complex environmental settings. A data set published this summer on gas phase concentrations of HNO4 and of HNO_3 in costal Antarctica during winter gives the first and unique opportunity to show how well the agreement between laboratory and field investigation is. The authors of the field study have carefully selected specific days from their data set where photochemistry and ventilation by wind is absent. They hypothise and show that only ad- and desorption from the snow can explain the observed gas-phase concentrations.

Here, I go a significant step further and support the author's conclusion by discussing the equilibrium partitioning of HNO₃ and HNO4 that one would expect based on selected laboratory data. Both, adsorption to the surface of the snow and uptake to the bulk forming a solid solution are discussed (HNO₃ only). Further, I address the question, if the snow holds enough HNO₃ and HNO4 at its surface or in its bulk (HNO₃ only) to fuel the observed emissions. Thus both equilibrium conditions and molecular flux budgets are discussed.

These calculations show that adsorption/desorption can indeed explain the observed mixing ratio in the Antarctic boundary layer. Release from a solid solution seems to be too slow.

Jones, A. E., Brough, N., Anderson, P. S., & Wolff, E. W. (2014). HO₂NO₂ and HNO₃ in the coastal Antarctic winter night: a "lab-in-the-field" experiment. Acpd, 14(9), 12771–12796.

Interactive comment on " HO_2NO_2 and HNO_3 in the coastal Antarctic winter night: a "lab-in-the-field" experiment" by A. E. Jones et al. , T. Bartels-Rausch thorsten.bartels-rausch@psi.ch Received and published: 11 July 2014

Legrand, M., Preunkert, S., Frey, M., Bartels-Rausch, T., Kukui, A., King, M. D., et al. (2014). Large mixing ratios of atmospheric nitrous acid (HONO) at Concordia (East Antarctic plateau) in summer: a strong source from surface snow? Atmospheric Chemistry and Physics Discussions, 14(8), 11749–11785. doi:10.5194/acpd-14-11749-2014

Ulrich, T., Ammann, M., Leutwyler, S., & Bartels-Rausch, T. (2012). The adsorption of peroxynitric acid

on ice between 230 K and 253 K. Atmospheric Chemistry and Physics, 12(4), 1833–1845. doi:10.5194/acp-12-1833-2012