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Multiphase modeling of nitrate photochemistry in the quasi-liquid layer (QLL): implications for NO_x release from the Arctic and coastal Antarctic snowpack

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Abstract. We utilize a multiphase model, CON-AIR (Condensed Phase to Air Transfer Model), to show that the photochemistry of nitrate (NO_3^-) in and on ice and snow surfaces, specifically the quasi-liquid layer (QLL), can account for NO_x volume fluxes, concentrations, and $[NO]/[NO_2]$ ($\gamma=[NO]/[NO_2]$) measured just above the Arctic and coastal Antarctic snowpack. Maximum gas phase NO_x volume fluxes, concentrations and γ simulated for spring and summer range from 5.0×10^4 to 6.4×10^5 molecules cm^{-3} s^{-1}, 5.7×10^8 to 4.8×10^9 molecules cm^{-3}, and ~0.8 to 2.2, respectively, which are comparable to gas phase NO_x volume fluxes, concentrations and γ measured in the field. The model incorporates the appropriate actinic solar spectrum, thereby properly weighting the different rates of photolysis of NO_3^- and NO_2^-. This is important since the immediate precursor for NO, for example, NO_2^-, absorbs at wavelengths longer than nitrate itself. Finally, one-dimensional model simulations indicate that both gas phase boundary layer NO and NO_2 exhibit a negative concentration gradient as a function of height although $[NO]/[NO_2]$ are approximately constant. This gradient is primarily attributed to gas phase reactions of NO_x with halogens oxides (i.e. as BrO and IO), HO_x, and hydrocarbons, such as CH_3O_2.

Final Revised Paper (PDF, 757 KB)   Supplement (114 KB)   Discussion Paper (ACPD)

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