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A mechanism for the photochemical transformation of nitrate and nitrite in snow

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The photolysis of nitrate in snow has been the subject of several field and laboratory studies. It is now well known that the photolysis leads to the formation of nitrogen oxides, which are released to the gas phase. In addition, field studies demonstrated that nitrous acid could be released from surface snow. This has also been attributed to the photolysis of nitrate. To investigate the possible mechanism leading to the formation of the different products of the nitrate photolysis, we performed photolysis experiments with artificial snow samples. We prepared snow samples with varying amounts of nitrate and nitrite, the anion of nitrous acid. These samples were irradiated using a high-pressure mercury lamp emitting radiation in the UV, visible and IR wavelength range. Starting with high nitrate and low nitrite concentrations led to the depletion of nitrate and the formation of nitrite in the snow, which subsequently also decayed. However, in the experiments with high nitrite and low nitrate initial concentrations significant amounts of nitrate were formed. This indicates that reaction cycles involving the recycling of nitrite to nitrate occurred. A simple kinetic mechanism was developed involving reactions of the nitrogen oxides in the liquid-like surface layer of the snow crystals to describe the observed experimental data sets. In a fitting procedure using the FACSIMILE software the unknown experimental photolysis rates of nitrate and nitrite were adjusted to give the best agreement between calculated and experimental time series of nitrate and nitrite concentrations with four different initial concentrations. The reaction mechanism and the obtained rate constants will be presented and discussed in detail.