

## **Air–snow exchange of nitrate: a modelling approach to investigate physicochemical processes in surface snow at Dome C, Antarctica**

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Snowpack is a multiphase (photo)chemical reactor that strongly influences the air composition in polar and snow-covered regions. Snowpack plays a special role in the nitrogen cycle, as it has been shown that nitrate undergoes numerous recycling stages (including photolysis) in the snow before being permanently buried in the firn. However, the current understanding of these physicochemical processes remains very poor. Several modelling studies have attempted to reproduce (photo)chemical reactions inside snow grains, but these required strong assumptions to characterise snow reactive properties, which are not well defined.

Physical processes such as adsorption, solid state diffusion and co-condensation also affect snow chemical composition. We developed a model including a physically based parameterisation of these air–snow exchange processes for nitrate. This modelling study divides into two distinct parts: firstly, surface concentration of nitrate adsorbed onto snow is calculated using existing isotherm parameterisation. Secondly, bulk concentration of nitrate in solid solution into the ice matrix is modelled. In this second approach, solid state diffusion drives the evolution of nitrate concentration inside a layered spherical snow grain. A physically-based parameterisation defining the concentration at the air–snow interface was developed to account for the the co-condensation process.

The model uses as input a one-year long time series of atmospheric nitrate concentration measured at Dome C, Antarctica. The modelled nitrate concentration in surface snow is compared to field measurements. We show that on the one hand, the adsorption of nitric acid on the surface of the snow grains fails to fit the observed variations. During winter and spring, the modelled adsorbed concentration of nitrate is 2.5 and 8.3-fold higher than the measured one, respectively. A strong diurnal variation driven by the temperature cycle and a peak occurring in early spring are two other major features that do not match the measurements. On the other hand, the combination of bulk diffusion and co-condensation incorporation processes allows a good reproduction of the measurements (correlation coefficient  $r=0.95$ ), especially with a correct amplitude and timing of summer peak concentration of nitrate in snow. During wintertime, nitrate concentration in surface snow is mainly driven by thermodynamic equilibrium, whilst the peak observed in summer is explained by the kinetic process of co-condensation.

For the first time, this study elucidates the air–snow transfer function of nitrate, based on a process-resolving modelling approach. This study also demonstrates that the co-condensation is the most important process to explain nitrate incorporation in snow subject to temperature gradients.