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Investigating the Uptake Mechanisms of Hydrogen Peroxide to Single and Polycrystalline Ice with a Novel Flow Tube System

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Air-ice chemical interactions are important for describing the distribution and subsequent chemical fate of trace atmospheric gases within ice and snow and determining the oxidative capacities of these environments. The nature of this interaction is governed by a compound's physicochemical properties as well as ice microstructure. Hydrogen peroxide (H_2O_2), a reservoir of HO_x radicals in the atmosphere and an important chromophore in snow and ice, is a trace gas that demonstrates complex uptake behaviour to frozen aqueous media by the reversible, fast adsorption to the air-ice interface, aggregation, and lateral interactions, and a slower process, ostensibly via uptake into the bulk. However, the exact mechanism and kinetics for the slow uptake of H_2O_2 and the size of this reservoir is unknown. It is important to describe and quantify this loss term, over environmentally-relevant timescales, accommodation of H_2O_2 into the bulk may be the dominant process which controls the composition and chemistry of the snow and overlying atmosphere. We hypothesize that the slow uptake of H_2O_2 occurs by diffusion into the grain boundaries of ice.

To provide mechanistic insight to the macroscopic phenomenon of atmospheric gas uptake to ice, and discern various mechanisms including adsorption to air-ice interface and accommodation into the bulk through uptake into grain boundaries, we design, machine, and validate a novel flow reactor system featuring a Drilled Ice Flow Tube (DIFT). Our flow reactor system is uniquely suited to testing these uptake mechanisms: by controlling the degree of grain boundaries present in the DIFT (ie. monocrystalline or polycrystalline), we can directly observe the effect of the ice microstructure on the adsorptive and bulk uptake of trace atmospheric gases over long timescales (eg. on the order of hours).

Here, we describe method development of the DIFT and demonstrate using polarised microscopy imagery that our experimental set-up allows for the direct interrogation of adsorption and uptake to ice grain boundaries. Using our novel DIFT reactor system, we present laboratory kinetic results for the fast and slow uptake of H_2O_2 and infer the environmental significance of these processes.