EGU21-1410
https://doi.org/10.5194/egusphere-egu21-1410
EGU General Assembly 2021

Molecular dynamics approach to assess aqueous alteration of potassium-rich feldspar surfaces

Anand Kumar, Allan K. Bertram, and Grenfell N. Patey
Department of Chemistry, The University of British Columbia, Vancouver (Canada) V6T 1Z1

Ice clouds play an important role in the Earth's radiative budget and hence climate. Heterogeneous ice nucleation, a major pathway for ice formation in cirrus and mixed-phase clouds, is induced by active sites present on atmospheric aerosol particles termed as ice-nucleating particles. Feldspars have been shown to be highly ice nucleation active. Despite the importance of mineral dusts for ice nucleation, the role of atmospheric aging (e.g. surface alteration due to interactions with chemical species) on their ice nucleation efficiency is largely unknown. This is primarily due to the lack of microscopic level insight into nucleation from laboratory/field-based experiments, due to the inability to experimentally access the small spatial and temporal scales at which nucleation process occurs – a problem that can be potentially tackled with computer simulations. We utilize direct Molecular Dynamics simulations (GROMACS 5.1.4) to investigate the interactions of solutes with different surfaces of potassium feldspar mineral (microcline) and the corresponding interfacial water structure at a microscopic scale. We investigated the interactions of monovalent cations (H_3O^+, $(NH_4)^+$, Li^+, K^+, Cs^+) with various surfaces of microcline, and subsequent effects on the near-surface water structure at 300 K. The investigated surfaces include the perfect cleavage planes, (001) and (010), as well as the high energy plane (100) of microcline. Feldspar is modeled as semi-rigid (lattice atoms fixed expect K^+ and H of surface OH) and as fully flexible (all lattice atoms free to move) with the CLAYFF force field, and the TIP4P/Ice model is employed for water. Results show that on simulation timescales, lattice vibration is necessary for ion exchange between added cation and lattice K^+, albeit at different exchange rates for the 3 planes. None of the 3 flexible surfaces show any preference for over K^+ in terms of ion exchange within the simulation timescale. Both the semi-rigid and flexible surfaces show higher adsorption of molecular cations ($(NH_4)^+$ and H_3O^+) compared with the simple spherical cations. In addition, we do not observe ice nucleation on modified microcline surfaces (both semi-rigid and flexible) at a supercooled temperature of 230 K within the simulation timescale. To conclude, the presented work provides an improved understanding of the processes modifying the feldspar surfaces in water and aqueous solutions and its possible relevance for ice formation.