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Spontaneous formation of oxidants at liquid-ice interface

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Recently, intensive new particle formation (NPF) events have been observed in the upper troposphere/lower stratosphere (UTLS), where ice formation and deep convection are predominant. These NPFs require atmospheric oxidants for reducing the volatility of condensation vapors and thus induce the gas-to-particle conversion. Meanwhile, the source of atmospheric oxidants should be strong enough to compete with the condensation sinks induced by the preexisting particles. The underlying formation mechanisms of atmospheric oxidants including HO_x radicals and H_2O_2 remain poorly understood. We propose that the ice formation followed by water condensation on ice surface are sources of H_2O_2 or HO_x radicals in the UTLS driving NPF. The Workman-Reynold effect that freezing of salt containing water producing strong internal electric field at liquid-ice interface supports this hypothesis. However, the formation efficiency of oxidants via liquid-ice interface currently is completely unknown. In this work, we will determine the formation efficiency of oxidant via freezing salt containing water by indirect and direct measurement methods. The titration reactions such as the reaction of terephthalate with OH radical to form a fluorescent product hydroxyterephthalate will be adopted for quantitatively and qualitatively determining the OH radical formation in ice or liquid phases. A high-sensitive fluorescence detector (AERO laser model AL 2021) will be used for directly measuring the H_2O_2 production in liquid phase. The factors like freezing rate, PH values and salt solution composition, controlling the formation efficiency of oxidants will be investigated. Subsequently, we will explore if the formed oxidants together with the injected gas precursors (SO_2 or VOCs) can lead to NPF in a custom-in ice-coated flow tube. The gas and particle species will be measured by various instruments (PTR-MS, SMPS and EESI-Orbitrap). This study can improve our ability to describe the multiphase oxidation processes of the UTLS regions.