HO₂ uptake onto deliquesced inorganic aerosols and doping impacts of transition metal ions

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The uptake of HO₂ radicals onto aerosols potentially affects VOC oxidation, ozone formation, and SOA generation. Also, the uptake process changes the aerosol properties such as size, oxidative capacity, and nucleation which further impacts the climate. However, such a process has been poorly understood and most multiphase reactions remain complicated. Previous studies used an aerosol flow tube combined with a radical detector and such approaches require manual controlling of the injection and thereby is in low time resolution. Our work combines a chemical conversion (CC) method with laser-pump and laser-induced fluorescence (LP-LIF) technique to detect the loss rate of HO₂ onto deliquesced inorganic aerosols as OH reactivity (the inverse of OH lifetime) by converting HO₂ into OH. The detection interval is 4 min and the aerosol surface area was measured simultaneously for uptake coefficient calculation.

The CC/LP-LIF technique was applied jointly with an atomizer for aerosol uptake of HO₂ under high RH conditions. Polydisperse salt aerosols were produced from (1) NaCl, (2) $(NH_4)_2SO_4$, (3) Na₂SO₄, and (4) Seawater. The 0.03% w/v reagent alone or contained with 0.0015% w/v additions (Cu(II), or Fe(II)) was dissolved in ultra-pure water for target aerosols. The detected radical loss rate can be explored by a single exponential fitting equation assuming pseudo-first order reactions. Gas-phase diffusion effect was removed to estimate the real uptake coefficient (). The determined from different aerosols was generally consistent with respective reported values. The detection limit of this CC/LP-LIF method is ~0.01 determined from the uncertainty of HO₂ reactivity of zero-air measurement and the maximal total surface concentration of aerosols generated from the atomizer. For the first time, we measured from Na₂SO₄ and two seawater samples. The enhancement effect was confirmed from ion-doped aerosols regardless of the type of parent aerosols. In all cases, from with Cu(II) was larger than that with Fe(II). Seawater samples showed quite big variations from two sites, and this may indicate the ionic strength influence which varies naturally across the aquatic ecosystem. The result of this study can complement current IUPAC report and also be embedded in model for oxidant production simulations.