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Measurements of Surrogate Respiratory Sessile Droplet pH and Implications for Exhaled Respiratory Aerosol and Airborne Disease Transmission

The respiratory aerosol pH has been proposed as a major driver for the infectivity loss of SARS-CoV-2 viruses and influenza A virus in exhaled aerosols, thus affecting the airborne transmission of respiratory diseases. A pH of 9-11 within aerosol droplets was considered driving the loss of SARS-CoV-2 infectivity (Haddrell et al., 2023). Sodium bicarbonate acts as a principal buffer in biological systems, regulating blood pH and balancing the CO_2 content in the gas and liquid phases. Upon exhalation, changes in gas phase conditions affect the aerosol composition and pH. Despite several studies utilising Raman spectroscopy to quantify atmospherically relevant aerosol pH, there is limited understanding of the kinetics of CO_2 partitioning and pH variability within respiratory fluid-relevant droplets.

The aim of this study is to explore the HCO_3^-/CO_3^{2-} equilibrium in surrogate respiratory fluid sessile droplets to elucidate the pH evolution of exhaled respiratory aerosol, yielding crucial insights for understanding the aerosol processes that are central to airborne disease transmission. In situ direct measurements are made using Raman spectroscopy, which is applied to study the kinetics of CO_2 evaporation from sodium chloride-sodium bicarbonate microlitre sessile droplets with and without the enzyme carbonic anhydrase (CA), respectively. In particular, we explore the impact of gas flow rate, droplet volume, gas phase CO_2 concentration, and enzymatic catalyst concentration on the CO_2 evaporation kinetics. The experimental results are used to benchmark the Respiratory Aerosol Model (ResAM), which simulates respiratory picolitre droplet thermodynamics and pH evolution (Luo et al., 2023).

Our measurements show that the CO_2 evaporates slowly in sessile droplets of volume of 40 µL, taking ~200 minutes for the droplet to reach pH 9 from an initial level 7.6. The effect of changing droplet size and gas flow rate to the pH change is within the experimental error. However, the gas phase CO_2 level controls the final pH profoundly. The ResAM simulates the pH evolution profile and is highly comparable to the experimental measurements. When scaled to respiratory aerosol droplet sizes of 0.004 and 65 pL, the pH occurs over ~27 min. The simulated pH evolution profiles of picolitre droplets show size independence. Simulations for both sessile droplet and respiratory aerosol show that the presence of CA can significantly increase the rate of pH increase, and the gas phase CO_2 level is also important for determining the final droplet pH.

This study represents the first direct in situ investigations into the pH dynamics of an aqueous sodium chloride-sodium bicarbonate droplet and CO_2 evaporation kinetics using Raman spectroscopy. The composition of human respiratory fluid is dynamic, reflecting the true nature and physicochemical properties of exhaled aerosols is challenging. However, this study highlighted the timescale differences for the pH increase in microlitre sessile droplet and aerosol droplets of respirable size, that are responsible for fomite and airborne transmission, respectively. This information is crucial for understanding the aerobiological pathway for virus transmission and providing evidence to inform indoor airborne control strategies.

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