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Fine particle pH and gas-particle partitioning during winter fog in Delhi, India

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Comprehensive measurements were conducted to simultaneously monitor the trace gases (HCl, HONO, HNO_3, SO_2, and NH_3) and inorganic chemical constituents (Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Ca^{2+}, and Mg^{2+}) of fine particulates (PM_1 and $PM_{2.5}$) at hourly resolution during the Winter Fog Experiment (WIFEX) field campaign, Delhi, India, for the winter period of 2017-2018. The measurements were performed using the instrument called Monitor for AeRosols and Gases in Ambient air (MARGA-2S) to study the role of chemical composition and gas-particle interplay chemistry in the life cycle of fog, i.e., formation, development, and dissipation phase. In the past, the variation of fine particle acidity (pH) and its impact on fog has not been studied explicitly and quantitatively over Delhi. The pH is a fundamental property of aerosol that plays a significant role in the chemical behavior and composition of particles, but it is very challenging and difficult to measure directly. Particulate water is also a significant component of aerosol and can serve as a medium for aqueous-phase reactions under foggy conditions. The pH depends on the particle water amount, as pH represents the concentration of H^+ per liquid water volume (i.e., particulate water). Whereas, H^+ concentration per unit volume of air is defined as the particulate proton loading.

Using the measured gas-phase and particle-phase concentrations and meteorological observations (T, RH), the particulate water and pH were estimated from the thermodynamic model ISORROPIA-II. In this study, the gas phase NH_3, HNO_3, and HCl and particle-phase NH_4^+, NO_3^-, Cl^-, and SO_4^{2-} species were estimated using ISORROPIA-II, and model predictions of these species were validated by using the measured gas and particle-phase species. The predictions were confirmed by a good agreement between predicted and measured ammonia concentrations (r=0.94) and aerosol species concentrations ammonium (r=0.97) chloride (r=0.61), nitrate (r=0.61), and sulfate (r=0.74). The predicted $PM_{2.5}$ pH ranged from 2.55 to 6.54, with mean pH of 4.55 ± 0.51. This was consistent with the findings of previous studies. It is concluded that high particle water content, higher acidic pH, and abundant ammonia concentrations can promote the gas-particle partitioning and formation of more secondary particles under foggy conditions. The scattering cross-section of these secondary fine hygroscopic particles increases under high humidity conditions due to water uptake, resulting in visibility degradation.