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Aerosol acidity and its neutralization in the eastern Indo-Gangetic Plain: implications for water-soluble organic carbon

Bijay Sharma[1], Anuraag J. Polana[1], Prashant Rawat[1], and Sayantan Sarkar[1,2]
[1]Department of Earth Sciences, Indian Institute of Science Education and Research, Kolkata, India
[2]Centre for Climate and Environmental Studies, Indian Institute of Science Education and Research, Kolkata, India

Aerosol acidity plays an important role in influencing precipitation pH, which has impacts on the environment as well as human health. It also has significance in shaping aerosol chemistry, including the catalytic formation of water-soluble organic carbon (WSOC), which in turn affects the hygroscopicity of aerosols. Past studies on aerosol acidity in the Indian subcontinent, mostly conducted in biomass burning (BB) source regions in the northwestern and central Indo-Gangetic Plain (IGP) and in western India, have identified Ca^{2+} and Mg^{2+} sourced from desert dust to be the predominant neutralizing agents. However, the prevalence of desert dust decreases progressively along the IGP corridor and is potentially rendered insignificant in the eastern IGP (eIGP). As such, there exists a critical weakness in our understanding of the processes governing aerosol acidity and its neutralization in the eIGP. To address this, the present study reports the seasonal variability of ionic species, WSOC and associated aerosol acidity in ambient $PM_{2.5}$ from a rural receptor site in the eIGP. To this end, a total of 88 $PM_{2.5}$ samples collected during the summer, post-monsoon and winter seasons of 2018 were analyzed for SO_4^{2-}, NO_3^-, Cl^-, Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, F^-, PO_4^{3-} and WSOC, followed by estimation of strong acidity. Across all seasons, the aerosol phase was dominated by SO_4^{2-}, NH_4^+ and NO_3^-, with values increasing by factors of 1.8-1.9, 1.4-2.9 and 1.8-11, respectively, for the regional BB-dominated post-monsoon and winter seasons as compared to summer. Significant positive Cl^- depletion in summer pointed towards the influx of marine air while negative depletion in post-monsoon and winter suggested a BB source, which was further supported by concentration-weighted trajectory analysis. The averaged pH of the aerosol extract decreased progressively from summer (5.5±0.4) to winter (4.5±0.2). NH_4^+ was observed to be the major acid-neutralizing agent across all seasons, with dust-derived Ca^{2+} and Mg^{2+} playing only minor roles. In general, WSOC formation is known to be catalyzed by the presence of excess acidity; however, during winter, it appeared that the regional transport of organic acids in the BB plume contributed to aerosol acidity at this receptor site ($r=0.92$; $p<0.01$ for WSOC and H^+). BB-derived K^+ appeared to perform a dual function of neutralizing acidity as well as producing it via reactions with WSOC during atmospheric transport. The wintertime acidity was also strongly governed by aerosol NO_3^- sourced from BB emissions and possibly accentuated via nighttime atmospheric chemistry at lower ambient temperatures, resulting in the formation of haze. These observations of the NO_3^- and WSOC-driven wintertime acidity, the dual function of K^+ and the dominant role of NH_4^+ in neutralization points to complex atmospheric processing of the IGP outflow during its transport to the eastern end of the corridor, which warrants further

investigation.

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