ABSTRACT

This is in an age of rampant climate change. There are huge uncertainties in the quantification of effective radiative forcings due to the interaction of aerosols with both clouds and solar radiations as well as precipitation by GCMs (Global Climatic Models). This work is a small effort to contribute to reduce these uncertainties in the long run. This was made possible by (a) adding to the estimates of hygroscopicity parameter (κ) and cloud condensation nuclei (CCN) in response to the dearth of such data, especially in the Indian sub-continent, and (b) by adding to the scientific understanding of these parameters. These have been accomplished as a result of two campaigns, both carried out at New Delhi, which is a highly polluted continental site in southern Asia home to anthropogenic emissions locally and long-range transfer of pollutants.

The first campaign was a long-term study from Jan 2017- Mar 2018 based on time-resolved estimates of κ and CCN in the PM_1 (Particulate Matter upto 1µm; refers to aerosol particles with an aerodynamic diameter within 1 µm regime) size range obtained from chemical composition (NH_4^+, SO_4^{2-}, NO_3^-, Cl^-, and organics that constitute the Non-refractory PM_1 ($NRPM_1$) species) and size distribution, for the first time. Air mass originating from southern Asia (SA) was the most polluted with high Cl^- and organic loading. SA was followed by the Bay of Bengal air mass (BB) with higher organics and NO_3^- with contamination exceeding that of the Arabian Sea air mass (AS). The dominant sources included biomass-burning and emissions from thermal power plants, industries, and vehicles. The average κ was reported to be 0.3 (ranging from 0.13-0.77) for all the air masses. The source activities by way of impacting the aerosol chemical composition impacted κ's diurnal pattern. SA> BB >AS was seen in terms of (a) $NRPM_1$ loading, (b) number concentrations (N_{CN}) in the bulk, Aitken (N_{Aitken}), and accumulation (N_{Accu}) modes, (c) means of CCN number concentration (N_{CCN}; 3669–28926 cm^{-3}) and, (d) the activated fraction (a_f; 0.19–0.87), for supersaturations (SS) ranging from 0.1- 0.8 %. Size impacted N_{CCN} more than chemical composition directly, being governed by N_{Aitken} or N_{Accu} depending on SS and critical diameter (D_c). a_f was ruled by the geometric mean diameter (GMD). The world's highest a_f (0.71±0.14 for the most dominant sub-branch of the SA air mass- R1- at 0.4 % SS) for a continental site is reported owing to very low D_c (25–130 nm, for 0.1- 0.8 % SS). Indirectly, N_{CCN} and a_f were impacted by the chemical properties via their impact on the diurnal behaviour of N_{Aitken}, N_{Accu}, κ, and D_c.

The first study was based on the most prevalent assumption of ammonium salts being the dominant salts at continental sites other than NaCl at a marine location. It is hypothsized that this assumption may lead to a huge underestimation of κ for a polluted site subject to heavy mineral dust loading like New Delhi. Accordingly, the second campaign which is also a long-term study was conducted from Dec 2012–2014 wherein detailed chemical speciation of $PM_{2.5}$ aerosols of inorganic ions (NH_4^+, Na^+, K^+, Ca^{2+}, Mg^{2+} SO_4^{2-}, NO_3^-, and Cl^-) and organic carbon (OC) was carried out based on filter-paper measurements of day and night. The validation of this hypothesis necessitated the quantification of mineral dust on κ. To accommodate the components of mineral dust in κ estimation, two new mathematical approaches differing in the sequence of salt formation based on Mann-Kendall's test were coined. The inclusion of mineral dust components resulted in an average increment of 57% and 63.5% in κ for day and night respectively. κ which was inclusive of mineral dust was inter-compared with those deduced using other analytical methods. This approach challenges the traditional κ estimation κ based on the dominance of ammonium salts while ignoring (a) the other components, their individual and collective impact on κ, and (b) meteorological parameters like temperature (T) and Relative Humidity (RH). The impact of mineral dust on κ decreased with an increase in water-soluble organic carbon to organic carbon ratio. The consideration of insoluble calcium salts lessened the effect of mineral dust on the one hand, while on the other hand resulted in $MgCl_2$ formation which led to an increase in the estimated κ. κ*'s* highest value was reported as 0.82 (on 23/7/2014 during the night). The significant contribution of mineral dust to κ was further verified for the cities of Durg and Kanpur in India, and Xian and Beijing in China for PM_1 and $PM_{2.5}$ aerosols.

The high values of κ, N_{CCN}, and a_f in the PM_1 regime can severely impact Delhi's rainfall, the radiation budget, and aerosol indirect effect, and require a thorough investigation to quantify this impact. The high impact of mineral dust on κ at polluted sites as seen in the $PM_{2.5}$ regime for Delhi, and both PM_1 and $PM_{2.5}$ regimes for the other cities can be of crucial relevance for CCN estimation and thus impact precipitation quantification by GCMs, lead to visibility deterioration and affect the indirect radiative forcing.