Investigation of climate relevant properties of aerosol and contribution of sources in arid region, Bikaner, India

Name: Sayantee Roy

Entry Number: 2017CEZ8086

Abstract. The concern of deteriorating air quality and the problem of climate change has been linked to dynamics of atmosphere aerosols (or particulate matter, PM). Due to the ubiquitous presence and dynamic nature of aerosols from source to sink, large uncertainties arise in their assessment related to regional and global climate impacts and health effects. As the behaviour are sensitive to a combination of factors like source and meteorology that determines its residence time and transformations in the atmosphere, detailed study in topographically and climatologically distinct regions of the world, and the inclusion of the findings in climate models is sensed useful to reduce prediction uncertainties. Moreover, strategic planning for policy making in order to address local air quality requires baseline understanding of emission sources and impacts. Therefore, this research work is the first set of measurements designed to study the variations, interactions and interrelations of aerosol properties in a typical arid area in the western region of India, where local and regional pollution mixes with natural crustal components. To achieve the designed objectives a top-down approach for identification of probable source of airborne PM is followed in this study, that includes a continuous particle sampling for a two-year period, and measurement of the suite of chemical constituents (elemental and organic carbon, ions, trace metals), followed by source identification by receptor modelling. To address a comprehensive physico-chemical characterization of aerosols and its effect of optical properties that regulates the local air and regional climate, the period of typically substantial PM load seasons was studied.

Exploration of the chemical constituents of $PM_{2.5}$ revealed strong seasonality for $PM_{2.5}$ and its constituents. Highest OC and EC concentrations ($\mu g\ m^{-3}$) were found during post-monsoon (15±17 and 5±3) and winter (15±10 and 6±4), respectively, and lowest (2±1 and 0.5±0.9), respectively, in the monsoon. The study site located in an arid region mixed with urban establishment. The favourable atmosphere (temperature >30˚C and humidity ~50%) during monsoon and availability of precursors to instigate photooxidation through OH radicals led to transformation of highly volatile organic fractions (OC1) to less volatile higher molecular weight organic fractions and increased the water solubility of OC to form WSOC i.e water-

soluble organic carbon (66% of OC). The production of water soluble in-organic ions (μg m^{-3}) through aqueous phase chemistry governed $PM_{2.5}$ mass in winter (45.4±31.8; 51%) and post-monsoon (32.3±18.5; 42%). Another interesting finding is the chemical neutralization during dust prone seasons were regulated by cations of crustal origin, whilst, in colder seasons by NH_4^+. Majorly, an ammonia-rich environment was prevalent in winters and post-monsoon, when likely formations of ammonium salts were seen. On contrary, compounds of crustal origin were prevalent at ammonium-poor conditions (warmer months). Saline soil and playa lakes of Thar desert has been a consistent natural source of NaCl and Na_2SO_4. Overall mean concentrations (μg m^{-3}) of major (Mg, S, Si, Al, Cl, Ca, Fe and K) and minor trace metals were found as 19±17 and 0.5±0.4 respectively. Non-water-soluble Ca and Mg existed as $CaCO_3$, MgO and $Mg(OH)_2$. Inhibited photochemical dissociation causing elevated ambient HNO_3 during colder days facilitated the formation of highly soluble and hygroscopic $Ca(NO3)_2$, deliquescence at >10% RH, which could catalyse CCN and denser haze formations.

To identify the source contributions based on the chemical composition USEPA-PMF5.0 was run and the analysis resolved seven factors which represents seven sources identified by their chemical profiles together with tracer species. The sources resolved were: Secondary sulphate (25.2%), resuspended dust (23.6%), residential cookstove (13.4%), smelter and non-tail-pipe emissions (11.3%), vehicular emissions (10.8%), lake and sea salt (8.5%), secondary nitrate (4.8%), and brick kilns (2.2%). Winter and post-monsoon PM concentrations in the region were majorly regulated by secondary sulphate and residential cookstove emissions, contributing to 22% and 22-27%, respectively. Dust storms prevalent in the region during pre-monsoon and monsoon have resulted in higher resuspended dust contribution (30-43%) from crustal sources during these seasons. Salt lakes in the region have influenced the fine PM mass with its effect aggravated during the monsoon by the sea-salts from the Arabian Sea. The fine PM mass has been impacted by salt lakes in the area, which is exacerbated during the monsoon by sea-salts from the Arabian Sea. Smelters, non-tailpipe emissions, and vehicular emissions were substantial contributors to moderately polluted to poor conditions ($PM_{2.5}$ loadings >60 μg m^{-3}). Furthermore, highest $PM_{2.5}$ loading (>90 μg m^{-3}) marking the very poor conditions in the region was contributed by residential cookstove emissions and secondary sulphate. While vehicular emissions were found to be primarily from vehicles (i.e., local sources), numerous smelters from northern states had potential influence on PM mass. The prominent seasonality in the source contributions observed in this study is particularly suggestive of substantial influence of meteorology on PM loading in the region.

The inter-relation between physiochemical and optical characteristics of aerosols during the post-monsoon (Oct, Nov, Dec) and winter (Jan-2021) were inspected by categorizing the days of these seasons into clear (37%), light (33%) and high (31%) pollution periods based on horizontal visibility and measured $PM_{2.5}$ concentrations. Elemental and organic carbon (50±15 μ gm^{-3}; 31%) and secondary inorganics (53±21 μ gm^{-3}; 33%) dominated the $PM_{2.5}$ mass (160±4 μ gm^{-3}) during high pollution period with low dust (14±7 μ gm^{-3}; 8%) content. Interestingly, the clear pollution period was also influenced by carbonaceous fraction (19±8 μ gm^{-3}; 32%) and secondary inorganics (19±5 μ gm^{-3}; 32%), but the $PM_{2.5}$ concentrations (59±9 μ gm^{-3}) were ~one-third as compared to high pollution period. High scattering coefficients were observed which were comparable to highly polluted Indian city like Delhi. An exponential increase in non-absorbing material was observed and showed a clear influence on light absorption capacity of EC and dust due to coating/mixing. Higher absorption Ångström exponent (AAE) >0.6 was observed for substantial BrC (Brown carbon), BC (Black carbon) and dust while increase in non-absorbing to absorbing components ratio >4 and low amount of EC (<4%) and BrC reduces AAE below 0.4. Higher mass absorption cross-section (>30 m^2g^{-1} of EC) was observed when 4-10% EC fraction of $PM_{2.5}$ associated with 2.5-3.5 times non-absorbing components to total absorbing components. Likewise, absorption enhanced by three to five-fold compared to uncoated EC for low EC fraction (3-6%) in $PM_{2.5}$, but high non-absorbing to absorbing component ratio (>2.5). Interestingly, absorption was minimally amplified for nominal coating fraction associated with significant core materials or vice-versa. These findings have implications not only in regional climate assessment but also for other regions with comparable geography and source-mixes.

Overall, the study puts forward a complete understanding of the chemical composition and source contribution of fine particulate matter in an arid desert region, with urban influences. Distinct aerosol properties in the study region revealed from the study emphasizes the importance to include region specific aerosol properties in the climate models to reduce uncertainties. The results of this work are expected to serve as database to address global climate change in response to long-range transport to and from a desert region with urban influence. Additionally, this work is expected to provide a valuable understanding for researchers, and climate modellers to incorporate the inputs exclusive to similar regions in climate models to obtain more accurate region-specific predictions of future climate change.