



Size resolved fog water chemistry and its atmospheric implications

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Fog is a natural meteorological phenomenon that occurs throughout the world. It usually contains substantial quantity of liquid water and results in severe visibility reduction leading to disruption of normal life. Fog is generally seen as a natural cleansing agent but it also has the potential to form Secondary Organic Aerosol (SOA) via aqueous processing of ambient aerosols. Size-resolved fog water chemistry for inorganics were reported in previous studies but processing of organics inside the fog water and quantification of aqSOA remained a challenge. To assess the organics processing via fog aqueous processing, size resolved fog water samples were collected in two consecutive winter seasons (2012-13, 2013-14) at Kanpur, a heavily polluted urban area of India. Caltech 3 stage fog collector was used to collect the fog droplets in 3 size fraction; coarse (droplet diameter $> 22 \mu\text{m}$), medium ($22 >$ droplet diameter $> 16 \mu\text{m}$) and fine ($16 >$ droplet diameter $> 4 \mu\text{m}$). Collected samples were atomized into various instruments such as Aerosol Mass Spectrometer (AMS), Cloud Condensation Nucleus Counter (CCNc), Total Organic Carbon (TOC) and a thermo denuder (TD) for the physico-chemical characterization of soluble constituents. Fine droplets are found to be more enriched with different aerosol species and interestingly contain more aged and less volatile organics compared to other coarser sizes. Organics inside fine droplets have an average $O/C = 0.87$ compared to O/C of 0.67 and 0.74 of coarse and medium droplets. Metal chemistry and higher residence time of fine droplets are seemed to be the two most likely reasons for this outcome from as the results of a comprehensive modeling carried out on the observed data indicate. CCN activities of the aerosols from fine droplets are also much higher than that of coarse or medium droplets. Fine droplets also contain light absorbing material as was obvious from their 'yellowish' solution. Source apportionment of fog water organics via PMF (Positive matrix factorization) revealed presence of some very highly oxidized OA inside fog water samples. From PMF results a method for aqSOA estimation is developed and aqSOA was found to be substantially contributing to total SOA. These findings indicate that light fog with large number of fine droplets can process the ambient aerosols more efficiently than very dense fog with larger droplets where scavenging becomes more important. These findings also highlight the need of incorporating fog size resolved chemistry along with metal chemistry into global models for accurately predicting aqSOA formation and contribution to total organic aerosol loading.