

Processing of ambient aerosols during fog events

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Fog is a major processing and removal agent of ambient aerosols. Enhanced secondary organic aerosol (SOA) production has been reported during fog events and there is a growing understanding that SOA formed via this aqueous processing can contribute significantly to ambient aerosol loading apart from traditional gaseous SOA.

The present study which was carried out during winter of 2012-13 in a heavily polluted city of Kanpur situated in Indo-Gangetic plane of India, focuses on identification and quantification of this aqueous processing of the ambient aerosols by natural fog. Although many smog chamber studies have been carried out to understand this aqueous processing and formation of SOA but often these studies were carried out in conditions not representative of complex ambient system and since very few ambient studies were reported till now so there is a need to evaluate what is happening in real ambient condition to properly understand this complex process.

During this campaign from 24 Nov, 2012 to 10 January, 2013, 12 fog events were captured from 20 Dec to 10 January and termed as foggy period while remaining as non foggy period. Foggy period typically showed very high RH and low temperatures than non foggy period with the highest RH (~95%) and lowest temperature (~3°C) occurring during actual fog events. We had deployed an array of instruments during this campaign for real time measurement of aerosol physicochemical properties – High Resolution Time of Flight Aerosol Mass Spectrometer (HR-ToF-AMS) for measurement of size resolved chemical composition of non refractory submicron aerosol, Scanning Mobility Particle Sizer (SMPS) for size distribution of submicron aerosol, Cloud Combination Probe (CCP) to measure fog droplet size distribution and liquid water content (LWC), Cloud Condensation Nuclei counter (CCN) to measure droplet forming potential of ambient aerosols, PM1 sampler and fog water collector, Vaisala RH & T sensor. This is the first time AMS is used in India and to our knowledge AMS has never used so extensively in ambient foggy condition before.

Average aerosol loading during foggy period was $104 \pm 44 \mu\text{g}/\text{m}^3$, much higher than that of non foggy period which was $70 \pm 43 \mu\text{g}/\text{m}^3$, but during actual fog events the loading was lowered to $85 \pm 23 \mu\text{g}/\text{m}^3$ indicating overall aerosol removal by fog. Overall aerosol composition during both the period was dominated by organics which constitutes about 70% of total AMS mass followed by Nitrate, but during foggy period Sulfate was found to be increased many fold.

AMS and SMPS measured aerosol mass compared well. An average aerosol density of 1.3 g/cc was assumed for conversion from SMPS measured number to mass based on the measured aerosol chemical composition of 70% organics and 30% inorganic. Droplet size distribution and LWC revealed that every fog events consists of multiple cycles of dense and thin periods with highest LWC as high as $0.160 \text{ g}/\text{m}^3$ and average droplet size of $25 \pm 4 \mu\text{m}$ during densest periods.

HR-ToF-AMS measured aerosol O:C ratio was also analyzed to understand the real time variation in ambient organic aerosol oxidation level. Noticeable differences for the diurnal average values of O:C ratio between foggy and non foggy period are found. Although in both the period diurnal O:C ratio was higher around noontime typically representing photochemical activity but during foggy period the days were mostly hazy with very little and weak Sunlight, so fog processed aerosol may have retained their already oxidized products. During foggy period, night to early morning O:C ratio was 0.54 ± 0.05 , higher than that of non foggy period 0.43 ± 0.03 , clearly indicating enhanced oxidation activity, despite the fact that oxidized organics are preferentially scavenged by fog. Biomass burning activity doesn't seem to contribute much during any of the period as $m/z = 60, 73$ signals of AMS spectra which are the markers of biomass burning aerosol, remain low and almost the same. To our knowledge this is the first reporting of real time evolution of ambient oxidation level under foggy condition. AMS also showed that mode size of all the species specially organics and sulphate had shifted to a higher diameter during foggy period an indication of aging and mass addition via secondary production.

During foggy period the acidity of ambient aerosol which is measured by H^+ concentration was almost twice as high as non foggy period, mainly due to enhanced sulphate production. Although during actual fog events the acidity was lowered because of enhanced removal of sulphate which normally associated with larger droplets. Total organics from both the period showed very good correlation with aerosol acidity.

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