Supplementary Information

Size-resolved Compositional Analysis and Source Apportionment of Submicron Aerosol during Lockdown Period using HR-ToF-AMS

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Text S1. High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToF-AMS)

The ambient atmospheric aerosol enters through an orifice and the aerodynamic lens (Liu et al., 2007) focuses the particles into a narrow beam of approximately 1 µm diameter using a series of apertures. The lens has 100% transmission efficiency for the particles of diameter 40-600 nm (Jayne et al. 2000; Zhang et al., 2004). At this stage, most of the atmospheric gases are removed from the sampled air through differential pumping. During the measurement mode, the AMS generally alters between two sampling modes, MS mode to monitor the average mass spectrum of particle constituents and in particle time-of-flight (P-ToF) mode to give the size-resolved mass spectra of the constituents. The high-resolution aerosol mass spectra (mass to charge ratio of 4–400) data were stored every 1 min. In the P-ToF mode, a rotating mechanical chopper wheel is positioned to partially block the particle beam to get the mass size distribution. Particles that pass the chopper impact a resistively heated surface (600°C) and get flash vaporized under a high vacuum around 10⁻⁸ torr pressure. The vaporized particles are termed Non-refractory (NR) particles. These particles are then ionized by 70 eV electron impact, and the ions get detected in the detection chamber. The HR-ToF-AMS can quantify the mass concentrations of non-refractory (NR) species, including total organic matter (Org), and sulfate (SO₄²⁻), nitrate (NO₃⁻), ammonium (NH₄⁺), and chloride (Cl⁻) of submicron particles.

The observed high-resolution mass concentration data were acquired by data acquisition software (DAQ) and were analyzed in Igor-Pro (Wavemetrics, OR, USA) using the standard ToF-AMS data analysis program SQUIRREL. The PIKA data analysis program analyzed the size-resolved mass spectrometer data (DeCarlo et al., 2006). A number of calibrations were carried out during the measurement period to make the mass spectra quantitative. Mass-based ionization efficiency (mIE) is carried out using ammonium nitrate (NH₄NO₃) particles to calibrate the signal to mass ratio (m/z) of
the instrument. A detailed explanation of the calibration procedure can be found elsewhere (Ng et al., 2011).

**Text S2. Source apportionment**

HR-ToF-AMS instrument measures mass spectra in the minute resolution that results in two-dimensional matrices of time series (TS, rows) and mass spectra (MS, columns). The Igor PMF evaluation tool (PET) was used to conduct PMF analysis on this dataset and interpret its results. In the process, only organic spectral data at a specific set of m/z values between m/z 10 and m/z 140 were selected. This approach is the most commonly used, and the reasons for selecting the specific set of m/z values have been described in Zhang et al., 2005. Within the PET analysis tool, variables with a signal-to-noise ratio < 0.2 (SNR < 0.2) were down-weighted by a factor of 2. Within the PET, we removed spikes from the dataset, down-weighted mass contribution data at selected weak m/z values by a factor of 2 (SNR < 2) as suggested in Zhang et al., 2005. PMF runs were conducted for one to six factors to explore the solution space using FPEAK and SEED tools (Ulbrich et al., 2009; Ng et al., 2011a). The optimal number of factors was chosen following the criteria that extra factors provided no meaningful information and instead resulted in a splitting of the existing factors. The chosen factors solution is discussed in detail in the results section. In this approach, the concentrations of inorganic species measured by the AMS were used only as external tracers for interpreting organic PMF factors.

**Text S3. Potential Source Contribution Function (PSCF)**

Back trajectories and cluster analysis can provide information about the origins and transport pathways of air parcels but cannot provide information about the contribution of the areas through which the air parcels passed. To better understand the probability and relative significance of the
spatial distribution of potential source regions of pollutants at the receptor site, an additional statistical analytical tool like potential source contribution function (PSCF) was applied (Ashbaugh et al., 1985). By combining the cluster analysis and PSCF model, we can better understand the impacts of regional sectors on PM$_1$ concentrations.

To evaluate the PSCF and do the statistical analysis and visualization of air mass trajectories, a geographic information system (GIS) based software application called TrajStat was used (Wang et al., 2009). Previously calculated back trajectories by HYSPLIT and hourly pollutant concentrations at the receptor site were incorporated into TrajStat for the PSCF analysis. PSCF divides the whole geographic region covered by the backward trajectories into a gridded $i$by $j$ array keeping the measurement location at the center (Gao et al., 1993; Hopke et al., 1995; Polissar et al., 1999; Cheng and Lin, 2001). Mathematically, the PSCF is defined as

$$PSCF = \frac{m_{ij}}{n_{ij}}$$

where $n_{ij}$ is the total number of air masses falling into the $ij$th cell during the study period. The number $m_{ij}$ is the number of endpoints in the $ij$th cell-associated with a trajectory that arrives at the sampling site.
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**Figure Captions:**

**Figure S1:** Location of the measurement site at the Indian Institute of Tropical Meteorology, Pune, India.

**Figure S2:** Temporal variation of mass concentration (µg m⁻³) of Org and inorganic constituents of NR-PM₁ during the pre-lockdown and the lockdown periods.

**Figure S3:** Mass fractions of four factors contributing to the OA components obtained from PMF analysis during the (a) pre-lockdown, (b) lockdown 1 (L1) and (c) lockdown 2 (L2) periods.
Figure S2: Temporal variation of mass concentration (µg m⁻³) of Org and inorganic constituents of NR-PM₁ during (a) pre-lockdown, (b) lockdown 1 (L1) and (c) lockdown 2 (L2) periods.
Figure S3: Mass fractions of four factors (BBOA, HOA, SVOOA, and LVVOOA) contributing to the OA components obtained from PMF analysis during the (a) pre-lockdown, (b) lockdown 1 (L1) and (c) lockdown 2 (L2) periods.