Understanding coupling between aerosol chemistry and Indian summer monsoon: An Indian Perspective

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Some Intriguing research questions

1. Aerosol microphysical and chemical processes: Impact on *cloud condensation nucleation*, *optical properties* and *rainwater chemistry*  
   (focus on carbonaceous aerosols including black carbon and dust)

2. Aerosol radiative impact on surface energy budget over the Himalaya
   ➢ Radiative *(forcing)* impact of aerosols on atmospheric stability and surface energy balance
Carbonaceous aerosols CA)

- **Elemental carbon (EC):**
  - Also c/a graphitic carbon (GC), soot, black carbon (BC), light absorbing carbon (LAC)
  - 2-10% of aerosol mass
  - mainly absorbing in nature, thus, have +ve radiative forcing (warming)

- **Organic Carbon (OC):**
  - ~10-30% of aerosol mass
  - mostly scattering, thus, have -ve radiative forcing (cooling)
  - Significant amount is water-soluble organic carbon (WSOC): ~30-60% of OC

**Major Sources:** (a) Primary emission
- Biomass burning emission
- Fossil-fuel emission

(b) Secondary organic aerosols (SOA)

Volatile organic compounds (VOCs) + OH, O₃, NO₃ chemistry → Oxygenated compounds, diacids and nitrates
Carbonaceous aerosols: Indian scenario

**INDOEX: INDidian Ocean EXperiment** (Lelieveld et al., Science, 2001)

Jan-Mar 1999 (winter monsoon) over the Indian Ocean (IO)

**Observations/Results:**

- High level of pollution and aerosol loading (up to 20 µg m$^{-3}$) over IO
- Chemical composition: large contribution from anthropogenic sources
- $\text{SO}_4^{2-}: 32\%; \text{Organics: 26}\%; \text{BC:14}\%, \text{mineral aerosols: 10}\% \text{ (fine-mode)}$

**Venkataraman et al., Science 2005**

Measured emissions from the combustion of biofuels: 1$^{\text{st}}$ measurements

Biofuels combustion as the largest source of BC emissions in India

**Sources of Black Carbon**

Biofuel or Fossil Fuel?

- Novakov et al., GRL, 2000: Fossil Fuel dominant
- Venkataraman et al., Science, 2005: a couple of papers from their group
- Gustafsson et al., Science, 2009;
  Srinivas, Ram JGR 2016, 17: $^{14}$C data
- Our studies: Biofuel dominant
  $K^+, \text{OC/EC}, K^+/\text{OC}, 210^{\text{Po}}$

**BC aerosols and radiative forcing:**

many studies over India

OC~15-45% of PM$_{2.5}$ mass (as our data suggest): Very little information on SOA
Study locations

- High aerosol loading from anthropogenic emissions and secondary formation
- Hot spot of CA and located within the source region: BL dynamics plays imp. role in trapping pollutants
- Foggy-hazy weather formation during wintertime

Manora Peak/Nainital (2005-2008):
- High-altitude, less influenced by anthropogenic emission, dominated by mineral aerosols
- To study long-range transport of aerosols and mineral dust chemistry

In addition, samples collected from Mt Abu (May 05-Feb 06); Hisar (HSR), Allahabad (ALB) and Manora Peak (MNP), all during Dec-2004, were analyzed and have been used for inter-comparison of the data

MODIS image: Dec. 2004
Source: MODIS/NOAA
Measurements in India: OC-EC concentrations & ratios at urban vs high-altitude site

**Sampling sites:**
- HSR
- KNP
- ALB
- Mt Abu
- MNP

**OC (µg C m⁻³):**
- High-altitude
- Urban
- Ocean

**EC (µg C m⁻³):**
- High-altitude
- Urban
- Ocean

**WSOC/OC ratio:**
- High-altitude
- Urban

**OC/EC ratio:**
- High-altitude
- Urban

**Sampling sites**

A case study: Fog-haze formation over IGP during wintertime

Oct 2008, Kanpur
- Emission sources and meteorological conditions
- High Relative humidity (RH)
- Shallower Boundary layer height

Chemical composition

Chemical analysis suggest that ~80% of aerosol mass is composed of carbonaceous and inorganic aerosols.

TCA (=1.6 × OC + EC) ~60% and water-soluble inorganic species (WSIS) ~20% of aerosol mass

Ram and Sarin, Atmos. Environ. (2011)

Nature, 2014
Average (OC/EC)_{Day}=6.7; (OC/EC)_{Night}=5.6; Probably indistinguishable

Average ratios: (WSOC/OC)_{Day}=0.66; (WSOC/OC)_{Night}=0.46, (WSOC/OC)_{winter}=0.30-0.40 (representative of primary emissions in IGP)

Relatively higher WSOC/OC ratios suggest enhanced SOA formation during daytime.

Over high-altitude, higher WSOC/OC ratios are due to chemical aging and SOA formation during the transport.

Ram and Sarin, Atmos. Environ. (2011)
Enhancement in SIA concentrations during fog-haze events

Ram and Sarin et al, AAQR (2012)
The IGP is characterized by intense biomass burning activities during winter and transport of dust in summer and thus, strong changes in composition and optical properties.

Question: Do varying composition and secondary formation have any impact on microphysical and optical properties of aerosols?
Enhancement in absorption due to mixing processes

Radiative impact of black carbon (BC) aerosols strongly depends on the accurate measurements of its mass and absorption coefficient.

- Varying amount of absorption for same quantity of BC mass each year
- Due to coating of other non-absorbing hygroscopic materials from secondary formation (organics as well as inorganics) over BC
The average mass absorption efficiency of EC ($\sigma_{\text{abs}}$) during daytime (11.7±2.5 m$^2$g$^{-1}$) is about factor of two higher than that during nighttime (5.7±1.3 m$^2$g$^{-1}$).

Relatively higher $\sigma_{\text{abs}}$ values obtained during daytime can be attributed to formation of internal mixture produced by coating of secondary aerosols.

Changes in the optical properties at Kanpur

<table>
<thead>
<tr>
<th>Oct 2008</th>
<th>CCN/CN</th>
<th>$b_{\text{abs-678 nm}}$</th>
<th>$\sigma_{\text{abs}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(@0.5%)</td>
<td>Mm$^{-1}$</td>
<td>m$^2$g$^{-1}$</td>
<td></td>
</tr>
<tr>
<td>Day</td>
<td>0.17±0.05</td>
<td>72.1±16.9</td>
<td>11.7±2.5</td>
</tr>
<tr>
<td>Night</td>
<td>0.18±0.06</td>
<td>63.9±9.9</td>
<td>5.7±1.3</td>
</tr>
</tbody>
</table>

Ram et al, AE, 2014
CCN exhibit fairly good correlation with AIS (Anthropogenic inorganic aerosols; WSOC + NO$_3^-$ + SO$_4^{2-}$)

Negative correlation b/w Critical diameter and WSIS/EC ratio

Critical diameter is defined as the diameter above which all particles will be activating

The CCN/CN ratios are relatively lower (range: 0.11—0.33) vs global values

Suppressed activation and hygroscopic growth in highly polluted environment of the IGP

Ram, Tripathi, Sarin et al, AE, 2014
Dust events over Manora Peak and its implication on rain water chemistry

Impact on Rain water composition (Bisht, Ram et al, ESPR, 2017):

- A total of 55 rainwater samples were collected during June-Sept 2012
- 2 in pre-monsoon and 53 in post-monsoon
- Average ionic concentration is ~3 times higher during pre-monsoon ($986 \pm 101 \, \mu\text{eq/l}$; pH=$6.25 \pm 0.50$) compared to ($373 \pm 37 \, \mu\text{eq/l};$ pH=$5.6 \pm 0.30$) during pre-monsoon, mainly due to the presence of mineral aerosols in pre-monsoon.
AOD at Manora Peak

Ram et al, ACP, 2010
Variability of optical properties at Manora Peak

Dust events

Supported by:

5-day air mass back-trajectories analysis

& increase in Ca$^{2+}$ concentration

(Srivastava, Ram, et al. STE, 2015)
Variability in RF and heating rates at Manora Peak

(Srivastava, Ram et al. STE, 2015)
Conclusions

- Carbonaceous aerosols and dust plays an important role in the modification of chemical composition and optical properties of aerosols as well as rainwater.

- Elevated RF and heating rate during summer in Himalayas are due to high BC and dust concentrations which may affect atmospheric instability.

- Absorbing nature of dust/ brown carbon and its quantification is important.

- We are studying isotopic characteristics of rainwater ($^{18}$O and D) to get more insight on source and transport pattern of moisture/water.
Thank you all for your patience listening

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