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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)

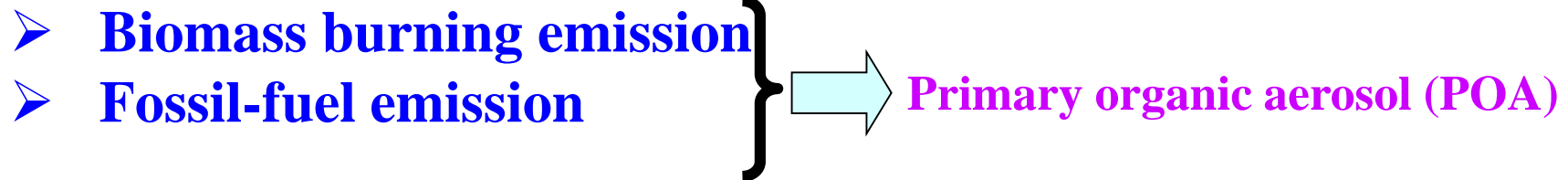
❖ Elemental carbon (EC):

- 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**)

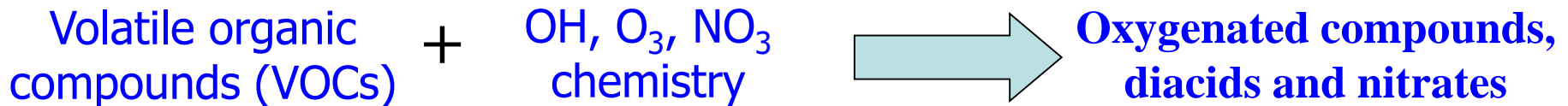
❖ Organic Carbon (OC):

- ❖ ~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



(b) Secondary organic aerosols (SOA)



Carbonaceous aerosols: Indian scenario

INDOEX: INDian 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 $20 \mu\text{g m}^{-3}$) over IO
- ❖ Chemical composition: large contribution from **anthropogenic sources**
- ❖ SO_4^{2-} : 32%; Organics: 26%; BC:14%, mineral aerosols: 10% (fine-mode)

Venkataraman et al., Science 2005

Measured emissions from the combustion of biofuels: 1st measurements

Biofuels combustion as the largest source of BC emissions in India

BC aerosols and radiative forcing: many studies over India
OC~15-45% of $\text{PM}_{2.5}$ mass (as our data suggest): Very little information on SOA

- **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^+ , OC/EC, K^+ /OC, ^{210}Po

Kanpur (2007- 2009):

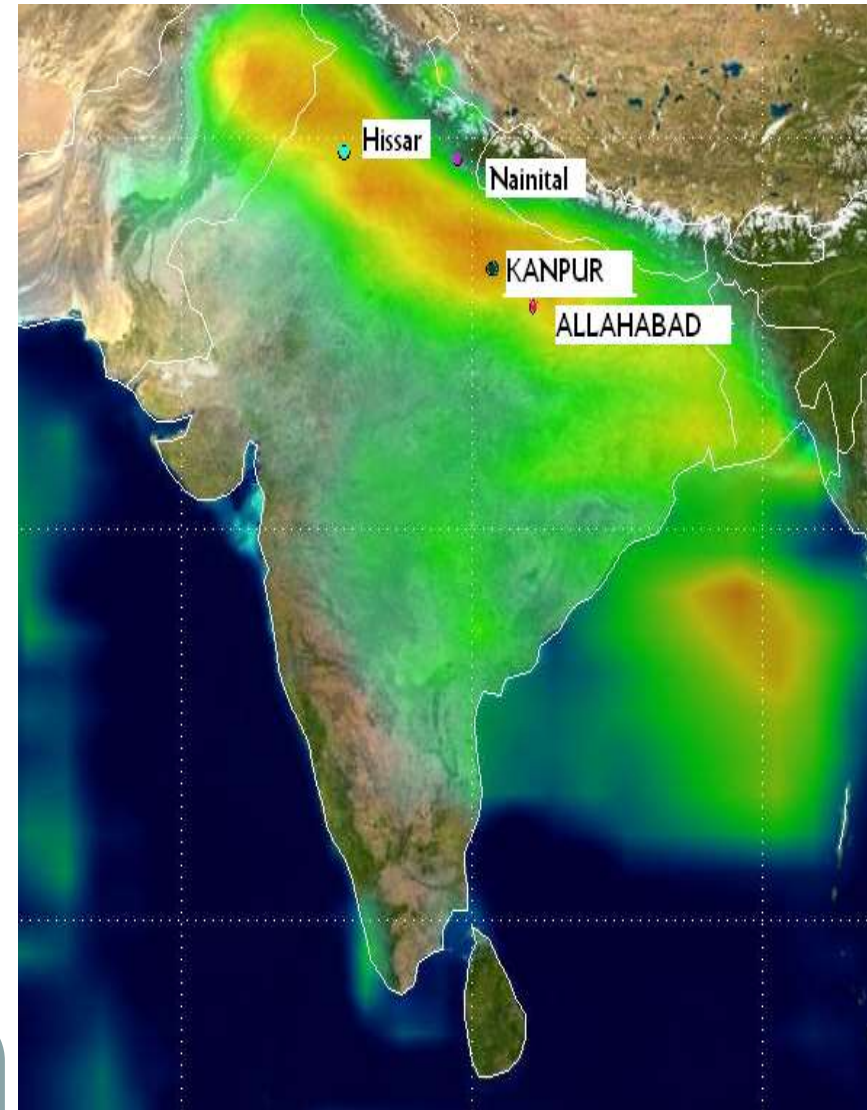
- High aerosol loading from anthropogenic emissions and secondary formation
- Hot spot of CA and located within the source region: BL dynamics plays imp. role in trapping pollutants
- Foggy-hazy weather formation during wintertime

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

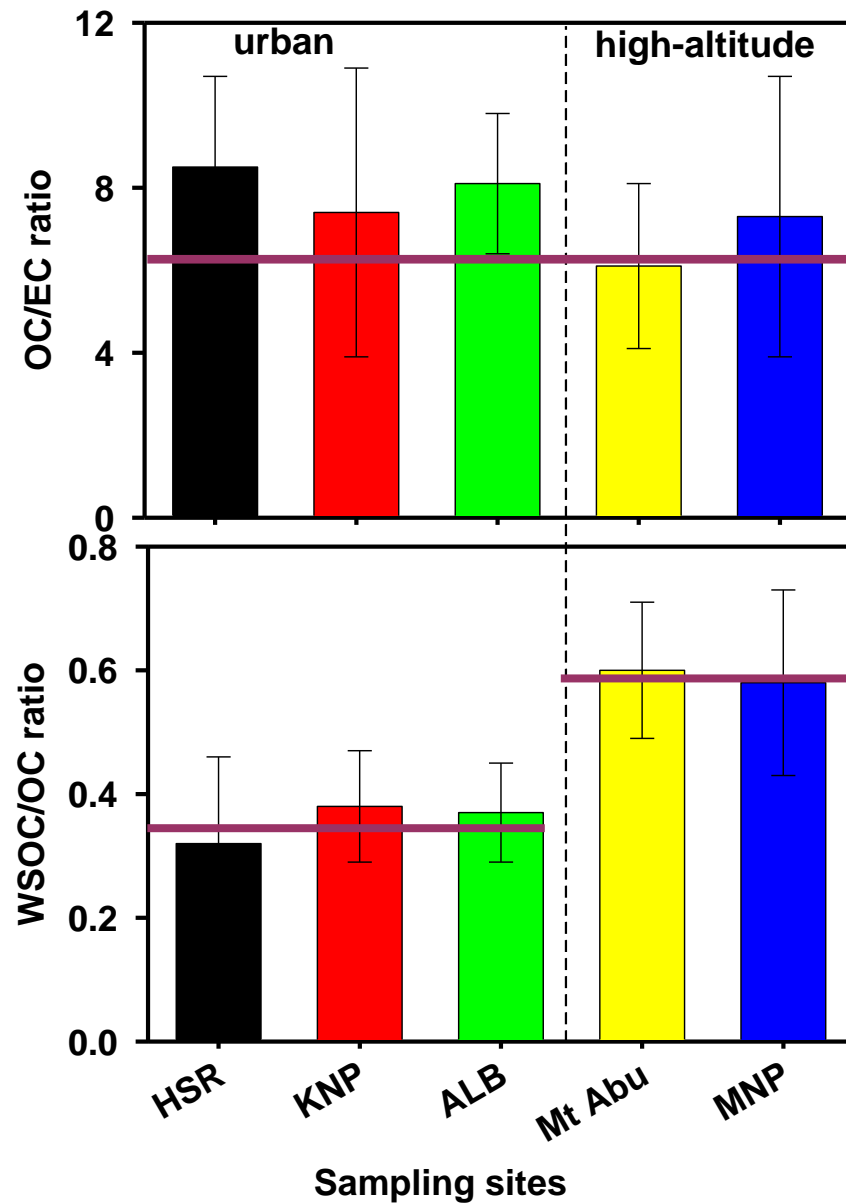
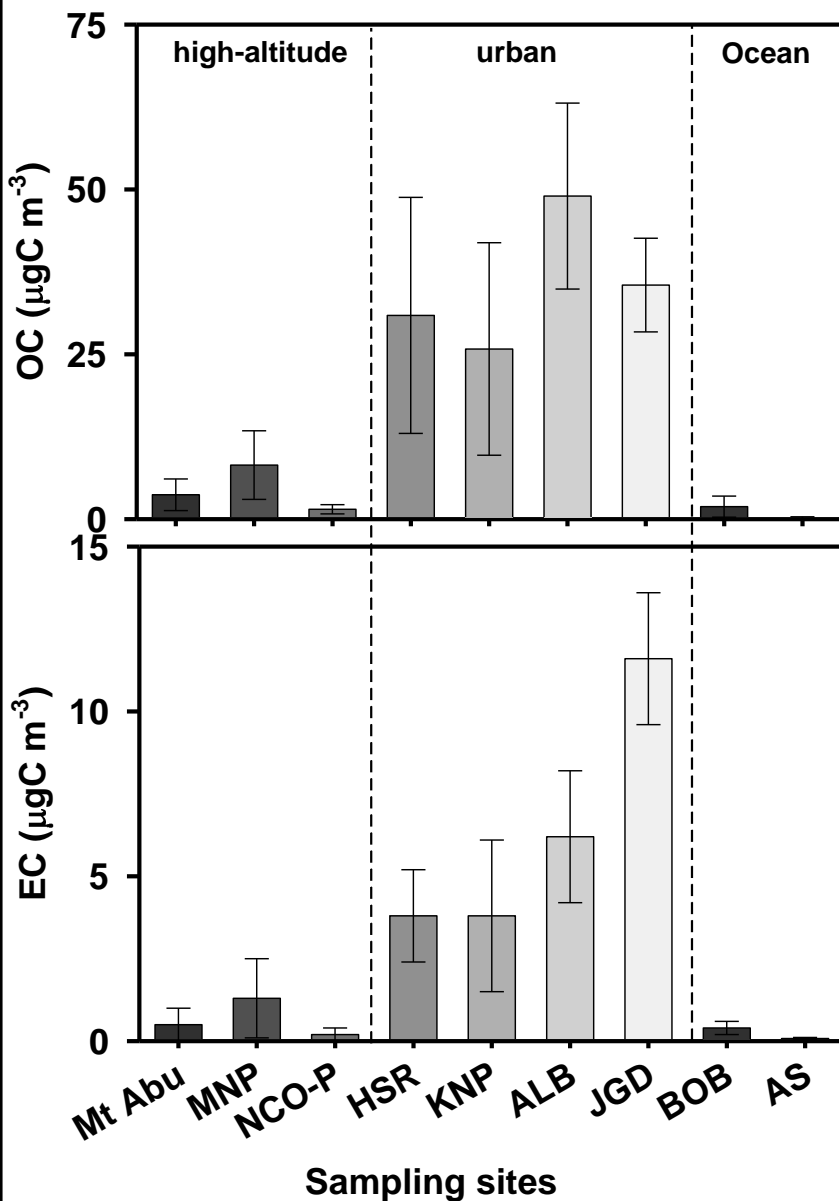
In addition, samples collected from **Mt Abu** (May 05-Feb 06); **Hisar (HSR)**, **Allahabad (ALB)** and **Manora Peak (MNP)**, all during Dec-2004), were analyzed and have been used for inter-comparison of the data

Study locations



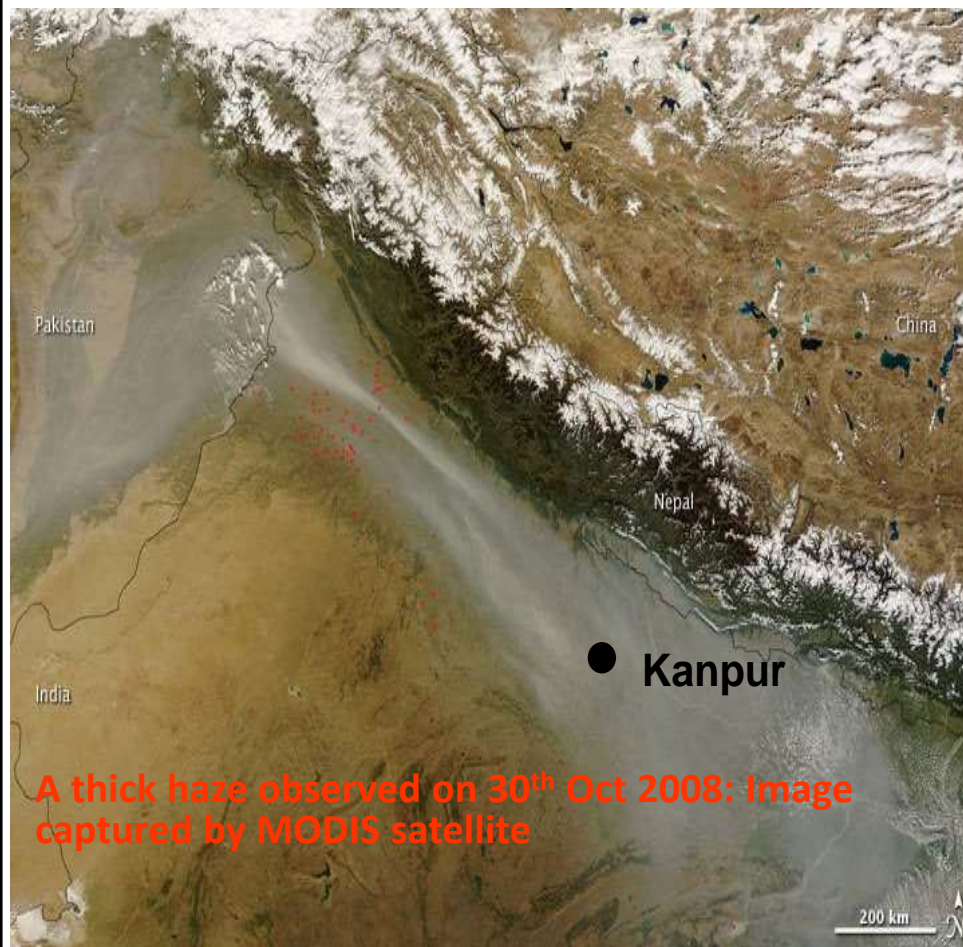
MODIS image: Dec. 2004
Source: MODIS/NOAA

Measurements in India: OC-EC concentrations & ratios at urban vs high-altitude site



AE (2008), JAS (2010), ACP (2010), JGR (2010)

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



A thick haze observed on 30th Oct 2008: Image captured by MODIS satellite

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 ($=1.6 \times \text{OC} + \text{EC}$) ~60% and water-soluble inorganic species (WSIS) ~20% of aerosol mass

Ram and Sarin, *Atmos. Environ.* (2011)

LETTER

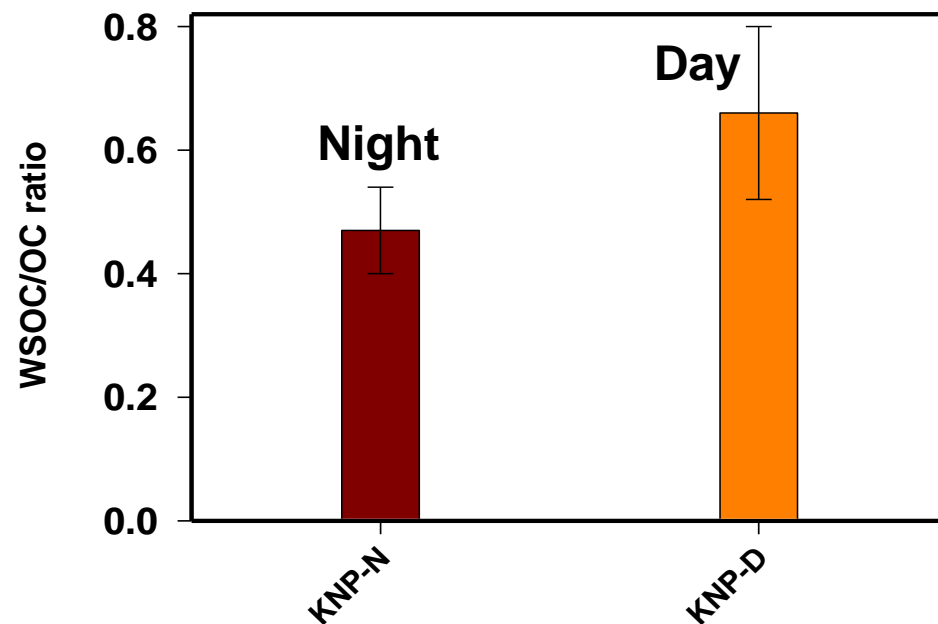
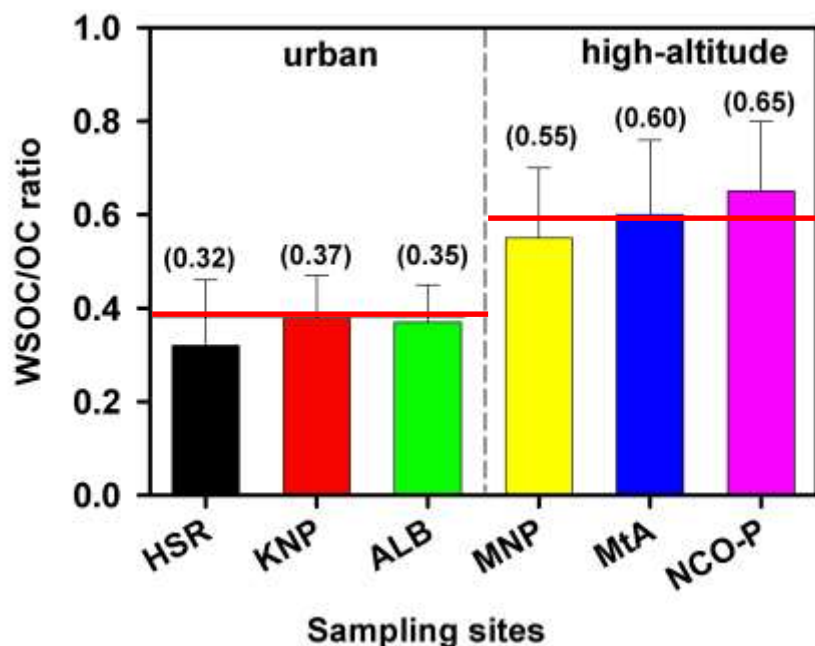
Nature, 2014

doi:10.1038/nature13774

High secondary aerosol contribution to particulate pollution during haze events in China

Ru-Jin Huang^{1,2*}, Yanlin Zhang^{3,4}, Carlo Bozzetti¹, Kin-Fai Ho⁵, Jun-Ji Cao², Yongming Han², Kaspar R. Daellenbach¹,

Evidence of Secondary OA formation over IGP

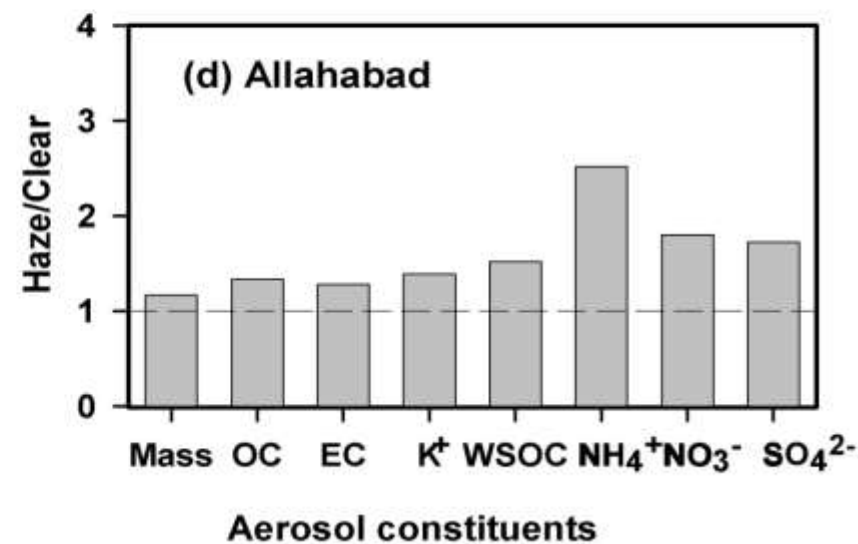
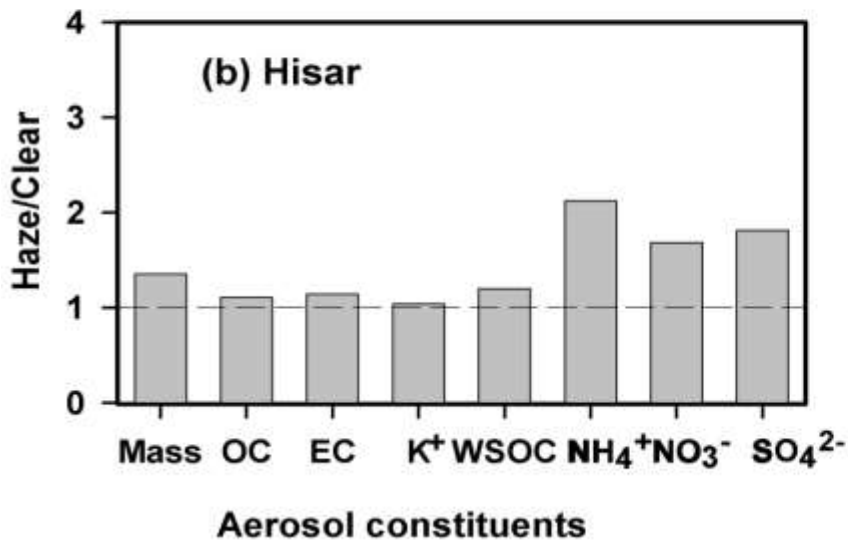
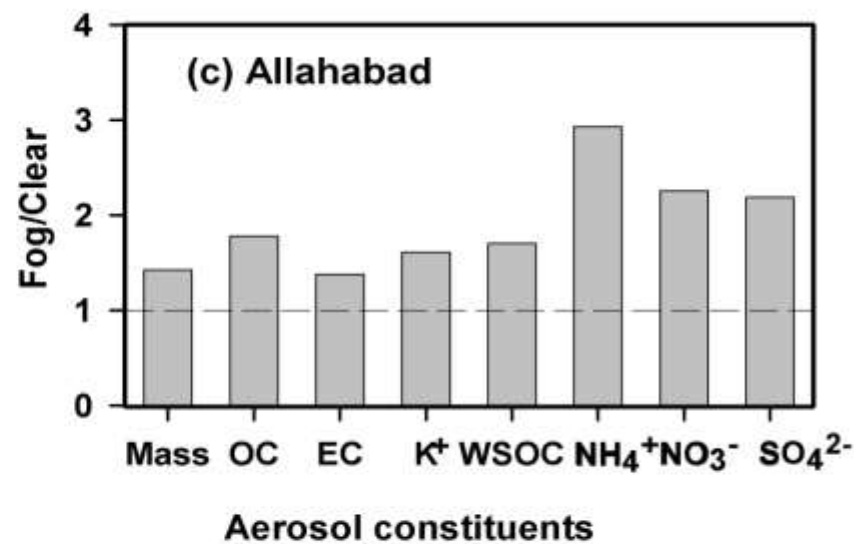
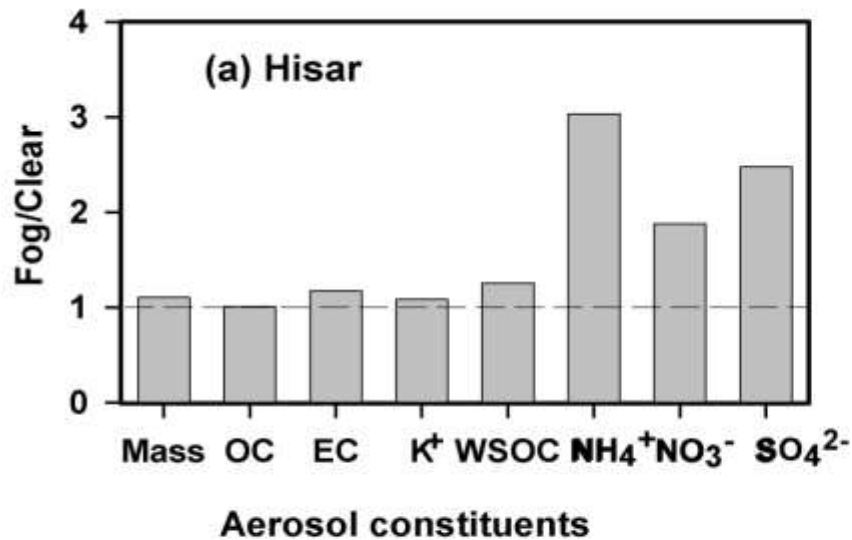


- Average $(OC/EC)_{Day}=6.7$; $(OC/EC)_{Night}=5.6$; Probably indistinguishable
- Average ratios: $(WSOC/OC)_{Day}=0.66$; $(WSOC/OC)_{Night}=0.46$, $(WSOC/OC)_{winter}=0.30-0.40$ (representative of primary emissions in IGP)
- Relatively higher WSOC/OC ratios suggest enhanced SOA formation during daytime.
- 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)

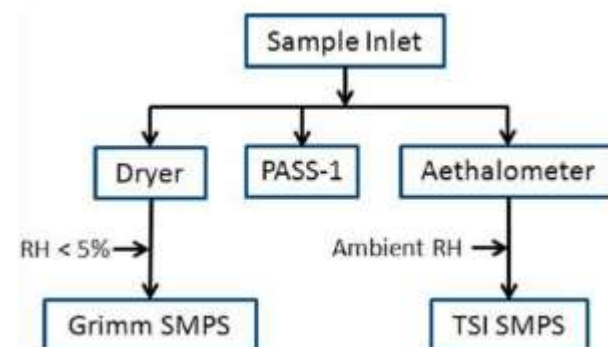
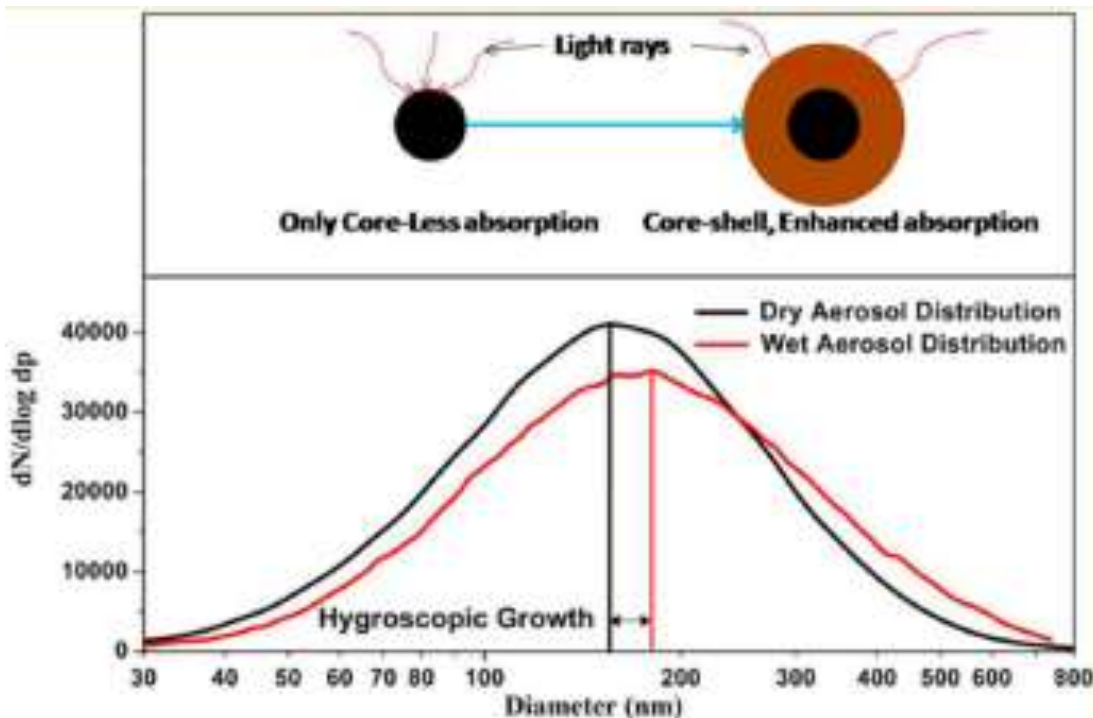


The IGP is characterized by intense biomass burning activities during winter and transport of dust in summer and thus, strong changes in composition and optical properties

Question: Do varying composition and secondary formation have any impact on microphysical and optical properties of aerosols?

Enhancement in absorption due to mixing processes

Shamjad, Tripathi....**Ram**, EST (2012)

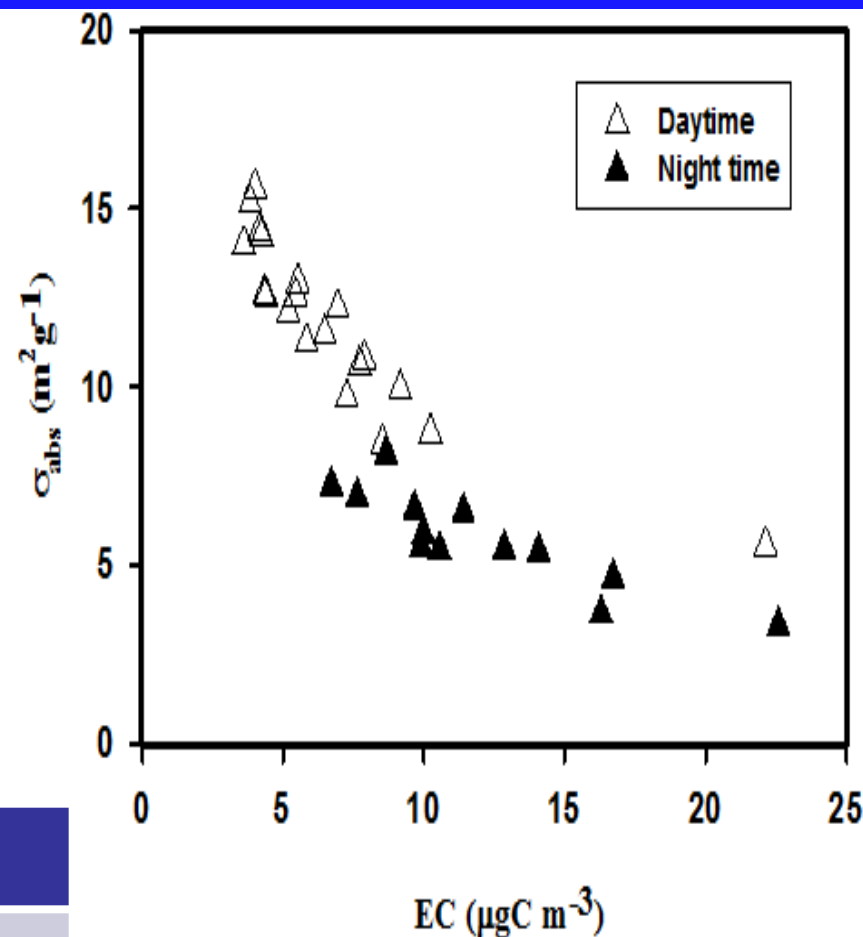


- Radiative impact of black carbon (BC) aerosols strongly depends on the accurate measurements of its mass and absorption coefficient
- Varying amount of absorption for same quantity of BC mass each year
- Due to coating of other non-absorbing hygroscopic materials from secondary formation (organics as well as inorganics) over BC

Changes in the optical properties at Kanpur

The average mass absorption efficiency of EC (σ_{abs}) during daytime ($11.7 \pm 2.5 \text{ m}^2\text{g}^{-1}$) is about factor of two higher than that during nighttime ($5.7 \pm 1.3 \text{ m}^2\text{g}^{-1}$).

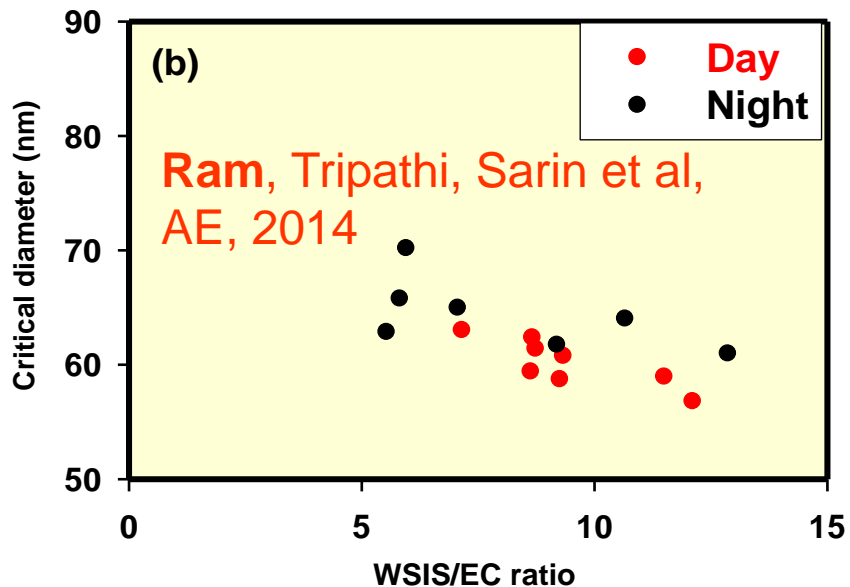
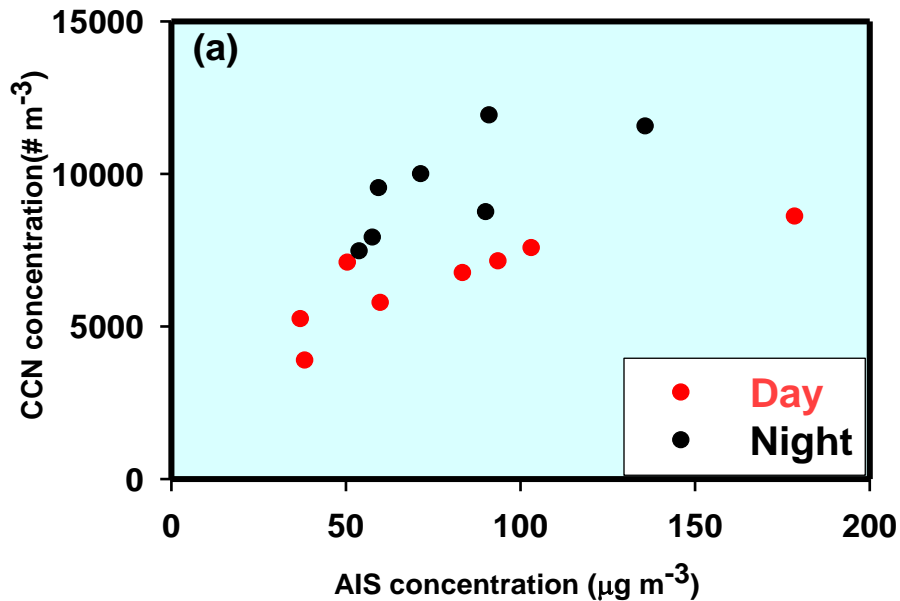
Relatively higher σ_{abs} values obtained during daytime can be attributed to formation of internal mixture produced by coating of secondary aerosols.



Ram et al, AE, 2014

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

Microphysical properties at Kanpur



CCN exhibit fairly good correlation with AIS (Anthropogenic inorganic aerosols; WSOC + NO₃⁻ + SO₄²⁻)

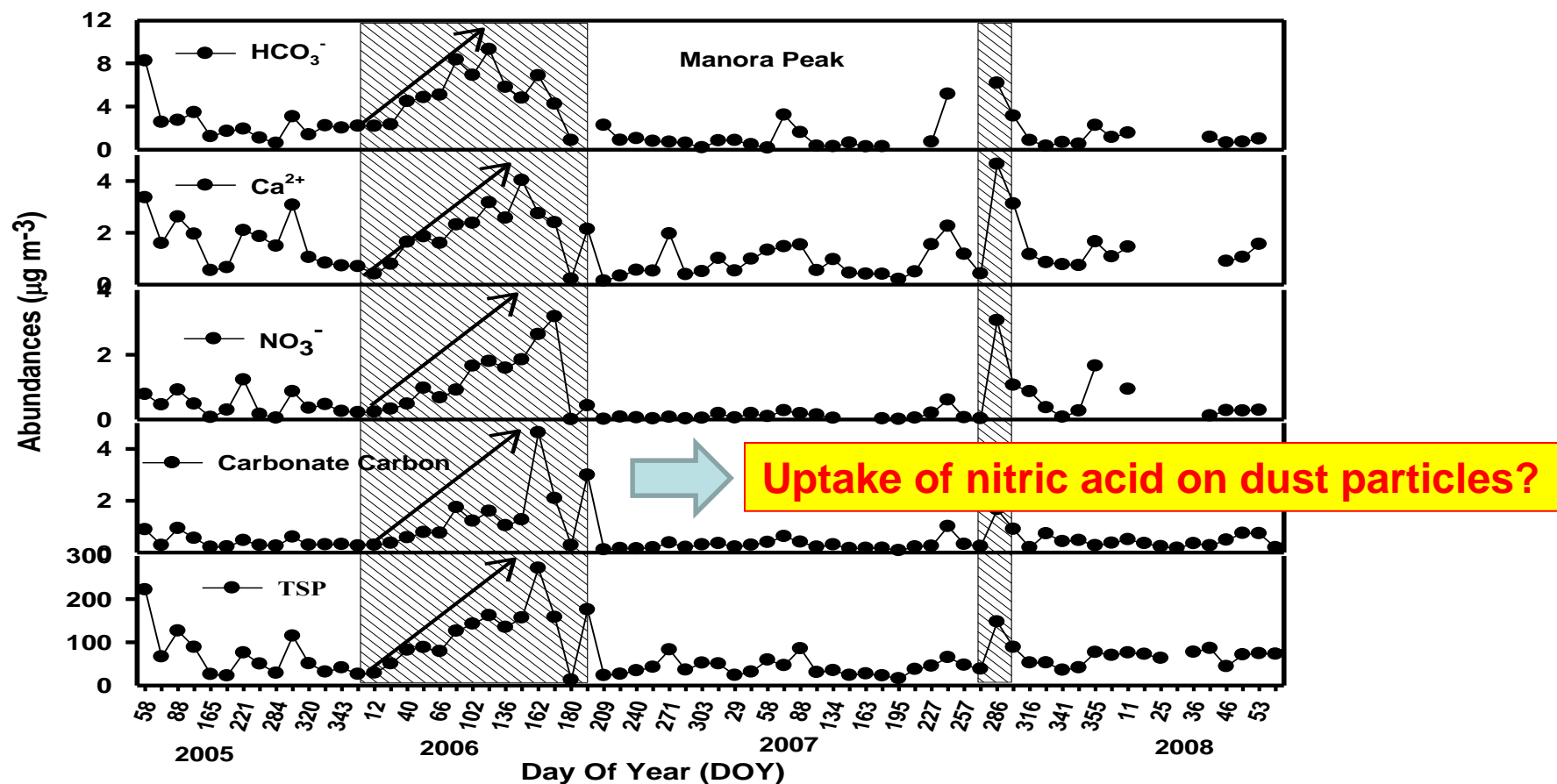
Negative correlation b/w Critical diameter and WSIS/EC ratio

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

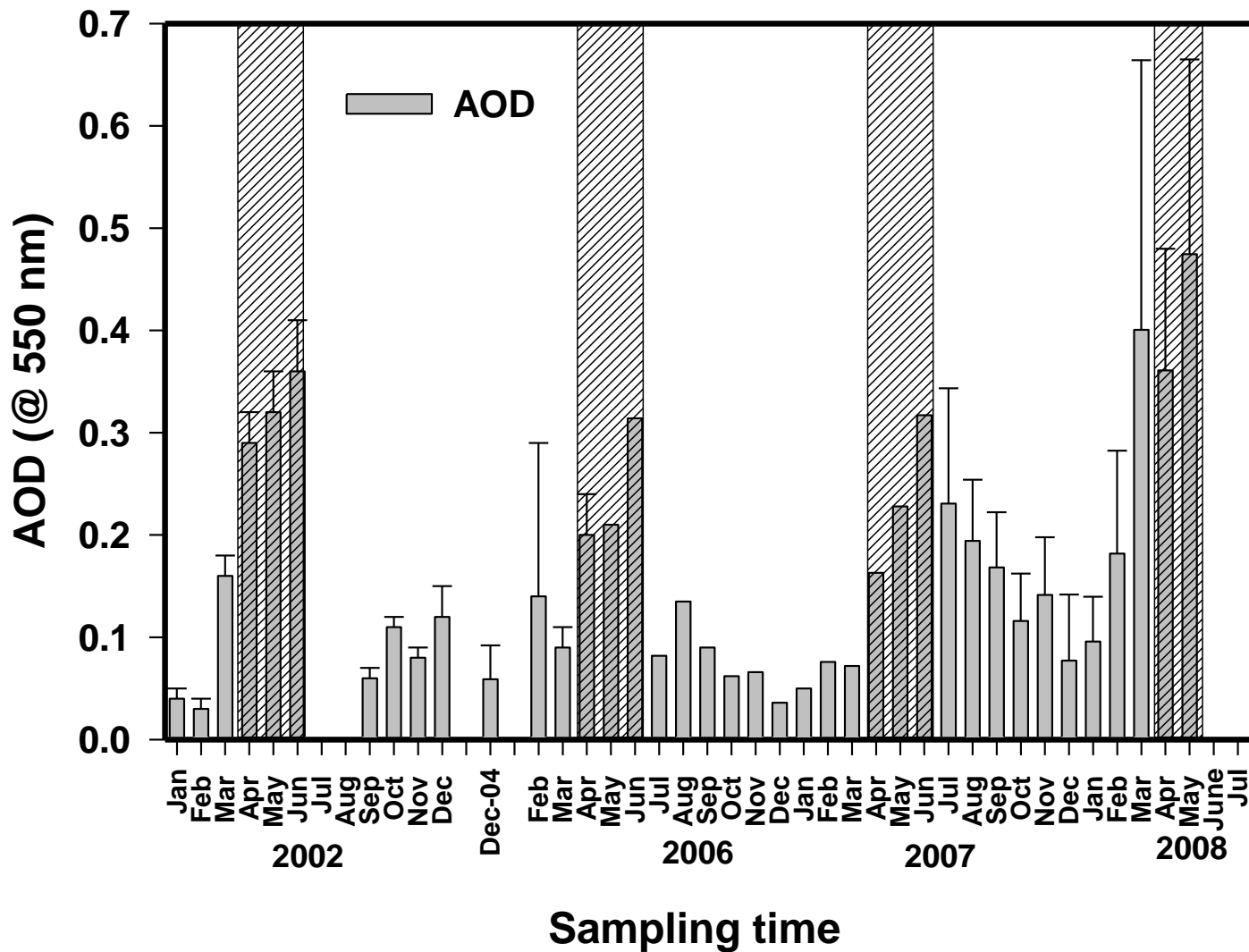
Suppressed activation and hygroscopic growth in highly polluted environment of the IGP

Dust events over Manora Peak and its implication on rain water chemistry

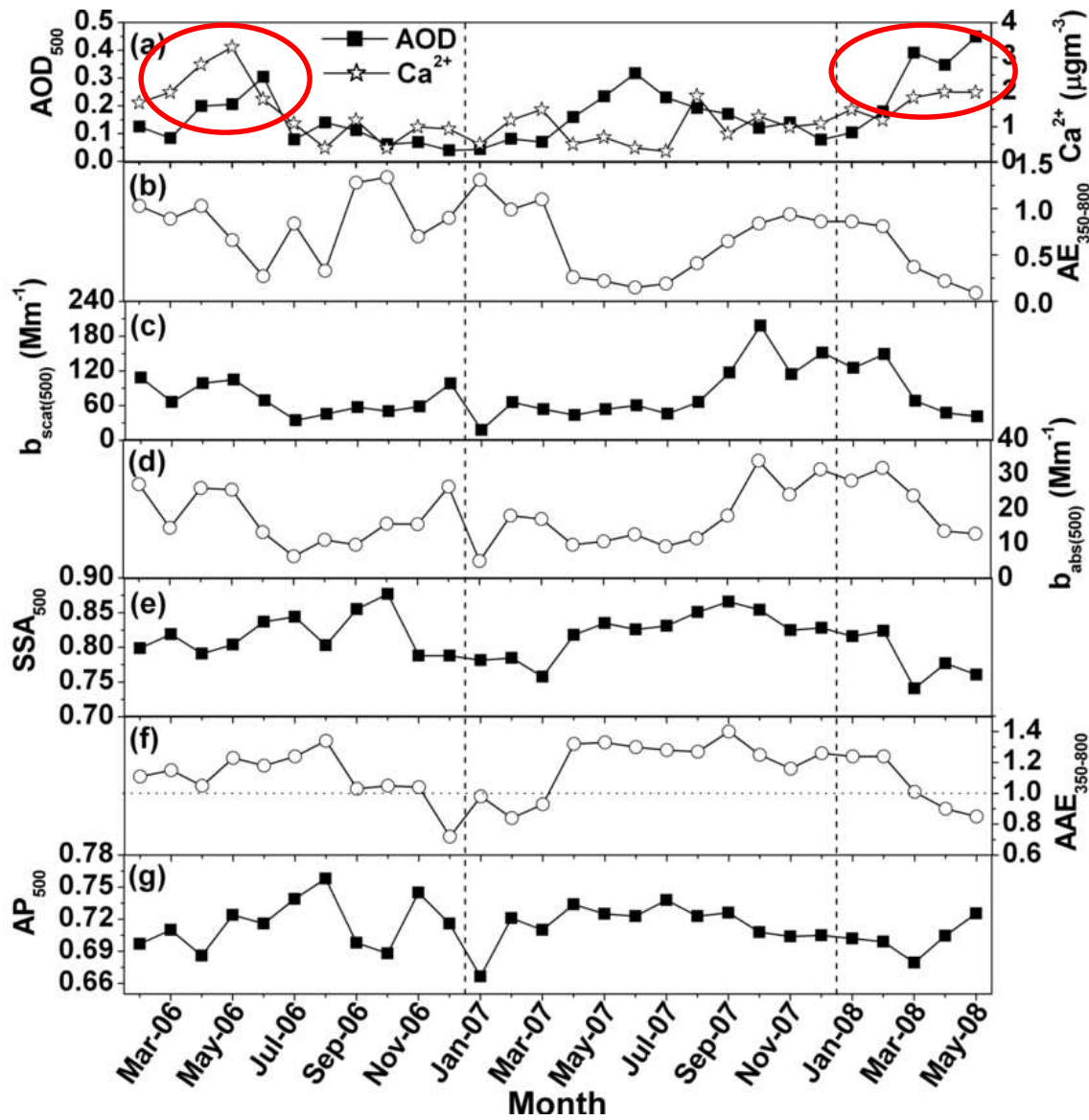


- **Impact on Rain water composition (Bisht, Ram et al, ESPR, 2017):**
- A total of 55 rainwater samples were collected during June-Sept 2012
- 2 in pre-monsoon and 53 in post-monsoon
- Average ionic concentration is ~ 3 times higher during pre-monsoon ($986 \pm 101 \mu\text{eq/l}$; $\text{pH}=6.25 \pm 0.50$) compared to ($373 \pm 37 \mu\text{eq/l}$; $\text{pH}=5.6 \pm 0.30$) during post-monsoon, mainly due to the presence of mineral aerosols in pre-monsoon

AOD at Manora Peak



Variability of optical properties at Manora Peak



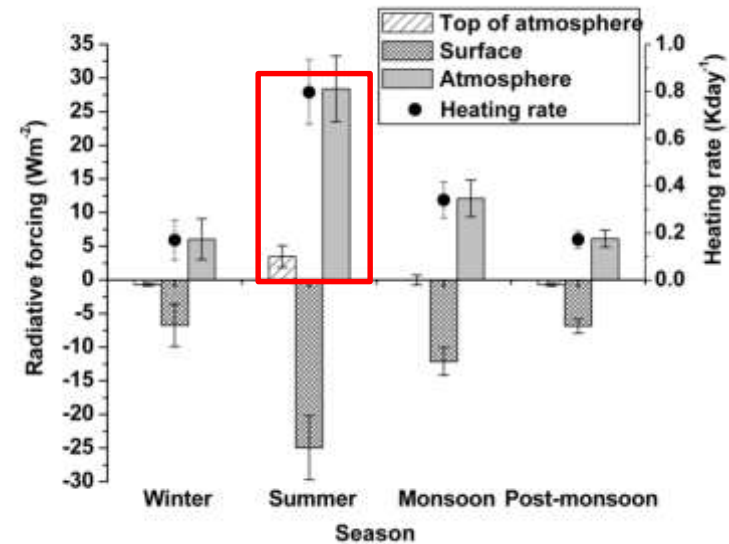
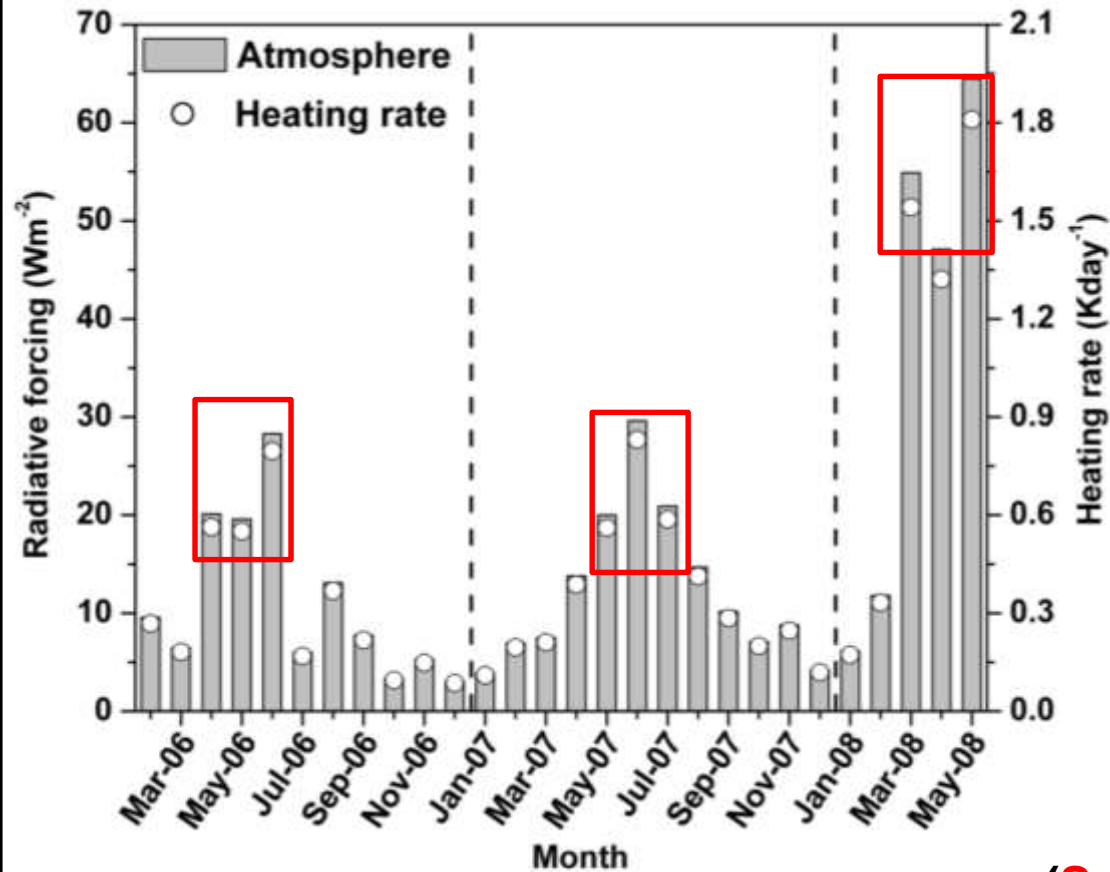
Dust events

Supported by:

5-day air mass back-trajectories analysis

& increase in Ca^{2+} concentration

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) get more insight on source and transport pattern of moisture/water

Thank you all for your patience listening

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