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Closure between measured and predicted Cloud Condensation Nuclei concentration at Kanpur, Indo-Gangetic Basin

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Introduction

- Atmospheric particles play a significant role in the Earth climate system indirectly by acting as cloud condensation nuclei (CCN) depending on their size and chemical composition.
- Sensitivity of CCN closure to various parameters like, size distribution, chemical composition or hygroscopicity in CCN activity considering bulk/ size-resolved chemical composition from filters/AMS and different mixing states has been studied.
- Several closure studies have been performed in the past but a very few studies have predicted CCN concentration within uncertainty levels (± 20%).

Objectives

- **To examine the effect of organics and their hygroscopicity on CCN activity of ambient aerosols.**
- **To study the sensitivity of CCN closure to aerosol chemical composition, mixing state and soluble organic fraction.**
- **To explore the effect of degree of oxygenation on hygroscopicity of organics.**

Methodology

- Measurements were conducted from Nov 7- 27, 2012 at the IIT Kanpur (80° 20'E, 26° 26'N), India using set of instruments given in Table 1.
- Chemical composition from AMS is used in Köhler theory to calculate critical diameter, Dc, assuming different mixing states (external/ internal). Soluble organic fraction (OOA, LVOOA, SVOOA, BBOA and their combinations) obtained from AMS-PMF analysis are used.
- CCN derived hygroscopicity parameter is calculated using κ-Köhler theory (Petters and Kreidenweis, 2007) and its correlation with O:C ratio is studied.

Table 1 : List of instruments used and the measured property

Instruments	Measured property
SMPS	Size distribution (14.6 <D< 685 nm)
CPC	Total particle number concentration (#/cc)
Aerodyne HR-ToF-AMS	Chemical composition (bulk & size-resolved)
CCNc	CCN concentration (#/cc) (SS=0.2-1%)

Results and Discussion

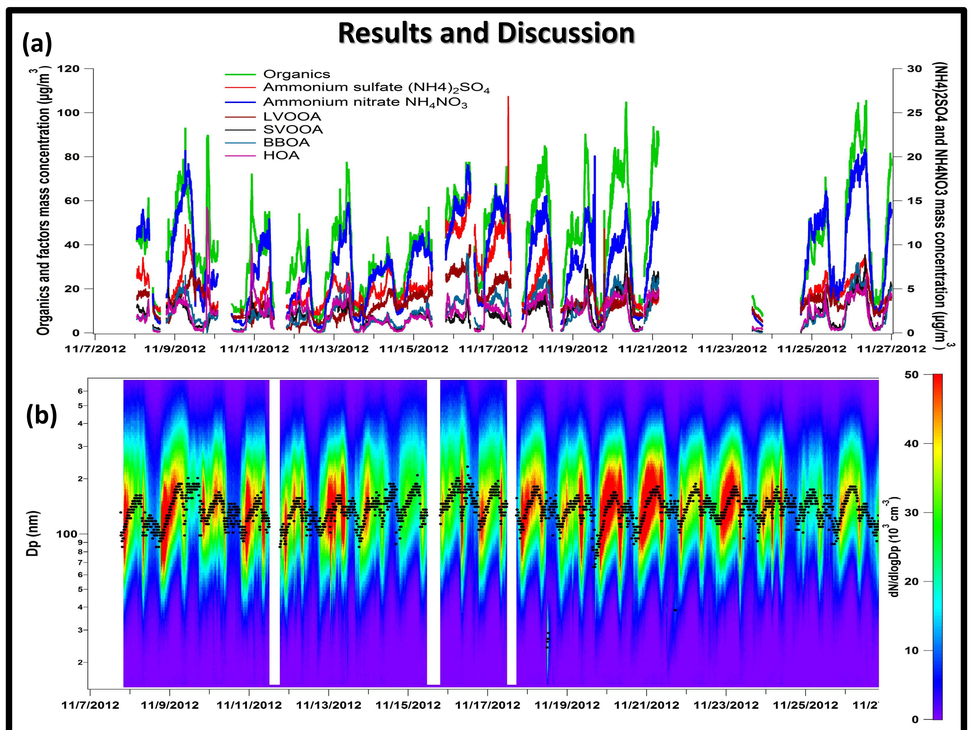

Figure1: Time series of (a) bulk chemical composition from HR-ToF-AMS and AMS-PMF derived factors (LVOOA. SVOOA, BBOA and HOA; collection efficiency of 1) and (b) SMPS size distribution during the measurement period. Most of the time, volume fraction of organics is greater than 50% with maxima at night time. The concentration of small sized particles present in the morning hours (around 7 -9 am) and evening time is also consistent with HOA. However, the dilution by rising boundary layer, lower emission rates and slightly more mixing in the evening hours suppressed the total aerosol concentration. Night time aerosols were not completely neutralized by the available NH_4^+ concentration leaving them acidic . LVOOA is the most dominating fraction of total organics.

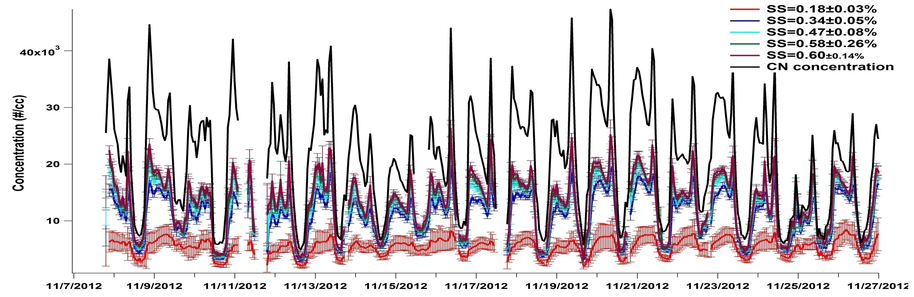

Figure2: Times series of CCN and CN concentration (#/cc) at 5 different depleted SS (0.18-0.60%). Maximum activation fraction (CCN/CN) observed at daytime due to enhanced photochemical activity is in accordance with O:C ratio. Also, less HOA and BC fraction, and dilution by rising boundary layer, are the major causes in lower CCN and CN concentration. But higher activated fraction is observed because of the condensation of more hygroscopic species on the existing aerosols. Few exceptions have been observed at the end of the sampling period when fraction of SVOOA is comparable to LVOOA.

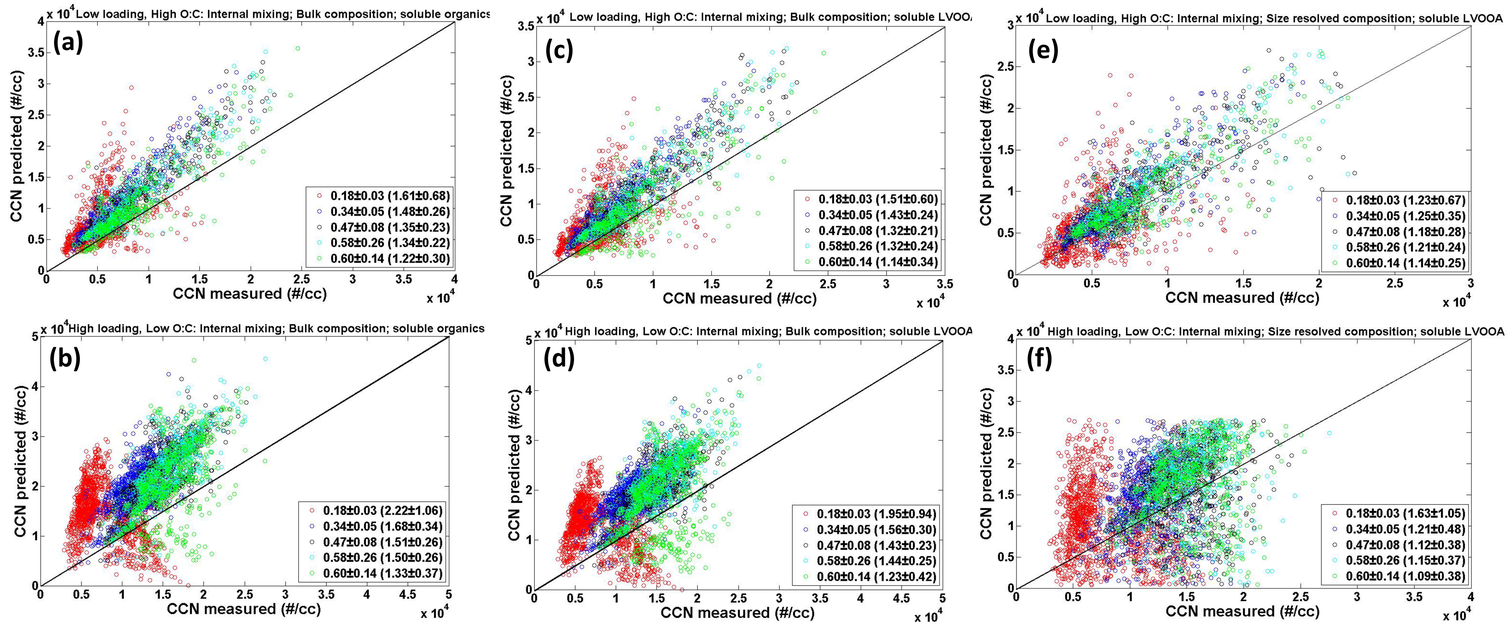

Figure3: Predicted Vs measured CCN concentration at SS=0.18-0.60%. Different models were considered on the basis of assumptions involved. The assumptions are as follows: Internally mixed aerosols; bulk/ size resolved chemical composition; solubility of organics, surface tension same as of pure water. Real-time volume fraction of LVOOA is considered as soluble organics . It has been observed that closure ratio improves as the SS increases showing less sensitivity to chemical composition. Closure ratio improved by 8-10% by inclusion of LVOOA as soluble organics (Fig a, c). The scenario of externally mixed aerosols further improved the closure by 10-25% (not shown here). Further, size resolved chemical composition of aerosols achieved the closure ratio of 1.14-1.25 at SS=0.18-0.60% for low loading and high O:C ratio case. However, in high loading and low O:C ratio , there is still an over-prediction of 9-63% (SS=0.18-0.60%) which can further be improved by application of size resolved mixing state.

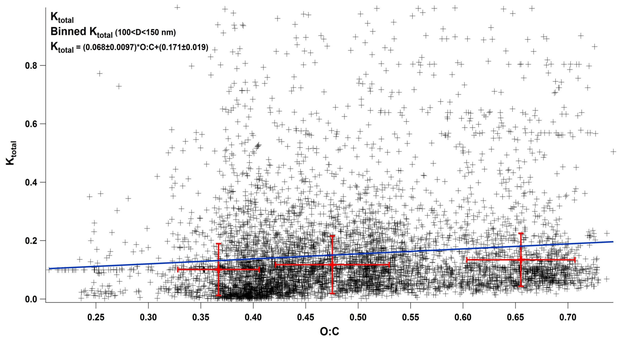

Figure4: Correlation between hygroscopicity parameter, κ (CCN derived) and O:C ratio (AMS) shows small increase in hygroscopicity with the degree of oxygenation. O:C ratio ranges from 0.2-0.8 with maxima at 1400-1500 hrs. Further, size binned data showed increasing trend in κ with O:C for particles only in size range of 100-150 nm. It is possible that the for other size bins, aerosol composition may be changing in a way that O:C ratio alone is not able to capture the changing trend.

Key Results

- Higher externally mixed HOA fractions during high loading and low O:C ratio were present (0.24±0.08) compared to opposite conditions (0.11±0.06) that caused less effective condensation of hygroscopic species which resulted into higher CCN over-prediction.
- LVOOA is the best representative of soluble organics and can be used in CCN closure studies.
- Overall, κ is slightly dependent on O:C in the range 0.2< O/C <0.8.

References

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