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Editor

Dr. Sagnik Dey, IIT Delhi

Editorial Office:

Centre for Atmospheric Sciences, Indian Institute of Technology Delhi

Phone: +91-11-2659-1315/Fax: +91-11-2659-1386

E-mail: sagnik@cas.iitd.ac.in

Website: www.iasta.org.in

Cover Photo: MODIS aerosol optical depth averaged over 2001-2010. Pie charts show the relative contributions of various individual aerosol species as estimated by global model (Courtesy: Myhre et al., 2013, Nature Education Knowledge, 4(5), 7)

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RESEARCH NEWS

1. First surface and aircraft based cloud condensation nuclei (CCN) closure studies in India

Deepika Bhattu and S. N. Tripathi (IIT Kanpur)

Abstract

First surface and aircraft based CCN closure studies were carried out using a CCN (cloud condensation nuclei) counter (Droplet Measurement Technology) and Scanning Mobility Particle Sizer (TSI), in the years 2008 and 2009 at Kanpur, North India and in 2009 during Indian Continental Tropical Convergence Zone (CTCZ) campaign consisting of total 6 flight sorties, respectively. Due to higher loading of CCN the supersaturation depletion correction was applied. Significant intra-seasonal variability was observed in N_{CCN} and CCN/CN ratio due to different air masses coming from north-west, east, and central parts of India. Higher CCN/CN ratio was observed during winter season when the air mass came from north-west, central, and eastern landmass of India which was due to relatively high accumulation mode particle concentration and large number of forest fires observed in those regions. The polluted continental air masses lead to a significant increase in CCN concentrations over the winter months, most likely due to increased anthropogenic activities, i.e., increased fuel usage, large biomass burning coupled with lower mixed boundary layers. CCN closure study slightly over-predicted the CCN concentration by $21\% \pm 18\%$ which was due to lack of size dependent chemical composition and assumption of internal mixing. However, in aircraft CCN closure (average over-prediction of 1.375), the closure ratio also varied with height and depended greatly on measured CN concentrations. Results of CCN closure improved significantly (overprediction improved by 37.5% and 34.6% for Pantnagar and Gaya, respectively) with the assumption of internally mixed aerosols composed of ammonium sulfate and insoluble organics. Hygroscopicity parameter calculated for the selected

sorties ($\kappa = 0.51$ and 0.5) indicated the presence of moderately hygroscopic organic species along with some inorganic content.

Introduction

Aerosols are defined as fine solid or liquid particles suspended in gas which originated from natural sources viz. windborne dust, sea spray and anthropogenic activities such as combustion of fossil fuels, with size ranging from $0.001\mu\text{m}$ to $100\mu\text{m}$. The particles which have sufficient soluble mass to grow or activate into cloud droplets at atmospheric supersaturation (SS) are termed as the cloud condensation nuclei (CCN). The concentration of CCN can significantly affect cloud microphysical processes, and in turn, several aspects of weather and climate. Higher concentration of CCN leads to formation of droplets of smaller mean diameter, which increases the cloud reflectivity and hence cooling effect (Twomey effect). The second indirect effect is related to the inhibition of precipitation in clouds with small mean droplet diameters, which affects the extent and lifetime of clouds. The uncertainty in estimating the indirect radiative forcing is due to incomplete description of spatial and vertical distributions of anthropogenic and other type of aerosols.

Physico-chemical processes involved in the activation of particles into CCN are governed by the droplet curvature and solute effects on the water vapour pressure. The Köhler theory, composed of both these competing effects, is the theoretical basis to predict CCN activity. It determines the “critical” supersaturation (S_c in %) required by a particle to behave as a CCN and finally activate into cloud droplets. For a given aerosol particle, S_c depends on two important parameters, the dry diameter and chemical composition. Comparison of modelled CCN with the observed CCN concentration is the ultimate test of Köhler theory. Recently, some modifications to Köhler theory have been proposed to include additional effects that organic compounds, soluble, or insoluble, have on surface tension of droplet solution. CCN closure study has been performed for many years with varying success rates all over the world which depends on the aerosol characteristics and ambient conditions.

In India, efforts have been made to study aerosol microphysical and optical properties in various environments, but no study was carried out to understand CCN behaviour and closure study. This is the first study dealing with CCN properties of aerosols in India. In this study, inter and intra-seasonal variation of CCN concentrations, role of long-range transported aerosols and CCN closure study over Kanpur covering all major seasons was presented for two years 2008 and 2009. In addition, in-situ aircraft observations were carried out during the early monsoon season of 2009 across the Indian Continental Tropical Convergence Zone region (CTCZ) to understand the vertical and spatial variation of key aerosol parameters.

Results

The ground based CCN study revealed that there is considerable intra seasonal variability of CCN and CCN/CN ratio ($N_{\text{CCN}}/N_{\text{CN}}$) due to differences in the directions of air masses in all the

seasons. For both winter and pre-monsoon seasons, highest CCN concentrations were found for eastern continental transport routes whereas the highest CCN/CN ratio (0.30) was found in winter when the air masses came from central and eastern continental parts of India while in year 2009, the CCN/CN ratio was 0.53 for the similar air mass trend. Strong seasonal variability of CCN was found with highest CCN concentrations in winter and lowest in monsoon season due to cloud and precipitation scavenging. Supersaturation depletion corrections were applied to the seasons with very high concentration to reduce the uncertainty in CCN prediction. CCN closure study done using organics as insoluble fraction showed average over-prediction of $21\pm 18\%$ at all SS measured (with under prediction of $5\pm 30\%$ for lower SS (0.13%) and over prediction of higher SS ($11\pm 32\%$ and $47\pm 38\%$ at SS of 0.33% and 0.64%, respectively) (Figure 1). The prediction error is due to also be due to lack of size dependent chemical composition and assumption of internal mixing. On the other hand, in-situ aircraft measurements of CCN concentration at SS (0.84%) carried out at 6 sorties at different altitudes (0.6-6.7 km) during the CTCZ aircraft campaign. It was observed that aerosol CCN activation depends not only on the solute volume fraction but also on the number concentration and size distribution of aerosols obtained from SMPS, and their mixing state. The closure analysis assuming aerosols is composed of pure ammonium sulfate showed a very good agreement between the CCN measured and CCN predicted concentrations with an average closure ratio of 1.375, $n = 229$ and $R^2 = 0.80$. For these sorties, the CCN closure ratio varied greatly with altitude suggesting that the sampled air mass has different characteristics at different altitudes. The sampled air mass was influenced by the vehicular emissions and biomass burning aerosols from local source or long-range transport. The closure ratio also varies with CN concentrations. It has been observed that CCN closure improved significantly (over-prediction reduced by 37.5% and 34.6% for Pantnagar and Gaya, respectively) with the assumption of internally mixed aerosols composed of ammonium sulfate and insoluble organics (Figure 2). Also, the effective hygroscopicity parameter κ , calculated by simple mixing rule is also consistent with the assumption and suggests that CCN predicted concentration is highly sensitive for aerosols with high insoluble organic fraction ($\sim 80\%$ for selected two sorties).

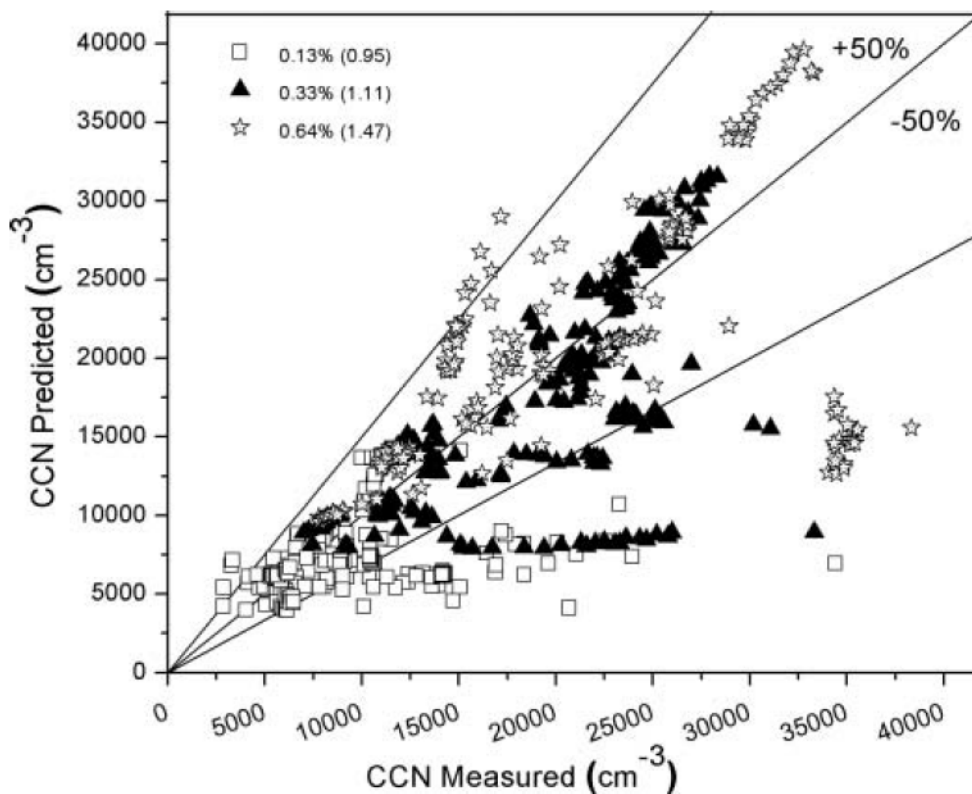


Figure 1: CCN predicted versus CCN measured at three SS (0.13%, 0.33%, 0.64%).

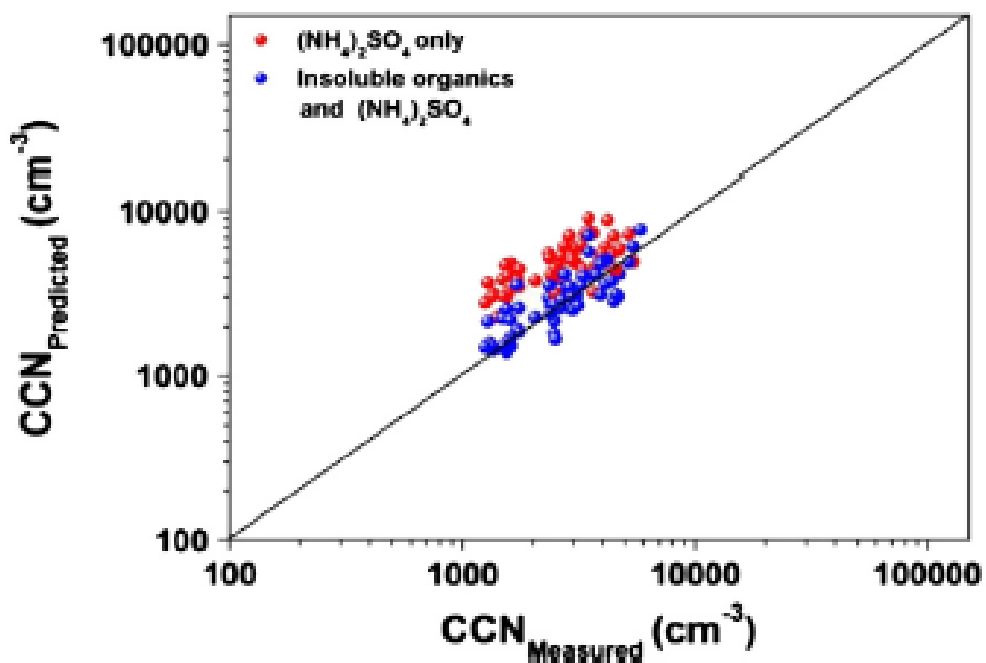


Figure 2: Change in closure ratio with assumption of internally mixed insoluble organic fraction for Gaya and Pantnagar sortie.



CCN closure analysis on surface and airborne measurements over the Indian region is a step towards understanding the dependence of CCN efficiency on the size distribution and chemical composition of the aerosol, which is one of the key linkages between aerosols, clouds and ultimately climate. It also improves our understanding of hygroscopic properties of aerosols of Indian origin.

Acknowledgements

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Srivastava, M., Tripathi, S.N., Dwivedi, A.K., Dalai, R., Bhattu, D., Bharti, P.K., Jaidevi., J., and Gupta T., CCN closure results from Indian Continental Tropical Convergence Zone (CTCZ) aircraft experiment, *Atmospheric Research*, 132-133,

2. Black Carbon science and impacts: Needs for India

Chandra Venkataraman (IIT Bombay)

Carbonaceous particles containing black carbon, BC, or refractory, light-absorbing carbon (commonly called “soot”), are formed from incomplete combustion in flames. They also contain organic carbon, OC, a multitude of organic chemical compounds. BC has a unique and important role in the Earth’s climate system because it strongly absorbs solar radiation, influences cloud processes, and alters the melting of snow and ice cover. An important property of BC is its ability to strongly absorb higher-energy shortwave radiation of the ultraviolet and visible spectrum. BC is a short-lived atmospheric constituent with a mean atmospheric lifetime of about a few days to weeks.

There are important uncertainties in the science of atmospheric BC, particularly relevant to regional climate change in South Asia, which must be addressed:

- In India, BC emission rates appear to be underestimated from both energy-use (residential cooking, using biomass fuel chulhas, residential lighting using kerosene lamps, diesel transport and industry, particularly brick production in traditional kilns) and the open burning of agricultural residue in fields following harvest.
- The atmospheric abundance of BC and its mixing with other aerosol constituents, like OC or sulphate, must be better understood to accurately estimate atmospheric warming from BC.
- BC removal from the atmosphere, particularly wet removal, needs further study, which is governed by water uptake by emitted particles.



- Non-homogeneous heating and cooling by BC aerosols can change regional circulation patterns. The effects of BC on liquid clouds, from cloud absorption and heating, could alter cloud lifetime. BC could induce changes in precipitation patterns and the Indian monsoon on various time scales.
- The deposition of absorbing aerosol particles including BC and dust decreases reflectivity of snow and ice surfaces. Recently, climate model simulations have shown such snow-pack changes to propagate to greater depths, resulting in larger heating over snow surfaces, in the Himalaya and Arctic regions.

A national programme to address these knowledge gaps must focus on large-scale field measurements and modelling needed to better estimate sectoral BC emission magnitudes and their uncertainty, better infer BC source strength from in-situ and satellite observations and receptor modelling and better constrain BC climate impacts from historical and future climate model simulations.

Uncertainty in BC climate effects raises the question, “what action can we take now?” Attempts to bring BC into climate assessment frameworks, using metrics common to greenhouse gases, have led to estimates of 100-year global-warming-potential (BC-GWP-100) of 900 (140 to 1700 range) including all forcing mechanisms. Caution is needed since BC and CO₂ emission amounts, which exert equivalent 100-year GWPs, have different impacts on temperature and rainfall, with different timing of these impacts. Also, mitigating BC addresses short-term climate change, but mitigating carbon dioxide is required to address long-term climate change. Yet, addressing BC provides an opportunity to slow climate change, as highlighted by international groups such as the Climate and Clean Air Coalition. For BC-rich sectors, discussed earlier in the Indian context, climate forcing by short-lived species (particulate matter and ozone precursor gases) is substantial (up to 75%) in comparison with their co-emissions of long-lived greenhouse gases (e.g., CO₂ and CH₄).

Addressing select BC sources could yield immediate reduction in short-term atmospheric warming. Importantly, any BC-rich source category is a promising candidate for reducing particle matter concentrations, with important co-benefits of reduced adverse public health impacts of air pollution. Other arguments relate to poverty alleviation. It is generally argued that increasing income leads to increasing energy consumption, concurrent CO₂ emissions, and an increasing climate footprint. However, at the lowest societal income levels, increase in income would lead to a shift away from high-BC-emitting energy technologies, such as biomass cooking stoves and kerosene wick lamps, leading to reduced climate footprint. Thus, climate mitigation from addressing BC emissions could be linked to strong development benefits.

INTERNATIONAL RESEARCH NEWS

1. Large uncertainty in aerosol forcing attributed to lack of knowledge about natural aerosols

A new study (*Carslaw et al., 2013*) has revealed that 45% of the variance observed in the estimated aerosol forcing relative to the pre-industrial era of 1750 can be attributed to



uncertainties in natural emissions of marine dimethylsulphide, volcanic sulphur dioxide, biogenic volatile organic carbon, biomass burning and sea spray. Remaining 34% of the variance is associated with the uncertainties in anthropogenic emissions. The results point out to the importance of understanding the long term changes in natural aerosol loading in addition to most of the recent efforts being focused on to accurately estimate anthropogenic aerosol properties.

Reference: Carslaw, K. S., L. A. Lee, C. L. Reddington, K. J. Pringle, A. Rap, P. M. Forster, G. W. Mann, D. V. Spracklen, M. T. Woodhouse, L. A. Regayre and J. R. Pierce (2013), Large contribution of natural aerosols to uncertainty in indirect forcing, *Nature*, 503, 67-71.

2. Educational article on aerosols

An educational article (*Myhre et al.*, 2013) has summarized the role of aerosols to global climate and its sensitivity. The article discusses the source and composition of aerosols and their global distributions, their role in climate and recent progress. The discussion with the help of simplified illustrations is an ideal thought-provoking article for the youngsters, who are at the beginning of their career in aerosol science.

Reference: Myhre, G., C. E. Lund Myhre, B. H. Samset and T. Storelvmo (2013), Aerosols and their relation to global climate and climate sensitivity, *Nature Education Knowledge*, 4 (5), 7.

REPORT OF INTERNATIONAL WORKSHOP

1. Changing Chemistry in Changing Climate (C4) -

1-3 May 2013 [Indian Institute of Tropical Meteorology (IITM), Pune]

Gufran Beig, Indian Institute of Tropical Meteorology (IITM), Pune, India
(beig@tropmet.res.in)

John P. Burrow, University of Bremen, Bremen, Germany (burrows@iup.physik.uni-bremen.de)

Manish Naja, Aryabhata Research Institute of Observational Sciences (ARIES), Nainital, India (manish@aries.res.in)

This workshop was a part of the regional activity of iCACGP and additionally sponsored by IGAC, IITM, MoES (India), SPARC, GURME (WMO) and MAIRS. The inauguration ceremony was also marked by the inauguration of SAFAR-Pune (System of Air Quality Forecasting and Research for Indian Metro cities) which is an operational services of air quality and weather information for the host metropolitan city Pune. The workshop attracted a huge response from scientists, young researchers and students from all over the world and a total of 147 abstracts were selected for oral and poster presentation under four themes. Total



number of registered participants who have attended the workshop was 211. Majority of the participants were from the region, representing 19 different states in India. Around 40 foreign scientists were also participated from countries like China, Germany, Ivory Coast, Kenya, Malaysia, Nepal, Nigeria, Pakistan, Switzerland, United Kingdom, and USA. In the course of these sessions, 21 invited talks were delivered by eminent scientists from all over the world.

The opening ceremony of the workshop witnessed the presence of Dr. Shailesh Nayak, secretary to Govt. of India, MoES, along with Prof. John Burrows, President, iCACGP who have made the inaugural speeches and Dr. Liisa Jalkanen, Chief, WMO, who gave the opening speech. Prof. B. N. Goswami, Director IITM, delivered the welcome speech followed by Dr. Gufran Beig and Dr. Manish Naja, conveners, C4, who gave the background on the conference and made the introductory remarks, respectively. Dr. Nayak, emphasized on the importance of studying the atmospheric chemistry with respect to the changes in the atmospheric composition and chemistry so as to see its impacts on the human health and the other social systems in the region. He also stressed on the need of ground based observation in this tropical region with main focus on long-term time series observations.

A panel discussion on "**Black Carbon and Climate Paradox in Asia**" was held. Concerns were expressed about the exaggerated role of South Asian black carbon in climate change. It was opined that organic carbon and other form of carbonaceous aerosols are equally important to understand the complete story but they are not given as much importance as BC. At the end it was concluded that there is a need to develop micro-level emission inventory of BC and OC in this region along with systematic measurements of its concentration and flux near the land, ocean and glaciers.

All the talks were delivered under following four themes and focussed on the past, ongoing and future needs of research in the field of atmospheric chemistry, aerosols, Asian monsoon, air quality and its health impacts.

Theme A – Atmospheric Chemistry in South Asia

Discussions were made on the tropospheric chemistry utilizing observed spatial-temporal variations of ozone and other trace gases from different network sites, balloon-borne, aircraft-borne, space-borne and ship-borne observations, micro-meteorological parameters, biomass burning, regional and long-range transport of trace gases, influences of stratosphere-troposphere exchange, land – atmosphere exchange of ozone, methane emissions over India along with the chloromethane and the dichloromethane measurements over land and sea. The role of Asian monsoon on UTLS ozone exchange was also discussed.

Capabilities of different models like WRF – Chem were also demonstrated by simulations of chemical fields in South Asia and gaps in our knowledge regarding the chemical, physical and dynamical processes were highlighted. Following are suggestions for improving our understanding in this region.



1. Long-term observations of some of the basic gases, particulate matter, radiation and meteorological parameters at regionally representative sites.
2. An intensive campaign having observations of many gases and radicals those play an important role in tropospheric chemistry and are not measured so-far in this region.
3. Air-borne observations to better understand the convective uplifting of air pollutants, particularly during summer-monsoon period. Attention is also needed in studying the UT/LS region.
4. Preparation of high resolution emission inventories and meteorological parameters.
5. Forming a wide spread regional observational network and interlinked with global scale network.

Theme-B - Black Carbon and Aerosols in Asia

This theme focussed on the aerosols especially the black carbon, and other particulates. Majority of the papers presented dealt with the studies on climate effects and health effects of aerosols. The climate effects were discussed with reference to the direct and indirect radiative forcing.

Studies showed that the black carbon radiative forcing is about 50% higher than in the Fourth IPCC Report, when black carbon effects on clouds and snow layers are included. Reduction of black carbon concentration is therefore a win-win situation for both air quality and climate warming. It has been observed through a study on aerosols that thick smoke plumes have altered the microphysical properties of aerosols such as effective radius and absorption coefficient.

Certain recommendations suggested a need to measure all aerosol species separately, vertical profiling of aerosol optical properties and studying their relationship with columnar physico-chemical and radiative properties as a short term plan. Spatio-temporal scale variations in aerosol composition and the influence of meteorology, constituent-segregated radiative forcing estimations against composite aerosol forcing, aerosol-cloud-precipitation-climate interactions, both regional and global aspects are some of the topics that need to be studied on a long term basis. The effects of aerosols other than *direct RF*, such as vertical temperature profiles, clouds (especially for mixed phase and ice clouds) are not well understood. An enhanced participation of regional stations in the Global Atmosphere Watch (GAW) program should be encouraged.

Theme-C - Air Quality and Weather

Papers presented in this theme ranged from studies on ambient/indoor air quality, using various techniques and impacts assessment on living beings and vegetations. Air quality in terms of trace constituents in the atmosphere including CO, CO₂, Ozone, SO₂, NO_x, Particulates (PM₁₀ & PM_{2.5}), Carbonaceous particles is measured using online analyzers or filter samples for OC and BC. The impacts of various extreme anthropogenic or even natural activities like, biomass burning, fireworks during Deepawali, forest fires and dust storms are



presented. Influences of the dynamical processes, like the boundary layer evolution and their seasonal variations are studied in the context of the air quality and the eventual impact on the public health. A study showed that the inhaled air pollutants are transferred from the mother to the growing foetus via the umbilical cord. It was also determined that burning of biomass leads to the exceeding levels of ozone.

Some issues that needed to be addressed in near future in this theme were, developing a monitoring network at the urban as well as the rural scale to identify in particular the sources of specific pollutants. Models for air quality forecasting need to be updated for which region specific or South Asia specific data in the form of emission inventories, PBL conditions need to be collected to make the models suitable for South Asian countries instead of using the global stats.

Theme-D - Climate and Monsoon

This section saw investigations on interactions between air constituents and climate and linking climate change with the Asian monsoon. The challenges in predicting Asian monsoon, models being used and the impacts of various atmospheric parameters like air quality, aerosols, impacts of aerosols on cloud formation and cloud properties, study of long term wind patterns, uplift of trace gases (e.g. water vapour) into UTLS regions and their radiative impact, long range transport at higher altitudes, troposphere-stratosphere exchange and linkages of Indian Ocean to world oceans have been discussed.

A study held at IITM has shown that the spatial structure of the summer ISOs have certain similarity with that of the summer seasonal mean. A common spatial mode of sub-seasonal and inter annual variability and that the seasonal mean of ISO anomaly can influence seasonal mean if frequency of occurrence of active and break phases are different. In order to have better understanding on linking climate and monsoon, an integrated measurement plan of ground-based, airborne and satellite observations is needed together with and comparisons with model results. The workshop ended with a rapporteur's session and an award ceremony for the best oral presentation (2) and best posters (8).



**FORTHCOMING EVENTS*****1. 9th International Aerosol Conference" -***

The 9th International Aerosol Conference will be held at Busan, Korea from Aug 31 - Sep 5, 2014. The conference covers all major topics of aerosol science and technology. The deadline for "Abstract Submissions" is February 28, 2014. More detailed information about this workshop can be found out in the conference website: www.iac2014.net/sub03_05.php.