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Seasonal differences in aerosol abundance and radiative forcing in months of contrasting emissions and rainfall over northern South Asia

P. Sadavarte[a], C. Venkataraman[a,d,*], R. Cherian[f], N. Patil[d], B.L. Madhavan[e], T. Gupta[b], S. Kulkarni[c,1], G.R. Carmichael[c], B. Adhikary[c,2]

[a] Department of Chemical Engineering, Indian Institute of Technology Bombay, Powai, Mumbai, India
[b] Department of Civil Engineering, Indian Institute of Technology Kanpur, India
[c] Center for Global and Regional Environmental Research, University of Iowa, Iowa City, IA, USA
[d] Interdiciplinary Program in Climate Studies, Indian Institute of Technology Bombay, Powai, Mumbai, India
[e] Remote Sensing of Atmospheric Processes, Leibniz Institute for Tropospheric Research, Leipzig, Germany
[f] Institute for Meteorology, University of Leipzig, Leipzig, Germany

HIGHLIGHTS

- WRF-STEM simulations show larger BC and OC burden in Apr, SO_4 in Jul & Sep.
- Influence of agricultural field burning in Apr, incursion from East Asia in Sep.
- Aerosol atmospheric forcing of 32–48 W m^{-2}, lower in Sep than in Apr & Jul.
- Anthropogenic fraction of atmospheric forcing, 45–74% over Ganga plain and Tibet.
- BC transport to Tibet, at elevated levels, from S Asia in Apr & E Asia in Sep.

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ABSTRACT

A modeling framework was used to examine gaps in understanding of seasonal and spatial heterogeneity in aerosol abundance and radiative forcing over northern South Asia, whose glimpses are revealed in observational studies. Regionally representative emissions were used in chemical transport model simulations at a spatial resolution of 60 × 60 km^2, in April, July and September, chosen as months of contrasting emissions and rainfall. Modeled aerosol abundance in northern South Asia was predominantly found to be dust and carbonaceous in April, dust and sulfate in July and sulfate and carbonaceous in September. Anthropogenic aerosols arose from energy-use emissions (from industrial sources, residential biofuel cooking, brick kilns) in all months, additionally from field burning in April, and incursion from East Asia in September. In April, carbonaceous aerosols were abundant from open burning of agricultural fields even at high altitude locations (Godavari), and of forests in the eastern Gangetic Plain (Kolkata). Direct radiative forcing and heating rate, calculated from OPAC-SBDART, using modeled aerosol fields, and corrected by MODIS AOD observations, showed regionally uniform atmospheric forcing in April, compared to that in other months, influenced by both dust and black carbon abundance. A strong spatial heterogeneity of radiative forcing and heating rate was found, with factor of 2.5–3.5 lower atmospheric forcing over the Tibet plateau than that over the Ganga Plain and Northwest in July and September. However, even over the remote Tibet plateau, there was significant anthropogenic contribution to atmospheric forcing and heating rate (45% in Apr, 75% in Sep). Wind fields showed black carbon transport from south Asia in April and east Asia in September. Further evaluation of the transport of dust and anthropogenic emissions from various source regions and their deposition in the Himalaya and Tibet, is important in understanding regional air quality and climate change over this ecosystem.

1. Introduction

The understanding of aerosols in northern South Asia, including the Ganga Plain, Tibetan Plateau and Himalayan foothills, is important both from their potential health effects on the large resident population (Stone et al., 2010; Smith et al., 2004) and their still

* Corresponding author. Interdisciplinary Programme in Climate Studies, Indian Institute of Technology Bombay, Powai, Mumbai 400 076, India.
E-mail address: chandra@iitb.ac.in (C. Venkataraman).
[1] Present address: Air Quality Planning and Science Division, California Environmental Protection Agency, Sacramento, USA.
[2] Present address: ICIMOD, Khumaltar, Kathmandu, Nepal.

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uncertain climate effects. Northern South Asia, home to about half a billion people, experiences large aerosol abundances almost all year around. Aerosol climatology using satellite observations reveals persistently high aerosol optical depth (AOD) in this region (Dey et al., 2012; Dey and Di Girolamo, 2010; Ramachandran and Cherian, 2008; Jethva et al., 2005) with a seasonally varying anthropogenic fraction, implying both spatial and seasonal heterogeneity in aerosol sources. High aerosol concentrations, especially those of secondary species (e.g., sulfates and nitrates) predominate throughout the winter during fog events (Kaul et al., 2011; Tare et al., 2006; Moorthy et al., 2005). In-situ observations of aerosols have been carried out during several field campaigns, from ground stations, ship-borne and air-borne sensors (e.g. Ramanathan et al., 2001; Tripathi et al., 2006; Moorthy et al., 2008; Jaidevi et al., 2011; Kulkarni et al., 2012). However, gaps remain in the understanding of the spatial and seasonal distribution of aerosols over the Indian subcontinent.

Few studies have been made of chemical transport and climate modeling over India. Chemical transport model simulations assimilated with satellite observations (Carmichael et al., 2009; Adhikary et al., 2008, 2007) were able to reproduce the large surface concentrations reported by observational studies. Recently, modeling platforms for trace gas chemistry, were developed with further evaluation of the sensitivity of ozone formation to NO_x emissions (Kumar et al., 2012a,b; Ghude et al., 2013). In regard to aerosols, important observational features, including AOD and vertical profiles were captured in pre-monsoon simulations of 2008 (Michael et al., 2015), while a large underestimation of AOD and black carbon (BC) surface concentrations compared to measurements was found (Nair et al., 2012), which was attributed both to emission uncertainty and model process bias.

There is also increased interest in air pollution effects on sensitive ecosystems like the Himalaya. Recently, significant BC concentrations were measured at Hanle (~4.52 km above msl) and other high altitude locations in the Himalayan foothills (Babu et al., 2011). Satellite lidar detection of aerosols, using Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) measurements (Kuhlmann and Quaas, 2010) detections reveal spatial heterogeneity in aerosols and induced heating over northern South Asia over the Tibetan plateau and the Gangetic plain. The occurrence of “polluted dust” plumes over the Tibetan plateau, suggests anthropogenic influence in remote regions. Thus, it is relevant to better understand the transport of anthropogenic pollutants to the Himalaya and Tibet regions.

In this context, the specific objectives of the present study include chemical transport model simulations over northern South Asia to investigate seasonal contrast and spatial heterogeneity in (i) surface and columnar aerosol species abundance in months of contrasting emissions and rainfall, over northern South Asia and selected sites (ii) evaluation of transport pathways of anthropogenic aerosols to the Himalayan and Tibet region (iii) estimation of monthly mean radiative forcing and atmospheric heating over three regions of northwest South Asia, Ganga plain and Tibet.

2. Modeling approach and validation

2.1. Chemical transport model – STEM

Aerosol simulations were made using the Weather Research and Forecasting model (WRF-Version 2.2) coupled with chemical transport model STEM (Sulfur Transport and dEposition Model, Tracer Version 2K1). The STEM model has been extensively used and validated with both tracer and full chemistry versions during various field campaigns, namely INTEX-B (Adhikary et al., 2010), TRACE-P/ACE-ASIA over east Asia (Carmichael et al., 2009). Studies over south-Asia have shown that the model was able to capture the seasonal variability in aerosol loading especially sulfate, dust, sea-salt, BC and organic carbon (OC) (Adhikary et al., 2007). The model was able to simulate the high surface aerosol concentrations measured over Indo-Gangetic Plain.

The STEM tracer model was run in forecast mode and simulated the mass concentration of sulfate, BC (hydrophobic and hydrophilic), OC (hydrophobic and hydrophilic), dust (fine and coarse) and sea salt (fine and coarse) aerosols. The meteorological fields from the National Center for Environmental Prediction (NCEP) Global Forecasting System (GFS) were used as initial and boundary conditions along with ice sheet coverage to model. The Weather Research Forecasting meteorological model provided the five day forecast. The WRF model was simulated with module settings such as the NOAH land surface model (Chen and Dudhia, 2001), MYJ boundary layer parameterization scheme (Mellor and Yamada, 1982) and the Kain-Fritsch subgrid scale cumulus scheme based on recommendations of Hines and Bromwich (2008). The details about WRF modeling schemes and setting are also described in D'Allura et al. (2011). Both the STEM and WRF models used a horizontal resolution of 60 × 60 km^2 with 30 vertical layers extending up to 10 hPa. Initially all the boundary conditions were set to zero at the start. Model simulations were made on the domain in a polar stereographic projection system which is centered over the Arctic region and extended until 35°N over the regions of East Asia, North America, Europe and part of South Asia (Fig. 1). The simulations were made on daily temporal scale with instantaneous values stored every 6 hourly basis from March–September 2008. In the present study, analysis is focused on northern South Asia (24°N–38°N, 68°E–98°E). The STEM meteorological pre-processor extracts the topography and land-use variables from the meteorology model WRF along with meteorological parameters necessary for calculating chemical transport. Similarly, the emission pre-processor takes the emission inventory inputs with different resolutions and maps them to the STEM domain. The pre-processor also converts all the temporal and spatial information into a standard time and grid-point format along with a consistent flux unit dimensions (molecules/sec/cm^2).

An emission inventory developed for ARCTAS mission of 2008 (Jacob et al., 2010) was used in this study, which included emissions from INTEX-B (Zhang et al., 2009) developed over south and south-east Asia. Indian emissions covered in INTEX-B were scaled from Reddy and Venkataraman (2002), which includes energy sectors of industry, power, residential and transport. Reddy and Venkataraman (2002a,b) emissions were scaled using the fuel type and sectoral energy consumption from IEA energy data set over India from 1996 to 2006. Further details about the Indian emissions in INTEX-B domain can be found in Zhang et al. (2009). Real-time emissions of BC, OC and SO_2 from open burning of forests and agricultural residues were included from RAQMS (Real-time Air Quality Modeling System) (D'Allura et al., 2011). The sea-salt and dust emissions were parameterized respectively based on Gong (2003) and Tang et al. (2003), with size resolution detailed in Adhikary et al. (2007).

2.2. Aerosol optical depth and radiation transfer calculations

STEM model derived aerosol concentrations were input to the Optical Properties of Aerosols and Clouds (OPAC 3.1) database (Hess et al., 1998), to calculate the AOD and single scattering albedo (SSA) along with the asymmetry parameter. Model calculated species were mapped to OPAC categories as follows: sulfate to water-soluble, BC to soot, OC to insoluble, both bins of sea-salt to sea-salt-coarse, dust-fine to mineral dust (nucleation) and dust-coarse to mineral (transported) from matching size-fractions. OPAC AOD was calculated on a monthly mean time scale at eight selected sites in the domain, representing urban and non-urban lo-