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A methodology to estimate source-specific aerosol radiative forcing

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ABSTRACT

This paper presents a novel approach to estimate source-specific radiative forcing by combining source apportionment results for particulate matter mass with satellite (moderate resolution imaging spectroradiometer (MODIS)) derived aerosol optical depth (AOD). Positive matrix factorization (PMF) was applied to particulate matter (PM) mass and its chemical constituents measured during a winter intensive study (December 2004) at Hisar, Haryana, India. The model resolved four factors including carbonate rich dust, combustion rich aerosol, secondary sulfate/nitrate, and an unidentified factor likely to be emission from polymer industries. Carbonate rich dust was the highest contributor to the measured PM mass closely followed by combustion rich aerosol with their average contributions accounting for 34.0% and 33.6%, respectively. Model apportioned species concentrations corresponding to each factor were then used to estimate factor specific optical and radiative properties, and radiative transfer calculations were performed for the shortwave regime. During the study period, although carbonate rich dust and combustion rich aerosol mass contributions were comparable, carbonate rich dust contributed to only 22% of top of the atmosphere forcing while combustion rich aerosol contributed nearly 56%. Overall, the results suggested that the aerosol radiative forcing was primarily governed by the aerosol optical and radiative properties, while the mass concentrations played a secondary role.

1. Introduction

Prediction of climate and its variations in different time scales is a major area of research, globally. Atmospheric aerosols influence the earth's climate in many important ways. Aerosols interact directly and indirectly with solar and infrared radiation in the atmosphere and give rise to radiative forcing. The direct and indirect aerosol radiative forcings (ARFs) remain a significant uncertainty for climate studies (Intergovernmental Panel for Climate Change (IPCC), 2007). Studies on specieswise determination of ARF from in-situ measurements of optical, physical and chemical properties of aerosols are quite rare. A recent study apportions assimilated monthly mean aerosol optical depth (AOD) values of seven species from a model simulation over Hanimaadhoo and Gosan (Adhikary et al., 2008). Another study during the Indian Ocean Experiment (INDOEX) simulated ARFs using a Monte Carlo radiation model for several species (Podgorny & Ramanathan, 2001). However, no studies on the source-specific aerosol extinctions and/or source apportioned ARFs exist over the tropics, and perhaps over the globe, to the best of our knowledge.

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The sources of aerosols can be natural (e.g., sea salt, biogenic, volcanic, and dust) or manmade (e.g., combustion of fossil fuel from urban/industrial processes and biomass burning), and they differ on a regional basis leading to regional variations in the Earth's radiation budget. Aerosols are abundant near source regions, however, they can impact global climate as their radiative influence gets transported due to changes in the mean atmospheric circulation patterns. The potential for aerosol forcing of climate can vary according to regional differences in aerosol columnar concentration, chemical composition and the age of air mass (Spencer, Holecek, Corrigan, Ramanathan, & Prather, 2008). The combined effects of aerosols produced by local sources and transported from long-range in different seasons may alter large-scale heating and induce changes in the atmospheric general circulation, thereby affecting the processes of generation of clouds and rainfall (Lau et al., 2008). Information on radiative effects of different kinds of aerosols is still not available in order to permit the examination of aerosol effects on hydrological cycle and monsoon climate variability. Studies focusing on the source-specific aerosol radiative forcing will be useful in examining the dynamical changes due to aerosols and their effect on climate.

The Indo-Gangetic plain in Northern India is a densely populated region. Emissions from biomass burning, industries, power plants and vehicles are dominant contributors to ambient aerosols throughout the year. Consequently, aerosol optical depths are higher over this region throughout the year compared to other regions in India (Ramachandran & Cherian, 2008). Dense foggy conditions accompanied by reduced visibility occur during winter over this region when cold air from higher latitudes mixes with the local moist air. Low temperatures and frequent temperature inversions during winter enhance fog formation resulting in accumulation of pollutants near the surface. Thus, to document the levels and variations in aerosols and trace gases, a major land campaign was conducted over the Indo-Gangetic plain in December 2004 during which a variety of aerosol and trace gas related measurements were made at eight fixed stations including Delhi, Hisar, and Kanpur (Ganguly, Jayaraman, Rajesh, & Gadhavi, 2006; Ramachandran, Rengarajan, Jayaraman, Sarin, & Das, 2006; Rengarajan, Sarin, & Sudheer, 2007). In this study, aerosol optical depth (AOD), aerosol size distribution, mass concentrations, and aerosol chemical composition measured during a land campaign in the Indo-Gangetic plain over Hisar (29.1°N, 75.7°E), Haryana during December 2004 were used to perform source apportionment of mass, and derive source-specific aerosol extinction and radiative forcing.

2. Measurements, data analysis, and approach

Hisar is a semi-urban location with a population of about 1.25 million and is an industrial town with several cotton and steel industries. Hisar is influenced by industrial emissions, traffic, and agricultural stubble burning to some extent. Details of the sampling site, prevailing meteorology during winter and measurement details are described in Ramachandran et al. (2006) and Rengarajan et al. (2007). Aerosol optical depths were measured using an indigenously built hand held sun photometer in the 0.4–1.02 μm wavelength range (Ramachandran et al., 2006). Ambient bulk aerosol samples (integrated over 8–10 h) drawn through pre-combusted (at 450 °C) Whatman quartz fiber filters using an Andersen high volume sampler (HVS) were measured by Rengarajan et al. (2007). These integrated filter samples were then used to determine the total aerosol mass and the chemical composition. Samples were analyzed for water soluble ionic species, carbonaceous aerosol and the results are reported in Rengarajan et al. (2007).

2.1. Mass and chemical composition

2.1.1. Data description and quality

Concentrations of total suspended particles (TSP) mass and 12 chemical species reported in Tables A1 and B1 of Rengarajan et al. (2007) were used for source apportionment of aerosols over Hisar. The data set was first examined for mass closure (Malm, Sisler, Huffman, Eldred, & Cahill, 1994). In this study, mass closure was not achieved. An earlier source apportionment study (Larson et al., 2006) suggested that when mass closure is not achieved the 'missing mass' may be calculated and included as a model input. Thus, missing mass was calculated as the difference between TSP mass and the sum of ionic species, elemental carbon (EC), carbonate carbon (CC), and organic matter (OM). It must be noted that organic carbon (OC) was only measured and OM was estimated as 1.8 times OC. Since several mineral dust marker elements (Si, Fe, Al, Ti) were not measured in this study, it is suggested that the missing mass is likely to be mineral dust. It was desirable to retain 'missing mass' as a chemical species for source apportionment since it serves as a surrogate for mineral dust.

Positive matrix factorization (PMF) was applied to the measured chemical species concentrations to identify the sources of TSP mass. The key statistics for all of the chemical species used as model inputs, and TSP mass are shown in Table 1. In this table, DL indicates detection limit and S/N indicates signal to noise ratio. Detection limits for all ions were reported in Rengarajan et al. (2007). The S/N ratio is useful in assessing the quality of data points and is discussed in detail by Paatero and Hopke (2003). Since all species had S/N ratios greater than 2, none of the data points were downweighted (Paatero & Hopke, 2003). Thus, a total of 13 species (including missing mass) and 31 samples (collected during December 2004) were retained for the analysis.