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Positive matrix factorization and trajectory modelling for source identification: A new look at Indian Ocean Experiment ship observations

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Abstract

The sources of aerosols on a regional scale over India have only recently received attention in studies using back trajectory analysis and chemical transport modelling. Receptor modelling approaches such as positive matrix factorization (PMF) and the potential source contribution function (PSCF) are effective tools in source identification of urban and regional-scale pollution. In this work, PMF and PSCF analysis is applied to identify categories and locations of sources that influenced surface concentrations of aerosols in the Indian Ocean Experiment (INDOEX) domain measured on-board the research vessel Ron Brown [Quinn, P.K., Coffman, D.J., Bates, T.S., Miller, T.L., Johnson, J.E., Welton, E.J., et al., 2002. Aerosol optical properties during INDOEX 1999: means, variability, and controlling factors. Journal of Geophysical Research 107, 8020, doi:10.1029/2000JD000037]. Emissions inventory information is used to identify sources co-located with probable source regions from PSCF. PMF analysis identified six factors influencing PM concentrations during the INDOEX cruise of the Ron Brown including a biomass combustion factor (35–40%), three industrial emissions factors (35–40%), primarily secondary sulphate–nitrate, balance trace elements and Zn, and two dust factors (20–30%) of Si- and Ca-dust. The identified factors effectively predict the measured submicron PM concentrations (slope of regression line $= 0.90 \pm 0.20$; $R^2 = 0.76$). Probable source regions shifted based on changes in surface and elevated flows during different times in the ship cruise. They were in India in the early part of the cruise, but in west Asia, south-east Asia and Africa, during later parts of the cruise. Co-located sources include coal-fired electric utilities, cement, metals and petroleum production in India and west Asia, biofuel combustion for energy and crop residue burning in India, woodland/forest burning in north sub-Saharan Africa and forest burning in south-east Asia. Significant findings are equivalent contributions of biomass combustion and industrial emissions to the measured aerosol surface concentrations, the origin of carbonaceous aerosols largely from biomass combustion and the identification of probable source regions in Africa, west Asia, the Arabian peninsula and south-east Asia, in addition to India, which affected particulate matter concentrations over parts of the INDOEX domain covered by the Ron Brown cruise.

Keywords: Positive matrix factorization (PMF); Potential source contribution function (PSCF); Source identification; INDOEX; Atmospheric aerosols

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1. Introduction

Observational studies during the Indian Ocean Experiment or INDOEX (e.g., Ramanathan et al., 2001; Ramachandran and Jayaraman, 2002) and more recent field campaigns (e.g., Ramachandran, 2005; Moorthy et al., 2005) show widespread existence of aerosols and trace gases of anthropogenic origin over the Indian continent and ocean regions adjoining India. Interior continental regions have a predominance of coarse-mode dust aerosols, significant particulate black carbon concentrations at the surface and in elevated layers (e.g., Tripathi et al., 2005) and high anthropogenic aerosol load especially in the Indo-Gangetic plain (e.g., Ramachandran et al., 2006). These studies point to large spatial and temporal variability of surface and elevated aerosols over the Indian subcontinent and adjoining oceans, the contribution of both local emissions and long-range transport, and regionally specific aerosol chemistry, potentially mediated by dust aerosols.

The possible sources of aerosols on a regional scale over India have only recently received attention. Using back trajectory information INDOEX investigators identified channels of flow from different geographical regions influencing their observations (e.g., Quinn et al., 2002) and qualitatively linked aerosol chemical and optical properties to broad source categories of anthropogenic, dust and sea-salt aerosols. Further, general circulation model simulations with newly available regional emissions information for atmospheric simulations over south Asia during the INDOEX period (e.g., Verma et al., 2007) indicated the possible contribution of regions outside India to INDOEX aerosols.

Receptor modelling for quantitative source apportionment of aerosols includes the chemical mass balance (Friedlander, 1973) and positive matrix factorization (PMF) (Paatero, 1997) approaches. Additionally, identification of source locations by combining back trajectory and composition information for a given aerosol dataset includes methods such as the potential source contribution function (PSCF) (Hopke et al., 1995). Both PMF and PSCF have found wide application on an urban scale (Pekney et al., 2006a, b) and are effective tools in source identification of long-range transported pollution as well (e.g., Kim and Hopke, 2004). In this work, we attempt quantitative source identification of INDOEX aerosols towards developing an understanding of the south Asian aerosol budget, i.e., the relative influence of emissions from within the region and from long-range transport from distant regions, on the south Asian aerosol load. We apply PMF and PSCF analysis to identify categories and locations of sources that influenced the surface concentrations of aerosols in the INDOEX domain measured on-board the research vessel Ron Brown (Quinn et al., 2002).

2. Modelling approach

2.1. Aerosol dataset

2.1.1. Data description

Aerosol chemical data were collected aboard the Ron Brown during its cruise in the Indian Ocean as part of the INDOEX (Quinn et al., 2002) from 22 February to 30 March 1999. These included submicron ($D_{\mathrm{aero}} < 1.1\,\mu\mathrm{m}$) and super micron ($1.1\,\mu\mathrm{m} < D_{\mathrm{aero}} < 10\,\mu\mathrm{m}$) particle fractions, collected using a two-stage impactor (Quinn et al., 2002). This work is limited to source identification for the submicron fraction, more likely to be subject to regional-scale transport. Particle samples for different chemical analysis were collected for different sampling durations: inorganic ions on a 6-h duration (four samples per day), carbon on 12-h duration (two samples per day) and trace elements on 24-h duration (one sample per day). Chemical species included in the concentration matrix input to PMF analysis were K^+, Na^+, NH_4^+, Ca^{2+}, methyl sulphonic acid (MSA), Cl^-, NO_3^-, non-sea-salt sulphate (nss-SO_4^{2-}), carbon fractions of organic carbon (OC) and elemental carbon (EC), and trace elements Al, Si, P, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As and Pb (Table 1). Analysis methods used (Quinn et al., 2002) were ion chromatography (IC) for ions, a thermographic method for OC and EC (Neususs et al., 2002), and X-ray fluorescence (XRF) spectroscopy for trace elements. The thermographic method for carbon analysis did not use laser transmission to identify the OC/EC split or correct for pyrolytic OC, per standard TOT/TOR methods, which could lead to overestimation of EC and corresponding underestimation of OC. The primary data were averaged to give 24-h mean concentrations and uncertainty estimated by propagation of variance, to give a concentration data matrix of 27 daily mean values for 23 chemical species.