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Study on particulate polycyclic aromatic hydrocarbons over Bay of Bengal in winter season

Mohit Saxena [a,*], D.P. Singh [b], T. Saud [a], Ranu Gadi [b], S. Singh [a], S.K. Sharma [a], T.K. Mandal [a]

[a] CSIR — National Physical Laboratory, Dr. K S Krishnan Road, New Delhi 110 012, India
[b] Guru Govind Singh Indraprastha University, New Delhi 110 006, India

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ABSTRACT

Particulate polycyclic aromatic hydrocarbon (PAH) concentration was estimated during Integrated Campaign on Aerosol, Gases and Radiation Budget (ICARB-W) in winter (2008–2009) on board the Sagar Kanya over Bay of Bengal (BOB). The concentration of particulate PAH (Σ_{11} 6.65 ± 2.86 ng/m^3) over BOB ranged from 0.28 ng/m^3 to 1.15 ng/m^3. The concentration of middle and high molecular weight PAHs (4–6 rings), accounted for >50% of total PAHs. Analysis of photochemical oxidation using ratios of BaA/Chy and BaP/BbF suggested the fresh release of air masses from local sources or from the nearest coastal areas of BOB. Principal component analysis (PCA) and correlation between tPAHs, nss-K^+, nss-SO_4^{2-}, NO_3^-, organic carbon (OC) and elemental carbon (EC) suggested that motor vehicle emission, biomass burning and coal combustion may be the possible principal sources of PAHs over BOB during the study period.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of complex organic compounds, formed by incomplete combustion or high-temperature pyrolysis of fossil fuel and biomass fuels (Serio et al., 1987; Wornat et al., 1987, 1988). PAHs are present in the ambient air as vapor (2–3 rings) or adsorbed into airborne particulate matter (Bodzek et al., 1993). Gas-to particle partition of PAHs (4 rings) depends on the molecular weight of the compounds, temperature, humidity and precipitation (Subramanyam et al., 1994; Vanjaarsveld et al., 1997).

PAHs, due to their lipophilicity and hydrophobicity, may partition and accumulate in organisms, leading to carcinogenic and mutagenic activity (Larsen and Larsen, 1998; Okona-Mensah et al., 2005; Perera et al., 2005). PAHs present in the atmosphere are subject to complex physico-chemical reactions and transformations in the atmosphere. Polyaromatic hydrocarbons are well mixed with secondary inorganic particulates that may result in the formation of nitro-PAHs, which are potentially more mutagenic and carcinogenic than the parent PAHs (Ramdahl et al., 1982; Valerio et al., 1984; Li et al., 2010). Long term exposure of PAHs may cause cataracts, kidney and liver damage while repeated contact with skin may induce redness and skin inflammation. Naphthalene, a specific PAH, can cause the breakdown of red blood cells, if inhaled or ingested in large amounts (http://www.health.sa.gov.au/pehs/PDF-files/ph-factsheet-PAHs-health.pdf). In a recent case study in pediatrics, increased serum PAHs were found to be associated with allergy, asthma, or respiratory symptoms (Al-Daghri et al., 2013). PAHs are also known to cause lung cancer, anemia, leukemia, and lymphoma, and disorders in reproductive function and the nervous system (Baird et al., 2007; Colombo et al., 2006).

Several extensive investigations and coordinated field campaigns have been carried out to assess the impact of particulates on climate (Pratesi et al., 2007; Vecchi et al., 2007). Arabian Sea Monsoon Experiment (ARMEX) (Sanjeeva

* Corresponding author at: Radio and Atmospheric Sciences Division, CSIR — National Physical Laboratory, Dr. K S Krishnan Road, New Delhi 110 012, India. Tel.: +91 11 45609448; fax: +91 11 45609310.
E-mail address: mohitnpl@gmail.com (M. Saxena).

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Rao, 2005), Bay of Bengal Monsoon Experiment (BOBMEX) (Bhat, 2001) and Indian Ocean Experiment (INDOEX) (Ramanathan et al., 2001) are some examples. Particulate PAHs in the urban and rural areas were mainly from automobile exhausts and/or combustion of fossil fuel (Mai et al., 2003; Ohura et al., 2004) whereas, in the Arctic, it has been reported that both biomass burning and coal combustion have contributed to PAHs (Ding et al., 2007). Similarly, Crimmins et al. (2004) assessed the continental impact of combustion derived particulate matter and pointed out that fossil fuel and biomass combustion were the predominant sources of PAHs in the marine atmosphere of the Indian Ocean and Atlantic Ocean during Aerosol 99 and INDOEX projects.

The occurrence and sources of PAHs in the atmosphere have been widely studied, both in urban and rural areas (Sienra et al., 2005; Wang et al., 2006; Lee et al., 2011; Liu et al., 2013; Tan et al., 2011; Chattopadhyay et al., 1998; Mohanraj and Azeez, 2003; Nizzetto et al., 2008; Pandit et al., 1996; Raiyani et al., 1993; Singh et al., 2011), but only limited study has been conducted on the analysis of particulate PAHs in the marine aerosol, notably over Bay of Bengal (BOB). To explore the physical, chemical and optical properties of aerosol in the entire BOB, coordinated experimental campaign (ICARB-W) was carried out for the period of December 2008 to January 2009. Results of ICARB-W which covers different aspects of aerosol and trace gases, have been published in several articles (Kharol et al., 2011; Sinha et al., 2011a,b; Kumar et al., 2011; Sharma et al., 2012a, b) so far. In the present study, characterization, distribution and possible sources of particulate bound polycyclic aromatic hydrocarbon over BOB are discussed in detail.

2. Experimental details

2.1. Sampling area

Sampling area and meteorological description of the Integrated Campaign on Aerosol, Gases and Radiation Budget (ICARB-W) during the winter period (2008–2009) have been published elsewhere (Kharol et al., 2011; Sinha et al., 2011a, b; Kumar et al., 2011; Sharma et al., 2012a,b). During the campaign, particulate matter (PM_{10}) was collected using a Respirable Dust Sampler (RDS; Envirotech 460 NL for PM_{10}) on board the Sagar Kanya (SK-254) over BOB. Sampling track of SK-254 cruise for winter season over BOB is shown in Fig. 1. The cruise started on 27 December, 2008 from Chennai (13.12°N, 80.3°E) and ended in Kochi (9.96°N, 76.3°E) on 31st January, 2009.

2.2. Ambient air sampling

PM_{10} samples were collected during the winter season from 27 December 2008 to 25 January 2009 on a pre-heated (550 °C for about 5 h) Whatman quartz microfiber filter paper (QMA: 20.3×25.4 cm^2), using a Respirable Dust Sampler (RDS; Envirotech 460 NL for PM_{10}) kept on the deck of the ship about 11 m high from the mean sea level. In the present study, samples were collected upwind in the uppermost front deck to avoid contamination from the ship exhaust. In the present study, a total of twenty two samples were collected from site 1 (ten samples), site 2 (six samples) and site 3 (six samples) of BOB having sampled period of around 18 h over the span of 24 h, but PAHs were detected only in eight samples from site 1, three samples from site 2 and two samples from site 3, whereas in the remaining nine samples, either PAHs were not present or their concentrations were below the detection limit. The collected samples were kept in polythene bags and stored in a refrigerator at −20 °C till analysis. The field blank sample was also collected using similar type of Respirable Dust Sampler (RDS; Envirotech 460 NL for PM_{10}) following the same protocol as opted for sample analysis.

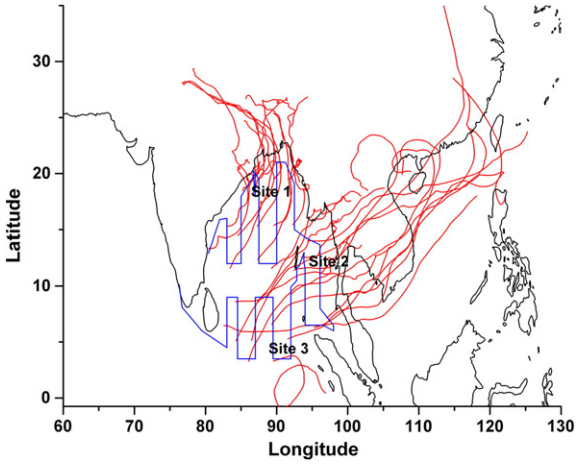

Fig. 1. Cruise track of the Integrated Campaign on Aerosols, Gases and Radiation budget during winter (ICARB-W) over the BOB and backward trajectory analysis (HYSPLIT model) at 500 m.

2.3. Carbonaceous aerosol & water soluble inorganic components (WSIC)

Experimental details for analysis of organic carbon (OC), elemental carbon (EC) and water soluble inorganic component (WSIC) were discussed in our earlier published work (Saud et al., 2012; Sharma et al., 2012b). Briefly, organic carbon (OC) and elemental carbon (EC) were analyzed using a Thermal/Optical Carbon Analyzer (Model: DRI-2001; M/s. Atmoslytic Inc., Calabasas, CA, USA) following the IMPROVE A (Interagency Monitoring of Protected Visual Environments) thermal/optical reflectance (TOR) protocol (Chow et al., 2001; Fung et al., 2002). Cations and anions present in the water soluble fraction of PM_{10} were analyzed by Ion Chromatograph (DIONEX ICS-3000). Eluent used for the separation of anion and cation was 25 mM NaOH (50% w/w) and 5 mM MSA at a flow rate of 1.5 and 1 ml/min respectively. Suppressed conductivity detection was used to monitor the eluted analyte. Working standards were prepared from stock standard solutions procured from M/S Dionex. All the standard solutions were filtered using 0.45 μm nylon membrane filters (Millipore) and degassed by ultrasonication.

2.4. Extraction of PAHs and analysis by GC–MS

Concentrations of particulate PAHs, deposited over quartz filter paper, were determined by cutting a piece of it and extracting it three times with 30 ml of HPLC-grade