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Atmospheric polybrominated diphenyl ethers (PBDEs) and Pb isotopes at a remote site in Southwestern China: Implications for monsoon-associated transport

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ABSTRACT

A 13-month sampling campaign was conducted at a remote site in southwestern China from October, 2005 to December, 2006. An integrated approach with lead isotopes and air back trajectory analysis was used to investigate the monsoon-associated atmospheric transport of PBDEs in tropical/subtropical Asia regions. The air concentration of PBDEs ranged from 1.6 to 57.5 pg m^{-3} (15.9 ± 12.0 pg m^{-3}), comparable to reported levels at other remote sites in the world. BDE-209, followed by BDE-47 and -99 dominated the PBDE compositions, indicating a mixed deca- and penta-BDE source. Air mass back trajectory analysis revealed that the major potential source regions of BDE-47 and -99 could be southern China and Thailand, while those of BDE-209 are widely distributed in industrialized and urbanized areas in tropical Asia. The different lead isotope compositions of aerosols between trajectory clusters further substantiated the observation that the South Asian monsoon from spring to summer could penetrate deep into southwestern China, and facilitate long-range transport of airborne pollutants from South Asia.

1. Introduction

The sweeping industrial development and urbanization in Asian countries in recent decades has already led to a dramatic increase in the emissions of airborne pollutants. The emitted pollutants can undergo atmospheric transport and result in deterioration of air quality in distant areas. Monsoon is one of the important factors that influence the long-range transport of airborne pollutants. Studies on the Tibetan Plateau (Cheng et al., 2007; Gong et al., 2010) and northeastern Asia (Tian et al., 2009) demonstrate that Asian monsoon can deliver significant amount of persistent pollutants to regions far away from their source areas. The high concentrations of airborne organic pollutants in tropical Asia have been widely reported (Chen et al., 2006; Muenhor et al., 2010; Zhang et al., 2008). Under the regime of South as well as East Asian monsoons, the outflow in tropical Asia could be a pollution source of global significance.

PBDEs are a class of semi-volatile organic pollutants widely used as flame retardants in commercial and industrial products. Owing to their persistency in the environment, biologically cumulative effect and tendency to undergo atmospheric transport, PBDEs have become ubiquitous environmental contaminants. Over the past several years, PBDE pollution in Asia has been widely reported (Chen et al., 2009; Hayakawa et al., 2004; Moon et al., 2007). Precise data on the production and consumption of PBDEs in tropical Asia are not available, but a large quantity of PBDEs may come from unregulated e-wastes recycling and disposal. Of the Asian countries, China, India and Pakistan have long been suspected of receiving and disposing the bulk of the world's obsolete computers and electronic components (Martin et al., 2004). Primitive treatment of the e-waste items often includes open combustion which could introduce large amounts of PBDEs into the atmosphere, both in gaseous and particle phases (Chen et al., 2009). Since the atmosphere is considered to be a key environmental vector for the transport of PBDEs (Wurl et al., 2006), those compounds have the potential for long-range atmospheric transport (LRAT) via Asian monsoon.

LRAT of pollutants can be traced with a variety of tools/indicators, which normally include compositional variations of target pollutants, diagnostic information from other chemical tracers, as well as meteorological analysis of the air mass movement. Lead has four naturally occurring stable isotopes, ^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb. The relative abundances depend upon the age and initial composition of ^{238}U, ^{235}U and ^{232}Th in the source regions thus exhibits distinguishable geographic variations. Meanwhile, Pb isotopic ratios remain constant during industrial and environmental processes because of relatively heavy atomic masses, making the ratios ideal indicators to track down sources of aerosol pollution (Lee et al., 2007). Air mass back trajectory and potential source contribution function (PSCF) have been widely employed to identify source regions of airborne pollutants (Hafner and Hites, 2003; Hoh and Hites, 2004). The back trajectory model is designed to simulate the trajectories of atmospheric transport of pollutants. With

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the calculated air parcel trajectories and on-site observation, the PSCF model can quantitatively identify source regions of airborne pollutants, and map the probability of each area as sources. In this study, a combination of PBDE composition/levels, Pb isotopes, air mass back trajectory and PSCF analysis was used to assess the atmospheric transport behavior of PBDE in tropical Asia.

Tengchong Mountain (TM) is a remote field site in the tropical zone of the Global Climate Observing System (GCOS), World Meteorological Organization (WMO). The site is located on a subtropical monsoon region controlled by East Asian monsoon as well as South Asian monsoon. During the winter season, the air mass travels from the east along the coasts of China and Vietnam, resulting in transportation of pollutants from southern China to TM. During the summer season, the moisture-laden South Asian summer monsoon driven by the low pressure over the northern and central Indian subcontinent penetrates deeply into southwestern China, taking pollutants from South Asia to TM site. Therefore, a variety of measurements were conducted here to study the atmospheric transport of various pollutants excluding POPs in Mainland Southeast Asia (MSA) and China (Chan et al., 2006). TM site and its neighboring areas in China and Myanmar are mostly forest covered. Local sources of PBDEs are negligible compared with source regions, which favors the observation of LRAT in tropical Asia.

The objective of this study was to examine the monsoon-facilitated LRAT behavior of PBDEs in southern China (SC), India and MSA regions. Background levels, temporal variation pattern and potential source regions of PBDEs in the tropical Asia were also investigated.

2. Material and methods

2.1. Sampling site and collection

A modified Anderson-type high volume sampler was installed on the top of Tengchong Mountain (24.95°N, 98.48°E, 1960 m a.s.l), located in Yunnan Province of southwestern China, close to the eastern part of the Tibetan Plateau (Fig. 1). Details on the sampling site can be found elsewhere (Tang et al., 2009). Local climate is characterized by high temperatures all year round, heavy rainfall in summer and intense solar radiation in spring.

Fifty-nine 24-h successive air samples (about 400 m^3) were collected once a week from 22 October 2005 to 27 December 2006. The quartz microfiber filters (QFF) (QMA, 20.3×25.4 cm, Whatman, Maidstone, England) and pre-cleaned polyurethane foam (PUF) plugs (6.5 cm diameter, 7.5 cm length, 0.030 g cm^{-3} density) were used to collect the particulate and gas phase samples, respectively. The QFFs were baked in a muffle furnace at 450 °C for 4 h. The PUF plugs were precleaned twice by an Accelerated Solvent Extraction with a mixture (1:1, V/V) of acetone and dichloromethane (DCM) at 100 °C and 1500 psi for 15 min. After sampling, all the samples were wrapped with clean aluminum foil, sealed in Teflon bags, and kept at −18 °C before analysis.

Three surface soil samples were taken along a transect between TM and the nearest town (Tengchong town) with different altitudes and one soil sample was collected at the nearest lead mine area. Each soil sample was composed of nine subsamples obtained from 0 to 5 cm depth. Soil samples were freeze-dried and grinded to test Pb isotope.

2.2. Pb isotope analysis

One quarter of aerosol-laden filter samples were submerged with 10 ml of concentrated high-purity HNO_3 and 2.5 ml of concentrated $HClO_4$ in acid-cleaned Pyrex test tubes and digested in an aluminum heating block. The temperature was set at 50 °C for 3 h, 75 °C 0.5 h, 100 °C 0.5 h, 125 °C 0.5 h, 150 °C 3 h, 175 °C 2 h and 190 °C until dry. 15 ml 5% (v/v) high-purity HNO_3 was added into each test tube and heated at 60 °C for 1 h. After cooling down, the solution was transferred into plastic tubes and centrifuged before analysis. Soil samples (0.125 g) were digested with a mixture of 8.0 ml of concentrated high-purity HNO_3 and 2.0 ml of concentrated $HClO_4$.

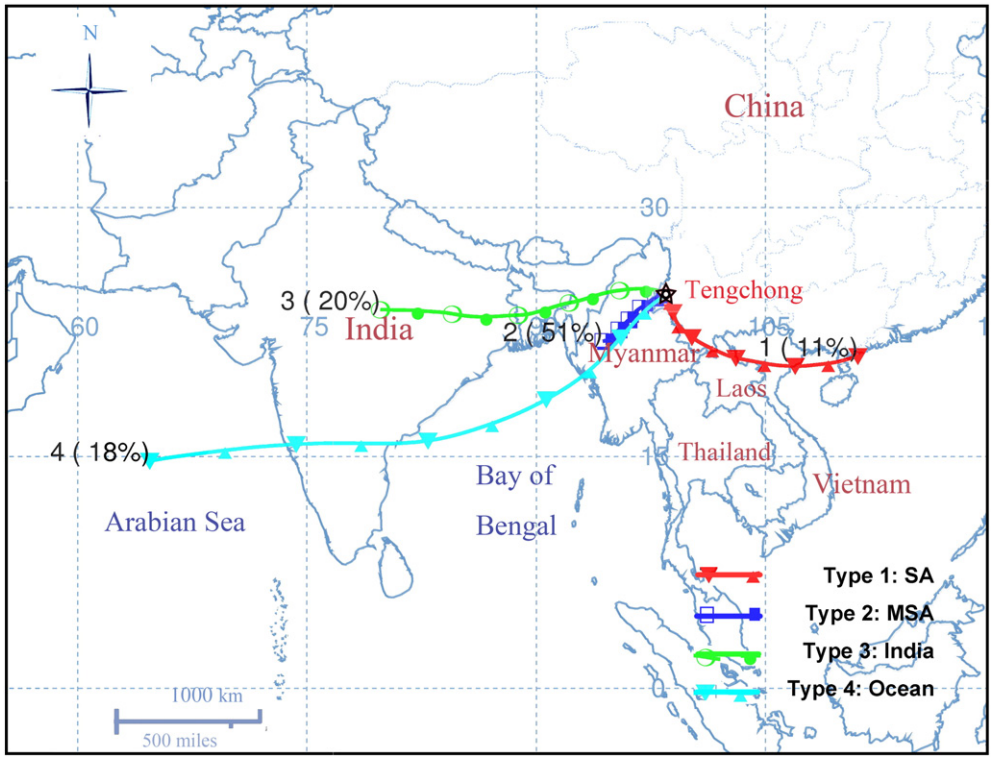

Fig. 1. Sketch map showing the TM sampling station and four types of clusters of air masses backward trajectories.