Atmospheric Environment 115 (2015) 124–131

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Long-range atmospheric transport of particulate Polycyclic Aromatic Hydrocarbons and the incursion of aerosols to the southeast Tibetan Plateau

Xiaoping Wang [a, b, *], Ping Gong [a, b], Jiujiang Sheng [a], Daniel R. Joswiak [a], Tandong Yao [a, b]

[a] Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing, 100101, China
[b] CAS Centre for Excellence in Tibetan Plateau Earth Sciences, China

HIGHLIGHTS

- Concentrations of particulate-PAHs were higher in the winter–spring season.
- The source region of particulate-PAHs was the Indo-Gangetic Plain.
- Smoke in the southern Himalayas could penetrate the barrier of Himalayas.

ARTICLE INFO

Article history:
Received 9 December 2014
Received in revised form
20 April 2015
Accepted 22 April 2015
Available online 27 May 2015

Keywords:
Polycyclic Aromatic Hydrocarbons
Tibetan Plateau
Westerly winds
Himalayas

ABSTRACT

The long range atmospheric transport of Polycyclic Aromatic Hydrocarbons (PAHs) from heavily polluted regions to remote areas has been well-known. However, there are limited studies focussing on the Tibetan Plateau (TP). To assess the temporal transport patterns of PAHs and particulate matters (PMs), a ground-based observation program (2008–2011) was conducted in the southeast TP. Relatively high atmospheric concentrations of particulate-PAHs (0.2–5.0 ng m^{-3}, sum of 15 compounds) and Total suspended Particles (TSP, 4.1–46.7 μg m^{-3}) were observed. Concentrations of particulate-PAHs/TSP exhibited seasonality with higher levels in the winter–spring season and lower levels in summer. Using the potential source contribution function model, the source region of both particulate-PAHs and particles was attributed to the Indo-Gangetic Plain (IGP), suggesting the co-transport of particulate-PAHs and aerosols. The aerosol incursion, penetrating the Himalayas and reaching the TP was further captured by Moderate Resolution Imaging Spectroradiometer and Cloud-Aerosol Lidar Infrared Pathfinder Satellite Observations.

1. Introduction

Polycyclic Aromatic Hydrocarbons (PAHs) are potentially carcinogenic and mutagenic, and they are commonly known as persistent organic pollutants (POPs) (Masiol et al., 2012). PAHs have attracted much attention due to their toxicity and ability to undergo long-range atmospheric transport (LRAT) (Ding et al., 2007). For example, LRAT is the most efficient way for PAHs released in the lower latitudes to reach the Arctic (Friedman and Selin, 2012). Furthermore, numerous studies have attributed the high atmospheric PAH concentrations in western North America to the biomass burning events that occurred in Russia/eastern Asian because the emissions were carried by trans-Pacific air (Genualdi et al., 2009a, 2009b; Tamamura et al., 2007).

Due to the rapid population growth and the associated energy demand, PAH emissions have become extensive in developing counties (Lang et al., 2008; Zhang and Tao, 2009). Based on the global emission inventories, China and India were the top two countries with the highest PAH emissions (Zhang and Tao, 2009; Devi et al., 2014; Mashi et al., 2010). The outflow of the PAHs from China has been comprehensively studied (Lang et al., 2008). The densely populated South Asia subcontinent is another important pollutant source region in the world (Ramanathan et al., 2005;

* Corresponding author. Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing, 100101, China.
E-mail address: wangxp@itpcas.ac.cn (X. Wang).

http://dx.doi.org/10.1016/j.atmosenv.2015.04.050
1352-2310/

Ali et al., 2014). In the winter–spring season, atmospheric brown clouds (ABCs) gather at the Indo-Gangetic Plain (IGP) on the south side of the Himalayas (Ram et al., 2011; Ramanathan et al., 2005). Black Carbon (BC), dust, sulphates, nitrates, fly ash, PAHs and other organic pollutants constitute ABCs (Bonasoni et al., 2010; Seinfeld, 2008). So far, BC has been widely detected in Himalayan glaciers and even in the lake sediment of the central Tibetan Plateau (TP) (Cong et al., 2013; Ming et al., 2008; Xu et al., 2009).

The IGP is embedded between the Hindu Kush and Himalaya Mountains. High PAH concentrations (Bhargava et al., 2004; Masih et al., 2010; Sharma et al., 2007; Singh et al., 2011) and aerosol loadings (Prasad and Singh, 2007) were previously reported for the IGP region. The south branch of westerly jet plays an important role in delivering pollutants during the non-monsoon season (October–May) (Bonasoni et al., 2010). Although the high peaks of the Himalayas can serve as a barrier to the direct transport of pollutants from South Asia to the TP, the Yarlung Tsangpo River valley cuts through the Himalayas and is considered a "leaking wall" that contaminates the southeast TP (Sheng et al., 2013; Cao et al., 2010). Jointly influenced by the Indian monsoon system and the south branch of the westerlies, the southeast TP is suggested as a receptor site for South Asian pollutants (Wang et al., 2014b; Xu et al., 2009). We conducted a continuous ambient air-monitoring program at the Southeast Tibet Observation and Research Station (STORS), operated by the Chinese Academy of Sciences. We previously found that seasonal variation of atmospheric DDTs and PCBs in the southeastern TP are strongly related to the cyclic patterns of the Indian monsoon (Sheng et al., 2013). In the present study, levels of PAHs and particulate matters (PM) and their seasonal variations at STORS were studied and we aim to address in detail the role of another climate pattern, namely the westerlies, on the PAHs/PM transport. This study will investigate (i) whether the transport of PAHs/PM over the Himalaya Mountains occurs and (ii) the locations of the important source regions for PAHs/PM in the southeast TP. A full understanding of these processes will help to elucidate how air circulation patterns affect contaminant transport.

2. Materials and methods

2.1. Climate of the sampling area

The TP, with an average elevation of 4000 m, is the largest and highest plateau in the world. The plateau climate is dominated by the monsoon system in summer and westerly winds in winter (Tian et al., 2007). During the winter months, the TP blocks the mid-latitude westerly jet stream and splits it into two currents (Xu et al., 2009). The north branch of the westerly winds is located at 40°N, around the region of the Altai and Qilian Mountains. From October to May, the south branch westerly jet stays mainly between 26°N and 28°N (the Himalaya–Hindu Kush range) (Han et al., 2008). After May, with the onset of the Indian monsoon system, this south jet moves dramatically northwards and integrates with the north jet (Han et al., 2008), and then the monsoon climate controls the south plateau (from July to September). In October, the Indian monsoon is fading away, and the south branch of the westerly winds rapidly retreats southwards and sweeps over the Himalaya-Hindu Kush area again (Kang et al., 2010). Therefore, the southeast TP is jointly influenced by the Indian monsoon system (in summer) and the south branch of the westerlies (in winter).

2.2. Sampling site and programs

The location of the sampling site (94.73°E, 29.77°N) is shown in Fig. 1. A low-volume air sampler was deployed on the observation field of the STORS (3300 m). This station is located at Lulang, southeast TP, which is 40 km west of the Yarlung Tsangpo Grand Canyon. Lulang is a very remote region with an extremely low population (0.3 person/square km). Two typical air circulation patterns over this region are depicted in the Supporting Information (SI, Figure SI-1). The sampling period was from November 2008 to September 2011, and the samples were collected every 2 weeks. The air stream passed first through quartz microfibre filters (QM-A, Whatman International Ltd) to collect the particulate phase and then through polyurethane foam (PUF) adsorbents, which retain the PAHs in gas phase (Smith et al., 2001). Before and after sampling, filters were weighed to obtain the total suspended particle (TSP) concentrations. An automatic weather station at the STORS recorded the meteorological parameters. Details regarding the sampling time and the average temperature/wind speed for each sampling period are given in Table SI-1.

2.3. Sample extraction and analysis

The collected quartz fibre filters and PUF were separately spiked with pryene-D10 and benzo(ghi)perylene-D12 as the recovery standards and Soxhlet extracted with dichloromethane (DCM) for 16 h. Then, the extracts were cleaned-up according to a previously published method (Liu et al., 2013). Briefly, extracts were concentrated and solvent-exchanged with hexane and then purified on a chromatography column (from the top to bottom: 1 g of anhydrous sodium sulphate, 2 g of 3% deactivated alumina, and 3 g of 6% deactivated silica gel). The column was eluted with 30 mL of a mixture of DCM and hexane (1:1). The elute was further cleaned using gel-permeation chromatography (GPC, containing 6 g of Biobeads SX3), and the samples were finally solvent-exchanged and concentrated in 20 μL of dodecane containing a known quantity of anthracene-D10 and perylene-D12 as the internal standards. The PAHs were analysed on a gas chromatograph (GC) with an ion-trap mass spectrometer (MS) (Finnigan Trace GC/PolarisQ) using a DB-5MS column (60 m × 250 μm, film thickness 0.25 μm); details on the gas chromatographic temperature program are given in text SI-1. The following fifteen compounds were measured and quantified: acenaphthylene (Acel), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (Bbf), benzo[k]fluoranthene (Bkf), benzo[a]pyrene (BaP), dibenz[a, h]anthracene (DahA), benzo[g, h, i]perylene (BghiP), and indeno[1,2,3-cd]pyrene (IcdP). These compounds are the "EPA16" without naphthalene.

2.4. Quality assurance/Quality control (QA/QC)

All analytical procedures were monitored using strict QA/QC measures. Laboratory blanks and field blanks were extracted and analysed in the same way as the samples. Acel, Ace, Phe and Ant were detected in the field blanks. Details on the compound concentrations for each field blank sample and the method detection limits (MDLs) are given in Table SI-2. The recoveries were between 65 and 105% for pryene-D10 and 85–120% for benzo(ghi)perylene-D12. These recoveries are in good agreement with previous studies (Liu et al., 2013; Wan et al., 2006). All of the reported values were field blank corrected (mean blank concentrations were subtracted) but not corrected for the recovery rates.

2.5. Satellite data

Moderate Resolution Imaging Spectroradiometer (MODIS) data retrieved from the Terra satellite has been widely used to investigate the spatial and temporal variations in aerosol particles over large scales (Remer et al., 2008). The MODIS science team has