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Influence of atmospheric circulation on the long-range transport of organochlorine pesticides to the western Tibetan Plateau

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ABSTRACT

Atmospheric circulation now is considered key to understanding global dispersion of organochlorine pesticides (OCPs). Many studies reported that the Indian monsoon and westerly wind played important roles on the transport of OCPs to the Tibetan Plateau. Previously, few observation campaigns have focused on the joint effect of Indian monsoon and westerly wind on long-range atmospheric transport of OCPs in a transition area of the Tibetan Plateau. In this study, air samples were collected for 7 months in the Ngari region, a westerly wind and Indian monsoon transition area in the western Tibetan Plateau. Westerly wind controls the climate of this transition area from March to May, and the levels of DDTs, HCHs, and endosulfans in the Ngari region were <10 pg/m^3, related to OCP transport by the upper-level westerly. From August to October, Indian monsoon and the airflow from north carried elevated concentrations of DDTs, HCHs, and endosulfans, and the levels of endosulfans in the Ngari region even reached hundreds of pg/m^3. In addition, due to the competition and conversion of Indian monsoon and westerly in June, the low levels of atmospheric OCPs reached minimum in the Ngari region. The seasonal shift of the westerly wind and the Indian monsoon resulted in complex seasonal patterns of the OCPs in this transition region, and due to the complex atmospheric circulations, the OCPs in the transition region originated from northwestern India and Northwest China (Xinjiang).

1. Introduction

Organochlorine pesticides (OCPs), including hexachlorobenzene (HCB), hexachlorocyclohexanes (HCHs), dichlorodiphenyltrichloroethanes (DDTs), and endosulfans are all considered persistent organic pollutants (POPs). Due to their persistence and toxicity, DDTs, technical HCH, and HCB have been banned for agricultural use since the 1980s in many countries (Breivik et al., 2004). For example, in China, the use of technical HCH and DDTs was banned after 1983 (Wei et al., 2007). However, DDTs remain in-use as anti-malaria precautions in tropical regions (e.g. India, western Africa) (UNEP, 2011a) and endosulfans and lindane (γ-HCH), are still currently in use in most of the countries (UNEP, 2009, 2011b). Despite of the source regions (Ozcan and Aydin, 2009; Pozo et al., 2006, 2008, 2011; Shunthirasingham et al., 2010), OCPs are still widely detected in remote regions to which they are transported (e.g. the Arctic, the remote ocean, and high mountains) (Bidleman et al., 2010; Choi et al., 2008; Gong et al., 2010; Su et al., 2008; van Drooge et al., 2004; Wania and Dugani, 2003; Xing et al., 2010). Long-range atmospheric transport, strongly influenced by atmospheric circulation, is one of the main pathways through which OCPs reach remote regions (Estellano et al., 2008; Huang et al., 2010; Li et al., 2007; Tian et al., 2009; Wania and Mackay, 1995, 1996, 1999).

The Tibetan Plateau, with its high altitude, is one of the big remote regions in world. The climate of the Tibetan Plateau is controlled by two atmospheric circulations: the westerly wind and the Indian monsoon (Yao et al., 2012; Zhang et al., 2009). In winter, the Tibetan Plateau blocks mid-latitude westerly wind, splitting the jet into two currents that flow south and north of the plateau. From May, with the Indian Monsoon onset, the westerly wind moves northward and the southern branch gradually breaks down (Ding, 1992; Kang et al., 2003). The monsoon climate dominates the southern Tibetan Plateau. Meteorological studies (Ding, 1992; Wang and LinHo, 2002) have found that the northern boundary of the Indian monsoon over the Tibetan Plateau has fluctuated between 30°N and 35°N, dependent on the relative strength of the westerly winds and the Indian monsoon. This "belt region" is regarded as the transition region between the westerly winds and the

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Indian monsoon. The northwestern Tibetan Plateau is a typical region influenced by westerly wind, while the southeastern Tibetan Plateau is monsoon influenced. Due to the complex climate system, complicated seasonal patterns of wet precipitation in the transition region have been found. For example, peak δ^{18}O values in precipitation occurred in June and September, related to the shifting influence of the westerly winds and Indian monsoon (Tian et al., 2001, 2007; Yao et al., 2013). Regarding the pollutants, the seasonal patterns of black carbon in ice cores from the northwestern Tibetan Plateau showed a distinct peak in winter and spring (Xu et al., 2009). Due to the influence of Indian monsoon in the southeastern Tibetan Plateau, gaseous POPs showed peak concentrations in summer (Sheng et al., 2013). Based on observation of the seasonal pattern of atmospheric POPs, many studies have suggested that both the Indian monsoon (Gong et al., 2010; Sheng et al., 2013; Xiao et al., 2010) and westerly wind (Cheng et al., 2007; Gai et al., 2014) are the main driving factors of POPs transport to the Tibetan Plateau. Wang et al. (2010) reported that the western Tibetan Plateau may be influenced by both Indian monsoon and westerly wind. However, few previous observations have aimed to investigate the influence of air circulation on the seasonal pattern and transport of POPs in the transition regions of westerly wind and Indian monsoon.

The Ngari region (28–35°N, 78–84°E) is located in the western Tibetan Plateau, which is surrounded by the POPs source regions of Pakistan (Nasir et al., 2014), northern India (Chakraborty et al., 2010, 2013; Pozo et al., 2011; Zhang et al., 2008), Central Asia (Zhao et al., 2013), and Northwest China (Jia et al., 2009; Liu et al., 2009). The westerly wind and the Indian monsoon, the main atmospheric circulations over the western Tibetan Plateau, may carry OCPs emitted in these source regions to Ngari (Wang et al., 2010). In the case of a weak Indian monsoon period, the invaded southward air flows (Chen et al., 2014; Li and Han, 2008) may carry pollutants emitted in Northwest China to the Ngari region.

In this study, air samples were collected from March to October 2010 using a high volume active sampler, and atmospheric OCPs were measured in the Ngari region. The objectives of the current study were to obtain the levels and seasonal pattern of atmospheric OCPs in the Ngari region, and to diagnose the sources of OCPs and investigate the influence of atmospheric circulation on long-range atmospheric transport of OCPs.

2. Materials and methods

2.1. Site description and sampling

An active air sampler was deployed at the Ngari Station for Desert Environment Observation and Research (NASDE), operated by the Chinese Academy of Sciences (33.39°N, 79.72°E, 4250 m asl), located in the western Tibetan Plateau (Fig. 1). The annual precipitation and average temperature at NASDE is only 69 mm and 0.5 °C, respectively (Hu et al., 2012), and the highest temperature occurs in July (Fig. S1 in Supplementary materials). Due to the high altitude, a low density of human activity occurs in this region (<0.5 person/km^2).

The sampling period encompassed March to June, and August to October 2010, with samples collected every 2 weeks. The air sampling was carried out in every alternative day for the 2 weeks. A total of 13 samples were collected in this study. Air samples were taken with a high-volume sampler with dual sampling modules, filter holder and glass cartridge. Particles were collected on a glass fiber filter with a 94 mm diameter (Whatman GFF, 1.6 mm). The filters were heated at 450 °C for 4 h before sampling. Gas-phase compounds were adsorbed on polyurethane foam (PUF) plugs (6 cm diameter and 5 cm long, density = 0.02 g/cm^3). PUF plugs were pre-cleaned by dichloromethane (DCM). Three PUF plugs without sampling were kept and mailed with the other plugs as field blanks, and five plugs were cut horizontally into front and back plugs for detecting possible breakthrough of chemicals. A total flow of about 1000 m^3 for each sampling period was obtained. After sampling, the filters and PUF plugs were wrapped in pre-cleaned aluminum foil (baked at 450 °C for 4 h) and stored in a tin container. The samples were stored at −20 °C prior to analysis.

2.2. Extraction, cleanup, and analysis

Both PUF plugs and filters were analyzed in this study. Each sample was transferred to a Soxhlet body and spiked with a mixture of surrogate standards [2,4,6-trichlorobiphenyl (PCB-30) and Mirex]. The samples were Soxhlet extracted using DCM for 24 h. The extracts were concentrated and solvent exchanged with hexane, prior to purification on a chromatography column. From top to bottom this consisted of 1 g anhydrous sodium sulfate, 2 g 3% deactivated alumina, and 3 g 6% deactivated silica gel. The column was eluted with 30 ml of a mixture

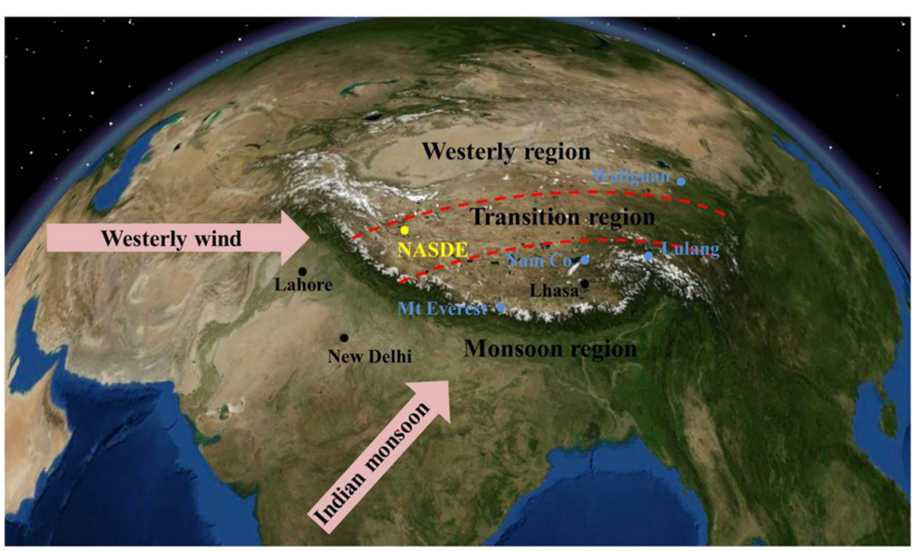

Fig. 1. Map of the sampling site. In this map, the yellow point is where the air samples of Nagri were collected (Nagri Station for Desert Environment Observation and Research, NASDE), and the blue points are the sampling sites in other remote regions in the Tibetan Plateau. The black points show the main cities in or around the Tibetan Plateau. The two red dotted lines divide the westerly region (up the lines), monsoon region (down the lines), and the transition region of westerly and monsoon (between the lines) in the Tibetan Plateau (Yao et al., 2013).