



Seasonal variation and source analysis of persistent organic pollutants in the atmosphere over the western Tibetan Plateau

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Abstract

Over the past few decades, the Tibetan Plateau (TP) region has become gradually contaminated by persistent organic pollutants (POPs). The picture regarding POPs is clear in the central and southern parts of the TP; however, few observational campaigns have focused on the western TP. To clarify the concentrations, seasonal trends and source regions of POPs in the western TP, a first study of POPs in Muztagh Ata (westerly region) and a long-term (5 years) monitoring program in Ngari (transect region influenced by both the Indian monsoon and westerly climate) were conducted. Except for hexachlorobenzene (HCB) and polychlorinated biphenyls (PCBs), relatively low POP levels were observed in the western TP. In Muztagh Ata, dichlorodiphenyltrichloroethanes (DDTs) showed higher concentrations in winter and lower ones in summer, whereas at Ngari, higher DDTs and hexachlorocyclohexanes (HCHs) concentrations were observed in summer as compared with winter. Source diagnosis indicated that Xinjiang and central Asia were the main source regions for POPs in Muztagh Ata and that westerly winds play a key role in transporting POPs from central Asia. No correlation was found between the height of the atmospheric boundary layer and the concentrations of POPs over the TP.

Keywords Persistent organic pollutants · Western Tibetan Plateau · Seasonality · Source regions · Atmospheric transport · Westerly wind

Introduction

The semi-volatility, persistence, and long-range atmospheric transport (LRAT) of persistent organic pollutants (POPs) determine their wide existence throughout the world, even the remote polar regions and high-elevation mountain areas are also inevitably affected by POPs (Arellano et al. 2011; Baek

et al. 2011; Choi et al. 2008; Kallenborn et al. 2013; Yu et al. 2015). Accordingly, interest has risen in the transport of POPs to remote areas. The European Monitoring and Evaluation Programme and the Arctic Monitoring and Assessment Programme found that POPs produced in the mid-latitude regions can undergo LRAT to the Arctic (Bailey et al. 2000; Gibson et al. 2016; Huang et al. 2007; Wang et al. 2010a). Therefore, the atmospheric circulation is important in the LRAT of POPs (Huang et al. 2010).

Similar to other remote regions, the Tibetan Plateau (TP) has limited human activity because of its high elevation and cold temperatures. However, being mainly controlled by westerly winds and the Indian monsoon (Gong et al. 2015; Wang et al. 2016a; Yao et al. 2012), which first pass through areas with high usage and emissions of POPs (e.g., Europe, central Asia, China's Sichuan Province, and India), the TP is therefore vulnerable to the effects of these pollutants. The Indian monsoon has been confirmed as the most important mechanism for the delivery of POPs to the southern TP from South Asia (Gong et al. 2010; Li et al. 2006a; Sheng et al. 2013), but the influence of westerly winds on the presence of POPs in the TP region remains less clear. In the western TP, Gong et al.

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(2015) has only done the short-term (7 months in 2010) POPs study in Ngari using active atmospheric sampler (AAS). This is insufficient for an understanding of the seasonal trends and source regions of POPs in the western TP.

Because of their high levels of efficiency and accuracy, high-volume AASs are widely used in the atmospheric monitoring of POPs. However, in the TP region, the sampling of POPs is very difficult because of its remoteness and harsh environment. As an alternative, passive air samplers (PASs) are simpler and less expensive than AASs and therefore have been highlighted as being more suitable for use in remote areas (Klanova et al. 2006; Ren et al. 2014; Tuduri et al. 2012), such as the TP. Long-term and continuous passive air sampling can reflect the seasonal trends of POPs well. For example, Ren et al. (2014) deployed polyurethane foam (PUF) PASs in the southeastern TP for a 3-year period and found higher dichlorodiphenyltrichloroethane (DDT) levels in the monsoon season than the non-monsoon season. However, studies of this type in the western TP remain limited.

The height of the atmospheric boundary layer (ABL) is believed to affect atmospheric circulation (Davy and Esau 2016) and the concentration of POPs (Bogdal et al. 2014; Dien et al. 2016). For example, the ABL height has a strong inverse relationship with gas-phase and fine-particle-phase polybrominated diphenyl ethers (PBDEs) (Dien et al. 2017). In the TP, the height of the ABL ranges from a few hundred meters to 5000 m (Chen et al. 2013; Li et al. 2006b; Yang et al. 2004; Zhang et al. 2003; Zuo et al. 2005). However, whether a correlation exists between the POPs and the ABL height over the TP remains unknown.

Therefore, in this study, AAS and PAS instruments were deployed in Muztagh Ata and Ngari (in the western TP), respectively, to quantify the concentration levels of POPs and investigate their seasonal trends and potential sources in this region. Then, through comparison with similar observations in Everest, Lulang, and Nam Co, the differences in the features of POPs in the west and other areas of the TP were analyzed, and the role of the westerly wind circulation on the transport of POPs to the western TP was explored. Lastly, the ABL height of the TP was investigated to test if significant relationships exist between ABL height and the concentrations of POPs in this region.

Materials and methods

Sampling sites and programs

Five sites in the TP were chosen for this study (Fig. 1). Muztagh Ata (3650 m) is located in the northwestern TP and close to central Asia, where the climate is relatively cold and dry. The average annual temperature is below 0 °C, and the lowest temperature is close to −15 °C in winter. Owing to the

dry climate and low vegetation coverage, the annual average wind speed is 3.57 m/s, with instantaneous wind speeds reaching 14 m/s. Ngari (4270 m), in the western TP, has similar climate characteristics to Muztagh Ata. The average temperature is only 0.5 °C (Hu et al. 2012). Wind speeds in this region are around 2.72 m/s, which is lower than that of Muztagh Ata. Nam Co (4730 m) is located in the central TP, where the climate is cold and dry in winter but warm and wet in summer. The average wind speed at Nam Co is about 3.11 m/s. Located at the southern TP, Everest (4276 m) is relatively warm and windy, with a mean annual temperature of 4 °C and an average wind speed of 4.49 m/s. Lulang (3326 m, in the southeastern TP) has higher temperatures (average of 5.38 °C) and lower wind speeds (1.42 m/s), because of its low altitude (3326 m) and large areas of forest (close to 50% of Lulang is covered by forest), respectively. A detailed summary of each site's meteorological parameters during the sampling period is provided in Table SI-1.

Despite of the difference regarding temperature and wind speed, precipitation situation in each site is also different. From east to west, the precipitation decreases, with the maximum in Lulang (880 mm/year) and minimum in Ngari (less than 200 mm/year). The differences among basic meteorological parameters suggest that climate of all these five sites is different. A common feature of the five sampling sites, because of their high altitude and harsh climate, is low population density and industrial and agricultural activity. The population density in five sampling sites is less than 10 person/km², far below the Chinese average value (130 person/km²). In Muztagh Ata, Everest, and Nam Co, the population density is about 7, 4.5, and 4 person/km², respectively. In Ngari and Lulang, the population density is less than 0.5 person/km², suggests that the local emission of pollutants can be ignored.

An AAS was installed at Muztagh Ata for 2-week periods. Because of its high sampling frequency, this was sufficient to meet the requirements of the potential source contribution function (PSCF) model to trace the source area—a method used widely in identifying the possible source regions of POPs (see Text SI-1 for more information on the PSCF model used in this study). Besides, owing to the instability of the power supply across the TP, duplicate PUF-PASs were deployed at Ngari, Everest, Nam Co, and Lulang for 2-month periods, the aim being to clarify the seasonal variation of POPs through long-term and continuous sampling. Field blanks were prepared to investigate the potential contamination during the handling, transport, and storage of the samples. In total, 209 (163 passive samples, 17 active samples, and 29 field blanks) air samples were collected across the five sites. Further detail regarding each sampling period is provided in Table SI-2, and Text SI-2 provides an in-depth description of the sample preparation procedures before the sampling was performed, as well as the storage methods after sampling.

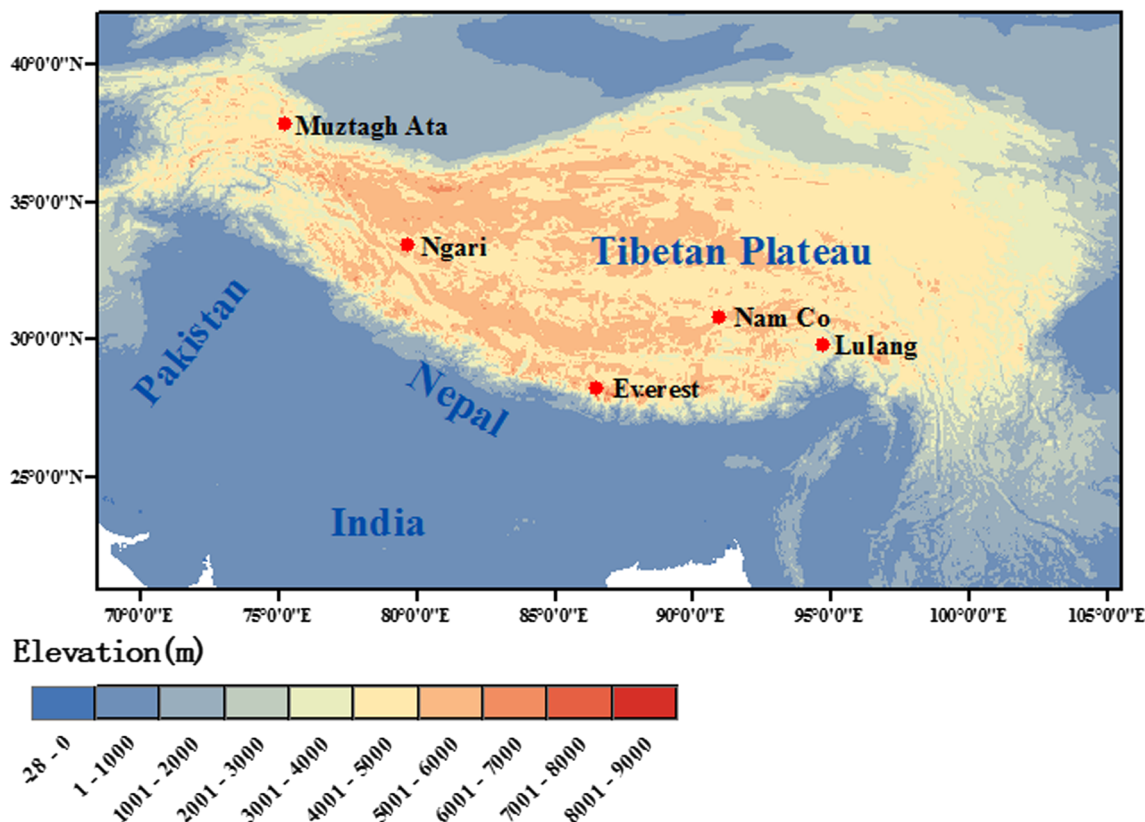


Fig. 1 Locations of the sampling sites

Sample extraction and analysis

The extraction and cleanup process for all samples are explained as follows. Firstly, the passive and active air samples were spiked with a mixture of 2,4,5,6-tetrachloro-*m*-xylene (TCmX) and decachlorobiphenyl (PCB-209) as recovery surrogates and were Soxhlet extracted with 200 mL of dichloromethane (DCM) for 16 h. Secondly, the samples were concentrated and solvent exchanged with hexane. Thirdly, the purification was finished on a chromatography column (from the top to bottom: 1 g of the anhydrous sodium sulfate, 2 g of the 3% deactivated alumina, and 3 g of the 6% deactivated silica gel), and the column was eluted with 30 mL of a mixture of DCM and hexane (1:1). Lastly, the samples were solvent exchanged and concentrated in 20 μ L of dodecane containing a known quantity of pentachloronitrobenzene (PCNB) as internal standards.

After extraction and cleanup, the extracts were analyzed using a gas chromatograph (GC) with an ion-trap mass spectrometer (MS) (Finnegan Trace GC/PolarisQ), using a CP-Sil 8CB capillary column (50 m, 0.25 mm, 0.25 mm) and operating under the MS–MS mode. Details on the gas chromatographic temperatures are given in Text SI-3. The following compounds were measured and quantified: hexachlorocyclohexanes (HCHs, including α -HCH, β -HCH, γ -HCH, and δ -

HCH), DDTs [*o,p'*-dichlorodiphenyldichloroethane (DDD), *p,p'*-DDD, *o,p'*-DDT, *p,p'*-DDT, *o,p'*-dichlorodiphenyldichloroethylene (DDE) and *p,p'*-DDE], hexachlorobenzene (HCB), and polychlorinated biphenyls (PCBs) (PCB-28, -52, -101, -138, -153, -180).

Quality assurance/quality control

All analytical procedures were performed under strict quality assurance and quality control measures. Procedure blanks were prepared to detect the potential contamination and loss in experiments. Procedure and field blanks were extracted and analyzed in the same way as the samples. Most of the POPs were not detected in the procedure blanks, indicating contamination was negligible during analysis, but trace amounts of POPs were detected in field blanks. The method detection limits for the PAS and AAS instruments were derived as the mean concentrations of the field blanks plus three times the standard deviation. The recoveries were 60–108% for 2,4,5,6-tetrachloro-*m*-xylene (TCmX) and 70–110% for PCB-209 in passive samples and between 70 and 125% for TCmX and between 73 and 120% for PCB-209 in active samples. All the reported values were field blank corrected (mean blank concentrations were subtracted) but not corrected for the recovery rates. The average relative standard deviation (RSD)

between the duplicate samples was 40% for POPs in all passive samples (see Table SI-3 for the average RSD at Ngari). This value is slightly higher than the results reported in Wang et al. (2016b) and Gong et al. (2017) using polystyrene-divinylbenzene copolymer resin (XAD)-based PASs.

PUF-PAS sampling rate

Passive air sampling is widely used around the world. The sampling rate (R , m^3/day) is needed to determine the volumetric air concentration (pg/m^3) of pollutants in the PAS. Details of the calculation of the site-specific sampling rates are included in Text SI-4.

The PAS is considered to have higher uncertainty than the AAS, because the former is more likely to be disturbed by variation in the environmental conditions; for example, the temperature and wind speed can influence the losses of deposition compounds (DCs). A correction based on DCs can minimize the effect of wind speed, meaning only the temperature needs to be considered (Moeckel et al. 2009). The sampling rate R was 5.16 ± 3.34 , 5.76 ± 1.95 , 5.48 ± 1.74 , and 5.19 ± 2.53 m^3/day at Ngari, Everest, Lulang, and Nam Co, respectively (Table SI-4). The rate was higher at Everest because of the higher wind speeds in this area than at the other sampling sites. The R values from this study were slightly higher than the value (4.28 m^3/day) at Sygera Mountain reported by Ren et al. (2014), which was also attributed to the influence of wind speed. Tuduri et al. (2006) found the sampling rates of PUF-PASs can increase at high wind speeds. Based on R , the concentrations of POPs at four sampling sites were obtained (Table SI-5b-e).

The concentrations were credible and comparable with those from AAS (Ren et al. 2014; Wang et al. 2017). In our previously published study, the data derived from PAS using R and concentrations directly produced by AAS was compared and good correlations were generally observed between two datasets (details about this comparison is presented in Text SI-5). This indicated that the R obtained in the current study would be proper to converting POPs sequestered in a PUF disk (pg/sample) to an ambient concentration (pg/m^3).

ABL height data collection

Data on ABL height were acquired from the European Centre for Medium-Range Weather Forecasts interim reanalysis (ERA-Interim) dataset (<http://apps.ecmwf.int/datasets/data/interim-full-mode/levtype=sfc/>). ERA-Interim is the latest global atmospheric reanalysis (Dee et al. 2011), which produces gridded data including a large variety of three-hourly surface parameters and six-hourly upper-air parameters covering the troposphere and stratosphere (Dee et al. 2011; von Engel and Teixeira 2013). In this study, the data used were regridded to a $0.5^\circ \times 0.5^\circ$ horizontal resolution, and the

monthly mean values of ABL height covering the entire sampling period were obtained. Also, the mean value of every 2 months in Ngari, Nam Co, Everest, and Lulang was calculated to match the period of the PAS sampling.

Results and discussion

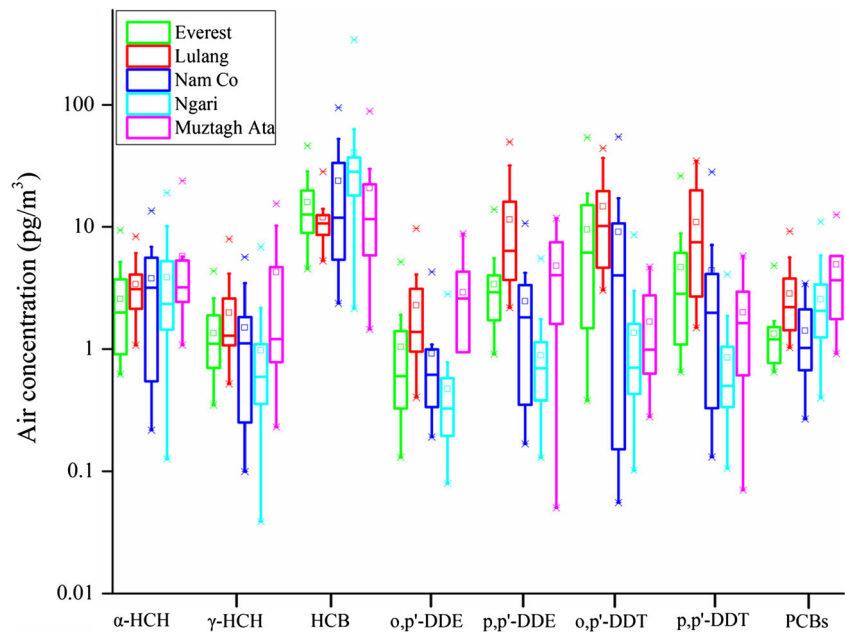
General comments on POP concentrations

The concentrations of POPs at every sampling point are presented in Table SI-5a-e. The data reported in Table SI-5 mainly included the concentrations of gas-phase compounds. For AAS sampling, the gas-phase and particles-phase compounds were adsorbed on PUF plugs and glass fiber filter, respectively. However, our previous study found that the concentrations of particulate POPs were negligible because of their very low values (Sheng et al. 2013). This may be due to the sampling sites were located in remote area, while urban particles are usually burdened by high concentrations of pollutants (Odabasi et al. 2015). Therefore, although the PAS collected both gas-phase and particle-phase POPs (Markovic et al. 2015), the low concentration of POPs in the particulate phase will exert less influence on the final POP concentrations.

The dominant chemicals in the atmosphere over the TP were HCB followed by DDT-class chemicals, i.e., *o,p'*-DDT, *p,p'*-DDT, and *p,p'*-DDE (Fig. 2 and Table SI-5), which is consistent with the results reported by Wang et al. (2016b). Also, the low molecular weight (LMW) PCBs were the abundant PCB congeners at every sampling site; the ratio between the sum of PCB-28, -52, and -101 to the concentration of total PCBs ($\Sigma_6\text{PCB}$) was $>70\%$. The similar results were found in air from urban sites, which were explained as the “urban fractionation effect” (Lu and Liu 2015; Wang et al. 2008). These high molecular weight (HWM) PCBs (PCB-138, -153, -180) are deposited in urban areas, and LMW PCBs can stay in the atmosphere for a long time and transport to other regions.

A comparison of the POP concentrations in this study with those recorded in other studies is presented in Table 1. The results in this study are comparable to other results from the TP (Wang et al. 2016b). Compared with other remote areas, the HCB concentrations over the TP found in this study are slightly lower than over the Antarctic (average value: 33.6 pg/m^3) (Khairy et al. 2016) and two to three times lower than over the Arctic (Hung et al. 2010), western Canada (Daly et al. 2007), and European mountain regions (van Drooge et al. 2004) (Table 1), although they are the highest concentrations among other studies in the TP. HCB is the most persistent chemicals in the global environment. Their average half-life is around 9 years (Barber et al. 2005) and is a “flyer” that can reach remote areas via LRAT and exist permanently in these places. Given that PUF disks can approach equilibrium with air for some of the more volatile compounds like HCB, the

Fig. 2 Box and whisker plot showing the general distribution of POPs at Everest, Lulang, Nam Co, Ngari, and Muztagh Ata (the lower and upper limits of the whiskers indicate the minimum and maximum; the lower and upper limits of the boxes indicate the 25th and 75th percentiles; the horizontal line in the box indicates the median; the small square in the box represents the mean; and stars denote outliers)



relatively low concentrations of HCB in the current study might be partly due to overestimation of the effective air volume. The HCH concentrations found in this study are akin to those over the Antarctic but lower than at other background sites around the world (Baek et al. 2011; Hung et al. 2010; van Drooge et al. 2004) (Table 1), which implies the remoteness of the TP. However, the DDT concentrations in this study, especially the two parent DDTs over the TP, are higher than those in the polar regions (Baek et al. 2011; Hung et al. 2010) and other remote areas (van Drooge et al. 2004). Additionally, the mean concentration levels of *o,p'*-DDT and *p,p'*-DDT in the southern TP (Lulang and Everest) are about two orders of magnitude higher than those in the Arctic (Table 1). This can be largely attributed to the closer proximity of Lulang to DDT source regions (South Asia).

In contrast with adjacent regions, the total concentrations of POPs (organochlorine pesticides (OCPs) + PCBs) in the TP are much lower than those in adjacent urban areas (one to two orders of magnitude), such as Chengdu in China (Pozo et al. 2009), India (Chakraborty et al. 2010; Zhang et al. 2008), Pakistan (Nasir et al. 2014), and Nepal (Pokhrel et al. 2018) (Table 1). These areas have a long history of POP use, especially India, where OCPs are still used today in agriculture and vector control (Chakraborty et al. 2010). Therefore, these urban areas may be potential sources of POPs in the TP.

Spatial distribution of POPs over the TP

Figure 2 presents the major POPs at each sampling site to illustrate the spatial distribution patterns. Higher levels of POPs were found at Lulang and Everest (southern TP), followed by Nam Co in the central TP; however, low

concentrations of POPs existed at Ngari and Muztagh Ata in the western TP. Therefore, the spatial distribution of POPs shows a decreasing gradient from the south and east to the west of the TP, which is similar to the result reported by Wang et al. (2016b).

In detail, higher levels of HCB were found at Nam Co and Ngari (Fig. 2), while DDTs were higher at Lulang, Everest, and Nam Co. The total PCBs (Σ_6 PCB) were high at Muztagh Ata, despite Muztagh Ata being a relatively clean site with low levels of POPs. The proportion of Σ_6 PCB accounting for total POPs at Muztagh Ata was found to be 20%, whereas proportions below 7% were found at other sampling sites in this study. These differences across the five sites may largely be attributable to their different sources. Besides, the different sampling sites are influenced by different atmospheric circulation systems, which also likely contributes to the spatial differences of POPs.

Seasonality of POPs

POPs over the TP derive mainly from LRAT driven by the atmospheric circulation (Gong et al. 2015; Sheng et al. 2013). The Indian monsoon in summer and the prevailing westerlies in winter influence the seasonal variation of POPs over the TP by carrying them from different sources. The seasonality of POPs was monitored at the five sites across the TP. Then, through comparing the seasonal variation at each site, the influence of the Indian monsoon and westerly winds on the characteristics of POPs over the TP was explored.

Muztagh Ata Westerly winds control the climate of Muztagh Ata. To clarify the situation regarding POPs in the atmosphere

Table 1 Comparison of the POP concentrations found in this study with those in the literature reported for other remote areas and their surrounding countries (units: pg/m³)

Sampling site	Sampling period	Sampler	DDT				HCH		HCB	PCBs	Reference
			o,p'-DDE	p,p'-DDE	o,p'-DDT	p,p'-DDT	α-HCH	γ-HCH			
This study											
Muztagh Ata	2015	AAS	2.90	5.00	1.65	2.07	4.40	4.20	17.91	9.70	This study
Ngari	2011–2016	PUF-PAS	0.47	0.89	1.35	0.85	3.87	0.97	32.04	2.55	
Lulang	2012–2015	PUF-PAS	2.29	11.44	14.62	10.90	3.40	1.99	11.93	2.85	
Everest	2012–2015	PUF-PAS	1.16	3.98	11.73	5.49	2.71	1.58	20.10	1.49	
Nam Co	2013–2015	PUF-PAS	0.92	2.46	9.09	4.41	3.78	1.50	23.79	1.41	
Previous studies in the TP											
Lulang	2008–2011	AAS	1.69	6.39	19	9.08	12.05	3.21	7.87		Sheng et al. (2013)
	2007–2012	XAD-PAS	1.20	2.00	12.90	5.20	2.50	0.60	11.20	0.80	Wang et al. (2016a, 2016b)
Ngari	2010	AAS	0.59	1.99	3.71	1.43	4.94	2.01			Gong et al. (2015)
	2007–2012	XAD-PAS	0.4	0.9	2.5	1.2	2.5	3.5	13.00	0.90	Wang et al. (2016a, b)
Everest	2006	AAS		5.10	5.10	3.70	19.20	7.70			Li et al. (2006a)
	2007–2012	XAD-PAS	0.20	0.80	5.00	2.50	3.40	1.90	18.30	0.80	Wang et al. (2016a, b)
Nam Co	2012–2014	AAS	1.20	1.90	3.60	1.20	4.00	2.10	20.00	2.50	Ren et al. (2017)
	2013	PUF-PAS	0.50	1.10	4.20	1.40	4.30	1.30	27.80		Wang et al. (2017)
	2007–2012	XAD-PAS	0.80	0.40	3.40	1.60	1.70	0.60	11.40	0.80	Wang et al. (2016a, b)
Muztagh Ata	2009–2012	XAD-PAS	0.10	1.30	0.80	1.20	2.60	0.40	26.80	0.40	Wang et al. (2016a, b)
Previous studies in other remote areas											
Alert, Arctic	2005	AAS	0.03	0.30	0.22	0.14	13	1.7	52		Hung et al. (2010)
King George Island, Antarctic	2005–2007	XAD-PAS	BDL	BDL	BDL	BDL	2.36	0.7	BDL		Baek et al. (2011)
Antarctic	2010	AAS	0.10	0.60		0.1	1.3	1.3	33.6		Khairy et al. (2016)
Rocky Mountains, western Canada	2003–2004	XAD-PAS		5.30		10	19.7	3.3	51		Daly et al. (2007)
High Tatras, European mountains	2001/2002	AAS		5.00		BDL	15	30	85		Van Drooge et al. (2004)
Previous studies in adjacent urban areas											
Cheng du, China	2005	PUF-PAS		BDL-56		BDL	145–176	68–142			Pozo et al. (2009)
New Delhi, India	2006–2007	AAS		110–1040	410–4430	100–1050	600–2330	1130–3400			Chakraborty et al. (2010)
Mumbai, India	2006	PUF-PAS		925.00	524.00	188.00	637.00	912.00			Zhang et al. (2008)
Lahore, Pakistan	2011	XAD-PAS	9–22	68–154	43–82	14–77	54–115	75–108			Nasir et al. (2014)
Karachi, Pakistan			7–19	24–116	6–66	3–57	6–61	27–66			
Kathmandu, Nepal	2014–2015	PUF-PAS	1–10	4–157	4–77	3–121	3–45	4–133	7–186	2.1–29.2	Pokhrel et al. (2018)
Pokhara, Nepal	2014–2015	PUF-PAS	1–9	8–306	3–64	6–540	1–7	3–23	4–28	1.6–16.6	

of this region, a full year of data was obtained from AAS measurements. Concentrations of DDT-class compounds at Muztagh Ata exhibited obvious seasonal fluctuation, with peaks occurring in September to February and valleys in May to July (Fig. 3a). The DDT levels were less than 10 pg/m³ in the high-concentration period (September–February) and even below the detection limit in the low-value period

(May–July) (Table SI-5a). In contrast to DDTs, the HCH values were high from April to June, and the highest peak appeared in April, while relatively low from January to March and from July to October (Fig. 3b). The high-value period is an important farming season (springtime) in the mid-latitudes of the northern hemisphere (Peltonensainio et al. 2009). Therefore, the high HCH values can be attributed

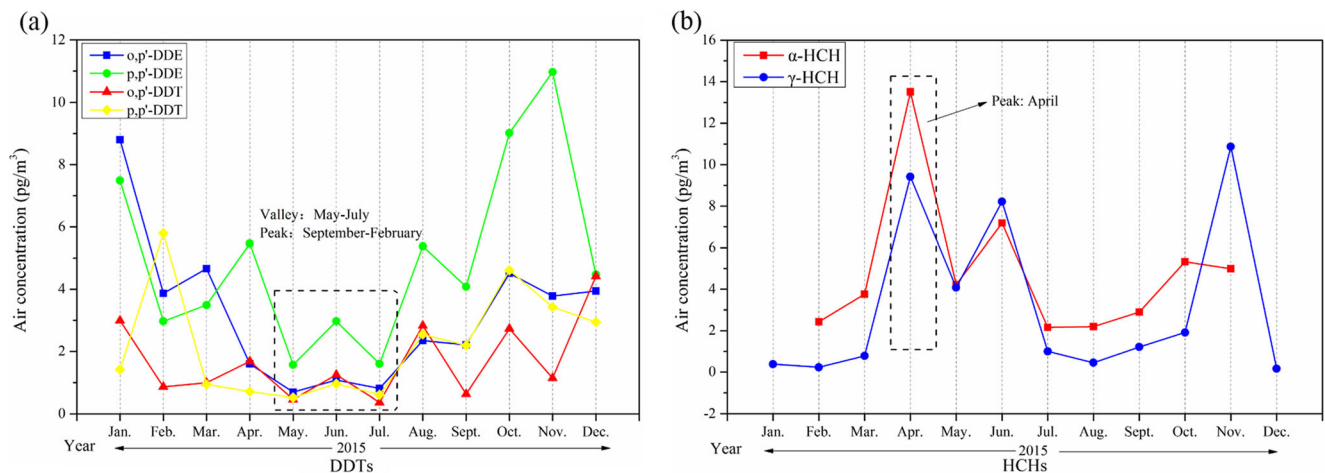


Fig. 3 Seasonal patterns of atmospheric **a** DDTs and **b** HCHs at Muztagh Ata

to the use of HCHs in agricultural production from spring. For HCB and PCBs, no obvious seasonal trends were found (Fig. SI-2 a, b).

Ngari This site is a westerly winds and Indian monsoon transition area in the western TP, for which 5 years of continuous sampling was carried out. DDT concentrations exhibited clear seasonality, with the higher levels occurring in the Indian monsoon season (May–September) and the lower levels occurring in the non-monsoon season (November–March) (Fig. 4a). A similar trend was also observed for HCHs (Fig. 4b). These seasonal variations of DDTs and HCHs demonstrated good repeatability and regularity during the 5-year monitoring period, which further confirms the credibility of the seasonal trends. Besides, different from DDTs and HCHs, the levels of HCB often peaked in the winter, especially in 2011 and 2012 (Fig. SI-3a). HCB mainly produced in industrial and agricultural use and combustion (Bailey 2001; Shin-ichi et al. 2001). At Ngari, there is an abundant release from biomass combustion in winter for heating. Similar results had been observed by AAS sampling in Lulang and Nam Co (Ren et al. 2014; Ren et al. 2017). Therefore, the high concentrations of HCB in winter may be attributable to local sources. Besides, the great PUF disk adsorption capacity during low temperature might also lead to the high value of HCB in winter. However, no consistent seasonality for PCBs is apparent (Fig. SI-3b).

Everest, Nam Co, and Lulang These three areas are mainly influenced by the Indian monsoon and therefore have higher concentrations of DDTs and HCHs during the Indian monsoon season and lower concentrations in the non-monsoon season (Fig. SI-4). Also, the levels during the high-concentration period were more than twice those in the low-value period (Table SI-5c, d, e). This trend is consistent with the results reported by Ren et al. (2014) in the southeastern TP.

Furthermore, Sheng et al. (2013) found the same seasonal change in DDTs at Lulang.

Based on the previous findings, we can conclude that Muztagh Ata has a different seasonality to Ngari, Everest, Nam Co, and Lulang, not only for DDTs but also for HCHs. Therefore, the POPs found at Muztagh Ata have different sources to the other sites. Muztagh Ata neighbors central Asia and Xinjiang, and it is possible that POPs transported to Muztagh Ata derive from these regions, which is an aspect we discuss further as follows. Ngari had a similar trend of change in POPs to Lulang, Everest, and Nam Co, indicating that POPs at Ngari may derive from the same sources as these three other sites.

Identification of the sources of POPs

Muztagh Ata

A PSCF model combines the concentrations of POPs with air mass trajectories to trace the sources of pollutants accurately. In this study, a PSCF model was used to identify the source regions of POPs at Muztagh Ata. The maps shown in Fig. 5 and SI-5 indicate that western Xinjiang and Tajikistan in central Asia were the main source regions of POPs at Muztagh Ata, with low levels from northern Pakistan, Kyrgyzstan, and Afghanistan. In Xinjiang, large quantities of DDTs and HCHs were used in agricultural production as insecticides before the 1980s, and POPs have been observed in water, sediments, and needles, mainly from historical application (Chen et al. 2011; Lei et al. 2015). The isomeric ratios of p,p'-DDT/p,p'-DDE in air can be used to evaluate whether residues are from historical usage (< 1) or fresh inputs (> 1). At Muztagh Ata, the ratios being less than 1 (average 0.54) (Table SI-8) further verified the DDTs there were mainly from historical emissions in its source areas. Tajikistan is another major source area of POPs for Muztagh Ata. This is an area of extensive historical

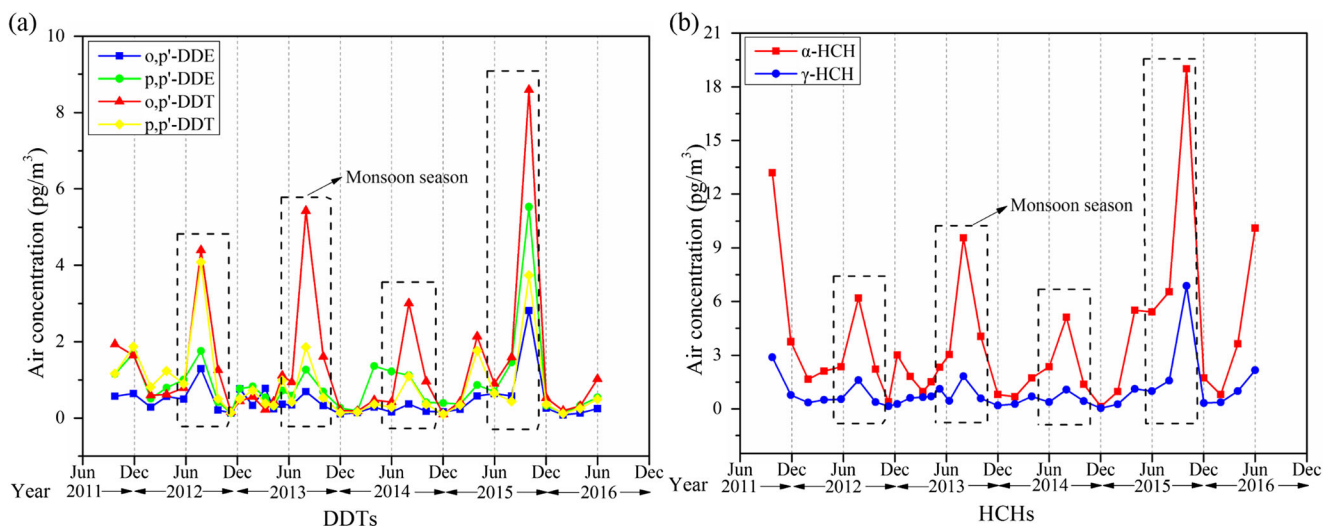


Fig. 4 Seasonal patterns of atmospheric **a** DDTs and **b** HCHs at Ngari

agricultural pesticide use, as well as large-scale burials of banned chlorinated insecticides. Greater than 10 ppm of OCPs have been found in the soil; plus, POPs have also been detected in plants and food (Barron et al. 2017). For PCBs, high PSCF values of LMW PCBs occurred in southern Kyrgyzstan and western Tajikistan (Fig. SI-5a), while HMW PCBs arriving at Muztagh Ata originated from western Xinjiang (Fig. SI-5b), close to the sampling site, which further implies that PCBs derive mainly from LRAT.

Mount Muztagh Ata is located in the eastern Pamirs and the western TP, where climate is dominantly controlled by westerly winds. Based on the previous ice core results, pollutants in ice core mainly came from anthropogenic emissions from European and central Asia areas (Li 1999; Wang et al. 2010b; Wang et al. 2014) and high concentrations of deposited pollutants in the glacier ice were closely associated with strong westerly winds (Wang et al. 2010b). In the present study, we calculated the monthly averaged westerly winds index (WWI) from the NCEP/NCAR reanalysis 1 (the range is 30–45° N, 60–75° E) (Table SI-7) and correlated the WWI with DDT and HCH concentrations in air of Muztagh Ata. Positive correlations were found (Figure SI-6), which demonstrated that input of pollutants in Muztagh Ata is truly related to the strength of westerly winds.

Ngari

For Ngari, the source areas of POPs have also been traced in previous studies using the same PSCF model (Gong et al. 2015). The results displayed that northwestern India and Xinjiang were the main sources of OCPs (Gong et al. 2015). Moreover, similar isomeric ratios indicate the same sources of POPs. Ngari has similar p,p'-DDT/p,p'-DDE and o,p'-DDT/p,p'-DDT values as Everest, Lulang, and Nam Co (Table SI-8),

and India is the major source region of POPs for these three locations (Ren et al. 2017; Sheng et al. 2013; Wang et al. 2007). Therefore, the results confirm that India is one of the sources of POPs at Ngari. Besides, the values of p,p'-DDT/p,p'-DDE and o,p'-DDT/p,p'-DDT are comparable to northern Pakistan (Nasir et al. 2014) and Nepal (Pokhrel et al. 2018) (Table SI-8), indicating a proportion of the POPs at Ngari derive from these two regions. Pokhrel et al. (2018) and Gong et al. (2010, 2015) also uncovered evidence that POPs from Pakistan and Nepal can be transported to the TP.

Comparing Muztagh Ata with Ngari, the same source was found in Xinjiang. While some POPs at Muztagh Ata derive from central Asia, India is the dominant source of POPs at Ngari. The difference of source regions mainly attributes to the different atmospheric circulation. Muztagh Ata is influenced by westerly winds throughout the year. However, Ngari can be regarded as a transition area between the Indian monsoon and westerly winds, meaning that the transportation of POPs takes place under the combined effects of these two atmospheric circulations (Gong et al. 2015).

Correlation between ABL height and atmospheric POP concentrations

The height of the ABL is considered as a key parameter influencing the sources, dispersion, and concentrations of pollutants (Collaud Coen et al. 2014). An inverse relationship between the ABL height and the concentrations of BDE-47, -99, and -183 has been found in Japan by Dien et al. (2017). It is therefore worth studying whether a correlation also exists between the ABL height and the other pollutants, like DDTs and HCHs. Besides, different from the local emissions in Japan, POPs in the TP derive mainly from LRAT, and the TP is regarded as a “cold trap” or “convergence zone” for

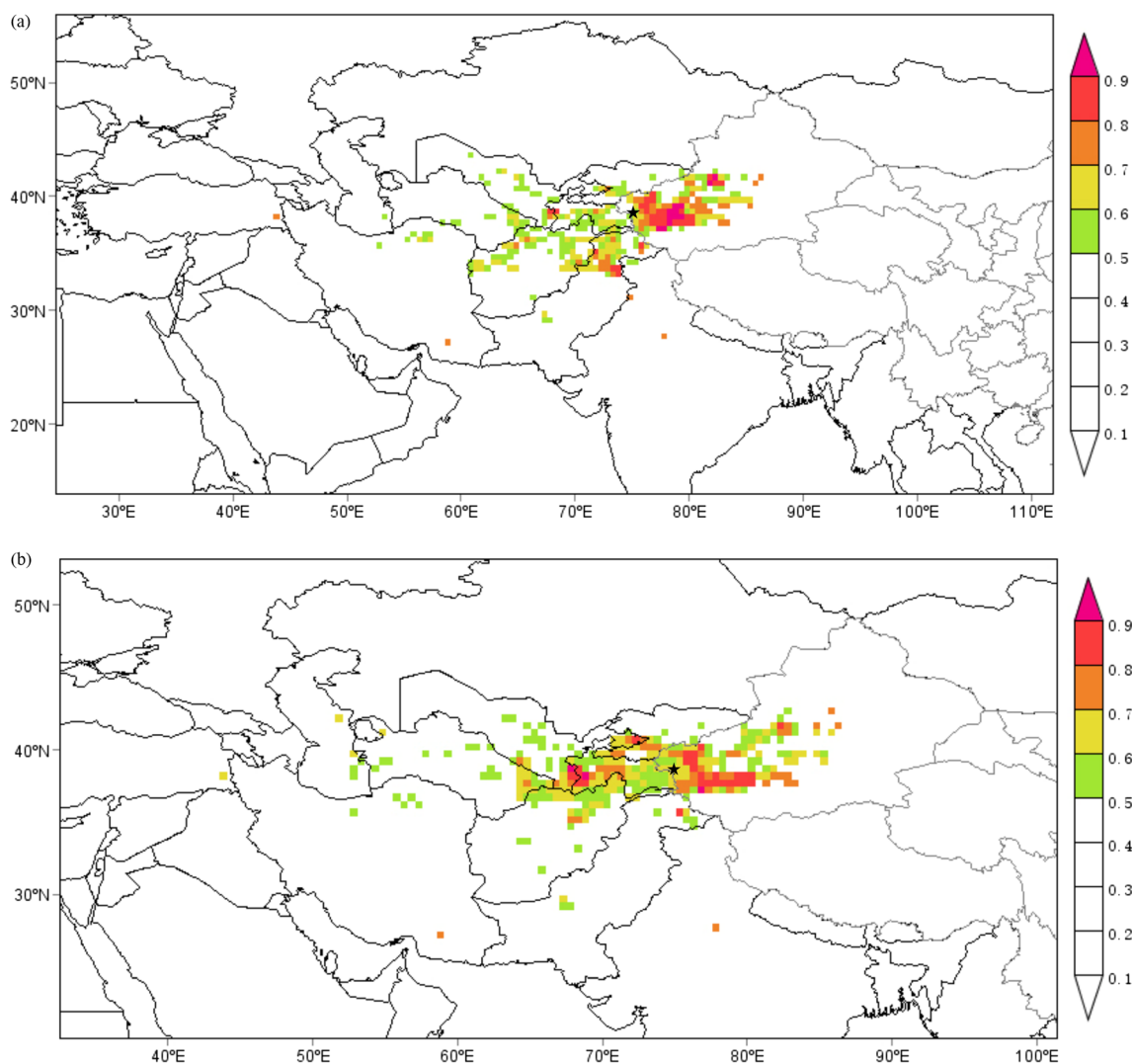


Fig. 5 PSCF maps for **a** DDTs and **b** HCHs at Muztagh Ata

POPs (Chen et al. 2009; Wang et al. 2016a). Whether such correlation is present over the TP is also of interest.

In the sampling period, the heights and seasonal change of the ABL were acquired. The highest ABL was found at Ngari (378–1764 m; median 1082 m), followed by Nam Co (586–1455 m; median 903 m) (Fig. SI-7f). Similar ABL heights were found for Muztagh Ata, Everest, and Lulang, at 240–918, 380–974, and 367–797 m, respectively (Fig. SI-7f). This is in agreement with the results from the central TP (700 and 940 m in the dry and wet season, respectively) reported by Song et al. (2006), but the highest value was lower than the results reported by Chen et al. (2013) (5000 m) and Li et al. (2006b) (3888 m). Figure SI-7 (a–e) indicates the seasonal trends of the ABL at the five sites. The monthly mean ABLs during the summer half of the year (April–September) were higher than those during the winter half of the year (October–March) at Muztagh Ata and Ngari (Fig. SI-7a and b). Other

studies have also pointed out that the highest ABLs appeared in summer and the lowest values appeared in winter over land (Dien et al. 2017; Pal and Haefelin 2016; Yang et al. 2013). However, the peak ABL values occurred in the spring, before the monsoon season, at Everest and Nam Co (Fig. SI-7c and d); and no obvious seasonal trend was found at Lulang (Fig. SI-7e).

Using Pearson correlation analysis on the concentrations of POPs and the ABL height, no consistent significant relationship was found at the sampling sites (Table SI-9). In Dien et al. (2017), the ABL height only had an inverse relationship with gas-phase and fine-particle PBDEs and demonstrated weak correlation with coarse-particle PBDEs. Also, the correlation was more likely to be found between gas-phase POPs and the ABL. Accordingly, we tried to find a correlation between the ABL and POPs using the data from Gong et al. (2015) at Ngari and Sheng et al. (2013) at Lulang, which were obtained

using AASs and only included gas-phase POPs. However, the correlation still could not be found for gas-phase POPs (Table SI-9), meaning there are no consistent correlations between the ABL and POP concentrations over the TP, indicating that ABL height is not the key factor influencing concentrations of POPs in the TP.

The inconsistency between the results of this study and those reported by Dien et al. (2017) possibly stems from the direction of air–surface exchange. In the TP, POPs mainly derive from LRAT; soil, forest, grass, and lakes have a strong ability to intercept and store POPs; and the dominant flux direction for POPs is deposition from the air to the surface (Ren et al. 2014; Ren et al. 2017; Wang et al. 2012; Wang et al. 2015; Wang et al. 2016a). On the other hand, air transport to the TP would be high under turbulent atmospheric conditions when winds are high and ABL is also high; and LRAT in air would be slow under the opposite conditions with stable or low ABL height. Combined with the deposition of POPs from air to surface and turbulent atmospheric conditions, they may explain the lack of correlation of POP concentrations in air (C_{air}) and ABL height. Contrary to this, in urban areas, POPs from local sources and pollutants follow an upward diffusion process, especially for gas-phase and fine-particle-phase compounds (Bentzen et al. 2010). Therefore, in this process, a high ABL has a dilution effect for POPs and thus led to low C_{air} . Taking all the above into account, we conclude that different levels of correlation between ABL height and POP concentrations may occur in different areas and for different compounds.

Conclusion and implications

This study confirms that POPs over the western TP are associated with westerly wind-driven atmospheric transport. Source diagnosis indicated that Xinjiang and central Asia are the major contributors of POPs to the western TP. Clear correlation between ABL height and POP concentrations was not found over the TP, despite being identified in other regions. Relevant research on POPs in the source areas (Xinjiang and central Asia) is lacking. Even the Global Atmospheric Passive Sampling network has no sampling sites deployed in these regions. Therefore, future work should focus on studying POPs in Xinjiang and central Asia. As the source regions of POPs in the western TP, more knowledge regarding these regions will provide more comprehensive evidence about the transport of POPs to the TP.

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