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Review

A review of current knowledge and future prospects regarding persistent organic pollutants over the Tibetan Plateau

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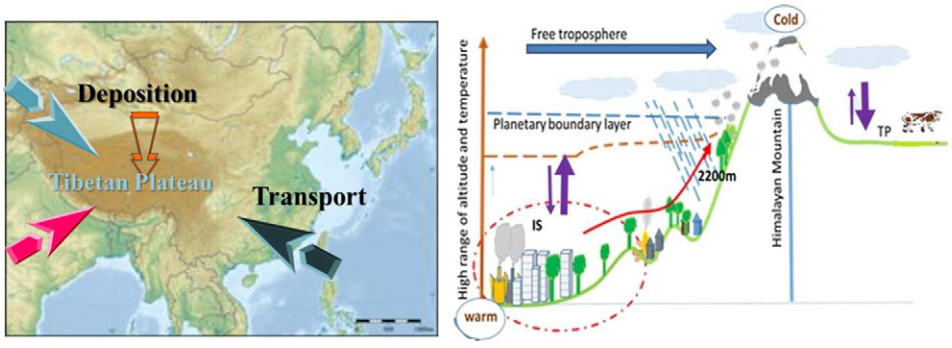
HIGHLIGHTS

- Levels of legacy and selected emerging persistent organic pollutants (POPs) over the Tibetan Plateau are reviewed.
- Atmospheric transport pathways for POPs are summarized.
- Factors affecting the capacity of soil and vegetation to accumulate POPs are elucidated.
- Data gaps and future perspectives regarding POPs over the Tibetan Plateau are discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Since the turn of the century, our understanding of the quantities, transport pathways, and fate of persistent organic pollutants (POPs) over the Tibetan Plateau (TP), the largest and highest plateau on Earth, has greatly enhanced. We begin in this article by reviewing the available literature on the levels of POPs over the TP. In general, the levels of most POPs are similar or lower than values reported for other background regions. However, dichlorodiphenyltrichloroethane (DDT) and hexachlorocyclohexane (HCH) levels in air and soil far exceed those measured in other mountainous areas. The East Asian monsoon, Indian Monsoon and westerly winds are responsible for the long-range atmospheric transport (LRAT) and arrival of POPs over the TP. Surface soil and vegetation act as "final sinks" for DDTs and other high molecular weight POPs. Linked to the continuous use of POPs in surrounding counties, LRAT and "cold trapping" by the TP can happen following emission–transport–deposition events, leading to the enrichment of POPs in the TP environment. Bioaccumulation of DDTs and high chlorinated PCBs have been found in Tibetan terrestrial and aquatic food chains, and newly emerging compounds such as polyfluoroalkyl substances and hexabromocyclododecanes have been widely detected in wild fish species. The corresponding ecological risks should be of great concern.
Climate change, such as increased temperatures and changing coverage of snow and glaciers, has the potential to affect the behavior and distribution of POPs. Therefore, long-term monitoring data are required. Ineffective regulation regarding POPs has been reported for countries in South Asia, emissions patterns, the outflow of POPs, and their seasonal and inter-annual variability should therefore be clarified.

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Estimating the loading of POPs, as well as how they move, within the TP, especially under the impact of glacial melt and global warming, should be a priority.

Contents

1. Introduction

Though mountain ecosystems are remote, they are more often than not surrounded by populated areas home to much anthropogenic activity. Thus, these ecosystems are no longer a pristine environment, free of anthropogenic contaminants, such as persistent organic pollutants (POPs) (Carrera et al., 2002; Daly et al., 2007; Daly and Wania, 2004; EscartÍn and Porte, 1998; Jaward et al., 2005; Morrissey et al., 2005; Schmid et al., 2011; Usenko et al., 2007; Wania and Westgate, 2008). Accordingly, interest has risen regarding the identification and quantification of organic contaminant levels in mountain regions (Daly and Wania, 2004; Wania and Westgate, 2008). POPs, such as organochlorinated pesticides (OCPs), polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs), have been observed at high altitudes in Europe (the Alps) (Arellano et al., 2011; Bogdal et al., 2010; Domine et al., 2007; Fernández et al., 2003, 1999; Finizio et al., 2006; Gabrieli et al., 2010; Gasic et al., 2009; Hafner et al., 2005; Hansen et al., 2006; Herbert et al., 2005; Hong et al., 2009; Levy et al., 2009; Roland, 2006; Villa et al., 2006) and western North America (the Rocky Mountains) (Bartrons et al., 2011; Bizzotto et al., 2009; Davidson et al., 2002; Hageman et al., 2006; Klasmeier et al., 2005; Landers et al., 2010; Waite et al., 2005).

During 2001–2016, a series of POP studies were conducted in the Tibetan Plateau (TP) region, which, being the highest and most extensive plateau on Earth, is often referred to as "the third pole of the world". The TP has an area of over 2.5×10^6 km^2 and an average elevation of >4000 m. Generally, due to its sparse human population and minimal industrial activities, the TP environment remains relatively pristine. However, there are three up-wind air masses that act upon the TP: one moving from west to east and pluming over countries in Europe and central Asia; another consisting of maritime air from the Indian Ocean, which comes across the Indian subcontinent and Bay of Bengal before reaching the south side of the TP; and a third from southeast to west deriving from the lower terrain of China, such as Sichuan basin and Yangtze Plain. The TP is surrounded and in close vicinity to areas with relatively much higher population density (such as Sichuan Province, China, and the Indian subcontinent), as well as intense agricultural and extensive industrial activities (i.e., the Indo-Gangetic Plain on the south side of the Himalaya, as well as the countries of Europe). The TP is therefore an ideal place to investigate the mechanisms of remote pollution sources and the atmospheric transport of POPs.

Owing to their semi-volatility, POPs can migrate from relatively warmer regions to colder climates through repeated evaporation, atmospheric transport, and deposition (Gouin et al., 2004). This process has been termed "cold condensation" or "grass-hopping". High mountains are usually cold and regarded as traps, reservoirs or sink for POPs. Consequently, the accumulation of POPs has the potential to pose problems for alpine ecosystems, which is the primary reason for studying organic contaminants in such regions. More specifically, one may ask: does the TP act as a cold trap for POPs? And what role does the TP play in the global cycling of POPs?

Melting glaciers have been shown to be a major source of semi-volatile POPs for sub-alpine lakes (Blais et al., 2001a, 2001b; Schmid et al., 2011). Glaciers, which cover large parts of the northern polar regions, as well as mountainous areas such as the Alps, Canadian Rockies and the TP, represent the largest reservoir of fresh water on Earth, and are

undergoing massive change through melting. Given that melting snow and glaciers over the TP supplies water for approximately one sixth of the world's population (Loewen et al., 2005), POP contamination of snow, ice, lake water and glaciers in the TP is of great concern because of its potential impact on drinking water supplies for people residing in adjacent lowland areas.

In addition, POPs in regions in and around Tibet have the potential to affect the mountain ecosystems themselves. Aside from the bioaccumulation of POPs in aquatic ecosystems, which may be accelerated by the greater longevity and slower growth of fish species in lakes, biomagnification and bioaccumulation through terrestrial food webs may pose considerable threats to predators. Local inhabitants may suffer from POP-related risks by consuming milk, butter and fat from grazing animals.

Thus, the study of POPs in the TP region is of particular interest, not least because of the possible amplification of certain contaminants in this region and their potential current and future impacts. Given the importance of this topic, the present paper compiles all existing information on POPs over the TP, paying particular attention to the levels of contaminants, their distributions, sources and transport pathways, as well as the related air–surface exchange processes and environmental issues. In doing so, the intention is to assist in our understanding of the mechanisms operating at large scales and the influence of various environmental parameters. We also identify knowledge gaps and suggest further avenues of research for the future.

2. POP concentrations over the TP

2.1. Air

2.1.1. Monitoring stations and programs

For global-scale monitoring programs, such as UNEP (the United Nations Environment Programme), the Arctic Monitoring and Assessment Programme (AMAP) and the European Monitoring and Evaluation Programme (EMEP), levels of pollutants in the air are particularly important. This is because such data are necessary when it comes to improving our understanding of atmospheric transport and validating atmospheric POP transport models (Pozo et al., 2009). The atmospheric monitoring of POPs traditionally relies upon high volume active air samplers (AASs). Even though it is often logistically difficult to deploy AASs at high altitudes, taking advantage of the Tibetan Observation and Research Platform (TORP), atmospheric POP concentrations have been reported for several sites on the TP (Fig. 1).

Short-term monitoring has been conducted at Waliguan Baseline Observatory [WBO, northern Tibet (Cheng et al., 2007)], Mt. Everest [southern Tibet (Li et al., 2006)], Gar [western Tibet, (Gong et al., 2014)], and Lhasa [central Tibet (Gong et al., 2010; Li et al., 2008)]. A relatively long-term observational campaign (starting in 2008) was implemented in southern Tibet [Lulang—a site close to the Yarlung Tsangpo Grand Canyon (the rift valley cutting through the Himalaya); Fig. 1]. Moreover, complementary AAS programs are currently being conducted at Muztagata, western Tibet, and at Namco, central Tibet (TORP; Fig. 1).

Aside from monitoring using AASs, passive air samplers (PASs) have also been developed and used for monitoring POPs over the TP (Wang et al., 2010a). PASs can be simple and cheap to use, and are therefore suitable for gaining a picture of the levels of POPs at larger scales, such as the whole plateau scale. To date, three types of PAS instruments have been deployed, using different sampling frequencies and spatial scales. Polyurethane foam (PUF) disk PASs tend to be used for shorter deployment periods [e.g., bi-monthly (Ren et al., 2014)], whereas XAD-resin PASs have been deployed for up to a year in determining long-term trends (Wang et al., 2010a). In 2007, based on XAD-resin PAS measurements, a passive air monitoring network comprising 16 sampling sites across the TP was established (Wang et al., 2010a). This network provides data on the levels of POPs at a large spatial scale, offering insights into both the spatial and temporal trends of POPs (Wang et al., 2016b). In another approach, a flow-through PAS was deployed at a remote research station located close to Nam Co Lake in the central TP (Xiao et al., 2010). Although this sampler use a flow-tube and has higher uptake rate as compared with PUF-PAS and XAD-PAS, it hasn't been widely used.

2.1.2. POP concentrations

2.1.2.1. Dichlorodiphenyltrichloroethane and hexachlorocyclohexane. Measurements of POPs in mountain air over the TP by both AAS and PAS instruments are summarized and compared with the results of other mountain regions in Table 1. The concentrations of hexachlorocyclohexanes (HCHs)

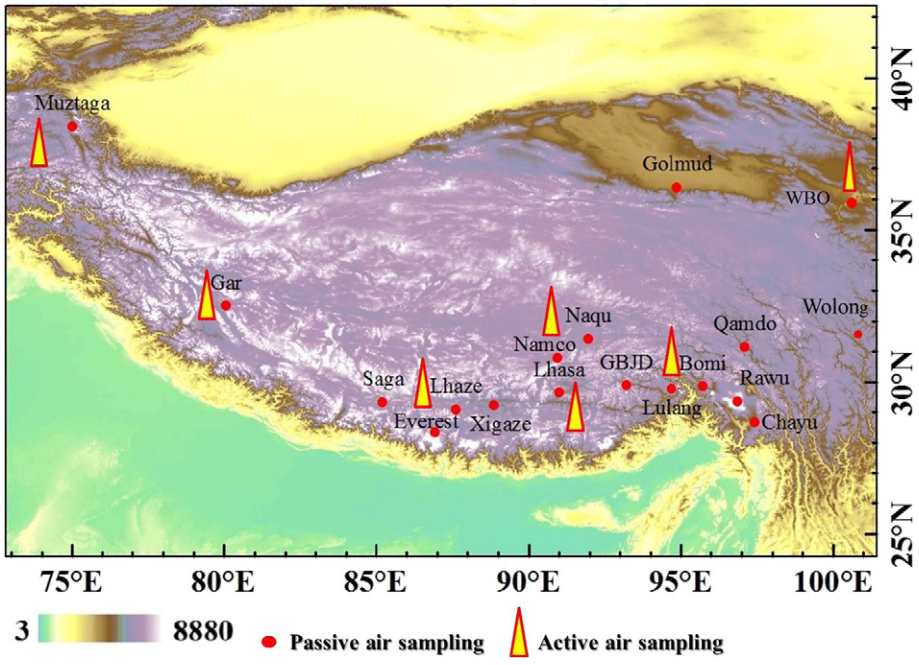

Fig. 1. Map showing the sampling sites of the Tibetan POP monitoring program (TORP) across the TP. The values in the legend are in meters.

Table 1
Comparison between the results of air sampling programs and various studies from remote areas.

Location	Date	Elevation	Concentration pg m^{-3}							Reference
			α-HCH	γ-HCH	HCB	o-p′-DDT	p-p′-DDT	∑PCB	∑PBDE	
Active air sampling										
WBO	2 April–23 May 2005	3816 m	21–350	40–250	4.6–51	17.9	4.42			Cheng et al. (2007)
Everest	May 31–June 11, 2002	4400 m–4976 m	8.7–25.3	4.0–9.4	4.8–12.6	ND-7.3	ND-5.3			Li et al. (2006)
Lulang	November 2008–September 2011	3330 m	0.6–51.0	BDL-7.1	0.05–27	BDL-60.5	BDL-33.6	BDL-16.7[a]		Sheng et al. (2013)
Gar	March–June, August–October 2010	4250 m	1.5–10.2	0.5–4.3		0.1–8.8	0.1–3.9			Gong et al. (2015)
Lhasa	August 6–August 12, 2006	3600 m	30–89	84–272	48–383	25–769	5–705			Li et al. (2008)
Lhasa	August–November 2006, June–July 2007	3600 m	BDL-9.2	3.7–20.9		BDL-34.3	BDL-9.7			Gong et al. (2010)
Passive air sampling										
TORP (XAD-PAS)	2007–2008		0.1–16.6	ND-18.9	2.8–80.0	1.9–39.4	0.6–12.7	1.8–8.2[a]	0.1–8.3	Wang et al. (2010a)
TORP (XAD-PAS)	2008–2012		0.1–17.7	0.1–20.1	3.0–85.0	0.1–44.5	0.1–26.1	0.1–3.9[a]		Wang et al. (2010b)
Lulang (PUF-PAS)	2008–2011	3330 m–4400 m	0.23–4.4	0.08–1.9	BDL-13.2	0.59–22.1	0.27–14			Ren et al. (2014)
Namco (flow-through PAS)	October 2006–February 2008	4700 m	5.7–91	1.52–15.8	9.69–25.2	0.73–11.8	0.66–6.63		0.83–5.2	Xiao et al. (2010, 2012)
Other mountain region										
Rocky Mountains	2003–2004		8–47	2–8	21–149		0.4–45			Daly et al. (2007)
Central Pyrenees (AAS)	2001/2003		2–25	5–91	36–98			16–70[b]		Van Drooge et al. (2002)
High Tatras (AAS)	2001/2002		7–34	11–86	62–119			23–64[b]		Van Drooge et al. (2002)
Lys Valley (PUF-PAS)	2003			0.86–4.8	28–46			7.5–52[c]		Jaward et al. (2005)
Arctic										
Alert (AAS)	2005		5.2–33	0.67–3.7	0.06–120			2–38[d]		Hung et al. (2010)

[a] Σ6PCBs.
[b] Σ15PCBs.
[c] Σ8PCBs.
[d] Σ7PCBs.

and dichlorodiphenyltrichloroethanes (DDTs) over the TP are generally higher than over other high-altitude mountain regions (Table 1). This may be due to the closer proximity of the TP to source regions of HCHs and DDTs. For example, γ-HCH concentrations are unusually high in WBO, especially when the prevailing air mass is from Russia and Kazakhstan (Cheng et al., 2007). Similarly, the concentrations of DDTs over the southeastern TP present similar levels to those over the Indian wetlands, and are apparently higher than those of other mountain areas (Table 1). The neighboring regions of the Indian subcontinent, central Asia and China have used DDTs and HCHs extensively (Santillo et al., 1997); therefore, a dominance of DDTs and HCHs in the atmosphere over the TP is reasonable and expected. Both AAS (Sheng et al., 2013) and PAS results (Wang et al., 2010a, 2016b) indicate that the atmosphere over southern Tibet receives a considerable contribution of DDTs. This suggests that AAS and PAS programs complement one another and provide robust results regarding the concentrations and distribution patterns of POPs.

2.1.2.2. Polychlorinanted biphenyls. Total polychlorinated biphenyl (PCB) concentrations are several pico-grams per meter cubed, which is comparable to concentrations reported from the Arctic but lower than those from mountain regions in Europe. The dominant PCB congeners are PCB 28, 52 and 101 (Wang et al., 2010a; Sheng et al., 2013; Ren et al., 2014). The heavier congeners with lower vapor pressures (PCB 138, 153 and 180) are generally below detection limits. A feature of note is the presence of major tetra-chlorinated congeners in air, which is similar to the situation observed over rural sites in India, where there is an abundance of tetra-PCBs. The reason for this presence of tetra-PCBs in the atmosphere over the TP has been attributed to their greater atmospheric transport potential (Wang et al., 2010a).

2.1.2.3. Hexachlorobenzene and polybrominated diphenyl ethers. The concentrations of hexachlorobenzene (HCB) reported by either AAS and PAS instruments are, overall, lower than the average (52 pg m^{-3}) for the Arctic region. There is, however, greater variability across the TP, suggesting the presence of some primary emissions, possibly attributable to inefficient combustion caused by low oxygen levels over the TP (Wang et al., 2010a; Sheng et al., 2013; Ren et al., 2014). Only a few studies have reported the concentrations of polybrominated diphenyl ethers (PBDEs) in the air over the TP. The concentrations of total PBDEs in air range from 0.10 to 8.3 pg m^{-3} (Wang et al., 2010a; Xiao et al., 2010, 2012), and this concentration range is one order of magnitude lower than that reported for Arctic Alert (1.2–55 pg m^{-3}; Hung et al., 2010); atmospheric PBDE concentrations over the TP are among the lowest values ever reported.

2.2. Soil

2.2.1. Soil type and POP concentrations

Previous studies have established that soils play an important role in receiving POPs from the atmosphere (Dalla Valle et al., 2005; Meijer et al., 2002; Nam et al., 2008a; Schuster et al., 2011). Background soils solely receive inputs of POPs via atmospheric deposition and have been shown to contain the bulk of the POPs inventory in terrestrial ecosystems. Early in 2001, Fu et al. (2001) reported the concentrations of DDTs and HCHs for Tibetan soil samples that were collected from 1993 to 1994. Very low DDT and HCH concentrations (from <Below Detection Limit to 2.8 ng/g and from 0.18 to 5.38 ng/g, respectively) were observed in their study. However, limited by quality control procedures, instrumental methods (electron capture detector), and only eight samples having been measured, this study was unable to provide detailed information regarding POP residues in soil.

According to the climate and vegetation zones over the TP (Fig. 2), soil can be classified into eight groups: mountain forest; alpine-burozems; sub-alpine scrub-meadow; alpine meadow; mountain shrubby steppe; alpine steppe; sub-alpine desert; and alpine desert (Wang et al., 2012). The soil POP concentrations for each soil type are summarized in Table 2. From the data in Table 2, we can see that the highest POP concentrations are to be found in forest soil and the lowest in desert soil. ∑DDTs and ∑PAHs concentrations vary by two orders of magnitude from forest soil to desert soil, ∑HCHs and HCB concentrations vary by one order of magnitude, and ∑PCBs only change by a factor of three. Similar to the aforementioned results for air, the ∑PBDE concentrations are generally lower compared with other POPs (Table 2). POPs are generally stored in soil organic carbon (SOC), which can vary from <0.1% to even >40% in the TP region (Wang et al., 2012; Yuan et al., 2014a, 2014b, 2015). The large variation in reported POP concentrations may be

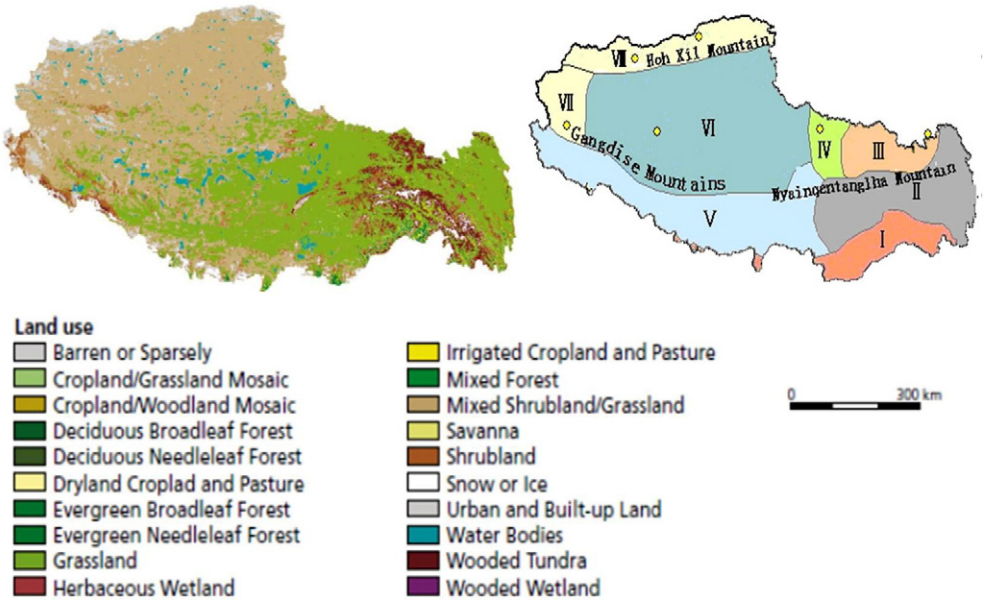

Fig. 2. Map showing the land use and vegetation cover of Tibet, and the corresponding eight soil zones: soil I: forest-latosols; soil II: alpine-burozems; soil III: sub-alpine scrub-meadow soils; soil IV: alpine meadow soils; soil V: mountain shrubby steppe soils; soil VI: alpine steppe soils; soil VII: sub-alpine desert soils; soil VIII: alpine desert soils.

attributable to the large difference in SOC. Forest soil is broadly regarded as a "final sink" for various POPs (Moeckel et al., 2009a). In the case of Tibetan forest, because of the low temperatures, the decomposition of forest litterfall and organic matter in soil is fairly slow, which provides a greater capacity of organic storage for POPs (Wang et al., 2014c). Basically, the SOC-driven concentration gradients observed for various soils of the TP (Table 2) agree with other observations reported for global background soils. Recently, studies conducted in alpine steppe soils (SOC < 1%), which are mainly located in the central TP (Yuan et al., 2012a,b, 2014a,b,c, 2015), have suggested that clay minerals play a key role in the accumulation of OCPs and PAHs in barren soil. This had also been previously asserted via a laboratory-based test (Torrents and Jayasundera, 1997).

2.2.2. Comparison with other background soils

By comparing the abovementioned soil studies with various other studies on background soils, we find that DDTs and HCH levels measured in TP soils far exceed those measured in the Pyrenees (Grimalt et al., 2004) and European Alps (Moeckel et al., 2008; Tremolada et al., 2008; Belis et al., 2009; Kirchner et al., 2009); in particular, HCB and PBDE levels are much lower than those reported from European background soils (Hassanin et al., 2004; Meijer et al., 2002, 2003). The average PCB concentration is about half the value (424 pg g^{-1}) reported for remote Chinese soils (Ren et al., 2007). Moreover, the average PAH concentration is around one third of the average reported for global background soil (Nam et al., 2009, 2008b). This comparison highlights the point that TP soils might accumulate considerable quantities of DDTs and HCHs due to its proximity to other countries in Asia.

2.3. Vegetation

Plant biomass plays a significant role in global environmental partitioning and plants are good indicators of tropospheric contamination levels. Plant biomass is believed to accumulate and circulate the residues of pollutants (Moeckel et al., 2009a; Ockenden et al., 1998; Schrlau et al., 2011). Therefore, vegetation should offer indirect evidence of atmospheric POP levels in remote areas (Jiao et al., 2009; Roland, 2006; Schmid et al., 2007). Studies have focused on the accumulation of POPs in the waxy cuticula of needles and leaves in the vegetation of the TP. For example, spruce needle samples [*Picea smithiana* (Wall.) Boiss, *Tsuga dumosa*], pine needles (*Pinus densata*) and cypress needles (*Cupressus torulosa*) have all been collected and measured (Wang et al., 2006, 2007b, 2007c; Yang et al., 2008), as have a number of different grass species, including Jidou (*O. glacialis* Benth. ex Bge), Rouzi (*T. rupifragum* Schrenk), *Kobresia* spp., *Polygonum* spp., and *Stipa purpurea*, as well as lichens and mosses (Zhu et al., 2014, 2015). Concentrations of POPs in these vegetation species have generally been found to be quite low, and broadly comparable to reported values for Arctic lichen (Kelly and Gobas, 2001), Antarctic mosses (Fuoco et al., 2009) and European spruce needles (Offenthaler et al., 2009; Shen et al., 2009).

2.4. Yak tissues

Although concentrations of POPs in grasses are low, their biomagnification along the lichen–caribou–wolf terrestrial food chain has been found in the Arctic (Kelly and Gobas, 2001). Similarly, given that grazing yaks may intake grasses as their main food, POPs will certainly be transferred into yak tissues.

Pan et al. (2014) collected yak muscle, liver and milk samples from pastures on in eastern TP (Ruoergai highland prairie) and reported OCP and PCB concentrations for these tissues. Other chemicals, such as polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), dioxin-like polychlorinated biphenyls (dl-PCBs), and polychlorinated naphthalenes (PCNs) were also found in yak muscle and fat from the Wolong high mountain area (Pan et al., 2013), along the eastern edge of the TP. By analyzing 30 domestic butter samples collected across the TP, Wang et al. (2009) found that $\sum_{25}$PCB concentrations in butter ranged from 137 to 2518 pg g^{-1}, and the average concentration of $\sum_{12}$PBDE was 125 pg g^{-1} — far lower than those found for butter from other regions in the world (Jafari et al., 2008; Kalantzi et al., 2001). Apart from this feature of lower POP concentrations, the common point among these studies was that chemical compositions in yak tissues and butters were mainly dominated by the more stable congeners, i.e., HCB, β-HCH, p-p′-DDE, and medium molecular weight PCBs and PCDDs (Wang et al., 2015a; Pan et al., 2014).

Table 2
POP concentrations (ng/g dw) in different soil types of the TP.

Soil type	∑DDTs	∑HCHs	HCB	∑PCBs	∑PBDEs	∑PAHs	Reference
Forest soil	21.6	2.09	0.489	0.284			Wang et al. (2014b)
				0.163	0.026		Zheng et al. (2011)
	0.55	0.57	0.106				Zheng et al. (2009)
Alpine-burozems	1.65	0.335	0.114	0.292	0.009		Wang et al. (2012)
						104	Wang et al. (2014a)
Sub-alpine scrub-meadow soils	0.887	0.157	0.183	0.596	0.014		Wang et al. (2012)
	2.85	0.4	0.1			201	Yang et al. (2013)
						46.1	Wang et al. (2014a)
Alpine meadow soils	0.435	0.279	0.057	0.198	0.007		Wang et al. (2012)
						60.2	Wang et al. (2014a)
Mountain shrubby steppe soils	0.301	0.084	0.036	0.302	0.009		Wang et al. (2012)
				0.186	0.011		Wang et al. (2009)
						43.0	Wang et al. (2014a)
Alpine steppe soils	0.119	0.183	0.53	0.257	0.002		Wang et al. (2012)
				0.100			Yuan et al. (2015)
					0.080		Yuan et al. (2012a, 2012b)
						23.1	Wang et al. (2014a)
	0.348	1.09	0.054			7.39	Yuan et al. (2014b)
	0.313	3.02	0.060				Yuan et al. (2014a)
Sub-alpine desert soils	0.129	0.201	0.25	0.193	0.00028		Wang et al. (2012)
						40.6	Wang et al. (2014a)
Alpine desert soils	0.134	0.094	0.50	0.121			Wang et al. (2012)
						12.6	Wang et al. (2014a)
	0.096	0.091				9.79	Tao et al. (2011)

2.5. Lake and river water

As compared with other mediums, POP concentration data reported for the Tibetan lake and river water were very limited. Zhang et al. (2003) analyzed the DDT and HCH isomers in water from Yamdrok and Co Ngoin Lake and found ∑HCH concentrations ranged from 1.8 to 3.8 ng l^{-1}, while ∑DDTs concentration varied from 0.2 to 0.3 ng l^{-1}. The Yangtze River is the longest river in China and originated from the glacier runoff in central TP. ∑DDT and ∑HCH concentrations in headwater of Yangtze River were reported in the range of below detection limit −0.2 ng l^{-1} and 0.7–2.1 ng l^{-1}, respectively (Liu et al., 2011b). After comparison, we found HCH values in the above-mentioned studies are much higher than the levels reported for other remote areas [from 1.4 to 139 pg l^{-1} (Fernández et al., 2005; Lohmann et al., 2009)]. POP level in Tibetan water body is expected to be low and these reported high levels might be due to the uncertainties caused by extraction (liquid-liquid extraction) and instrumental methods (electron capture detector) during measurements. Apparently, future studies on POPs contamination in lake water and glacier runoff are needed.

2.6. Wild fish

The TP is home to the highest and largest high-altitude lakes in the world. Because of the oligotrophic conditions and lower temperatures of these lakes, hundreds of endemic fish species (e.g., *Gymoncypris namensis, Gymoncypris waddellii, Ptychobarbus dipogon, Schizopygopsis stoliczkae, Schizopygopsis younhusbandi, Schizopygopsis microphalus, Oxygymnocypris stewartii*) breed. To date, OCPs, PCBs, PBDEs and polyfluoroalkyl substances (PFASs), and hexabromocyclododecanes (HBCDs), have all been detected in the wild fish populations of Tibetan lakes (Shi et al., 2010; Yang et al., 2008, 2010, 2011; Zhu et al., 2013). Fig. 3 summarizes and compares the concentrations of each POP class in fish muscle. Among the various legacy and emerging POPs, the average concentrations follow the order: ∑DDTs > ∑HCHs > ∑PFASs > ∑PCBs > HCB > ∑PBDEs > ∑HBCD. Because of the oligotrophic conditions and lower temperatures of alpine lakes, Tibetan fish generally have slower growth rates and live for longer, which may lead to the accumulation of higher concentrations of pollutants (Braune et al., 1999). Following a global comparison, the ∑DDTs and ∑HCHs concentrations are comparable to the values reported for fish species of the Canadian Rockies (Demers et al., 2007). Furthermore, similar to the results reported for Tibetan yaks, the metabolites of p,p′-DDE and β-HCH are the most abundant in the DDT and HCH family, respectively (Yang et al., 2010).

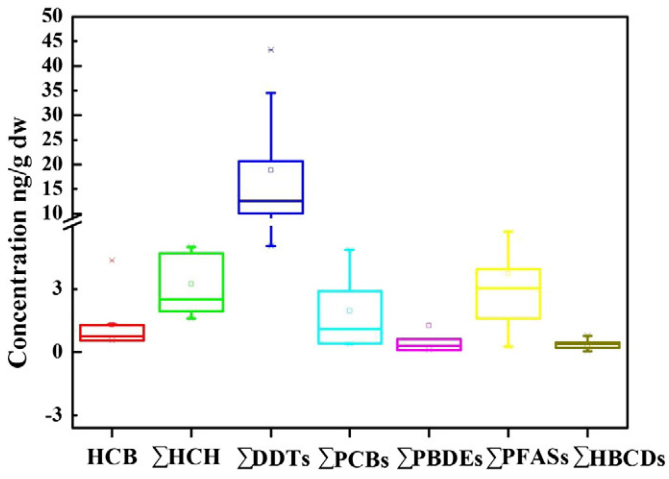

Fig. 3. POP concentrations (ng/g dw) in wild fish of the TP.

In conclusion, due to its remoteness, the concentrations of various POPs in the air, soil, vegetation, and yak/fish tissue of the TP are relatively low. Nevertheless, the very fact that pollutants can be found in these various mediums over the TP suggests a role played by long-range atmospheric transport (LRAT). We turn to this topic in the next section.

3. Atmospheric transport

3.1. Indian monsoon

Since the TP region has been relatively less impacted by human activities, the POPs in its air must derive from LRAT (Xu et al., 2009). Recently, Sheng et al. (2013) presented evidence of monsoon-driven POP transport. Taking p-p′-DDT as a model chemical, its concentrations were found to vary synchronously with the fluctuation of the Indian Monsoon Index. The seasonal fluctuation of the monsoon wave and DDT waves showed a similar wave width and wave frequency (Sheng et al., 2013), revealing that the delivery of DDTs by the Indian Monsoon is direct and fast. Alongside this temporal proof, the spatial pattern of DDTs displays a relatively high concentration in the southeastern TP (Wang et al., 2010a, 2016b), where the Indian monsoon enters the region (Wang et al., 2010a). Taken together, the temporal and spatial trends of DDTs in the atmosphere over the TP provide us with evidence for considerable quantities of DDTs being transported into the TP by the Indian monsoon.

Given that POPs over the TP mainly originate from adjacent polluted regions, determining the source areas and delivery paths accurately is of great interest. To do so, we need to trace air mass plumes, and for this the most common approach is to examine the backward trajectories via the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model, which can be downloaded (www.arl.noaa.gov/ready/hysplit4.html) and used for free. Accordingly, the possible transport paths for POPs arriving over the TP have been well studied (Cheng et al., 2007; Cong et al., 2013; Gong et al., 2010, 2015; Li et al., 2006; Liu et al., 2010). However, the HYSPLIT model does not associate the identified paths with particular chemical types and measurements, and it only considers meteorological data. To address this, the Potential Source Contribution Function (PSCF) model, which combines chemical measurements with air trajectories, was developed (Cheng et al., 1993; Hafner and Hites, 2003). This model focuses mainly on those trajectories showing abnormally high or low chemical concentrations, and it calculates the fraction of high hourly points in each geographical cell. Ultimately, the PSCF model presents the probability of an air parcel resulting in a high measured concentration for each given geographical cell (Hafner and Hites, 2003). Using this approach, the source of OCPs and PCBs at Lulang (southeastern TP) has been traced to the eastern Indian coastal region and Bay of Bengal (Fig. 4; Sheng et al., 2013), and the source of PAHs and particulates at Lulang to the Indo-Gangetic Plain (Fig. 4; Wang et al., 2015b). Based on relevant published studies (Cong et al., 2013; Gong et al., 2010; Li et al., 2006; Liu et al., 2010; Sheng et al., 2013), we can conclude that both the HYSPLIT and PSCF models have provided comprehensive confirmation of the transport of POPs to the TP by the Indian monsoon.

3.2. Westerly flow and the East Asian monsoon

The TP is a vast area with complicated climate and air circulation patterns. As explained in the previous section, the southern part of the TP is clearly and directly influenced by the Indian monsoon. However, further north, the monsoon is blocked by the high elevation of the plateau. Elsewhere, in the extreme western and eastern parts of the plateau, westerly winds and the East Asian monsoon play a key role in delivering POPs, respectively. For example, for the Ngari region (28°–35°N, 78°–84°E), which is located in the western TP, HYSPLIT model results show that air masses from northern Pakistan, Iran, the Middle East,

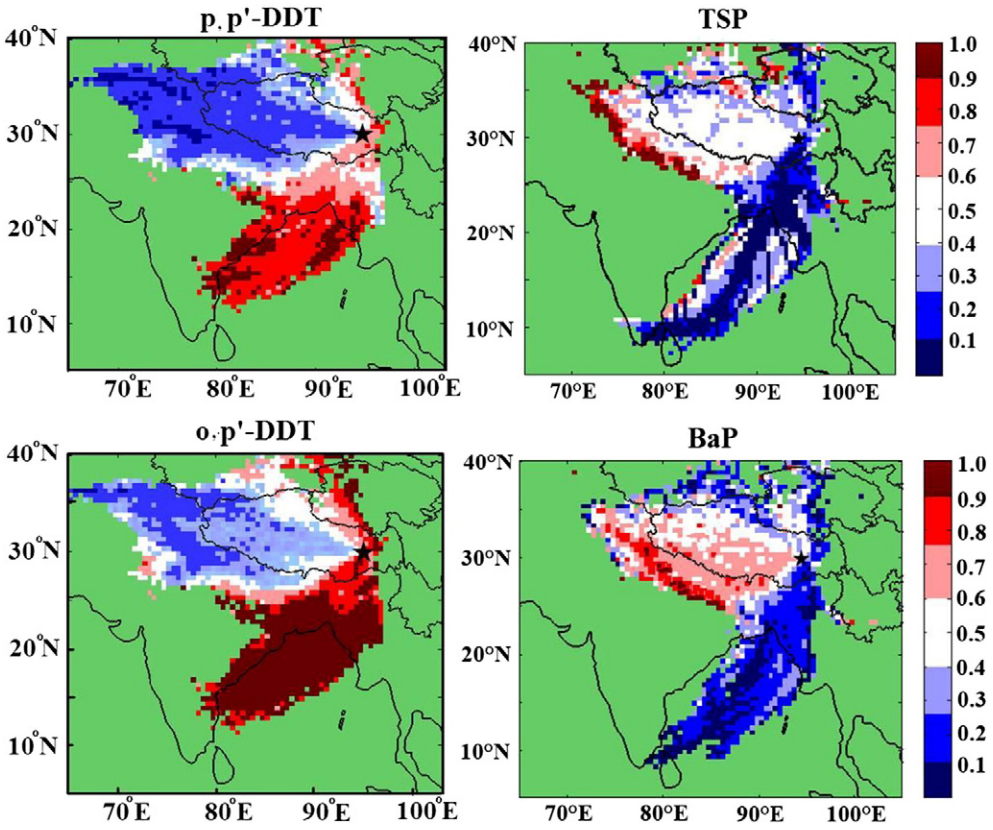

Fig. 4. PSCF maps for p,p′-DDT, o,p′-DDT, TSP and BaP. Dark red represents PSCF values from 0.91 to 1.0, with shades of red ranging from 0.61 to 0.15; white represents 0.41 to 0.60. Dark blue represents 0 to 0.10, with shades of blue ranging from 0.40 to 0.15. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

and even Europe, can reach it (Gong et al., 2015). Therefore, POPs emitted in these areas can be transported to the western TP. On the eastern edge of the TP, the air monitoring at Wolong Mountain offers crucial data because of its close proximity to densely populated and highly industrialized parts of China (Chengdu Plain). Air mass trajectories reveal that, prior to arriving at Wolong Mountain, the air had been present over eastern China and had passed over the western part of the Sichuan basin, including the Chengdu Plain (Liu et al., 2010). Consequently, similar summer to winter POP ratios have been observed for the Chengdu Plain and Wolong Mountain, indicating the Chengdu basin is an important source region of POPs for the eastern TP, and the East Asian monsoon is responsible for this transport (Liu et al., 2010).

Compared with other air circulation systems, transport by westerly winds can only contribute minor amounts of POPs to the western TP. Levels of OCPs in the Ngari region have been reported to be less than 10 pg m^{-3} (Gong et al., 2015), while areas close to India (Sheng et al., 2013) and on the Chengdu Plain (Liu et al., 2010) demonstrate much higher OCP concentrations of up to 40 and 30 pg m^{-3}, respectively. In addition, a recent study (Wang et al., 2016b) found that HCB is the dominant chemical in the air over the western TP (European sources), while DDTs are more predominant over the southern TP (Indian sources). This suggests that different air circulation systems and different source regions will exert different influences on either the levels or composition of POPs. On the basis of composition/fingerprint analysis of POPs, three zones were classified by Wang et al. (2016b). These zones are broadly in line with the three major climatic zones (monsoon region, westerly region, and transition region) discovered by long-term observations of $\delta^{18}O$ in precipitation. This consistency demonstrates that the levels, composition and spatial distribution patterns of POPs over the TP are strongly influenced by atmospheric circulation (Wang et al., 2016b).

3.3. The Himalaya

The Himalaya is a mountain range bordered to the north by the TP, and to the south by the Indian subcontinent. Because of its tremendous height, the Himalaya acts as a natural wall splitting the westerly flow into two branches; plus, it imposes a remarkable influence on precipitation over the Indian subcontinent. By investigating the differences in the concentrations and compositions of POPs along the southern and northern slopes of the Himalaya, it has been found that high elevation sites on the southern slope often show the impact of long-range synoptic-scale winds, with similar relative compositions of POPs to distant source regions (Gong et al., 2014). However, lower sites are more influenced by local emissions. This feature that high-elevation locations are affected more by long-range atmospheric transport is in line with observations in other parts of the world, such as South Island, New Zealand (Lavin and Hageman, 2013). Furthermore, such results indicate that the Himalaya is unable to completely block the synoptic-scale transport of pollutants, despite its enormous height.

There are three deep mountain valleys that cut through the Himalaya: Split valley, Zhangmu valley and Yarlung Tsangpo Grand Canyon. The latter is most famous, being the deepest and largest canyon on Earth. Moreover, this canyon has been described as a "leaking wall" that allows the transport of various pollutants, such as black carbon (Cao et al., 2011) and POPs (Sheng et al., 2013; Wang et al., 2015b). Relatively high concentrations of atmospheric POPs (up to 40 pg m^{-3}) have been reported there (Sheng et al., 2013), and the incursion of particulate aerosols through the canyon can be clearly captured by the MODIS (Moderate Resolution Imaging Spectroradiometer) data retrieved from the Terra satellite, as well as CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) (Wang et al., 2015b; Xu et al., 2014).

Although there have been no studies regarding the transport of pollutants along other Himalayan valleys, it is reasonable to assume that these too provide channels for the transport of POPs.

Another mechanism is that vegetation grown on the Himalaya can act like an organic partition that absorbs atmospheric POPs, storing them in their tissues. A study on the atmospheric transport of POPs along the southern slopes of the Himalaya demonstrated that the presence of vegetation and precipitation results in considerable quantities of POPs being removed from the overlying air, enhancing the deposition of POPs at the mountain surface (Pokhrel et al., 2016). In addition, there are discernable differences between sites on the northern and southern slopes of the Himalaya; Gong et al. (2014) found a sharp decrease in absolute values of atmospheric POP concentrations from south to north, with the difference ranging from hundreds of pg m^{-3} (in the south) to several pg m^{-3} (in the north). These results suggest that the Himalaya hinder the direct transport of POPs from the south Indian subcontinent to the TP. As such, this process differs from the previous two processes mentioned above, which favor the trans-Himalaya transport of POPs rather than highlight the important role of precipitation and vegetation on the upslope transport of POPs along the mountain slopes of the Himalaya (Pokhrel et al., 2016). A schematic diagram summarizing the three transport mechanisms is presented in Fig. 5.

4. Atmospheric deposition

Atmospheric deposition, including wet deposition via rain and snow, dry deposition of particles, and gaseous deposition, is a major contributor to the elimination of POPs in source regions, and becomes a significant POPs input pathway for the occurrence of these compounds in remote areas (Venier and Hites, 2010). Despite the deposition by rain occurring along mountain slopes, researchers have found that scavenging of POPs by rain in the Bay of Bengal precipitation center reduces the LRAT potential of water soluble POPs (i.e., HCHs) and leads to lower atmospheric concentrations of HCHs over the TP (Sheng et al., 2013). On the contrary, once POPs have undergone LRAT and reach the TP, wet and dry deposition associated with the extremely cold temperatures will promote the accumulation of POPs in the Tibetan environment (Loewen et al., 2005). The accumulation in snow and the air–surface deposition of POPs are reviewed in this section.

4.1. Snow scavenging

Snow/rain events scavenge pollutants from the air, meaning POPs can accumulate and condense in snow packs (Arellano et al., 2011). A few field studies have been conducted that reported PCB, DDT and HCH quantities in the eastern Rongbuck glacier (Kang et al., 2009; Wang et al., 2007a, 2008b) and Dasuopu glacier (Wang et al., 2008a). However, the data obtained by these studies are not comparable, mainly because of the different analytical methods they employed. For example, some studies used the solid-phase microextraction technique to quantify pesticide concentrations in melted snow samples (Wang et al., 2008b), while others (Kang et al., 2009) used the traditional liquid-liquid extraction method. Limited by the extremely high elevation of such glaciers, only a small number of snow samples can be collected, which results in relatively high analytical uncertainties. Nonetheless, DDT metabolites (p-p′-DDD) and p-p′-DDT were detected in the firn core of Dasuopu glacier (Wang et al., 2008a), and seasonal differences were observed for α- and γ-HCH, with concentrations increasing from samples taken in the non-monsoon to the monsoon season (snowpack in the eastern Rongbuk glacier) (Kang et al., 2009).

Apart from reported concentrations in fresh snow, POP concentrations combined with the water equivalent of ice cores can provide data on deposition fluxes. Among the literature to date, a relatively comprehensive study was conducted in the eastern Rongbuk glacier in which the annual atmospheric deposition fluxes of ∑DDT, ∑HCH and ∑PAHs were obtained (Wang et al., 2008b). Due to the close proximity of the eastern Rongbuk glacier to India, the larger deposition fluxes generally corresponded to periods of extensive use of these pollutants in India. Recently, atmospheric deposition fluxes of an emerging group of compounds — the perfluoroalkyl acids (PFAAs) — have been reported for two glaciers influenced by the Indian monsoon and westerly flow, respectively (Wang et al., 2014b). Although the monsoon region glacier showed larger annual water equivalent accumulation, large ∑PFAAs fluxes were found in the westerly-influenced glacier, which is closer in proximity to European countries (Wang et al., 2014b), where PFAAs and their precursors have been widely used. This result supports the argument that source regions play a key role in the atmospheric deposition fluxes of chemicals, rather than simply the snow accumulation rate. Compared to absolute concentrations, atmospheric deposition fluxes (in units of pg cm^{-2} yr^{-1}) retrieved from ice cores can provide a number of advantages in terms of quantifying the levels

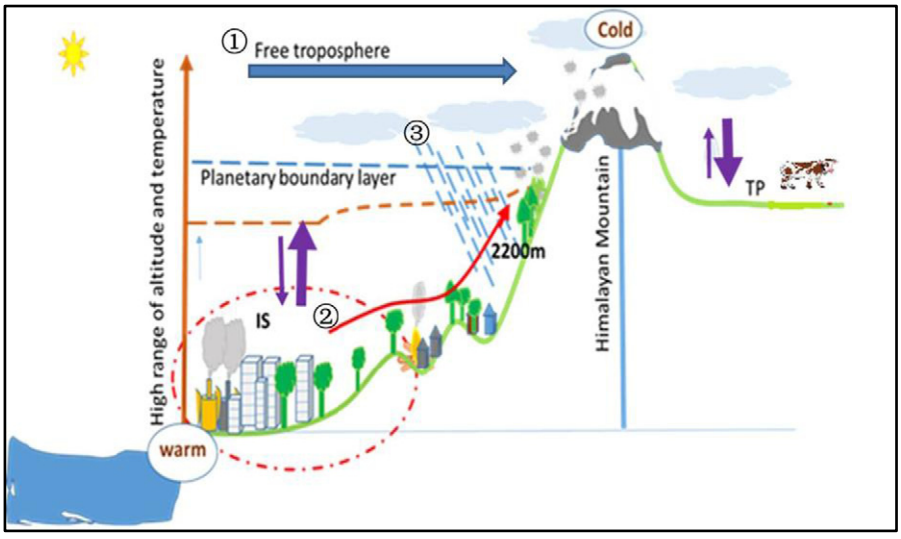

Fig. 5. Schematic illustration of the POP transport mechanisms operating across the TP: ① long-range synoptic-scale transport; ② valley transport; ③ precipitation scavenging and vegetation sorption of POPs.

of POPs deposited on the glacier surface; this allows scientists to estimate the total amount of POPs stored in the glacier as a whole (Wang et al., 2008b).

4.2. Air–surface exchange

It has been suggested that a global "cold condensation" phenomenon exists in which semi-volatile chemicals migrate to colder regions of the planet (Gouin et al., 2004). Owing to its cold temperatures, which promote the condensation of atmospheric gas-phase POPs to the mountain surface, the TP is conceptually regarded as one such "cold trap" or "convergence zone" for POPs (Chen et al., 2008). However, with the rise of global warming, secondary emissions of POPs from soils and water bodies have complicated the picture. The exchange processes of POPs between air and the land surface are bidirectional, comprising gaseous deposition from the air and evaporation from the land surface.

4.2.1. Direction of air–soil exchange

A few studies to date have focused on the air–soil exchange of POPs. The following questions have been addressed: (1) Do all chemicals experience a net deposition from air to soil on the TP? (2) Does TP soil act as a final sink or secondary source during the global cycling of POPs? (3) Which factors control the exchange direction? (4) What quantities of chemicals can be deposited in, or evaporate from, soil?

By comparing the fugacities of POPs in surface media and ambient air, the direction of air–surface exchange can be inferred. Equations regarding the fugacity fraction (*ff* — the ratio of soil fugacity to the sum of soil and air fugacity) and net exchange fluxes (the difference between deposition flux and evaporation flux) have been put forward (Backe et al., 2004; Mackay and Paterson, 1991; Mackay et al., 1992). By comparing *ff* values and considering analysis uncertainties, an *ff* of between 0.3 and 0.7 can be considered to differ significantly from the equilibrium. Meanwhile, an *ff* of greater than 0.7 indicates net volatilization from soil into air, whereas values less than 0.3 indicate net deposition from air to soil (Li et al., 2009). Based on this threshold, it has been determined that Tibetan soil is an overall sink for DDT and DDE, whereas it is a partial contributor (secondary source) for HCB, low molecular weight PCBs (4-Cl PCBs) and HCHs observed in air (Fig. 6; Wang et al., 2012). In another case study from Balang Mountain in Wolong Nature Reserve (eastern edge of the TP) (Liu et al., 2010), air and soil were found to be in equilibrium with respect to HCHs and HCB, whereas a situation of net atmospheric deposition prevailed for DDT-class chemicals. Similarly, the air–soil exchange state showed that Tibetan soils remain as a sink for high molecular weight PAHs, but likely become a potential secondary source for low molecular weight PAHs (Fig. 6; Wang et al., 2014a). All the above findings suggest that the fate of POPs in the TP environment strongly relies upon these chemicals' properties.

4.2.2. Soil–air partition coefficient

Soil retention capacity governs how much of a chemical is able to accumulate in soil. The sorptive capacities of soil for different POPs can be explored by calculating the soil–air partition coefficient (K_{SA}) value according to Eq. (1) (Li et al., 2009). The K_{SA} governs air–soil exchange patterns and is proportional to soil organic matter (SOM) content and K_{OA} (octanol/air partition coefficient):

$$K_{SA}=\varphi SOM\ K_{OA} \quad (1)$$

PCB congeners, i.e., -28, -101 and -180, are frequently used as model compounds to address the possible fate of POPs with different volatilities. Li et al. (2009) produced a global gridded map of log K_{SA} for PCB-28, -101 and -180 using meteorological (http://www.cdc.noaa.gov/Composites/) and SOC (http://islscp2.sesda.com/ISLSCP2_1) data. Using a similar concept, K_{SA} values on the basis of realistic temperature and SOC data (40 sampling sites), measured by Wang et al. (2012), were calculated and interpolated (see Fig. 7). The log K_{SA} values for PCB-28, -101 and -180 are 2–3 orders of magnitude higher than those reported in Li et al. (2009). Limited by the reanalysis data, the temperature data used by Li et al. (2009) for the Tibet region ranged from 20 °C to 30 °C, which were much higher than the measured data [−7 °C to 15 °C (Wang et al., 2012)]. Furthermore, the SOC data (<0.1%) used by Li et al. (2009) were lower than the measured data [0.1%–40% (Wang et al., 2012)]. This is probably the reason for the underestimation of log K_{SA} values. Based on the K_{SA} map for the TP (Fig. 7), Tibetan soil shows a strong ability to hold POPs, with a similar extent to boreal regions. This feature far exceeds traditional viewpoints. Although Tibetan soil is generally SOC-poor, as compared with that in boreal regions, cold temperatures play an important role in the retention of POPs in soil on the TP.

With regard to different chemicals, higher K_{SA} values have been observed for high molecular weight PCB (high K_{OA}), suggesting chemicals with high Koa may become enriched in cold climate soil (Růžičková et al., 2008). This may explain why Tibetan soils remain a "sink" for DDT-class chemicals and high molecular weight PAHs. Concerning different regions of the TP, soils from north and central TP generally have a greater ability to hold PCBs (Fig. 7), which can be attributed to the relatively high SOC and low temperatures. Because regions with elevated log K_{SA} values (above 8) may act as longer-term "traps", the northern and central TP will act like a "sink" for deposited POPs. However, sampling sites (southern TP) with moderate or low log K_{SA} values (around 6 or 7) can only be temporary or transient sinks for chemicals. This spatial variation demonstrates that caution should be applied when regarding the whole plateau as a "trap" for all POPs.

4.2.3. Air–soil gas exchange fluxes

Quantification of the air–soil exchange flux can provide detailed data on POP amounts volatilized from air or deposited in soil. The available

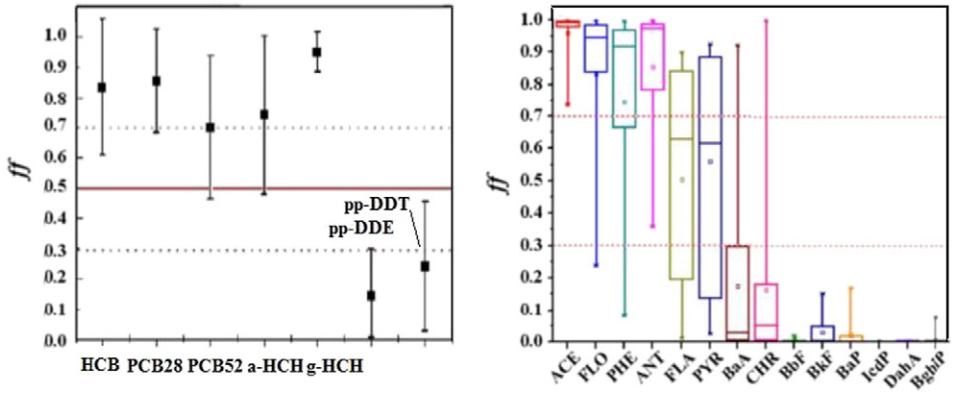

Fig. 6. Air–soil exchange directions (fugacity fractions) of different chemicals.

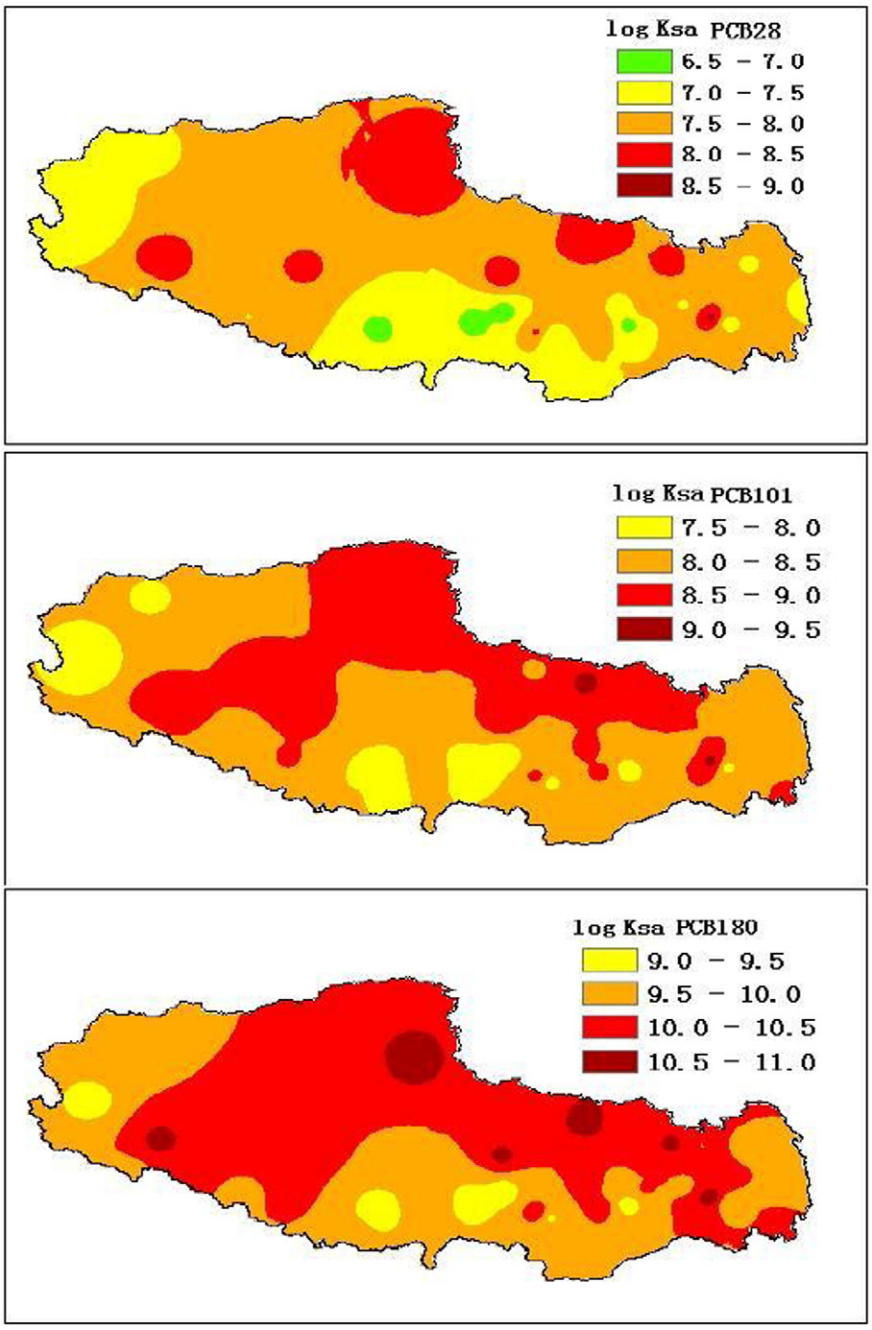

Fig. 7. Soil and air partition coefficient K_{SA} across the TP for PCB-28, -101, and -180.

literature shows that the annual average air–soil gas exchange fluxes were −0.3, −0.7, 0.5 and 0.3 ng m^{-2} h^{-1} for HCB, γ-HCH, *p, p′*-DDE and *p, p′*-DDT, respectively (Wang et al., 2012, 2016b). A negative flux suggests the net air–soil exchange of POPs is one of deposition from soil to air, and a positive flux suggests the opposite, i.e., evaporation from air to soil. Although numerous studies have evaluated the air–soil exchange directions (*ff*) of POPs (Backe et al., 2004; Bidleman and Leone, 2004; Kaya et al., 2012; Liu et al., 2011a, 2011b; Masih et al., 2012; RůŽičková et al., 2008; Syed et al., 2013), the exchange fluxes themselves have seldom been reported. Here, after unit conversion to pg cm^{-2} y^{-1}, the reported gas deposition fluxes of DDTs (*p, p′*-DDE + *p, p′*-DDT) were found to be three times higher than their wet deposition fluxes retrieved by the above-mentioned ice core study (Wang et al., 2008b). Combined with the contribution of both air–soil gas deposition and wet deposition (downward transport), soil will continuously be the largest sink for DDT class chemicals. With respect to the more water-soluble chemicals like HCHs, the retrieved wet deposition flux (hundreds of pg cm^{-2} y^{-1}) is comparable to the soil-to-air evaporation flux (Wang et al., 2016b, 2008b), indicating these two fluxes will offset one another, ultimately giving rise to a status of equilibrium.

4.2.4. Air–vegetation partition

Vegetation can provide an extensive organic surface for the partitioning of POPs in the atmosphere, and enhance the net atmospheric deposition of POPs (McLachlan and Horstmann, 1998). Forest regions cover more than 1.2 million square kilometers and almost half the area of the TP is covered by grassland. As such, the capacities of forest and grassland to accumulate POPs have been an important focus of study.

Forest canopies can filter and intercept atmospheric POPs, resulting in a marked reduction in quantities of POPs reaching beneath such canopies. The ratio of the POP concentration in the forest to that in clearings (DF) has been used to investigate the capacity of the forest canopy in filtering POPs. Ren et al. (2014) reported the DF values (0.50 and 0.39 for DDE and DDT, respectively) for Tibetan forest. The results indicate that the Tibetan forest canopy can intercept approximately 50% of atmospheric DDTs, and this depletion ability is more effective than those found in the coniferous forests of the European Alps [DF range of 0.64–0.80 for DDTs (Jaward et al., 2005)]. Integrating the influence of wet and dry POP deposition, POPs deposited in the form of litterfall, and the accumulation of gaseous POPs by surface litter, Wang et al. (2014c) estimated air-to-forest ground fluxes of POPs and reported high flux values of up to 660 pg cm^{-2} y^{-1} for ΣDDT. This value is approximately one order of magnitude higher than that reported for the Italian Alps (Moeckel et al., 2009b). Given forest is dominantly located in the mountain valleys of the southeastern TP, which is also the entrance point for POPs arriving via the Indian monsoon, the risk to the environment in this region should be of great concern.

The dynamic uptake of POPs by grass can be investigated by collecting grass samples during its growth period. The concentrations of POPs in grass can then be plotted against growth time using the equation

$$y = A\left(1-e^{-\left(\frac{1}{k}\right)x}\right) + y_0, \quad (2)$$

where y is the concentration of the POP in grass (pg g^{-1}), A is the POP concentration in grass when absorption reaches equilibrium (pg g^{-1}), k represents the equilibrium time (day), x is the growing time (days), and y_0 is the primary concentration in grass (pg g^{-1}). Equations of best fit show that the mathematical equilibrium times for γ-HCH and HCB are 24 and 11 days, respectively, and for DDE and DDT they are 153 and 232 days, respectively (Wang et al., 2015a). Since these estimated equilibrium times are much longer than the real growing season (less than 120 days), the continuous accumulation of DDTs occurs for the whole growth period. Likewise, Wang et al. (2016a) also found atmospheric deposition to be a major uptake mechanism for the accumulation of POPs in Tibetan crops (Wang et al., 2016a), because the isomer ratios in crops — including (DDE + DDD)/DDT and α/γ-HCH — were very close to those observed for lichens, conifer needles and pasture grasses in Tibet, and rice in India (Wang et al., 2016a). Therefore, the POPs in the crops also came from the same source as those in other vegetation of the TP; namely, atmospheric deposition.

5. Bioaccumulation of POPs in terrestrial and aquatic food chains of the TP

The aforementioned air–vegetation partition leads to the accumulation of POPs in pasture and farmland crops. Consequently, grazing yaks and local residents will take in POPs through food consumption. Aside from the reported concentration levels in yak meat, milk and butter samples, the accumulation of POPs via air to yak butter has also been investigated (Wang et al., 2015a). First, the enrichment of more stable POPs, such as β-HCH, p,p′-DDE and heavier PCB congeners increased along the air–grass–yak (butter) food chain (Wang et al., 2015a). On the other hand, the air–butter transfer factor ($TF_{A:B}$) can represent the volume of air that each gram of lipid can accumulate (m^3 air g^{-1} lipid). On the basis of measured air and butter POP concentrations, Tibet-specific $TF_{:AB}$ was calculated and provided an effective tool to predict air concentrations from butter concentrations (and vice versa) at the TP scale (Wang et al., 2015a).

Based on a survey of local peoples' dietary habits, Pan et al. (2014) calculated the estimated daily intake (EDI) of OCPs and compared the results with the World Health Organization's (WHO) acceptable daily intakes (ADIs). EDIs were found to be generally lower than the corresponding ADIs, indicating there were no immediate risks from the regular dietary intake of meat and milk (Pan et al., 2014). Also, taking into account all toxicity equivalency quantities (TEQs) of PCDD/Fs, dl-PCBs and PCNs, the daily intake of ∑TEQs via dietary consumption of yak muscle was below the WHO's tolerable daily intake (Pan et al., 2013). In fact, the foods consumed by local inhabitants also include crops (approximately 50%). On the basis of the method recommended by the US Environmental Protection Agency, the exposure risks to humans via the intake of crops have been estimated (Wang et al., 2016a). Generally, residents will be exposed to negligible cancer risks by consuming agricultural food, and only a few sites show low cancer risks (Wang et al., 2016a). In conclusion, although the environment of the TP (i.e., air, soil and vegetation) has been contaminated by various toxic POPs, and biomagnification of POPs along the air–grass–yak food chain has been observed (Wang et al., 2015a), obvious harmful consequences of POP consumption by local residents have yet to be seen.

POPs can also be transferred along the aquatic food chain. Aside from atmospheric deposition, as snow melts, snow-sequestrated POPs can enter alpine lakes via influxes of meltwater (Blais et al., 2001a). Owing to the relatively short food chains in alpine lakes and the lower trophic levels of fish, significant biomagnification of POPs has not been observed in the alpine lake food chain. However, recently, Ren et al. (under review), applied a fugacity-based dynamic bioaccumulation model to the top predator (the fish *Gymnocypris namensis*) of the second largest lake over the TP with localized parameters. They found that DDEs, DDDs, PCB-138, -153 and -180 displayed significant positive correlations with trophic level, and confirmed that gastrointestinal uptake was the key mechanism that drove the biomagnification of POPs in the high-altitude aquatic food chain. This result challenged the traditional viewpoint that relatively short food chains in alpine lakes result in weak or no biomagnification of POPs. However, due to religious reasons, the local residents do not consume fish as food. Therefore, the human health risk in this region posed by fish can be ignored.

6. Temporal trends

Mountain glaciers and lake sediments act as “natural archives” for studying historical trends of pollution (Hong et al., 2009; Van de Velde et al., 1999). POP records reflected by ice and sediment cores are effective proxies for the revelation and reconstruction of the historical usage of POPs (Muir et al., 1996; Villa et al., 2006). Concentrations of total DDTs in ice cores (East Rongbuk glacier) (Wang et al., 2008b) show maxima of approximately 2 ng l^{-1} corresponding to the mid-1970s, which was the peak time for malaria cases in India (in 1976). After the banning of DDTs and HCHs in India, their concentrations can generally be seen to have decreased after the 1990s. However, concentrations of total PAHs in ice cores sharply increase after 1990, when India entered a period of rapid industrialization and urbanization. In western Tibet, ice core records of emerging compounds, ΣPFAAs, show concentrations continuously increased from 1980 to 1999, which is in broad agreement with the time series of global extensive emissions (Wang et al., 2014b). Compared with ice core records, which mainly provide up to 50 years of data, sediment records can provide a picture of pollution trends on a time scale of hundreds of years. PAHs in sediments from remote lakes of the TP (Wang et al., 2010b; Yang et al., 2016) show a remarkable increase in PAH concentrations taking place from the 1980s, followed by an even faster rate of increase after the 1990s. This temporal pattern is generally in line with the stages of economic development of countries around the TP.

With the support of TORP, a long-term atmospheric POPs monitoring program across the TP has been conducted since 2008. In a recently published study based on this program (Wang et al., 2016b), the annual averages of POPs, including OCPs and PCBs, from 2007 to 2012, across the TP, were reported. This contemporary air monitoring campaign provides temporal patterns that can be used to evaluate the effectiveness of regional regulatory measures for POPs (Wang et al., 2016b). For

instance, significant decreases in concentrations of α-HCH, γ-HCH, ∑PCBs and HCB were observed, while concentrations of two parent DDTs remained stable during the 5-year period (Wang et al., 2016b). The results reflected the effectiveness of the Stockholm Convention in reducing emissions of HCHs, PCB and HCB substances in Asian countries to the background atmosphere. Similar results have been reported for European Alps (Kirchner et al., 2016). Given the source of DDTs in the atmosphere of the TP is mainly the Indian subcontinent, the stable DDT concentrations indicated that the regulation of DDTs in India might be less effective (Sharma et al., 2014).

7. Research needs

Although research in this field conducted over the past decade has improved our understanding of POPs over the TP, there are still many areas where additional efforts are required to fill knowledge gaps. These recommended future avenues of research can be summarized as follows:

The PAS is an ideal instrument for the atmospheric monitoring of POPs over the TP, where power supplies are lacking. However, the sampling rate of POPs can be influenced by wind speed and temperature differences, which vary vastly in different locations over the TP. It is therefore necessary to carry out careful calibrations with respect to rates of uptake. Improving our understanding of how PASs perform under different environmental conditions on the TP will enhance the reliability of this instrument as a monitoring tool on the continental or global scale.

More measurements are needed in terms of the seasonal variations of POPs in the atmosphere over the TP. To build upon what is already known regarding the transport of contaminants via LRAT requires the setting up of more active air monitoring stations, especially at border sites between the TP and India, and between the TP and Pakistan. To date, AMAP has produced 20 consecutive years of data on various legacy and emerging POPs and PAHs (Hung et al., in press). Meanwhile, the EMEP POPs monitoring network started from six sites in 1999 and had expanded to 34 sites by 2012 (Gusev et al., 2009). Compared with the full and informative data of these international programs, the POPs data of TORP are relatively short-term and lower quality. Using AASs to conduct continual POPs sampling across the large spatial scale of the TP would provide high-resolution and long-term seasonal trend information regarding POPs, and further offer the chance to investigate the possible links between climate change/variations and the regional cycling of POPs.

POP concentrations have not yet been measured for all component parts of the TP environment. Data coverage for lakes remains sporadic, both temporally and spatially. There have been no synoptic surveys with which to constrain a particular lake's budget. The spatial coverage of surface water concentration, vertical profiles of POPs in the water column, cycling and fate of POPs in aquatic ecosystems, contaminant burdens in suspended particulates, and sediment burial, are all processes that still remain unclear.

Organic carbon pools represent major active stores of pollutants. Therefore, the burial and storage of POPs in terrestrial surface layers and forest ecosystems across the TP should be of concern. The storage capacities of forest foliage, litter fall, and soil humus, merit careful evaluation. Plant foliage has a relatively large surface area, and their waxy cuticles have a high affinity for many organic pollutants. The uptake capacity and kinetics among different species, ages and seasons need further investigation.

A significant proportion of the TP's annual precipitation occurs as snow, and a snowpack covers the mountains for most of the year. Thus, snow has an important influence on the extent and timing of contaminant delivery to mountain ecosystems. A snowpack receives contaminants from the atmosphere via both wet and dry deposition, and may lose them via volatilization, wind relocation, and meltwater percolation. Our understanding of the occurrence and relative importance of these processes is still lacking. Moreover, measurements of POPs in rainfall/snowfall can provide evidence for the mountain cold trapping mechanism.

Snowpack melt is a major hydrologic event across the TP. Accelerated snow and glacier melting may result in a substantial release of POPs into lakes or the surface water system. Against the background of global warming, glacial melt is regarded as a secondary source of POPs. Improved knowledge of the hydrological cycle of POPs in this complicated snow/glacier–lake system is essential for studying and understanding the "source–sink" relationships of POPs, and will also provide important insights into their global cycling.

Similar to the North and South Pole, the TP is a region of particular interest for studying both climate change and pollution contamination. However, the impact of climate change is complicated, not least because it is expected to alter human activity and the associated emission patterns of pollutants, thus — in the context of the present review — exerting an influence on the transport and fate of POPs. Therefore, regional-scale chemical-fate models that quantify the impact of climate change on the emissions, fate and future use of POPs and their transport to the TP, are essential.

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