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Altitude-dependent accumulation of short chain chlorinated paraffins in fish from alpine lakes and Lhasa river on the Tibetan Plateau

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ABSTRACT

High mountain cold-trapping effects can play important roles in the global long-range transport of persistent organic pollutants (POPs). Short chain chlorinated paraffins (SCCPs) have recently been included into the Stockholm Convention as a new class of POPs. However, the long-range transport behavior and environmental fate of SCCPs still remain largely unknown in high-altitude mountain areas. In this study, a total of 51 fish samples were collected from five high-altitude mountain lakes and Lasha river across the Tibetan Plateau. SCCPs were positively detected in all fish samples, and the concentrations ranged from 3.9 to 107 $ng\,g^{-1}$ dry weight (dw) with an average of 26.6 $ng\,g^{-1}$ dw. Compared to aquatic organisms from the Artic and Antarctica, the SCCP levels found in alpine fish from the Tibetan Plateau were lower. A significant increasing trend in accumulation levels of SCCPs in alpine fish with the increasing altitude was found on the Tibetan Plateau ($r = 0.98$, $p < 0.001$). Shorter chain congener group C10 showed a significant increase in percentage contribution to total SCCPs with increasing altitude, but a contrary tendency was found for longer chain congener group C13. The widespread occurrence of SCCPs in Tibetan fish was mainly sourced from the long-range atmospheric transport, and the altitude-dependent distribution of SCCPs was due to the mountain cold-trapping effects and potential susceptibility to bioaccumulation. To our knowledge, this is the first report regarding the altitude-dependent accumulation of SCCPs in biota in the polar environment.

1. Introduction

Chlorinated paraffins (CPs) are complex technical mixtures used in a wide variety of commercial products (van Mourik et al., 2016). The commercial CPs are generally subdivided into short chain (SCCPs, C_{10-13}), medium chain (MCCPs, C_{14-17}), and long chain CPs (LCCPs, C_{18-30}) according to carbon atom number (Tomy et al., 1998). As high production volume chemicals in the world, CPs have been detected in diverse environmental compartments including air, water, dust, soil, sediments and sludge, as well as in biota and the human in the recent decade (van Mourik et al., 2016; Wei et al., 2016). Among the CPs, SCCPs with carbon chain 10-13 are classified as priority hazardous substances, and have been paid particular attentions owing to their environmental persistence (Iozza et al., 2008; Zeng et al., 2013), long-range transport ability (Li et al., 2016; Li et al., 2017; Ma et al., 2014; Tomy et al., 2000; Wu et al., 2017), bioaccumulation and biomagnification (Zeng et al., 2017a; Zeng et al., 2015), and potential carcinogenic effect and endocrine disrupter property (Kobeticova and Cerny, 2018; UNEP, 2016; Zhang et al., 2016). In view of the potential adverse effects on the environment and human health, SCCPs were ultimately listed as a new class of persistent organic pollutants (POPs) by the Stockholm Convention in May 2017 (UNEP, 2017), and now the produce and use of SCCPs begin to be banned worldwide.

SCCPs have similar vapor pressures ($0.028–2.8 \times 10^{-7}$ Pa) to those of conventional POPs, and also have persistence in air with half-lives of 0.81–10.5 days (UNEP, 2016). Hence, SCCPs could undergo long-range atmospheric transport. Several earlier studies reported that SCCPs were detected in the sediments of remote Arctic lakes far from local sources of contamination (Tomy et al., 1999), as well as in Arctic biota, such as beluga whale, ringed seal, walrus (Tomy et al., 2000), seabird and char (Reth et al., 2006). CPs were also found to be widely distributed in spruce and humus needles collected from the Alps, the great mountain range of

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Europe (Iozza et al., 2009). Several recent studies demonstrated the occurrence of SCCPs in the atmosphere and biota not only in the Arctic (Letcher et al., 2018; Li et al., 2017) but also in the Antarctica (Li et al., 2016; Ma et al., 2014). These results indicate that SCCPs have substantial abilities to undergo long-range atmospheric transport.

High mountains can act as an important cold trap for atmospheric pollutants owing to barrier effects, low temperatures and high precipitation rates, and thus may play important roles in the global long-range transport of semi-volatile organic compounds (Daly and Wania, 2005). The Tibetan Plateau is one of the most remote and isolated regions in the world, and often referred to as the “Third Pole” or the “Roof of the World” due to its high altitude and large size of its glaciers (Yang et al., 2010a). It is far away from modern industrial and anthropogenic activities, and is thus considered to be a pristine region. The unique geographical and meteorological conditions make it be an ideal indicator region to explore the long-range atmospheric transport and environmental fates of POPs. The lakes and rivers on the Plateau may be particularly sensitive to atmosphere-transported POPs because the sparse vegetation and thin soil have little retention abilities for organic contaminants. A few previous studies (Wang et al., 2009; Wang et al., 2012; Wang et al., 2016; Yang et al., 2013; Zhu et al., 2015) reported that some semi-volatile organic pollutants, such as organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and polycyclic aromatic hydrocarbons (PAHs), were widely presented in various environmental matrices on the Tibetan Plateau. In addition, OCPs, PCBs, PBDEs as well as perfluorinated compounds (PFCs) and hexabromocyclododecane (HBCD) have also been frequently detected in alpine fish from the Tibetan Plateau (Ren et al., 2017; Shi et al., 2010; Yang et al., 2011; Yang et al., 2010b; Zhu et al., 2013). Recently, Wang's group (Wu et al., 2018; Wu et al., 2017) observed an altitude-dependent distribution of SCCPs and MCCPs in atmosphere at the Shergyla mountain and Lhasa on the Tibetan Plateau. However, to our knowledge, there is still no report on SCCPs in Tibetan biota, and the bioaccumulation behavior of SCCPs in high altitude environments also remains largely unknown.

Alpine lakes or rivers provide natural exposure environments of air-transported SCCPs to aquatic organisms. SCCPs accumulated in organisms from high-altitude aquatic environment may be uniquely associated with the atmosphere in this region. Alpine fish may be an important indicator of SCCP accumulation in such region due to their slow growth rate and high lipid content under oligotrophic status and low temperatures (Kidd et al., 1995). In this study, a total of 51 fish samples were collected from five alpine lakes and Lhasa river across the entire Tibetan Plateau. The aims of this work was to investigate the levels, distribution patterns and altitude-dependent accumulation behavior of SCCPs, and to further improve our understanding of long-range transport and fate of SCCPs in the remote alpine region.

2. Materials and methods

2.1. Sampling

A total of fifty-one fish samples were collected from five mountain lakes, namely Qinghai Lake, Basum Lake, Palgon Lake, Yamdro Lake and Manasarovar Lake, as well as Lasha River on the Tibetan Plateau of China. The altitudinal and meteorological information of the sampling sites and the detailed information of these samples including fish species are summarized in Table 1. Other related geographic information of the sampling sites and the characteristic parameters of the fish samples and conditioning factors were reported in previous studies (Yang et al., 2011; Yang et al., 2010b). The five alpine lakes and the Lasha River are located on the Tibetan Plateau at latitudes between 28.9°N and 37.3°N, and longitudes between 79.9°E and 100.3°E, with altitudes ranging from 3225 to 4588 m. Among the alpine lakes, Yamdro Lake and Manasarovar Lake are famous holy lakes of the Tibet Plateau. Yamdro Lake is the biggest inner lake beyond the north foot of the Himalayas at an altitude of 4441 m, while Manasarovar Lake is one of the highest freshwater lakes at an altitude of 4588 m in the world. All investigated lakes are natural origin and regarded to be far from production and usage areas or known pollution source of SCCPs.

Fish samples were collected using fishing nets during 2007–2010. All the catched fish belong to the same family of *Cyprinidae* and subfamily of *Schizothoracinae*, which are unique fish species in the Tibetan Plateau (Yang et al., 2010b). All the fish taken from a single lake were of the same species. In order to avoid potential contaminations, strict control quality measures were taken during the sample collection. Each fish was individually wrapped with solvent-rinsed and prebaked foil, placed into a CP-free polyethylene bag, stored in an ice-cooled box, and then immediately transported to the laboratory. In laboratory, the skin of each fish was removed and only muscle tissue was taken as an individual sample. The muscle tissue was homogenized and freeze-dried, and stored at −20 °C until analysis.

2.2. Sample preparation

Approximately 1 g dry weight of fish sample were spiked with 10 ng surrogate standard $^{13}C_{10}$-*trans*-chlordane and extracted by accelerated solvent extraction (ASE 200, Dionex USA) with n-hexane/dichloromethane (1:1, v/v). The extract was concentrated to 10 mL and an aliquot of the extract (1/10) was taken for gravimetric determination of the lipid content. About 3–10 mL of concentrated sulfuric acid was added into the remnant extract to remove most of the lipids. The cleaned extract was concentrated to a final volume of approximately 2 mL before further cleanup on a 1.5 cm i.d. silica-Florisil composite column, which was self-packed with 14 g of Florisil (deactivated with 1.5% of water), 2 g of neutral

Table 1
Sample information and concentration of SCCPs in alpine fish from lakes and river on the Tibetan Plateau.

Sites	Altitude (m)	Precipitation (mm/yr)[a]	Species	*n*	Age (year)	Lipid (%)	$\sum$SCCPs (ng g^{-1}, dw)		$\sum$SCCPs (ng g^{-1}, lw)	
							mean ± SD	min–max	mean ± SD	min–max
Qinghai Lake	3225	337	*Gymnocypris przewalskii*	10	nd[b]	2.9 ± 1.4	6.5 ± 2.2	3.9–10.9	281 ± 160	93.8–569
Basum Lake	3538	646	*Racoma biddulphi Gunther*	5	7.3 ± 1.0	18.0 ± 4.1	64.6 ± 15.6	38.1–79.4	367 ± 102	271–480
Palgon Lake	4242	61	*Racoma tibetanus*	8	9.9 ± 2.2	3.1 ± 1.3	14.6 ± 11.2	4.4–35.0	622 ± 603	75.7–1610
Yamdro Lake	4441	373	*Gymoncypris waddellii*	17	9.1 ± 2.1	3.1 ± 1.7	17.5 ± 12.3	5.0–55.6	637 ± 349	122–1430
Manasarovar Lake	4588	169	*Gymoncypris waddellii*	6	6.3 ± 0.8	8.6 ± 2.5	58.5 ± 25.0	38.8–107	709 ± 325	445–1340
Lhasa River	3650	372–739	*Oxygymnocypris stewartii*	5	nd[b]	15.9 ± 8.6	40.9 ± 9.0	20.0–48.8	354 ± 275	170–833

[a] Data for alpine lake were from the reference (Xiang and Zheng, 1989); data for river were from the reference (Zhu et al., 2013).
[b] Not determined.

silica gel, 10 g of 44% acidified silica gel and 4 g of anhydrous sodium sulfate from bottom to top. The column was conditioned with 50 mL of *n*-hexane. After the sample was loaded, the first fraction that contained PCBs, toxaphene and most of organochlorine pesticides was pre-eluted with 60 mL of *n*-hexane, followed by 6 mL of dichloromethane. The second fraction containing the target CPs and a part of HCHs was futher eluted with 120 mL of *n*-hexane/dichloromethane (1:1, v/v). The second fraction was collected, concentrated to near dryness under a gentle nitrogen flow, and solvent exchanged to 200 μL of cyclohexane. Prior to analysis by GC/MS, 10 ng of ε-HCH was added as an internal standard.

2.3. Instrumental analysis, identification and quantification

Instrumental analysis was performed using a high-resolution gas chromatography coupled with an electron capture negative ionization - low resolution mass spectrometer (HRGC/ECNI-LRMS, Agilent 7890B-7000D, USA) based on our previously developed method (Zeng et al., 2011a; Zeng et al., 2011b). The detailed procedures for identification and quantification can be also found in our previous publications (Zeng et al., 2017a; Zeng et al., 2017b; Zeng et al., 2015; Zeng et al., 2011a). Briefly, in order to ensure accurate quantification, CP congeners were first identified by carefully comparing the retention time range, signal shape and isotope ratio with their reference standards. The actual relative integrated signals for some congeners that suffered from mass overlapping interference, were corrected by chemical calculation using isotopic abundance and theoretical isotope ratios. The details on chemical calculation procedure have been described previously (Zeng et al., 2011a). The total SCCPs ($\sum$SCCPs) quantification was performed using the procedure described by Reth et al. (2005) on the basis of linear relationship between degrees of chlorination and response factors. A multi-level standard calibration with a coefficient of determination (R^2) ≥ 0.97 was used to quantify.

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

All glassware was ultrasonic rinsed with solvent and heated at 450 °C overnight prior to use. One procedural blank (anhydrous sodium sulfate as blank matrix) was included in each batch of eight samples to monitor the possible blank contamination. SCCPs levels in the blanks were below or close to the detection limits, and therefore the final concentrations of SCCPs reported in this study were not blank-corrected. The recoveries of SCCPs and ^{13}C-*trans*-chlordane in matrix-spiked samples were in the range of 81.0%–96.0% and 80.0%–98%, respectively, with the relative standard deviations (RSDs) below 12.0% ($n = 6$). The surrogate recoveries of ^{13}C-*trans*-chlordane in all field fish samples were between 79.0% and 97.0%. The method detection limits (MDLs) for total SCCPs ($\sum$SCCPs) defined as three times the standard deviation (SD) of the procedure blanks ($n = 6$), were estimated at 3 ng g^{-1} dry weight (dw) and 50 ng g^{-1} lipid weight (lw) in fish samples.

3. Results and discussion

3.1. Concentrations and congener group patterns of SCCPs in alpine fish

Descriptive statistics of concentrations of $\sum$SCCPs in fish samples from five alpine lakes and Lasha River are summarised in Table 1. Of the 51 fish samples, SCCPs were detected in all the samples at concentrations ranging from 3.9 to 107 ng g^{-1} dw (mean: 26.6 ng g^{-1} dw) and from 75.7 to 1610 ng g^{-1} lw (mean: 519 ng g^{-1} lw). The average lipid content in the fish samples varied from 2.9% to 18%. Significant positive relationship was observed between percent lipid content and dry basis concentrations of $\sum$SCCPs ($r = 0.686$, $p < 0.05$), suggesting that lipid content still played an important role in controlling alpine bioaccumulation of SCCPs. This relationship was coincident with the previous findings in other non-plateau environments (Yuan et al., 2012; Zeng et al., 2017a; Zeng et al., 2011b). However, there were no relationships between the age, body weight or length of fishes ($p > 0.05$). Fish samples with the highest (107 ng g^{-1} dw) and lowest (3.9 ng g^{-1} dw) dry basis concentrations of $\sum$SCCPs were found in from Manasarovar Lake and Qinghai Lake, respectively. These two lakes have the relatively highest and lowest altitudes among the investigated sampling sites. Similarly, fish samples with the maximum (709 ng g^{-1} lw) and minimum (281 ng g^{-1} lw) average lipid basis concentrations of $\sum$SCCPs were also found from Manasarovar Lake and Qinghai Lake, respectively. These results indicated that altitude is an important factor affecting bioaccumulation levels of SCCPs in alpine fish.

This is the first report on the detection of SCCPs in biota on the Tibetan Plateau. The average dry basis concentrations of SCCPs determined in alpine fish from the Tibetan Plateau were found to be lower than previously detected levels in cod (mean: 208 ng g^{-1} dw) and gammarids (518 ng g^{-1} dw) from the Svalbard in the Arctic (Li et al., 2017), and also slightly lower than the reported levels in the Antarctic fish Notothenia coriiceps (mean: 69.9 ng g^{-1} dw and 1.50 μg g^{-1} lw) (Li et al., 2016). However, the average lipid basis concentrations of SCCPs in the present study were apparently higher than those in the cod samples from the European Arctic (Reth et al., 2006). In addition, the range of the lipid basis concentrations of SCCPs in fish samples from the Tibetan Plateau was comparable to the early detected levels in marine mammals (129–1580 ng g^{-1} lw) from the Arctic (Tomy et al., 2000). These results indicated SCCPs are prevalent in aquatic ecosystems in the polar environments, and the bioaccumulation behavior and fate of SCCPs merit further investigation.

Congener group patterns of SCCPs can offer penetrating insights into their environmental behavior. Fig. 1 illustrates the congener group abundance profiles of SCCPs for the alpine fish from different lakes and river on the Tibetan Plateau. All profiles showed a similar distribution pattern. Relative to commercial CP formulations (Gao et al., 2018), the congener group C10 with 6–7 chlorine atoms was found to be the most abundant in all alpine fish samples, accounting for 36%–50% of the total composition of SCCPs. The second most abundant congener group was C11, followed by C12 and C13. The significant predominance of shorter carbon chain congeners in the profiles were consistent with the recently reported results, which provided important signals on the long-range atmospheric transport of SCCPs to this region (Wu et al., 2017). Generally, SCCPs show a trend of increasing vapor pressure (VPs) with decreasing carbon chain length and degree of chlorination (Drouillard et al., 1998). The present observation on the enrichment of more shorter chain congeners implied that these volatile components could be more easily transported and cold-trapped in the alpine regions. This specific distribution pattern of SCCP congeners in the fish may be as a result of the congener fractionation during environmental processes of long-range atmospheric transport, mountain cold-trapping effect and bioaccumulation on the basis of their differential behaviors. The congener group patterns of SCCPs for alpine fish from the Tibetan Plateau were also in agreement with those of aquatic organisms from the Arctic and Antarctica (Li et al., 2016; Li et al., 2017). These results provide new proofs on the contamination of SCCPs to the Tibetan Plateau mainly by long-range atmospheric transport.

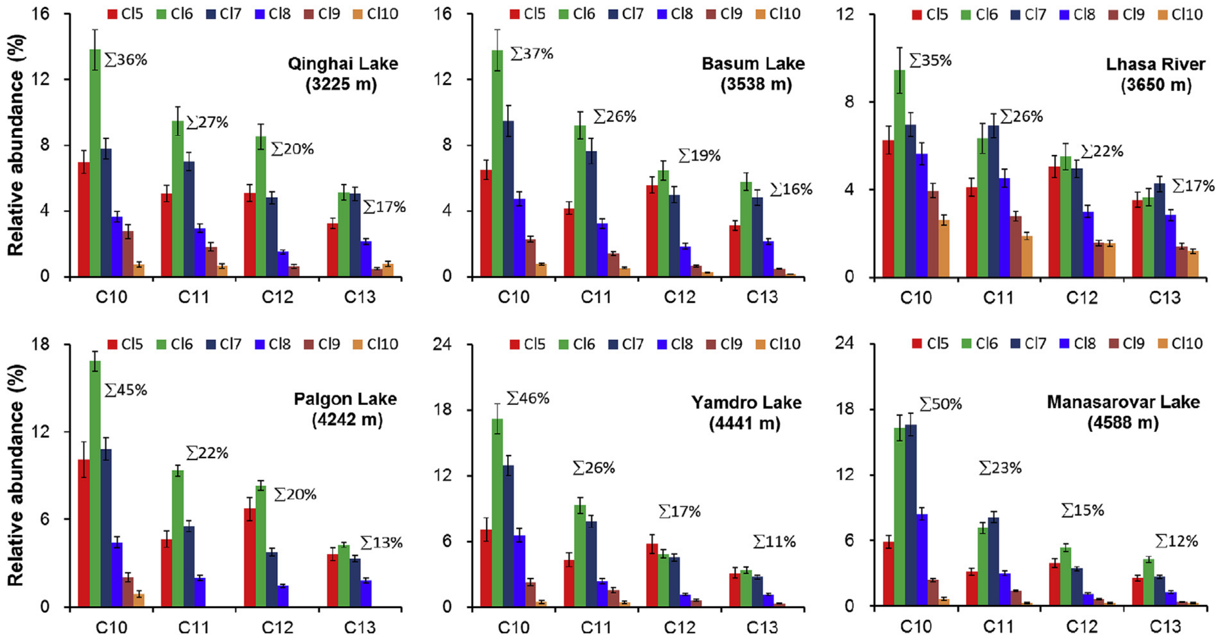

Fig. 1. Congener group abundance profiles of SCCPs in alpine fish from lakes and river on the Tibetan Plateau.

3.2. Comparison of the levels of SCCPs with those of conventional POPs in alpine fish

The levels of conventional POPs including PCBs, PBDEs, HBCDs, hexachlorobenzene (HCB), hexachlorocyclohexanes (HCHs), dichlorodiphenyltrichloroethane and its metabolites (DDTs), perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) in the same fish samples from the Tibetan Plateau have been reported in previous studies (Shi et al., 2010; Yang et al., 2011; Yang et al., 2010b; Zhu et al., 2013). A comprehensive comparison of the levels of SCCPs with those of these conventional POPs was conducted. As illustrated in Fig. 2, SCCPs showed the highest concentrations detected in the same fish samples among the nine POPs. The mean concentrations of SCCPs were approximately two orders of magnitude higher than those of HCB (a mean of 0.21 ng g^{-1} dw with a range of 0.08–0.55 ng g^{-1} dw), PCBs (a mean of 0.32 ng g^{-1} dw with a range of 0.06–0.97 ng g^{-1} dw) and HBCDs (a mean of 0.38 ng g^{-1} dw with a range of 0.04–1.2 ng g^{-1} dw) (Yang et al., 2010b; Zhu et al., 2013), and were several ten times higher than those of HCHs (a mean of 0.55 ng g^{-1} dw with a range of 0.27–0.98 ng g^{-1} dw), PBDEs (a mean of 0.65 ng g^{-1} dw with a range of 0.05–10.5 ng g^{-1} dw) and PFOA (a range of 0.5–1.86 ng g^{-1} dw) (Shi et al., 2010; Yang et al., 2011), and were also several times higher than those of DDT (a mean of 4.0 ng g^{-1} dw with a range of 0.84–10.1 ng g^{-1} dw) and PFOS (a mean of 5.2 ng g^{-1} dw with a range of 1.3–7.54 ng g^{-1} dw) (Shi et al., 2010; Yang et al., 2010b). These comparative results indicated SCCPs were identified to be the most predominant POPs in alpine fish of the Tibetan Plateau until now. Considering potential adverse effects on the environment and human health similar to other POPs, the alpine behavior and fate of SCCPs in biota on the Tibetan Plateau need to be paid adequate attentions, which will provide a better understanding on the transport mechanism and potential ecological risks of SCCPs in the polar environments.

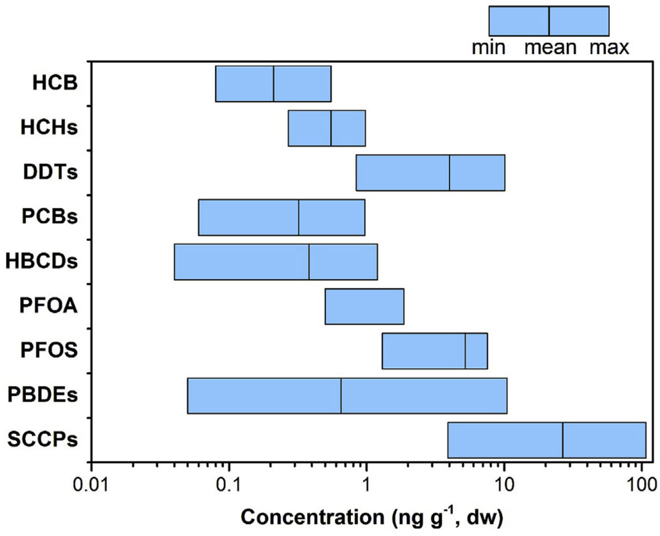

Fig. 2. Comparison of the levels of SCCPs with those of conventional POPs in alpine fish from the Tibetan Plateau. Data were from previous studies (Shi et al., 2010; Yang et al., 2011; Yang et al., 2010b; Zhu et al., 2013).

3.3. Altitude-dependent accumulation of SCCPs in alpine fish

The regression of lipid-normalized $\sum$SCCP concentrations in alpine fish samples against altitude is presented in Fig. 3. A significant linear correlation between $\sum$SCCP average concentrations and attitudes was found ($r = 0.98$, $p < 0.001$). However, the big error bar could be observed in Fig. 3 due to the presence of the only or two outliers at some sampling sites. Therefore, further regression analysis using individual non-outlier $\sum$SCCP concentrations of fish was performed (Fig. S1), and a similarly significant relationship was also obtained ($r = 0.90$, $p < 0.001$). As shown in Fig. 3, a slope of the linear regression was 308 ng g^{-1} km^{-1}, indicating an obviously increasing trend in the concentrations of $\sum$SCCPs in Tibetan fish with increasing altitude of the sampling sites. Two recent studies (Wu et al., 2018; Wu et al., 2017) also reported a similar altitudinal trend of SCCPs and MCCPs in air at Shergyla Mountain and Lasha on the Tibetan Plateau. To our knowledge, the present study is the first

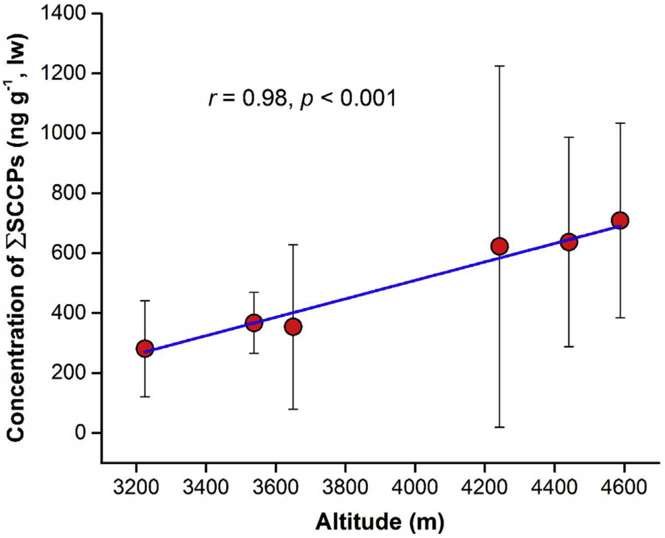

Fig. 3. Altitude-dependent accumulation of average SCCPs in alpine fish from the Tibetan Plateau.

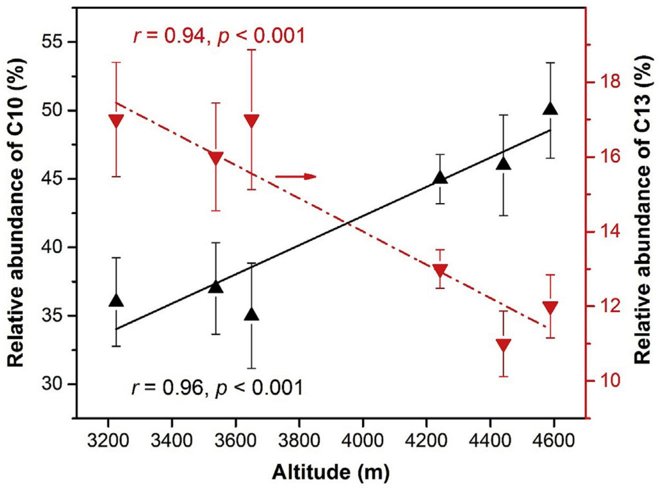

Fig. 4. Trends of percentage relative abundance of C10 (black line) and C13 (red dash dot line) homologues in alpine fish with increasing altitude on the Tibetan Plateau. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

report about the altitude-dependent distribution of SCCPs in fish from the Tibetan Plateau. Such a positive altitudinal gradient of other organic pollutant concentrations in the same fish samples was also found for HCHs, HCB, and methyl mercury (Yang et al., 2011; Yang et al., 2010b), but not found for DDTs, PBDEs, PCBs, PFOA, PFOS, and HBCD (Shi et al., 2010; Yang et al., 2011; Yang et al., 2010b; Zhu et al., 2013). In addition, a few previous studies have also shown that in the mountain slopes of the Plateau (e.g. southeast Tibet and Himalayas), environmental concentrations of some POPs in various mediums, such as soil, lichen, bark, and conifer, pine and spruce needles, increased significantly with increasing altitude (Wang et al., 2009; Wang et al., 2006; Yang et al., 2008; Yang et al., 2013), but opposite results were observed for polycyclic aromatic hydrocarbons (PAHs) (Guzzella et al., 2011; Yang et al., 2013). In general, an altitude-dependent concentration distribution of contaminants in typical high-altitude mountain regions might be associated with the mountain cold-trapping effect, caused by repeating the temperature-driven air surface exchanges (Grimalt et al., 2001; Wania and Westgate, 2008). This differential distribution pattern for different contaminants would be as a combined result of their cold-trapping abilities, persistence during long-range transport, and potential local source.

Furthermore, the changes of percentage contribution of different congener groups ($C_{10}-C_{13}$) to $\sum$SCCPs with altitude were also systematically explored. As shown in Fig. 4, strong linear correlations between the relative abundance of C10 or C13 groups and the altitude of sampling sites were found. Obviously, shorter chain group C10 showed a significant increasing trend in percentage relative abundance with increasing altitude ($r = 0.96$, $p < 0.001$), while longer chain group C13 showed a significant decreasing trend in percentage relative abundance with increasing altitude ($r = 0.96$, $p < 0.001$). No significant correlations were observed for C11 and C12 congener groups. According to the slope values of linear regresses for the relative abundance versus the altitude (Fig. 4), an increasing percentage contribution to $\sum$SCCPs for C10 and a decreasing percentage contribution to $\sum$SCCPs for C13 groups was about 11% and 4.4% per kilometer, respectively, indicating that shorter chain SCCPs, the more volatile components, could be preferably enriched at higher elevation. Wu et al. (2017) recently reported the levels of C10 congener group in Tibetan air increased faster with increasing altitude compared with other SCCP congener groups. Our present findings in Tibetan fish are well in agreement with the previous preliminary results (Wu et al., 2017). It has been confirmed that shorter chain group C10 can be more easily transported to higher altitudinal areas, which provide a strong explanation on C10 as the predominant congener groups in the SCCP distribution patterns.

3.4. Implications for long-range atmospheric transport and mountain cold-trapping effects

The Tibetan Plateau is located in the eastern Eurasia and lies to the north of the Himalayas. As the “third pole” or the “roof of the world”, the average elevation of the Tibetan Plateau was over 4000 m above sea level. Similar to the polar environments in the Artic and Antarctica, high-altitude mountain regions on the Tibetan Plateau have very low temperatures and deep snow accumulation. The water of the mountain lakes in these regions sourced mainly from the snow and ice of surrounding high mountains. Due to sparse population and few modern industry, remote high-altitude mountains are believed to be far away from anthropogenic/industrial activities and local emission sources. In the present study, since there were no major industrial/anthropogenic activities at/around the sampling sites, it was implied that long-range atmospheric transport was the primary external source of SCCPs. Similar to other conventional POPs, SCCPs released into the environment can arrive to the remote or polar regions, and the potentials of long-range atmospheric transport have been confirmed in some previous studies (Ma et al., 2014; Wu et al., 2017).

Behind the long-range atmospheric transport of SCCPs to the Tibetan Plateau, precipitation might be easily considered as an efficient pathway for capturing both particulate and gaseous SCCPs from air to lake water. In this study, no significant correlations were found between the concentrations of SCCPs in fish from the lakes and the annual precipitation of the sampling sites, implying that the main source of SCCPs in most of the sampling sites might be not via wet/dry deposition during the long-range atmospheric transport. The results were also in consistent with the findings of a previous study for most of OCP contaminants (Yang et al., 2010b). Considering that the sampling sites have polar-like environments, the repeating distillation and cold condensation effects, namely known mountain cold-trapping effect, can easily take place. As the most key finding in the present study, the altitude-dependent accumulation of SCCPs in Tibetan fish fitly reflected the mountain cold-trapping effects, which were characterized by an increase in pollutant concentrations corresponding to an increasing altitudinal gradient. Moreover, a clear altitude-dependent distribution of SCCPs in atmosphere at Shergyla Mountain (Wu et al., 2017) also reflected the orographic cold-trapping effects for SCCPs being particularly pronounced on the Tibetan Plateau, and herein further strengthened our present finding. Therefore, it can be concluded that widespread occurrences of SCCPs in alpine fish from the Tibetan Plateau were mainly sourced from the long-range atmospheric transport, and the altitude-dependent distribution of SCCPs was due to the mountain cold-trapping effects and potential susceptibility to bioaccumulation.

4. Conclusion

This study demonstrated that SCCPs were widely accumulated in alpine fish from five lakes and Lasha river on the Tibetan Plateau. The concentrations of SCCPs in fish from the Tibetan Plateau were lower than those found in aquatic organisms from the Artic and Antarctica. Congener group patterns were dominated by shorter chain group C10, the more volatile components of SCCPs. Comparison of the levels of SCCPs with those of conventional POPs indicated that SCCPs were currently the most abundant organic pollutants in alpine fish from the Tibetan Plateau. Significant altitude-dependent accumulation of SCCPs in alpine fish were found. Shorter chain group C10 showed a significant increase trend in percentage contribution to the total SCCPs with increasing altitude, but longer chain group C13 showed an opposite trend. Widespread occurrences of SCCPs in alpine fish from the Tibetan Plateau were mainly sourced from the long-range atmospheric transport, and the altitude-dependent distribution of SCCPs was attributed to a result of the mountain cold-trapping effects and potential susceptibility to bioaccumulation. More investigations are urgently needed to gain a comprehensive understanding on SCCP transport process and accumulation behavior in various environmental mediums and biota on the Tibetan Plateau.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2019.04.059.

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