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Long-range atmospheric transport and alpine condensation of short-chain chlorinated paraffins on the southeastern Tibetan Plateau

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ABSTRACT

Pristine alpine regions are ideal regions for investigating the long-range atmospheric transport and cold trapping effects of short chain chlorinated paraffins (SCCPs). The concentrations and alpine condensation of SCCPs were investigated in lichen samples collected from the southeastern Tibetan Plateau. The concentrations of SCCPs ranged from 3098 to 6999 ng/g lipid weight (lw) and appeared to have an increasing trend with altitude. For congeners, C_{10} dominated among all the congener groups. The different environmental behavior for different congener groups was closely related to their octanol-air partition coefficient (K_{oa}). C_{10} congeners showed an increasing trend with altitude, whereas C_{13} congeners were negatively correlated with altitude. Volumetric bioconcentration factors (BCF) of SCCPs reached 8.71 in lichens, which were higher than other semivolatile organic compounds (SVOCs) such as organochlorine pesticides (OCPs), polybrominated diphenyl ethers (PBDEs), polychlorinated biphenyls (PCBs), and hexabromocyclododecane (HBCD). These results suggested that SCCPs were prone to accumulate in the lichen from the air and provided evidence for the role of lichens as a suitable atmospheric indicator in the Tibetan Plateau.

Introduction

Chlorinated paraffins (CPs) are a multiplex group of industrial mixtures produced primarily by direct chlorination of unbranched alkane mixtures (De Boer et al., 2010). CPs have been widely used in industrial applications, such as additives for extreme pressure lubricants, flame retardants, plasticizers and coating additives (Wei et al., 2016). Short-chain chlorinated paraffins (SCCPs, $C_{10} - C_{13}$) are believed to have carcinogenic effects in rats and mice, and there is also evidence that they affect the digestive and endocrine organs (UNEP, 2016). Additionally, SCCPs are persistent in the environment and are supposed to have bioaccumulation and long-range transport potential similar to those of other persistent organic pollutants (POPs) (UNEP, 2016). SCCPs were listed as emerging POPs in Annex A of the Stockholm Convention in 2017 (UNEP, 2017).

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SCCPs in the environment are primarily derived from the process of production, usage, disposal and recycling of products containing CPs (van Mourik et al., 2016). These CPs discharged into the environment might be transported over long distances in the atmosphere and eventually migrate to the polar regions and experience cold condensation, causing global pollution (Wang et al., 2019). Similar to the "cold condensation" occurring in polar regions, orographic cold trapping in high-altitude areas may remarkably affect the distribution and long-range transport of POPs (Yang et al., 2013). As the world's "third pole", the Tibetan Plateau has an average altitude of higher than 4000 m and is one of the highest and coldest regions on earth (Li et al., 2019). The local source of SCCP is not significant due to the low population density and the scarce industry in the Tibetan Plateau, which makes the Tibetan Plateau an ideal natural laboratory for studying the long-range atmospheric transport and cold condensation behavior of SCCPs.

In consideration of the unavailable electrical power in the remote area of the Tibetan Plateau, different kinds of plants have been extensively used as passive sampling matrices to monitor atmospheric contamination of semivolatile organic compounds (SVOCs) since the 1980s (Eriksson et al., 1989). Pine needles, mosses, tree bark, and lichens are the frequently used plants owing to the relatively high lipid content, widely seasonal availability and large surface area (Zhu et al., 2015). Due to the lack of root-like and barrier structures, lichens and mosses, are considered to absorb pollutants directly from the atmosphere (Yogui et al., 2011) and accumulate air pollution with the passage of time (Simonich and Hites, 1995). Previous results suggested that the profiles of SVOCs in the lichens were similar to the XAD-air-samplers, but the profiles of SVOCs in the mosses were similar to those of the soil (Zhu et al., 2015). Compared with mosses, lichens were totally exposed to the atmosphere and had less interference from soil (Appendix A Fig. S1), which made lichens superior in revealing the atmospheric pollution in remote mountain regions.

The main target of present study was to investigate the contamination levels of SCCPs in lichens and to investigate the volumetric bioconcentration factors (BCF) of SCCPs between lichen and air in the southeastern Tibetan Plateau. The contamination level, spatial distributions, and cold trapping effect of SCCPs in lichen samples were measured to understand the environmental behavior and transmission mechanism of SCCPs in high altitude areas. Furthermore, this study aimed to verify the hypothesis that lichens act as a passive sampler, indicating that the contamination of SCCPs in the southeastern Tibetan Plateau, and to improve scientific understanding of the role that lichens play in delivering SCCPs and other SVOCs from the atmosphere to plant.

1. Materials and methods

1.1. Sampling

The sampling area included both sides of Shergyla Mountain, which is located in the southeastern of the Tibetan Plateau. Compared with other parts of the Tibetan Plateau, the sampling area is featured by Plateau valley terrain, involving the densest virgin forests. This area is dominated by westerly wind in winter and Indian monsoon in summer, which could bring POPs from nearby potential sources into the Tibetan Plateau (Sheng et al., 2013). In this study, lichen samples were collected at 22 sites, with 12 sites in the western slopes and 10 sites in the eastern slopes (**Fig. 1**). Each lichen sample was collected from 1.5 or 2.0 m above the ground and mixed six different spots on the target tree in June 2011. All samples were packed in aluminum foil, sealed in plastic bags, and stored in insulated iceboxes during the sampling campaign. Lichen samples were taken back to the laboratory of Tibet University, then immediately lyophilized and ground, and finally transported to our laboratory in Beijing and kept at −20°C until chemical analysis.

1.2. Analytical procedures

The sample analysis procedures contained the extraction of target compounds using accelerated solvent extraction (Dionex ASE 350, USA) and the purification using a multilayer silica-Florisil column. In short, about 2 g of lichen samples mixed with 10 g of anhydrous sodium sulfate and added with 1 ng of $^{13}C_{10}$-*trans*-chlordane, then extracted with dichloromethane and hexane (1:1, *V/V*) using ASE. The collected extract was rotary-evaporated and then purified on a multilayer silica-Florisil composite column that consisted of 3 g Florisil, 2 g activated silica gel, 5 g acid silica gel (30%, W/W), and 4 g anhydrous sodium sulfate from the bottom to the top. Finally, 10 ng ε-HCH was added before instrumental analysis. The 1 μL of extract were analyzed on a 7890A gas chromatograph in electron-capture negative-ion (ECNI) mode coupled with a 7000B triple quadruple mass spectrometer in single quad mode (Agilent, USA). Detailed information for instrumental analysis, identification and quantification, and lipid content analyses can be found in our previous studies (Li et al., 2016, 2019).

1.3. Quality assurance/quality control

The analytical procedures were performed in lab fume hood and procedure blanks and spiked blanks were used to assess possible contamination. Anhydrous sodium sulfate was used as field blank control throughout the whole sampling process. The concentrations of SCCPs detected in procedural blanks (blank controls throughout the sampling and treatment process) were 2.66–3.38 ng/g, which were less than 5% of the sample concentrations. The limit of detection was defined as the mean concentration in the procedural blanks plus three times the standard deviation, which was estimated to be 4.36 ng/g. The recovery of $^{13}C_{10}$-*trans*-chlordane was between 56% and 91%, with the mean value of 82%.

2. Results and discussion

2.1. Concentrations and altitudinal distribution of SCCPs

SCCPs have been detected in all lichen samples (Table 1), which revealed that SCCPs were pervasively presented in the southeastern Tibetan Plateau. The SCCP concentrations in the lichen samples ranged from 226 to 668 ng/g dry weight (dw), with an average of (445 ± 135) ng/g dw (mean concentration ± standard deviation). The SCCP concentrations in the present study were higher than those in moss samples collected from Antarctica (24.9–62.8 ng/g dw) (Li et al., 2016) and spruce needle samples from the remote Alps (26–460 ng/g dw) (Iozza et al., 2009) and terrestrial plants from the Arctic (52.8–434 ng/g dw) (Li et al., 2017). Compared to the industrial areas in China, SCCP concentrations in the present study were much lower than those in bark samples (320–4270 ng/g dw) and needle samples (400–4010 ng/g dw) collected from Beijing (Wang et al., 2015). Compared to some typical POPs, SCCP concentrations were approximately 10-1000 times higher than the concentrations of organochlorine pesticides (OCPs) (34 ng/g), polychlorinated biphenyls (PCBs) (0.36 ng/g), polybrominated

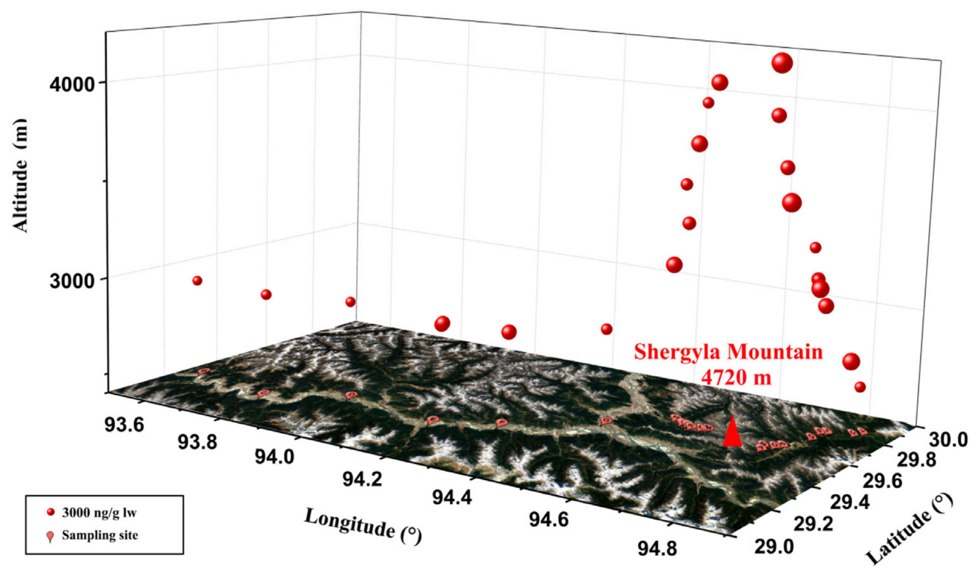

Fig. 1 – Geographic location and altitude of sampling sites and concentrations of SCCPs in lichen samples on the southeastern Tibetan Plateau. Detailed information about the location and altitude is summarized in Appendix A Table S1.

Table 1 – Detailed parameters and concentrations of SCCPs in lichen samples.

Sampling site	Altitude (m)	Lipid content (%)	SCCPs concentration (ng/g dw)[a]	SCCPs concentration (ng/g lw)[b]
1	2596	6.97	245	3509
2	2736	8.31	457	5507
3	2935	9.79	451	4609
4	2943	9.10	310	3414
5	2956	10.4	483	4647
6	2970	7.09	226	3181
7	2994	7.72	239	3098
8	2998	9.42	317	3360
9	3022	11.8	562	4770
10	3123	10.6	599	5642
11	3161	11.7	395	3387
12	3246	7.03	369	5247
13	3334	11.1	395	3562
14	3458	9.76	401	4108
15	3574	7.47	426	5698
16	3644	9.99	426	4268
17	3748	12.9	607	4678
18	3846	11.0	583	5285
19	4000	13.5	641	4763
20	4040	10.2	367	3606
21	4144	12.9	668	5172
22	4250	8.74	612	6999

[a] Dry weight-basis concentrations of short-chain chlorinated paraffins (SCCPs);
[b] Lipid weight-basis concentrations of short-chain chlorinated paraffins (SCCPs).

diphenyl ethers (PBDEs) (0.16 ng/g), and hexabromocyclododecane (HBCD) (0.14 ng/g) in the lichens collected from the same sampling area (Zhu et al., 2015). This conclusion implied that SCCPs might be the most dominant environmental pollutants in the Tibetan Plateau.

The altitudes of the sampling locations varied from 2596 to 4250 m above sea level, and the present of SCCPs in the Shergyla Mountain mainly derived from long-range atmospheric transport from the surrounding industrial areas, such as industrial parks in Pakistan, India, and China (Chaemfa et al., 2014). The SCCP concentrations increased by a factor of approximately 2.73 with the elevation increased from 2596 to 4144 m; correlation analysis indicated that SCCP concentrations in the lichen samples positively correlated with the sampling altitudes ($p<0.01$, Appendix A Fig. S2a), which is similar to the previous finding on SCCPs in air samples and OCPs and PCBs in lichens in the same sampling area (Zhu et al., 2015; Wu et al., 2017). Due to the hydrophobic nature of organic pollutants, they could partition between gaseous and lipophilic substances. In the present study, the lipid contents for the lichen samples were in the range of 6.97% to 13.5% with an average value of 9.89% (Table 1). Positive correlation was found

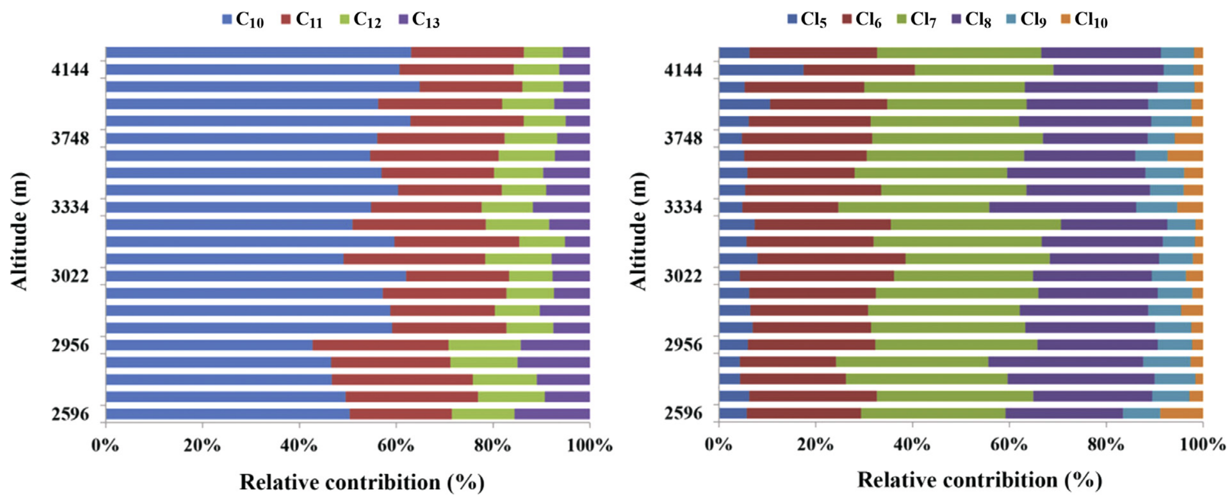

Fig. 2 – Relative contribution of SCCP congener groups in lichen samples from the Tibetan Plateau.

between the lipid contents and the concentrations of SCCPs (p<0.01, Appendix A Fig. S2b). The lipid-normalized concentrations of SCCPs also showed similar increasing trends by altitude (p<0.05, Appendix A Fig. S2c). This result could be due to the cold-trapping effect in the Tibetan Plateau, caused by the temperature-driven air surface exchanges in the typical mountain areas (Wania and Westgate, 2008).

The eastern and western slopes of Shergyla Mountain have complicated and different climates due to the abundant moisture passage from the Yarlung Tsangpo Valley and mountains block. In the present study, the concentrations of SCCPs on the eastern slopes (446 ng/g dw) were comparable with those on the western slopes (443 ng/g dw), which is similar to our previous study on PCBs and PBDEs in moss and soil collected from the same sampling region (Zhu et al., 2015). However, Wu et al. (2017) found relatively higher SCCP concentrations in air samples from the western slopes. The SCCP concentrations showed increasing trend with altitude on both the western slopes and the eastern slopes, and the increase rate on the western slopes was more obvious than that on the eastern slopes. The SCCP concentrations in the western slopes was positive correlated to the altitude (R^2=0.68, p<0.01, Appendix A Fig. S3a), but the correlation on the eastern slopes was not significant (p>0.05, Appendix A Fig. S3b). Based on the previous study, the turbulent water vapor flux from the Yarlung Tsangpo Valley could increase the mixing rates of POPs in the atmosphere above the eastern slopes (Zhu et al., 2015; Wu et al., 2017), causing relatively varied gradient values of SCCPs compared with the western slopes.

2.2. *Congener profiles of SCCPs*

The congener profiles of SCCPs in the Tibetan lichen samples are shown in Fig. 2. The present analysis method (GC-ECNI-LRMS) can only detect 5-10 chlorine atom numbers; in this section, C_{10} congener represents the C_{10} with $Cl_{5\text{-}10}$ and the same with C_{11}, C_{12}, C_{13}. The chlorine content of SCCPs ranged from 62.5% to 64.2% in lichens (Appendix A Table S1). The C_{10} congener groups and Cl_7 congener groups were the most predominant congeners in all the Tibetan lichen samples (C_{10} congener: 56.2%, Cl_7 congeners: 31.6%, Fig. 2). The profiles of SCCPs congener groups in the present study were consistent with those of the plant samples from the Nam Co Lake Basin (Li et al., 2019) and in air samples at Shergyla Mountain (Wu et al., 2017), which suggested that there was a similar source of SCCPs in the Tibetan Plateau (from long range atmospheric transport). Previous studies reported that SCCPs were dominated by relative long-chain congener groups (C_{12} and C_{13}) in urbanized and industrial samples (Sun et al., 2017; Zeng et al., 2017). The difference of SCCPs profiles between the industrial region and Shergyla Mountain might be due to the fractionation process of SCCPs during the long-range transport. The different profiles of SCCP congener groups between the remote and industrial areas could also be influenced by the physicochemical properties of different congener groups. Shorter carbon chain congeners of SCCPs are more easily migrated to remote areas due to their high volatility and low K_{oa} values (Drouillard et al., 1998; Li et al., 2019).

The C_{10} congeners in lichen samples showed an increasing trend with altitude (p<0.05), while C_{13} congeners were negatively correlated with altitude (p<0.05, Appendix A Table S2). Moreover, considering the number of chlorine substitutions, no relationship was found between altitude and $Cl_{5\text{-}10}$ congeners (p>0.05, Appendix A Table S2) in the present study. This result implied that the SCCP congeners with shorter carbon chains had higher transport abilities to higher altitudes. The octanol-air partition coefficient (K_{oa}) is a key physicochemical parameter describing the distribution behavior of organic pollutions in air and organic phase. The average K_{oa} values of the C_{10} to C_{13} congeners were 10.4, 11.2, 11.6 and 12.1 (Wu et al., 2017), which suggests that longer chain congener groups of SCCPs with higher logK_{oa} values are more inclined to accumulate in low altitude than shorter chain congener groups.

2.3. *Lichen–air accumulation of SCCPs and other SVOCs*

Forest regions cover almost half the area of the southeastern Tibetan Plateau. Lichens can provide an organic surface for the partitioning of SCCPs and other SVOCs in the atmosphere and enhance the net atmospheric deposition of these organic compounds (McLachlan and Horstmann, 1998). Our previous studies investigated OCPs, PCBs, PBDEs, and HBCD in lichen and air samples in the same area (Zhu et al., 2014, 2015), and we also reported the SCCPs in air samples from the southeastern Tibetan Plateau (Wu et al., 2017). Volumetric bioconcentration factors (BCFs) were calculated as the ratio of the concentrations of SCCPs and other SVOCs in lichens to their concentrations in air samples from the Shergyla Mountain using the

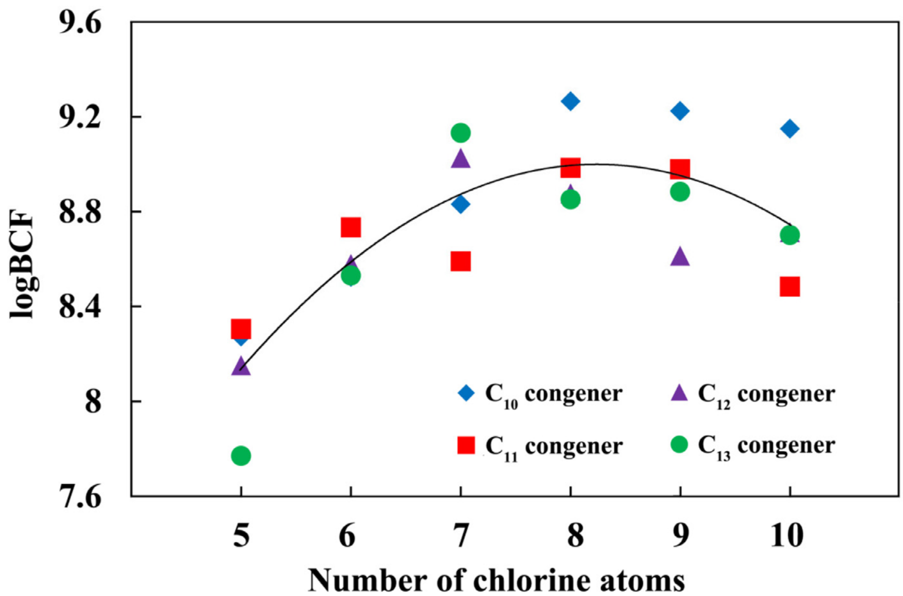

Fig. 4 – Relationships between logBCF and the number of chlorines (logBCF = 3.38 + 1.36 × Number of chlorine atoms – 0.0828 × Number of chlorine atoms^2, R^2 = 0.663, p < 0.01).

following equation (Morris et al., 2018):

$$BCF = \frac{C_{lichen} \times \rho_{lichen} \times 10^6}{C_{air}}$$

where, C_{lichen} (pg/g ww) is the wet weight concentrations of SCCPs and other SVOCs in lichen samples, ρ_{lichen} (0.64 g/cm^3) is the density of the lichen (Kelly and Gobas, 2001), 1.0 × 10^6 is the pg/cm^3 to pg/m^3 conversion factor, and C_{air} (pg/m^3) is the air concentration of SCCPs and other SVOCs (Zhu et al., 2014; Wu et al., 2017).

The logBCF values of different SCCPs congeners and other SVOCs showed a relatively large difference (Appendix A Table S3). The greatest logBCF values were observed for BDE-154 (logBCF = 9.58), whereas the smallest logBCF values were found for pentachlorobenzene (logBCF = 6.49). The average log BCF values of OCPs, HBCD, PCBs, PBDEs and SCCPs were 7.78, 8.14, 8.35, 8.53, and 8.71, respectively. The high logBCF values of SCCPs suggested that SCCPs might be more prone to accumulate in the lichens from the air than OCPs, HBCD, PCBs, and PBDEs. Physicochemical properties play important roles in the bioaccumulative capacity of POPs. The logBCF values of SCCPs and other SVOCs were positively correlated with their logK_{oa} and logK_{ow} (Fig. 3), suggesting that lichens were more efficient for accumulating more lipophilic POPs as the atmospheric indicator.

The log BCF values for individual SCCPs congeners varied within a relatively small range among different carbon atoms. However, a parabolic correlation was found between the log BCF values and the chlorine atom numbers of SCCPs (Fig. 4). The vertex for the curve was found at #Cl = 8.21, which indicated that the Cl_8 congener groups were more likely accumulated from the air to the vegetation. These can be explained that SCCP congeners with fewer chlorine atoms presented greater biodegradation ability than the congeners with more chlorine atoms (Javorska et al., 2009). Whereas, more-chlorinated congener groups of SCCPs with the large molecular volume restricted contaminant penetration into vegetation cellular membranes due to the molecular steric hindrance (She et al., 2013).

3. Conclusions

In this study, we first investigated the occurrence and environmental behavior of SCCPs in lichens collected from the southeastern Tibetan Plateau. This work confirmed the potential mountain cold trapping ability of SCCPs in the Tibetan Plateau, similar to other legacy POPs. The different environmental behaviors for different congener groups are closely related to the physicochemical properties of SCCPs, such as logK_{oa}. Although our previous study has shown that SCCPs are trophic dilution in the Plant–Plateau Pika–Eagle Food Chain, this study shows that SCCPs can be accumulated in lichens with high lipid content, which may be related to the lack of enzymes that metabolize SCCPs in lichens. Lichen−air accumulation of SCCPs emphasizes that SCCPs might more prone to accumulate in the lichens from the air than other four kinds of SVOCs. These results provided evidence for the role of lichens as a suitable atmospheric indicator in the Tibetan Plateau.

Declaration of Competing Interest

The authors declare no conflicts of interest in this work.

Acknowledgments

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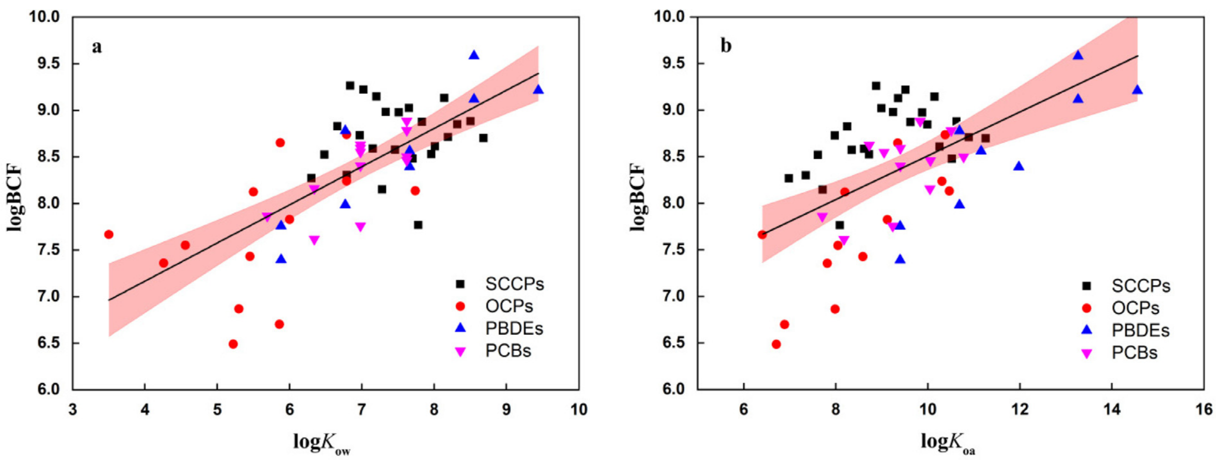

Fig. 3 – Relationships between logBCF and logK_{ow} of organic pollutants (a: logBCF = 5.53 + 0.410 × logK_{ow}, R^2 = 0.503, $p < 0.01$) and logK_{oa} (b: logBCF = 6.17 + 0.235 × logK_{oa}, R^2 = 0.320, $p < 0.01$).

Appendix A. Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2020.06.023.

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