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Occurrence of short- and medium-chain chlorinated paraffins in soils and sediments from Dongguan City, South China☆

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ABSTRACT

As a group of emerging organic pollutants, chlorinated paraffins (CPs) have attracted rising global attention due to their persistence and toxicity. In this study, we have investigated the concentration levels and profiles of short-chain chlorinated paraffins (SCCPs) and medium-chain chlorinated paraffins (MCCPs) in soils and sediments from Dongguan City, an industrial area in South China, and have also screened very short-chain chlorinated paraffins (vSCCPs) by means of ultra-high resolution liquid chromatograph coupled with an Orbitrap Fusion Tribrid mass spectrometer. The results indicated that total SCCP concentrations ranged from 6.75 to 993 ng/g (mean 172 ng/g) in soils and from 4.00 to 613 ng/g (mean 153 ng/g) in sediments, respectively. Higher MCCP levels were observed with a range of 23.9 −2427 ng/g (mean 369 ng/g) in soils and 14.0−1581 ng/g (mean 493 ng/g) in sediments, respectively. The results indicated that MCCPs dominated over SCCPs in the studied region. The dominant homologues in soils and sediments were $C_{13}Cl_{6-7}$ and $C_{14}Cl_{7-8}$, $C_{13}Cl_7$, and $C_{14}Cl_{7-8}$, respectively. Furthermore, six vSCCP homologues (C_8Cl_{7-8} and C_9Cl_{5-8}) in soils and four vSCCPs (C_9Cl_{5-8}) in sediments have been identified. Because of their higher detection frequencies, further studies should focus on the transformation mechanisms and toxicities of these vSCCPs in environmental media and biota.

1. Introduction

Chlorinated paraffins (CPs) are important industrial additives and have been widely used as plasticizers, leather fat liquors, sealants, and flame retardants since the 1930s (Bayen et al., 2006; Wang et al., 2017; Yuan et al., 2017a). According to their carbon chain lengths, CPs can be divided into three categories: short-chain (SCCPs, C_{10-13}), medium-chain (MCCPs, C_{14-17}), and long-chain (LCCPs, C > 18) (Bayen et al., 2006). During their production, storage, transportation, and usage, CPs may be released from materials, allowing their diffusion into various environmental media. As a result, they have become ubiquitous global organic pollutants (Van Mourik et al., 2015). Specifically, SCCPs were listed as new persistent organic pollutants (POPs) in the Stockholm Convention as a result of their environmental persistence, bioaccumulation, long-range transportation and toxicity (POPRC, 2017). Consequently, they have been gradually phased out in Japan, the United States, Canada, and Europe since the end of last century (Gluge et al., 2016; WCC, 2013; WCC, 2014). Restriction on the application of SCCPs may have led to increased usage of alternatives such as MCCPs. However, the PBT properties (persistence, bioaccumulation potential, and toxicity) of MCCPs have been less well studied and are still a matter of debate. MCCPs are more difficult to quantify accurately than SCCPs (Gluge et al., 2018). Therefore, limited data are available on the co-occurrence of SCCPs and MCCPs and their environmental behavior.

During the last decades, numerous studies have been concerned with SCCP levels and profiles in soils, sediments, biota, and humans (Bayen et al., 2006; Van Mourik et al., 2016). Some researchers have also observed very short-chain chlorinated paraffins (vSCCPs) with carbon chains shorter than C_{10} in soils, sediments, indoor dust and even in biotas (Friden et al., 2011; Xia et al., 2019; Zhou et al., 2019). Interestingly, since 2010, most research on CPs has been conducted in China, and the reported SCCP levels in China seemingly exceed those in other countries (Du et al., 2018; Gluge et al., 2018; Van

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Mourik et al., 2016; Zhuo et al., 2019). This may be related to China being the largest producer at present, and the production capacity reached 1,600,000 tonnes/year in 2013 (Van Mourik et al., 2016; WCC, 2014). It should be noted that commercial CP products in China are classified according to chlorine mass rather than carbon chain length (Li et al., 2018b). In other words, the mainly used CP products, such as CP42 (ca. 42% chlorine), CP52 (ca. 52% chlorine), and CP70 (ca. 70% chlorine), contain both SCCPs and MCCPs simultaneously (Li et al., 2018b). Considering that SCCPs could cause liver toxicity in rainbow trout at high exposure levels (Fisk et al., 2000), and also show hepatotoxicity and immunomodulatory effects in rats (Geng et al., 2019; Wang et al., 2019), however, a recent study has suggested that MCCPs may exhibit similar or even higher bioaccumulative potential than the already restricted SCCPs (Castro et al., 2018). Therefore, it is necessary to investigate the concentration levels and profiles of both SCCPs and MCCPs in environmental media.

Dongguan City, located in the east central part of the Pearl River Delta Economics Zone, has undergone rapid industrialization and urbanization since the end of the 1970s (Cai et al., 2010). It is a highly industrialized manufacturing area, including electrical machinery, textile and garment, furniture, toy, and papermaking industries (Zeng et al., 2014). In this study, we collected soil and sediment samples from Dongguan City. The objectives of this study were: 1) to investigate the levels and profiles of SCCPs and MCCPs in soils and sediments, 2) to trace their possible source in the studied region, and 3) to screen and identify vSCCPs.

2. Experimental

2.1. Chemicals and materials

SCCP standards (51.5%, 55.5%, and 63% chlorine contents) and MCCP standards (42%, 52%, and 57% chlorine contents) in cyclohexane (100 ng μL^{-1}) were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). $^{13}C_{12}$-labeled α-hexabromocyclododecane (α-HBCD), purchased from Cambridge Isotope Laboratories (Andover, MA, USA), was used as a surrogate standard.

All solvents used in extraction and analytical procedures were HPLC grade. Hexane, methanol, and acetonitrile were obtained from Merck (Darmstadt, Germany). Dichloromethane (CH_2Cl_2) was obtained from CNW Technologies (Düsseldorf, Germany). Florisil (60–100 mesh) was purchased from CNW Technologies (Düsseldorf, Germany) and activated at 550 °C before use. Silica gel (70–230 mesh), purchased from Merck (Darmstadt, Germany). Silica gel used in Step 1 (column A) was activated at 130 °C for 10 h, while was activated at 550 °C for 10 h for Step 2 (column B). Alumina (100–200 mesh), purchased from Guangzhou Chemical Reagent Factory (Guangzhou, China), was soxhlet-extracted with methanol and dichloromethane, each for 48 h, then activated at 250 °C and deactivated with 3% ultrapure water. Sodium sulfate was baked at 450 °C for 4 h and stored in sealed desiccators.

2.2. Sample collection

Between September and December of 2011, a total of forty-nine surface soil samples (SO1–49) and seventeen sediment samples (SD1–17) were collected from Dongguan City, South China (Fig. 1). Topsoil samples were mainly from different industrial zones and their surrounding areas, including garbage- and coal-fired power plants, electronics factories, hardware factories, landfills, toy factories, furniture factories, and textile mills. Each surface soil sample (0–20 cm) was collected using a pre-cleaned stainless steel shovel and was then packed in aluminum foil. Sediment samples (0–5 cm) were collected from reservoirs (SD1-7) and rivers (SD8-17) using a Van Veen stainless steel grab, and stored in solvent-rinsed aluminum containers. After collection, all samples were carried back to the laboratory, freeze dried, ground, and homogenized, passed through a 1 mm sieve, and stored in brown bottles at −20 °C until analysis. Total organic carbon (TOC) contents in soil and sediment samples were determined with a total organic carbon analyzer (VARIO EL cube, Germany).

2.3. Extraction, cleanup, and instrumental analysis

The analytical method for extraction, purification, and determination of the target CPs was published in our previous study (Wu et al., 2019). Here, it is described in brief. Approximately 10 g of a soil/sediment sample was spiked with a known amount of $^{13}C_{12}$-labeled α-HBCD, and then soxhlet-extracted with CH_2Cl_2 (200 mL) for 72 h. After concentration, the residue was taken up in hexane (ca. 1 mL), and the extract was subjected to clean-up on a multilayer silica gel/alumina column (column A, i.d. 1 cm) that had been sequentially wet-loaded with neutral silica gel (130 °C), acidic silica gel (concentrated sulfuric acid/silica, 44:56, w/w, 130 °C), neutral alumina, and anhydrous Na_2SO_4 from bottom to top. The target CP fraction was collected and subjected to further clean-up on a multilayer silica gel/Florisil column (column B, i.d. 1 cm), which had been wet-packed with Florisil (550 °C), neutral silica gel (550 °C), acidic silica gel (concentrated sulfuric acid/silica, 30:70, w/w, 550 °C), and anhydrous Na_2SO_4 from bottom to top. The collected fraction was concentrated to near dryness under a gentle stream of nitrogen and redissolved in methanol (400 μL) prior to analysis.

Instrumental analysis was conducted with a Thermo Ultimate 3000 ultra-high-resolution liquid chromatograph coupled with an Orbitrap Fusion Tribrid mass spectrometer (Orbitrap Fusion TMS, Thermo Fisher Scientific, USA). Samples (1 μL) were separated on a Thermo Scientific Hypersil Gold C_{18} column (2.1 × 100 mm; 1.9 μm). The column was maintained at a flow rate of 0.3 mL min^{-1} with a mobile phase consisting of (A) water (containing 0.02% acetic acid and 5 mM ammonium acetate), (B) methanol, and (C) acetonitrile. The Orbitrap Fusion TMS was operated in electrospray ionization (ESI) in negative ion mode with the following parameters: detector type, Orbitrap; mass range, 200–700 m/z; MS resolution, 120,000 FWHM; spray voltage, 3900 V; ion transfer tube temperature, 275 °C; vaporizer temperature, 300 °C. The most two abundant m/z signals of $[M-H]^-$ of CPs and $^{13}C_{12}$-labeled α-HBCD were used for quantification and qualification with 5 ppm mass tolerance. Data processing of CP chromatographic peaks was performed with Xcalibur Qual and Quan Browser software.

2.4. Quality assurance and quality control

Quality control and quality assurance were performed to ensure the accuracy of the results. Laboratory blanks, spiked blanks (spiked standards in solvents), and spiked matrices (spiked standards in pre-extracted soils/sediments) were analyzed in the same way as real samples. The average laboratory blank contents of SCCPs and MCCPs were 5.91 and 8.51 ng/g, respectively. Method detection limits (MDLs) of SCCPs and MCCPs were defined as their average concentrations in the laboratory blanks plus three times the standard deviation, and amounted to 8.90 and 12.8 ng/g for SCCPs and MCCPs in sediment/soil samples, respectively. The average recoveries of SCCPs (55.5% Cl) and MCCPs (52% Cl) were 105 ± 3.6% and 98.6 ± 5.2% from the spiked blanks, and 96.2 ± 6.6% and 92.3 ± 6.2% from the spiked matrices, respectively. The average recovery rate of the surrogate from soil and sediment samples was 94.6 ± 7.1%. The reported SCCP and MCCP concentrations in this study are not corrected for recovery rate, but the corresponding background values (the average SCCP and MCCP concentration in

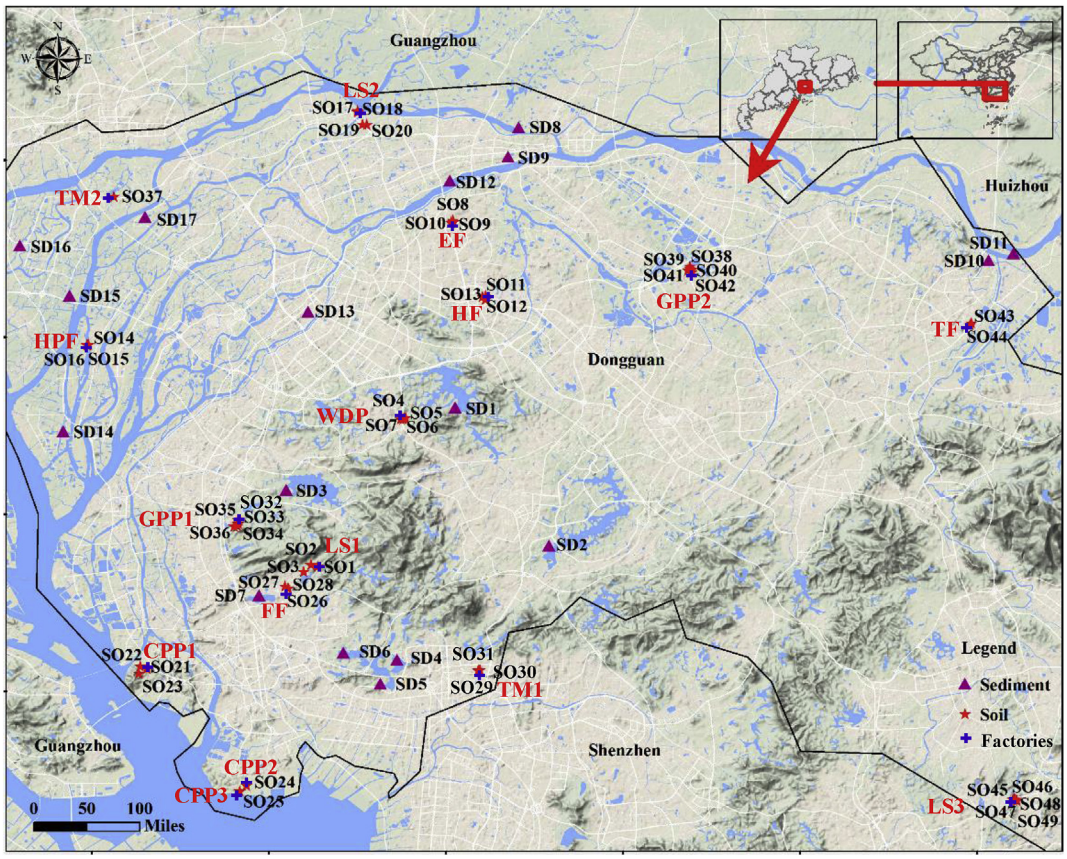

Fig. 1. Sampling sites of soils (SO) and sediments (SD) from Dongguan City, China (LS: Landfill site; WDP, Waste disposal plant; EF, Electronics factory; HF, Hardware factory; HPF, Hardware and plastic factory; CPP, Coal-fired power plant; FF, Furniture factory; TM, Textile mill; GPP, Garbage-fired power plant; TF, Toy factory).

Laboratory blanks) of SCCPs and MCCPs have been subtracted.

Moreover, in order to assure the accuracy of quantification for CPs, we also calculated the goodness of fit (R^2) of the reconstructed CP patterns in all soils/sediments. The results showed that 90.9% of these samples with R^2 value greater than 0.5 for SCCPs (Range, 0.32−0.86; median, 0.64), and 100% of these samples with R^2 value greater than 0.8 for MCCPs (Range, 0.87−0.99; median, 0.98), indicating the obtained CP values in this paper were reliable (Wu et al., 2019).

3. Results and discussion

3.1. Levels and possible sources of SCCPs and MCCPs in soils and sediments

3.1.1. SCCPs and MCCPs in soils

As shown in Fig. 2 and Table 1, both SCCPs and MCCPs were detected in all soil samples. The total concentrations of SCCPs and MCCPs in soils ranged from 6.75 to 993 ng/g (mean 172 ng/g) and from 23.9 to 2427 ng/g (mean 369 ng/g), respectively. The highest SCCP level of 993 ng/g was found in the soil close to a coal-fired power plant (SO24), which may have been associated with coal-mining and power plant operation, including leachate from a conveyer belt and lubricant (Brandsma et al., 2019; Gao et al., 2012). The highest MCCP level of 2427 ng/g was found in the soil close to a factory that main produce hardware and plastic (SO15), which may have been attributable to nearby industrial activities of plastic and hardware production (Pribylova et al., 2006). This result is consistent with a report published by Pribylova et al. who found a high MCCP concentration (5575 ng/g) in river sediment collected near a factory involved in hardware manufacture in the Czech Republic (Pribylova et al., 2006). CPs are widely used as plasticizers/flame retardants in plastics, in paints and other coatings, leathers, textiles, and sealing compounds (Fiedler, 2010; Van Mourik et al., 2016; Wei et al., 2016), as well as in poly(vinyl chloride) (PVC) (Wang et al., 2018b). Consequently, it is reasonable that considerably high levels of CPs were found in soils close to versatile factories, such as furniture factory (SO26, SCCPs, 883 ng/g; MCCPs, 1306 ng/g), textile factory (SO30, SCCPs, 842 ng/g; MCCPs, 1727 ng/g), solid waste incineration power plant (SO35, SCCPs, 433 ng/g; MCCPs, 1323 ng/g), and toy factory (SO43, SCCPs, 331 ng/g; MCCPs, 1389 ng/g). In addition, waste incineration also contributed to the load in SO35.

To our knowledge, the classification of commercial CP products in China is based on chlorine content rather than carbon chain length (Li et al., 2018b). In other words, the commonly used CP products CP42, CP52, and CP70 contain both SCCPs and MCCPs, albeit in different ratios (Li et al., 2018b). Thus, to some extent, the ratio of MCCPs to SCCPs (M/S) can reflect the compositions of commercial CP formulations commonly used in recent years (Li et al., 2018a). The M/S ratios in most soil samples were above 1, in the range 0.55−4.97 (mean 2.23). The results indicated that MCCPs dominated over SCCPs in the studied region, suggesting the use of commercial CP formulations containing more MCCPs in recent years. This is consistent with results obtained for soil samples from the Pearl River Delta (PRD) and Shanghai, China (Wang et al., 2017; Wang et al., 2013b). Moreover, significant correlation was observed between the level of SCCPs and the corresponding level of MCCPs ($R^2 = 0.77$, $p < 0.01$, Fig. S1), indicating that both SCCPs and MCCPs have a similar emission source input, which is also consistent with reported results for soil samples from the PRD,

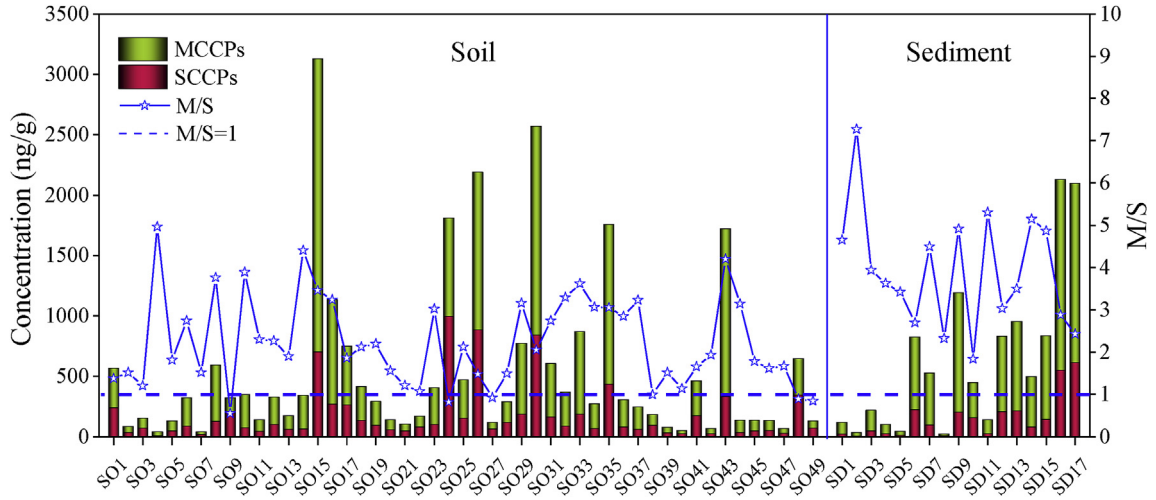

Fig. 2. Concentrations of SCCPs and MCCPs and the ratios of MCCPs to SCCPs (M/S) in soils and sediments.

Table 1
Comparison of SCCPs and MCCPs in soils and sediments with those studies reported.

Sample information		SCCPs			MCCPs			Reference
Soil (ng/g)								
Type and sites	N[a]	Mean	Median	Range	Mean	Median	Range	
Top soil, Dongguan, China	49	172	85.4	6.75–993	369	204	23.9–2427	This study
Surface soil in a CPs plant, Dalian, China	2	1421		1018–1824	2051		2028–2075	Xu et al. (2016)
Surface soil from the surrounding of a CPs plant, Dalian, China	23	142	107	24.8–482	199	94.7	19.3–1461	Xu et al. (2016)
Surface soil in a CPs plant, Shandong Province, China	6	–[b]	–	28,000–554,000	–	–	–	Wang et al. (2018b)
Surface soil from farmlands and villages around a CPs plant, Shandong Province, China	8	–	–	102–441	–	–	–	Wang et al. (2018b)
Farmland soils irrigated with wastewater, Beijing, China	13	–	–	159.9–1450	–	–	–	Zeng et al. (2011)
Paddy and upland soils, Liaohe River Basin, China	14	114	110	56.9–189	–	–	–	Gao et al. (2012)
Paddy soils from e-waste dismantling area, Taizhou, China	32	80.2	–	30.4–530	–	–	–	Yuan et al. (2017b)
Urban surface soil, Shanghai, China	75	39.4	15.7	ND–615	15.5	7.98	1.95–188	Wang et al. (2014)
Suburb surface soil, Shanghai, China	101	18.8	3.52	ND–697	42.3	15.3	ND–666	Wang et al. (2017)
Surface soils, Guangzhou, China	44	10.3	–	1.45–25.5	–	–	–	Huang et al. (2016)
Surface soils, Chengdu, China	30	1.43	–	0.218–3.26	–	–	–	Huang et al. (2016)
Archived soils, Switzerland	35	–	–	3.0–35	–	–	5.1–160	Bogdal et al. (2017)
Background soils, UK	26	16	–	<0.8–179	–	–	–	Halse et al. (2015)
Background soils, Norway	32	12	–	<0.8–281	–	–	–	Halse et al. (2015)
Sediment (ng/g)								
Type and sites	N.	Mean	Median	Range	Mean	Median	Range	
Reservoir sediment, Dongguan, China	7	59.8	21.6	4.00–223	206	96.6	29.1–601	This study
River sediment, Dongguan, China	10	219	179	6.01–613	694	658	14.0–1581	This study
River/Coastal sediment, Shenzhen, China	8	317	401	14.7–574	960	541	10.9–2500	Zeng et al. (2017)
River/estuary sediment, Liaohe River, China	42	212	–	39.8–480	–	–	–	Gao et al. (2012)
River sediment, Yellow River, China	13	–	80	11.6–9760	44.8	25.8	8.33–168	Qiao et al. (2016)
River sediment, Czech Republic	31	27.2	–	ND–347	345	–	ND–5575	Pribylova et al. (2006)
Marine sediment, Hong Kong, China	35	22	12.2	<LOD–75.9	58.7	12.5	<LOD–26.0	Zeng et al. (2017)
Marine sediment, East China Sea, China	48	25.9	–	5.8–64.8	–	–	–	Zeng et al. (2012)
Marine sediment, Bohai and Yellow Sea, China	117	38.4	–	14.5–85.2	–	–	–	Zeng et al. (2013)
Marine sediment, Tokyo Bay, Japan	8	10.3	8	1.3–27.4	19.2	16.6	3.2–56.8	Zeng et al. (2017)
Marine sediment, North and Baltic Seas	20	75	26	5–377	79	29	5–499	(Huttig and Oehme, 2005)

[a] Number of samples.
[b] Not reported.

China (Wang et al., 2013b).

Compared with reported levels of CPs in soils worldwide, the CP concentrations in soils from Dongguan City are at a medium level (Table 1). The SCCP concentrations in soils in the present study are comparable to those reported from different regions of China, such as soil in the Liaohe River Basin (56.9–189 ng/g; mean 114 ng/g) (Gao et al., 2012) and an e-waste dismantling area (30.4–530 ng/g; mean 80.2 ng/g) (Yuan et al., 2017b), but much lower than those

from CP production plants (range 1018–554,000 ng/g) (Wang et al., 2018a; Xu et al., 2016) and wastewater irrigated farmland (159–1450 ng/g) (Zeng et al., 2011). At the same time, the measured SCCP levels are higher than those from other countries, including Switzerland (3.0–35 ng/g) (Bogdal et al., 2017), the UK (<0.8–179 ng/g; mean 16 ng/g) (Halse et al., 2015), and Norway (<0.8–281 ng/g; mean: 12 ng/g) (Halse et al., 2015).

For MCCPs, the levels detected in the studied region were also lower than those from the CP production plant (2028–2075 ng/g; mean 2051 ng/g), but comparable to those in soils surrounding the plant (19.3–1461 ng/g; mean 199 ng/g) (Xu et al., 2016), and higher than those in urban soils (1.95–188 ng/g; mean 15.5 ng/g) from Shanghai (Wang et al., 2014; Wang et al., 2017) and archived soils from Switzerland (5.1–160 ng/g) (Bogdal et al., 2017).

3.1.2. SCCPs and MCCPs in sediments

The detection percentages of SCCPs and MCCPs in all sediments were 100%, indicating their ubiquity. The total concentrations of SCCPs and MCCPs in sediments varied in the ranges 4.00–613 ng/g (mean 153 ng/g) and 14.0–1581 ng/g (mean 493 ng/g), respectively (Fig. 2 and Table 1). It is noteworthy that the median levels of SCCPs and MCCPs in sediments from reservoirs (SD1-7, SCCPs, 21.6 ng/g; MCCPs, 96.6 ng/g) were 8-fold and 7-fold lower than those from rivers (SD8-17, SCCPs, 179 ng/g; MCCPs, 658 ng/g). This might be ascribed to the fact that these reservoirs act as important drinking water sources for local residents, and therefore strict regulation on these areas is imposed by local government. Meanwhile, the studied river received treated/untreated industrial effluent, leading to elevated pollutant concentrations in the sediment (Feng et al., 2012). For example, considerably high concentrations of SCCPs and MCCPs were found in SD16 (SCCPs, 549 ng/g; MCCPs, 1581 ng/g) and SD17 (SCCPs, 613 ng/g; MCCPs, 1485 ng/g). This may have been associated with some factories on both banks upstream of the two sampling sites, which were engaged in the production of leather, textiles, paper, and hardware, as well as a vehicle maintenance site, and may have discharged treated/untreated industrial wastewater into the rivers (Fiedler, 2010; Heath et al., 2006; Wang and Wang, 2008).

The M/S ratios in all sediments were also greater than one, with a range from 1.84 to 7.27 (mean 3.90). The results were similar to those for soils, however significant difference between the mean of M/S value in sediments and soils was observed (T-test, $p < 0.01$). Similar findings were obtained in other studies of sediments from the PRD (Zeng et al., 2017), and even in analyses of marine mammals from the South China Sea (Zeng et al., 2015). These results again suggested that MCCPs are the major components in Chinese commercial CPs. In addition, a significant correlation was again observed between the levels of SCCPs and the corresponding levels of MCCPs in sediment samples ($R^2 = 0.90$, $p < 0.01$, Fig. S1), consistent with results obtained for all surface sediments from the PRD and Hong Kong, China ($R^2 = 0.93$, $p < 0.001$) (Zeng et al., 2017).

The mean concentrations of SCCPs in sediments from Dongguan City were comparable to those detected in sediments from around China, including Shenzhen (14.7–574 ng/g, mean 317 ng/g) (Zeng et al., 2017), Liaohe River (39.8–480 ng/g, mean 212 ng/g) (Gao et al., 2012), and Yellow River (11.6–9760 ng/g, median 80 ng/g) (Qiao et al., 2016), but were higher than those from Hong Kong (<LOD–75.9 ng/g, mean 22 ng/g) (Zeng et al., 2017), the East China Sea (5.8–64.8 ng/g, mean: 25.9 ng/g) (Zeng et al., 2012), and Bohai Bay and the Yellow Sea (14.5–85.2 ng/g, mean 38.4 ng/g) (Zeng et al., 2013). Compared to other countries, the SCCP levels in sediments examined in this study were comparable to those from the North and Baltic Seas (5–377 ng/g, mean 75 ng/g) (Huttig and Oehme, 2005), but higher than those from Tokyo Bay, Japan (1.3–27.4 ng/g, mean 10.3 ng/g) (Zeng et al., 2017) and the Czech Republic (ND–347 ng/g, mean 27.2 ng/g) (Pribylova et al., 2006).

The MCCP levels in sediments in the present study were at the same level as those from Shenzhen, China (10.9–2500 ng/g, mean 900 ng/g) (Zeng et al., 2017) and the Czech Republic (ND–5575 ng/g, mean 345 ng/g) (Pribylova et al., 2006), but higher than those from the Yellow River, China (8.33–168 ng/g, median 25.8 ng/g) (Qiao et al., 2016), Hong Kong, China (<LOD–26.0 ng/g, mean 58.7 ng/g) (Zeng et al., 2017), Tokyo Bay, Japan (3.2–56.8 ng/g, mean 19.2 ng/g) (Zeng et al., 2017), and the North and Baltic Seas (5–377 ng/g, mean 75 ng/g) (Huttig and Oehme, 2005).

3.2. Composition profiles of SCCPs and MCCPs

Fig. 3 shows the average carbon and chlorine homologue group profiles of SCCPs and MCCPs in soils and sediments examined in this study. In general, C_{13} homologues were predominant in all SCCP samples, accounting for 39.4% and 47.4% in soils and sediments, respectively. The relative abundances of carbon chains in SCCPs in soils decreased in the order $C_{13} > C_{12} \approx C_{11} > C_{10}$. Similar distributions were observed in urban and suburban soils from Shanghai (Wang et al., 2014, 2017). The relative carbon chain distribution of SCCPs in sediments decreased in the order: $C_{13} > C_{12} > C_{11} > C_{10}$. The SCCP homologue patterns were similar to those found in sediments from Shenzhen by Zeng et al. (2017). However, the profiles were different from those found in the PRD in an earlier study by Chen et al. (2011), whereby C_{10}, C_{11}, C_{12}, and C_{13} homologues contributed in similar proportions. This difference might be ascribed to the abovementioned shift towards the production and usage of commercial CP formulations containing more long-chain homologues (Zeng et al., 2017). Compared to the SCCP patterns in soils/sediments in present study, short-chain SCCPs (C_{10-11}) were once observed in the air samples by Wang et al. this might be attributable to the different physiochemical properties of SCCP homologues, short-chain SCCPs are more easily emitted to gas phase (Wang et al., 2013b).

For MCCPs, similar carbon chain distributions were observed in soil and sediment samples from the studied region, both of which were dominated by C_{14} groups, followed by C_{15}, C_{16}, and C_{17} groups. The results were similar to those obtained for soils and sediments from the PRD (Chen et al., 2011; Wang et al., 2013b; Zeng et al., 2017).

Among the chlorine homologue group profiles of SCCPs, we found Cl_{6-7} (64.2%) to be dominant in soils. Similar profiles were found in the surrounding soil of a CP plant (Xu et al., 2016). However, the chlorine homologue profiles in this study differed from that observed in soils from Chongming Island, Shanghai, China, where the dominant homologue groups were Cl_{7-8} (Wang et al., 2013a). For the sediments, Cl_7 (39.1%) homologues were the major chlorine homologue groups, distinct from those from Lake Thun, where the dominant components were Cl_{9-10} (Iozza et al., 2008), and Liaohe River Basin, China, where the dominant components were Cl_{5-6} (Gao et al., 2012). In terms of chlorine homologue groups in MCCPs, Cl_{6-8} and Cl_{7-8} prevailed in soils and sediments, contributing 78.7% and 62.0% to the total MCCPs, respectively. The results were similar to those for soils and sediments obtained from the PRD, China (Wang et al., 2013b; Zeng et al., 2017).

Principal component analysis (PCA) was performed for SCCP and MCCP homologues in soils and sediments, respectively. As shown in Fig. S2, the first two main factors were extracted, which accounted for 81.8% of the total variation. From the scatter plot, it can be seen that SCCP homologues with longer carbon chains (C_{12-13}) and higher chlorine contents (Cl_{7-10}) tended to be present in the sediments, whereas homologues with shorter carbon chains and lower chlorine contents tended to be present in soils. Similar results were also observed for the MCCP homologues (Fig. S3); homologues with

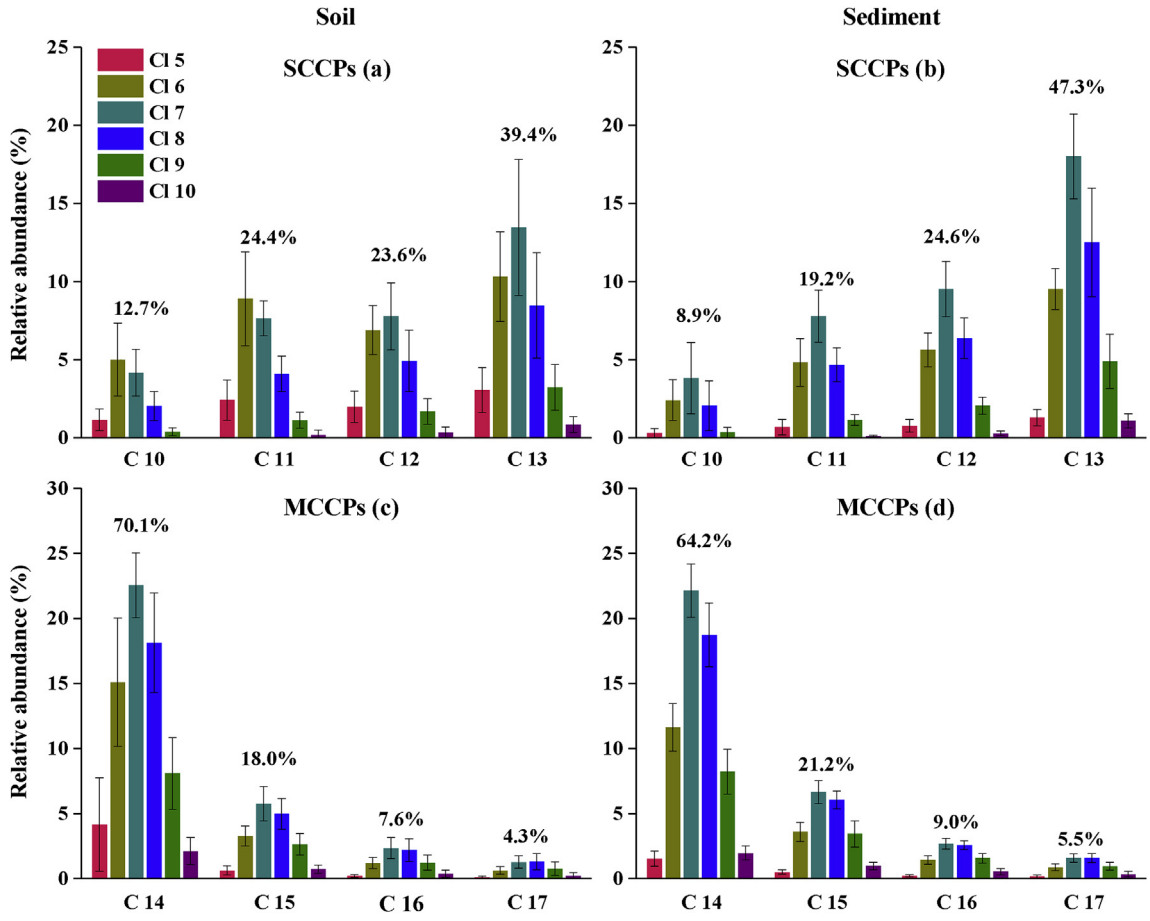

Fig. 3. Average composition profiles of SCCPs and MCCPs in soils (a and c) and sediments (b and d).

shorter carbon chains (C_{14}) and lower chlorine contents ($Cl_{5,6}$) tended to be present in soil, whereas those homologues with longer carbon chains (C_{15-17}) and higher chlorine contents (Cl_{7-10}) tended to be present in sediment. This difference might be ascribed to their physicochemical properties and resultant environmental behaviors. Considering that the sampling sites were close to the emission sources, the CPs probably entered the soil through direct input by dry/wet deposition, without experiencing complicated environmental geochemical processes in the terrestrial ecosystem (Wang et al., 2013b). Conversely, the main source of CPs in sediment was wastewater discharge (Zeng et al., 2017), and furthermore, during transportation along the river, CP homologues underwent a continuous partitioning process between water, particulates, and sediment. Since the log K_{ow} values of CP homologues increase with their degree of chlorination and carbon chain length, those CPs with high log K_{ow} values (high chlorination degree and/or long chain) are expected to be mostly adsorbed by sediments (Hilger et al., 2011; Pan et al., 2018).

3.3. Screening and semi-quantification of vSCCPs in soils and sediments

Recently, Xia et al. reported the concentrations of C_9-CPs in soil and biota from the Antarctic, suggesting that they can undergo long-range transport and are global pollutants (Xia et al., 2019). In our previous work, we also identified these C_9-CPs in soil, sediment, and indoor dust samples (Wu et al., 2019). Due to their typical characteristics of POPs (Xia et al., 2019), they have attracted the attention of researchers (Xia et al., 2019). Based on the method established in our previous work (Wu et al., 2019), we screened these compounds in soil and sediment samples after quantification of SCCPs and MCCPs. The identification of vSCCPs were based on the retention time (shorter or similar to SCCPs) and accurate mass (the most two abundant $[M\text{-}H]^-$ ions with 5 ppm mass tolerance), and we finally identified six vSCCP homologues (C_8Cl_{7-8} and C_9Cl_{5-8}) in soils and four vSCCPs (C_9Cl_{5-8}) in sediments, respectively (Table 2, Fig. S4, Fig. S5, Fig. S6). C_8Cl_{7-8} homologues were only found in two samples collected from the inside lawns of two coal-fired power plants, suggesting their emission might related to coal combustion during coal-fired power generation. During coal-mining and coal-crushing processes, the coal could be contaminated by the conveyor belt and lubricant, which might contain these vSCCPs (Brandsma et al., 2019; Gao et al., 2012). In our previous work, we identified several vSCCPs in commercial CP products, such as C_9Cl_{5-8} (Wu et al., 2019). Alternatively, these vSCCPs might be derived from the thermal degradation of higher CPs, given the high temperatures in the whole operation process of power plants (Xin et al.,

Table 2
Detection frequency (%) of CPs with carbon chains shorter than C10 in soils and sediments.

Samples	C_8Cl_7	C_8Cl_8	C_9Cl_5	C_9Cl_6	C_9Cl_7	C_9Cl_8
Soils	4.1	4.1	4.1	83.7	26.5	4.1
Sediments	–	–	17.6	47.1		

Mean not detected

2017). Among these chemicals, the detection frequency of C_9Cl_6 reached up to 83.7% in soils and 47.1% in sediments, followed by C_9Cl_7 with corresponding detection frequencies of 26.5% and 35.3%. The results imply that these C_9 compounds are ubiquitous CPs in the environment.

We also further semi-quantified these vSCCPs in soils and sediments assumed that they would have similar intensity to SCCP homologues on the UHPLC-Orbitrap Fusion TMS. The results revealed that the total concentration of vSCCPs was <2.85 ng/g (median 0.16 ng/g) in soils and <1.56 ng/g (mean 0.21 ng/g) in sediments. To date, only one study has reported C_9-CPs in soils from Antarctica and sediments from Yellow River, China, with mean concentrations of 14.0 ng/g and 20.3 ng/g, respectively (Xia et al., 2019). Considering their high detection frequencies and concentrations, further studies should be carried out to determine their occurrence, toxicity, and ultimate fate in the environment.

3.4. Correlation between CP concentrations and TOC contents

In general, CP homologues have relatively high log K_{oc} values (organic carbon sorption coefficients, K_{oc}), in the ranges 4.1–5.4 for SCCPs and 5.0–6.2 for MCCPs (Environment Canada, 2008). Hence, the SCCP and MCCP concentrations should theoretically associate with TOC contents in soils and sediments (Wang et al., 2017). In this study, the TOC contents varied from 0.07 to 6.42% in soils and from 0.18 to 3.91% in sediments, respectively. The results indicated no significant relationships between SCCP or MCCP concentrations and the corresponding TOC contents in soils (SCCPs, $R^2 = 0.26$, $p < 0.01$; MCCPs, $R^2 = 0.36$, $p < 0.01$; Fig. S7 a, b), suggesting that TOC contents have limited impact on the occurrence of CPs in soils from Dongguan City. This result was different to that for soil cores for sewage-irrigated farmland (Zeng et al., 2011). This difference might be explained as follows: the CPs in soils from Dongguan are mainly continuously influenced by the emission source through dry/wet deposition, whereas Zeng et al. focused on the vertical migration of CPs in soil cores, whereby the TOC present could play a critical role (Zeng et al., 2011).

Similarly, no obvious relationship could be established between SCCP/MCCP concentrations and TOC content (SCCPs, $R^2 = 0.15$, $p > 0.01$; MCCPs, $R^2 = 0.16$, $p > 0.01$; Fig. S7 c, d), implying that TOC content has limited influence on the adsorption of CP homologues on sediment from Dongguan City. A possible reason may be that CP concentrations in sediments are based on emission sources from industrial activities, and equilibrium partition of CPs between water, particulates, and sediment has yet to be established (Gao et al., 2012; Qiao et al., 2016). A similar phenomenon was also observed in sediments from the middle reaches of the Yellow River (Qiao et al., 2016) and from the Liaohe River Basin, China (Gao et al., 2012).

4. Conclusion

SCCPs and MCCPs in soils and sediments from Dongguan City have been measured. Total SCCP and MCCP concentrations were found in the ranges 6.75–993 ng/g and 23.9–2427 ng/g in soils, and 4.00–613 ng/g and 14.0–1581 ng/g in sediments, respectively. MCCPs predominated over SCCPs in most soil samples and in all sediment samples, implying that the former constitute the main components of Chinese commercial CP products. Homologues with more carbon and chlorine atoms tend to adsorb on sediment due to their physicochemical properties. Some vSCCPs have been identified and semi-quantified in this study, and our results suggest their ubiquity in soils and sediments. Much more attention should be paid to their occurrence and potential risks in further studies, because of their higher toxicity. No significant relationships have been found between CP concentrations and TOC contents in soils and sediments, which might be related to the continuous effect of local industrial activities.

Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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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.2020.114181.

Author statement

Yang Wu: Sample collection, sample pretreatment, instrumental analysis. Shutao Gao: Methodology, sample collection. Bingjing Ji: Sample pretreatment, instrumental analysis. Zhiyang Liu: Sample collection, Data analysis. Xiangying Zeng: Writing-reviewing. Zhiqiang Yu: Writing- Reviewing and Editing,

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