Occurrence, Distribution and Health Risk of Short-Chain Chlorinated Paraffins (SCCPs) in China: A Critical Review

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Abstract: With being listed in the Stockholm Convention, the ban on short-chain chlorinated paraffins (SCCPs) has been put on the agenda in China. Based on the literature over the past decade, this study comprehensively analyzed the occurrence, distribution of and human exposure to SCCPs in China, aiming to provide a reference for the changes in SCCPs after the ban. SCCPs were ubiquitous in environmental matrices, and the levels were considerably higher than those in other countries. SCCPs from the emission region were 2–4 orders of magnitude higher than those in the background area. Environmental processes may play an important role in the SCCP profiles in the environment, and C_{10} and Cl_{6} were identified as potential factors distinguishing their spatial distribution. River input was the dominant source in the sea areas, and atmospheric transport was the main source in the remote inland areas. Ingestion and dermal absorption and food intake may pose potential risk to residents, especially for children and infants. More studies are needed on their temporal trend, source emission and environmental degradation. The enactment of the restriction order will have a great impact on China’s CP industry; nevertheless, it will play a positive role in the remediation of SCCP pollution in the environment.

Keywords: POPs; occurrence; spatial distribution; SCCPs; human exposure; health risk

1. Introduction

Chlorinated paraffins (CPs) are industrial products derived from the chlorination of n-alkanes, with a carbon backbone length of 10–30 carbon atoms and a general degree of chlorination between 30% and 70%, also known as polychlorinated n-alkanes (PCAs) [1]. Since 1930s, CPs have been widely used as additives in lubricants and cutting fluids for metal working, flame retardants in the rubber industry, paints, sealing materials and other applications [2]. The cumulative global production of CPs reached 13.1 million metric tons (Mt) between 1935 and 2012, much above that of other anthropogenic chemicals [3]. During production, transportation and use, CPs are inevitably released into the environment, and they have been detected in various environmental matrices, such as air, water, sediments, soils, biota and humans [4]. Compared to medium-chain CPs (MCCPs, C_{14}–C_{17}) and long-chain CPs (LCCPs, C_{18}–C_{30}), short-chain CPs (SCCPs, C_{10}–C_{13}) have received more concerns due to their persistence, long-range transport potential, bioaccumulation and toxicity [5]. In 2017, SCCPs were listed in Annex A in the Stockholm Convention at the eighth meeting of the Conference of the Parties as a group of new POPs [6]. In May 2022,
The General Office of the State Council issued an action plan on controlling new pollutants, which explicitly included SCCPs in the control scope.

Developed countries, such as the United States, Canada, European countries and Japan, have recognized the impact of SCCPs on the environment and humans and have even stopped their production and use (except for some exempted industrial products) [7]. However, SCCPs are still produced and used in some developing countries, including China, India and Brazil [8]. China is the largest producer and consumer of CPs in the world. There are 100–140 CP producers in China, with annual production capacity reaching 1.6 Mt [9], accounting for 20–30% of the total global production [10]. In 2019, 236.4 kilotons (kt) of SCCPs were used in China and a total of 4.2 kt SCCPs were emitted into the environment [11]. However, the concern of SCCPs in China is relatively late, mainly due to the lack of attention to the environmental and health problems caused by new pollutants [12].

With the breakthrough in detection technology, Chinese scholars paid more and more attention to SCCPs [13]. Numerous studies have been conducted in air [14–16], dust [17–19], water [20–22], sewage sludge [22–24], soil [25–27], sediment [28–30], biota [31–33] and humans [34–36]. However, few studies have been reported for the comprehensive analysis of SCCPs in China [37], which is unfavorable for the understanding of the pollution status and potential risk of SCCPs. Especially in the past five years, great changes have taken place in international policies on the production and use of SCCPs, with the most significant event being the inclusion of SCCPs in the controlled list of POPs [6].

In the present study, we critically reviewed the literature published over the past decade, with emphasis on the occurrence, distribution and health risk of SCCPs, to outline the environmental status and potential risk of SCCPs in China, and provide a reference database for evaluating the changes in SCCPs after being banned.

2. Literature Survey and Analysis

The literature survey was performed by retrieving the Web of Science (WoS) database (http://apps.webofknowledge.com/; accessed on 1 July 2020) and the China Academic Journal Network Publishing Database (https://www.cnki.net/; accessed on 1 July 2020) until 30 June 2020. The keyword we searched was “SCCPs”, from which the literature about China was screened out. A total of 223 publications were initially obtained, among which 164 and 59 papers were found in the WoS database and the CAJD, respectively. We examined these publications individually and picked out the ones that matched the topic. Finally, 130 relevant publications were selected for analysis, including 113 papers obtained from the WoS database and 17 papers from the CAJD.

Figure 1 shows the number of publications on SCCPs in China per year and the percentages of English and Chinese publications during the period 2010–2020. The number of publications has been increasing continuously, with a significant increase since 2017, due to the fact that SCCPs were listed as a group of POPs in 2017 [6] and began to attract widespread attention. Most of the papers were published in English journals (87%), indicating a good international communication of SCCPs’ research in China. These articles were mainly concentrated in China’s coastal provinces, while relatively few studies were conducted on remote areas in the central and western regions (Figure S1A). In addition, human exposure (22%) and pollution sources (20%) were two major domains carried out (Figure S1B), implying more concerns on the health risk caused by SCCPs.
3. Levels and Distribution of SCCPs in Environmental Matrices

3.1. Air

Atmosphere is an important medium for long-range transport of SCCPs. SCCPs in gas phase (0.13–1350 ng/m³), particle phase (0.31–454 ng/m³) and sum of gas and particle phases (1.11–1442 ng/m³) throughout China (Table S1) were significantly higher than those from other countries and regions, such as Canada [38,39], the UK [40], Norway [41,42], Switzerland [43], Japan [14], South Korea [14], India [44], Pakistan [44] and Australia [45]. SCCPs in the atmosphere were mainly distributed in the gas phase (0.46–45.7 times the concentration of the particle phase) [20,46–51] and can be partitioned between the gas and particle phases. Gaseous SCCPs in summer were significantly higher than those in winter [15,50,52] and spring [20], whereas particle-bound SCCPs in summer were significantly lower than those in winter [53], indicating that the atmospheric SCCPs and their partitioning in two phases were probably affected by temperature. SCCPs in particle phase tend to volatilize into gas phase with the increase in temperature [54].

We summarized the atmospheric SCCPs in different regions of China, as shown in Table 1 and Figure S2A. The spatial distribution varied significantly between regions, mostly depending on SCCPs production and consumption, as well as their physicochemical properties and meteorological variables [37]. Overall, SCCPs in coastal areas were higher than those in central and western regions, which coincided with the regional difference in GDP per capita and population density, as well as the geographical distribution of CP manufacturing plants (Figure S3). SCCPs in northern China and central China were high, followed by eastern China, northeastern China and southern China. Meanwhile, SCCPs in southwestern China, northwestern China and China sea areas were relatively low, and the lowest was found in the Shergyla Mountain, which accordingly can be considered as a background area in China. SCCPs in the emission region (defined in Table 1) were three orders of magnitude higher than those in the background area, indicating that industrial activities had a critical impact on local atmospheric SCCPs.
Table 1. Concentrations of SCCPs in air, water, soil, sediment and biota samples collected from different regions of China.

<table>
<thead>
<tr>
<th>Region</th>
<th>Air (ng/m^3)</th>
<th>Water (ng/L)</th>
<th>Soil (ng/g dw)</th>
<th>Sediment (ng/g dw)</th>
<th>Biota (ng/g dw)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Gas Phase</td>
<td>Particle Phase</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Northeastern China</td>
<td>4.04–165</td>
<td>0.52–10.5</td>
<td>4.10–1490</td>
<td>56.9–189</td>
<td>ND–13,800</td>
</tr>
<tr>
<td>Northern China</td>
<td>0.40–1350</td>
<td>1.40–87.7</td>
<td>162–176</td>
<td>160–1450</td>
<td>690–9120</td>
</tr>
<tr>
<td>Eastern China</td>
<td>6.08–63.2</td>
<td>2.36–105</td>
<td>15.0–1978</td>
<td>ND–697</td>
<td>ND–2020</td>
</tr>
<tr>
<td>Central China</td>
<td>-</td>
<td>2.98–89.4</td>
<td>-</td>
<td>-</td>
<td>4.19–9760</td>
</tr>
<tr>
<td>Southern China</td>
<td>0.95–106</td>
<td>1.60–51.8</td>
<td>61.0–460</td>
<td>1.45–541</td>
<td>ND–6600</td>
</tr>
<tr>
<td>Northwestern China</td>
<td>-</td>
<td>4.36–27.5</td>
<td>-</td>
<td>ND–100</td>
<td>-</td>
</tr>
<tr>
<td>Southwestern China</td>
<td>1.10–14.4</td>
<td>3.82–39.9</td>
<td>-</td>
<td>0.22–948</td>
<td>ND–680</td>
</tr>
<tr>
<td>China sea areas</td>
<td>2.80–29.0</td>
<td>0.31–3.60</td>
<td>11.0–110</td>
<td>4.40–1757</td>
<td>9.30–9100</td>
</tr>
<tr>
<td>Background area</td>
<td>0.13–1.27</td>
<td>-</td>
<td>81.6 ± 31.1</td>
<td>ND–59.0</td>
<td>3.90–300</td>
</tr>
<tr>
<td>Emission region</td>
<td>81.7–988</td>
<td>7.40–454</td>
<td>27.0–4700</td>
<td>30.4–554,161</td>
<td>32.5–350,000</td>
</tr>
</tbody>
</table>

\(^a\) dry weight (dw); \(^b\) limited data reported on dry weight, and the concentration based on lipid weight were 620–250,000 ng/g

Indoor SCCPs had similar spatial distribution characteristics (Table S1). However, they were significantly higher than outdoor ones [16,48], which revealed possible emission sources in indoor environment, such as furniture, flooring, plastic products and clothing [55]. Indoor dust is deposited by PM in indoor air, resulting in the similar spatial distribution of indoor SCCPs (Table S2). Significant high SCCPs were found in e-waste recycling sites [56,57] and CPs production plants [49], which were much higher than those from Australia, Canada and Sweden [58].

3.2. Water

SCCPs have low water solubility, with logarithmic octanol–water partition coefficients (LogK_{ow}) ranging from 4.01 to 8.67 [59]. They are prone to being adsorbed on sediment when they are discharged into aquatic environment according to the Level III fugacity model [60,61]. SCCPs in freshwater, seawater and wastewater around China were 15.0–1640 ng/L, 4.10–1978 ng/L and 27.0–4700 ng/L, respectively (Table S3), which were considerably higher than those in North America [62], France [63] and Japan [64], but comparable to those in industrial areas from the UK [65]. Industrial wastewater was the main source of SCCPs in sewage treatment plants (STPs), and effluents from STPs had significant effects on the occurrence of SCCPs in the surrounding water [22].

The levels in the coastal waters of the Yellow and Bohai Seas [66] were largely higher than those in the South China Sea [32]. Meanwhile, lower SCCPs were detected in the central Bohai Sea [20] and the Liaodong Bay [67], which indicated that SCCPs occurred in the Bohai Sea potentially came from the river input in the Shandong Peninsula (especially the Yellow River). SCCPs in eastern China and northeastern China were higher than those in southern China and northern China (Table 1; Figure S2B). This differed from atmospheric SCCPs (Figure S2A), implying the possible difference in the sources of SCCPs in air and water. However, clear information on the spatial distribution of SCCPs in water is not available due to the limited data.

3.3. Soil

Soil is a reservoir for SCCPs via atmospheric deposition, sewage sludge application and wastewater irrigation. SCCPs in ambient soils, agricultural soils and industrial soils in China were ND–948 ng/g dw, 0.22–1609 ng/g dw and 30.4–554,161 ng/g dw, respectively (Table S4), which were significantly higher than those in the UK [68], Norway [68], Switzerland [69] and Antarctica [70]. Soil SCCP contamination can be caused by anthropogenic...
activities such as e-waste dismantling and industrial manufacture as high levels were detected in these regions. In addition, SCCPs in farmland soils were obviously higher than those in ambient soils, suggesting that wastewater irrigation and sewage sludge application may input additional SCCPs.

The spatial distribution of soil SCCPs was similar to that of atmospheric SCCPs, showing that the SCCPs in northern China were higher than those in eastern China and northeastern China, followed by southern China (Table 1; Figure S2C). This finding indicated that atmospheric deposition was the predominant source of SCCPs in soils. Interestingly, high SCCPs were observed in the soils from Yunnan [71], located in the southwest border of China, mainly due to human activity and atmospheric deposition. Comparable SCCPs were found on the Tibetan Plateau [72,73], which is a remote high altitude mountain region located in the southwest of China with a relatively sparse human population and minimal industrial activities and can be acted as a background area. These results demonstrated that atmospheric transport may play an important role in the occurrence of SCCPs in soils in southwestern China. Remote high altitude mountain areas can trap SCCPs through distillation and cold condensation based on the weather conditions of low temperature and high precipitation, known as “mountain cold-trapping” [74].

3.4. Sediment

Sediment can be a reservoir of SCCPs because SCCPs tend to deposit into sediment in aquatic environment. SCCPs in freshwater sediments, marine sediments and contaminated sediments throughout China were ND–9760 ng/g dw, ND–13,800 ng/g dw and 32.5–350,000 ng/g dw, respectively (Table S5), significantly higher than those in North America [75,76], the UK [77], Czech Republic [78], Switzerland [79], Japan [64,80] and the North and Baltic Seas [81], while they were comparable to those in industrial areas from the UK [65] and Spain [82,83]. High SCCPs were observed in freshwater sediments from e-waste dismantling areas. Likewise, discharge of wastewater from STPs resulted in SCCP contamination in the receiving sediments [22]. SCCPs in freshwater sediments were generally higher than those in marine sediments (except sediments from Dalian Bay), which was consistent with the results from other regions [64,80].

The spatial distribution of sedimentary SCCPs was roughly consistent with those in air and soil, which ranked as northern China > southern China > northeastern China > central China > eastern China > southwestern China > northwestern China (Table 1; Figure S2D). In coastal waters, the highest SCCPs were found in estuaries areas [29,32,80,84], and they were characterized by declining and dispersing trend towards offshore and decreasing with depth [85,86], testifying that river input was the major source of SCCPs in marine sediments. Additionally, sedimentary SCCPs in China sea areas were higher than those in remote areas of southwest and northwest. The lowest concentration was detected in Qinghai Lake, located on the Tibetan Plateau [30], which can be used as a background area.

3.5. Biota

Given the ubiquitous of SCCPs in environmental matrices and their persistence and lipophilicity, SCCPs will accumulate in organisms, resulting in possible risk to ecology and human health. SCCPs in biota were ND–30,000 ng/g dw, with 9.30–20,320 ng/g dw, 3.90–30,000 ng/g dw and ND–13,600 ng/g dw in marine organisms, freshwater organisms and terrestrial organisms, respectively (Table S6). Further, SCCPs in plants, zooplankton, invertebrates, fish, reptiles, birds and mammals were ND–13,600 ng/g dw, 3600–5600 ng/g dw, 11.1–25,000 ng/g dw, 3.90–30,000 ng/g dw, 110–1400 ng/g dw, <28.0–2300 ng/g dw and 280–9100 ng/g dw, respectively. SCCPs in freshwater organisms were higher than those in marine organisms, which was consistent with the distribution of sedimentary SCCPs. Moreover, SCCPs in aquatic organisms were generally higher than those in terrestrial organisms. Habitat and feeding habits may be key factors driving SCCPs accumulation in organisms. Sediment feeding and filter feeding led to the accumulation of SCCPs by invertebrates from sediments. Meanwhile, bioaccumulation resulted in slightly higher
SCCPs in fish than invertebrates [67,87,88]. In terrestrial organisms, plants contained high levels, mainly because they can absorb SCCPs not only through roots, but also by leaves [73,89,90]. However, there was a significant negative correlation between SCCPs and trophic levels in some aquatic and terrestrial food webs, suggesting that SCCPs may undergo trophic dilution [72,91].

For spatial distribution, SCCPs in biota were consistent with those in environmental medium, illustrating that organisms were significantly affected by SCCP exposure in the environment. The highest levels were found in eastern China, followed by northeastern China, northern China and southern China (Table 1). Industrial activities such as e-waste recycling had a significant impact on the SCCP exposure of organisms in the surrounding environment [92–95]. It is worth noting that the highest SCCPs were detected in fish from the Dianshan Lake, a major drinking water source in Shanghai [96], suggesting that the water source may be potentially polluted by SCCPs. Comparable levels were occurred in organisms from paddy fields in the YRD [31], showing possible contamination by irrigation water. Similar regional distribution of SCCPs was found in fish from the China sea areas [97]. However, SCCPs in organisms from the estuary areas were higher than those from other sea areas [32,98,99], which indicated that their contents were significantly impacted by regional sources. The lowest SCCPs in organisms were found in the Tibetan Plateau (Table 1), which can be considered as a background area of China, and they were two orders of magnitude lower than those in the emission region. Nevertheless, these levels were obviously higher than those from the Arctic [100] and Antarctic [70], implying that the Tibetan Plateau, known as the third pole of the world, was more strongly influenced by anthropic activity. However, there is no information on biological SCCPs from central China, northwestern China and southwestern China.

3.6. Regional Characteristics

SCCPs in environmental matrices from different regions were normalized using min-max normalization method to analyze the regional characteristics of SCCPs, and identify the high level regions. The formula for this method is as follows:

\[
x^* = \frac{x - \text{min}}{\text{max} - \text{min}}
\]

where \(x^*\) represents the normalized value, \(x\) represents the original value, \(\text{min}\) represents the minimum value, and \(\text{max}\) represents the maximum value.

In order to make the data conducive to analysis, logarithmic transformation was performed on the original data before calculation. Although there were regional differences in SCCP concentrations in environmental matrices, the percentiles of SCCPs were high in northeastern, northern and eastern China (43.5–88.6%), and middle in central, southern, northwestern and southwestern China (16.3–68.7%), but low in China sea areas and background area (0–62.3%; Figure 2). Thus, more attention should be focused on the high level regions such as northeastern, northern and eastern China.
4. SCCP Homologue Profiles

4.1. SCCP Profiles

Characterizing the compositions of SCCPs in the environment is crucial to understand their sources and fates. Table S7 shows the profiles of SCCPs in the atmosphere. The SCCP homologues were dominated by C\textsubscript{10} (55.2\%) and Cl\textsubscript{6} (45.7\%) in the gas phase and C\textsubscript{11} (33.5\%) and Cl\textsubscript{7} (30.6\%) in the particle phase (Figures 3A and 4A). Gas–particle partitioning of SCCPs significantly affects their environmental behaviors (e.g., atmospheric transport and deposition). Shorter carbon chain and lower chlorinated SCCPs are more likely to partition in gas phase, leading to their persistent existence and long-distance atmospheric transport [54]. As high as 66.1\% of C\textsubscript{10} SCCPs was detected in the background area (Figure S4A). However, no similar distribution was observed for chlorine homologues (Figure S5A).

The C\textsubscript{10} and C\textsubscript{11} homologues were the predominant carbon chain groups in water, accounting for 30.5\% each (Table S8). Specially, the carbon chain was dominated by C\textsubscript{10} (34.0\%) in seawater and C\textsubscript{11} (27.2\%) in freshwater (Figure 3B). Meanwhile, Cl\textsubscript{7} was the most abundant congener in seawater and freshwater (Figure 4B). The carbon profiles in freshwater (lake water and river water) were similar to those in effluent from STPs (Figure S4B), indicating that freshwater inland may be directly affected by local source discharge [22]. Shorter carbon chain SCCPs are more inclined to migrate with water, resulting in the increase in C\textsubscript{10}\% in seawater. Similar but less significant phenomena were observed for the chlorine homologues (Figure S5B).
Figure 3. Carbon profiles of SCCPs in different environmental matrices in China. (A) Carbon profiles of SCCPs in air. (B) Carbon profiles of SCCPs in water. (C) Carbon profiles of SCCPs in soil. (D) Carbon profiles of SCCPs in sediment. (E) Carbon profiles of SCCPs in biota.
Figure 4. Chlorine profiles of SCCPs in different environmental matrices in China. (A) Chlorine profiles of SCCPs in air. (B) Chlorine profiles of SCCPs in water. (C) Chlorine profiles of SCCPs in soil. (D) Chlorine profiles of SCCPs in sediment. (E) Chlorine profiles of SCCPs in biota.

SCCP profiles in different soils were listed in Table S9. Similar carbon profiles were found in farmland and ambient soils, and C_{10} congeners were dominant in both types of soils, contributing to 33.9% and 32.5%, respectively (Figure 3C). Cl_{6} congeners (26.5%) were the most abundant groups in farmland soil and Cl_{7} (27.6%) was the largest group of chlorine homologues in ambient soil (Figure 4C). This difference was mainly due to the sources of SCCPs in soils. Consistent with SCCP congeners in the atmosphere, we found that C_{10} (53.1%) accounted for the highest proportion in the background area (Figure S4C), indicating that atmospheric transport and deposition was the main source of SCCPs in the background area. However, in contrast to the carbon profiles, the chlorine homologues...
were dominated by Cl₈ (34.8%) and Cl₇ (31.2%), which was different from those in the other regions (Figure S5C). This may be influenced by atmospheric deposition of particulate SCCPs as a major contributor. Additionally, in high altitude areas, higher chlorinated SCCPs are more likely to enter the soil through particle-bound deposition [73].

The carbon homologues were dominated by C₁₀ (32.9%) in sediment (Table S10), while the proportion of C₁₀ in marine sediment (36.8%) was higher than that in freshwater sediment (29.5%) (Figure 3D). Cl₆ (30.8%) was the most abundant congener group in marine sediment and Cl₇ (24.0%) and Cl₈ (22.1%) were the predominant groups in freshwater sediment (Figure 4D). The abundance of shorter carbon chain and lower chlorinated SCCPs in marine sediment was significantly higher than that in freshwater sediment, implying that they are more soluble and can be transported more easily. In addition, the SCCP profiles of sediment and water in marine environment were more similar, mainly due to the frequent exchange of SCCPs between seawater and marine sediment caused by tides. Nevertheless, the highest abundance of C₁₀ was found in sediments from the Yangtze River and the Yellow River (Figure S4D), possibly because they received large amounts of C₁₀ homologues migrating from polluted tributaries. Similar SCCP profiles were found in northern, eastern and southern China, as well as in China’s coastal waters (Figures S4D and S5D), reflecting the consistency of SCCP sources in these regions [24].

C₁₀ was the dominant group of carbon homologues in biota, with abundance of 35.8%, 40.7% and 37.3% in marine organisms, freshwater organisms and terrestrial organisms, respectively (Table S11; Figure 3E). Meanwhile, Cl₆ and Cl₇ were the predominant chlorine homologues in biota, accounting for 50.3%, 61.5% and 59.1% in marine organisms, freshwater organisms and terrestrial organisms, respectively (Figure 4E). No regional characteristics were occurred in the homogenous composition of SCCPs in organisms (Figures S4E and S5E), mainly due to their selective absorption and metabolism of SCCPs. Marine bivalves had high potential for bioaccumulation, and higher chlorinated SCCPs were more prone to being accumulated [29,67]. Likewise, the logarithmic bioaccumulation factors (Log BAFs) of SCCPs in marine fish had significant positive correlations with carbon chain length, chlorine contents and Kₐw [87]. However, the accumulation of SCCPs was significantly negatively correlated with carbon chain length, chlorine contents and Kₐw in freshwater aquatic organisms [101] and alpine fish [102]. Alpine vegetation had a high proportion of C₁₀, corresponding to SCCP profiles in the environmental matrices, but the percentage of C₁₀ in the higher-trophic organisms was reduced due to the dilution of SCCPs [72,73]. Nevertheless, the content of C₁₀ congeners in fish from the Tibetan Plateau had a significant positive correlation with altitude [102].

4.2. Principal Component Analysis (PCA) by Carbon and Chlorine Profiles

To clarify the regional differences in SCCP homologues, PCA was applied to identify the spatial distribution of SCCPs based on carbon and chlorine profiles, and to obtain the major factors for their differentiation. SCCPs in gas and particle phases can be obviously distinguished based on carbon and chlorine profiles (Figure 5A). However, it was difficult to implement the spatial distribution of atmospheric SCCPs because this information was masked by the difference in SCCP profiles between gas and particle phases (Figure S6A). The highest contributions to PC1 (66.6%) were C₁₃, C₁₀, C₁₂ and Cl₆, and the most contributing descriptors to PC2 (13.5%) were Cl₅ and Cl₇. These SCCP homologues may be the dominant factors for identifying the gas–particle distribution of SCCPs in the atmosphere.
SCCPs between seawater and freshwater can be effectively distinguished by carbon and chlorine profiles, except for effluent from STPs, which represents the fingerprint profiles of industrial wastewater (Figure 5B). Additionally, the spatial distribution of SCCPs in water can be effectively distinguished, although the data was limited (Figure S6B). PC1 explained 53.0% of variance, and C_{10}, Cl_{6}, Cl_{8}, Cl_{9} and C_{12} were the most contributors, while PC2 yielded 20.6% of explainable results, with C_{11} and Cl_{5} loading heavily.

Figure 5. PCA of SCCP congeners in different environmental matrices in China. (A) PCA of SCCP congeners in air (G: gas phase; P: particle phase). (B) PCA of SCCP congeners in water (SW: seawater; FW: freshwater; STP: wastewater from sewage treatment plant). (C) PCA of SCCP congeners in soil (FS: farmland soil; AS: ambient soil). (D) PCA of SCCP congeners in sediment (FWS: freshwater sediment; MS: marine sediment). (E) PCA of SCCP congeners in biota (MO: marine organism; FO: freshwater organism; TO: terrestrial organism).
addition, SCCPs in soil and sediment can be spatially identified (Figure S6C,D). Meanwhile, SCCPs between farmland soil and ambient soil, as well as between marine sediment and freshwater sediment can be effectively identified using carbon and chlorine profiles (Figure 5C,D). With regard to soil, Cl$_{9}$, Cl$_{8}$, Cl$_{6}$ and Cl$_{5}$ had the highest contributions to PC1 (45.4%), and C$_{10}$, C$_{12}$ and C$_{13}$ contributed the most to PC2 (20.4%). For sediment, C$_{12}$, C$_{10}$, Cl$_{6}$ and Cl$_{8}$ were the most contributing descriptors to PC1 (59.5%), while C$_{11}$ and Cl$_{5}$ were loading highest in PC2 (16.3%).

However, the SCCPs of organisms could not be effectively distinguished between different regions (Figure S6E). In addition, biological SCCPs classified according to the species of marine organisms, freshwater organisms and terrestrial organisms also cannot be obviously differentiated (Figure 5E). This finding indicated that SCCP compositions in organisms are more complex than those in environmental matrices.

4.3. Environmental Implications of SCCP Profiles

The composition of SCCP congeners in the environment is influenced by emission sources and environmental processes. However, the extent of their influence is still controversial. For example, by simulating the environmental fate of SCCPs, Chen et al. [103] concluded that the subtle difference in physicochemical properties between SCCP homologues caused only minor differences in the fate and transport of SCCPs. However, it is found that there was obvious regional difference in the composition of SCCPs in soils and sediments from an e-waste dismantling area, indicating that environmental processes, such as transportation via atmosphere and/or water, and deposition, resulted in different SCCP profiles in environmental matrices [104]. We confirmed the impact of environmental processes on the SCCP profiles in the environment, as they were significantly different from those in the commercial mixtures (C$_{13}$-congener group was predominant in CP-42, CP-52 and CP-70) [105] and domestic polymeric products (C$_{11}$ and C$_{13}$ were the predominant carbon homologue groups) [104] in China. Moreover, we concluded that C$_{10}$ and Cl$_{6}$ may be potential factors to identify their spatial distribution. Nevertheless, it is necessary to further explore the influence of source emissions on SCCP profiles and the mechanism of migration and transformation on the spatial distribution of SCCPs.

5. Daily Intake and Human Health Risk of SCCPs

5.1. Levels and Profiles of SCCPs in Food and Human Tissue

Dietary exposure is one of the main external exposure routes of SCCPs, and the SCCPs in human tissue show the internal exposure. We summarized the concentrations and compositions of SCCPs in food throughout China. The SCCPs in aquatic foods, meat, cereals, legumes, vegetables, eggs, milk, culinary oil and fruits were 37,100–487,000 ng/g lipid weight (lw), 25,300–88,300 ng/g lw, 37,200–88,600 ng/g lw, 41.7–801 ng/g wet weight (ww), 132,000–605,000 ng/g lw, ND–150,000 ng/g lw, 130–5770 ng/g lw, 892–1320 ng/g lw and 6.34–168 ng/g ww, respectively (Table S12). E-waste recycling activities significantly affected the SCCPs in local food [56,95,106–108]. Higher SCCPs were found in aquatic foods than those in other food categories [109,110], and the levels in shrimp and shellfish were generally higher than those in fish [111]. Moreover, SCCPs in farmed fish were significantly lower than those in river fish, but obviously higher than those in sea fish [112,113]. SCCPs in cooked food were significantly lower than those in raw food [114], revealing that Chinese traditional cooking can effectively eliminate SCCPs in diet. SCCP profiles in food had no regional features due to the complexity of origin (Table S13).

SCCPs in human tissues including placenta, breast milk, blood, plasma and serum have been detected, with levels at 98.5–3771 ng/g lw, ND–16,120 ng/g lw, 370–35,000 ng/g lw, ND–203 ng/g ww and 1670–57,800 ng/g lw, respectively (Table S14). The internal exposure of fetus to SCCPs can be elucidated by analyzing SCCPs in maternal serum, cord serum and placenta during gestation. Significantly lower SCCPs in cord serum (roughly one-third) were detected compared to maternal serum [34,115,116], indicating that SCCPs were effectively retained by the placental barrier. In addition, lower chlorinated and shorter
carbon chain SCCPs can be transported more efficiently across placenta. The determination of SCCPs in breast milk can show infants’ exposure to SCCPs during lactation. SCCPs in breast milk from China were significantly higher than those from Korea, Japan, Sweden and Norway [117,118]. Moreover, SCCPs in breast milk from urban areas [119] were significantly higher than those from rural areas [36]. Exposure amount of SCCPs during the lactation period was > 100 times higher than during the gestation period [116], indicating that more attention should be paid to SCCPs’ exposure of infants. There was no significant regional difference in SCCP profiles in human tissues (Table S15), as diets are considered to be the main sources of SCCPs.

5.2. Daily Intakes of SCCPs via Inhalation, Ingestion and Dermal, and Dietary Exposure

Humans can be exposed to SCCPs via their diets, dermis and by breathing. The estimated daily intake (EDI) of SCCPs (mean and 95th percentile) through inhalation, ingestion and dermal, and dietary exposure was calculated to evaluate the levels of human exposure to SCCPs (Table S16). The EDIs of SCCPs via inhalation exposure for children were higher than those for adults [16,51,53,120,121]. Meanwhile, children had higher exposure levels to SCCPs via indoor air [16]. Industrial employees from a CP production plant had the highest EDI [49], showing that occupation had a significant influence on SCCP exposure. Nevertheless, these EDIs were much lower than the tolerable daily intake (100 µg/kg body weight (bw)/day) suggested by the International Program on Chemical Safety [121].

Ingestion and dermal absorption are suggested to be two dominant routes of human exposure to SCCPs in indoor dust. The exposure to SCCPs in indoor dust was significantly higher than that via inhalation, and children (including infants and toddlers) had higher EDIs than those for teenagers and adults [18,19,120]. In addition, the exposure doses of SCCPs via ingestion were 2.80–10.2 times higher than those through dermal absorption, especially for children [18]. Workers and residents in e-waste dismantling area were confronted with high exposure levels, implying e-waste recycling activities may pose potential health risk to local residents [56].

Humans are exposed to more SCCPs through their diets than air and indoor dust. The EDIs of SCCPs from meat, aquatic foods, cereals, legumes, milk, vegetables and fruits were 130–155 ng/kg bw/day, 32.0–872 ng/kg bw/day, 1390–5189 ng/kg bw/day, 529 ng/kg bw/day, 29.7 ng/kg bw/day, 97.0 ng/kg bw/day and 36.9 ng/kg bw/day, respectively (Table S16). Aquatic products may be the largest contributor to dietary exposure except for cereals, and consumption of farmed freshwater fish was confronted with greater exposure to SCCPs compared with wild sea fish [112]. Infants suffered greater levels of SCCPs via breast milk than adults and children [116–119,122], which were higher than those from Korea, Japan, Sweden and Norway [117,118]. In addition, the body burden of postnatal exposure during the lactation period was about 124 times higher than that of prenatal exposure during the gestation period [116]. The intake of SCCPs by infants was negatively correlated with months of age, and the EDIs of SCCPs for neonates were higher than the recommended value (11.0 µg/kg bw/day) by the World Health Organization (WHO) [116,122].

Using the data from Beijing [114,120], we examined the contribution of SCCP intake via inhalation, ingestion and dermal, and dietary exposure for adults and children, as well as the proportion of various foods to dietary exposure (Figure 6). Dietary exposure was dominant in SCCP intake for adults and children, accounting for 84.7% and 57.6%, respectively. However, the proportion of children exposed to SCCPs by ingestion and dermal absorption (41.0%) was significantly higher than that of adults (13.7%), which was mainly caused by children’s living habits. Inhalation exposure contributed the least to SCCP intake in adults (1.58%) and children (1.40%). In addition, meat (51.9%) was the predominant source of dietary exposure, followed by fish (12.6%), seafood (11.8%), fruit (10.0%) and vegetable (8.06%), while the contribution of grain and cooking oil was relatively low (5.54%). This differed from the results of EDIs for food categories, probably due to the inconsistency between the recommended intake and the actual dietary intake. The intake
of SCCPs in animal food is higher than that in plant food, mainly because of the high fat content and high trophic level of animals, which leads to a greater accumulation of SCCPs than plants [110]. Moreover, Chinese people consume more meat than aquatic products, resulting in higher SCCPs exposure via meat than aquatic products [123]. Of course, the dietary structure of residents varies between regions; therefore, regional difference in dietary exposure needs to be further clarified.

![Figure 6. Contribution of SCCPs intake via three main exposure routes. (A) Estimated daily intake (EDI) of SCCPs for adults. (B) EDI of SCCPs for children. (C) The relative contribution of various foods to dietary exposure.](image)

### 5.3. Human Health Risk of SCCPs

The margin of exposure (MOE) developed by the European Food Safety Authority (EFSA) was used to assess the health risk of SCCPs via inhalation, ingestion and dermal, and dietary exposure, which is defined as the ratio of benchmark dose lower confidence limit (BMDL) to EDI [124]. The BMDL corresponding to an additional risk of 10% (BMDL$_{10}$) is 2.3 mg/kg bw/day, and MOE $>$ 1000 (MOE $^{-1}$ $<$ 0.001) represents low risk for human health [124]. In this study, the EDI used in the formula was the 95th percentile or maximum value. Although risk assessment implied low risk to residents via inhalation exposure, ingestion and dermal absorption and food intake may pose a high risk, especially for children and infants (Figure 7). This differed from many studies [16,36,53,110,114,120,125,126] that used the no–observed–adverse–effect level (NOAEL) for risk assessment, a reference point of which was 100 mg/kg bw/day for SCCPs (a NOAEL of 10 mg/kg bw/day was recommended in the latest report) [124]. BMDL$_{10}$ is considered to be a more appropriate method for health risk assessment [127], and the European Food Safety Authority (EFSA) Panel on Contaminants in the Food Chain (CONTAM) recommends the use of BMDL$_{10}$ to evaluate the health risk of SCCPs to human body [124]. Therefore, our study provided a new insight into the risk of SCCP exposure confronted by the Chinese population and revealed the severity of SCCP contamination in China. In addition, this study and previous studies found that occupational workers and local residents had a high risk of exposure to SCCPs due to industrial activities [49,56]. Overall, there is a need to strengthen the risk assessment and prevention and control of SCCPs, especially for special groups, such as occupational workers, children and infants.
few studies have analyzed the levels and compositions of SCCPs on a time scale, which is not conducive to a comprehensive understanding of the changes in SCCPs in the environment. Moreover, there was limited information on the source emissions and environmental behaviors of SCCPs, such as their industrial sources and degradation processes, which are unfavorable to the pollution control of SCCPs. To this end, it is necessary to strengthen the research on the aspects mentioned above.

With the exemption period coming to an end, China is phasing out the use of SCCPs. CP products in China are mainly classified according to chlorine content, which makes it difficult to remove SCCPs in mixed products. CP-52 is the main type of CP products produced in China, accounting for 80–90% of the total production [128], with SCCP content of 24.9% [129]. However, CP products with SCCP content of less than 0.6% are considered acceptable. This requires enterprises to improve the production process, or choose medium-
and long-chain paraffins as raw materials, which increases the production cost. According to statistics, the annual production of CPs in China is 1.2 Mt, with an output value of CNY 6 billion \[9,130\]. Therefore, the enactment of the restriction order will have a great impact on China's CP industry. Nevertheless, it will also play a positive role in the remediation of SCCP pollution in the environment.

Recent studies showed that MCCPs have similar toxicity and fate to SCCPs, and it is expected that MCCPs will also be added to the Stockholm Convention on POPs in the future \[131,132\]. First, China should strengthen the research on the source, occurrence and risk of MCCPs in the environment and make efforts for MCCPs to be included in the list of POPs. On the other hand, China should develop new alternatives to prepare for the ban of MCCPs in the future.

**Supplementary Materials:** The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/separations9080208/s1, Figure S1: Regional distribution (A) and domain proportion (B) of SCCP research in China over the past decade; Figure S2: Concentrations of SCCPs (mean value) in environmental matrices in China; Figure S3: GDP per capita (A), population density (B) and distribution of CP manufacturing plants (C) in China; Figure S4: Carbon composition profiles of SCCP congeners in different environmental matrices in China. (A) The congener composition of SCCPs in air. (B) The congener composition of SCCPs in water. (C) The congener composition of SCCPs in soil. (D) The congener composition of SCCPs in sediment. (E) The congener composition of SCCPs in biota; Figure S5: Chlorine composition profiles of SCCP congeners in different environmental matrices in China. (A) The chlorine abundance of SCCP congeners in air. (B) The chlorine abundance of SCCP congeners in water. (C) The chlorine abundance of SCCP congeners in soil. (D) The chlorine abundance of SCCP congeners in sediment. (E) The chlorine abundance of SCCP congeners in biota; Figure S6: PCA of SCCP congeners in different regions of China. (A) PCA of SCCP congeners in air. (B) PCA of SCCP congeners in water. (C) PCA of SCCP congeners in soil. (D) PCA of SCCP congeners in sediment. (E) PCA of SCCP congeners in biota. NE: Northeastern China; N: Northern China; E: Eastern China; C: Central China; S: Southern China; NW: Northwestern China; SW: Southwestern China; CS: China sea areas; BA: Background area; Table S1: Concentrations of SCCPs in air samples in China; Table S2: Concentrations of SCCPs in dust samples in China; Table S3: Concentrations of SCCPs in water samples in China; Table S4: Concentrations of SCCPs in soil samples in China; Table S5: Concentrations of SCCPs in sediment samples in China; Table S6: Concentrations of SCCPs in biota samples in China; Table S7: Comparison of the concentrations (ng/m$^3$) and compositional profiles (%) of SCCPs in air; Table S8: Comparison of the concentrations (ng/L) and compositional profiles (%) of SCCPs in water; Table S9: Comparison of the concentrations (ng/g dw) and compositional profiles (%) of SCCPs in soil; Table S10: Comparison of the concentrations (ng/g dw) and compositional profiles (%) of SCCPs in sediment; Table S11: Comparison of the concentrations (ng/g dw) and compositional profiles (%) of SCCPs in biota; Table S12: Concentrations of SCCPs in food; Table S13: Comparison of the concentrations (ng/g dw) and compositional profiles (%) of SCCPs in food; Table S14: Concentrations of SCCPs in human tissue; Table S15: Comparison of the concentrations (ng/g lw) and compositional profiles (%) of SCCPs in human tissue; Table S16: Estimated daily intake (EDI) of SCCPs via various exposure routes. References \[133–169\] can be found in supplementary materials.

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