平成27年11月発行　Chemosphere　第138巻 104頁～111頁掲載

Short-chain chlorinated paraffins in cooking oil and related products from China

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ABSTRACT

Short-chain chlorinated paraffins (SCCPs) are emerging persistent organic pollutants. It has been found that dietary intakes of SCCPs in China have recently increased and are now higher than in Japan and Korea. The contribution of cooking oil to dietary exposure to SCCPs in China was evaluated by analyzing SCCPs in cooking oil, raw seeds used to produce cooking oil, and fried confectionery products collected in China in 2010 and 2012. Detectable amounts of SCCP homologs were found in 48 out of the 49 cooking oil samples analyzed, and the SCCP concentrations varied widely, from <9 to 7500 ng g^{-1}. Estimated dietary intakes of total SCCPs in cooking oil ranged from <0.78 to 38 μg d^{-1}. The estimated dietary intake of SCCPs was relatively high (mean 14.8 μg d^{-1}) for residents of Beijing. Fried confectionery was found to contain SCCP concentrations of 11–1000 ng g^{-1}. Cooking oil might therefore be one of the sources of SCCPs to Chinese diets. SCCPs were also detected in raw seeds used to produce cooking oil, but the concentrations varied widely. The SCCP homolog patterns in the raw seed and cooking oil samples were different, implying that the seeds used to produce the oil (and therefore the soil on which the seeds were produced) were unlikely to be the sources of SCCPs in cooking oil. Further investigations are needed to determine the routes through which cooking oil becomes contaminated with SCCPs during the production and processing of the oil.

Word count: 5842 words

Keywords: short-chain chlorinated paraffins; exposure; food; homolog analysis; China.

1. Introduction

Chlorinated paraffins (CPs) are used in a wide range of industrial applications including in plasticizers, flame retardants, cutting fluids, and lubricants (European Chemicals Bureau, 2008). The CPs are typically categorized into three groups according to the lengths of the carbon chains, and these groups are short-chain CPs (SCCPs, with $C_{10–13}$ components), medium-chain CPs ($C_{14–17}$), and long-chain CPs ($C_{18–30.}$). SCCPs appear to be persistent in the environment and have the potential to strongly accumulate in biota (Persistent Organic Pollutants Review Committee, 2010; Fisk et al., 1996, 1998; Houde et al., 2008; Iozza et al., 2008). The International Agency for Research on Cancer has classed SCCPs as group 2B compounds (International Agency for Research on Cancer, 1990), and SCCPs are also classified as Carc. 2 in Annex VI of Regulation (EC) NO. 1272/2008 in European Union (EU), possibly carcinogenic to humans (EC, 2008). Recently, Geng et al. (2015) found that SCCPs can stimulate β-oxidation, and SCCPs are therefore considered to be peroxisome proliferators. SCCPs are currently under review as candidates for inclusion in the Stockholm Convention list of persistent organic pollutants (Persistent Organic Pollutants Review Committee, 2010).

CPs have been produced around the world. Notably, the total amount of CPs produced in China has continually increased in recent years, 1,000,000 t being produced in China in 2009 (Chen et al., 2011; Tong et al., 2009). Three of the commercial CP formulations are called CP-42, CP-52, and CP-70, and they have chlorine contents of 42%, 52%, and 70%, respectively. More than 80% of the total amount of CPs produced in China in 2008 was of CP-42 and CP-52, which are used as plasticizers in poly(vinyl chloride) (China Chemical Reporter, 2004; 2009). CP-42 and CP-52 contain SCCPs but also contain CPs with longer carbon chain lengths that are classified as medium- and long-chain chlorinated paraffins (Yuan et al., 2010).

The release of SCCPs can occur during the production, storage, transportation, use, disposal, and recycling of CP-containing products (Persistent Organic Pollutants Review Committee, 2010). Most SCCP emissions probably occur during the formulation and manufacture of products containing SCCPs (de Boer et al. 2010; de Boer and El-Sayed Ali, 2010). The release of SCCPs into sewers has been found to result in SCCPs accumulating in sewage sludge (Zeng et al., 2012) and the aquatic environment (Gao et al., 2012; Tomy et al., 1997). It has also been suggested that SCCPs enter the environment through evaporation and are transported in the vapor phase, because SCCPs have been found in air samples collected at remote sites (Li et al., 2012), soil in remote non-industrial areas such as Chongming Island, China (Wang et al., 2013), and even in marine mammals in the Arctic (Tomy et al.,2000). SCCPs have been detected in bivalves (Yuan et al., 2012), fish (Ma et al., 2014; Reth et al., 2005), birds (Reth et al., 2006), and human milk (Thomas et al., 2006), so it has been concluded that biota can be exposed to SCCPs through the food chain and that SCCPs can bioaccumulate.

In a previous investigation (Harada et al., 2011) we found that the dietary exposure of residents of Beijing to SCCPs (geometric mean 620 ng (kg body weight)$^{-1}$ d^{-1} in 2009) increased by two orders of magnitude between 1993 and 2009. This finding raised questions about the food items that contributed most to SCCP intakes in Beijing. The consumption of fish is considered to be a major source of lipophilic pollutants to humans (Feo et al., 2009; Ma et al., 2014; Yuan et al., 2012; Zeng et al., 2011). Oils and fats could also be important dietary sources of SCCPs because the estimated log K_{ow} values of CPs show that CPs will partition strongly into lipophilic matrices (Hilger et al., 2011). Out of 11 types of food from Japan that were analyzed for SCCPs, the SCCP concentrations were highest in the oil and fat samples (Iino et al., 2005). A feature of Chinese food culture is that a great deal of cooking oil is used for stir-frying and deep frying. Because of this, and its large population (nearly 1.4 billion people), China probably has one of the world's largest markets for oil seeds and cooking oil. Chinese people, especially northern Chinese people, often consume ready-prepared fried food, such as fried dough sticks and twists, fried vegetable balls, fried peanuts, and fried chicken, as a snack or as part of a meal. Cooking oil and fried confectionery are consumed in relatively large amounts in almost all parts of China, and an average per capita oil and fat consumption of 32.7 g d^{-1} has been reported for Chinese people (Ministry of Public Health of China, 2004). Furthermore, it has been reported that recycled cooking oil called "gutter oil" has been on the market in China and actually is assumed to be used by small restaurants and street food vendors (BBC News, 2011). We supposed that there could be a possibility of contamination by gutter oil in some cooking oil which may show a unique SCCP homolog profiles.

In the study presented here, we assessed the exposure of Chinese people to SCCPs in food. There were two specific aims of the study. First, we aimed to evaluate the contribution of SCCPs in oil and fat to the total dietary exposure to SCCPs of people in different parts of China. To achieve this we analyzed SCCPs in cooking oil and fried confectionery products collected in China in 2010 and 2012. Second, we aimed to estimate the importance of different sources of SCCPs to cooking oil. SCCPs in cooking oil can originate in the raw vegetables or seeds used to produce the oil or can enter the oil during the production process if inappropriate procedures (such as gutter oil) are used.

2. Materials and methods

2.1. Samples

Various types of cooking oil (n=49), fried confectionery (n=20), and raw seeds (n=13) were purchased from markets and supermarkets in Beijing, Fushun, Hong Kong, Shanghai, and Shenyang in 2010 and 2012. The raw seed samples that were collected were all cultivated in northern China. SCCP concentrations were also determined in several types of oil that had been produced in China and exported to Japan, and these samples were obtained from the China Town in Yokohama. The sample types and source areas of the samples that were analyzed are summarized in Table 1.

2.2. Analytical methods

The analytical methods are described in detail in the Supplementary Material (SM). Briefly, a 1 g aliquot of cooking oil or a 5 g aliquot of a fried confectionery or raw seed sample was extracted with hexane, then the extract was partitioned with dimethyl sulfoxide. The dimethyl sulfoxide was then mixed with saturated saline and hexane. The hexane layer, which contained the SCCPs, was collected and purified by passing it through an activated Florisil column, as described by Tomy et al. (1997) and Chen et al. (2013), from which the SCCPs were eluted with a 1:4 mixture of dichloromethane and hexane. Recovery standards were not used because, at the time of analysis, isotope-labeled SCCP standards were not available. The cleaned extract was concentrated under a stream of nitrogen and analyzed by the high-resolution gas chromatography and high-resolution mass spectrometry with electron-capture negative ionization (Harada et al., 2011). A short, thin capillary gas chromatography column(15 m long, 0.25 mm i.d., 0.1 μm film thickness; Agilent Technologies, Santa Clara, CA, USA) was used, and each sample was injected using the on-column injection technique. Chemical ionization was performed using methane as the reagent gas. A 1:1:1 mixture of reference solutions containing SCCPs with Cl contents of 45%, 55%, and 65% was prepared for each carbon chain length (C_{10}, C_{11}, C_{12}, and C_{13}). These mixtures were analyzed and the data used to construct a calibration curve for each carbon chain length and Cl content. The $[M–Cl]^-$ ion peak was monitored for each SCCP chain length and Cl content. The calibration curves were linear, and the correlation coefficients were >0.998.

The method detection limit (MDL) was defined as the mass of analyte injected into the gas chromatograph that gave a signal with a signal-to-noise ratio of 3. The MDLs were 0.08–20 ng g^{-1} (wet weight) for the oil samples and 0.008–2 ng g^{-1} (wet weight) for the confectionery and raw seed samples. The recoveries of the SCCPs through the extraction and sample preparation processes were evaluated by analyzing fortified samples (i.e., with the same matrices as the samples, to allow matrix effects to be evaluated), and were 81–134%. No significant differences were found in the recoveries achieved for the three different matrices. Procedural blanks (samples in which the SCCP concentrations were below the MDL) were processed with each batch of seven samples to determine if SCCP contamination occurred during the extraction and sample preparation processes.

2.3. Statistical analyses

Each value below the MDL was given a value of half of the MDL when the summary statistics were calculated. The data were tested using the Tukey–Kramer honestly significant difference test after the analysis of variance (ANOVA) method had been performed, using Student's t-test for parametric analysis or the Steel–Dwass test for nonparametric analysis. Factor analysis was performed to help identify the sources of the

SCCPs in the samples. Homolog pattern analyses were performed on samples that contained more than nine detectable SCCP homologs.

The contribution of each homolog with a specified number of chlorine atoms to the total SCCP concentration was calculated for each sample. In the factor analysis, eigenvalues >1 were taken into account and the normalized varimax rotation was applied to the eigenvectors. Statistical significance was considered to be indicated when $p<0.05$. Statistical analyses were performed using JMP version 11 software (SAS Institute Incorporated, Cary, NC, USA).

3. RESULTS

3.1. Cooking oil samples

Detectable amounts of SCCP homologs were found in 48 of the 49 cooking oil samples analyzed, and the SCCP concentrations in these samples varied widely, from <9 to 7500 ng g^{-1} (Fig. 1 and Table S1 in SM). The predominant SCCP components were the polychlorinated tridecanes (36.0% of the total SCCP concentrations), followed, in decreasing order, by the polychlorinated undecanes (27.0%), polychlorinated dodecanes (19.5%), and polychlorinated decanes (17.5%). The hexachlorinated homologs were the most abundant components for each chain length except the polychlorinated decanes (Fig. 2A; n=28).

Higher total SCCP concentrations were found in samples from Beijing (1100 ng g^{-1} in soybean oil from a market) and Fushun (1200 ng g^{-1} in a blended nut and seed oil in a package collected in 2012) than in the other samples (Fig. 1). Although these high concentrations were found in soybean oil and blended nut and seed oil, samples of raw seeds did not contain high SCCP concentrations. The highest SCCP concentrations found in samples from Hong Kong, Shanghai, and Shenyang were 230 ng g^{-1} (maize oil), 240 ng g^{-1} (soybean oil), and 210 ng g^{-1} (peanut oil), respectively. Two of the nine samples of oil that were produced in China and exported to Japan contained high concentrations of all the SCCP homologs (giving total SCCP concentrations of 7500 and 3100 ng g^{-1}). These oils were intended for use as flavorings rather than for frying, and they were excluded from our dietary intake estimates because the amounts that would be consumed at each use were considered to be negligible.

The dietary intakes of SCCPs through the consumption of cooking oil in China were estimated assuming that a typical Chinese person consumes vegetable oil at a rate of 32.7 g d^{-1} (Table 2). The estimated dietary SCCP intakes were <0.78–38 μg d^{-1}. The estimated SCCP intake was relatively high for Beijing (mean 14.8 μg d^{-1}), but this was not statistically significantly higher than the estimated SCCP intakes for the other areas (ANOVA, p=0.12). Dietary intakes of SCCPs in the whole diet (including beverages) in Beijing were estimated to be 26.3–69.4 μg d^{-1} (mean 46 μg d^{-1}) in a study performed in 2009 (Harada et al., 2011). Combining the results of this study and the previous study suggested that cooking oil might make a significant contribution (around 32.2%) to the exposure of Beijing residents to SCCPs. The estimated SCCP intakes through the consumption of cooking oil (mean dietary intake: 212 ng (kg body weight)$^{-1}$ d^{-1}) in China were below the tolerable daily intake that has been set for the non-neoplastic effects of SCCPs (100 μg (kg body weight)$^{-1}$ d^{-1}) (WHO/IPCS, 1996).

3.2. Fried confectionery samples

Fried confectionery samples were collected from Beijing, Fushun, and Shenyang (Table 1), and SCCPs were detected in all these samples (Table S2 in SM). The highest total SCCP concentrations were found in the samples from Fushun (31–1000 ng g^{-1}), and, of all the samples from Fushun, a fried peanut sample contained the highest SCCP concentration (1000 ng g^{-1}). The samples from Beijing contained the second highesttotal SCCP concentrations (11–160 ng g^{-1}) and the samples from Shenyang contained the lowest concentrations. However, there were no significant differences between the SCCP concentrations in the fried confectionery samples from Beijing, Fushun, and Shenyang (ANOVA, p=0.45).

The predominant SCCP components in the fried confectionery samples were the polychlorinated undecanes (29.5% of the total SCCP concentrations), followed, in decreasing order, by the polychlorinated decanes (25.9%), polychlorinated tridecanes (24.5%), and polychlorinated dodecanes (20.0%). The pentachlorinated homologs were the most abundant components for each chain length except the polychlorinated tridecanes (Fig. 2B; n=20).

The estimated dietary intakes of SCCPs in fried confectionery were 0.59–49.7 μg d^{-1}, and they are shown in Table 3. The mean estimated SCCP intake was higher in Fushun (mean 9.3 μg d^{-1}) than in Beijing and Shenyang because one sample (fried peanuts) from Fushun contained a particularly high SCCP concentration, as mentioned above. The geometric mean and median estimated intakes for all three cities were comparable. The estimated SCCP intakes were lower for fried confectionery than for cooking oil, but, according to the mean SCCP intake in fried confectionery in Beijing (4.7 μg d^{-1}), fried confectionery was found to contribute a considerable proportion (10.2%) of the total dietary intake of SCCPs in parts of China.

3.3. Raw seeds used to produce cooking oil

The SCCP concentrations found in raw seeds cultivated in the north of China, purchased in Fushun and Shenyang, are shown in Table S3 in SM. SCCPs were detected in 11 of the 13 samples analyzed. The total SCCP concentrations were <2–68 ng g^{-1} (Table S3 in SM). The total SCCP concentrations did not correlate with the fat contents of the samples (Pearson's product moment correlation, p=0.97). The predominant SCCP components were the polychlorinated undecanes (34.4% of the total SCCP concentrations), followed, in decreasing order, by the polychlorinated decanes (26.2%), polychlorinated tridecanes (20.6%), and polychlorinated dodecanes (18.9%). The pentachlorinated SCCP homologs were the predominant homologs for all of the chain lengths (Fig. 2C; n=6). The SCCP concentrations were somewhat lower in the raw seed samples than in the cooking oil and fried confectionery samples.

We determined the proportion that each SCCP homolog with a specified number of chlorine atoms contributed to the total SCCP concentration for each of the seed samples (n=6) and the cooking oils (n=5) made from the same types of seeds (Fig. S1 in SM). These samples came from Shenyang and Fushun, both in Liaoning Province. The northern part of China is a grain and oil-seed producing area, and has a well-developed grain and oil-seed commodity market. The raw seeds and cooking oil purchased in Shenyang and Fushun and analyzed in this study were all from northern China. The predominant SCCP components in the cereal oils (maize (n=1) and rice (n=1)) were the polychlorinated tridecanes and polychlorinated dodecanes, but the predominant SCCP components in the raw maize seed (n=2) and rice seed (n=1) samples were the polychlorinated undecanes and polychlorinated decanes. The predominant SCCP components in the nut and seed oils (peanut (n=2) and sesame (n=1)) were the polychlorinated dodecanes and polychlorinated undecanes, but the predominant components in the peanut seeds (n=1) were the polychlorinated undecanes and polychlorinated decanes. It can be seen that the SCCP homolog patterns in the raw seed and corresponding cooking oil samples, except for sesame seeds and oil, were different.

3.4. Factor analysis of the SCCP homologs

Factor analysis was performed to attempt to identify the potential sources of the SCCP homologs to the three sample types. Factors 1 and 2 accounted for 93% of the total variance (with eigenvalues >1) (Table S4 in SM). After varimax rotation had been performed, the first factor had high loadings for most of the SCCP homologs except the highly chlorinated decanes and undecanes. However, the second factor also had high loadings for the highly chlorinated undecanes and decanes. The first factor score was higher for the cooking oil samples than the other samples. The cooking oil samples and fried confectionery samples had significantly different ($p<0.05$, Steel–Dwass test) median scores for factor 1. The factor 2 scores were lower for the raw seeds than for the other sample types, and the median factor 2 scores for the raw seed and cooking oil samples were significantly different ($p<0.05$, Steel–Dwass test). These results suggest that the samples could have been contaminated with SCCPs from at least two sources and that the contributions of the sources may have been different for the three different sample types.

4. Discussion

In the present study, we assessed the exposure of humans in China to SCCPs in three types of food. SCCPs were detected in cooking oil samples (<9–7500 ng g^{-1}) and fried confectionery products (11–1000 ng g^{-1}). Raw seed samples contained detectable but relatively low concentrations of SCCPs, This is the first systematic study in which SCCP concentrations in cooking oil, raw seeds, and fried confectionery products from China have been determined.

Cooking oil consumption rates in China have changed significantly in recent years, the annual per capita consumption of cooking oil having increased by a factor of three (from 7.7 to 21.2 kg) between 1996 and 2011 (Wang, 2012). The total amount of cooking oil consumed in China reached 25.15×10^{6} t in 2011.

The SCCP concentrations found in fats (beef tallow, butter, margarine, mayonnaise, salad oil, and other products) analyzed in a market basket study in Japan were summarized in a previous publication (Iino et al., 2005). In this study, we determined SCCP concentrations in other food items. We used the earlier study as a reference, and analyzed cooking oil, fried products, and raw seeds separately. We found that cooking oil produced in China contained SCCP concentrations in the order of nanograms to micrograms per gram, indicating that cooking oil will be one of the sources of dietary exposure to SCCPs in China (contributing around 32.2% of the total SCCP intake). The fried confectionery products that were analyzed contained comparable SCCP concentrations to the oil samples, suggesting that relatively contaminated cooking oil might be used by both private consumers and local confectionery makers. Some of the raw seed samples contained SCCPs at concentrations in the order of nanograms per gram, meaning that cooking oil made out of them could contain SCCPs. The dietary intake of SCCPs in cooking oil and confectionery was estimated to be 19.5 μg d^{-1} and to account for 42.4% of the dietary intake of SCCPs from all sources, indicating that cooking oil and confectionery are two sources of human exposure to SCCPs.

The dietary intake of SCCPs was found to be more than 10 times higher in Beijing than in cities in Japan and Korea in a food duplicate study that was previously performed (Harada et al., 2011). The SCCP intake in contaminated cooking oil estimated in the study presented here could explain the different dietary intakes in China, Japan, and Korea that were found in the previous study. The SCCPs in the food duplicate samples from Beijing collected in 2009 and analyzed in the previous study had comparable contributions from the penta- and hexa-chlorinated decanes, undecanes, dodecanes, and tridecanes. These homolog patterns support our conclusion that the SCCPs in much of the food in China could originate in Chinese cooking oil.

Cooking oil samples from different Chinese cities were found to contain detectable concentrations of SCCPs. The SCCP concentrations in the samples from the different cities were not, however, statistically significantly different. The SCCP concentrations in different samples from the same city varied widely, and the variations between the cooking oil samples from different manufacturers and sources, and from different brands, are summarized in Table 4. There were 40 samples from different parts of China (excluding the samples collected from the China Town in Yokohama). Three of the seven samples containing the highest SCCP concentrations (>500 ng g^{-1}) were purchased in markets (Dongjiao Market and Xijiao Market in Beijing) and had no formal brand name, and their sources were not indicated. In contrast, 80% of the samples containing SCCPs at concentrations of <100 ng/g (including <MDL) were produced by large companies. These results indicate that oil sold in markets with little information on its source and oil produced by small local companies are more likely to contain detectable concentrations of SCCPs than is oil produced by large companies.

We also assessed the carbon chain homolog profile of oil samples into 2 groups to compare with the results of other studies in China (Table 5). Group 1 (n= 25) were all produced by large companies: C_{11} and C_{12} were predominant homolog accounting for 59.5%. The homolog distributions of group 1 were similar to those in lake water and fish samples (where the percentage of C_{11} and C_{12} was relatively high) collected from Gaobeidian Lake in Beijing (Zeng, L. X., et al., 2011). Group 2 (n= 15) were produced by small companies and markets. The relative abundance for C_{10}, C_{11}, C_{12}, and C_{13} homologs were 18.3%, 29.6%. 16.9%, and 35%. The results could not be comparable to any other studies in Table 5, indicating that group 2 could contain more complex homologs and might be produced in inappropriate procedures like “gutter oil”.

For convenience, Chinese people frequently buy fried confectionery, such as fried dough sticks (especially for breakfast), fried meat on skewers, and fried vegetable balls. These types of fried confectionery are sold and often cooked by supermarkets, shops, and street market traders. Such ready-prepared fried confectionery is eaten not only as a snack but also frequently as a side dish to a main meal. Street market traders may use recycled oil purchased in bulk to decrease their costs, and this could be one of the reasons that SCCPs were found in the fried confectionery samples that were analyzed.

The raw seed samples contained detectable concentrations of SCCPs, but these concentrations were, on the whole, relatively low. Different SCCP concentrations were found even in samples of the same kind of cereal, implying that the SCCP sources were heterogeneous. SCCPs are released from items used in manufacturing activities, such as metalworking fluid and items containing plasticizers and flame retardants. It is not easy to find alternatives to SCCPs that are available from the chemical industries in developing countries even though SCCPs are fat-soluble and potentially bioaccumulative (Fisk et al., 1996, 1998; Houde et al., 2008; Iozza et al., 2008). It seems that the current SCCP concentrations in food in China may have been caused by increasing releases of SCCPs from products containing CPs, and that these emission have been increasing because of the recent remarkable economic development of China. The presence of SCCPs in raw seeds could have been caused by their transfer to the seeds from contaminated soil, as has been found in several previous studies (Gao et al., 2012; Wang et al., 2013; Wang et al., 2014; Zeng et al., 2011). SCCP concentrations in soil can be affected by local industrial activities, which may explain the large variations found in the SCCP concentrations in the raw seeds that were analyzed. However, the SCCP homolog patterns in the raw seeds and cooking oil samples did not match. Polychlorinated decanes and polychlorinated undecanes were predominant in the raw seed samples, but polychlorinated tridecanes were predominant in the cooking oil samples. It is possible that specific SCCP homologs are accumulated or lost during the processing of seeds to produce oil. However, it is possible that some cooking oil is contaminated with SCCPs because of inappropriate operating procedures.

There is a lack of data to compare our data with because little information on human dietary exposure to SCCPs is currently available. There are also some factors that limit the abilities of laboratories to analyze CPs effectively, such as the use of electron capture negative ionization mass spectrometry in CP analyses, which can give poor sensitivity for the less chlorinated homologs ($<Cl_5$). We only chose samples that were produced in China, so we cannot identify differences in SCCP concentrations in food from different East Asian countries. Raw seed samples were only collected in Shenyang and Fushun, in northern China, and further investigations into the contamination of raw seeds with SCCPs are required. Statistics on the consumption of fried confectionery were

not available, so we tentatively assumed that a typical Chinese person consumes 50 g d^{-1}. Accurate information on Chinese eating habits is required to allow the impact of the consumption of fried confectionery on exposure to SCCPs to be assessed.

In conclusion, SCCPs were detected in cooking oil samples and samples related to cooking oil from several cities in China. SCCPs in cooking oil and fried confectionery could be the dietary sources of SCCPs to Chinese people.

Acknowledgments

This study was mainly supported by a Grant-in-Aid for Health Sciences Research from the Japanese Ministry of Health, Labour and Welfare (H21-Food-003 and H24-Food-003). The funding bodies had no input into the study design, data collection and analysis, decision to publish, or manuscript preparation. Takumi Takasuga and Chihiro Nouda are employees of Shimadzu Techno-Research Incorporated. The other authors declare they have no competing financial interests.

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Table 1. Sample categories, the sampling locations, the years the samples were collected, the numbers of samples, and the sample types

	Area	Year	n	Individual items
Cooking oil	Shanghai	2010	6	peanut, maize, rapeseed, soybean, sunflower seed oil
	Beijing	2010	7	soybean, mustard, sesame seed, olive oil
	Fushun(1)	2010	8	peanut, maize, soybean, sunflower seed oil
	Fushun(2)	2012	8	nut blend, maize, soybean, sunflower, sesame, mixed oil
	Shenyang	2012	6	peanut, maize, soybean, sunflower seed, mixed oil
	Hong Kong	2010	5	peanut, maize, olive oil
	Japan	2010	9	peanut, sesame seed, pepper oil
Fried confectionery	Beijing	2010	6	noodle, dough twists, sesame seed, rice cracker
	Fushun	2012	7	vegetable balls, dough twists, sesame seed, peanut, soybean
	Shenyang	2012	7	vegetable balls, donuts, dough twists, sesame seed, cake, soybean, mutton slices
Raw seeds for vegetable oil	Fushun	2012	6	peanut seed, soybean, maize, rice, sesame, sunflower seed
	Shenyang	2012	7	peanut seed, peanut, sesame, maize, rice, soybean, sunflower seed

Fushun (1) was collected in 2010; Fushun (2) was collected in 2012; Japan: the samples were produced in China and exported to Japan, and were collected from China Town in Yokohama.

Table 2. Estimated dietary intakes (in μg d^{-1}) of short-chain chlorinated paraffins (SCCPs) by Chinese people through consuming cooking oil produced in China

Sampling area	Shanghai	Beijing	Fushun		Shenyang	Hongkong	Japan [a]
Year	2010	2010	2010	2012	2012	2010	2010
Range	<0.78–8.0	1.26–36	<0.78–26	<1.60–38	<1.60–7.5	<0.78–7.3	<0.78–6.4
Q2	1.1	17.0	3.7	1.9	2.0	5.7	1.3
Mean ± SD	2.7±2.9	14.8±12.3	6.5±8.4	10.7±16.6	3.3±2.5	4.9±2.6	2.5±2.1
GM (GSD)	1.75 (2.62)	9.06 (3.47)	3.47 (3.30)	3.65 (4.31)	2.59 (2.04)	3.89 (2.46)	1.87 (2.23)

The SCCP intakes were estimated assuming that a Chinese person consumes vegetable oil at a rate of 32.7 g d^{-1}

Japan: The samples were produced in China and exported to Japan, and were collected from China Town in Yokohama; [a] Two oil samples used for flavoring only were excluded because of their low consumption rates.

Q2: median; GM: geometric mean; GSD: geometric standard deviation.

Table 3. Dietary intakes (in μg d^{-1}) of short-chain chlorinate paraffins (SCCPs) by Chinese people through the consumption of fried confectionery produced in China

Sampling area	Beijing	Shenyang	Fushun
Year	2010 (6/6)	2012 (7/7)	2012 (7/7)
Range	0.59–-7.8	0.96–-4.5	1.52–-49.7
Q2	4.9	1.7	4.0
Mean ± SD	4.7±3.0	2.5±1.4	9.3±16.4
GM (GSD)	3.47 (2.72)	2.18 (1.77)	4.56 (2.83)

The SCCP intakes were estimated using the tentative assumption that a Chinese person consumes fried confectionery at a rate of 50 g d^{-1} ; Q2: median; GM: geometric mean; GSD: geometric standard deviation.

Table 4. Total short-chain chlorinated paraffin concentrations in cooking oil samples produced by different Chinese manufacturers

Concentration (ng g^{-1})	n	Class of manufacturer*
>500	7	3 handmade, 2 from large companies, 2 from small companies
100–500	13	6 from large companies, 7 from small companies,
<100	4	4 from large companies
<MDL (not detected)	16	2 handmade, 13 from large companies, 1 from a small company

*Handmade: collected from a market or farmer, with no particular brand or other information marked. Large company: a company with factories in several cities, the products of which consumers can purchase in most Chinese cities. Small company: a local company with a factory in the suburbs of a small city and no factories in other cities

Table 5. Comparison of relative homolog patterns found in the environment/ biota in China

Name of samples	Relative SCCP homolog group abundance profiles (%)($\Sigma C_{10\text{-}13}$=100%)				Region	Reference
	CP-C10	CP-C11	CP-C12	CP-C13		
Oil	17.5	27	19.5	36	Different cities	This study
(Group 1)	19.2	32.9	26.6	21.1	Different cities	This study
(Group 2)	18.3	29.6	16.9	35	Different cities	This study
Fried confectionery	25.9	29.5	20	24.5	Different cities	This study
Raw seeds	26.2	34.4	18.9	20.6	Different cities	This study
Seawater	42	40.6	13	4.3	Liaodong Bay, North China	Ma, X. D. et al., 2014
Sediments	34.6	34.9	23.7	6.7	Liaodong Bay, North China	Ma, X. D. et al., 2014
Organism	37.4	44.9	12.5	5.2	Liaodong Bay, North China	Ma, X. D. et al., 2014
Sewage sludge	27	34	23	16	North China	Zeng, L. X. et al., 2012
Sediment	36.7	32.9	20.2	10.2	Bohai Sea	Ma, X. D.,et al., 2014
Bivalve	28.7	37.7	19.4	14.2	Bohai Sea	Ma, X. D.,et al., 2014
Bird (White wagtail)	27	29	27	17	South China	Luo, X. J. et al., 2015
Bird (Red-folanked bluetail)	28	27	24	20	South China	Luo, X. J. et al., 2015
Bird (Goldfinch)	30	27	25	18	South China	Luo, X. J. et al., 2015
Bird (Oriental magrie-robin)	24	25	26	25	South China	Luo, X. J. et al., 2015
Bird (Long-tail shrike)	31	26	21	22	South China	Luo, X. J. et al., 2015
Bird (Great tit)	23	22	29	26	South China	Luo, X. J. et al., 2015

Bird (Grey-backed trush)	31	29	24	16	South China	Luo, X. J. et al., 2015
Bark-winter	23	25	25	27	Beijing	Wang, T. et al., 2015
Bark-summer	39	30	17	14	Beijing	Wang, T. et al., 2015
Needle-winter	29	27	22	22	Beijing	Wang, T. et al., 2015
Needle-summer	41	30	16	13	Beijing	Wang, T. et al., 2015
Sediment	40.4	39.4	15.4	4.8	Liaohe River Basin	Gao, Y. et al., 2012
Paddy soil	41.3	40.1	13.9	4.7	Liaohe River Basin	Gao, Y. et al., 2012
Upland soil	42.7	38.7	14.7	3.9	Liaohe River Basin	Gao, Y. et al., 2012
Mollusks (Rapana venosa)	34	34.4	14.5	17.1	Bohai Sea	Yuan, B. et al., 2012
Mollusks (Neverita didyma)	34.8	31.5	21.9	11.8	Bohai Sea	Yuan, B. et al., 2012
Mollusks (Chlamys Farreri)	28.5	26.8	27.2	17.5	Bohai Sea	Yuan, B. et al., 2012
Mollusks (Mya arenaria)	20.1	32.7	18.8	28.4	Bohai Sea	Yuan, B. et al., 2012
Soil (site B)	23.1	19.9	25.6	31.4	Liangshui River, Tongzhou	Zeng, L. X. et al., 2011
Soil (site C)	37.6	27.1	18.3	17	Liangshui River, Tongzhou	Zeng, L. X. et al., 2011
Soil (site G)	30.8	24.2	23.7	21.3	Liangshui River, Tongzhou	Zeng, L. X. et al., 2011
Soil (site J)	54	23	13.2	9.8	Liangshui River, Tongzhou	Zeng, L. X. et al., 2011
Fish (Leather catfish)	19.2	31.5	33.2	16.1	Liaobeidian Lake, Beijing	Zeng, L. X. et al., 2011
Fish (Common carp)	17.4	33.5	35.9	13.2	Liaobeidian Lake, Beijing	Zeng, L. X. et al., 2011
Fish (Chinese softshell turtle)	12.7	39.9	35.7	11.7	Liaobeidian Lake, Beijing	Zeng, L. X. et al., 2011
Fish (Java tilapia)	13.9	38.8	38	9.3	Liaobeidian Lake, Beijing	Zeng, L. X. et al., 2011
Lake water from upstream	14.6	34.9	35.5	15	Liaobeidian Lake, Beijing	Zeng, L. X. et al., 2011
Lake water from STP outfall	14.9	35.4	35.2	14.5	Liaobeidian Lake, Beijing	Zeng, L. X. et al., 2011

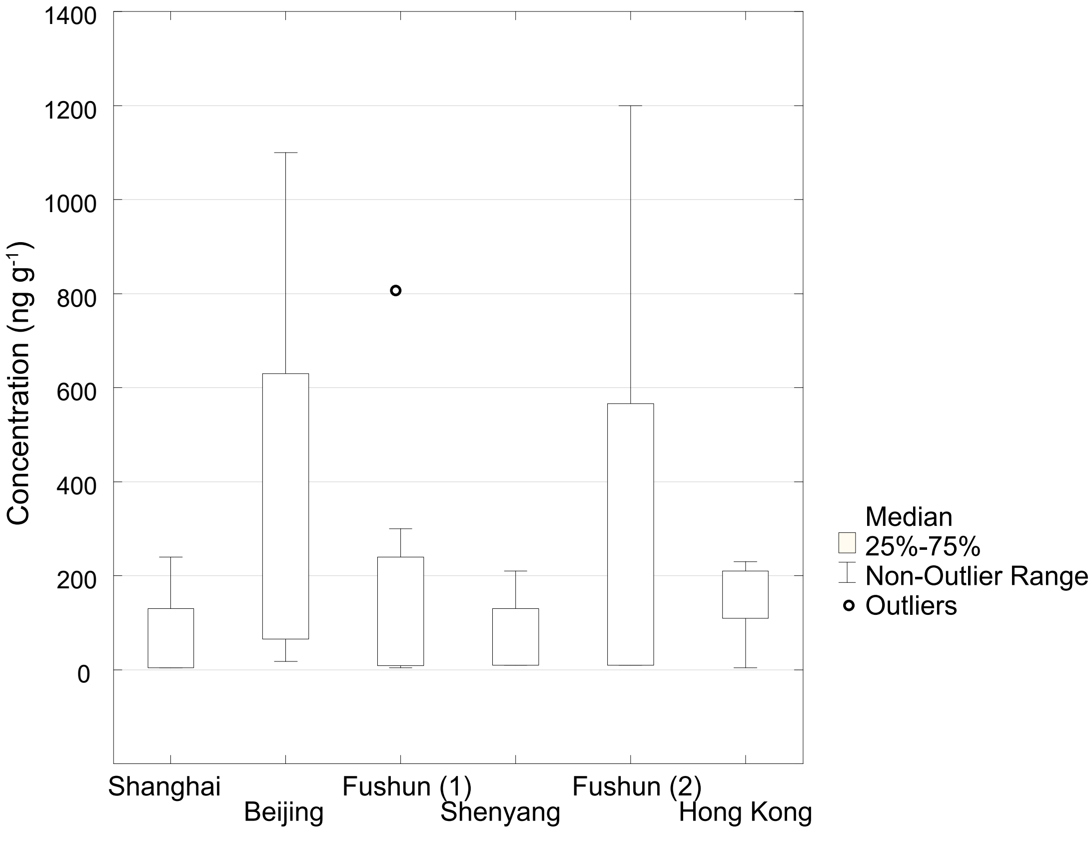
Lake water from downstream	15.8	34.8	34.4	15	Liaobeidian Lake, Beijing	Zeng, L. X. et al., 2011
Raw sewage	39.1	27.2	17.2	16.5	Sewage treatment plant, Beijing	Zeng, L. X. et al., 2012
Secondary effluent	50.8	29	13.1	7.1	Sewage treatment plant, Beijing	Zeng, L. X. et al., 2012
Woodland soil	29.3	26	23.4	22	Guangzhou	Chen, L. et al. 2013
Vegetable field soil	29.9	29.1	23.3	17.6	Guangzhou	Chen, L. et al. 2013
Paddy soil	28.4	30.2	19.8	21.5	Guangzhou	Chen, L. et al. 2013
Background soil	26	28	22.2	13.7	Guangzhou	Chen, L. et al. 2013
Air	34	34	20	12	Dongguan	Wang, Y., et al., 2013
Air	37	36	17	10	Guangzhou	Wang, Y., et al., 2013
Air	39	34	16	11	Huizhou	Wang, Y., et al., 2013

Group 1 was the samples produced by large companies; Group 2 was the samples produced by small companies and markets.

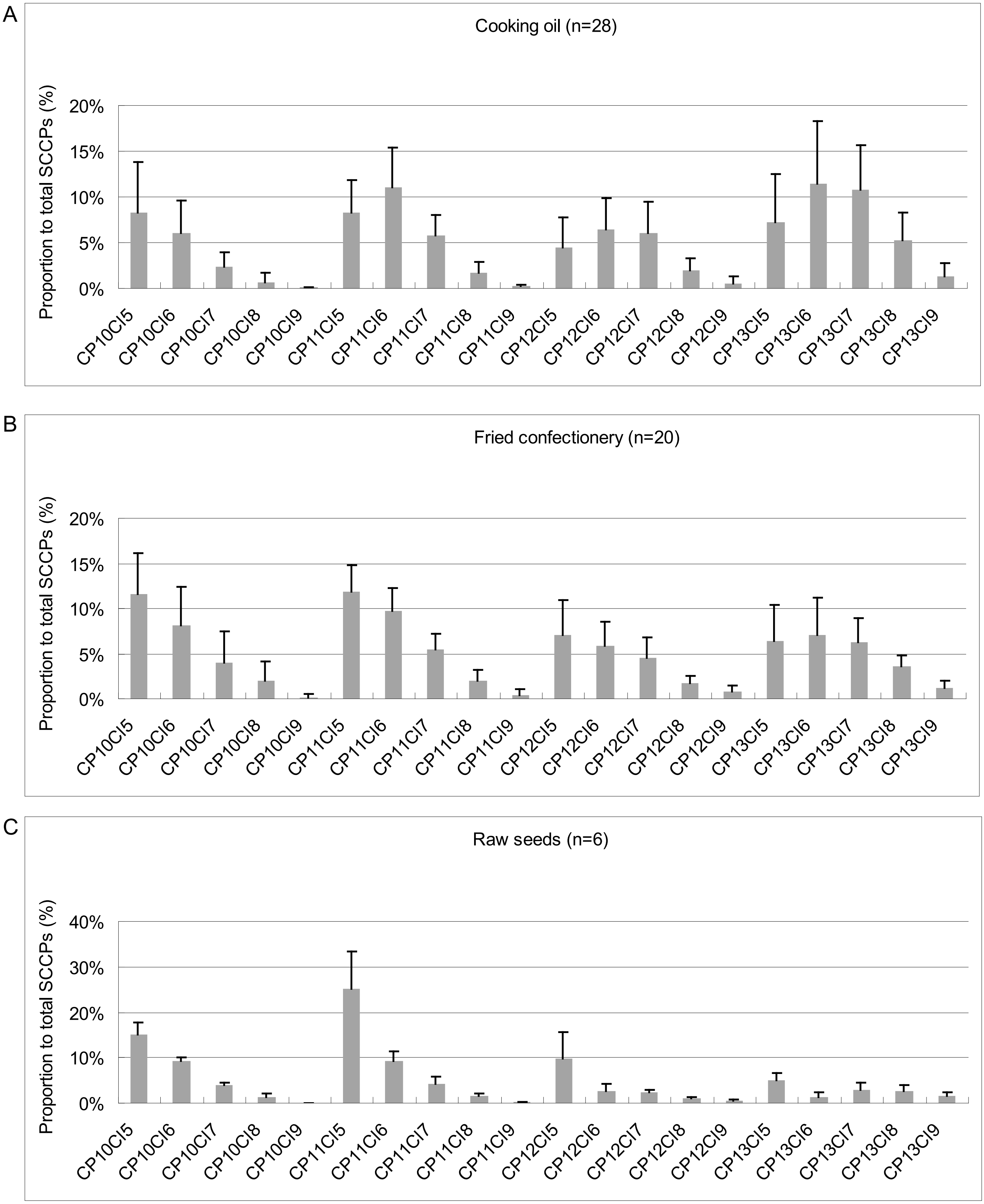
Figure legends

Figure 1. Box-and-whisker plot of the total short-chain chlorinated paraffin concentrations found in the cooking oil samples. Each box represents the first, second, and third quartiles. The lower whisker indicates the lowest value within the −1.5 interquartile range of the first quartile. The upper whisker indicates the highest value within the +1.5 interquartile range of the third quartile. Fushun (1) was collected in 2010, and Fushun (2) was collected in 2012.

Figure 2. Contributions of the short-chain chlorinated paraffin (SCCP) homologs to the total SCCP concentrations in the (A) cooking oil samples (n=28), (B) fried confectionery samples (n=20), and (C) raw seeds that are used to produce vegetable oil (n=6). Only samples containing more than nine detectable SCCP homologs were selected. The contribution of each homolog to the total SCCP concentration was calculated for each sample separately. The bars indicate the means and the whiskers the standard deviations.



1400
1200
1000
800
600
400
200
0
Concentration (ng g-1)
Shanghai
Beijing
Fushun (1)
Shenyang
Fushun (2)
Hong Kong
Median
25%-75%
Non-Outlier Range
Outliers


A
Cooking oil (n=28)
B
Fried confectionery (n=20)
C
Raw seeds (n=6)
Proportion to total SCCPs (%)
0%
5%
10%
15%
20%
30%
40%
CP10Cl5
CP10Cl6
CP10Cl7
CP10Cl8
CP10Cl9
CP11Cl5
CP11Cl6
CP11Cl7
CP11Cl8
CP11Cl9
CP12Cl5
CP12Cl6
CP12Cl7
CP12Cl8
CP12Cl9
CP13Cl5
CP13Cl6
CP13Cl7
CP13Cl8
CP13Cl9

Supplementary Material

Analytical methods

Chemicals

Polychlorinated decanes (with Cl contents of 44.82%, 55.00%, and 65.02%), polychlorinated undecanes (with Cl contents of 45.50%, 55.20%, and 65.25%), polychlorinated dodecanes (with Cl contents of 45.32%, 55.00%, and 65.08%), and polychlorinated tridecanes (with Cl contents of 44.90%, 55.03%, and 65.18%) were obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany). The internal standard (syringe spike), $^{13}C_{12}$-labeled 2,3,3′,5,5′-pentachlorobiphenyl (CB-111), was obtained from Cambridge Isotope Laboratories (Andover, MA, USA). Acetone, hexane, dimethyl sulfoxide, and sodium sulfate were purchased from Kanto Chemical Company Incorporated (Tokyo, Japan).

Extraction and clean-up procedure

A 1 g aliquot of cooking oil or a 5 g aliquot of a fried confectionery or raw seed sample was extracted with 20 mL of hexane for 10 min, using a shaker. A 2 mL aliquot of the cooking oil extract or a 4 mL aliquot of the fried confectionery or raw seed extract was taken and shaken with 2.5 mL hexane-saturated dimethyl sulfoxide for 4 min. The dimethyl sulfoxide layer was transferred to a new tube and shaken with 1 mL hexane for 2 min. The dimethyl sulfoxide layer was removed and combined with 10 mL hexane-washed water, 0.5 mL saturated saline, and 2 mL hexane. The hexane layer was removed and passed through a sodium sulfate column.

The crude extract was loaded onto an 8 g activated Florisil (Wako Pure Chemicals, Osaka, Japan) column that had been preconditioned with 90 mL of a 1:4 (v/v) mixture of dichloromethane and hexane. The short-chain chlorinated paraffins (SCCPs) were eluted with 90 mL of a 1:4 mixture of dichloromethane and hexane. The eluate was concentrated to 50 μL of decane under a stream of nitrogen, then 250 pg of $^{13}C_{12}$-labeled CB-111 (used as an internal standard) was added before the extract was analyzed by high-resolution gas chromatography and high-resolution mass spectrometry with electron-capture negative ionization (HRGC/ECNI/HRMS). The molecular weight of $^{13}C_{12}$-labeled CB-111 is similar to the molecular weight of the C_{12} CPs, so the labeled CD-111 may have interfered with the SCCP determination. However, a significant effect for this homolog was not identified in the factor analysis, so we concluded that there was little likelihood that the $^{13}C_{12}$-labeled CB-111 interfered with the results of the SCCP analyses. An isotope-labeled SCCP standard was not available at the time of analysis, so a surrogate standard was not used in the extraction and cleanup procedure.

Instrumental analysis and quality control

The HRGC/ECNI/HRMS system comprised a Hewlett Packard 6890 Series HRGC system (Agilent Technologies, Palo Alto, CA, USA) and a Thermo Fisher Scientific Finnigan MAT 95 XL HRMS system (Thermo Fisher Scientific Incorporated, Yokohama, Japan). The HRGC system was equipped with a DB-5MS capillary column (15 m long, 0.25 mm i.d., 0.1 μm film thickness; Agilent Technologies), and the carrier gas, used at a flow rate of 1.0 mL min^{-1}, was helium (99.9999% pure; Air Liquide Japan Ltd., Tokyo, Japan). A 2 μL aliquot of each sample extract was injected using the on-column injection technique. The initial injector temperature was 100 °C, and it was increased to 300 °C at 100 °C min^{-1}. The initial oven temperature was 100 °C, which was held for 1 min, then the temperature was increased to 300 °C at 10 °C min^{-1}. The transfer line temperature was 300 °C. The ECNI reagent gas was methane (99.9999% pure; Air Liquide Japan Ltd.), and the flow rate was 2 mL min^{-1}. The ion source temperature was 130 °C. The ionizing voltage and emission current were 40 eV and 250 μA, respectively.

A 1:1:1 mixture of reference solutions containing SCCPs with Cl contents of 45%, 55%, and 65% was prepared for each carbon chain length (C_{10}, C_{11}, C_{12}, and C_{13}). These mixtures were analyzed and the data were used to construct a calibration curve for each SCCP homolog with same number of chlorine atoms, using the compositions determined using electron impact ionization mass spectrometry (EI/MS) (Harada et al., 2011). The homolog concentrations in the mixtures were assumed to be proportional to the relative peak areas determined by EI/MS.

The samples were analyzed by ECNI/HRMS, in which the highest $[M–Cl]^-$ ion peak was used to quantify each homolog with the same number of chlorine atoms because this would be a relatively specific ion fragment. The calibration curves were constructed using five dilutions of the 1:1:1 SCCP mixtures (at total CP concentrations of 20–2000 ng mL^{-1}). Each calibration curve was linear, and the correlation coefficients (r) were all >0.998. When a sample concentration exceeded the upper limit of the relevant calibration curve the sample was diluted to bring the concentration into the calibration curve range.

The instrumental detection limit (IDL) was defined as the injected mass of the analyte that produced a signal with a signal-to-noise ratio of 3. No SCCPs were detected in the procedural blank samples, so the method detection limit (MDL) value was considered to be equal to the IDL (Martin et al. Anal. Chem. 2002, 74, 584-590). The MDLs were 0.08–20 ng g^{-1} for the oil samples and 0.008–2 ng g^{-1} for the confectionery and raw seed samples. The detected values were used even if they were below the quantification limit (a signal-to-noise ratio of 10).

The recoveries through the extraction and cleanup processes were evaluated by analyzing seven fortified 1 g aliquots of samples that were uncontaminated with SCCPs. A total of 570 ng of SCCPs was added to each of these fortified samples. The recoveries were 81–134% (mean±SD 96±16%). No significant differences were found between the recoveries for the three matrices. The recoveries were around 100% and isotope-labeled SCCP standards were not available, so the measured values were not corrected for the recoveries. Procedural blanks (samples that were uncontaminated with SCCPs) were processed with each batch of seven samples to check for any contamination that occurred during the extraction and cleanup processes.

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Table S1. Short-chain chlorinated paraffin (SCCP) concentrations (in ng g^{-1}) in the cooking oil samples produced in China

SCCP homologs	Shanghai 2010 Range (n >MDL)	n=6 Q2	Beijing 2010 Range (n >MDL)	n=7 Q2	Fushun 2010 Range (n >MDL)	n=8 Q2	2012 Range (n >MDL)	n=8 Q2	Shenyang 2012 Range (n >MDL)	n=6 Q2	Hong Kong 2010 Range (n >MDL)	n=5 Q2	Japan 2010 Range (n >MDL)	n=9 Q2
$C_{10}H_{17}Cl_5$	<4–7.4(2)	<4	5.1–140(7)	24	<4–70(5)	9	<4–110(2)	<4	<4–16(1)	<4	<4–35(4)	11	<4–410(5)	4.8
$C_{10}H_{16}Cl_6$	<2–4.6(3)	<2	2.8–100(7)	22	<2–43(5)	6.1	<2–140(4)	2.9	<2–14(3)	2.7	<2–23(4)	5.8	<2–290(6)	2.7
$C_{10}H_{15}Cl_7$	<0.3–1.4(4)	0.58	0.75–26(7)	8.5	<0.3–11(5)	2.4	<1–75(7)	3.9	<1–8.4(4)	3.9	<0.3–6.8(4)	1.6	0.54–83(9)	0.8
$C_{10}H_{14}Cl_8$	<0.2–0.63(4)	0.21	<0.2–2.3(6)	1.5	<0.2–1.6(5)	0.36	<0.9–28(2)	<0.9	<0.9–3.7(4)	2.7	<0.2–1.1(4)	0.3	<0.2–11(4)	<0.2
$C_{10}H_{13}Cl_9$	<0.08–0.41(3)	0.08	<0.08–0.44(4)	0.09	<0.08–0.19(3)	<0.08	<0.1–0.82(2)	<0.1	<0.1–0.31(3)	0.1	<0.08–0.097(2)	<0.08	<0.08–0.77(3)	<0.08
total $C_{10}Cl_x$	<4–14(2)	<4	9.3–270(7)	56	<4–130(5)	19.5	<4–350(5)	6.3	<4–42(3)	7	<4–66(4)	19	<4–790(5)	8.3
$C_{11}H_{19}Cl_5$	<5–12(2)	<5	<5–140(6)	32	<5–76(5)	8.9	<5–98(2)	<5	<5 (0)	<5	<5–27(4)	16	<5–680(5)	5.3
$C_{11}H_{18}Cl_6$	<3–13(3)	<3	4.9–180(7)	78	<3–86(6)	14.5	<2–150(2)	<2	<2–17(2)	<2	<3–32(4)	25	<3–1000(5)	3.7
$C_{11}H_{17}Cl_7$	0.59–6.5(6)	1.45	2.4–65(7)	40	0.88–29(8)	6.5	<2–130(6)	3.6	<2–14(4)	3.4	0.79–13(5)	10	1.4–390(9)	2.2
$C_{11}H_{16}Cl_8$	<0.4–1.8(5)	0.69	0.79–11(7)	3.6	<0.4–4.9(6)	1.24	<0.5–47(7)	2.3	<0.5–7.1(5)	2.8	<0.4–3.3(4)	1.9	<0.4–72(7)	0.6
$C_{11}H_{15}Cl_9$	<0.1–0.66(4)	0.25	<0.1–0.82(5)	0.19	<0.1–0.43(6)	0.13	<0.4–3.9(3)	<0.4	<0.4–1.2(4)	0.73	<0.1–0.32(4)	0.2	<0.1–3.7(5)	0.1
total $C_{11}Cl_x$	<5–33(3)	<5	10.8–390(7)	170	<5–200(6)	42	<5–430(5)	6	<5–39(4)	6.3	<5–71(4)	56	<5–2100(5)	10
$C_{12}H_{21}Cl_5$	<6–6.2(1)	<6	<6–26(4)	6.1	<6–48(3)	<6	<20–75(2)	<20	<20(0)	<20	<6–17(3)	7.4	<6–390(2)	<6
$C_{12}H_{20}Cl_6$	<4–14(2)	<4	<4–61(5)	11	<4–71(6)	8.6	<4–110(2)	<4	<4–10(2)	<4	<4–31(4)	15	<4–720(3)	<4
$C_{12}H_{19}Cl_7$	<2–14(2)	<2	<2–70(6)	10	<2–49(7)	8.3	<0.9–130(4)	2.4	<0.9–15(3)	1.48	<2–22(4)	12	<2–560(6)	2.3
$C_{12}H_{18}Cl_8$	<0.8–3.7(2)	<0.8	<0.8–22(6)	3.5	<0.8–8.6(6)	1.7	<0.4–53(6)	2.3	<0.4–9.8(5)	2.0	<0.8–5.4(4)	3	<0.8–120(5)	0.8
$C_{12}H_{17}Cl_9$	<0.4–0.96(4)	0.56	<0.4–3.5(5)	1.4	<0.4–0.99(2)	<0.4	<0.3–11(4)	1.08	<0.3–4.4(4)	2.4	<0.4–1.0(3)	0.5	<0.4–14(2)	<0.4
total $C_{12}Cl_x$	<6–38(2)	<6	<6–170(6)	26	<6–180(6)	21.5	<20–380(2)	<20	<20–39(2)	<20	<6–75(4)	38	<6–1800(3)	<6
$C_{13}H_{23}Cl_5$	<9–33(1)	<9	<9–84(5)	16	<9–70(3)	<9	<20 (0)	<20	<20 (0)	<20	<9–13(2)	<9	<9–520(3)	<9
$C_{13}H_{22}Cl_6$	<7–63(2)	<7	<7–130(6)	39	<7–110(5)	10.4	<4–49(2)	<4	<4–15(2)	<4	<7–24(4)	11	<7–990(5)	13
$C_{13}H_{21}Cl_7$	<2–61(4)	3.7	2.8–110(7)	33	2.3–80(8)	11.2	<3–84(3)	<3	<3–25(3)	3.5	<2–22 (4)	13	<2–920(7)	12
$C_{13}H_{20}Cl_8$	<2–21(3)	<2	<2–35(6)	11	<2–24(5)	5.8	<2–85(4)	4	<2–36(4)	5.6	<2–10(4)	5.8	<2–320(6)	4.9
$C_{13}H_{19}Cl_9$	<0.5–3.3(5)	0.76	<0.5–7.3(6)	2.7	<0.5–3.3(5)	1.08	<0.8–24(4)	2.15	<0.8–12(5)	3.9	<0.5–2.4(4)	1.1	<0.5–51(5)	0.8
total $C_{13}Cl_x$	<9–180(2)	<9	<9–360 (6)	130	<9–290 (5)	28.5	<20–240(3)	<20	<20–88(2)	<20	<9–71(4)	31	<9–2800(5)	31
Total$C_{10\text{-}13}Cl_x$	<9–240 (2)	<9	18–1100 (7)	520	<9–800 (6)	105	<20–1200 (3)	<20	<20–210(2)	<20	<9–230(4)	170	<9–7500 (6)	94

MDL: Method detection limits; Q2: median

Japan: These samples were produced in China and exported to Japan, and were collected from China town in Yokohama.

Table S2. Short-chain chlorinated paraffin (SCCP) concentrations (in ng g^{-1}) in the fried confectionery samples produced in China

SCCP Homologs	Beijing 2010 Range (*n* >MDL)	n=6 Q2	Fushun 2012 Range (*n* >MDL)	n=7 Q2	Shenyang 2012 Range (*n* >MDL)	n=7 Q2
$C_{10}H_{17}Cl_5$	2.2–28(6)	9.1	2.9–130(7)	10	1.4–8.4(7)	4.0
$C_{10}H_{16}Cl_6$	0.9–12(6)	5.5	2.4–210(7)	6.4	1.4–4.0(7)	3.0
$C_{10}H_{15}Cl_7$	0.24–4(6)	1.5	2–170(7)	2.5	1.0–2.3(7)	1.4
$C_{10}H_{14}Cl_8$	0.036–1.1(6)	0.29	0.77–79(7)	1.2	0.51–1.4(7)	0.9
$C_{10}H_{13}Cl_9$	<0.008–0.23(5)	0.047	0.022–2.7(7)	0.1	0.024–0.23(7)	0.1
total $C_{10}Cl_x$	3.4–41(6)	17	8.7–590.0(7)	21	5.4–15.0(7)	10
$C_{11}H_{19}Cl_5$	1.8–19(6)	10	3.3–57(7)	8.4	2.2–9.5(7)	5.5
$C_{11}H_{18}Cl_6$	1.6–23(6)	9.1	2.2–96(7)	7.5	1.4–7.9(7)	3.3
$C_{11}H_{17}Cl_7$	0.53–9.1(6)	3.8	1.9–97(7)	3.7	1.2–4.1(7)	2.3
$C_{11}H_{16}Cl_8$	0.12–1.9(6)	1.09	0.8–42(7)	1.2	0.47–1.2(7)	1.1
$C_{11}H_{15}Cl_9$	0.015–0.25(6)	0.096	0.1–4.4(7)	0.2	0.10–0.55(7)	0.2
total $C_{11}Cl_x$	4.1–53.0(6)	24.5	8.5–300(7)	21	6.3–23(7)	12
$C_{12}H_{21}Cl_5$	<0.6–10(5)	5.1	<2-6.9(6)	5.1	<2-12(5)	4.6
$C_{12}H_{20}Cl_6$	0.72–16(6)	5.6	1.7–17(7)	2.9	0.81–11(7)	1.7
$C_{12}H_{19}Cl_7$	0.48–14(6)	4.3	1.2–12(7)	2.3	0.85–8.1(7)	1.4
$C_{12}H_{18}Cl_8$	0.13–3.6(6)	1.09	0.63–3.7(7)	1.0	0.58–2.2(7)	0.7
$C_{12}H_{17}Cl_9$	<0.04–0.45(5)	0.21	0.3–1(7)	0.5	0.31–0.67(7)	0.5
total $C_{12}Cl_x$	1.3–44.0(6)	16	5.1–51.0(7)	13	3.9–34.0(7)	8.3
$C_{13}H_{23}Cl_5$	0.9–12(5)	6.4	<2-15 (5)	6.4	<2-6.7 (3)	<2
$C_{13}H_{22}Cl_6$	1.2–19(6)	9.6	2.4–14(7)	3.4	<0.4–5.2(6)	1.8
$C_{13}H_{21}Cl_7$	0.79–15(6)	9.3	1.8–15(7)	3.2	0.87–5.5(7)	1.6
$C_{13}H_{20}Cl_8$	0.25–5.3(6)	3.4	1.0–11(7)	2.0	0.89–4.0(7)	1.6
$C_{13}H_{19}Cl_9$	0.06–0.98(6)	0.69	0.51–2.5(7)	0.7	0.49–1.0(7)	0.7
total $C_{13}Cl_x$	2.3–49 (6)	28	8.0–56(7)	17	3.6–22 (7)	5.6
Total$C_{10\text{-}13}Cl_x$	11-160 (6)	100	31-1000 (7)	80	19-89 (7)	34

MDL: Method detection limits; Q2: median.

Table S3. Short-chain chlorinated paraffin (SCCP) concentrations (in ng g^{-1}) in the raw seeds samples from Shenyang and Fushun

SCCP homologs	Shenyang peanut seeds	peanut seeds	sesame seeds	maize seeds	soybean seeds	sunflower seeds	rice seeds	Fushun peanut seeds	sesame seeds	maize seeds	soybean seeds	sunflower seeds	rice seeds	Total median
$C_{10}H_{17}Cl_5$	3.4	<0.40	2.1	4.1	<0.40	<0.40	2	8.4	7.1	3.5	0.74	0.49	12	2.1
$C_{10}H_{16}Cl_6$	2	<0.20	1.1	2.4	<0.20	0.51	1.2	5.9	6.1	2.2	0.4	0.35	7.3	1.2
$C_{10}H_{15}Cl_7$	0.84	<0.10	0.54	1	<0.10	0.34	0.48	1.1	3.2	0.64	0.29	0.26	1.1	0.54
$C_{10}H_{14}Cl_8$	0.37	<0.09	<0.09	0.41	<0.09	<0.09	<0.09	0.23	1.5	0.23	<0.09	<0.09	<0.09	<0.09
$C_{10}H_{13}Cl_9$	<0.01	<0.01	<0.01	0.019	<0.01	<0.01	<0.01	<0.01	0.077	<0.01	<0.01	<0.01	<0.01	<0.01
total $C_{10}Cl_x$	6.7	<0.4	3.7	7.9	<0.4	0.84	3.7	16	18	6.6	1.4	1.1	21	6.6
$C_{11}H_{19}Cl_5$	4.9	<0.50	3	3.9	<0.50	<0.50	5.4	20	18	8.3	<0.50	<0.50	29	3.9
$C_{11}H_{18}Cl_6$	1.6	<0.20	1.1	1.7	<0.20	0.47	1.5	7.3	7.2	2.7	<0.20	0.33	7.6	1.5
$C_{11}H_{17}Cl_7$	0.8	<0.20	0.42	0.96	<0.20	0.21	0.46	1.4	4.8	0.72	<0.20	<0.20	1.4	0.46
$C_{11}H_{16}Cl_8$	0.37	<0.05	0.25	0.42	<0.05	<0.05	<0.05	<0.05	1.4	0.19	<0.05	<0.05	<0.05	<0.05
$C_{11}H_{15}Cl_9$	<0.04	<0.04	<0.04	0.044	<0.04	<0.04	<0.04	<0.04	0.14	<0.04	<0.04	<0.04	<0.04	<0.04
total $C_{11}Cl_x$	7.7	<0.5	4.8	7	<0.5	0.68	7.4	28	32	12	<0.5	<0.5	38	7.7
$C_{12}H_{21}Cl_5$	3.6	<2.00	<2.00	3.9	<2.00	<2.00	<2.00	<2.00	3.2	<2.00	<2.00	<2.00	<2.00	<2.00
$C_{12}H_{20}Cl_6$	0.91	<0.40	<0.40	0.98	<0.40	<0.40	<0.40	<0.40	1.9	<0.40	<0.40	<0.40	<0.40	<0.40
$C_{12}H_{19}Cl_7$	0.43	<0.09	0.38	0.53	<0.09	<0.09	<0.09	<0.09	1.8	0.36	<0.09	<0.09	<0.09	<0.09
$C_{12}H_{18}Cl_8$	0.21	<0.04	0.17	0.25	<0.04	<0.04	<0.04	<0.04	0.77	0.16	<0.04	<0.04	<0.04	<0.04
$C_{12}H_{17}Cl_9$	0.17	<0.03	<0.03	0.17	<0.03	<0.03	<0.03	<0.03	0.37	<0.03	<0.03	<0.03	<0.03	<0.03
total $C_{12}Cl_x$	5.4	<2	<2	5.8	<2	<2	<2	<2	8	<2	<2	<2	<2	<2
$C_{13}H_{23}Cl_5$	<2.00	<2.00	<2.00	<2.00	<2.00	<2.00	<2.00	<2.00	2.9	<2.00	<2.00	<2.00	<2.00	<2.00
$C_{13}H_{22}Cl_6$	<0.40	<0.40	<0.40	<0.40	<0.40	<0.40	0.8	<0.40	2	<0.40	<0.40	<0.40	<0.40	<0.40
$C_{13}H_{21}Cl_7$	0.99	<0.30	0.52	0.61	<0.30	<0.30	0.42	<0.30	2.3	<0.30	<0.30	<0.30	<0.30	<0.30
$C_{13}H_{20}Cl_8$	0.73	<0.20	0.52	0.64	<0.20	<0.20	0.38	<0.20	2.2	<0.20	<0.20	<0.20	<0.20	<0.20
$C_{13}H_{19}Cl_9$	0.41	<0.08	0.36	0.4	<0.08	<0.08	<0.08	<0.08	0.78	<0.08	<0.08	<0.08	<0.08	<0.08
total $C_{13}Cl_x$	2.1	<2	<2	<2	<2	<2	<2	<2	10	<2	<2	<2	<2	<2
Total$C_{10\text{-}13}Cl_x$	22	<2	8.5	21	<2	<2	11	44	68	19	<2	<2	59	22
(ng $g_{lipid\ weight}^{-1}$)	44	<4.0	17.5	5500	<20	<4.0	3900	98	126	10000	<18.0	<4.2	12700	44
Fat content (%)	49.9	49.8	48.5	0.4	9.9	50.3	0.3	45.1	53.8	0.2	11.1	47.9	0.5	

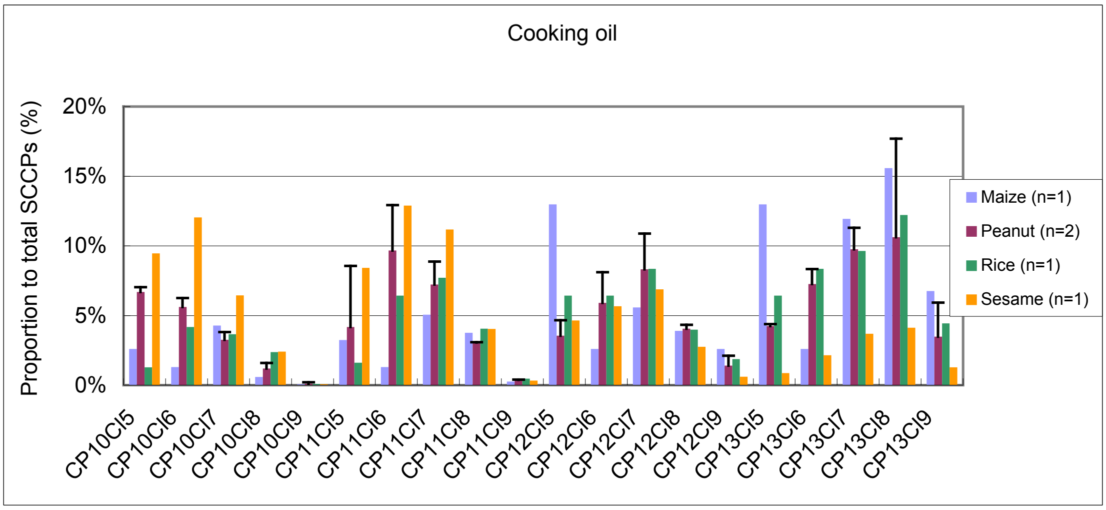
Table S4. Factor analysis of the short-chain chlorinated paraffins data

	Initial solution		Varimax rotated	
	F1	F2	F1	F2
Eigenvalue	15.49	3.15		
Contribution (%)	77.4	15.8		
Eigenvector				
$C_{10}H_{17}Cl_5$	**0.93**	0.15	**0.87**	0.38
$C_{10}H_{16}Cl_6$	**0.85**	0.44	**0.71**	0.64
$C_{10}H_{15}Cl_7$	0.54	**0.79**	0.32	**0.90**
$C_{10}H_{14}Cl_8$	0.22	**0.92**	-0.02	**0.95**
$C_{10}H_{13}Cl_9$	0.33	**0.84**	0.10	**0.90**
$C_{11}H_{19}Cl_5$	**1.00**	-0.03	**0.97**	0.23
$C_{11}H_{18}Cl_6$	**1.00**	0.00	**0.97**	0.25
$C_{11}H_{17}Cl_7$	**0.97**	0.21	**0.89**	0.45
$C_{11}H_{16}Cl_8$	**0.83**	0.51	0.68	**0.71**
$C_{11}H_{15}Cl_9$	0.61	**0.75**	0.40	**0.88**
$C_{12}H_{21}Cl_5$	**0.96**	-0.03	**0.94**	0.21
$C_{12}H_{20}Cl_6$	**0.97**	-0.07	**0.96**	0.18
$C_{12}H_{19}Cl_7$	**0.968**	-0.033	**0.945**	0.213
$C_{12}H_{18}Cl_8$	**0.929**	0.065	**0.882**	0.297
$C_{12}H_{17}Cl_9$	**0.787**	0.201	**0.710**	0.393
$C_{13}H_{23}Cl_5$	**0.959**	-0.133	**0.961**	0.114
$C_{13}H_{22}Cl_6$	**0.963**	-0.125	**0.964**	0.123
$C_{13}H_{21}Cl_7$	**0.969**	-0.102	**0.964**	0.146
$C_{13}H_{20}Cl_8$	**0.970**	-0.012	**0.942**	0.233
$C_{13}H_{19}Cl_9$	**0.926**	0.088	**0.873**	0.319

Factor score (mean±standard deviation [median])	F1	F2
Cooking oil (n=28)	0.26±1.34(-0.15)^{a}	0.06±0.78(-0.22)^{a}
Fried confectionery (n=20)	-0.28±0.21(-0.25)^{b}	0.01±1.32(-0.28)^{ab}
Raw seeds for cooking oil (n=5)	-0.25±0.01(-0.25)^{ab}	-0.37±0.05(-0.39)^{b}

The factors in bold indicate the most significant correlations.The median factor scores in the same columns but without the same superscripts differ significantly (Steel-Dwass test, p<0.05). The factor scores with the same superscripts or without superscripts do not differ significantly (p>0.05). F1: 1st factor; F2: 2nd factor

A



B

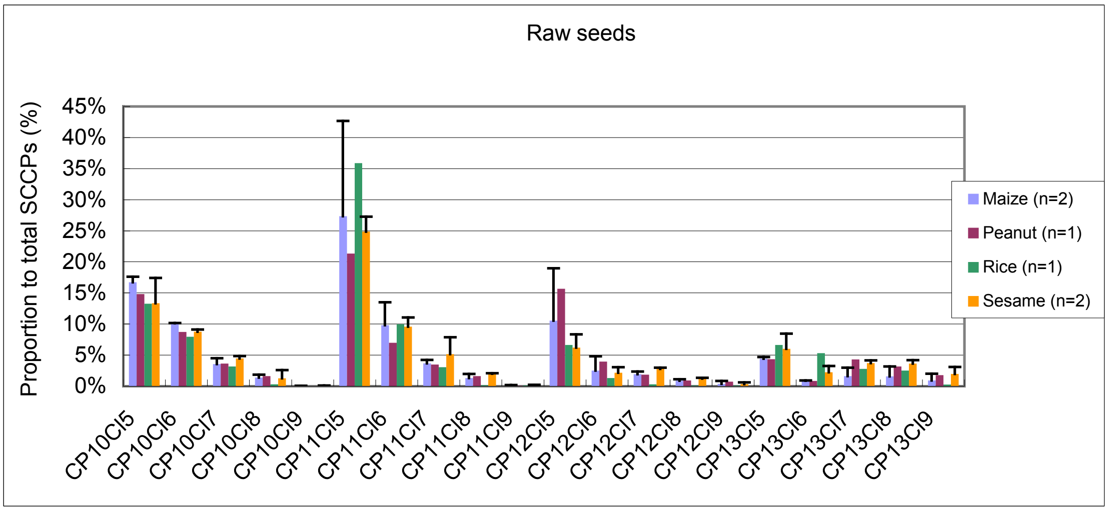


Figure S1. Contributions of the short-chain chlorinated paraffin (SCCP) homologs to the total SCCP concentrations in the (A) cooking oil samples and (B) raw seeds that are used to produce vegetable oil from Shenyang and Fushun. Only samples containing more than nine detectable SCCP homologs were selected (n=5 for cooking oil and n=6 for raw seeds). The contribution of each homolog to the total SCCP concentration was calculated for each sample separately. The bars indicate the means and the whiskers the standard deviations for groups with n≥2.