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Combined Effects of Dust and Dietary Exposure of Occupational Workers and Local Residents to Short- and Medium-Chain Chlorinated Paraffins in a Mega E-Waste Recycling Industrial Park in South China

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Supporting Information

ABSTRACT: Four types of dust samples and nine categories of locally produced staple foods were collected from a mega e-waste recycling industrial park and its surrounding regions, and simultaneously analyzed for short-chain and medium-chain chlorinated paraffins (CPs) to estimate dust and dietary exposure and their combined effects on occupational workers and local residents. All samples related to e-waste activities contained considerably high concentrations of CPs. The highest dust concentration was found in e-waste workshops. CPs were highly accumulated in local plant and animal origin foods, most markedly in fish, vegetables, and rice. The main contribution to CP intake under a median exposure scenario was from the diet, and vegetables, fish, and rice were the three largest dietary intake sources. Only the combined dust and food exposure from the present study has approached or even exceeded the highest tolerable daily intake (TDI) set up by the International Program on Chemical Safety (IPCS). However, due to lack of official threshold values for CP exposure on adverse human health, there are limitations on accurate risk assessment. Considering the presence of other exposure pathways, CPs' endocrine disrupter properties, as well as the multicomponent chemical "cocktails" effects, potential high risks from CP exposure may be posed to e-waste workers and local residents.

INTRODUCTION

Chlorinated paraffins (CPs) are a class of extremely complex technical mixtures produced and used in large amounts worldwide for several decades.[1] According to carbon chain length, CPs are divided into short chain (SCCPs, $C_{10\text{-}13}$), medium chain (MCCPs, C_{14-17}), and long chain chlorinated paraffins (LCCPs, $C_{>17}$). Due to their excellent thermal and chemical stability, over 200 CP formulations worldwide have been utilized in a large number of commercial and industrial applications, including use as flame retardants (FRs) and plasticizers in plastics, especially in polyvinyl chloride (PVC),[2] and as additives in metal-working fluids, rubber, paints, coating, and sealants.[3,4] China has been producing CPs since the 1950s and is the largest producer, consumer, and exporter in the world with a production capacity up to 1600 kilotons in 2014.[5] Of the CPs, SCCPs are classified as hazardous chemicals to the environment and are paid special attention, since they have persistence,[6−8] bioaccumulation abilities,[9−11] long-range transport potential,[12−14] and potential carcinogenic and other toxic effects.[15] In addition, SCCPs also have potential endocrine disrupter properties and have been regulated in the EU, Japan, USA, and Canada.[15,16] Widespread occurrences of SCCPs in various matrices in the world especially from the remote areas raise environmental concerns regarding the compound class in recent years.[12−14] In 2016, the Review Committee of Persistent Organic Pollutants (POPs) considered that SCCPs met the screening criteria of POPs,[15] and the eighth Conference of Parties of Stockholm Convention (SC) ultimately decided to list SCCPs in Annex A as new POPs in SC in May, 2017.[17] Relative to SCCPs, as concomitant pollutants, data on MCCPs in the environment and human exposure are very scarce. More information is urgently needed to confirm whether or not MCCPs should also be considered as a common concern.[18]

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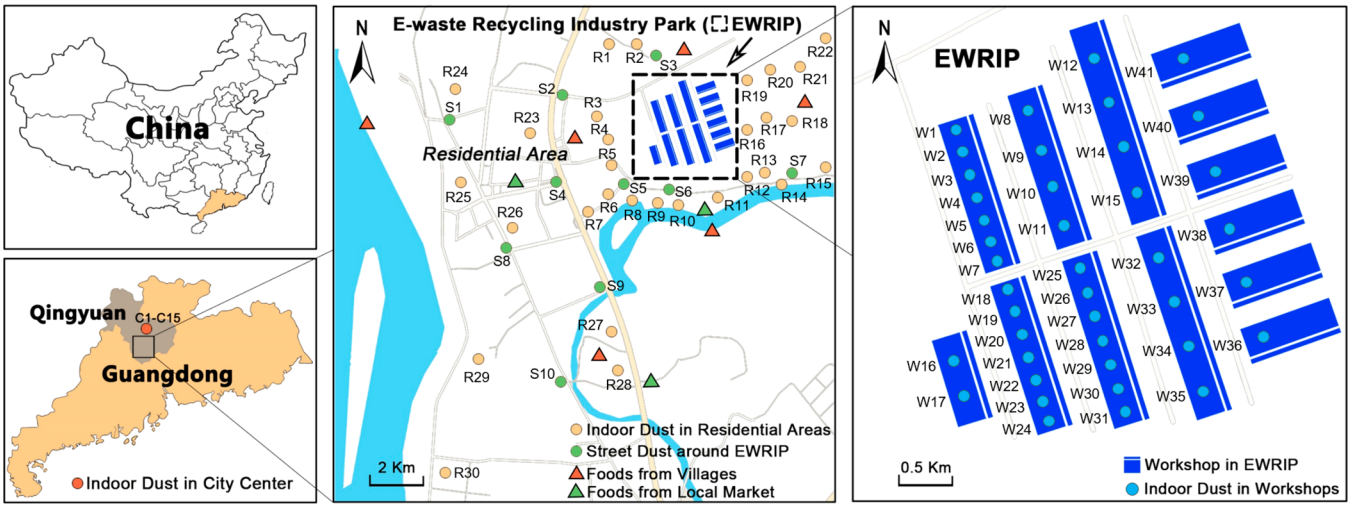

Figure 1. Map of the study areas (e-waste recycling cluster area, residential areas, exterior street surfaces, and city center) and sampling sites.

China is a major processor of e-waste, with both imported and domestic e-waste being important sources.[19] Although e-waste imports were restricted in recent years, a previous report[20] indicated that domestic sources in China overtook the U.S. as a source of e-waste. E-waste recycling has been extensively identified to be a main source of many toxic substances[19,21,22] such as polybrominated diphenyl ethers (PBDEs) and organophosphate esters, to surrounding environments that can result in severe pollution and health hazards. CPs are largely used as flame retardants and plasticizers in electronic PVC plastics. The latest study[23] reported that concentrations of SCCPs and MCCPs in PVC cables were found to be up to 191 mg g^{-1} and 145 mg g^{-1}, respectively. According to statistics, e-wastes contain approximately 30% plastics, and 12 million tons of plastic wastes are generated annually from e-products worldwide.[24,25] Rudimentary operations including unprotected manual dismantling and thermal treatment employed in the recycling of FR- and plasticizer-containing PVC e-waste inevitably result in extensive releases of CPs into the local environment. There are a few studies[26−31] indicating severe pollution of CPs in several e-waste recycling areas in China. Luo's group found that CPs can accumulate at high levels in terrestrial bird species,[26] home-produced eggs,[27,28] and typical freshwater food webs[29] from e-waste recycling areas in South China. Yuan et al.[30] reported relatively high levels of SCCPs present in soil and paddy seeds in an e-waste dismantling area in Eastern China. Collectively, Yuan's[30] and Luo's[26−29] studies implied that diet may be a potentially important human exposure pathway for CPs in e-waste recycling areas. Zeng et al.[31] reported elevated levels of SCCPs in surface particulates from workshop floors of e-waste recycling sites in China, implying that dust is a potentially significant human exposure pathway of CPs for occupational workers in e-waste processing facilities.

Two additional recent studies[32,33] indicated that CPs can be easily released from PVC plastics and transformed to more toxic POPs, such as polychlorinated biphenyls (PCBs) and polychlorinated naphthalenes (PCNs), during the thermal processes, which further raises environmental concerns over CPs at present. We speculate that thermochemical emission and transformation may occur in the thermal processes of e-waste recycling operations, which can facilitate the release of CPs from e-waste to dust and air in typical recycling workshops. The released CPs can be easily transported over a short-range and dispersed into surrounding multimedia,[34] and subsequently bioaccumulate in terrestrial agricultural food chains. Despite the substantial studies above indicating that serious environmental pollution by CPs in e-waste recycling areas of China is a result of e-waste dismantling, systematic research on human dust and dietary exposure to CPs around such sites has not been carried out.

External exposure assessments from recent research[35] have identified dust and dietary intake to be two major exposure pathways of SCCPs and MCCPs for the general population. To further explore human exposure to CPs in occupational environments and their effects on the natural environment, we carried out the most comprehensive dust and dietary intake assessments of CPs to date, from a mega e-waste recycling industrial park in China. Specific emphasis was placed on elucidating the differences between e-waste pollution and nonpollution areas. Four kinds of dust were collected from e-waste workshops, exterior street surfaces, local residential homes, and control sites. A large number of locally produced staple food items covering the main food consumption were collected from the villages around the investigated e-waste recycling industrial park. The samples were simultaneously analyzed for SCCPs and MCCPs to estimate dust and dietary exposures and their combined effects on occupational workers and local residents. The results are expected to fill in the current gaps relevant to the exposure of the occupational population and local population to CPs in e-waste recycling areas.

MATERIALS AND METHODS

Background of the Study Area. The study area (N 23.5°, E 113.0°) is located in Shijiao Town, Qingyuan City, Guangdong Province (Figure 1). Detailed information on the background of the study area is given in the Supporting Information (SI). The selected typical e-waste recycling industrial park is one of four newly established mega industrial parks in Qingyuan with electronic PVC plastics as the major recycling resource.

Sample Collection, Storage, Handling, and Preparation. A total of 81 dust samples and 92 locally produced staple food samples were collected from a mega e-waste recycling industrial park and its surrounding residential areas (Figure 1).

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Table 1. Overall Descriptive Statistics of SCCP and MCCP Concentrations in Dust Samples ($\mu g\ g^{-1}$, dw) from E-waste Recycling Workshops, Local Residential Homes, Exterior Street Surfaces, and Control Homes in Qingyuan City Center

sampling site	compound	range	mean	geomean	median	P95[a]
e-waste recycling workshops (n = 41)	∑SCCPs	246−19900	5600	3760	4180	18500
	∑MCCPs	874−48000	17800	13000	13900	44900
local residential homes (n = 30)	∑SCCPs	34.5−2030	580	370	412	1920
	∑MCCPs	79.2−6510	1760	1130	1250	5490
exterior street surfaces (n = 10)	∑SCCPs	32.4−982	501	359	516	898
	∑MCCPs	55.1−1570	772	567	767	1530
control homes (n = 15)	∑SCCPs	27.8−173	59.0	49.0	46.5	160
	∑MCCPs	74.0−539	185	162	166	351

[a]The 95th percentile.

Detailed information on sample collection, storage, handling, and preparation is presented in the SI.

Instrumental Analysis, Identification, and Quantification. SCCPs and MCCPs were simultaneously determined using a high-resolution gas chromatograph coupled to an electron capture negative ionization-low resolution mass spectrometer (HRGC/ECNI-LRMS, Agilent 7890B-7000D, USA) based on the previously developed method,[9−11,36,37] which has been further validated by GC-Q-TOF HRMS and obtains good comparability of results with a variation factor of 0.8−1.2. Detailed information with regard to instrumental analysis, identification, and quantification is given in the SI.

Quality Assurance and Quality Control (QA/QC). All glassware was soaked in a cleaning agent (decon90) for 12 h and then rinsed thoroughly with ultrapure water. After being carefully dried, the glassware was rinsed with solvent and then heated at 450 °C overnight prior to use. Each batch of eight samples was followed by two procedural blanks. Field blanks for dust were prepared by suction of anhydrous sodium sulfate with a vacuum cleaner in the same manner as collecting field dust samples. SCCPs and MCCPs in the procedure and field blanks contained <5% of the CP concentration in all the samples, and thus the concentrations of CPs were not blank corrected. In a recovery efficiency test, house dust samples from control homes and fish samples collected from a lake in the Tibetan Plateau, known to have very low levels of CPs, were selected as matrix-spiked samples. Recoveries of SCCPs (63.0% Cl) and MCCPs (57.0% Cl) in matrix-spiked samples with spiked concentrations of 0.1 and 1 $\mu g\ g^{-1}$ were 75−103% and 70−112%, respectively (Table S1). Precision was evaluated by analysis of matrix spike duplicates (n = 5), and the relative standard deviations (RSDs) in duplicate sample tests were ≤12%. The surrogate recoveries of ^{13}C-*trans*-chlordane in all field samples were 68% to 115%. The method detection limits (MDLs) for total SCCPs (∑SCCPs) and total MCCPs (∑MCCPs) were defined as three times the standard deviation of blank values. The MDLs in dust, food, and drinking water were estimated to be 110 ng g^{-1} dry weight (dw), 10 ng g^{-1} dw, 5.5 ng L^{-1} for ∑SCCPs, and 180 ng g^{-1} dw, 16 ng g^{-1} dw, and 8.6 ng L^{-1} for ∑MCCPs, respectively.

Estimation of Dust and Dietary Exposures and Their Combined Healthy Risk. For the estimation of total dust intake, two exposure pathways including dust ingestion and dermal absorption were both considered in this study. The estimated daily intake (EDI) via these two pathways for adults (occupational workers and residents) and children (residents) were calculated:[35,38−40]

$$\mathrm{EDI}_{\mathrm{ingestion}} = \sum \frac{C_{\mathrm{dust}} \times \mathrm{IR} \times \mathrm{EF}}{\mathrm{BW}}$$

$$\mathrm{EDI}_{\mathrm{dermal\ absorption}} = \sum \frac{C_{\mathrm{dust}} \times \mathrm{SA} \times \mathrm{AF} \times \mathrm{ABS} \times \mathrm{EF}}{\mathrm{BW}}$$

$$\mathrm{EDI}_{\mathrm{total\ dust}} = \mathrm{EDI}_{\mathrm{ingestion}} + \mathrm{EDI}_{\mathrm{dermal\ absorption}}$$

where $\mathrm{EDI}_{\mathrm{ingestion}}$ and $\mathrm{EDI}_{\mathrm{dermal\ absorption}}$ are the estimated daily intake via dust ingestion and dermal absorption, respectively ($\mu g\ kg^{-1}\ day^{-1}$), $\mathrm{EDI}_{\mathrm{total\ dust}}$ is the estimated total dust intake ($\mu g\ kg^{-1}\ day^{-1}$), C_{dust} is SCCP or MCCP concentration in dust samples in a certain kind of environment ($\mu g\ g^{-1}$, dry weight), IR is the daily ingestion rate of dust (mg day^{-1}), EF is the exposure fraction (hours spent over a day in a certain environment), SA is the surface area of the skin in contact with dust ($cm^2\ day^{-1}$), AF is the adherence factor of dust on skin (mg cm^{-2}), ABS is the dermal absorption fraction, and BW is the body weight for adults or children (kg). The values of IR and AF were estimated to be at least two times higher for occupational workers than the general population (local residents). All the parameters used in the calculation of dust intake for adults and children are summarized in Table S2, and the daily activity pattern and the time spent per day by occupational workers and local residents in different kinds of environment are presented in Table S3 according to our survey.

Total dietary intake was calculated assuming that occupational workers and local residents of the investigated e-waste area derived all their intake of food from local sources. This assumption was based on the information from questionnaires completed by occupational workers and local residents during the sampling period. In the questionnaires, local residents confirmed that most if not all of their produce was either consumed by themselves or sold for consumption within their own local communities, and occupational workers confirmed that their food consumption pattern was the same as that of local residents. The average daily consumption rates of each type of food for adults and children are shown in Table S4 based on information from the questionnaires and published data. Dietary exposure was calculated:[19,41]

$$\mathrm{EDI}_{\mathrm{total\ diet}} = \sum_{i=1}^{10} \left(\frac{\mathrm{Ci} \times \mathrm{CRi}}{\mathrm{BW}} \right)$$

where $\mathrm{EDI}_{\mathrm{total\ diet}}$ is the estimated total dietary intake ($\mu g\ kg^{-1}\ day^{-1}$), Ci is SCCP or MCCP concentration in each food group ($\mu g\ g^{-1}$, wet weight), and CRi is the daily consumption rate of each food group and drinking water (g day^{-1}).

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Table 2. Overall Descriptive Statistics of SCCP and MCCP Concentrations in Local Food (in $\mu g\ g^{-1}$, lw and in Parentheses, $\mu g\ g^{-1}$, ww) and Drinking Water Samples (ng L^{-1}) from the E-waste Recycling Area

food group	food item	compound	range	mean	median
fish	mrigal carp	∑SCCPs	285−487	408 (3.46)	416 (3.54)
	(n = 5)[a]	∑MCCPs	284−563	443 (3.76)	458 (3.89)
	yellow catfish	∑SCCPs	94.3−148	118 (5.64)	114 (5.54)
	(n = 5)	∑MCCPs	116−156	134 (6.51)	131 (6.50)
	white amur bream	∑SCCPs	113−163	132 (1.57)	119 (1.65)
	(n = 5)	∑MCCPs	116−172	141 (1.68)	135 (1.86)
shellfish	Asian clam	∑SCCPs	37.1−66.9	49.8 (1.11)	45.5 (1.06)
	(p = 3)[b]	∑MCCPs	35.5−59.9	46.6 (1.04)	44.5 (1.03)
shrimp	river prawn	∑SCCPs	76.5−118	94.3 (0.898)	87.9 (1.01)
	(p = 3)	∑MCCPs	102−133	114 (1.12)	107 (1.42)
meat	pork	∑SCCPs	37.2−58.6	49.2 (0.881)	50.3 (0.766)
	(n = 5)	∑MCCPs	48.3−69.3	58.6 (1.05)	58.8 (0.926)
poultry	chicken	∑SCCPs	25.3−84.2	43.4 (2.71)	32.0 (2.06)
	(n = 5)	∑MCCPs	31.9−72.0	46.9 (2.92)	41.8 (2.70)
	duck	∑SCCPs	40.7−88.3	66.1 (2.03)	67.7 (1.90)
	(n = 5)	∑MCCPs	48.9−111	73.9 (2.24)	67.7 (1.79)
egg	chicken egg	∑SCCPs	3.37−6.84	4.84 (0.412)	4.94 (0.391)
	(n = 10)	∑MCCPs	5.29−10.6	7.96 (0.687)	7.95 (0.684)
	duck egg	∑SCCPs	1.62−2.91	2.04 (0.267)	1.95 (0.245)
	(n = 10)	∑MCCPs	2.07−4.26	3.01 (0.399)	2.93 (0.398)
vegetable	mustard	∑SCCPs	132−167	148 (0.466)	145 (0.457)
	(p = 3)	∑MCCPs	245−331	291 (0.915)	297 (0.937)
	lettuce	∑SCCPs	493−541	519 (3.54)	524 (3.56)
	(p = 3)	∑MCCPs	582−690	635 (4.36)	633 (4.31)
	sweet potato	∑SCCPs	247−263	256 (1.63)	258 (1.65)
	(p = 3)	∑MCCPs	296−322	305 (1.95)	299 (1.90)
	Chinese radish	∑SCCPs	236−605	402 (0.661)	394 (0.657)
	(p = 3)	∑MCCPs	383−747	547 (0.885)	512 (0.853)
cereal	rice	∑SCCPs	37.2−88.6	62.8 (0.397)	67.4 (0.379)
	(n = 8)	∑MCCPs	55.4−108	87.8 (0.543)	91.4 (0.564)
culinary oil	peanut oil	∑SCCPs	0.892−1.32	1.10	1.09
	(n = 5)	∑MCCPs	1.22−1.76	1.46	1.42
drinking water	well water	∑SCCPs	54.2−80.4	67.2	68.1
	(n = 6)	∑MCCPs	42.3−96.5	63.1	59.9
	tap water	∑SCCPs	29.6−45.3	37.2	39.0
	(n = 5)	∑MCCPs	23.7−40.8	31.0	32.5

[a] n = number of individual samples. [b] p = number of pooled samples.

Two scenarios were considered for the exposure assessment via combined dust and dietary intakes. Median and high-end estimates of daily ∑SCCP or ∑MCCP intakes were calculated based on the 50th and 95th percentile (P95) concentrations in both dust and food samples.[19] The total exposure dose (EDI_{total}) was the sum of $EDI_{total\ dust}$ and $EDI_{total\ diet}$. Hazard quotients (HQ) were applied to assess the combined health risks, calculated by the following equation:[39,42]

$$HQ = \frac{EDI_{total}}{TDI}$$

where TDI is the tolerable daily intake (TDI) of target contaminations. A TDI of 100 $\mu g\ kg^{-1}\ day^{-1}$ for SCCPs and MCCPs recommended by the International Program on Chemical Safety (IPCS) was adopted in this study.[43]

RESULTS AND DISCUSSION

Concentrations and Homologue Profiles of SCCPs and MCCPs in Dust. Descriptive statistics of ∑SCCP and ∑MCCP concentrations (range, mean, geomean, median, and P95) in four kinds of dust collected from e-waste recycling workshops in the industrial park, local residential homes around the industrial park, exterior street surfaces, and control homes are summarized in Table 1. SCCPs and MCCPs were detected in all the dust samples, and both of their concentrations exhibited the order of e-waste workshops > surrounding residential homes > street surfaces > control homes. Figure S1 in the Supporting Information presents the box-whisker-plots of ∑SCCPs and ∑MCCPs in the four kinds of dust samples. The concentrations of SCCPs and MCCPs detected in dust from the e-waste workshops ranged from 246 to 19 900 (mean: 5600) $\mu g\ g^{-1}$ and 874−48000 (mean: 17 800) $\mu g\ g^{-1}$, respectively. The maximum values of 19 900 $\mu g\ g^{-1}$ for ∑SCCPs and 48 000 $\mu g\ g^{-1}$ for ∑MCCPs in the present study were found to be the highest records of CP dust concentrations among all the published data to date. Dust concentrations of SCCPs and MCCPs from the surrounding residential homes near the park were in the range of 34.5−2030 (mean: 580) $\mu g\ g^{-1}$ and 79.2−6510 (mean: 1760) $\mu g\ g^{-1}$, respectively, which were about 10 times lower than the e-waste workshops, but 10 times higher than control homes from Qingyuan city center (SCCPs: range of 27.8−173 $\mu g\ g^{-1}$ with

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a mean of 59.0 $\mu g\ g^{-1}$ and MCCPs: range of 74.0−539 $\mu g\ g^{-1}$ with a mean of 185 $\mu g\ g^{-1}$). Dust concentrations of SCCPs and MCCPs from exterior street surfaces ranged from 32.4 to 982 (mean: 501) $\mu g\ g^{-1}$ and 55.1−1570 (mean: 772) $\mu g\ g^{-1}$, respectively, which were comparable or slightly lower than those of local residential homes.

Comparison of SCCP and MCCP concentrations in dust reported here with those from other regions of China and developed countries worldwide is summarized in Table S5. The mean concentrations of SCCPs and MCCPs in indoor dust from local residential homes in the e-waste recycling area were between 2 and 10 times higher than those from various indoor environments in other regions of China, for example, Harbin (SCCPs: 53.6 $\mu g\ g^{-1}$),[39] Beijing (SCCPs and MCCPs: 148 and 139 $\mu g\ g^{-1}$),[35] and Dalian (SCCPs and MCCPs: 227 and 205 $\mu g\ g^{-1}$),[40] and several to several dozen times higher than developed countries including Sweden (SCCPs and MCCPs: 6.7 and 308 $\mu g\ g^{-1}$),[3,44] Germany (8.5 and 229 $\mu g\ g^{-1}$),[4] the UK (92.7 and 463 $\mu g\ g^{-1}$),[44] Canada (49.3 and 176 $\mu g\ g^{-1}$),[44] and Australia (61.2 and 180 $\mu g\ g^{-1}$).[44] The dust concentrations of SCCPs and MCCPs from households in Qingyuan city center as control homes in this study are comparable to those reported in Harbin,[39] Beijing,[35] and Dalian.[40] In all dust samples, MCCPs exhibited 2−3 times higher levels than SCCPs. A significant positive relationship was observed between ∑SCCPs and ∑MCCPs (R^2 = 0.72, p < 0.01), consistent with the findings in sediment and biota from our previous studies.[10,11,37]

The homologue profiles and congener group abundance profiles of SCCPs and MCCPs in four kinds of dust samples are illustrated in Figures S2 and S3, respectively. C_{13} was the predominant homologue group within SCCPs in all dust samples, accounting for 40% or more of the total composition of SCCPs, followed by $C_{12} > C_{11} > C_{10}$. An obviously higher proportion of C_{13} group was found in dust from the workshops and surrounding homes than exterior street surfaces and control homes. The high proportion of C_{13} (∼40%) found in indoor dust from exterior street surfaces and control homes is in agreement with the reported results (∼35% or more) in indoor dust reported in three previous studies conducted in China.[35,39,40] However, the higher proportion of C_{13} (49−51%) in the dust samples found in the workshops and surrounding household areas is more similar to the composition of C_{13} (∼75%) detected in a commercial CP52 technical mixture, which is widely applied as flame retardants and plasticizers in electronic PVC plastics in China.[35] MCCPs shared a similar homologue profile in all dust samples with C_{14} as the most abundant group. This distribution pattern is similar to the results from other studies.[3,4,35,39,40] The high proportion of C_{14} is also consistent with the composition of the commercial CP52 technical mixture.[35] In addition, similar to CP52, Cl_{7-8} congener groups predominated in both SCCP and MCCP compositions. Statistical analysis of significance test in the homologue distribution pattern between dust samples and the CP52 technical mixture indicated no significant difference using the t-test (p > 0.05). Therefore, on the basis of the analytical results, we infer that SCCPs and MCCPs in dust samples mainly originated from more emissions of the CP52 technical mixture.

Concentrations and Homologue Profiles of SCCPs and MCCPs in Local Staple Foods. Descriptive statistics of ∑SCCP and ∑MCCP concentrations (mean, median, and range) in locally produced staple food and drinking water samples are summarized in Table 2. To enable comparison with previously obtained data, ∑SCCPs and ∑MCCPs are reported in Table 2 and illustrated in Figure S4 on both a lipid weight (lw) and wet weight (ww) basis. The corresponding dry weight concentrations are presented in Table S6. On the basis of available limited data about a few food items reported previously, a comparison of CP concentrations in aquatic products, meat products, and eggs originating from the e-waste recycling areas with those reported from other countries and regions of China is summarized in Supporting Information, Table S7. In all food samples, comparable or higher levels of MCCPs than SCCPs can be observed with the ratios (MCCPs/SCCPs) of 1−2, lower than the ratios of 2−3 in dust samples. Significant positive relationships also exist between lipid-normalized ∑SCCPs and ∑MCCPs (R^2 = 0.81, p < 0.01), indicating the common sources and similar accumulation.

As presented in Table 2, average or median ∑SCCPs and ∑MCCPs expressed on a lipid weight basis were highest in the samples of vegetable, followed by the samples of fish, shrimp, rice, duck, pork, shellfish, chicken, and egg. On a wet weight basis, however, fish exhibited the highest average or median concentrations, followed by chicken, duck, vegetable, pork, shellfish, shrimp, egg, and rice. The maximum ∑SCCP and ∑MCCP lipid weight concentrations were recorded in vegetables, while the maximum wet weight concentration was found in fish.

Fish, Shellfish, and Shrimp. Among three local fish species, the highest lipid weight-average concentrations of ∑SCCPs and ∑MCCPs was detected in mrigal carp at 408 and 443 $\mu g\ g^{-1}$ lw, followed by white amur bream and yellow catfish, while the highest wet weight-average concentrations of ∑SCCPs and ∑MCCPs were found in yellow catfish at 5.64 and 6.51 $\mu g\ g^{-1}$ ww. The concentrations were found to be the highest accumulation levels to date compared to all previously reported data in fish (Table S7), indicating that fish produced locally within the vicinity of e-waste recycling industrial park have been heavily contaminated with CPs. The concentrations of SCCPs and MCCPs are several to several dozen times higher than previously reported for fish from non-e-waste related regions of China, but about 3 orders of magnitude higher than values reported previously for marine or freshwater fish in Europe,[45] northern America,[46] and Hong Kong.[11] Chan et al.[22] and Labunska et al.[19] reported elevated accumulations of PBDEs in food, especially in fish, at e-waste recycling sites compared to non-e-waste sites, which were well in line with our present findings for CPs.

Concentrations in our shellfish samples exceeded, by a considerable margin, those in mollusks from the Chinese Bohai Sea[47] and Pearl River Estuary,[48] similar to those from Dianshan Lake, China.[49] Concentrations in river prawns from the e-waste recycling area exceeded by 1 order of magnitude those in shrimps from the Pearl River Estuary, China,[48] and were twice those in shrimps from Dianshan Lake, China.[49]

Pork, Chicken, Duck, and Egg. Average ∑SCCP and ∑MCCP lipid weight concentrations were 49.2 and 58.6 $\mu g\ g^{-1}$ lw for pork, 43.4 and 46.9 $\mu g\ g^{-1}$ lw for chicken, and 66.1 and 73.9 $\mu g\ g^{-1}$ lw for duck, while higher wet weight concentrations were found in chicken than duck and pork (Table 2). Until now, no information exists on concentrations of SCCPs and MCCPs in pork, chicken, and duck. In comparison to the only one study on composited mean

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Table 3. Estimated Daily Intake (EDI, $\mu g\ kg^{-1}\ day^{-1}$) of SCCPs and MCCPs via Food Consumption and Dust Intake for Occupational Workers and Local Residents (Adult and Children) in a Mega E-waste Recycling Industrial Park in Qingyuan, South China

	occupational workers				local residents (adult)				local residents (children)			
	SCCPs		MCCPs		SCCPs		MCCPs		SCCPs		MCCPs	
	median[a]	high[b]	median	high	median	high	median	high	median	high	median	high
fish	2.38	2.73	2.72	3.20	2.38	2.73	2.72	3.20	4.13	4.72	4.71	5.54
shellfish	0.124	0.155	0.121	0.140	0.124	0.155	0.121	0.140	0.209	0.262	0.205	0.236
shrimp	0.119	0.143	0.167	0.168	0.119	0.143	0.167	0.168	0.201	0.240	0.282	0.283
meat	0.998	1.83	1.21	2.15	0.998	1.83	1.21	2.15	3.18	5.85	3.84	6.84
poultry	0.97	1.96	1.10	1.97	0.97	1.96	1.10	1.97	3.11	6.25	3.53	6.29
egg	0.151	0.214	0.258	0.352	0.151	0.214	0.258	0.352	0.651	0.922	1.11	1.51
vegetables	7.95	9.37	10.1	12.2	7.95	9.37	10.1	12.2	17.3	20.3	21.8	26.5
cereal	2.26	3.40	3.37	4.11	2.26	3.40	3.37	4.11	4.30	6.45	6.39	7.80
culinary oil	0.393	0.472	0.512	0.619	0.393	0.472	0.512	0.619	1.09	1.31	1.43	1.72
drinking water	0.002	0.002	0.001	0.002	0.002	0.002	0.001	0.002	0.003	0.003	0.002	0.003
$EDI_{total\ diet}$	15.4	20.3	19.5	24.9	15.4	20.3	19.5	24.9	34.1	46.4	43.3	56.8
dust ingestion	1.62	7.13	5.31	17.6	0.21	0.80	0.54	2.32	1.79	6.85	4.64	20.0
dermal absorption	3.02	13.3	9.89	32.7	0.39	1.48	1.00	4.32	2.34	8.95	6.06	26.1
$EDI_{total\ dust}$	4.64	20.4	15.2	50.3	0.60	2.28	1.54	6.64	4.13	15.8	10.7	46.0
EDI_{total}	20.0	40.7	34.7	75.2	16.0	22.6	21.1	31.6	38.3	62.2	54	103

[a]Median exposure, the 50th percentile. [b]High end exposure, the 95th percentile.

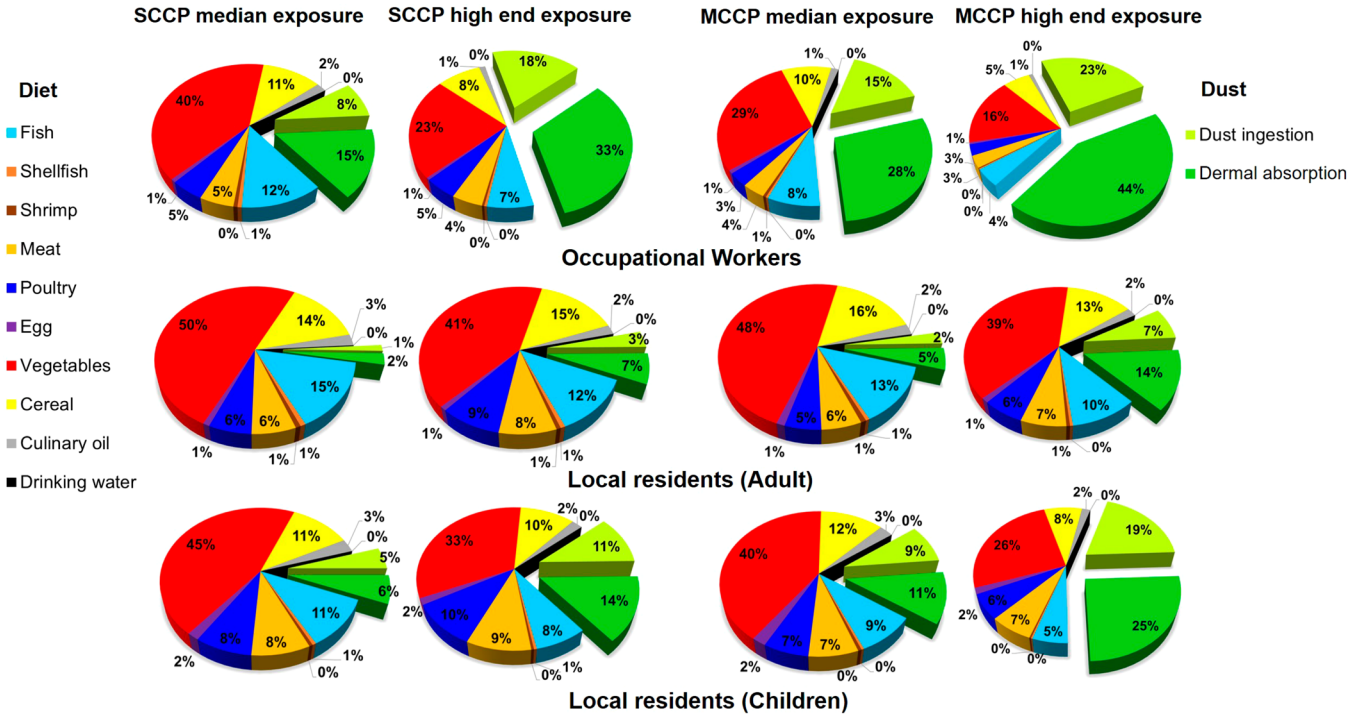

Figure 2. Percentage contribution of food groups, drinking water, and dust intake to total daily exposure dose of SCCPs and MCCPs.

products from 20 Chinese provinces, our current data on pork, chicken, and duck from the e-waste recycling area exceeded by 2 orders of magnitude the national average.[50] Average concentrations of ∑SCCPs and ∑MCCPs were 4.84 and 7.96 $\mu g\ g^{-1}$ lw for chicken eggs and 2.04 and 3.01 $\mu g\ g^{-1}$ lw for duck eggs. The accumulation levels in chicken eggs in the present study are similar to those in home-produced eggs from another e-waste polluted area of Qingyuan, China.[27,28]

Culinary Oil and Drinking Water. The peanut oil that was not produced locally contained relatively low concentrations of CPs compared to other local foods analyzed, but were still at the high end of the concentration range of CPs in cooking oil in China.[51] ∑SCCPs and ∑MCCPs in local well water were evidently higher than in local tap water, and were 2−4 times higher than those in drinking water from Beijing, China.[35]

Vegetables and Rice. This is the first report on highly accumulated concentrations of CPs in vegetables. The highest ∑SCCP and ∑MCCP concentrations were recorded in local lettuce with 3.54 and 4.36 $\mu g\ g^{-1}$ ww, respectively, followed by sweet potato, Chinese radish, and mustard. There are no reported concentrations of CPs in vegetables to date that can be used for comparison. Average ∑SCCPs and ∑MCCPs in

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rice were detected of 0.397 and 0.543 $\mu g\ g^{-1}$ ww, respectively. The concentrations of SCCPs exceeded by 1 order of magnitude those in rice from e-waste recycling sites in Taizhou, China.[30] Except for intake from soils and pore water, vegetation can accumulate CPs through both particulate-bound and gaseous depositions from the polluted atmosphere, which is likely to be responsible for the high accumulation of CPs in vegetables and rice.

In view of no regulatory limit of CPs in food at present, the measured concentrations cannot be directly compared and assessed. However, the highly elevated accumulation of CPs found in food from contaminated areas should be paid special attention, and the related policies/criteria are urgently needed in order to safeguard human health.

The homologue and congener group abundance profiles of SCCPs and MCCPs in food samples are illustrated in Figures S5−6 and S7−S8, respectively. In animal origin foods, C_{10}, C_{11}, C_{12}, and C_{13} shared an almost equal contribution to ∑SCCPs except for chicken and duck eggs, which was different from the distribution pattern in dust. In plant origin foods, species-specific accumulation profiles of SCCPs can be observed. For lettuce, sweet potato, and rice, SCCP congener group abundance profiles also showed a comparable contribution for four carbon chain groups. However, for Chinese radish and mustard, SCCP congener group abundance profiles were predominated by long-chain C_{13}, which was roughly similar to the distribution pattern in dust. MCCP homologue profiles in most food samples were still characterized by C_{14} as the main group, but with higher proportions of C_{15-17} compared to dust samples. Congeners with Cl_{7-8} and Cl_{8-10} groups predominated in the composition of SCCPs and MCCPs, respectively. Some discrepancies in the homologue pattern between dust and food samples may imply differential bioavailability of individual CP homologues and their species-specific bioaccumulation or biotransformation mechanisms, which merits further investigation in the near future.

Estimation of Daily Dust and Dietary Exposure and Their Combined Health Risk. On the basis of the SCCP and MCCP concentrations in different dust and food categories, EDIs were estimated under two scenarios for occupational population and local population in/around the mega e-waste recycling industrial park. Table 3 presents the comprehensive estimates of daily median and high-end exposure to SCCPs and MCCPs via dust and dietary intake for occupational workers and local residents (adults and children). Figure 2 illustrates the percentage contributions of food groups, drinking water, and dust intake to total daily exposure dose.

Estimated total median exposures to SCCPs via combined diet and dust intake were 20.0, 16.0, and 38.3 $\mu g\ kg^{-1}\ day^{-1}$ for e-waste workers, local adults and children, respectively. Total median ∑SCCP exposure for occupational workers was 1.25 times higher than that for local adults, but much lower than that for local children, indicating that children are more susceptible to CP exposure compared to workers in e-waste recycling area. The principal contribution to total median ∑SCCP exposure originated from diet rather than dust, but a significantly higher percentage contribution of dust could be observed for occupational workers than local adult residents and children. As shown in Figure 2, vegetables, fish, and rice were the three largest contributors to total dietary intake, which were identified as the main sources of dietary exposure, followed by poultry and meat. Shellfish, shrimp, eggs, and culinary oil exhibited relatively low contributions. Drinking water intake was almost negligible compared to food groups due to the low water solubility of CPs. Our median dietary exposure to SCCPs for local adult residents (15.4 $\mu g\ kg^{-1}\ day^{-1}$) was up to 26 times higher than that for the general population in Beijing,[35] and over 2 orders of magnitude higher than the dietary exposure level in Japan.[52,53] Regarding total dust intake, dermal absorption exhibited a greater contribution ratio than ingestion. Our median dust exposure to SCCPs for local adult residents (0.60 $\mu g\ kg^{-1}\ day^{-1}$) was 3−13 times higher than indoor dust exposure for the general population in Beijing,[35] Dalian,[40] and Harbin, China,[39] and exceeded by 1 order of magnitude what was reported in Stockholm, Sweden.[3] Taking combined dust and dietary exposures into account, total median ∑SCCP exposure for local adult residents in our present study was around 20 times higher than a previous estimate for the general population in Beijing, China.[35] If high-end exposure is considered, the difference in estimated SCCP exposures between our study and the previous study increased further. High-end exposures to SCCPs via combined diet and dust intake were 40.7, 22.6, and 62.2 $\mu g\ kg^{-1}\ day^{-1}$ for occupational workers, local adults, and children, respectively, 1.4−2 times higher than median exposure. Under a SCCP high-end exposure scenario, relative contribution of dust to total intake significantly increased, and most markedly for occupational workers, in which case dust contributed to more than 50% of total exposure and was the main source.

Compared with SCCP exposure, MCCP exposure receives more contribution from dust intake due to the higher dust concentrations. Estimated median exposure to MCCPs via combined diet and dust intake were 34.7, 21.1, and 54 $\mu g\ kg^{-1}\ day^{-1}$ for e-waste workers, local adults, and children, respectively, which were 1.3−1.7 times higher than the corresponding median exposure to SCCPs. Children exhibited substantially higher exposure than workers and adults, further supporting that children are more susceptible to CP exposure. Median MCCP exposure for local adults was around 25 times higher than that for the general population in Beijing, China.[35] Under a MCCP high-end exposure scenario, the exposure levels doubled those of the corresponding median exposure. It is especially notable that dust was the predominant exposure source of MCCPs for occupational workers contributing 77% to total exposure.

The International Programme on Chemical Safety (IPCS) proposed that a TDI for SCCPs, MCCPs and LCCPs was 100 $\mu g\ kg^{-1}\ day^{-1}$.[43] On the basis of the TDI, the combined health risk was evaluated by the hazard quotient (HQ) under median and high-end exposure scenarios. As shown in Figure 3, the HQ values ranged from 0.16 to 1.03, indicating a potential health risk for occupational workers and local residents. Significantly higher health risks were found to be present in local children and occupational workers than in local adult residents. Especially for local children, a HQ value of 1.03 indicated that the current MCCP high-end exposure for children exceeds the TDI value and poses a high health risk to them. All other HQs are close to 1, implying that the combined dust and food exposures (EDI_{total}: 16.0−75.2 $\mu g\ kg^{-1}\ day^{-1}$, Table 3) for e-waste workers and local residents are approaching the TDI proposed by IPCS, which should be a matter of concern. The Environment Canada reported a much lower TDI of 10 and 6 $\mu g\ kg^{-1}\ day^{-1}$ for SCCPs and MCCPs, respectively.[54] If these TDI values were used for comparison, all the combined exposures would substantially exceed these TDI values and pose high health risks to the occupational

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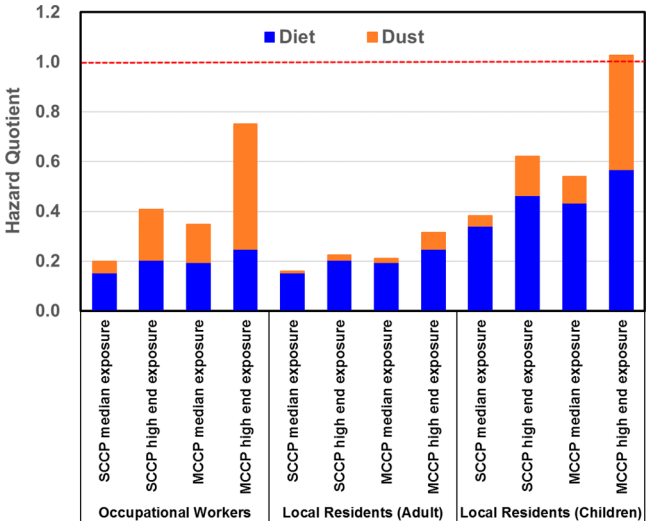

Figure 3. Hazard quotient of occupational workers and local residents' exposure to SCCPs and MCCPs in the e-waste recycling area.

workers and local residents. However, it should be noted that, because of the lack of the official threshold values for assessing human health effect due to CP exposure, it is hard to conduct a comprehensive assessment on health risk due to the exposure to CPs. In addition, considering that SCCPs are potential endocrine disrupting chemicals, standard toxicity tests would fail to detect low-dose effects and non-monotonic dose response.[55] Therefore, there are limitations on the risk assessment by the comparison only with the outdated TDI values.

In the present study, if air inhalation was included in the combined exposure assessment, the health risks would increase further. On the other side, if considering the synergistic effect or combined toxicity of SCCPs and MCCPs due to their similar physicochemical properties, the HQs would double at least. Moreover, if taking into account the multicomponent chemical "cocktails" effects as well, the HQs may elevate more significantly and pose a substantial risk to the target group. Therefore, our results highlight the potential for adverse human health impacts arising from exposure to CPs at e-waste recycling areas and is of a currently great concern. With more frequent use and shorter lifecycle of e-products nowadays, e-waste recycling is highly relevant to the circular economy to achieve resource recycling. The presence of hazardous chemicals including CPs in raw electronic materials or products will end up in e-wastes to be recycled, which will pose a risk to e-waste workers and contaminate more items in the future market. Thus, source control and labeling of hazardous chemicals need to be further strengthened and implemented. Environmental risk management is urgently needed to reduce the potential adverse impacts on the environment and human health. Further investigations with greater extensive monitoring of local food contamination and human body burdens of CPs, as well as long-term health outcomes on the exposed population, should be conducted in the future.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b02625.

Additional information on sample extraction and cleanup; analysis and quantification; percent recoveries (Table S1); parameters and data for EDI calculations and risk assessment (Tables S2−S4); concentration comparison in dust samples worldwide (Table S5); lipid content, water content, and dry weight concentrations in foods (Table S6); concentration comparison in some food categories worldwide (Table S7); box-whisker-plots of CP concentrations in dust (Figure S1); homologue profiles in dust (Figures S2 and S3); statistical histogram of CP concentrations in food (Figure S4); and homologue profiles in food (Figures S5−S8) (PDF)

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The authors declare no competing financial interest.

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■ REFERENCES

(1) Fiedler, H. Short-chain chlorinated paraffins: production, use and international regulations. In *Chlorinated Paraffins. The Handbook of Environmental Chemistry*; de Boer, J., Ed.; Springer-Verlag: Berlin, Heidelberg, 2010.

(2) Zhang, B.; Zhao, B.; Xu, C.; Zhang, J. Emission inventory and provincial distribution of short-chain chlorinated paraffins in China. *Sci. Total Environ.* **2017**, *581-582*, 582−588.

(3) Friden, U. E.; McLachlan, M. S.; Berger, U. Chlorinated paraffins in indoor air and dust: Concentrations, congener patterns, and human exposure. *Environ. Int.* **2011**, *37* (7), 1169−1174.

(4) Hilger, B.; Fromme, H.; Völkel, W.; Coelhan, M. Occurrence of chlorinated paraffins in house dust samples from Bavaria, Germany. *Environ. Pollut.* **2013**, *175*, 16−21.

(5) World Chlorine Council. *International Chlorinated Alkanes Industry Association (ICAIA) Newsletter*, September 2014. http://www.eurochlor.org/media/88258/20140908_icaia_newsletter_03_final.pdf.

(6) Iozza, S.; Muller, C. E.; Schmid, P.; Bogdal, C.; Oehme, M. Historical profiles of chlorinated paraffins and polychlorinated biphenyls in a dated sediment core from Lake Thun (Switzerland). *Environ. Sci. Technol.* **2008**, *42* (4), 1045−1050.

(7) Zeng, L. X.; Zhao, Z. S.; Li, H. J.; Han, W. Y.; Liu, Q.; Xiao, K.; Du, Y. G.; Wang, Y. W.; Jiang, G. B. Distribution of Short Chain Chlorinated Paraffins in Marine Sediments of the East China Sea: Influencing Factors, Transport and Implications. *Environ. Sci. Technol.* **2012**, *46* (18), 9898−9906.

(8) Zeng, L. X.; Chen, R.; Zhao, Z. S.; Wang, T.; Gao, Y.; Li, A.; Wang, Y. W.; Jiang, G. B.; Sun, L. G. Spatial Distributions and Deposition Chronology of Short Chain Chlorinated Paraffins in

DOI: 10.1021/acs.est.8b02625
Environ. Sci. Technol. 2018, 52, 11510−11519

Marine Sediments across the Chinese Bohai and Yellow Seas. *Environ. Sci. Technol.* **2013**, *47*, 11449−11456.
(9) Zeng, L. X.; Wang, T.; Wang, P.; Liu, Q.; Han, S. L.; Yuan, B.; Zhu, N. L.; Wang, Y. W.; Jiang, G. B. Distribution and Trophic Transfer of Short-Chain Chlorinated Paraffins in an Aquatic Ecosystem Receiving Effluents from a Sewage Treatment Plant. *Environ. Sci. Technol.* **2011**, *45* (13), 5529−5535.
(10) Zeng, L. X.; Lam, J. C. W.; Wang, Y. W.; Jiang, G. B.; Lam, P. K. S. Temporal Trends and Pattern Changes of Short- and Medium-Chain Chlorinated Paraffins in Marine Mammals from the South China Sea over the Past Decade. *Environ. Sci. Technol.* **2015**, *49* (19), 11348−11355.
(11) Zeng, L. X.; Lam, J. C. W.; Chen, H.; Du, B. B.; Leung, K. M. Y.; Lam, P. K. S. Tracking Dietary Sources of Short- and Medium-Chain Chlorinated Paraffins in Marine Mammals through a Subtropical Marine Food Web. *Environ. Sci. Technol.* **2017**, *51* (17), 9543−9552.
(12) Tomy, G. T.; Muir, D. C. G.; Stern, G. A.; Westmore, J. B. Levels of C_{10}-C_{13} polychloro-n-alkanes in marine mammals from the Arctic and the St. Lawrence River estuary. *Environ. Sci. Technol.* **2000**, *34* (9), 1615−1619.
(13) Wu, J.; Gao, W.; Liang, Y.; Fu, J.; Gao, Y.; Wang, Y.; Jiang, G. Spatiotemporal Distribution and Alpine Behavior of Short Chain Chlorinated Paraffins in Air at Shergyla Mountain and Lhasa on the Tibetan Plateau of China. *Environ. Sci. Technol.* **2017**, *51* (19), 11136−11144.
(14) Letcher, R. J.; Morris, A. D.; Dyck, M.; Sverko, E.; Reiner, E. J.; Blair, D. A. D.; Chu, S. G.; Shen, L. Legacy and new halogenated persistent organic pollutants in polar bears from a contamination hotspot in the Arctic, Hudson Bay Canada. *Sci. Total Environ.* **2018**, *610-611*, 121−136.
(15) *Report of the Persistent Organic Pollutants Review Committee on the Work of Its Twelfth Meeting: Risk Management Evaluation on Short-Chain Chlorinated Paraffins*, UNEP/POPS/POPRC.12/11/Add.3; Rome, September 2016.
(16) Kobeticova, K.; Cerny, R. Ecotoxicity assessment of short- and medium-chain chlorinated paraffins used in polyvinyl-chloride products for construction industry. *Sci. Total Environ.* **2018**, *640−641*, 523−528.
(17) UNEP, *Recommendation by the Persistent Organic Pollutants Review Committee to List Short-Chain Chlorinated Paraffins in Annex a to the Convention and Draft Text of the Proposed Amendment*, UNEP/POPS/COP.8/14, 2017.
(18) Wang, Y.; Gao, W.; Jiang, G. Strengthening the Study on the Behavior and Transformation of Medium-Chain Chlorinated Paraffins in the Environment. *Environ. Sci. Technol.* **2017**, *51* (18), 10282−10283.
(19) Labunska, I.; Harrad, S.; Wang, M.; Santillo, D.; Johnston, P. Human Dietary Exposure to PBDEs Around E-Waste Recycling Sites in Eastern China. *Environ. Sci. Technol.* **2014**, *48* (10), 5555−5564.
(20) Duan, H.; Miller, T. R.; Gregory, J.; Kirchain, R. Quantitative Characterization of Domestic and Transboundary Flows of Used Electronics. Analysis of Generation, Collection, and Export in the United States, 2013
(21) Lu, S. Y.; Li, Y. X.; Zhang, T.; Cai, D.; Ruan, J. J.; Huang, M. Z.; Wang, L.; Zhang, J. Q.; Qiu, R. L. Effect of E-waste Recycling on Urinary Metabolites of Organophosphate Flame Retardants and Plasticizers and Their Association with Oxidative Stress. *Environ. Sci. Technol.* **2017**, *51* (4), 2427−2437.
(22) Chan, J. K. Y.; Man, Y. B.; Wu, S. C.; Wong, M. H. Dietary intake of PBDEs of residents at two major electronic waste recycling sites in China. *Sci. Total Environ.* **2013**, *463-464*, 1138−1146.
(23) Wang, C.; Gao, W.; Liang, Y.; Wang, Y.; Jiang, G. Concentrations and congener profiles of chlorinated paraffins in domestic polymeric products in China. *Environ. Pollut.* **2018**, *238*, 326−335.
(24) Huisman, J.; Magalini, F.; Kuehr, R. M. C.; Ogilve, S.; Poll, J.; Delgado, C.; Artim, E.; Szlezak, J.; Stevels, A. *2008 Review of Directive 2002/96 on Waste Electrical and Electronic Equipment (WEEE): Final Report*; United Nations University: 2007; p 347.
(25) Chen, H. X.; Qin, Y. F.; Zeng, M. M.; Guo, Y.; Xia, X. F. Discussion on actuality and foreground of waste electrical and electronic equipment disposal industry in China. *China Environ. Prot. Ind.* **2011**, *2*, 37−39.
(26) Luo, X.-J.; Sun, Y.-X.; Wu, J.-P.; Chen, S.-J.; Mai, B.-X. Short-chain chlorinated paraffins in terrestrial bird species inhabiting an e-waste recycling site in South China. *Environ. Pollut.* **2015**, *198*, 41−46.
(27) Zeng, Y.; Huang, C.; Luo, X.; Liu, Y.; Ren, Z.; Mai, B. Polychlorinated biphenyls and chlorinated paraffins in home-produced eggs from an e-waste polluted area in South China: Occurrence and human dietary exposure. *Environ. Int.* **2018**, *116*, 52−59.
(28) Zeng, Y.-H.; Luo, X.-J.; Tang, B.; Mai, B.-X. Habitat- and species-dependent accumulation of organohalogen pollutants in home-produced eggs from an electronic waste recycling site in South China: Levels, profiles, and human dietary exposure. *Environ. Pollut.* **2016**, *216*, 64−70.
(29) Sun, R.; Luo, X.; Tang, B.; Chen, L.; Liu, Y.; Mai, B. Bioaccumulation of short chain chlorinated paraffins in a typical freshwater food web contaminated by e-waste in south china: Bioaccumulation factors, tissue distribution, and trophic transfer. *Environ. Pollut.* **2017**, *222*, 165−174.
(30) Yuan, B.; Fu, J.; Wang, Y.; Jiang, G. Short-chain chlorinated paraffins in soil, paddy seeds (Oryza sativa) and snails (Ampullariidae) in an e-waste dismantling area in China: Homologue group pattern, spatial distribution and risk assessment. *Environ. Pollut.* **2017**, *220*, 608−615.
(31) Zeng, Y.-H.; Tang, B.; Luo, X.-J.; Zheng, X.-B.; Peng, P.-A.; Mai, B.-X. Organohalogen pollutants in surface particulates from workshop floors of four major e-waste recycling sites in China and implications for emission lists. *Sci. Total Environ.* **2016**, *569-570*, 982−989.
(32) Zhan, F.; Zhang, H.; Wang, J.; Xu, J.; Yuan, H.; Gao, Y.; Su, F.; Chen, J. Release and Gas-Particle Partitioning Behaviors of Short-Chain Chlorinated Paraffins (SCCPs) During the Thermal Treatment of Polyvinyl Chloride Flooring. *Environ. Sci. Technol.* **2017**, *51* (16), 9005−9012.
(33) Xin, S.; Gao, W.; Wang, Y.; Jiang, G. Thermochemical emission and transformation of chlorinated paraffins in inert and oxidizing atmospheres. *Chemosphere* **2017**, *185*, 899−906.
(34) Xu, J.; Gao, Y.; Zhang, H.; Zhan, F.; Chen, J. Dispersion of Short- and Medium-Chain Chlorinated Paraffins (CPs) from a CP Production Plant to the Surrounding Surface Soils and Coniferous Leaves. *Environ. Sci. Technol.* **2016**, *50* (23), 12759−12766.
(35) Gao, W.; Cao, D.; Wang, Y.; Wu, J.; Wang, Y.; Wang, Y.; Jiang, G. External Exposure to Short- and Medium-Chain Chlorinated Paraffins for the General Population in Beijing, China. *Environ. Sci. Technol.* **2018**, *52* (1), 32−39.
(36) Zeng, L. X.; Wang, T.; Han, W. Y.; Yuan, B.; Liu, Q. A.; Wang, Y. W.; Jiang, G. B. Spatial and Vertical Distribution of Short Chain Chlorinated Paraffins in Soils from Wastewater Irrigated Farmlands. *Environ. Sci. Technol.* **2011**, *45* (6), 2100−2106.
(37) Zeng, L. X.; Lam, J. C. W.; Horii, Y.; Li, X. L.; Chen, W. F.; Qiu, J. W.; Leung, K. M. Y.; Yamazaki, E.; Yamashita, N.; Lam, P. K. S. Spatial and temporal trends of short- and medium-chain chlorinated paraffins in sediments off the urbanized coastal zones in China and Japan: A comparison study. *Environ. Pollut.* **2017**, *224*, 357−367.
(38) Li, J.; Zhang, Z.; Ma, L.; Zhang, Y.; Niu, Z. Implementation of USEPA RfD and SFO for improved risk assessment of organophosphate esters (organophosphate flame retardants and plasticizers). *Environ. Int.* **2018**, *114*, 21−26.
(39) Liu, L.-H.; Ma, W.-L.; Liu, L.-Y.; Huo, C.-Y.; Li, W.-L.; Gao, C.-J.; Li, H.-L.; Li, Y.-F.; Chan, H. M. Occurrence, sources and human exposure assessment of SCCPs in indoor dust of northeast China. *Environ. Pollut.* **2017**, *225*, 232−243.

DOI: 10.1021/acs.est.8b02625
Environ. Sci. Technol. 2018, 52, 11510−11519

(40) Shi, L.; Gao, Y.; Zhang, H.; Geng, N.; Xu, J.; Zhan, F.; Ni, Y.; Hou, X.; Chen, J. Concentrations of short- and medium-chain chlorinated paraffins in indoor dusts from malls in China: Implications for human exposure. *Chemosphere* **2017**, *172*, 103−110.
(41) Zhang, H.; Vestergren, R.; Wang, T.; Yu, J.; Jiang, G.; Herzke, D. Geographical Differences in Dietary Exposure to Perfluoroalkyl Acids between Manufacturing and Application Regions in China. *Environ. Sci. Technol.* **2017**, *51* (10), 5747−5755.
(42) Wang, Y.; Hu, J.; Lin, W.; Wang, N.; Li, C.; Luo, P.; Hashmi, M. Z.; Wang, W.; Su, X.; Chen, C.; Liu, Y.; Huang, R.; Shen, C. Health risk assessment of migrant workers' exposure to polychlorinated biphenyls in air and dust in an e-waste recycling area in China: Indication for a new wealth gap in environmental rights. *Environ. Int.* **2016**, *87*, 33−41.
(43) IPCS *Environmental Health Criteria 181: Chlorinated Paraffins*; International Programme on Chemical Safety, Geneva, Switzerland, http://www.inchem.org/documents/ehc/ehc/ehc181.htm, Accessed date: January 2018. 1996.
(44) Wong, F.; Suzuki, G.; Michinaka, C.; Yuan, B.; Takigami, H.; de Wit, C. A. Dioxin-like activities, halogenated flame retardants, organophosphate esters and chlorinated paraffins in dust from Australia, the United Kingdom, Canada, Sweden and China. *Chemosphere* **2017**, *168*, 1248−1256.
(45) Reth, M.; Zencak, Z.; Oehme, M. First study of congener group patterns and concentrations of short- and medium-chain chlorinated paraffins in fish from the North and Baltic Sea. *Chemosphere* **2005**, *58* (7), 847−854.
(46) Houde, M.; Muir, D. C. G.; Tomy, G. T.; Whittle, D. M.; Teixeira, C.; Moore, S. Bioaccumulation and trophic magnification of short- and medium-chain chlorinated paraffins in food webs from Lake Ontario and Lake Michigan. *Environ. Sci. Technol.* **2008**, *42* (10), 3893−3899.
(47) Yuan, B.; Wang, T.; Zhu, N. L.; Zhang, K. G.; Zeng, L. X.; Fu, J. J.; Wang, Y. W.; Jiang, G. B. Short Chain Chlorinated Paraffins in Mollusks from Coastal Waters in the Chinese Bohai Sea. *Environ. Sci. Technol.* **2012**, *46* (12), 6489−6496.
(48) Sun, R.; Luo, X.; Tang, B.; Li, Z.; Huang, L.; Wang, T.; Mai, B. Short-chain chlorinated paraffins in marine organisms from the Pearl River Estuary in South China: Residue levels and interspecies differences. *Sci. Total Environ.* **2016**, *553*, 196−203.
(49) Zhou, Y.; Yin, G.; Du, X.; Xu, M.; Qiu, Y.; Ahlqvist, P.; Chen, Q.; Zhao, J. Short-chain chlorinated paraffins (SCCPs) in a freshwater food web from Dianshan Lake: Occurrence level, congener pattern and trophic transfer. *Sci. Total Environ.* **2018**, *615*, 1010−1018.
(50) Huang, H.; Gao, L.; Zheng, M.; Li, J.; Zhang, L.; Wu, Y.; Wang, R.; Xia, D.; Qiao, L.; Cui, L.; Su, G.; Liu, W.; Liu, G. Dietary exposure to short- and medium-chain chlorinated paraffins in meat and meat products from 20 provinces of China. *Environ. Pollut.* **2018**, *233*, 439−445.
(51) Cao, Y.; Harada, K. H.; Liu, W.; Yan, J.; Zhao, C.; Niisoe, T.; Adachi, A.; Fujii, Y.; Nouda, C.; Takasuga, T.; Koizumi, A. Short-chain chlorinated paraffins in cooking oil and related products from China. *Chemosphere* **2015**, *138*, 104−111.
(52) Iino, F.; Takasuga, T.; Senthilkumar, K.; Nakamura, N.; Nakanishi, J. Risk assessment of short-chain chlorinated paraffins in Japan based on the first market basket study and species sensitivity distributions. *Environ. Sci. Technol.* **2005**, *39* (3), 859−866.
(53) Harada, K. H.; Takasuga, T.; Hitomi, T.; Wang, P. Y.; Matsukami, H.; Koizumi, A. Dietary Exposure to Short-Chain Chlorinated Paraffins Has Increased in Beijing, China. *Environ. Sci. Technol.* **2011**, *45* (16), 7019−7027.
(54) Government of Canada, Environment Canada, Health Canada. *Canadian Environmental Protection Act. Priority Substances List Assessment Report, Chlorinated Paraffins*, ISBN 0-662-205154, 1993.
(55) Vandenberg, L. N.; Colborn, T.; Hayes, T. B.; Heindel, J. J.; Jacobs, D. R., Jr.; Lee, D.-H.; Shioda, T.; Soto, A. M.; vom Saal, F. S.; Welshons, W. V.; Zoeller, R. T.; Myers, J. P. Hormones and Endocrine-Disrupting Chemicals: Low-Dose Effects and Nonmonotonic Dose Responses. *Endocr. Rev.* **2012**, *33* (3), 378−455.

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