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# Tracking Dietary Sources of Short- and Medium-Chain Chlorinated Paraffins in Marine Mammals through a Subtropical Marine Food Web

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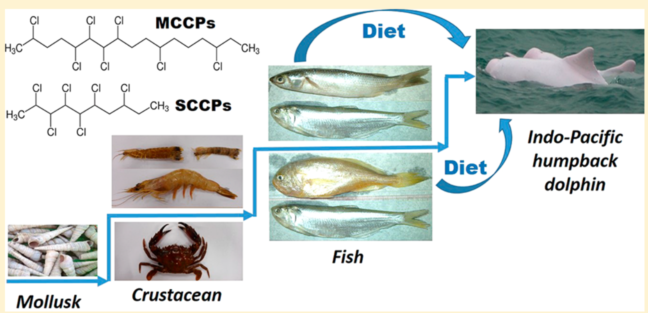
Supporting Information



**ABSTRACT:** Our previous study revealed an elevated accumulation of short-chain chlorinated paraffins (SCCPs) and medium-chain chlorinated paraffins (MCCPs) in marine mammals from Hong Kong waters in the South China Sea. To examine the bioaccumulation potential and biomagnification in these apex predators, we sampled the dietary items of marine mammals and tracked the sources of SCCPs and MCCPs through a marine food web in this region. Sixteen fish species, seven crustacean species, and four mollusk species were collected, and the main prey species were identified for two species of marine mammals. Concentrations of ∑SCCPs and ∑MCCPs in these collected species suggested a moderate pollution level in Hong Kong waters compared to the global range. Lipid content was found to mediate congener-specific bioaccumulation in these marine species. Significantly positive correlations were observed between trophic levels and concentrations of ∑SCCPs or ∑MCCPs ($p < 0.05$). Trophic magnification factors for ∑SCCPs and ∑MCCPs were 4.29 and 4.79, indicating that both of them have trophic magnification potentials. Elevated biomagnification of SCCPs and MCCPs from prey species to marine mammals was observed. This is the first report of dietary source tracking of SCCPs and MCCPs in marine mammals. The elevated biomagnification between prey and marine mammals raises environmental concerns about these contaminants.




## INTRODUCTION

Chlorinated paraffins (CPs) are high molecular weight polychlorinated *n*-alkanes that have been used as additives, secondary plasticizers, and flame retardants in a variety of industrial applications for several decades.[1] CPs have been produced on an industrial scale since the 1930s. The Chinese production capacity of CPs had increased to 1600 kilotons in 2014,[2] and China is the largest producer, consumer, and exporter of CPs in the world.[3] As large production volume chemicals, CPs have been detected in various environmental matrices worldwide in the past decade.[4−9] By their chain length, CPs are divided into short-chain (SCCPs, $C_{10-13}$), medium-chain (MCCPs, $C_{14-17}$), and long-chain (LCCPs, $C_{>17}$) chlorinated paraffins.[10] Of the CPs, SCCPs have attracted the most extensive concern due to their bioaccumulation, long-range transport potential, and high aquatic and mammalian toxicity.[11] In May 2017, SCCPs had been listed as a group of persistent organic pollutants (POPs) under the Stockholm Convention.[12] However, scientific studies on the occurrence and behavior of SCCPs in biota[13−22] and humans[23,24] are still limited. Among the available studies, information about MCCPs is especially scarce, but they may also deserve the same attention as concomitant pollutants such as SCCPs.[23−25]

Because of their high hydrophobicity (log $K_{OW}$ 5−8) and resistance to metabolism, SCCPs and MCCPs tend to bioaccumulate in biota and have the potential to biomagnify in food webs.[26,27] However, limited studies are available to date on the bioaccumulation and trophic magnification of CPs, especially MCCPs, which is one key criterion for evaluating their POP characteristics. To our knowledge, there are only a few studies that have focused on the biomagnification of SCCPs

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Table 1. Trophic Level (TL), Lipid Content (%), and ∑SCCP and ∑MCCP Concentrations [dry weight basis (dw) and lipid weight basis (lw)] in Fish, Crustacean, and Mollusk Species from Subtropical Hong Kong Waters

| species | n[a] | TL[b] | lipid[b] (%) | ∑SCCPs[b] (ng g-1, dw) | ∑MCCPs[b] (ng g-1, dw) | ∑SCCPs[b] (ng g-1, lw) | ∑MCCPs[b] (ng g-1, lw) |
|---|---|---|---|---|---|---|---|
| | | | | Fishes | | | |
| *A. fasciatus* | 1[a] | 3.17 | 4.6 | 45.8 | 168 | 1010 | 3800 |
| *C. thrissa* | 13 | 2.32 ± 0.07 | 41.1 ± 10.3 | 298 ± 115 | 793 ± 338 | 720 ± 175 | 1960 ± 677 |
| *C. arel* | 1[a] | 2.68 | 7.9 | 47.2 | 64.0 | 598 | 810 |
| *D. russelii* | 1[a] | 3.56 | 7.0 | 73.6 | 146 | 1050 | 2090 |
| *E. cardinalis* | 1[a] | 3.06 | 13.0 | 106 | 131 | 812 | 1000 |
| *I. japonica* | 1[a] | 3.14 | 1.6 | 15.3 | 41.4 | 955 | 2590 |
| *J. heterolepis* | 4 | 3.11 ± 0.16 | 7.4 ± 3.8 | 64.2 ± 39.7 | 128 ± 110 | 886 ± 106 | 1660 ± 561 |
| *L. brevirostris* | 6 | 3.31 ± 0.12 | 10.0 ± 0.8 | 69.4 ± 9.1 | 155 ± 68.7 | 693 ± 59.2 | 1540 ± 630 |
| *P. macracanthus* | 1[a] | 2.88 | 5.4 | 31.9 | 68.6 | 622 | 1230 |
| *P. argentata* | 1[a] | 2.95 | 6.0 | 54.2 | 75.3 | 903 | 1260 |
| *P. sextarius* | 1[a] | 3.25 | 8.7 | 90.3 | 134 | 1000 | 1470 |
| *P. indicus* | 1[a] | 3.35 | 4.5 | 53.7 | 103 | 1220 | 2370 |
| *R. richardsonii* | 1[a] | 2.88 | 4.0 | 31.3 | 54.6 | 781 | 1320 |
| *S. canaliculatus* | 7 | 2.92 ± 0.19 | 13.8 ± 7.2 | 85.0 ± 45.7 | 225 ± 140 | 739 ± 373 | 2040 ± 1320 |
| *S. ovata* | 1[a] | 3.08 | 7.7 | 56.3 | 103 | 739 | 1320 |
| *T. vagina* | 1[a] | 3.04 | 3.9 | 28.5 | 57.3 | 761 | 1480 |
| | | | | Crustaceans | | | |
| *H. harpax* | 4 | 3.18 ± 0.26 | 8.9 ± 1.7 | 52.4 ± 15.7 | 88.4 ± 39.9 | 586 ± 94.3 | 975 ± 330 |
| *M. affinis* | 5 | 3.11 ± 0.04 | 5.1 ± 0.7 | 23.6 ± 11.1 | 26.6 ± 23.6 | 464 ± 177 | 503 ± 272 |
| *M. nepa* | 1[a] | 2.45 | 4.0 | 11.1 | 18.8 | 278 | 471 |
| *M. ensis* | 1[a] | 2.67 | 4.3 | 18.1 | 22.9 | 413 | 525 |
| *P. sanguinolentus* | 5 | 2.86 ± 0.17 | 5.8 ± 1.2 | 21.9 ± 15.2 | 28.2 ± 9.6 | 368 ± 186 | 496 ± 151 |
| *P. pelagicus* | 1[a] | 2.72 | 4.4 | 20.8 | 37.6 | 474 | 855 |
| *P. trituberculatus* | 1[a] | 2.74 | 10.5 | 26.9 | 42.7 | 261 | 392 |
| | | | | Mollusks | | | |
| *A. ferruginea* | 1[a] | 2.21 | 10.7 | 38.9 | 63.8 | 363 | 596 |
| *B. rana* | 1[a] | 2.79 | 10.9 | 44.4 | 82.8 | 408 | 754 |
| *M. trapa* | 1[a] | 2.28 | 7.7 | 21.8 | 43.9 | 280 | 563 |
| *T. bacillum* | 1[a] | 2.11 | 6.3 | 18.8 | 33.0 | 302 | 515 |

[a]Three to ten individuals were pooled to form a composite sample for some species with small size. [b]Average ± standard deviation.

until now, and the results are not consistent.[13,16,17,19,28] SCCPs have been found to be biomagnified in aquatic food webs in Lake Ontario and Lake Michigan (Canada),[13] Gaobeidian Lake (China),[28] Bohai Bay (China),[16] Pearl River Estuary (China),[17] and Antarctica,[19] but trophic dilution has also been observed in cod and gammarid species from the Arctic,[29] in benthic mollusks from Bohai Bay,[30] and in a freshwater food web in an e-waste recycling site in South China.[31] The complex results may be attributed to the biomagnification processes generally being affected by organisms, depuration rate, feeding habit, food web structure, and even environmental parameters, such as temperature and suspended particles.[32] Therefore, further research is necessary to reveal the potential of SCCPs and factors influencing their biomagnification in different food webs. Due to the similar structures and physicochemical properties of CP congeners, MCCPs exhibit behaviors similar to those of SCCPs in the environment. However, little is known about the bioaccumulation and biomagnification pathway of MCCPs,[13] especially in marine food webs.

Hong Kong is situated at the southeastern part of the Pearl River Estuary (PRE) in the Pearl River Delta (PRD), South China. As the PRD region is one of the fastest growing industrialized and urbanized regions in China, recent studies have demonstrated that this region has become an area heavily contaminated by CPs.[17,33−35] Hong Kong waters are located in the downstream of the PRE, and it is conceivable that the waters may be contaminated by CPs discharged from the PRD. Our recent monitoring study carried out in Hong Kong waters revealed that SCCPs and MCCPs are widespread in marine sediments and much higher levels are in the PRE.[35] Moreover, our previous study[14] revealed elevated accumulation of SCCPs and MCCPs in two species of marine mammals, finless porpoises (*Neophocaena phocaenoides*) and Indo-Pacific humpback dolphins (*Sousa chinensis*), which are the two resident cetacean species in the coastal waters of Hong Kong in the South China Sea.[36,37] Marine mammals are the top predators of the marine food chain and feed on a variety of aquatic prey.[38] We suspect that the elevated levels of these recalcitrant and lipophilic chemicals are strongly associated with their dietary intake in marine food webs. Understanding the feeding ecology of marine mammals is crucial for understanding how they accumulate high contaminant burdens from SCCPs and MCCPs. However, up to now, the dietary sources and bioaccumulation pathways of CPs in marine mammals were still unknown. Such information is important for assessing the ecological and health risks of these substances.

In this study, we sampled the dietary items of these marine mammals from the Hong Kong waters of the South China Sea. Sixteen fish species, seven crustacean species, and four species of mollusk were collected to simultaneously analyze SCCPs and MCCPs. The aim was to further track the dietary sources of SCCPs and MCCPs in marine mammals through a specific marine food web and examine the bioaccumulation potential and biomagnification of SCCPs and MCCPs in these apex



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marine predators. It is hoped that the results can provide a better understanding of the behavior and fate of SCCPs and MCCPs in marine ecosystems.

## ■ MATERIALS AND METHODS

**Sample Collection.** Aquatic organisms including 4 mollusk species, 7 crustacean species, and 16 fish species were collected from the subtropical Hong Kong waters of the South China Sea from August to November, 2012 (Table 1). Invertebrates and fishes were caught with a bottom trawl. The 16 fish species included *Apogon fasciatus* (*A. fasciatus*), *Clupanodon thrissa* (*C. thrissa*), *Cynoglossus arel* (*C. arel*), *Dendrophysa russelii* (*D. russelii*), *Evynnis cardinalis* (*E. cardinalis*), *Inegocia japonica* (*I. japonica*), *Johnius heterolepis* (*J. heterolepis*), *Leiognathus brevirostris* (*L. brevirostris*), *Priacanthus macracanthus* (*P. macracanthus*), *Pennahia argentata* (*P. argentata*), *Polydactylus sextarius* (*P. sextarius*), *Platycephalus indicus* (*P. indicus*), *Repomucenus richardsonii* (*R. richardsonii*), *Signaus canaliculatus* (*S. canaliculatus*), *Solea ovata* (*S. ovata*), and *Trypaucehn vagina* (*T. vagina*). The seven species of crustacean included *Harpiosquilla harpax* (*H. harpax*), *Metapenaeus affinis* (*M. affinis*), *Miyakea nepa* (*M. nepa*), *Meapenaeus ensis* (*M. ensis*), *Portunus sanguinolentus* (*P. sanguinolentus*), *Portunus pelagicus* (*P. pelagicus*), and *Portunus trituberculatus* (*P. trituberculatus*). The four species of mollusk included *Anadara ferruginea* (*A. ferruginea*), *Bufonaria rana* (*B. rana*), *Murex trapa* (*M. trapa*), and *Turritella bacillum* (*T. bacillum*). These marine species together with two cetacean species (*Neophocaena phocaenoides* and *Sousa chinensis*) from this region[14] constitute a marine food web with regard to low-trophic-level marine organisms to the top marine mammals. The two species of marine mammals mainly prey on fishes,[36,37] and most fish species prey on small fishes and benthic invertebrates (fishbase.org). The prey items for the two species of marine mammals are identified in Table 2,[36,37] and feeding habits for other organisms are presented in Table S1 of the Supporting Information (SI). The food web structure was similar to that of a previous study.[16] The sampling area covered the west, south, and east of Hong Kong waters, as shown in Figure 1. All collected samples were wrapped in aluminum foil and stored in ice-cooled boxes onboard. After they were transferred to the laboratory, their species were identified. For some species of a small size, 3−10 individuals were pooled to form a composite sample. The muscles of the fish and soft tissues of the invertebrates were dissected, freeze-dried, homogenized, and then stored at −20 °C before chemical analysis. Detailed information on the samples and sampling sites is presented in Tables S1 and S2 (SI), respectively.

**Instrumental Analysis, Identification, and Quantification.** SCCPs and MCCPs were simultaneously analyzed using high-resolution gas chromatography coupled with an electron capture negative ionization low-resolution mass spectrometer (HRGC/ECNI-LRMS, Agilent 7890A/5975C) based on our previously developed method.[14,39] SCCP and MCCP congeners containing 10−13 carbon and 5−10 chlorine atoms were determined for all biota samples. The most and second-most abundant isotope ions $[M - Cl]^-$ of each individual homologue were monitored under a selected ion monitoring mode for quantification and confirmation, respectively.[40,41] To ensure the instrument's sensitivity and to minimize mutual interference of CP congeners, all monitored ions of SCCPs and MCCPs were divided into four groups ($C_{10}$−$C_{15}$, $C_{11}$−$C_{16}$, $C_{12}$−$C_{17}$, and $C_{13}$−$C_{14}$) and subjected to four individual injections for each sample.[14]

Table 2. Biomagnification Factors (BMFs) of ∑SCCPs and ∑MCCPs from Prey Fishes to Two Cetacean Species of Marine Mammals

| cetacean species | prey items | BMF ∑SCCPs | BMF ∑MCCPs |
|---|---|---|---|
| finless porpoise (*N. phocaenoides*) | *A. fasciatus* | 3.1 | 1.4 |
| | *C. arel* | 5.2 | 6.7 |
| | *D. russelii* | 2.9 | 2.6 |
| | *J. heterolepis* | 3.5 | 3.3 |
| | *L. brevirostris* | 4.5 | 3.5 |
| | *P. argentata* | 3.4 | 4.3 |
| | *M. affinis* | 6.7 | 11 |
| Indo-Pacific humpback dolphin (*S. chinensis*) | *C. thrissa* | 24 | 24 |
| | *C. arel* | 29 | 58 |
| | *D. russelii* | 17 | 23 |
| | *J. heterolepis* | 20 | 28 |
| | *L. brevirostris* | 25 | 31 |
| | *P. macracanthus* | 28 | 38 |
| | *P. argentata* | 19 | 38 |
| | *S. canaliculatus* | 24 | 23 |
| | Chinese herring[a] (*Ilisha elongata*) | 15 | –[b] |
| | Sardine[a] (*Sardinella jussieu*) | 13 | – |
| | Silver pomfret[a] (*Pampus argenteus*) | 36 | – |
| | Tapertail anchovy[a] (*Coilia mystus*) | 38 | – |
| | Bombay duck[a] (*Harpadon nehereus*) | 11 | – |
| | Squid[a] (*Loligo tagoi*) | 18 | – |

[a]Data are from a previous study conducted in the Pearl River Estuary.[17] [b]Not available.

During CP analysis with LRMS, the SCCP congener can be disturbed by the MCCP congener with five carbon atoms more and two chlorine atoms less due to mass overlap.[42] To ensure valid quantification, CP congener groups were first identified by comparing retention time range, signal shape, and isotope ratio with their reference standards.[43] The actual relative integrated signals for CP congener that suffered from mass overlapping interference were corrected by chemical calculation using isotopic abundance and theoretical isotope ratios. The detailed chemical calculation procedure has been described in our previous work.[39] The total SCCP and MCCP quantification was performed using the method described by Reth et al.[44] The coefficients of determination ($R^2$) of five-point calibration curves for both SCCPs and MCCPs were ≥0.97.

**Quality Assurance and Quality Control (QA/QC).** A procedural blank was included in each batch of eight samples to monitor the possible interference or contamination. All targeted SCCPs and MCCPs in blanks were below or close to the detection limits, and the final concentrations of CPs reported in this study were not blank-corrected. The recoveries of SCCPs (51.5%, 55.5%, and 63.0% Cl) and MCCPs (42.0, 52.0, and 57.0% Cl) in matrix-spiked samples were 81.0−96.0% and 83.0−101%, respectively, and the relative standard deviations (RSDs) were ≤13.0% ($n$ = 5). The surrogate recoveries of $^{13}$C-*trans*-chlordane in all samples were 79.0−97.0%, and the concentration data were corrected by the


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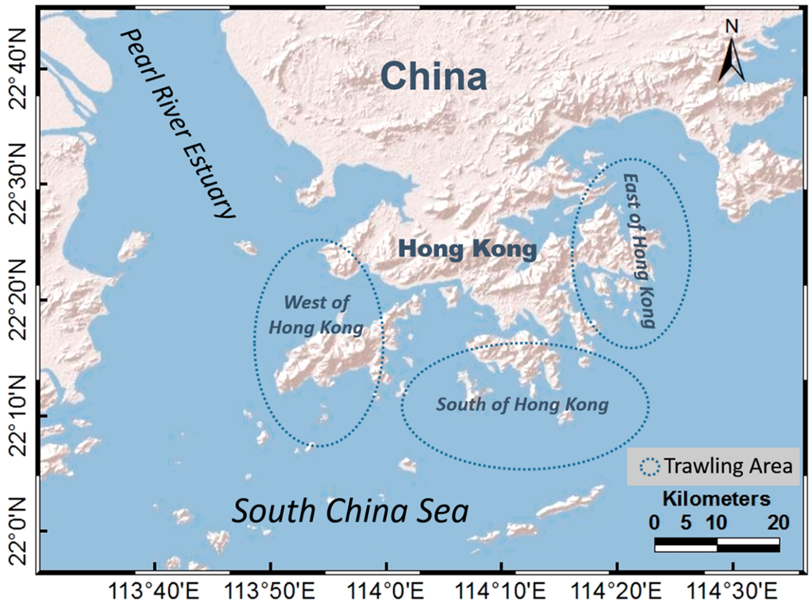


**Figure 1.** Sampling map of fishes, crustaceans, and mollusks in Hong Kong waters.

surrogates. The method detection limits (MDLs) for total SCCPs (∑SCCPs) and MCCPs (∑MCCPs) were defined as three times the standard deviation (SD) of the mean procedural blanks ($n = 8$). The MDLs in biota were estimated at 10 ng $g^{-1}$ dry weight (dw) and 40 ng $g^{-1}$ lipid weight (lw) for ∑SCCPs and 16 ng $g^{-1}$ dw and 60 ng $g^{-1}$ lw ∑MCCPs.

**Stable Nitrogen Isotope Analysis and Trophic Level (TL) Calculation.** The muscle for stable isotope analysis was lyophilized and ground into an ultrafine powder. Stable isotope measurement was performed by a Thermo DELTA V Advantage isotope ratio mass spectrometer interfaced with a Flash EA 112 series elemental analyzer. Stable isotope abundance was expressed as $\delta^{15}N$ (‰) = $[(^{15}N/^{14}N)_{sample}/(^{15}N/^{14}N)_{standard} - 1] \times 1000$ (‰). The $(^{15}N/^{14}N)_{standard}$ values were based on atmospheric $N_2$ and the analytical precision was ±0.2‰. The trophic level (TL) was estimated on the basis of the measured nitrogen isotope ratios in food web samples using the following formula: $TL_{consumer} = (\delta^{15}N_{consumer} - \delta^{15}N_{primary\ consumer})/3.8 + 2$, where $\delta^{15}N_{primary\ consumer}$ is the stable nitrogen isotope value of the zooplankton with an average of 9.7‰, 3.8 is the isotopic trophic enrichment factor, and the TL of zooplankton (mainly amphipods and copepods) was assumed to be 2 according to previous studies.[16,17,45]

**Biota−Sediment Accumulation Factor (BSAF), Biomagnification Factor (BMF), and Trophic Magnification Factor (TMF) Assessment.** The BSAF was assessed by the following equation: $BSAF = C_{biota}/C_{sediment}$, where $C_{biota}$ and $C_{sediment}$ are the average lipid-normalized CP concentrations in benthic organisms [ng $g^{-1}$, lipid weight (lw)] and the average organic-carbon-normalized concentrations in sediment (ng $g^{-1}$, TOC), respectively.[16] The BMF was defined as the ratio of the average lipid-normalized concentration between marine mammals (predator) and fishes (prey).[13,17] For BSAF and BMF calculations, the data set for sediment and marine mammals was published previously.[14,35] The trophic magnification factor (TMF) was used to describe the biomagnification of SCCPs and MCCPs in this study. TMF was calculated on the basis of the correlations between the TLs and lipid-normalized SCCP or MCCP concentrations as per the following equations: $\log C_{SCCPs/MCCPs} = a + b \times TL$, where $a$ and $b$ represent the constant and the slope of the linear regression, respectively. Slope $b$ was used to calculate TMF by the equation $TMF = 10^b$.[28,32]

## ■ RESULTS AND DISCUSSION

SCCPs ($C_{10-13}Cl_{5-10}$) and MCCPs ($C_{14-17}Cl_{5-10}$) were detected in all fish, crustacean, and mollusk samples, suggesting that they are ubiquitous pollutants in marine organisms in subtropical Hong Kong waters. One-way ANOVA (SPSS 20.0) indicated no significant differences ($p > 0.05$) of ∑SCCP concentrations and ∑MCCP concentrations in marine species with similar TLs among different sampling areas. Therefore, the same species from three sampling areas (i.e., eastern, southern, and western waters) were pooled for analysis.

**Accumulation Levels of ∑SCCPs and ∑MCCPs in Marine Organisms.** ∑SCCP and ∑MCCP concentrations (dw and lw) for each species are summarized in Table 1 and illustrated in Figure S1 (SI). Detailed data for individuals of the species are shown in Table S3 (SI). Quantified individuals of SCCPs and MCCPs are also presented in Table S4 (SI). The lipid content varied largely between marine species, and the values ranged from 1.6% to 41%. Significant positive correlations were found between the lipid content and dry basis concentrations of ∑SCCPs or ∑MCCPs ($R^2 = 0.87$ and 0.78, $p < 0.05$; Figure S2, SI), indicating that lipid content plays a key role in the bioaccumulation of SCCPs and MCCPs, which coincides with our previous reports[14,28,30] and other studies.[16,19,29,46] Therefore, the concentrations are expressed in ng $g^{-1}$ lw below.

∑SCCP concentrations for fishes, crustaceans, and mollusks ranged from 280 to 1940 [mean ± standard deviation (SD), 801 ± 253; 95% confidence interval (CI), 730−872], 202−694 (mean ± SD, 422 ± 162; 95% CI, 348−496), and 259−506 (mean ± SD, 328 ± 79.0; 95% CI, 262−394), ng $g^{-1}$ lw, respectively. Average levels of ∑SCCPs among the three taxonomic groups were found in fish > crustacean > mollusk. Among these marine species, the highest average levels of ∑SCCPs were found in fish species *P. indicus* and *D. russelii*, which are known to prey on fishes and crustaceans. A lower average concentration of ∑SCCPs was detected in *C. arel*, a


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tongue sole known to feed on benthic crustaceans and bivalves. The lowest average concentrations of ∑SCCPs were found in crustacean species *P. sanguinolentus* and *M. nepa*, which are known to feed on small fish and crustaceans but are also scavengers and deposit feeders. Accumulation levels of ∑SCCPs in the fish, crustaceans, and mollusks from eastern Hong Kong waters were much lower than the recent reported levels in similar species from the PRE (210–21 000 ng $g^{-1}$ lw).[17] The PRE, situated at the upstream of the northwestern waters of Hong Kong, has been proved to be an important reservoir of SCCPs derived from the PRD.[33] Higher accumulation levels of ∑SCCPs in marine organisms from the PRE are attributable to heavier water pollution in this region than in Hong Kong waters.[35] It is noteworthy that ∑SCCP concentrations in the fish (133–2230 ng $g^{-1}$ lw) were much lower than in the two cetacean species of finless porpoise (570–5800 ng $g^{-1}$ lw) and Indo-Pacific humpback dolphins (920–24 000 ng $g^{-1}$ lw) that dwell in Hong Kong waters,[14,34] which provides a valuable opportunity to study the biomagnification potential of SCCPs in marine mammals.

When comparing ∑SCCP concentrations in marine organisms reported here with those from other regions of China and other countries/regions worldwide, we can conclude that accumulated concentrations of ∑SCCPs in marine organisms from Hong Kong waters are higher than the levels in fish from the North Sea and Baltic Sea (39–670 ng $g^{-1}$, lw),[20] in top predatory fish across Canada (12–288 ng $g^{-1}$, lw),[18] and in cod samples from the European Arctic (28–540 ng $g^{-1}$, lw)[22] and Norwegian Arctic (10 ng $g^{-1}$, wet weight)[47] but lower than the recently reported concentrations in fishes from the Svalbard in the Arctic (4100–9700 ng $g^{-1}$, lw)[29] and King George Island in Antarctica (1500 ng $g^{-1}$, lw)[19], and much lower than previously reported levels in marine organisms from Bohai Bay in north China (4800–32 900 ng $g^{-1}$, lw).[15,16,46] The range of ∑SCCPs in marine organisms from Hong Kong waters overlaps the concentrations reported for whole fish from Lake Michigan and Lake Ontario (172–1030 ng $g^{-1}$, lw)[13] and from the Ebro River Delta (Spain) (172–3840 ng $g^{-1}$ lw).[48] The anthropogenic activities, regional pollution level, and species differences may be the major factors influencing the accumulation levels of CPs in global biota. These comparisons indicate that Hong Kong waters are at the medium level of SCCP pollution internationally, while other regions in China (e.g., Bohai Bay and the PRD) show heavier contamination by SCCPs.

∑MCCP concentrations for fishes, crustaceans, and mollusks ranged from 502 to 4770 (mean ± SD, 1820 ± 934; geometric mean, 1624; 95% CI, 1550–2080), 205–1190 (mean ± SD, 593 ± 306; 95% CI, 454–732), and 464–874 (mean ± SD, 603 ± 132; 95% CI, 492–713) ng $g^{-1}$ lw, respectively. As shown in Table 1, among these marine species, the lowest average level of ∑MCCPs was detected in *P. trituberculatus*, while the highest levels of ∑MCCPs were found in *A. fasciatus*, a cardinalfish known to feed on small crustaceans. Similar to ∑SCCPs, ∑MCCP concentrations in the fish species (205–7530 ng $g^{-1}$, lw) from Hong Kong waters were much lower than in the finless porpoises (670–11 000 ng $g^{-1}$, lw) and Indo-Pacific humpback dolphins (1400–56 000 ng $g^{-1}$, lw) from this region,[14] indicating that the biomagnification potential of MCCPs warrants further attention. Until now, there has been a lack of information on the bioaccumulation of MCCPs. ∑MCCP concentrations in fish from Hong Kong waters were found to be higher than those in fishes from northern Europe (14–1600 ng $g^{-1}$, lw),[22] the North Sea and Baltic Sea (19–691 ng $g^{-1}$, lw),[20] Lake Ontario and Lake Michigan (nd–2220 ng $g^{-1}$, lw),[13] and freshwater bodies in Canada (13–130 ng $g^{-1}$, lw).[18] Owing to the limited information on MCCPs in the world's aquatic biota, presently it is impossible to make a global comparison on MCCP levels. In Hong Kong waters, all samples exhibited relatively higher levels of ∑MCCPs than ∑SCCPs. The results were in agreement with our previous reports concerning marine mammals[14] and sediments[35] but opposite to the findings in fishes from Lake Ontario and Lake Michigan.[13] This indicates that MCCP contamination might be heavier in Asia and requires further attention.

Similar to the previous results,[14,35] the dry basis concentrations of ∑SCCPs correlated well with those of ∑MCCPs ($R^2$ = 0.94, $p$ < 0.01; Figure S3A, SI). The root mean squared error (RMSE) was 73.2 ng $g^{-1}$ dw, and the deviations for 87.6% fitting data to regression values were within one RMSE. Significant positive relationships between lipid-normalized ∑SCCPs and ∑MCCPs were also observed ($R^2$ = 0.66, $p$ < 0.01; Figure S3B, SI). The RMSE was 560 ng $g^{-1}$ lw and the deviations for 76.5% fitting data to regression values were within one RMSE. It has been reported that SCCPs and MCCPs are the two common components of CPs in most technical mixtures in China, and they are not strictly grouped by the chain length of *n*-alkane feedstock during chlorination processes.[6,33,49] The significant correlations further reveal that SCCPs and MCCPs could be sharing the same sources and similar accumulation, transfer, and transformation.

**Homologue Profiles and Lipid-Mediated Congener-Specific Accumulation.** The carbon and chlorine homologue abundance profiles of SCCPs and MCCPs in these marine species are shown in Figures S4 and S5 (SI), respectively. The ranges of relative abundance of carbon lengths for every species are also shown in Table S5 (SI). Most species with low lipid content shared similar SCCP and MCCP homologue abundance profiles. Differences were only observed between high- and low-lipid species. Higher relative abundances of shorter $C_{10-11}$ were found in most low-lipid species (53–75%) than in the high-lipid species *C. thrissa* (47–59%). Regarding chlorine substitution, the chlorine content of SCCPs ranged from 59% to 63%. $Cl_6$ and $Cl_7$ were the two dominant congeners, with an average abundance of 24% and 34%, respectively (Figure S5, SI). The homologue patterns of SCCPs were generally consistent with those in sediments from this region[35] but obviously different from those in marine mammals from Hong Kong waters[14] and other marine species from the adjacent PRE.[17] By comparison, higher abundances of longer-chain groups ($C_{12-13}$) along with higher SCCP concentrations were frequently found in marine organisms from the PRE[17] and especially in marine mammals from Hong Kong waters.[14] As discussed above, the PRE receives large volumes of industrial discharge from the PRD, and thus, the difference of the SCCP pattern may result from the diverse sources of pollution and species collected for studies between the PRE and Hong Kong waters. In contrast to the prey and other marine organisms, longer-chain groups dominating the SCCP homologue profile and elevated accumulation in marine mammals may be attributed to high trophic position, lipid content, and accumulation capacities. In addition, SCCP homologue patterns in the present study were also different from those observed in fish from the North Sea and Baltic Sea ($C_{13}$ dominating),[20] the European Arctic,[22] and Canada ($C_{11-12}$



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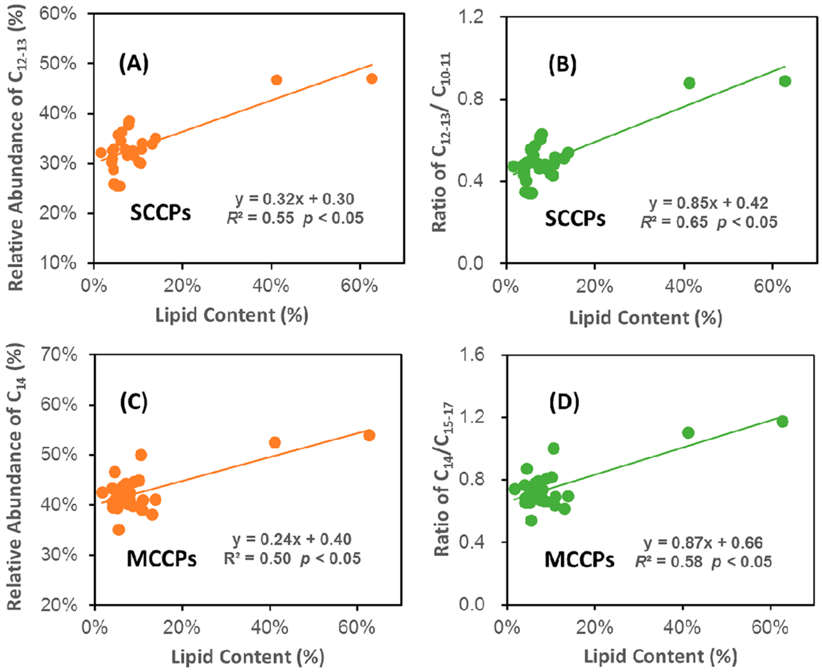


**Figure 2.** Correlations of lipid content vs relative abundance of $C_{12-13}$ (A), lipid content vs ratio of $C_{12-13}/C_{10-11}$ (B), lipid content vs relative abundance of $C_{14}$ (C), and lipid content vs ratio of $C_{14}/C_{15-17}$ (D). Each point in the figure is the average of each marine species from Hong Kong waters. Data of marine mammals collected in 2012 from our previous study[14] were also integrated into the figure for correlation analysis.

dominating),[18] while they were generally similar to the patterns reported in marine species from the Arctic,[29] Antarctica,[19] and the Chinese Bohai Sea ($C_{10-11}$ dominating).[16,46] Notwithstanding potential species-specific differences, the discrepancy in the SCCP homologue pattern may be primarily due to the different used CP formulation.

Most low-lipid species exhibited a more common MCCP homologue distribution, with $C_{14}$ as the most abundant group with a range of 35−54% (Table S5, SI), followed by $C_{15}$, $C_{16}$, and $C_{17}$. Compared to most low-lipid species, the high-lipid *C. thrissa* was found to contain a higher abundance of $C_{14}$ with a range of 50−54%. Regarding chlorine substitution, the chlorine content of MCCPs ranged from 49% to 53%. $C_{14}Cl_{5-7}$ predominated in all the samples, which was slightly different from the finding that $C_{14}Cl_{6-8}$ dominated in Hong Kong sediments.[35] The MCCP homologue patterns were generally similar to the composition profiles of commercial MCCP mixtures in China. Comparing the MCCP patterns with those of marine mammals, it is apparent that a significantly higher proportion of $C_{14}$ was observed in finless porpoises (ranging from 50% to 65%, average 56%) and Indo-Pacific humpback dolphins (ranging from 41% to 51%, average 44%) than their prey and other marine organisms in Hong Kong waters ($p < 0.05$), implying that $C_{14}$ has larger bioavailability and bioaccumulation potential when compared with $C_{15-17}$.

Generally, log $K_{OW}$ of SCCP congeners increases with an increase in carbon chain length, which may lead to facilitating enrichment of longer carbon groups in marine species with higher lipid content.[10] Several previous studies have indicated that the bioaccumulative capacities of SCCP congeners increase with increasing carbon chain length from 10 to 13,[16,17,21,28] and that the number of carbon atoms is the primary factor influencing the bioaccumulation of SCCPs.[29] However, the bioaccumulative behavior of MCCP congeners with a carbon chain length >13 is still unknown. To explore the biological and physicochemical factors influencing the bioaccumulation of CPs, average congener group abundance profiles of SCCPs and MCCPs in typical marine species are illustrated in Figure S6 (SI). From the comparisons among the species, a higher relative abundance of $C_{12-13}$ of $\sum$SCCPs (47%) and $C_{14}$ of $\sum$MCCPs (53%) were found in *C. thrissa*, which was characterized by high lipid content but relatively low trophic level. As discussed above, compared with all the sampled marine species in Hong Kong waters, significantly higher proportions of $C_{12-13}$ and $C_{14}$ were also found in marine mammals characterized by higher lipid content and their top trophic position. It is, therefore, conceivable that longer-chain groups $C_{12-13}$ within SCCPs and short-chain group $C_{14}$ within MCCPs may prefer to accumulate in high-lipid organisms. To test the hypothesis, the relationships between lipid content and relative abundance of $C_{12-13}$ or $C_{14}$ were further analyzed. As shown in Figure 2, significant positive correlations between lipid content and relative abundance of $C_{12-13}$ or the ratio of $C_{12-13}/C_{10-11}$ were observed in all the marine organisms, including marine mammals ($p < 0.05$). Similarly, significant correlations were also observed between lipid content and relative abundance of $C_{14}$ or the ratio of $C_{14}/C_{15-17}$ ($p < 0.05$). Most of the available studies[14,16,19,28−30,46,50] have indicated that lipid content is the key factor influencing the concentration/accumulation of SCCPs in biota, and two of these studies also mentioned that more hydrophobic longer-chain congeners within SCCPs are present in marine animal species[19] and seafood[50] with higher lipid content. In this study, we demonstrated first that lipid content can mediate congener-specific bioaccumulation and found that carbon chain groups $C_{12-13}$ within SCCPs and $C_{14}$ within MCCPs can be preferentially enriched in high-lipid marine species. Second, the physicochemical properties of CP congeners (e.g., log $K_{OW}$ = 5−8) and their bioavailability together with species-differentiated degradation, transformation, and excretion could also contribute to this congener-specific bioaccumulation, but this requires further study.



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**Transfer and Trophic Magnification of both SCCPs and MCCPs.** Our recent study[35] has provided information on the source of discharge and release of SCCPs and MCCPs (i.e., levels and composition profiles) in sediment from Hong Kong waters and information on the magnitude and geographic extent of an effect observed in the benthic community, which is of great importance to the aquatic ecosystem of this region. Furthermore, the transfer of SCCPs and MCCPs from sediment to benthic invertebrates and then to fishes was estimated in this study. Benthic invertebrates (crustaceans and mollusks) are frequently exposed to sediments by ingesting sediment particles, which can accumulate sediment-associated CPs and then transfer them to fishes and apex predator marine mammals. Therefore, the biota-sediment accumulation factors (BSAFs) of SCCPs and MCCPs were simultaneously assessed in Hong Kong waters. As shown in Table S6 (SI), BSAFs of ∑SCCPs and ∑MCCPs ranged from 0.6 to 1.4 and from 0.9 to 2.3, respectively. Among seven crustacean and four mollusk species, only three species showed BSAF values greater than 1 for ∑SCCPs, but nine species did so for ∑MCCPs. The BSAF > 1 indicated that SCCPs and MCCPs can be bioaccumulated in some benthic invertebrates. The discrepancy in BSAFs among the species might result from the difference in feeding habits, lipid composition, metabolism, and selective excretion abilities.[51] The BSAF ranges of SCCPs for crustaceans and mollusks in Hong Kong waters overlap those reported for seawater (0.27−2.3)[16,46] and freshwater invertebrates (0.28−4.53).[31] To our knowledge, there are no available BSAFs of MCCPs for comparison.

The feeding habits of finless porpoises and Indo-Pacific humpback dolphins in Hong Kong waters were identified in two previous studies through analysis of stomach contents collected from stranded cetaceans.[36,37] Additional information on the feeding habits of the two cetaceans can also be found in other research reports.[52−54] Finless porpoises and humpback dolphins around Hong Kong mainly prey on fishes, cephalopods, and occasionally shrimps, and fishes are the most commonly found prey. The diet of the two cetaceans residing in Hong Kong waters overlaps to some extent, as important preys (e.g., *J. heterolepis*, *C. arel*, *L. brevirostris*) are shared by them. However, dolphins favor prey species common in the estuary, whereas porpoises exploit more pelagic habitats for food. These prey preferences appear to be reflected in their distribution around Hong Kong.[14] Porpoises occur mainly in the southern and eastern waters of Hong Kong, but dolphins are seen mostly in northwestern waters close to the PRE.[55] On the basis of the available dietary information, we identified 7 prey species for porpoises and 14 for dolphins by integrating the reported marine organisms from the PRE[17] that are also the prey species for dolphins.

The prey list and calculated BMFs for the two cetacean species are shown in Table 2. Detailed data are compiled in Table S7 (SI). The evaluated BMFs for ∑SCCPs and ∑MCCPs between prey species and finless porpoises were all above 1 and in the range of 3.1−6.7 and 1.4−11, respectively. Higher BMFs for ∑SCCPs and ∑MCCPs between prey species and Indo-Pacific humpback dolphins were observed in the range of 11−38 and 23−58, respectively, signifying that biomagnification of SCCPs and MCCPs was more than 10-fold from the studied prey to dolphins. Because Indo-Pacific humpback dolphins prefer murky, brackish waters of the estuary, whereas finless porpoises prefer more clear, saline, and colder water,[36] the higher BMFs found in dolphins than in porpoises may result from different feeding and habitat preferences. The BMFs for ∑SCCPs in finless porpoises were slightly higher than those reported between prey and predators from Lake Ontario and Lake Michigan (0.54−3.6),[13] the PRE (1.1−3.4),[17] and the Fildes Peninsula of Antarctica (0.4−3.5),[19] while the BMFs for ∑SCCPs in Indo-Pacific humpback dolphins were much higher than all the reported values.[13,17,19] Houde et al.[13] reported that the BMFs for MCCP congeners from *Diporeia* to sculpin in Lake Ontario were between 2.7 and 14, comparable to the BMFs for ∑MCCPs found in finless porpoises but much lower than those in Indo-Pacific humpback dolphins. In addition, nearly equivalent or even larger BMFs for MCCPs than SCCPs were found in the two cetaceans.

Trophic transfers of SCCPs and MCCPs in the food web of Hong Kong waters were investigated to assess their biomagnification. As shown in Figure 3, significant positive relationships were found between trophic levels and lipid-normalized concentrations of ∑SCCPs ($R^2$ = 0.66, $p < 0.05$, RMSE = 0.21) in these marine species including marine mammals.[14,56] The deviations for 75.9% fitting data to regression values were within one RMSE. Similar significant relationships were also found between trophic levels and lipid-normalized concentrations of ∑MCCPs ($R^2$ = 0.56, $p < 0.05$, RMSE = 0.28).[14,56] The deviations for 79.3% fitting data to regression values were within one RMSE. The trophic magnification factors (TMFs) were calculated to be 4.29 and

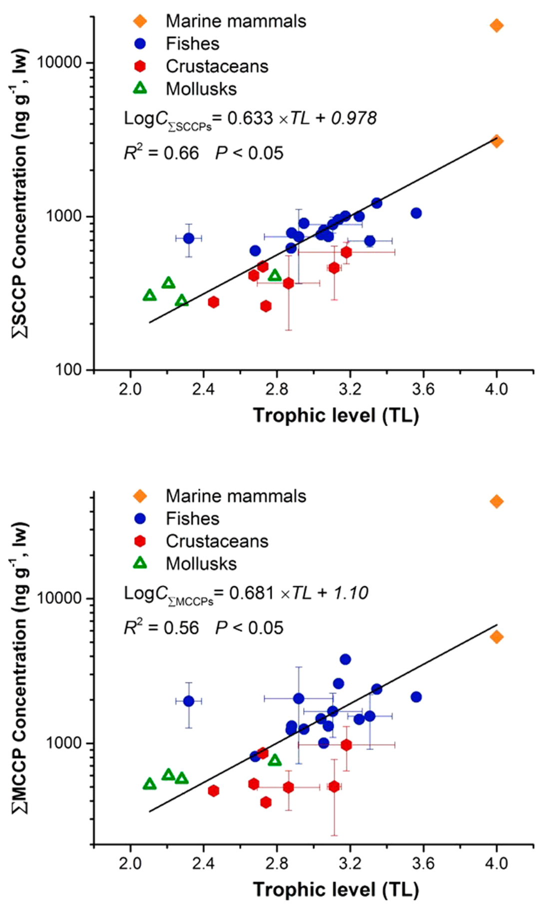


**Figure 3.** Trophic magnification of lipid-normalized concentrations of SCCPs (A, top) and MCCPs (B, bottom) with the estimated trophic level in the marine food web for marine mammals based on the results of stable isotope analysis.



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4.79 for SCCPs and MCCPs, respectively. The results of the two TMFs > 1 indicated that both SCCPs and MCCPs have trophic magnification potentials in the marine food web in Hong Kong waters. The observed TMF for ∑SCCPs in the present study was higher than that reported in a marine food web in Bohai Sea (2.38)[16] and in two freshwater food webs in Lake Gaobeidian (1.61)[28] and Lake Michigan (1.20).[13] The TMF values for MCCPs (4.79) were much higher than that (0.22) in freshwater food webs from Lake Ontario,[13] where MCCPs did not show trophic magnification. No other available field data on TMF for MCCPs can be used for further comparison.

Generally, the TMF values were highly dependent on food web configuration[57] and can be influenced by interspecific variations in ecological (e.g., food intake) and organismal parameters, including metabolism, reproductive status, migration, and age.[16] To our knowledge, among the available studies on biomagnification of SCCPs, only three[13,16,28] of the eight[13,16,17,19,28−31] food webs showed trophic magnification of SCCPs with TMF > 1. The previous study indicated that the TMFs of SCCP formula groups ($C_{10-13}$) displayed an increasing trend with increasing carbon chain length from 10 to 13 due to an increase of their log $K_{OW}$ values.[16] In this study, a slightly higher TMF of ∑MCCPs ($C_{14-17}$) than ∑SCCPs ($C_{10-13}$) was found, indicating that MCCPs dominated by $C_{14}$ may have a larger potential for trophic transfer and biomagnification.

In this study, we for the first time reported the elevated BMFs of CPs in marine mammals and corroborated the high biomagnification potential of not only SCCPs but also MCCPs in marine mammals by dietary bioaccumulation and trophic transfer. The elevated BMFs found in marine mammals should be a cause for concern regarding CP contamination. Our results point to the fact that organisms at higher trophic levels in the marine ecosystem, such as marine mammals, are potentially at higher risk associated with exposure to SCCPs and MCCPs, particularly MCCPs, which have higher biomagnification potential than SCCPs. Longer-chain CPs, including MCCPs and LCCPs, are considered as potential replacements for the banned SCCPs; however, the current status of these compounds in the emerging Asian regions is still unclear, and thus, investigation of their occurrence, distribution in the marine food web, and toxicological information is urgently needed. Further investigations are also recommended to fully assess the ecological and health risks of SCCPs and MCCPs in the marine ecosystem.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b02210.

Additional information on sample extraction and cleanup, sample information for analysis (Tables S1 and S2), detailed concentrations of individuals of the species (Table S3), concentrations of chain length groups (Table S4), relative abundance values or ranges of carbon chain groups (Table S5), BSAF and BMF data (Tables S6 and S7), concentration comparison among these marine species (Figure S1), concentration correlation analysis (Figures S2 and S3), carbon and chlorine homologue patterns (Figures S4 and S5), and average congener group abundance profiles (Figure S6) (PDF)

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### Notes

The authors declare no competing financial interest.

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