# Levels of short- and medium-chain chlorinated paraffins in edible insects and implications for human exposure

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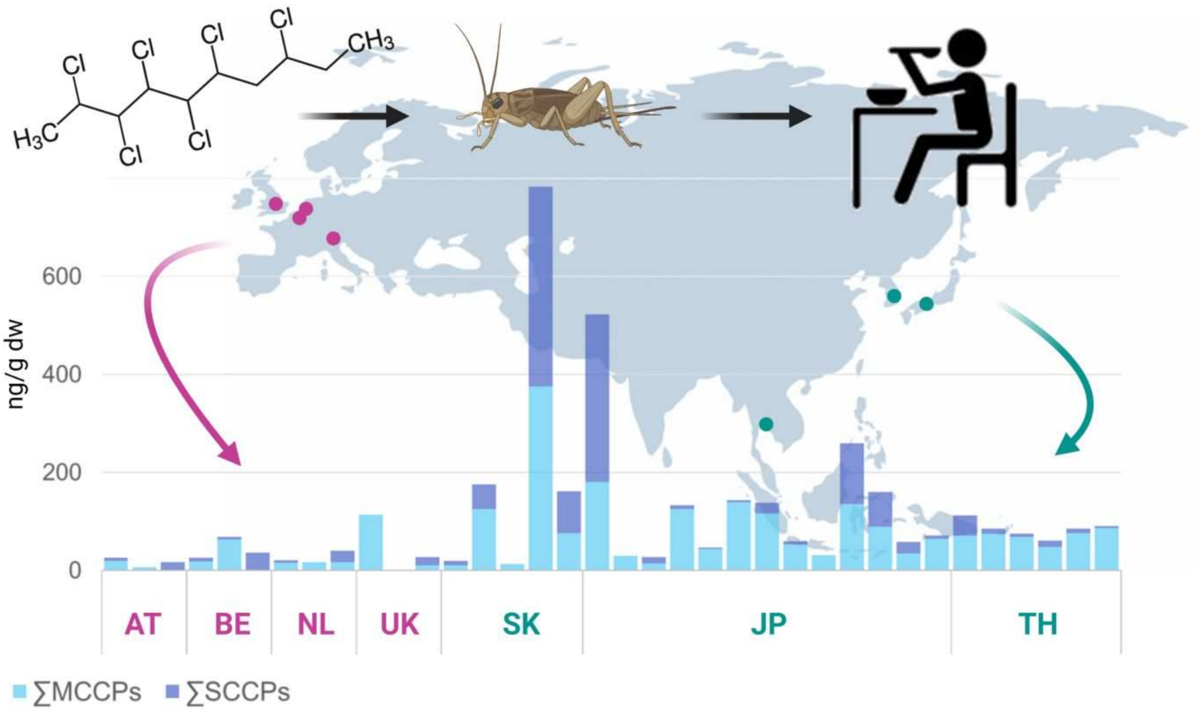

# Abstract

This study reports on the occurrence and distribution of short- and medium-chain chlorinated paraffins (SCCPs and MCCPs, respectively) in edible insects purchased from Asia and Europe. A total of 36 edible insect samples (*n*=24 from Asia, *n*=12 from Europe) authorized and prepared for human consumption were purchased and analyzed for SCCPs and MCCPs via gas chromatography and mass spectrometry. SCCPs were detected in 83% of all edible insect samples with an overall median ∑SCCP concentration of 8.7 ng/g dry weight (dw) and a range of <2.0 to 410 ng/g dw, while MCCPs were present in 92% of samples with a median ∑MCCP concentration of 51 ng/g dw and a range of <6.0 to 380 ng/g dw. Median ∑SCCP and ∑MCCP levels in edible insects purchased in Asia were approximately two- and four-times higher, respectively, than those from Europe, while the difference was statistically significant for ∑MCCPs ($p < 0.001$). Differences in homologue patterns were also observed between Asian and European samples to suggest diverse sources of CP contamination to insects which may include environmental accumulation, industrial processing equipment and food additives. Estimated daily intake of SCCPs and MCCPs via consumption of edible insects suggested that adverse health outcomes were very unlikely, but that continued monitoring of insect farming and processing practices are warranted.


Keywords: Chlorinated paraffins (CPs); SCCPs; MCCPs; edible insects; human exposure; Europe; Asia

Synopsis: The occurrence and distribution of SCCPs and MCCPs were investigated in edible insects. Health risks for the adult population via consumption of edible insects are likely to be low, but industrially produced insect-based foods require continuous monitoring.

# Introduction

The demand for food necessary to feed the growing world population is expected to increase by 75% by 2050.[1] Accordingly, the demand for livestock products and the increase in animal protein production is likely to add extra pressure on environmental systems, including water and land use. There is thus the urgent need to find alternative animal protein sources to protect food safety.[2] Among such alternatives, edible insects have attracted significant research interest and are praised for their elevated nutritional value[3] and environmental sustainability, compared with traditional livestock.[4]

Despite their beneficial health and environmental traits, the chemical and microbiological safety of edible insects remains a major factor determining their inclusion in the human diet. While edible insects have historically been harvested from the wild, insect farming is a more recent production method that allows the rearing of insects in controlled settings, where conditions can be constantly monitored.[5, 6] As with other foodstuffs, however, insects may accumulate potentially harmful contaminants such as persistent organic pollutants, veterinary drugs, mycotoxins, flame retardants, plasticizers and toxic metals during the rearing and processing phases. [6-8]

Chlorinated paraffins (CPs) have recently attracted increasing attention as a class of organic contaminants which can accumulate in foodstuffs.[9-11] CPs are chlorinated linear chain alkanes with the general formula $C_xH_{(2x+2-y)}Cl_y$. They are typically divided into three classes based on the number of carbon atoms present, namely short-chain CPs (SCCPs; $C_{10-13}$), medium-chain CPs (MCCPs; $C_{14-17}$) and long-chain CPs (LCCPs; $C_{>17}$), and by degree of chlorination varying between approximately 30% and 70% by weight.[12] CPs have been produced since the 1930s for a wide variety of industrial applications, mainly as plasticizers and flame retardants in polyvinyl chloride, rubber, lubricants, sealants, and other additives in consumer products.[13] Due to their potential toxicity,[14] environmental persistence[12] and ability to bioaccumulate and biomagnify in the food chain,[15] SCCPs have been recently classified as persistent organic pollutants (POPs) under the Stockholm Convention.[16] Such legal restrictions have consequently opened the way to the use of industrial replacements like MCCPs and LCCPs. However, since MCCPs have high global production volumes and exhibit similar physico-chemical properties and toxicity profiles to SCCPs (Ren et al., 2019), they too may represent threats to public health and the environment.[17]

Dietary intake is among the most prevalent human exposure pathways for CPs, alongside inadvertent dust ingestion, inhalation, and dermal absorption.[11, 18] Foodstuffs may become contaminated with CPs via bioaccumulation in the environment,[11] release from food processing equipment like hand blenders[19] or baking ovens,[20] and migration from plastic food packaging.[21]

Edible insects often undergo industrial processing via methods that may contribute to CP contamination such as freeze-drying, oven-drying or milling techniques to improve shelf life and acceptance as food,[4, 22] and are generally characterized by an elevated fat content (ranging on average from 13.4% for Orthoptera to 33.4% for Coleoptera),[3] which make them particularly prone to accumulation of CPs.

The aim of this study was to investigate, for the first time, the presence and homologue patterns of SCCPs and MCCPs in edible insects purchased from different countries in Asia and Europe. This research also evaluated the potential for exposure to CPs associated with insect consumption among adult populations to assess the safety of edible insects as a sustainable dietary alternative for animal-derived proteins.

# Experimental section

## *Sample collection*

Edible insects from Asia (South Korea-SK and Japan-JPN) and Europe (Austria-AT, Belgium-BE, the Netherlands-NL and United Kingdom-UK) were collected between September 2017 and January 2020 in the frame of previous investigations,[6, 8] while insects from Thailand-TH were collected in February 2021 (Table 1). All samples (*n*=36) were purchased from supermarkets and online shops and were authorized and ready for human consumption. Insects were characterized by different life stages (adult, *n*=22; larva, *n*= 10; pupae, *n*=4), rearing process (farmed, *n*=32; collected from the wild, *n*=4), and purchasing status (natural, *n*=24; seasoned, *n*=7; milled, *n*=5). “Natural” samples refer to those which were boiled and dehydrated, “seasoned” samples have been boiled and dehydrated followed by addition of flavorings like sugar or soy sauce to improve flavor, and “milled” samples have been boiled and dehydrated followed by grinding to flour. Individual insects included in each purchased sample were pooled, homogenized in a mortar, and stored in precleaned polypropylene tubes at - 20 °C pending analysis. The lipid content of each sample was determined gravimetrically, as detailed by Poma, et al. [8].

## *Sample preparation and extraction*

Homogenized insect samples (5 g) were weighed into 50 mL glass centrifuge vials and spiked with 5 ng of internal standard 1,5,5,6,6,10-hexachloro[$^{13}C_{10}$]decane ($^{13}$C-HCD). Samples were then vortexed for 1 min and ultrasonicated for 10 min in 20 mL of a 3:1 solution of *n*-hexane:dichloromethane. Following ultrasonication, the samples were centrifuged for 3 min at 2000 rpm and the supernatants transferred to clean 50 mL centrifuge vials. The extraction process was repeated once more with fresh solvent. Clean-up of sample extracts was achieved via direct addition of 6 mL concentrated sulphuric acid and vortexing for 1 min. A second clean-up step was performed by addition of 6 g of acidified silica (44% w/w sulphuric acid) to each sample extract. Purified extracts were then concentrated to approximately 0.5 mL and fractionated on prepacked 500 mg Agilent Bond Elut silica cartridges, which were first preconditioned with 6 mL of dichloromethane, positive-pressure dried, and then 6 mL *n*-hexane. After loading the sample, the first fraction was eluted with 6 mL of *n*-hexane to remove potentially interfering organohalogen contaminants (discarded) and the second fraction of 12 mL dichloromethane contained the CPs and internal standard. The final extract was concentrated to near dryness and reconstituted in 100 µL of *iso*-octane containing 5 ng of 6-methoxy-2,3,3’,4,4’,5’-hexabromodiphenyl ether (6-MeO-BDE-157) and ε-hexachlorocyclohexane (ε-HCH) external standards. Full details of standards and reagents are detailed in Section S1 of the Supporting Information (SI).

Table 1. Descriptive information for edible insect samples.

| ID | Order | Common name | Scientific name | Life stage | Purchase country/region | Origin country/region | Status | Farmed/ wild | Lipid content (%) |
|---|---|---|---|---|---|---|---|---|---|
| SK-1 | Orthoptera | grasshopper | *Oxya japonica Thunberg* | adult | South Korea | China | natural | farmed | 1.0 |
| SK-2 | Orthoptera | cricket | *Nemobius sylvestris* | adult | South Korea | China | natural | farmed | 4.0 |
| SK-3 | Coleoptera | mealworm | *Tenebrio molitor* | larva | South Korea | South Korea | natural | farmed | 12 |
| SK-4 | Coleoptera | mealworm | *Tenebrio molitor* | larva | South Korea | China | natural | farmed | 4.7 |
| SK-5 | Lepidoptera | silkworm | *Bombyx mori* | larva | South Korea | South Korea | seasoned | farmed | 8.6 |
| JPN-1 | Lepidoptera | silkworm | *Bombyx mori* | pupae | Japan | Japan | seasoned | farmed | 9.2 |
| JPN-2 | Trichoptera | caddisfly | *Stenopsyche marmorata* | larva | Japan | Japan | seasoned | wild | 0.4 |
| JPN-3 | Orthoptera | house cricket | *Acheta domesticus* | adult | Japan | Japan | milled | farmed | 2.6 |
| JPN-4 | Orthoptera | house cricket | *Acheta domesticus* | adult | Japan | Thailand | milled | farmed | 6.4 |
| JPN-5 | Orthoptera | black cricket | *Gryllus Bimaculatus* | adult | Japan | Thailand | milled | farmed | 7.8 |
| JPN-6 | Orthoptera | Jamaican field cricket | *Gryllus assimilis* | adult | Japan | Thailand | milled | farmed | 14 |
| JPN-7 | Orthoptera | mole cricket | *Gryllotalpa brachyptera* | adult | Japan | Thailand | natural | wild | 12 |
| JPN-8 | Coleoptera | diving beetle | *Dytiscus marginalis* | adult | Japan | Thailand | natural | wild | 8.1 |
| JPN-9 | Coleoptera | small june beetle | *Rhomborrhina japonica* | adult | Japan | Thailand | natural | wild | 1.8 |
| JPN-10 | Lepidoptera | silkworm | *Bombyx mori* | pupae | Japan | Thailand | milled | farmed | 8.2 |
| JPN-11 | Lepidoptera | silkworm | *Bombyx mori* | pupae | Japan | Thailand | natural | farmed | 16 |
| JPN-12 | Lepidoptera | shea caterpillar | *Cirina butyrospermi* | larva | Japan | Africa | natural | farmed | 6.8 |
| JPN-13 | Lepidoptera | shea caterpillar | *Cirina butyrospermi* | larva | Japan | Africa | seasoned | farmed | 16 |
| TH-1 | Lepidoptera | silkworm | *Bombyx mori* | pupae | Thailand | Thailand | natural | armed | 24 |
| TH-2 | Hymenoptera | weaver ant | *Oecophylla smaragdina* | adult | Thailand | Thailand | natural | farmed | 27 |
| TH-3 | Orthoptera | house cricket | *Acheta domesticus* | adult | Thailand | Thailand | natural | farmed | 16 |
| TH-4 | Orthoptera | house cricket | *Acheta domesticus* | adult | Thailand | Thailand | seasoned | farmed | 21 |
| TH-5 | Orthoptera | house cricket | *Acheta domesticus* | adult | Thailand | Thailand | seasoned | farmed | 12 |
| TH-6 | Orthoptera | house cricket | *Acheta domesticus* | adult | Thailand | Thailand | seasoned | farmed | 11 |
| AT-1 | Orthoptera | cricket | *Acheta domesticus* | adult | Austria | Austria | natural | farmed | 6.4 |
| AT-2 | Orthoptera | grasshopper | *Locusta migratoria* | adult | Austria | Austria | natural | farmed | 6.1 |
| AT-3 | Coleoptera | mealworm | *Tenebrio molitor* | larva | Austria | Austria | natural | farmed | 9.7 |
| BE-1 | Orthoptera | cricket | *Acheta domesticus* | adult | Belgium | Netherlands | natural | farmed | 5.4 |
| BE-2 | Orthoptera | grasshopper | *Locusta migratoria* | adult | Belgium | Netherlands | natural | farmed | 4.4 |
| BE-3 | Coleoptera | mealworm | *Tenebrio molitor* | larva | Belgium | Netherlands | natural | farmed | 10 |
| NL-1 | Orthoptera | cricket | *Acheta domesticus* | adult | Netherlands | Netherlands | natural | farmed | 7.9 |
| NL-2 | Orthoptera | grasshopper | *Locusta migratoria* | adult | Netherlands | Netherlands | natural | farmed | 8.1 |
| NL-3 | Coleoptera | mealworm | *Tenebrio molitor* | larva | Netherlands | Netherlands | natural | farmed | 9.9 |
| UK-1 | Orthoptera | cricket | *Acheta domesticus* | adult | United Kingdom | Netherlands | natural | farmed | 8.5 |
| UK-2 | Orthoptera | grasshopper | *Locusta migratoria* | adult | United Kingdom | Netherlands | natural | farmed | 6.8 |
| UK-3 | Coleoptera | mealworm | *Tenebrio molitor* | larva | United Kingdom | Netherlands | natural | farmed | 8.6 |

### *Instrumental analysis and quantification*

All analyses were conducted on an Agilent 7000D gas chromatograph-mass spectrometer (GC-MS) using electron capture negative ionization (ECNI) and single ion monitoring (SIM) mode. Injections of 2 µL were performed with a multimode inlet at a starting temperature of 92˚C, which was held for 0.04 min before ramping at a rate of 700˚C/min to 300˚C. An Agilent DB5-MS capillary column (15 m, 0.25 mm internal diameter, 0.1 µm film thickness) was fitted to the GC for analyses and the oven programmed to hold for 1.25 min at 90˚C, ramp at 25˚C/min to 180˚C and then 10˚C/min to 325˚C and hold for 6 min. Helium was the carrier gas applied at a flow rate of 1 mL/min for 17.85 min and then increased to 2.5 mL/min. The ECNI source was operated at 150˚C, the transfer line at 300˚C, and the quadrupoles set to 150˚C each. Acquisition of CP homologues ranging from $C_{10}$ to $C_{17}$ and $Cl_5$ to $Cl_{10}$ (24 SCCPs and 24 MCCPs) was achieved by monitoring the two most abundant isotopes of $[M-HCl]^-$ ions for the $Cl_{5-6}$ SCCPs and $[M-Cl]^-$ for all other groups (Table S1). The most abundant isotope for each homologue was used for quantification and the second isotope for confirmation reliant on CP 'peaks' exceeding a signal to noise ratio of 3. Quantification of ∑SCCPs and ∑MCCPs was performed using a modified version of the chlorine-content calibration procedure first described by Reth, et al. [23]. In brief, SCCP and MCCP concentrations in sample extracts were determined separately by first analyzing five-point chlorine-content gradient calibration curves constructed using technical mixtures of SCCPs (51.5, 55.5 and 63%Cl) and MCCPs (42, 52 and 57%Cl). The area of the most abundant isotope for each homologue was divided by the area of the 6-MeO-BDE-157 external standard to derive relative responses, which were summed to determine total relative responses per ∑SCCP and ∑MCCP technical standard. The total chlorine content was also determined for each standard as the mass fraction of Cl among the measured homologues. Exponential equations were fit to the relationship between the total relative response and the measured chlorine-content for each of the SCCP and MCCP calibration standards and used to determine the concentrations in analyzed sample extracts. Example calibration curves and detailed explanation of the quantification procedure is described by McGrath, et al. [24].

### *Statistical analysis*

Statistical analyses were carried out in Excel version 16 and SPSS 26. ∑SCCP and ∑MCCP levels have been expressed in ng/g dry weight (dw) of insect rather than per lipid content to represent the concentrations in the samples as purchased and with the assumption that all moisture was removed during the drying processes described on package labelling. Furthermore, while CPs may accumulate more readily in foods with higher lipid content, associations between the lipid content of edible insect samples and CP concentrations were weak and not statistically significant (∑SCCPs - Spearman ρ = 0.150, p = 0.486; MCCPs - Spearman ρ = 0.249, p = 0.211). None of the sample concentrations qualified as upper bound outliers, defined as concentrations exceeding the third quartile level by three times the interquartile range among all samples. Values determined to be below limits of quantification (LOQ) were replaced with half the LOQ for calculation of statistics (medium bound) except human exposure assessment (see Section S2). Q-Q plots determined SCCP and MCCP concentration data to each approximate a lognormal distribution and, thus, the data was log transformed prior to statistical comparison testing. Comparisons of CP concentrations between descriptive sample categories displayed in Table 1 were performed by ANOVA with a Tukey post-hoc test with $p < 0.05$ considered as statistically significant. Estimated daily intake (EDI) and Margin of Exposure (MOE) for CPs via consumption of edible insects were calculated for adult populations in Europe and Asia using the ∑SCCP and ∑MCCP concentrations measured in the insect samples and the European Food Safety Authority's (EFSA) recommended lower bench mark dose ($BMDL_{10}$).[25] Full details of the human exposure assessment calculations are provided in Section S2.

### *Quality assurance and quality control*

The analytical method employed has undergone comprehensive validation testing to confirm the accuracy and precision of SCCP and MCCP measurement in a variety of foods, as described by McGrath, et al. [24]. Continued method performance was ensured by the inclusion of a procedural blank and positive control sample with each batch of 12 edible insect samples. While no CP contamination was observed among the procedural blanks (*n*=3) in the present study, LOQs for SCCPs and MCCPs were set to meet a 95% confidence interval above the consistent low CP levels observed in 10 procedural blanks analyzed during comprehensive method validation.[24] LOQs were 2.0 ng/g dw insects for ∑SCCPs and 6.0 ng/g dw insects for ∑MCCPs according to the calculations described by McGrath, et al. [24]. Positive control samples (*n*=3) involved fortification of procedural blanks with 300 ng of both SCCP 55.5%Cl and MCCP 57%Cl technical mixtures, which equates to 60 ng/g dry weight (dw) each in insects. The European Union Reference Laboratory's (EURL) accuracy criteria[26] was met in each of the positive controls with ∑SCCPs ranging from 92 to 128% and ∑MCCPs ranging from 62 to 99% and relative standard deviation (RSD) of 18 and 25%, respectively. The $^{13}C$-HCD internal standard recovery ranged from 82 to 128% in all samples to demonstrate good extraction efficiency.

# Results and discussion

## *SCCP concentrations*

SCCPs were detected in 83% of all edible insect samples with an overall median ∑SCCP concentration of 8.7 ng/g dw and range of <2.0 to 410 ng/g dw (Figure 1, Table S3). The median level of ∑SCCPs measured in samples purchased in Asian countries ($n$=24), 9.3 ng/g dw, was slightly higher than the median observed among edible insects purchased in Europe ($n$=12), 5.5 ng/g dw, although the difference was not statistically significant ($p$ = 0.123). Of the samples analyzed in the present study, all edible insects purchased in European countries were also produced within Europe (Austria and the Netherlands), while all but two of the samples purchased in Asian countries also originated from Asia (China, South Korea, Japan, and Thailand). The overall maximum concentration of 410 ng/g dw was measured in a sample purchased from South Korea (SK-4) and produced in China. The elevated level observed in sample SK-4 may have been influenced by the high degree of CP manufacturing in China, which is the world's leading producer of CPs with production volumes estimated to have exceeded 1,000,000 t/y as of 2013 and approximately 150 CP production plants located throughout the country to act as potential point sources to the environment.[27, 28]

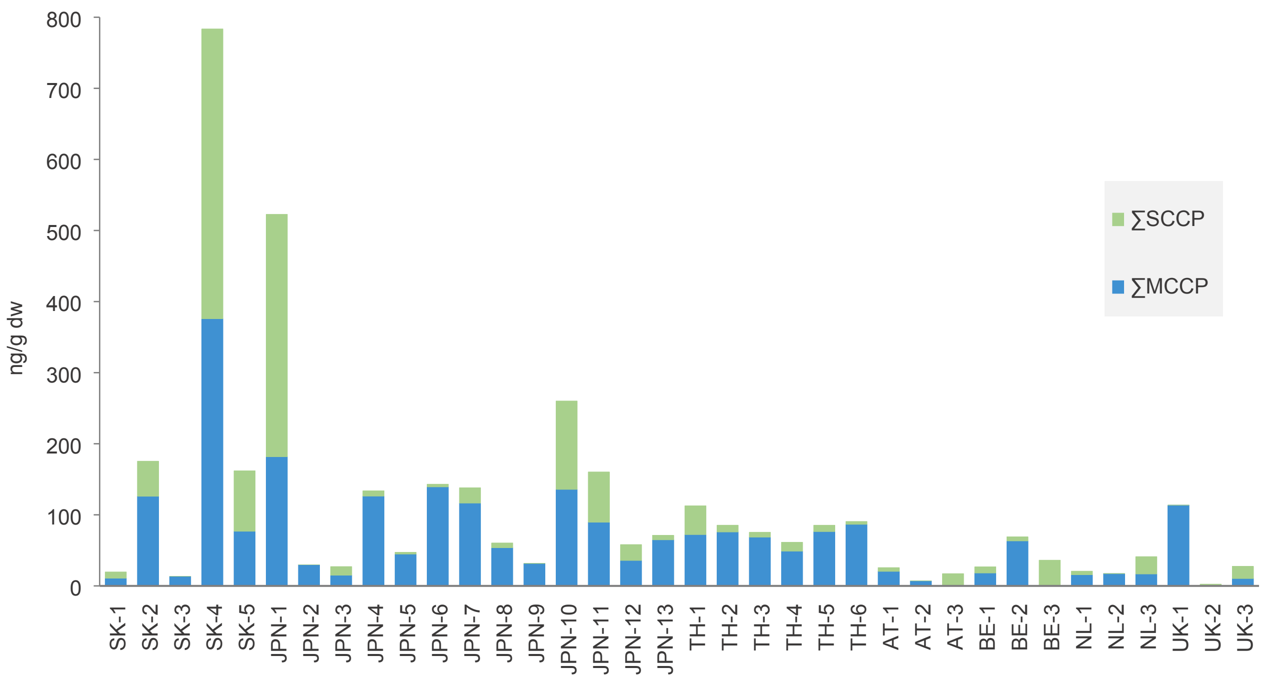


Figure 1. Concentrations (ng/g dw) of ∑SCCPs and ∑MCCPs in edible insect samples. SK= South Korea, JPN= Japan, TH= Thailand, AT= Austria, BE=Belgium, NL= Netherlands and UK= United Kingdom.

There were also two samples purchased from Japan in which ∑SCCP levels were particularly elevated with respect to the overall distribution, namely sample JPN-1 (340 ng/g dw) and JPN-9 (120 ng/g dw). Both samples have been subjected to post-harvest processing other than drying, in the form of seasoning for JPN-1 and milling for JPN-10. Seasonings may be an additional source of CP contamination in edible insects as many seasoning flavors are rich in lipids prone to accumulate CPs.[9] Milling of insects to flour also has the potential to contaminate insect products due to contact and processing by blenders shown to contain CPs within the lubricants and internal components of electric motors.[19] Too few samples were available for statistical comparison in this study between samples which have been seasoned (n=7) or milled (n=5) and those which have not. Given that SCCP concentrations in seasoned and milled samples were not consistently elevated within the dataset, it reasons that if contamination is indeed derived by such processing methods, it may only occur on a case-by-case basis depending on the specific additives or equipment involved. Finally, the contamination of edible insects with SCCPs could have been affected by other factors, such as intrinsic species-specific differences in accumulation and biotransformation capabilities and the insect life stage.

Due to the lack of previous CP data in edible insects, the concentrations of ∑SCCPs measured in this study were compared with those reported in common foods of animal origin (including meat, fish and seafood, eggs, and dairy products) and biota (Table 2).

Table 2. ∑SCCPs and ∑MCCPs concentrations in foods of animal origin and biota measured in this and other studies. To facilitate comparisons among the different studies, an estimation of wet weight CP concentrations (ng/g ww) in edible insects was derived based on an average unprocessed insect dry fraction of 0.5.

| Food type | ΣSCCPs Mean (±SD) | ΣSCCPs Median | ΣSCCPs Range | ΣMCCPs Mean (±SD) | ΣMCCPs Median | ΣMCCPs Range | Ref. |
|---|---|---|---|---|---|---|---|
| Edible insects (ng/g dw) | 38±87 | 8.7 | <2.0 - 410 | 66±71 | 51 | <6.0 - 380 | |
| *Edible insects (ng/g ww)* | *19±44* | *4.4* | *<1.0 - 210* | *33±35* | *26* | *<3.0 - 190* | This study |
| *Edible insects (ng/g lw)* | *570±1500* | *100* | *<10 - 8700* | *1100±1700* | *580* | *<30 - 8000* | |
| Infant food - infant formula (ng/g ww) | | 0.2 | | | 2.3 | | Perkons, et al. [29] |
| Infant food - meat or fish meal (ng/g ww) | | 0.9 | | | 1.6 | | |
| Meat (ng/g ww) | | <LOQ | <LOQ - 2.3 | | 7.7 | <LOQ - 27 | |
| Fish (ng/g ww) | | <LOQ | <LOQ - 2.7 | | 6.0 | <LOQ - 73 | McGrath, et al. [9] |
| Milk and dairy products (ng/g ww) | | <LOQ | <LOQ - 5.1 | | 5.1 | <LOQ - 22 | |
| Eggs (ng/g ww) | | | ND | | 10 | 3.3 - 16 | |
| Meat (ng/g ww) | | | <4.5 - 37* | | | | |
| Fish (ng/g ww) | | | <3.7 - 200* | | | | Krätschmer, et al. [30] |
| Milk and dairy products (ng/g ww) | | | <11 - 49* | | | | |
| Eggs (ng/g ww) | | | <3.5 - 40* | | | | |
| Infant food - infant formula (ng/g ww) | | 16.3 | 7.41 - 54.2 | | 11.1 | 3.04 - 20.9 | Han, et al. [31] |
| Infant food - meat puree (ng/g ww) | | 6.68 | 4.21 - 8.43 | | 2.14 | 1.24 - 5.41 | |
| Wild grasshoppers (ng/g dw) | | 300 | | | 410 | | Liu, et al. [32] |
| Wild crickets (ng/g dw) | | 920 | | | 580 | | |
| Meat (ng/g ww) | | 89.9 | | | | | |
| Fish (ng/g ww) | 180 | 159 | 19.1 - 629 | | | | Lee, et al. [10] |
| Dairy products (ng/g ww) | | 301 | | | | | |
| Eggs (ng/g ww) | | 286 | | | | | |
| Cow milk (industrial areas) (ng/g lw) | 1670 | | | 190 | | | Dong, et al. [33] |
| Cow milk (non-industrial areas) (ng/g lw) | 490 | | | 72 | | | |
| Freshwater fish (ng/g ww) | | | 0.3 - 10.6 | | | 1.3 - 73 | Labadie, et al. [34] |
| River fish (ng/g ww) | 50±34 | | 11 - 150 | | | | |
| Farmed freshwater fish (ng/g ww) | 40±34 | | 3.9 - 200 | | | | Huang, et al. [35] |
| Wild sea fish (ng/g ww) | 26±13 | | 8.1 - 65 | | | | |
| Milk powder (ng/g dw) | 18.3 | 18.1 | 16.2 - 20.5 | 14.2 | 17.6 | 1.70 - 23.3 | Gao, et al. [18] |
| Aquatic foods (ng/g ww) | 1472 | | 215 - 4200 | 80.5 | | 9.0 - 586 | Wang, et al. [36] |
| Meat and meat products (ng/g ww) | 129±4.1 | | | 5.7±0.6 | | | Huang, et al. [11] |

*data are for total ΣCPs.
SD = standard deviation

SCCP concentrations in edible insects were generally higher than those measured in foods from Belgium,[9] Germany,[37] in animal-based foods for infants from different European countries[29] and freshwater fish from France.[34] They were comparable to SCCP levels detected in animal-based foods for infants,[31] milk powder[18] and dairy products[33] from China, and lower than the levels found in other animal foods from Korea[10] and China.[11, 35, 36] In addition, the concentrations of SCCPs found in edible insects were one to two orders of magnitude lower than the levels measured in wild grasshoppers and crickets collected from an e-waste recycling site in South China.[32] This could be expected, due to high detection frequency and contamination of organisms collected from e-waste recycling regions,[38] and it highlights the relevance of rearing edible insects in controlled conditions.

### MCCPs concentrations

MCCPs were detected in 92% of insect samples with an overall median ∑MCCP concentration of 51 ng/g dw and a range of <6.0 to 380 ng/g dw (Figure 1, Table S3). ∑MCCP concentrations were significantly higher ($p < 0.001$) in the edible insects purchased from Asia (median, 74 ng/g dw; range, 11 to 380) than those from Europe (median, 16 ng/g dw; range <6.0 to 110 ng/g dw). The detection frequency of MCCPs in the Asian samples also exceeded that of the European insects, with rates of 100 and 75%, respectively. As with SCCPs it is possible that large-scale manufacturing of CPs in China has influenced MCCP concentrations in insect samples produced within China or in nearby South Korea and Japan due to long range atmospheric transport of pollutants.[12, 39] However, only a third of the samples purchased in Asia were produced in these three countries while 14 samples were from Thailand and another 2 from an un-specified country in Africa. The cause of significantly higher ∑MCCP concentrations among edible insects purchased in Asia remains, thus, unclear.

∑MCCP concentrations in edible insects were generally higher than those measured in animal-based foods for infants,[29, 31] common animal foods from Belgium,[9] dairy products from China,[18, 33] freshwater fish from France[34] and meat products from China.[11] Levels of MCCPs in edible insects from this study were, however, lower than those found in aquatic foods from China[36] and in wild grasshoppers and crickets from around an e-waste recycling site in South China[32] (Table 2).

### Homologue patterns

Concentrations of ∑MCCPs were greater than those of ∑SCCPs in 81% of samples with an average SCCP/MCCP ratio of 0.26. Spearman correlation analyses between the levels of ∑SCCPs and ∑MCCPs in the samples showed only weak associations (Spearman $\rho = 0.358$, $p = 0.052$) to indicate that contamination of edible insects by the two homologue groups is not likely to derive from a common source. The distribution of individual homologues (Figure ) was evaluated on the basis of relative response corrected for the fractional isotopic abundance of the monitored *m/z* values and Cl number according to the methods of Tomy, et al. [40]. While carbon-chain distributions varied to some degree between all samples, the SCCP patterns were somewhat distinct between samples purchased in Asia and Europe. The median proportions of $\sum C_{10}$, $\sum C_{11}$, $\sum C_{12}$ and $\sum C_{13}$ homologue groups were 20, 23, 22 and 33 % in the Asian samples and 27, 48, 19 and 7.2% in the European samples. While $C_{13}$ homologues have been observed to be dominant in SCCP technical products manufactured in Germany (Witaclor) and the UK (Cereclor),[41] a number of MCCP and LCCP technical mixtures manufactured in China (CP-42, CP-52 and CP-70) have also exhibited large proportions of $C_{13}$.[28] The greater levels of $C_{13}$ homologues observed in the edible insects from Asia may reflect the presence of these congener groups in MCCP and LCCP technical products manufactured in China, even as SCCP products are currently phased-out. The difference between SCCP homologue patterns in Asian and European insects may also hint at different sources of contamination. The shift toward shorter chain SCCPs in the European samples with respect to typical technical mixture profiles may suggest SCCP bioaccumulation to have occurred in the rearing environment, as preferential uptake of $C_{10}$ and $C_{11}$ homologues has been evidenced in other organisms.[42] Conversely, the similarities between homologue patterns in the Asian insects and commercial CP products might indicate a more direct transfer via processing equipment such as drying ovens or the lubricants in mechanical mixers, each of which have been evidenced to release SCCPs during operation.[19, 20] MCCP carbon-chain distributions were more consistent across samples from the two continents, with median $\sum C_{14}$, $\sum C_{15}$, $\sum C_{16}$ and $\sum C_{17}$ proportions of 47, 28, 18 and 5.8 % in insects purchased in Asia and 52, 27, 20 and 3.8 % in those from Europe. These homologue patterns were broadly representative of those in MCCP technical products from both China and Europe.[28, 41] The overall calculated chlorine content in the samples ranged from 61.1 to 64.2 % (median; 62.9 %) for SCCPs and 54.9 to 60.6 % (median; 58.1 %) for MCCPs. Typically, higher degrees of chlorination were apparent for SCCPs in the Asian samples, with median $\sum Cl_6$, $\sum Cl_7$, $\sum Cl_8$, $\sum Cl_9$ and $\sum Cl_{10}$ proportions of 17, 30, 33, 15 and 3.2 %, respectively, while a greater dominance of lower chlorinated homologues was observed in European samples with median proportions of 31, 41, 21, 5 and 1.4 %, respectively. $Cl_5$ SCCPs were only detected in one sample (SK-2) with a low proportional abundance of 2%. The predominance of lighter chlorine groups among the European samples may further indicate that preferential uptake of the less chlorinated SCCPs has occurred in the environment.[42] The chlorine distribution of MCCPs in edible insects from Asia were relatively consistent from sample to sample, with median proportions of $\sum Cl_6$, $\sum Cl_7$, $\sum Cl_8$, $\sum Cl_9$ and $\sum Cl_{10}$ measuring 7.9, 27, 36, 20 and 6.9 %, respectively (Figure 2). $Cl_5$ MCCPs were not detected in any of the samples. Although median proportions of the MCCP chlorine groups were broadly similar in European edible insects (11, 33, 32, 18 and 0.0% for $\sum Cl_6$, $\sum Cl_7$, $\sum Cl_8$, $\sum Cl_9$ and $\sum Cl_{10}$, respectively), the patterns differed greatly between individual samples from Europe, perhaps indicating specific sources relating to separate edible insect producers.

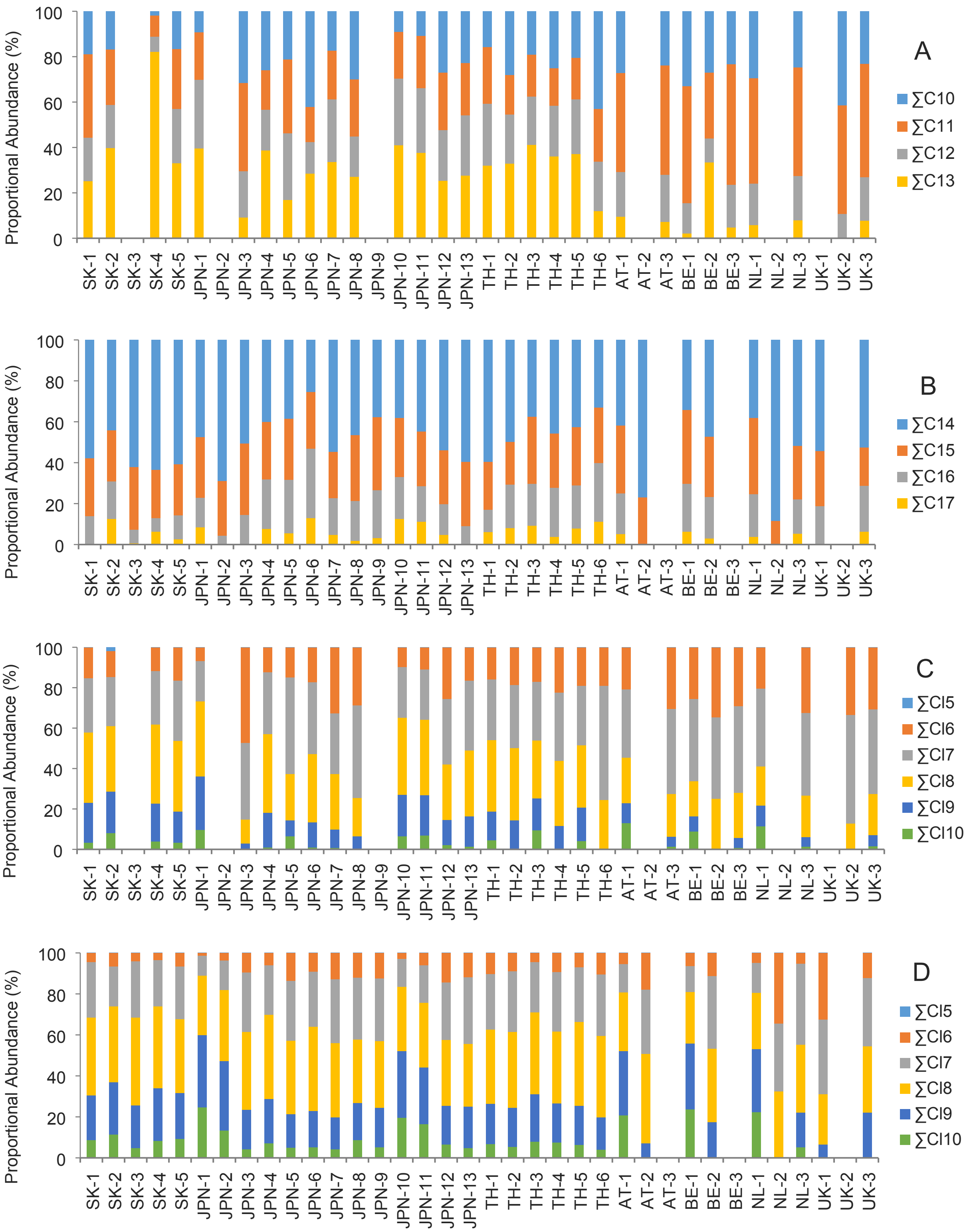


Figure 2. Proportional abundance of carbon-chain groups (A and B) and chlorine numbers (C and D) for SCCPs and MCCPs, respectively, in edible insect samples. The homologue distribution of SCCPs and MCCPs were calculated by correcting individual homologue relative responses for fractional isotopic abundance of monitored m/z species and chlorine number according to Tomy, et al. [40]. SK= South Korea, JPN= Japan, TH= Thailand, AT= Austria, BE=Belgium, NL= Netherlands and UK= United Kingdom. No data is shown for samples in which SCCPs or MCCPs were <LOQ.

### *Human exposure assessment*

An estimate of human exposure to SCCPs and MCCPs via consumption of edible insects was conducted for adult populations in Asia and Europe (Table 3). Based on the calculations of scenario A, for which it was assumed that the insect consumption habits of adults in Thailand are representative of adults in each of the other countries in this study, $50^{th}$ percentile deterministic EDIs for ∑SCCPs were 0.19 and 0.092 ng/kg bw/d for Asian and European adult populations, respectively. The $50^{th}$ percentile deterministic intakes of ∑MCCPs were estimated to be 1.5 and 0.27 ng/kg bw/d for Asian and European adults, respectively. The probabilistic $50^{th}$ percentile EDIs showed similar trends. In case 1 (*per capita* insect consumption maintained constant throughout the year and SCCP and MCCP concentrations varied from day to day), EDIs were 0.92 and 0.18 ng/kg bw/d for ∑SCCPs and 1.9 and 0.39 ng/kg bw/d for ∑MCCPs for Asia and European adults, respectively. In case 2 (both *per capita* insect consumption and SCCP and MCCP concentrations were constant throughout the year), EDIs were 0.28 and 0.093 ng/kg bw/d for ∑SCCPs and 1.2 and 0.23 ng/kg bw/d for ∑MCCPs in the Asian and European adult populations, respectively. Although the uniform application of Thai edible insect consumption rates may overestimate CP exposure for European populations, scenario A intake of SCCPs and MCCPs appears to represent very small contributions to total exposure to CPs via the diet. CP intake via edible insect consumption at the $50^{th}$ percentile EDI represented proportions of less than about 1% of the overall dietary CP exposure estimated for several Asian and European countries.[10, 18, 30, 43] Given the potential of edible insects to be a sustainable alternative for meat and other animal proteins, a second insect consumption scenario, scenario B, was considered to provide an estimation of human exposure in the instance that edible insects replaced all animal-derived protein in the adult diet. $50^{th}$ percentile deterministic CP intakes according to scenario B were approximately 65 times higher than those of scenario A, with Asian and European adult population exposure respectively estimated to be 12 and 6.3 ng/kg dw/d for ∑SCCPs and 94 and 19 ng/kg bw/d for ∑MCCPs. The large range of CP concentrations measured in edible insect samples resulted in much higher exposure rates estimated according to $95^{th}$ percentile concentrations, up to a maximum of 390 ng/kg bw/d ∑SCCPs in Asia.

For probabilistic estimates, in case 1, the $95^{th}$ percentile EDI was estimated at 110 ng/kg bw/d for ∑SCCPs and 210 ng/kg bw/d for ∑MCCPs in Asia, while it was much lower in Europe, with values of 23 and 49 ng/kg bw/d for ∑SCCPs and ∑MCCPs, respectively. Also, in case 2 (more conservative), the $95^{th}$ percentile EDI reached 230 ng/kg bw/d for ∑SCCPs and 380 ng/kg bw/d for ∑MCCPs in Asia, higher than in Europe (45 ng/kg bw/d for ∑SCCPs and 93 ng/kg bw/d for ∑MCCPs). According to estimates using scenario B, rates of

CP intake via consumption of edible insects were generally lower than intake from animal products in other studies. Animal products were reported to account for approximately half of SCCP total dietary exposure for adults in South Korea, equating to a median of ~490 ng/kg bw/d for 30-49 year old males, for example.[10] Among Chinese adults, median estimates of ∑SCCP intake via meat and meat products were also approximately an order of magnitude above those in the insects, 130 ng/kg bw/d, although ∑MCCP intake in the same study were much lower, 4.7 ng/kg bw/d.[11] In Germany, adult exposure to ∑SCCPs and ∑MCCPs was estimated to be 53 and 28 ng/kg bw/d, respectively, from mean CP levels in meat, eggs, dairy and fish.[30]

Table 3. Deterministic and probabilistic estimations of ∑SCCP and ∑MCCP intake (ng/kg bw/d) via consumption of insects among Asian and European populations.

| Insect consumption rate | | | CP concentration | Asia | | Europe | |
|---|---|---|---|---|---|---|---|
| | | | | ∑SCCPs | ∑MCCPs | ∑SCCPs | ∑MCCPs |
| Scenario A | Deterministic estimates | | 50$^{th}$ percentile | 0.19 | 1.5 | 0.092 | 0.27 |
| | | | 95$^{th}$ percentile | 6.3 | 3.6 | 0.49 | 1.4 |
| | Probabilistic estimates | Case 1 | 50$^{th}$ percentile | 0.92 | 1.9 | 0.18 | 0.39 |
| | | | 95$^{th}$ percentile | 1.7 | 3.2 | 0.33 | 0.69 |
| | | Case 2 | 50$^{th}$ percentile | 0.28 | 1.2 | 0.093 | 0.23 |
| | | | 95$^{th}$ percentile | 3.8 | 6.2 | 0.61 | 1.2 |
| Scenario B | Deterministic estimates | | 50$^{th}$ percentile | 12 | 94 | 6.3 | 19 |
| | | | 95$^{th}$ percentile | 390 | 220 | 34 | 98 |
| | Probabilistic estimates | Case 1 | 50$^{th}$ percentile | 58 | 110 | 13 | 25 |
| | | | 95$^{th}$ percentile | 110 | 210 | 23 | 49 |
| | | Case 2 | 50$^{th}$ percentile | 17 | 78 | 6.7 | 18 |
| | | | 95$^{th}$ percentile | 230 | 380 | 45 | 93 |

Scenario A; the mean edible insect intake reported for adults in Thailand, 1.3 g/d, was applied for adults in both the Asian and European population categories. Scenario B; insect consumption replaces all protein intake from animal products, equating to 80 and 88 g dw/d insects for Asian and European populations, respectively. Based on the deterministic estimations, probabilistic estimates were conducted by Monte Carlo simulations. Those estimates were conducted in the following two cases for both scenario A and B: in case 1, a group of 1,000 people where *per capita* insect consumption was constant throughout the 365 days and SCCP and MCCP concentrations varied from day to day; in case 2, a group of 1,000 people where both *per capita* insect consumption and SCCP and MCCP concentrations were constant throughout the 365 days (a more conservative estimate).

The margin of exposure (MOE) approach was applied to evaluate potential health risks posed by intake of CPs via consumption of edible insects, according to recommendations of EFSA[25]. 50$^{th}$ percentile MOEs based on deterministic EDIs were considerably higher than the $1.0 \times 10^3$ threshold above which health concerns are considered unlikely,[25] ranging from $1.2 \times 10^7$ to $1.3 \times 10^8$ for scenario A and $2.1 \times 10^5$ to $2.0 \times 10^6$ for scenario B (Table S4). Deterministic MOEs calculated from 95$^{th}$ percentile concentrations were also more than 2 orders of magnitude above the $1.0 \times 10^3$ threshold in all instances except for scenario B exposure to

SCCPs in Asia ($5.9x10^3$) and Europe ($6.8x10^4$). Results consistently above the $1.0x10^3$ threshold were also obtained when considering the 50th and 95th percentile MOEs of probabilistic estimates (Table S4), suggesting that the health risks posed by SCCP and MCCP ingestion via insect consumption are very low. It should be noted that the EDIs calculated in this study represent only a proportion of total human exposure to CPs such that MOEs reported here can only provide approximate indications of health risks. Toxicokinetic data is also currently limited for SCCPs and MCCPs, such that health risk assessments should be reconsidered upon the release of new findings. This study provides, for the first time, occurrence data on SCCPs and MCCPs in edible insects from several countries in Asia and Europe and an evaluation of the potential human exposure implications. Edible insects are considered as a more sustainable source of dietary protein compared to other animal-based foods, though few studies so far have examined the safety of insects in a chemical contaminant context. This research suggests that SCCPs and MCCPs are widespread in insect-based foods from both Asia and Europe, with higher concentrations among Asian-bought products indicating region-specific contamination sources. An overall dominance of MCCPs among samples may reflect international legislative actions to reduce SCCP usage while the incidence of elevated SCCP levels in a small number of samples illustrates that specific point-sources to insects persist. Analysis of CP homologue distribution in the samples suggests that complex sources of contamination may include environmental bioaccumulation, industrial processing equipment and food additives. While human exposure and risk assessments indicated that edible insects are unlikely to pose health threats, exposure rates estimated according to the most conservative scenarios suggested that continued monitoring of edible insect farming and processing practices is warranted.

## Supporting Information

Additional experimental details (GC-ECNI/MS SIM acquisition parameters for SCCPs and MCCPs) and materials and methods (standards and reagents); Concentrations of ∑SCCPs and ∑MCCPs in individual samples; Additional information concerning edible insect consumption and calculated margins of exposure.

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## References

(1) van Huis, A.; Dicke, M.; van Loon, J. J. A. Insects to feed the world. *Journal of Insects as Food and Feed* **2015**, *1* (1), 3-5. DOI: 10.3920/JIFF2015.x002.

(2) Lee, J. H.; Kim, T.-K.; Jeong, C. H.; Yong, H. I.; Cha, J. Y.; Kim, B.-K.; Choi, Y.-S. Biological activity and processing technologies of edible insects: a review. *Food Science and Biotechnology* **2021**, *30* (8), 1003-1023. DOI: 10.1007/s10068-021-00942-8.

(3) Rumpold, B. A.; Schlüter, O. K. Nutritional composition and safety aspects of edible insects. *Molecular Nutrition & Food Research* **2013**, *57* (5), 802-823. DOI: 10.1002/mnfr.201200735.

(4) Baiano, A. Edible insects: An overview on nutritional characteristics, safety, farming, production technologies, regulatory framework, and socio-economic and ethical implications. *Trends in Food Science & Technology* **2020**, *100*, 35-50. DOI: 10.1016/j.tifs.2020.03.040.

(5) Papastavropoulou, K.; Koupa, A.; Kritikou, E.; Kostakis, M.; Proestos, C. Edible insects: Benefits and potential risk for consumers and the food industry. *Biointerface Research in Applied Chemistry* **2022**, *12* (4), 5131-5149. DOI: 10.33263/BRIAC124.51315149.

(6) Poma, G.; Fujii, Y.; Lievens, S.; Bombeke, J.; Gao, B.; Jeong, Y.; McGrath, T. J.; Covaci, A. Occurrence, patterns, and sources of hazardous organic chemicals in edible insects and insect-based food from the Japanese market. *Food and Chemical Toxicology* **2021**, *154*, 112311. DOI: 10.1016/j.fct.2021.112311.

(7) Meyer, A. M.; Meijer, N.; Hoek-van den Hil, E. F.; van der Fels-Klerx, H. J. Chemical food safety hazards of insects reared for food and feed. *Journal of Insects as Food and Feed* **2021**, *7* (5), 823-831.

(8) Poma, G.; Yin, S.; Tang, B.; Fujii, Y.; Cuykx, M.; Covaci, A. Occurrence of Selected Organic Contaminants in Edible Insects and Assessment of Their Chemical Safety. *Environmental Health Perspectives* **2019**, *127* (12), 127009. DOI: 10.1289/EHP5782.

(9) McGrath, T. J.; Limonier, F.; Poma, G.; Bombeke, J.; Winand, R.; Vanneste, K.; Andjelkovic, M.; Van Hoeck, E.; Joly, L.; Covaci, A. Concentrations and distribution of chlorinated paraffins in Belgian foods. *Environmental Pollution* **2021**, *291*, 118236. DOI: 10.1016/j.envpol.2021.118236.

(10) Lee, S.; Choo, G.; Ekpe, O. D.; Kim, J.; Oh, J. E. Short-chain chlorinated paraffins in various foods from Republic of Korea: Levels, congener patterns, and human dietary exposure. *Environmental Pollution* **2020**, *263*. DOI: 10.1016/j.envpol.2020.114520.

(11) 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. *Environmental Pollution* **2018**, *233*, 439-445. DOI: 10.1016/j.envpol.2017.10.022.

(12) Wei, G. L.; Liang, X. L.; Li, D. Q.; Zhuo, M. N.; Zhang, S. Y.; Huang, Q. X.; Liao, Y. S.; Xie, Z. Y.; Guo, T. L.; Yuan, Z. J. Occurrence, fate and ecological risk of chlorinated paraffins in Asia: A review. *Environment International* **2016**, *92-93*, 373-387. DOI: 10.1016/j.envint.2016.04.002.

(13) van Mourik, L. M.; Gaus, C.; Leonards, P. E. G.; de Boer, J. Chlorinated paraffins in the environment: A review on their production, fate, levels and trends between 2010 and 2015. *Chemosphere* **2016**, *155*, 415-428. DOI: 10.1016/j.chemosphere.2016.04.037.

(14) Ren, X.; Geng, N.; Zhang, H.; Wang, F.; Gong, Y.; Song, X.; Luo, Y.; Zhang, B.; Chen, J. Comparing the disrupting effects of short-, medium- and long-chain chlorinated paraffins on cell viability and metabolism. *Science of The Total Environment* **2019**, *685*, 297-307. DOI: 10.1016/j.scitotenv.2019.05.388.

(15) Yuan, B.; Vorkamp, K.; Roos, A. M.; Faxneld, S.; Sonne, C.; Garbus, S. E.; Lind, Y.; Eulaers, I.; Hellström, P.; Dietz, R.; Persson, S.; Bossi, R.; de Wit, C. A. Accumulation of short-, medium-, and long-chain chlorinated paraffins in marine and terrestrial animals from Scandinavia. *Environmental Science & Technology* **2019**, *53* (7), 3526-3537. DOI: 10.1021/acs.est.8b06518.

(16) UNEP. *UNEP/POPS/COP.8/SC-8/11. SC-8/11, Listing of Short-Chain Chlorinated Paraffins.* 2017. http://chm.pops.int/Portals/0/download.aspx?d=UNEP-POPS-COP.8-SC-8-11.English.pdf (accessed 20/5/2020).

(17) Glüge, J.; Schinkel, L.; Hungerbühler, K.; Cariou, R.; Bogdal, C. Environmental risks of medium-chain chlorinated paraffins (MCCPs): A review. *Environmental Science and Technology* **2018**, *52* (12), 6743-6760. DOI: 10.1021/acs.est.7b06459.
(18) 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. *Environmental Science and Technology* **2018**, *52* (1), 32-39. DOI: 10.1021/acs.est.7b04657.
(19) Yuan, B.; Strid, A.; Darnerud, P. O.; de Wit, C. A.; Nyström, J.; Bergman, Å. Chlorinated paraffins leaking from hand blenders can lead to significant human exposures. *Environment International* **2017**, *109*, 73-80. DOI: 10.1016/j.envint.2017.09.014.
(20) Sprengel, J.; Rixen, S.; Kappenstein, O.; Vetter, W. Transport of chlorinated paraffins (CPs) from baking oven doors into the food. *Food Chemistry: X* **2021**, *10*, 100122. DOI: 10.1016/j.fochx.2021.100122.
(21) Wang, C.; Gao, W.; Liang, Y.; Jiang, Y.; Wang, Y.; Zhang, Q.; Jiang, G. Migration of chlorinated paraffins from plastic food packaging into food simulants: Concentrations and differences in congener profiles. *Chemosphere* **2019**, *225*, 557-564. DOI: 10.1016/j.chemosphere.2019.03.039.
(22) Ayieko, I. A.; Onyango, M.; Ngadze, R. T.; Ayieko, M. A. Edible Insects as New Food Frontier in the Hospitality Industry. *Frontiers in Sustainable Food Systems* **2021**, *5*.
(23) Reth, M.; Zencak, Z.; Oehme, M. New quantification procedure for the analysis of chlorinated paraffins using electron capture negative ionization mass spectrometry. *Journal of Chromatography A* **2005**, *1081* (2), 225-231. DOI: 10.1016/j.chroma.2005.05.061.
(24) McGrath, T. J.; Poma, G.; Bombeke, J.; Limonier, F.; Van Hoeck, E.; Joly, L.; Covaci, A. Optimization and validation of an analytical method for the quantification of short- and medium-chained chlorinated paraffins in food by gas chromatography-mass spectrometry. *Food Control* **2021**, *119*, 107463. DOI: 10.1016/j.foodcont.2020.107463.
(25) EFSA CONTAM Panel; Schrenk, D.; Bignami, M.; Bodin, L.; Chipman, J. K.; del Mazo, J.; Grasl-Kraupp, B.; Hogstrand, C.; Hoogenboom, R.; Leblanc, J.-C.; Nebbia, C. S.; Ntzani, E.; Petersen, A.; Sand, S.; Schwerdtle, T.; Vleminckx, C.; Wallace, H.; Brüschweiler, B.; Leonards, P. E. G.; Rose, M.; Binaglia, M.; Horvath, Z.; Ramos Bordajandi, L.; Nielsen, E. Risk assessment of chlorinated paraffins in feed and food. *EFSA Journal* **2020**, *18* (3), 220. DOI: 10.2903/.
(26) EURL. *Interlaboratory Study on Determination of Chlorinated Paraffins in Coconut Fat*; European Union Reference Laboratory for Halogenated POPs in Feed and Food, Freiburg, Germany, 2018.
(27) Glüge, J.; Wang, Z.; Bogdal, C.; Scheringer, M.; Hungerbühler, K. Global production, use, and emission volumes of short-chain chlorinated paraffins – A minimum scenario. *Science of The Total Environment* **2016**, *573*, 1132-1146. DOI: 10.1016/j.scitotenv.2016.08.105.
(28) Li, T.; Gao, S.; Ben, Y.; Zhang, H.; Kang, Q.; Wan, Y. Screening of Chlorinated Paraffins and Unsaturated Analogues in Commercial Mixtures: Confirmation of Their Occurrences in the Atmosphere. *Environmental Science and Technology* **2018**, *52* (4), 1862-1870. DOI: 10.1021/acs.est.7b04761.
(29) Perkons, I.; Abdulajeva, E.; Bartkiene, E.; Zacs, D. Short- and medium-chain chlorinated paraffins in commercial complementary baby food produced in different European countries: Occurrence, congener group profiles, portion-based dietary intake, and risk assessment. *Science of The Total Environment* **2022**, *814*. DOI: 10.1016/j.scitotenv.2021.152733.
(30) Krätschmer, K.; Schächtele, A.; Vetter, W. Short- and medium-chain chlorinated paraffin exposure in South Germany: A total diet, meal and market basket study. *Environmental Pollution* **2021**, *272*. DOI: 10.1016/j.envpol.2020.116019.
(31) Han, X.; Chen, H.; Deng, M.; Du, B.; Zeng, L. Chlorinated paraffins in infant foods from the Chinese market and estimated dietary intake by infants. *Journal of Hazardous Materials* **2021**, *411*. DOI: 10.1016/j.jhazmat.2021.125073.
(32) Liu, Y.; Luo, X.; Zeng, Y.; Wang, Q.; Tu, W.; Yang, C.; Mai, B. Trophic magnification of short- and medium-chain chlorinated paraffins in terrestrial food webs and their bioamplification in insects and amphibians during metamorphosis. *Environmental Science and Technology* **2020**, *54* (18), 11282-11291. DOI: 10.1021/acs.est.0c03096.
(33) Dong, S.; Zhang, S.; Li, X.; Wei, S.; Li, T.; Zou, Y.; Zhang, W.; Cheng, J.; Wang, R.; Wang, P.; Su, X. Occurrence of short- and medium-chain chlorinated paraffins in raw dairy cow milk from five Chinese provinces. *Environment International* **2020**, *136*, 105466. DOI: 10.1016/j.envint.2020.105466.
(34) Labadie, P.; Blasi, C.; Le Menach, K.; Geneste, E.; Babut, M.; Perceval, O.; Budzinski, H. Evidence for the widespread occurrence of short- and medium-chain chlorinated paraffins in fish collected from the Rhône River basin (France). *Chemosphere* **2019**, *223*, 232-239. DOI: 10.1016/j.chemosphere.2019.02.069.
(35) Huang, Y.; Qing, X.; Jiang, G.; Chen, L.; He, Q.; Meng, X. Z.; Gao, B. Short-chain chlorinated paraffins in fish from two developed regions of China: Occurrence, influencing factors and implication for human exposure via consumption. *Chemosphere* **2019**, *236*. DOI: 10.1016/j.chemosphere.2019.07.048.
(36) Wang, R.; Gao, L.; Zheng, M.; Tian, Y.; Li, J.; Zhang, L.; Wu, Y.; Huang, H.; Qiao, L.; Liu, W.; Su, G.; Liu, G.; Liu, Y. Short- and medium-chain chlorinated paraffins in aquatic foods from 18 Chinese provinces: Occurrence, spatial distributions, and risk assessment. *Science of The Total Environment* **2018**, *615*, 1199-1206. DOI: 10.1016/j.scitotenv.2017.09.327.

(37) Krätschmer, K.; Schächtele, A.; Vetter, W. Chlorinated paraffins in baby food from the German market. *Food Control* **2021**, *123*. DOI: 10.1016/j.foodcont.2020.107689.

(38) Guan, K. L.; Liu, Y.; Luo, X. J.; Zeng, Y. H.; Mai, B. X. Short- and medium-chain chlorinated paraffins in aquatic organisms from an e-waste site: Biomagnification and maternal transfer. *Science of The Total Environment* **2020**, *708*, 134840. DOI: 10.1016/j.scitotenv.2019.134840.

(39) Vorkamp, K.; Rigét, F. F. A review of new and current-use contaminants in the Arctic environment: Evidence of long-range transport and indications of bioaccumulation. *Chemosphere* **2014**, *111*, 379-395. DOI: 10.1016/j.chemosphere.2014.04.019.

(40) Tomy, G. T.; Stern, G. A.; Muir, D. C. G.; Fisk, A. T.; Cymbalisty, C. D.; Westmore, J. B. Quantifying C10-C13 polychloroalkanes in environmental samples by high-resolution gas chromatography/electron capture negative ion high-resolution mass spectrometry. *Analytical Chemistry* **1997**, *69* (14), 2762-2771.

(41) Bogdal, C.; Alsberg, T.; Diefenbacher, P. S.; Macleod, M.; Berger, U. Fast quantification of chlorinated paraffins in environmental samples by direct injection high-resolution mass spectrometry with pattern deconvolution. *Analytical Chemistry* **2015**, *87* (5), 2852-2860. DOI: 10.1021/ac504444d.

(42) 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. *Science of The Total Environment* **2018**, *615*, 1010-1018. DOI: 10.1016/j.scitotenv.2017.10.026.

(43) FPS. *Presence of chlorinated paraffins in foodstuffs and intake of the Belgian population*; Federal Public Service, Health, Food Chain Safety and Environment, Brussels, Belgium, 2021.