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Environmental Risks of Medium-Chain Chlorinated Paraffins (MCCPs): A Review

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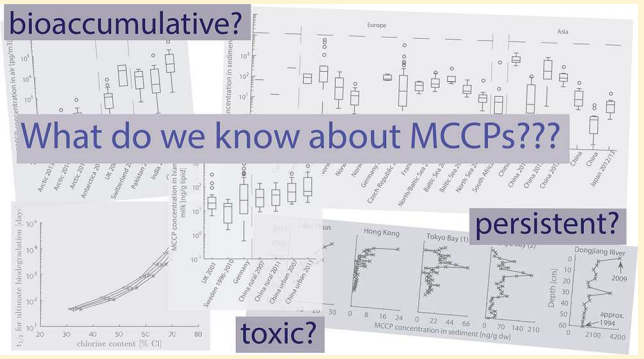
Supporting Information

ABSTRACT: Chlorinated paraffins are industrial chemicals that can be subdivided into short-chain (SCCP), medium-chain (MCCP), and long-chain (LCCP) chlorinated paraffins. The global production volumes of MCCPs are nowadays suspected to be much higher than those of S- and LCCPs, and the few available studies on the environmental occurrence of chlorinated paraffins report often higher MCCP concentrations than S- or LCCP concentrations in the environment. The present review focuses, therefore, on MCCPs specifically and provides a literature overview and a data analysis of the production volumes, PBT properties (persistence, bioaccumulation potential, and toxicity), and the worldwide measured concentrations of MCCP in environmental samples, biota, and humans. Furthermore, we include our own measurements of technical CP formulations from China, the major global producing country, to estimate the global production amounts of MCCPs. The key findings from this review are that (1) MCCPs are toxic to the aquatic environment, and the available data suggest that they are also persistent; (2) available time trends for MCCPs in soil, biota, and most of the sediment cores show increasing time trends over the last years to decades; and (3) MCCP concentrations in sediment close to local sources exceed toxicity thresholds (i.e., the PNEC). Our study shows that overall, MCCPs are of growing concern, and regulatory actions should be considered seriously.



INTRODUCTION

Polychlorinated *n*-alkanes, also known as chlorinated paraffins (CPs), are complex industrial chemicals with the molecular formula $C_nH_{2n+2-x}Cl_x$ and a chlorination degree varying from 30% to 70% by weight. According to their carbon chain length, CPs are subdivided into short-chain CPs (SCCPs, C_{10-13}), medium-chain CPs (MCCPs, C_{14-17}), and long-chain CPs (LCCPs, C_{18-30}). Depending on the alkane chain lengths and chlorination degrees, different technical mixtures of S-, M-, and LCCPs have different physicochemical properties[1] and are, therefore, used in various applications. In general, CPs are mainly used as flame retardants, plasticizers, and extreme-pressure additives.[2] Production and use volumes of total CPs are nowadays higher than ever before and exceeded 1 000 000 t in 2013.[3−5] Therefore, CPs are among the industrial chemicals with the highest production volumes.

In the last years, the focus of the research community and of regulatory authorities, with respect to CPs, was mainly on SCCPs.[6,7] SCCPs have shown to be persistent,[8−10] bioaccumulative,[11] and toxic,[12] whereas for MCCPs, these PBT properties are less studied and still a matter of debate. MCCPs are more difficult to quantify accurately than SCCPs,[13] and many studies have reported only SCCP (but no MCCP) concentrations.

The available data suggest, however, that especially MCCPs could be of critical concern to the environment. The global production amounts of MCCPs are nowadays suspected to be much higher than those of SCCPs,[14] and MCCPs are quite often measured in higher concentrations in the environment than SCCPs. The U.S. Environmental Protection Agency,[15,16] Environment and Health Canada,[17,18] the European Union (EU),[19,20] and the European Chemicals Agency (ECHA)[21] have assessed the risks of MCCPs, but no production or use prohibitions or restrictions are yet in force. This is different for SCCPs that have been substantially restricted in the EU in 2000 and have also been listed in Annex A of the Stockholm Convention on Persistent Organic Pollutants (POPs) (meaning a global ban with time-limited specific exemptions) in 2017. In addition to these global treaties, several national and regional restrictions on the production and use of SCCPs have been enforced.[14] It is expected that due to their listing in Annex A of the Stockholm Convention, the production and use of SCCPs will decrease in the future, and SCCPs will be replaced in most

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applications. The risk management evaluation adopted by the Persistent Organic Pollutants Review Committee proposed alternative products and processes for SCCPs and suggested, among other chemicals, also MCCPs as alternatives for SCCPs.[2] However, they stated clearly that most of the alternatives that have been identified in the risk management evaluation have not been assessed under the Convention and that it is unknown if some of these would exhibit POP-like properties.

The present review aims, therefore, to support the ongoing risk assessment of MCCPs and provides a literature overview and a data analysis of the production volumes, PBT properties, and the worldwide measured concentrations of MCCP in environmental samples, biota, and humans after 1995. Furthermore, we include measurements of the composition of technical CPs formulations from the major global producing country, China. So far, only a few technical mixtures have been analyzed for S- and MCCPs. Additional information on the composition of the technical CPs formulations is, therefore, highly important for estimating global production volumes of MCCPs.

Previous scientific reviews focused always on both SCCPs and MCCPs,[6,22−24] but the amount of information on SCCPs always surpassed those on MCCPs and limited a critical assessment of the MCCPs. Our review focuses, therefore, specifically on MCCPs and shows information on SCCPs only for comparison.

PRODUCTION AND USE

Production amounts for MCCPs have been reported for North America (17 800 t in 1998),[25] the United Kingdom (40 000 t in 1991),[26] and Russia (21 000 t in 2007 and 27 000 t in 2011)[4] (Table S1). Production capacities (but no production amounts) have been reported for the EU (45 000−160 000 t in 2004)[19] and Thailand (20 000 t in 1994).[27] Production amounts for MCCPs from the Asian market are not available because manufacturer in Asia do not differentiate between S-, M-, and LCCPs. However, some information is available on the produced technical CP mixtures in China. The International Chlorinated Alkanes Industry Association (ICAIA) stated for example that nearly 90% of the CP mixtures produced in China in 2012 were CP-52.[3] CP-52 is characterized by a chlorine content of 52% and can include CPs of all chain length. One study has analyzed the fractions of S-, M-, and LCCPs in three CP-52 mixtures,[28] and four more measurements are available for the fraction of SCCPs in CP-52.[29−32] We analyzed ourselves 11 CP-52 mixtures, obtained in 2017 from nine different producers in China and determined the fractions of S-, M-, and LCCPs (section S1.2 in the Supporting Information). MCCPs in CP-52 ranged (with one exception) between 29% and 67%, with a mean value of 57%. LCCPs were in all CP-52 mixtures below 12% with a mean of 4% and SCCPs ranged (with the one exception) between 24% and 63%. We do not know how representative the samples are, given that there are more than 120 CP producers in China,[14] but the measurements give at least a first indication of the range of the MCCPs in CP-52 as they cover around 10% of the producers. If we take the latest available production figure from China (1 050 000 t in 2013)[5] and assume that the manufacturer did not change the composition of CP-52 in the last five years to a great extent, MCCPs might have been produced in the order of 600 000 t in China in 2013. This number is much larger than any of the production amounts reported in literature for North America, Russia, or the EU and indicates that China was the largest producer of MCCPs in 2013. Data for the last five years are missing, but CP production amounts were increasing in China before 2013,[14] and there is no indication that this trend has stopped or reversed.

Information on the use of MCCPs is very scare and restricted to the United States,[33] the EU,[19,26,34−38] Norway,[35,39] and Japan[40,41] (Table S1). It is known that the main uses of MCCPs are as secondary plasticizers in polyvinyl chloride (PVC), as extreme pressure additives in metal working fluids, and as additives to paints, adhesives, sealants, rubbers and other polymeric materials.[19] However, not much is known about the application areas in the specific countries. Numbers reported by the European Chemicals Bureau[19] indicate that MCCPs were mainly used as secondary plasticizers in PVC in the EU between 1994 and 1997. The only data further available are for Japan for 2001 and indicate a MCCP usage of around 50% in PVC and 40% as extreme pressure additives in metal working fluids.[40] More information on the uses of MCCPs is therefore urgently needed, especially if emission-reduction strategies will become necessary in the future.

CHEMICAL PROPERTIES

MCCPs consists of thousands of congeners and the chain length and chlorination degree of the molecules influence most of the chemical properties. Therefore, for most of the properties exists not one "true" value but rather a range of values.

Physicochemical Properties. Measurements for some physicochemical properties of MCCPs are available,[42−46] but they do not cover all groups of constitutional isomers nor all properties. Therefore, we derived in our previous work a set of property data for SCCPs and MCCPs based on calculations with the quantum-chemical model COSMOtherm and validated with data from literature.[1] In brief, the calculated logarithmic octanol−water partition coefficients range for MCCPs between 6.77 (for a C_{14}−CP with 35% chlorine) and 9.85 (for a C_{17}−CP with 70% chlorine) (Table S5 and Figure S2c), which shows the large preference of MCCPs for organic phases such as the lipid tissue of organisms compared to the aqueous phase. The values also indicate that soil and sediment are likely to be relevant environmental reservoirs for MCCPs. Specific data for the congener groups and measured octanol−water partition coefficients are provided in Table S5. The calculated vapor pressure ranges for MCCPs between 6.64×10^{-12} Pa (for a C_{17}−CP with 70% chlorine) and 1.31×10^{-2} Pa (for a C_{14}−CP with 35% chlorine) (Table S4 and Figure S2a). A vapor pressure of 2.27×10^{-3} Pa has been reported for MCCPs with a chlorine content of 45%,[19] and a vapor pressure of $1.3–2.7 \times 10^{-4}$ Pa was reported for a $C_{14–17}$-CP with 52% chlorine.[42] The calculated solubilities in water range for MCCPs between 0.02 μg/L (for a C_{17}−CP with 70% chlorine) and 40.4 μg/L (for a C_{14}−CP with 35% chlorine) (Table S4 and Figure S2b). Madeley and Gillings[43] determined the solubility of a C_{15}−CP with 51% chlorine to be 5 μg/L, and Campbell and McConnell[42] reported the solubility of a C_{16}−CP with 52% chlorine to be 10 μg/L. With these chemical properties ranging over several order of magnitudes, MCCPs are prone to a complex behavior and fate in the environment. Thus, MCCPs partition in the environmental compartments, such as in the atmosphere (gaseous and particulate phases) and in water (dissolved and particulate phases). Therefore, a review about the occurrence of MCCPs in the environment needs to consider all environmental matrices and their subphases.

Degradation in the Environment. The degradation of MCCPs in the environment has been reported in water and secondary activated sludge. Degradation in air through the

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reaction with OH radicals and degradation in soil and sediment through biodegradation might be possible, but studies that confirm these degradation pathways for MCCPs are missing. Estimated half-lives in air and soil from EPI Suite are provided in section S2.1 in the Supporting Information; however, it is unclear how representative the data are.

Photochemical Degradation. The degradation of CPs in water through photochemical dechlorination was investigated by Koh and Thiemann.[47] The obtained half-lives in water for CP mixtures with 30−53% chlorine ranged between 9.6 h (for C_{17-24}-CPs with 35% chlorine) and 12.8 h (for C_{12-18}-CPs with 52% chlorine). However, these half-lives are only relevant for chemicals in the uppermost water layer. MCCPs in deeper water layers cannot be photochemically degraded due to a lack of UV radiation.

Biodegradation. Ready biodegradation tests in secondary activated sludge reported by the MCCP manufacturer in the EU showed no biodegradation for C_{14-17}-CPs with 63.2% chlorine and C_{15}−CPs with 51% chlorine. Only substances and mixtures with a chlorine content below 46% were readily biodegradable.[48] According to these data, MCCP congeners with more than 46% chlorine have to be considered as persistent, which is in line with the estimated data from EPI Suite (Table S6). Reliable studies on the biodegradation of MCCPs in water, soil, or sediment are missing.

Transformation Pathways and Products. Bergman et al.[49] reported that synthetic CPs and commercial CP products are transformed into a large number of aromatic hydrocarbons as well as numerous polychlorinated aromatic compounds at temperatures above 300 °C. They stated that dehydrohalogenation of the aliphatic structure, followed by ring formation, is most probably the explanation for the formation of aromatic compounds. Schinkel et al.[50] confirmed dehydrohalogenation reactions of chlorotridecanes (and the formation of chlorinated alkenes) at temperatures above 160 °C. Furthermore, photochemical and bacterial dechlorination of MCCPs have been observed.[47,51] No study has observed yet degradation pathways that could lead to a chain length reduction. However, a breakdown of carbon−carbon bonds is considered a critical reaction pathway because this would transform long- and medium-chain CPs to short-chain CPs.

Conclusions. Not much has been published so far on the degradation of MCCPs. However, the available data from the ready biodegradation tests in secondary activated sludge point out that MCCP congeners with more than 46% chlorine have to be considered as persistent.

Bioaccumulation and Biomagnification Potential. A review of the bioaccumulation potential of MCCPs in the aquatic environment is available from Thompson and Vaughan[52] from 2013. The following paragraphs summarize the findings of Thompson and Vaughan[52] and describe additional studies published after 2013.

BCF and BAF. No study has reported so far reliable bioconcentration or bioaccumulation factors for MCCPs. Available studies (but omitted in the present discussion due to the limited reliability) are summarized in Table S9.

BMF. Laboratory-derived biomagnification factors were reported by Fisk et al.[53−55] However, the results show no clear trend: BMFs for C_{14-17}-CPs ranged between 0.3 and 5, with a mean of 1.6 and a median of 0.99. A detailed data analysis is provided in section S2.3 of the Supporting Information. Field-derived BMFs for MCCPs were reported by Houde et al.[11] and Zeng et al.[56] BMFs reported by Houde et al.[11] were generally below one. Field-derived BMFs reported by Zeng et al.[56] were above one but questionable (Table S9).

TMF. No bioaccumulation of MCCPs was found in the food webs studied by Houde et al.,[11] NIVA,[57−59] and Huang et al.[60] Miljodirektoratet[61] and Zeng et al.[56] otherwise calculated trophic magnification factors above one, which would indicate an increase in the chemical concentration in the biota with increasing trophical level. However, the TMFs in both studies were derived from concentrations in different tissues and are, therefore, questionable.

Conclusions. It remains so far unclear whether or not MCCPs are bioaccumulative and there is, therefore, an urgent need for reliable studies that investigate the bioaccumulation and biomagnification potential of these chemicals. Studies on the BMF and TMF should pay attention to compare concentrations in similar tissues. Also, care should be taken that animals are representative of the same food web.

ECHA[62] considered C_{14}−CPs with 40% to 56% Cl by weight and C_{15}−CPs with 51% Cl by weight as potentially bioaccumulative and proposed that the bioaccumulation potential of MCCPs is decreasing with increasing carbon chain length and chlorine content. However, if we consider the available measured data for SCCPs and MCCPs (section S2.2 of the Supporting Information), no clear trends are visible. ECHA requested aqueous and dietary exposure tests from the registrant manufacturers for C_{14}−CPs with 50−52% and 55−60% chlorine until September 2018,[63] but tests for MCCPs with other carbon chain lengths and chlorination degrees will most probably be necessary to conclude whether or not MCCPs (or single-congener groups of the MCCPs) should finally be considered as bioaccumulative.

Eco-toxicity. *Toxicity to Soil Organisms.* To date, the highest sensitivity of organisms to MCCPs in soil has been observed for the earthworm *Eisenia fetida*.[12,64] The data show that a statistically significant decrease in reproduction occurred at 900 μg/g dry weight (dw) soil. The predicted no-effect concentration (PNEC) in soil for *E. fetida* reproduction was estimated to be 28 μg/g dw soil based on a no-observed-effect concentration (NOEC) of 280 μg/g dw and an assessment factor of 10.[12] The organic carbon (OC) content in the study was 4.7%, which gives a PNEC of 5.9 mg/g OC. The study was conducted with a C_{14-17}-CP with 52% chlorine.

Toxicity to Sediment Organisms. Among the organisms tested, *Hyalella azteca* and *Lumbriculus variegatus* have the highest sensitivity to MCCPs in sediment. *H. azteca* showed a statistically significant reduction in mean weight at 270 mg/g dw sediment, and *L. variegatus* showed a statistically significant reduction in the mean total weight of worms per replicate at 410 mg/g dw sediment.[12,19,65,66] The PNEC in sediment was estimated to be 13 μg/g dw for both organisms based on a NOEC of 130 μg/g dw sediment and an assessment factor of 10.[12] The organic carbon content in both studies was around 5%, which gives a PNEC of 260 μg/g OC. Both studies were conducted with a C_{14-17}-CP with 52% chlorine.

Toxicity to Aquatic Invertebrates. MCCPs show toxicity to *Daphnia magna* in both short- and long-term exposure.[12] Most of the data available have been generated using a C_{14-17}-CP mixture with 52% chlorine. A 48 h EC_{50} of 5.9 μg/L was reported for *D. magna*,[12,67] although other studies suggest that the EC_{50} might be higher than this value.[12] Long-term studies with *D. magna* suggest that the 21 day LOEC for MCCPs is at around 18 μg/L and the NOEC is around 10 μg/L.[12,68] The PNEC for MCCPs in water for aquatic invertebrates was estimated to be 1 μg/L based

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on the NOEC of 10 μg/L for *D. magna* and an assessment factor of 10.[12]

Toxicity to Fish. So far, only one study has reported the toxicity of MCCPs to fish. Cooley et al.[69] exposed rainbow trout to different concentrations of C_{14}−CPs in food and found behavioral effects as well as effects on liver and thyroid histology for the C_{14}−CP with 55% chlorine at concentrations of 0.22 and 1.3 μg/g wet weight (ww) whole fish. No effects were observed for the C_{14}−CP with 48% chlorine at concentrations between 0.02 and 0.11 μg/g ww whole fish. We propose here, therefore, a PNEC for the toxicity of MCCPs to rainbow trout of 11 ng/g ww whole fish based on the NOEC of 0.11 μg/g ww whole fish and an assessment factor of 10. A lipid content of 0.24 g lipid/g ww whole fish[70] gives a PNEC of 46 ng/g lipid whole fish for rainbow trout.

Conclusions. MCCPs meet the toxicity threshold defined under REACH (chronic NOEC or EC_{10} for freshwater organisms below 10 μg/L)[71] and should, therefore, be considered toxic to the environment. This is in line with the European Classification, Labelling and Packaging Regulation, which classified MCCPs as acute and chronic toxic to the aquatic environment.[72]

OCCURRENCE OF MCCPS IN THE ENVIRONMENT, BIOTA, AND HUMANS

The following subsections discuss the worldwide measured concentrations of MCCPs in the environment, biota, and humans. However, the first subsection discusses the sources of errors in the measurements to illustrate the uncertainties in the data.

Sources of Errors in the Measurements. So far, no standard method for analyzing chlorinated paraffins has been available, which means that, basically, each laboratory has been using its own method. We have recorded the instrumental technique and the quantification method used in the different studies (Tables 1−6), but it is beyond this review to evaluate the different methods, as this has been addressed before.[73] However, we would like to point out the largest uncertainties associated with the measurements and discuss the consequences for data interpretation. First, it is important to note that almost all reported MCCP concentrations were obtained by gas chromatography coupled to mass spectrometry (GC−MS) and that MCCPs with 16 and 17 carbon atoms were hardly detected due to their low vapor pressure, which results in long retention times and their inseparability in the GC. The use of direct liquid injection mass spectrometry as applied in the new method of Bogdal et al.[74] is an elegant alternative to GC−MS for analyzing MCCPs (and LCCPs) in the future. Also, methods based on liquid chromatography are used now more often[28,75,76] and are interesting alternatives to methods based on GC−MS. Gas chromatography coupled to electron capture negative ionization mass spectrometry (GC−ECNI−MS), as used by most of the laboratories, also has the disadvantage that lower chlorinated CPs ($<Cl_5$) are not detected.[6,23] The reported chlorination degrees of low-chlorinated mixtures are, therefore, systematically too high.[77] Another disadvantage of this method is that the external standard mixture used for quantification must have an average molecular mass and a chlorination degree as similar as possible to the CP mixture in the sample. Basing the quantification on a standard with a significantly different chlorination degree than the sample, can appreciably affect the results and easily falsify the measurements by up to an order of magnitude.[77] Reth et al.[77] suggested deriving a linear relationship between instrumental response and the chlorine content of the external standard mixture to correct for the inevitable difference in the chlorination degree between the standard mixture and the field sample. We have indicated in Tables 1 to 6 under "compensation" whether the authors of the studies used such a linear correlation to compensate for differences in the chlorine content of sample and standard. An alternative is to use specific chain-length standards and response factors for each congener group, as recently suggested by Yuan et al.[78] Another important aspect to note is that around 40% of the studies conducted measurements with low-resolution mass spectrometry (LRMS). However, mass spectrometers with a resolution below 10 000 (full width at half-maximum) are not able to resolve the commonly occurring mass interferences from different CP congener groups (for example, interferences between $^{12}C_{18}{}^{1}H_{30}{}^{35}Cl_4{}^{37}Cl_4$ (533.97 Da) and $^{12}C_{16}{}^{1}H_{25}{}^{35}Cl_8{}^{37}Cl_1$ (533.91 Da)). LCCP congener groups that cannot be resolved from the MCCP congener groups, therefore, increase the MCCP concentration by at least 15% (for the calculation, see section S3 in the Supporting Information.) An increased response factor of the longer-chained CPs will increase this percentage even further.[74] In addition to the commonly occurring mass interferences, other mass interferences from, for example, transformation products[79] or unexpected fragment ions formed during the ionization[80] can complicate the analysis even more. However, Schinkel et al.[79,81] and Yuan et al.[80] showed that it is possible to identify the target congeners when applying mass spectral deconvolution procedures. Zeng et al.[82] presented a method in which they used the isotope abundance of the interferential congeners and an algebraic equation group to obtain the true integrated ion signals. Both procedures (deconvolution and the algebraic equation group) are options to decrease the interferences computationally, and we highly recommend to use one of these procedures for the analysis of MCCPs with LRMS.

General Remarks. If we go through the Tables 1 to 6, where we have listed all of the studies that reported MCCP concentrations in the environment, biota, and humans and check (a) which studies have used HRMS, or LRMS with a mathematical procedure to reduce the interferences between the congeners (methods of Bogdal et al.[74] or Zeng et al.)[82] and (b) which studies have compensated for the differences between chlorine content in standard and sample, not many studies are left. Taking all the other possible error sources into account, which we have listed above, we have to assume that most of the reported concentrations might not be very accurate. We believe, however, that the overall picture from the whole set of measurements and studies is (at least at the order of magnitude) correct and will give valuable insights into the environmental contamination with MCCPs. This is also very reasonable if we consider the results from the available interlaboraty studies with SCCPs.[83−85] Tomy et al.[83] found differences between the assigned values and the reported concentrations from −30% to 310%. The latest interlaboratory study of van Mourik et al.[85] reported differences of up to 54%. Therefore, we will take a more general approach and interpret ranges of the measured concentrations rather than single values. The ranges discussed in the following paragraphs refer always to the 25th to 75th percentile of the reported data of each study. Thus, outliers that might be due to measurement errors are cut out. We also excluded some of the studies that measured obviously unrealistic concentrations (Table S9). Also, studies before 1995 were excluded because only semiquantitative analytical methods were available.[86] Values below the limit of detection reported in the

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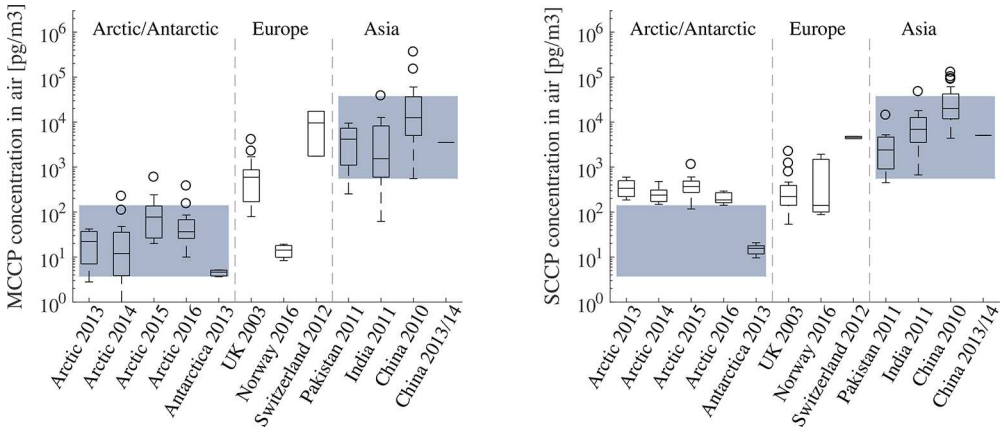

Figure 1. MCCP and SCCP concentrations in air. The blue rectangles indicate the MCCP concentration ranges in the specific regions. The data from the Arctic are only semiquantitative.

original study were predicted using the Robust Regression on Order Statistics implemented in R.[87] This method performs a regression on the data assuming log-normal quantiles. The regression line is used to predict unknown observations and summary statistics are computed based on the predicted and reported observations. This approach was used because it ensures that measurements below the limit of detection are included in the assessment and are not arbitrarily substituted or omitted.

MCCP Concentrations in Air. MCCP concentrations in air (in the vapor and particulate phase) have been measured in the Arctic, Antarctic, Europe, and Asia (Figure 1 and Tables 1 and S10). However, measurements from other areas of the world like North or South America, Africa, or Australia are also needed to get a more-complete picture of the global distribution of the MCCPs in air. The reported MCCP concentrations in air are lowest in the Arctic and Antarctic (4−140 pg/m^3) and highest in Asia (600−36 500 pg/m^3). Peak concentrations were measured in China[88] with up to 360 000 pg/m^3. MCCP concentrations in air measured in Asia and Europe are in the same order of magnitude as SCCP concentrations measured at the same locations and points in time (Figure 1 and Table S10). MCCP concentrations in air in the Arctic are, however, around one order of magnitude lower than the SCCP concentrations.[89−92] This indicates a slightly lower long-range atmospheric transport potential of MCCPs compared with SCCPs. However, the data from the Arctic[89−92] have to be considered as semiquantitative because the contribution of possible contamination during sampling and analyses has not been fully validated and blank levels in some samples were quite high.[92] Also, only C_{14} and C_{15}−CP congeners have been measured. Thus, final conclusions on the long-range atmospheric transport potential will only be possible if qualitatively better data are available.

MCCP Concentrations in Water. MCCP concentrations in water bodies that are not directly contaminated by a point source (background sites) are only available for Lake Ontario, Canada[11] and for stormwaters in Oslo, Norway[57,59,93] (Figure 2 and Tables 1 and S10). However, the measured MCCP concentrations in Lake Ontario were conducted with filtered water and are not representative because MCCPs are also attached to particles. MCCP concentrations measured in the stormwaters in Oslo were in the range from 15 to 130 ng/L water and, thus, slightly lower than the SCCP concentrations at the same sites and points in time (Figure 2). One study reported also MCCP concentrations in a water body close to a local source in the

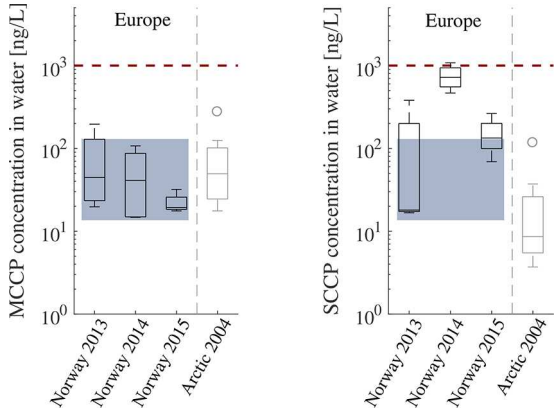

Figure 2. MCCP and SCCP concentrations in surface water. The blue rectangles indicate the MCCP concentration ranges in the background sites. The box plot in gray represents a site close to a local source. The horizontal dashed lines show the PNECs for MCCPs and SCCPs, respectively.

Canadian Arctic. The MCCP concentrations ranged between 24.5 and 102 ng/L and were a factor of five higher than the SCCP concentrations from the same site. The concentrations from both studies were one order of magnitude lower than the reported PNEC for MCCPs in water (1000 ng/L).[12] However, the studies are not representative for other areas and, given that MCCPs are widely discharged to water bodies after their use as cutting fluids in metal working applications,[19] more measurements of MCCP concentrations in water are urgently needed. Most important would be measurements close to industrial sites to see whether or not MCCPs pose a risk to aquatic organisms close to these sites.

MCCP Concentrations in Soil. MCCP concentrations in soil are available for Europe, South Africa, and China (Figure 3 and Tables 1 and S10). The measured concentrations do not show large differences between regions, particularly in comparison with the ranges observed in air. Measured MCCP concentrations at background sites range between 1 and 37 ng/g dw in Europe and China and between 15 and 305 ng/g dw in South Africa. The measured MCCP concentrations from Xu et al.,[94] which are from sites close to a CP production plant in China, are in the same range as the background soil concentrations in South Africa (50 to 310 ng/g dw). The MCCP concentrations measured by Bogdal et al.[96] are shown here as one box plot; however, they encompass archived soil samples from six sampling sites in Switzerland, covering the

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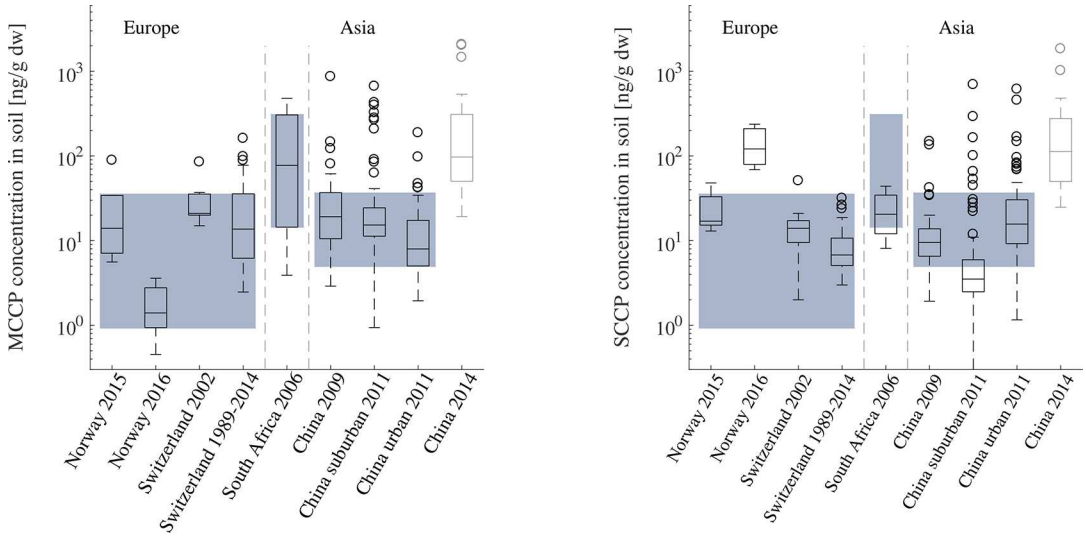

Figure 3. MCCP and SCCP concentration in soil. The blue rectangles indicate the MCCP concentration ranges in background soil in the specific regions. The box plot in gray represents a site close to a local source.

period from 1989 to 2014. The authors could show that MCCP concentrations in soil increased during the whole time period by a factor of two to three, with the highest concentrations occurring in the last sample from 2014.

MCCP concentrations in soil in Europe (except Norway 2016) and China are in the same range as the SCCP concentrations measured at the same sites and points in time (Figure 3). Although, the measured MCCP concentrations are still well below the estimated PNEC for soil (28 μg/g dw), continuous monitoring of sites close to local sources and background soils would be important to ensure that levels of MCCPs are not further increasing.

MCCP Concentrations in Surface Sediment. Similar to MCCP concentrations in soil, available MCCP concentrations in surface sediment do not show large differences between regions (Figure 4 and Tables 2 and S11). Concentrations at background sites are in the range of 14−290 ng/g dw (Canada), 1−403 ng/g dw (Europe and South Africa), and 5−1300 ng/g dw (Asia). MCCP concentrations in background surface sediment are, thus, one order of magnitude higher than MCCP concentrations in background soil. MCCP concentrations in sediment close to local sources in Australia and China range from 1140 to 9760 ng/g dw (Figure 5) and are more than one order of magnitude higher than concentrations in background sediment. Moreover, some of the measured concentrations in Australia in 2001[97] and in the Pearl River in China in 2010[98] exceeded the PNEC (13 μg/g dw) for MCCPs in sediment. Although these sediment samples are from heavily industrialized and urbanized areas, the high concentrations should not be overlooked because they are from open water bodies that are habitats for aquatic invertebrates, fish, and humans. The high MCCP concentrations in the Pearl River in 2010 were also confirmed by measurements from Zeng et al.[41] in 2012 and 2013 who measured only slightly lower levels than Chen et al.[98] in 2010. The MCCP concentrations in sediment close to local sources are also up to one order of magnitude higher than the SCCP concentrations from the same sites and points in time (Figure 5). The high sediment concentrations close to local sources are very alarming and more measurements are needed in heavily industrialized and urban areas to assess the risks of MCCPs and to determine the emission sources of the MCCPs in these regions.

MCCP Concentrations in Sediment Cores. MCCP concentrations were reported in eight sediment cores from Switzerland, Sweden, China, and Japan (Figure S7 and Table S14). The core from Lake Thun, Switzerland,[99] two of the cores from Sweden (Baltic Sea),[100] and the two cores from China all showed increasing MCCP levels, whereas one of the cores from the Baltic Sea[100] and the two cores from Tokyo Bay, Japan,[41] showed decreasing or nondiscernible time trends. These trends match the reported production and use data of MCCPs with decreasing CP levels in Japan and increasing CP levels in China.[14] The cores from Sweden were from urban (decreasing trend) and industrial (increasing trends) sites and show the increasing use of MCCPs in industrial applications in Europe. The occurrence of MCCPs in sediment slices from the 1940s and 50s shows furthermore that MCCPs persist in sediment over decades and degrade only slowly.

MCCP Concentrations in Fish. MCCP concentrations in fish (Figure 6 and Tables 3 and S12) are available from the Arctic, Canada, the North Atlantic, Europe, and China. The reported concentrations are similar in the Arctic, Canada, Europe, and the Liaodong Bay in Northeastern China and range between 12 and 700 ng/g lipid. MCCP concentrations measured in the South China Sea[56] and in paddy fields in the Yangtze River Delta in China[101] were higher and ranged between 1260 and 2600 ng/g lipid. MCCP concentrations in fish from a lake on Bear Island in the Arctic[102] were in the upper 50th percentile of the observed concentrations from Canada and Europe. The contamination of this remote island was explained by two reasons.[103] First, the lake is in a mountainous area and receives a lot of precipitation. Second, large seabird colonies use the lake as resting area and input of guano from these birds causes an increase in POP levels. These observations and the relatively high MCCP concentrations found in the Arctic fish show once more that MCCPs are able to undergo long-range atmospheric transport. The MCCP concentrations measured in Lake Ontario (Canada) between 1979 and 2004[104] are shown here as one box plot; however, they encompass measurements from every four to six years from 1979 to 2004. MCCP concentrations increased by a factor of almost four between 1979 and 1998 and decreased afterward. However, the decrease in concentrations after 1998 was most likely due to large changes in the Lake Ontario food web and is not statistically

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Environ. Sci. Technol. 2018, 52, 6743−6760

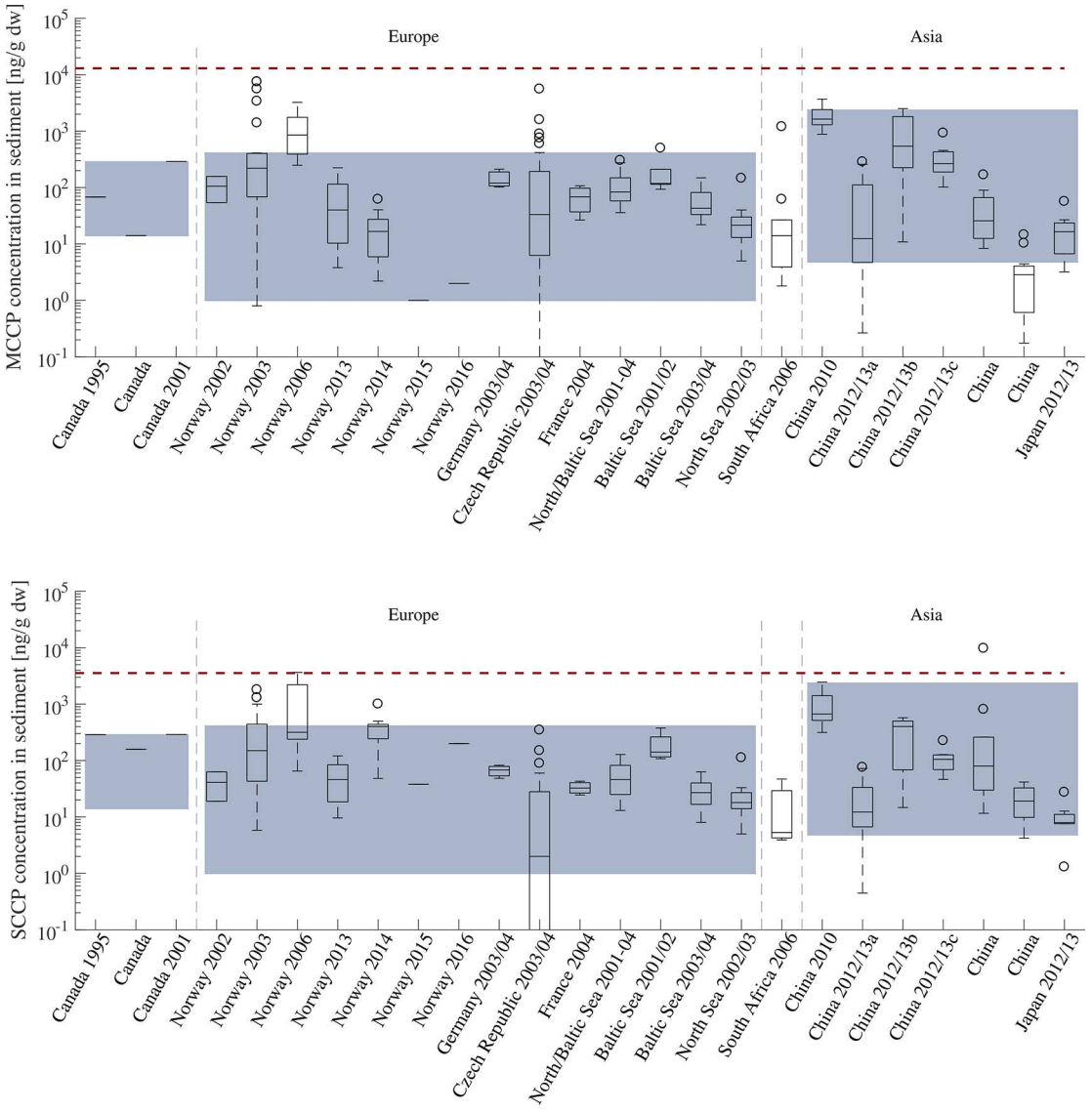

Figure 4. MCCP and SCCP concentrations in surface sediment (background sediment). Studies without an indicated year did not report the sampling year. The blue rectangles indicate the MCCP concentration ranges in the background surface sediment in the specific regions. The horizontal dashed lines show the PNECs for MCCPs and SCCPs, respectively.

significant anymore when corrected for the food-web effect.[104] MCCP concentrations are in the same range as SCCPs in most of the sampled fish (Figure 6). Toxicological data are only available for MCCPs in rainbow trout; however, none of the studies measured MCCPs in this fish species. Future research should focus, therefore, on more toxicological data to assess the risk of MCCPs to fish. Also, studies of food webs to investigate the trophic magnification and biomagnification of MCCPs in aquatic biota would be very important.

MCCP Concentrations in Birds. Measured MCCP concentrations in bird eggs and bird tissue are available from the Arctic, Norway, and China (Figure 7 and Tables 4 and S12). The concentrations in bird eggs are very similar in the Arctic and in Norway, ranging between 2.1 and 170 ng/g lipid. Lipid normalized concentrations in blood and muscle/liver are around one order of magnitude higher (19−1660 ng/g lipid) than the MCCP concentrations in bird eggs. Data from Søndre Skjßlholmen (Osloford, Norway)[57,59,93,105] in eggs and blood were taken every year between 2013 and 2016; however, the measured concentrations show inconsistent time trends. The MCCP concentrations in bird eggs and bird tissue were in the same range or slightly lower than the SCCP concentrations measured in the same animals and points in time (Figure 7).

MCCP Concentrations in Mammals. MCCP concentrations in mammals are available for ringed seals and polar bears from the Arctic[61] and finless porpoise, Indo-Pacific humpback dolphins,[106] and yellow weasels[101] from China (Figure 8 and Tables 5 and S13). MCCP concentrations in the plasma of ringed seals and polar bears ranged between 74 and 600 ng/g lipid and were slightly lower than the SCCP concentrations from these animals.[61] MCCP concentrations in the finless porpoises (South China Sea) were in the range from 990 to 9200 ng/g lipid and, thus, two times higher than the SCCP concentrations. MCCP concentrations in the Indo-Pacific humpback dolphins (Pearl River Estuary) ranged between 1500 and 51 500 ng/g lipid and were up to 3.5 times higher than the SCCP concentrations. The MCCP concentrations in the finless porpoises and the humpback dolphins showed a statistically significant increase between 2004 an 2014. The higher MCCP concentrations present in humpback dolphins compared to finless porpoises were attributed to

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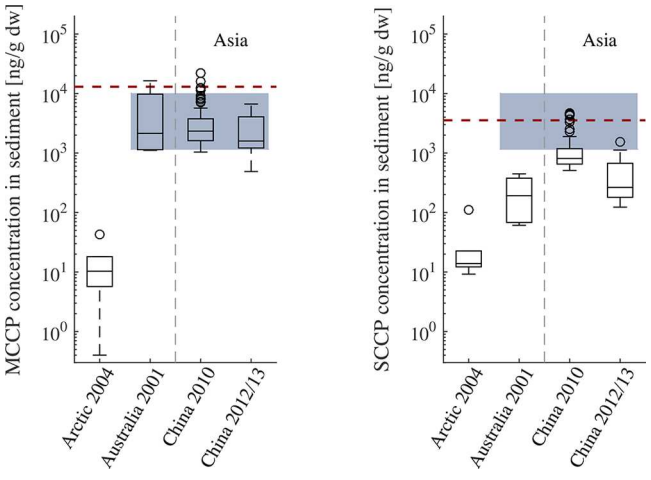

Figure 5. MCCP and SCCP concentrations in surface sediment close to local sources. The blue rectangle indicates the MCCP concentration range in surface sediment close to local sources in Australia and Asia. The horizontal dashed lines show the PNECs for MCCPs and SCCPs, respectively.

species—species differences in feeding habits and biotransformation as well as exposure levels in their living habitats.[106] MCCP concentrations in yellow weasel from the Yangtze River Delta were in the same order of magnitude as the MCCP concentrations in the humpback dolphins and show that MCCPs accumulate also in terrestrial mammals.

The available studies give a first impression of the levels of MCCPs in mammals, but more measurements in marine and terrestrial mammals from around the world are needed to evaluate the contamination of these animals with MCCPs. Moreover, there is, to our knowledge, no study so far that has examined the level above which MCCPs pose a risk to the health of mammals. Such a study would be extremely important to judge whether or not the measured MCCP concentrations affect the health of the animals. However, independent of these questions, it is for sure that the MCCP levels add to the levels of other toxic pollutants and can, for example, increase the risk of cancer and reproductive problems in whales and dolphins.

MCCPs in Human Breast Milk, Placenta, and Human Blood. MCCP concentrations in human breast milk (Figure 9 and Tables 6 and S13) are available from Europe and China. The measured concentrations show neither temporal nor spatial trends. Also, no significant differences could be observed

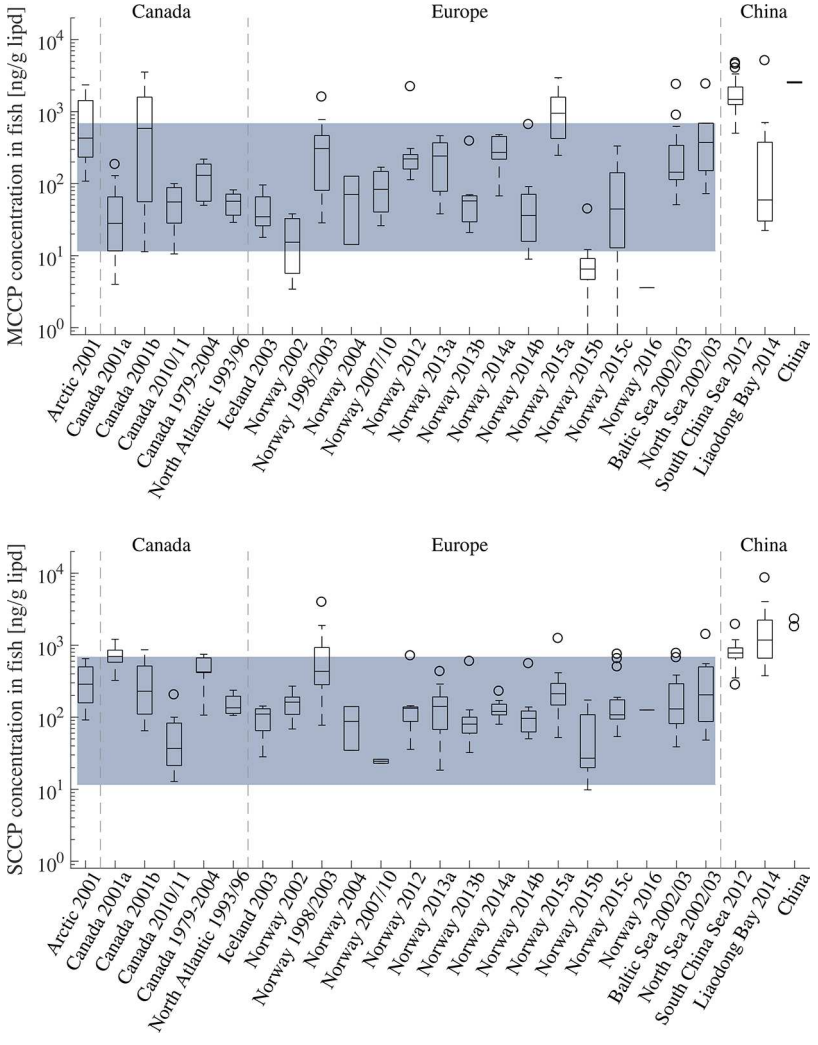

Figure 6. MCCP and SCCP concentrations in fish. The blue rectangle indicates the MCCP concentration range in fish in the Arctic, Canada, and Europe.

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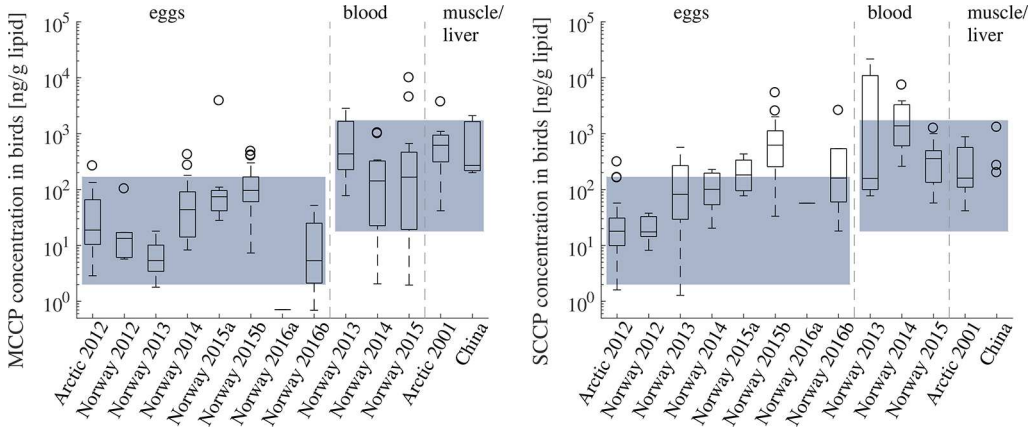

Figure 7. MCCP and SCCP concentrations in birds. The blue rectangles indicate the MCCP concentration ranges in eggs, in blood, and in muscle and liver.

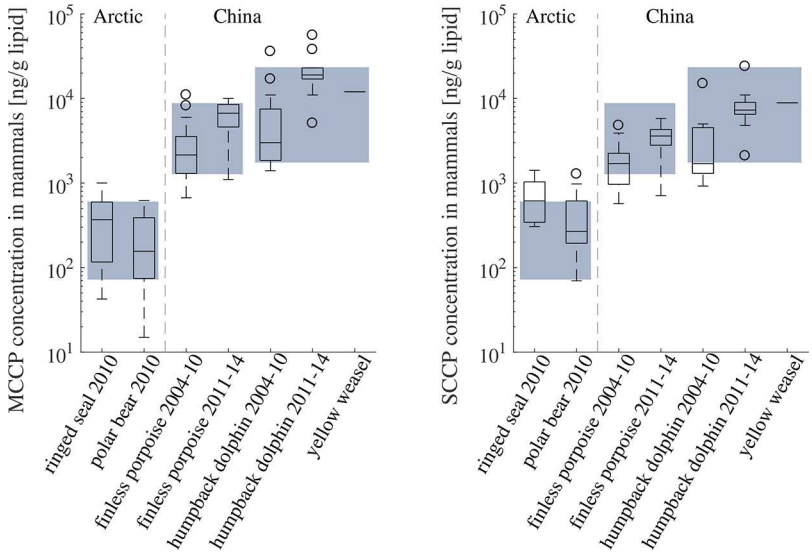

Figure 8. MCCP and SCCP concentrations in mammals. The blue rectangles indicate the MCCP concentration ranges in the specific species and regions.

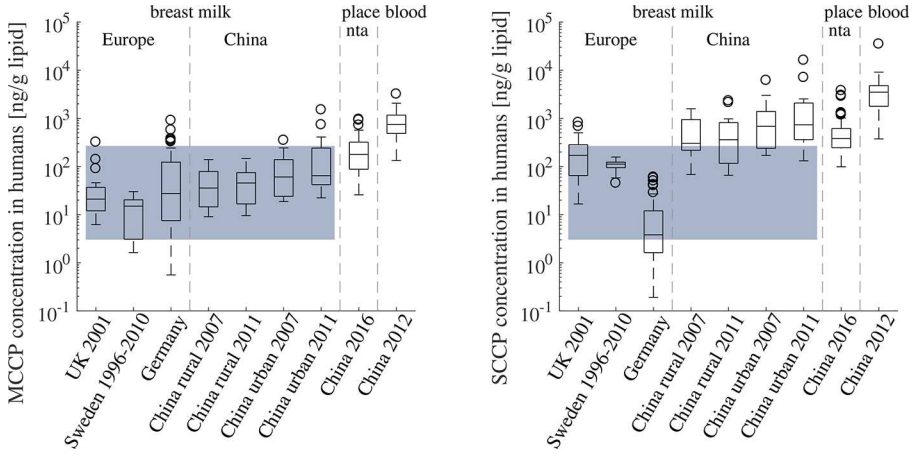

Figure 9. MCCP and SCCP concentrations in human breast milk, placenta, and human blood. The study in Germany did not report the sampling year. The blue rectangle indicates the MCCP concentration range in human breast milk.

between rural and urban areas in China.[107,108] The measured MCCP concentrations range between 3.10 and 240 ng/g lipid and are, thus, one order of magnitude lower than the SCCP concentrations measured in the same samples (with the exception of the concentrations measured in Germany)[109] (Figure 9). MCCP concentrations measured in human placenta are slightly higher,[110] and MCCP concentrations measured in human blood[76] are around one order of magnitude higher than

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Table 1. Information on the Concentrations Shown in Figures 1−3[a]

country	year	month	specific location	type of site	instrumental technique	quantification	compensation[b]	sample no.	literature source
concentrations in air (pg/m³)									
Arctic	2013	1−12	Zeppelin, Ny-Ålesund	remote	GC−ECNI−HRMS	T	no	12	NILU[89]
Arctic	2014	1−12	Zeppelin, Ny-Ålesund	remote	GC−ECNI−HRMS	T	no	12	NILU[90]
Arctic	2015	1−12	Zeppelin, Ny-Ålesund	remote	GC−ECNI−HRMS	T	no	12	NILU[91]
Arctic	2016	1−12	Zeppelin, Ny-Ålesund	remote	GC−ECNI−HRMS	T	no	12	NILU[92]
Antarctic	2013	1, 2	Georgia King Island	remote	GC−ECNI−LRMS	Z + R	yes	10	Ma et al.[113]
U.K.	2003	4, 5	various	rural−urban	GC−ECNI−HRMS	T	no	40	Barber et al.[114]
Norway	2016	7−9	Oslo	urban	GC−HRMS	−	−	3	NILU[115]
Switzerland	2012	8, 9	Zurich	urban	GC−ECNI−HRMS	T	yes	2	Bogdal et al.[74]
Pakistan	2011	1	various	rural−urban	GC−ECNI−LRMS	R	yes	10	Chemefa et al.[116]
India	2006	1	various	rural−urban	GC−ECNI−LRMS	R	yes	20	Chemefa et al.[116]
China	2010	1−3, 7−9	Pearl River Delta	urban	GC−ECNI−LRMS	R	yes	40	Wang et al.[88]
China	2013/14	1−12	Shenzhen	urban	UPLC−ESI−HRMS	R	yes	24	Li et al.[28]
concentrations in water (ng/L)									
Norway	2013		Oslo (river, stw)	urban	GC−HRMS	−	−	4	NIVA[93]
Norway	2014		Oslo (river, stw)	urban	GC−HRMS	−	−	4	NIVA[59]
Norway	2015		Oslo (river, stw)	urban	GC−HRMS	−	−	4	NIVA[57]
Arctic	2004		Iqaluit (river)	dump site	GC−ECNI−HRMS	T	no	9	Dick et al.[117]
concentrations in soil (ng/g dw)									
Norway	2015		Oslo	urban	GC−HRMS	−	−	5	NILU[118]
Norway	2016		Oslo	urban	GC−HRMS	−	−	5	NILU[115]
Switzerland	2002		various	rural−urban	GC−ECNI−LRMS	R	yes	9	Iozza[119]
Switzerland	1989−2014		various	rural−urban	GC−ECNI−HRMS	T	yes	36	Bogdal et al.[96]
South Africa	2006		Vaal Triangle	rural−urban	GC−EI−HRMS	−	−	12	Quinn et al.[95]
China	2009		Pearl River Delta	urban	GC−ECNI−LRMS	R	yes	59	Wang et al.[88]
China	2011		Shanghai	suburban	GC−ECNI−LRMS	R	yes	101	Wang et al.[120]
China	2011		Shanghai	urban	GC−ECNI−LRMS	R	yes	75	Wang et al.[121]
China	2014		Dalian city	industrial	GC−ECNI−LRMS	Z + R	yes	25	Xu et al.[31]

[a]The column "compensation" states whether the chlorine content differences between standard and sample were compensated according to Reth et al.[77] [b]−, not stated. stw, stormwater. R, quantified according to Reth et al.[77] T, quantified according to Tomy et al.[86] and Tomy and Stern.[13] Z, quantified according to Zeng et al.[82]

the measured MCCP concentrations in human breast milk. Studies on adverse effects of MCCPs in humans are missing, so it is currently impossible to assess the risk of MCCPs to humans. However, it has repeatedly been shown that toxic pollutants can affect the development of the fetus,[111] and it cannot be precluded that MCCPs also have negative developmental effects.

CONGENER GROUP PROFILE

Most of the available data for congener groups originates from studies with LRMS. The group congener profiles are, therefore, more qualitative than quantitative, and the differences between the profiles should be interpreted with caution. However, we find that the shorter-chained (C_{14}) and medium-chlorinated CPs (Cl_{7-8}) are the most prevalent MCCPs in environmental matrices (Figures S8−S10). $C_{14}Cl_{7-8}$ was also the most prevalent MCCPs in our measured CP-52 technical mixtures (Figure S2). MCCPs with 16 and 17 carbon atoms made up to 30% of the total concentrations in those environmental samples in which they could be measured; Pribylová et al.[112] even found a C_{16} content of up to 60% in sediment samples from the Czech Republic. However, robust statements about the congener groups profiles will only be possible if more studies with direct injection or liquid chromatography are available. The congener group profile will otherwise always be affected by the low vapor pressure of the longer-chained MCCPs.

CONCLUSIONS AND OUTLOOK

The accurate quantification of chlorinated paraffins and, especially, the quantification of the longer-chained MCCPs and LCCPs is very challenging. This is visible by the fact that many measured concentrations are affected by technical failures. Data generated, for example, following the method of Tomy and Stern[13] without compensation for the chlorine content differences between standard and sample might substantially under- or over-estimate the real concentrations. Concentrations that have been measured with LRMS, without using mathematical procedures to reduce the interferences between the congeners, most probably affected by mass interferences. Studies that quantified only C_{14}- and C_{15}−CPs underestimated the MCCP concentrations. However, we believe that, despite these failures, the overall picture from the whole set of measurements and studies is (at least at the order of magnitude) correct and gives valuable insights into the environmental contamination with MCCPs. For future studies, we recommend to use HRMS and the newest analytical methods (for example, Bogdal et al.[74] or Yuan et al.)[80] for analyzing CPs because these are less susceptible to interferences and allow the quantification of MCCPs with all chain lengths and LCCPs. Laboratories that have only LRMS available are still encouraged to measure MCCPs, but we highly

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Environ. Sci. Technol. 2018, 52, 6743−6760

Table 2. Information on the Sediment Concentrations Shown in Figures 4 and 5[a]

country	year	specific location	type of site	instrumental technique	quantification	compensation	sample no.	literature source
concentrations in surface sediment (background) (ng/g dw)								
Canada	1995	Lake Erie	industrial	GC−ECNI−HRMS	T	no	3	Tomy[122]
Canada	−	Hamilton harbor	industrial	GC−ECNI−HRMS	−	−	1	Muir et al.[123]
Canada	2001	Lake St. Clair		GC−ECNI−HRMS	−	−	20	Gewurtz et al.[124]
Norway	2002	Tromso		GC−ECNI−LRMS	T	no	2	Umweltbundesamt[125]
Norway	2003	various	−	GC−MS	−	−	23	NIVA[126]
Norway	2006	various	−	GC−ECNI−LRMS	T	no	10	Petersen et al.[127]
Norway	2013	Oslofjord[1]		GC−HRMS	−	−	9	NIVA[93]
Norway	2014	Oslofjord[1]	−	GC−HRMS	−	−	9	NIVA[59]
Norway	2015	Oslofjord[2]	−	GC−HRMS	−	−	1	NIVA[57]
Norway	2016	Oslofjord[2]	−	GC−HRMS	−	−	1	NIVA[105]
Germany	2003/04	Hamburg harbor	−	GC−ECNI−LRMS	T	no	3	Umweltbundesamt[125]
Czech Republic	2003/04	various	rural−urban	GC−ECNI−LRMS	C	no	12	Pribylová et al.[112]
France	2004	River Seine	−	GC−ECNI−LRMS	T	no	3	Umweltbundesamt[125]
North/Baltic Sea	2001−04	various	−	GC−CH_4/CH_2Cl_2−NICI−LRMS	T	n. n.	18	Umweltbundesamt[125]
Baltic Sea	2001/02	various	−	GC−EI−MS/MS	H	n. n.	6	Hüttig and Oehme[128]
Baltic Sea	2003/04	various	−	GC−ECNI−LRMS	R	yes	11	Hüttig and Oehme[129]
North Sea	2002/03	various	−	GC−EI−MS/MS	H	n. n.	14	Hüttig and Oehme[128]
S. Africa	2006	various river	3	GC−EI−HRMS	−	−	9	Quinn et al.[95]
China	2010	Pearl River Delta	4	GC−ECNI−LRMS	T	no	39	Chen et al.[98]
China	2012/13	Hong Kong (sea)	urban	GC−ECNI−LRMS	R	yes	35	Zeng et al.[41]
China	2012/13	Shenzhen (sea)	urban	GC−ECNI−LRMS	R	yes	8	Zeng et al.[41]
China	2012/13	Pearl River Delta	4	GC−ECNI−LRMS	R	yes	7	Zeng et al.[41]
China		Yellow River	rural−urban	GC^2−ECNI−HRMS	R	yes	13	Qiao et al.[130]
China		Yangtze River	rural−urban	GC^2−ECNI-HRMS	R	yes	13	Qiao et al.[131]
Japan	2012/13	Tokyo Bay	urban	GC−ECNI−LRMS	R	yes	8	Zeng et al.[41]
concentrations in surface sediment close to local sources (ng/g dw)								
Arctic	2004	Iqaluit, river	dump site	GC−ECNI−HRMS	T	no	6	Dick et al.[117]
Australia	2001	Yarraville (sea)	industrial, 5	CSR−GC	−	n. n.	4	Kemmlein et al.[97]
China	2010	Pearl River Delta	industrial	GC−ECNI−LRMS	T	no	76	Chen et al.[98]
China	2012/13	Pearl River Delta	industrial	GC−ECNI−LRMS	R	yes	9	Zeng et al.[41]

[a]The column "compensation" states whether the chlorine content differences between standard and sample were compensated according to Reth et al.[77] −, not stated. n.n., not necessary. 1, Alne, Bekkelaget, Frognerkilen. 2, Cm21. 3, agriculture and industrial region. 4, low industry activity area. 5, area influenced by CP manufacturer. C, quantified according to Coelhan.[132] H, quantified according to Hüttig and Oehme.[133] R, quantified according to Reth et al.[77] T, quantified according to Tomy et al.[86] and Tomy and Stern.[13]

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Table 3. Information on the MCCP Concentrations in Fish (Nanograms per Gram of Lipid) Shown in Figure 6[a]

country	year	specific location	sample medium	fish type	instrumental technique	quantification	compensation	sample no.	literature source
Arctic	2001	Bear Island, Lake Ellasjoen	liver, muscle	a	GC−ECNI−LRMS	R	yes	4	Reth et al.[102]
United States	1995	Lake Erie	whole fish	b, c	GC−ECNI−HRMS	T	no	2	Tomy and Stern[13]
Canada	2001	Lake Michigan	whole fish	d−f	GC−ECNI−HRMS	T	no	11	Houde et al.[11]
Canada	2001	Lake Ontario	whole fish	d, f−h	GC−ECNI−HRMS	T	no	13	Houde et al.[11]
Canada	2010/11	various lakes	whole fish	f, i, j	GC−NCI−HRMS	T	no	84	Saborido Basconcillo et al.[134]
Canada	1979−04	Lake Ontario	whole fish	f	GC−ECNI−HRMS	T	no	29	Ismail et al.[104]
North Atlantic	1993−06	−	whole fish	k−o	GC−ECNI−LRMS	−	no	5	Lahaniatis et al.[135]
Iceland	2003	Akureyri, Vestmanna.	liver	p	GC−ECNI−LRMS	R	yes	4	Reth et al.[102]
Norway	2002	various	liver	p	GC−MS	−	−	42	NIVA[126]
Norway	1998/03	various	various	various	GC−MS	−	−	163	NIVA[126]
Norway	2004	Lofoten	liver	p	GC−ECNI−LRMS	R	yes	2	Reth et al.[102]
Norway	2007/10	Lake Stensjön, Sännen, and Tjultr.	liver	a, q	−	−	−	39	Nyberg et al.[136]
Norway	2012	various	liver	p	GC−MS	−	−	165	NIVA[137]
Norway	2013	various	liver	p	GC−MS	−	−	82	NIVA[138]
Norway	2013	Oslofjord, Lysakerfj.	liver	r	GC−HRMS	−	−	15	NIVA[93]
Norway	2014	various	liver	p	GC−MS	−	−	119	NIVA[58]
Norway	2014	Oslofjord, Sollerud skole	liver	r	GC−HRMS	−	−	15	NIVA[59]
Norway	2015	various	liver	p	GC−MS	−	−	153	NIVA[139]
Norway	2015	Inner Oslofjord	muscle, liver	m, p	GC−HRMS	−	−	30	NIVA[57]
Norway	−	various	−	q, s	−	−	−	70	NIVA[140]
Norway	2016	Inner Oslofjord	liver	p	GC−HRMS	−	−	13	NIVA[105]
Baltic Sea	2002/3	−	liver	p, r, t	GC−ECNI−LRMS	half T, half R		29	Umweltbundesamt[125]
North Sea	2002/3	−	liver	p, r, t	GC−ECNI−LRMS	T	no	35	Umweltbundesamt[125]
China	2012	South China Sea	muscle and soft tissue	various	GC−ECNI−LRMS	R	yes	51	Zeng et al.[56]
China	2014	Liaodong Bay	whole fish	various	GCxGC−ECNI−HRMS	R	yes	73	Huang et al.[60]
China	−	Yangtze River Delta	muscle	u, v	GC−NCI-LRMS	B	n. n.	10	Du et al.[101]

[a]The column "compensation" states whether the chlorine content differences between standard and sample were compensated according to Reth et al.[77] −, not stated. n. n., not necessary. a, Arctic char. b, yellow perch. c, channel catfish. d, alewife. e, deepwater sculpin. f, lake trout. g, slimmy sculpin. h, rainbow smelt. i, Walleye. j, brook trout. k, sprat. l, redfish. m, herring. n, mackerel. o, halibut. p, cod. q, perch. r, flounder. s, trout. t, North sea dab. u, pond loach. v, rice field eel. B, quantified according to Bogdal et al.[74]. R, quantified according to Reth et al.[77]. T, quantified according to Tomy et al.[86] and[13].

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Environ. Sci. Technol. 2018, 52, 6743−6760

Table 4. Information on the MCCP Concentrations in Birds (Nanograms per Gram of Lipid) Shown in Figure 7[a]

country	year	specific location	sample medium	bird type	instrumental technique	quantification	compensation	sample no.	literature source
Arctic	2012	Svalbard	egg	a, b	GC−HRMS	−	−	24	Miljodirektoratet[61]
Norway	2012	Sklinna, Rost	egg	b, c	GC−ECNI−MS	−	−	24	Huber et al.[141]
Norway	2013	Oslofj., Søndre Skjßlh.	egg	c	GC−HRMS	−	−	15	NIVA[93]
Norway	2014	Oslofj., Søndre Skjßlh.	egg	c	GC−HRMS	−	−	14	NIVA[59]
Norway	2015	Oslofj., Søndre Skjßlh.	egg	c	GC−HRMS	−	−	15	NIVA[57]
Norway	2015	Oslo	egg	d−f	GC−HRMS	−	−	30	NILU[118]
Norway	2016	Oslofj., Søndre Skjßlh.	egg	c	GC−HRMS	−	−	15	NIVA[105]
Norway	2016	Oslo	egg	f	GC−HRMS	−	−	10	NILU[92]
Norway	2013	Oslofj., Søndre Skjßlh.	blood	c	GC−HRMS	−	−	3	NIVA[93]
Norway	2014	Oslofj., Søndre Skjßlh.	blood	c	GC−HRMS	−	−	10	NIVA[59]
Norway	2015	Oslofj., Søndre Skjßlh.	blood	c	GC−HRMS	−	−	13	NIVA[57]
Arctic	2001	Bear Island	muscle, liver	a, g	GC−ECNI−LRMS	R	yes	8	Reth et al.[102]
China	−	Yangtze River Delta	muscle	h−j	GC−NCI−LRMS	B	n. n.	18	Du et al.[101]

[a]The column "compensation" states whether the chlorine content differences between standard and sample were compensated according to Reth et al.[77] −, not stated. n. n., not necessary. a, kittiwake. b, common eider. c, herring gull. d, sparrowhawk. e, tawny owl. f, fildfare. g, little auk. h, peregrine falcon. i, collared scops owl. j, common cuckoo. B, quantified according to Bogdal et al.[74] R, quantified according to Reth et al.[77]

Table 5. Information on the MCCP Concentrations in Mammals (Nanograms per Gram of Liquid) Shown in Figure 8[a]

country	year	specific location	sample medium	mammal	instrumental technique	quantification	compensation	sample no.	literature source
Arctic	2010	Svalbard	plasma	ringed seal	GC−HRMS	−	−	9	Miljodirektoratet[61]
Arctic	2012	Svalbard	plasma	polar bear	GC−HRMS	−	−	20	Miljodirektoratet[61]
China	2004−14	South China Sea	blubber	finless porpoise	GC−ECNI−LRMS	Z	yes	50	Zeng et al.[106]
China	2004−14	Pearl River Estuary	blubber	Indo-Pacific humpback dolphin	GC−ECNI−LRMS	Z	yes	25	Zeng et al.[106]
China	−	Yangtze River Delta	muscle	yellow weasel	GC−NCI-LRMS	B	n. n.	5	Du et al.[101]

[a]The column "compensation" states whether the chlorine content differences between standard and sample were compensated according to Reth et al.[77] −, not stated. n. n., not necessary. B, quantified according to Bogdal et al.[74] Z, quantified according to Zeng et al.[82]

Table 6. Information on the MCCP Concentrations in Human Breast Milk, Placenta, and Human Blood (Nanograms per Gram of Liquid) Shown in Figure 9[a]

country	year	specific location	type of site	instrumental technique	quantification	compensation	sample no.	literature source
U.K.	2001	Lancaster, London	urban	GC−ECNI−HRMS	T	no	25	Thomas et al.[142]
Sweden	1996−10	Uppsala county	−	GC−ECNI−HRMS	T	no	523	Darnerud et al.[143]
Germany	−	Bavaria	rural−urban	GC−ECNI−LRMS	R	yes	60	Hilger et al.[109]
China	2007	various	rural	GCxGC−ECNI−HRMS	R	yes	452	Xia et al.[107]
China	2011	various	rural	GCxGC−ECNI−HRMS	R	yes	960	Xia et al.[107]
China	2007	various	urban	GCxGC−ECNI−HRMS	R	yes	570	Xia et al.[108]
China	2011	various	urban	GCxGC−ECNI−HRMS	R	yes	800	Xia et al.[108]
China	2016	Henan	−	GC−NCI−HRMS	G	yes	54	Wang et al.[110]
China	2012	Shenzhen	urban	UPLC−ESI−LRMS	R	yes	50	Li et al.[76]

[a]The column "compensation" indicates whether the chlorine content differences between standard and sample were compensated according to Reth et al.[77] −, not stated. R, quantified according to Reth et al.[77] T, quantified according to Tomy et al.[86] and Tomy and Stern.[13] G, quantified according to Gao et al.[30]

recommend the usage of the deconvolution procedure to decrease errors due to interferences.

If we look at the obtained overall picture of the environmental contamination with MCCPs, we see that MCCPs have been detected in all environmental compartments as well as in fish, birds, mammals, and human tissues, and they are often measured in higher concentrations than SCCPs. Most alarming to us are the sediment concentrations that reach or exceed the PNEC in sediment, as well as the increasing time trends observed for the MCCPs in various locations worldwide. We also observe the potential of the MCCPs to undergo long-range atmospheric transport and their high potential for chronic toxicity to aquatic invertebrates. There is, therefore, an urgent need to generate data on the degradation of MCCPs in soil and sediment as well as reliable data on the bioaccumulation potential of MCCPs. ECHA requested aqueous and dietary exposure tests for C_{14}−CPs and aerobic and anaerobic transformation tests in aquatic sediment systems for C_{14} and C_{15}−CPs from the registrant manufacturers until September 2018.[63] However, additional data for MCCPs with other carbon-chain lengths and chlorination degrees will most probably be necessary to draw final conclusions on the MCCPs. Unfortunately, those test are currently difficult to conduct because only standards of MCCPs mixtures but no chain-length or single-congener standards are available. The development of those standards would be a big step forward and would also allow the development of response factors for

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Environ. Sci. Technol. 2018, 52, 6743−6760

congener groups or single congeners, as done by Yuan et al.[78] for the quantification of SCCPs.

It would, in general, also be very important to study whether MCCPs can degrade into SCCPs or other (potentially unknown) compounds of concern and, if so, to what extent. There is also a necessity for more long-term monitoring studies in remote and non-remote regions to monitor temporal trends of MCCPs in air, soil, sediment, and biota. Decision makers should otherwise not wait too long for more data to come because there is already a lot of evidence supporting the conclusion that MCCPs with more than 46% chlorine are persistent and toxic, and if we apply the precautionary principle, regulatory actions should be considered seriously.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b06459.

Information about the reported production and use amounts of MCCPs and LCCPs; the physicochemical and kinetic properties; the mass interferences between CP congeners; and the measured concentrations in the environment, biota, and humans. (PDF)

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Supplementary Material: Environmental risks of medium-chain chlorinated paraffins (MCCPs) - A review

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Contents

List of Figures

List of Tables

S1 Production and use

S1.1 Information on the reported production and use amounts of MCCPs and LCCPs

Table S1: Production and usage of MCCPs [tons/year]. The percentages under 'comments' refer to the fractions of MCCPs compared to total CPs. '–' means no data available.

year	production	literature source	usage									literature source	comments
			total	metal work	PVC	leather liquors	rubber	textile	sealant	paint	other		
Denmark													
	–		28	–	–	–	–	–	–	–	–	EC [2005]	
European Union													
1985	–		46200	–	–	–	–	–	–	–	–	IPEN [2007]	84% $MCCPs_{use}$
1992	–		77000	–	–	–	–	–	–	–	–	Willis et al. [1994]	70% $MCCPs_{use}$
1994	–		56555	2611	45476	1614	2479	–	3079[1]		1296[2]	EC [2005]	
1995	–		58671	2765	48640	1270	2767	–	2392[1]		837[2]	EC [2005]	
1996	–		59306	3302	49240	1172	2324	–	2638[1]		630[2]	EC [2005]	
1997	–		65256	5953	51827	1048	2146	–	3541[1]		741[2]	EC [2005]	
2002	–		–	8000	30000	1550	–	–	–	–	–	EC [2005] [3]	
2003	–		55074	8036	33687	1415	3541	–	8321[1]		74[2]	EC [2005] [4]	
2004	45000-160000[5]	EC [2005] [6]	–	–	–	–	–	–	–	–	–		capacity from 5 manufacturing sites
2006	–		63700	9907	34676	708	7077	–	11323		–	COWI [2010] [7]	
2013	–		40000	–	–	–	–	–	–	–	–	Davis [2013] [3]	
France													
2010	–		–	–	0	–	–	–	–	0	—	INERIS [2012]	
Germany													
2006	–		6681	1136	1136	<66.81	1670		2272		401	COWI [2010] [7,3]	Germany & Austria
Japan													
2001	–		7617	3199	3619	–	–	–	–	–	799	Tsunemi [2010] [10]	64% $MCCPs_{use}$
2002	–		–	1552	–	–	–	–	–	–	–	Zeng et al. [2017b] [14]	

year	production	literature source	usage									literature source	comments
			total	metal work	PVC	leather liquors	rubber	textile	sealant	paint	other		
2003	–		–	1996	–	–	–	–	–	–	–	Zeng et al. [2017b] [14]	
2004	–		–	2588	–	–	–	–	–	–	–	Zeng et al. [2017b] [14]	
2005	–		–	2664	–	–	–	–	–	–	–	Zeng et al. [2017b] [14]	
2006	–		–	2756	–	–	–	–	–	–	–	Zeng et al. [2017b] [14]	
2007	–		–	2642	–	–	–	–	–	–	–	Zeng et al. [2017b] [14]	
2008	–		–	1990	–	–	–	–	–	–	–	Zeng et al. [2017b] [14]	
2009	–		–	1192	–	–	–	–	–	–	–	Zeng et al. [2017b] [14]	
Latvia													
2008	–		–	–	–	–	–	–	2000	–	–	Fridmanis et al. [2011]	
Lithuania													
2007	–		69	–	–	–	–	–	–	–	–	Kruopiene et al. [2014]	import
2008	–		71	–	–	–	–	–	–	–	–	Kruopiene et al. [2014]	import
North America													
1998	17800	Muir et al. [2003] [8]	–	–	–	–	–	–	–	–	–		46% $MCCPs_{prod}$
Norway													
2005	–		54–64	5	–	–	15–20	–	30–35	1	3	COWI [2010] [9]	
2007	–		78	3	–	–	4	–	46	2	23	EFTA [2010] [9]	
2009	–		205–409	3	130–260	3–10	34–101	–	32		–	COWI [2010]	
Russia													
2007	21000	ICAIA [2012]	–	–	–	–	–	–	–	–	–		
2011	27000	ICAIA [2012]	–	–	–	–	–	–	–	–	–		
Sweden													
1990	0	KEMI [1991]	3200[11]	–	–	–	–	–	–	–	–	KEMI [1991]	
1995	0	KEMI [1991]	644	–	–	–	–	–	–	–	–	KEMI [2016] [12]	55% $MCCPs_{use}$
1996	0	KEMI [1991]	358	–	–	–	–	–	–	–	–	KEMI [2016] [12]	50% $MCCPs_{use}$
1997	0	KEMI [1991]	100	–	–	–	–	–	–	–	–	KEMI [2016] [12]	23% $MCCPs_{use}$
1998	0	KEMI [1991]	109	–	–	–	–	–	–	–	–	KEMI [2016] [12]	16% $MCCPs_{use}$

year	production	literature source	usage									literature source	comments
			total	metal work	PVC	leather liquors	rubber	textile	sealant	paint	other		
1999	0	KEMI [1991]	90	–	–	–	–	–	–	–	–	KEMI [2016] [12]	17% $MCCPs_{use}$
2000	0	KEMI [1991]	127	–	–	–	–	–	–	–	–	KEMI [2016] [12]	24% $MCCPs_{use}$
2001	0	KEMI [1991]	265	–	–	–	–	–	–	–	–	KEMI [2016] [12]	52% $MCCPs_{use}$
2002	0	KEMI [1991]	180	–	–	–	–	–	–	–	–	KEMI [2016] [12]	34% $MCCPs_{use}$
2003	0	KEMI [1991]	222	–	–	–	–	–	–	–	–	KEMI [2016] [12]	49% $MCCPs_{use}$
2004	0	KEMI [1991]	183	–	–	–	–	–	–	–	–	KEMI [2016] [12]	44% $MCCPs_{use}$
2005	0	KEMI [1991]	235	–	–	–	–	–	–	–	–	KEMI [2016] [12]	31% $MCCPs_{use}$
2005	0	KEMI [1991]	94.1	65.8	3.5	–	–	–	22.8		2	COWI [2010] [7]	
2006	0	KEMI [1991]	335	–	–	–	–	–	–	–	–	KEMI [2016] [12]	36% $MCCPs_{use}$
2007	0	KEMI [1991]	294	–	–	–	–	–	–	–	–	KEMI [2016] [12]	47% $MCCPs_{use}$
2008	0	KEMI [1991]	192	–	–	–	–	–	–	–	–	KEMI [2016] [12]	31% $MCCPs_{use}$
2009	0	KEMI [1991]	112	–	–	–	–	–	–	–	–	KEMI [2016] [12]	26% $MCCPs_{use}$
2010	0	KEMI [1991]	94	–	–	–	–	–	–	–	–	KEMI [2016] [12]	21% $MCCPs_{use}$
Thailand													
1994	20000	ICIS [1995]	–	–	–	–	–	–	–	–	–		capacity
United Kingdom													
1991	40000	Willis et al. [1994][12]	12000	1200	8400	–	–	–	1200	1200	–	Willis et al. [1994] [13]	80% $MCCPs_{prod}$
2003	–		9968	1500	8000	0	100	–	300		100	COWI [2010] [7]	
United States													
2007	–		45300	–	–	–	–	–	–	–	–	EPA [2009]	66% $MCCPs_{use}$
World													
1977	–		110000	–	–	–	–	–	–	–	–	Campbell and McConnell [1980]	48% $MCCPs_{use}$
1980	–		–	–	–	–	–	–	–	–	–	Mukherjee [1990]	50% MCCPs

year	production	literature source	usage									literature source	comments
			total	metal work	PVC	leather liquors	rubber	textile	sealant	paint	other		

[1] approx. split 2/3 used in sealants and 1/3 used in paints
[2] solvent for carbonless copy paper
[3] numbers provided by Euro-Chlor
[4] cites Entec (2004). Risk reduction strategy and analysis of advantages and drawbacks for medium chain chlorinated paraffins. Stage 3 Report. Unpublished report produced under contract for the Department for Environment, Food and Rural Affairs (UK).
[5] 40% of the amount produced in the EU is exported (ECHA [2008] cites Defra 2008)
[6] from IUCLID – International Uniform Chemical Information Database (existing substances)
[7] cites Entec (2008). Environmental risk reduction strategy and analysis of advantages and drawbacks of medium-chain chlorinated paraffins (MCCPs) – updated report. For Department for Environment, Food and Rural Affairs (DEFRA), November 2008
[8] unpublished data from CPIA (Chlorinated Paraffin Industry Association) (2000).
[9] numbers from the Norwegian Product Register
[10] FRCJ Japan (2003). Hearing survey by AIST, flame retardant chemicals association of Japan
[11] 50% in electrical cables as flame retardant, 50% in floor covering
[12] numbers from the Swedish Product Register
[13] numbers provided by ICI Chemicals and Polymers Limited
[14] numbers from the Japan Lubricating Oil Society. Report of fund for environmental protection of lubricating oil in 2002 (for 2002 data), in 2003 (for 2003 data), in 2004 (for 2004 data), in 2005 (for 2005 and 2006 data), and in 2008 (for data from 2007 to 2009), respectively

Table S2: Production and usage of LCCPs [tons/year]. The percentages under 'comments' refer to the fractions of LCCPs compared to total CPs. '–' means no data available.

year	production	literature source	usage									literature source	comments
			total	metal work	PVC	leather liquors	rubber	textile	sealant	paint	other		
Canada													
2001	–		200	–	–	–	–	–	–	–	–	Canada [2012]	6.7% $LCCPs_{use}$
European Union													
1985	–		6600	–	–	–	–	–	–	–	–	IPEN [2007]	12% $LCCPs_{use}$
1992	–		16500	–	–	–	–	–	–	–	–	Willis et al. [1994]	15% $LCCPs_{use}$
1998–2004	–		5000–10000	–	–	–	–	–	–	–	–	Brooke et al. [2009]	% for 2004
Japan													
2001	–		3721	390	966	–	–	–	–	–	2365	Tsunemi [2010] [1]	32% $LCCPs_{use}$
2002	–		–	135	–	–	–	–	–	–	–	Zeng et al. [2017b] [4]	
2003	–		–	125	–	–	–	–	–	–	–	Zeng et al. [2017b] [4]	
2004	–		–	180	–	–	–	–	–	–	–	Zeng et al. [2017b] [4]	
2005	–		–	207	–	–	–	–	–	–	–	Zeng et al. [2017b] [4]	
2006	–		–	198	–	–	–	–	–	–	–	Zeng et al. [2017b] [4]	
2007	–		–	123	–	–	–	–	–	–	–	Zeng et al. [2017b] [4]	
2008	–		–	134	–	–	–	–	–	–	–	Zeng et al. [2017b] [4]	
2009	–		–	180	–	–	–	–	–	–	–	Zeng et al. [2017b] [4]	
North America													
1998	12700	Muir et al. [2003] [2]	–	–	–	–	–	–	–	–	–		33% $LCCPs_{prod}$
Sweden													
1990	0	KEMI [1991]	–	–	–	–	5	–	–	–	–	KEMI [1991]	
1998	0	KEMI [1991]	400	–	–	–	–	–	–	–	–	KEMI [2011] [3]	60% $LCCPs_{use}$
1999	0	KEMI [1991]	363	–	–	–	–	–	–	–	–	KEMI [2011] [3]	69% $LCCPs_{use}$
2000	0	KEMI [1991]	374	–	–	–	–	–	–	–	–	KEMI [2011] [3]	71% $LCCPs_{use}$
2001	0	KEMI [1991]	233	–	–	–	–	–	–	–	–	KEMI [2011] [3]	43% $LCCPs_{use}$
2002	0	KEMI [1991]	301	–	–	–	–	–	–	–	–	KEMI [2011] [3]	60% $LCCPs_{use}$
2003	0	KEMI [1991]	254	–	–	–	–	–	–	–	–	KEMI [2011] [3]	51% $LCCPs_{use}$

year	production	literature source	usage									literature source	comments
			total	metal work	PVC	leather liquors	rubber	textile	sealant	paint	other		
2004	0	KEMI [1991]	233	–	–	–	–	–	–	–	–	KEMI [2011] [3]	54% $LCCPs_{use}$
2005	0	KEMI [1991]	285	–	–	–	–	–	–	–	–	KEMI [2011] [3]	53% $LCCPs_{use}$
2006	0	KEMI [1991]	336	–	–	–	–	–	–	–	–	KEMI [2011] [3]	49% $LCCPs_{use}$
2007	0	KEMI [1991]	315	–	–	–	–	–	–	–	–	KEMI [2011] [3]	51% $LCCPs_{use}$
2008	0	KEMI [1991]	414	–	–	–	–	–	–	–	–	KEMI [2011] [3]	67% $LCCPs_{use}$
2009	0	KEMI [1991]	300	–	–	–	–	–	–	–	–	KEMI [2011] [3]	71% $LCCPs_{use}$
2010	0	KEMI [1991]	336	–	–	–	–	–	–	–	–	KEMI [2011] [3]	76% $LCCPs_{use}$
World													
1977	–		60000	–	–	–	–	–	–	–	–	Campbell and McConnell [1980]	26% $LCCPs_{use}$

[1] FRCJ Japan (2003). Hearing survey by AIST, flame retardant chemicals association of Japan
[2] unpublished data from CPIA (Chlorinated Paraffin Industry Association) (2000).
[3] numbers from the Swedish Product Register
[4] from Japan Lubricating Oil Society. Report of fund for environmental protection of lubricating oil in 2002 (for 2002 data), in 2003 (for 2003 data), in 2004 (for 2004 data), in 2005 (for 2005 and 2006 data), and in 2008 (for data from 2007 to 2009), respectively

S1.2 Technical mixtures

Sample preparation Technical CP-52 formulations were dissolved in cyclohexane (1-2 mg/mL). Aliquots were cleaned on a small 1 g silica column (silica gel 60, Merck). Silica was cleaned with 2 mL dichloromethane and conditioned with 4 mL n-hexane. Then, 100 μg per formulation were applied. The elution series consisted of, first, pure n-hexane (4 mL), second, a 1:1 mixture (5 mL) of n-hexane and dichloromethane (DCM) and, third, a respective 2:8 mixture (5 mL). The fractions were separately collected. All CPs eluted in the second fraction. Solvent was evaporated and redissolved in 1 mL of cyclohexane. Aliquots of 50 ng/μL in acetonitrile were prepared for chemical analysis. No blank issues are reported compared to the high CP concentrations that were processed.

Instrumental Analysis Ultra-performance liquid chromatography (UPLC) coupled to electrospray ionization coupled to an Orbitrap high resolution mass spectrometer (HRMS, R = 140 000 FWHM at m/z 200), was operated in full scan negative ion mode. $[M+acetate]^-$ adduct ions were monitored. Settings were applied according to Cariou et al. [2016]. The injected volume was 5 μL. Chromatographic separation was performed using a Hypersil Gold column (100 mm * 2.1 mm, 1.9 μm) at a flow rate of 0.4 mL/min using 10 mM ammonium acetate in water (A) and in acetonitrile (B). The LC gradient was applied as follows: (A/B) 50:50 for 1 min, ramped to 0:100 over 9 min, hold for 5 min and returned to 50:50, followed by equilibration. Following MS settings were applied: capillary temperature 300 °C, source heater temperature 150 °C and spray voltage 2.5 kV.

Data processing Target compounds were chlorinated paraffins with sum formula $C_nH_{2n+2-x}Cl_x$ ($10 \leq n \leq 30$ and $5 \leq x \leq n$). The theoretical $[M+acetate]^-$ mass clusters for all homologues were derived from Envipat web 2.2 [Loos et al., 2015]. The target ions were extracted from the recorded full scan mass spectra using Thermo Xcalibur 4.0.27.19 QuanBrowser. The extracted ion intensities were corrected for their contribution to the full mass cluster of the respective homologue. It is expected that the chain length influences the response of CPs as shown by Bogdal et al. [2015]. Thus, response factors were approximated for SCCPs, MCCPs and LCCPs as followed: Three Ehrenstorfer CP mixtures with chlorination degrees close to CP-52 (SCCP 51.5 wt %Cl, MCCP 52 wt %Cl and LCCP 49 wt %Cl) were analyzed. For each Ehrenstorfer mixture, sum intensities (of all present homologues) were derived and normalized for the sum intensity of the analyzed SCCP mixture. This gave response correction factors of 0.24 for homologues of chain length C_{14} to C_{17} (MCCPs) and 0.20 for homologues of chain length $C_{\leq 18}$ (LCCPs). This procedure is an approximate response factor correction to roughly compensate for the higher response of MCCPs and LCCPs compared to SCCPs.

Results The results of the analysis for the sum of SCCPs, MCCPs, and LCCPs are shown in Table S3. The congener profile for the S- and MCCPs (averaged over the eleven samples) is shown in Fig. S1.

Table S3: Fractions of S-, M-, and LCCPs in the technical mixture CP-52 in percent. All samples originate from Chinese producers. Sample IDs with different letters indicate samples from different producers.

Sample ID	fraction SCCPs	fraction MCCPs	fraction LCCPs	reference
A-52	42	58	0	this study
B-52	0	100	0	this study
C-52	42	56	2.5	this study
D-52a	63	29	7.7	this study
D-52b	55	45	0	this study
E-52	52	40	8.3	this study
F-52	48	40	12	this study
G-52a	24	65	11	this study
G-52b	40	59	1.4	this study
H-52	34	66	0.3	this study
I-52	33	67	0	this study
J-52	25	-	-	Gao et al. [2012]
K-52	40	-	-	Gao et al. [2016]
L-52	11	-	-	Xu et al. [2016]
M-52	66	-	-	Zou et al. [2017]
N-52	33	41	27	Li et al. [2018]
O-52	2.3	47	50	Li et al. [2018]
P-52	53	35	12	Li et al. [2018]

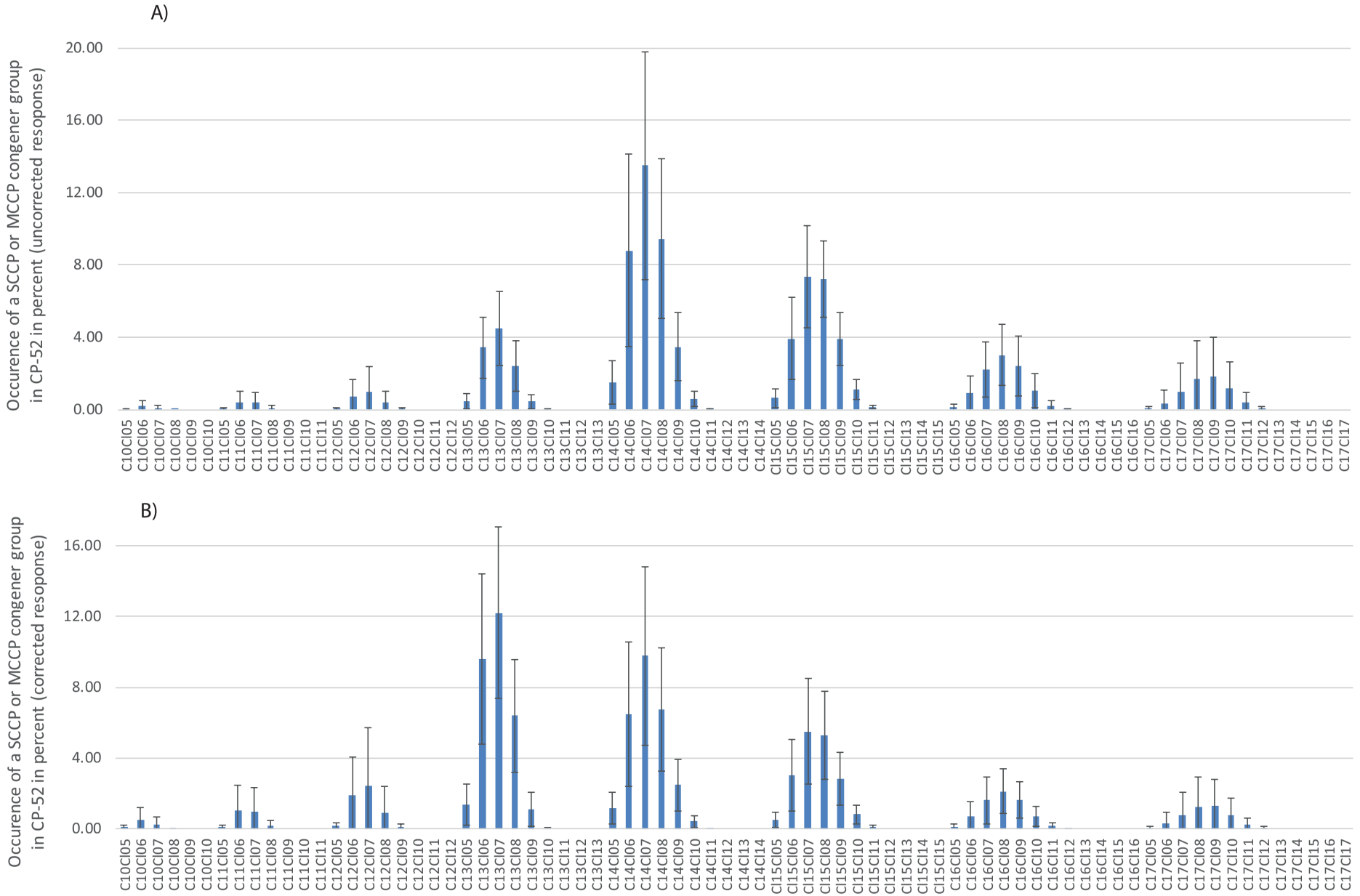

Figure S1: Occurerence of a SCCP or MCCP congener group in CP-52 (average over eleven samples) with A) uncorrected response and B) corrected response. SCCPs had a response factor correction of 1, MCCPs of 0.24.

S2 Chemical properties

S2.1 Calculated properties

Table S4: Physicochemical properties of MCCPs calculated with COSMOtherm. Vapor pressure (P_L), and solubility in water (S_W) and in octanol (S_O) were obtained with COSMO*therm*, for more details see Glüge et al. [2013]. Data given in square brackets represent the range of the isomers.

Molecular formula	Cl [%]	P_L [Pa] COSMO*therm*	S_W [μg/L] COSMO*therm*	S_O [g/L] COSMO*therm*
$C_{14}H_{27}Cl_3$	35.3	[$1.31\cdot10^{-4}$, $1.30\cdot10^{-2}$]	[5.02, 40.4]	[297, 412]
$C_{14}H_{25}Cl_5$	47.9	[$7.95\cdot10^{-5}$, $1.48\cdot10^{-3}$]	[3.86, 34.3]	[329, 550]
$C_{14}H_{23}Cl_7$	56.5	[$8.79\cdot10^{-7}$, $1.47\cdot10^{-4}$]	[3.16, 44.5]	[219, 524]
$C_{14}H_{21}Cl_9$	62.8	[$5.23\cdot10^{-8}$, $1.47\cdot10^{-5}$]	[2.78, 272]	[366, 560]
$C_{14}H_{19}Cl_{11}$	67.6	[$1.46\cdot10^{-8}$, $6.61\cdot10^{-7}$]	[2.60, 87.0]	[431, 700]
$C_{14}H_{18}Cl_{12}$	69.6	[$8.38\cdot10^{-9}$, $1.65\cdot10^{-7}$]	[1.90, 68.2]	[291, 668]
$C_{15}H_{29}Cl_3$	33.7	[$1.03\cdot10^{-4}$, $3.89\cdot10^{-3}$]	[1.56, 9.88]	[297, 403]
$C_{15}H_{27}Cl_5$	46.1	[$7.55\cdot10^{-6}$, $5.14\cdot10^{-4}$]	[1.12, 13.8]	[252, 486]
$C_{15}H_{25}Cl_7$	54.7	[$3.47\cdot10^{-7}$, $5.30\cdot10^{-5}$]	[0.93, 17.6]	[228, 484]
$C_{15}H_{23}Cl_9$	61.1	[$2.88\cdot10^{-8}$, $5.56\cdot10^{-6}$]	[0.72, 7.38]	[300, 579]
$C_{15}H_{21}Cl_{11}$	66.0	[$1.09\cdot10^{-8}$, $2.57\cdot10^{-7}$]	[0.89, 68.1]	[384, 596]
$C_{15}H_{19}Cl_{13}$	69.8	[$6.37\cdot10^{-10}$, $3.73\cdot10^{-8}$]	[0.50, 16.0]	[287, 639]
$C_{16}H_{31}Cl_3$	32.3	[$1.50\cdot10^{-5}$, $1.39\cdot10^{-3}$]	[0.38, 3.13]	[296, 390]
$C_{16}H_{29}Cl_5$	44.5	[$5.21\cdot10^{-6}$, $1.66\cdot10^{-4}$]	[0.29, 4.81]	[263, 541]
$C_{16}H_{27}Cl_7$	53.1	[$1.33\cdot10^{-7}$, $1.64\cdot10^{-5}$]	[0.28, 6.89]	[206, 534]
$C_{16}H_{25}Cl_9$	59.5	[$1.33\cdot10^{-8}$, $2.01\cdot10^{-6}$]	[0.22, 11.8]	[294, 547]
$C_{16}H_{23}Cl_{11}$	64.4	[$1.45\cdot10^{-9}$, $1.94\cdot10^{-7}$]	[0.19, 27.8]	[333, 522]
$C_{16}H_{21}Cl_{13}$	68.4	[$1.93\cdot10^{-10}$, $1.86\cdot10^{-8}$]	[0.15, 42.2]	[289, 1070]
$C_{17}H_{33}Cl_3$	31.0	[$1.31\cdot10^{-5}$, $4.77\cdot10^{-4}$]	[0.10, 0.84]	[292, 374]
$C_{17}H_{31}Cl_5$	43.0	[$1.18\cdot10^{-6}$, $5.02\cdot10^{-5}$]	[0.08, 1.51]	[230, 556]
$C_{17}H_{29}Cl_7$	51.6	[$3.96\cdot10^{-8}$, $6.17\cdot10^{-6}$]	[0.08, 3.28]	[179, 497]
$C_{17}H_{27}Cl_9$	58.0	[$3.50\cdot10^{-9}$, $7.54\cdot10^{-7}$]	[0.08, 12.7]	[364, 557]
$C_{17}H_{25}Cl_{11}$	63.0	[$9.39\cdot10^{-10}$, $7.26\cdot10^{-8}$]	[0.04, 11.5]	[318, 587]
$C_{17}H_{23}Cl_{13}$	67.0	[$4.77\cdot10^{-11}$, $6.63\cdot10^{-9}$]	[0.04, 5.37]	[289, 508]
$C_{17}H_{21}Cl_{15}$	70.3	[$6.64\cdot10^{-12}$, $4.70\cdot10^{-10}$]	[0.02, 1.71]	[259, 980]

Table S5: Physicochemical properties of MCCPs. Octanol-water (K_{OW}), air-water (K_{AW}), and octanol-air (K_{OA}) partition coefficient for MCCPs were obtained with COSMO*therm*, for more details see Glüge et al. [2013]. The particle bound fractions were calculated according to Eqs. (S1) to (S3). Data given in square brackets represent the range of the isomers.

Molecular formula	Cl [%]	K_{OW} [-] exp. values	K_{OW} [-] COSMO*th.*	log K_{AW} [-] COSMO*th.*	log K_{OA} [-] COSMO*th.*	particle–bound fraction [%]
$C_{14}H_{27}Cl_3$	35.3		[6.77, 7.80]	[-3.40, -0.50]	[8.43, 10.3]	[0.08, 6.08]
$C_{14}H_{25}Cl_5$	47.9	6.3 ± 0.01[a], 5.52[b]	[6.88, 7.89]	[-3.44, -1.24]	[9.33, 10.6]	[0.64, 10.6]
$C_{14}H_{23}Cl_7$	56.5	5.47[b]	[6.57, 7.97]	[-5.45, -2.08]	[10.3, 12.3]	[5.12, 86.7]
$C_{14}H_{21}Cl_9$	62.8		[6.21, 7.99]	[-6.41, -2.96]	[11.2, 13.8]	[29.9, 99.5]
$C_{14}H_{19}Cl_{11}$	67.6		[6.87, 8.09]	[-7.40, -4.22]	[12.5, 14.4]	[91.0, 99.9]
$C_{14}H_{18}Cl_{12}$	69.6		[6.94, 8.03]	[-7.51, -4.66]	[12.9, 14.6]	[96.1, 99.9]

$C_{15}H_{29}Cl_3$	33.7		[7.37, 8.30]	[-2.87, -0.54]	[9.88, 11.4]	[2.23, 40.9]
$C_{15}H_{27}Cl_5$	46.1		[7.14, 8.41]	[-4.07, -1.14]	[10.7, 12.3]	[13.1, 86.3]
$C_{15}H_{25}Cl_7$	54.7		[6.99, 8.48]	[-5.44, -1.98]	[11.6, 13.6]	[55.4, 99.1]
$C_{15}H_{23}Cl_9$	61.1		[7.61, 8.55]	[-6.08, -2.78]	[12.5, 15.0]	[90.5, 100]
$C_{15}H_{21}Cl_{11}$	66.0		[6.88, 8.54]	[-7.41, -4.16]	[13.9, 15.4]	[99.5, 100]
$C_{15}H_{19}Cl_{13}$	69.8		[7.53, 8.62]	[-7.97, -4.70]	[14.4, 16.6]	[99.9, 100]
$C_{16}H_{31}Cl_3$	32.3	7.2[c]	[7.86, 8.87]	[-3.19, -0.31]	[10.3, 12.2]	[5.48, 82.0]
$C_{16}H_{29}Cl_5$	44.5		[7.61, 8.96]	[-3.75, -1.03]	[11.2, 12.5]	[30.3, 90.9]
$C_{16}H_{27}Cl_7$	53.1		[7.34, 9.00]	[-5.43, -1.95]	[12.1, 14.2]	[79.9, 99.8]
$C_{16}H_{25}Cl_9$	59.5	6.81 ± 0.02[a]	[7.27, 9.06]	[-6.61, -2.69]	[12.9, 15.0]	[96.2, 100]
$C_{16}H_{23}Cl_{11}$	64.4		[7.20, 9.09]	[-7.52, -3.59]	[13.8, 16.2]	[99.5, 100]
$C_{16}H_{21}Cl_{13}$	68.4	7.36 [c]	[7.45, 9.12]	[-8.82, -4.47]	[14.7, 17.4]	[99.9, 100]
$C_{17}H_{33}Cl_3$	31.0		[8.41, 9.43]	[-2.66, -0.22]	[10.7, 12.2]	[13.7, 83.3]
$C_{17}H_{31}Cl_5$	43.0	8.21[b]	[8.04, 9.52]	[-3.88, -0.95]	[11.7, 13.1]	[57.9, 97.2]
$C_{17}H_{29}Cl_7$	51.6	8.01[b]	[7.59, 9.53]	[-5.62, -1.81]	[12.5, 14.6]	[90.9, 100]
$C_{17}H_{27}Cl_9$	58.0		[7.35, 9.58]	[-7.21, -2.67]	[13.4, 15.8]	[98.8, 100]
$C_{17}H_{25}Cl_{11}$	63.0		[7.57, 9.72]	[-7.69, -3.34]	[14.2, 16.4]	[99.8, 100]
$C_{17}H_{23}Cl_{13}$	67.0		[7.87, 9.69]	[-8.03, -4.31]	[15.2, 17.6]	[100, 100]
$C_{17}H_{21}Cl_{15}$	70.3		[8.74, 9.85]	[-8.92, -5.20]	[16.2, 18.8]	[100, 100]

[a] Hilger et al. [2011b] [b] Renberg et al. [1980] - original min/max range attributed to C_{14} and C_{17} [c] Fisk et al. [1998b]

The fractions of particle–bound MCCPs in air were calculated as:

$$\phi = \phi_{\mathrm{pf}} + \phi_{\mathrm{pc}}, \tag{S1}$$

$$\phi_{\mathrm{pf}} = \frac{c_{\mathrm{pf}} V_{\mathrm{pf}}}{c_{\mathrm{pf}} V_{\mathrm{pf}} + c_{\mathrm{pc}} V_{\mathrm{pc}} + c_{\mathrm{a}} V_{\mathrm{a}}} = \frac{1}{1 + v_{\mathrm{pc}}/v_{\mathrm{pf}} \cdot K_{\mathrm{pcpf}} + 1/(v_{\mathrm{pf}} K_{\mathrm{pfa}})}, \tag{S2}$$

$$\phi_{\mathrm{pc}} = \frac{c_{\mathrm{pc}} V_{\mathrm{pc}}}{c_{\mathrm{pf}} V_{\mathrm{pf}} + c_{\mathrm{pc}} V_{\mathrm{pc}} + c_{\mathrm{a}} V_{\mathrm{a}}} = \frac{1}{1 + v_{\mathrm{pf}}/(v_{\mathrm{pc}} K_{\mathrm{pcpf}}) + 1/(v_{\mathrm{pc}} K_{\mathrm{pca}})}, \tag{S3}$$

where ϕ_{pf} is the fraction bound to fine particles (or aerosols), and ϕ_{pc} the fraction bound to coarse particles (or aerosols). These two fractions represent in a simplified way the size distribution of particles in the atmosphere. Here, coarse and fine particles are defined as particulate matter with a diameter of 2.5 μm–10 μm and <2.5 μm, respectively.
The partition coefficients K_{pca}, K_{pfa}, and K_{pcpf} were calculated as:

$$K_{\mathrm{pca}} = 1.22 \cdot K_{\mathrm{oa}} \cdot f_{\mathrm{ompc}} \cdot \rho_{\mathrm{pc}}/1000, \tag{S4}$$

$$K_{\mathrm{pfa}} = 1.22 \cdot K_{\mathrm{oa}} \cdot f_{\mathrm{ompf}} \cdot \rho_{\mathrm{pf}}/1000, \tag{S5}$$

$$K_{\mathrm{pcpf}} = K_{\mathrm{pca}}/K_{\mathrm{pfa}}, \tag{S6}$$

where f_{ompc} and f_{ompf} are the fractions of organic matter, and ρ_{pc} and ρ_{pf} are the densities of the coarse and fine particles, respectively. The fractions of organic matter were set to 0.08 and 0.22 [Putaud et al., 2004], the densities to 1930 kg/m^3 and 1620 kg/m^3, respectively [Hu et al., 2012]. The volume fractions of coarse and fine particles in air (v_{pc}, v_{pf}) were set to $3 \cdot 10^{-12}$ and $9 \cdot 10^{-12}$, respectively. These values are representative for natural background in Europe [Putaud et al., 2004].

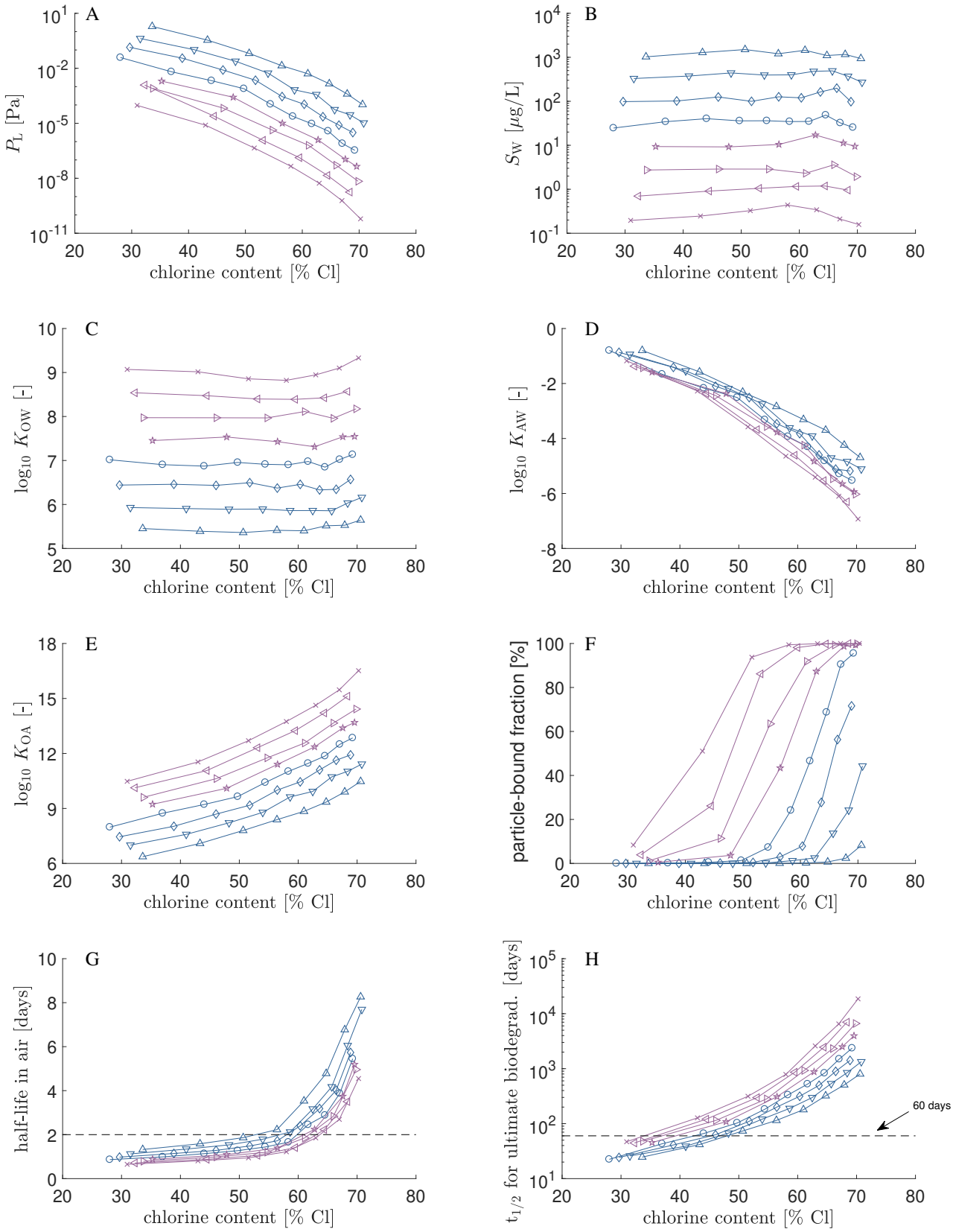

Figure S2: Dependence of the averaged (geometric average) property values calculated with COSMO*therm* (A−E) and EPI Suite™ (G−H) on the chlorine content. Data in Subfigure F were calculated according to Eqs. (S1) to (S3). Symbols in blue represent SCCPs (△ C_{10}; ▽ C_{11}; ◇ C_{12}; o C_{13}), symbols in violet MCCPs (⋆ C_{14}; ▷ C_{15}; ◁ C_{16}; x C_{17}). Figures A−E are adapted from Glüge et al. [2013].

Table S6: Calculated half-lifes in air and biodegradation half-lifes for MCCPs. Data were obtained from EPISuite™ with AOPWIN (half-lifes in air), Biowin2 (fast biodegradation), Biowin3 (ultimate biodegradation), and Biowin4 (primary biodegradation). Data given in square brackets represent the range of the isomers considered.

Molecular formula	Cl [%]	half-life in air [days]	probab. for fast biodegradation	ultimate biodegradation[a] [days]	primary biodegradation[a] [days]
$C_{14}H_{27}Cl_3$	35.3	[0.74, 1.06]	[0.00, 0.04]	[26.4, 79.4]	[3.90, 10.5]
$C_{14}H_{25}Cl_5$	47.9	[0.91, 1.29]	[0.00, 0.00]	[66.3, 199]	[6.78, 18.3]
$C_{14}H_{23}Cl_7$	56.5	[1.18, 1.78]	[0.00, 0.00]	[288, 498]	[19.4, 31.8]
$C_{14}H_{21}Cl_9$	62.8	[1.66, 3.01]	[0.00, 0.00]	[721, 1248]	[33.7, 55.3]
$C_{14}H_{19}Cl_{11}$	67.6	[2.80, 5.34]	[0.00, 0.00]	[3128, 3128]	[96.3, 96.3]
$C_{14}H_{18}Cl_{12}$	69.6	[4.28, 6.75]	[0.00, 0.00]	4950	127
$C_{15}H_{29}Cl_3$	33.7	[0.67, 0.95]	[0.00, 0.03]	[28.0, 84.1]	[4.05, 10.9]
$C_{15}H_{27}Cl_5$	46.1	[0.81, 1.26]	[0.00, 0.00]	[70.2, 211]	[7.04, 19.0]
$C_{15}H_{25}Cl_7$	54.7	[1.02, 1.44]	[0.00, 0.00]	[176, 528]	[12.3, 33.0]
$C_{15}H_{23}Cl_9$	61.1	[1.36, 2.55]	[0.00, 0.00]	[763, 1322]	[35.0, 57.4]
$C_{15}H_{21}Cl_{11}$	66.0	[2.04, 4.24]	[0.00, 0.00]	[1911, 3311]	[60.9, 99.9]
$C_{15}H_{19}Cl_{13}$	69.8	[4.12, 6.37]	[0.00, 0.00]	8296	174
$C_{16}H_{31}Cl_3$	32.3	[0.62, 0.84]	[0.00, 0.03]	[29.6, 89.0]	[4.20, 11.3]
$C_{16}H_{29}Cl_5$	44.5	[0.74, 1.10]	[0.00, 0.00]	[74.3, 223]	[7.31, 19.7]
$C_{16}H_{27}Cl_7$	53.1	[0.90, 1.21]	[0.00, 0.00]	[186, 559]	[12.7, 34.3]
$C_{16}H_{25}Cl_9$	59.5	[1.15, 1.73]	[0.00, 0.00]	[808, 1399]	[36.3, 59.6]
$C_{16}H_{23}Cl_{11}$	64.4	[1.61, 2.94]	[0.00, 0.00]	[2024, 3506]	[63.2, 104]
$C_{16}H_{21}Cl_{13}$	68.4	[2.67, 4.87]	[0.00, 0.00]	8784	180
$C_{17}H_{33}Cl_3$	31.0	[0.57, 0.76]	[0.00, 0.02]	[31.4, 94.2]	[4.36, 11.8]
$C_{17}H_{31}Cl_5$	43.0	[0.67, 1.08]	[0.00, 0.00]	[78.6, 236]	[7.59, 20.4]
$C_{17}H_{29}Cl_7$	51.6	[0.80, 1.11]	[0.00, 0.00]	[197, 591]	[13.2, 35.6]
$C_{17}H_{27}Cl_9$	58.0	[1.00, 1.47]	[0.00, 0.00]	[494, 1482]	[23.0, 61.9]
$C_{17}H_{25}Cl_{11}$	63.0	[1.33, 2.80]	[0.00, 0.00]	[2143, 3712]	[65.6, 108]
$C_{17}H_{23}Cl_{13}$	67.0	[1.97, 3.95]	[0.00, 0.00]	[5368, 9300]	[114, 187]
$C_{17}H_{21}Cl_{15}$	70.3	[3.84, 5.72]	[0.00, 0.00]	23300	326

[a] The scores for primary and ultimate biodegradation, calculated by EPISuite™, were converted into half-lives in water according to Scheringer et al. [2012].

S2.2 Bioconcentration factors for SCCPs

EC [2000] summarized the available studies for the bioconcentration factors (BCF) for SCCPs and we describe the most important studies (which describe BCFs at steady state) here briefly. The study of Madeley and Maddock (1983a) exposed rainbow trout (*Oncorhynchus mykiss*) to measured concentrations of 0.033, 0.1, 1.07 and 3.05 mg/L of a C_{10-12}, 58% wt Cl for 60 days. The concentrations were determined by means of a ^{14}C-labeled chlorinated *n*-undecane mixed into the commercial product. In addition, parent compound analysis was also undertaken at various times during the test. Whole body BCFs of 1173−7816 were determined based on radioactivity measurements in the fish and BCFs of 574−7273 were determined based on the parent compound analysis. The BCFs were found to increase with decreasing exposure concentration.

Madeley and Maddock (1983b) conducted also a second study were they exposed rainbow trout (*Oncorhynchus mykiss*) again to a SCCP (C_{10-12}, 58% wt) at concentrations of 3.1 and 14.3 μg/L. Exposure

was for 168 days using a flow-through system. The bioconcentration was measured by means of a ^{14}C-labeled chlorinated *n*-undecane mixed into the commercial product. The whole fish BCF was estimated to be 3600−5300. These BCFs were based on the amount of ^{14}C-labeled material present in the various organs. A limited number of parent compound analyses were also carried out at various times during the tests, and these indicated that some of the 14C-label present in the liver and viscera may not have been the parent chlorinated paraffin. Therefore, these measured BCFs are likely to represent maximum values.

Additional information were submitted by Japan to the Persistent Organic Pollutant Review Committee [POPRC, 2009]. The Japanese study tested a C_{13}-CP with 48.7% wt. Cl. Three main components were identified in the mixture: $C_{13}H_{23}Cl_5$ (49.8% wt. Cl), $C_{13}H_{22}Cl_6$ (54.5% wt. Cl) and $C_{13}H_{21}Cl_7$ (58.4% wt. Cl). The fish used in the test were Common Carp (*Cyprinus carpio*) and the exposure period was 62 days. The test was carried out using nominal concentrations of 0.01 mg/L and 0.001 mg/L and a flow-through system. Samples of water and fish were analyzed by parent compound analysis and steady state was reached after approximately 28 days. Lipid normalized BCFs ranged between 1962 and 2150 for the C_{13}-CP with 49.8% wt. Cl, between 2100 and 2530 for the C_{13}-CP with 54.5% wt. Cl, and between 3000 and 3630 for the C_{13}-CP with 58.4% wt. Cl.

The reported BCFs for the SCCPs and MCCPs are also shown in Fig. S3. The experimental values do not show clear trends - neither for the carbon chain nor for the chlorine content. The BCF is decreasing from C_{11} 58% Cl to C_{13} 58% Cl and from C_{13} 50% Cl to C_{15} 51% Cl, but increasing for C_{13} 50% Cl to C_{14} 45% Cl. The BCF is increasing from C_{13} 50% Cl to C_{13} 58% Cl. The study of Bengtsson et al. [1979] [EC, 2000] shows decreasing BCFs with increasing chlorine content, but given the high concentrations they used and an exposure time of only 14 days, the BCFs were most probably not determined at steady state.

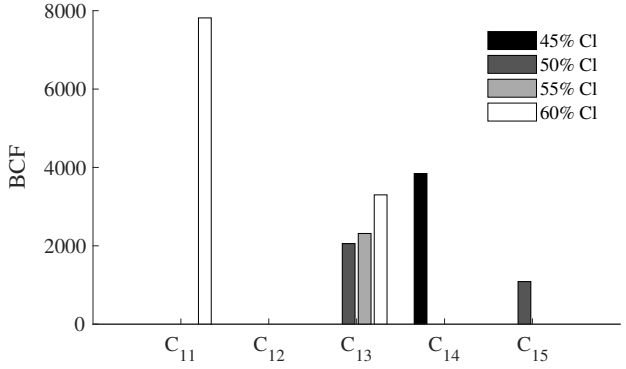

Figure S3: Reported BCFs for SCCPs

S2.3 Laboratory-derived biomagnification factors

Fisk et al. [1996, 1998a, 2000] exposed juvenile rainbow trout (*Oncorhynchus mykiss*) to different chlorinated paraffins in food, followed by a depuration phase with clean food. The CP concentrations were corrected for growth dilution and lipid normalized for all bioaccumulation parameters. CP concentrations in fish reached in most of the cases no steady state. The biomagnification factor (BMF) was therefore calculated as:

$$\mathrm{BMF} = \alpha \cdot F/k_\mathrm{d} \tag{S7}$$

where α is the assimilation efficiency, F is the feeding rate, and k_d is the depuration rate. Depuration rates were calculated by fitting the depuration phase data to a first-order decay curve (ln concentration= $a + k_\mathrm{d}$·time in days, where a is a constant). The feeding rate was assumed to be 1.5% of the body weight of the fish and corrected for the lipid percentage of the food (which was 14%; determined in the same manner as the lipid percentage in the fish). The assimilation efficiencies were calculated by fitting the concentration data to the integrated form of the kinetic rate equation for constant dietary exposure using iterative non-linear regression and the equation:

$$C_\mathrm{fish} = (\alpha F C_\mathrm{food}/k_\mathrm{d}) \cdot (1 - \exp[-k_\mathrm{d} t]) \tag{S8}$$

where C_{fish} is the concentration in the fish (lipid basis and growth corrected), C_{food} is the concentration in the food (on a lipid basis), and t is the time of uptake in days. The obtained BMFs are shown in Fig. S4

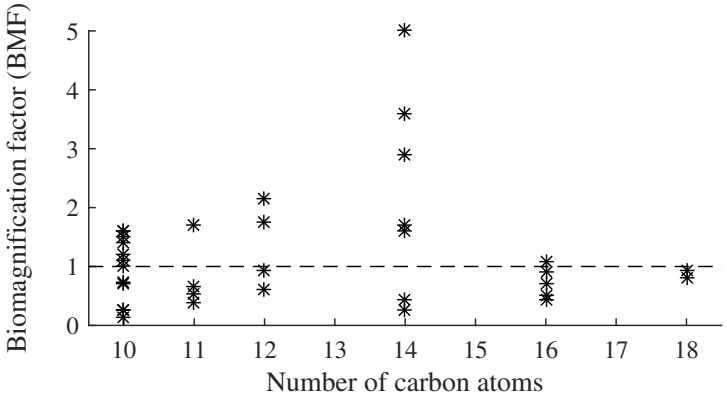

Figure S4: Relationship between laboratory-derived BMFs of chlorinated paraffins by fish and the number of carbon atoms. Data are from the studies of Fisk et al. [1996, 1998a, 2000] using the kinetically derived assimilation efficiency.

Fisk et al. [1998a, 2000] found in their two last publications that the coefficient of determination for the assimilation efficiency models tended to be low (Fig. S5). They stated in this context 'previous work on other hydrophobic organochlorines suggests that for compounds with a log K_{OW} between 4.5 and 7.0, assimilation efficiencies of organochlorines by fish are approximately 50%' and calculated additionally BMFs

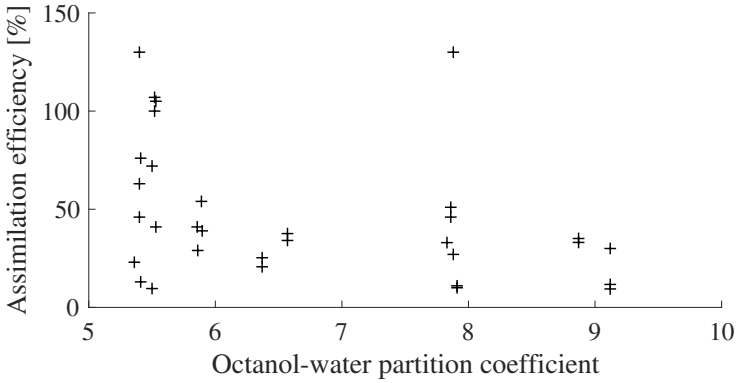

Figure S5: Relationship between assimilation efficiencies of chlorinated paraffins by fish [Fisk et al., 1996, 1998a, 2000] and the octanol-water partition coefficient [Glüge et al., 2013].

with an assumed assimilation efficiency of 50%. However, Fisk et al. [1998b] and also Arnot and Gobas [2004] showed that the assimilation efficiency of hydrophobic organochlorine compounds is decreasing for compounds with an octanol-water partition coefficient larger than seven. We think, therefore, that the lower assimilation efficiencies observed by Fisk et al. [1998a, 2000] are mainly due to the high octanol-water partition coefficient of the compounds (MCCPs have log K_{OW}s between 6.77 and 9.85) and that the BMFs calculated with the kinetically derived assimilation efficiencies are more realistic than the BMFs with an assumed assimilation efficiency of 50%.

S3 Mass interferences between CP congeners

Typical interferences occur from $C_{n+2}H_{2(n+2)+2-(x-1)}Cl_{x-1}$ when analysing $C_nH_{2n+2-x}Cl_x$. We demonstrate this here on an example. The target MCCP is $C_{16}H_{25}Cl_9$, the interference is the LCCP $C_{18}H_{30}Cl_8$. Table S7 shows the molar mass and probability of the natural occurrence of the different isotopes, Table S8 shows the abundances of the different masses of $C_{18}H_{30}Cl_8$ and $C_{16}H_{25}Cl_9$. The abundance of the signal (A_S) was calculated as:

$$A_S = \sum_{i}^{\text{isotopes}} p_i^{n_i}, \tag{S9}$$

where p_i is the probability of the occurrence of the stable isotopes ^{12}C, ^{1}H, ^{35}Cl, and ^{37}Cl. n_i is the number of the isotopes in the molecule of interest. The number of permutations (n_{per}) was calculated as:

$$n_{per} = \frac{(n_{^{35}Cl} + n_{^{37}Cl})!}{n_{^{35}Cl}! \cdot n_{^{37}Cl}!}. \tag{S10}$$

Table S7: Molar mass and probability for the different isotopes

i		^{12}C	^{1}H	^{35}Cl	^{37}Cl
Molar mass	m_i	12.00000	1.00783	34.9688527	36.9659
Probability	p_i	0.9889	0.9998	0.7578	0.2422

Table S8: Calculation of the abundance of the different masses of $C_{18}H_{30}Cl_8$ and $C_{16}H_{25}Cl_9$.

^{12}C	^{1}H	^{35}Cl	^{37}Cl	mass	abundance of signal	permu-tations	abundance	rel. abun-dance	minimum resolution
$n_{^{12}C}$	$n_{^{1}H}$	$n_{^{35}Cl}$	$n_{^{37}Cl}$	m	A_S	n_{per}	A	A_{rel} *	R_{min} **
$C_{18}H_{30}Cl_8$									
18	30	8	0	525.99	0.0884247	1	0.08842	35.0%	
18	30	7	1	527.98	0.0282614	8	0.22609	89.4%	
18	30	6	2	529.98	0.0090326	28	**0.25291**	**100%**	
18	30	5	3	531.98	0.0028869	56	0.16167	63.9%	
18	30	4	4	533.97	0.0009227	70	0.06459	25.5%	
18	30	3	5	535.97	0.0002949	56	0.01651	6.5%	
18	30	2	6	537.96	0.0000943	28	0.00264	1.0%	
18	30	1	7	539.97	0.0000301	8	0.00024	0.1%	
18	30	0	8	541.96	0.0000096	1	$9.63{\cdot}10^{-6}$	0.0%	
$C_{16}H_{25}Cl_9$									
16	25	9	0	531.92	0.0685895	1	0.06859	27.2%	8658
16	25	8	1	533.91	0.0219219	9	0.19730	78.2%	8690
16	25	7	2	535.91	0.0070064	36	**0.25223**	**100%**	8723
16	25	6	3	537.91	0.0022393	84	0.18810	74.6%	8755
16	25	5	4	539.90	0.0007157	126	0.09018	35.8%	8788
16	25	4	5	541.90	0.0002287	126	0.02882	11.4%	8820
16	25	3	6	543.90	0.0000731	84	0.00614	2.4%	
16	25	2	7	545.89	0.0000234	36	0.00084	0.3%	
16	25	1	8	547.89	0.0000075	9	$6.72{\cdot}10^{-5}$	0.0%	
16	25	0	9	549.89	0.0000024	1	$2.39{\cdot}10^{-6}$	0.0%	

* Abundance normalized to the most intensive abundance signal in the isotopic cluster

** Values of $m/\Delta m$ measured for the two peaks of equal hight in a mass spectrum at m and $m\pm\Delta m$ that are separated by a valley which at its lowest point is 10% of the height of either peak

The abundance, A, was then calculated as:

$$A = A_S \cdot n_{per}. \tag{S11}$$

Figure S6 shows the abundance of the different masses for $C_{18}H_{30}Cl_8$ and $C_{16}H_{25}Cl_9$ graphically.

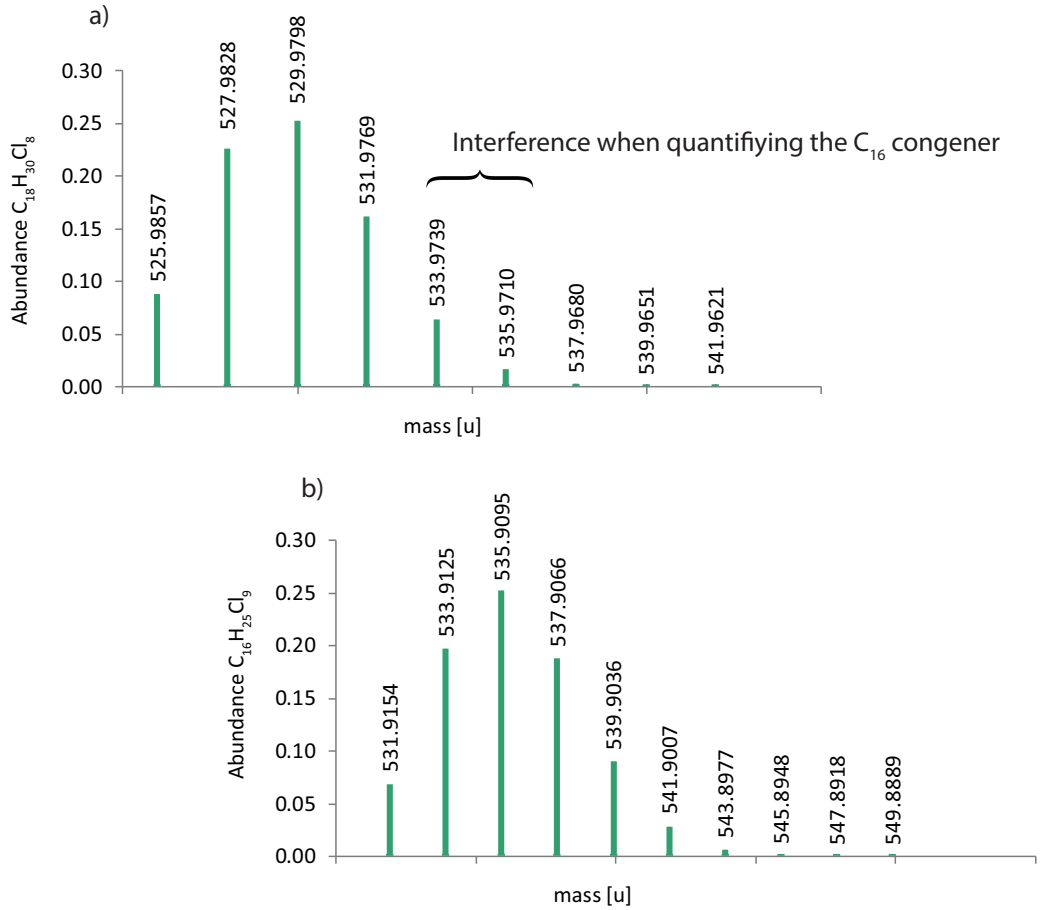

Figure S6: Abundance of the different masses of a) $C_{18}H_{30}Cl_8$ and b) $C_{16}H_{25}Cl_9$.

The quantification is normally based on the two highest isotope signals, this would be mass 535.91 Da and mass 533.91 Da for $C_{16}H_{25}Cl_9$. An amount of one gives a signal of 0.4495 (0.2522 + 0.1973). At a same amount one, the interfering masses of $C_{18}H_{30}Cl_8$ (535.97 Da and 533.97 Da) give a signal of 0.0811 (0.0165 + 0.0646). The contribution of the interfering congener ($C_{18}H_{30}Cl_8$) to the measured signal is therefore 15%.

The minimum resolution to resolve the peaks of $C_{18}H_{30}Cl_8$ and $C_{16}H_{25}Cl_9$ with mass spectrometry is shown in Table S8 under minimum resolution. The values were calculated as:

$$R_{min} = \frac{m_{C_{16}}}{|m_{C_{18}} - m_{C_{16}}|}, \tag{S12}$$

where $m_{C_{16}}$ is one of the masses of $C_{16}H_{25}Cl_9$ and $m_{C_{18}}$ is the interfering mass of $C_{18}H_{30}Cl_8$. Minimum resolutions for isobaric interferences are provided in Yuan et al. [2016].

S4 Information on the measured concentrations in the environment, biota, and in humans

S4.1 Omitted studies

Table S9: Omitted studies in the review and the reasons for omitting them.

Omitted study	Reason
Air, soil, birds, human tissue	
All available studies included	
Water	
Houde et al. [2008]	Measured only filtered water, not the particulate phase
Willis et al. [1994]	Measured before 1995 (only semi-quantitative analytical methods)
WHO [1996]	Measured before 1995 (only semi-quantitative analytical methods)
BUA [1992]	Measured before 1995 (only semi-quantitative analytical methods)
Petersen et al. [2006]	Only two values, one of them below the limit of detection
NIVA [2017]	Measured only filtered water, not the particulate phase
Surface sediment	
EPA [1988]	Measured before 1995 (only semi-quantitative analytical methods)
BUA [1992]	Measured before 1995 (only semi-quantitative analytical methods)
WHO [1996]	Measured before 1995 (only semi-quantitative analytical methods)
Fish	
Miljodirektoratet [2013]	Too many no detects
Bennie et al. [2000]	Measurements were probably too high due to coeluting interferences
Mammals	
NILU [2016a]	Too many no detects
Bennie et al. [2000]	Measurements were probably too high due to coeluting interferences
Bioegradation in sediment	
Fisk et al. [1998b]	The estimated half-lives for biodegradation were based on the difference between toluene-extractable ^{14}C measurements (taken to represent unchanged chlorinated paraffin) and total ^{14}C measurements (taken to represent degraded chlorinated paraffin). However, the results of this test should be treated with caution as the identity of the ^{14}C present in the samples was not determined, and it was only assumed that the non-extractable ^{14}C represented metabolites [EC, 2005].
BCF	
Thompson et al. [2000]	The test was based on radiolabeled residues and may therefore have included metabolites of the parent substance [Thompson and Vaughan, 2013].
Vaughan and Hurd [2010]	The test was based on radiolabeled residues and may therefore have included metabolites of the parent substance [Thompson and Vaughan, 2013].
BAF	
Houde et al. [2008]	Values were based on filtered water only. Also water samples were taken at different locations and time points compared to the biota samples.

BMF	
Zeng et al. [2017b]	The lipid-normalized concentrations in the marine mammals were based on blubber samples, while the lipid-normalized concentrations in the fish and crustaceans were based on muscle and soft tissue samples. Some of the derived BMFs even compared concentrations in marine mammals and fish from very different sites.

S4.2 Included studies

Table S10: Information on the concentrations shown in Figures 3, 4, and 5 in the main text. The column congeners indicates if congener data were available, either from the publication or on request. Ratio stands for the MCCP to SCCP ratio of the medians.

country	year	congeners	MCCPs median	MCCPs 25th and 75th quantile	SCCPs median	SCCPs 25th and 75th quantile	ratio	literature source
Concentrations in air [pg/m^3]								
Arctic	2013	C_{14-15}[a]	22.1	[7.05, 37.2]	338	[220, 497]	0.07	NILU [2014]
Arctic	2014	C_{14-15}[a]	12.0	[3.86, 35.6]	239	[172, 309]	0.05	NILU [2015]
Arctic	2015	C_{14-15}[a]	77.5	[26.5, 137]	370	[274, 489]	0.21	NILU [2016b]
Arctic	2016	C_{14-15}[a]	36.5	[26.0, 67.5]	186	[162, 269]	0.20	NILU [2017b]
Antarctic	2013	C_{14-17}	4.62	[3.81, 5.10]	15.6	[11.7, 17.8]	0.30	Ma et al. [2014]
UK	2003	no	594	[170, 869]	220	[142, 391]	2.70	Barber et al. [2005]
Norway	2016	no	14.3	[9.88, 18.1]	140	[100, 1483]	0.10	NILU [2017a]
Switzerland	2012	C_{14-16}	9575	[1750, 17400]	4550	[4270, 4830]	2.10	Bogdal et al. [2015]
Pakistan	2006	no	4168	[1106, 7309]	2403	[909, 4639]	1.73	Chaemfa et al. [2014]
India	2011	no	1533	[596, 8216]	6922	[3532, 12617]	0.22	Chaemfa et al. [2014]
China	2010	C_{14-17}	12490	[5047, 36479]	19962	[11818, 42164]	0.63	Wang et al. [2013]
China	2013/4	C_{14-17}	3530		5060		0.70	Li et al. [2018]
Concentrations in water [ng/L]								
Norway	2013	no	44.5	[23.4, 129]	18	[17.3, 200]	2.48	NIVA [2014a]
Norway	2014	no	41.2	[14.9, 87.2]	719	[554, 938]	0.06	NIVA [2015b]
Norway	2015	no	19.3	[18.3, 25.8]	134	[99.5, 201]	0.15	NIVA [2016c]
Arctic	2004	no	49.4	[24.5, 101]	8.60	[5.47, 26.1]	5.74	Dick et al. [2010]
Concentrations in soil [ng/g dw]								
Norway	2015	no	14.0	[7.10, 34.3]	17.0	[15.3, 33.0]	0.82	NILU [2016a]
Norway	2016	no	1.40	[0.94, 2.78]	121	[79.5, 210]	0.01	NILU [2017a]
Switzerland	2002	no	21.0	[20.0, 35.5]	14.0	[9.50, 17.3]	1.50	Iozza [2010]
Switzerland	1989-2014	C_{14-17}	13.7	[6.20, 35.7]	6.77	[5.08, 10.7]	2.02	Bogdal et al. [2016]
S. Africa	2006	no	77.5	[14.5, 305]	20.5	[12.1, 34.5]	3.78	Quinn et al. [2009]
China	2009	C_{14-17}	19.2	[10.5, 36.9]	9.52	[6.55, 13.8]	2.01	Wang et al. [2013]
China subur.	2011	C_{14-17}	15.3	[11.3, 24.4]	3.52	[2.49, 5.95]	4.35	Wang et al. [2017]
China urban	2011	C_{14-17}	7.98	[5.03, 17.4]	15.7	[9.22, 30.4]	0.51	Wang et al. [2014]
China	2014	no	97.1	[50.3, 310]	113	[49.9, 277]	0.86	Xu et al. [2016]

[1] personal communication

Table S11: Information on the concentrations shown in Figures 6 and 7 in the main text. The column congeners indicates if congener data were available, either from the publication or on request. Ratio stands for the MCCP to SCCP ratio of the medians.

country	year	congeners	MCCPs median	MCCPs 25^{th} and 75^{th} quantile	SCCPs median	SCCPs 25^{th} and 75^{th} quantile	ratio	literature source
Concentrations in surface sediment (background) [ng/g dw]								
Canada	1995	C_{14-17}	68.0	[68.0, 68.0]	288	[288, 288]	0.24	Tomy [1997]
Canada	-	no	14.1	[14.1, 14.1]	158	[158, 158]	0.09	Muir et al. [2000]
Canada	2001	C_{14-16}	290	[290, 290]	290	[290, 290]	1.00	Gewurtz et al. [2007]
Norway	2002	C_{14-15}	106	[54.0, 158]	41.0	[19.0, 63.0]	2.58	Umweltbundesamt [2008]
Norway	2003	no	220	[68.5, 403]	150	[43.0, 440]	1.47	NIVA [2004]
Norway	2006	C_{14-17}	852	[390, 1760]	316	[238, 2208]	2.70	Petersen et al. [2006]
Norway	2013	no	40.0	[10.3, 115]	46.0	[18.5, 84.3]	0.87	NIVA [2014a]
Norway	2014	no	16.7	[5.93, 27.4]	401	[244, 438]	0.04	NIVA [2015b]
Norway	2015	no	1.00	[1.00, 1.00]	38.0	[38.0, 38.0]	0.03	NIVA [2016c]
Norway	2016	no	2.00	[2.00, 2.00]	200	[200, 200]	0.01	NIVA [2017]
Germany	2003-04	C_{14-15}	121	[107, 189]	68.0	[53.4, 78.9]	1.77	Umweltbundesamt [2008]
Czech Rep.	2003/4	C_{14-16}	33.3	[6.26, 193]	2.00	[0.04, 28.0]	16.6	Přibylová et al. [2006]
France	2004	C_{14-15}	68.5	[37.0, 97.8]	32.5	[26.5, 40.4]	2.11	Umweltbundesamt [2008]
North/Baltic Sea	2001-04	C_{14-15}	83.5	[58.0, 149]	46.0	[25.0, 82.0]	1.82	Umweltbundesamt [2008]
Baltic Sea	2001/02	C_{14-15}	119	[114, 211]	142	[116, 262]	0.84	Hüttig and Oehme [2005]
Baltic Sea	2003/04	no	43.0	[33.3, 81.8]	27.0	[16.8, 40.0]	1.59	Hüttig and Oehme [2006]
North Sea	2002/03	no	21.5	[13.0, 30.0]	18.0	[14.0, 27.0]	1.19	Hüttig and Oehme [2005]
S. Africa	2006	no	14.0	[3.90, 26.8]	5.30	[4.25, 29.3]	2.64	Quinn et al. [2009]
China	2010	C_{14-17}	1634	[1296, 2416]	669	[514, 1410]	2.44	Chen et al. [2011]
China	2012/13	some	12.5	[4.70, 112]	12.2	[6.68, 33.3]	1.02	Zeng et al. [2017b]
China	2012/13	some	542	[225, 1816]	401	[68.6, 503]	1.35	Zeng et al. [2017b]
China	2012/13	some	266	[189, 432]	105	[69.1, 126]	2.53	Zeng et al. [2017b]
China	-	C_{14-17}	25.8	[12.6, 66.4]	80.0	[29.9, 260]	0.32	Qiao et al. [2016]
China	-	C_{14-17}	2.85	[0.61, 4.06]	19.2	[9.81, 32.6]	0.15	Qiao et al. [2017]
Japan	2012/13	some	16.6	[6.70, 23.7]	7.95	[7.60, 11.1]	2.08	Zeng et al. [2017b]
Concentrations in surface sediment close to local sources [ng/g dw]								
Arctic	2004	no	10.3	[5.70, 18.1]	13.9	[12.2, 22.6]	0.74	Dick et al. [2010]
Australia	2001	C_{14-17}	2138	[1138, 9756]	192	[68.0, 377]	11.2	Kemmlein et al. [2002]
China	2010	C_{14-17}	2333	[1613, 3764]	808	[651, 1187]	2.89	Chen et al. [2011]
China	2012/13	no	1590	[1215, 4063]	264	[180, 671]	6.02	Zeng et al. [2017b]

Table S12: Information on the concentrations shown in Figures 8 and 9 in the main text. The column congeners indicates if congener data were available, either from the publication or on request. Ratio stands for the MCCP to SCCP ratio of the medians.

country	year	congeners	MCCPs median	MCCPs 25^{th} and 75^{th} quantile	SCCPs median	SCCPs 25^{th} and 75^{th} quantile	ratio	literature source
Concentrations in fish [ng/g lipid]								
Arctic	2001	no	429	[233, 1425]	288	[158, 500]	1.49	Reth et al. [2006]
Canada	2001	C_{14-15}	28.1	[11.6, 65.5]	697	[581, 854]	0.04	Houde et al. [2008]
Canada	2001	C_{14-16}	585	[56.0, 1591]	230	[110, 514]	2.55	Houde et al. [2008]
Canada	2010/11	C_{14-16}	55.7	[28.3, 88.1]	36.9	[21.4, 83.0]	1.51	Saborido B. et al. [2015]
Canada	1979-04	no	131	[57.0, 187]	425	[416, 666]	0.31	Ismail et al. [2009]
N. Atlantic	1993-06	C_{14-15}	57.0	[36.5, 71.5]	135	[113, 196]	0.42	Lahaniatis et al. [2000]
Iceland	2003	no	34.7	[26.0, 65.6]	111	[65.1, 131]	0.31	Reth et al. [2006]
Norway	2002	no	15.4	[5.68, 32.7]	162	[110, 190]	0.10	NIVA [2004]
Norway	1998/03	no	307	[80.8, 465]	434	[283, 928]	0.71	NIVA [2004]
Norway	2004	no	70.7	[14.3, 127]	87.6	[34.7, 141]	0.81	Reth et al. [2006]
Norway	2007/10	no	83.3	[40.4, 148]	24.5	[22.9, 26.1]	3.40	Nyberg et al. [2012]
Norway	2012	no	220	[160, 253]	133	[87.5, 139]	1.65	NIVA [2013]
Norway	2013	no	242	[78.4, 369]	141	[67.5, 191]	1.71	NIVA [2014b]
Norway	-	no	57.6	[29.5, 67.8]	80.2	[60.1, 100]	0.72	NIVA [2014a]
Norway	2014	no	272	[219, 453]	120	[108, 152]	2.27	NIVA [2015a]
Norway	2014	no	36.1	[15.8, 71.1]	96.4	[62.5, 122]	0.37	NIVA [2015b]
Norway	2015	no	951	[424, 1591]	212	[148, 295]	4.49	NIVA [2016a]
Norway	2015	no	6.52	[4.68, 9.11]	27.0	[19.9, 108]	0.24	NIVA [2016c]
Norway	-	no	44.4	[12.8, 142]	108	[94.2, 174]	0.41	NIVA [2016b]
Norway	2016	no	3.60	[3.60, 3.60]	126	[126, 126]	0.03	NIVA [2017]
Baltic Sea	2002/03	no	144	[114, 341]	130	[81.6, 292]	1.11	Umweltbundesamt [2008]
North Sea	2002/03	no	373	[152, 691]	205	[87.2, 500]	1.82	Umweltbundesamt [2008]
China	2012	C_{14-17}	1480	[1257, 2198]	779	[666, 917]	1.90	Zeng et al. [2017a]
China	2014	C_{14-17}	59.3	[30.3; 376]	1179	[660; 2233]	0.05	Huang et al. [2017]
China	-	C_{14-17}	2550	[2500, 2600]	2050	[1800, 2300]	1.24	Du et al. [2018]
Concentrations in birds [ng/g lipid]								
Arctic	2012	no	18.8	[10.4, 65.7]	18.0	[9.93, 30.9]	1.05	Miljodirektoratet [2013]
Norway	2012	no	13.4	[6.09, 17.0]	17.4	[14.5, 32.8]	0.77	Huber et al. [2015]
Norway	2013	no	5.36	[3.44, 10.1]	82.0	[29.3, 267]	0.07	NIVA [2014a]
Norway	2014	no	43.7	[14.1, 90.9]	101	[53.5, 197]	0.43	NIVA [2015b]
Norway	2015	no	74.3	[41.7, 96.1]	182	[95.2, 336]	0.41	NIVA [2016c]
Norway	2015	no	96.4	[60.6, 168]	623	[256, 1125]	0.15	NILU [2016a]
Norway	2016	no	0.71	[0.71, 0.71]	56.6	[56.6, 56.6]	0.01	NIVA [2017]
Norway	2016	no	5.33	[2.12, 25.0]	160	[60.0, 540]	0.03	NILU [2017b]
Norway	2013	no	435	[227, 1663]	158	[99.8, 10930]	2.75	NIVA [2014a]
Norway	2014	no	142	[22.5, 324]	1379	[606, 3289]	0.10	NIVA [2015b]
Norway	2015	no	167	[19.3, 466]	357	[135, 496]	0.47	NIVA [2016c]
Arctic	2001	no	620	[313, 940]	160	[110, 567]	3.88	Reth et al. [2006]
China	-	C_{14-17}	270	[218, 1643]	270	[218, 1043]	1.00	Du et al. [2018]

Table S13: Information on the concentrations shown in Figures 10 and 11 in the main text. The column congeners indicates if congener data were available, either from the publication or on request. Ratio stands for the MCCP to SCCP ratio of the medians.

country	year	congeners	MCCPs median	MCCPs 25^{th} and 75^{th} quantile	SCCPs median	SCCPs 25^{th} and 75^{th} quantile	ratio	literature source
Concentrations in mammals [ng/g lipid]								
Arctic	2010	no	369	[116, 596]	614	[343, 1032]	0.60	Miljodirektoratet [2013]
Arctic	2012	no	156	[74.4, 389]	267	[194, 614]	0.58	Miljodirektoratet [2013]
China	2004-10	C_{14-17}	2150	[1300, 3550]	1700	[965, 2250]	1.26	Zeng et al. [2015]
China	2011-14	C_{14-17}	6700	[4600, 8500]	3600	[2800, 4300]	1.86	Zeng et al. [2015]
China	2004-10	C_{14-17}	3000	[1850, 7500]	1700	[1300, 4525]	1.76	Zeng et al. [2015]
China	2010-14	C_{14-17}	19000	[17000, 23000]	7300	[6500, 9000]	2.60	Zeng et al. [2015]
China	-	C_{14-17}	12000		8900		1.35	Du et al. [2018]
Concentrations in human breast milk, placenta, and human blood [ng/g lipid]								
UK	2001	no	21.0	[12.0, 36.8]	170	[64.8, 283]	0.12	Thomas et al. [2006]
Sweden	1996-10	no	15.0	[3.08, 20.3]	112	[94.3, 123]	0.13	Darnerud et al. [2012]
Germany	-	no	27.5	[7.49, 123]	3.80	[1.62, 12.0]	7.22	Hilger et al. [2011a]
China	2007	C_{14-17}	35.7	[14.5, 78.7]	304	[218, 942]	0.12	Xia et al. [2017a]
China	2011	C_{14-17}	45.4	[16.7, 74.5]	360	[116, 812]	0.13	Xia et al. [2017a]
China	2007	C_{14-17}	60.5	[24.0, 139]	681	[239, 1390]	0.09	Xia et al. [2017b]
China	2011	C_{14-17}	64.4	[41.9, 241]	733	[362, 2081]	0.09	Xia et al. [2017b]
China	2018	C_{14-15}	178	[87,5, 320]	379	[245, 614]	0.47	Wang et al. [2018]
China	2012	no	741	[485, 1167]	3490	[1776, 4780]	0.21	Li et al. [2017]

Table S14: Information on the concentrations shown in Figure S7. The column compensation indicates if the chlorine content differences between standard and sample were compensated according to Reth et al. [2005]. The column congeners indicates if congener data were available, either from the publication or on request.

country	time period	water body	specific location	type of site	instrumental technique	quantification	compensation	congener data	literature source
Concentrations in sediment cores [ng/g dw]									
Switzerland	1961−2004	lake	Lake Thun	rural	GC-ECNI-LRMS	R	yes	yes	Iozza et al. [2008]
Sweden	1881−2015	ocean	Baltic Sea (1)	urban	APCI-QTOF-HRMS	B	n. n.	yes	Yuan et al. [2017]
Sweden	1954−2015	ocean	Baltic Sea (2)	industrial	APCI-QTOF-HRMS	B	n. n.	yes	Yuan et al. [2017]
Sweden	1882−2008	ocean	Baltic Sea (3)	industrial	APCI-QTOF-HRMS	B	n. n.	yes	Yuan et al. [2017]
China	1951−2004	ocean	Hong Kong	not remote	GC-ECNI-LRMS	R	yes	no	Zeng et al. [2017b]
China	approx. 1994−2009	river	Dongjiang River	industrial	GC-ECNI-LRMS	T	no	no	Chen et al. [2011]
Japan	1959−2010	ocean	Tokyo Bay (1)	not remote	GC-ECNI-LRMS	R	yes	no	Zeng et al. [2017b]
Japan	1940−2012	ocean	Tokyo Bay (2)	not remote	GC-ECNI-LRMS	R	yes	no	Zeng et al. [2017b]

- not stated n. n. not necessary [B] quantified according to Bogdal et al. [2015] [R] quantified according to Reth et al. [2005] [T] quantified according to Tomy et al. [1997] and Tomy and Stern [1999]

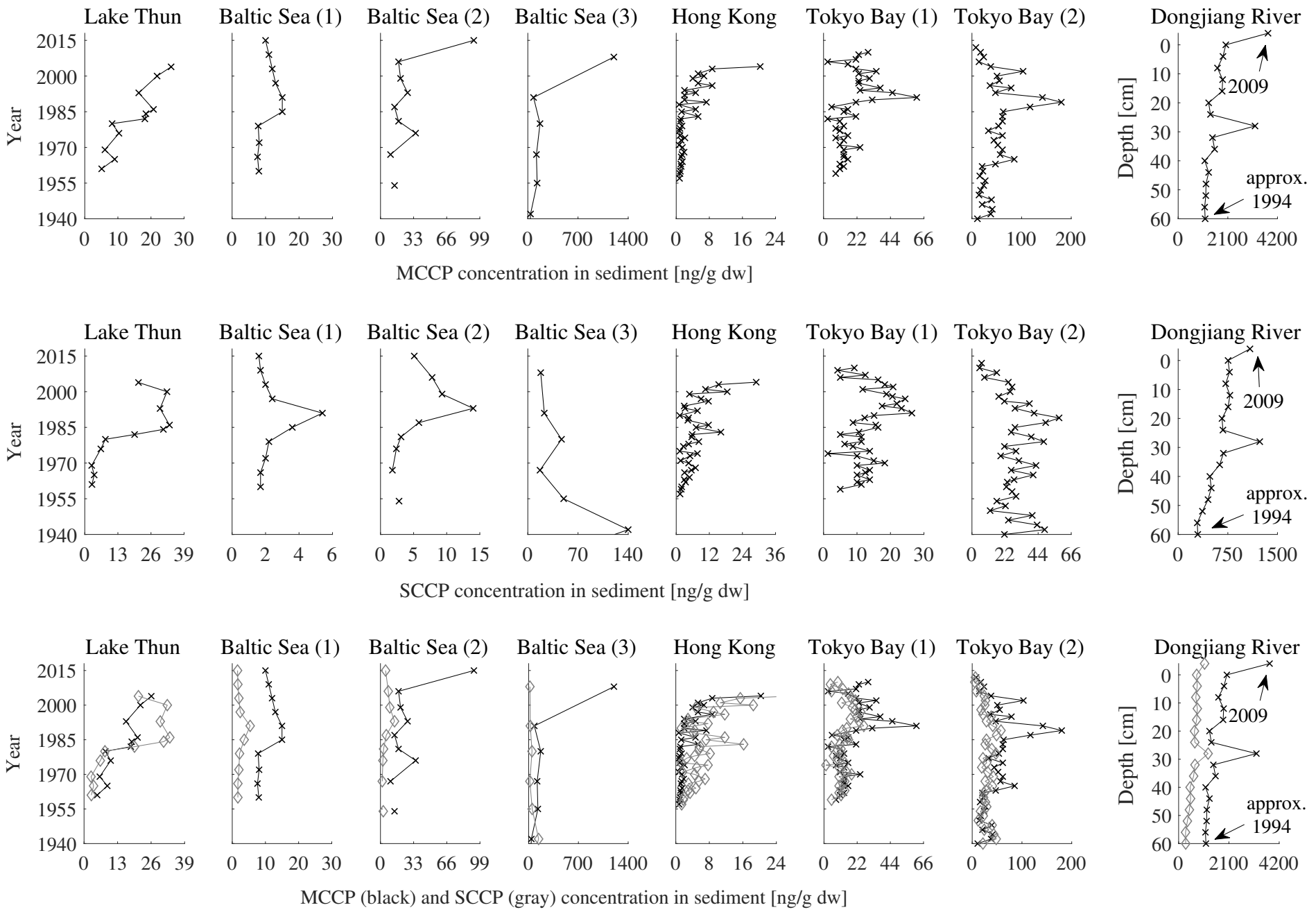

Figure S7: MCCP and SCCP concentrations in sediment cores

S5 Group congener profiles

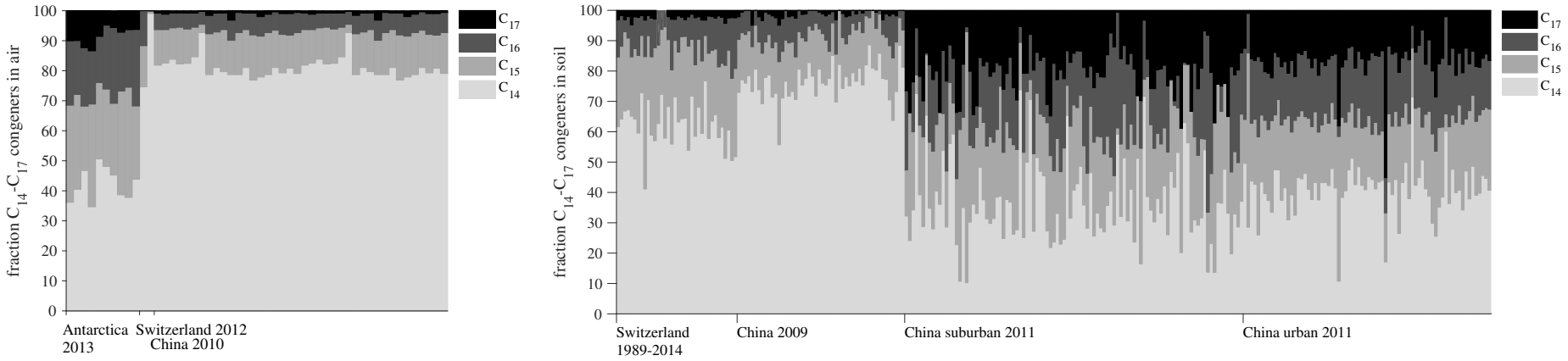

Figure S8: Group congener profiles for MCCPs with C_{14} to C_{17} congeners (1)

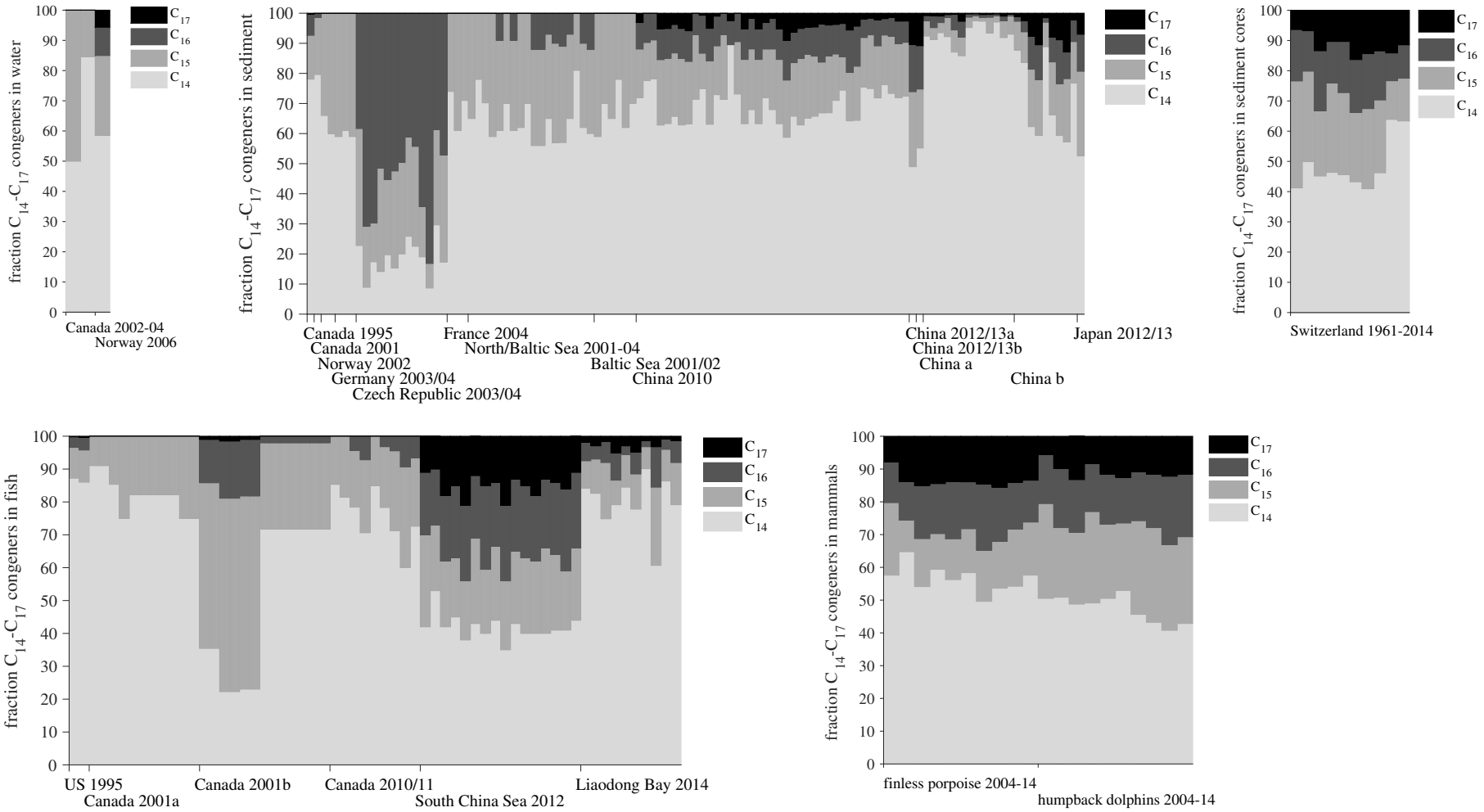

Figure S9: Group congener profiles for MCCPs with C_{14} to C_{17} congeners (2)

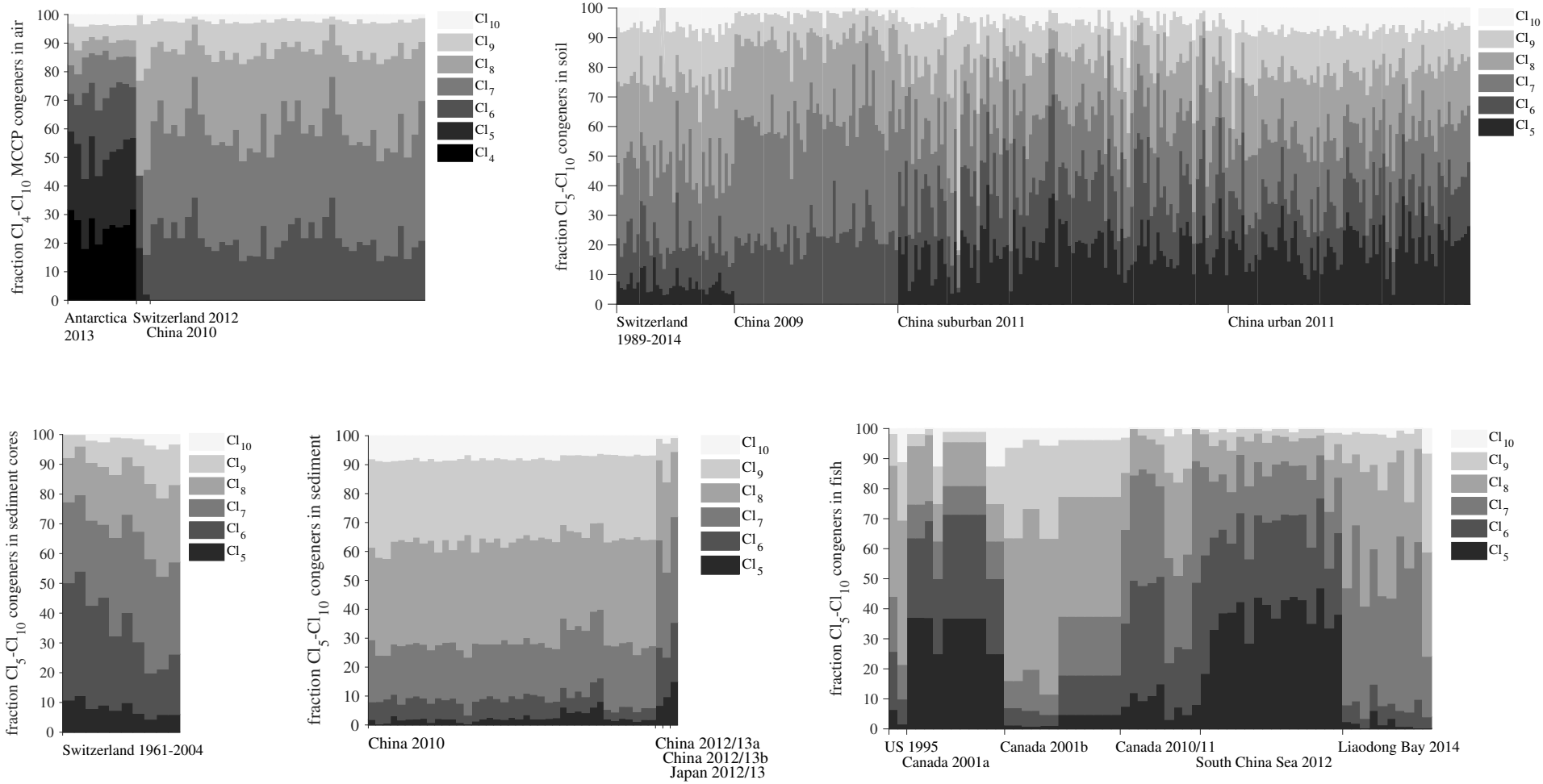

Figure S10: Group congener profiles for MCCPs with Cl_4 to Cl_{10} congeners

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