



Short-, medium-, and long-chain chlorinated paraffins in South African indoor dust and cat hair

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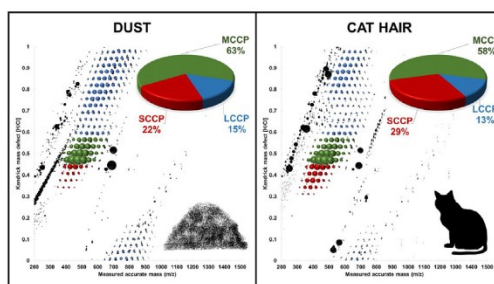
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HIGHLIGHTS

- First report on SCCPs, MCCPs, and LCCPs in the South African indoor environment.
- MCCPs were the dominant CPs followed by SCCP and LCCPs in indoor dust and cat hair.
- SCCPs, MCCPs and LCCPs were found in cat hair from an indoor environment contaminated with CPs.
- CP levels in cat hair were approximately 10-fold lower than in dust for all three CP classes.
- SCCPs with shorter carbon chains and lower chlorine substitution were observed in cat hair.

GRAPHICAL ABSTRACT



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ABSTRACT

Polychlorinated n-alkanes or chlorinated paraffins (CPs) contain a magnitude of structural isomers and are categorized as short-chain (SCCPs), medium-chain (MCCPs), and long-chain (LCCPs) CPs, according to the carbon chain lengths. In this study the Σ SCCPs, Σ MCCPs, and Σ LCCP concentrations are reported for South African indoor dust and pet cat hair. The median concentrations of the Σ CPs (C_9 – C_{37}) ranged from 33 to 663 $\mu\text{g/g}$ for freshly collected dust (FD), 36–488 $\mu\text{g/g}$ for dust collected from household vacuum cleaner bags (VD), and 1.2–15 $\mu\text{g/g}$ for cat hair (CH) samples. MCCPs were the dominant CP group, followed by SCCPs and LCCPs. The Σ MCCP concentration ranged from 13 to 498 $\mu\text{g/g}$ in dust and 0.6–6.5 $\mu\text{g/g}$ in cat hair. SCCPs with shorter carbon chains and lower chlorine substitution were observed in cat hair. LCCPs with carbon chains $> C_{20}$ were detected in dust and hair samples, possibly indicating the use of wax grade LCCP formulations. Non-traditional Kendrick mass defect plots were used to obtain information on the magnitude of CPs and provide evidence of possible interfering compounds. This is the first report on the occurrence of SCCPs, MCCPs, and LCCPs in the South African indoor environment.

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1. Introduction

Chlorinated paraffins (CPs) are complex mixtures of synthetic polychlorinated n-alkanes with various degrees of chlorination. Based on the carbon chain length, CPs are classified as short-chain (SCCPs, $\leq C_{13}$), medium-chain (MCCPs, C_{14} to C_{17}), and long-chain (LCCPs, $\geq C_{18}$). The LCCPs with chain lengths longer than C_{20} are also referred to as wax grade or very long-chain CPs (Mukherjee, 1990; Schinkel et al., 2018). International conventions and regulatory agencies have classified SCCPs as environmentally hazardous substances and restricted their production and use, and in some cases, MCCPs and LCCPs are produced as alternatives (Glüge et al., 2016; UNEP, 2012; van Mourik et al., 2016). There are currently no immediate actions on the production and use of SCCPs in South Africa. According to a report published in 2015, South Africa produced approximately 10,000 tonnes of CPs per year of which 50% is exported (ICIS, 1995). In South Africa, four MCCP formulations are produced with chlorine contents from 45% to 55% (<http://www.ncp.co.za/chlorinated-parrafin-plasticlor>). Concerns regarding the potential toxicity of MCCPs exist for aquatic environments, due to their possible persistence and the temporal concentration increase in soil, biota, and most sediment cores (Glüge et al., 2018). Chlorinated paraffins are mainly used as flame retardants and plasticizer additives in polyvinyl chloride (PVC), lubricating oils, paints, coatings, sealants, and adhesives. They are thus closely associated with the indoor use of these products in vinyl flooring, carpet backing, textiles and fabrics, floor polishes, furniture, wallpaper, and kitchen equipment and appliances (Coelhan and Hilger, 2014; de Boer et al., 2010; Gallistl et al., 2018; Mukherjee, 1990; van Mourik et al., 2016; Wang et al., 2018; Yuan et al., 2017b). There is currently no evidence of natural sources of CPs in the environment and the anthropogenic releases of CPs into the environment are mainly through volatilization, wash-off, and abrasion (de Boer et al., 2010). As a result of their widespread use, levels for CPs have been reported for different environmental compartments including dust, air, water, sewage sludge, soil, sediment, and biota (Coelhan and Hilger, 2014; Tomy et al., 1997; van Mourik et al., 2016). It is proposed that SCCPs are more likely to migrate through the vapour phase due to the dominance of C_{10} and C_{11} carbon chain SCCPs in air, while longer chain CPs and/or higher chlorine contents are likely adsorbed to particles, like dust (Coelhan and Hilger, 2014; Zhou et al., 2018). It was also suggested that CPs might evaporate (at a low evaporation rate) from plastic materials and partition to dust. (Olofsson et al., 2013). There is, however, limited information on LCCPs in indoor dust: recent results showed that LCCPs were the dominant congeners in dust from Sweden (Wong et al., 2017). CP levels are considerably higher than those of other flame retardants in outdoor and indoor environments (de Boer et al., 2016). Dust is one of the most frequently studied matrices in the indoor environment and inhalation (by adults) and dust ingestion (by toddlers) are suggested to be significant routes for human exposure to CPs (Fridén et al., 2011). Hair has been presented as an indicator for the detection of atmospheric exposure, and possible estimation of corresponding air concentrations (Schramm, 2008). Pet cats share a common environment with toddlers and cat hair is also directly exposed to the environment and may accumulate contaminants from indoor air and dust. Cats have been selected as good sentinels for human indoor exposure to flame retardants (Dye et al., 2007; Norrgran Engdahl et al., 2017). Being a non-invasive matrix, hair might, therefore, have relevance as indoor exposure model for children.

Aspects of the analysis of CPs in environmental matrices are discussed in the review from van Mourik et al. (2015). One of the major advances in CP analysis by Zencak and Oehme (2004) introduced chlorine adduct formation using negative ion

atmospheric pressure chemical ionization (APCI) hyphenated with liquid chromatography. Bogdal et al. (2015) employed a direct flow injection method using this unique ionization technique coupled to a high-resolution time-of-flight mass spectrometer (HR-TOF-MS). CPs were quantified by reconstructing the CP patterns in the samples into a linear combination of CP patterns from technical formulations using a mathematical deconvolution algorithm. In addition to the improved selectivity and sensitivity, the accurate mass information obtained from the HR-TOF-MS analysis can also be used to comprehensively explore data from complex samples by constructing mass defect (MD) plots. Kendrick (1963) introduced a smart approach to identify organic ions belonging to a homologous series. The use of non-traditional Kendrick MD plots corresponding to the substitution of chlorine for hydrogen (H/Cl) was previously used to facilitate the identification of halogenated contaminants in environmental samples (Cariou et al., 2016; Jobst et al., 2013; Taguchi et al., 2010).

In this study, concentrations for the Σ SCCP, Σ MCCP, and Σ LCCP in South African indoor dust and cat hair (CH) were determined using APCI coupled to a high-resolution quadrupole time-of-flight mass spectrometry (qTOF-MS) with chlorine-induced adduct formation and the accurate mass full scan data were evaluated using non-traditional Kendrick MD plots. Two indoor dust types were collected from houses as fresh or "active" dust (FD) and dust taken from household vacuum cleaner bags (VD). The FD represents samples from a single room over a short time-span and VD contamination over periods of months from multiple rooms. In addition to the dust, hair samples were obtained for some of the indoor pet cats, which share a common environment with toddlers.

2. Materials and methods

Information about the standards, chemicals, and suppliers is provided in the Supporting Information (SI) of this manuscript.

2.1. Sample collection

Dust samples ($n = 20$) were collected from homes in Pretoria, South Africa. Details on the sample collection are provided in Brits et al. (2019) and samples associated with each home are provided in the supporting information (Table S1). Briefly, FD ($n = 9$) was collected from the living room, as a mixture of floor dust and elevated surface dust, and VD ($n = 11$) was collected from household vacuum cleaner bags or canisters (for vacuum cleaners without bags). During sample collection, sodium sulphate (Na_2SO_4) spread on aluminium foil, was collected using the same method employed to collect dust samples as field blanks ($n = 6$). The samples were wrapped in aluminium foil, placed in plastic resealable bags and transported to the laboratory for processing. Dust and field blank samples were sieved (stainless steel sieve, $500 \mu\text{m}$) and stored in amber vials at room temperature until chemical analysis. The CH samples ($n = 10$) were collected from Persian cats at a pet grooming service, representing six homes. Glass wool was exposed to the air during hair sampling to simulate field blanks ($n = 3$). The samples were wrapped in aluminium foil, placed in resealable plastic bags, and stored at room temperature until chemical analysis.

2.2. Sample pre-treatment

Dust ($\sim 50 \text{ mg}$) and cat hair ($\sim 500 \text{ mg}$) was extracted using accelerated solvent extraction (ASE) with hexane/acetone (3:1, v/v) as previously described by Brits et al. (2019) Three blanks and three SRM 2585 samples were analysed together with each batch of nine to eleven samples. The cat hair samples were subjected to a

freezing-lipid precipitation step with methanol to reduce the presence of polar wax-like compounds prior to fractionation as previously employed for complex lipid-rich samples (Liu et al., 2018). The extracts were fractionated on pre-cleaned glass columns fitted with a glass wool plug and filled from the bottom with 0.5 g silica gel, 0.5 g florisil and 0.5 g anhydrous Na₂SO₄. The column was conditioned with 40 mL hexane. The extracts, solvent exchanged to hexane, were quantitatively transferred to the glass column. The CPs were eluted from the column with 15 mL hexane and 15 mL dichloromethane/hexane (1:1, v/v). The combined eluents were evaporated to near dryness at 30 °C under low nitrogen flow and reconstituted to 0.5 mL with acetonitrile, followed by the addition of ¹³C₁₀-anti-Dechlorane Plus® as injection standard.

2.3. Instrumental analysis

The CP analysis was based on the analytical method previously described by Brandsma et al. (2019). Flow injection mass spectrometry was performed into an APCI source coupled to a qTOF-MS (Compact, Bruker, Bremen, Germany). The sample extracts (10 µL) were injected using an Agilent 1290 infinity HPLC system (Agilent Technologies, Amstelveen, The Netherlands) with a mixture of acetonitrile/dichloromethane (10:1, v/v) as mobile phase at a flow rate of 250 µL/min. Detailed settings of the qTOF-MS are given in the Supporting Information. An average full scan mass spectrum of the peak in the chromatogram was background subtracted and accurate masses, with a signal-to-noise of more than 3 at an absolute intensity threshold of 100 counts, were extracted using Bruker Compass DataAnalysis software. The accurate mass list was exported to Microsoft Excel software for quantitation.

2.4. Quantitation and deconvolution

Strict criteria were set to assign peak areas to each CP congener group to confirm positive identification and accurate quantitation of the CP congeners. The two most abundant [M+Cl]⁻ ion pairs were used as quantitation and qualifier ions (Table S2). Peak areas of the congeners were only included when, (1) the unique [M+Cl]⁻ ions were present at a signal-to-noise higher than 10 with a mass accuracy of ≤5 ppm from the theoretical mass, (2) the ion pair intensity ratio was within 10% of the theoretical ratios. In total 1278 target ions for CPs ranging from C₉H₁₇Cl₃ to C₄₀H₅₆Cl₂₆ were considered. The quantitation of the CPs was performed using the deconvolution algorithm previously applied by Bogdal et al. (2015). Detailed information on the deconvolution procedure for SCCPs, MCCPs, and LCCPs can be found in the supporting information of Brandsma et al. (2017). The goodness-of-fit was calculated between measured patterns and deconvoluted patterns and expressed as the coefficient of determination (R²). CP concentrations for samples with an R² < 0.5 should be taken as indicative values (Bogdal et al., 2015; Brandsma et al., 2017). The degree of chlorination calculated for the seven standard technical CP formulations compared well with the stated chlorination degree of the manufacturer (Table S3).

2.5. Quality assurance and quality control

Field blanks for dust and hair samples were included in the analysis of each batch of nine to eleven samples. No LCCPs were detected in the eight field blanks. The ΣSCCP and ΣMCCPs in the blanks were 10 ng and 43 ng absolute, which amount to 4% of the lowest concentration found in the samples, and hence no background subtraction was performed. The limits of quantification (LOQ) were calculated as the mean values plus ten times the standard deviation of the congeners in the blanks. Based on an average sample intake of 0.05 g dust and 0.5 g hair, the LOQs for

ΣSCCPs were 60 ng/g and 6.0 ng/g and the LOQs for ΣMCCPs were 387 ng/g and 39 ng/g for the respective matrices. As no LCCPs were detected in the blanks, the LOQ was set at the concentration of the lowest calibration standard, which was 198 ng/g for dust and 20 ng/g for the hair samples. Linearity was assessed using five-point (10–200 ng) calibration curves for each CP formulation and the correlation coefficients (R²) for the formulations were better than 0.998. The CP recoveries for both matrices were assessed through triplicate spiking experiments. Dust and hair samples were spiked with a mixture containing 500 ng of SCCP (SCCP 55.5% Cl), MCCP (MCCP 42% Cl), and LCCP (LCCP 36% Cl) standards and underwent the same sample pre-treatment and analysis procedures as the samples. The relative recoveries in the dust were 99 ± 11% for ΣSCCPs, 107 ± 12% for ΣMCCPs, and 107 ± 14% for ΣLCCPs. The recoveries from the cat hair were 90 ± 13% for ΣSCCPs, 94 ± 10% for ΣMCCPs and 92 ± 11% for ΣLCCPs. The R² for the ΣSCCPs and ΣMCCPs were higher than 0.51 for all samples (Table S5). The deconvolution method applied in this study resulted in low R² values for the ΣLCCPs, due to the lack of standard technical mixtures containing carbon chain lengths > C₂₀. Limitations regarding analytical standards were previously discussed by Schinkel et al. (2018).

3. Results and discussion

3.1. CP concentrations in house dust SRM 2585

There is currently no reference material for CPs in dust or hair. The concentrations of ΣSCCPs, ΣMCCPs, and ΣLCCPs for the NIST SRM 2585 material in this study were 8.65 ± 0.16 µg/g (R² = 0.82), 11.98 ± 0.24 µg/g (R² = 0.73), and 19.26 ± 0.56 µg/g (R² = 0.02), respectively (additional information on the method validation and CP composition in the NIST SRM 2585 material are provided in Table S4 and Fig. S1). The results obtained from our study were lower than the first report on concentrations of CPs in the NIST SRM 2585 (n = 2), which were 102 and 119 µg/g for ΣSCCPs, 116 and 121 µg/g for ΣMCCPs, and 87 and 112 µg/g for ΣLCCPs (Wong et al., 2017). Shang et al. (2019) reported concentrations of 7.58 ± 0.43 µg/g for ΣSCCPs and 16.4 ± 2.1 µg/g for ΣMCCPs for the SRM 2585. Brandsma et al. (2019) recently reported average levels of 7.1 ± 0.2 µg/g for the ΣSCCPs, 10 ± 0.2 µg/g for ΣMCCPs and 16 ± 0.4 µg/g for ΣLCCP. The concentrations reported in our study compare well with these two studies, taking into account that the between-laboratory coefficient of variation reported for the same sample in a recent inter-laboratory comparison was 68% (van Mourik et al., 2018). The congener profile of the ΣSCCP was dominated by carbon chain lengths of C₁₂ (followed by C₁₁ and C₁₃) and chlorine substitution of Cl₆ and Cl₇ (followed by approximately equal contributions of Cl₅ and Cl₈) similar to the profile previously reported by Wong et al. (2017). The ΣMCCPs were dominated by equal contributions of C₁₄ and C₁₅ and chlorine substitution of Cl₆ followed by Cl₇ and Cl₅. The ΣLCCP profile showed a higher prevalence of C₂₂ to C₂₅ carbon chain lengths and chlorine substitution of Cl₆ and Cl₇. The mean chlorine contents in ΣSCCPs, ΣMCCPs, and ΣLCCPs were 59%, 52%, and 40% respectively. This was similar to the degree of chlorination previously reported (59% for ΣSCCPs, 52% for ΣMCCPs, and 43% for ΣLCCPs) (Brandsma et al., 2019).

3.2. CP concentrations in indoor dust and cat hair

SCCPs, MCCPs, and LCCPs were detected in all samples analysed and large variations were observed for CP congener profiles between matrices and for the individual samples (see Table S5 and Figs. S13–S43). The ΣCP concentration in the FD ranged from 33 to 663 µg/g, with a median concentration of 94 µg/g and the

concentration in the VD ranged from 36 to 488 $\mu\text{g/g}$, with a median concentration of 68 $\mu\text{g/g}$ (Fig. 1). The ΣMCCPs were the dominant CP group contributing between 58% and 64% to the CP concentration, followed by ΣSCCPs (20–29%) and ΣLCCPs (13–17%). The median concentrations of the ΣSCCPs were 17 $\mu\text{g/g}$ in the FD and 14 $\mu\text{g/g}$ in the VD, with concentration ranging from 5.4 to 353 $\mu\text{g/g}$ and 5.1–214 $\mu\text{g/g}$ for the respective dust matrices (Fig. 1). The ΣSCCP concentrations in the dust samples were higher than previously reported in house dust from Australia (13 $\mu\text{g/g}$) (He et al., 2019), Canada (6.2 $\mu\text{g/g}$) (Shang et al., 2019), Germany (5 $\mu\text{g/g}$) (Hilger et al., 2013) and Sweden (5–9 $\mu\text{g/g}$) (Wong et al., 2017) and lower than reported for Australia (Wong et al., 2017), China (Chen et al., 2018; Liu et al., 2017; Shi et al., 2017; Wong et al., 2017), and the UK (Wong et al., 2017) (Table S6). The median concentrations of the ΣMCCPs were 47 $\mu\text{g/g}$ in the FD and 46 $\mu\text{g/g}$ in the VD with concentrations ranging from 21 to 498 $\mu\text{g/g}$ and 13–200 $\mu\text{g/g}$ respectively (Fig. 1). The ΣMCCP concentration in the South African dust was higher than reported for Canada (19 $\mu\text{g/g}$) (Shang et al., 2019) but lower than reported for China (Chen et al., 2018; Shi et al., 2017; Wong et al., 2017) and Germany (Hilger et al., 2013) (Table S6). The median ΣMCCP concentration was similar to results reported for indoor dust from Australia (He et al., 2019).

The median concentration of the ΣLCCPs in the VD was 13 $\mu\text{g/g}$, which ranged from 4.3 to 74 $\mu\text{g/g}$, and 9.4 $\mu\text{g/g}$ in the FD, ranging

from 1.9 to 108 $\mu\text{g/g}$ (Fig. 1). The ΣLCCP concentrations from indoor dust samples were previously reported in house dust from five countries (Wong et al., 2017). The concentrations of the ΣLCCP ranged from 92 to 1995 $\mu\text{g/g}$, which were higher than the levels found in this study (Table S6). The ΣLCCPs were higher than recently reported for house dust from Australia (He et al., 2019).

The median concentrations of the ΣCPs in the cat hair were 7.0 $\mu\text{g/g}$, with concentrations ranging from 1.2 to 15 $\mu\text{g/g}$. There is, however, no other literature on the occurrence of CPs in human or pet cat hair. The median concentration of ΣSCCPs , ΣMCCPs , and ΣLCCPs in the CH samples were 2.0 $\mu\text{g/g}$, 4.0 $\mu\text{g/g}$, and 0.9 $\mu\text{g/g}$, respectively. So, SCCPs, MCCPs and LCCPs were found in house dust and cat hair from an indoor environment contaminated with CPs. Considering that the samples sizes limit meaningful statistical analysis, the levels in cat hair were found to be roughly 10-fold lower than in dust for all three CP classes.

3.3. CP congener group profiles

Non-traditional Kendrick MD plots were used to assess the carbon chain lengths and degree of substitution of SCCPs, MCCPs, and LCCPs in the dust and hair samples. As shown in Fig. 2, the chlorine adduct formation using negative ion APCI improved the selectivity and sensitivity for halogen-containing compounds. The

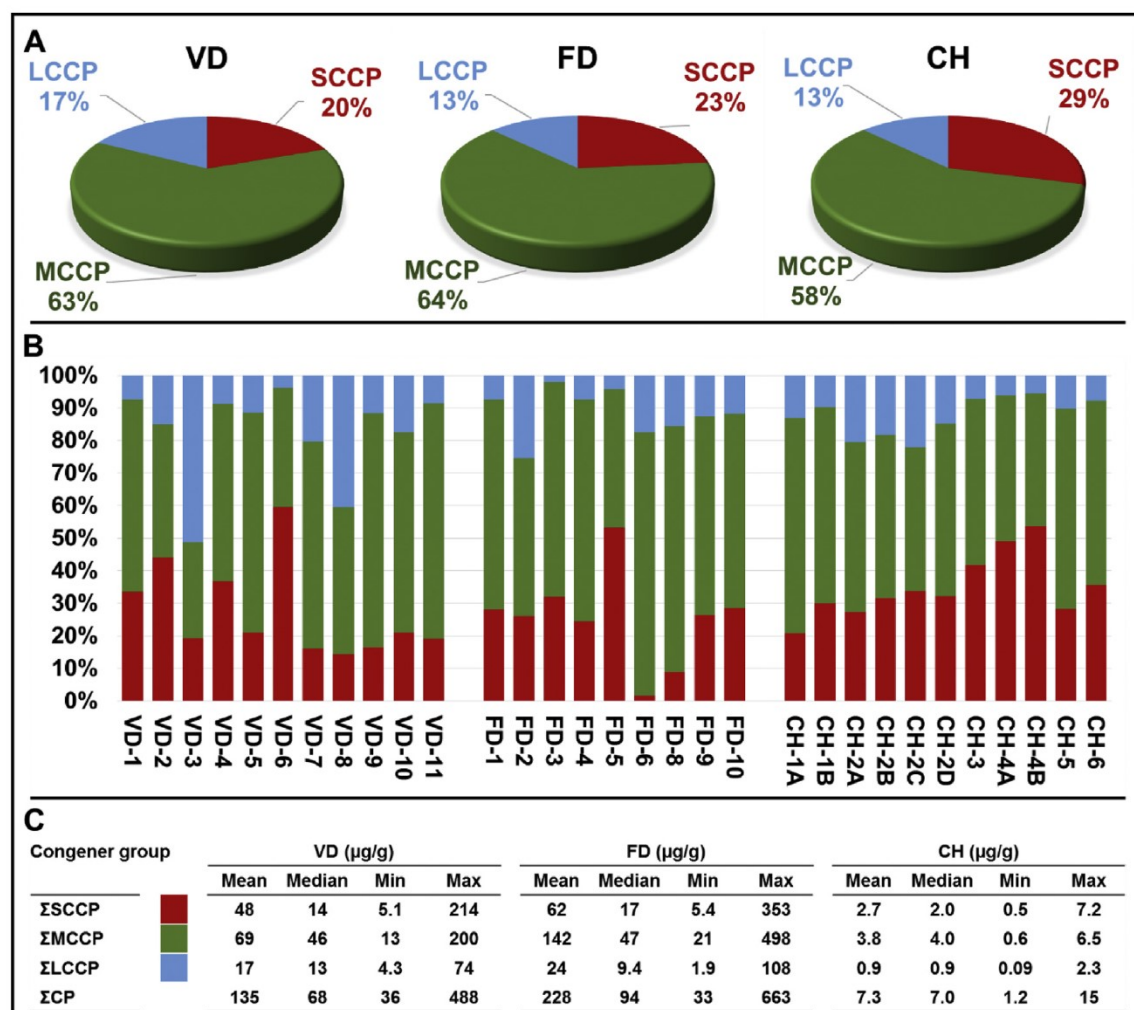


Fig. 1. CP congener profiles of (A) the median ΣSCCPs , ΣMCCPs and ΣLCCPs in VD, FD and CH, (B) the relative abundances (%) in the individual VD, FD and CH samples. (C) Summary of the mean, median, and concentration range ($\mu\text{g/g}$).

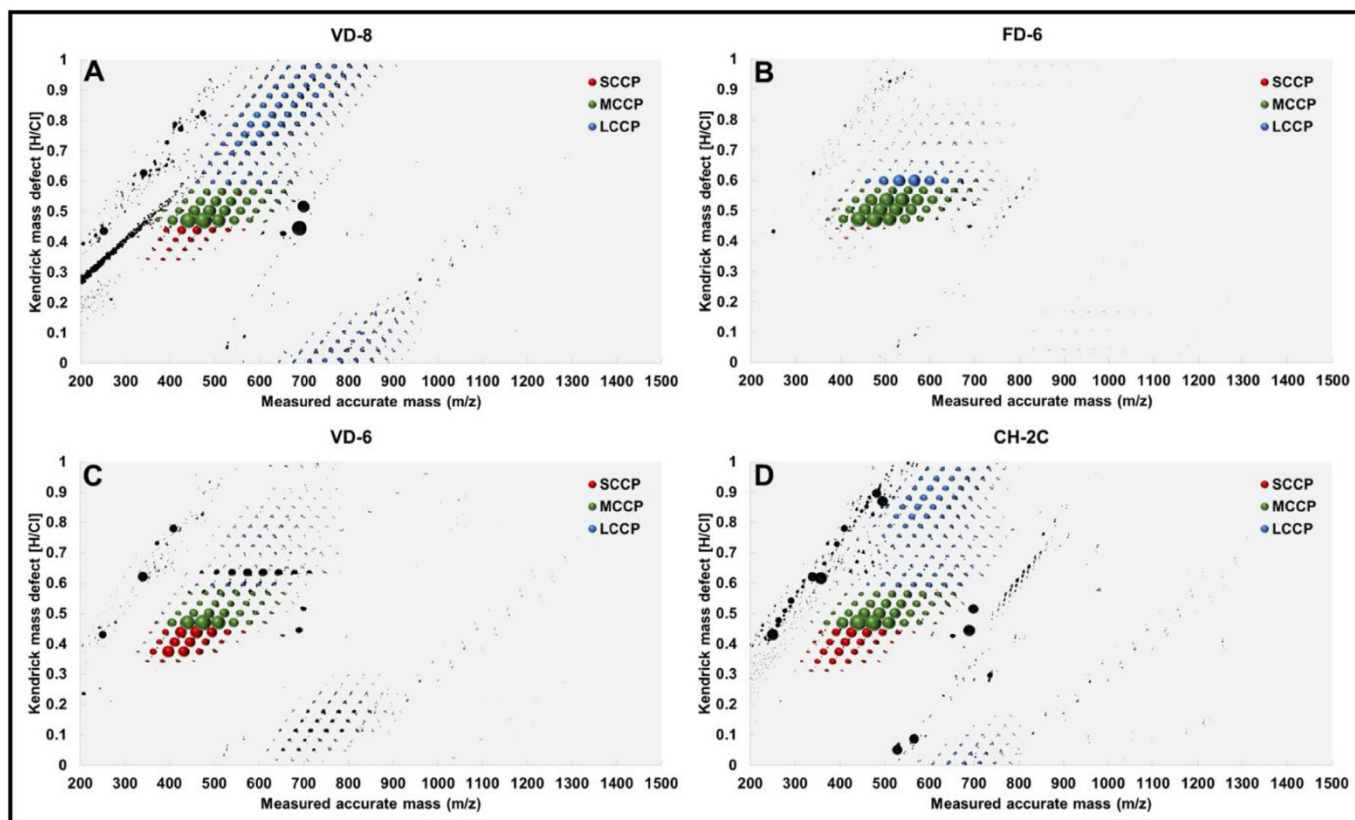


Fig. 2. Non-traditional Kendrick MD plots constructed for (A) sample VD-8, (B) sample FD-6, (C) sample VD-6, (D) sample CH-2C. The red spheres represent the quantifier and qualifier $[M+Cl]$ ions for SCCPs, the green for MCCPs and the blue for LCCPs, the sphere sizes represent the instrument response. (See supporting information for details). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

non-traditional Kendrick MD plots could constructively be used to provide an informative picture of the CP contamination present in the dust and hair samples. A detailed discussion on the construction of these plots is provided in the supporting information (Fig. S2). As shown in Fig. 2A, the carbon chain lengths for the CP congeners in the dust samples went up to C_{37} . The CP congeners in the CH samples (Fig. 2D), ranged from C_9 to C_{36} . Screening studies have previously reported the identification of LCCPs up to C_{35} in European eel samples (Cariou et al., 2016). In addition to obtaining information on the magnitude of CPs, which aids in the selection of the target list for quantitation, the MD plots also provide evidence on possible interferences. As shown in Fig. 2C (and Fig. S2), the pattern observed for VD-6 shows abundant C_{19} and prominent congeners $\geq C_{30}$. The difference between measured and theoretical masses for the C_{19} homologue group were within 10 ppm of the theoretical masses. The ratios of the quantitation and qualifier ions do however vary by more than 40% from the theoretical ion pair intensity ratios (examples of mass scale-expanded segments for mass spectra are provided in Fig. S3). This emphasises the use of very strict mass accuracy criteria and the use of ion ratios to distinguish between positive identified CPs and the presence of possible interferences. This pattern was also observed for the cat hair (CH-6) taken from the same household.

As shown in Fig. 3, comparable patterns were observed for the dust, with carbon chain lengths of C_{13} dominating the Σ SCCP profile. The chlorine substitution for the Σ SCCPs ranged from Cl_4 to Cl_{10} and was dominated by Cl_6 and Cl_7 followed by approximately equal contributions of Cl_5 and Cl_8 . Although C_{13} carbon chain lengths were also dominant in cat hair, C_9 to C_{11} congeners with lower

chlorine substitution (Cl_4 and Cl_5) were observed. Zhou et al. (2018) previously showed that longer-chain and higher chlorinated SCCP congeners adsorb to small particles and shorter-chain and lower chlorinated CP congeners can migrate from small particles to larger particles. This could imply that these shorter-chain and lower chlorinated CP congeners could accumulate through volatilization and condensation processes on the large surface area of hair from the small dust particles. In addition, lower molecular weight SCCP congeners are more prone to volatilization (Wang et al., 2013; Wang et al., 2015). The volatile shorter chain SCCPs with fewer chlorine atoms were previously reported to be dominant in indoor air (Fridén et al., 2011; Wang et al., 2013) and it was reported that these congener groups can be taken up by the protective epicuticular wax layer of pine needles through kinetically limited gaseous deposition (Wang et al., 2015). Although gaseous exchange and atmospheric particle deposition play a role in CP transport, generally, higher levels are found in dust compared to air. This might suggest that SCCPs with shorter carbon chains and lower chlorine substitution could be adsorbed to the waxy sebum layer on cat hair over time, possibly due to contributions from indoor dust or direct migration from possible source in the indoor environment. Toxicokinetic studies further showed that distributions of SCCPs congener groups in blood, urine, and faeces from Sprague Dawley rats depended on octanol/water partition coefficients (K_{ow}) (Geng et al., 2016). Lower chlorinated Cl_5 -SCCPs were associated with blood and urine, and Cl_{8-10} -SCCPs were eliminated via the faeces. Although no data are available for the unmetabolised CPs partitioning between blood and hair we cannot exclude that internal exposure may have a substantial contribution. The current

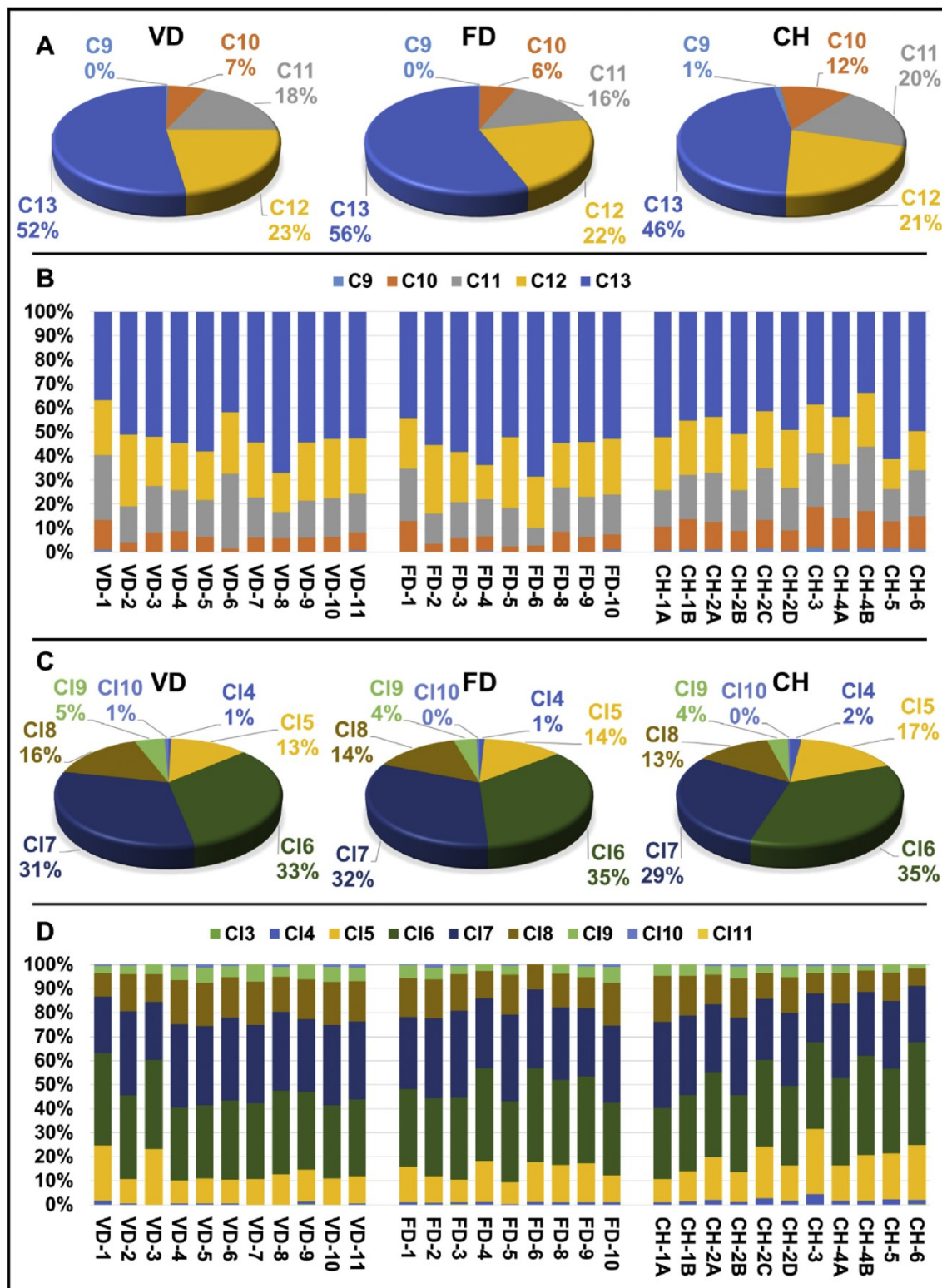


Fig. 3. Congener profiles of the median Σ SCCPs in the dust and hair samples where (A), is the composition based on carbon chain length, (B) is the carbon chain length in the individual samples, (C) is the composition based on chlorine substitution, (D) is the chlorine substitution in the individual samples.

analytical method did not allow to distinguish between internal/absorbed levels and external/adsorbed levels on the cat hair. Theoretically, internal blood levels of CPs could be transported to the hair follicles and further stored in the hair structure. The

influences of atmospheric deposition, direct migration from sources, or endogenous mechanisms on the hair concentrations are not fully understood, and factors may vary for different CP congeners. Further research is needed to investigate the migration and

leaching behaviour of CPs from products to evaluate the potential human exposure to CPs. The carbon chain length profiles for dust were similar to previously reported for SCCPs in dust samples from Canada (Shang et al., 2019; Wong et al., 2017), China (Chen et al., 2018; Wong et al., 2017), and Australia (He et al., 2019; Wong et al., 2017). The chlorine distribution calculated in this study does, however, differ from studies in house dust from Canada (Shang et al., 2019), China (Chen et al., 2018; Liu et al., 2017; Shi et al., 2017), and Germany (Hilger et al., 2013), where Cl₇ and Cl₈ dominated the ΣSCCP profile. This is probably an indication that different commercial CP mixtures are used in these countries. The degree of chlorination of the ΣSCCPs ranged from 56% to 59% in all samples analysed, similar to results reported for house dust from Canada (ranging from 53.8% to 57.2%) (Shang et al., 2019) and lower than results for Germany (60% and 62.5%) (Hilger et al., 2013).

As shown in Fig. 4, similar patterns for the ΣMCCPs carbon chain length profiles were observed for all samples, with C₁₄ dominating the congener profile followed by C₁₅, C₁₆, and C₁₇. The MCCP chlorine homologue groups ranged from Cl₄ to Cl₁₁ and were dominated by Cl₇ followed by nearly equal contributions for Cl₆ and Cl₈. Similar carbon chain length profiles were previously reported for dust samples from Canada (Shang et al., 2019; Wong et al., 2017), China (Chen et al., 2018; Shi et al., 2017; Wong et al., 2017), and Australia (He et al., 2019; Wong et al., 2017). The profile from Sweden was dominated by C₁₇ carbon chain lengths (Wong et al., 2017). The chlorine distribution calculated in this study differs from studies from Canada where Cl₈ was the dominant group (Shang et al., 2019), Cl₇ and Cl₈ were the dominant contributors in China (Chen et al., 2018; Shi et al., 2017), and Cl₆ was dominant for Germany (Hilger et al., 2013). The degree of chlorination for the ΣMCCPs ranged from 54 to 56% in all the samples analysed, similar to results reported for house dust samples from Canada (ranging from 53.8 to 57.2%) (Shang et al., 2019) and higher than results for Germany (44.5 and 49.5%) (Hilger et al., 2013).

The ΣLCCP congener profiles differ between the sample types and large variation could be observed for the individual samples (see Fig. 5). The C₁₈ congeners dominate the profiles followed by C₁₉ and C₂₀. The chlorine homologue groups for LCCPs in the dust samples were dominated by Cl₈ and Cl₇ followed Cl₉. The profile in CH was dominated by equal contributions from Cl₇ and Cl₈, followed by Cl₆ and Cl₉. As shown in the Kendrick MD plot (Figs. 2B), and Fig. 1, the CP profile for FD-6 consists mostly of MCCPs and C₁₈-LCCPs. The ΣLCCP congener profile for this sample (Fig. S29) shows a unique profile which was dominated by C₁₈ carbon chain lengths; no C₂₁ was observed and from C₂₆ to C₃₆, only even carbon chain lengths were observed. This CP profile in this dust sample could be influenced by the presence of small particles from products which contain a CP formulation of which the LCCPs could be possible impurities. The analysis of household consumer products could provide more information on the type of CP formulations present in these products. The LCCP congener profile in sample FD-2 and the cat hair samples (CH-2A to 2D) taken from the same house were dominated by C₂₅ carbon chain lengths which might confirm the use of wax grade LCCP formulations. This profile was also observed for two of the VD samples (VD-3 and VD-8). We also observed low and high chlorination for the LCCP carbon chain lengths C₂₄ to C₂₇ in sample VD-2 and to a lesser extent in FD-4 (Figs. S14 and S27), this "double" chlorination pattern could indicate that combinations of CP formulations, present in products, might contribute to this congener profile. The ΣLCCPs with carbon chain lengths longer than C₂₁ contributed more to the ΣLCCP concentration in VD as compared with the FD and CH samples. This variation between the dust types might be due to various factors that influence the partitioning of CPs from products to suspended particles and deposition on dust. As the VD samples represent dust from multiple

rooms, it might include particles from products which contain CPs such as flexible PVC or polyurethane insulation foams.

3.4. Implications

The occurrence of CPs in the South African indoor environments indicates that humans and pets are exposed to these compounds. To date, there is limited data on CPs, especially LCCPs in the indoor environment, and more research is needed to understand indoor exposures in order to estimate the risk and fate of CPs. Dust ingestion was suggested to be a significant CP exposure route for toddlers (Fridén et al., 2011; Gao et al., 2018; Shi et al., 2017). Toddlers spend more time indoors and are usually in close proximity to indoor dust; their crawling activity and frequent hand-mouth contact enhances the direct contact and exposure to dust and subsequently contaminants found in dust. While this pilot study may somewhat limit conclusions, the results represent an important baseline for developing larger studies to assess CP exposure estimates. Health-based reference values for CP congeners are lacking and it is unclear if the positions of chlorine substitution and the degree of chlorination would have a substantial impact on health effects. Indoor environments such as child care centers and schools have to be taken into consideration when exposure to CPs are characterized for children.

SCCPs have been listed as persistent organic pollutants (POPs) under the Stockholm Convention of the United Nations. However, for this class of chemicals an unusual long list of exemptions was made. They may still be used as adhesives in the rubber industry; rubber products in industries related to mining and forestry, waterproofing paints, adhesives, lubricant additives, secondary plasticizers in flexible PVC products and flame retardant in paints and plastics (UNEP, 2012). The effectiveness of the 'POP label' for SCCPs can therefore be questioned. Given the very high global production volumes of CPs and the recently published UN Global Chemicals Outlook II (ICIS, 1995), which predicts a doubling of the total amount of chemicals used worldwide within the coming decade, it seems imperative for the UN to reduce the current restrictions on the SCCPs in the Stockholm Convention and in addition consider MCCPs for a POP label. ΣSCCP, ΣMCCP, and ΣLCCP levels have recently been reported in sewage sludge (Brandtsma et al., 2017), sediment (Yuan et al., 2017a), human plasma (Li et al., 2017), and terrestrial animals (Du et al., 2018; Yuan et al., 2019). These studies indicate that not only SCCPs but also MCCPs and LCCPs have a bioaccumulative potential. With the current regulatory situation in South Africa, adequate environmental monitoring data are available for some of the POP pesticides. CPs have, however, not received attention and only limited data is available. It was previously reported that background soil concentrations for ΣMCCPs in South Africa (Quinn et al., 2009), were as high as soil collected from sites close to a CP production plant in China (Glüge et al., 2018). The congener profiles reported in this study show that the ΣMCCPs are the major CP group in the indoor environment. Congener profiles of local commercial formulations are needed to confirm if the MCCPs found, could be from a locally produced technical MCCP mixture. However, the contribution from the ΣSCCPs and ΣLCCPs may be due to formulations used in imported products. International restrictions of SCCPs may result in an increased use of MCCPs and LCCPs, which underlines the importance of including and monitoring of these two groups to support the enforcement of regulations.

4. Conclusions

We have reported for the first time the occurrence of SCCPs, MCCPs, and LCCPs in the South African indoor environment. The

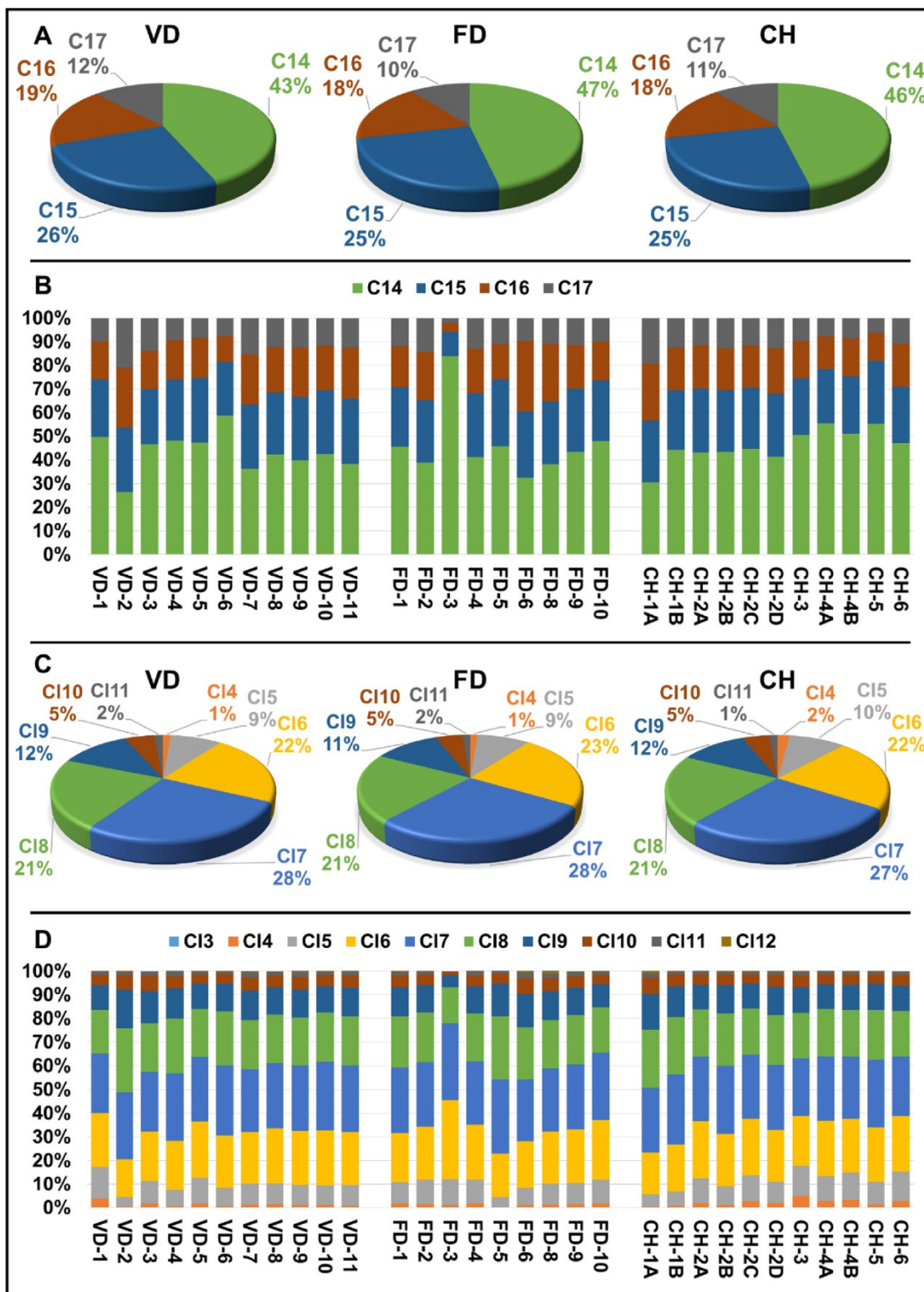


Fig. 4. Congener profiles of the median Σ MCCPs in the dust and hair samples where (A), is the composition based on carbon chain length, (B) is the carbon chain length in the individual samples, (C) is the composition based on chlorine substitution, (D) is the chlorine substitution in the individual samples.

results of the study emphasise the need for strict identification criteria to distinguish between positively identified CPs and possible interferences. The Σ MCCPs were the major CP group in the samples followed by Σ SCCPs and Σ LCCPs. The congener profiles for the Σ LCCP confirms the presence of wax grade LCCP formulations.

C₉ to C₁₁ SCCPs with lower chlorine substitution (Cl₄ and Cl₅) were observed in the cat hair samples, suggesting that SCCPs with shorter carbon chains and lower chlorine substitution could be favourably adsorbed to the hair. This might indicate that indoor dust partly contributes to the pattern observed in the hair. The

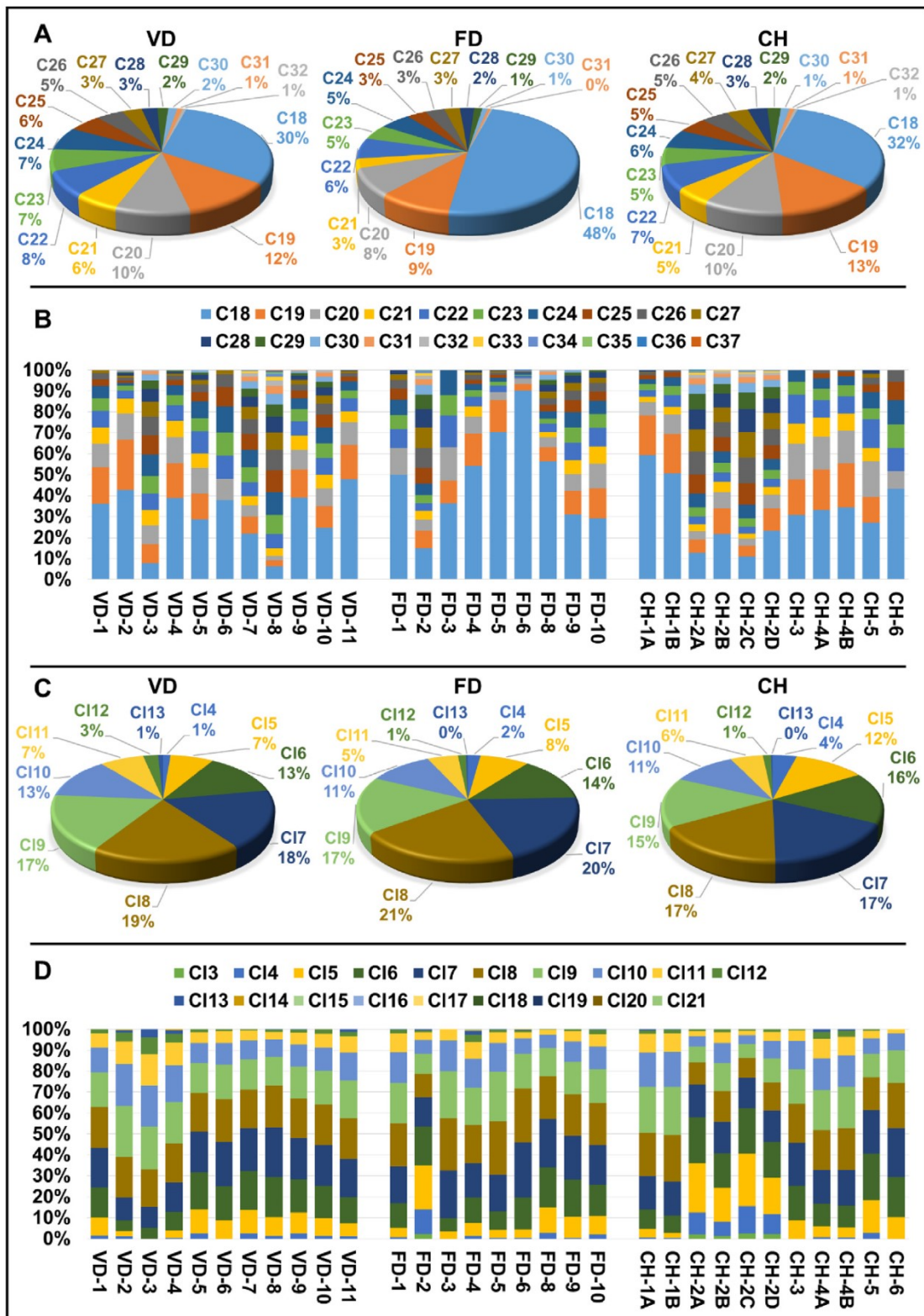


Fig. 5. Congener profiles of the median Σ LCCPs in the dust and hair samples where (A), is the composition based on carbon chain length, (B) is the carbon chain length in the individual samples, (C) is the composition based on chlorine substitution, (D) is the chlorine substitution in the individual samples.

differences in congener profiles between cat hair and dust may be of importance considering that dust ingestion rates are regularly used for risk assessments. Cat hair provides specific information on indoor exposure and could be seen as a non-invasive passive

sampler to continued exposure of CPs in the indoor environment. This study further supports the analytical capability to provide reliable data on present and future levels of chlorinated paraffins in South African environmental matrices.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2019.124643>.

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