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Chlorinated paraffins leaking from hand blenders can lead to significant human exposures

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ABSTRACT

Background: Chlorinated paraffins (CPs, polychlorinated *n*-alkanes) are versatile, high-production-volume chemicals. A previous study indicated that hand blenders leak CPs into prepared food.
Objectives: (1) to estimate exposure to CPs from hand blender use compared to background CP exposure from diet; (2) to assess the risk from human dietary exposure to CPs from hand blender use; (3) to investigate how hand blenders leak out CPs.
Methods: CPs were analyzed in food market baskets, in cooking oil/water samples (1 g oil/100 mL water) mixed using 16 different hand blenders, and in dismantled components of the hand blenders.
Results: Dietary intake of CPs from food market baskets was calculated to be 4.6 μg/day per capita for Swedish adults. Total CP amounts in oil/water leakage samples ranged from < 0.09 to 120 μg using the hand blenders once. CP leakage showed no decreasing levels after 20 times of hand blender usage. CP profiles in the leakage samples matched those of self-lubricating bearings and/or polymer components disassembled from the hand blenders.
Conclusions: Usage of 75% of the hand blenders tested will lead to increased human exposure to CPs. The intake of CPs for Swedish adults by using hand blenders once a day can raise their daily dietary intake by a factor of up to 26. The 95th percentile intake of CPs via using the hand blenders once a day exceeded the TDI for Swedish infants with a body weight < 7.2 kg. CP leakage came from blender components which contain CPs. The leakage may last several hundred times of hand blender use.

1. Introduction

Chlorinated paraffins (CPs, C_6–C_{38}), or polychlorinated *n*-alkanes, have numerous applications, e.g. as plasticizers, flame retardants, lubricants and in metal cutting fluids (USEPA, 2009). The annual global production currently exceeds 1 million tons (Glüge et al., 2016). CP technical products are complex mixtures, conventionally categorized into short-chain (C_{10}–C_{13}, SCCPs), medium chain (C_{14}–C_{17}, MCCPs), and long chain (≥ C_{18}, LCCPs) CPs, and sub-defined according to their chlorination degree (30% Cl – 70% Cl by weight) (EnvironmentCanada, 1993). < C_{10} CPs are considered impurities of SCCPs (Reth and Oehme, 2004). The toxicity of CPs has been evaluated using SCCP, MCCP or LCCP products. SCCPs are potential carcinogens (IARC, 1990) and Persistent Organic Pollutants (POPs) under Stockholm Convention (UNEP, 2017). MCCPs have been registered under the REACH Regulation and are included in the EU endocrine disruptor candidates list as well as SCCPs (EU, 2015). MCCPs have been shown to cause hemorrhaging effects via suspected vitamin K deficiency in rats (DanishEPA, 2014). Rats exposed to LCCPs were observed an increase in liver weight (Bucher et al., 1987; Nilsen et al., 1981). Recently, the U.S. EPA took strong action in regulating the production and import of SCCPs, MCCPs and LCCPs into the U.S. (ACC, 2016).

CPs up to C_{18}, especially those with high chlorination degree, have shown bioaccumulation (Darnerud et al., 1982; Fisk et al., 2000) and biomagnification potentials (Houde et al., 2008). SCCPs, MCCPs and LCCPs were identified in human tissues by (Campbell and McConnell, 1980) and in human blood (Li et al., 2017). SCCPs and MCCPs were analyzed in mothers' milk (Thomas et al., 2006; Xia et al., 2017b) with a median total concentration up to 797 ng/g lipid (Xia et al., 2017a). The US EPA considers ingestion as a primary non-occupational exposure

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route to SCCPs (USEPA, 2009), and MCCPs/LCCPs are assumed to be without any risks to humans via drinking water or fish ingestion at the present exposure levels (USEPA, 2015). However, a previous study showed that foodstuff prepared for feeding experiments of cats was contaminated by CPs coming from use of some hand blenders sold on the Swedish market, indicating these may pose a risk for human exposure to CPs (Strid et al., 2014).

Hand blenders are common kitchen appliances used to make soups, smoothies, sauces, and in particular baby food. The hand blender motor drives the cutter blade in a bell-shaped casting at the other end of the blender through a working shaft.

The finding of CPs leaking out of hand blenders was unexpected. This leads to an urgent need to assess the health risk from dietary exposure to the leaking CPs and to understand how CPs leak from the hand blenders. To address the needs, we measured CP amounts in the leakage from the hand blenders' use and estimated the background daily dietary intake using Swedish food market baskets. We further tested if the amount of CP leakage changed with usage time. Finally, we dismantled all the hand blenders and compared CP patterns in the components with the patterns in the leakage to search for the source(s).

2. Methods

2.1. Hand blenders and other food processors

2.1.1. Sample collection

All food processors (see the Supplementary material) were purchased from retailers in Sweden. The prices ranged from ca. 99 to 2000 Swedish kronor (equivalent to ca. 10 to 250 US dollars), see Table 1. Ten different brands were tested. Twelve hand blenders were purchased in 2014, and four in 2016, two of which were upgrade models of their 2014 models. A kitchen glass jar blender and a coffee mill, representing different types of food processors, were purchased in 2016.

2.1.2. Leakage test

New hand blenders and the other two food processors were first cleaned according to the written instruction manuals from the manufacturers (see the Supplementary material). As a standardized surrogate for food, approximately 1 g of CP-free cooking oil was mixed with 100 mL of water at room temperature (20 °C) using a hand blender once. The mixing time was under the recommended maximum running time, varying from 10 s to 2 min, for fluids or soft foodstuffs specified in the instruction manual of respective hand blenders. In cases where no such information was available the hand blenders were used for 1 min. For the coffee mill, 2 g of activated silica were ground as coffee-bean surrogate and collected for analysis.

2.1.3. Wearage test

Three hand blenders (H1, H2 and H11) were run twice to test whether CP leakage was a one-off occurrence. It was not, therefore wearage tests on two more hand blenders were performed (H5 and H16). Two blenders were tested repeatedly by mixing new samples of oil and water for 1 or 2 min, according to the recommended use time in the instruction manual for the respective blender. Oil-water mixtures from the fifth, tenth, fifteenth and twentieth runs were extracted and analyzed for CP leakage. Since there was no significant decrease of CP leakage, we did not rerun the other blenders.

2.1.4. Extraction and clean-up

The oil-water mixture was spiked with Dechlorane-603 as a surrogate standard and extracted using isohexane:acetone (3:1). Lipids were removed using concentrated sulfuric acid. The extracts were further cleaned up by applying to a sulfuric acid silica column. The eluent solvent was evaporated off first and then 50 ng α-HBCDD in 100 μL of isooctane was added as volumetric standard.

2.1.5. Dismantling

Complete information and details on the dismantling procedures are given in the Supplemental material. Briefly, the bell-shaped end (Fig. 1) was detached from the hand blender. The working shaft was then taken apart from the bell-shaped casting. The shaft was sawed off for components, including bearing(s), washers and polymer coating.

2.1.6. Component analysis

Analyzed components were wiped clean, submerged in acetone in respective test tubes and placed in an ultrasonic bath for 30 min. Extracts were solvent-exchanged to hexane and cleaned up using a sulfuric acid silica column. To reduce the number of tests, the analysis started with bearings which contained visible lubricant. Polymer washers were then analyzed if the CP pattern of the bearing did not fit the pattern of the leakage samples. Finally, polymer coatings were analyzed if neither pattern fits that of the leakage samples.

2.2. Daily dietary intake of CPs

Food market baskets consisting of 13 food categories were purchased by the National Food Agency, Sweden from five different Swedish food grocery chains during May–June 2015. Each food category contained a varying number of food items (up to 31 items per category: meat) (see Table S3), and the contributing weight of each food item to the total homogenate was based on food consumption data from the Swedish Board of Agriculture. For each food category, the per capita intake was obtained by combining concentration data and consumption data, and addition of all 13 food categories gave the total per capita dietary intake. In this report we have used the calculated daily mean (per capita) dietary intake of CPs as a benchmark of background dietary intake.

Samples were stored in glass containers at −25 °C until analysis. Due to economic constraints samples from the five grocery chains were pooled before analysis. The extraction and cleanup procedure is based closely on that described previously (Wong et al., 2017; Yuan et al., 2017; Zhou et al., 2016). A method overview is shown in Fig. S1. Detailed information on the food categories, cleanup procedure and daily intake estimation is given in the Supplemental material.

2.3. Instrumental analysis

CPs were analyzed at a chemical resolution of carbon-chlorine congener groups, i.e. CP congeners with a fixed chain length (n) and number of chlorines (m) (denoted as C_nCl_m). Analytes were directly injected into an APCI-QTOF-MS (QTOF Premier, Waters, UK). Instrument settings as described previously (Bogdal et al., 2015) were applied with several adjustments (Yuan et al., 2016). Detector responses of m/z ratios (Table S6) corresponding to 277 congener groups from C_6Cl_4 to $C_{31}Cl_{12}$ were considered to form a congener group pattern.

2.4. Quantification

The CP quantification method developed by (Bogdal et al., 2015) was used. CP congener group patterns of a set of CP technical products (n = 44) were initially analyzed, and a sub-set of 18 products were selected for quantification in this study, consisting of 6 SCCPs, 9 MCCPs and 6 LCCPs (Table S5). The CP congener group pattern of each sample was reconstructed by a pattern-deconvolution algorithm from CP patterns of the selected products. The goodness-of-fit between CP patterns was evaluated by the coefficient of determination R^2, with $R^2 = 1$ for a perfect fit. In this study, CP patterns in the tested hand blenders were satisfactorily reconstructed ($R^2 > 0.80$). Relative contributions of the products were then used to calculate instrument response factors of SCCPs, MCCPs and LCCPs respectively in the sample. An example of quantification of CPs from blender H8 is given in Fig. 2. Detailed