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Distribution of polychlorinated naphthalenes (PCNs) in the whole blood of typical meat animals

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ABSTRACT

The concentrations and distribution of polychlorinated naphthalenes (PCNs) in the whole blood of eight typical terrestrial meat animals (chicken, duck, rabbit, pig, cattle, sheep, horse and donkey) consumed daily in our life were investigated. The total concentrations (on a liquid volume basis) of PCNs were in a range from 305 to 987 pg/L. Donkey blood contained the highest PCN concentrations. Mono-CNs were the dominant homolog group, accounting for 38%–71% PCNs. Apart from the mono-CNs and tri-CNs homolog groups, two hepta-CNs (mean: 9.5%) contributed most, followed by tetra-CNs (mean: 6.5%). The congeners CN1, 5/7, 24/14, 27/30, 52/60, 66/67, and 73 were the most abundant congeners or congener groups. The highest toxicity equivalencies (TEQs) were observed in cattle blood (117.4 fg TEQ/L) then chicken blood (117.1 fg TEQ/L). CN73 contributed 65% to total TEQs, followed by CN70 (20%) and CN66/67 (14%). The dietary intakes of PCNs were also estimated. Chicken meat, which forms the second largest component of meat product consumption in China, contributed most to the total TEQs (61%), followed by beef (27%) and pork (5.9%). The consumption of chicken might pose the highest risk from exposure to PCNs than other types of meat to populations who prefer to eat chicken meat.

Introduction

Polychlorinated naphthalenes (PCNs), a mixture of up to 75 congeners, are persistent organic pollutants listed under the Stockholm Convention that have been commercially applied or used in the production of traded goods, which have been banned for several decades because of their high potential adverse ecological effects on the environment (Yamashita et al., 2003). Although the manufacture of PCNs has ceased, they are still being emitted from their former application and use in products, with simultaneous secondary pollution and emission as by-products during thermal processes (Fernandes et al., 2017). PCNs were detected everywhere, including air, water, marine sediments, and biota (Kunisue et al., 2009). Several PCN congeners have been reported to interact with aryl hydrocarbon receptors (AhR) to produce dioxin-like responses in humans such as dermal lesions, mortality, and carcinogenicity, indicating that PCN exposure can cause toxic effects on the environment and human health (Blankenship et al., 2000; Falandysz et al., 2014; Villeneuve et al., 2000).

Foods of animal origin, including meat, eggs, milk, and fish, form a dietary transfer pathway, which is considered as the major route of human exposure to PCNs, accounting for 90% of their daily individual intake (Falandysz, 2003).

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Marti-Cid et al. (2008) reported PCNs in local foodstuffs randomly collected from 12 representative cities of Catalonia, Spain. The highest concentrations of PCNs were found in fish and seafood (47.1 ng/kg ww (wet weight)), followed by oils and fats (21.5 ng/kg ww), bakery products (15.3 ng/kg ww), and dairy products (11.7 ng/kg ww). Fernandes et al. (2011) investigated PCNs in 100 foods commonly consumed by the local population of Ireland: the highest PCNs were in fish (21.8 ng/kg ww), followed by animal fat (3.69 ng/kg ww), and dairy products (1.81 ng/kg ww).

Determination of PCNs in marine ecosystems has indicated the bio-magnification of some congeners of PCN in the food chain of a marine mammal and black cormorant and some lower food chain animals from the Baltic Sea (Falandysz and Rappe, 1996, 1997; Lundgren and Tysklind, 2002) and general accumulation in fish (Falandysz et al., 1997; Kannan et al., 2000). Chickens were found more contaminated with PCNs than pigs (Guruge et al., 2004). Besides, PCNs were also detected in the human body, including breast milk, adipose tissue, liver, and blood (Fernandes et al., 2017). At present, there is very little information on the analysis of PCNs in blood and the internal exposure of PCNs in animals to humans. Therefore, the present study aims to determine the levels of PCNs in the blood of typical meat animals, and thus contribute to understanding how humans are exposed to PCNs through consuming animal tissues.

1. Materials and methods

1.1. Sample collection

Whole blood samples, treated with ethylene diamine tetraacetic acid (EDTA) as an anticoagulant, and collected from eight species of meat animals commonly consumed by humans (chicken, duck, rabbit, pig, cattle, sheep, horse and donkey), were purchased from Bersee Technology Co. Ltd. (Beijing, China). These whole blood samples were taken at random from healthy adult animals raised on large farms in the suburbs of Beijing. The fresh blood was stored in a refrigerator at 4–8°C then analyzed within three weeks.

1.2. Sample extraction and analysis

The analytical procedures for PCNs used an established isotope dilution high-resolution gas chromatography/high-resolution mass spectrometry method (Li et al., 2017; Liu and Zheng, 2013; Wang et al., 2016). As information on the pretreatment of animal blood is limited, the samples were analyzed on the basis of studies related to human blood (Wittsiepe et al., 2007) with some modifications. Briefly, 50 mL of the whole blood samples was diluted with 50 mL of deionized water then shaken for 30 min. Then 50 mL of aqueous saturated ammonium sulfate solution was added to the samples and shaken for 1 min followed by the addition of 50 mL of absolute ethanol then shaking for 1 min. Finally, the samples were extracted using 100 mL of *n*-hexane which was dried by passing through anhydrous sodium sulfate.

The extraction was spiked with an internal standard (ECN-5102, Cambridge Isotope Laboratories Inc., Tewksbury, MA, USA). The clean-up was treated using a multilayer silica gel column and a basic alumina column. Each extract was evaporated to approximately 20 μL using a rotary evaporator, placed under a gentle stream of nitrogen, then spiked with recovery standards (ECN-5260, Cambridge Isotope Laboratories Inc., Tewksbury, MA, USA). The PCNs were analyzed by a gas chromatograph (GC) coupled to a DFS mass spectrometer (Thermo Fisher Scientific, Hudson, NH, USA) using an electron impact (EI) source.

1.3. Quality assurance and quality control

One laboratory blank sample was analyzed with each batch of samples (2 or 3 samples), following the same procedure as in Section 1.2. The PCN peaks in the chromatogram were identified by comparing their retention times with those of the internal standards and by their ion ratios. The method performance was evaluated using the limit of detection (LOD) and the limit of quantification. The LODs of the PCNs were 0.01–0.29 pg/g. The recoveries of the PCN internal standards in the samples were 42%–97%, which met the requirements for trace PCN analysis (Fernandes et al., 2017) but this fell to only 15% for PCN congener concentrations near the LODs.

2. Results and discussion

2.1. Concentrations of PCNs

PCNs were detected in most of the animal whole blood samples analyzed. A total of 75 mono- to octa-CN congeners were quantified. The total concentrations of PCNs in the different animal whole blood samples are described in Fig. S1. The concentrations (on a liquid volume basis) of PCNs ranged from 305 to 987 pg/L (median: 826 pg/L, mean: 756 pg/L) in the eight species of commonly-consumed animals examined. Donkey blood contained the highest PCN concentrations (987 pg/L), which was 3.2 times higher than those in pig blood (305 pg/L), 1.7 times those in sheep blood (576 pg/L), and 1.4 times those in chicken blood (725 pg/L). The average concentration of PCNs in the horse, duck, rabbit, and cattle blood samples was (863 ± 50) pg/L, with only slight differences between these animals.

Guruge et al. (2004) found that the concentration of PCNs in pig fat (160 pg/g lipid weight) was 40% of that in chicken fat (400 pg/g lipid weight), perhaps because of the rapid growth in the weight and amount of fat of pigs. The concentration of PCNs in the whole blood of the pigs in the latter study was just 42% that of chicken found in the present study, with similar distributions of PCNs in the fat of pig and chicken meat. Fernandes et al. (2011) found that the average total PCNs in duck, ovine, bovine, and porcine fat from Ireland were in a decreasing order: 3.8, 3.6, 2.3, and 1.7 ng/kg ww, respectively. Many studies have correlated the concentration of PCNs in marine mammals, fish, with those of food products in the diet such as milk, eggs, liver, and dairy, detecting the highest level of PCNs in fish, followed by animal fat (Braune and Muir, 2017; Falandysz et al., 1997; Li et al., 2015; Rotander et al., 2012).

The patterns of PCN (mono- to octa-CNs) homologs in the animal whole blood samples are shown in Fig. 1. Of the

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