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Original Article

Concentrations of perfluoroalkyl substances in foods and the dietary exposure among Taiwan general population and pregnant women

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ABSTRACT

This study quantified five perfluorocarboxylic acids (PFCAs) and two perfluorosulfonic acids in cereals, meats, seafood, eggs, pork liver, and milk in Taiwan using ultra-performance liquid chromatography-tandem mass spectrometry and evaluated the dietary exposure of the general population and pregnant women using per capita consumption and a questionnaire, respectively. Perfluorooctanoic acid (PFOA) and PFCAs of 10–12 carbons were found in almost all of the samples in considerable concentrations in rice and pork liver, reaching as high as 283 ng/g (PFOA in pork liver); the levels are two to three orders of magnitude higher than previous reports. Perfluorooctane sulfonate (PFOS), the most frequently mentioned perfluoroalkyl substance, was rarely detected in many food items (detection frequencies <20% in rice, flour, pork, chicken, salmon, squid, eggs, and milk) at <0.4 ng/g, except for beef, pork liver and some seafood (detection frequencies: 100%, GMs: 0.05–3.52 ng/g). Compared to populations in Western countries, people in Taiwan are exposed to much more perfluorohexanoic acid, PFOA, perfluorodecanoic acid, and perfluoroundecanoic acid (11.2, 85.1, 44.2, and 4.45 ng/kg b.w./day, respectively), mainly due to the higher contaminations in food. The exposure of 8.0 μg PFOA/person/day in the 95 percentile of pregnant women was due to their frequent consumption of pork liver.

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1. Introduction

Perfluoroalkyl substances (PFASs), including perfluorocarboxylic acids (PFCAs), perfluorosulfonic acids (PFSAs), and their salts are chemically-stable, water-proof, oil-proof, and heat-resistant, and thus they are widely used in the industry [1,2]. PFASs and their impurities are released into the environment in all stages of production, use, and disposal. It is estimated that 45 thousand tons of perfluorooctane sulfonate (PFOS) may have entered the air or environmental bodies of water between 1970 and 2012 globally [3]. Perfluorinated compounds have been gradually phased out in the world. However, due to their persistency and global ocean circulation, previously released PFASs have accumulated in the environment. The bioaccumulation factors of PFASs in fish are positively associated with fluorinated carbon chain lengths, and range from 1000 to 5000 in fish [4]. In general, PFOS is the most frequently detected and is found at higher concentrations than other PFASs in wild animals, followed by PFOA [5–9]. As high as 225 and 380 ng/g of PFOS has been observed in wild fish in Germany and in the United States, respectively [10,11].

Toxicological studies, mostly focusing on PFOS and PFOA, have reported PFASs to have adverse effects on reproductive, endocrine, and immune systems, and to be carcinogenic [12–15]. In humans, PFAS exposure was positively correlated with triglyceride, total cholesterol, low-density lipoprotein cholesterol levels, and the increase in bladder cancer mortality [16,17]. In addition, high PFAS levels in blood have been associated with prolonged time to pregnancy and low semen quality in adults, as well as preterm birth and low birth weight in the new-born [18–20].

The major route of exposure to PFASs is through the ingestion of contaminated foods, especially long-chain, large-molecule PFASs from foods of animal origin [21–24]. These compounds tend to bind cytosolic proteins and thus they are frequently found in meat, seafood, and dairy products [4,25–27]. In China, Gulkowska et al. found sub-to low ng/g of PFOS in all their samples of fish, molluscs, crabs, shrimps, and other shellfish and observed PFOA and perfluoroundecanoic acid (PFUnDA) in more than half of their samples [28]. In Norway, Haug et al. detected up to 12 PFASs in cod liver, fish, eggs, chicken, pork, and milk, and found PFOS and PFUnDA to be the most abundant [29]. A similar result was reported by Eriksson et al., showing PFUnDA to be the most abundant in fish (<24–290 pg/g) and milk (<130–190 pg/g), followed by PFOA (<100–240 pg/g and <67–77 pg/g, respectively) [30]. Regarding the effect of dietary frequency, one market basket study in Sweden showed that people who consume more meat and fish are exposed to higher amounts of perfluorodecanoic acid (PFDA) and perfluorononanoic acid (PFNA) as well as PFOS and PFUnDA than those who eat more cereals and vegetables [31]. In a study of a one-day composite of diet samples containing more seafood in South Korea, Kim et al. reported higher concentrations of long-chain PFASs (C_{11}–C_{14}) than those of short-chain PFASs such as perfluorohexanoic acid (PFHxA) [32].

Among the PFASs with various carbon-chain lengths, PFOA and PFOS in foods are the most frequently detected and reported to date, especially for seafood and meat. Besides, compared to the Western countries, people in East and Southeast Asia consume less meat [33], more rice than wheat, and more seafood than those in Europe and America. An investigation on the exposure to PFASs through the East Asian diet is needed. This study measured seven PFASs in 140 samples of 14 foods primarily consumed in Taiwan, and estimated the daily intake of PFASs based on these measured concentrations and the diet frequency obtained from per capita consumption and a questionnaire.

2. Methods

2.1. Samples

Ten batches of 14 types of food (rice, flour, pork, chicken, beef, salmon, grass carp, oysters, clams, shrimps, squids, eggs, pork liver, and milk) were collected between September 2010 and April 2011. The 14 foods were chosen based on per capita consumption data for 2008 obtained from the Taiwan Council of Agriculture [34], which lists the most consumed foods in the four categories: (1) cereals: rice (59%) and wheat (41%); (2) meats: pork (51%), poultry (41%), and beef (5%); (3) seafood: fish (45%) and shellfish (38%) and (4) dairy products: milk (78%, Table 1). Because liver is a target organ of PFASs [16,35] and pork liver is a food very commonly consumed by pregnant women in Taiwan, pork liver was also included in the samples. The foods, except for milk, were purchased from two traditional markets in Taipei City, the capital of Taiwan. The milk was purchased from convenience stores. There are very limited agricultural activities in Taipei City, where raw food and food products are from the entire Taiwan, which is a small island (area 35,883 km^2) with well-organized food transportation systems.

2.2. Chemicals and reagents

The standard solutions of seven analytes including (a) five PFCAs: PFHxA, PFOA, PFDA, PFUnDA, and perfluorododecanoic acid (PFDoDA) and (b) two PFSAs: perfluorohexane sulfonate (PFHxS) and PFOS, as well as five stable isotope-labeled compounds (perfluoro-*n*-[1,2,3,4,6-$^{13}C_5$] hexanoic acid, perfluoro-*n*-[1,2,3,4-$^{13}C_4$]octanoic acid, perfluoro-*n*-[1,2,3,4-$^{13}C_4$]undecanoic acid, sodium perfluoro-1-hexane[$^{18}O_2$]sulfonate, and sodium perfluoro-1-[1,2,3,4-$^{13}C_4$] octanesulfonate) all had > 98% of purity at 50 μg/mL in 1.2 mL in methanol and were purchased from Wellington Labs (Guelph, Ontario, Canada).

Methanol of LC/MS grade was purchased from J.T. Baker (Phillipsburg, NJ, USA) as the organic mobile phase on liquid chromatography. Methanol, acetonitrile, acetone, *n*-heptane, and dichloromethane of LC grade, and ammonium hydroxide (28–30%), potassium hydroxide (85%), and analytical-grade formic acid (88%), which were used for sample preparation, were also purchased from J.T. Baker. *N*-methylmorpholine of analytical grade (95.5%) was purchased from Sigma–Aldrich (Saint Louis, MO, USA).

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