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

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\textbf{A B S T R A C T}

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 \textmu g 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 perflurocarboxylic acids (PFCAs), perfluorosulfonic acids (PFSSAs), 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 persistence 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, PFOS 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, Gukowska 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 (C11–C14) 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 km²) with well-organized food transportation systems.

2.2. Chemicals and reagents

The standard solutions of seven analytes including (a) five PFCAs: PFHxA, PFOA, FFD, PFUnDA, and perfluorododecanoic acid (PFDoDA) and (b) two PFSSAs: perfluorohexane sulfonate (PFHxS) and PFOA, as well as five stable isotope-labeled compounds (perfluoro-n-[1,2,3,4,6-13C5]hexanoic acid, perfluoro-n-[1,2,3,4-13C4]octanoic acid, perfluoro-n-[1,2,3,4-13C3]undecanoic acid, sodium perfluoro-1-hexane[18O2]sulfonate, and sodium perfluoro-1-[1,2,3,4-13C4]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).
2.3. Analytical methods

The qualification and quantification of PFASs were described in a previous study [36]. Briefly, rice was ground to powder with mortar and pestle while meats and seafood were homogenized using a laboratory blender. One-gram sample was spiked with 100 μL of the 0.5-ng/μL isotope-labeled internal standards and was digested for 2 h in a methanol-prewashed homogenization tube (DT-20, IKA Works, Staufen, Germany) with 10 mL of 0.5-N potassium hydroxide in methanol on Ultra-Turrax Tube Disperser (IKA Works). Afterward the sample was centrifuged at 3000 rpm (1410 × g) for 30 min and the supernatant was collected and diluted with 500-mL acidified Milli-Q water (pH 3.5). As for the milk, a 25-mL sample was added with 100 μL of the 0.5-ng/μL internal standards and 450 mL of 0.5-N potassium hydroxide, shaken for 30 min, and acidified to pH 3.5 with nine-mL formic acid. After filtering with glass fiber filters (pore size 0.45 μm), the acidified samples were extracted using conditioned Atlantic HLB disks (47 mm, 450 mL of 0.5-N potassium hydroxide, Phenomenex, Torrance, CA, USA) using automated solid phase extractors (SPE-DEX 4790, Horizon Technology). The eluent of 20-mL 0.1% ammonium hydroxide in methanol was filtered through a nylon membrane and was concentrated to one mL. Isotope dilution techniques were used to quantify the PFASs. A process blank, a duplicate of a food sample, was reduced. Isotope dilution techniques were used to quantify the PFASs. A process blank, a duplicate of a food sample, and a quality control (QC) sample spiked with 100 μL of 0.1-ng/μL PFASs were included in each batch of 14 samples for all the ten batches. The limits of detection (LODs) mostly ranged from 0.01 ng/g to 0.07 ng/g, except for PFHxA in beef and oyster (0.28 and 0.13 ng/g, respectively), PFUnDA in eggs (0.10 ng/g), and PFOS in pork liver (0.31 ng/g, Table S1) [36]. The background levels obtained from process blanks ranged from < LOD to sub-ng/g for PFUnDA, PFDoDA, PFHxS, and PFOS, and at low ng/g levels for PFHxA, PFOA, and PFDA. The randomly selected duplicates for each batch in the ten batches turned out to be oyster, liver, rice, salmon, liver, clam, oyster, oyster, and liver, which proved precise quantification with most relative percent differences (RPDs) less than 15% between duplicates. The spiked QC matrixes for the ten batches were clam, beef, clam, grass carp, oyster, oyster, liver, salmon, beef, and clam, and the average differences in concentrations between the spiked and non-spiked samples ranged from 8.67 (PFDoDA) to 14.8 ng/g (PFUnDA), which was accurate comparing with the spiked level of 10 ng/g.

2.4. Quality control and data analysis

To prevent contamination, laboratory materials containing PFASs were avoided. Glassware was cleaned with tap water and Milli-Q water, followed by rinsing with acetone, n-heptane, dichloromethane, and methanol, respectively. Filter membranes were prewashed with methanol. Methanol was injected three times before each batch subjected to instrumental analysis to ensure that any residual PFAS in the UPLC-MS/MS system was removed and background interference was reduced. Isotope dilution techniques were used to quantify the PFASs. A process blank, a duplicate of a food sample, and a quality control (QC) sample spiked with 100 μL of 0.1-ng/μL PFASs were included in each batch of 14 samples for all the ten batches. The limits of detection (LODs) mostly ranged from 0.01 ng/g to 0.07 ng/g, except for PFHxA in beef and oyster (0.28 and 0.13 ng/g, respectively), PFUnDA in eggs (0.10 ng/g), and PFOS in pork liver (0.31 ng/g, Table S1) [36]. The background levels obtained from process blanks ranged from < LOD to sub-ng/g for PFUnDA, PFDoDA, PFHxS, and PFOS, and at low ng/g levels for PFHxA, PFOA, and PFDA. The randomly selected duplicates for each batch in the ten batches turned out to be oyster, liver, rice, salmon, liver, clam, oyster, oyster, oyster, and liver, which proved precise quantification with most relative percent differences (RPDs) less than 15% between duplicates. The spiked QC matrixes for the ten batches were clam, beef, clam, grass carp, oyster, oyster, liver, salmon, beef, and clam, and the average differences in concentrations between the spiked and non-spiked samples ranged from 8.67 (PFDoDA) to 14.8 ng/g (PFUnDA), which was accurate comparing with the spiked level of 10 ng/g.
2.5. Evaluation of daily intake

Daily intake of each food for each PFAS per person was calculated based on per capita consumption data for the year 2008 (Table 2) and the geometric mean (GM) concentrations of PFASs. In order to avoid overestimation, the PFASs with positive rates lower than 50% in a food item were not included in the estimation of daily intake; all the included data were those detected in nine or all of samples (n = 10). We calculated both the GMs and the medians of PFASs because the concentrations did not follow normal distributions; GMs instead of medians were chosen in the following assessments in this study because GMs would be more appropriate for representing central values of the measured concentrations. PFAS intakes from wheat were calculated using their concentrations in flour. The intake from cereals represented those from rice and flour. We summed the intake from salmon (a wild oceanic fish) and grass carp (a farmed freshwater fish) to constitute the total intake of fish. Similarly, meat included pork, poultry, and beef; shellfish covered oyster, shrimp, clam, and squid. The daily intake per person was divided by the average body weight of an adult in Taiwan (65 kg) to calculate the exposure dose per kilogram body weight.

In addition to the general population, we also evaluated the exposure in pregnant women in Taiwan. This study was approved by the Research Ethics Committee, National Taiwan University Hospital (#200812144R), and informed consent was obtained from each individual. Sixty-two questionnaires on food consumption were administered to pregnant women visiting gynecology clinics of National Taiwan University Hospital between October 2010 and June 2011. The questionnaire was composed of two sections, basic information and diet. The former contained the participants’ weight, height, ethnic group, and education. The latter was a food intake questionnaire, in which the participants answered the consumption frequency and amount of each food item in a week. For example, they checked the frequency of oceanic fish consumption among “always”, “frequent”, “sometimes”, “seldom”, and “never”, and the amount for each consumption among “< 1”, “1–2”, “2–3”, “3–4”, and “> 5” portions, which one portion was pre-defined and illustrated with both a picture and its weight on the questionnaire. The results were used to calculate the participants’ daily intake of each food item. Batch Fit analyses were conducted using Oracle Crystal Ball Fusion Edition Release 11.1.2.1.0 (Oracle Corporation, Redwood Shores, CA, USA) to estimate the optimized exposure model. Based on the model, the GM concentrations of PFASs were applied to one million times of Monte Carlo trials to obtain the cumulative intake from foods of 25%, 50%, 75%, and 95% probability among pregnant women.

3. Results and discussion

3.1. Distributions in foods

PFCAs were detected in most foods at GM concentrations ranging from sub-to < 30 ng/g with detection frequencies of 90% or 100% (Table 2). The GM and median concentrations were comparable (Table 2). PFOA was found in all of the samples and was the most abundant PFCA in most food items with GM concentrations ranging from 5.73 ng/g (eggs, n = 10) to 12.1 ng/g (beef, n = 10), except for 1.44 ng/mL in milk (n = 10). PFOA concentrations in salmon (GM: 7.48 ng/g), oyster (GM: 8.56 ng/g), pork liver (GM: 10.0 ng/g), and eggs (GM: 5.73 ng/g) were only lower than those of PFDA (GMs: 7.99, 12.3, 20.7, and 22.2 ng/g, respectively, Table 2). Compared with PFCCAs, the two PFASs were less frequently detected; for example, PFHxS was observed in five beef, one squid, one liver, and three milk samples, and PFOS was found in none to three samples of rice, flour, pork, chicken, salmon, oyster, squid, egg, and milk (n = 10 for each food item, Table 3). The concentrations of PFASs were found at GMs ranging from 0.04 to 0.73 ng/g, with the exception of PFOS in squid (1.06, detection frequency 10%) and pork liver (3.52 ng/g, detection frequency 100%, Table 3).

In general, this study found higher concentrations than those in previous reports. The concentrations of PFHxA and PFOA in rice (GMs: 1.10 and 8.90 ng/g, respectively) and flour (GMs: 1.33 and 8.84 ng/g, respectively) were two to three orders of magnitude higher than those reported for cereals in Sweden, which PFHxA and PFOA were at 0.004–0.011 ng/g and 0.011–0.062 ng/g, respectively [31]. PFOA in meats (GMs: 8.78–12.1 ng/g) were two orders of magnitude higher than those in Spain, Italy, and Norway, where PFOA was not detectable or lower than 0.1 ng/g in pork, beef, and chicken [27,29,37]. The concentrations of C10–C12 PFCCAs in meats (GMs: 3.06–3.80 ng/g, 0.13–0.24 ng/g, and 1.19–1.58 ng/g, respectively) were ten to a few hundred times higher than those reported by Haug et al. for Norway [29]. In this study, the detection frequencies and concentrations of PFCCAs found in fish (mostly 100%, GMs: 0.62–7.99 ng/g) were much higher than those reported for freshwater fish in Canada and for rainbow trout in Sweden (<0.102 ng/g) [26,38]; the PFHxA (GMs: 0.90–1.06 ng/g) and PFOA (GMs: 7.48–9.67 ng/g) in seafood were at least five times higher than those reported in China [25]. The concentrations of the PFASs in pork liver (GMs: 0.73–20.7 ng/g) were higher than those of PFHxA, PFOA, and PFOS reported by Jogsten et al. for lamb liver (<0.05–0.33 ng/g) [39], and those for the liver of wild animals including fish, birds, and marine mammals [40–42].

In contrast to considerable concentrations of PFOS in foods found by other studies [28,29,43], PFOS was only frequently detected in beef, grass carp, shrimp, and clam at low levels (GMs: 0.12, 0.53, 0.06, and 0.05 ng/g, respectively; n = 10, Table 3) and in pork liver at 3.52 ng/g (n = 10) in this study. The PFOS concentrations we found in beef (0.03–0.35 ng/g, GM: 0.12 ng/g) was more than ten times lower comparing with that in Italy (mean: 2.11 ng/g) but was similar to that of a beef sample in Norway (0.06 ng/g) [27,29]. Concentrations of PFOS in fish and other seafood were often reported higher than those of other PFASs [27,29,38,44]; however, we could rarely find it in salmon, oyster, and squid. This discrepancy could result from local differences in contamination [45]. The information on environmental concentrations of PFOS in Taiwan is limited to rivers that receive industrial or domestic wastewater [46,47]; the concentrations vary a lot with 64.4–7165 ng/L in river water, 95–1828 ng/g in river fish muscle, and 110 to 28,933 ng/g in river fish liver [47]. Because the above river water is not
Concentrations of perfluorocarboxylic acids in foods (ng/g w.w.; ng/mL for milk; n = 10).

\[ \text{Concentrations of perfluorosulfonic acids in foods (ng/g w.w.; ng/mL for milk; n = 10).} \]

\[
\begin{align*}
\text{Food item} & \quad \text{PFHxA} & \quad \text{PFOA} & \quad \text{PFDA} & \quad \text{PFUnDA} & \quad \text{PFDoDA} \\
\text{Rice} & \quad 100\% & \quad 1.10 & \quad 1.07 & \quad 100\% & \quad 8.90 & \quad 5.37 & \quad 100\% & \quad 5.67 & \quad 5.12 & \quad 100\% & \quad 0.48 & \quad 0.50 & \quad 100\% & \quad 1.62 & \quad 1.63 \\
\text{Flour} & \quad 100\% & \quad 1.33 & \quad 1.39 & \quad 100\% & \quad 8.84 & \quad 6.44 & \quad 90\% & \quad 2.47 & \quad 2.11 & \quad 90\% & \quad 0.11 & \quad 0.17 & \quad 90\% & \quad 1.98 & \quad 1.87 \\
\text{Pork} & \quad 100\% & \quad 1.31 & \quad 1.02 & \quad 100\% & \quad 10.7 & \quad 5.91 & \quad 100\% & \quad 3.22 & \quad 3.63 & \quad 100\% & \quad 0.18 & \quad 0.18 & \quad 90\% & \quad 1.32 & \quad 1.68 \\
\text{Beef} & \quad 100\% & \quad 1.16 & \quad 1.21 & \quad 100\% & \quad 12.1 & \quad 8.27 & \quad 100\% & \quad 3.80 & \quad 3.70 & \quad 100\% & \quad 0.24 & \quad 0.30 & \quad 90\% & \quad 1.58 & \quad 1.63 \\
\text{Chicken} & \quad 100\% & \quad 1.17 & \quad 1.02 & \quad 100\% & \quad 8.78 & \quad 6.49 & \quad 100\% & \quad 3.06 & \quad 3.35 & \quad 100\% & \quad 0.13 & \quad 0.19 & \quad 90\% & \quad 1.19 & \quad 1.30 \\
\text{Salmon} & \quad 100\% & \quad 1.06 & \quad 1.08 & \quad 100\% & \quad 7.48 & \quad 5.39 & \quad 100\% & \quad 7.99 & \quad 8.55 & \quad 100\% & \quad 0.62 & \quad 0.72 & \quad 90\% & \quad 1.44 & \quad 1.78 \\
\text{Grass Carp} & \quad 100\% & \quad 1.02 & \quad 1.04 & \quad 100\% & \quad 7.84 & \quad 5.95 & \quad 100\% & \quad 6.79 & \quad 7.59 & \quad 100\% & \quad 1.17 & \quad 1.19 & \quad 90\% & \quad 1.60 & \quad 1.78 \\
\text{Oyster} & \quad 100\% & \quad 0.94 & \quad 1.20 & \quad 100\% & \quad 8.56 & \quad 5.71 & \quad 90\% & \quad 12.3 & \quad 10.5 & \quad 100\% & \quad 1.39 & \quad 1.39 & \quad 90\% & \quad 5.38 & \quad 6.28 \\
\text{Shrimp} & \quad 100\% & \quad 0.90 & \quad 0.87 & \quad 100\% & \quad 7.75 & \quad 8.23 & \quad 100\% & \quad 6.58 & \quad 7.36 & \quad 100\% & \quad 0.80 & \quad 0.78 & \quad 90\% & \quad 3.69 & \quad 4.63 \\
\text{Clam} & \quad 100\% & \quad 1.05 & \quad 0.98 & \quad 100\% & \quad 8.25 & \quad 6.40 & \quad 100\% & \quad 6.60 & \quad 7.05 & \quad 100\% & \quad 0.80 & \quad 0.86 & \quad 90\% & \quad 2.30 & \quad 3.07 \\
\text{Squid} & \quad 100\% & \quad 1.05 & \quad 1.07 & \quad 100\% & \quad 9.67 & \quad 6.40 & \quad 100\% & \quad 5.94 & \quad 7.84 & \quad 100\% & \quad 5.94 & \quad 7.84 & \quad 90\% & \quad 6.90 & \quad 9.93 \\
\text{Egg} & \quad 100\% & \quad 0.95 & \quad 0.82 & \quad 100\% & \quad 5.73 & \quad 5.35 & \quad 100\% & \quad 22.2 & \quad 23.1 & \quad 100\% & \quad 0.94 & \quad 0.95 & \quad 100\% & \quad 6.49 & \quad 7.87 \\
\text{Liver} & \quad 100\% & \quad 0.30 & \quad 0.03 & \quad 100\% & \quad 1.44 & \quad 1.70 & \quad 100\% & \quad 0.94 & \quad 1.53 & \quad 100\% & \quad 0.06 & \quad 0.02 & \quad 10\% & \quad 0.07 & \quad 0.07 \\
\text{Pork} & \quad 100\% & \quad 0.07 & \quad 0.07 & \quad 100\% & \quad 0.03 & \quad 0.03 & \quad 100\% & \quad 0.23 & \quad 0.23 & \quad 100\% & \quad 0.23 & \quad 0.23 & \quad 90\% & \quad 0.23 & \quad 0.23 \\
\text{Salmon} & \quad 100\% & \quad 0.07 & \quad 0.07 & \quad 100\% & \quad 0.03 & \quad 0.03 & \quad 100\% & \quad 0.23 & \quad 0.23 & \quad 100\% & \quad 0.23 & \quad 0.23 & \quad 90\% & \quad 0.23 & \quad 0.23 \\
\end{align*}
\]

DF: detection frequencies; number of detectable samples divided by the sample number of a food item (n = 10). GM: geometric means of detectable concentrations.

PFOA: perfluorooctanoic acid.
PFDA: perfluorodecanoic acid.
PFUnDA: perfluoroundecanoic acid.
PFHxA: perfluorohexanoic acid.
PFDoDA: perfluorododecanoic acid.

The PFAS profiles were different among food categories, with higher proportions of C10–C12 PFCAs in eggs, pork liver, and seafood but more PFOA in cereals and meat (Fig. 1). The concentrations would not be directly relevant to those in foods in our study. Our results may also reflect the overall decline of PFOS in the environment, which was brought about by reducing the production and use by companies such as 3M since 2002 as well as by the fact that PFOS was added to the list of controlled compounds of Stockholm Convention in 2009 [45,49].

C10–C12 PFCAs were the most abundant in eggs (81%), followed by pork liver (71%), shellfish (57%), fish (52%), milk (42%), cereals (36%), and meat (29%). Meat was predominated by PFOA (62%) and contained the least C10–C12 PFCAs. Lau et al. demonstrated that the elimination of PFASs differs between terrestrial species and is not necessarily dependent on the chain lengths [16]; however, the comparisons were with rodents, dogs, and primates, and were limited to C6, C8, and C9 PFASs. Cereals also contained relatively lower proportions of C10–C12 PFCAs than most foods of animal origin; this observation is consistent with the report of Blaine et al., showing that crops accumulate more shorter-chain PFASs than longer-chain PFASs [50]. Liver contained not only high proportions of C10–C12 PFASs but also the highest of the PFDoDA (29%) and PFOS (6.5%) than other food items. This trend was somewhat consistent with a previous study on cattle liver, which contained higher proportions of PFUnDA (10.8%) and PFOS (58.6%) than cattle muscle (PFUnDA was lower than the method detection limit (MDL) and PFOS was 56.6%, respectively) [51].

3.2. Profiles in specific food categories

The studied PFASs, except for PFOS, were found in almost all the samples of rice and flour at GMs ranging from 0.04 ng/g (PFHxA) to 8.90 ng/g (PFOA) and 0.07 ng/g (PFHxA) to 8.84 ng/g (PFOA), respectively (Tables 2 and 3). The concentrations, especially at shorter-chain PFASs, are comparable or even higher than those in foods of animal origin. Rice is one of the staples in Asia, and flour is used to make noodles as well as bread. However, the PFAS contaminations of these two staples have been rarely reported. D’Hollander et al. studied 14 PFASs in Europe and all the analytes were below the LODs in rice, and there were only 0.18 ng/g of PFHxA, <0.01 ng/g of PFOA, and 0.02 ng/g of PFDA in wheat [52]. Haug et al. reported the
concentrations of 10 PFASs in bread of Oslo, Norway, instead of flour, ranged from <0.0015 ng/g (perfluorobutane sulfonic acid) to 0.051 ng/g (PFOA) [29]. Eriksson et al. analyzed eight PFASs in potato and none of them was detected [30]. Pérez et al. reported sub ng/g of total PFASs in cereal mixtures according to the consumption in Brazil, Saudi Arabia, Serbia, and Spain [53]. The above findings would not be applicable to East or Southeast Asian countries.

Fig. 1 – The proportion of each perfluoroalkyl substance (PFAS) by food categories. PFDA: perfluorodecanoic acid, PFDoDA: perfluorododecanoic acid, PFHxA: perfluorohexanoic acid, PFHxS: perfluorohexane sulfonate, PFOA: perfluorooctanoic acid, PFOS: perfluorooctane sulfonate, PFUnDA: perfluoroundecanoic acid.

Table 4 – Daily perfluoroalkyl substance (PFAS) intake (ng/person/day) and the proportion of total PFASs (%) among the general population in Taiwan.

| Food item | PFHxA | PFOA | PFDA | PFUnDA | PFDoDA | PFHxS | PFOS | Total PFASs |
|-----------|-------|------|------|--------|--------|-------|------|-------------|
| Cereals   |       |      |      |        |        |       |      |             |
| Rice      | 145 (6.18) | 1175 (50.0) | 748 (31.8) | 63.4 (2.70) | 214 (9.10) | 5.28 (0.22) | -<sup>b</sup> | 2351 (20.1<sup>c</sup>) |
| Flour     | 131 (9.29) | 870 (61.7) | 194 (13.8) | 10.8 (0.77) | 195 (13.8) | 6.89 (0.49) | - | 1408 (12.0) |
| Meats     |       |      |      |        |        |       |      |             |
| Pork      | 137 (7.79) | 1117 (63.7) | 336 (19.2) | 18.8 (1.07) | 138 (7.85) | 8.35 (0.48) | - | 1755 (15.0) |
| Beef      | 13.3 (6.11) | 139 (63.7) | 43.7 (20.0) | 2.76 (1.26) | 18.2 (8.32) | - | 1.38 (0.63) | 218 (1.86) |
| Chicken   | 97.1 (8.11) | 728 (60.9) | 254 (21.2) | 10.8 (0.90) | 98.7 (8.25) | 7.47 (0.62) | - | 1196 (10.2) |
| Fish      |       |      |      |        |        |       |      |             |
| Salmon    | 54.0 (5.68) | 381 (40.1) | 407 (42.8) | 31.6 (3.32) | 73.3 (7.72) | 3.56 (0.38) | - | 950 (8.12) |
| Grass carp| 51.9 (5.37) | 399 (41.2) | 346 (35.7) | 59.6 (6.15) | 81.4 (8.42) | 3.05 (0.32) | 27.0 (2.79) | 968 (8.27) |
| Seafood   |       |      |      |        |        |       |      |             |
| Oyster    | 13.8 (3.22) | 126 (29.4) | 181 (42.2) | 20.5 (4.77) | 79.2 (18.4) | 1.18 (0.27) | - | 422 (3.60) |
| Shrimp    | 13.2 (4.49) | 114 (38.7) | 96.9 (32.8) | 11.8 (3.99) | 54.3 (18.4) | 3.24 (1.10) | 0.88 (0.30) | 294 (2.51) |
| Clam      | 15.5 (5.48) | 121 (43.1) | 97.2 (34.4) | 11.8 (4.18) | 33.9 (12.0) | 1.62 (0.57) | 0.74 (0.26) | 282 (2.41) |
| Squid     | 4.86 (4.37) | 44.8 (40.2) | 27.5 (24.7) | 2.27 (2.04) | 31.9 (28.7) | - | - | 111 (0.94) |
| Liver     | - | 0.80 (18.9) | 1.67 (39.6) | 0.17 (3.98) | 1.21 (28.6) | - | 0.32 (7.57) | 4.17 (0.036) |
| Egg       | 42.8 (2.61) | 258 (15.8) | 999 (61.1) | 42.3 (2.59) | 292 (17.8) | 2.25 (0.14) | - | 1636 (14.0) |
| Milk      | 1.25 (1.21) | 60.0 (58.3) | 39.2 (38.1) | 2.50 (2.43) | - | - | - | 103 (0.88) |
| All foods | 721 (6.15) | 5534 (47.3) | 3771 (32.2) | 289 (2.47) | 1311 (11.2) | 42.9 (0.37) | 30.3 (0.26) | 11,698 |

PFDA: perfluorodecanoic acid.
PFDoDA: perfluorododecanoic acid.
PFHxA: perfluorohexanoic acid.
PFHxS: perfluorohexane sulfonate.
PFOA: perfluorooctanoic acid.
PFOS: perfluorooctane sulfonate.
PFUnDA: perfluoroundecanoic acid.

<sup>a</sup> The proportion of the compound to the total PFASs in the single food item.
<sup>b</sup> A PFAS in which its detection frequency did not exceed 50% in a food item was not included in the estimation of daily intake.
<sup>c</sup> The proportion of the total PFASs in a food item to those of summed PFASs from all items.
In terms of seafood, PFOA and PFDA were found at higher concentrations (GMs: 7.48–9.67 ng/g and from 5.94 to 12.3 ng/g, respectively) than the other three PFCAs (low ng/g levels, Table 2). Tittlemier et al. also reported higher concentrations of PFDA in seafood than longer-chain PFUnDA and PFDoDA, although they did not find PFOA [38]. On the other hand, the levels of PFOA and PFDA in our study were six to 30 times lower than those previously reported for tilapia and oysters in a polluted wastewater area in Taiwan [54].

The concentrations of PFASs found in pork liver were higher than those in any other food item (except for PFOA) (Tables 2 and 3). This finding is consistent with other reports on the liver of cattle and aquatic animals [41,55]. In Taiwan, pork liver is occasionally consumed by the general population, but is consumed much more often by pre- and postnatal women as a nutritious supplement. The high amount of PFASs in pork liver may pose health risks to pregnant women and there are several studies have suggested that PFASs could be transmitted to their offspring through cord blood and breast milk [56–58].

3.3. Intake from foods

The estimated total intake of PFASs was 11.7 μg/person/day in the general population in Taiwan based on the studied foods (Table 4). The predominant compounds were PFOA (5.5 μg/person/day, 47.3% of the total PFASs) and PFDA (3.8 μg/person/day, 32.2%). People there ingest most of the PFASs from rice (2.4 μg/person/day, 20.1%), pork (1.8 μg/person/day, 15.0%), and eggs (1.6 μg/person/day, 14.0%, Table 4). The major contributed food categories were cereals (32%), meats (27%), and fish (16%). The total daily intake for PFOA and PFDA in this study was 179 and 290 times higher, respectively, than those reported by Haug et al. for the general Norwegian population [29]. In our study, the daily ingestion of PFOA from meats (1.98 μg/person/day) was comparable to that from cereals (2.05 μg/person/day). This finding is inconsistent with those of Vestergren et al., which PFOA intake could be dominated by large consumption of cereals, vegetables, and dairy products rather than meats and fish products [31]. This difference may result from a relatively lower ratio of PFOA from cereals to that of meats and fish products.

| Compound | Country | Daily intake (ng/kg body weight/day) | Reference |
|----------|---------|------------------------------------|-----------|
| PFHxA    | Taiwan (this study) | 11.2 | Fromme et al. (2007) [21] |
|          | Germany | 4.30 | Zhang et al. (2010) [61] |
|          | Korea   | 85.1 | Heo et al. (2014) [62] |
|          | Germany | 8.63 | Fromme et al. (2007) [21] |
|          | UK      | 70.0 | U.K. Food Standards Agency, (2006) [44] |
|          | North America and Europe | 1–130 | Trudel et al. (2008) [23] |
|          | Canada  | 0.10–0.50 | Ostertag et al. (2009) [42] |
|          | Canada  | 0.20–0.40 | Ostertag et al. (2009) [60] |
|          | Norway  | 0.60 | Haug et al. (2010) [63] |
|          | Italy   | 5.74 (for men) | Renzi et al. (2013) [45] |
|          |         | 4.10 (for women) |         |
| PFDA     | Taiwan (this study) | 44.2 | Ostertag et al. (2009) [60] |
|          | Canada  | 0.10–0.30 |         |
| PFUnDA   | Taiwan (this study) | 4.45 | Haug et al. (2010) [63] |
|          | Norway  | 0.34 |         |
| PFHxS    | Taiwan (this study) | 0.65 | Fromme et al. (2007) [21] |
|          | Germany | 2.00 |         |
|          | China   | 1.19 | Zhang et al. (2010) [61] |
|          | Korea   | 0.60–3.03 | Heo et al. (2014) [62] |
|          | Germany | 1.40 | Fromme et al. (2007) [21] |
|          | UK      | 100 | U.K. Food Standards Agency, (2006) [44] |
|          | Spain   | 1.07 | Ericson et al. (2008) [37] |
|          | North America and Europe | 3–220 | Tittlemier et al. (2008) [38] |
|          | Canada  | 4.00 | Ostertag et al. (2009) [42] |
|          | Canada  | 0.20–2.40 | Ostertag et al. (2009) [60] |
|          | Canada  | 0.10–0.20 (1990) |         |
|          |         | 0.80–2.00 (1998–2004) |         |
|          | Norway  | 1.50 | Haug et al. (2010) [63] |
|          | Italy   | 5.71 (for men) | Renzi et al. (2013) [45] |
|          |         | 4.08 (for women) |         |

PFDA: perfluorodecanoic acid.  
PFHxA: perfluorohexanoic acid.  
PFHxS: perfluorohexane sulfonate.  
PFOA: perfluorooctanoic acid.  
PFOS: perfluorooctane sulfonate.  
PFUnDA: perfluoroundecanoic acid.
The exposure to PFASs among the general population in Taiwan ranged from 0.46 ng/kg b.w./day for PFOA to 85.1 ng/kg b.w./day for PFOS (Table 5). The exposure to PFHxS and PFOS in Taiwan ranged from 0.46 ng/kg b.w./day for PFOS to 85.1 ng/kg b.w./day. The prevalence of PFHxS and PFOS in Taiwan was similar to ours, but are dissimilar to other reports. However, the current study revealed that people in Taiwan are exposed to much higher levels of PFASs (Table 5), although they consume a comparable amount of meat to those in European Union and North America [21,23,29,37,38,42,44,45,60–63]. The estimated daily intake of PFOA in China (9.83 ng/kg b.w./day) and in Korea (0.17–1.68 ng/kg b.w./day) did not include cereals; excluding the sources of rice and flour, the exposure to PFOA in Taiwan (53.7 ng/kg b.w./day) remained five to 300 times higher than that in China and Korea [61,62]. The higher exposure might be mainly attributed to the higher concentrations of PFASs in foods found in the current study than those in previous reports. Moreover, the estimated average daily intake of PFOA in this study was only 18 times lower than the tolerable daily intake (TDI) suggested by European Union and 35 times lower than that suggested by the U.K. (1500 and 3000 ng/kg b.w./day, respectively) [44,64]. Because the high exposure group may exceed these TDIs, the exposure of the Taiwanese to PFOA deserves further attention.

Regarding pregnant women in Taiwan, PFOA was the highest exposed PFASs; the upper limit of 95% probability from each food item ranged from 69.0 (eggs) to 2517 ng/person/day (rice), in a total of 4390 ng/person/day (Table 6). The next was PFDA, which ranged from 104 (squid) to 1604 ng/person/day (rice), in a total of 3502 ng/person/day. The 95% upper limit of total PFASs from liver for pregnant women (740 ng/person/day in total) were at least two orders of magnitude higher than that of the general population (4.17 ng/person/day in total). Liver was a major source of PFOS consumed by the pregnant women. It also contributed significantly to the exposure to other PFASs (Tables 4 and 6). The upper limits of PFOS of 25% and 50% probability of exposure for pregnant women were 1671 ng/person/day and 2634 ng/person/day, respectively (Fig. 2), indicating that these women had at least 75-percent probability of being exposed to PFOS at an amount of 1.7 μg/person/day and 50-percent likelihood of being exposed to ≥2.5 μg/person/day or approximately 40 ng/kg b.w./day.

Levels of PFOS in the serum of women in Taiwan have been similar from 2001 to present, but those of PFOS decreased gradually. Wang et al. reported median concentrations of 12.7 ng/mL for PFOS and 2.39 ng/mL for PFOA in the serum of pregnant women collected in 2001 [65]. The serum GM of PFOS and PFOA in the women aged 20–30 in 2006–2008 were 8.21 ng/mL and 2.52 ng/mL, respectively [66]. In the serum of Taiwan females collected in 2011, the PFOS levels further decreased (median 6.01 ng/mL) but the PFOA levels remained unchanged (median 2.48 ng/mL) [67]. The gradual drop of PFOS in serum would indicate reduced exposure of Taiwan women to PFOS in the past years. Although the PFOS concentrations in the serum collected in 2011 were still higher than those of PFOA, this may result from previous long-term exposure and the long half-life of PFOS (5.4 years) in humans [16]. Given the overriding PFOA intake and the overall PFAS exposure found in this study, more studies are needed to develop dietary

| Food item | PFFxS | PFOA | PFDA | PFUnDA | PFDoDA | PFHxS | PFOS |
|-----------|-------|------|------|--------|--------|-------|------|
| Rice      | 311   | 2517 | 1604 | 136    | 458    | 11.3  | —    |
| Flour     | 157   | 1045 | 233  | 7.09   | 234    | 8.27  | —    |
| Pork      | 95.6  | 780  | 235  | 13.1   | 96.3   | 3.65  | —    |
| Beef      | 58.0  | 605  | 190  | 12.0   | 78.9   | —     | 6.00 |
| Chicken   | 68.2  | 511  | 178  | 6.41   | 69.3   | 5.24  | —    |
| Ocean fish| 50.1  | 353  | 377  | 29.3   | 68.1   | 2.36  | —    |
| Fresh fish| 29.8  | 228  | 198  | 34.1   | 46.7   | 0.58  | 15.5 |
| Oyster    | 42.2  | 384  | 552  | 62.4   | 241    | 3.59  | —    |
| Shrimp    | 40.3  | 347  | 294  | 35.9   | 165    | 9.86  | 2.69 |
| Clam      | 47.1  | 370  | 296  | 35.9   | 103    | 4.04  | 2.24 |
| Squid     | 18.5  | 170  | 104  | 37.8   | 121    | —     | —    |
| Liver     | —     | 142  | 297  | 29.9   | 214    | —     | 56.9 |
| Egg       | 11.4  | 69   | 267  | 11.3   | 78.1   | 0.60  | —    |
| Milk      | 8.97  | 431  | 281  | 17.9   | —      | —     | —    |
| Total     | 938   | 4390 | 3502 | 469    | 1973   | 49.5  | 83.3 |

PFDA: perfluorodecanoic acid.
PFDoDA: perfluorododecanoic acid.
PFFxS: perfluorohexane sulfonate.
PFOA: perfluorooctanoic acid.
PFOS: perfluorooctanoic acid.
PFUnDA: perfluoroundecanoic acid.

a A PFAS which its detection frequency did not exceed 50% in a food item was not included in the estimation of daily intake.
guidelines to pregnant women to reduce risks of vertical transmission of these compounds and their potential adverse development effects [20,57,68].

4. Conclusion

This study measured five PFCAs and two PFASs in fourteen of the most often consumed foods in Taiwan using UPLC-MS/MS, and found PFHxA, PFOA, PFDA, PFUnDA, and PFDoDA to be ubiquitous in most of the foods. These levels were much higher than those of most previous reports, especially in the long-chain PFCAs in foods of animal origin. This study observed considerable PFAS concentrations in the matrices of rice and pork liver, which are seldom studied in western countries, but rice is a major staple in Asia and pork liver is a specialty food commonly consumed by pregnant and postnatal women in Taiwan. This is the first daily intake estimation of these PFASs in foods including staple for East and Southeast Asia, and the findings differ from those reports from western countries with varied ingestion amounts and profiles of PFASs in foods.

Conflicts of interest statement

The authors declare no conflicts of interest.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.jfda.2017.12.011.

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