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Perfluorinated compounds in milk, milk powder and yoghurt purchased from markets in China

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The exposure to perfluorinated compounds (PFCs) through the diet for humans is of great concern. Among the diet, dairy products are in great demand. This paper reports the study on the levels of 9 perfluorinated acids and 2 fluorotelomer acids in milk, milk powder and yoghurt purchased from Chinese markets from 2008 to 2009. The analytes were quantitated by high performance liquid chromatography/electrospray tandem mass spectrometry (HPLC-ESI-MS/MS). In milk samples, perfluoroheptanoic acid (PFHpA) and perfluorononanoic acid (PFNA) were detected frequently, both in 68% of samples. In milk powder samples, perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA) and PFNA were the only detected PFCs and none of them was observed in more than 35% of samples. In yoghurt, PFOA was the most frequently detected compound, found in 69% of samples. None of fluorotelomer acids was observed in any samples. The mean concentrations of total PFCs were 178 pg/g (wet weight) in milk, 98 pg/g (dry weight) in milk powder and 42 pg/g in yoghurt (wet weight). It is notable that the data of this study indicate significant differences (P<0.001) among three kinds of packaging of milk in the concentration of total PFCs. A preliminary human health risk assessment of milk and dairy products consumption was conducted in this study. For adults, the mean daily intake of PFOS and total PFCs was equal to or lower than 23 and 167 pg kg^{-1} d^{-1} (body weight), respectively.

Perfluorinated compounds, milk, milk powder, yoghurt

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Perfluorinated compounds (PFCs) are a class of synthetic perfluorochemicals that serves as a kind of surfactants used in a variety of consumer and industrial applications for more than 50 years due to their oleophobic and hydrophobic properties [1]. In May 2009, 9 new chemicals including perfluorooctane sulfonate (PFOS) were listed under the Stockholm Convention as persistent organic pollutants (POPs) in Geneva. PFCs have generated notable interest because of their high persistence, biomagnification and potential toxicity [2,3]. PFCs are widely distributed in the environment and have been detected in water, soil, air and animals [2−5]. In recent years, the concentrations of PFCs have been extensively measured and studied for human exposure and risk evaluation. Various PFCs have been detected in human blood and milk from North America [6,7], Europe [8,9] and Asia [10,11]. A study shows that both serum albumin proteins and β-lactoglobulin proteins had a strong binding affinity for some PFCs [12]. Moreover, a strong association was found between concentrations of PFOS in maternal serum and milk collected from 12 primiparous women in Sweden [8].

So far, a great number of studies focus on the sources of human exposure to PFCs. There was a report indicating that food, drinking water, air and indoor dust were potential sources of human exposure to PFCs [13]. Generally, dietary ingestion is considered as the main route of exposure to PFCs even though there is no sufficient evidence [13,14]. In recent years, the levels of PFCs in a series of foodstuffs have been investigated in several countries such as the United Kingdom [15], Canada [13,16], Germany [9] and

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Spain [14]. Halldorsson et al. [6] investigated the correlation between dietary variables and plasma levels of PFOS among 1076 pregnant women. The results show that the PFOS level has positive correlation with intakes of red meat, milk, animal fat, potatoes and snacks without the adjustment analyses for parity, smoking, maternal age, prepregnancy body mass index and socio-occupational status. Meanwhile, studies have been launched to evaluate human exposure from dietary sources to PFCs. For an adult, the range of average intake level of PFOS is from 1.07 (Spain) [14] to 100 (the United Kingdom) [15] ng kg^{-1} d^{-1} (body weight).

A recent study has shown that high levels of PFOS (45.0–86.9 ng/g wet weight) were detected in chicken eggs in China [17]. Nevertheless, limited data are available to estimate dietary exposure of Chinese to PFCs. Milk and dairy products as a class of basilic foods are consumed by humans. Recently, EPA found record PFOS, PFOA levels in Alabama grazing fields where the sludge had been applied to grasslands used for grazing animals for over 10 years [18]. Cows may pick up PFCs from the sludge when they graze. Thus, milk and dairy products are one possible source of PFOS, because PFOS can bind to β-lactoglobulin proteins [12]. Moreover, PFCs have been found in milk or dairy products in several countries [14,16]. However, to our knowledge, there is no report yet on the occurrences of PFCs in milk and dairy products from Chinese markets.

The purpose of the present study was to investigate the levels of 9 perfluorinated acids and 2 fluorotelomer acids in a variety of milk, milk powder and yoghurt purchased from Chinese markets and to provide these data for a preliminary risk assessment of Chinese exposure through consuming these products.

1 Materials and methods

1.1 Samples

From August 2008 to February 2009, cow milk (n=84), cow milk powder (n=36) and yoghurt (n=32) samples were purchased randomly from markets in three cities (Beijing, Tianjin and Wuhan) of China. Most of them were not necessarily produced in local places, especially for the samples from Beijing. These samples are comprehensive and representative, almost containing all kinds of products people can acquire in local markets, supermarkets and grocery stores. There are 11, 26 and 5 company brands for milk, milk powder and yoghurt samples, respectively. Over 80 percent of milk and yoghurt samples were made by 4 companies, and the liquid milk production of the 4 companies accounted for nearly 60% of the total in China. Milk samples are divided into three groups on the basis of packaging: Bailey (polyethylene; shelf life: 30 d), Tetra Fino Aseptic (laminate of paper, polyethylene and aluminium foil; shelf life: 30 d) and Tetra Brik Aseptic (laminate of paper, polyethylene and aluminium foil; shelf life: 6–8 months). All the samples are kept in their original packaging at 4°C and analyzed in their shelf lives.

1.2 Chemicals

Perfluoroheptanoic acid (PFHpA, >98%), perfluorooctanoic acid (PFOA, 95%), perfluorononanoic acid (PFNA, 97%), perfluorodecanoic acid (PFDA, 97%), perfluorododecanoic acid (PFDoDA, 96%) and perfluorotetradecanoic acid (PFTA, 96%), 2-Perfluorooctyl ethanoic acid (FOEA, >98%), and 2H-perfluoro-2-decenoic acid (FOUEA, >98%) were purchased from Alfa Aesar (Lancashire, England). Perfluoroundecanoic acid (PFUnDA, 95%), potassium salts of perfluorohexane sulfonate (PFHxS, ⩾98%) and perfluorooctane sulfonate (PFOS, ⩾98%) were obtained from Sigma-Aldrich (Oakville, ON, Canada). Sodium perfluoro-1-[1,2,3,4-$^{13}C_4$]octanesulfonate (MPFOS, ⩾98%) and perfluoro-n-[1,2,3,4-$^{13}C_4$] octanoic acid (MPFOA, ⩾98%) were purchased from Wellington Laboratories (Canada) as the internal standards.

Methanol (HPLC grade) was obtained from Fisher Scientific (Pittsburgh, PA, USA). Ammonium acetate (NH_4OAc, >97%) and ammonium hydroxide (analytical grade; v/v, 50%) were purchased from Alfa Aesar. All water (>18.2 MΩ/cm) used in the method was prepared with Milli-Q Advantage A10 system (Millipore, USA).

1.3 Sample extraction and purification

The extraction method described by Ericson [13] was used, with some modifications. For milk and yoghurt samples, a 1-g aliquot was taken and placed in a polypropylene (PP) centrifuge tube, and precleaned with MeOH. Internal standards (2 ng MPFOS and 2 ng MPFOA) were added into the tube, followed by 4 mL MeOH. Samples were mixed well on vortex before sonicated for half an hour. After that, they were centrifuged for 15 min at 3000 r/min. The supernatant was transferred into precleaned PP tube and the extraction was repeated with another 4 mL MeOH. For milk powder, 0.5 g sample was extracted in sequence with 8 mL Methanol and 8 mL 0.01 mol/L HCl in Methanol and other procedures were similar to the above. Supernatants were combined and concentrated to 1 mL with a gentle stream of N_2. The sample was diluted with 40 mL water in a PP container and loaded onto Waters Oasis® WAX single-use cartridge (6 cc/150 mg) previously conditioned with 4 mL MeOH and 4 mL water. Vacuum was used to speed up the concentrations of water samples. SPE cartridge was washed with 4 mL acetate buffer solution (pH 4) and 4 mL MeOH and eluted with 2% NH_4OH in MeOH (target fraction). The eluate was concentrated to 1 mL under nitrogen for injection. The flow rate of all steps was controlled at 1 drop/s. Blanks of MeOH (HPLC grade), treated as real samples in all steps, were processed with each batch.

1.4 Instrumental analysis

Analysis of 9 perfluorinated acids and 2 fluorotelomer acids was performed using a high performance liquid chromatography (P680 pump, Ultimate 3000 autosampler, Dionex, US)/electrospray ionization tandem mass spectrometry (API 3200; Applied Biosystems/MDS SCIEX, US). Experimental conditions of electrospray tandem mass spectrometry are shown in Table 1. A 10 μL aliquot was injected into Dionex Acclaim 120 C18 column (5 μm, 4.6 mm i.d.×150 mm length; Dionex, Sunnyvale, CA, USA). Methanol (A) and 50 mmol/L NH_4OAc (B) were delivered at a flow rate of 1.0 mL/min. For milk and milk powder samples, the 10 min program was chosen for separation. However, there was matrix interference for yoghurt samples, so the 15 min program was chosen to improve the separation. The gradient started at 28% B followed by a 4 min ramp to 5% B, and returned to initial condition after holding for 3 min (6 min for yoghurt samples), and was kept at the level until 10 min (15 min).

1.5 Quality assurance and data analysis

Quantification was performed using the internal standard method. A standard calibration composed of ten points of serial concentrations (0.01–50 μg/L) was performed using a $1/x^2$ weighted regression. MPFOS and MPFOA were used as internal standards. The limits of detection (LOD) were calculated as three standard deviations from the average concentration in the extraction blanks. LOD were in a range of 0.002–0.031 μg/L (Table 1). Correlation coefficient is greater than 0.99 for each analyte. Recoveries of 11 spiked PFCs (2 ng each of standards) ranged from (84±9)% to (113±4)% in milk, from (81±10)% to (111±4)% in milk powder and from (80±8)% to (118±5)% in yoghurt. The standard calibration was conducted before each batch of analysis and standards were run as the quality control sample, which were measured after every 10 injections to check for instrumental drift. If the standards were not within ±20% of its initial value, a new calibration curve would be constructed.

To minimize the background, all potential sources of instrumental and procedural contamination were eliminated. Polytetrafluorethylene (PTFE) materials had been removed from laboratory equipments. All the containers employed during the sample preparation and analysis procedure were washed thoroughly with methanol. One method blank sample was analyzed with every batch of eight samples. All analytes detected in the method blanks were below the LOD.

Nondetectable levels were treated as zero. Data were analyzed using the statistical software SPSS 13.0 (SPSS Inc., 2005). Kruskal-Wallis *H* test was applied to examine the relationship between the packaging of milk and PFC concentrations.

2 Results and discussion

2.1 PFC concentrations in milk, milk powder and yoghurt

PFHpA, PFOA and PFNA were the predominant compounds found in all milk samples. PFHpA and PFNA were both detected in 68% of the milk samples, followed by PFOA in 46% (Table 2). PFOS was only observed in 36% of the milk samples, at a mean concentration of 24 pg/g. PFDA, PFUnDA and PFTA were found occasionally. In milk powder samples, only three PFCs (PFOS, PFOA and PFNA) were found, and their detection frequencies were lower than 35%. In yoghurt samples, PFOA was the most

Table1 Experimental conditions of electrospray tandem mass spectrometry

Analytes	Parent ion Q1 (*m/z*)	Daughter ion Q3 (*m/z*)	Time *T*(ms)	Declustering potential *V*(V)	Entrance potential *V*(V)	Collision cell entrance potential *V*(V)	Collision energy *E*(eV)	Collision cell exit potential *V*(V)	Internal mark	LOD (pg/g)
PFHxS	398.7	79.9	50	−70	−5.5	−14.93	−67	−10	MPFOS	2
PFHpA	362.9	319	50	−24	−3.0	−12.61	−12	−12	MPFOA	13
PFOA	412.8	369	50	−22	−3.5	−35.45	−22	−10	MPFOA	18
PFOS	498.8	79.9	50	−80	−8.0	−20.64	−80	−10	MPFOS	5
PFNA	462.8	419	50	−27	−3.5	−15.30	−12	−16	MPFOA	27
PFDA	512.8	469	50	−30	−4.0	−15.15	−15	−11	MPFOA	15
PFUnDA	562.9	519	50	−32	−4.0	−16.00	−14	−12	MPFOA	15
PFDoDA	612.9	569	50	−20	−5.0	−30.86	−25	−15	MPFOA	21
PFTA	712.9	669	50	−30	−4.5	−21.56	−17	−15	MPFOA	31
FOEA	476.9	393	50	−20	−3.0	−16.83	−19	−10	MPFOA	11
FOUEA	456.9	393	50	−28	−3.0	−15.09	−18	−10	MPFOA	7
MPFOS	502.9	79.9	50	−80	−8.5	−19.79	−90	−10		
MPFOA	416.8	372	50	−23	−3.5	−35.60	−21	−15		

Table 2 Summary of PFCs concentrations (pg/g) in milk, milk powder and yoghurt samples from Chinese Markets [a)]

	PFHpA	PFOA	PFOS	PFNA	PFDA	PFUnDA	PFTA	Total PFCs
milk, *n*=84								
Range	<13–312	<18–178	<5–695	<27–476	<15–44	<15–40	<31–144	
Mean	54	26	24	67	7			178
Median	44			52				
Samples above LOD (%)	68	46	36	68	21	1	5	
milk powder, *n*=36								
Range	<26	<36–482	<10–175	<54–192	<30	<30	<62	
Mean		46	22	30				98
Median								
Samples above LOD (%)	0	33	25	22	0	0	0	
yoghurt, *n*=32								
Range	<13–106	<18–229	<5–32	<27–256	<15–100	<15–21	<31–34	
Mean	7	32	3					42
Median		28						
Samples above LOD (%)	13	69	13	6	3	3	3	

a) Mean values were not calculated when more than 90% of the samples had values below LOD. PFHxS, PFDoDA, FOEA and FOUEA were not found in any of the samples above LOD. All values were not corrected for recoveries of PFCs.

predominant compound and the detection frequency was 69% with concentrations ranging from below LOD to 229 pg/g. Other PFCs were detected in only a few yoghurt samples. Fluorotelomer acids were not found in any samples. The mean concentrations of total PFCs were 178 pg/g (wet weight) in milk, 98 pg/g (dry weight) in milk powder and 42 pg/g in yoghurt (wet weight).

The PFC levels found in the milk and dairy products are similar to or somewhat higher than those found in several previous studies [10,14,16]. Ericson [14] determined PFC levels in 36 composite samples from the Catalan (Spain) Market. Only 3 PFCs (PFOS, PFOA and PFHpA) were detectable. The total concentrations of PFCs were 71 pg/mL in whole milk and 121 pg/mL in dairy products composed of cheese, yoghurt, cream caramel and custard. PFOS, PFOA and related fluorinated chemicals were analyzed in composite samples of food groups from the 2004 Total Diet Study (TDS) by the U.K. Food Standard Agency [15]. PFOS and PFOA were not found at a level above LOD in milk (both 0.5 ng/g of fresh weight) and dairy products (both 5 ng/g of fresh weight). Another study showed that PFCs were neither observed in the formula based on milk from the United States except a few samples [10]. There was a study investigating the levels of perfluorooctanesulfonamides in food composites collected over 12 years in Canada [17]. The average concentrations of N-methylperfluorooctanesulfonamide and N-ethylperfluorooctanesulfonamide in dairy products were 68 and 31 pg/g (wet weight), respectively.

2.2 Composition profiles of PFCs

The composition profiles of relative levels of PFHpA, PFOA, PFOS, PFNA and PFDA using mean concentration values are shown in Figure 1. In milk samples, PFHpA and PFNA contributed up to 29% and 37%, respectively, of the total composition, while ∑PFOA+PFOS only accounted for 27%. PFDA was a small contributor of PFCs in milk and yoghurt and it was not detected in milk powder. The proportions of PFOA in both milk powder and yoghurt were higher than that in milk. In whole milk from Spain, PFOS was not detectable [14]. The proportion of PFHpA was lower than PFOA in whole milk from Spain, which was in agreement with that in yoghurt but different from that in milk from China.

2.3 Correlation between the PFC concentrations and the packaging of milk

No significant differences in the levels of PFCs were among various company brands. In addition, there were many kinds (such as whole and skimmed milk) and tastes (such as chocolate and fruits) in the milk and yoghurt samples and no difference in concentrations of these samples was observed. However, significant differences among three kinds of packaging of milk in the concentrations of PFHpA, PFNA and total PFC were found and the values of *P* (Kruskal-Wallis H) were 0.002, 0.001 and <0.001, respectively. The PFC levels in milk for three different packaging are shown in Figure 2. Among these packaging, the levels of PFCs in milk packaged with Bailey are notable higher than the levels with other two packaging. The total PFC

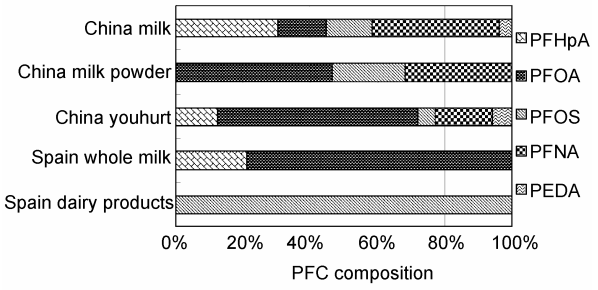

Figure 1 Composition profile of five PFCs in milk (data from this study and Ref. [14]).

concentrations in some samples exceed 600 pg/g. PFC levels in milk with Tetra Fino Aseptic are similar to the levels with Tetra Brik Aseptic. The total PFC concentrations in all the samples with Tetra Fino Aseptic and Tetra Brik Aseptic are below 300 pg/g. In addition, PFOS levels in milk of the two packaging are close to zero.

There were some reports [13,16,19] suggesting that food packaging might serve as a source of PFCs, used as repellents of water and grease, in food. Therefore, we consider that the difference of PFCs concentration may be related to the milk packaging or processing. Begley et al. [19] demonstrated that perfluorochemicals and polyfluoroalkyl phosphoric acids (PAPs) would migrate into food simulants from food-contact paper. PFOA migrated from a microwave popcorn bag into oil at a concentration as high as 300 ng/g. PAPs are a class of commercial fluorinated surfactants and used in human food-contact paper products. The occurrences of PAPs in human serum indicated that food-contact materials may be a source of PAPs that will migrate to food and be consumed [20,21]. However, in another study reported by Bradley et al. [22], it was noted that the coating materials of cookware products containing polytetrafluoroethylene (PTFE) were not considered as significant sources of PFCs, because the levels of PFCs in food were too low to be detected. With regard to it, Jogsten et al. [23] also investtigated the influence of food packaging on the concentrations of PFCs and indicated it was uncertain whether some foods packaging could contribute to an exposure to PFCs. Therefore, further research needs to be carried out to verify which types of food packaging are correlated with the concentrations of PFCs in food.

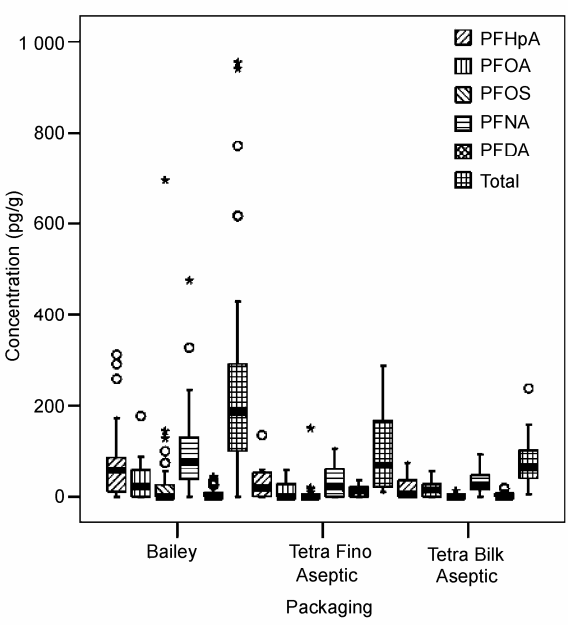

Figure 2 Box plot of concentrations of PFHpA, PFOA, PFOS, PFNA, PFDA and total PFC in milk on the basis of different packaging. The data indicate significant differences (P<0.001) among three kinds of packaging of milk in the concentration of total PFCs.

2.4 Daily intake (DI) of PFCs through milk and dairy products

According to a recent study [24], the food composition was investigated in 12 provinces of China in 2007. In that study, milk and milk products including cow milk, cow milk powder, yoghurt, and sheep milk were mentioned as one of the 13 food groups. The mean consumption was 59.2 g/d for an adult male, 18–45 years of age and 63 kg of body weight. On the basis of this value, we estimated the DI of PFCs through the consumption of milk and dairy products. If milk is supposed to be the only contributor of the consumption (Not detected =0), the mean DI of PFOS and total PFCs are 23 and 167 pg kg^{-1} d^{-1}, respectively. If the milk powder and yoghurt are taken into account, the intake values of total PFC will get lower.

In the United States, the maximum DI of PFOS via infant formula and dairy milk were estimated to be 1.4 and 0.7 ng kg^{-1} d^{-1} [10], respectively. In several other Asian countries, the average DI of PFOS and total PFCs (sum of PFOS, PFOA, PFHxS, PFBS, PFHpA and PFNA) by infants through breastfeeding were 11.8 and 18.2 ng kg^{-1} d^{-1}, respectively [10]. Among these countries, the average DI of total PFCs in Japan was the highest (40.5 ng kg^{-1} d^{-1}), and the lowest was in India (11.0 ng kg^{-1} d^{-1}). Another study assessed the health risks in infants associated with exposure to PFCs via the consumption of mothers' breast milk from Zhoushan, China [11]. The mean DI (n=19) of PFOS to the child was 10 ng kg^{-1} d^{-1}.

In 2006, provisional tolerable daily intake (TDI) of 300 and 3000 ng kg^{-1} d^{-1} for PFOA and PFOS, respectively, were recommended by the U.K. Food Standards Agency Committee on Toxicology (FSA), on the basis of a thorough review of currently toxicity studies [15]. In addition, the German Federal Institute for Risk Assessment suggested a TDI of 100 ng kg^{-1} d^{-1} for both PFOA and PFOS for the whole population, applying toxicity an uncertainty factor of 1000 to a no-observed-adverse-effect-level (NOAEL) value from a two-generation reproductive toxicity study in rats [25]. In 2008, European Food Safety Authority (EFSA) established a TDI of 150 ng kg^{-1} d^{-1} for PFOS and 1,500 ng kg^{-1} d^{-1} for PFOA [26]. In our study, the calculated DI of PFCs for adults or infants via milk and dairy products from China is far below the values recommended. Therefore, there is little potential risk of PFCs for Chinese through consuming these products.

3 Conclusions

In summary, this is the first study on the occurrence of PFCs in milk, milk powder and yoghurt purchased from

Chinese markets. PFHpA, PFOA, PFOS, PFNA, PFDA, PFUnDA and PFTA are found in these products. The PFC levels in this study are comparable to or somewhat higher than those in previous studies in Spain and the U.S.A. A pilot risk assessment of exposure to PFCs based on the consumption of milk and dairy products is evaluated. The findings indicate that Chinese are exposed to relatively low concentrations of PFCs via consumption of these products. However, further studies should be carried out to determine the levels in other foodstuffs. Moreover, the routes of PFCs' entry into food need to be investigated.

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