Perfluorooctane sulfonate (PFOS) and other fluorochemicals in viscera and muscle of farmed pigs and chickens in Beijing, China

WANG JieMing, SHI YaLi, PAN YuanYuan & CAI YaQi*

State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China

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Eleven perfluorinated compounds (PFCs) were determined in tissue samples from farmed pigs and chickens ($n=143$) that were purchased in Beijing in March 2009. The total PFCs mean concentration was the highest in pig liver (3.438 ng g^{-1} wet weight (ww)), followed by pig kidney (0.508 ng g^{-1} (ww)), pig heart (0.167 ng g^{-1} (ww)), chicken liver (0.098 ng g^{-1} (ww)), chicken heart (0.050 ng g^{-1} (ww)), pork loin (0.018 ng g^{-1} (ww)), and chicken breast (0.012 ng g^{-1} (ww)). Among the PFCs analyzed, perfluorooctane sulfonate (PFOS) was the predominant compound in viscera and muscle of farmed pigs and chickens. In addition, a strong linear correlation ($r = 0.932$) was observed between the concentrations of PFOS and perfluoroundecanoic acid (PFUnDA) in pig liver. Our preliminary conclusion is that there is little potential risk of exposure to PFCs via the consumption of these products in Beijing.

perfluorinated compounds, viscera, muscle, dietary intake

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Perfluorinated compounds (PFCs) are a group of manmade fluorinated organic compounds, which are used extensively in food packaging, textile coatings, lubricants, surfactants, fire-fighting foams, and nonstick coatings for cookware [1,2]. Concern about the environmental persistence and potential toxicity of PFCs has grown over the past several decades [2]. Consequently, research has increasingly focused on PFCs and their effects on environmental pollution, toxicology and food safety. These compounds are widely distributed in air, dust, water, soil, wild animals, and humans [3–7]. In 2000, the main manufacturer of perfluorooctane sulfonate (PFOS) in the United States, 3M, voluntarily phased-out its production. However, PFOS is still produced in China and annual production has increased in recent years [2]. In May 2009, the Conference of the Parties 4 (COP-4) of the Stockholm Convention classified PFOS and perfluorooctane sulfonyl fluoride as persistent

organic pollutants.

PFOS can bind to proteins [8] and is mainly distributed in liver, blood serum, kidney, heart and muscle [9]. PFOS can bioaccumulate and biomagnify through the food chain [10–12]. The environmental distribution of a suite of PFCs, including PFOS and perfluorocarboxylates, has been investigated in the food web of the bottlenose dolphin [11]. Trophic magnification factors for PFOS and some perfluorocarboxylates indicated biomagnification occurred in the marine food web. Another study indicated that accumulation of PFOS was comparable with that of short and medium chained fatty acids [13].

In recent years, the occurrence of PFCs in human serum has attracted much attention [14,15]. Research has also increasingly focused on the routes of human exposure, including food, water, air, and indoor dust [16–22]. Tittlemier et al. [18] showed that humans are mainly exposed to PFOS through food consumption, which accounts for 60% of exposure. Furthermore, the levels of PFCs in meat products

*Corresponding author (email: caiyaqi@rcees.ac.cn)

were found to be relatively high among all the foodstuffs analyzed. High levels of PFOS have also been found in the liver of both wild and farmed animals in Japan [23]. While PFCs have been detected in various meat products from some countries [18,20,24], very little information is available on the PFCs in these products from China.

Beijing is the political and cultural capital of China and a center of international activity. By 2008, the population of Beijing had exceeded 20 million. Pork and chicken are the main meats consumed by Chinese people and account for up to 88% of meat consumption among pork, chicken, beef, and mutton [25]. The aim of the present study was to investigate the levels of PFOS and 10 other fluorochemicals in meat products (farmed pigs and chickens) readily available in Beijing. The data could be used for a pilot exposure assessment of consumption of these products.

1 Materials and methods

1.1 Samples

In March 2009, pig hearts, pig livers, pig kidneys, pork loin, chicken hearts, chicken livers and chicken breast samples were purchased randomly from 14 markets in eight districts in downtown Beijing, China (Figure S1). All the samples were kept at -20°C in their original packages until analysis.

1.2 Chemicals

Perfluoroheptanoic acid (PFHpA, >98%), perfluorooctanoic acid (PFOA, 95%), perfluorononanoic acid (PFNA, 97%), perfluorodecanoic acid (PFDA, 97%), perfluorododecanoic acid (PFDoDA, 96%), 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, $\geq 98\%$) and PFOS ($\geq 98\%$) were obtained from Sigma-Aldrich (Oakville, ON, Canada). Sodium perfluoro-1-[1,2,3,4- $^{13}\text{C}_4$] octanesulfonate (MPFOS, $\geq 98\%$) and perfluoro-n-[1,2,3,4- $^{13}\text{C}_4$] octanoic acid (MPFOA, $\geq 98\%$) were purchased from Wellington Laboratories (Canada) for use as internal standards.

Tetra-*n*-butylammonium hydrogen sulfate (TBA) was purchased from Acros Organics (Morris Plains, NJ, USA). Methyl *tert*-butyl ether (MTBE) was purchased from J. T. Baker (Phillipsburg, NJ, USA). Methanol (HPLC grade) was obtained from Fisher Scientific (Pittsburgh, PA, USA). Ammonium acetate (NH_4OAc , >97%) and ammonium hydroxide (analytical grade; 50%, v/v) were purchased from Alfa Aesar (Ward Hill, MA, USA). Potassium hydroxide (super grade, >98%) and sodium carbonate (super grade, $\geq 99.8\%$) were obtained from the Beijing Chemical Reagent Factory (China).

All water ($>18.2\text{ M}\Omega\text{ cm}^{-1}$) used in this study was pre-

pared with a Milli-Q Advantage A10 system (Millipore, USA).

1.3 Analytical method

Two different modified extraction methods, ion-pairing and alkaline digestion [26], were used for viscera samples (pig heart, pig liver, pig kidney, chicken heart, and chicken liver) and muscle samples (pork loin and chicken breast), respectively. Before extraction, samples were thawed and homogenized. For chicken heart, at least 20 samples were pooled and homogenized as one extraction sample, while each chicken liver extraction sample was composed of at least three samples. For viscera samples, a 1-g aliquot was taken and placed in a polypropylene (PP) centrifuge tube, which was precleaned with methanol. Internal standards (MPFOS and MPFOA) were added into the tube, followed by 1 mL of 0.5 mol/L TBA solution and 2 mL of buffer (pH 10, 0.25 mol/L sodium carbonate). After mixing, 5 mL of MTBE was added, and the mixture was shaken for 20 min at 250 r/min. The organic and aqueous layers were separated by centrifugation at 3000 r/min for 15 min. Then, 4 mL of MTBE was removed and transferred to a second 15-mL PP tube. The extraction was repeated twice as described above, except that 5 mL of MTBE was removed each time instead of 4 mL in the subsequent extractions. All three extracts were combined. The final extract was concentrated to 1 mL under nitrogen gas after the addition of 1 mL of methanol. For muscle samples, 1 g of each sample was freeze-dried and ground into powder before extraction. The sample was spiked with internal standards and extracted by shaking for 16 h at 250 r/min with 10 mL of 10 mmol L^{-1} KOH in methanol in a PP tube, and then centrifuged. The supernatant was transferred into a PP tube and the residue was extracted with 3 mL of methanol. The supernatants were combined and concentrated to 1 mL under nitrogen gas. Each sample (viscera or muscle) was diluted to 40 mL using water in a PP container and loaded onto a Waters Oasis[®] WAX single-use cartridge (6 cc/150 mg), which was pre-conditioned with 4 mL of methanol and 4 mL of water. The sample was loaded onto the pre-conditioned cartridge with a positive pressure pump. The SPE cartridge was washed with 4 mL of acetate buffer solution (pH 4) and 4 mL of methanol, and eluted with 2% NH_4OH in methanol (target fraction). The flow rate of all steps was controlled at 1 drop s^{-1} . The eluate was concentrated to 1 mL under nitrogen for injection. Blanks of methanol (HPLC grade), treated as real samples in all steps, were processed with each batch.

Analysis of 9 perfluorinated acids and 2 fluorotelomer acids was performed using a high performance liquid chromatography (HPLC, P680 pump, Ultimate 3000 autosampler, Dionex, Sunnyvale, CA, USA) electrospray ionization tandem mass spectrometry (ESI-MS/MS, API 3200, Applied Biosystems/MDS SCIEX, USA). The experimental

conditions for ESI-MS/MS are shown in Table S1. A 10 μL aliquot was injected into a Dionex Acclaim 120 C18 column (5 μm , 4.6 mm i.d. $\times$ 150 mm length). Methanol (A) and 50 mmol L^{-1} NH_4OAc (B) were delivered at a flow rate of 1.0 mL min^{-1} . The separation time was 10 min. The solvent gradient started at 28% B, was ramped over 4 min to 5% B, held for 3 min, increased to 28% B, and held at this level until 10 min.

1.4 Quality assurance and data analysis

Quantification was performed using the internal standard method. A 10-point standard calibration curve was constructed using serial dilutions from 0.01–50 $\mu\text{g L}^{-1}$ and fit with a $1/x^2$ weighted regression. MPFOS and MPFOA were used as internal standards. The limits of detection (LODs) were calculated as three standard deviations from the average concentration in the extraction blanks. LODs were in a range of 0.002–0.032 $\mu\text{g L}^{-1}$ (Table S2). The correlation coefficient was >0.99 for each analyte. Recoveries of 11 spiked PFCs and two internal standards are shown in Table S2. The standard calibration curves were conducted before analysis and mixed standard (2 ng) was run as the quality control concentration, which was measured after every 10 injections to check for instrumental drift. If the standards were not within $\pm 20\%$ of their initial values, a new calibration curve was constructed.

To minimize the background, all potential sources of instrumental and procedural contamination were eliminated. Polytetrafluorethylene (PTFE) materials were removed from the laboratory equipment. All the containers employed

during the sample preparation and analysis procedures were washed thoroughly with methanol prior to use. One method blank sample was analyzed with every batch of eight samples. All analytes detected in the method blanks were below the LOD.

For samples where the compounds were not detected, the levels were treated as zero in statistical analysis. Data were analyzed by SPSS 13.0 (SPSS Inc., Chicago, IL, USA, 2005). Spearman’s correlation analysis was applied to examine the relationships among the PFCs.

2 Results

2.1 Concentrations of PFCs in raw animal tissue samples

The levels of PFCs detected in raw animal tissue samples from Beijing are summarized in Tables 1 and 2. The mean levels of total PFCs were in the order pig liver (3.438 ng g^{-1} wet weight (ww)) $>$ pig kidney (0.508 ng g^{-1} (ww)) $>$ pig heart (0.167 ng g^{-1} (ww)) $>$ chicken liver (0.098 ng g^{-1} (ww)) $>$ chicken heart (0.050 ng g^{-1} (ww)) $>$ pork loin (0.018 ng g^{-1} (ww)) $\approx$ chicken breast (0.012 ng g^{-1} (ww)). The total PFCs level in pig liver was over 30-fold higher than that in chicken liver. However, the level in pig heart was only 3-fold higher than that in chicken heart. Among these tissues, significantly lower levels of PFCs were observed in chicken breast and pork loin than other tissues. The PFCs, and in particular PFOS (0.094–11.30 ng g^{-1}), levels in pig liver samples were notably higher than in other tissue samples. Both PFOA and PFOS could be quantified in all of the

Table 1 Concentrations (ng g^{-1} , wet weight) of perfluorinated compounds (PFCs) detected in various pig tissues^{a)}

	PFHxS	PFHpA	PFOA	PFOS	PFNA	PFDA	PFUnDA	PFDoDA	PFTA	Total PFCs
Pig heart, $n=17$										
Range	<LOD	<LOD-0.062	<LOD-0.642	<LOD-0.136	<LOD-0.096	<LOD-0.068	<LOD-0.098	<LOD	<LOD	
Mean		0.008	0.107	0.024	0.009	0.005	0.014			0.167
Median			0.030	0.010						
Positive (%)		18	65	65	18	12	24			
Pig liver, $n=20$										
Range	<LOD-0.020	<LOD-0.060	0.034-1.790	0.094-11.30	<LOD-0.528	<LOD-0.614	<LOD-2.560	<LOD-0.536	<LOD-0.154	
Mean	0.002	0.012	0.241	2.163	0.161	0.150	0.560	0.124	0.025	3.438
Median			0.130	0.481	0.083	0.065	0.097			
Positive (%)	10	35	100	100	80	80	90	45	25	
Pig kidney, $n=24$										
Range	<LOD	<LOD-0.042	<LOD-4.979	<LOD-0.718	<LOD-0.216	<LOD-0.071	<LOD-0.162	<LOD-0.037	<LOD	
Mean		0.012	0.326	0.102	0.045	0.009	0.014			0.508
Median			0.059	0.055	0.030					
Positive (%)		38	83	88	58	25	25	4		
Pork loin, $n=20$										
Range	<LOD	<LOD	<LOD-0.097	<LOD-0.081	<LOD	<LOD	<LOD-0.043	<LOD	<LOD	
Mean			0.007	0.007			0.004			0.018
Median										
Positive (%)			10	25			10			

a) Mean values were not calculated when $>90\%$ of the samples had values below the LOD. Median values are given when $>50\%$ of the samples had values above the LOD. FOEA and FOUEA were not detected in any of the samples above LOD.

Table 2 Concentrations (ng g⁻¹, wet weight) of perfluorinated compounds (PFCs) detected in various chicken tissues^{a)}

	PFHpA	PFOA	PFOS	PFNA	PFDA	PFUnDA	PFDoDA	Total PFCs
Chicken heart, <i>n</i> =19								
Range	<LOD-0.060	<LOD-0.031	<LOD-0.273	<LOD-0.041	<LOD-0.043	<LOD-0.177	<LOD-0.034	
Mean	0.008	0.007	0.026	0.004	0.005			0.050
Median								
Positive (%)	16	32	47	16	11	5	5	
Chicken liver, <i>n</i> =19								
Range	<LOD-0.034	<LOD-0.061	<LOD-0.288	<LOD-0.038	<LOD-0.042	<LOD-0.166	<LOD-0.039	
Mean	0.003	0.009	0.063	0.009	0.003	0.011		0.098
Median			0.043					
Positive (%)	11	21	68	32	11	16	5	
Chicken breast, <i>n</i> =24								
Range	<LOD-0.017	<LOD-0.035	<LOD-0.137	<LOD	<LOD	<LOD	<LOD	
Mean			0.012					0.012
Median								
Positive (%)	8	4	21					

a) Mean values were not calculated when >90% of the samples had values below the LOD. Median values are given when >50% of the samples had values above the LOD. PFHxS, PFTA, FOEA and FOUEA were not detected in any of the samples above the LOD.

pig liver samples. Our data suggest that pigs may accumulate PFCs mainly in the liver. Additionally, PFHxS was only found in two of the twenty pig liver samples. In chicken liver, PFOS was the predominant compound, and was detected in 68% of the samples. In the other two chicken tissues sampled, PFOS was also the most frequently detected compound. The two fluorotelomer acids were not detected in any of the samples. In general, the levels of PFCs detected in chicken tissues were lower than those in pig tissues.

2.2 Composition profile of PFCs

The composition profiles of mean concentrations of the seven main PFCs in the tissues are shown in Figure 1. In pig liver, PFOS contributed up to 63% of the total composition of PFCs. Aside from PFOS, PFUnDA was the most prevalent PFC, contributing 16% of the total. The PFC-profile of pig kidney was very similar to that of pig heart, and PFOA was dominant, accounting for about two thirds of the total PFCs in the 2 tissues. In comparison, fewer PFCs were found in pork loin than pig viscera, with only 3 PFCs detected. In chicken liver and heart, PFOS contributed over half of the total PFCs. PFUnDA was observed in chicken liver, but not in chicken heart. In chicken breast samples, PFOS was the only compound detected.

2.3 Correlations between PFCs in pig liver

The levels of PFCs in pig liver were significantly higher than those in the other tissues and 5 PFCs (PFOA, PFOS, PFNA, PFDA and PFUnDA) were detected in over 80% of the samples. We found these five PFCs were significantly ($P<0.001$ – 0.04) correlated with each other, except for PFOA/PFDA. The strongest linear correlation ($r = 0.932$) was observed between the concentrations of PFOS and

Figure 1 Composition profiles of the seven main perfluorinated compounds detected in pig and chicken tissue samples: perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorooctane sulfonate (PFOS), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), and perfluorododecanoic acid (PFDoDA).

PFUnDA (Figure 2), which indicated that exposure to these two PFCs might be from the same source. In pig kidney, PFOA and PFOS were detected in over 80% of the samples, but no statistically significant correlation ($P>0.05$) was observed between them.

3 Discussion

3.1 Comparison with other studies

In general, the levels of PFCs observed in Beijing meat products were similar to or lower than those observed in other studies. In Japan, liver samples from farm animals (cattle, pigs, and chickens) were analyzed for 10 PFCs [23]. Among the analyzed PFCs, PFOS was dominant with mean liver concentrations of 67 ng g⁻¹ in chickens, 54 ng g⁻¹ in pigs, and 34 ng g⁻¹ in cattle. The PFOS levels in pig and chicken livers in the Japanese study were much higher than in the corresponding samples in our study. In contrast, the levels of other PFCs were not significant and were compa-

Figure 2 Correlation between perfluorooctane sulfonate (PFOS) and perfluoroundecanoic acid (PFUnDA) concentrations (ng g^{-1}) in pig livers.

table with those observed in our survey. It should be noted that the animals in the Japanese study had longer life spans (chickens 2.5–7 months, pigs 7 months) than in our study (chickens 40 d, pigs 5 months) before slaughter.

In samples from Catalonia (Spain), levels of PFCs were determined in 36 composite samples of randomly purchased foodstuffs [20]. In pork and chicken samples, PFOS was the only compound detected. The mean concentration in pork (0.045 ng g^{-1}) was somewhat higher than in our study, while the mean concentration in chicken (0.02 ng g^{-1}) was consistent with our results. In another study from Catalonia, Jogsten et al. [27] determined 11 PFCs in composite samples of meat products including pork loin, chicken breast, pig liver and lamb liver. The results indicated that although PFOS was the most frequently detected compound (8/20 food items), the highest PFOS level was only 0.33 ng g^{-1} (fresh weight) in lamb liver. In comparison with our results, none of PFCs were above the LOD in pork loin, chicken breast or pig liver.

In the United Kingdom, 20 composite food groups from the 2004 Total Diet Study (TDS) were analyzed for 15 PFCs, including PFOS [28]. PFOS was found in only 4 food groups, and the remaining PFCs ($0.5\text{--}20 \text{ ng g}^{-1}$ fresh weight) were detected in very few samples above the LOD. However, in the TDS, the LOD were notably higher than in our study. In Canada, Tittlemier et al. [18] analyzed 54 solid food composite samples of meat or other animal-derived food items for PFOS and perfluorocarboxylates. PFOS was predominant with a concentration of 2.7 ng g^{-1} (ww). In a recent study, the levels of nine PFCs were determined in the livers of 60 skip-jack tuna (*Katsuwonus pelamis*) collected from offshore waters and the open ocean along the Pacific Rim [29]. Both PFOS and PFUnDA were predominant compounds at concentrations of $<1\text{--}58.9$ and $<1\text{--}31.6 \text{ ng g}^{-1}$ (ww), respectively. The pig liver in our survey had similar composition profiles for these two PFCs. However, PFOA was rarely detected in the tuna livers, unlike the liver samples in our study.

3.2 Dietary intake of PFCs through meat products

In a recently published survey, the relative contributions of major food categories to food composition in Beijing were described [30]. Meat consumption for body weight (bw) was $1.6 \pm 1.3 \text{ g kg}^{-1} \text{ d}^{-1}$. On the basis of this value, we preliminarily estimated the dietary intake (DI) of PFCs through the consumption of meat products. Assuming pork is the only consumed meat product (not detected=0), the mean DI of PFOS and total PFCs are 0.011 and $0.029 \text{ ng kg}^{-1} \text{ d}^{-1}$, respectively. In a previous study, seven types of seafood collected from fish markets in two coastal cities of China were analyzed for nine PFCs [31]. The DI of PFOS and PFOA through fish consumption were $1.7\text{--}2.8$ and $1.9 \text{ ng kg}^{-1} \text{ d}^{-1}$, respectively. In comparison, the DI of PFCs via pork consumption is much lower. However, the DI of PFCs will increase when the consumption of animal viscera are taken into account. Although Chinese people do not consume much pig or chicken viscera, the levels of PFCs in viscera are much higher than those in muscle. The UK Food Standards Agency (UK FSA) Committee on Toxicology has established provisional tolerable daily intake (TDI) levels of 300 and $3000 \text{ ng kg}^{-1} \text{ d}^{-1}$ for PFOS and PFOA, respectively [32,33]. The European Food Safety Authority (EFSA) has derived a TDI of $150 \text{ ng kg}^{-1} \text{ d}^{-1}$ for PFOS and $1500 \text{ ng kg}^{-1} \text{ d}^{-1}$ for PFOA [34]. Thus, for the population of Beijing, the DI of PFCs via meat products is far below the recommended values.

Some countries have estimated the DI of PFOS. The UK FSA study showed that the estimated average adult dietary intakes from the whole diet in 2004 were $100 \text{ ng kg}^{-1} \text{ d}^{-1}$ for PFOS and $70 \text{ ng kg}^{-1} \text{ d}^{-1}$ for PFOA [28]. In Catalonia, the DI was estimated to be $62.5 \text{ ng kg}^{-1} \text{ d}^{-1}$ (ND=0) [20]. In Canada, the average DI of total perfluorocarboxylates and PFOS for Canadians was estimated to be $250 \text{ ng kg}^{-1} \text{ d}^{-1}$, based on results from the 2004 TDS composites study [18]. Another study from Canada showed that DI was $210\text{--}610 \text{ ng kg}^{-1} \text{ d}^{-1}$ in Nunavut [24]. Moreover, caribou meat contributed 43%–75% of daily dietary exposure to PFCs.

In summary, the levels of PFCs in animal viscera appeared higher than those in muscles. The levels in pig tissues were higher than those in the corresponding chicken tissues. Our results indicate that there is little potential risk of exposure to PFCs for the Beijing population via consumption of these products. However, further studies are required to investigate the source of PFCs in farmed animals. Additionally, the contribution of dietary intake to the total intake of PFCs should be investigated.

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Supporting Information

Figure S1 Fourteen sampling sites (red pentagrams) in the downtown of Beijing

Table S1 Experimental conditions of electrospray tandem mass spectrometry

Table S2 Recovery results ($n=3$) for the various matrices

The supporting information is available online at csb.scichina.com and www.springerlink.com. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.