# Species-specific accumulation of perfluorinated compounds in farm animals from Japan

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### Introduction

Fluorinated organic compounds (FOCs) are constituents in a wide range of applications such as liquid repellants for paper, packaging, textile, leather, adhesives, insecticides and other industrial products. Occurrences of perfluorinated chemicals in the environment recently have brought public concerned as a new group of pollutants. It was reported that FOCs were found in environmental compartments including water, sediment and wild life in various locations including some remote areas. Concentrations of FOCs in non-occupational exposed human were reported in various countries. Potential health effects of these compounds were reported such as liver damage, hypolipidemia, peroxisome proliferation inducing several liver enzyme activities and tumor promotion in laboratory experiments with mammals.<sup>1</sup>

FOC concentrations were reported in edible fish from various countries.<sup>2</sup> However, data are scarce for farm animals.<sup>3</sup> Farm animal originated food also contributes considerable portion of persistent organohalogen compounds to human. Therefore, it is important to determine concentration of FOCs in farm animals and their products. Hence, purpose of present study was to report, suitable analytical method to determine FOCs in domestic animals and to elucidate current status of contamination in farm animals in Japan.

#### Experimental Procedures

Animals were randomly selected from various regions from Japan, which including cattle, chicken, pig, and horse. Animal blood and liver were collected during year 2002 to 2004. Sera were prepared by centrifuging at 3000 RPM for 15 min. and kept –20°C until FOCs analysis. Samples were analyzed for 11 FOCs: perfluorohexanesulfonate (PFHxS), perfluorooctanesulfonate (PFOS), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnA), perfluorododecanoic acid (PFDoA) and perfluorooctanesulfonylamide (PFOSA). One ml of serum/plasma sample was employed for extraction by an ionpairing method, which described elsewhere. <sup>3-5</sup>For the extraction of liver samples, a homogenate of 1 g of liver in 5mL of distilled water was prepared. A 1-mL aliquot of the liver sample homogenate was extracted according to the procedure described above. Analysis of FOCs were performed using a high performance liquid chromatographtandem mass spectrometer (HPLC-MS/MS), operated in the electrospray negative ionization (ESNI) mode.<sup>4,5</sup>

#### **Results and Discussion**

The limits of quantification (LOQ) for PFHxS and PFOSA were 2 pg/ml, while for PFOS, PFHxA, PFHpA, PFNA, PFDA, PFDA, PFUnA, PFDoA were 10 pg/ml. The LOQ for PFPeA was 50 pg/ml, while mean blank concentration for PFOA was 49 pg/ml. The mean procedural recovery for analytes were ranged between 70±13 to 99±16 (n=8). The recovery of <sup>13</sup>PFOA spiked to calf serum and liver was 96±5 and 98±5, respectively. The concentrations of selected FOCs in sera and liver are given in Table 1. The concentrations of PFHxS, PFPeA, PFHxA, PFHpA, PFOSA and were very low and not frequently detected in blood and liver samples, hence those data are not presented. The detection frequency for PFOS was 100% for sera and liver samples.

Table 1.Fluorinated organic compounds (pg/ml for sera; pg/g for liver) in farm animals from Japan

Animal	Sample	No of	PFOS	PFDoDA	PFUnDA	PFDA	PFNA	PFOA
		samples						
Cattle	Serum	77	3049	5.5	95	215	405	= 0
	Liver	13	658 -	<10 -	<10 -	<10 -	165	50
	2.001		10300	38	503	1433	<10 -	<49 -
			33672	18	100	79	943	239
			33072	10	100	19	20	<49
			8783 -	<10 -	<10 -	<10 -	40.00	40
			71810	114	495	320	<10 - 98	<49
		22	5801	22	539	27	73	148
		14	558 -		<10 -			
			18600	<10 -	294	<10 -	<49 -	<49 -
				89		130	365	965
	~		66538	4.0	37			40
Chicken	Serum		39300 -	<10	<10 -	4.1	6	<49
Chicken	Liver		91850	<10	82	<10 - 12	<10 - 17	<49
Pig	Serum			33	214			176
			374			68	103	
	Liver			<10 –	<10 -			<49 –
			109 - 991	88	595	<10 - 624	<10 - 943	300
		27	53990	86	461			44
						68	65	
		6	44328 - 65900	62 - 127	345 - 748	49 - 100	34 - 93	<49 - 64
Horse	Serum		00000	121	62	-5 100	07 00	
10130	Seruiti		707	26	02	25	18	<49
		12			43 -			
			289 - 870	19 - 57	134	<10 - 63	<10 - 51	<49

The mean PFOS concentration was greater in chicken serum compared to that in all other animals. The PFOS concentration in pig serum was 2-15 folds lower than other animals. The mean PFOS concentration in chicken liver was found to be the highest among all the animals. The liver/serum partitioning was similar in cattle and chicken. It can be suggested that animal feeds and species-specificity were mainly influenced to current FOCs accumulation pattern in farm animals.

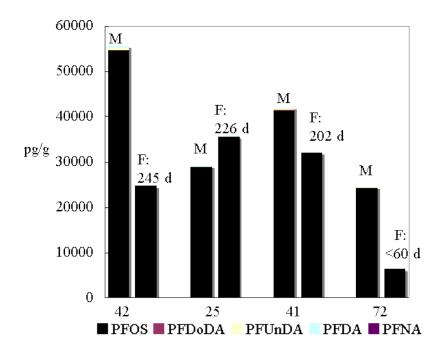


Fig 1.Concentrations (pg/g) of FOCs in mother (M) and fetus (F) liver of cattle. X-axis indicates age of mother in months. Fetus age: days.

Additionally, we analyzed 4 pairs of mother and fetus liver samples and data are shown in Figure 1. The percentage detection of PFOS and PFDA were 100% for mother and fetus lever, while, PFNA, PFUnA, PFDoA were not detected in most fetus liver. PFOA was not detected in any of liver samples (<49 pg/ml). This data suggested that PFOS could cross the placental barrier to enter fetal circulation and accumulate in liver as a target organ. PFOS was also reported in human cord blood, which highly correlated with maternal blood concentration, indicated that human fetus might also susceptible to PFOS exposure.<sup>6</sup>

In the current analytical method, 1 ml of blood or 0.2 g of liver is enough to measure FOCs in farm animals, however it is important to measure FOCs in other animal products in order to establish human daily intake of those compounds via animal products. This study provides evidence for the current background concentrations of farm animals in Japan. The current levels of FOCs in farm animals were seems to be lower than those in fish and wild animals from inland and coastal areas in Japan.<sup>2</sup>

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#### Reference

1. J. P. Giesy and K. Kannan, Environ. Sci. Technol., 2002, 36, 146A.

2. S. Taniyasu, K. Kannan, Y. Horii, N. Hanari and N. Yamashita, Environ. Sci. Technol., 2003, 37, 2634-2639

3. K. S. Guruge, S. Taniyasu, S. Miyazaki, N. Yamanaka and N. Yamashita, OrganohalogenCompds., 2004. 66, 4029-4034

4. N. Yamashita, K. Kannan, S. Taniyasu, Y. Horii, T. Okazawa, G. Petrick and T. Gamo. *Environ. Sci. Technol.* 2004. 38, 5522-5528

5. K. S. Guruge, S. Taniyasu, N. Yamashita, S. Wijeratna, K. M. Mohotti, H. R. Seneviratne, K. Kannan, N. Yamanaka,

andS. Miyazaki. J. Environ. Monit., 2005, 7, 371-377.

6. K. Inoue, F. Okada, R. Ito, S. Kato, S. Sasaki, S. Nakajima, A. Uno, Y. Saijo, F. Sata, Y. Yoshimura, R. Kishi and H. Nakazawa. *Environ Health Perspect*. 2004.112, 1024-1207