This article was downloaded by: On: 15 January 2011 Access details: Access Details: Free Access Publisher Taylor & Francis Informa Ltd Registered in England and Wales Registered Number: 1072954 Registered office: Mortimer House, 37- 41 Mortimer Street, London W1T 3JH, UK

Chemistry and Ecology

Publication details, including instructions for authors and subscription information: <http://www.informaworld.com/smpp/title~content=t713455114>

A review of spatial and temporal assessment of PFOS and PFOA contamination in China

Chunli Chen^{ab}; Yonglong Luª; Xiang Zhang^{ab}; Jing Geng^{abc}; Tieyu Wangª; Yajuan Shiª; Wenyou Hu^{ab}; Jing Liab

a State Key Laboratory of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing, China ^b Graduate University of Chinese Academy of Sciences, Beijing, China ^c Business Administration College, Taiyuan University of Technology, Taiyuan, China

To cite this Article Chen, Chunli , Lu, Yonglong , Zhang, Xiang , Geng, Jing , Wang, Tieyu , Shi, Yajuan , Hu, Wenyou and Li, Jing(2009) 'A review of spatial and temporal assessment of PFOS and PFOA contamination in China', Chemistry and Ecology, 25: 3, $163 - 177$

To link to this Article: DOI: 10.1080/02757540902918321 URL: <http://dx.doi.org/10.1080/02757540902918321>

PLEASE SCROLL DOWN FOR ARTICLE

Full terms and conditions of use:<http://www.informaworld.com/terms-and-conditions-of-access.pdf>

This article may be used for research, teaching and private study purposes. Any substantial or systematic reproduction, re-distribution, re-selling, loan or sub-licensing, systematic supply or distribution in any form to anyone is expressly forbidden.

The publisher does not give any warranty express or implied or make any representation that the contents will be complete or accurate or up to date. The accuracy of any instructions, formulae and drug doses should be independently verified with primary sources. The publisher shall not be liable for any loss, actions, claims, proceedings, demand or costs or damages whatsoever or howsoever caused arising directly or indirectly in connection with or arising out of the use of this material.

A review of spatial and temporal assessment of PFOS and PFOA contamination in China

Chunli Chen^{a,b}, Yonglong Lu^{a*}, Xiang Zhang^{a,b}, Jing Geng^{a,b,c}, Tieyu Wang^a, Yajuan Shi^a, Wenyou Hu^{a,b} and Jing Li^{a,b}

^aState Key Laboratory of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing, 100085, China; ^bGraduate University of Chinese Academy of Sciences, Beijing, 100039, China; ^cBusiness Administration College, Taiyuan University of Technology, Taiyuan, 030024, China

(*Received 15 February 2009; final version received 24 March 2009*)

The current state of concentrations of perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in China is presented. While products that are known to degrade to either PFOS or PFOA have been used in China, concentrations in environmental media have been reported to be relatively low across China. Greater concentrations of PFOS and PFOA were observed in southern and eastern China than in other areas of China. Concentrations of PFOS and PFOA were relatively great in the Huangpu River, with concentrations of 20.5 ng l−¹ and 1590 ng l−1, respectively. Surface waters of Dongguan and Shanghai were more contaminated by PFOS and PFOA than that of other cities. Dongguan was the only city in China in which PFOS value in surface water exceeded the water quality criterion, while PFOA concentration in Shanghai was 152 ng l^{−1}. Similar to other contaminants, point-source pollution was also the common pattern of PFOS and PFOA contamination. Concentrations of PFOS in human blood in China were relatively greater in China than other countries, with drinking water contamination given as the most likely source. Concentrations of PFOS in human blood have increased from the 1980s to the 2000s, while such a trend was not observed for PFOA.

Keywords: persistent organic pollutants (POPs); perfluorinated organic pollutants (PFOS; PFOA); fresh water systems; humans; organisms

1. Introduction

Perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) are two of the more prominent perfluorinated compounds (PFCs) widespread globally in many environmental media, including remote regions far from sources, such as the Arctic and Antarctic [1–3]. Perfluorinated acids, including PFOS and PFOA, are heat resistant and water and oil-repellent. Due to their unique properties, they have been used since the 1970s as surfactants and surface protectors in stain-resistant coatings for fabrics, carpet and leather, and in grease and oil-resistant coatings for paper products and food containers, such as fast-food packaging [4–6].

ISSN 0275-7540 print*/*ISSN 1029-0370 online © 2009 Taylor & Francis DOI: 10.1080*/*02757540902918321 http:*//*www.informaworld.com

^{*}Corresponding author. Email: yllu@rcees.ac.cn

164 *C. Chen* et al.

PFCs have been found in both freshwaters and oceans, in almost every wildlife species in which they were measured, and in the blood serum of both occupationally and non-occupationally exposed people across the world [7–14]. Meanwhile, PFCs have also been found in sewage, sludge, guillemot eggs, and fresh and tap water [15–19].Among several PFCs, PFOS was the predominant compound found in biota [20,21], while PFOA was predominant in environmental matrices [20]. Furthermore, they have been shown to elicit toxic effects in exposed laboratory animals [22,23]. PFOS and PFOA were resistant to biotransformation and were ubiquitously found in human blood with half-lives of several years [24]. Following *in utero* exposure in rodents [25,26], PFOS and PFOA can reduce viability and cause bodyweight deficits and other postnatal effects on rat pups.

PFCs in the environment originate from the production, use and disposal of products containing PFCs [27]. The major sources of PFOS and PFOA in the environment seem to be their dissemination with wastewater and their release in traces from consumer products [24]. It has also been reported that residential histories may be a determinant of concentrations of PFOS and PFOA in the blood of people from Japan [28,29], USA [10] and China [30]. Here we provide a brief overview of the research status concerning PFOS and PFOA in environmental media of China. This review summarises the data on concentrations of PFOS and PFOA in multienvironmental media across China, and discusses the information from both spatial and temporal perspectives. The concentrations observed are also compared and contrasted with concentrations observed in other countries. Data were obtained from peer-reviewed articles published before July 2008. The available data consisted mainly of arithmetic or geometric mean and data ranges or medians. The midpoint was used if only ranges were given. Due to a lack of information on the statistical distribution of the data in most studies, the means for sample size or variance were not weighted. The information is compiled in tables available in the Supporting Information (SI) section (online only).

2. Production and use of PFCs in China

Relatively large amounts of industrial chemicals are manufactured and used in China, especially in the textile, leather and paper industries. The demand for PFCs in the country was met primarily through importation, with less than 1% was produced by domestic suppliers. The major fluoropolymer products of China include polytetrafluoroethylene (PTFE), fluorinated ethylene propylene (FEP), polyvinylidene fluoride (PVDF) and polychlorotrifluoroethylene (PCTFE), among which PTFE accounts for approximately 90%. Most of the manufacturers of fluoropolymers were located in southern and eastern China, especially in Hubei, Zhejiang, Fujian, Guangdong and Jiangsu provinces [31,32]. The spatial distribution of the manufacturing provinces and sampling sites is shown in Figure 1.

In 2003, the seven major manufacturers of fluoropolymers in China were responsible for more than 90% of domestic production. The PTFE production status in China is shown in Table 1 [33]. Approximately 149,800 tonnes of PTFE were produced in 2003, while the total production capacity of all the manufacturers could achieve up to 240,000 tonnes yr−1. Approximately 90% of the fluoropolymers used in China was PTFE.

3. PFOS and PFOA pollution in environmental media

Like other persistent organic pollutants (POPs), most of the research in environmental contamination for PFOS and PFOA in China focused on the eastern and southern parts of the country [34], especially on the estuaries of the Pearl River Delta and Yangtze River.

Figure 1. Spatial distribution of the manufacturing provinces and sampling sites in China.

Table 1. Production of PTFE in 2003 in China (tonnes yr^{-1}).

Manufacturer	Production capacity		Output/Tonnes Location/Province
Zhejiang Juhua Group Corporation	5000	3600	Zhejiang
Shanghai 3F Co., Ltd	4000	2900	Shanghai
Jiangsu Myland Group	3500	2800	Jiangsu
Darkin Fluorine Chemical (China) Co., Ltd	3000		Jiangsu
Shandong Dongyue Polymer Material Co., Ltd.	5000	3200	Shandong
Liaoning Fuxin Fluorin Chemical Co., Ltd.	1000	730	Liaoning
Second factory of Chenguang Research Institute of Chemical Industry	1000	650	Sichuan
Jinan 3F Fluoro. Chemical Co., Ltd.	1500	1100	Shandong
Total	240000	149800	

Note: Source: http://www.pka.com.cn/news/view.asp?id=3442.

3.1. *PFOS and PFOA in surface water*

3.1.1. *PFOS and PFOA in rivers*

In general, PFOS and PFOA were both detectable in all the main rivers in China. Concentrations of PFOS and PFOA in river water in China collected from the literature are presented in Figure 2 (Supplementary Table S1 – online only) [35–40]. If different researchers investigated the same area, means of the different results are given.

Among the 10 sites identified in Figure 2, concentrations of PFOS and PFOA were greater in some areas, especially the Huangpu River. Concentrations of both PFOS and PFOA at that location were the greatest values among all the sites, with concentrations greater than $20 \text{ ng } l^{-1}$

166 *C. Chen* et al.

Figure 2. PFOS and PFOA concentrations in rivers in China (ng l−1). Yangtze River [36]; Yangtze River estuary [35]; Three Gorges Reservoir areas of the Yangtze River [37]; Pearl River Delta [36,38]; Songhua River [39]; Huangpu River [35]; Hunhe River between Fushun and Shenyang City [40]; Yellow River in Jinan City [40]; Bohai Bay around Dalian [40]; Tianchi [40].

and 1000 ng l−¹ for PFOS and PFOA, respectively. These relatively great concentrations of PFOS and PFOA in the Huangpu River suggested sources associated with the urban and industrial areas in Shanghai, since the Huangpu River flows through Shanghai, and then empties into the East China Sea. In fact, there were many PFC manufacturers in and around Shanghai [31]. Therefore, one of the main reasons for the relatively great concentrations of PFOS and PFOA in the Huangpu River was the discharge of contaminants and wastewater from PFC factories. In addition, the water quality of the Huangpu River has been adversely affected by intense industrial and urban activities, so the second possible source for PFOS and PFOA contamination in the river was the upstream water pollution. Furthermore, the manufacturing industry cannot be neglected, and some research showed that it was one of the possible sources of organo-fluorine compounds [41]. Manufacturing in Shanghai is intensive, and the Huangpu River has been greatly influenced by the cities along its course. Therefore, the manufacturing industry concentrated in Shanghai was the third possible source of PFOS and PFOA contamination in the Huangpu River.

PFOS was detected in nine other sites, except the Yangtze River estuary, with concentrations ranging from 0.41–20.5 ng l^{−1}. Moreover, the mean concentration of PFOS was 0.41 ng l^{−1} in Tianchi, which is in Jilin province and relatively remote from the likely sources. PFOA concentrations in these sites ranged from $0.32-1590$ ng l⁻¹. Like PFOS, the greatest concentration was detected in the Huangpu River as well. The least concentration of PFOA of 0.32 ng l⁻¹ was observed in the Songhua River. Concentrations of PFOA were not determined for water of the Hunhe River between Fushun and Shenyang City, the Yellow River in Jinan City, Bohai Bay around Dalian, or Tianchi.

3.1.2. *PFOS and PFOA in coastal waters*

There is little information about PFOS and PFOA in the coastal waters of China, and most of the information that is available is for southern China.

Compared with other Asian countries, concentrations of PFOS and PFOA along China's coasts were less than those in Korea and Japan. PFOS concentrations from all the sampling sites in China ranged from 0.008 to 9.68 ng l⁻¹ [38,42,43], while in Korea, they ranged from 0.039–730 ng l⁻¹ [38,42], and in Japan from $12.7-25.4$ ng 1^{-1} [44]. Similarly, concentrations of PFOA from all the

sampling sites in China ranged from 0.16–37.6 ng l⁻¹ [38,42,43], compared to 0.239–317 ng l⁻¹ in Korea [38,42], and $154-192$ ng 1^{-1} in Japan [44].

3.1.3. *PFOS and PFOA in inland cities*

Information on concentrations of PFOS and PFOA in surface water is available for eight cities in China [36,37,40,45] (Figure 3; Supplementary Table S2 – online only). The greatest concentration of PFOS was found in Dongguan City, Guangdong Province, which is in the southern part of China. The city is famous for manufacturing and production of electronics, telecommunication equipment and machinery, which accounts for the majority of local gross industrial output (GIO). These industries have transformed the Dongguan area into a "world manufacturing base". The manufacturing processes involved in production of electronics, plastics and textiles were the most likely sources of PFOS and PFOA in Dongguan.

Concentrations of PFOS in Shenyang, Wuhan and Shanghai were similar, with a mean of 5.00 ng l−1. Among all the cities studied, concentrations of PFOS in Beijing were moderate (approximately 1.75 ng l^{-1}), while those in Chongqing, Yichang and Nanjing were relatively less (*<*1.00 ng l−1). The mean concentration of PFOS from these eight cities was 1.29 ng l−1, which was less than those in Japan and New York City, where concentrations of PFOS were 6.88 ng l⁻¹ and 87.3 ng l^{-1} , respectively.

The greatest concentration of PFOA, which was approximately 152 ng l^{-1} , occurred in Shanghai. The PFOA concentration in Chongqing was $21.4 \text{ ng } 1^{-1}$, the second greatest value among the data, while PFOA concentrations in the six other cities were all below $10 \text{ ng } l^{-1}$. Excluding Shengyang, for which there was no data from the literature, the mean value of PFOA in the

Figure 3. Concentrations of PFOS and PFOA in surface water in China cities (ng l^{−1}). Beijing [45]; Shenyang [40]; Dongguan [36]; Chongqing [36]; Yichang [36]; Nanjing [36]; Shanghai [36]; Wuhan [37].

seven other cities was 29.2 ng l^{−1}. This was half of the PFOA concentration reported for Japan $(58.6 \text{ ng } l^{-1})$, but about 15-fold greater than New York City $(1.98 \text{ ng } l^{-1})$ [20,46].

3.1.4. *PFOS and PFOA in tap water*

Concentrations of PFOS and PFOA in tap water in China are summarised in Figure 4 (Supplementary Table S3 – online only) [39,40,47]. Among these 11 cities studied, the greatest concentration of PFOS of 13.2 ng l−¹ occurred in Kunming, followed by Shenzhen, with a concentration of approximately 7.50 ng l−1. Concentrations of PFOS in tap water of other cities were less, with concentrations ranging from $0.06-1.62$ ng l^{-1} . PFOA was analysed in tap water from only four cities, and the greatest concentration of approximately $109 \text{ ng } l^{-1}$ was observed in Hangzhou. This concentration was more than 50-fold greater than those in Shenzhen, Kunming or Harbin. In general, concentrations of PFOS in tap water were greater in southern than northern China. This is most likely due to the fact that most of the PFC manufacturers that produced or used PFC are situated in southern China.

Concentrations of PFOS and PFOA in tap water in China were comparable to those in Japan, but much greater than those reported for Malaysia, Canada, Sweden, Thailand and Vietnam (Table 2) [40,47,48].

3.1.5. *Hazard assessment of PFOS exposure to aquatic species*

Water-quality criteria for PFOS for the protection of aquatic animals have been represented as the secondary maximum concentration (SMC) and secondary continuous concentration (SCC)

Figure 4. PFOS concentration in tap water in China (ng l−1). Shenyang [40]; Dalian [40]; Changchun [40]; Jinan [40]; Yinchuan [40]; Beijing [40]; Shanghai [40]; Harbin [39]; Shenzhen [47]; Kunming [47]; Hangzhou [47].

Location	PFOS $(ng L^{-1})$	PFOA	Reference	
China	$0.56 - 13.2$	$1.10 - 109$	[40, 47]	
Malaysia	$0.00 - 0.10$	$0.00 - 0.10$	[47]	
Canada	n.d. ^a	0.20	[47]	
Japan	$0.16 - 22.0$	$2.30 - 84.0$	[48]	
Sweden	$0.30 - 0.80$	$0.00 - 1.30$	[47]	
Thailand	$0.10 - 1.90$	$0.00 - 4.60$	[47]	
Vietnam	n.d.	n.d.	[47]	

Table 2. Concentrations of PFOS and PFOA in tap water throughout the world.

Note: ^aNot detectable.

[49]; values for the SMC and SCC are 8.50 and 1.20 µg of PFOS 1^{-1} , respectively. A preliminary estimate of risk was determined by comparing the concentrations of PFOS in surface water to the SMC and SCC. None of the PFOS concentrations in China exceeded either SMC or SCC. In fact, even the greatest measured concentration of PFOS ($94.0 \text{ ng } 1^{-1}$) was approximately 13-fold less than the SCC and 90-fold less than the SMC.

The bioaccumulation factor (BAF) of PFOS for fish has been estimated to vary between 6300 and 125,000 [50]. Based on these estimates of BAF, the potential for adverse effects of PFOS on trophic level IV organisms was assessed by comparing measured concentration of PFOS with the water quality criterion (WQC) that was calculated to be protective for avian wildlife. The WQC for wildlife was estimated to be 43.0 ng of PFOS l^{-1} [51]. Only the concentration of PFOS in surface water from Dongguan City exceeded the wildlife WQC and was approximately 2-fold greater than the wildlife WQC. The PFOS concentration in the Huangpu River was approximately half that of the wildlife WOC (43.0 ng l⁻¹). However, concentrations at other sites were less than the wildlife WQC. The conservative nature of the risk analysis was used to extrapolate from birds to safe water concentrations [52]. Therefore, none of the concentrations of PFOS in surface water of China were expected to cause adverse population-level effects, but necessary attention should be paid to some locations.

3.2. *PFOS and PFOA in organisms*

Compared with other developed countries, the available surveys on concentrations of PFOS and PFOA in organisms of China were very deficient. The limited data on PFOS and PFOA concentrations in organisms are given in Table 3 [53–56].

PFOS concentrations in livers of skipjack tuna in the East China Sea (Taiwan) were approximately half of the mean value found in skipjack tuna in Japan, but comparable to that found in India [53]. Seven types of seafood collected from fish markets in Guangzhou and Zhoushan were analysed by Gulkowska et al. (2006) [54], including fish, molluscs, crabs, shrimp, oysters, mussels, and clams. Fluorochemicals were detectable in all the samples, and concentrations of PFOS in seafood samples ranged from 0.30–13.9 ng g^{-1} ww. The greatest concentration of PFOS was found in mantis shrimp, while the greatest concentration of PFOA was found in swimming crab (1.67 ng g^{-1} wet weight). PFOS and PFOA were detected in mussels and oysters from coastal waters of southern China [55]. Concentrations of PFOS and PFOA in blood serum of giant pandas and red pandas housed in zoos and animal parks from six provinces in China were reported by Dai et al. (2006) [56]. The results showed that PFOS was the predominant PFC in pandas. Concentrations of PFOS in serum of red pandas was comparable to those observed in polar bears collected from Alaska [57], while concentrations of PFOS in serum of giant pandas was one third of those observed in polar bear collected from Alaska [57].

Location	Species	Tissue	PFOS	PFOA	Reference
Southern China	Mussel	Soft tissue	$0.113 - 0.352$	$< 0.204 - 0.328$	$[55]$
East China Sea (Taiwan)	Fish	Liver	$5.10 - 7.10$	n.d. ^a	$[53]$
Zhoushan	Fish	Soft tissue	$0.38 - 1.77$	n.d.	[54]
Zhoushan	Mollusc	Soft tissue	$0.96 - 1.32$	$0.31 - 0.35$	[54]
Zhoushan	Crab	Soft tissue	$0.94 - 2.80$	$0.34 - 0.87$	[54]
Zhoushan	Shrimp	Soft tissue	$1.28 - 1.80$	n.d.	$[54]$
Zhoushan	Shellfish	Soft tissue	$0.42 - 0.51$	$0.29 - 0.48$	$[54]$
Guangzhou	Fish	Soft tissue	$0.67 - 2.93$	n.d.	$[54]$
Guangzhou	Mollusc	Soft tissue	$0.87 - 1.07$	$0.31 - 0.43$	$[54]$
Guangzhou	Crab	Soft tissue	$2.02 - 4.59$	$0.42 - 1.67$	$[54]$
Guangzhou	Shrimp	Soft tissue	$0.58 - 13.9$	$< 0.25 - 0.45$	$[54]$
Guangzhou	Shellfish	Soft tissue	$0.33 - 0.54$	$< 0.25 - 0.34$	$[54]$
All of China	Red panda	Whole blood	$0.80 - 73.8$ ^b	$0.33 - 8.20b$	[56]
All of China	Giant panda	Whole blood	$0.76 - 19.0b$	$0.32 - 1.56^b$	[56]

Table 3. Concentration of PFOS and PFOA in organisms in China (ng g^{-1} ww).

Note: ^aNot detectable; ^bValues are μ g l⁻¹.

3.3. *PFOS and PFOA in humans*

3.3.1. *PFOS and PFOA in human blood*

The world's first report of organic fluorinated compounds in human blood was published in the 1960s [58], more than 40 years earlier than the first report of the compounds being found in human blood in China.A comparison of PFOS and PFOA in human blood serum among different countries was summarised as box-plot charts for both PFOS and PFOA (Figure 5 and 6; Supplementary Table S4 – online only) [59–69]. According to an experiential relationship suggested by Ehresman et al. (2007) [70], the concentrations of PFOS and PFOA reported for wholeblood were converted to blood serum values by multiplying by a factor of 2, and plasma values were converted to blood serum values by multiplying by a factor of 1.

To date, most measurements of PFCs in human blood have been conducted in Europe, Asia, North America and Australia, while there are no available reports on PFCs in human blood in Africa. The greatest concentrations of PFOS in human blood were for China, and the greatest concentrations of PFOA were for Korea [60]. The concentrations of PFOS reported in these studies were likely overestimated due to calibration method used [71]. However, from a global perspective, PFOS and PFOA concentrations were comparatively greater in Europe than those in other areas. Just like most of the other countries, the PFOS concentration in human blood in China was greater than the PFOA concentration. Research has shown that the main source of PFOS and PFOA contamination in human blood originates from drinking water [72,73]. Therefore, the high level of PFCs in the blood of the Chinese population reflected the possible higher level of PFCs in the drinking water in China.

In order to compare concentrations of PFOS and PFOA in blood of men and women in the countries studied, drop-line charts are given (Figure 7 and 8; Supplementary Table S4 – online only). In most of these 16 countries for which data is available, the PFOS concentration in human blood serum differs between males and females, with males generally having greater concentrations. In addition, significant differences in concentrations of PFOS in blood serum were observed between males and females from China, Canada and Korea. However, the distribution of PFOS in males and females in Canada and Korea was different from that observed in China and most of other countries, because concentrations of PFOS in females were greater than those in males from these two countries. Concentrations of PFOA were relatively less in people from China than other countries, and there was no obvious difference between males and females. Like PFOS, the PFOA concentration in human serum differs according to sex in most of these

Figure 5. Box-plot charts of PFOS in human serum in different countries. China [59]; Korea [60]; Japan [60,61,62]; India [60]; Malaysia [60]; Sri Lanka [63]; Sweden [69]; Germany [64]; Spain [65]; Poland [60]; Italy [60]; Belgium [60]; United Kingdom [66]; United States [67]; Canada [68]; Brazil [60]; Colombia [60]; Australia [69].

16 countries, again with greater concentrations observed in males. However, there was a relatively great difference in concentrations in blood of males and females in Japan, Korea and Poland, as opposed to China and other countries.

3.3.2. *PFOS and PFOA in breast milk*

Until now, there was only one research study on PFOS and PFOA in human breast milk in China. So et al. (2006) assessed the health risks in infants associated with exposure to perfluorinated compounds in human breast milk from Zhoushan, China. PFOS and PFOA were the two dominant PFCs detected in all milk samples and ranged from 45–360 ng l⁻¹ (median, 100 ng l⁻¹) and $47–210$ ng l⁻¹(median, 110 ng l⁻¹), respectively [74]. In addition, there was no statistically significant correlation between concentrations of either PFOS or PFOA and maternal age, weight, or infant weight. Furthermore, the results indicated that there may be a small potential risk of PFOS for the infants in Zhoushan via the consumption of breast milk [74].

Concentrations of PFOS in human breast milk in China were comparable to those observed in Germany (median, 118 ng l^{−1}) [75], the USA (median, 106 ng l^{−1}) [76], Malaysia (median, 111 ng l^{−1}) [77] or the Philippines (median, 104 ng l^{−1}) [77], but less than that observed in Japan (median, 196 ng l^{−1}) [77], Hungary (median, 330 ng l^{−1}) [75], or Sweden (median, 166 ng l^{−1}) [78]. Concentrations of PFOA in human breast milk in China were greater than those observed in the USA (median, 36.1 ng l⁻¹) [76] and Japan (median, 67.3 ng l⁻¹) [77].

3.4. *Temporal trend of PFOS and PFOA pollution in China*

The monitoring of PFCs in the environment is not adequate because of limited funding and little policy support. In addition, most of the places were investigated only once. Limited data collected

Figure 6. Box-plot charts of PFOA in human serum in different countries. China [59]; Korea [60]; Japan [60,61,62]; India [60]; Malaysia [60]; Sri Lanka [63]; Sweden [69]; Germany [64]; Spain [65]; Poland [60]; Italy [60]; Belgium [60]; United Kingdom [66]; United States [67]; Canada [68]; Brazil [60]; Colombia [60]; Australia [69].

Figure 7. Drop-line charts of PFOS in human serum in different countries. China [59]; Korea [60]; Japan [60,61,62]; India [60]; Malaysia [60]; Sri Lanka [63]; Sweden [69]; Germany [64]; Spain [65]; Poland [60]; Italy [60]; Belgium [60]; United Kingdom [66]; United States [67]; Canada [68]; Brazil [60]; Colombia [60]; Australia [69].

over time for some of the places are presented (Figure 9; Supplementary Table S5 – online only). Between 2002 and 2004, there was a relatively great increase of the concentration of PFOS in blood of people living in Shenyang. Concentrations of PFOS in blood serum in 2004 were approximately 400-fold greater than that those in blood serum collected in 1987. This suggests that people in

Figure 8. Drop-line charts of PFOA in human serum in different countries. China [59]; Korea [60]; Japan [60,61,62]; India [60]; Malaysia [60]; Sri Lanka [63]; Sweden [69]; Germany [64]; Spain [65]; Poland [60]; Italy [60]; Belgium [60]; United Kingdom [66]; United States [67]; Canada [68]; Brazil [60]; Colombia [60]; Australia [69].

Shenyang experienced a relatively high level of exposure to PFOS during these years, and that residential histories may be a determinant for PFOS [30]. This was in contrast to observations made in Japan and the USA [28,29,79]. PFOS concentrations in the USA were significantly greater in human serum collected in 1989 compared to that collected in 1974. However, additional data collected in 2001 did not suggest any increases in PFOS concentrations in blood plasma since 1989 [79].

Temporal trends in concentrations of PFOS and PFOA in water of the Pearl River Delta showed that PFOS increased from 3.49–23.1 ng l⁻¹ in one year, and increased about 6-fold from 2003 to 2004. The significant increase indicated that there was some source along the Pearl River Delta. At the same time, PFOA did not show any increase there.

Figure 9. Temporal trend of PFOS and PFOA levels in China. Human serum in Shenyang [30,59]; Pearl River Delta [36,38].

4. Conclusion

In general, concentrations of PFOS and PFOA in environmental media in China were relatively less than those in other areas. This may be partly due to the fact that PFCs were not mass produced in China. On a regional scale, concentrations of PFOS and PFOA were greater in southern and eastern China, especially the Huangpu River,Yangtze River estuary and Pearl River Delta, than in other areas. This indicates concentrations of PFOS and PFOA were closely related to the spatial distribution of PFCs manufacturing provinces, and also indicates the strong influence of river discharge on the magnitude and extent of PFCs contamination in southern and eastern China. Waters in the Huangpu River, Dongguan and Shanghai were seriously polluted, and the greatest PFOA concentration in tap water was found in Hangzhou City. All of these areas are in the provinces which have PFCs manufacturers. Moreover, the PFOA level in Hangzhou was more than 50-fold greater than other cities in China. Furthermore, concentrations of PFOS and PFOA in China's coastal waters were less than those in other Asian countries, such as Korea and Japan. Also, up to this point, there have been no reports on PFCs in the soil and sediment. Furthermore, concentrations of PFOS in human blood serum was greater in China compared to other countries, which indicated that PFOS levels in drinking water in China are relatively greater than in other countries. In addition, local geographic, economic and historical factors also had a marked effect on the diffusion of PFOS and PFOA contamination.

Acknowledgements

This study was supported by the National Basic Research Program of China ("973" Research Program) with Grant No. 2007CB407307 and the Knowledge Innovation Program of the Chinese Academy of Sciences, with Grant No. KZCX2-YW-420-5, and No. KZCX1-YW-06-05-02.

The authors would like to thank Prof. John Giesy (Department of Veterinary Biomedical Sciences & Toxicology Center, University of Saskatchewan) for language polishing, editing and constructive suggestions.

References

- [1] J.P. Giesy and K. Kannan, *Global distribution of perfluorooctane sulfonate in wildlife*, Environ. Sci. Technol. 35 (2001), pp. 1339–1342.
- [2] J.W. Martin, D.M. Whittle, D.C.G. Muir, and S.A. Mabury, *Perfluoroalkyl contaminants in a food web from Lake Ontario*, Environ. Sci. Technol. 38 (2004), pp. 5379–5385.
- [3] S. Wei, L.Q. Chen, S. Taniyasu, M.K. So, M.B. Murphy, N. Yamashita, L.W.Y. Yeung, and P.K.S. Lam, *Distribution of perfluorinated compounds in surface seawaters between Asia and Antarctica*, Mar. Pollut. Bull. 54 (2007), pp. 1813–1818.
- [4] B.D. Key, R.D. Howell, and C.S. Criddle, *Fluorinated organics in the biosphere*, Environ. Sci. Technol. 31 (1997), pp. 2445–2454.
- [5] E. Sinclair, S-K. Kim, and K. Kannan, *Quantitation of gas phase perfluoroalkyl surfactants and fluorotelomer alcohols released from non-stick cookware and microwave popcorn*, Environ. Sci. Technol. 41 (2007), pp. 1180–1185.
- [6] E. Kissa, *Fluorinated surfactants and repellents*, 2nd edn. Marcel Dekker, New York, 2001.
- [7] J. Falandysz, S. Taniyasu, A. Gulkowska, N. Yamashita, and U. Schulte-Oehlmann, *Is fish a major source of fluorinated surfactants and repellents in humans living on the Baltic coast?* Environ. Sci. Technol. 40 (2006), pp. 748–751.
- [8] A.M. Calafat, L.Y. Wong, Z. Kuklenyik, J.A. Reidy, and L.L. Needham, *Polyfluoroalkyl chemicals in the US population: Data from the National Health and Nutrition Examination Survey (NHANES) 2003–2004 and comparisons with NHANES 1999–2000*, Environ. Health Persp. 115 (2007), pp. 1596–1602.
- [9] L. Maestri, S. Negri, M. Ferrari, S. Ghittori, F. Fabris, P. Danesino, and M. Imbriani, *Determination of perfluorooctanoic acid and perfluorooctanesulfonate in human tissues by liquid chromatography/single quadrupole mass spectrometry*, Rapid Commun. Mass Spectrom. 20 (2006), pp. 2728–2734.
- [10] G.W. Olsen, P.W. Logan, K.J. Hansen, C.A. Simpson, J.M. Burris, M.M. Burlew, P.P. Vorarath, P. Venkateswarlu, J.C. Schumpert, and J.H. Mandell, *An occupational exposure assessment of a perfluorooctanesulfonyl fluoride production site*, Biomon. Aiha. J. 64 (2003), pp. 651–659.
- [11] K. Kannan, J.C. Franson, W.W. Bowerman, K.J. Hansen, J.D. Jones, and J.P. Giesy, *Perfluorooctane sulfonate in fish-eating water birds including bald eagles and albatrosses*, Environ. Sci. Technol. 35 (2001), pp. 3065–3070.
- [12] P.T. Hoff, K. Van de Vijver, W. Van Dongen, E.L. Esmans, R. Blust, and W.M. De Coen, *Perfluorooctane sulfonic acid in bib* (Trisopterus luscus) *and plaice* (Pleuronectes platessa) *from the Western Scheldt and the Belgian North Sea: Distribution and biochemical effects*, Environ. Toxicol. Chem. 22 (2003), pp. 608–614.
- [13] K.I. Van de Vijver, P.T. Hoff, K. Das, W. Van Dongen, E.L. Esmans, T. Jauniaux, J.M. Bouquegneau, R. Blust, and W.M. De Coen, *Perfluorinated chemicals infiltrate ocean waters: Link between exposure levels and stable isotope ratios in marine mammals*, Environ. Sci. Technol. 37 (2003), pp. 5545–5550.
- [14] Z. Kuklenyik, J.A. Reich, J.S. Tully, L.L. Needham, and A.M. Calafat, *Automated solid-phase extraction and measurement of perfluorinated organic acids and amides in human serum and milk*, Environ. Sci. Technol. 38 (2004), pp. 3698–3704.
- [15] S. Takagi, F. Adachi, K. Miyano, Y. Koizumi, H. Tanaka, M. Mimura, I. Watanabe, S. Tanabe, and K. Kannan, *Perfluorooctanesulfonate and perfluorooctanoate in raw and treated tap water from Osaka, Japan*, Chemosphere 72 (2008), pp. 1409–1412.
- [16] N. Wang, B. Szostek, P.W. Folsom, L.M. Sulecki, V. Capka, R.C. Buck, W.R. Berti, and J.T. Gannon, *Aerobic biotransformation of C-14-labeled 8-2 telomer B alcohol by activated sludge from a domestic sewage treatment plant*, Environ. Sci. Technol. 39 (2005), pp. 531–538.
- [17] E. Sinclair, and K. Kannan, *Mass loading and fate of perfluoroalkyl surfactants in wastewater treatment plants*, Environ. Sci. Technol. 40 (2006), pp. 1408–1414.
- [18] D. Skutlarek, M. Exner, and H. Farber, *Perfluorinated surfactants in surface and drinking water*, Environ. Sci. Pollut. R. 13 (2006), pp. 299–307.
- [19] R. Loos, J. Wollgast, T. Huber, and G. Hanke, *Polar herbicides, pharmaceutical products, perfluorooctanesulfonate (PFOS), perfluorooctanoate (PFOA), and nonylphenol and its carboxylates and ethoxylates in surface and tap waters around Lake Maggiore in Northern Italy*, Anal. Bioanal. Chem. 387 (2007), pp. 1469–1478.
- [20] K. Senthilkumar, E. Ohi, K. Sajwan, T. Takasuga, and K. Kannan, *Perfluorinated compounds in river water, river sediment, market fish, and wildlife samples from Japan*, B. Environ. Contam. Tox. 79 (2007), pp. 427–431.
- [21] M. Houde, T.A.D. Bujas, J. Small, R.S. Wells, P.A. Fair, G.D. Bossart, K.R. Solomon, and D.C.G. Muir, *Biomagnification of perfluoroalkyl compounds in the bottlenose dolphin* (Tursiops truncatus) *food web*, Environ. Sci. Technol. 40 (2006), pp. 4138–4144.
- [22] M.E. Austin, B.S. Kasturi, M. Barber, K. Kannan, P.S. MohanKumar, and S.M.J. MohanKumar, *Neuroendocrine effects of perfluorooctane sulfonate in rats*, Environ. Health. Persp. 111 (2003), pp. 1485–1489.
- [23] G.L. Kennedy, J.L. Butenhoff, G.W. Olsen, J.C. O'Connor, A.M. Seacat, R.G. Perkins, L.B. Biegel, S.R. Murphy, and D.G. Farrar, *The toxicology of perfluorooctanoate*, Crit. Rev. Toxicol. 34 (2004), pp. 351–384.
- [24] P.H. Roos, J. Angerer, H. Dieter, M. Wilhelm, D. Wölfle, and J.G. Hengstler, *Perfluorinated compounds (PFCs) hit the headlines: meeting report on a satellite symposium of the annual meeting of the German Society of Toxicology*, Arch. Toxicol. 82(1) (2008), pp. 57–59.
- [25] W.M. Henderson and M.A. Smith, *Perfluorooctanoic acid and perfluorononanoic acid in fetal and neonatal mice following in utero exposure to 8-2 fluorotelomer alcohol*, Toxicol. Sci. 95 (2007), pp. 452–461.
- [26] C.J. Wolf, S.E. Fenton, J.E. Schmid, A.M. Calafat, Z. Kuklenyik, X.A. Bryant, J. Thibodeaux, K.P. Das, S.S. White, C.S. Lau, and B.D. Abbott, *Developmental toxicity of perfluorooctanoic acid in the CD-1 mouse after cross-foster and restricted gestational exposures*, Toxicol. Sci. 95 (2007), pp. 462–473.
- [27] A. Karrman, J.F. Mueller, B. Van Bavel, F. Harden, L.M.L. Toms, and G. Lindstrom, *Levels of 12 perfluorinated chemicals in pooled Australian serum, collected 2002–2003, in relation to age, gender, and region*, Environ. Sci. Technol. 40 (2006), pp. 3742–3748.
- [28] K. Harada, N. Saito, K. Inoue, T. Yoshinaga, T. Watanabe, S. Sasaki, S. Kamiyama, and A. Koizumi, *The influence of time, sex and geographic factors on levels of perfluorooctane sulfonate and perfluorooctanoate in human serum over the last 25 years*, J. Occup. Health. 46 (2004), pp. 141–147.
- [29] K. Harada, A. Koizumi, N. Saito, K. Inoue, T. Yoshinaga, C. Date, S. Fujii, N. Hachiya, I. Hirosawa, S. Koda, Y. Kusaka, K. Murata, K. Omae, S. Shimbo, K. Takenaka, T. Takeshita, H. Todoriki, Y. Wada, T. Watanabe, and M. Ikeda, *Historical and geographical aspects of the increasing perfluorooctanoate and perfluorooctane sulfonate contamination in human serum in Japan*, Chemosphere 66 (2007), pp. 293–301.
- [30] Y.H. Jin, N. Saito, K.H. Harada, K. Inoue, and A. Koizumi, *Historical trends in human serum levels of perfluorooctanoate and perfluorooctane sulfonate in Shenyang, China*, Tohoku. J. Exp. Med. 212 (2007), pp. 63–70.
- [31] S.F. Mei, *Production and usage of PFOS and PFOA in China*, Organofluorine Ind. 1 (2008), pp. 21–25 [in Chinese].
- [32] Q.Q. Xie, *Related technical regulations and measurement of PFOS in the world*, Chin. Petrol. Chem. St. Qual. 28(2) (2008), pp. 22–27 [in Chinese].
- [33] China, Information Center of KA packaging market in China, Dynamic News, 2004. Available from: http://www. pka.com.cn/news/view.asp?id=3442.
- [34] Y. Xing, Y.L. Lu, R.W. Dawson, Y.Y. Shi, H. Zhang, T.Y. Wang, W.B. Liu, and H.C. Ren, *A Spatial temporal assessment of pollution from PCBs in China*, Chemosphere 60 (2005), pp. 731–739.
- [35] Q. Zhang, C.J. Zhang, Q. Zhou, and L. Chen, *Quantitative characterization of PFOS and PFOA in the surface water by SPE, HPLC/MS*, Sichuang Environ. 25(5) (2006), pp. 10–13 [in Chinese].
- [36] M.K. So, Y. Miyake, W.Y. Yeung, Y.M. Ho, S. Taniyasu, P. Rostkowski, N. Yamashita, B.S. Zhou, X.J. Shi, J.X. Wang, J.P. Giesy, H. Yu, and P.K.S. Lam, *Perfluorinated compounds in the Pearl River and Yangtze River of China*, Chemosphere 68 (2007), pp. 2085–2095.
- [37] Y.H. Jin, M. Ding, C. Di, L. Wang, G.H. Dong, W.Q. Shu, andY.H. Zhang, *Monitoring the status of PFOS and PFOA pollution in Sanxia reservoir area of the Yangtze river and surface water of Wuhan*, Ecol. Environ. l5 (3) (2006), pp. 486–489 [in Chinese].

176 *C. Chen* et al.

- [38] M.K. So, S. Taniyasu, N.Yamashita, J.P. Giesy, J. Zheng, Z. Fang, S.H. Im, and P.K.S. Lam,*Perfluorinated compounds in coastal waters of Hong Kong, South China, and Korea*, Environ. Sci. Technol. 38 (2004), pp. 4056–4063.
- [39] B. Liu, Y.H. Jin, Q.L. Yu, K. Wang, G.H. Dong, H.Y. Li, N. Saitou, and K. Sasaki, *Investigation of perfluorooetane sulfonate (PFOS) and perfluorooetanoic acid (PFOA) pollution in the surface water of the Songhua River*, Acta Scientiae Circumstantiae 27(3) (2007), pp. 480–486 [in Chinese].
- [40] Y.H. Jin, X. Liu, H.M. Qin, Y.X. Ma, Y.O. Fan, Y.H. Zhang, N. Saitou,K. Sasaki,and A. Koizumi, *The status quo of perfluorooctane sulfonate (PFOS) pollution in tap water and different waters in partial areas of China*, Chin. Environ. Sci. 24(2) (2004), pp. 166–169 [in Chinese].
- [41] Q.X. Zhou and X.G. Hu, *Researching progresses in environmental pollution behavior, toxic effects and mechanisms of PFOS/PFOA*, Chin. J. Environ. Sci. 28(10) (2007), pp. 2153–2162 [in Chinese].
- [42] N. Yamashita, K. Kannan, S. Taniyasu, Y. Horii, G. Petrick, and T. Gamo, *A global survey of perfluorinated acids in oceans*, Mar. Pollut. Bull. 51 (2005), pp. 658–668.
- [43] X.D. Ju, Y.H. Jin, K. Sasaki, and N. Saito, *Perfluorinated surfactants in surface, subsurface water and microlayer from Dalian Coastal waters in China*, Environ. Sci. Technol. 42 (2008), pp. 3538–3542.
- [44] N. Yamashita, K. Kannan, S. Taniyasu, Y. Horii, T. Okazawa, G. Petrick, and T. Gamo, *Analysis of perfluorinated acids at parts-per-quadrillion levels in seawater using liquid chromatography-tandem mass spectrometry*, Environ, Sci. Technol. 38 (2004), pp. 5522–5528.
- [45] X.L. Zhao, J.D. Li, Y.L. Shi, Y.Q. Cai, S.F. Mou, and G.B. Jiang, *Determination of perfluorinated compounds in wastewater and river water samples by mixed hemimicelle-based solid-phase extraction before liquid chromatography-electrospray tandem mass spectrometry detection*, J. Chromatogr. A. 1154 (2007), pp. 52–59.
- [46] E. Sinclair, D.T. Mayack, K. Roblee, N. Yamashita, and K. Kannan, *Occurrence of perfluoroalkyl surfactants in water, fish, and birds from New York State*, Arch. Environ. Con. Tox. 50 (2006), pp. 398–410.
- [47] N.P.H. Lien, S. Fujii, S. Takana, M. Nozoe, W. Wirojanagud, A. Anton, and G. Lindstrom, *Perfluorinated substances in tap water of Japan and several countries and their relationship to surface water contamination*, Environ. Eng. Res (Japanese Journal) 43 (2006), pp. 611–618.
- [48] S. Takagi, F. Adachi, K. Miyano, Y. Koizumi, H. Tanaka, M. Mimura, I. Watanabe, S. Tanabe, and K. Kannan, *Perfluorooctanesulfonate and perfluorooctanoate in raw and treated tap water from Osaka, Japan*, Chemosphere 72 (2008), pp. 1409–1412.
- [49] S.A. Beach, J.L. Newsted, K. Coady, and J.P. Giesy,*Ecotoxicological evaluation of perfluorooctanesulfonate (PFOS)*, Rev. Environ. Contam. Toxicol. 186 (2006), pp. 133–147.
- [50] C.A. Moody, J.W. Martin, W.C. Kwan, D.C.G. Muir, and S.A. Mabury, *Monitoring perfluorinated surfactants in biota and surface water samples following an accidental release of fire-fighting foam into Etobicoke Creek*, Environ. Sci. Technol. 36 (2002), pp. 545–551.
- [51] J.L. Newsted, J.P. Giesy, and K.K. Coady, *Avian toxicity reference values (TRVs), predicted no effect concentrations (PNECs) and water quality criteria for perfluorooctane sulfonate (PFOS)*, Project No. 178401; 3M Co.: St. Paul, MN, 2004.
- [52] P. Rostkowski, N. Yamashita, I.M.K. So, S. Taniyasu, P.K.S. Lam, J. Falandysz, K.T. Lee, S.K. Kim, J.S. Khim, S.H. Im, J.L. Newsted, P.D. Jones, K. Kannan, and J.P. Giesy, *Perfluorinated compounds in streams of the Shihwa industrial zone and Lake Shihwa, South Korea*, Environ. Toxicol. Chem. 25 (2006), pp. 2374–2380.
- [53] K. Hart, K. Kannan, L. Tao, S. Takahashi, and S. Tanabe, *Skipjack tuna as a bioindicator of contamination by perfluorinated compounds in the oceans*, Sci. Total Environ. 403 (2008), pp. 218–224.
- [54] A. Gulkowska, Q.T. Jiang, M.K. So, S. Taniyasu, P.K.S. Lam, and N. Yamashita, *Persistent perfluorinated acids in seafood collected from two cities of China*, Environ. Sci. Technol. 40 (2006), pp. 3736–3741.
- [55] M.K. So, S. Taniyasu, P.K.S. Lam, G.J. Zheng, J.P. Giesy, and N. Yamashita, *Alkaline digestion and solid phase extraction method for perfluorinated compounds in mussels and oysters from south China and Japan*, Arch. Environ. Con. Tox. 50 (2006), pp. 240–248.
- [56] J.Y. Dai, M. Li, Y.H. Jin, N. Saito, M.Q. Xu, and F.W. Wei, *Perfluorooctanesulfonate and perfluorooctanoate in red panda and giant panda from China*, Environ. Sci. Technol. 40 (2006), pp. 5647–5652.
- [57] K. Kannan, J. Koistinen, K. Beckmen, T. Evans, J. F. Gorzelany, K. J. Hansen, P. D. Jones, E. Helle, M. Nyman, and J. P. Giesy, *Accumulation of perfluorooctane sulfonate in marine mammals*, Environ. Sci. Technol. 35 (2001), pp. 1593–1598.
- [58] D. R. Taves, *Evidence that there were two forms of fluoride in human serum*, Nature 217 (1968), pp. 1050–1051.
- [59] L.W.Y. Yeung, M.K. So, G.B. Jiang, S. Taniyasu, N. Yamashita, M.Y. Song, Y.N. Wu, J.G. Li, J.P. Giesy, K.S. Guruge, and P.K.S. Lam, *Perfluorooctanesulfonate and related fluorochemicals in human blood samples from China*, Environ. Sci. Technol. 40 (2006), pp. 715–720.
- [60] K. Kannan, S. Corsolini, J. Falandysz, G. Fillmann, K.S. Kumar, B.G. Loganathan, M.A. Mohd, J. Olivero, N. Van Wouwe, J.H. Yang, and K.M. Aldous, *Perfluorooctanesulfonate and related fluorochemicals in human blood from several countries*, Environ. Sci. Technol. 38 (2004), pp. 4489–4495.
- [61] S. Masunaga, K. Kannan, R. Doi, J. Nakanishi, and J. P. Giesy, *Levels of perfluorooctane sulphonate (PFOS) and other related compounds in the blood of Japanese people*, Organohalogen. Compd. 59 (2002), pp. 319–322.
- [62] S. Taniyasu, K. Kannan, Y. Horii, N. Hanari, and N. Yamashita, *A survey of perfluorooctane sulfonate and related perfluorinated organic compounds in water, fish, birds, and humans from Japan*, Environ. Sci. Technol. 37 (2003), pp. 2634–2639.
- [63] K.S. Guruge, S. Taniyasu, N. Yamashita, S. Wijeratna, K.M. Mohotti, H.R. Seneviratne, K. Kannan, N. Yamanaka, and S. Miyazaki, *Perfluorinated organic compounds in human blood serum and seminal plasma: A study of urban and rural tea worker populations in Sri Lanka*, J. Environ. Monit. 7 (2005), pp. 371–377.
- [64] H. Fromme, O. Midasch, D. Twardella, J.Angerer, S. Boehmer, and B. Liebl, *Occurrence of perfluorinated substances in an adult German population in southern Bavaria*, Int. Arch. Occup. Environ. Health 80 (2007), pp. 313–319.
- [65] I. Ericson, M. Gomez, M. Nadal, B. van Bavel, G. Lindstrom, and J.L. Domingo, *Perfluorinated chemicals in blood of residents in Catalonia (Spain) in relation to age and gender: A pilot study*, Environ. Int. 33 (2007), pp. 616–623.
- [66] A. Karrman, I. Langlois, B. van Bavel, G. Lindstrom, and M. Oehme, *Identification and pattern of perfluorooctane sulfonate (PFOS) isomers in human serum and plasma*, Environ. Int. 33 (2007), pp. 782–788.
- [67] G.W. Olsen, D.C. Mair, W.K. Reagen, M.E. Ellefson, D.J. Ehresman, J.L. Butenhoff, and L.R. Zobel, *Preliminary evidence of a decline in perfluorooctanesulfonate (PFOS) and perfluorooctanoate (PFOA) concentrations in American Red Cross blood donors*, Chemosphere 68 (2007), pp. 105–111.
- [68] C. Kubwabo, N. Vais, and F.M. Benoit, *A pilot study on the determination of perfluorooctanesulfonate and other perfluorinated compounds in blood of Canadians*, J. Environ. Monit. 6 (2004), pp. 540–545.
- [69] A. Karrman, B. van Bavel, U. Jarnberg, L. Hardell, and G. Lindstrom, *Perfluorinated chemicals in relation to other persistent organic pollutants in human blood*, Chemosphere 64 (2006), pp. 1582–1591.
- [70] D.J. Ehresman, J.W. Froehlich, G.W. Olsen, S.C. Chang, and J.L. Butenhoff, *Comparison of human whole blood, plasma, and serum matrices for the determination of perfluorooctanesulfonate (PFOS), perfluorooctanoate (PFOA), and other fluorochemicals*, Environ. Res. 103 (2007), pp. 176–184.
- [71] W.K. Reagen, M.E. Ellefson, K. Kannan, and J.P. Giesy, *Comparison of extraction and quantification methods of perfluorinated compounds in human plasma, serum, and whole blood*, Anal. Chim. Acta. 628 (2008), pp. 214–221.
- [72] E.A. Emmett, F.S. Shofer, H. Zhang, D. Freeman, C. Desai, and L.M. Shaw, *Community exposure to perfluorooctanoate: relationships between serum concentrations and exposure sources*, J. Occup. Environ. Med. 48 (2006), pp. 759–770.
- [73] K. Harada, N. Saito, K. Sasaki, K. Inoue, and A. Koizumi, *Perfluorooctane sulfonate contamination of drinking water in the Tama River, Japan: Estimated effects on resident serum levels*, Bull. Environ. Contam. Toxicol. 71 (2003), pp. 31–36.
- [74] M.K. So, N. Yamashita, S. Taniyasu, Q.T. Jiang, J.P. Giesy, K. Chen, and P.K.S. Lam, *Health risks in infants associated with exposure to perfluorinated compounds in human breast milk from Zhoushan*, China, Environ. Sci. Technol. 40 (2006), pp. 2924–2929.
- [75] W. Volkel, O. Genzel-Boroviczeny, H. Demmelmair, C. Gebauer, B. Koletzko, D. Twardella, U. Raab, and H. Fromme, *Perfluorooctane sulphonate (PFOS) and perfluorooctanoic acid (PFOA) in human breast milk: Results of a pilot study*, Int. J. Hyg. Environ. Health 211 (2008),pp. 440–446.
- [76] L. Tao, K. Kannan, C.M. Wong, K.F. Arcaro, and J.L. Butenhoff, *Perfluorinated compounds in human milk from Massachusetts, USA*, Environ. Sci. Technol. 42 (2008), pp. 3096–3101.
- [77] L. Tao, J. Ma, T. Kunisue, E.L. Libelo, S. Tanabe, and K. Kannan, *Perfluorinated compounds in human breast milk from several Asian countries, and in infant formula and dairy milk from the USA*, Environ. Sci. Technol. 42 (2008), pp. 8597–8602.
- [78] A. Karrman, I. Ericson, B. van Bavel, P.O. Darnerud, M. Aune, A. Glynn, S. Lignell, and G. Lindstrom, *Exposure of perfluorinated chemicals through lactation: Levels of matched human milk and serum and a temporal trend, 1996–2004, in Sweden*, Environ. Health Persp. 115 (2007), pp. 226–230.
- [79] G.W. Olsen, H.Y. Huang, K.J. Helzlsouer, K.J. Hansen, Butenhoff, J.L., and J.H. Mandel, *Historical comparison of perfluorooctanesulfonate, perfluorooctanoate, and other fluorochemicals in human blood*, Environ. Health Persp. 113 (2005), pp. 539–545.