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Review

# Environmental contamination, human exposure and body loadings of perfluorooctane sulfonate (PFOS), focusing on Asian countries

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## HIGHLIGHTS

- In Asia, PFOS were mostly detected in water systems, wild mammals and seafood.
- Higher concentrations were detected in human samples in China, Japan and Korea.
- Major human exposure pathways of PFOS were via oral intake and inhalation.
- Concentrations were associated with gender and dietary habit (seafood consumption).

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## ABSTRACT

Perfluorinated compounds (PFCs) are man-made fluorinated hydrocarbons, which are very persistent in the environment. Since the early 1980s, the usage of PFCs has sharply increased for a wide array of industrial and commercial applications. Being the most important PFC, perfluorooctane sulfonate (PFOS) has received much attention. In the past decades, increasing surveys have been focused on this compound, to study its sources, fates and effects in the environment. According to the large production volume and wide usage in industrial and commercial products in the past, PFOS can be detected in various environmental media and matrix, even in human tissues. This article attempted to review the current status of PFOS contaminations in Asia, focusing on water systems, sediments, wide animals and human tissues. A special section is devoted to examine the pathways of human exposure to this compound, as well as human body loadings of PFOS and their possible association with diseases.



**Contents**



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## 1. Introduction

Perfluorinated compounds (PFCs) are man-made fluorinated hydrocarbons, which have been manufactured by the 3M Company since the late 1940s (3M, 1999a). They include a large group of chemicals which are characterized by a fully fluorinated hydrophobic linear carbon chain, attached with different hydrophilic functional groups (Fromme et al., 2009). These products possess distinctive surface-active properties, which allowed their wide applications in coating formulations, fire-fighting foams and lubricants. The strong C-F bond formed inside provides PFCs ability of resisting various thermal, chemical and biological degradation (Kissa, 2001), and render PFCs very persistent in the environment.

Recently, PFCs have received worldwide attention because they are frequently detected in various environmental matrices, even in the Arctic region (Chaemfa et al., 2010; Sonne, 2010). Among these compounds, PFOS is the most representative and commonly studied one, because its persistence, with over 8 years of half-life when remained in human bodies, its unique chemical properties allows diverse applications in fabric, leather and paper surface protection, as well as fire fighting foams, mining and oil well surfactants (OECD, 2002). Recently, PFOS has been added to Annex B of the Stockholm Convention on Persistent Organic Pollutants in May 2009 (Geneva: Stockholm Convention Secretariat, 2009).

Perfluorooctane sulfonyl fluoride (POSF) is also considered as a synthetic PFC. It can synthesize PFOS directly or indirectly through chemical or enzymatic hydrolysis (3M, 1999b). POSF-derived products can be ultimately degraded to PFOS. In 2002, 3M Company (it is the dominant global POSF manufacturer) manufactured about 3665 t POSF, accounted for ~80% of the estimated global POSF production of 4650 t between 1970 and 2002, the two largest production sites are in the United States and Belgium, followed by Italy, and small scale manufacturers in Japan and Germany (OECD, 2002), with some smaller ones distributed in Europe (6, with 4 in EU member states), Asia (6, with 4 in Japan) and South America (1) (Paul et al., 2009).

Before 2003, PFOS was the major ingredient of Scotchgard, a fabric, furniture and carpets protector made by 3M Company. However, PFOS was replaced by perfluorobutane sulfonate (PFBS) after 2003 because it was found to be harmful to the environment (Renner, 2006). PFOS can be commercially synthesised by two major processes: electro-chemical fluorination (ECF) (3M, 1999a), and telomerization (Schultz et al., 2003).

The existence of PFOS in the environment arises from direct and indirect sources. The direct sources are derived from manufacture and application of PFOS, POSF and their-derived products and byproducts, which generated a large amount of waste in aqueous and solid form, with most of the solids eventually migrated into the aqueous form (Paul et al., 2009). The indirect sources come from chemical reaction impurities or breakdown from its precursor. It has been estimated that 85% of indirect emissions are resulted from the release from consumer products during use and disposal (3M, 2000). Due to their global distribution and toxicity, productions of PFOS and their related products were ceased in 2002 by the 3M Company (3M, 2008).

Fluorinated organic compounds (FOCs), especially perfluorinated compounds (PFCs), have received less attention than other halogenated compounds, such as chlorinated and brominated compounds, due to the great difficulty in measurement (Giesy and Kannan, 2001). Belisle and Hagen (1978) described a procedure for the determination of total fluorine in biological samples by gas chromatography (GC), including whole blood, serum as well as plasma samples, but only ionic or acid-labile fluoride can be measured. Two years later, Belisle and Hagen (1980) successfully quantitated perfluorooctanoic acid (PFOA) in plasma, urine and liver tissue by GC with electron capture detection (ECD) for the first time. Similarly, by using GC, Yamamoto et al. (1989) detected the free ionic, ionizable, and organic fluorine in human whole blood and serum samples, and subsequently Ylinen and Auriola (1990) efficiently analyzed C9 perfluorinated carboxylic acids (PFCAs) in rats serum and tissue samples by GC with flame ionization. Later on, a method for C8-C10 PFCAs determination in biota tissue was developed using gas–liquid chromatography (GLC) with ECD by Kudo et al. (1998). At the same year, a method for the quantitative determination of C6-C10 PFCAs in biota samples was described by using high-performance liquid chromatography (HPLC) with fluorescence detection, which is more sensitive than the GC methods reported previously (Ohya et al., 1998). Historically, low-level detection of fluorochemicals has been limited to relatively insensitive or nonmass-specific detection methods, until 2001, Hansen et al. (2001) developed a new method for several low-level (ng $mL^{-1}$ or ng $g^{-1}$) PFCs in sera and liver tissue, using high-performance liquid chromatography with electro-spray tandem mass spectrometry (HPLC/MS/MS). Since then, a number of studies have been conducted, based on this analytical technique.

Most of the PFOS extraction methodologies for solid samples, such as food, sediment, biota and human tissue samples, involved ion-pairing extraction, followed by clean up steps by solid phase extraction (SPE) and ENVI-Carb (Moody and Field, 1999; Powley et al., 2005), besides the alkaline digestion was also applied for the biota sample extraction (So et al., 2006a). For liquid samples, including water and human milk, the SPE was usually directly used as extraction and cleanup method. The different extraction and clean-up method, extraction solvent and tubes, detection equipment used, as well as the laboratory contamination background, are the factors that led to limit of detections (LODs) which caused discrepancies in different studies. These differences would effectively influence the calibration, detection and final average concentration among studies. In the present review, there were two problems when PFOS concentration are summarized and compared among different studies: the different LOQs and recoveries for each research. To lower the LOQs, most studies used PP (polypropylene) and stainless steel vessels during sample collection, preparation and analyzing, and the accessible fluoropolymer tubes/parts of HPLC were replaced by PEEK (polyether ether ketone) to minimize the background signals (Yamashita et al., 2004). On the other hand, increasing the sample amount or decreasing the final extract volume was also found to be helpful, however this higher enrichment factor of the samples can increase the instrumental matrix effects, which can cause to

ion-suppression or enhancement of mass spectrum signal. The recovery of PFOS for each study reviewed was different, even for similar samples, and most authors did not mention if the final concentration for the recoveries was corrected. Due to this circumstance, the concentration comparison in this review were done while ignoring the recoveries difference.

In the previous reviews carried out, Houde et al. (2006) summarizes and compared the PFCs concentration and contamination profiles among wildlife and human samples, evaluated the bioaccumulation/biomagnifications in the environment, and discussed possible sources. While Martin et al. (2010) focused on the prediction of PFOS exposure models through manufacturing, emissions and degradation mechanisms of its various forms of precursor (PreFOS), as well as exposure trends, environmental monitoring and biomonitoring of PreFOS and PFOS. D'eon and Mabury (2011) discussed the PFCs production, human exposure sources and trends, as well as the biotransformation processing. With the aforementioned related reviews in mind, the major aim of this study was to review PFOS contamination in Asia up to 2010, with emphasis on studies related to their concentrations detected in different environmental media, such as water, air, sediment, and wild animals. An attempt was also made to encapsulate the pathways of human exposure to PFOS, based on their human body loadings, and their potential health impacts. Besides, the trends of PFOS were also discussed.

## 2. Levels of PFOS contamination in different environmental media and biota

### 2.1. Water systems

During the manufacturing process, PFOS can be released into the environment, mainly in solid forms, of which about 80% will finally deposit in water (3M, 1999b). It has been found in water and sediment samples collected from interior rivers to deep sea oceans, as well as in snow water from highly developed urban areas to the Arctic region. Therefore, monitoring PFOS in water system may provide the information on its long-range atmospheric and hydrospheric transport from continental source regions. This is the main reason that scientists around the world have been engaged in detecting PFOS in water systems, including surface and ground freshwater, seawater and precipitations.

### 2.2. Fresh and sea water

In Asia, there are six plants producing and supplying PFOS related substances, with 4 of them located in Japan (Paul et al., 2009). In past researches, Japan did the most comprehensive surveys of water, especially in Tokyo Bay. With relatively sufficient data collected from this region, the model of sources, diffusion and fate of PFOS in industrialized and urbanized area can be built up. Tokyo Bay receives water discharges from several rivers, including the river flow through suburban and metropolitan areas of Tokyo (Taniyasu et al., 2003). In this study, higher PFOS concentration was found in Tokyo Bay (8 to 59 ng $L^{-1}$, mean: 26 ng $L^{-1}$) area than other coastal locations in Japan, indicating that one of the PFOS sources in surface and coastal waters may concentrate in industrial and metropolitan region. This hypothesis was proved by the fact that PFOS was detected at the similar range in Tokyo Bay (12.7–25.4 ng $L^{-1}$) (Yamashita et al., 2004) and 10 Tokyo Rivers (0.5–58 ng $L^{-1}$) during 2004–2005 (Takazawa et al., 2009), as well as it was found in all rivers and secondary effluents in Tokyo Bay (42–635 ng $L^{-1}$) in 2005, with secondary effluents much higher than rivers (Murakami et al., 2008). Generally, the lower PFOS concentration was found in the rural area while the higher ones was located in the downstream sites, in addition, the discharge of municipal wastewater has been shown to be an important route into the Tokyo Bay water system (Takazawa et al., 2009). Later on, for the first time, Ahrens et al. (2009) reported the PFOS concentration in pore water sample in Tokyo Bay. Interestingly, PFOS was found to be on the basis of the same distribution in all sample pore ($n = 2$): it was slightly lower in the surface water, and increased according to the depth, after reaching their maximum, they rapidly decreased (lower than the surface), and more importantly, in the deeper layer, PFOS was the only PFCs that can be found in the water sample (Ahrens et al., 2009). Sakurai et al. (2010) collected seawater from Tokyo Bay during 2004–2006 and similarly found that the PFOS concentration in the lower layer (2.5 ng $L^{-1}$, <0.4–14 ng $L^{-1}$) was slightly less than that in the upper layer (5.5 ng $L^{-1}$, 0.78–17 ng $L^{-1}$). Additionally, the authors found there is a negative correlation between the PFOS concentration and salinity, indicating the freshwater inputs were the predominant source of PFOS to the Tokyo Bay (Sakurai et al., 2010). Up to 2010, based on the horizontal and vertical PFOS data collected in Tokyo Bay, two clear clues can be concluded in developed and urban areas, one is that fresh water was one of the most important sources of PFOS, which came from industrial wastewater and domestic sewage; the other is PFOS contamination is widespread across Japan, even in the deep ocean water. This finding can be further proved by the study of PFOS was detectable in all groundwater and spring samples in Tokyo (0.28–133 ng $L^{-1}$) (Murakami et al., 2009). However, the former transport was due to the ocean current, while the later one owing to leakage of the contaminated wastewater or street runoff. Although the 3M Company announced PFOS and its related products were phased out since 2002, their overseas manufacturing was still in progress. For example, time trend study of PFOS concentration was conducted in Kamogawa River (Japan) in the year of 2004 and 2007. It was found that the PFOS concentration detected in 2007 (4.1–10 ng $L^{-1}$) (Senthilkumar et al., 2007) was 100-fold higher than the data obtained by a previous study (0.09–0.14 ng $L^{-1}$) (Saito et al., 2004). Factors such as consistent sources and rising production all contributed to the PFOS pollution in Kamogawa River sharply increasing during the short period.

In China, fluorochemical manufacturing facilities are mainly concentrated in central China right now, pond and effluent water was supposed to contain much more pollutants around these plants. The effluents flow out from fluorochemical manufacturing facilities contributes the majority of PFOS contamination, which may also be responsible for the high PFOS concentration in surrounding river. For example, around a manufacture plant in Wuhan (Yangtze River pass by) (Wang et al., 2010a), a very high concentration of PFOS was found in the effluents from the wastewater treatment pond (mean: 1021 μg $L^{-1}$) and pond water near plant less than 200 m away (mean: 984 ng $L^{-1}$), although in the pond water more than 500 m of the plant, the PFOS concentration falls to the 14.1 ng $L^{-1}$, a similar distribution pattern and concentration level was also detected in Taiwan, in a semiconductor fabrication plant, waste water, final effluent and pure water contained PFOS of 12566670, 128670 and 36.7 ng $L^{-1}$, respectively, and in nearby river system it was found to be 45–5440 ng $L^{-1}$ (Lin et al., 2009). It is evident that PFOS cannot be effectively removed from industrial wastewater, a plant located in the upstream of Yangtze River, Pan and You (2010) revealed that PFOS concentrations in downstream of Yangtze River Estuary ranged from 36.3–703.3 ng $L^{-1}$, which is much higher than that in other Asian river and sea water systems, even higher than the downstream (75–144 ng $L^{-1}$) of Tennessee River, in which there was also a manufacturing plant. Additionally, PFOS cannot be effectively removed by the wastewater treatment, PFOS contamination in this area may come from the domestic sewerage from the nearby metropolitan instead of the PFOS releasing from manufacturing wastewater.

**Table 1**
The comparison of PFOS concentration in water samples from China.

| Location | Sample type | PFOS concentration ($ng\ L^{-1}$) | References |
|---|---|---|---|
| Dalian | Coastal water | nd-0.96 | Ju et al. (2008) |
| Yangtze River | Riverwater | <0.01–14.0 | So et al. (2007) |
| Pearl River, Guangzhou | Riverwater | 0.90–99.0 | |
| PRD, Hong Kong, South China | Coastal water | 0.02–12 | So et al. (2004) |
| Jilin Province | Surfacewater | 0.41 | Jin et al. (2004) |
| Liaoning Province | Surfacewater | 1.98 | |
| | Groundwater | 0.32–1.32 | |
| Shandong Province | Surfacewater | 4.2 | |
| | Groundwater | 0.82–3.96 | |
| | Riverwater | 1.68 | |
| Yangtze River Estuary | Riverwater | 36.3–703 | Pan et al. (2009) |
| Three Gorges Reservoir Area | Riverwater | 0.10–37.8 | Jin et al. (2006) |
| Wuhan | Surfacewater | 2.30–25.5 | |

The authors also observed that the concentration of PFOS was high (143.9 $ng\ L^{-1}$) at low salinity area, but relatively low (47.1 $ng\ L^{-1}$) at high salinity area, except for the reason that freshwater inputs were the predominant source of PFOS, but also due to the fact that the solubility of PFOS is 40 times higher in freshwater than that in seawater (Moore, 2003). However in the Pearl River Estuary (South China), owing to there are no manufacturing plants around this area, the PFOS concentration detected in Hong Kong coastal water and open sea of South China showed much lower value (0.02–12 $ng\ L^{-1}$) (So et al., 2004). The concentration of PFOS from seawater collected from Victoria Harbor (central part of Hong Kong) was moderately high, as the seawater within the harbor (a semi-enclosed bay) is primarily influenced by domestic sewage and industrial effluent from the nearby highly urbanized and industrialized areas of Hong Kong Island and Kowloon (So et al., 2004). In addition, higher concentrations were detected in winter than that in summer (So et al., 2004), this seasonal difference was also recognized by Tsuda et al. (2010) in the river of Japan. Table 1 compares PFOS concentrations in water samples collected from different parts of China. Overall, PFOS concentrations in north China were lower than that in central, east and south China. The highest concentrations measured in Songhua River, Liaoning, Jiling and Shandong Province (all in north China) were less than 8.04 $ng\ L^{-1}$, while much higher concentrations were noted in the Pearl River (99 $ng\ L^{-1}$) and Guangzhou (both south China), and Yangtze River Estuary (701 $ng\ L^{-1}$) (central China).

In South Korea, PFOS was detected in stream and lake of Shihwa industrial zone, ranging from 2.24 to 651 $ng\ L^{-1}$ (Rostkowski et al., 2006); whereas the surface waters from estuarine and coastal areas of South Korea had a range of 4.11–450 $ng\ L^{-1}$ (Naile et al., 2010). Based on these results, it seems that PFOS contamination in South Korea is more serious than Japan, as the values detected matched the Japanese secondary effluents (Murakami et al., 2008), and the Chinese Yangtze River Estuary, a grossly polluted water body, which receives discharges from this industrialized and urbanized mega delta (Pan and You, 2010). Except for the highly populated and developed areas, PFOS analysis was also conducted in some relatively rural sites: in India, the PFOS concentrations in water systems were all lower than 3.91 $ng\ L^{-1}$ (Yeung et al., 2009b), and it was <0.2–5 $ng\ L^{-1}$ in the river of Thailand (Kunacheva et al., 2010), these values reach as low as the Lake water collected from Canadian Arctic (1.2–1.8 $ng\ L^{-1}$) (Butt et al., 2010), comparatively lower than those reported in other Asian countries.

Overall, being an environmental sink of pollutants, the monitoring of PFOS in river and sea water clearly indicates the source and distribution of this chemical; it has large sample volume and is convenient to sample, so the study in this field provides the most sufficient data for PFOS contamination. Nevertheless, there is still a lot to learn about the presence the distribution of PFOS, particularly in undeveloped and developing region of Asia, which can provide more spatial and temporal records for the future work.

### 2.3. Precipitation

Precipitation is considered as the most effective scavenger for removing particulates and atmospheric contaminants (Omer Ali, 2005). PFOS is a water-soluble compound, it can dissolve in cloud droplets, besides, it also adsorb to the particulates surface in the atmosphere (Kwok et al., 2010). The concentrations of these pollutants can reflect local atmospheric contamination directly. Analyzing of PFOS concentration in precipitation, including rain and snow water, were centralized in China, Japan and India. In 2009, Liu et al. (2009c) determined the PFOS concentration in snow and rain in Dalian, which is a coastal city located in north eastern of China. The snow and rain samples in that study were collected during 2006–2007. PFOS was found in all snow samples, ranged from 37.5–182 (mean: 145) $ng\ L^{-1}$ while rain samples were from 9.92 to 113 $ng\ L^{-1}$ (Liu et al., 2009c). Interestingly, PFOS concentration was roughly the same in snow and rain samples, this may suggest that PFOS exists and is transported in precipitation form, and not related with the temperature and state (liquid or solid). Moreover, the precipitation in Dalian contained the much higher PFOS concentration than all later studies in Asia. The authors explains that there are no fluorochemical manufacturing facilities in Dalian, and the high concentration may resultant from local industrial and domestic contamination and transport via the atmosphere from other areas, however, these do not appear to be the main reasons because Shenyang contains much lower PFOS concentrations in its precipitation samples (<0.38–51 (mean: 5.4) $ng\ L^{-1}$) (Liu et al., 2009b) and it is adjacent to Dalian and has similar urbanization and industrial development patterns with it. Nevertheless, the precipitation in these two cities are still much heavier than that in Tsukuba (residential area) and Kawaguchi (industrial area) of Japan, Hong Kong (business area of China) and Patna (India agricultural area). PFOS were <0.1–1.34 (mean: 0.36) $ng\ L^{-1}$ in Tsukuba and <0.1–4.21 (mean: 0.81) $ng\ L^{-1}$ in Kawaguchi, while in Hong Kong it was <0.1–0.7 (mean: 0.32) $ng\ L^{-1}$ and <0.1–0.08 (mean: 0.04) $ng\ L^{-1}$ in Patna (Kwok et al., 2010). Therefore, it is necessary to find out the potential resources in northern China cities, especially in Dalian, such as various monitoring need to be done in different sites and water body of the cities. During the same study, the authors also analyzed the PFOS concentration in USA (<0.1–0.64 (mean: 0.27) $ng\ L^{-1}$ in Slingerlands (business area) and <0.1–0.29 (mean: 0.18) $ng\ L^{-1}$ in Downtown Albany (residential area)) and France (<0.1–0.23 (mean: 0.11) $ng\ L^{-1}$ in Toulouse (business area) (Kwok et al., 2010). Except for the agricultural area in India which contained relatively lower concentration, the PFOS in other cities were at the same level, although it was slightly higher in the Japanese industrial area than that of other residential and business areas. The PFOS concentrations in this study were much lower than that of Dalian (179–3625 folds in mean value) and Shenyang (7–135 folds in mean value).

### 2.4. Sediments

During the past 5 years, there has been an increase in number of reports concerning PFCs in fresh and coastal water sediments, which may act as an important sink and reservior for most pollutants including PFOS. In Japan, a study showed that the Kamogawa

River sediments (south of Japan) contained PFOS ranging between <0.33–11 ng $g^{-1}$ dry weight (dw) (Senthilkumar et al., 2007). However, more studies were conducted on marine sediments, e.g. Tokyo Bay, where it is the most populated area in Japan, and the estimated annual input of PFOS form the local rivers was 74–346 kg $year^{-1}$. It was observed at 0.3–0.9 ng $g^{-1}$ dw (Ryosuke and Shigeki, 2006) and 0.45–1.79 ng $g^{-1}$ dw (Zushi et al., 2010) at 2004; 0.06–3.6 ng $g^{-1}$ dw during 2004–2006 (Sakurai et al., 2010). The range of 0.09–0.14 ng $g^{-1}$ dw PFOS was detected in other Japanese marine sediments of Ariake Sea (Nakata et al., 2006). Generally, there is no big difference in the PFOS concentration between fresh water and seawater sediment, but the sample was slightly higher in river than that from marine. In the research on Tokyo Bay core sediment in 2008, the largest concentration was found in the 0–15 cm depth (0.095–0.128 ng $g^{-1}$ dw, deposit between 1998 and 2008), then sharply fall into 0.039 ng $g^{-1}$ dw in 17 cm depth, until 79 cm the PFOS concentration was roughly range between 0.01–0.02 ng $g^{-1}$ dw (Ahrens et al., 2009). Also in the deep seawater detection in Suruga Bay (800–850 m undersea), Tosa Bay (50–200 m undersea) and Nankai Tough (4010 m undersea), the profile of PFOS distribution was roughly the same: became low as the water depth became deeper, but in Suruga Bay, it was high in the 0–5 cm core depth and then decreased until 30.5 cm (Harino et al., 2009). The results were in line with the observation made in Yangtze River Estuary, China (Pan et al., 2009), with 536.7 ng $g^{-1}$ dw at 0–17 cm depth, then decreased to 138.1 ng $g^{-1}$ dw at 34–50 cm. This phenomenon reflects the historical enrichment of PFOS in sediments. The data indicated freshwater sediments contained the highest concentrations (Kamogawa River), followed by marine sediments of Tokyo Bay, while the marine sediments of the open sea (Tosa Bay) contained the lowest concentrations, which reflected the intensity of human activities.

In China, most sediment research was done in the Huangpu River, because it is adjacent to Shanghai City (largest metropolis in east China), and is the Yangtze River Estuary (longest river in China). When sampling in 2007, Li et al. (2010) detected 1.57–8.78 ng $g^{-1}$ dw PFOS in samples collected from Huangpu River, this value was decreased after 2 years later, which is <0.03–0.46 ng $g^{-1}$ dw (Bao et al., 2010), the authors also found the relevant but slightly higher concentration in Zhujiang River (south China) at <0.03–3.1 ng $g^{-1}$ dw. These two sampling reveal roughly the same level of PFOS in Huangpu River, strangely, PFOS contamination seems to be more serious in central China estuarine in the year of 2008, in which PFOS concentration was obtained in the range of 72.9–536.7 ng $g^{-1}$ dw in all sediments of it (Pan et al., 2009). Pan et al. (2009) and Li et al. (2010) share the same extraction method and all of them using HPLC/MS/MS to do the analysis samples, but the concentration variation was largely different within 2007–2009, so the further research needs to be conducted to find out in which level the PFOS contamination is at. So far it is uncertain why the PFOS concentration in Huangpu River sharply increased a hundred and a thousand times, which may be resultant from the acute large quantity of PFOS or its precursor releasing comes from fluorochemical plant located in the middle portion of Yangtze River during 2008. In the same study, it was observed that salinity was an important parameter to determine PFOS concentration, with relatively high concentration of PFOS contained in sediment (196.2 ng $g^{-1}$ dw), at higher salinity, but lower concentration at lower salinity (72.9 ng $g^{-1}$) (Pan et al., 2009); later on, You et al. (2010) improved this observation, the authors found that sorption and desorption of PFOS on sediment were significantly dependent on salinity. In the same year (2008), Bao et al. (2009) also revealed the range of <0.12–0.37 ng $g^{-1}$ dw PFOS in Daliao River sediments of north China, the pollution level was similar as in Huangpu River in 2007 and 2009, as well as in Zhujiang River, so the highly PFOS contaminated in Huangpu River can be suggested as an exception. PFOS concentrations detected in sediments samples collected from estuarine and coastal areas of South Korea were generally less than the 2.0 ng $g^{-1}$ dw (Naile et al., 2010). In general, sediments collected from Japan, China and South Korea were considered low (below 11 ng $g^{-1}$, dw), except the sediments from Yangtze River Estuary of China (72.9–536.7 ng $g^{-1}$, dw). The low concentration of PFOS in sediment sample may be due to the relatively higher water solubility of this compound and its consequent partitioning to the water. As the important portion of the water system, PFOS can be deposited into sediment naturally, the sorption and desorption of PFOS in sediment can be influenced by many factors, such as salinity, pH and cationic and anionic surfactants, besides hydrophobic partitioning was also found was the main function group to the sorption of PFOS in sediment (Pan et al., 2009; You et al., 2010).

Overall, the assessment of PFOS in the media of water system clearly reflect the widespread distribution of this chemical. However, further studies and monitoring are needed, particularly in newly developing fluoropolymer manufacturer and undeveloped areas of Asia. Through monitoring the PFOS concentration within these regions, a clear map of PFOS accumulation and fluctuation in Asia can be drawn up to find out more about the transport and fate of PFOS in environmental system.

### 2.5. Wild animals

Since early 1980s, the production and usage of PFOS increased dramatically, and they have been detected in wild animals near urbanized and industrial regions. However, recent studies showed that PFOS were also detected in animals thriving in remote areas, such as polar bears and water bird eggs from open sea (Holmstrom et al., 2005; Kannan et al., 2005; Smithwick et al., 2006). PFOS were primarily found in the liver, serum and kidneys of wild animals (Volkel et al., 2008), with liver and blood regarded as preferred tissues in monitoring PFOS in wildlife. Wild animal tissue samples can be extracted when in wet or dry conditions after freeze or air dry, so the existing paper usually calculate the sediment PFOS concentration in ng $g^{-1}$ dw (dry weight) and ng $g^{-1}$ ww (wet weight).

### 2.6. Mammals

Dolphins and porpoise are the top predators in the marine ecosystem, they may accumulate PFOS from surroundings or through consumption of the preys. Liver samples of melon-headed whales were collected in 1982, 2001/2002 and 2006, which covered periods of before, during and after the POSF-based production phase-out. PFOS can be detected in all liver samples above limit of detections (LODs). In 1982, the PFOS concentrations in liver samples were ranged from 4.6 to 10.2 (mean: 7.6) ng $g^{-1}$ ww, while these values were ranged between 17.8–117 (mean: 51.1) ng $g^{-1}$ ww in 2001/2002 samples and 49.2–60.6 (54.1) ng $g^{-1}$ ww in 2006 samples (Hart et al., 2008a). Obviously, PFOS concentrations in 2001/2002 and 2006 samples were much higher than in 1982, in which PFOS-related products only flourished nearly 10 years; after decades of manufacturing, its concentration were nearly 7-fold higher than in 1982. Additionally, the values were at the same level in 2001/2002 and 2006 because of PFOS was phased-out by the major manufacturer at 2003. Yeung et al. (2009a) collected the liver samples of Indo-Pacific humpback dolphin and finless porpoise during 2003 and 2007 from Hong Kong (south China), and analyzing the PFOS concentrations of them. PFOS was found to be in all dolphin and porpoise livers at concentrations ranging from 26–693 (mean: 251) and 51.3–262 (mean: 151) ng $g^{-1}$ ww, respectively, but there was no significant difference between these two species (Yeung et al., 2009a). In the same year, Yeung et al. (2009b) also determined the PFOS

concentration of dolphin liver (mean: 27.9 ng $g^{-1}$ ww) in Ganges River of India. Moon et al. (2010) noted all the liver samples of minke whales contained PFOS 2.8–162 ng $g^{-1}$ ww and long-beaked common dolphins 18–152 ng $g^{-1}$ ww, based on a survey conducted in South Korean coastal waters, in 2006 (Moon et al., 2010). Fluoropolymer manufacturing does not exist in India, the PFOS contamination here is relatively low when compared with other Asian countries, such as Japan and China, its concentration in water system is similar or even lower than that of remote marine locations, as a result, the mean concentration of 27.9 ng $g^{-1}$ ww in Indian dolphin liver were much lower (5–9-fold) than that in Japanese areas, even lower than the whale's liver (mean: 37.3 ng $g^{-1}$ ww) collected from the Arctic marine organisms of Canada (Kelly et al., 2009). Being both a predator in the marine ecosystem, the POPs accumulation pattern of dolphin and whale are similar, so it is expected that the PFOS in these two mammals are roughly in the same level.

For blood samples, PFOS were detected in serum samples of 27 individuals of red panda (20.36 ± 4.35, 0.80–73.80 μg $L^{-1}$) and giant panda (11.10 ± 2.01, 0.76–19.0 μg $L^{-1}$), collected from seven different locations of China (Dai et al., 2006). The data showed there was no age- or sex- related differences observed between PFOS and panda sera, but variation was significant (92-fold among samples), depending on the panda species and sample locations (Dai et al., 2006). The difference in PFOS concentration in animal body may arise from less exposure and/or better metabolism and excretion ability of PFOS (Yeung et al., 2006). Studies related to PFOS in the serum of other wild animals were also conducted and the results showed that PFOS in serum of Amur tiger ranged 0.10–9.82 ng $mL^{-1}$, Bengal tiger 0.863–1.43 ng $L^{-1}$, and African lion 2.24–3.03 ng $mL^{-1}$, with the concentrations tested in two species of tigers similar, but almost 2-fold lower than African lion (Li et al., 2008a,b).

### 2.7. Bird

In addition to wild mammals, PFOS are also widely distributed in tissues (liver, blood and egg) of wild birds. In 2002, liver samples of six species of birds collected from 1997–1999 across Japan contained PFOS <19–450 ng $g^{-1}$ ww, compared to those of 10 species collected from 1993–1994 over South Korea <10–500 ng $g^{-1}$ ww. No significant correlations were noted between sex or length/weight with PFOS in this study (Kannan et al., 2002). A later study conducted in 2003 (Taniyasu et al., 2003), obtained 11–167 ng $mL^{-1}$ and 68–1200 ng $g^{-1}$ ww of PFOS in bird whole blood and liver, respectively, and the values were substantially higher than those reported in the early studies (Kannan et al., 2002). Three species of waterbird egg samples collected from Hong Kong (2006), Xiamen and Quanzhou Bay (2004) for PFOS analyzing, ranged from 14.4 to 343 ng $g^{-1}$ ww (Wang et al., 2008). The wide range of concentration may depend on the different species waterbird instead of location: night heron (mean: 127 ng $g^{-1}$ ww) has the relatively high PFOS concentration than other two species (little egret (mean: 58 ng $g^{-1}$ ww) and great egret (mean: 20.4 ng $g^{-1}$ ww)). Compared to the domestic fowl eggs (mean: 0.08 ng $g^{-1}$ ww (chick egg) and mean: 0.34 ng $g^{-1}$ ww (duck egg)) (Zhang et al., 2010a), the wild waterbird eggs are over 60–700-fold higher, this great disparity may result from the age, food pattern, as well as different trophic level of these birds.

### 2.8. Fish (aquatic animal)

Taniyasu et al. (2003) detected the blood and liver PFOS concentrations in fish, which were 1–834 ng $mL^{-1}$ and 3–7900 ng $g^{-1}$ ww, respectively, with higher concentrations obtained in carnivorous and near-bottom feeders. A study conducted in Japan showed that PFOS concentrations in bird livers ranged from 0.15 ng $g^{-1}$ ww (large-bill crow) to 238 ng $g^{-1}$ ww (cormorants), compared with fish liver which ranged from 0.8 to 2.6 ng $g^{-1}$ ww, reflecting that higher tropic animals tend to accumulate larger amounts of PFOS (Senthilkumar et al., 2007). Based on these more recent values, it seems to be an indication that PFOS concentrations contained in the Japanese wildlife are decreasing, when compared to samples collected during 1997–2003.

During the same period, Yoo et al. (2009) detected relatively higher concentrations of 15–93 ng $mL^{-1}$ and 18–260 ng $g^{-1}$ ww respectively in fish blood and liver samples, but these values were all lower than that report (<10–500 ng $g^{-1}$ ww) obtained in an early study (Kannan et al., 2002).

Tseng et al. (2006) determined the muscle (mean: 200 ng $g^{-1}$ ww) and liver (mean: 310 ng $g^{-1}$ ww) samples of tilapia, and liver (mean: 260 ng $g^{-1}$ ww) samples of Japanese seaperch purchased in a Taiwan fish market. PFOS can be found in all samples, to some extent indicates this chemical is ubiquitous in Taiwan fish products (Tseng et al., 2006).

Skipjack tuna is distributed in tropical and temperate waters around the world, which is an important carnivorous fish, and is a suitable bioindicator for monitoring global oceanic pollution. During 1997–1999, skipjack tuna were collected from Japan, East China Sea (off Taiwan), off Indonesia, North Pacific Ocean and the Indian Ocean (Hart et al., 2008b). The authors analyzed the livers of these samples, and found PFOS can be detected in all samples from Japan (1.5–58.9 ng $g^{-1}$ ww) and East China Sea (5.1–7.1 ng/ww), and in the liver samples from off Indonesia, North Pacific Ocean and the Indian Ocean were <1–7.5, <1–2.1 and <1, respectively. The mean PFOS concentration in tuna livers from offshore (Japan, East China Sea and Indonesia) was much higher than that in open-sea (North Pacific and Indian Ocean), which suggested the large variation of PFOS concentration between these two sites may effectively influenced by nearby coastal areas.

PFOS can be found not only in the developed coastal regions but also in the fish muscles (<0.15–7.54 ng $g^{-1}$ dw) of inland rivers. Shi et al. (2010) collected nine species of fish from six sites of Qinghai-Tibetan Plateau, where it is far away from modern industrial or commercial activities. Although the PFOS concentration in Qinghai-Tibetan Plateau is lower than most other fish muscles in Asia, the PFOS pollution in this remote area was formed (Shi et al., 2010).

In addition to fish liver and muscle, Li et al. (2008c) detected the serum samples of five fish species from Gaobeidian Lake (North China) during 2005–2007. The PFOS concentrations were above the LODs in all samples, ranged from 4.81–84.4 ng $mL^{-1}$, in addition, the zooplankton collected from the same lake was detected at 4.18 ng $g^{-1}$ ww of PFOS, this study further prove the significant correlations between tropic levels and PFOS (Li et al., 2008c).

### 2.9. Others

Tissue distribution of PFOS concentration in animal organs can help scientists to clarify the accumulation and metabolism of this chemical in biota, the experimental animal feeding of $^{35}S$-labeled PFOS was conducted by Borg et al. (2010). In which study revealed that mass-labeled PFOS contained in the liver, lung and blood was higher than that of kidney and brain, this confirmed that PFOS was inclined to bind to liver and blood in previous studies. A tissue distribution was conducted by using wild Chinese Sturgeon, the authors detected the PFOS concentrations in 10 tissues of this animal. PFOS can be detected in all tissues, ranged from largest to the lowest were egg > liver > gallbladder > kidney > intestine > heart > ovary > stomach > gill > muscle, within these organ, except for egg (14.6 ng $g^{-1}$ ww) and liver (5.8 ng $g^{-1}$ ww), the other organs contain PFOS concentration no more than 0.52 ng $g^{-1}$ ww

(Peng et al., 2010). The result clearly showed together with liver, egg also the target that PFOS can highly accumulated, which indicating that most PFOS might pass to the next generation through pregnant sturgeon. Through this inheritance, concern may that fetus would be the most vulnerable part when exposure to PFOS.

## 3. Human exposure to PFOS pollution via different environmental media

To assess the human health risk exposure to PFOS, different pathways need to be considered. For adults, oral and inhalation exposure are the two major pathways. Oral exposure is mainly through consumption of contaminated food (Haug et al., 2010; Jogsten et al., 2009; Ostertag et al., 2009), and drinking water (Mak et al., 2009; Murakami et al., 2009); while inhalation exposure is mainly through inhalation of indoor and outdoor contaminated air and dust (Strynar and Lindstrom, 2008; Zhang et al., 2010a).

### 3.1. Air

There seems to be a severe lack of information related to PFOS concentration from Asia. Only measurement of PFOS in the outdoor air of Japan has been conducted. An average concentration of 5.3 pg $m^{-3}$, and 0.6 pg $m^{-3}$ was detected in urban region and rural region of Japan, respectively (Sasaki et al., 2003). The results were comparable to the study conducted in 2005, with mean PFOS concentration 5.6 pg $m^{-3}$ detected in urban area and 0.7 pg $m^{-3}$ in rural area of Japan (Harada et al., 2005). However, in the next year, it was found that the PFOS concentration was almost decreased by half in the urban area (2.9 pg $m^{-3}$), but increased about 3 times in rural region of Japan, especially at busy roads (up to 6.8 pg $m^{-3}$) (Harada et al., 2006). The highest mean PFOS concentration of 7.3 pg $m^{-3}$ in Wako City of Japan was subsequently detected (since 2003) (Sugita et al., 2007). Within the period of 2003–2007, there seemed to be dramatic changes with regards to PFOS concentrations in urban and rural area of Japan. There has been a decreasing trend in some urban areas and an increasing trend in rural areas, which indicated the sources of PFOS have been gradually shifted from urban to rural regions. However, the PFOS concentrations are still increasing for some industrialized and densely populated cities such as Wako City.

### 3.2. Drinking water

During the manufacturing process, PFOS can be released into the environment in both liquid and solid forms (Paul et al., 2009). PFOS were commonly detected in tap water and bottled water worldwide, as they cannot be effectively removed from raw water (surface or ground fresh water) (Jin et al., 2009; Kunacheva et al., 2010; Takagi et al., 2008).

According to Table 2, a range of 0.2–12 ng $L^{-1}$ was detected in surface water samples in Japan (Saito et al., 2004); while an average level generally less than 4 ng $L^{-1}$ (0.1–4 ng $L^{-1}$) was obtained for drinking water, except for a few samples (44 and 51 ng $L^{-1}$) supplied by the Kinuta Waterworks, this is due to contamination by the raw water from Tama River (with PFOS 0.7–157 ng $L^{-1}$) (Harada et al., 2003). Mak et al. (2009) noted the range of 0.066–4.9 ng $L^{-1}$, which was comparable to that obtained from previous studies conducted in 2003 and 2004 (Harada et al., 2003; Saito et al., 2004). A subsequent study showed that the concentrations in tap water of Japan ranged from 0.16 to 22 ng $L^{-1}$ (Takagi et al., 2008).

In Asia, there are totally six plants producing and supplying PFOS related substances, with 4 of them located in Japan (Paul et al., 2009), which explained that concentrations of PFOS in Japanese drinking water are generally higher than those of other countries: India (<0.040–8.4 ng $L^{-1}$), Vietnam (<0.05–0.1 ng $L^{-1}$), Thailand (0.13–1.9 ng $L^{-1}$) and Malaysia (0.025–0.1 ng $L^{-1}$) (Fujii et al., 2007). A later survey conducted in Thailand showed that although PFOS was detected in all samples, the average concentration was rather low (0.17 ng $L^{-1}$) (Kunacheva et al., 2010).

**Table 2**
The PFOS concentration and their Risk Quotient (RQ) in tap water samples from Asia.

| Country | PFOS concentration (ng $L^{-1}$) | Risk quotient (RQ) | References |
|---|---|---|---|
| Japan | 0.1–4 | 0.0005–0.02 | Harada et al. (2003) |
| | 0.066–4.9 | 0.0003–0.02 | Mak et al. (2009) |
| | 0.16–22 | 0.0008–0.11 | Takagi et al. (2008) |
| China | 0.042–11 | 0.0002–0.06 | Mak et al. (2009) |
| | <0.1–14.8 | <0.0005–0.07 | Jin et al. (2009) |
| India | <0.04–8.4 | <0.0002–0.04 | Fujii et al. (2007) |
| Vietnam | <0.05–0.1 | <0.0003–0.0005 | |
| Thailand | 0.13–1.9 | 0.0006–0.01 | |
| Malaysia | 0.025–0.1 | 0.0001–0.0005 | |

During 2006–2008, Mak et al. (2009) also conducted a survey of tap water collected from 10 cities of China. Results showed that PFOS ranged from 0.042–11 ng $L^{-1}$, which matched an early survey conducted in 2007, with a range of <0.1–14.8 ng $L^{-1}$ (Jin et al., 2009). The values were similar with those of the Japanese tap water. In fact, PFOS concentrations in one area's drinking water are usually similar to the surface water of the same area, reflecting that PFOS are not effectively removed by water treatment process (Skutlarek et al., 2006).

Mak et al. (2009) conducted a risk assessment based on concentrations of PFOS in tap water. The level of risk related to PFOS-contaminated tap water is characterized by risk quotient (RQ), based on comparing the detected PFOS concentration in tap water with the guideline value (for PFOS) issued by the U.S.EPA, which is 200 ng $L^{-1}$ (Mak et al., 2009). If the value of RQ less than the unity, it means the PFOS in tap water would not pose an immediate health risk, but if the value of RQ larger than the unity, it means the PFOS concentration in tap water may impose adverse health effects. According to Table 2, all RQs measured in Asian tap water samples were less than unity, indicating that the PFOS concentrations contained in tap water may not give rise to immediate health risk. However, PFOS is not the only PFCs, and also not the only toxic chemical contained in the tap water, other toxic chemicals should also be taken into account.

### 3.3. Food consumption

Food consumption is regarded as the most important route for human exposure to chemical contaminants (Brustad et al., 2008; Dovydaitis, 2008; Genuis, 2008), and for PFOS, consumption of contaminated food can contribute over 60% of total exposure in all intake sources (Tittlemier et al., 2007). However, with the exception of data on fish and/or shellfish, information on PFOS concentrations in other food items is limited. It is envisaged that in coastal areas with heavy industrial activities, fish and/or shellfish cultured around the areas may be contaminated with PFOS, due to the release of PFOS into the aquatic systems, bioaccumulation and biomagnification along food chains, leading to higher exposure to local people, especially those with a seafood dietary preference.

### 3.4. Seafood

Five types of seafood (fish, crab, mollusk, shrimp and shell fish) were collected from Guangzhou (South China) and Zhoushan (East

China) local markets for analyses. Results indicated that PFOS can be detected in all seafood samples, which ranged from 0.33 (shellfish) to 13.9 (shrimp) ng $g^{-1}$ ww (Gulkowska et al., 2006). Based on these values, the Average Daily Intake (ADI) of PFOS was lower than the PFOS Reference Dose (RfD) (0.025 μg $kg^{-1}$ $d^{-1}$) (for non-cancer health risk assessment). However, information related to intake of PFOS via consumption of other food items, and through other exposure pathways is still largely unknown.

So et al. (2006a) showed that PFOS concentrations in mussel and oyster samples from South China and Japan ranged 0.11–0.59 ng $g^{-1}$ ww. The eleven mollusk species (soft tissues) collected from nine coastal cities in the Bohai Sea region (north China), had lower concentrations and frequency of detection (61%) (Pan et al., 2010b).

### 3.5. Daily food

Zhang et al. (2010a) analyzed samples of daily food items, namely meat, animal liver, blood cake, chicken and duck eggs from 17 cities of China analyzed for PFOS and obtained the following results: 0.05 ± 0.02 ng $g^{-1}$ dw (beef), 0.08 ± 0.06 ng $g^{-1}$ dw (chicken and goat); 0.32 ± 0.18 ng $g^{-1}$ dw (chicken liver), 1.99 ± 2.00 ng $g^{-1}$ dw (pork liver), 0.08 ± 0.05 ng $g^{-1}$ ww (chick egg) 0.34 ± 0.38 ng $g^{-1}$ ww (duck egg) and < 0.12–0.08 ± 0.03 ng $g^{-1}$ dw (animal blood cakes). Even based on the dry weight, the daily food studied by Zhang et al. (2010a) had relatively slight PFOS contamination than most seafood and wildlife. Besides, based on the wet weight of unit sample, Wang et al. (2010c) determined PFOS in different tissue samples from farmed pigs and chickens that were purchased in Beijing in 2009, for pig: 0.024 ng $g^{-1}$ ww (heart), 2.163 ng $g^{-1}$ ww (liver), 0.102 ng $g^{-1}$ ww (kidney) and 0.007 ng $g^{-1}$ ww (loin); for chick: 0.026 ng $g^{-1}$ ww (heart), 0.063 ng $g^{-1}$ ww (liver) and 0.012 ng $g^{-1}$ ww (breast). Among these results, with the exception of pig liver being comparable with seafood, the other edible tissues were about 10 times lower than that. These indicate that Chinese consumers (non-seafood diets) are exposed to a low level of PFOS contamination from food. PFOS was also detected in dairy products from Beijing, Tianjin and Wuhan (three major cities of northern and central China) by Wang et al. (2010b), but it was only observed in 36%, 25% and 13% of 84 milk, 36 milk powder and 32 yoghurt samples above the LODs (0.005 ng $g^{-1}$), respectively. In milk samples, it ranged from less than 0.005–0.695 (mean: 0.024) ng $g^{-1}$, in milk powder was less than 0.005–0.175 (mean: 0.022) ng $g^{-1}$ and less than 0.005–0.032 (mean: 0.003) ng $g^{-1}$ in yoghurt.

### 3.6. Dust ingestion

In general, there are two pathways for human exposure to PFOS through dust ingestion, via indoor and outdoor. The first paper concerning PFOS in indoor dust revealed an average PFOS concentration of 24.5 (11–2500) ng $g^{-1}$ in household dust (Moriwaki et al., 2003), while a later similar study showed a range of 7.0–41 ng $g^{-1}$ in indoor dust (Tsunenobu et al., 2006). Zhang et al. (2010a) obtained a range of 1.11 ± 1.27 to 10.7 ± 11.9 ng $g^{-1}$ PFOS in indoor dust collected from 17 cities of China, with a detection frequency of 68%. The range of <0.2–11 ng $g^{-1}$ in street dust was found in Japan (Murakami and Takada, 2008). Based on the above data, there seems to be considerable variation on the concentrations detected from various places, possibly due to the distinct PFOS sources of these samples sites and different sampling and detection methodologies adopted. However the most concerned would be exposure to elevated PFOS in the confined indoor area, with higher PFOS levels. While in the area of occupational exposure area, Wang et al. (2010a) studied the dust samples collected from the perfluorochemical manufacturing plant in Wuhan (central China). The PFOS concentration ranged from 10.49 μg $g^{-1}$ (plant road) to 4691.94 μg $g^{-1}$ (sulfonation workshop). Obviously, indoor PFOS contamination, especially in the manufacture plant, was much higher than outdoor, and it was considered as an important exposure pathway imposed to workers, and if without appropriate treatment and protection, the employees here may have adverse effects by inhalation of these highly contaminated dust. Compared to the other solid particles, the PFOS contained in dust was higher than that in sediment, may suggesting that PFOS can be more effective transport by atmosphere than hydrosphere.

## 4. Body loading of PFOS

### 4.1. Human blood

Intake of PFOS through oral consumption and inhalation can be primarily found in the liver, serum and kidney (Volkel et al., 2008). It has been noted that the concentration of PFOS in cord blood was negatively associated with birth weight, ponderal index, and head circumference of new born babies (Tao et al., 2008a). Information about the toxicokinetics of PFOS in humans is limited, but it is commonly recognized that PFOS in human serum is the predominant indicator of body loadings of PFOS. In historical research, most studies were focused on the human serum and whole blood PFOS concentration instead of plasma samples. The 1:1 ratio between human serum and plasma PFOS concentration was found, while it was approximately 2:1 ratio when compared serum or plasma to whole blood (Ehresman et al., 2007).

Firstly, PFOS concentration in human blood has large variation mainly according to the sampling area and period, Fig. 1 shows the data related to a study on serum PFOS concentrations from 10 countries (including United States, Colombia, Brazil, Italy, Poland, Belgium, India, Malaysia, Korea and Japan) (Kannan et al., 2004). Higher values were generally detected in developed than developing countries. The highest mean PFOS concentration was found in the samples from the United States, followed by Poland, Korea and Japan, while lower values were obtained from India, followed by Italy and Colombia. Similarly, Fig. 2 compares PFOS concentrations in human serum samples from different parts of the world, which also confirmed that lower average PFOS concentrations were detected in human serum samples from developing countries, such as Peru, Sri Lanka and Vietnam (0.70, 5.03 and 3.20 ng $mL^{-1}$, respectively) (Guruge et al., 2005; Rylander et al., 2009). This distribution pattern also can be found in some Chinese cities. Two similar surveys were conducted by analyzing human whole blood samples collected from of Chinese 9 cities in 2004 and 12 cities in 2006–2008, both results showed that the highest PFOS was obtained in industrial and populated city (Shengyang and Shijiazhuang), and interestingly both of them located in the north China, moreover, Shenyang City was reported contains high PFOS concentration in its precipitation (Liu et al., 2009b) before,

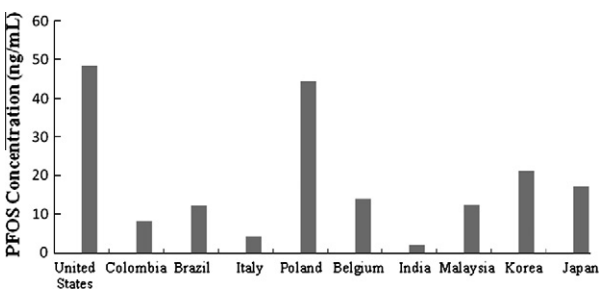


**Fig. 1.** Human serum PFOS concentrations in different countries (Kannan et al., 2004).

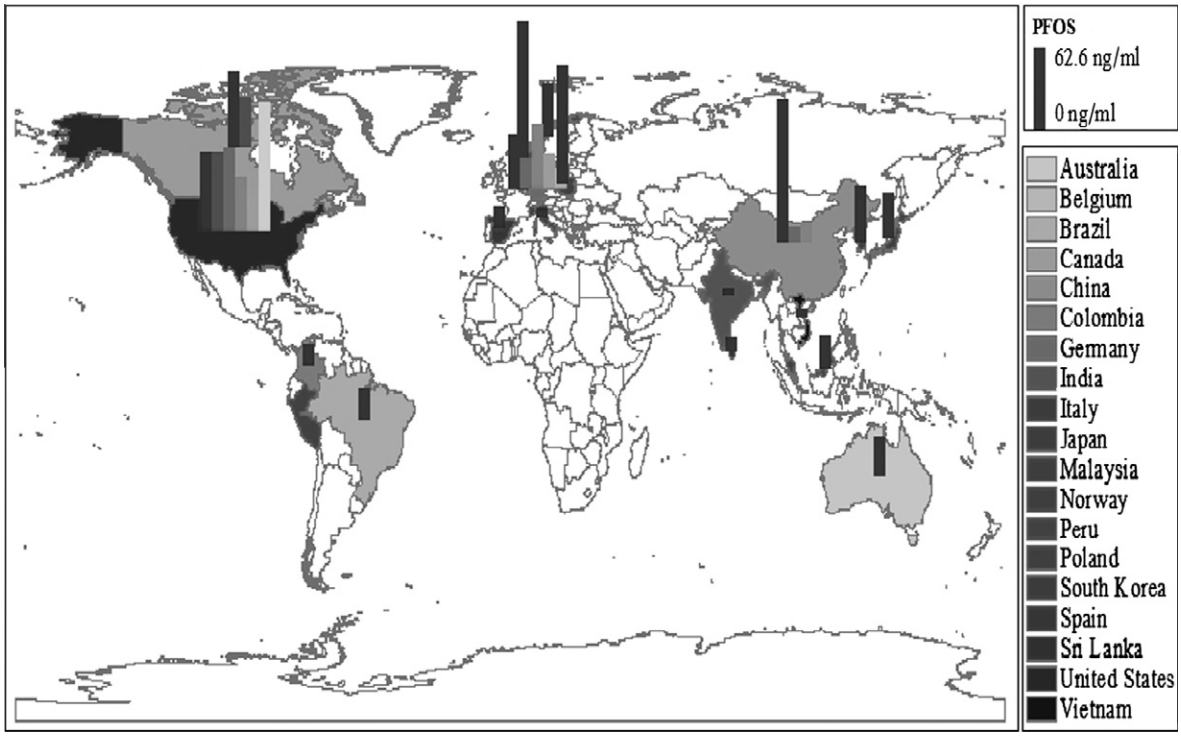


**Fig. 2.** The PFOS concentration in human serum collected from global countries. Australia (collected from 2002–2003) (Karrman et al., 2006); Colombia, Brazil, Italy, Poland, India, Malaysia, Korea and Japan (collected from 2004) (Kannan et al., 2004), United States (from left to right, collected from 1974–1989, 1990–2002, 2003–2004, 2003 and 2004, respectively) (Calafat et al., 2006a, 2007a,b; Kannan et al., 2004); China (from left to right, collected from 2007, 2008 and 2009,respectively) (Yeung et al., 2006, 2008); Canada (from left to right, collected from 2002 and 2009) (Kubwabo et al., 2004); German (from left to right, collected from 1977–2004, 2005, 2006, 2007, 2008, respectively) (Wilhelm et al., 2009); Belgium (from left to right, collected from 2004 and 2002–2005) (Kannan et al., 2004; Roosens et al., 2010); Peru (collected from 2003) (Calafat et al., 2006b); Norway (collected from 2004) (Rylander et al., 2009); Sri Lanka (collected from 2005) (Guruge et al., 2005); Spain (collected from 2008) (Karrman et al., 2010) and Vietnam (collected from 2009) (Rylander et al., 2009).

while the lowest concentration was found in Jintan and Kunming, one of which is a small city in east China and the other is southeast city of China and far away from industry (Pan et al., 2010a; Yeung et al., 2006). Besides, in other Asian countries, higher serum PFOS concentration (1.2–8 times) was found in the urban population than that of rural worker in Sri Lanka (Guruge et al., 2005), Karrman et al. (2009) found PFOS concentration in women serum was significantly higher in Osaka than that in Miyagi of Japan, Afghanistan human serum samples contained relatively low PFOS (1.2–5.5 ng $mL^{-1}$ for males, 0.2–11.8 for females, 0.5–3.9 for children), with man and urban residents had significantly higher levels than that in women and villagers, respectively ($p < 0.001$) (Hemat et al., 2010). The large variability of PFOS percentage in human blood among different countries indicates that there may be diverse exposure sources and pathways of PFOS to humans in different countries. In China, PFOS contributes about 60–90% of PFCs in human blood in China (Pan et al., 2010a; Yeung et al., 2008), and only 24% of PFCs in human blood in Korea (Harada et al., 2010). Therefore, in the main cities of Asia, such as Osaka, Takayama and Sendai of Japan, Busan and Seoul of Korea and Hanoi of Vietnam, the PFOS concentrations were roughly the same, ranging from 6.19–8.43 ng $mL^{-1}$ (Harada et al., 2010), however, in China its concentration can reach 34 ng $mL^{-1}$ in Shenyang during the same period (Pan et al., 2010a).

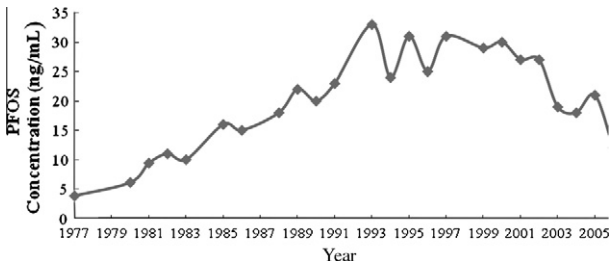


**Fig. 3.** Time trend of PFOS concentration in human sera collected from Norway during the year of 1977–2005 (Haug et al., 2009).

PFOS and its related products were widely used before 2002 in industrial and commercial areas, but they were phased out by the 3M company (the largest PFOS manufacturing company) after 2002, leading to lower bioaccumulation and biomagnifications in food chains (3M, 2008). Fig. 3 shows that the PFOS concentrations in the serum samples in Norway increased 8-fold from 1977 to 1993 (from 3.8 ng $mL^{-1}$ to 33), then fluctuated during 1993 to 2000, and finally sharply decreased after 2002. Similarly, in Norway, Harada et al. (2007) also found serum concentration of PFOS reached plateau levels in Japan in the late 1980s. However, the PFOS concentration in Korea and China seems trace the other way, there is significantly increasing of PFOS concentration in Busan City of Korea from 2000–2008 (Harada et al., 2010), additionally, the time trend of PFOS concentration in China was reported by Jin et al. (2007), the authors detected the human serum samples collected from the year of 1987, 1990, 1999 and 2002 in Shenyang (north China), with mean concentrations of 0.03, 0.02, 1.8 and 12.9 respectively. The low concentration before 1999 revealed the rare usage of PFOS at that time, but sharply increased 10 times at 2002, indicating acute exposure of PFOS in Shenyang, moreover, it was increased three times (34 ng $mL^{-1}$) in 2006, detected by Pan et al. (2010a). Unlike the trend in Norway, in which the PFOS concentration in human serum decreased after 2002, it was still increasing after 2002, might suggest that the manufacture and application of PFOS was moved to China after 2002.

Secondly, since PFOS is a persistent contaminant that is hard to degrade, it might be expected that this chemical in body loading would increase with age (Duarte-Davidson and Jones, 1994). Liu et al. (2009a) found the effect of age on blood PFOS levels has positively correlation with age ($p < 0.01$) in Liaoning Province (north China), as well as Pan et al. (2010a) also found this correlation in all samples collected from 12 cities of China. Zhang et al., 2010b revealed in Nanchang City (central China) that PFOS concentration was 4.04 ng mL$^{-1}$ in teenagers (age < 18 yrs), which was 2 times lower than that in adults (age > 18 yrs), showing the significantly increasing ($r = 0.468$, $p < 0.01$) with age. In Japan and Korea, PFOS also detected in human serum samples collected from 1994, 2000, 2003–2004 and 2007–2008 ($p < 0.01$) (Harada et al., 2007, 2010).Thirdly, gender-related discrepancy also can be found in some research. Significantly higher ($p < 0.05$) concentrations of PFOS were observed in males than females in 9 cities of China, which contained highly populated and rural cities (Yeung et al., 2006). (Harada et al., 2007) found serum samples in Japanese male collected from 2003–2004 has significantly higher means for PFOS (1.5-fold) than those from female ($p < 0.01$). Pan et al. (2010a) also confirmed PFOS concentration was significantly influenced by gender in the 20–29 years old group of in 12 Chinese Cities.

### 4.2. Human milk and other tissue

The lactational transfer of PFOS has been suggested as an important source for infant exposure to PFOS, with newborns particularly vulnerable to PFOS exposure (Toms et al., 2009). Fig. 4 shows the concentrations of PFOS in human milk samples examined in different countries.

Tao et al. (2008b) analyzed a total of 184 human milk samples from seven Asian countries (Japan, India, Malaysia, Philippines, Indonesia, Vietnam and Cambodia) and observed that PFOS was the predominant PFCs, in 85% of the Indian samples and 100% samples of other countries. The PFOS of all samples ranged from <0.011 ng mL$^{-1}$ (India) to 0.523 (Japan), with the lowest mean of 0.046 ng mL$^{-1}$ (India) and the highest mean of 0.232 ng mL$^{-1}$ (Japan). Significantly higher PFOS were noted in samples collected from Japan than all other countries, with samples from Malaysia, Philippines and Indonesia significantly higher than those from Vietnam, Cambodia and India. This reflected the degree of industrialization of these countries. The distribution pattern of PFOS in human milk is similar with those of human serum, with PFOS the lowest in Indian human milk, similar to the trend observed in human serum samples (Kannan et al., 2004).

Human milk samples collected from 12 provinces of China indicated the mean concentration of PFOS was 0.046 (0.006–0.137) ng mL$^{-1}$, and the concentrations detected generally reflected the degree of industrialization and urbanization of different provinces, with higher levels detected in Liaoning and Shanghai Provinces, especially the former (So et al., 2006b). A later study indicated a reduction of PFOS concentration with a range of 0.006–0.137 mg mL$^{-1}$ in samples from Zhoushan City of Zhejiang Province (Liu et al., 2010), compared with the range of 0.045–0.36 ng mL$^{-1}$, also in samples collected from the same city a few years ago (So et al., 2006b). This may be due to the sharp decrease of PFOS production, until the production was finally ceased in 2002 (Paul et al., 2009).

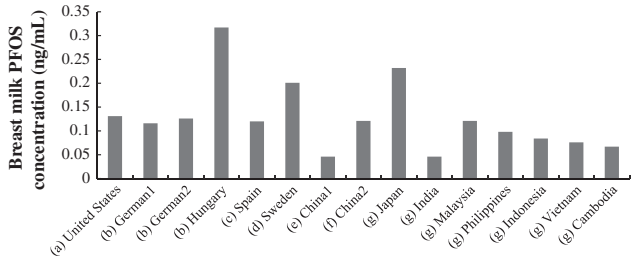


**Fig. 4.** The comparison of PFOS concentrations in human breast milk from global countries. (a) Tao et al. (2008a); (b) Volkel et al. (2008); (c) Karrman et al. (2010); (d) Karrman et al. (2007); (e) Liu et al. (2010); (f) So et al. (2006b); (g) Tao et al. (2008b). *German1*: Munich; *German2*: Leipzig; *China1*: Heilongjiang, Liaoning, Hebei, Henan, Shanxi, Ningxia, Jiangxi, Fujian, Shanghai, Hubei, Sichuan and Guangxi; *China2*: Zhoushan.

Fig. 4 also shows that the PFOS concentrations in human milk was much lower than that in human serum, which confirmed an early study conducted in Sweden, indicating the serum/milk ratio of PFOS of 113 (Karrman et al., 2007). Globally, the highest concentration was recorded from Hungary milk samples (mean: 0.317 ng mL$^{-1}$; range: 0.096–0.639 ng mL$^{-1}$), based on 13 milk samples collected during 1996–1997. The values were substantially higher than samples collected from Munich (0.116 ng mL$^{-1}$) and Leipzig (0.126 ng mL$^{-1}$) in 2006 (Volkel et al., 2008), although it was concluded that it would not impose any associated risks even if children are breast fed (Volkel et al., 2008).

There is limited information concerning levels of PFOS in human tissues (e.g. adipose and liver) other than blood and milk, due to obvious reasons. Xu et al. (2010) determined the levels of PFOS both in urban and rural children fingernails. Samples from urban children have significantly higher values (mean: 328 ng g$^{-1}$) than those in rural children (mean: 27 ng g$^{-1}$), also higher PFOS concentration were found in children aged <9 years than in children aged $\geqslant$9 years. (Xu et al., 2010). The results showed that the PFOS contained in children fingernails is relatively higher when compared to blood and human milk samples, may reflect that apart from abdominal organs, fingernails tissue also accumulate PFOS, besides, further studies such as research on hair PFOS detection could be conducted. Additionally, the higher concentration found in the group <9, indicating the exposure discrepancy between two groups, may suggest that for children <9 oral digestion of PFOS may be likely.

### 4.3. Diseases related to PFOS exposure

Being an ubiquitous PFC, PFOS may act as endocrine disruptors, neurotoxic agents, and fetal development perturbing substances and may also be carcinogenic (Pirali et al., 2009). However, the information related to human diseases caused by PFOS exposure is very limited.

Pirali et al. (2009) measured PFOS in 28 patients undergoing thyroid surgery for benign and malignant thyroid disorders, and found that PFOS can be detected in all surgical specimens of thyroid tissue, and a significant correlation between the serum and thyroid tissue levels of PFOS was found in all patients. However, according to these observations, it cannot be concluded that PFOS are actively concentrated in the thyroid, and caused the disease directly.

Melzer et al. (2010) examined the associations between serum PFOS concentration and thyroid disease in representative samples of the general population in United States and observed that higher PFOS serum concentrations were associated with thyroid diseases. However, more work is needed to investigate the mechanisms involved in the relationship between these diseases and PFOS serum concentration.

Furthermore, there are great concerns that PFOS can be transferred to the developing organisms through placenta and milk. Based on a study, after feeding pregnant rats with 3.2 mg PFOS/kg throughout gestation and lactation period, it was noted that hypothyroxinemia was induced in rat pups, suggesting that PFOS can be transferred to infants though maternal milk, and also exposure in utero during the postnatal period (Yu et al., 2009).

In general, the relationships between PFOS concentration in human or animal bodies and diseases are not clear, and further studies are essential to determine if PFOS can disrupt or affect target cells, tissues and organs of humans or animals. It is also necessary to obtain more epidemiological data concerning this deadly compound (Pirali et al., 2009).

## 5. Conclusion

The existence of PFOS in the environment is derived from manufacture and application of PFOS and derived products and byproducts, which generate a large amount of waste in aqueous and solid form. In Asia, PFOS are mostly detected in water systems, wild mammals and seafood, with concentrations generally higher in samples collected from industrialized regions than rural areas. In addition, higher concentrations of PFOS are detected in human samples (including human serum and milk) in China, Japan and Korea than that in India, Vietnam and Sri Lanka. The major exposure pathways of PFOS are mainly via oral intake and inhalation for human beings, and the concentrations are commonly associated with gender and dietary habit (especially those with a higher consumption of seafood). PFOS and related products were widely used in industrial and commercial areas from 1977 to 1993, with relatively higher concentrations detected in the biota samples during this period. After PFOS was phased-out in 2002, its bioaccumulation and biomagnifications in food chains gradually decreased. However, PFOS is still exists in the environment which could exert harmful effects to biota and human beings.

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