

Progress and perspective of perfluorinated compound risk assessment and management in various countries and institutes

Yasuyuki Zushi · Jonathan Nartey Hogarh ·
Shigeki Masunaga

Received: 30 September 2010 / Accepted: 1 April 2011 / Published online: 20 April 2011
© Springer-Verlag 2011

Abstract Perfluorooctane sulfonate (PFOS) and related compounds have recently been designated as target chemicals for regulation by the Stockholm Convention on Persistent Organic Pollutants (POPs). Many countries have investigated and tried to implement various countermeasures in response to this decision. In this article, we collect reports concerning regulations and risk evaluations of perfluorinated compounds (PFCs) and review the current PFC management practiced in various countries. The first part of this review contains a comprehensive collection of proposed standard PFC values, including provisional tolerable daily intakes (pTDI), drinking water guidelines, and predicted non-effect concentrations (PNEC). The pTDI values ranged from 0.1 to 0.3 $\mu\text{g}/\text{kg}/\text{day}$ for PFOS, and there are wide margins of safety for adults. Health risks for plant workers exposed to PFCs and for infants are of particular concern. The application of these proposed values in controlling PFC pollution is one approach that may effectively control human health risk without unduly sacrificing the benefits from PFC use. The second part of this review contains a collection and review of a number of regulations and countermeasures, such as an EU directive, regulation in Canada, and the Significant New Use Rule (SNUR), including voluntary control (i.e., production phase-out by 3M, stewardship programs, regulation in the semiconductor industry). Most of these regulations are based principally on the precautionary principle. However, they may not be as effective in pollution reduction as intended because the chemicals in question are already widely distributed in the

environment owing to their use and mobility in the environment. In addition, these types of regulations would be non-operative in developing countries because rapidly growing economies place great demand on high performance materials, including PFCs. Further development of risk assessment methods that allow the evaluation of the counter risks of PFC alternatives and the loss of benefits from the PFC ban is necessary because of the possible continuous use of PFCs, especially in developing countries.

Keywords Perfluorinated compounds (PFCs) · Guideline · Risk assessment · Management · Regulation · TDI

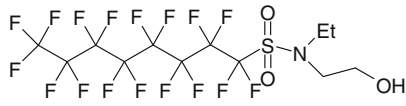
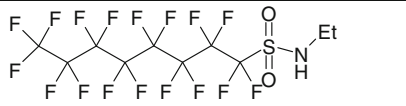
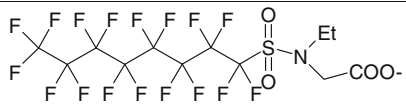
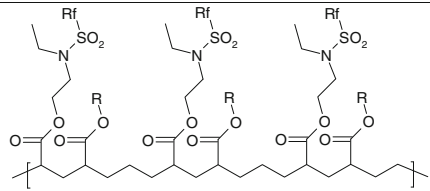
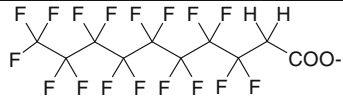
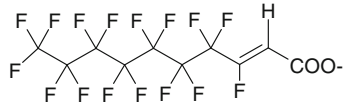
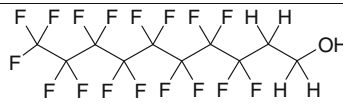
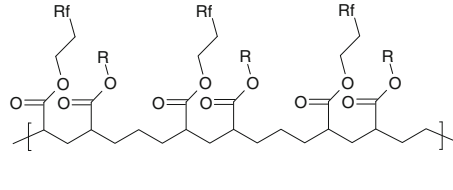
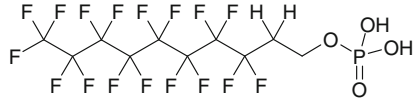
Introduction

Perfluorinated compounds (PFCs), including perfluorooctane sulfonate (PFOS), perfluorooctanoate (PFOA), and other related compounds, are class of chemicals with a polyfluorinated alkyl chain in their molecular structure (Table 1). Perfluorinated compounds exhibit characteristics of fluoroalkyl chains, such as low molecular polarizability, short C–F bond length, and large C–F bond binding energy. These characteristics contribute to the variety of useful and interesting PFC features, including the oil and water repellency, physical and chemical stability, and reduced solvent surface tension of materials that employ PFCs as polymer building blocks. Because of these qualities, PFCs have been used since the 1950s in various applications: upholstery, textiles, fire-fighting materials, hydraulic fluid, photo-resist, emulsifying agents for polytetrafluoroethylene (PTFE) production, and others.

Global PFOS and PFOA pollution was first observed in 2001 (Giesy and Kannan 2001). Their accumulation in

Y. Zushi · J. N. Hogarh · S. Masunaga (✉)
Graduate School of Environment and Information Sciences
Yokohama National University, 79-7 Tokiwadai, Hodogaya-ku,
Yokohama, Kanagawa 240-8501, Japan
e-mail: masunaga@ynu.ac.jp

Table 1 Classification and chemical structures of PFCs

Classification	Example	Structure	Remark
PFSA	PFOS ($n = 7$)	$\text{CF}_3(\text{CF}_2)_n\text{SO}_3^-$	$n = 3-4$ are conceivable
PFCA	PFOA ($n = 6$)	$\text{CF}_3(\text{CF}_2)_n\text{COO}^-$	$n = 1-16$ are conceivable
FOSE	<i>N</i> -MeFOSE, <i>N</i> -EtFOSE (see structure)		Precursors of PFOS/PFOA
FOSA	FOSA, <i>N</i> -MeFOSE, <i>N</i> -EtFOSE (see structure)		
FOSAA	<i>N</i> -MeFOSAA, <i>N</i> -EtFOSAA (see structure)		
FOSA Acrylate polymer	FOSA Acrylate polymer (see structure, Rf: perfluoroalkyl chain, R: alkyl chain)		Possibly degrade to PFOS/PFOA
FTCA	8:2 FTCA (see structure), 10:2 FTCA		Precursors of PFCA
FTUCA	8:2 FTUCA (see structure), 10:2 FTUCA		
FTOH	8:2 FTOH (see structure), 10:2 FTOH		
FT Acrylate polymer	FT Acrylate polymer (see structure, Rf: perfluoroalkyl chain, R: alkyl chain)		Possibly degrade to PFCA
PAPS	8:2 mono PAPS (see structure), 8:2 di PAPS		

wildlife and the human body prove that they are persistent in the environment, and they may be toxic to organisms. Following this finding, many studies have reported PFC contamination in various environmental media, including aquatic ecosystems, drinking water, outdoor and indoor air environments, and food products.

High PFC concentrations have been found in river and coastal water near fluorochemical facilities (Hansen et al. 2002; Saito et al. 2004), sewage treatment plants (Zushi et al. 2008; Ahrens et al. 2009), and urban sites emanating from road surfaces (Murakami et al. 2009b; Zushi and Masunaga 2009a, b). Ground water pollution has also been observed in urban regions (Murakami et al. 2009a) and near military bases at which high levels of PFC-containing fire-fighting materials are used for training (Moody et al. 2003).

High levels of PFCs in drinking water and human blood near the 3M PFC manufacturing facilities' disposal sites, in the Oakdale and Washington County Landfill in Lake Elmo, Minnesota, were reported by the Minnesota Department of Health (MDH 2008). The pollution of drinking water by illegally recycled hazardous waste, especially agricultural soil with high PFOA levels, was reported in the Ruhr area of Germany (Skutlarek et al. 2006). People living near the contaminated site were consequently exposed to high PFOA levels (Hölzer et al. 2009).

High levels of atmospheric pollution by fluorotelomer alcohols (FTOHs), perfluorooctane sulfonamide (FOSA), and perfluorooctane sulfonamido ethanols (FOSE), which degrade into PFOA and PFOS, have been reported in North America near carpet production and treatment facilities (Stock et al. 2004). Higher levels of FOSE contamination have been identified in indoor air than in outdoor air, suggesting that indoor air is a significant source of PFC exposure for humans (Shoeib et al. 2005). In addition to precursors such as FTOH, FOSA and FOSE, PFOS and PFOA have been detected in the particle phase, suspended in the atmosphere (Harada et al. 2005b).

Food contamination by PFCs has been observed (FSA 2006; Ministry of the Environment in Japan 2006; Tittlemier et al. 2006; Fromme et al. 2007) and has been suggested as the dominant pathway for human PFC exposure among such candidates as indoor air, outdoor air, house dust, and drinking water (Fromme et al. 2009).

As described above, PFC pollution is prominent in various environmental matrices. Although PFCs exhibit unparalleled performance in many applications, concerns over their adverse effects on human health and the environment have increased. These chemicals must be managed appropriately.

On May 2009, PFOS and perfluorooctyl sulfonyl fluoride (PFOSF) were added to the list of the Stockholm Convention on Persistent Organic Pollutants (POPs), which

prohibit or restrict the production and use of chemicals that have persistent, bioaccumulative, toxic, and long-range transportable natures in the ratifying countries (169 participants in 2010). The treaty includes three categories of regulation for the chemicals listed: the elimination of production and use (Annex A), the restriction of production and use (Annex B), and the reduction of unintentional release (Annex C). PFOS and PFOSF are listed in Annex B, permitting their production and use for specific purposes only, such as photo-imaging, photo-resist and anti-reflective coatings for semi-conductors, aviation hydraulic fluids, hard metal plating (only in closed-loop systems), medical devices, fire-fighting foam, and insect baits for control of leaf-cutting ants (UNEP 2009b). Although PFOS and PFOSF are included in the convention and have already been partially regulated, Wang et al. (2009) indicated the limitations and disadvantages of such regulation for developing nations. These disadvantages include the inadequacies of such precautionary policies from the aspect of the demand for economic growth (large economic losses for the phase-out of PFOS/PFOSF) and the inability to seek alternative materials. Thus, we have to find acceptable pollution control and management schemes for developing countries, rather than a complete ban on use and production.

In this article, we summarize various reports concerning PFC regulation and risk evaluation and review the current situation of PFC management. This work might help in the development of optimal PFC management.

Information on current regulations and guideline values

Current regulation of PFC production/usage including voluntary control

As mentioned in the previous section, the listing of PFOS and PFOSF in Stockholm Convention on POPs was a symbolic event in the regulation of PFC production and use. Additionally, a number of regulations including voluntary control have been applied to stop PFC pollution in various countries and bodies, as described below.

It has been announced that PFOS and its derivatives will be regulated on the market or only used as a substance or constituent of preparations in the EU Directive (2006). Under this directive, PFOS is used in un-substitutable applications, including photolithography processes, photographic coatings, mist suppressants for non-decorative hard chromium (VI) plating/wetting agents in controlled electroplating systems (pollution prevention and control are required), and hydraulic fluids for aviation. The regulation started in June 2008 within EU member countries (27 countries in 2010). Fire-fighting foams, which had been on

the market before 27 December 2006, would be banned after 27 June 2011.

Canada announced on 17 June 2006 that it would prohibit the manufacture, use, sale, offer for sale, and importation of FTOHs (FTOHs are not produced in Canada). Environment Canada and Health Canada considered that FTOHs might degrade into long chain PFCAs that are bioaccumulative and suspected to cause cancer and other adverse effects (Ammoniums 2006; Government of Canada 2006). This action was based on the precautionary principle and preventative measure. PFOS and its salts have been added to the “virtual elimination list”, whose addition requires the identification of the level of quantification and the publication of regulations prescribing the quantity or concentration of PFOS, after receiving royal assent on 17 April 2008 in Canada. Also, “Perfluorooctane sulfonate and its salts and certain other compounds regulations” came into force on 29 May 2008 (Government of Canada 2009). The purpose of this regulation was to prevent the harm that would be imposed on Canada’s environment from the use or release of PFOS and its related compounds by prohibiting the manufacture, use, sale, offer for sale, import of PFOS, and manufactured items containing PFOS.

U.S. Environmental Protection Agency (USEPA) listed PFOS, its salts, PFOSF, their longer and shorter chain homologues, and related compounds, including polymers containing the above-described chemicals, as substructures in the significant new use rule (SNUR), which requested manufacturers and importers to notify USEPA 90 days before their use under the Toxic Substances Control Act (TSCA) from 2003 (USEPA 2002).

Following consultation between USEPA and 3M, the major producer of PFOS/PFOSF in the world, it was declared in 2000 that 3M would phase out the production of PFOS and of its related chemicals by 2003 (3M 2000). DuPont, which is one of the major producers of PFOA, paid 16.5 million dollars as a penalty because it glossed over important risk information concerning PFOA. Afterward, DuPont (2005) made a plan to reduce their emission of those chemicals by more than 85% (compared with the baseline year of 1999) by 2007. As a stewardship program, eight major PFOA producers (Arkema, Ashahi, Ciba, Clariant, Daikin, 3M/Dyneon, DuPont, and Solvay Solexis) aimed to reduce the emission of PFOA, its precursors and longer chain homologues by 95% (compared with the amount in the baseline year of 2000) by 2010, and by 100% by 2015. According to the progress report of the stewardship program by USEPA, four of the eight companies in the U. S. operations satisfied their 2010 target by the end of 2008 (USEPA 2009a). No company in non-U.S. operations has satisfied this target, and certain companies increased their emission of PFOA precursors from the base line year. Also, the substitution of PFOA by PFCs with shorter alkyl

chain (e.g., perfluorohexanoate, PFHxA and perfluorobutane sulfonate, PFBS), which are considered to be lower in bioaccumulation and toxicity, have been adopted by certain companies.

Voluntary action to reduce PFOS emission in the semiconductor production process by the Semiconductor Industry Association (SIA) and Semiconductor Equipment and Materials International (SEMI) has been implemented (WSC 2009).

Regulations concerning the production and usage of PFOS, PFOA, and their homologues based on the concept of the precautionary principle have been introduced around the timing of the listing of PFOS by the Stockholm Convention on POPs. However, it is difficult for developing countries to change their practice, and it is not practical for those countries to stop production, usage, and import/export just to reduce environmental PFOS pollution level. In fact, China increased its production of PFOS and related compounds during 2003–2006 (UNEP 2008). In addition to this practical difficulty, it is also doubtful that the regulations would significantly decrease the environmental PFC levels because PFCs have been used in many applications and have been widely distributed. The reports concerning the non-point source of PFCs reveal the difficulty surrounding the rapid reduction in contamination levels by the regulation of production only. Moreover, such regulation would diminish the advantages of the use of PFCs. Thus, we need to seek risk reduction while maintaining benefits. Science-based risk assessment for PFCs is necessary to propose acceptable guidelines for PFC levels in the environment that can tolerate the use of PFCs for some specific applications and to help achieve good PFC management in our society.

Important toxicological values and guideline values of PFCs

In this section, we collected and reviewed the guideline/reference values proposed by various institutions, which are based on toxicological data or scientific investigation. These include tolerable daily intake (TDI) and drinking water levels for protecting human health and coastal/river water pollution levels for sound aquatic ecosystems.

Provisional tolerable daily intake (pTDI)

Provisional tolerable daily intakes (pTDI) were established by the Federal Institute for Risk Assessment in Germany (BfR) for PFOS (BfR 2006) and by the Committee on Toxicity (COT) in the U.K., mainly organized by experts, for PFOS and PFOA. The value for PFOS suggested by BfR was calculated to be 0.1 $\mu\text{g}/\text{kg bw}/\text{day}$ based on the No Observed Adverse Effect Level (NOAEL) value (100 $\mu\text{g}/\text{kg bw}/\text{day}$ for decrease of body weight) in a

two-generation reproductive study in rats with uncertainty values for inter-specific extrapolation (UF: 10), intra-species extrapolation (UF: 10), and an additional factor of 10. The value for PFOS by COT is 0.3 $\mu\text{g}/\text{kg}$ bw/day based on the NOAEL value (0.03 mg/kg bw/day for decreased serum triiodothyronine (T3) levels) from the 26-week capsule study of cynomolgus monkeys with uncertainty values for inter-specific and intra-species extrapolations (UF: 10, respectively) (COT 2006a). The PFOA value was suggested as 3 $\mu\text{g}/\text{kg}$ bw/day based on effects on the liver, kidney, haematological, and immune systems with an uncertainty factor of 100 (COT 2006b). Recently, the European Food Safety Authority (EFSA) has set a pTDI value of 0.15 $\mu\text{g}/\text{kg}$ bw/day for PFOS and 1.5 $\mu\text{g}/\text{kg}$ bw/day for PFOA based on a subchronic study of cynomolgus monkeys using the UF of 200 (100 for inter and intra-species differences, 2 for uncertainties laying in the relatively short duration of the key study and the internal dose kinetics) (EFSA 2008). Regarding PFOA, COT members decided to follow the EFSA approach on pTDI derivation, which they considered justifiable. The COT therefore adopted the pTDI derived by the EFSA for PFOA (from 3.0 to 1.5 $\mu\text{g}/\text{kg}$ bw/day) (COT 2009).

So et al. (2006) suggested pTDIs for PFOS and PFOA for infants as 25 and 333 ng/kg-bw/day, respectively, though these values include a large extent of uncertainty factors, such as extrapolation from animal to human, from average human to sensitive human, Lowest Observed Adverse Effect Level (LOAEL) to NOAEL, sub-chronic to chronic and so on, in their derivation. In general, PFOS is regarded to be more toxic than PFOA by one order of magnitude.

Drinking water

Drinking water guideline values have been introduced by various agencies/institutes. The Minnesota Department of Health (MDH) provided health-based values (HBVs) of PFOS, PFOA, and perfluorobutanoate (PFBA) concentrations in drinking water for protecting public health and gradually tightened the values beginning in 2002 (MDH 2008). The HBV for PFOS was chosen as 0.3 $\mu\text{g}/\text{l}$ based on the toxicological studies conducted on cynomolgus monkeys taking decreases in serum high-density lipoprotein and thyroid hormones as endpoints. During the derivation of HBV with a UF of 100, intake rate of drinking water and relative source contribution were taken into account. HBVs for PFOS and PFBA were derived in the same manner as 0.5 and 7 $\mu\text{g}/\text{l}$, respectively. The drinking water guideline values of the Drinking Water Inspectorate (DWI, UK), which were set for drinking water suppliers, are slightly different from those of MDH (DWI 2007). In this case, the drinking water guideline values for PFOS and PFOA were

set in three tiers, and tighter actions were required in higher tiers. For PFOS, guideline value was set at 0.3 $\mu\text{g}/\text{l}$ in tier 1 based on 10% of the pTDI proposed by COT and assuming that a 1-year-old child weighing 10-kg drinks 1 L of water in a day. If the concentration of PFOS exceeds this level, the water suppliers are requested to consult with local health professionals and monitor the level of PFOS in their drinking water. In tier 2, 3.0 $\mu\text{g}/\text{l}$ of PFOS in drinking water was set as the guideline value, taking into consideration the worst case estimates of dietary intakes for a small adult. If the concentration of PFOS exceeds this level, the action in tier 1 plus additional actions are required to reduce the concentration to below 1.0 $\mu\text{g}/\text{l}$. In tier 3, 9.0 $\mu\text{g}/\text{l}$ was set as the guideline value based on 100% of the pTDI for adult. If the concentration of PFOS exceeds this level, the action in tier 2 plus other actions are required. Additionally, consultation with local health professionals should take place as soon as possible and the necessary action undertaken to reduce exposure from drinking water within 7 days. The guideline values for PFOA have been set as 0.3, 10.0, and 90.0 $\mu\text{g}/\text{l}$ in tiers 1, 2, and 3, respectively. Afterward, following the COT recommendation that the pTDI for PFOA should be reduced from 3.0 to 1.5 $\mu\text{g}/\text{kg}$ -bw/day, the guideline values for PFOA have been reduced to 0.3, 5.0, and 45.0 in tiers 1, 2, and 3, respectively (DWI 2009). Although the value of pTDI for PFOA is higher than that for PFOS, the guideline values in tier 1 are the same because the elevated concentrations of PFOA indicate potential contamination by other perfluorinated chemicals.

The German Drinking Water Commission (DWC) suggested a drinking water guideline value for both PFOS and PFOA of 0.3 $\mu\text{g}/\text{l}$. This value is based on the NOAEL, referring to toxicological studies on rats, rhesus monkeys, and cynomolgus monkeys with setting the given UF values, considering 10% of pTDI is contributed to exposure from drinking water. They also suggested lifelong health-based precautionary value for non- or low-potency genotoxic substances (HPV₁, 0.1 $\mu\text{g}/\text{l}$). This HPV₁ also applies to PFOA, PFOS, and other perfluorinated compounds. The precautionary action value for infants and adults (requiring immediate action to reduce adults' intake of PFOS/PFOA in drinking water) have been also chosen as 0.5 and 5.0 $\mu\text{g}/\text{l}$, respectively (DWC 2006).

Recently, USEPA developed provisional health advisories for PFOS (200 ng/l) and PFOA (400 ng/l) for drinking water (USEPA 2009b). The value of PFOA was based on a sub-chronic toxicological study in mice, which resulted in a Benchmark Dose Level (BMDL₁₀) of 0.46 mg/kg/day, and the value of PFOS was based on a sub-chronic study in monkeys, which led to a NOAEL of 0.03 mg/kg/day. In deriving the reference dose (RfD) for PFOS, uncertainty factor of 2,430 was used, dividing into 10 for variations in

the dose–response among humans, 3 for differences in toxicodynamics (default value), and 81 for differences in the rate of clearance (toxicokinetics) between mice and humans, which is derived from the differences in PFOA half-life between mice and humans. An uncertainty factor of 390 for deriving the RfD of PFOS was set in the same way as PFOA, only that of toxicokinetics was different, for which 13 was chosen based on the clearance study of PFOS. Finally, the RfD was converted into the drinking water guideline value, considering a 10 kg child drinking 1 l/day of water with relative source contribution factor of 0.2.

The State of New Jersey recommended the lowest guideline value of drinking water for PFOA as 0.04 µg/l, which was developed by New Jersey Department of Environmental Protection (NJDEP) (2007; Post et al. 2009). This guideline value was derived from NOAEL in the serum of experimental animals, not from NOAEL of external dose, because the half lives of PFOA (and PFOS) in body are largely different between human and experimental animal. Thus, for the derivation of a guideline value of drinking water for PFOA, the serum level of PFOA at NOAEL (endpoint: decreased body weight and haematological effects) in a chronic diet test against adult female rats (1800 µg/l) was converted to NOAEL for human (18 µg/l) with the UF of 100 (10 for intra-species, 10 for inter-species). The NOAEL for human was next converted to the drinking water guideline value for PFOA (rounded guideline value of 0.04 µg/l), considering a relative source contribution factor of 0.2 and a factor of 100, which is used for the conversion of serum level of PFOA to drinking water level of PFOA (The ratio of PFOA concentration in human serum and drinking water is approximately 100:1).

Predicted no-effect concentration (PNEC)

Predicted no-effect concentration is used as a threshold value for protecting wildlife and as a criterion for the assessment of ecological risk of pollutants by comparing this number with the predicted environmental concentration (PEC). A PEC/PNEC ratio larger than one indicates a potential risk for target organisms.

PNECs have been suggested by some institutions based on toxicological studies. The UK Environment Agency suggested a PFOS PNEC of 25 µg/l in freshwater, including an assessment factor of 10 as a toxicological study for the three taxonomic groups (UK Environment Agency 2004). In addition, a PFOS PNEC of 2.5 µg/l was suggested for marine environments, including an assessment factor of 100 as a toxicological study for two taxonomic groups. The PNEC of PFOS in terrestrial environments was suggested as 373 mg/kg soil dry weight based on short-term tests against earthworms. PFOS PNEC

value for oral exposure was derived as 0.0167 mg/kg in food, based on toxicological tests on male rats and consideration of secondary poisoning through bioaccumulation in food chain. Also, PNECs of PFOS in sediment were suggested based on the results in aquatic ecosystem using K_d values and the equilibrium partitioning method. However, K_d varies with sediment conditions, such as organic contents and ionic substances (Higgins and Luthy 2006; Zushi et al. 2010), thus, further development of the PNEC in sediment is required. The Oslo and Paris Conventions for the Protection of the Marine Environment of the North-East Atlantic (OSPAR) used the same PNEC values suggested by the UK Environment Agency, though the values in sediment was not defined (OSPAR Commission 2006).

Environment Canada derived an Estimated No-Effects Value (ENEV) of PFOS, which is similar to PNEC, based on a 2-year dietary study in which the histopathological effects in the livers of male and female rats were determined to be 0.06–0.23 and 0.07–0.21 mg/kg bw/day, respectively (Environment Canada 2006).

The Ministry of the Environment in Japan derived a PNEC of PFOS in an aquatic environment in their report, referencing various toxicological studies (Ministry of the Environment in Japan 2008). They derived PNEC based on chronic toxicity studies consisting of the lowest No-Observed Effect Concentrations (NOEC) for three groups of organisms (algae, shellfish, and fish). The lowest NOEC (shellfish) was used for PNEC by dividing NOEC by an assessment factor of 10, used when the chronic toxicity studies were available for the three groups of organisms, and the obtained PNEC value was 23 µg/l. All the PNECs introduced by various environmental agencies are in the order of ppb to sub-ppm; however, Newsted et al. (2005) established the lowest PNEC value based on a sub-chronic toxicity study on male and female bobwhite quail. The PNEC value in trophic level IV fish-eating birds was derived by monitoring various toxicological endpoints including reproduction, such as egg production, fertility, hatchability and survival, and growth of offspring. The values for males and females were derived as 1.5 and 0.08 µg/g wet weight in liver, respectively. Rostkowski et al. (2006) calculated a PNEC value of 50 ng/l in water based on the lowest PNEC value for trophic level IV fish-eating birds and considering the biomagnification and bioconcentration of PFOS. The information concerning the PNEC of PFOA is scarce compared with PFOS. Based on the result of a toxicological study in rats, the severity of PFOA toxicity was generally lower than that of PFOS. In addition, the bioaccumulation potential of PFOA seems low; thus, the consideration of secondary poisoning may not be taken into account for the PNEC derivation.

Regulations, guideline values, and other items are summarized in Table 2.

Table 2 Summary of the regulation/guideline values for PFCs

Regulation/guideline	Country/institute	Contents	Reference
Production, use, import			
Stockholm convention on POPs	Ratifying countries	PFOS and PFOSF: Partially regulated	UNEP (2009b)
EU Directive	EU member countries	PFOS and the derivatives: partially regulated	EU Directive (2006)
Regulation in Canada	Canada	FTOH, PFOS and its salts: prohibition	Government of Canada (2006, 2009)
SNUR	USEPA	PFOS and its salts, PFOSF, their higher and lower homologue: required to notify when they are manufactured or imported	USEPA (2002)
Water guideline value			
Drinking water guideline value	MDH (Minnesota, USA)	PFOS, PFOA and PFBA: 0.3, 0.5, and 7 µg/l, respectively	MDH (2008)
Drinking water guideline value	DWI (UK)	PFOS and PFOA: 0.3 µg/l in tier 1	DWI (2007)
Drinking water guideline value	DWC (Germany)	PFOS and PFOA: 0.3 µg/l	DWC (2006)
Drinking water guideline value	USEPA	PFOS and PFOA: 0.2 and 0.4 µg/l, respectively	USEPA (2009b)
Drinking water guideline value	NJDEP (New Jersey, USA)	PFOA: 0.04 µg/l	NJDEP (2007)
PNEC	Environment Agency (UK)	PFOS: 25 µg/l in fresh water and 2.5 µg/l in marine environment	UK Environment Agency (2004)
PNEC	–	PFOS: 50 ng/l in water environment considering secondary poisoning	Rostkowski et al. (2006)
pTDI value			
pTDI	BfR (Germany)	PFOS: 150 µg/kg bw/day (for adult)	BfR (2006)
pTDI	COT (UK)	PFOS and PFOA: 0.3 and 1.5 µg/kg bw/day, respectively (for adult)	COT (2006a, 2009)
pTDI	EFSA (EU)	PFOS and PFOA: 0.15 and 1.5 µg/kg bw/day, respectively (for adult)	EFSA (2008)
pTDI	–	PFOS and PFOA: 0.025 and 0.333 µg/kg bw/day, respectively (for child)	So et al. (2006)
Others			
Voluntary phase-out	3M Company	PFOS and its related chemicals: stop their production	3M (2000)
Voluntary reduction of emission	SEMI	PFOS: reduce its emission in the process of the production	WSC (2009)
2010/2015 PFOA Stewardship program	USEPA	PFOA, its precursors and longer chain homologues: reduction of their emission in production process	USEPA (2009a)

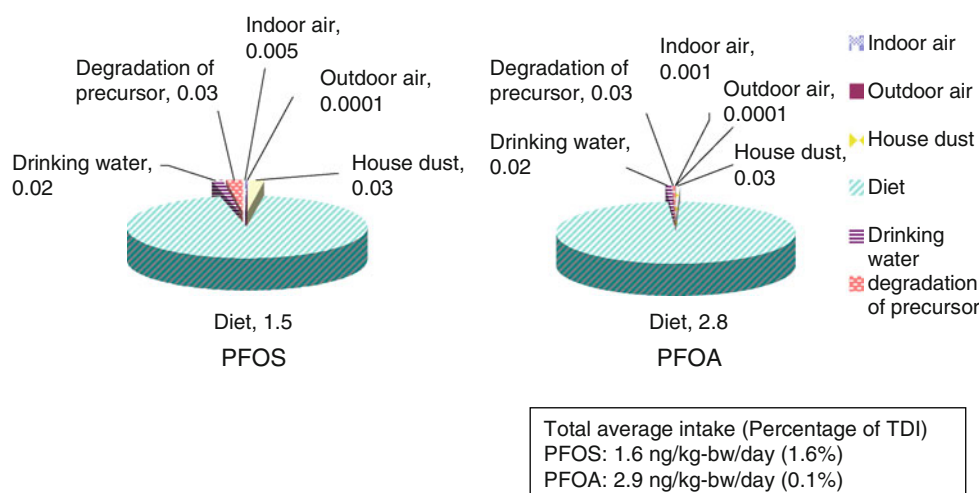
PFC risk assessment

Human health risk

The risk of PFCs, especially PFOS, has been evaluated in various reports, and the risk for human adults is low based on the margin of exposure (MOE), which has been derived from the ratio of the pTDI and the amount of intake. Fromme et al. (2009) evaluated the average (and high) daily intake of PFOS and PFOA, including the degradation contribution from their precursors, as 1.6 (11.0) and 2.9 (12.7) ng/kg bw/day, respectively, and concluded that

these levels are adequately lower than the pTDI of PFOS (100 ng/kg bw/day by BfR) and PFOA (3000 ng/kg bw/day by COT) for the general adult population. The main exposure pathway was judged to be dietary intake, which was supported by the amount of exposure estimated from the blood PFOS level in humans using a one-compartment toxicokinetic model (Harada et al. 2005a; Fromme et al. 2007) (Fig. 1). For residents in highly polluted areas, the intake of polluted drinking water was regarded as the significant exposure pathway (Kärman et al. 2009). Even in this case, the intake of PFOS and PFOA through polluted drinking water was not considered to pose a significant

Fig. 1 Pathway-specific daily intakes of PFOS and PFOA (ng/kg bw/day) (Fromme et al. 2009)



health risk for the general adult population. However, the risk for workers who are exposed to PFCs at work and the risk for infants are of concern. The daily intake of PFOS for plant workers who have been highly exposed would exceed the pTDI.

In an epidemiological study, it was reported that mortality rate from disease in the bladder and other urinary organs for workers who were highly exposed to PFC in PFOSF-based fluorochemical production facility was significantly higher than the expected mortality rate in ordinary group (Alexander et al. 2003). As for infants, daily PFOS and PFOA intakes from breast milk were calculated to be 30 and 17 ng/kg bw/day, respectively, based on data from China (So et al. 2006). The daily PFOS intakes in several Asian countries were reported by Tao et al. (2008) as 11.8 ng/kg bw/day for the overall average with the highest average, 28.7 ng/kg bw/day, from Japanese samples. Thus, highly exposed individuals would exceed the pTDI for infants as suggested by So et al. (2006). Higher exposure per body weight for infants compared with adults was also reported from a scenario-based approach (Trudel et al. 2008).

Some epidemiological studies indicate the possibility of adverse effects on infants by PFCs. A negative correlation between PFOS/PFOA concentration in cord serum and body weight of newborns in USA has been reported (Apelberg et al. 2007). PFOA concentrations in the plasma of pregnant women and the body weights of their newborns were negatively correlated, though a significant PFOS correlation was not observed in Denmark (Fei et al. 2007). On the other hand, a negative correlation between PFOS serum concentration and newborn body weight was observed in Japan, though a significant correlation for PFOA was not observed (Washino et al. 2009). Those results do not necessarily indicate a relationship between PFC and body weight because there are other potential triggers for the decrease of newborn body weight that may

correlate with PFC concentration. However, the risk of PFCs for infants should be further studied.

Ecological risk

The risks for aquatic organisms have been evaluated using PEC/PNEC ratios. The UK Environmental Agency predicted PFOS concentrations in the river water around background areas (regional areas) and downstream of PFOS sources, such as chromium plating, photography, aviation, fire-fighting foam production, photolithography, fabrics, paper treatment, and coating industries (highly polluted area), using the European Union System for the Evaluation of Substances (EUSES) based on seven scenarios that represent degradation of PFOS-related compounds (UK Environment Agency 2004). The calculated PEC/PNEC ratio for the river polluted by fire fighting foams production was as high as 4 for aquatic organisms. The ratios in the regional areas were lower than 0.004 in all scenarios. The agency also evaluated the risk for high trophic level organisms using PNEC for secondary poisoning, which is used for assessment of the top predator because of their increased level of chemical exposure through the food chain. As a result, PEC/PNEC values in all areas, including both regional area and the highly polluted area, for all scenarios exceeded 10 in the freshwater food chain. The agency concluded that major attention should be paid to secondary poisoning.

OSPAR conducted the risk evaluation, especially for marine organisms, in a similar fashion to the UK Environment Agency and reached a similar conclusion (OSPAR Commission 2006). Environment Canada estimated risk by the ratio of the environmental level and the no-effect level (EEV/ENEV), such as PEC/PNEC. The risk quotient of 9.2 for PFOS was obtained using the maximum exposure concentration of 3,770 µg/kg wet weight in the liver of South Hudson polar bear, the top predator in the Arctic,

with an ENEV of 408 $\mu\text{g}/\text{kg}$. It was concluded that the greatest potential risk from PFOS exists for higher trophic mammals.

The results described above show that while the PFOS risk is minimal in regional area of the water environment, the risk for higher trophic animals is significant when the bioaccumulation through the food chain is taken into account. A PNEC value of 50 ng/l in water was suggested for PFOS considering the bioaccumulation in organisms (Rostkowski et al. 2006). Water bodies exceeding this value are not rare. For example, a PFOS survey of rivers in a large EU area revealed that the 90th percentile of the PFOS concentration in EU rivers was 73 ng/l with an average concentration of 39 ng/l (Loos et al. 2009). These values are comparable to the PNEC of 50 ng/l .

Although the available information is limited, the ecological risk posed by PFOA is estimated to be minimal except for special cases of high exposure because the PNEC for PFOA is higher than that of PFOS by an order of magnitude, and the environmental levels are comparable with PFOS.

Further development of risk assessment

For a detailed risk assessment that can be used in practical and reasonable PFC management, the development of PEC and PNEC derivation is required. Also, considering the counter risk and loss of benefits caused by a halt in PFC use is important because these compounds are valuable in industry and in our society for their previously noted special properties. The cost caused by PFOS regulation was evaluated by Risk & Policy Analysts Limited and BRE Environment (RPA & BRE) and the Ministry of the Environment in Canada. Those results have influenced the decision to include PFOS in the Stockholm Convention on POPs, especially the preparation of Annex F (socio-economic considerations) (RPA & BRE 2004; Canada Gazette 2008). Although Canada has suggested that net benefit would be obtained by PFOS regulation, their evaluation did not include the adverse effects of PFCs on ecosystem or the benefits to our lifestyles. Thus, the conclusion is not applicable to developing countries because the use of these chemical would be in high demand for improving their society in addition to the lack of information on the risk to the ecosystem. The benefits to our society from PFOS use in addition to the cost from PFOS risk, such as the cost of replacing the source of the water supply to avoid exposure, should be considered for the PFOS risk management. The development of this analysis, including the further development of risk assessment, would aid decision making in our society, given the concurrent nature of the problems at hand. Further development of the analysis would be more important in developing countries, as mentioned above.

PFC contamination in developing countries

To evaluate the PFC risk level and risk-based management for PFCs in developing countries, information on PFC pollution in developing countries was collected and reviewed in this section.

Unlike in developed countries, relatively few studies have been conducted to investigate PFC contamination of environmental matrices in developing countries. As a consequence, the data are limited, and there is a poor understanding of the sources, distribution, and exposure risks of PFCs in developing countries. Nevertheless, it is becoming evident that PFCs are leaving similar footprints in developing countries as those in their developed counterparts. In a recent study involving nine countries, PFOS was detected in significant measures in the blood of individuals from both developed and developing countries (Kannan et al. 2004). In this study, PFOS levels were highest in blood samples collected from the United States and Poland (>30 ng/l), moderate in samples from Belgium, Italy, Korea, Brazil, Malaysia, and Columbia (3–29 ng/l), and lowest in samples from India (<3 ng/l). The risk to the general population in developing countries is minimal based on the result of risk estimation in developed countries.

In China, the level of PFC contamination was greater in urban rivers than in rural rivers, with urban rivers containing up to 14.1 and 30.8 ng/l PFOS and PFOA, respectively (Jin et al. 2009). Jin et al. (2009) also reported that the PFOS and PFOA levels in drinking water were mostly below 10 ng/l and thus much lower than that in the rivers. However, these samples were collected during 2002–2003, and the production amount of PFOS and related compounds increased dramatically during 2003–2006 (UNEP 2008). Thus, a higher contamination of tap water in China is predicted nowadays than the previously reported values. Rivers in Korea had higher PFOS levels (450 ng/l), while fish in these rivers were found to bioaccumulate PFOS to the level of 612 ng/g dry weight (Naile et al. 2010). Naile et al. (2010) suggested that PFOS contamination in Korean rivers was due to point sources. Similarly, Orata et al. (2009) have attributed PFOS and PFOA contamination in Lake Victoria in Kenya to point sources. Results showed that on average, rivers draining into Lake Victoria contained PFOA (0.4–96.4 ng/l) and PFOS (<0.4 –13.2 ng/l), which were higher levels than in the lake itself (PFOA, 0.4–11.7 ng/l and PFOS, 0.4–2.5 ng/l) (Orata et al. 2009). Earlier, Orata et al. (2008) reported considerable PFC bioaccumulation in some fish in Lake Victoria (Orata et al. 2008). These studies in Kenya represent the only investigations conducted on PFCs in Africa so far.

Studies in South American countries also reported PFC presence in water and biota. For example, drinking water in

Brazil reportedly contained PFOS (0.58–6.70 ng/l), PFOA (0.35–2.82 ng/L), and several related chemicals (Quinete et al. 2009). Untreated river water contained less of PFOS and PFOA in Brazil (Quinete et al. 2009). This seems unusual and indeed contrary to the observation made by Jin et al. (2009) in China that PFCs levels in drinking water were generally below the levels in river water samples. Bioaccumulation and biomagnification of PFCs have also been reported in aquatic animals in South America, with the highest concentrations found in sea mammals such as dolphins (Dorneles et al. 2008; Leonel et al. 2008; Quinete et al. 2009).

Clearly, developing countries are equally exposed to the problems of environmental PFC contamination, though at lower levels than developed countries. Further research is required to fully understand PFC contamination in developing countries. Information is presently lacking regarding PFC accumulation in sediments, soils, and other wildlife besides aquatic organisms in developing countries. Information on airborne levels of these pollutants are also lacking in developing countries; while in Africa, human exposure to these chemicals has yet to be investigated. As has been described in earlier sections of this review, PFCs were widely used in many consumer products (such as surface treatment in carpets, textiles, furniture, non-stick cooking utensils, fire-fighting foams, and insecticides), many of which are presently used in developing countries. Most probably, the various regulations stipulated by the Stockholm Convention, the EU and other countries concerning the production, use and import of PFCs might impact severely on the economics or innovation of technology in developing countries, even though benefit gained in ecological improvement might be quite limited. Thus, the regulation used in developed countries might not be necessarily operative in developing countries for an issue like the management of beneficial chemicals like PFCs. We therefore find it a step in the right direction that the development of alternative materials and the transfer of this knowledge to developing countries is under consideration by UNEP (2009a). Risk-based chemical management is one hope in this difficult and conflictive problem.

Conclusion

Many kinds of regulation concerning the use, production, and import of PFOS, its derivatives, and their related compounds have come into effect, as described in this article. PFC pollution, however, has been widespread due to the extensive use of those chemicals in industrial and consumer products, and also due to their environmental mobility. Those chemical characteristics may make the introduced regulations less effective in developed

countries. There may be further difficulties in developing countries because these countries may not have the economical ability to introduce alternative technologies.

The risk posed by PFOS/PFOA is limited to developing countries except for some areas of high contamination from a point source, though the impact of their homologues and the impact for vulnerable population needs to be investigated.

Although a precautionary method of chemical management is now adopted in various bodies, we need to develop PFC risk assessment that includes their homologues, on which information is lacking. Risk assessment for people who are exposed to high levels of PFCs under specific conditions or for the people vulnerable to PFC exposure, like infants, needs to be improved. Risk-based PFC management needs to be developed in developing countries in particular because PFCs are quite beneficial to these countries. Rather than totally prohibiting the use of PFCs, optimal PFC management strategies should be sought based on the real-world risk evaluation and efforts to maintain the PFC risk level at an acceptable level should be undertaken.

References

- Ahrens L, Felizeter S, Sturm R, Xie Z, Ebinghaus R (2009) Polyfluorinated compounds in waste water treatment plant effluents and surface waters along the River Elbe. *Ger Mar Pollut Bull* 58:1326–1333
- Alexander BH, Olsen GW, Burriss JM, Mandel JH, Mandel JS (2003) Mortality of employees of a perfluorooctanesulphonyl fluoride manufacturing facility. *Occup Environ Med* 60:722–729
- Ammoniums (2006) Canada bans import of four fluorotelomers. *Chem Eng News* 84(26):29
- Apelberg BJ, Goldman LR, Calafat AM, Herbstman JB, Kuklennyik Z, Heidler J, Needham LL, Halden RU, Witter FR (2007) Determinants of fetal exposure to polyfluoroalkyl compounds in Baltimore, Maryland. *Environ Sci Technol* 41:3891–3897
- BfR (2006) High levels of perfluorinated organic surfactants in fish are likely to be harmful to human health. http://www.bfr.bund.de/en/press_information/2006/21/high_levels_of_perfluorinated_organic_surfactants_in_fish_are_likely_to_be_harmful_to_human_health-8172.html
- Canada Gazette (2008) Perfluorooctane sulfonate and its salts and certain other compounds regulations, vol 142, No. 12
- COT (2006a) COT statement on the tolerable daily intake for perfluorooctane sulfonate. <http://cot.food.gov.uk/pdfs/cotstatementpfos200609.pdf>
- COT (2006b) COT statement on the tolerable daily intake for perfluorooctanoic acid. <http://cot.food.gov.uk/pdfs/cotstatementpfoa200610.pdf>
- COT (2009) Committees on toxicity, mutagenicity, carcinogenicity of chemicals in food, consumer products and the environment. <http://cot.food.gov.uk/pdfs/cotcomccoreport2009.pdf>
- Dorneles PR, Lailson-Brito J, Azevedo AF, Meyer J, Vidal LG, Fragoso AB, Torres JP, Malm O, Blust R, Das K (2008) High accumulation of perfluorooctane sulfonate (PFOS) in marine Tucuxi dolphins (*Sotalia guianensis*) from the Brazilian Coast. *Environ Sci Technol* 42:5368–5373

- DuPont (2005) DuPont global PFOA strategy—comprehensive source reduction. U.S. EPA public Docket AR226-1914
- DWC (2006) Provisional evaluation of PFT in drinking water with the guide substances perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) as examples. <http://www.umweltbund.esamt.de/uba-info-presse-e/hintergrund/pft-in-drinking-water.pdf>
- DWI (2007) Guidance on the water supply (water quality) regulations 2000/01 specific to PFOS (perfluorooctane sulphonate) and PFOA (perfluorooctanoic acid) concentrations in drinking water
- DWI (2009) Guidance on the Water Supply (Water Quality) Regulations 2000 specific to PFOS (perfluorooctane sulphonate) and PFOA (perfluorooctanoic acid) concentrations in drinking water. http://www.dwi.gov.uk/stakeholders/information-letters/2009/10_2009annex.pdf
- EFSA (2008) Scientific opinion of the panel on contaminants in the food chain on perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA) and their salts. http://www.efsa.europa.eu/EFSA/Scientific_Opinion/contam_ej_653_PFOS_PFOA_en.pdf?ssbinary=true
- Environment Canada (2006) Ecological Screening Assessment Report on perfluorooctane sulfonate, its salts and its precursors that contain the C₈F₁₇SO₂ or C₈F₁₇SO₃, or C₈F₁₇SO₂N moiety. [http://www.ec.gc.ca/CEPARRegistry/documents/subs_list/PFOS_SAR/PFOS_EC_SAR_June_2006_\(en\).pdf](http://www.ec.gc.ca/CEPARRegistry/documents/subs_list/PFOS_SAR/PFOS_EC_SAR_June_2006_(en).pdf)
- EU Directive (2006) Directive 2006/122/ECOF of the European Parliament and of the Council of 12 December 2006. http://eur-lex.europa.eu/LexUriServ/site/en/oj/2006/l_372/l_37220061227en00320034.pdf
- Fei C, MacLaughlin Jk, Tarone RE, Olsen J (2007) Perfluorinated chemicals and fetal growth: a study within the Danish national birth cohort. *Environ Health Perspect* 115:1677–1682
- Fromme H, Schlummer M, Moller A, Gruber L, Wolz G, Ungewiss J, Bohmer S, Dekant W, Mayer R, Liebl B, Twardella D (2007) Exposure of an adult population to perfluorinated substances using duplicate diet portions and biomonitoring data. *Environ Sci Technol* 41:7928–7933
- Fromme H, Tittlemier SA, Vökel W, Wilhelm M, Twardella D (2009) Perfluorinated compounds—exposure assessment for the general population in western countries. *Int J Hyg Environ Heal* 212:239–270
- FSA (2006) Fluorinated chemicals: UK dietary intakes. <http://www.food.gov.uk/multimedia/pdfs/fsis1106.pdf>
- Giesy JP, Kannan K (2001) Global distribution of perfluorooctane sulfonate in wildlife. *Environ Sci Technol* 35:1339–1342
- Government of Canada (2006) Order adding toxic substances to schedule 1 to the Canadian environment protection act, 1999. *Canada Gazette* 140, <http://gazette.gc.ca/archives/p1/2006/2006-06-17/html/reg1-eng.html>
- Government of Canada (2009) Regulations adding perfluorooctane sulfonate and its salts to virtual elimination list. *Canada Gazette* 143, <http://canadagazette.gc.ca/rp-pr/p2/2009/2009-02-04/html/sor-dors15-eng.html>
- Hansen KJ, Johnson HO, Eldridge JS, Butenhoff JL, Dick LA (2002) Quantitative characterization of trace levels of PFOS and PFOA in the Tennessee River. *Environ Sci Technol* 36:1681–1685
- Harada K, Inoue K, Morikawa A, Yoshinaga T, Saito N, Koizumi A (2005a) Renal clearance of perfluorooctane sulfonate and perfluorooctanoate in humans and their species-specific excretion. *Environ Res* 99:253–261
- Harada K, Nakanishi S, Saito N, Tsutsui T, Koizumi A (2005b) Airborne perfluorooctanoate may be a substantial source contamination in Kyoto area. *Jpn Bull Environ Contam Toxicol* 74:64–69
- Higgins CP, Luthy RG (2006) Sorption of perfluorinated surfactants on sediments. *Environ Sci Technol* 40:7251–7256
- Hölzer J, Goen T, Rauchfuss K, Kraft M, Angerer Jg, Kleeschulte P, Wilhelm M (2009) One-year follow-up of perfluorinated compounds in plasma of German residents from Arnsherg formerly exposed to PFOA-contaminated drinking water. *Int J Hyg Environ Heal* 212:499–504
- Jin YH, Liu W, Sato I, Nakayama SF, Sasaki K, Saito N, Tsuda S (2009) PFOS and PFOA in environmental and tap water in China. *Chemosphere* 77:605–611
- Kannan K, Corsolini S, Falandysz J, Fillmann G, Kumar KS, Loganathan BG, Mohd MA, Olivero J, Wouwe NV, Yang JH, Aldous KM (2004) Perfluorooctanesulfonate and related fluorochlorinated chemicals in human blood from several countries. *Environ Sci Technol* 38:4489–4495
- Kärman A, Harada KH, Inoue K, Takasuga T, Ohi E, Koizumi A (2009) Relationship between dietary exposure and serum perfluorochemical (PFC) levels—a case study. *Environ Int* 35:712–717
- Leonel J, Kannan K, Tao L, Fillmann G, Montone RC (2008) A baseline study of perfluorochemicals in Franciscana dolphin and Subantarctic fur seal from coastal waters of Southern Brazil. *Mar Pollut Bull* 56:778–781
- Loos R, Gawlik BM, Locoro G, Rimaviciute E, Contini S, Bidoglio G (2009) EU-wide survey of polar organic persistent pollutants in European river waters. *Environ Pollut* 157:561–568
- 3M (2000) Phase-out Plan for POSF-Based Products. U.S. EPA public Docket AR226-0600
- MDH (2008) Perfluorochemical contamination in lake Elmo and Oakdale, Washington county, Minnesota. http://www.health.state.mn.us/divs/eh/hazardous/sites/washington/lakeelmo/phael_mooakdale.pdf
- Ministry of the Environment in Japan (2006) Chemicals and the Environment. <http://www.env.go.jp/chemi/kurohon/index.html> (in Japanese)
- Ministry of the Environment in Japan (2008) Perfluorooctane sulfonate and its salts. <http://www.env.go.jp/chemi/report/h19-03/index.html> (in Japanese)
- Moody CA, Strauss SH, Field JA (2003) Occurrence and persistence of perfluorooctanesulfonate and other perfluorinated surfactants in groundwater at a fire-training area at Wurtsmith Air Force Base, Michigan, USA. *J Environ Monit* 5:341
- Murakami M, Kuroda K, Sato N, Fukushi T, Takizawa S, Takada H (2009a) Groundwater pollution by perfluorinated surfactants in Tokyo. *Environ Sci Technol* 43:3480–3486
- Murakami M, Shinohara H, Takada H (2009b) Evaluation of wastewater and street runoff as sources of perfluorinated surfactants (PFSs). *Chemosphere* 74:487–493
- Naile JE, Khim JS, Wang T, Chen C, Luo W, Kwon B-O, Park J, Koh C-H, Jones PD, Lu Y, Giesy JP (2010) Perfluorinated compounds in water, sediment, soil and biota from estuarine and coastal areas of Korea. *Environ Pollut* 158:1237–1244
- Newsted JL, Jones PD, Coady K, Giesy JP (2005) Avian toxicity reference values for perfluorooctane sulfonate. *Environ Sci Technol* 39:9357–9362
- NJDEP (2007) Guidance for PFOA in drinking water at Pennsgrove Water Supply Company. <http://www.nj.gov/dep/watersupply/pfoa.htm>
- Orata F, Quinete N, Werres F, Wilken R-D (2008) Perfluorooctanoic and perfluorooctane sulfonate in Nile Perch and tilapia from gulf of Lake Victoria. *Afr J Pure Appl Chem* 2:75–79
- Orata F, Quinete N, Werres F, Wilken R-D (2009) Determination of perfluorooctanoic acid and perfluorooctane sulfonate in Lake Victoria Gulf Water. *Bull Environ Contam Toxicol* 82:218–222
- OSPAR Commission (2006) Perfluorooctane sulphonate (PFOS). hazardous substances series. http://www.ospar.org/documents/dbase/publications/P00269_BD%20on%20PFOS%20_2006%20version.pdf

- Post GB, Louis JB, Cooper KR, Boros-Russo BJ, Lippincott RL (2009) Occurrence and potential significance of perfluorooctanoic acid (PFOA) detected in New Jersey public drinking water systems. *Environ Sci Technol* 43:4547–4554
- Quinete N, Wu Q, Zhang T, Yun SH, Moreira I, Kannan K (2009) Specific profiles of perfluorinated compounds in surface and drinking waters and accumulation in mussels, fish, and dolphins from southeastern Brazil. *Chemosphere* 77:863–869
- Rostkowski P, Yamashita N, So IMK, Taniyasu S, Lam PKS, Falandysz J, Lee KT, Kim SK, Khim JS, Im SH, Newsted JL, Jones PD, Kannan K, Giesy JP (2006) Perfluorinated compounds in streams of the Shihwa industrial zone and Lake Shihwa, South Korea. *Environ Toxicol Chem* 25:2374–2380
- RPA & BRE (2004) Risk reduction strategy and analysis of advantages and drawbacks for perfluorooctane sulphonate (PFOS). http://www.pops.int/documents/meetings/cop_1/chemlisting/sweden/PFOS_Stage4_FinalReport.pdf
- Saito N, Harada K, Inoue K, Sasaki K, Yoshinaga T, Koizumi A (2004) Perfluorooctanoate and perfluorooctane sulfonate concentrations in surface water in Japan. *J Occup Heal* 46:49–59
- Shoeib M, Harner T, Wilford BH, Jones KC, Zhu J (2005) Perfluorinated sulfonamides in indoor and outdoor air and indoor dust: occurrence, partitioning, and human exposure. *Environ Sci Technol* 39:6599–6606
- Skutlarek D, Exner M, Farber H (2006) Perfluorinated surfactants in surface and drinking water. *Environ Sci Pollut Res* 13:299–307
- So MK, Yamashita N, Taniyasu S, Jiang Q, Giesy JP, Chen K, Lam PKS (2006) Health risks in infants associated with exposure to perfluorinated compounds in human breast milk from Zhoushan, China. *Environ Sci Technol* 40:2924–2929
- Stock NL, Ellis DA, Martin JW, Muir DCG, Mabury SA (2004) Polyfluorinated telomere alcohols and sulfonamides in the North American troposphere. *Environ Sci Technol* 38:991–996
- Tao L, Ma J, Kunisue T, Libelo EL, Tanabe S, Kannan K (2008) Perfluorinated compounds in human breast milk from several Asian countries, and in infant formula and dairy milk from the United States. *Environ Sci Technol* 42:8597–8602
- Tittlemier SA, Pepper K, Edwards L (2006) Concentrations of perfluorooctanesulfonamides in Canadian total diet study composite food samples collected between 1992 and 2004. *J Agric Food Chem* 54:8385–8389
- Trudel D, Horowitz L, Wormuth M, Scheringer M, Cousins IT, Hungerbuhler K (2008) Estimating consumer exposure to PFOS and PFOA. *Risk Anal* 28:251
- UK Environment Agency (2004) Environmental Risk Evaluation Report: perfluorooctanesulphonate (PFOS)
- UNEP (2008) Consideration of new information on perfluorooctane sulfonate (PFOS). http://www.unon.org/confss/doc/unep/pops/POPRC_04/POPRC_4_INF_17/K0841478%20POPRC-4-INF17.pdf
- UNEP (2009a) Annotated outline for a guidance document on perfluorooctane sulfonate alternatives. <http://chm.pops.int/Convention/POPsReviewCommittee/hrPOPRCMeetings/POPRC5/POPRC5Documents/tabid/592/language/en-US/Default.aspx>
- UNEP (2009b) Report of the conference of the parties of the Stockholm convention on persistent organic pollutants on the work of its fourth meeting. <http://chm.pops.int/Portals/0/Repository/COP4/UNEP-POPS-COP.4-38.English.pdf>
- USEPA (2002) Perfluoroalkyl sulfonates; significant new use rule; final and supplemental proposed rule. Federal Register 67, <http://www.epa.gov/fedrgstr/EPA-TOX/2002/March/Day-11/t5747.pdf>
- USEPA (2009a) 2010/15 PFOA Stewardship Program. <http://www.epa.gov/opptintr/pfoa/pubs/stewardship/index.html>
- USEPA (2009b) The toxicity of perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS). <http://www.epa.gov/opptintr/pfoa/pubs/activities.html>
- Wang T, Wang Y, Liao C, Cai Y, Jiang G (2009) Perspectives on the inclusion of perfluorooctane sulfonate into the Stockholm convention on persistent organic pollutants. *Environ Sci Technol* 43:5171–5175
- Washino N, Saijo Y, Sasaki S, Kato S, Ban S, Konishi K, Ito R, Nakata A, Iwasaki Y, Saito K, Nakazawa H, Kishi R (2009) Correlations between prenatal exposure to perfluorinated chemicals and reduced fetal growth. *Environ Health Perspect* 117:660–667
- WSC (2009) Joint statement of the 13th meeting of the world semiconductor council (WSC). <http://www.eeca.eu/data/File/ESIA%20WSC/Beijing%202009%20WSC%20Joint%20Statement%20Final.pdf>
- Zushi Y, Masunaga S (2009a) First-flush loads of perfluorinated compounds in stormwater runoff from Hayabuchi River basin, Japan served by separated sewerage system. *Chemosphere* 76:833–840
- Zushi Y, Masunaga S (2009b) Identifying the nonpoint source of perfluorinated compounds using a geographic information system based approach. *Environ Toxicol Chem* 28:691–700
- Zushi Y, Takeda T, Masunaga S (2008) Existence of nonpoint source of perfluorinated compounds and their loads in the Tsurumi River basin, Japan. *Chemosphere* 71:1566–1573
- Zushi Y, Tamada M, Kanai Y, Masunaga S (2010) Time trends of perfluorinated compounds from the sediment core of Tokyo Bay, Japan (1950s–2004). *Environ Pollut* 158:756–763