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The emergence of persistent organic pollutants in the environment: the occurrence and treatment of perfluorinated compounds

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Abstract

Perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) are two perfluorinated compounds (PFCs) used to produce everyday goods, including clothing, carpets, textiles, upholstery, paper, packaging and cleaning products. PFOS and PFOA belong to the class of chemical contaminants known as persistent organic pollutants (POPs). They are particularly difficult to deal with once released into the environment because they do not break down easily, they can travel long distances in the air or water supply and they can accumulate in human and animal tissue. Scientists have linked these chemicals, which can be toxic for both humans and wildlife, with serious environmental and health risks. Researchers have reported PFC contamination in river, tap and bottled water in Japan, the US, Europe and in developing countries such as Thailand, Malaysia and Vietnam. Conventional treatment technologies do not remove PFCs effectively from our water supplies, compounding the problem. Some advanced methods have been found to be effective in treating PFOS and PFOA in the water environment. However, these technologies have yet to be applied on a large scale due to regulative and economical constraints on their development.

Keywords

Micropollutant, perfluorinated compounds, PFOS, PFOA, water treatment processes.

1. Introduction

The protection of our fresh water resources has become increasingly difficult because of growing water scarcity, the increasing demand for water and deterioration of water quality by polluting substances. Water contamination by new chemicals, particularly organic micropollutants, is emerging as a major concern. Persistent organic pollutants (POPs) are a class of organic compounds that resist photolytic, chemical and biological degradation and remain intact in the environment for long periods of time. Capable of travelling long distances through air and water, POPs can accumulate in the fatty tissue

of living organisms, where — if the level of accumulated chemical becomes high enough — they are toxic to humans and wildlife (United States Environmental Protection Agency (USEPA) 2006).

Organic micropollutants are not only persistent: they often occur in very low concentrations that are difficult to detect. For both these reasons, their management requires an approach that is different from the conventional treatments used to manage normal wastewater or other pollution problems caused by common pollutants such as nitrogen and phosphorous. The problems of managing organic micropollutants escalate when new micropollutants with unique characteristics are introduced into the environment because new treatment methods must be engineered to deal with them.

Some of the newest types of organic micropollutants are perfluorinated compounds (PFCs), a family of organofluorine compounds in which hydrogens are replaced by fluorines linked on a carbon chain. PFCs have been synthesized and used widely in industrial and commercial applications since the 1960s to produce everyday goods such as carpets, leather, textiles, paper and packaging, coating materials, cleaning products, pesticides, insecticides and fire-fighting foams (European Commission 2006). Perfluorooctane sulfate (PFOS) and perfluorooctanic acid (PFOA) are two of the predominant PFCs: PFOS carrying sulfonate as its terminal group, and PFOA carrying carboxylate as its terminal group (Figure 1). Specific applications of PFOS include surface treatment, paper protection and performance chemicals, while PFOA is used as an emulsifier and surfactant (Organization for Economic Cooperation and Development (OECD) 2002).

Production of perfluoroalkyl carboxylates (PFCAs) — the group of PFCs to which PFOA belongs — began in 1947, using an electrochemical fluorination process. Prevedouros et al. (2006) estimated the global historical production of PFCAs up to 2006 to be 4,400–8,000 tonnes, while the total global (both direct and indirect) emissions to the environment were 3,200–7,300 tonnes. The majority of these emissions came from the production of fluoropolymer, which is applied to manufactured metal parts to make them resistant to oil and water (for example, fluoropolymer is used in the non-stick coating applied to frying pans). Among the various forms of PFCAs, PFOA is the most frequently detected compound in the water environment and biota. PFOS-related
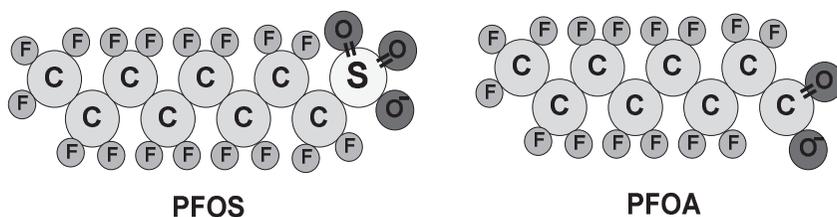

PFOS **PFOA**

Figure 1. The chemical structures of PFOS and PFOA
Source: Organization for Economic Cooperation and Development 2002

chemicals are produced in the US, Europe and Japan. In 2000, approximately 4,500 tonnes of PFOS-related chemicals were produced.

Used in the worldwide manufacture of various products for some 40 years, PFOS production is now in decline, owing largely to a decision made in 2000 by major US manufacturer 3M to phase out the manufacture and use of PFOS from 2001. PFOA is still being manufactured and used. Released into the water environment through point-source (industrial and sewage treatment plant) and non-point-source (surface run-off and atmospheric) discharges for many decades, the presence, persistence and bioaccumulative potential of both these micropollutants are now the cause of global concern. PFOS and PFOA have been detected in the livers, bladders and blood of humans and many animals, including fish, birds and marine mammals (International POPs Elimination Network (IPEN) 2005; Renner 2001). Fish-eating animals higher up the food chain, such as mink and bald eagles, contained higher concentrations, indicating the bioaccumulative properties of PFOS (Giesy and Kannan 2001). PFOS has been shown to be toxic in rats and rabbits in the laboratory (3M 2003; Renner 2001). A two-year experiment on rats has shown that exposure to PFOS results in liver tumors and thyroid follicular cell tumors (OECD 2002). The lowest observed adverse effect level was 0.4 mg/kg of body weight per day and the no-observed-adverse-effect level of PFOS was estimated to be 0.1 mg/kg of body weight per day (OECD 2002). Despite this mounting body of evidence, the PFOS and PFOA risk assessment for humans has yet to be understood. The exposure criteria of PFCs for human health are still unclear and there has been no agreement from the scientific community on what level of exposure is acceptable.

Despite scientific uncertainty, the potential environmental and health risks of increasing amounts of PFOS and PFOA in our water supply are real enough to prompt action. At the international level, in 2009, PFOS and related compounds were recognized as new POPs by the Stockholm Convention on Persistent Organic Pollutants, which classified them in the same class of POPs as dichlorodiph-enyltrichloroethane (DDT). The convention restricted the production of PFOS and related compounds for use in photo-imaging, fire-fighting foam, insect baits for leaf-cutting ants and specific exemptions for which substitute chemicals are not available, such as in the production of metal plating, leather and apparel, textiles and upholstery, paper, packaging, rubber and plastics (International Institute for Sustainable Development (IISD) 2009). One regional response came from the European Commission, which released a directive in December 2006 prohibiting the sale of products containing PFOS in Europe (EC 2006). In the US, at the national level, the US Environmental Protection Agency (USEPA) released the Initiate 2010/15 PFOA stewardship program in 2006. The purpose of this program was to help eight major PFC producers, including 3M and Daikin, to phase out their use of PFOA by 95 per cent in 2010 (USEPA 2006).

Again in the US, a number of states are also making their own precautions. The Minnesota Department of Health recommended 7 μg/L (micrograms per litre) in drinking water as the safe level of PFOA for human health in 2002, revising it to 0.5 μg/L in 2007 (Minnesota Department of Health 2007). Other PFCs were included in the criteria — 0.3 μg/L for PFOS, 1 μg/L for PFCA and 0.6 μg/L for perfluoroalkyl sulfonate (PFAS is the group of PFCs to which PFOS belongs) — because all of them have been detected in local surface water and drinking water. The state of New Jersey has released a guideline, recommending PFOA in drinking water be no more than 0.04 μg/L (New Jersey Department of Environmental Protection 2009).

2. Perfluorinated compounds in the water environment
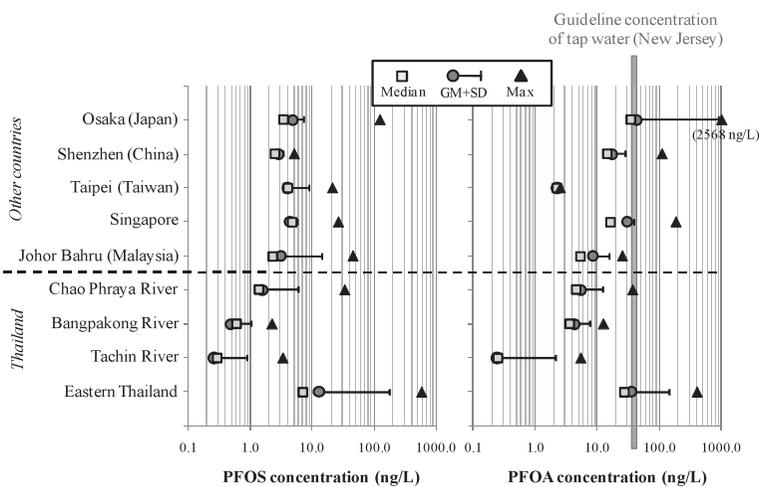
2.1 PFCs in surfacewater

Once PFOS and PFOA are released into the environment, they persist for many years and are able to travel long distances without easily breaking down. They are found in surface waters in developed and developing countries, including those of North America, Europe and Asia. In the US, Sinclair et al. (2004) reported that PFOS and PFOA were found in Michigan surface waters — the highest concentration of PFOS and PFOA reached 29 ng/L (nanograms per litre) and 36 ng/L, respectively, in areas where several paper mills were located. Sinclair et al. later found PFOS and PFOA concentrations in the surface water of New York State to be 0.8–30 ng/L and 10–173 ng/L respectively. In samples taken at Lake Onondaga, PFOS concentration was remarkably high (198–1090 ng/L) as a result of industrial discharges (Sinclair and Kannon 2006). In Northern Europe, Berger et al. (2004) found that different sources of water contained widely varying concentrations of PFOA: 5.2 ng/L in seawater, 7.8 ng/L in lake water and 13.1 ng/L in rainwater. PFOS concentration was very low in the samples that averaged less than 1 ng/L for sea, lake and rainwater samples.

Japan was the first Asian country to express concern about PFOS and PFOA contamination of the environment. Taniyasu et al. (2003), who wrote the first report on PFOS contamination of the Japanese water environment, found water in Tokyo Bay had a high PFOS concentration of 59 ng/L (the national average is 26 ng/L). Lower PFOS concentrations were detected in Osaka Bay (12 ng/L), Ariake Bay (9 ng/L), Lake Biwa (7.4 ng/L) and Seto Inland Sea (less than 4.3 ng/L). A later survey conducted by Saito et al. (2004) found PFOS concentrations in the range of 0.89–3.69 ng/L and PFOA concentrations in the range of 0.97–21.5 ng/L contaminating surface water at many locations in Japan. The highest PFOA concentration of 21.5 ng/L was found in the surface water of the Kinki area in western Japan. The contamination source was a sewage treatment plant that was discharging about 18 kg of PFOA per day into the nearby Kanzaki River, which flows into Osaka Bay. In addition, Lien (2007) found that

some sewage treatment plants discharged treated effluents containing PFOS and PFOA into the Yodo River, which also flows into Osaka Bay. PFOS and PFOA concentrations in these effluents were in the ranges of 3–76 ng/L and 25–922 ng/L respectively. Therefore, the amounts of PFOS and PFOA discharged into Osaka Bay through the Yodo River were estimated to be 64 g/d (grams per day) and 375 g/d respectively.

PFCs have been found in developing as well as developed countries. Kunacheva et al. (2009a and 2009b) investigated the contamination of PFCs in surface water in central and eastern Thailand. PFOS and PFOA concentrations in the Chao Phraya River mainstream varied in the range of 2.8–3.4 ng/L and 5.9–12.4 ng/L respectively. PFOS increased from 0.8 to 2.3 ng/L along the river, indicating the presence of a number of contamination (point and non-point) sources. Higher levels of PFCs were detected in industrialized areas in eastern Thailand. PFOS and PFOA concentration in rivers varied from 3.1 to 36.9 ng/L and 14.8–402.3 ng/L. The concentrations in reservoirs ranged from 5.3 to 572.5 ng/L for PFOS and 16.7–72.4 ng/L for PFOA. The sampling points of these surface waters were located downstream of industrial zones. The results show that industrial activities are a major source of PFC contamination in surface water.

The above results are shown in Figure 2, which compares PFOS and PFOA concentrations found in surface water in Thailand with concentrations of PFOS and PFOA found in the surface waters of other Asian countries. Bangpakong River and Tachin River are located in suburban areas in Central Thailand, where industrial

Note: GM＝Geometric mean, SD＝Standard deviation

Figure 2. Comparison of concentrations of PFOS and PFOA in surface water in Thailand and other Asian countries
Source: the author, based on Lien et al. 2008, Tanaka et al. 2008 and Kunacheva et al. 2009a and 2009b

activity is low. PFOS concentrations in these rivers were accordingly very low compared with concentrations found in other countries. However, PFOS levels in industrialized areas were comparable to those in other Asian countries — the highest concentration levels appearing in Eastern Thailand surface water, where sampling points were located near key industrial zones, suggesting higher discharges of PFCs.

The New Jersey Department of Environmental Protection (2009) set a guideline for a "safe" concentration of PFOA in drinking water at 40 ng/L. When we take into account that all the results for surface-water samples summarized in Figure 2 are the sources of tap water, the results shown in the figure are somewhat disturbing. Eastern Thailand's PFOA range of 14.8–402.3 ng/L is far above concentrations deemed safe by the New Jersey guideline and PFOA levels in samples taken in Osaka and Singapore are also above the guideline. If, in each case, conventional water treatment plants are not effective in removing PFOA from surface water when processing it for use as tap water, what effect might this compound have on the people living in the area?

2.2 PFCs in tap water

The occurrence of PFCs in drinking water is of great concern. Researchers have reported PFC contamination in river, tap and bottled water in Japan (Saito et al. 2004; Takagi et al. 2008), the US (Boulanger et al. 2004; Hansen et al. 2002) and Europe (Ericson et al. 2008; Skutlarek et al. 2006). Tap water in several Japanese cities was found to contain PFOS and PFOA levels ranging from non-detectable (below the limit of detection for the analytical equipment used) to 6.8 ng/L and 0.3–37.5 ng/L respectively (Lien 2007). Tap water in cities from other countries, including Canada, China, Malaysia, Sweden, Thailand and Vietnam, were also found to contain PFOS and PFOA in the ranges of non-detectable to 13.2 ng/L and non-detectable to 109 ng/L respectively (Tanaka et al. 2008).

A comprehensive case study of PFCs in water treatment plants recently conducted in Bangkok, Thailand, collected samples from water treatment plants, tap water and bottled water (Kunacheva et al. 2010). The average PFOS and PFOA concentrations in tap water were found to be 0.2 ng/L and 3.6 ng/L respectively. The tap water results also showed that PFOS and PFOA concentrations were not similarly detected in all areas of the city. The concentrations of PFOS and PFOA detected across the city differed because they came from different water sources. Surprisingly, bottled water — surface water that has been treated for sale commercially — in Bangkok contained higher levels of PFCs than tap water, with average concentrations of 0.2 ng/L (PFOS) and 10.6 ng/L (PFOA).

These results are summarized in Figure 3, which compares PFOS and PFOA concentrations in tap water and bottled (drinking) water in Bangkok with tap-water concentrations detected in other Asian cities. The figure shows that PFOS
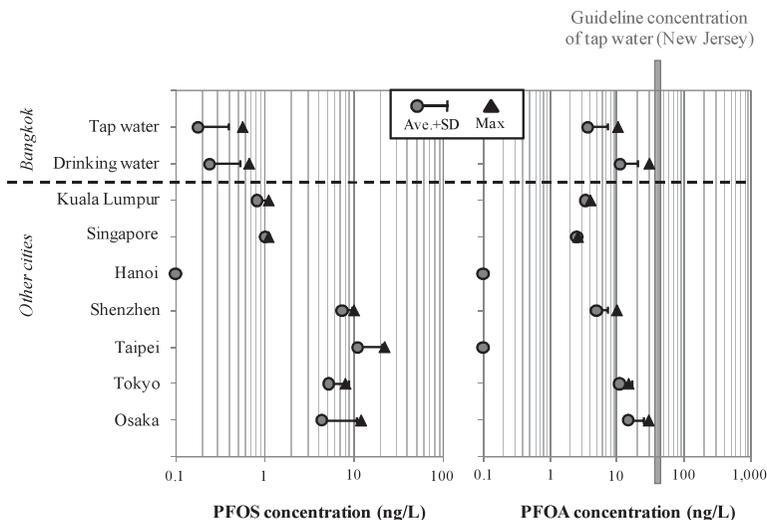
Figure 3. Comparison of concentrations of PFOS and PFOA found in tap and
drinking water in Bangkok and other Asian cities
Source: Kunacheva et al. 2010

concentrations varied from 0.2 to 7.3 ng/L across the south-east and east Asian region.
Bangkok tap and drinking-water samples in the detectable range showed lower PFOS
concentration on average, while the highest levels of PFOS detected were found in
Taipei. PFOS concentrations detected in tap water in the south-east Asian cities of
Bangkok, Kuala Lumpur, Singapore and Hanoi were lower than concentrations found in
Shenzhen, Taipei, Osaka and Tokyo. In each case, the trends of PFOS concentrations
are comparable to those seen already in Figure 2, which showed PFOS concentrations
for surface waters in Bangkok and other cities across south-east and east Asia.

Figure 3 also shows PFOA concentrations ranging from 2.5 to 14.8 ng/L across the
samples taken in the eight cities surveyed. The range of PFOA concentrations
detected in Bangkok was the highest among the other cities in south-east Asia — the
average PFOA concentration in Bangkok tap water was comparable to that in
Shenzhen although significantly lower than in Osaka and Tokyo. It should be noted,
however, that even the highest PFOA concentrations detected in tap and drinking-
water samples in all the cities surveyed were within the limits set in the 2009 guideline
by the New Jersey Department of Environmental Protection.

Occurrences of PFCs in water treatment plants during treatment processes were
also studied by Kunacheva et al. in 2010. The average rates of the removal of PFOS and
PFOA during the conventional treatment processes in the liquid phase (without
suspended solids in water) were 45 per cent and −4 per cent respectively. These
removal rates were comparable to those recorded in an earlier report by Takagi et al.
(2008) in Japan, which showed conventional treatment processes having a limited effect

on the removal rates for PFCs and noted that the breakdown of larger chemicals into PFOS and PFOA during the treatment processes could actually increase the concentrations of PFOS and PFOA in some cases (shown above as a minus percentage in the removal rate). Both studies showed that although some PFOS can be removed by current treatment processes, the removal rates of PFCs are lower than 50 per cent in most water treatment plants because conventional treatment processes are not designed for treating PFCs. The treatment process did not remove PFCs completely, particularly in the liquid phase samples. The results of both Takagi et al. (2008) and Kunacheva et al. (2010) indicate that current treatment processes cannot remove PFCs completely.

2.3 PFCs in wastewater

PFC contamination of domestic wastewater generated by household activities has been reported in many countries. In the US, Boulanger et al. (2005) have reported PFOS and PFOA contamination of domestic wastewater in Iowa, which contained 26 ng/L and 22 ng/L respectively. Similar PFC levels were also found in the liquid waste or effluent discharged from municipal wastewater treatment plants in the Pacific northwest, where the range of PFOS and PFOA concentrations were in the lower range of 12–27 ng/L and 7–16 ng/L respectively (Schultz et al. 2006). Higher PFCs concentrations were detected in six wastewater treatment plants in New York State. PFOS and PFOA concentrations varied widely, from 4 to 68 ng/L and 58–1,050 ng/L respectively (Sinclair and Kannan 2006).

Wastewater treatment plants receive wastewater primarily from domestic and commercial activities. The results cited above suggest that PFCs can occur at hundreds of ng/L concentrations without influence from industrial activities. A study of PFC contamination in domestic wastewater was also conducted in Switzerland's Glatt Valley watershed. Huset et al. (2008) reported the PFC concentration in influents and effluents from seven wastewater treatment plants. PFOS and PFOA ranged from 18–449 ng/L to 5–9 ng/L respectively in the influents, and 16–303 ng/L and 12–35 ng/L in the effluents. The results show that PFOS and PFOA were not removed efficiently in all seven wastewater treatment plants, leaving them to contaminate the water environment downstream and possibly enter the local food chain.

In Asia, PFC contamination of wastewater was reported in Japan and Singapore. Murakami et al. (2008) collected samples from the influents and effluents of a number of wastewater treatment plants in the Kanto region of Japan. PFOS and PFOA concentrations were found in the influents in the range of 14–336 ng/L and 14–41 ng/L and 42–635 ng/L and 10–68 ng/L respectively. PFC concentrations in the effluents were higher than in the influents. These effluent concentrations are comparable to those recorded in other studies in Japan (Qiu 2007) and other countries (Schultz et al.

2006; Sinclair and Kannan 2006). The cause is probably because PFCs are produced through the biodegradation of larger chemicals. In Singapore, Yu et al. (2009) reported PFOS and PFOA concentrations from two wastewater treatment plants, which receive wastewater consisting of 95 per cent domestic wastewater and 5 per cent industrial and commercial wastewater. Due to seasonal variation, the range of PFOS and PFOA concentrations were 11–461 ng/L and 16–1,057 ng/L respectively.

According to studies conducted in developing countries, industrial wastewater contained higher levels of PFCs than domestic wastewater. In 2009, field studies were conducted in 10 industrial zones in central and eastern Thailand to identify the occurrence of PFCs in industrial wastewater. The studies detected elevated concentrations of PFCs in the wastewater generated by the electronics, textile, chemical and glass-making industries. Total PFC concentrations in the influent of wastewater treatment plants ranged from 40 to 3,344 ng/L (Kunacheva et al. 2009c). In total, 10 industrial zones released 188.41 g/d of PFCs, with PFOS and PFOA the dominant PFCs in all industrial zones. Figure 4 compares the PFOS and PFOA concentration in the influent and effluent of the 10 wastewater treatment plants surveyed. It is immediately evident from the figure that the concentration values are not significantly different before and after treatment: indeed, in some cases, the concentrations of PFOS and PFOA are higher in the effluent than in the influent. The wastewater treatment plants in all 10 industrial zones surveyed are using biological processes, such as activated sludge and the use of wetlands, which are not designed for treating PFCs. The results of this survey would seem to prove conclusively that biological processes such as these are ineffective at removing PFCs.
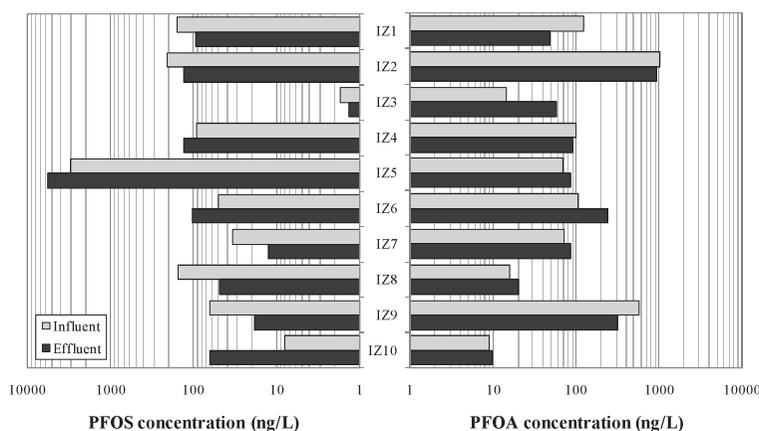

Figure 4. PFOS and PFOA concentration in the influent and effluent of
wastewater treatment plants in 10 industrial zones (IZ) in Thailand
Source: Kunacheva et al. 2009c

3. Treatment of perfluorinated compounds

The fact that conventional techniques are not sufficient to treat PFCs has encouraged the development of alternative treatments. The alternative advanced methods that have already been tested include UV irradiation (Hori et al. 2004), membrane process (Tang et al. 2006) and adsorption (Senevirathna et al. 2010a).

3.1 Oxidation of PFCs using UV irradiation

UV radiation can be an effective viricide and bactericide. Disinfection using UV radiation is commonly used in wastewater treatment applications and is increasingly used in the treatment of drinking water. Studies show that irradiation at UV254 nm (nanometers) and UV254＋UV185 nm can degrade PFCAs (Qiu 2007). They also show that PFASs can be degraded by UV254＋185 photolysis, although the products after decomposition have yet to be identified conclusively. The difficulty of this process is that the presence of natural organic matter in water reduces the performance of UV photolysis for PFCAs. Ozone-related processes were also studied but they were found to be ineffective in degrading PFC molecules (Qiu 2007).

3.2 Membrane filtration

Membrane filtration is one of the possible candidate mechanisms for separating PFCs in real-scale applications. Tang et al. (2006) investigated the feasibility of using reverse-osmosis membranes for treating semiconductor wastewater containing PFOS. Reverse osmosis is a filtration method that removes many types of large molecules and ions from solutions by applying pressure to the solution when it is on one side of a selective membrane. Tang et al. reported that the reverse-osmosis membranes generally rejected 99 per cent or more of the PFOS for feed concentrations of 0.5–1500 mg/L (milligrams per litre). Darling and Reinhard (2008) also measured PFC removal using four nano-filtration membranes. They reported that the amniotic membrane rejected more than 95 per cent of PFOS. It was noted that the performance decreased by up to 35 per cent if the pH of the water being treated was less than three.

3.3 PFC adsorption

In many cases, adsorption has been demonstrated to be an effective and economical method of removing many organic pollutants from water. Adsorption is the adhesion to a surface of atoms, ions, biomolecules or molecules of gas, liquid or dissolved solids. Researchers have reported the effectiveness of adsorption methods using anion exchange synthetic polymers (Qiang et al. 2009) and activated carbon (Qiang et al. 2009; Valaria and Reyes 2008; Qiu 2007) to eliminate some PFCs in environmental water. Senevirathna et al. (2010a) have tested the isotherm and kinetic characteristics of

adsorption methods using batch and column experiments.

Non-ion exchange synthetic resins are used effectively as adsorbents for different applications in the food industry. Melin (1999) has shown the effectiveness of non-ion exchange synthetic resins (including Dowex and Amberlite) as adsorbents to eliminate methyl tertiary butyl ether (MTBE) from US groundwater. Similar to granular activated carbon (GAC), granular synthetic resins can also be used in fixed bed filters, particularly to eliminate organic pollutants. The process flow configuration of a synthetic resin system is very similar to that of a GAC system. The main difference between the two is the provision of a regeneration process for resin systems because resins can be regenerated on site (Melin 1999). Synthetic resin columns can be operated in series, in parallel or as a combination of the two configurations, depending on a number of factors, such as the need for continuous operation, space constraints, effluent criteria, service-cycle time constraints, operation logistics and requirements for multi-barrier treatment. At present, the higher unit cost of resins compared with the more traditional sorbent GAC is the main reason why resin sorbents are not used more widely in drinking water treatment scenarios (Melin 1999).

Senevirathna et al. (2010b) have also investigated PFC adsorption on non-ion exchange resins. Their tests applied a comparatively higher flow rate of 15 mL/min, which ensured 1.3 minute retention time. Inflow PFOS concentration was kept constant at 10 μg/L. It was noticed for all polymers that the percentage removal efficiency is more than 99 per cent. Amb XAD4 showed excellent performance by removing more than 99.99 per cent of PFOS from the first 23,000 bed volumes that passed through it.

4. Conclusions

The global contamination of the water environment and, consequently, of humans and animals by PFOS and PFOA is an emerging environmental and health concern. These new chemical pollutants are persistent, bioaccumulative and can be toxic. They are found in surface and tap water in both developed and developing countries in the range of 1–1090 ng/L for PFOS and 10–922 ng/L for PFOA. Higher concentrations have been detected around industrialized areas, which are recognized as the major sources of PFC-contaminated water.

PFCs in tap and bottled water are also of great concern. Researchers have reported PFC contamination of surface, tap and bottled water in Japan, the US, Europe and developing countries such as Thailand, Malaysia and Vietnam. Surveys have shown high concentrations of PFOS and PFOA in tap water, ranging from non-detectable to 13.2 ng/L and non-detectable to 109 ng/L respectively.

Studies have also reported that the conventional technologies used in treatment

plants do not remove PFCs adequately. Water treatment plants in Bangkok removed less than 50 per cent of PFCs. Advanced methods such as UV-visible light irradiation, membrane processes and adsorption already exist as alternative methods to degrade or remove PFCs. Among them, membrane filtration and adsorption processes are proving particularly effective for removing PFOS, with reverse-osmosis membrane testing showing PFOS removal rates of up to 99 per cent and adsorption processes using Amb XAD4 removing more than 99.99 per cent of PFOS. However, these technologies have yet to be applied commercially.

Clearly, new studies are essential to assess how these advanced treatments might be applied on a real scale. Rigorous health-impact assessments to ascertain the effects of PFCs in animal and human tissue are an immediate necessity. These would enable more stringent national and international regulations to be implemented to limit the point-source contamination by industries that manufacture PFCs and related products and would discourage wastewater treatment plants from discharging PFC-contaminated effluents into nearby watercourses or on land. Studies to find harmless or less harmful alternatives to these chemical compounds should also be made a top priority.

References

3M (2003) 'Environmental and health assessment of perfluorooctane sulfonic acid and its salt', available online at http://solutions.3m.com/wps/portal/3M/en_US/PFOS/PFOA/Information/Health-Environment (Feb, 2010).

Berger, U., Järnberg, U., and Kallenborn, R. (2004) 'Perfluorinated alkylated substances (PFAS) in the European Nordic environment', *Organohalogen Compounds,* 66, pp. 4046–4052.

Boulanger, B., Vargo, J., Schnoor, J. L., and Hornbuckle, K. C. (2004) 'Detection of perfluorooctane surfactants in Great Lakes water', *Environmental Science Technologies,* 38 (2004), pp. 4064–4070.

Boulanger, B., Vargo, J. D., Schnoor, J. L., Hornbuckle, K. C. (2005) 'Evaluation of perfluorooctane surfactants in a wastewater treatment system and in a commercial surface protection product', *Environmental Science Technologies*, 39, pp. 5524–5530.

Darling, E. S. and Reinhard, M. (2008) 'Nanofiltration for trace organic contaminant removal: structure, solution and membrane fouling effects on the rejection of perfluorochemicals', *Environmental Science Technologies*, 42, pp. 5292–5297.

Ericson, I., Nadal, M., Van Babel, B., Lindstrom, G., and Domingo, J. L. (2008) 'Levels of perfluorochemicals in water samples from Catalonia, Spain: is drinking water a significant contribution to human exposure?', *Environmental Science Pollution and Research*, 15, pp. 614–619.

EUROPA (2006) 'POP — persistent organic pollutants', available online at http://ec.europa.eu/environment/pops/index_en.htm (June 2010).

European Commission (2006) 'Directive of the European parliament and of the council: relating to restrictions on the marketing and use of perfluorooctane sulfonates', (amendment of Council Directive 76/769/EEC). Commission of the European Communities.

Giesy, J. P. and Kannan, K. (2001) 'Global distribution of perfluorooctane sulfonate in wildlife', *Environmental Science Technologies,* 35(7), pp. 1339.

Hansen, K. J., Johnson, H. O., Eldridge, J. S., Butenhoff, J. L., and Dick, L. A. (2002) 'Quantitative characterization of trace levels of PFOS and PFOA in the Tennessee River', *Environmental Science Technologies*, 36, pp. 1681–1685.

Hori, H., Hayakawa, E. and Einaga, H. (2004) 'Decomposition of environmentally persistent perfluorooctanoic acid in water by photochemical approaches', *Environmental Science Technologies*, 38, pp. 6118–6124.

Huset, C. A., Chiaia, A. C., Barofsky, D. F., Jonkers, N., Kohler, H. E., Ort, C., Giger, W. and Field, J. A. (2008) 'Occurrence and mass flows of fluorochemicals in the Glatt Valley watershed, Switzerland', *Environmental Science Technologies*, 42, pp. 6369–6377.

International Institute for Sustainable Development (2009) 'Summary of the fourth conference of the parties to the Stockholm Convention on Persistent Organic Pollutants: 4–8 May 2009', available online at www.iisd.ca/chemical/pops/cop4/

International POPs Elimination Network (2005) 'Body burden community monitoring handbook: new POPs candidates', available online at www.oztoxics.org/cmwg/chemicals/new%20pop.html (February 2010).

Kunacheva, C., Boontanon, S. K., Fujii, S., Tanaka, S, Musirat, C., Ardsalee, N. and Wongwattana, T. (2009a) 'Contamination of perfluorinated compounds (PFCs) in Chao Phraya River and Bangpakong River, Thailand', *Water Science and Technology*, 60(4), pp. 975–982.

Kunacheva, C., Fujii, S., Tanaka, S., Boontanon, S. K., Musirat, C., Wongwatthana, T., and Shivakoti, B. R. (2009b) 'Occurrences of perfluorinated compounds in eastern Thailand', *Organohalogen Compounds*, 71, pp. 216–221.

Kunacheva, C., Fujii, S., Boontanon, S. K., Tanaka, S. and Shivakoti, B. R. (2009c) 'Performance of industrial estates' wastewater treatment plant on removal of perfluorinated compounds', proceeding of the third IWA-ASPIRE conference and exhibition, Taiwan, October 2009.

Kunacheva, C., Fujii, S., Tanaka, S., Boontanon, S. K., Poothong, S., Wongwatthana, T. and Shivakoti, B. R. (2010) 'Perfluorinated compounds contamination in tap water and drinking water in Bangkok, Thailand', *Journal of Water Supply: Research and Technology — AQUA*, 59(4), pp. 345–354.

Lien, N. P. H. (2007) 'Study on distribution and behavior of PFOS and PFOA in water environment', PhD thesis, Urban and Environmental Engineering, Kyoto University, Kyoto, Japan.

Lien, N. P. H., Fujii, S., Tanaka, S., Nozoe, M., Tanaka, H. (2008) 'Contamination of perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in surface water of the Yodo River Basin (Japan)', *Desalination* 226, pp. 338–347.

Melin, G. (1999) 'Evaluation of the applicability of synthetic resin sorbents for MTBE removal from water', Center for Groundwater Restoration and Protection, National Water Research Institute, California.

Minnesota Department of Health (2007) *Environmental Health Information — Perfluorochemicals and Health*, Minnesota, USA.

Murakami, M., Imamura, E., Shinohara, H., Kiri, K., Muramatsu, Y., Harada, A., Takada, H. (2008) 'Occurrence and sources of perfluorinated surfactants in rivers in Japan', *Environmental Science Technologies*, 42, 6566–6572.

New Jersey Department of Environmental Protection (2009) 'Division of water supply: guidance for PFOA in drinking water at Pennsgrove Water Supply Company', April 2009. Available online at www.state.nj.us/dep/watersupply/pfoa_dwguidance.pdf

Organization for Economic Cooperation and Development (2002) 'Cooperation on existing chemicals — hazard assessment of perfluorooctane sulfonate and its salts', Environment Directorate joint meeting of the Chemicals Committee and the Working Party on Chemicals, Pesticides and Biotechnology, OECD, Paris, 21 November 2002.

Prevedouros, K., Cousins, T. I., Buck, C. R., and Korzeniowski, H. S. (2006) 'Sources, fate and transport of perfluoro-carboxylates', *Environmental Science Technologies*, 2006, 40(1), pp. 32–34.

Qiang, Y., Ruiqi, Z., Shubo, D., Jun, H., and Gang Y. (2009) 'Sorption of perfluorooctane sulfonate and perfluorooctanoate on activated carbons and resin: kinetic and isotherm study', *Water Research*, 43, pp. 1150–1158.

Qiu, Y. (2007) 'Study on treatment technologies for perfluorochemicals in wastewater', doctoral thesis, Kyoto University.

Renner, R. (2001) 'Growing concern over perfluorinated chemicals', *Environmental Science Technologies*, 35(7), 154A.

Saito, N., Harada, K., Ionue, K., Sasaki, K., Yoshinaga, T., and Koizumi, A. (2004) 'Perfluorooctanoate and perfluorooctane sulfonate concentrations in surface water in Japan', *Journal of Occupational Health*, 46, pp. 49–59.

Schultz, M. M., Higgins, C. P., Huset, C. A., Luthy, R. G., Barofsky, D. F. and Field, J. A. (2006) 'Fluorochemical mass flows in a municipal wastewater treatment facility', *Environmental Science Technologies*, 40(23), pp. 7350–7357.

Senevirathna, S. T. M. L. D., Tanaka, S., Fujii, S., Kunacheva, C., Harada, H., Shivakoti, B. R. and Okamoto, R. (2010a) 'A comparative study of adsorption of perfluorooctane sulfonate (PFOS) on to granular activated carbon, ion-exchange polymers and non-ion-exchange polymers', *Chemosphere*, 80, 647–651.

Senevirathna, S. T. M. L. D., Tanaka, S., Fujii, S., Kunacheva, C., Harada, H., Ariyadasa, B. H. A. K. T. and Shivakoti, B. R. (2010b) 'Adsorption of perfluorooctane sulfonate (n-PFOS) on to ion-exchange polymers and granular activated carbon: batch and column test', *Desalination*, 260, pp. 29–33.

Sinclair, E., Taniyasu, S., Yamashita, N., and Kannan, K. (2004) 'Perfluoroctanoic acid and perfluorooctanesulfonate in Michigan and New York waters', *Organohalogen Compounds,* 66, pp. 4069–4073.

Sinclair, E. and Kannan, K. (2006) 'Mass loading and fate of perfluoroalkyl surfactants in wastewater treatment plants', *Environmental Contamination and Toxicology,* 40(5), pp. 1408–1414.

Skutlarek, D., Exner, M., and Färber, H. (2006) 'Perfluorinated surfactants in surface and drinking waters', *Environmental Science Pollution and Research,* 13, pp. 299–307.

Takagi, S., Adachi, F., Miyano, K., Koizumi, Y., Tanaka, H., Mimura, M., Watanabe, I., Tanabe, S., Kannan, K. (2008) 'Perfluorooctanesulfonate and perfluorooctanoate in raw and treated tap water from Osaka, Japan', *Chemosphere,* 72, pp. 1409–1412.

Tanaka, S., Fujii, S., Lien, N. P. H., Nozoe, M., Kunacheva, C., Kimura, K., and Binaya, S. (2008) 'Contamination of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) in water environment of 21 cities in 10 countries', *Journal of Japan Society on Water Environment,* 31(11), pp. 665–670

Taniyasu, S., Kannan, K., Horii, Y., Hanari, N., and Yamashita, N. (2003) 'A survey of perfluorooctane sulfonate and related perfluorinated organic compounds in water, fish, birds and humans from Japan', *Environmental Science Technologies,* 37 (12), 2634–2639.

Tang, C. Y., Fu, Q. S., Robertson, A. P., Criddle, C. S., Leckie, J. O. (2006) 'Use of reverse osmosis membranes to remove perfluorooctane sulfonate (PFOS) from semiconductor wastewater', *Environmental Science Technologies,* 40, pp. 7342–7349.

United States Environmental Protection Agency (2006) 'Persistent organic pollutants', available online at www.epa.gov/oppfead1/international/pops.htm (accessed January 2010).

Valeria, O. and Reyes, S. (2008) 'Removal of perfluorinated surfactants by sorption on to granular activated carbon, zeolite and sludge', *Chemosphere,* 72, pp. 1588–1593.

Yu, J., Hu, J., Tanaka, S. and Fujii, S. (2009) 'Perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in sewage treatment plants', *Water Research,* 40, pp. 2399–2408.

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