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Technical Note

Biodegradation of perfluorooctanesulfonate (PFOS) as an emerging contaminant

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HIGHLIGHTS

- We have identified the aerobic microorganism for the decomposition of PFOS.
- Ca. 67% of a 600 mg L^{-1} conc. of PFOS was biodegraded by *Pseudomonas aeruginosa* after 12 h.
- Our result is considered a major advancement in sustainable PFOS treatment.

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ABSTRACT

Perfluorooctanesulfonate (PFOS) is a compound of global concern because of its persistence and bioaccumulation in the environment. Nevertheless, little is known of the potential for PFOS biodegradation, even though the importance of characterizing the function and activity of microbial populations detected in the environment has been discussed. This study focused on the biodegradation of PFOS by a specific microorganism. Through this study, we have identified the aerobic microorganism for the specific decomposition of PFOS from wastewater treatment sludge, as a well-known sink for environmental PFOS. This species was *Pseudomonas aeruginosa* strain HJ4 with a 99% similarity, a mesophilic rod type bacteria (30–37 °C). A pH range of 7–9 was determined to be optimal for the growth of strain HJ4. In this study approximately 67% over a range of concentrations (1400–1800 μg L^{-1}) for PFOS was biologically decomposed by *P. aeruginosa* after 48 h incubation. This result is reported here for the first time, which strongly pertains to the efficient biodegradation of PFOS. Therefore, our study is considered a major advancement in sustainable PFOS treatment.

1. Introduction

The ubiquitous presence of anthropogenic perfluorinated compounds (PFCs) in the environment has been recognized as an important global environmental issue (Key et al., 1997; Liou et al., 2010; Clarke and Smith, 2011; Llorca et al., 2012; Naile et al., 2013; Zareitalabad et al., 2013). Amongst PFCs, perfluorooctanesulfonate (PFOS), having approximately 41 yr half-life in water (US EPA, 2012), has ignited widespread interest as a stable yet toxic product arising from the biodegradation, photooxidation, photolysis, and hydrolysis of PFCs (Boudreau et al., 2003; US EPA, 2012; Jacquet et al., 2012; Naile et al., 2013; Benskin et al., 2013), as well as being used directly in surfactants, insecticides, and protective coatings for paper, textile, carpets, and clothing (Renner, 2008). In particular, PFOS has been characterized as having either a potential or confirmed adverse impact on both human health, i.e., cancer risks, and the environment, i.e., bioaccumulation and/or biomagnifications within wildlife (Key et al., 1997; Giesy and Kannan, 2001; OECD, 2002; Boudreau et al., 2003; Clarke and Smith, 2011; US EPA, 2012). For these reasons, PFOS was listed as a persistent organic pollutant in Annex B of the Stockholm Convention in 2009 (Wang et al., 2009). PFCs have been produced since the 1950s (US EPA, 2012), yet there still is insufficient knowledge on the degradation of PFOS, because they have relatively recently been recognized as pollutants of importance (Howard and Muir, 2010). Thus, in recent years the development of sustainable

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treatment methods for PFOS degradation has become an important challenge.

Several process technologies for PFOS treatment in water have been reported. Previous studies have shown that most conventional degradation processes are ineffective for the degradation of PFOS, because this compound including high energy carbon–fluorine in PFOS molecule is inherently recalcitrant to chemical and biodegradation treatments (Key et al., 1998; Boudreau et al., 2003; Cheng et al., 2008; Vecitis et al., 2008; Takagi et al., 2011; Yu et al., 2009; Jacquet et al., 2012; Benskin et al., 2013). For example, advanced oxidation processes (AOPs), which commonly utilize ·OH such as direct photolysis in the presence of either H_2O_2 and Fe_2O_3, UV–ozonation, peroxone (O_3–H_2O_2), and Fenton's process, have been shown to be ineffective for PFOS degradation (3M, 2003; Vecitis et al., 2008). In a recent study, it has been reported that PFOS was decomposed by 68% after 10 d by direct photolysis at 254 nm light (Yamamoto et al., 2007). In contrast, the sonolytic process as an AOP could be effective in treating PFOS in aqueous solution by means of *in situ* pyrolytic reactions in the vapor and interfacial regions (Cheng et al., 2008; Vecitis et al., 2008). Thermal processes at high temperatures (above 300 °C) can be very effective method for decomposition of PFOS (Wang et al., 2013). Furthermore, PFOS could be separated from aqueous solutions by fresh activated carbon and reverse osmosis units (US EPA, 2012). However, the processes mentioned above do not represent a sustainable treatment method for PFOS decomposition, because of the large energy consumption as well as the secondary contamination, such as that as arising from the need for disposal of used activated carbon. As a result, these physico-chemical treatment processes can be inadequate and limited for the degradation of ubiquitous PFOS.

Until recently, PFOS has been perceived as being biologically inert (Key et al., 1997, 1998; OECD, 2002; Liou et al., 2010). Hence, there is a general understanding that PFOS is more difficult to biologically remediation, however, it is not biologically inactive by means of the effect of uncoupling in the presence of various monoamines and related compounds (Key et al., 1997). As a result, few reports on the biodegradation of PFOS as a highly sustainable treatment process are available (Key et al., 1998; Yu et al., 2009; Takagi et al., 2011). Therefore, further research is needed to evaluate the biodegradation of PFOS as a suitable alternative tool.

The objective of the present work is to assess whether PFOS is degradable by aerobic microbes arising from activated sludge and soil, which is part of an ongoing program of investigation of enhanced biodegradation of PFOS. In this study, we have identified the strain which is capable of degrading PFOS. Therefore, these data are important for assessing the fate of PFOS in the environment.

2. Materials and methods

2.1. Chemicals and media

PFOS (40% in H_2O, St. Louis, MO) was purchased from Aldrich. Isotopically labeled internal standard of $^{13}C_4$-PFOS and other PFCs, i.e., perfluorobutane sulfonate (PFBS) and perfluorohexane sulfonate (PFHxS) as products of PFOS biodegradation, were purchased from Wellington Laboratories (Guelph, ON, Canada). Other chemicals used in this study were ACS grade reagents and were obtained from Sigma–Aldrich. All solutions were made with high-purity water from a Millipore ultra-purification system (>18 MΩ cm).

Strains for the degradation of PFOS were routinely cultivated in two defined media, C medium and Luria Bertani (LB) medium, respectively. In order to cultivate and grow the strain, C medium as a liquid phase contained (g L^{-1}) $(NH_4)_2SO_4$, 5.0; KH_2PO_4, 1.0; K_2HPO_4, 2.0; $MgSO_4$ $7H_2O$, 0.2; NaCl, 2.0; $CaCl_2{\cdot}2H_2O$, 0.01; $FeSO_4{\cdot}7H_2O$, 0.01; and 2.0 mL trace elements solution. Trace elements solution contained (g L^{-1}) MoO_3, 0.001; $ZnSO_4{\cdot}7H_2O$, 0.007; $CuSO_4{\cdot}5H_2O$, 5.0×10^{-4}; H_3BO_3, 0.001; $MnSO_4{\cdot}5H_2O$, 0.001; $CoCl_2{\cdot}6H_2O$, 0.001; $NiSO_4{\cdot}7H_2O$, 0.001. The final pH of C medium was approximately 7.0.

In order to observe the form of the strain for the degradation of PFOS, LB medium as a solid phase contained (g L^{-1}) tryptone, 10; yeast extract, 5; NaCl, 10; agar, 15. The final pH of LB medium was approximately 7.0.

2.2. Collection of sludge and soil as inoculum sources

In this study, activated sludge, as a mixture of wastewater and sewage sludge, was collected from the complete mixing aeration tank of 15 municipal wastewater treatment plants in Gwangju and Seoul, Korea. Aerobic microbes for the biodegradation of PFOS can be found in treated sludge, because wastewater treatment sludge is a sink for environmental PFCs (Yu et al., 2009; Takagi et al., 2011) as well as a host of other synthetic chemicals. In addition, soil, as a potential inoculum source, was obtained from the surroundings of the wastewater treatment plants. Sludge and soil were sampled in 2 L polypropylene bottles using a metal spatula, and were kept on ice in the field and refrigerated at 4 °C in the laboratory prior to use.

2.3. Isolation of strain

The strain of aerobic microbes was isolated from samples of sludge and soil as mentioned above. In this study, soil slurry was diluted with aliquot of the autoclaved water. The strain for the biodegradation of PFOS was then grown and maintained by adding respective samples into C medium containing 600 mg L^{-1} PFOS as a substrate. These experiments were performed at a temperature of 30 ± 2 °C on an orbital shaker at 120 rpm. Growth of the strain was confirmed by the naked eye after a few days. This strain was inoculated with 1% (v/v) in the same medium again and was then subcultured more than 10 times in the same manner. The strain isolated from the experiment using C medium was inoculated by streaking on LB medium using a pure separation technique, and was then incubated at 37 °C for 1 d. Next, cultured dominant single colonies were separated and repeatedly subcultured several times in LB medium in the same manner. For subsequent experiments, 650 μL of the strain was well mixed with 350 μL of 80% glycerol solution, autoclaved and was then stored in a tube (1.7 mL, Microtubes MCT-175-C, Union City, CA) which stored kept at −70 °C.

2.4. Identification of strain

API 20E kit (API 20E Enterobacteriaceae, BioMerieux, France) was used to characterize a Gram-negative rod that is motile, catalase positive, and oxidase positive. Then, in order to evidently confirm the isolated strain identification, sequencing of the PCR amplified 16S rRNA gene was performed at a commercial laboratory (SolGent in Daejeon, Korea). In this analysis, the PCR reaction mixture consisted of 2 μL dNTP, 2 μL phosphate buffer with magnesium sulfate, 0.2 μL of 5′, 3′ 5 μM primer, 10 μL DNA template, and 5 μL rTaq. Finally, sequences were compared to the National Center for Biotechnology Information database using the Basic Local Alignment Search Tool (BLAST) to determine phylogenetic similarities.

2.5. Microbial activity test

After about 24 h of incubation in LB medium, the motility of separate microorganisms was investigated using an optical microscope (Olympus CX40, Japan). In addition, the microbial form was observed using Gram staining.

2.6. Growth conditions using isolated strain

To evaluate the growth of isolated strains under various conditions of pH, 100 mL C medium was poured into a Erlenmeyer flask (300 mL) and the strain was inoculated at a concentration of 1% (by volume) for cultures at each initial pH 3–10, as adjusted by solutions of 1 N NaOH and 2 N HCl. Growth controls consisted of inoculated medium without PFOS. Abiotic controls consisted of medium uninoculated with PFOS. These media were incubated aerobically at 30 °C and were then shaken on a rotary shaker at 120 rpm. The growth of the cultured strain was determined after 1 h at 660 nm wavelength using a UV/Vis spectrophotometer (Shimadzu UV 1601, Kyoto, Japan).

To evaluate the growth of the isolated strain under varying temperature conditions, 100 mL C medium was poured into a Erlenmeyer flask (300 mL) and the strain was inoculated at a concentration of 1% (by volume) for cultures at each defined temperature (25–50 °C). In this experiment, abiotic controls consisted of uninoculated medium with PFOS. Growth of cultured strain was determined spectrophotometrically every 1 h at 660 nm wavelength, as mentioned above.

2.7. Analytical methods

All samples were stored in PP bottles and were extracted using solid-phase extraction (SPE) disk cartridges (Waters Oasis HLB, Waters, Milford, MA) using previously described methods (Benskin et al., 2013). 100 μL of 50 ng mL^{-1} isotopically labeled internal standard of $^{13}C_4$-PFOS was added to the sample tube. SPE cartridges were pretreated by flushing the cartridges with 6 mL of HPLC-grade methanol followed by a rinse with 6 mL of deionized water. The pretreated SPE cartridge was topped with a SPE cartridge tube adaptor (Supelco, USA) fitted with a PP tubing connector. The water from the cartridge was then completely removed through the pretreated SPE cartridge using a vacuum pump (GAST Vacuum Pump, MFG) at a flow rate of approximately 1 drop s^{-1}.

Samples were extracted by eluting 4 mL of HPLC-grade methanol twice (a total of 8 mL) using the cartridge adaptor. The eluent was collected in a 30 mL PP bottle. This extract was reduced under a steady flow of ultrapure nitrogen (99.999%, Dae-Chang, Korea) to 1 mL and was then vigorously vortexed for 30 s. After filtering by using a 0.22 μm PP syringe filter (13 mm, HSW Norm-Ject model, Whatman), this extract was transferred to an autosampler vial, capped, and analyzed. The remaining extract was capped and placed into a cooler at 4 °C for storage for further instrumental analysis.

Analysis of extracts including PFOS was accomplished by liquid chromatography tandem mass spectrometry (LCMS-2010 model, Shimadzu, Japan). 1 μL extracts were injected onto an Eclipse XDB-C18 analytical column (3.5 μm, 150 × 2.1 mm) which was maintained at 40 °C. The mobile phase consisted of 30% ammonium acetate (10 mM) in water and 70% acetonitrile at a flow rate of 0.2 mL min^{-1}. The detector voltage was set at 1.5 kV; nebulizing gas flow, 1.5 L min^{-1}; curved desolvation line temperature, 250 °C. All analyses were performed with negative ESI mode and multiple reaction monitoring method on selected precursor and product ions.

The calibration was established with the internal standardization method. The limits of detection for PFOS, PFBS and PFHxS were determined as 4.3, 1.9, and 3.9 ng L^{-1}, respectively, based on the signal-to-noise three ($S/N = 3$). The recovery of the SPE extraction method from the pure water of 0.5 L ($n = 3$) was assessed with each spiked concentration (200 ng L^{-1}) of PFOS, PFBS and PFHxS. The recoveries of PFOS, PFBS and PFHxS were 100%, 62%, and 104%, respectively.

3. Results and discussion

3.1. Strain identification

To understand PFOS biodegradation processes, its phylogenetic identification is considered to be the primary step, which can be followed by isolation and laboratory pure-culture studies of the important microorganism as a predominant population. An important question that should be addressed in the pure-culture studies is the identification of microbial species which have become the important players in the biodegradation of PFOS. Hence, we monitored the populations of many wastewater facilities to examine their biodegradability of PFOS. Among several species, one potential strain that best adapts to the PFOS biodegradation was isolated from C medium through sub-culturing [Supplementary Material (SM), Fig. SM-1]. Preliminary studies were conducted using different media to ascertain the best suited microbial candidate for PFOS degradation, choosing C medium (data not shown). The strain cultured in our study was nominated arbitrarily with strain HJ4. Then, the growth of strain HJ4 was observed from C medium inoculated with PFOS. Strain HJ4 was a Gram negative rod type bacterium (Fig. SM-2). Approximately 700 bp of the large subunit 16S rDNA were amplified by PCR using primers targeting conserved 27F (5 μM) (5′-AGAGTTTGATCCTGGCTC) and 1492R (5 μM) (5′-ACGGCTACCTTGTTACGA) (data not shown). Results from BLAST searches in the GenBank database confirmed that sequences of isolated strains were homologous to *Pseudomonas aeruginosa*, with 99% similarity. The sequences obtained were deposited to GenBank with the accession numbers of HJ4. This sequence of isolated strains as bacteria is provided in Supplementary information (Table SM-1). Some researchers have demonstrated that PFCs can be biologically degraded by the identified D2 strain of *Pseudomonas fluorescens* (Key et al., 1998), but a bacteria species for PFOS biodegradation has not been reported up to now.

3.2. pH condition on strain HJ4

To gain an insight into the activity of the identified strain HJ4, as well as to determine optimal growth conditions, it was subjected to varying pH conditions. The optimal growth conditions of the isolated strain were investigated in the pH range of 3–10 and at a constant temperature of 35 °C in C medium. A series of experiments were carried out in C medium inoculated with PFOS.

The growth of strain HJ4 under varying pH values is shown in Fig. 1a. As shown in Fig. 1a, the growth of strain HJ4 at the initial stage was relatively slow, especially in the pH range of 7–9, but after 5 h, its growth steeply increased. Then, after 12 h, the growth of strain HJ4 reached a plateau. In contrast, as shown in Fig. 1a, the active growth of strain HJ4 was not observed within the pH range of 3–5, or at pH 10. This result indicates that a pH range of 7–9 is necessary for the growth of strain HJ4. Considering this result, all of the following experiments were performed at pH 7.

3.3. Temperature condition on strain HJ4

The optimal growth conditions of the isolated strain was investigated within the temperature range of 25–50 °C and at pH 7 in C medium inoculated with PFOS. The growth of strain HJ4 under

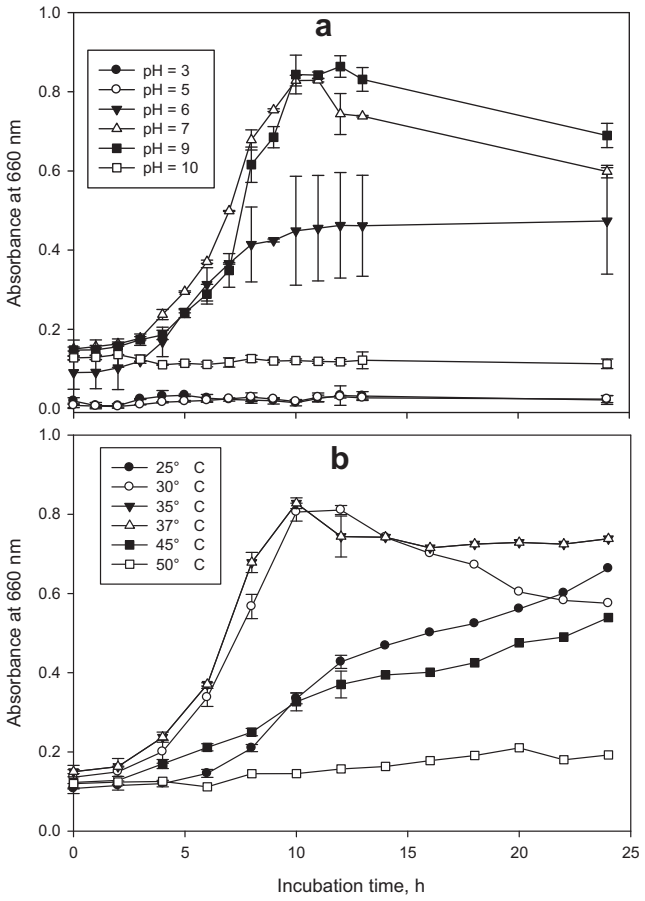

Fig. 1. (a) pH effect on the growth of isolated strain HJ4 by change in absorbance. (b) Temperature effect on the growth of isolated strain HJ4 by change in absorbance. *Note that a detailed explanation of the experimental conditions is mentioned in the text.

varying temperature values is shown in Fig. 1b. As shown in Fig. 1b, the growth of strain HJ4 at the initial stage was relatively very slow from 0 to 4 h incubation time, but after 4 h, its growth steeply increased when incubated at constant temperatures of 35–37 °C. Then, after approximately 12 h, the growth of strain HJ4 reached a plateau. In contrast, as shown in Fig. 1b, the growth of strain HJ4 was very slow and minimal above 45 °C over 24 h. Furthermore, the growth of strain HJ4 at a temperature range of approximately 30–35 °C increased gradually with increasing incubation time. This result indicates that a temperature range of 30–37 °C, i.e., 35–37 °C, is optimal for the growth of strain HJ4, as a mesophilic microorganism. Thus, all of the following experiments were performed at 35 °C.

3.4. Biodegradation of PFOS by strain HJ4

Strain HJ4 was grown with PFOS as a substrate. In this study, 0.1% (volume) glucose was typically added into aqueous PFOS and strain HJ4 solution. The addition of glucose has previously been shown to accelerate the growth of strain HJ4 for PFOS biodegradation and while its use was not essential, it supported the growth of strain HJ4. Based on this result, all experiments were performed at pH 7.0 and a temperature 35 °C in the presence of 0.1% glucose.

As shown in Fig. 2, PFOS over a range of concentrations (1400–1800 μg L^{-1}) was gradually degraded by strain HJ4. PFOS concentration consistently decreased with the vigorous growth of strain

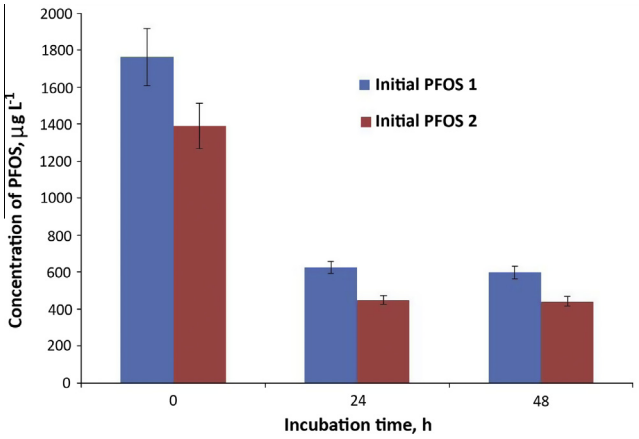

Fig. 2. PFOS biodegradation by strain HJ4 and its growth according to incubation time. *Note that a detailed explanation of the experimental conditions is mentioned in the text.

HJ4 over 48 h. At 48 h, PFOS was decomposed by approximately 67%, and thereafter its residual concentration was found to be constant. At the same time, the growth of strain HJ4 was observed to be constant (data not shown). Thus, it indicates that the biodegradation of PFOS as a highly sustainable treatment process can be very effective.

From this data, as mentioned above, further research needs to evaluate the biodegradation of PFOS as a suitable alternative tool. Hence, fluoride ions, which can be generated from the biodegradation of PFOS, were investigated by using ionic chromatography, as proposed by similar studies (Key et al., 1998; Liou et al., 2010). However, no defluorination was observed, even through several attempts at measurement. In addition, no change in fluoride level was observed in abiotic controls. Despite our extensive search for microbial PFOS biodegradation, no evidence on defluorination of PFOS biodegradation has been uncovered. However, in this study, PFBS and PFHxS were consistently detected as minor products. The concentrations of these PFCs were in the range of 4.0–26.0 ng L^{-1}. Furthermore, tandem MS analysis may show the formation possibility of several unknown products (Fig. SM-3). From this result, an alternative explanation would be the partial cleavage of C–C bonds in the PFOS molecule, instead of C–F bonds, having the rigidity conferred by the fluorine substituent (Key et al., 1998). As a result, further investigation of the mechanism of biodegradation of PFOS will require an understanding of the specific characteristics, i.e., the unknown enzyme(s) and cofactor(s) through crude cell free extracts of strain HJ4, involved in the transformation of PFOS. This experiment is now on-going.

4. Conclusion

This study showed the biodegradation of PFOS by a specific microorganism. We isolated the aerobic microorganism, strain HJ4, for the biodegradation of PFOS. This species had 99% similarity to *P. aeruginosa* strain HJ4, which is a gram negative and mesophilic bacteria (30–37 °C). Under optimal conditions, the concentration of PFOS was biologically decomposed by approximately 67% at incubation time 48 h. However, the formation of fluoride ion from PFOS biodegradation was not observed. Nonetheless, this study could contribute to the biodegradation of PFOS.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.chemosphere.2014.01.072.

References

3M, 2003. Environmental and Health Assessment of Perfluorooctane Sulfonic Acid and Its Salts, prepared by 3M, US EPA docket AR 226-1486.

Benskin, J.P., Ikonomou, M.G., Gobas, F.A.P.C., Begley, T.H., Woudneh, M.B., Cosgrove, J.R., 2013. Biodegradation of N-ethyl perfluorooctane sulfonamido ethanol (EtFOSE) and EtFOSE-based phosphate diester (SAmPAP diester) in marine sediments. Environ. Sci. Technol. 47, 1381–1389.

Boudreau, T.M., Wilson, C.J., Cheong, W.J., Sibley, P.K., Mabury, S.A., Muir, D.C.G., Solomon, K.R., 2003. Response of the zooplankton community and environmental fate of perfluorooctane sulfonic acid (PFOS) in aquatic microcosms. Environ. Toxicol. Chem. 22, 2739–2745.

Cheng, J., Vecitis, C.D., Park, H., Mader, B.T., Hoffmann, M.R., 2008. Sonochemical degradation of perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) in landfill groundwater: environmental matrix effects. Environ. Sci. Technol. 42, 8057–8063.

Clarke, B.O., Smith, S.R., 2011. Review of 'emerging' organic contaminants in biosolids and assessment of international research priorities for the agricultural use of biosolids. Environ. Int. 37, 226–247.

Giesy, J.P., Kannan, K., 2001. Global distribution of perfluorooctane sulfonate in wildlife. Environ. Sci. Technol. 35, 1339–1342.

Howard, P.H., Muir, D.C.G., 2010. Identifying new persistent and bioaccumulative organics among chemicals in commerce. Environ. Sci. Technol. 44, 2277–2285.

Jacquet, N., Maire, M.A., Landkocz, Y., Vasseur, P., 2012. Carcinogenic potency of perfluorooctane sulfonate (PFOS) on Syrian hamster embryo (SHE) cells. Arch. Toxicol. 86, 305–314.

Key, B.D., Howell, R.D., Criddle, C.S., 1997. Fluorinated organics in the biosphere. Environ. Sci. Technol. 31, 2445–2454.

Key, B.D., Howell, R.D., Criddle, C.S., 1998. Defluorination of organofluorine sulfur compounds by *Pseudomonas* sp. strain D2. Environ. Sci. Technol. 32, 2283–2287.

Liou, J.S.-C., Szostek, B., DeRito, C.M., Madsen, E.L., 2010. Investigating the biodegradability of perfluorooctanoic acid. Chemosphere 80, 176–183.

Llorca, M., Farré, M., Tavano, M.S., Alonso, B., Koremblit, G., Barceló, D., 2012. Fate of a broad spectrum of perfluorinated compounds in soils and biota from Tierra del Fuego and antarctica. Environ. Pollut. 163, 158–166.

Naile, J.E., Khim, J.S., Hong, S., Park, J., Kwon, B.-O., Ryu, J.S., Hwang, J.H., Jones, P.D., Giesy, J.P., 2013. Distributions and bioconcentration characteristics of perfluorinated compounds in environmental samples and collected from the west coast of Korea. Chemosphere 90, 387–394.

OECD, 2002. Co-operation on Existing Chemicals – Hazard Assessment of Perfluorooctane sulfonate and Its salts. Environment Directorate Joint Meeting of the Chemicals Committe and the Working Party on Chemicals, Pesticides and Biotechnology, Organisation for Economic Co-operation and Development, Paris, 21 November 2002.

Renner, R., 2008. PFOS phaseout pays off. Environ. Sci. Technol. 42, 4618.

Takagi, S., Adachi, F., Miyano, K., Koizumi, Y., Tanaka, H., Watanabe, I., Tanabe, S., Kannan, K., 2011. Fate of perfluorooctanesulfonate and perfluorooctanoate in drinking water treatment processes. Water Res. 45, 3925–3932.

US EPA, 2012. Emerging Contaminants – Perfluorooctane Sulfonate (PFOS) and Perfluorooctanoic Acid (PFOA). Emerginge Contaminants Fact Sheet – PFOS and PFOA, EPA 505-F-11-002.

Vecitis, C.D., Park, H., Cheng, J., Mader, B.T., Hoffmann, M.R., 2008. Kinetics and mechanism of the sonolytic conversion of the aqueous perfluorinated surfactants, perfluorooctanoate (PFOA), and perfluorooctane sulfonate (PFOS) into inorganic products. J. Phys. Chem. A 112, 4261–4270.

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.

Wang, F., Shih, K., Lu, X., Liu, C., 2013. Mineralization behavior of fluorine in perfluorooctanesulfonate (PFOS) during thermal treatment of lime-conditioned sludge. Environ. Sci. Technol. 47, 2621–2627.

Yamamoto, T., Noma, Y., Sakai, S.-I., Shibata, Y., 2007. Photodegradation of perfluorooctane sulfonate by UV irradiation in water and alkaline 2-propanol. Environ. Sci. Technol. 41, 5660–5665.

Yu, J., Hu, J., Tanaka, S., Fujii, S., 2009. Perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) in sewage treatment plants. Water Res. 43, 2399–2408.

Zareitalabad, P., Siemens, J., Hamer, M., Amelung, W., 2013. Perfluorooctanoic acid (PFOA) and perfluorooctanesulfonic acid (PFOS) in surface waters, sediments, soils and wastewater – a review on concentrations and distribution coefficients. Chemosphere 91, 725–732.