



# Spatial and temporal trends in perfluorooctanoic and perfluorohexanoic acid in well, surface, and tap water around a fluoropolymer plant in Osaka, Japan



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## HIGHLIGHTS

- PFOA around a fluoropolymer plant decreased greatly.
- Concentrations of PFHxA appear to have increased in well and surface water.
- Widespread groundwater contamination by PFOA was found around the plant.
- PFOA levels in tap water were lower than the EPA Lifetime Health Advisory value.

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## ABSTRACT

This study was conducted to clarify the spatial distributions of perfluorooctanoic acid (PFOA) and perfluorohexanoic acid (PFHxA) in well, surface and tap water around a fluoropolymer plant in Osaka between 2003 and 2016 and to predict the fate of those chemicals in these aquatic environments. We analyzed 44 well, six surface and six tap water samples collected within a 5 km radius of the plant. The PFOA concentrations in well water ranged from 45.2 to 7440 ng/L (median = 240 ng/L), while PFHxA concentrations ranged from 9.68 to 970 (median = 45.4 ng/L) in 2015–2016. The concentration of other perfluoroalkyl carboxylic acids were lower than PFOA and PFHxA in well water. Fixed-point observation showed that the levels of PFOA decreased greatly over the last few decades, whereas those of PFHxA increased in both well and surface water. Further monitoring and investigation are suggested to understand PFOA and PFHxA contamination and fate in the environment, as well as their potential for human exposure in this region.

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## 1. Introduction

Perfluorooctanoic acid (PFOA) has been widely applied for industrial and household purposes in surfactants and lubricants, and as an additive for fluoropolymer (FP) production. In recent years, PFOA has attracted growing attention because of its environmental persistence and bioaccumulative properties with potential adverse

health impacts (Abdellatif et al., 1991; Nilsson et al., 1991; Fei et al., 2009; Washino et al., 2009). In humans, PFOA has an average elimination half-life of about 3 years because of its high intestinal absorption rate and poor elimination from the kidneys (Harada et al., 2005; Kennedy et al., 2004; Steenland et al., 2010). As a result, PFOA has the potential to bioaccumulate, which has led to health concerns regarding its toxic and carcinogenic effects (Barry et al., 2013; Vieira et al., 2013). Recently, the eight major FP manufacturers have made voluntary efforts to eliminate the emission and production of PFOA (USEPA, 2014).

Our previous studies identified high levels of PFOA contamination in the Osaka area in the early 2000s. Moreover, the levels in

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### Abbreviations

|         |                                      |
|---------|--------------------------------------|
| PFOA    | perfluorooctanoic acid               |
| PFHxA   | perfluorohexanoic acid               |
| PFBA    | perfluorobutanoic acid               |
| PFPeA   | perfluoropentanoic acid              |
| PFHpA   | perfluoroheptanoic acid              |
| PFNA    | perfluorononanoic acid               |
| PFDA    | perfluorodecanoic acid               |
| PFUnDA  | perfluoroundecanoic acid             |
| PFDoDA  | perfluorododecanoic acid             |
| PFTriDA | perfluorotridecanoic acid            |
| PFTeDA  | perfluorotetradecanoic acid          |
| FP      | fluoropolymer                        |
| STPs    | sewage treatment plants              |
| GM      | geometric mean                       |
| PFCAs   | perfluorinated carboxylic acid       |
| GC-MS   | gas chromatography-mass spectrometry |
| MDL     | method detection limits              |
| SE      | standard error                       |
| HA      | health advisory                      |
| CI      | confidential interval                |

wastewater and ecological samples strongly suggested that FP plants in Settsu, Osaka Prefecture, were a source of PFOA (Saito et al., 2004; Morikawa et al., 2006). Daikin Industries, which is one of the eight major FP manufacturers and operates the only major FP plant in Settsu, announced that they voluntarily reduced PFOA emissions by more than 99% between 2000 and 2012 (Daikin, 2012). Furthermore, they set a target for complete cessation of PFOA emissions by 2015, which they achieved by replacing it with alternative substances such as perfluorohexanoic acid (PFHxA) (Daikin, 2016). The structure of PFHxA is similar to that of PFOA; however, it is known to have a shorter half-life than other long-chain perfluorinated carboxylic acids (PFCAs) (Fujii et al., 2015; Nilsson et al., 2010; Russell et al., 2013). However, like other PFCAs, PFHxA is difficult to degrade in the environment because its strong carbon-fluorine bonds (C-F) are widely distributed in the environment.

We previously demonstrated the remarkable reduction of PFOA emissions by approximately 85% from 2003 to 2008 around the FP plant in Settsu (Niisoe et al., 2010). Similar reductions were also confirmed in human serum samples collected in Settsu, for which geometric mean (GM) PFOA concentrations of 29.54 in 2004 to 13.46 ng/mL in 2008 were observed (Harada et al., 2011). Despite these reductions, the levels of PFOA remaining in the environment and emissions of its alternative substance, PFHxA, have not been thoroughly investigated in this area. Previous studies based on water samples collected from sewage treatment plants (STPs) in 2003 showed that wastewater discharged from the FP plant in Settsu was released into surface water (6.6 t/yr), air (2.3 t/yr) and soil (1.2 t/yr) (Niisoe et al., 2010). In many cases, surface water is a source of groundwater recharge; thus, pollution of surface water can cause deterioration of groundwater quality (Winter et al., 1998). Our preliminary test based on samples collected from wells located near the FP plant in 2006 suggested that the concentrations of PFOA in well water samples were high (57,000 ng/L and 8300 ng/L). Tap water, which is often used as drinking water in Japan, is also useful for observing PFOA contamination of municipal water supplies. Therefore, there are advantages for assessing well, river, and tap water to define the spatial and temporal occurrence of PFCAs in this area. In addition, it is

essential to assess the safety of water sources to prevent potential human health risks associated with drinking or using contaminated water and to provide evidence for decision making to decrease human, ecological and environmental risks. However, few studies have evaluated spatial and temporal trends in perfluorinated organic compounds near FP production plants. Therefore, this study was conducted to clarify the spatial distributions of PFOA and PFHxA in well, surface and tap water and predict the fate of those chemicals in water around an FP plant in Osaka.

## 2. Materials and methods

### 2.1. Study area

The authors identified potential affected sites within a 5-km radius from the FP plant in Settsu, including parts of Suita, Moriguchi, Kadoma, Neyagawa, and Ibaraki and all of Yodogawa Ward and Higashiyodogawa Ward in Osaka.

### 2.2. Sample collection

Samples were collected from well water, river water, and tap water between June 18, 2015 and April 6, 2016 (Fig. 1). Well water and river water samples were drawn from the surface using a tinsplate bucket. All samples consisted of 2 L of water collected into polyethylene terephthalate bottles. All instruments were thoroughly pretreated with distilled water and methanol. Samples were delivered to the laboratory immediately after collection, then stored at 4 °C for up to 1 wk until analysis.

To collect private well water, letters were sent out to assigned owners and persons responsible for disaster cooperation wells and religious wells of temples and shrines within the study areas. We identified and employed a list of disaster cooperation wells and religious institutions in Osaka Prefecture using government websites (<http://www.pref.osaka.lg.jp/kankyoeisei/saigaijikyoryokuido/> and <http://www.pref.osaka.lg.jp/shigaku/shukyo/>) and Google maps. Overall, 256 letters were sent to all of the subjects, along with stamped return envelopes, informed consent statements, and questionnaires. We received 96 responses, 20 of which indicated that well owners would cooperate. As a result, we collected 23 well water samples, including two from continually investigated wells (sites 00-1 and 00-2) and 21 samples from wells that had not previously been sampled (01-20; Fig. 1), with two wells at site 17. Some samples were sent directly from the well's owners to the laboratory (n = 8) after collection using pre-washed sampling bottles that we sent beforehand with instructions. To evaluate temporal trends in well water, archived samples collected in 2006, 2009, and 2011 from well 00-1 (Fig. 1), and those collected in 2006 and 2010 from well 00-2 (Fig. 1) were used. These samples had been stored at −20 °C in the Sample Bank of Kyoto University (Koizumi et al., 2009) since they were collected. The questionnaire interviews we conducted with well owners indicated that none of the wells were used for drinking water, but some had been used for household usage, such as watering plants, washing the pet supplies, or recharging private pond water.

Surface water samples were collected from the Ai River (Fig. 1, site 00-3) on June 18, 2015. To observe the temporal changes in surface water, we used six samples collected in 2003, 2008, 2009, 2010, 2011, and 2013 (one sample per year) from the same sampling sites (Fig. 1, site 00-3). These samples have also been stored at −20 °C in the Sample Bank of Kyoto University.

Tap water samples were collected from a public park near the Ai River (Fig. 1, site 00-3) in Osaka and a residence in Sakyo Ward, Kyoto Prefecture, which is located 34 km upstream of the FP plant.

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