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Occurrence and spatial distribution of perfluorinated compounds in groundwater receiving reclaimed water through river bank infiltration

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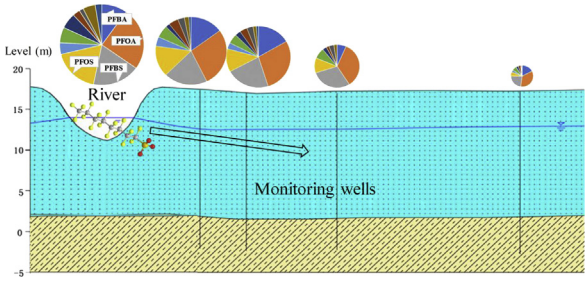
HIGHLIGHTS

- 11 out of 14 selected PFCs were detected in riverside groundwater.
- PFCs in river were likely the source for those in the riverside groundwater.
- PFC concentrations in groundwater decreased with increasing distances from river.
- Riverbank lithology was the main factor affecting the transport of PFCs.
- PFOS attenuated stronger than PFOA, PFBS and PFBA.

GRAPHICAL ABSTRACT



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ABSTRACT

Perfluorinated compounds (PFCs) in groundwater are of widespread concern due to their potential toxicity to human health and ecological systems. PFCs in rivers can infiltrate into groundwater through riverbank infiltration, potentially endangering the safety of drinking water and causing a deterioration in the groundwater environment. This study investigated the occurrence of PFCs in rivers and riverside groundwater from 2014 to 2017 in a city in north China. PFCs were detected in most of the groundwater samples, ranging from not detected to 64.8 ng L^{-1}. The predominant PFCs in both river and groundwater samples were perfluorooctane sulfonate, perfluorooctanoic acid, perfluorobutane sulfonate and perfluorobutanoic acid. The PFC concentrations and major compounds were consistent in both the river and riverside groundwater samples at each site, suggesting that the adjacent river was the source of the PFCs in the riverside groundwater. The spatial distribution of the PFCs in the riverside groundwater was affected by the hydraulic connection between the groundwater and the river, the lithology of the aquifer and the properties of the compounds. The results indicated that PFCs were attenuated during riverbank infiltration and the ability of different riverbank lithologies to remove PFCs was in the order sandy clay > fine sand > sandy gravel. Perfluorooctane sulfonate concentrations decreased sharply with increasing distances from river, whereas perfluorooctanoic acid, perfluorobutane sulfonate and perfluorobutanoic acid could by transported for greater distances in riverside groundwater. This study provides valuable information on PFCs in riverside groundwater affected by riverbank infiltration.

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1. Introduction

Perfluorinated compounds (PFCs) include perfluoroalkyl carboxylic acids, perfluoroalkane sulfonic acids and various precursors. They are widely used in commercial and industrial products (e.g. surfactants, fire-fighting foams, agrochemicals, textiles and cosmetics) due to their extreme thermal and chemical stability and unique amphiphilic properties (Kissa, 2001; OECD, 2002; Ahrens, 2011). However, these unique characteristics also make these substances highly persistent, widespread, bioaccumulative and potentially toxic (Lau et al., 2007; Conder et al., 2008; Lindstrom et al., 2011). PFCs have been detected in various environmental matrices, including air, water, soil, sediment, wildlife and even human blood and urine (Kannan et al., 2004; Plassmann and Berger, 2013; Liu et al., 2015; Xiao, 2017). Perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) are the most commonly detected and pose serious risks to both human health and the environment. The transport and fate of PFCs in the natural environment are of great concern and the production, use and emission of some PFCs are strictly regulated in countries such as Canada, Norway and the USA.

Many rivers in China have accepted or been recharged by reclaimed water as a result of water shortages. However, PFCs cannot be completely removed from wastewater in municipal wastewater reclamation plants and therefore these reclaimed waters could be a source of PFCs in natural waters. PFC concentrations in reclaimed wastewater in California ranged from 90 to 470 ng L^{-1} (Plumlee et al., 2008). PFOS and PFOA concentrations ranged up to 0.7 and 12 ng L^{-1} in the water reclamation plants in Australia (Thompson et al., 2011). The concentrations of PFOA and PFOS in wastewater treatment plant (WWTP) effluents in Shenyang were 18.4–41.1 and 1.69–3.85 ng L^{-1}, respectively (Sun et al., 2011). PFOA was the dominant PFC, with concentrations of 100–145 ng L^{-1} in six WWTPs in Tianjin (Sun et al., 2012). PFCs in surface waters were investigated in a number of major rivers, including the main streams and tributaries in China, and the main compounds detected were PFOS and PFOA (Wang et al., 2015). The total PFC concentration was 3.0–52 ng L^{-1} in the tributaries of the Pearl River in 2012; PFOA, PFBS, PFOS were the three most abundant PFCs (Zhang et al., 2013b). The total concentrations of 18 PFCs in the surface waters of the Yangtze River were 2.20–74.56 ng L^{-1} in 2013, with no obvious seasonal variation (Pan et al., 2014). Nine PFCs were investigated in the Haihe River and had a total concentration of 12–74 ng L^{-1} with perfluorohexanoic acid (PFHxA), PFOA and PFOS the most dominant (Li et al., 2011).

PFCs have also been widely detected in groundwater worldwide. A pan-European survey on polar organic persistent pollutants in groundwater showed that more than half of the groundwater samples were contaminated with PFCs (Loos et al., 2010). PFCs were detected in all groundwater samples, with PFOS concentrations of 0.28–133 ng L^{-1} and PFOA concentrations of 0.47–60 ng L^{-1} in Tokyo, Japan (Murakami et al., 2009). PFCs have now been reported in groundwater in China. The total concentration of 12 PFCs ranged from 2.7 to 9.6 ng L^{-1} in groundwater samples from the Huai River Basin (Zhu et al., 2017) and 17 PFCs in groundwater were found at concentrations of 5.3–615 ng L^{-1} in a rural area of eastern China (Chen et al., 2016b). The total PFC concentrations in groundwater were 0.32–8.3 ng L^{-1} in Tianjin (Qi et al., 2016) and from not detected to 17 ng L^{-1} on the Liaodong Peninsula (Li et al., 2016) in north China. A spatial distribution study around a large fluorochemical industrial park in China showed that the PFC levels in groundwater initially decreased sharply, followed by a gentle decrease with increasing distance from the source (Liu et al., 2016). Information about PFCs, especially their source and transport in groundwater, is still limited due to the limited number of study areas and field types.

Rivers are an important recharge source of groundwater in the alluvial fan area of north China (Dun et al., 2013), especially in the dry season when the groundwater table in the unconfined aquifer is lower than the river water table. Pollutants in rivers may infiltrate into groundwater during the recharge process (Lapworth et al., 2009). However, there is insufficient information on the transport of PFCs from rivers to groundwater through riverbank infiltration to understand the occurrence and spatial distribution of PFCs in groundwater, especially the effects of reclaimed water channel on PFC contamination in groundwater. We therefore selected several representative sites in north China and analyzed 14 PFCs sampled from four riverside cross-sections of rivers that had received reclaimed water and the adjacent groundwater. We compared the occurrence of PFCs in the riverside groundwater with those in the river water and evaluated the impacts of the river water on the distribution of PFCs in the groundwater. We also determined the spatial distribution and transport pattern of PFCs from the river to the groundwater through riverbank infiltration.

2. Materials and methods

2.1. Study area

The study area is located northeast and southeast of a city in north China and the sampling sites and monitoring wells are shown in Fig. 1. Site A is located at CB River, sites B and C at BY River and site D at LS River, which is a tributary of BY River. These rivers receive reclaimed water from different WWTPs, thus were chosen as representatives to investigate the impact of different sources on PFC occurrences in river and groundwater. They flow from northwest to southeast and eventually discharge into the Bohai Sea. The permeabilities of the aquifers at the four sampling sites are significantly different as a result of the different lithologies. This helps us to study the effect of geological condition on the PFC distribution in groundwater. The aquifer at site A is mainly composed of sandy gravel, which has a highest permeability of 200 m d^{-1}. The aquifers at sites B and C are composed of fine sand and have a permeability of about 20 m d^{-1}. The lithology of the aquifer at site D is sandy clay, with a poor permeability of 0.001 m d^{-1} (Fig. 1). The surface water table was higher than the groundwater at the four sampling times from 2014 to 2017, indicating that the groundwater was recharged from surface water.

2.2. Sampling

We collected river water samples and groundwater samples in January 2014, November 2015, October 2016 and May 2017. Information about the river and groundwater samples are listed in Table S1. Different distances groundwater monitoring wells were designed to investigate the migration ability of PFCs in riverside groundwater. The river water samples were collected from the surface (top 1–20 cm) using 1 L polypropylene bottles pre-rinsed with Milli-Q water and HPLC-grade methanol. For groundwater sampling, 1 L groundwater samples were collected in a polypropylene bottle after pumping at least 50 L of water. River and groundwater samples were unavailable at site D in 2015 and groundwater samples were unavailable at site B1 in 2016 due to the management and maintenance of the monitoring wells. Samples were stored in polypropylene bottles at 4 °C and all samples were extracted within one week.

2.3. Chemical reagents

Fourteen PFCs standards [perfluorobutanoic acid (PFBA),