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Contamination risk of raw drinking water caused by PFOA sources along a river reach in south-western Finland

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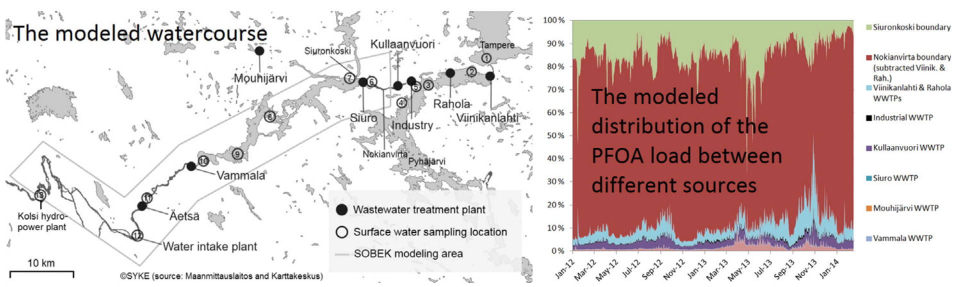
HIGHLIGHTS

- Transport of PFOA was simulated in River Kokemäenjoki in Finland.
- River and wastewater PFOA mass flows were determined.
- Communal wastewater treatment plants caused only 11% of the total PFOA load.
- The concentration of PFOA in raw drinking water remains on a safe level.

GRAPHICAL ABSTRACT



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ABSTRACT

Transport of perfluorooctanoic acid (PFOA) was simulated in the beginning of River Kokemäenjoki in Finland using one-dimensional SOBEK river model. River Kokemäenjoki is used as a raw water source for an artificial groundwater recharge plant, and the raw water intake plant is located near the downstream end of the model application area. Measured surface water and wastewater concentrations were used to determine the PFOA input to the river and to evaluate the simulation results. The maximum computed PFOA concentrations in the river at the location of the raw water intake plant during the simulation period Dec. 1, 2011–Feb. 16, 2014 were 0.92 ng/l and 3.12 ng/l for two alternative modeling scenarios. These concentration values are 2.3% and 7.8%, respectively, of the 40 ng/l guideline threshold value for drinking water. The current annual median and maximum PFOA loads to the river were calculated to be 3.9 kg/year and 10 kg/year respectively. According to the simulation results, the PFOA load would need to rise to a level of 57 kg/year for the 40 ng/l guideline value to be exceeded in river water at the raw water intake plant during a dry season. It is thus unlikely that PFOA concentration in raw water would reach the guideline value without the appearance of new PFOA sources. The communal wastewater treatment plants in the study area caused on average 11% of the total PFOA load. This raises a concern about the origin of the remaining 89% of the PFOA load and the related risk factors.

1. Introduction

Perfluorinated alkyl acids (PFAAs) are a group of emerging pollutants that have been used in industry and consumer products from the 1950s (Lau et al., 2007). Because of their fluorine-carbon chain PFAAs are inert, show resistance to high temperatures and repel oil and water, which makes them an ideal coating material for consumables,

Abbreviations: CWWTP, communal wastewater treatment plant; LOD, limit of detection; LOQ, limit of quantification; PFAA, perfluorinated alkyl acid; PFOA, perfluorooctanoic acid; PFOS, perfluorooctanesulfonic acid; WIP, water intake plant; WWTP, wastewater treatment plant.

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such as clothes and furniture (Kissa, 2001). Other common uses of PFAAs include aviation hydraulic fluids, firefighting foams, paints and metal plating industry (Renner, 2001).

The PFAA compound found the most frequently in European wastewaters is perfluorooctanoic acid (PFOA) (Loos et al., 2013). PFOA is extremely persistent in the environment, since it is not affected by biodegradation (Liou et al., 2010) or photodegradation (Vaalgamaa et al., 2011). It is not significantly removed in wastewater treatment (Schultz et al., 2006) and some studies have even reported higher PFOA concentrations in wastewater effluents than influents, presumably because of biodegradation of its precursors (Becker et al., 2008; Murakami et al., 2009; Schultz et al., 2008). Unlike most other persistent organic pollutants, PFOA is water soluble and found in animals in serum rather than in fat (Post et al., 2012), and its partitioning to sediment is low (Ahrens et al., 2010b, 2011). Because of these properties PFOA can be transported long distances with water. Traces of PFOA have been detected even in Arctic areas far from all emission sources (Butt et al., 2010; Lau et al., 2007). Another long range transport pathway for PFOA is suggested to be the transport of its volatile precursors in the atmosphere.

Zareitalabad et al. (2013) have reviewed a large number of PFOA concentrations in surface waters and wastewater around the world. Out of all the reported surface water concentrations for PFOA half were in the range of 0.8–13 ng/l and the median concentration was 3.1 ng/l. In Finland Perkola (2014) reported PFOA concentrations in five rivers to be lower, 0.08–1.51 ng/l. For wastewater Zareitalabad et al. (2013) reported a median PFOA concentration of 27 ng/l, and Loos et al. (2013) found the maximum, mean and median PFOA concentrations of 90 European wastewater treatment plants (WWTP) to be 15,900 ng/l, 255 ng/l and 12.9 ng/l, respectively. These studies reveal the wide occurrence of PFOA in the environment and the potential role of WWTPs as its point sources.

PFOA has also been detected in drinking water sources and finished tap water (Post et al., 2012), which raises concerns about the safety of drinking water, as exposure to PFOA has been linked to several adverse health effects in humans (Barry et al., 2013; Frisbee et al., 2010; Lam et al., 2014; Melzer et al., 2010; Steenland et al., 2010). Currently the use of PFOA is not regulated by any international agreements, but it has been proposed for restriction at the European Chemicals Agency (2014). Also in 2006 eight major manufacturers of PFOA committed to a voluntary program to reduce PFOA facility emissions and its content in finished products (USEPA, 2006). As reviewed by Zushi et al. (2012), some guideline values have been set for drinking water concentrations: 40 ng/l (NJDEP, 2007), 300 ng/l including both PFOA and PFOS (German Drinking Water Commission, 2006), 300 ng/l (UK Drinking Water Inspectorate, 2009) and 400 ng/l (USEPA, 2009). There is also an Italian guideline value of 500 ng/l (Regione del Veneto, 2014). PFOA is not regulated and no guideline values have been suggested for it in Finland.

PFOA can enter surface water or groundwater from several sources, including industrial air emissions, industrial and domestic wastewater, storm water runoff, land application of biosolids, and release of firefighting foams (Post et al., 2012). Methods that have been used to assess the importance of different PFAA sources include mass balance calculations (Filipovic et al., 2013; Scott et al., 2010; Takazawa et al., 2009), modeling applications (Earnshaw et al., 2014; Paul et al., 2012), and estimation of correlations between PFAA concentrations and for example population or catchment surface area (Müller et al., 2011; Murakami et al., 2008; Pistocchi and Loos, 2009; Takazawa et al., 2009). Different studies place varying emphasis on the importance of the known PFOA sources: communal wastewater treatment plants (CWWTPs) (Becker et al., 2008; Müller et al., 2011; Perkola and Sainio, 2013; Yu et al., 2009), industrial WWTPs (Murakami et al., 2008; Pistocchi and Loos, 2009; Takazawa et al., 2009) and atmospheric deposition (Filipovic et al., 2013; Scott et al., 2010) have all been reported to be major PFOA sources. According to one estimate, 60% of perfluorocarboxylates (including PFOA) are emitted originally from fluoropolymer factories, out of which 23% is distributed to air, 65% to water and 12% to land (Prevedouros et al., 2006).

The elevated levels of PFOA in the environment, especially in drinking water sources, and the uncertainty about the sources of PFOA, call for raw water intake risk assessment and modeling the fate and transport of PFOA. For example Harada et al. (2003) reported a case where Tama River was heavily contaminated by PFOS originating from a WWTP. Because the downstream of Tama River was used for intake of raw drinking water, also the drinking water was contaminated and PFOS concentrations in tap water were measured to be as high as 51 ng/l. Concerns about drinking water contamination and damage to the vulnerable esker ecosystem rose in Finland after a regional water company, Turku Region Water Ltd., in 1999 announced its plans to extract raw water from River Kokemäenjoki to be used in artificial groundwater recharge in the Virttaankangas esker (Lyytimäki and Assmuth, 2014). Despite the public concern the artificial groundwater recharge system (Fig. SD 1 in Supplementary data) was built and taken into use in 2011 to provide drinking water to the 285 000 inhabitants of the Turku region. The background information about the artificial groundwater recharge plant and the related public debate has been covered in detail by Lyytimäki and Assmuth (2014).

In this study the transport of PFOA and an artificial sweetener acesulfame was modeled using SOBEK river model that was parameterized to describe a 100 km distance of a water course in the Kokemäenjoki River basin. The aim was to assess the transport of the compounds from CWWTPs and the main tributaries to the downstream location where raw water is extracted for the artificial groundwater recharge plant. Since water soluble contaminants such as PFOA are not effectively removed during infiltration and percolation through soil layers, the contaminants in the raw water can remain in the finished drinking water produced at the plant (Davis et al., 2007). The main objectives of the study were to find out whether PFOA concentration in raw water can exceed a safe level and what percentage of the total PFOA load to River Kokemäenjoki is caused by the CWWTPs of the study area. Acesulfame, which was found in surface waters in much higher concentrations than PFOA, was used as a surrogate variable to assess the performance of the water quality model.

2. Materials and methods

2.1. SOBEK river model and the study area

SOBEK is a modeling suite developed for integral water solutions by Deltares in The Netherlands. It involves seven modules, which can be combined for different modeling purposes related to water quantity and quality (Deltares, 2014). This study utilized two modules, "D-Flow 1 D Open Water" and "D-Water Quality 1 D". "D-Flow 1 D Open Water" is a one-dimensional hydraulic model that can be used to model water velocity and level in rivers. It describes water flow as a numerical solution of the complete de Saint Venant equations. "D-Water Quality 1 D" is a one-dimensional water quality model that can simulate transport and mixing of substances using a numerical solution of the advection–diffusion equation. It also includes additional water quality processes, such as sorption and degradation, and supports simultaneous simulation of multiple substances.

The flow model was parameterized to describe water flow in a river reach and three connected lakes in the Kokemäenjoki area as a part of project CONPAT (Assmuth et al., 2015; Perkola et al., 2015). The flow model was available from the project for this study and it was tested by Happonen (2015), where the model performance was assessed against measured river and lake water levels and river flows. The flow model simulated the water velocity and water level for a 100 km distance from River Nokianvirta to the Kolsi hydropower plant that is located in River Kokemäenjoki (Fig. 1). The model included 676 river and lake bed cross sections, which were determined based on sonar data and water depth maps. The raw water intake plant (WIP) is located in