



River water contaminated with perfluorinated compounds potentially posing the greatest risk to young children

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HIGHLIGHTS

- ▶ Young children are at the highest risk of exposure to PFCs via water consumption.
- ▶ Residents reside near to Keya River are at highest risk of exposure to PFCs.
- ▶ River water were used to surrogate as tapwater.
- ▶ Deterministic and probabilistic risk assessment methods were used.
- ▶ Probability density functions for exposure factors were plotted.

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ABSTRACT

Although, humans are exposed to perfluorinated compounds (PFCs) from various media, water consumption could be an important source for the residents living near to contaminated areas. Since comprehensive multimedia exposure model has not been developed for PFCs, assessment of the potential risk due to exposure to PFCs through direct water consumption could be a conservative estimate. The human health risks derived from the exposure to PFCs through water consumption were assessed for different age groups of general population in Taiwan using probabilistic approach. Based on available data on concentrations of PFCs in river water, exposure to PFOS, PFOA and PFDA via water consumption for different age groups were calculated using deterministic and probabilistic risk assessment methods. The oral non-cancer risks from PFOS, PFOA and their combination, expressed as a Hazard Index (HI), was determined by comparing oral exposure dose (through water intake) with the oral Reference Dose (RfD). The average exposure to PFOS, PFOA and PFDA via water consumption for adults ranged from 0.16 to 220.15, 0.43 to 12.5 and 0.43 to 2.36 ng kg-bw⁻¹ d⁻¹ and for children 0.13–354.3, 0.35–20.17 and 0.35–3.79 ng kg-bw⁻¹ d⁻¹, respectively. Probabilistic values of total HIs for all age groups reside near to Keya River exceed the RfD 2.4–4.8 times, corresponding mainly to PFOS with a percentage of 97%. In conclusions, children aged 1–3 years old and the residents reside near to Keya River are at the highest risk of exposure to PFCs via water consumption.

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1. Introduction

Perfluorinated chemicals (PFCs) are all anthropogenic organic chemicals, which are used in a wide range of industrial and commer-

cial applications due to their persistence to biotic and abiotic degradation (OECD, 2002, 2005; Washburn et al., 2005; Fromme et al., 2009) and of significant health concern due to their adverse effects in animal toxicity and in human epidemiological studies such as, hepatotoxicity, developmental and reproductive toxicity, immunotoxicity, hormonal effects and carcinogenic potency (Thomford, 2001; OECD, 2002; 3M-Company, 2002; Seacat et al., 2002, 2003; Kennedy et al., 2004; Harada et al., 2005; USEPA, 2006; Lau et al., 2007; Andersen et al., 2008; Fei et al., 2008, 2009; Lin et al., 2009a, 2009b; Melzer et al., 2010; Lin et al., 2011; Wang et al., 2011; Bloom et al., 2010). PFCs spread globally in various environmental matrices, including air, surface water, sediments, aquatic invertebrates, fish, and other wildlife, predominantly in the aqueous environment (Martin et al., 2003). Several investigations suggested the discharge from industrial WWTPs as one of the significant point sources for

Abbreviations: CATT, C8 Assessment of Toxicity Team; CONTAM, The Scientific Panel on Contaminants in the Food Chain; EFSA, European Food Safety Authority; ELE/OPTO-A, electro/optoelectro A plants; EXPDs, exposure doses; HI, Hazard Index; HSP, Hsinchu Science Park; MDH, Minnesota Department of Health; PDFs, probability density functions; PFCs, perfluorinated compounds (chemicals); PFDA, perfluorodecanoic acid; PFOA, perfluorooctanoic acid; PFOS, perfluorooctane sulfonate; RfD, Reference Dose; SEM, semiconductor; TDI, Tolerable Daily Intake; USEPA, US Environmental Protection Agency; WHO, World Health Organization; WVDEP, West Virginia Department of Environmental Protection; WWTPs, waste water treatment plants.

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PFCs pollution of the aquatic environment (Bossi et al., 2008; Lin et al., 2009c, 2010; Kim et al., 2012).

As their environmental fate trend, PFCs have been revealed in drinking water in many countries in US, Europe and Asia Loos et al. (2007) exhibited PFOA and PFOS in tap water drawn from Lake Maggiore in Italy, which had similar concentrations to those in the lake. In some regions of China, concentrations PFOA and PFOS exceeded 10 ng L^{-1} , whereas the median concentrations were $4.2\text{--}5.4 \text{ ng L}^{-1}$ in rivers, which were comparable to those in the US, Europe and Japan (Saito et al., 2004; Jin et al., 2009). These investigations suggested that water treatment steps (sand filtration and chlorination) may not be efficient to remove the contamination of PFCs. (Skutlarek et al., 2006; Loos et al., 2007). Several extensive studies observed relatively high concentrations of PFCs in human serum (e.g. median 374 ng L^{-1} of PFOA) among those, who used water from drinking water supply, which was highly contaminated by those compounds (e.g. $1900\text{--}18600 \text{ ng L}^{-1}$ of PFOA) (U.S.EPA, 2001; LHWA, 2005; Skutlarek et al., 2006; Emmett et al., 2006; Loos et al., 2007). These data indicate that drinking water is the dominant source for the population resides near to contaminated areas. In recent years, some efforts utilizing activated carbon filtration, and more advanced water treatment processes succeeded for removals. Nevertheless, these successes are questionable in consistency over time (Bartell et al., 2009; Hölzer et al., 2009; Thompson et al., 2011).

As their ubiquitous distribution, humans can be exposed to PFCs through not only drinking water, but also other pathways, including the ingestion with food, such as fish, seafood, livestock, crops and vegetables and house dust; inhalation with air; and water consumption, such as showering, and cleaning. As our current knowledge, few studies have been conducted in risk assessment for PFCs, mainly estimated potential exposure to PFOA and PFOS from various pathways predicting daily intakes deriving the Reference Doses and health based values from drinking water for general adult population in US and Europe (Paustenbach et al., 2007; Fromme et al., 2009). Given that the comprehensive multimedia exposure model has not been developed for human exposure to PFCs due to limited available data on different media, we aimed to maximize the estimates of the potential risk of exposure to PFCs through water consumption using probabilistic assessment. Semiconductor, electrochemical and optoelectronic industries have been grown rapidly in Taiwan. Recent studies reported higher level of PFCs in water area, including industrial and municipal WWTP effluents, rivers and coastal water in Taiwan with the predominant and prevalent exhibition of PFOA, PFOS, PFHxA and PFDA (upto 310, 5440, 406 and 58.2 ng L^{-1} , respectively) compared to those reported in other countries (Tseng et al., 2006; Lin et al., 2009c, 2010). The objective of the present study was to assess the human exposure to perfluorinated compounds through water consumption for different age groups of general population in Taiwan using probabilistic approach. To the best of our knowledge, this is the first exposure assessment for PFDA through water consumption assessing the risk of all age groups, including children.

2. Materials and methods

2.1. Concentrations of PFCs in river water

For the purposes of exposure assessment, PFC concentrations in the river water were used as surrogates for tapwater concentration. Results of two papers reported the concentrations of PFCs in water area in Taiwan, including industrial and WWTP effluents, rivers and coastal water, were adopted for exposure concentrations for water consumption. First investigation covered the surface water collected from the downstream of Tour-Chyan and Nan-Kan

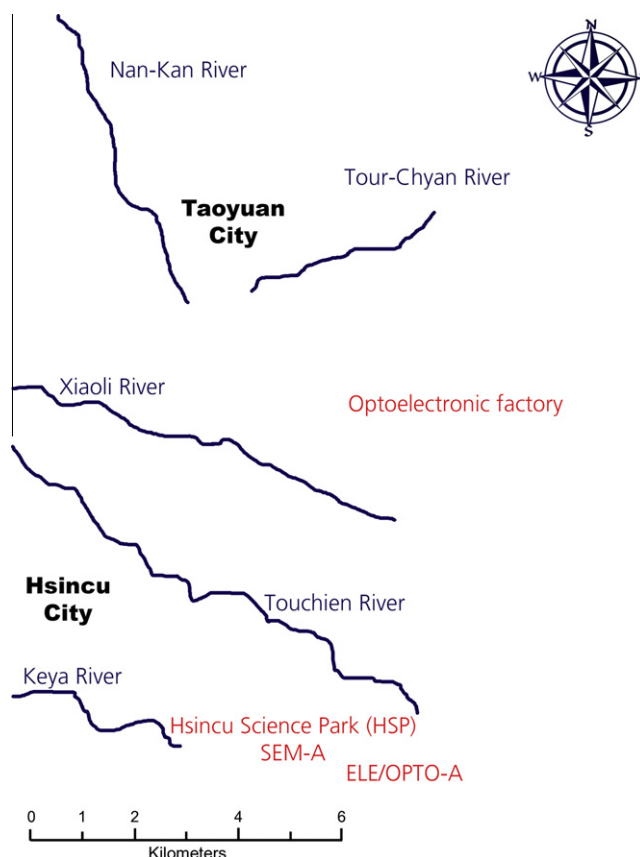


Fig. 1. Map of rivers of interest.

rivers located in northern Taiwan, which receive untreated municipal wastewater and agricultural wastewater directly from the cities and suburban area (Fig. 1) (Tseng et al., 2006). Second paper reported in 2009 by Lin et al. identified many different PFCs in river water from Keya, Touchien, and Xiaoli rivers located near to three major sources of potential PFC contamination (Lin et al., 2009c). The Keya River, serves as waste water drainage of semiconductor (SEM) fabrication plant, is greatly impacted by upstream industrial sources from Hsinchu Science Park (HSP). Correspondingly, The Touchien River, serves a population of 790,000, and the Xiaoli River, serves a population of 38,000, are impacted a half of its PFC load by upstream electronics/optoelectronics fabrication (ELE/OPTO) wastewaters and small optoelectronics factory, respectively (Fig. 1). PFOA, PFOS and PFDA were predominant and prevalent in all samples in both papers.

Exposure point concentrations and probability density functions (PDFs) of PFOS, PFOA and PFDA in surface water, as an environmental compartment in the risk assessment are summarized in Table 1.

2.2. Exposure assessment

Daily intakes of PFCs were calculated using both deterministic and probabilistic risk assessment methods for different age groups (<0.5 ; $0.5\text{--}0.9$; $1\text{--}3$; $4\text{--}12$; $13\text{--}18$; $19\text{--}64$; >65).

To evaluate the exposure doses for humans, PFC concentration values were converted into doses using U.S.EPA (1997) exposure equation, considering the concentration of contaminants in tapwater, the intake rate of tapwater, exposure frequency, exposure duration and body weight of an average individual (U.S.EPA, 1997). The equation is expressed as follows:

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