CURRENT STATUS OF ENVIRONMENTAL CONTAMINATION, HUMAN EXPOSURE AND EMISSION PERFLUORINATED COMPOUNDS (PFCs) IN SOUTH KOREA

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Introduction

Exposure and contamination by perfluorinated compounds (PFCs) are a global emerging concern due to their typical POPs properties including persistency, bioaccumulation, toxicity and long-range transportable potential[1,2]. In 2009, the Stockholm Convention on Persistent Organic Pollutants (POPs) included perfluorooctanesulfonate (PFOS) and its related substances in the category of new POPs[3].

PFCs consist of water-soluble, less volatile, and environmentally persistent groups, and a volatile, neutral, and environmentally degradable precursor group. The former includes perfluoroalkyl acids (PFAAs) such as perfluorocarboxylic acids (PFCAs) and perfluoroalkylsulfonates (PFASs), and the latter includes fluorotelomer alcohols (FTOHs) and perfluoroalkyl sulfonamide/sulfonamidoethanols (FOSAs/FOSEs)[1]. PFAAs, a major contributor to total global PFCs emission, are known to be directly emitted to the environment from manufacturing, chemical usage, and/or distribution processes, while some of PFAAs are indirectly emitted via transformation of PFC precursors[1,4]. Therefore, levels in and transfer among environmental compartments including air, water, soil, sediment, biota, and human are strongly associated with the relative composition of PFAAs and precursors in emission and their fates in the environment.

PFCs in the Korean environment have been monitored since the early 2000's when concern over PFAAs was initiated by their remarkably high levels measured in some ambient waters and some citizen sera[5,6]. Those elevated levels in the Korean environment reported by the early studies, which were frequently similar with or above the levels in the environment of developed countries and thus were in an opposite trend of existing POPs, evoked a potential risk of emerging POPs in Korea.

The early PFCs studies in Korea have been focused on relatively contaminated areas, the so-called "hot-spot", in which concerns of national PFCs status could be overestimated and/or be biased. Also, most of the early studies targeted PFAAs only, by which contamination and/or exposure mechanisms could not be explained. Research articles for existing POPs showed a decreasing trend after the maximum in the mid-2000's while sharply increasing trend for emerging POPs[7]. Also, national surveillance including regular monitoring for contamination level in water, soil, and sediment, human exposure, and emission estimation have been performed since the year of 2005. According to recent research trend and governmental efforts, it is expected possibly 1) to determine a representative of PFCs status in Korean environments, 2) to assess the transfer of PFCs among compartments, and 3) to evaluate whether there exists a harmonization among levels of emission, environmental contamination, and human exposure measured in Korean environment. This review paper presents the preliminary results on the basis of meta-analysis of the data collected for emission, environmental contamination, and human exposure.

Materials and methods

We collected PFCs data from the literature published during the periods of 2004-2012. Monitoring data of environmental contamination were collected for PFAAs in ambient waters, soils, sediment, and biota both in inland and marine environments.

Human exposure data were based on internal dose and external dose. Concentrations detected in sera of general population were used for internal dose. The serum concentrations were used to estimate daily intake of PFCs based on one-compartment steady-state pharmacokinetic model (PK model). For external dose, daily intakes of PFCs were also calculated from the production of concentrations in drinking waters, foodstuffs, indoor/outdoor air, and house-dust and uptake rate (and activity) via each exposure pathway. The probability distribution of daily intakes were obtained from Monte-Carlo simulation using Crystal-Ball® software in which

value of individual parameters was extracted from their distribution functions and thereafter was iteratively caclualted.

Emission of PFCs in Korea has been estimated for PFAAs in wastewater treatment plants as a point source or running waters as a sum of point and nonpoint sources. Collected emission data were compared simply and/or were used to recalculate the probability distribution of emission.

Results and discussion

Contamination levels in ambient waters

Median concentrations of PFOA and PFOS measured in Korean ambient waters were 7 ng/L and 4.5 ng/L, respectively (Figure 1). 51% of data were originated from "hot-spot" areas including the Nakdong River (NR) and the Shihwa Lake (SL) watersheds, indicating that average (and median) concentrations would be lower. Therefore, we expect that average contamination level in Korean aquatic environments is not likely to be higher when compared with those in other countries.

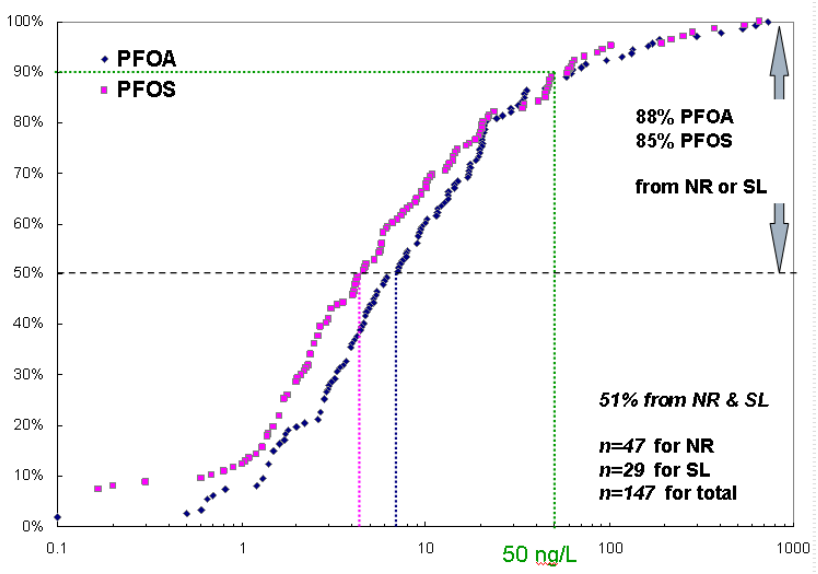

Figure 1. Distribution of PFOA and PFOS concentrations measured in Korean ambient waters

Human exposure levels

Extensive monitoring has been performed to determine average PFOA and PFOS in sera of general population. Meta-analysis results showed average concentrations of 2 ng/ml for PFOA and 5 ng/ml for PFOS. These levels were two times lower than those in developed countries. Korean daily intakes estimated by PK model were 0.42 ng/kg/day for PFOA and 0.69 ng/kg/day for PFOS (Figure 2), which were in similar range with Norwegian daily intakes but several to several tens times lower than those of other developed countries.

PFOA and PFOS intakes via inhalation and tap water drinking accounted for small portion of total exposure. This means that other exposure pathway including food uptake and dermal exposure could be principle contributor (Table 1). We calculate those two exposures (not shown).

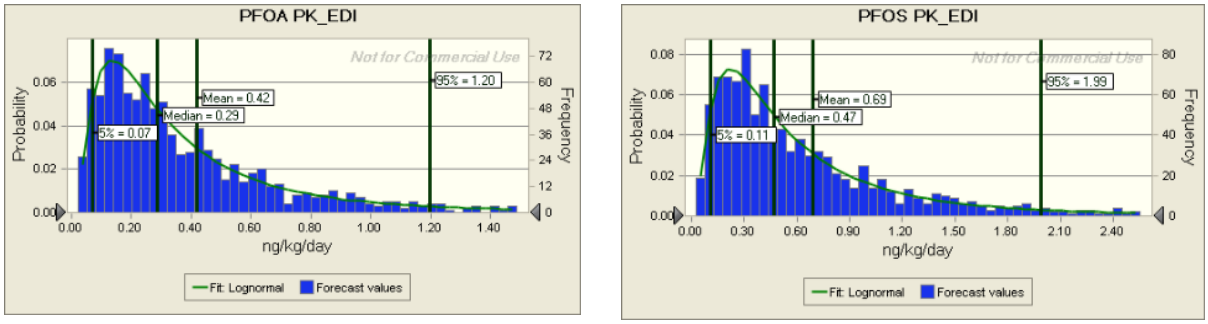

Figure 2. Probability distribution of daily intakes of PFOA and PFOS estimated by PK model from concentrations in Korean sera.

Table 1. Daily intakes of PFOA and PFOS estimated from the production of concentrations and uptake rate of each exposure pathway.

	Year	PFOA	PFOS
		Concentration of precursors	
Indoor	2009	5.13	0.05
Outdoor	2009	12.3	0.12
Tap waters	2009	5.4	1.2
		Exposure (ng/kg/day)	
Inhalation exposure	2009	0.007(1.7%)	0.003 (0.4%)
Drinking exposure	2009	0.12 (28.6%)	0.03 (4%)
PK-EDI exposure	2003-2008	0.42 (100%)	0.69 (100%)

Emission levels

We calculated emission factor (per capita daily emission) both for WWTPs-derived discharge and river-derived discharge (Table 2). Emission factors of Korea were a few to several ten times lower than those of Japan and European countries. Furthermore, contribution of point source (WWTPs) to aquatic PFCs seemed to be relatively minor when compared with river-derived emission factors which would reflect total emission factor from point and nonpoint sources.

PFCs discharged through rivers/streams enter coastal marine environments and transport to open oceans. Coastal waters of Korean peninsula seems to receive river waters different PFC composition pattern (Figure 3). The majority of PFASs such as PFHxS and PFOS were discharged from the Han River while most of PFCAs such as PFOA and PFNA were discharged from the Nakdong River. This indicates that Korean coastal waters could be influenced by different PFC contamination.

Table 2. Comparison of PFC emission factor.

μg/capita/day		PFOA	PFNA	PFHxS	PFOS	reference
river / stream	Korea (median)	23	4.4	4.4	18	this study
	Korea (average)	75	36	17	43	this study
	Japan	120	55.7		77.2	Murakami et al., 2008
	EU	78.2	1.42	--	--	McLachlan et al., 2007
	EU	82[a] (19.2)[b]	--	--	27.4	Pistocchi and Loos, 2009
	Switzerland	17.6	--	33.1	101	Huset et al., 2008
WWTP	Korea	3.17	0.32	1.84	1.68	Kim et al., 2011
	Denmark	2.5	0.34	1.83	1.42	Bossi et al., 2008
	USA	97	4.65	6.29	10.5	Loganathan et al., 2007
	Switzerland	11.7	0.14	15.2	57.2	Huset et al., 2008
	USA	12.4	0.82	2.46	9.54	Schultz et al., 2006
	Japan	28.7	33.2	--	154	Murakami et al., 2008

[a] from all data; [b] from data of <0.5 t/yr in Pitocchi and Loos, 2009
values for references were quoted or calculated using information provided

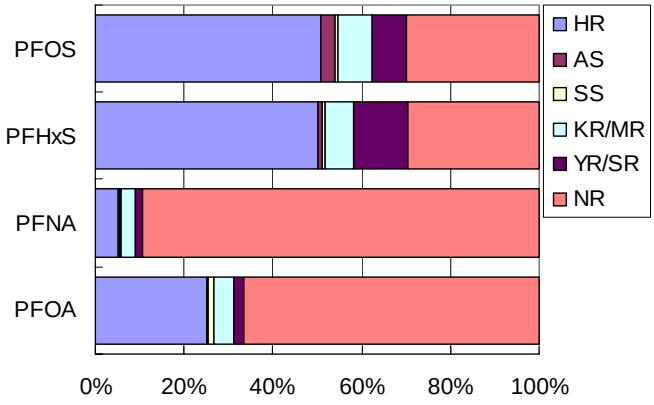

Figure 3. Relative contribution of major river/streams to coastal water contamination.

Acknowledgements
This work was supported by Incheon National University Research Grant in 2012 and was also a part of the project (11162KFDA725) funded by the Korea Food & Drug Administration in 2011.

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