METHOD DEVELOPMENT AND MONITORING OF PERFLUORINATED COMPOUNDS IN COASTAL WATERS, BIOTA, AND HUMAN BREAST MILK SAMPLES FROM ASIAN COUNTRIES

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Abstract

The increased awareness of global distribution of perfluorinated compounds (PFCs) in the environment has initiated a cooperative study between Japan and Hong Kong. This joint cooperation targeted the development of new methods for the analysis of PFCs in various matrices at low concentrations, and the subsequent monitoring of PFCs in different environmental matrices and human population in Asian countries.

An accurate and reliable analysis of PFCs has been impeded by the problems of procedural blanks and the lack of well-developed analytical methods. In the present study, the concentrations of PFCs in various laboratory equipment and parts of the analytical instruments were measured, and subsequent modifications were made to reduce the blank levels as necessary. New methods were also developed for the analysis of PFCs in water samples, mussel and oyster tissues, and human breast milk samples. Coastal and open ocean water samples were extracted by solid phase extraction (SPE) employing hydrophilic-lipophilic balance (HLB) cartridge. This method allows the measurement of perfluorooctane sulfonamide (PFOSA), perfluorooctansulfonate (PFOS), perfluorohexanesulfonate (PFHS) and perfluorobutanesulfonate (PFBS), 1H, 1H, 2H, 2H-perfluorooctanesulfonic acid (THPFOS), perfluorononanoic acid (PFNA) and perfluorooctanoic acid (PFOA) at pg/L levels. A combination of alkaline digestion and SPE was developed for the extraction of PFCs from soft tissues of mussels and oysters. This method is rapid and robust with acceptable recoveries for most of the target analytes. However, trace levels of certain PFCs in the procedural blanks restricted the applicability of this method to highly-contaminated samples. New extraction method by SPE employing weak-anion exchange (WAX) cartridge was developed for the analysis of human
breast milk. The achievement of low limits of quantifications (LOQs) for various PFCs makes this new method suitable for the analysis of milk samples with comparatively low PFC concentrations.

The newly developed methods were applied to the analysis of water, mussel, oyster, and human breast milk samples collected from Asian countries. Seawater samples were collected from Hong Kong (6 locations for both summer and winter seasons), China (8 and 6 locations for summer and winter locations, respectively), and Korea (11 locations). PFOS and PFOA were the two dominant chemicals found in all water samples. Ranges of concentrations of PFOS in coastal seawaters of Hong Kong, Pearl River Delta (PRD), including South China Sea, and Korea were 0.09 – 3.1, 0.02 – 12 and 0.04 – 730 pg/mL, respectively while those of PFOA were 0.73 – 5.5, 0.24 – 16 and 0.24 – 320 pg/mL, respectively. Seasonal variations of PFC concentrations were observed in surface seawaters in PRD and South China Sea indicating the influence of the Pearl River discharge on the extent of PFC pollution in China and, to a lesser extent, Hong Kong. In this study, maximum PFOS and PFOA concentrations were detected in a location within Kyeonngi Bay in Korea, which is heavily influenced by wastewater discharged from a number of local industries. The high PFHS concentration (52 pg/mL) in this location indicated a local source of this chemical around Kyeonngi Bay. Apart from this specific location, the PFC concentrations from other locations in Korea were comparable to those in Hong Kong and China.

This pioneer study indicated the potential discharges of PFCs in the environment of Hong Kong and China. As part of the continual monitoring survey, biota and human breast milk samples were collected in order to elucidate the distribution of PFCs in another environmental matrix as well as the human population. Mussel samples were
collected from six locations along the east coast of China and oyster samples were collected from Tokyo Bay in Japan. Concentrations of individual PFCs in mussels and oysters from south China and Japan ranged from 113.6 to 586.0 pg/g, wet weight (ww) for PFOS, 63.1 to 511.6 pg/g, ww for PFHS, <12.0 to 30.1 pg/g, ww for PFBS and 37.8 to 2,957.0 pg/g, ww for PFOSA. The analysis of perfluoroalkyl carboxylates was compromised by the presence of certain carboxylates in the procedural blank. The maximum PFOA concentrations (660.5 pg/g, ww) were detected in oyster samples from Tokyo Bay, while that of perfluoroheptanoic acid (PFHpA) (507.1 pg/g, ww) and perfluorohexanoic acid (PFHxA) (346.9 pg/g, ww) were detected in mussel samples from Bei Hai in China. Different composition profiles between mussel (from China) and oyster samples (from Japan) indicated clear differences in the pollution sources between China and Japan.

The extent of human exposure to PFCs in China was evaluated by the measurement of PFC concentrations in human breast milk samples collected from 19 primiparas mothers from Zhoushan, China. PFOS and PFOA were the two dominant PFCs in the breast milk samples. The maximum concentrations of PFOS and PFOA were 360 ng/L and 210 ng/L, respectively, which were much less than average concentrations detected in human blood or serum samples. No statistically significant correlation was found between concentrations of either PFOS or PFOA and maternal age, weight or infant weight. Although perfluoroundecanoic acid (PFUnDA), perfluorodecanoic acid (PFDA) and PFNA were found to have a significant positive correlation with fish consumption data, a definitive conclusion could not be drawn due to the small sample size in the present study. Future studies will be necessary to elucidate the possible exposure pathways of PFCs to human population.
The occurrence of various PFCs in water and biological samples from Hong Kong, China, Korea, and Japan indicated the contamination of PFCs in these areas. Different composition profiles in the samples suggested the existence of independent pollution sources of PFCs in each country. The comparatively smaller PFC concentrations in water and mussel samples from Hong Kong and China indicated that these regions are probably not heavily polluted by PFCs. The concentrations of PFOS in all the water samples from Hong Kong and China were less than those that would be expected to cause adverse effects to aquatic organisms and their predators. PFOS concentration in water samples from Kyeonngi Bay, Korea, nevertheless, was higher than the guideline value that is protective of avian wildlife. The PFOS concentration in this location is likely to pose adverse effects on aquatic animals and avian wildlife, and a more refined risk assessment is necessary in this area. The detection of PFCs in human breast milk suggested the possibility that various PFCs could accumulate in breast milk, and the subsequent maternal transfer to infants through breast-feeding. One out of 19 milk samples had a PFOS concentration greater than the safety guideline value. However, it should be noted that uncertainty factors were applied in the risk assessment process, and thus the actual risk may have been overestimated. Overall, there is generally a low risk for infants from PFCs through breast-feeding. It is recommended that future studies should be carried out to elucidate the possible exposure pathways of PFCs to human population.
Lists of publications


Papers published in conference proceedings


Falandysz, J., Jecek, L., Rostkowski, P., Gulkowska, A., Taniyasu, S., So, M.K., Yamashita, N. Concentration of perfluorinated carboxylic acids in river waters supply to the city of Gdansk, Poland. Proceedings of International Symposium on...

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