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Evaluation and Management of Emerging Chemicals by the Comprehensive Monitoring Using Local Networks

Takahiro NISHINO
Tokyo Metropolitan Research Institute for Environmental Protection
1-7-5 Shinsuna, Koto-Ku, Tokyo 136-0075 JAPAN
E-mail: nishino-t@tokyokankyo.jp

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In recent years, the number of chemicals developed and used has been increasing year by year not only in Japan but also worldwide, with the hundred-millionth chemical registered with the Chemical Abstract Service (American Chemical Society) in 2015. Particularly, in urban areas (e.g., Tokyo, Osaka, Nagoya, Fukuoka) there is more consumption of these chemicals, and the concentrations of these chemicals in several environmental media are higher there than in other areas. However, no surveys of environmental pollution by these chemicals have been carried out, and not enough has been done to assess the environmental risk.

To ascertain the state of environmental pollution with these chemicals, the Environment Research and Technology Development Fund study (5-1602) "Evaluation and Management of Emerging Chemicals by the Comprehensive Monitoring Using Local Networks," was conducted from FY2016 to FY2018.

In this study, a local network of regional environmental research institutes was used effectively to survey emerging chemicals in five large cities (Tokyo, Nagoya, Hyogo, Osaka, and Fukuoka) in Japan. First, a screening analysis of river water and sediment samples from these five cities was carried out using GC-MS and LC-QTOFMS at the Fukuoka Institute of Health and Environmental Sciences and Nagoya City Environmental Science Research Institute, respectively. Prior to the analysis, the water samples were extracted using a solvent extraction method with dichloromethane for GC-MS, and solid phase extraction method using OASIS HLB and Sep-Pak AC2 cartridges for LC-QTOFMS. Similarly, sediment samples were extracted using a solvent extraction method with acetone and dichloromethane. The data from the screening analysis and other information (Quantitative Structure-Activity Relationship (QSAR) and references) indicated which chemicals should be researched, preferentially, in each environmental medium. In the water samples, the main chemicals detected were pharmaceuticals and personal care products (PPCPs), such as antibiotics (e.g., clarithromycin) and antihypertensives (e.g., telmisartan). In the case of the sediment samples, the chemicals researched included polycyclic aromatic hydrocarbons (PAHs) (e.g., benzo[a]pyrene) and plasticizers (e.g., bis (2-ethylhexyl) phthalate). Next, quantitative analyses of these chemicals were carried out in three environmental media (air, water and sediment) as part of a risk assessment for aquatic organisms. The air samples taken during warm and cold seasons in the five cities were analyzed by the Osaka City Research Center of Environmental Science. Analyses of the water samples were carried out at the Tokyo Metropolitan Research Institute for Environmental Protection. Sampling points for water were mainly selected downstream of effluents (e.g., sewage treatment

plants), and 42 samples were collected from the five cities in each season. Sampling was carried out twice a day (AM and PM), taking account of daily variation in concentration of chemicals. Sediment samples were collected at 17 points in the cities and analyzed at the Hyogo Prefectural Institute of Environmental Sciences. The risk assessment for aquatic organisms was carried out by comparing the analyzed data with the Predicted No-Effect Concentration (PNEC) gathered from various sources. PNEC values vary widely depending on the reference. Therefore, the smallest value was adopted to be on the safe side for each chemical. For the water samples, a summary of chemical concentrations is given in Table 1, in which averaged concentration data between AM and PM is listed.

Table1 Concentrations of chemicals in the water samples (ng/L).

Compound		Clarithromycin		Erythromycin		Carbamazepine		Diclofenac		Triclosan	
(PNEC)		50		20		29.7		66.3		28	
Sampling point		Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer
Fukuoka	Najima B.	**190**	28	7.2	(2.1)	**31**	17	21	(4.3)	N.D.	N.D.
	Tonomono B.	**210**	5.3	7.7	N.D.	21	N.D.	**200**	10	N.D.	N.D.
Osaka	Shimoshiromi B.	**910**	**170**	**60**	**28**	**74**	**42**	**1,200**	**72**	**(42)**	N.D.
	Kema watergate	**130**	4.3	11	(1.4)	19	6.6	**92**	20	N.D.	N.D.
Hyogo	Tokura B.	**720**	**210**	**65**	**32**	**70**	**36**	**140**	65	**(53)**	N.D.
	Downstream of STP	18	**53**	(0.7)	(1.6)	11	3.7	**89**	**90**	N.D.	N.D.
Nagoya	Maizuru B.	**800**	**340**	**31**	**32**	**49**	**31**	**100**	**73**	**190**	**440**
	Nakatsuchito B.	**760**	**530**	**40**	15	**130**	**82**	**400**	**96**	**130**	**190**
Tokyo	Hino B.	**540**	**170**	**63**	**29**	**73**	**60**	**83**	42	**(41)**	N.D.
	Ryogoku B.	**170**	**180**	18	**23**	**31**	**39**	**91**	**120**	N.D.	N.D.

Compound		N,N-diethyl-m-toluamide		Trimethoprim		Caffeine		Bisphenol A		Fexofenadine	
(PNEC)		5,200		1,000		5,200		1,500		>25,600	
Sampling point		Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer	Winter	Summer
Fukuoka	Najima B.	28	80	25	14	180	40	(100)	N.D.	1,200	290
	Tonomono B.	29	140	21	N.D.	480	110	N.D.	N.D.	1,100	33
Osaka	Shimoshiromi B.	130	510	96	68	44	(22)	520	N.D.	5,000	1,500
	Kema watergate	58	63	11	3.7	14	(20)	N.D.	N.D.	840	110
Hyogo	Tokura B.	67	99	97	87	270	78	550	130	5,700	1,200
	Downstream of STP	10	24	19	6.6	53	(26)	N.D.	(86)	980	340
Nagoya	Maizuru B.	87	70	130	230	5.5	28	520	150	6,500	2,000
	Nakatsuchito B.	75	220	130	82	280	79	490	200	6,900	800
Tokyo	Hino B.	24	88	93	47	(12)	N.D.	400	(70)	5,500	590
	Ryogoku B.	55	180	31	39	120	71	(55)	N.D.	460	650

B.:Bridge
STP:Sewage Treatment Plant
Data are shown as the average values for AM and PM at each point.
Numbers highlighted in grey indicate that these concentrations exceeded 1/10 PNEC.
Numbers in bold indicate that these values exceeded the PNEC.

The results show that the concentration levels of five kinds of chemicals (clarithromycin, erythromycin, triclosan, diclofenac and carbamazepine) exceeded the PNEC in several of the water samples. Concentrations of antibiotics such as clarithromycin or antihistamines such as fexofenadine tended to be higher in the cold season than in the warm season. In contrast, insect repellents such as N, N-diethyl-m-toluamide tended to be higher in the warm season than in the cold season. It was thought that many of these chemicals were effluents from several kinds of factories and households, flowing into rivers or the sea directly or via sewage treatment plants. Table 2 gives a summary of the results on the sediment samples. In the samples, several kinds of PAHs and bis (2-ethylhexyl) phthalate were detected at concentrations higher than the low level of the effects range set by the National Oceanic and Atmospheric Administration (NOAA), or the PNEC calculated according to the equilibrium partitioning method. In all of the air samples, the same chemicals studied in the water and sediment samples were found at levels below the detection limit. This shows that the chemicals researched in this study

Table2 Concentrations of chemicals in the sediment samples (ng/g-dry)

Chemical / Sampling Point		Naphthalene	Acenaphthene	Fluorene	Phenanthrene	Fluoranthene	Pyrene	Benzo[a]Anthracene	Benzo[k]Fluoranthene	Benzo[k]Fluoranthene	Benzo[e]pyrene	Bis(2-ethylhexyl)phthalate
Fukuoka	Najima B.	17	3.3	4.0	31	28	30	8.2	5.0	5.1	11	1,600
Osaka	The estuary of the Yodogawa river	15	4.6	7.7	45	110	100	46	28	33	39	940
Hyogo	Samon B.	20	2.7	4.7	27	64	69	27	20	18	31	1,500
Nagoya	Chidori B.	21	5.1	5.1	32	91	100	41	34	30	59	5,000
Tokyo	The estuary of the Sumidagawa river	74	16	26	130	210	210	82	63	57	96	4,500
NOAA ERL		160	16	19	240	600	665	261	–	–	–	–
NOAA ERM		2,100	500	540	1,500	5,100	2,600	1,600	–	–	–	–
PNECsed		150	1,780	350	4,100	1,000	42	0	2	2	4	1,270

shows exceed the value of NOAA ERL.
shows exceed the value of PNECsed.

were either not transferred from an atmospheric medium, or were not volatilized into it.

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