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Original Article

Water Quality and Concentration of Alkylphenols in Rivers Used as Source of Drinking Water and Flowing through Urban Areas

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Abstract

Objectives: To examine nonylphenol (NP) and 4-*t*-octylphenol (OP) concentrations and general water quality indicators along a river in the greater Tokyo area and to specify the distribution and origin of alkylphenols.

Methods: Water was sampled from the Edogawa River, a main river in the greater Tokyo area, which is a source of public drinking water; and the Sakagawa River system, a tributary of the Edogawa River. To determine alkylphenol in river water, NP and OP concentrations were quantified using gas chromatography-mass spectrometry (GC-MS).

Results: The detection rates of NP above the detection limit were 100% in both rivers, and those of OP were 75.0% in the Edogawa River and 92.9% in the Sakagawa River system. The median NP and OP concentrations in the Edogawa River were 0.24 μg/l and 0.066 μg/l, and those in the Sakagawa River system were 0.87 μg/l and 0.19 μg/l respectively. Alkylphenol concentrations are significantly higher in the Sakagawa River system than in the Edogawa River. In the Sakagawa River system, the NP and OP concentrations were highest in the water along the nonindustrial area with an underdeveloped sewerage system.

Conclusions: NP and OP were detected widely in the Edogawa River and Sakagawa River system. Endocrine-disrupting chemical (EDC) pollution in a river by the inflow of urban wastewater was demonstrated. A systematic monitoring of alkylphenols in tributary rivers and streams as well as in main rivers will help control EDC pollution and protect the source of drinking water in urban areas.

Key words: nonylphenol, octylphenol, river, urban area, sewerage system

Introduction

Nonylphenol (NP) and 4-*t*-octylphenol (OP) are alkylphenols (APs) suspected to be endocrine-disrupting chemicals (EDCs) (1–3); both have been widely used as nonionic surfactant materials and additives in plastic materials. Concern has recently increased about the diffusion of these compounds into water environments (4–8). EDCs have been defined as artificial substances in the environment that interfere with the normal functioning of the endocrine hormone system when they are absorbed by the body; they are also referred to as "environmental hormones" (9, 10).

Jobling et al. (11) reported that the production of a vitellus precursor protein in blood named vitellogenin, which is normally found in mature female fish, is induced by NP in male fish. An association has been found between the decline in reproductive power and the biosynthesis of vitellogenin due to induction by OP, and an equivalent effect of degradation metabolites of alkylphenol polyethoxylates (APEs) has been demonstrated (12–14). Colborn et al. (1) stated in their book "Our Stolen Future" that estrogenic compounds like OP are generated in the degradation of APEs used in many industrial and household detergent formulations. APEs are also used as emulsifiers and wetting agents in household articles, toiletries, pesticide formulations, and many other industrial and agricultural products (15).

In Japan, 70% of tap water is provided from surface water, including river water (16). Rivers are the key to maintaining sanitary environments because they provide a source of tap water while at the same time receiving sewage. Consequently,

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at the lower reaches of a river running through urban areas, the mixing of wastewater to some extent with the source water for water works is inevitable.

Here, we present a systematic water survey of the Edogawa River and its tributary, the Sakagawa River system, which is believed to be the main source of water pollution at the lower reaches of the Edogawa River (17). Our aim is to specify the distribution and origin of EDCs in the source of tap water in urban areas. The distribution of APs and the association between APs and general water quality indicators are studied.

Materials and Methods

Survey areas and sampling points (Fig. 1)

The Edogawa River branches off from the Tonegawa River and flows along the border between three major prefectures in the greater Tokyo area, namely, the Saitama Prefecture and the Tokyo Metropolis on the right bank and the Chiba Prefecture on the left bank, branches into the Old Edogawa River and flows into Tokyo Bay. It is 54.7 km long, with a catchment area of 200.3 km^2. The Sakagawa River system rises from a plateau in the northern part of the Chiba Prefecture, receives rainwater from three river basin cities (Kashiwa, Nagareyama and Matsudo) and first meets the Edogawa River at a point of 24.3 km from the mouth. Its catchment area is 53.9 km^2 (18).

There are three water purification plants downstream of the Edogawa River, specifically downstream of the confluence region where the Sakagawa River system flows into the Edogawa River. These plants provide tap water to 3.2 million people. Therefore, the water quality of the Edogawa River and Sakagawa River system is of critical concerned being a major source of tap water for the greater Tokyo area (17).

The flow of the Edogawa River and Sakagawa River system in our study area has decreased because of a decline in incoming flow and also with the development of an urban sewerage system over the last 20 years. The decrease in flow

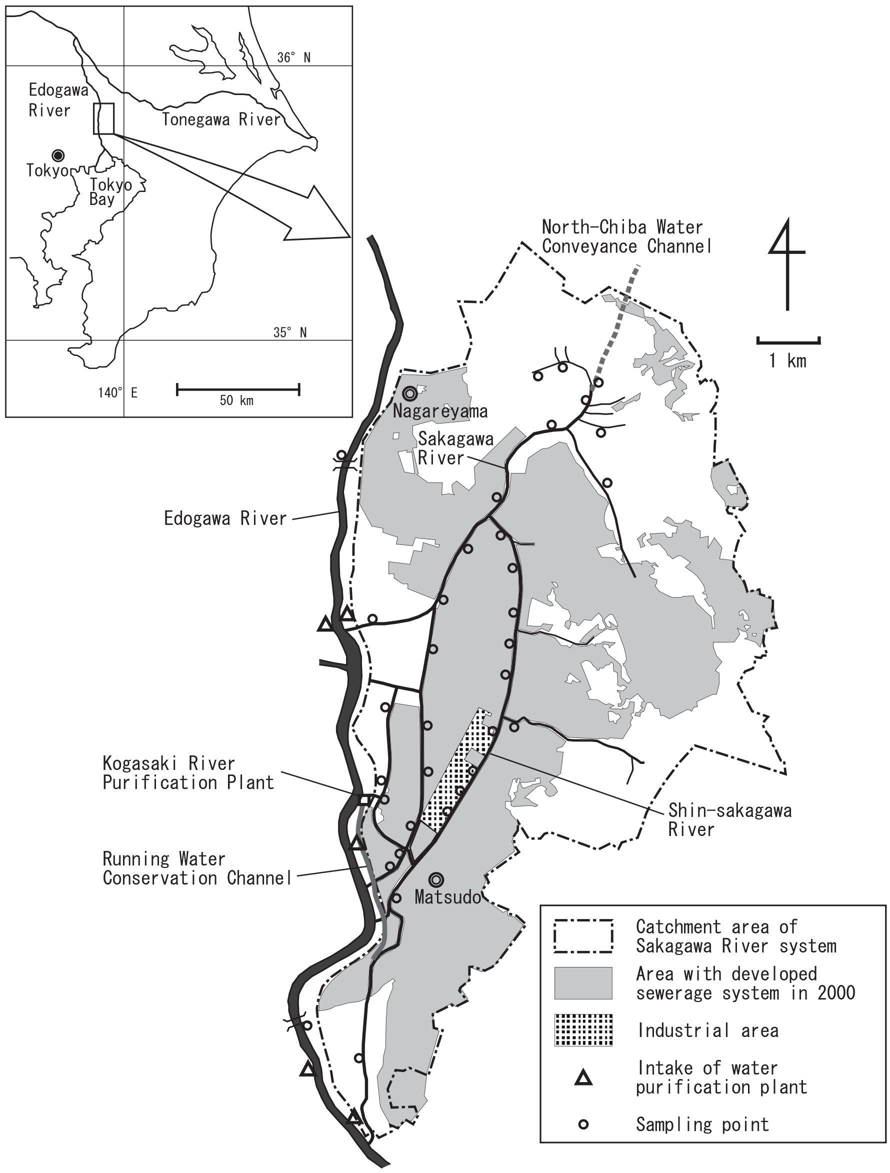

Fig. 1 Map of Edogawa River, Sakagawa River system and sampling points.

was more evident in the Sakagawa River system than in the Edogawa River. Records of the Ministry of Environment showed that the average 75th percentile in biochemical oxygen demand (BOD) of the Sakagawa River system is between 16.7 mg/l and 5.7 mg/l from 1990 to 2000. Although BOD has been declining in recent years, the level of water pollution in the Sakagawa River system remains high, compared with the average 75th percentile in BOD, i.e., between 3.2 mg/l and 2.0 mg/l, in the Edogawa River from 1990 to 2000. Pouring less polluted water into the upper reaches of the Sakagawa River through the North-Chiba Water Conveyance Channel started to solve these problems in April of 2000 (18).

Thirty-four sampling points were defined for this study either from (a) the Sakagawa River system and its own tributaries or (b) part of the Edogawa River near the Sakagawa River system (Fig. 1). Samples from the Sakagawa River system and its own tributaries area were further categorized as follows: (a-1) samples from a nonindustrial area with a developed sewerage system, (a-2) samples from an industrial area where each factory has installed a sewerage system, (a-3) samples from a nonindustrial area with an underdeveloped sewerage system that receives water from the North-Chiba Water Conveyance Channel, (a-4) samples from a nonindustrial area with an underdeveloped sewerage system and (a-5) others, which comprised samples from the North-Chiba Water Conveyance Channel and samples for which the above categorization is non applicable. Water samples were collected at a depth of 10 cm below the surface midstream using a stainless steel water sampler suspended from bridges between May and September 2000. They were stored in 3-liter amber glass bottles and transported to the laboratory in an ice box.

Identification of NP and OP

Sample preparation was based on the "Endocrine Disrupting Chemicals Interim Investigation Manual (Water, Sediment, and Aquatic Organisms)" published by the Japan Environmental Agency (19). One liter of sample water was adjusted to pH 3.0 by adding hydrogen chloride; 50 ml dichloromethane was then added and the sample was shaken for 10 min, after which the dichloromethane layer was extracted. This procedure was repeated and the two extracted dichloromethane layers were merged. The obtained dichloromethane extract was then dehydrated using an evaporator and a nitrogen stream until the volume becomes 1.0 ml. 1 μl of this solution was used to identify and qualify NP and OP by gas chromatography-mass spectrometry (GC-MS) under the following conditions. The gas chromatograph HP6890 (Hewlett-Packard, Palo Alto, CA, USA) and an HP-5MS column (30 m×0.25 mm i.d., 0.25 μm film thickness) (Hewlett-Packard, Palo Alto, CA, USA) were used. The initial temperature before injection was 50°C for 2 min; it was increased at a rate of 10°C/min to 280°C and held there for 5 min; the injection temperature was 280°C. The mass spectrometer HP5973 (Hewlett-Packard, Palo Alto, CA, USA) was used in the electron impact mode at 70 eV. We monitored ions having m/z values 107, 121, 135, 136, 149, 150, 163, 188 in the single ion monitoring (SIM) mode, and calculated NP and OP concentrations using the peak areas of the target materials and those of internal standards, i.e., naphthalene-d_8 (m/z=136) (Wako Pure Chemical Industries, Ltd., Osaka, Japan) and phenanthrene-d_{10} (m/z=188) (Wako Pure Chemical Industries, Ltd., Osaka, Japan).

Measurement of water quality indicators

We measured the following physicochemical water quality indicators within 6 hours from individual sampling: concentrations of ammonia nitrogen (NH_4^+-N), nitrite nitrogen (NO_2^--N), nitrate nitrogen (NO_3^--N) and phosphoric ion (PO_4^{3-}-P) by absorption photometry using an absorption photometer (HC-1000, Central Kagaku Corp., Tokyo, Japan); concentrations of total nitrogen (TN) and total phosphorus (TP) by following the testing methods for industrial wastewater (Japanese Industrial Standards-K0102) (20); and biochemical oxygen demand (BOD) by a microbiological membrane method using BOD-2000 (Central Kagaku Corp., Tokyo, Japan). In individual sampling sites, pH and dissolved oxygen (DO) were measured using a pH meter (HI-98111, HANNA Instruments, Padova, Italy) and DO meter (OM-12, Horiba, Ltd., Kyoto, Japan).

Statistical analysis

Statistical analysis was conducted by SPSS 10.0 for Windows. Indicators of the differences between the Edogawa River and the Sakagawa River system were compared by Mann Whitney's U-test. Correlations between indicators were examined using Spearman's correlation coefficients, since the distributions of APs were significantly different from the normal distribution. Differences in the measurements among the four groups in the Sakagawa River system were examined by the Kruskal Wallis test.

Results

Seventy samplings were conducted: 12 in the Edogawa River, 56 in the Sakagawa River system and 2 in the North-Chiba Water Conveyance Channel. Table 1 shows the AP concentrations and values of water quality indicators in our study. In the Edogawa River, the median concentration of NP was 0.24 μg/l, and that of OP was 0.066 μg/l; in the Sakagawa River system, these were 0.87 μg/l and 0.19 μg/l, respectively. The AP concentration was significantly higher in the Sakagawa River system than in the Edogawa River. The detection rates of NP above the detection limit (0.1 μg/l) were 100% in both rivers, and those of OP (detection limit: 0.01 μg/l) were 75.0% in the Edogawa River and 92.9% in the Sakagawa River system. The NP and OP concentrations of two samples from the North-Chiba Water Conveyance Channel were respectively 0.30 and 0.43 μg/l for NP, and 0.025 and 0.040 μg/l for OP; these values were similar to those in the Edogawa River.

As regards general water quality indicators, the mean total nitrogen, ammonia nitrogen, nitrite nitrogen, total phosphorus, phosphoric ion and BOD levels were significantly higher in the Sakagawa River system than in the Edogawa River. The mean DO was significantly lower in the Sakagawa River system. There were no significant differences between the mean pH and the mean nitrate nitrogen. The 75th percentile of BOD measured in the Edogawa River was 3.5 mg/l, and that measured in the Sakagawa River system was 18.9 mg/l. Neither

Table 1 Concentrations of nonylphenol (NP), 4-*t*-octylphenol (OP) and some indicators of water qualities in Edogawa River and Sakagawa River system

Indicator		Edogawa River (n=12)	Sakagawa River system (n=56)	
NP	μg/L	0.24 (0.12–0.18–0.35–0.38)	0.87 (0.29–0.74–1.5–9.5)	**
OP	μg/L	0.066 (<0.01–0.012–0.13–0.23)	0.19 (<0.01–0.090–0.43–1.7)	**
TP	mg/L	0.23±0.059 (0–0.52)	1.1±0.20 (0.18–7.9)	**
PO_4^{3-}-P	mg/L	0.030±0.0082 (0–0.080)	0.35±0.064 (0–3.5)	**
TN	mg/L	1.9±0.17 (0.66–2.5)	3.9±0.14 (2.3–7.6)	**
NH_4^+-N	mg/L	0.034±0.018 (0–0.18)	0.88±0.047 (0–2.1)	**
NO_2^--N	mg/L	0.037±0.015 (0–0.20)	0.21±0.028 (0–1.2)	**
NO_3^--N	mg/L	1.7±0.22 (0.47–2.5)	1.5±0.10 (0.32–3.3)	
BOD	mg/L	3.1±0.42 (1.5–6.8)	14±0.74 (4.2–23)	**
DO	mg/L	6.4±0.42 (4.5–8.5)	4.6±0.24 (1.3–9.2)	**
pH		7.5±0.95 (6.9–8.2)	7.4±0.036 (6.8–8.4)	
water temperature	°C	23.1±0.861 (16.1–27.0)	24.4±0.414 (17.5–29.0)	

The NP and OP concentrations are medians (min–25th percentile–75th percentile–max).
The values for the others are means±SE (min–max).
** p<0.01 by Mann-Whitney test.
NP=nonylphenol; OP=4-*t*-octylphenol; TP=total phosphorus; PO_4^{3-}-P=phosphoric ion; TN=total nitrogen; NO^{2-}-N=nitrite nitrogen; NO^{3-}-N=nitrate nitrogen; NH^{4+}-N=ammonia nitrogen; BOD=biochemical oxygen demand; DO=dissolved oxygen.

Table 2 Spearman's correlation coefficients of alkylphenol concentrations and indicators representing river water qualities in Edogawa River and Sakagawa River system

Indicator	Edogawa River (n=12)		Sakagawa River system (n=56)	
	NP	OP	NP	OP
NP	—	0.80**	—	0.61**
OP	0.80**	—	0.61**	—
TP	0.20	0.01	–0.06	–0.18
PO_4^{3-}-P	0.23	0.16	0.18	0.10
TN	0.86**	0.79**	–0.19	–0.23
NH_4^+-N	–0.08	–0.21	–0.23	–0.27*
NO_2^--N	–0.36	–0.31	0.09	0.04
NO_3^--N	0.86**	0.65*	0.11	0.36**
BOD	0.74**	0.44	0.24	0.27*
DO	–0.76**	–0.36	–0.15	–0.11
pH	0.36	0.51	–0.03	0.27*
water temperature	0.79**	0.70*	0.12	0.43**

* p<0.05, ** p<0.01.
NP=nonylphenol; OP=4-*t*-octylphenol; TP=total phosphorus; PO_4^{3-}-P=phosphoric ion; TN=total nitrogen; NO_2^--N=nitrite nitrogen; NO_3^--N=nitrate nitrogen; NH_4^+-N=ammonia nitrogen; BOD=biochemical oxygen demand; DO=dissolved oxygen.

value attained the environmental standards set by the Japanese Ministry of the Environment: not more than 2.0 mg/l for the Edogawa River (classified as category A in Japanese environmental standards) and not more than 10.0 mg/l for the Sakagawa River system (classified as category E).

Table 2 shows the correlation coefficients between AP concentrations and the values of general water quality indicators in the Edogawa River and Sakagawa River system. There was a significant positive correlation between NP and OP concentrations ($p<0.01$). Of the water quality indicators, BOD significantly positively correlated with NP concentration in the Edogawa River and with OP concentration in the Sakagawa River system. Nitrate nitrogen and water temperature significantly positively correlated with NP and OP concentrations in the Edogawa River and with OP concentration in the Sakagawa River system.

Table 3 shows NP and OP concentrations of the four categorized areas in the Sakagawa River system. NP and OP concentrations differed significantly between the four categorized areas. The NP and OP concentrations were highest in the samples from the nonindustrial area with an underdeveloped sewerage system. However, the NP and OP concentrations of the samples from the nonindustrial area with an underdeveloped sewerage system that receives water from the North-Chiba Water Conveyance Channel decreased to the same levels as those of the samples from the nonindustrial area with a developed sewerage system and the industrial area.

Discussion

This study demonstrated the distributions of APs in the Edogawa River and Sakagawa River system. NP and OP were detected from both the Edogawa River and the Sakagawa River system; the concentrations of both these compounds

Table 3 Comparison of nonylphenol (NP) and 4-*t*-octylphenol (OP) concentrations in four categorized areas in Sakagawa River system

Category of sampling area	n	NP (μg/L)		OP (μg/L)	
Nonindustrial area with developed sewerage system	27	0.86 (0.29–1.7)		0.16 (<0.01–1.7)	
Industrial area	5	0.81 (0.56–1.7)		0.14 (0.11–0.71)	
Nonindustrial area with underdeveloped sewerage system that receives water from North-Chiba Water Conveyance Channel	9	0.98 (0.35–1.8)	*	0.24 (<0.01–0.40)	**
Nonindustrial area with underdeveloped sewerage system	7	2.9 (0.55–9.5)		0.60 (0.22–1.6)	

The NP and OP concentrations are medians (min–max).
* $p<0.05$, ** $p<0.01$ in Kruskal Wallis test.

were significantly higher in the Sakagawa River system than in the Edogawa River. The NP and OP concentrations in the Sakagawa River system were highest in the water along the nonindustrial area with an underdeveloped sewerage system. The results indicate that endocrine-disrupting chemical (EDC) pollution in a river that is a source of the public drinking water is related to the inflow of urban wastewater into the river system.

It is generally accepted that 1.0 μg/l is an acceptable standard for NP for freshwater by taking into consideration the lowest observable effect level (LOEL) and a safety factor of 0.1 (2, 21–23). Twenty-two NP measurements of our samples from the Sakagawa River system (39.3% of the samples in the Sakagawa River system) exceeded 1.0 μg/l. The Japanese Ministry of the Environment reported that the predicted no effect concentrations (PNECs) of NP and OP on fish were 0.608 μg/l and 0.992 μg/l, respectively (24, 25). Forty-six and three measurements from our samples exceeded the PNECs of NP and OP, respectively. The NP and OP measurements clearly demonstrated widespread EDC pollution in the Edogawa River and Sakagawa River system.

A survey of major rivers in Japan conducted in 1999 by the Japan Environment Agency showed that the maximum concentration of NP detected was 4.6 μg/l, and that of OP detected was 0.61 μg/l among 124 samples (8). Two and seven samples respectively exceeded these values in our study.

In the Edogawa River, the detection rate of NP above the detection limit was 100%, and that of OP was 75.0%. These results indicate that the contamination by APs occurred in the river that is a source of public drinking water. The maximum concentration of NP detected in the Edogawa River in the survey by the Tokyo Metropolitan Government in 1999 was 0.1 μg/l (n=2) (26). Our measurements indicated a slightly higher NP concentration in the Edogawa River than that measured in the survey by the Tokyo Metropolitan Government. Although this difference might be due to factors related to sampling, the trend of increase in NP concentration could be possible if appropriate countermeasures are not taken.

To improve water quality in the Edogawa River, the Kogasaki River Purification Plant and Running Water Conservation Channel have been established (Fig. 1), and water gates at the confluence points between the Sakagawa River system and the Edogawa River have been controlled. As a result, since 1998, there has been no inflow of polluted water from the Sakagawa River system into the Edogawa River upstream of the intakes of the three water purification plants under normal-water-level conditions in the Sakagawa River system. These countermeasures have led to an improvement in water quality at the intakes of the three water purification plants (18, 27). However, inflow from the Sakagawa River system into the Edogawa River upstream of the intakes is still observed under high-water-level conditions in the Sakagawa River system, for example, after heavy rainfall. As long as the Edogawa River is a source of public drinking water, continuous monitoring of alkylphenols is necessary to provide safe drinking water.

The degree of variation in AP concentration in the Sakagawa River system was larger than that in the Edogawa River. Both NP and OP concentrations were highest in the samples from the nonindustrial area with an underdeveloped sewerage system. These results suggest that the NP and OP loads from the tributary rivers and streams in the area with an underdeveloped sewerage system were predominant in the Sakagawa River system. APs in the main rivers have been monitored annually since 1998 in Japan (3). Results of our study suggest that AP monitoring is necessary not only in main rivers, but also in tributary rivers and streams in urban areas.

The area with an underdeveloped sewerage system where higher NP and OP concentrations were observed was mainly located at the upper reaches of the Sakagawa River system and the concentrations decreased at the middle and lower reaches. These decreases can be explained as follows. The less polluted water from the North-Chiba Water Conveyance Channel has a dilution effect on the polluted water discharged from the upper reaches of the Sakagawa River system (17, 18), which has been clearly shown in this study. The sorption of NP and OP to the bed sediments was considered to be another cause of these decreases. River sediments are rich in organic matter, and NP and OP preferentially adsorb onto organic matter as a result of their lipophilicity (6, 28, 29). The reduction in the inflow of APs and APEs due to the development of the sewerage system at the middle and lower reaches is also presumed to play a role (18). The treated water in the Kogasaki River Purification Plant, which collects all the polluted water from the Sakagawa River system during normal-water-level, has been reinjected into the lower parts of the Sakagawa River system through the Running Water Conservation Channel since 1998 (27). This has also contributed to the improvement in water quality at the lower reaches of the Sakagawa River system. To improve water quality at the upper reaches of the Sakagawa River system, further development of the sewerage system as well as other river purification measures are necessary.

In relation to industries and AP concentration, the results for the Sakagawa River system indicate that the NP and OP concentrations of the samples from industrial area were similar to those of the samples from the nonindustrial area with an underdeveloped sewerage system that receives water from the North-Chiba Water Conveyance Channel and from the non-industrial area with a developed sewerage system. This means that the treated effluents from the factories located in the industrial area did not have significant effects on the NP and OP loads in the Sakagawa River system during our study. However, the small-scale cleaning industry and car washing industry are scattered along the Sakagawa River system (30). These industries could play a role in increasing AP concentration in the Sakagawa River system. The AP concentration reported from the Sumidagawa River, another river flowing through the greater Tokyo area, was considered to be related to the industries along the river (31, 32). Restriction on the industrial use of nonylphenol ethoxylates that are biodegraded to NP has already been applied in European countries and others on the basis of regulatory or voluntary initiatives (2, 21, 22). In Japan, restrictions on the use of APs and APEs as well as their substitutes have been promoted by some industries such as the soap, detergent, cleaning and car washing industries (24, 33). The feasible control of industrial sources of APs and APEs requires attention.

The contribution of household wastewater to the water pollution in the Sakagawa River system was estimated to be 65.9% (17). Possible sources of AP pollution in relation to household wastewater during our study period were household detergents, especially imported ones, commercial detergents used for family-run cleaning industries and restaurants, cosmetic products and agricultural chemicals for gardening (15, 33, 34). APEs contained in those products were considered as AP-causing compounds. Further investigations on the sources of APs in river waters would help develop appropriate countermeasures for controlling AP pollution in rivers running through urban areas.

Among the general water quality indicators in our study, the nitrate nitrogen level in the Edogawa River was similar to that in the Sakagawa River system although the levels of other nitrogen indicators in the Edogawa River were significantly lower than those in the Sakagawa River system. Nitrate is used mainly in inorganic fertilizers. The nitrate concentration in surface water can increase as a result of leaching or runoff from agricultural land or contamination from human or animal waste as a consequence of the oxidation of ammonia and similar sources (35). Agricultural land is predominant along the upper reaches of the Edogawa River. The following two factors are considered the reasons why the nitrate nitrogen level in the Edogawa river was similar to that in the Sakagawa River system in this study: 1) nitrate load from the upper reaches of the Edogawa River, and 2) nitrate formation by the oxidation of ammonia and similar sources owning to the fact that the Edogawa River had a more aerobic water condition than the Sakagawa River system.

There were significant correlations between AP concentration and some of the general water quality indicators. Water temperature had significant positive correlations with NP and OP concentrations in the Edogawa River and with OP concentration in the Sakagawa River. Isobe et al. reported that the NP concentration in the Sumidagawa River was higher in spring and summer than in autumn and winter (32). They discussed that a higher water temperature could promote the degradation of parent nonylphenol ethoxylates to produce lower ethoxymers and facilitate the biodegradation of nonylphenol monoethoxylate to NP in a benthic environment in the river and that the resuspension or desorption of APs generated in the bottom sediment is a considerable factor for the higher AP concentrations in the river. It seems that the same phenomenon could occur in our study area. Significant correlations between nitrate nitrogen and NP and OP concentrations in the Edogawa River, and OP concentration in the Sakagawa River suggest that nitrate and some fractions of NP, OP and their ethoxylates may be derived from agricultural land because nitrate is used in fertilizers and nonylphenol ethoxylates and octylphenol ethoxylates are discharged into the environment mainly through agricultural chemicals (34). Thomas et al. reported that NP has been identified from stream water in an agricultural area during a storm event (36). Significant correlations between BOD and NP concentration in the Edogawa River and OP concentration in the Sakagawa River suggest that NP, OP and their ethoxylates are released into the aquatic environment together with other biodegradable organic contaminants such as domestic wastewater, which is a main cause of the BOD load in our study area (17).

Interventions to improve the quality of drinking water provide significant health benefits. Source water protection is the first step in protecting drinking water quality. The planning and implementation of protection measures will require collaboration with many sectors, such as national and local authorities; agricultural, industrial and other commercial entities; and communities (35, 37). With respect to the control of water pollution by APs and APEs, a systematic monitoring system for these compounds, further development of a sewerage system and other river purification measures, the development of substitutes for them in the agricultural and industrial products, and risk communication to reduce the use of products containing them should be promoted through mutual cooperation of all sectors.

In summary, our study showed widespread AP pollution in a main river in the greater Tokyo area, which is a source of public drinking water, and its tributary rivers receiving urban wastewater. A systematic monitoring of APs in tributary rivers and streams as well as in main rivers will help control EDC pollution and protect the source of drinking water in urban areas.

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