Occurrence of Bisphenol A in Some Municipal Wastewater Treatment Plants' Effluents in Bangkok Region

Intira Pookpoosa, Ranjna Jindal, Daisy Morknoy and Kraichat Tantrakarnapa

***Abstract*—** Bisphenol A (BPA), implicated in endrocrine disruption, is used in the primary production of polycarbonate plastics and epoxy resins which are used in the plastics industry. Because of its large scale production and widespread usages, BPA is released into the environment through air, land, and water during manufacturing, processing, and leaching from end-of-life's treatment.

The world population is exposed to BPA due to its use in food and beverage packaging and storage containers. BPA leaches from plastics when exposed to heat or acidic environments. The concern over BPA has grown over the last few years; however, there is still much debate over the potential harmful effects of BPA. Because most of the current wastewater treatment plants (WWTPs) are not designed to treat emerging contaminants such as BPA, they can escape into the aquatic environment. This study aims at investigating the occurrence and fate of BPA in five municipal wastewater treatment plants (MWWTPs) in Bangkok region.

***Keywords*—** Bisphenol A, wastewater, Bangkok, HPLC.

I. Introduction

MANY of the thousands of anthropogenic chemicals currently released into the environment are endocrine-disrupting compounds. These are defined as exogenous chemicals or chemical mixtures that impact endocrine system structure or function and cause adverse effects [1]. Because of its large scale production and widespread usages, BPA is released into the environment through air, land, and water during manufacturing, processing, and leaching from end-of-life's treatment. As a chemical that has found widespread use for more than fifty years, BPA is now being tested extensively for its potential to cause biological harm to humans via contact with products made from BPA [1]-[3]. The concern over BPA has grown over the last few years; however, there is still much debate over the potential effects of BPA exposure. One issue is the lack of consistency in findings, and a number of studies contradict one another.

Bisphenol A (BPA) is an organic compound containing two equivalent phenol groups that was first synthesized in 1891. Many of the adverse effects that BPA has on human health were first accidentally discovered about 2 decades ago. In 1993, it was revealed that BPA leached from autoclaved polycarbonate flasks, leading to increased proliferation of breast cancer cells [4]. Exposure to BPA due to damaged polycarbonate cages led to disruption of meiosis in the oocyte of control female mice [5]. While the chemical companies are trying to protect their own interest, there are legitimate debates and differences of opinions over data interpretation and the potential toxicity of BPA. Even though the debate remains open, it is clear that BPA can act as an endocrine disruptor that can possibly disrupt a number of metabolic processes.

This study aims at investigating the occurrence and fate of BPA in five municipal wastewater treatment plants (MWWTPs) in Bangkok region. Results of the first sampling event conducted during 8-9 October, 2013 are presented in this paper.

II. Methodology

A. Site selection and sampling

At present, there are total of seven municipal wastewater treatment plants in and around Bangkok, Thailand. Only five full-scale MWWTPs were selected for this study: Rattanakosin (RK), Chong Non Si (CN), Din Daeng (DD), Nong Khaem (NK), and Thungkru (TK).

Locations of the MWWTPs are shown in Fig. 1. The basic information and the operating conditions of these plants are presented in Table I.

Grab samples were collected in the first sampling event in October 2013. For BPA measurement, amber glass bottles (500 ml) were used for collecting samples that were pre-rinsed several times in the laboratory with DI water, methanol and Milli-Q water, and were rinsed again with ambient water on site. Plastic bottles (1.5 L) were used for sampling to determine other conventional parameters. Samples were immediately placed on ice and brought to the laboratory within 8 h, and stored in a cold room (4 ^{o}C) until analysis. Analyses were carried out at the Environmental Engineering Laboratory, Mahidol University and the Environmental Research and Training Centre (ERTC) Laboratory.

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TABLE I
THE BASIC INFORMATION OF FIVE WASTEWATER TREATMENT PLANTS (WWTPS) [6]

WWTPs	Service Area (km^2)	Pipe length (km)	Population served (persons)	Capacity (m^3/d)	Treatment Area (m^2)	Outfall point	Treatment Process
Rattanakosin	4.1	16.3	76,000	40,000	6,683	Klong Banglampoo	Two-Stage Activated Sludge
Chong Non Si	28.5	51	580,000	200,000	32,000	Chaophraya River	Cyclic Activated Sludge System
Din daeng	37	64	1,080,000	350,000	27,200	Bueng Makkasan and Klong Sam Sen	Activated Sludge with Nutrients Removal
Nong Khaem	44	46	520,000	157,000	86,400	Klong Ratchaburana	Vertical Loop Reactor Activated Sludge
Thungkru	42	26	177,000	65,000	14,400	Klong Ban Jak	Vertical Loop Reactor Activated Sludge
Total	155.6	203.3	2,433,000	812,000	166,683		

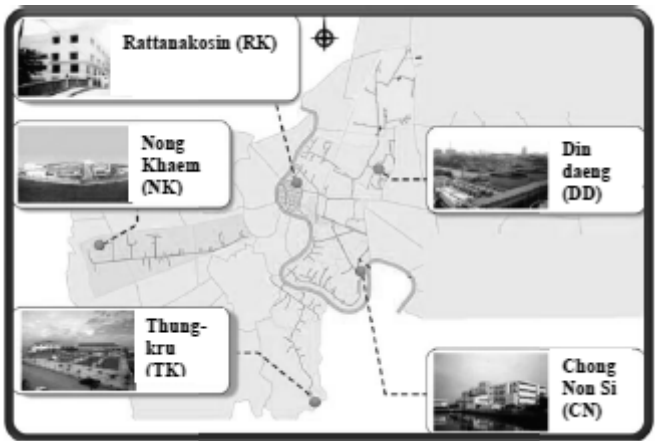

Fig. 1 The five selected wastewater treatment plants (WWTPs) Bangkok, Thailand

B. Samples preparation and analyses

Samples (500 ml) were first filtered with glass microfiber filter papers (Whatman, GF/B, 1.0 µm, Adelaide Co., Thailand). Solid-phase extraction (SPE) was done by using vacuum manifolds coupled with 6 cm^3, 500 mg, OASIS HLB cartridges (Waters-Millford, MA, USA) to concentrate the samples. The cartridges were conditioned with 10 mL of acetone, 10 mL of methanol, and 10 mL of Mili-Q water (+1% formic acid) in sequence. The flow rate of the equipment was maintained at approximately 3 mL/min. The cartridge washing was done twice with 6 mL of Mili-Q water at 5 min interval. Subsequently, the cartridges were allowed to dry under vacuum suction for 45 min. To extract BPA, the cartridges were rinsed with 2 mL of methanol (+1% formic acid) three time at 5 min intervals. A total of 6 ml of eluted fractions were collected in brown glass tubes and then evaporated under nitrogen gas. The dried residues were dissolved in 0.5 mL of methanol and were transferred to amber vials for HPLC analysis. Concentrated samples (25 µL) from the above procedure were injected into the HPLC for BPA measurement.

C. HPLC analysis

High performance liquid chromatography (HPLC) is an analytical technique in which a liquid mobile phase transports a sample through a column containing a liquid in stationary phase. The interaction of the sample with the stationary phase selectively retains individual compounds and permits separation of sample components. Detection of the separated sample compounds is achieved mainly though the use of absorbance detectors for organic compounds and through conductivity and electrochemical detectors for metal and inorganic components. HPLC has maximum efficiency at low flow rate because of typically slow diffusion rates between liquid phases.

In this study, all wastewater samples were analyzed using HPLC performed on a Shimadzu model LC-20AB equipped with a Shimadzu model RF-20A fluorescence detector, and a C18 reversed-phase column (Luna 5u, 250mm×4.6mm, phenomenex U.S.A). The Shimadzu model equipment (Shimadzu Corporation, Japan) consist of a communication bus module, a column oven and an auto sampler. The column oven temperature was held at 30^oC, fluorescence detection was carried out using 228 nm as the excitation and 313 nm as the emission wavelengths. An external calibration method used the calibration curve in the concentration range of 10-5000 ugL^{-1}. The limit of detection (LOD) and limit of quantification (LOQ) were defined as the concentration with a

signal-to-noise (S/N) ratio of 3 and 10, respectively. LOD and LOQ were 1.3 and 4.2 ugL^{-1}, respectively.

III. Results and Discussion

A. Influent and Effluent Water Quality

The measured conventional parameters for influents and effluents of the five MWWTPs are shown in Table II. All of these parameters in the effluents were within the domestic wastewater standards of Thailand. Base on the population served by the five MWWTPs as shown in Table I, the largest one is DD (population served 1,080,000 persons). However, the highest values for EC, TS, TSS, TDS, and BOD_5 were observed to be of the influent to NK (third ranking with respect to the size of population served). It could be due to fact that NK also receives the sludge of all the seven MWWTPs operated under Bangkok Metropolitan Administration (BMA). The nearest point where solid waste from the surrounding municipal area is collected is the Nong Khaem Solid Waste Disposal Center before sending to landfill. The runoff from this facility is also treated together with influent wastewater at NK. Therefore, all the influent water quality parameters of NK are at highest levels, especially in rainy season. Almost all the influent parameters of RK, such as EC, TS, and TDS were lowest as it is the smallest among the five MWWTPs and has the lowest size of the population served (76,000 persons).

The highest effluent parameters such as EC and TS were measured to be also of NK. Because of the highest influent values, the effluent parameters were also still high. However, the removal values for TS, TSS, TDS, and BOD_5 of NK were higher than the others. Although, both NK and TK use the Vertical Loop Reactor Activated Sludge for the treatment system. NK has the capacity and the treatment area higher than TK, thus the efficiency of NK is higher than it.

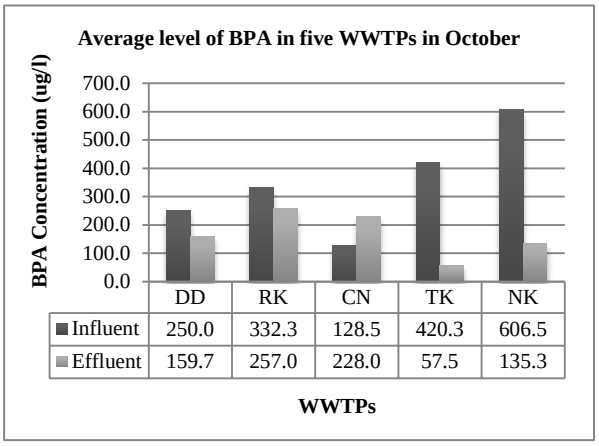

Fig. 2 Average level of BPA in five WWTPs in October, 2013

B. Occurrence of BPA

Fluorescence detection is the most frequently used non-MS-based method for LC determination of BPA because the fluorophore in the BPA molecule is fairly strong [7]. The BPA concentrations in the influents and effluents of five MWWTPs are shown in Table II and Fig. 2.

The influent concentrations ranged between 128.5 ngL^{-1} and 606.5 ngL^{-1}, and the BPA levels in the effluents ranged between 57.5 ngL^{-1} and 257.0 ngL^{-1}. Even though these WWTPs are not designed to remove BPA from wastewater, the effluent BPA concentrations in most of the five MWWTPs were lower than the influent levels except in CN which had the lowest influent concentration (128.5 ngL^{-1}). The highest influent BPA concentration was found to be 606.5 ngL^{-1} at NK.

The trend of influent BPA concentrations among the five MWWTPs was similar to the levels of the conventional parameters. The influent BPA concentration appeared to be

TABLE II
THE CONVENTIONAL PARAMETERS AND BPA CONCENTRATION OF FIVE WASTEWATER TREATMENT PLANTS (WWTPS) IN OCTOBER, 2013

Parameter	Din daeng (DD)		Rattanakosin (RK)		Chong Non Si (CN)		Thungkru (TK)		Nong Khaem (NK)	
	Influent	Effluent	Influent	Effluent	Influent	Effluent	Influent	Effluent	Influent	Effluent
DO (mgL^{-1})	6.50	8.90	3.80	8.60	6.00	7.20	5.70	8.60	3.10	6.63
pH	7.39	7.41	7.47	7.38	7.30	7.19	7.29	7.46	7.16	7.28
EC (uS/cm)	577	455	539	393	704	567	729	668	782	720
TS (mgL^{-1})	346.22	321.56	340.44	314.00	479.56	434.67	382.44	369.56	511.78	448.67
TDS (mgL^{-1})	343.87	294.93	308.53	302.80	347.87	327.73	427.33	410.80	484.40	379.73
TSS (mgL^{-1})	20.20	8.53	14.80	11.07	15.20	13.20	5.07	4.40	53.87	3.47
BOD_5 (mgL^{-1})	51.0	3.8	61.0	1.1	45.0	8.4	37.0	1.6	67.0	4.6
Temp. (^{o}C)	28.5	28.3	29.1	28.8	28.3	28.3	28.8	29.3	28.5	29.0
BPA Concentration (ngL^{-1})	250.0	159.7	332.3	257.0	128.5	228.0	420.3	57.5	606.5	135.3

Significantly correlated to DO level (R^2 = -0.7489). Thus, high BPA concentrations were observed in influent samples with low DO levels. It was reported in a study that BPA degradation in river waters could be under aerobic conditions [8].

C. BPA Removal in MWWTPs

BPA removal efficiencies of DD, RK, CN, TK and NK were 36.13%, 22.67%, -77.43%, 86.32%, and 77.69%, respectively.

The highest BPA removal efficiency was of TK followed by NK. The treatment process employed at both the WWTPs was vertical loop reactor activated sludge process. Thus, this process seems to be the best for BPA degradation. On the other hand, BPA concentrations in the effluents of CN were higher than its influent levels. It could probably be due to the treatment process employed at CN which might not be able to remove BPA. However, the results presented in this paper are only based on the first sampling event. More sampling and investigations are required to have a better picture.

D. Risk of BPA Occurrence in Aquatic Environments

Wastewater effluents containing BPA can be a source of contamination of the aquatic environment of receiving water bodies. Because BPA contaminate in aquatic environment, it also can contaminate aquatic organism. As indicated in a recent study, BPA is the most documented chemical reported among the phenolic compound in surface waters around the world [9]. Apart from its ocurrence in aquatic environments, BPA has also been found in meats of freshwater fish and seawater fishes [10]. Fishes are at the top of the aquatic ecological food chain as well as at the top of the consumers 'food products' list. Hence, they are also at the top of the sources of BPA contamination with bioaccumulations in their bodies. Fishes live in the contaminated water and consume BPA though their food. Moreover, BPA in seawater than in river water can continue for longer time with no degradation and the possibility of BPA contamination on a marine organism can be higher than that on freshwater organism [8], [11]. Although it has been reported that BPA is biodegradable, the leaching of BPA from plastic products, effluents from wastewater treatment plants and landfills contribute significant amounts of BPA in the freshwater systems. Human can get BPA into their systems though consuming not only these contaminated fish meats but also other aquatic species (such as shell, shrimp, and crab).

IV. Conclusions

Based on the results of the first sampling, it could be seen that BPA was presented in majority of the influent and effluent samples of the five MWWTPs being investigated in this study. The influent concentrations ranged between 128.5 ng L^{-1} and 606.5 ng L^{-1}, and the BPA levels in the effluents ranged between 57.5 ng L^{-1} and 257.0 ngL^{-1}. Influent samples having high level of EC, TSS, and BOD_5 showed high BPA concentrations. Even though these MWWTPs are not designed to remove BPA from wastewater, the effluent BPA concentrations in most of the five MWWTPs were lower than the influent levels. However, as these results are based on only one of the several sampling events to be conducted, more investigations are required to have the complete picture regarding the situation of occurrence of Bisphenol A in the municipal wastewater treatment plants' effluents in Bangkok region.

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References

[1] S. Flint, T. Markle, S. Thompson and E. Wallace, "Bisphenol A exposure, effects, and policy: A wildlife perspective," Journal of Environmental Management, no. 104, pp. 19-34, 2012. http://dx.doi.org/10.1016/j.jenvman.2012.03.021

[2] D. A. Crain, M. Eriksen, T. Iguchi, S. Jobling, H. Laufer, G. A. LeBlanc and L. J. Guilleyye Jr., "An ecological assessment of bisphenol-A: Evidence from comparative biology," Reproductive Toxicology, vol. 24, pp. 225-239, 2007. http://dx.doi.org/10.1016/j.reprotox.2007.05.008

[3] V. Santhi, N. Sakai, E. Ahmas and A. Mustafa, "Occurrence of bisphenol A in surface water, drinking water and plasma from Malaysia with exposure assessment from consumption of drinking water," Science of the Total Environment, Vols. 427-428, pp. 332-338, 2012. http://dx.doi.org/10.1016/j.scitotenv.2012.04.041

[4] L. N. Vandenberg, R. Hauser, M. Marcus, N. Olea and W. V. Welshons, "Human Exposure to Bisphenol A," Reprod Toxicol., vol. 24, no. 2, pp. 77-139, 2007. http://dx.doi.org/10.1016/j.reprotox.2007.07.010

[5] P. A. Hunt, K. E. Koehler, M. Susiarjo, C. A. Hodges, A. Ilagan, R. C. Voigt, S. Thomas, B. F. Thomas and T. J. Hassold, "Bisphenol A Exposure Causes Meiotic Aneuploidy in the Female Mouse," Current Biology, vol. 13, pp. 546-553, 2003 http://dx.doi.org/10.1016/S0960-9822(03)00189-1

[6] Water Quality Management Office under Drainage and Sewerage Department(DDS), BMA, "Water Quality Management and Remediation," 2008. [Online]. Available: http://dds.bangkok.go.th/wqm/English/inplementation.html. [Accessed 2 June 2012].

[7] X. Cao, "Backgroung Paper on Chemistry and Analytical Methods for Determination of Bisphenol A Food and Biological Samples," in FAO/WHO Expert Meeting on Bisphenol A (BPA), Ottawa, Canada, 2010.

[8] J.-H. Kang and F. Kondo, "Bisphenol A degradation in seawater is different from that in river water," Chenosphere, vol. 60, pp. 1288-1292, 2005. http://dx.doi.org/10.1016/j.chemosphere.2005.01.058

[9] K. K. Selvaraj, G. Shanmugam, S. Sampath, D. J. Larsson and B. R. Ramaswamy, "GC–MS determination of bisphenol A and alkylphenol ethoxylates in river water from India and their ecotoxicological risk assessment," Ecotoxicology and Environmental Safety, vol. 99, p. 13–20, 2013. http://dx.doi.org/10.1016/j.ecoenv.2013.09.006

[10] X. Wei, Y. Huang, M. H. Wong, J. P. Giesy and C. K. Wong, "Assessment of risk to humans of bisphenol A in marine and freshwater fish from Pearl River Delta, China," Chemosphere, vol. 85, no. 1, p. 122–128, 2011. http://dx.doi.org/10.1016/j.chemosphere.2011.05.038

[11] A. Belfroid, M. van Velzen, B. van der Horst and D. Vethaak, "Occurrence of bisphenol A in surface water and uptake in fish: evaluation of field measurements," Chemosphere, vol. 49, p. 97–103, 2002.
http://dx.doi.org/10.1016/S0045-6535(02)00157-1

[12] A. B. Bailey and E. J. Hoekstra, "Backgroung Paper on Sources and Occurrence of Bisphenol A Relevant for Exposure of Consumers," in FAO/WHO Expert Meeting on Bisphenol A (BPA), Ottawa, Canada, 2010.

[13] C. A. Staples, P. B. Dom, G. M. Klecka, S. T. O'Block and L. R. Harris, "A Review of The Environmental Fate, Effects, and Exposures of Bisphenol A," Chemosphere, vol. 36, no. 10, pp. 2149-2173, 1998.
http://dx.doi.org/10.1016/S0045-6535(97)10133-3

[14] M. Fürhacker, S. Scharf and H. Weber, "Bisphenol A: emissions from point sources," Chemosphere, vol. 41, pp. 751-756, 2000.
http://dx.doi.org/10.1016/S0045-6535(99)00466-X

[15] D. P. Mohapatra, S. K. Brar, R. D. Tyagi and R. Y. Surampalli, "Occurrence of bisphenol A in wastewater and wastewater sludge of CUQ treatment plant," Journal of Xenobiotics, vol. 1, no. 3, 2011.

[16] A. S. Stasinakis, G. Gatidou, D. Mamais, N. S. Thomaidis and T. D. Lekkas, "Occurrence and fate of endocrine disrupters in Greek sewage treatment plants," Water Research, vol. 42, p. 1796–1804, 2008.
http://dx.doi.org/10.1016/j.watres.2007.11.003

[17] N. Kuroda, Y. Kinoshita, Y. Sun, M. Wada, N. Kishikawa, K. Nakashima, T. Makino and H. Nakazawa, "Measurement of bisphenol A levels in human blood serum and ascitic fluid by HPLC using a fluorescent labeling reagent," Journal of Pharmaceutical and Biomedical Analysis, vol. 30, no. 6, p. 1743–1749, 2003..
http://dx.doi.org/10.1016/S0731-7085(02)00516-2