Screening analysis of 1170 organic micro-pollutants in sewage treatment plants in Kitakyushu, Japan

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Introduction: Around 100,000 man-made chemicals are currently used to facilitate modern lifestyles. Some of these chemicals are harmful and most chemicals are discharged into the aquatic environment after being used, so it is necessary to monitor the concentrations of these chemicals in sewage treatment plant (STP) influents and effluents to gain an understanding of the pollution statuses of aquatic systems and to protect human and ecosystem health. Kitakyushu is an industrial city with a population of one million in southern Japan, and it contains five STPs that use the activated sludge treatment method. These STPs treat 99.8% of the domestic wastewater produced in Kitakyushu. The objectives of this study were to determine the types and quantities of chemicals that are discharged in domestic wastewater in Kitakyushu and enter the aquatic environment from the STPs. To achieve this, we analysed 1170 chemicals in the influents and effluents of the STPs in Kitakyushu using two comprehensive analytical methods, gas chromatography-mass spectrometry (GC-MS) and liquid chromatography-time of flight mass spectrometry (LC-TOF-MS).

Experimental: A water sample was taken from each of three points (after the grit chamber (influent), after the first settling tank, and after the final settling tank (effluent)) in each STP on 29 October 2014. A 1-L aliquot of each sample was spiked with surrogate standards and liquid–liquid extracted with dichloromethane. The extract was dehydrated, concentrated, and then spiked with internal standards [1]. The extract was then analysed by GC-MS using the selected ion monitoring and scanning modes, and 900 target semi-volatile organic compounds were identified and quantified using the AIQS system [2]. A further 200-mL aliquot of each raw water sample was spiked with surrogate standards, filtered, and solid-phase extracted using Waters PS-2 and AC-2 cartridges. The eluate was concentrated and an internal standard was added, then the extract was analysed for 300 polar chemicals by LC-TOF-MS [3].

Results and Discussion

Detected compounds and concentrations: The numbers of chemicals that were detected and their concentrations are shown in Table 1. Out of the 1170 substances analysed, 133 were found at least once. A lot of pharmaceuticals and personal care products (PPCPs) and phenols were detected at high concentrations, and ubiquitous substances (such as *n*-alkanes, phthalates, antioxidants and their degradation products, degradation products of

Table 1 Concentrations (μg/L) of the chemicals found and the numbers of chemicals found (in parentheses)

Target substance	Influent	Effluents
Chemicals containing C and H only (190)	36 (44)	0.39 (28)
Chemicals containing C, H, and O only (159)	532 (37)	4.3 (22)
Chemicals containing N (118)	1.8 (3)	0.54 (4)
Chemicals containing S (12)	0.29 (2)	0.48 (3)
Chemicals containing P (9)	0.47 (3)	0.27 (3)
Pharmaceuticals and personal care products (108)	95 (30)	11 (24)
Pesticides (574)	0.11 (5)	0.11 (6)
Total (1170)	665 (124)	17 (90)

vulcanization accelerators and PAHs) and sterols found in human faeces and kitchen waste were also found. Organophosphorus flame retardants and pesticides were also detected at low concentrations. Most of these chemicals have also been detected in the aquatic environment in Japan [4]. The influent volumes and the concentrations of the chemicals that were found in the samples collected after the grid chambers were used to calculate the total amounts of the chemicals in the STP influents. The total amounts of the detected substances and the PPCPs in the influents of the five STPs were 279.5 and 42.2 kg/d, respectively, which equate to 279.5 and 42.2 mg/(capita d), respectively. The five prescription drugs that were found at the highest concentrations in the influents and effluents are shown in Fig. 1. These drugs appear to be used in large amounts in Japan because they have frequently been found in Japanese wastewater systems [5].

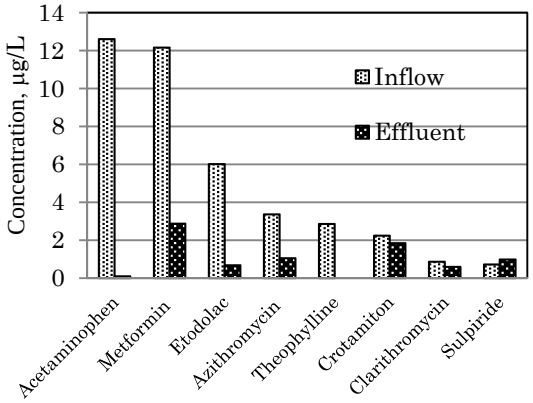

Fig. 1 Five drugs at the highest concentration

Removal of the detected substances: It has been shown that 99% or more of the biological oxygen demand is removed between the first and final settling tanks because of components being biodegraded and adsorbed onto the sludge. We compared the concentrations of the chemicals that were detected after the grit chamber and after the final settling tank, and we found that 97.4% of the chemicals were removed. This is almost the same as the removal ratio for the biological oxygen demand. However, the chemicals were separated into three groups, depending on the changes in their concentrations during the treatment process. These groups were the easily degradable (removable) chemicals, the persistent chemicals, and chemicals that were produced during the treatment processes. Natural materials, sterols, phenols, and phthalates were easily removed because they are highly biodegradable. Some of the drugs, pesticides, and organophosphorus flame retardants were not effectively removed by the activated sludge. The concentrations of 2,6-di-*tert*-butyl-4-benzoquinone, anthraquinone, 2(3H)-benzothiazolone, and 1,3-dicyclohexylurea were higher in the effluent than in the influent, so these substances appeared to be products of the degradation of other chemicals. The PPCPs contributed only 15% of the total chemical concentrations in the influents, but they were found at relatively high concentrations in the effluents and contributed 64% of the total chemical concentrations in the effluents. This indicates that we must be concerned about the potential effects of PPCPs on the aquatic environment.

References

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