Distribution of Pharmaceuticals in Clinical Wastewater from Medical Institutions and Development of Advanced Water Treatment Systems

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Scope of the lecture:

This lecture scopes for new environmental pollution problems in the water environment by pharmaceuticals and the authors want to show our recent findings for their occurrence and effectiveness of advanced water treatment system for reduce the pollution load to the water environment.

Learning objectives:

1. To learn the back ground and present status of pharmaceuticals in the water environment
2. Hospital effluent are major loading source for river in some pharmaceuticals.
3. Introduction of advanced water treatment system is efficient to decrease pollution load to the water environment.

Extended abstract:

Introduction

In recent years, the environmental pollution problems originated from pharmaceuticals have begun to receive an increasing amount of attention worldwide (Ferrando-Climent et al., 2014; Verlicchi et al., 2015).

Pharmaceuticals are designed to have specific physiological effects on targeted areas of the body. After ingestion their metabolites and remained unchanged are, however, discharged from the body due to their high polarity (Kolpin et al., 2002). Since more than 90% of urban areas are covered with sewerage systems in Japan (Japan Sewage Works Association 2015), both domestic waste and hospital effluent are transferred into sewerage systems and treated in sewage treatment plants (STPs) (Azuma et al., 2012). In addition, pharmaceuticals are constantly used in hospitals, and the quantity of pharmaceuticals used in Japan is the second largest in the world. Despite this situation, their occurrence in hospital effluent is largely unknown.

In this study, we conducted survey of pharmaceuticals in the hospital effluent together with influent and effluent of the STP which treats the wastewater from the targeted hospital, and river water which include the treated water. All sampling points are located in an urban area of Japan.

Material and Methods

Clinical wastewater from hospital effluent, STP influent and effluent were collected in glass bottles. The survey was conducted once in four different seasons from 2014 to 2017. Forty one pharmaceuticals were selected for analysis based on the previously detected level and frequency of detection in STPs and river water (Azuma et al., 2015). All analytical standards were of high purity (> 98%). Concentrations of targeted pharmaceuticals were determined by the multiresidue analytical method for multiple pharmaceuticals in environmental water samples based on the combination of solid phase extraction (SPE) and liquid chromatography-tandem mass spectrometry (LC-MS/MS) (Azuma et al., 2015).

Results and Discussion

Concentrations of pharmaceuticals surveyed in the hospital effluent, STP influent and effluent, and river water are shown in Figure 1 (Azuma et al., 2016a). Thirty eight compounds were detected in hospital effluent over a wide concentration range from ng/L to μg/L, with the highest at 92 μg/L. The overall orders of the pharmaceuticals detected in the hospital effluent are similar to those detected in the STP influent. This suggests similarity in the usage of the pharmaceuticals in hospital and household. However, three different profiles could be observed in the individual concentrations: one group includes antibacterials such as ciprofloxacin, anticancers such as cyclophosphamide and doxifluridine, analgesic-antipyretic compounds such as acetaminophen whose concentrations in the hospital effluent are 2 to 10 times higher than those detected in the STP influent; anticancers such as tegafur, antipruritic crotaminon, antivirals such as aciclovir, and psychotropics such as sulpiride are included in the second group whose concentration in the hospital effluent was 10 times lower than those in the STP influent; and the last group which includes remainders having similar profile.

The concentration ranges of pharmaceuticals detected in the hospital effluent and STP influent were also similar to those in the STP effluent. These results agreed with recalcitrant property of many pharmaceuticals against conventional water treatment system centered on biological treatment (Evgenidou et al., 2015). However, after additional ozone treatment at STP, mean concentrations of all targeted pharmaceuticals decreased to the range from N.D. to several tens of ng/L, which were roughly one-tenth to one-hundredth of the concentrations detected in STP effluent with chlorination after biological treatment. These results suggest that advanced water treatment system is effective for reduction of the pollution load to the water environment.

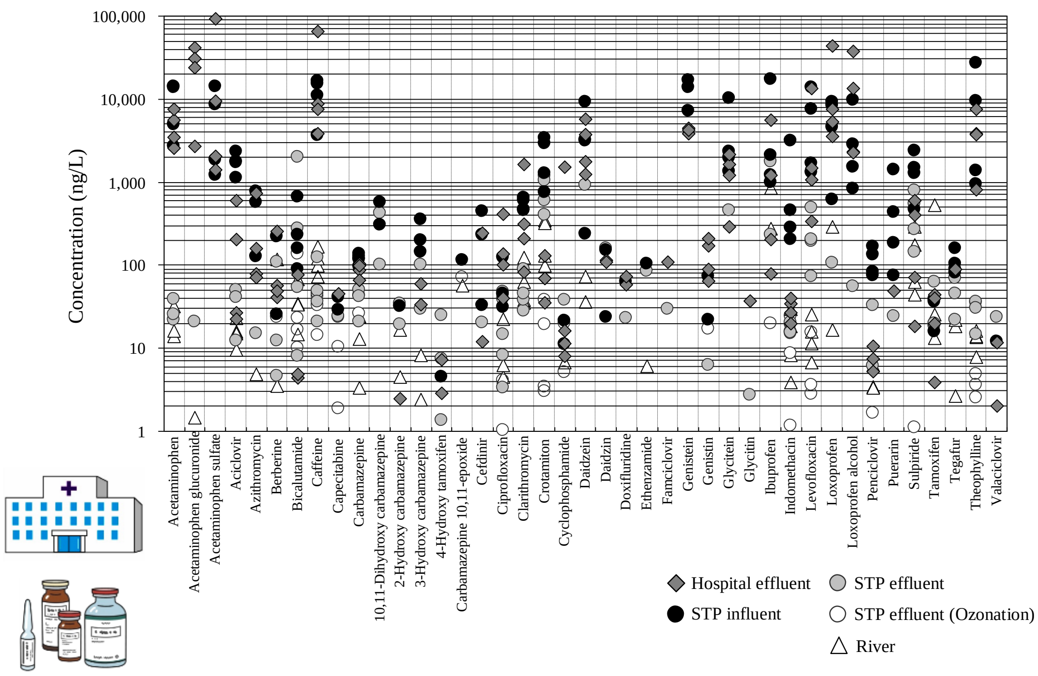

Figure 1. Distribution of pharmaceuticals in hospital effluent, STP influent and effluent, and river water (Azuma et al., 2016a).

Therapeutic group dependent contribution in the total detected concentration of pharmaceuticals was calculated and results are shown in Figure 2. In the case of the hospital effluent, 45% was occupied by antibacterials, followed by 30% occupation by analgesic-antipyretic compounds. The corresponding values of antiviral, phychotropic, and antipruritic compounds were, however, quite low as between 0.2% to 5%. In the STP influent, as high as 76% was occupied by analgesic-antipyretic (42%) and bronchodilator (34%) compounds. In contrast, degree of contribution of antiviral, anticancer, phychotropic, and antipruritic compounds were found to be quite low as ranging from 0.3% to 3%. The summed-up degree of contribution of the anticancer, phychotropic, analgesic-antipyretic, and antipruritic compounds in the STP effluent became 4% to 15% higher than the STP influent, meaning that these chemicals were recalcitrant against wastewater treatment in STPs and survived after treatment.

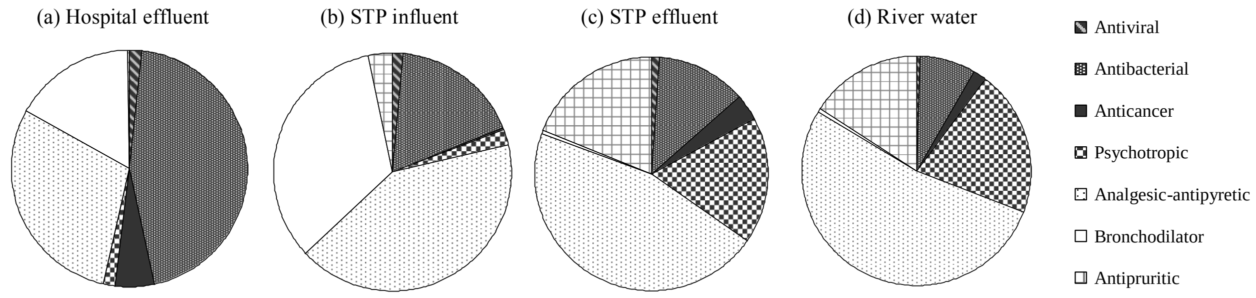

Figure 2. Contribution of pharmaceuticals in hospital effluent, STP influent and effluent, and river water (Azuma et al., 2016b).

Next, effectiveness of the advanced water treatment systems for removal of pharmaceuticals in water samples was analyzed. We applied UV, H_2O_2 (hydrogen peroxide), and ozone based treatments in addition to biological treatment. The results obtained as removal efficiencies for all water treatment systems are summarized in Table 1.

In biological treatment, higher efficiency than 90% was obtained for the third group of compounds, but many pharmaceuticals were resistant for treatment; the removal rate of carbamazepine, cyclophosphamide, ethenzamide, iomeprol, iopamidol, indomethacin, olmesartan, and tegafur were less than 20% (Azuma et al., 2016b). These results were largely in agreement with that of the field survey.

Table 1. Removal efficiency of pharmaceuticals in various forms of water treatment systems.

Therapeutic class	Treatment system							
	Biological treatment	UV	H_2O_2	UV/H_2O_2	O_3	O_3/UV	O_3/H_2O_2	$O_3/UV/H_2O_2$
Antiviral	△~○	×~△	×	△~○	○~◎	○~◎	○~◎	○~◎
Antibacterial	×~△	△~○	×~△	○~◎	○~◎	◎	◎	◎
Anticancer	△~◎	○~◎	×	△~○	○~◎	◎	◎	○~◎
Psychotropic	×~△	×~△	×	△~○	△~◎	○~◎	△~◎	○~◎
Analgesic-antipyretic	○~◎	△~○	×	○	○~◎	○~◎	○~◎	○~◎
Bronchodilator	×~△	×~△	×	○	◎	◎	◎	◎
Antipruritic	×~△	○	×	○	◎	◎	◎	◎
Herbal medicine	◎	△	×~△	○	◎	◎	◎	◎
Phytoestrogen	◎	△	×	○~◎	◎	◎	◎	◎

(×: Persistent; △: Gradually degradable; ○: Degradable; ◎: ≤ Readily degradable)

Similar tendency was observed for the UV treatment. Almost all of the compounds were recalcitrant to hydrogen peroxide. However, by combination of hydrogen peroxide with UV, removal efficiency was significantly improved (Azuma, 2017). These results suggest that contribution of active oxygen species such as hydroxyl radical which was generated by UV-induced decomposition of hydrogen peroxide (Keen et al., 2012).

Higher removal rates were obtained for all ozone treatments; most of the pharmaceuticals were removed within several minutes to 10 minutes. In addition, additional improvement of removal rates were achieved by combination of ozone and UV and/or hydrogen peroxide, as well as in the case of hydrogen peroxide and UV (Azuma, 2017).

This research indicated the present status of occurrence of pharmaceuticals in the hospital effluent and effectiveness of introduction of advanced water treatment systems not only for STPs but also for hospital effluent for reduction of pharmaceuticals in wastewaters. Further research for expanding target hospitals together with the measures for concerns about pathogenic microbes are expected to proceed in future.

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