Environment International 115 (2018) 89–96

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Select antibiotics in leachate from closed and active landfills exceed thresholds for antibiotic resistance development

S.S. Chung[a,*], J.S. Zheng[a], S.R. Burket[b], B.W. Brooks[b,c]

[a] Department of Biology, Hong Kong Baptist University, Kowloon Tong, Kowloon, Hong Kong, China
[b] Department of Environmental Science, Center for Reservoir and Aquatic Systems Research, Baylor University, Waco, TX, USA
[c] Institute of Biomedical Studies, Baylor University, Waco, TX, USA

ARTICLE INFO

Keywords:
Urbanization
Antibiotic resistance
Predicted no effect concentration
Landfill leachate
Hong Kong

ABSTRACT

Though antibiotic resistance (ABR) represents a major global health threat, contributions of landfill leachate to the life cycle of antibiotics and ABR development are poorly understood in rapidly urbanizing regions of developing countries. We selected one of the largest active landfills in Asia and two landfills that have been closed for 20 years to examine antibiotic occurrences in leachates and associated hazards during wet and dry season sampling events. We focused on some of the most commonly used human antibiotics in Hong Kong, one of the most populous Asian cities and the fourth most densely populated cities in the world. Seven antibiotics (cephalexin [CLX], chloramphenicol [CAP], ciprofloxacin [CIP], erythromycin [ERY], roxithromycin [ROX], trimethoprim [TMP], sulfamethoxazole [SMX]) were quantitated using HPLC-MS/MS generally following previously reported methods. Whereas CLX, CAP, ROX and SMX in leachates did not exceed ABR predicted no effect concentrations (PNECs), exceedances were observed for CIP, ERY and TMP in some study locations and on some dates. In fact, an ABR PNEC for CIP was exceeded in leachates during both sampling periods from all study locations, including leachates that are directly discharged to coastal systems. These findings highlight the importance of developing an advanced understanding of pharmaceutical access, usage and disposal practices, effectiveness of intervention strategies (e.g., leachate treatment technologies, drug take-back schemes), and contributions of landfill leachates to the life cycle of antibiotics and ABR development, particularly in rapidly urbanizing coastal regions with less advanced waste management systems than Hong Kong.

1. Introduction

By 2050 the global population is predicted to reach 9.8 billion people, many of which will be concentrated in Asia. Whereas Asia contributed 55% to the global population in 2014, 22 megacities are expected in Asia by 2030. Higher population densities are also observed in this region; for example, Hong Kong is presently the fourth most densely populated city in the world. Such population densities correspondingly result in a concentration of urban consumption of natural resources, including food, energy and water, and consumer products. In fact, in many urban areas of Asia and other regions, access to and consumption of consumer chemicals, including medicines, are occurring more rapidly than implementation of public health and environmental interventions, including advanced wastewater and solid waste management (Brooks, 2018). This couples with dramatically variable standards of waste management practices and treatment technologies among developed and developing regions, differential implementation and protection of environmental quality from risks posed by contaminants of historical and emerging concern is inevitable.

In recent years, pharmaceuticals have received attention as contaminants of emerging concern because they represent indicators of an urbanizing environment and water cycle (Brooks, 2014). Antibiotics, in particular, have been examined as environmental contaminants due to associated risks presented to aquatic and terrestrial ecosystems (Brooks et al., 2008). For example, Brain et al. (2008) examined adverse effects of antibiotics to aquatic plants and algae, and identified likely sensitive species resulting from increased evolutionary conservation of therapeutic targets for a number of antibiotic classes. Antibiotic occurrence in the environment has also received heightened attention due to influences on the development of antibiotic resistance (ABR), which is

Abbreviations: CLX, cephalexin; CAP, chloramphenicol; CIP, ciprofloxacin; ERY, erythromycin; ROX, roxithromycin; TMP, trimethoprim; SMX, sulfamethoxazole; PNECs, predicted no effect concentrations; ABR, antibiotic resistance; EPD, Environmental Protection Department; SW, Shuen Wan; PPV, Pillar Point Valley; WENT, West New Territories Landfill; SPE, solid phase extraction

* Corresponding author at: SCT 703, Cha Chi-Ming Science Tower, Ho Sin Hang Campus, 224 Waterloo Road, Kowloon, Hong Kong, China.
E-mail address: sschung@hkbu.edu.hk (S.S. Chung).

https://doi.org/10.1016/j.envint.2018.03.014
Received 8 January 2018; Received in revised form 7 March 2018; Accepted 11 March 2018
0160-4120/

now spreading (Qiao et al., 2018) and represents a leading threat to global public health (World Health Organization, 2015). For example, influences of antibiotics in the environment on the development of ABR microorganisms was recently identified as a priority global research need (Boxall et al., 2012; Rudd et al., 2014).

Numerous studies have reported the environmental introduction of antibiotics and other pharmaceuticals from manufacturing processes (Larsson, 2014), and following human use through discharge of municipal wastewater and land application of biosolids, and veterinary use in terrestrial agriculture and aquaculture operations (Qiao et al., 2018). Antibiotics and other medicines may also enter the environment from solid waste management processes, which has been the subject of several studies. For example, Holm et al. (1995) observed five sulfonamide compounds from 40 to 1600 μg/l in ground water influenced by an unlined landfill in Denmark. Masoner et al. (2016a, 2016b) found that contaminants of emerging concern, including antibiotics, were commonly found in raw and final leachate in US landfills. Wu et al. (2015) examined levels of 20 antibiotics in the leachate from two refuse transfer stations and one landfill site in Shanghai. Despite relatively elevated concentrations for all reported samples, these levels were generally higher in leachate from transfer stations than the landfill. Regarding the relationship between the age of the landfill and concentration of antibiotics in associated leachate plumbs, Wu et al. (2017) found that with the exception of sulfamethazine, lower levels were observed from older landfills. Whereas such findings appeared to conflict with a conclusion by Yu et al. (2016) that the levels of sulfapyridine and sulfadiazine increased with the age of the landfill, it was in fact not. This was because the oldest landfill studied by Yu et al. (2016) was just 6 years in age, which would be considered a middle-aged landfill by Wu et al. (2017). Beyond these recent studies, the environmental occurrence and risks of pharmaceuticals in general (Kristofco and Brooks, 2017; Saari et al., 2017) and antibiotics in particular are not well studied in many parts of Asia (see Table 1). Further, how landfill leachate, an important type of wastewater effluent, but highly variable in its compositions, contributes to the life cycle of antibiotics is still largely not understood.

In the present study, we selected Hong Kong, one of the most populous cities in Asia, for an initial study to determine whether commonly used human antibiotics (cephalexin [CLX], chloramphenicol [CAP], ciprofloxacin [CIP], erythromycin [ERY], roxithromycin [ROX], trimethoprim [TMP], and sulfamethoxazole [SMX]) were present in and/or differed among leachates of closed and active landfills during wet and dry seasons. Specifically, the Shuen Wan (SW) and Pillar Point Valley (PPV) landfills were selected because these restored facilities were closed in 1995 and 1996, respectively, whereas we studied the West New Territories Landfill (WENT) because it represented one of the largest active landfills in Asia, and then compared our findings to previous research (Table 1) that also examined these target analytes. We further identified whether presence of antibiotics exceeded proposed no effect concentrations for the development of ABR microorganisms.

2. Methods

2.1. Study locations

One of the largest active landfills in Asia, WENT opened in 1993, has a designed capacity of 61 million m^3, and is currently receiving domestic, industrial, commercial and construction waste (Environmental Protection Department, 2015b). Leachate from WENT is temporarily stored and aerated in leachate lagoons on site and then is piped for stripping of ammonia at 1000 °C and then treated in a biological sequencing batch reactor. Treated leachate is connected to the municipal sewer and discharged to North Western Water Control Zone in Hong Kong via a submarine outfall (EPD, personal communication, 10 August 2016).

PPV started operation in 1983 and was closed on 1996. A total of 13 million tons of domestic, construction, commercial and industrial wastes were landfilled there. Restoration started in 2004 when capping, treatment facilities, membrane, gas collection, surface drainage were added (EPD, 2015a). PPV contains an on-site ammonia stripping facility very similar to WENT. Treated leachate is transferred to the Pillar Point Sewage Treatment Works for chemical enhanced primary treatment before discharging to the North Western Water Control Zone via a submarine sewage outfall (EPD, personal communication, 10 August 2016). While requests for raw leachate samples from PPV were made, EPD explained that only treated leachate could be provided for study.

SW landfill started operation ten years earlier than PPV (in 1973), was closed in 1995, and then restoration started in 1996. It received a total of 15 million tons of domestic waste, commercial, industrial and construction waste during its operation (EPD, 2015a). Collected raw leachate is pumped to the Tai Po sewage treatment work for secondary treatment and the treated wastewater will be discharged to Victoria Harbor (Phase Two) Water Control Zone under the Tolo Harbor Effluent Export Scheme (EPD, personal communication, 10 August 2016). No treated leachate sample was available because there is no leachate treatment facility on-site. This restored landfill is now used as a golf driving range. It is important to note that prior to commissioning of a sewage sludge incinerator during 2015 (EPD, 2016), all sewage sludge was landfilled in Hong Kong. As a result, both PPV and SW received sewage sludge throughout their life spans and WENT continued to receive sewage sludge until April 2015. Fig. 1 shows locations of each of these landfills.

2.2. Sample collection

Because infiltration from precipitation influences production of landfill leachate, we selected June and January for sampling in Hong Kong, which typically corresponds to the highest annual rainfall in June and the lowest annual rainfall in January. Duplicate leachate samples of each location were collected from PPV, SW, and WENT on 26 June 2015 and 12 Jan 2016. In all cases, pre-cleaned 4 l amber colored glass bottles conforming to Environmental Protection Agency (EPA) contaminant free standard were used to hold samples. In the field, glass bottles were sample-rinsed three times immediately before sample collection. For the field blank, one pre-cleaned 4 l amber glass bottle was exposed at the raw leachate collection site (in the June 2015 campaign) and another at the treated leachate sampling site (in the January 2016 campaign).

2.3. Sample extraction

A one liter aliquot of each sample were sequentially filtered by 0.45 μm glass fiber filter and 0.22 μm nylon membrane using a standard Buchner funnel and flask filtration set up. Before extraction, for each 500 ml of filtered sample, 5 ml of 5% (w/v) Na_2EDTA was added as a chelating agent. 5 M formic acid was added to adjust the solution to pH 3.0–3.5. The filtered samples were then extracted with Oasis Hydrophilic–Lipophilic Balanced (HLB) solid phase extraction (SPE) cartridges and SPE manifold (Sigma-Aldrich Co. USA). Target analytes in each cartridge were eluted with methanol for determination by a high-performance liquid chromatograph–tandem mass spectrometer (HPLC–MS/MS).

2.4. Chemicals and standards

All antibiotics standards (TMP, CLX, CIP, SMX, CAP, ERY and ROX) were procured from Sigma-Aldrich with purities of ≥99.5% for CLX, SMX and ERY, ≥98% for TMP, CIP and CAP, and ≥90%for ROX. Individual stock solutions of antibiotics were prepared at concentrations of 1000 μg/l by dissolving appropriate amounts of antibiotics in methanol and stored in the dark at −20 °C. Nine concentrations of