Science of the Total Environment 463–464 (2013) 488–496

Contents lists available at ScienceDirect

Science of the Total Environment

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

Sewage pollution in urban stormwater runoff as evident from the widespread presence of multiple microbial and chemical source tracking markers

J.P.S. Sidhu [a,c,*], W. Ahmed [a,c], W. Gernjak [b], R. Aryal [b,f], D. McCarthy [d], A. Palmer [a], P. Kolotelo [d], S. Toze [a,e]

[a] CSIRO Land and Water, Ecosciences Precinct, 41 Boggo Road, Qld 4102, Australia
[b] The University of Queensland, Advanced Water Management Centre (AWMC), Qld 4072, Australia
[c] Faculty of Science, Health and Education, University of the Sunshine Coast, Maroochydore, DC, Qld 4558, Australia
[d] Monash Water for Liveability, Civil Engineering Department, Monash University, Clayton 3800, Australia
[e] School of Population Health, University of Queensland, Herston Road, Herston, Qld 4006, Australia
[f] Centre for Water Management and Reuse, University of South Australia, Mawson Lakes, 5095, SA, Australia

HIGHLIGHTS

- Presence of multiple MST and CST markers suggests ubiquitous sewage contamination.
- MST and CST markers suggest ubiquitous sewage contamination in urban environment.
- Good consensus (>80%) between the occurrence of MST and CST markers
- HF183 had high concurrence with human adenovirus and acesulfame.

ARTICLE INFO

Article history:
Received 30 April 2013
Received in revised form 3 June 2013
Accepted 5 June 2013
Available online 3 July 2013

Editor: Damia Barcelo

Keywords:
Stormwater
Sewage contamination
Adenovirus
Fecal source tracking
Micro-pollutants

ABSTRACT

The concurrence of human sewage contamination in urban stormwater runoff ($n = 23$) from six urban catchments across Australia was assessed by using both microbial source tracking (MST) and chemical source tracking (CST) markers. Out of 23 stormwater samples human adenovirus (HAv), human polyomavirus (HPv) and the sewage-associated markers; *Methanobrevibacter smithii nifH* and *Bacteroides* HF183 were detected in 91%, 56%, 43% and 96% of samples, respectively. Similarly, CST markers paracetamol (87%), salicylic acid (78%) acesulfame (96%) and caffeine (91%) were frequently detected. Twenty one samples (91%) were positive for six to eight sewage related MST and CST markers and remaining two samples were positive for five and four markers, respectively. A very good consensus (>91%) observed between the concurrence of the HF183, HAv, acesulfame and caffeine suggests good predictability of the presence of HAv in samples positive for one of the three markers. High prevalence of HAv (91%) also suggests that other enteric viruses may also be present in the stormwater samples which may pose significant health risks. This study underscores the benefits of employing a set of MST and CST markers which could include monitoring for HF183, adenovirus, caffeine and paracetamol to accurately detect human sewage contamination along with credible information on the presence of human enteric viruses, which could be used for more reliable public health risk assessments. Based on the results obtained in this study, it is recommended that some degree of treatment of captured stormwater would be required if it were to be used for non-potable purposes.

1. Introduction

Urban stormwater can be used to augment non-potable and potable water supplies within cities and other urban areas (Sidhu et al., 2012). However, stormwater may also contain a variety of chemicals, metals and fecal material of human and animal origin. There are several impediments to reuse of stormwater for non-potable and potable purposes in urban residential areas. The most significant issue appears to be associated with the presence of pathogens in stormwater, potentially originating from human sewage contamination (Cizek et al., 2008; Noble et al., 2006; Rajal et al., 2007; Sauer et al., 2011; Sercu et al., 2009).

There is a growing evidence that stormwater conveyance networks can be contaminated with sewage due to failing sewer infrastructure and cross connections between stormwater and sewage networks (Noble et al., 2006; Rajal et al., 2007; Sercu et al., 2009).

* Corresponding author at: CSIRO Land and Water, Ecosciences Precinct, 41 Boggo Road, Qld 4102, Australia. Tel.: +61 7 3833 5576; fax: +61 7 3833 5503.
E-mail address: Jatinder.Sidhu@csiro.au (J.P.S. Sidhu).

0048-9697/$ – see front matter. Crown Copyright © 2013 Published by Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.scitotenv.2013.06.020

Exposure to stormwater runoff impacted waters has been linked to increased risk of gastrointestinal (GI) diseases (Curriero et al., 2001; Gaffield et al., 2003). Human health risk assessment and remediation strategies for microbial contamination from stormwater can be more effectively implemented if sources of contamination are known.

Traditional fecal indicator bacteria (FIB) such as *Escherichia coli* and *Enterococcus* spp. are routinely monitored to assess the microbiological quality of surface waters, however, the presence of FIB does not necessarily correlate with the presence of pathogens especially viral and protozoan pathogens (Horman et al., 2004; McQuaig et al., 2009; Selvakumar and Borst, 2006). Furthermore, monitoring for the FIB numbers in stormwater does not provide definitive information on the possible sources of contamination which is a major shortcoming of such evaluations. As identification of sources of pollution is difficult, microbial source tracking (MST) and chemical source tracking (CST) methods have been developed and used to discriminate between human and non-human sources of fecal contamination in environmental waters (Glassmeyer et al., 2005; Nakada et al., 2008; Parker et al., 2010; Sauer et al., 2011).

MST methods based on polymerase chain reaction (PCR) can be used to detect the presence of specific genes associated with certain groups of bacteria (Bernhard et al., 2003; Scott et al., 2005) or viruses (Fong et al., 2005; McQuaig et al., 2009) from human and animal hosts. PCR based methods have been successfully used for the detection of sewage-associated *Bacteroides* HF183 and *nifH* markers in surface waters (Ahmed et al., 2012b; Sercu et al., 2011; Seurinck et al., 2005; Ufnar et al., 2006). Human adenovirus (HAv) and human polyomavirus (HPv) are known to be highly prevalent (10^2 to 10^5/l) in sewage contaminated surface waters (Hamza et al., 2009; Muscillo et al., 2008; Sauer et al., 2011) and due to their stringent host specificity they are considered as most accurate MST markers (Ahmed et al., 2012b; Fong et al., 2005; Wong et al., 2012). In addition, viral MST assays provide more reliable information on potential health risks from water resources.

During the past decade, a number of studies have extensively surveyed the prevalence of pharmaceuticals and personal care products in sewage effluent and aquatic environments (Benotti and Brownawell, 2007; Clara et al., 2004; Duan et al., 2013; Glassmeyer et al., 2005; Nakada et al., 2008; Verlicchi et al., 2012). A number of CST markers including fecal sterols/stanols (Gregor et al., 2002), caffeine (Buerge et al., 2003; Heberer et al., 2002), and artificial sweeteners (Nakada et al., 2008; Scheurer et al., 2011) have also been suggested as specific sewage markers. Persistent markers such as acesulfame are reported to be useful for tracing the pathways of treated sewage, whereas, biodegradable compounds such as caffeine are indicators of untreated wastewater ingress into fresh water (Buerge et al., 2006).

Each of the MST and CST marker described in the literature to date has advantages and disadvantages (Hagedorn and Weisberg, 2009; Scott et al., 2002). These limitations include inadequate host specificity, lack of prevalence of the markers in host groups, lack of temporal and geographical stability and their environmental persistence. The consequence of inaccurate source tracking based on false positive results (if a non-specific marker is used) may lead to expensive infrastructure improvements that may not actually improve the water quality in question. MST analysis approach involving several markers is reported to improve the accuracy of identification of polluting sources (Ahmed et al., 2012a; Boehm et al., 2003; Mauffret et al., 2012; Noble et al., 2006). To date, most published studies on the characterizing of fecal contamination in stormwater are limited to MST markers (Noble et al., 2010; Sauer et al., 2011; Sidhu et al., 2012; Surbeck et al., 2006). Only a few studies have evaluated the advantages of using both MST and CST markers for the assessment of human sewage contamination in surface water (Blanch et al., 2006; Gourmelon et al., 2010; Litton et al., 2010; Peeler et al., 2006; Sauve et al., 2012). Application of a set of markers may provide additional information such as confidence in source identification, differentiation between recent and prior sewage contamination events and accurate health risk assessments which are vital from a regulatory point of view.

In our previous study (Sidhu et al., 2012), human-specific HF183 *Bacteroides* marker was detected in most of the stormwater samples collected from Brisbane, Australia suggesting ubiquitous sewage contamination in the urban environment. This study was carried out to determine if the contamination of stormwater runoff with sewage is limited to sub-tropical Brisbane or is a broader issue in urban catchments in other major cities across Australia. Stormwater samples were collected from six residential and commercial catchments in Brisbane, Sydney and Melbourne were assessed by using a set of sewage associated MST and CST markers. The MST markers investigated included both bacterial and viral markers. The human specific *Bacteroides* HF183 and *Methanobrevibacter smithii nifH* were tested to detect presence of human origin fecal pollution. The enteric viruses, HAv and HPv were also tested due to their specificity as MST markers and as index virus for the presence of other human enteric viruses. The chemical markers proven as useful indicators of anthropogenic sewage pollution (Glassmeyer et al., 2005) including readily biodegradable (caffeine, paracetamol, and salicylic acid) and recalcitrant marker acesulfame were also tested.

The specific aims were to determine; (i) the frequency of occurrence of sewage pollution in stormwater runoff in urban catchments across Australia; (ii) to assess the efficacy of using a set of MST and CST markers for differentiation between recent and prior contamination event; (iii) to determine the concurrence of HAv and HPv in stormwater runoff. This was done with an aim to improve understanding of the extent of potential health risks associated with reuse of stormwater for non-potable and potable purposes.

2. Materials and methods

2.1. Stormwater sampling sites

The studied catchments differ with respect to the size of their drainage area, impervious area and land use. A brief site description and potential sources of contamination in the six catchments is presented in Table 1. Three catchments, Fitzgibbon (north of Brisbane), Banyan Creek (south of Melbourne) and Ku-Ring-Gai (north of Sydney), represented medium density residential catchments covering total area of 290 ha, 235 ha and 8.9 ha, respectively. The impervious surface coefficient was estimated by using an image classification and cadastral filtering of high-resolution visible aerial photography method and was determined to be 30–39%. The remaining three sites, Makerston Street (Brisbane), Hornsby (Sydney), and Smith Street (Melbourne) are located in high density commercial areas. The Makerston Street catchment covers a total area of 30 ha, Hornsby 1.1 ha and Smith Street 23 ha. Impervious area in these catchments was determined to be $\approx$90%. Site specific rainfall data was collected from the Australian Bureau of Meteorology (BOM) website which varied across catchments from 5.8 to 82 mm depending upon storm intensity (Table 3).

2.2. Stormwater sampling strategy

Multiple stormwater samples were collected from each of the six sampling sites after the storm events on multiple occasions. On each sampling occasion, volume proportional composite samples were taken using automated sampling infrastructure (ISCO 6700 or equivalent) triggered by automated flow measurement (either using a Doppler flowmeter or a weir, depending on site characteristics). The automatic samplers were programmed to the site specific requirements, overall allowing to reliably determined event mean concentrations via composite samples. These samplers were programmed to fill up to 20 l high density polyethylene containers (HDPE) (Food and Drug approved grade) during a storm event which were then mixed to obtain a composite