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Identification of sewage markers to indicate sources of contamination: Low cost options for misconnected non-stormwater source tracking in stormwater systems

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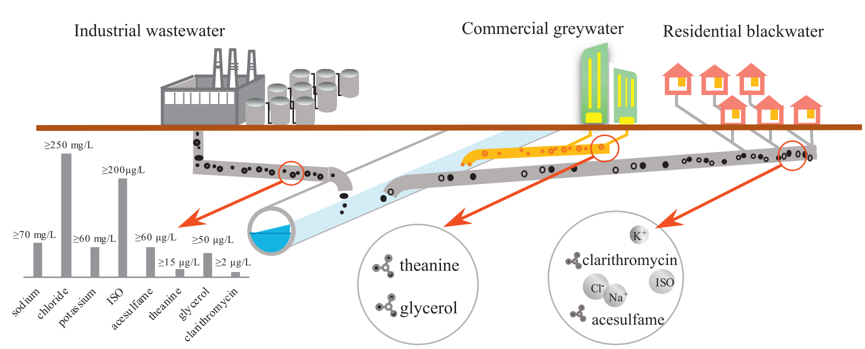
HIGHLIGHTS

- Concentrations of fifty-two markers through industrial WWTPs were investigated.
- Eight markers were found stable to indicate industrial and domestic contamination.
- Benchmark data to distinguish between industrial and domestic sewage was provided.
- Tracking of illicit domestic discharges was manifested using acesulfame and theanine.

GRAPHICAL ABSTRACT



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ABSTRACT

There has been increasing research focusing on the detection and occurrence of wastewater contamination in urban water systems. To find suitable markers to indicate industrial and domestic sewage flows inappropriately entering storm drains, this study investigated the occurrence and fate of 52 chemical markers through wastewater treatment facilities of manufacturers of agricultural and sideline products, beverage products, and pharmaceutical products, which are also consumed in our daily life. Of the 52 candidate markers, sodium, chloride, potassium, isomalto-oligosaccharide, acesulfame, theanine, glycerol, and clarithromycin were found to be conservative markers, with an average change in concentrations through the wastewater treatment processes of <30%. These markers are useful in identifying industrial and domestic sewage flow contamination in urban sewers. Specially, sodium, chloride, potassium, isomalto-oligosaccharide, acesulfame, and clarithromycin exhibited higher concentrations in blackwater than in greywater, with detected average concentrations of 43.8 mg/L, 189 mg/L, 37.3 mg/L, 123 μg/L, 37.2 μg/L, and 0.99 μg/L in blackwater, respectively. In contrast, theanine and glycerol were observed with higher concentrations in greywater than in blackwater (average 10.1 μg/L and 19.5 μg/L in greywater, respectively). The benchmark concentrations to discriminate between industrial and domestic sewage were also presented. A study in a storm drainage system of downstream Taihu catchment, China

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demonstrated the usefulness of the markers as low-cost options to trace and quantify misconnected wastewater entries into storm drains, while denoting priority areas for misconnected entries correction.

1. Introduction

For watershed managers, the location of potential sources of contamination is an important step in addressing urban water quality concerns. In the urban river catchment, where separate sewer and storm networks have been developed, inappropriate or illicit non-stormwater discharge from the storm drainage system may account for significant pollution of the receiving waters, including wastewater, which is be an important source of dry-weather flow (Pitt et al., 1993; Field et al., 1994; Brown et al., 2004; Xu et al., 2014, 2016; Revitt and Ellis, 2016). Additionally, on wet-weather days, contaminated urban stormwater runoff consists of traditional precipitation that drains from city surfaces as well as the waters from illicit and/or inappropriate wastewater flows into the storm drainage system, thus aggravating the overflow pollution (Zhang et al., 2015, 2016; Yin et al., 2017).

Efficient implementation of substantial efforts to eliminate the illicit connections would require a thorough assessment of sewer conditions; furthermore, prioritization of high-risk areas would allow the utilities to focus their efforts and proactively treat problems. Most of the city-wide assessments so far have relied upon physical methods, such as CCTV (closed circuit television), temperature based thin-film cabling examination, and flow measurements. However, these methods are often too detail-focused and labor-intensive to achieve system-wide tracking of contaminated sources in sewer dry-weather flow.

Optionally, a few marker species could be used to identify and quantify sewage source discharge, which is usually less costly to perform than by thorough physical inspections. Recently various chemical markers have been utilized to detect wastewater contamination in surface waters and groundwater, and these markers can be generally classified as pharmaceutical and personal care products (PPCPs), artificial sweeteners, fluorescent whitening agents (FWAs), sterols/stanols, isotopic compositions, and other alternative potential markers (Lim et al., 2017). Thirty-two chemical markers, including 12 PPCPs, 3 artificial sweeteners, 4 FWAs, 8 sterols/stanols, 2 isotopic compositions, and 3 other alternative potential markers, have been investigated. For example, in recent studies, researchers demonstrated that PPCPs and artificial sweeteners could serve as promising markers of sewage contamination (Nakada et al., 2008; Buerge et al., 2009; Kasprzyk-Hordern et al., 2009; Gasser et al., 2011; Daneshvar et al., 2012; Fono and Sedlak, 2005; Kuroda et al., 2012; Tran et al., 2014a, Tran et al., 2014b; Sun et al., 2016; Yang et al., 2017).

Although these reported markers have shown to be useful in indicating anthropogenic domestic sewage contamination in surface water bodies (i.e., rivers, canals, lakes, and coastal waters) and groundwater, it is still challenging to further distinguish raw sewage sources, considering pollution may result from various domestic and industrial activities. For example, flow discharge of higher concentrations may be observed as a result of illicit industrial wastewater entry into the storm pipes of an urbanized area, where food, beverage, and pharmaceutical manufacturing industries are present. Considering these products are consumed in daily life, these potential markers associated with a variety of industries may also be effective indicators of domestic sewage discharge. From this perspective, identifying new markers will facilitate the identification and quantification of industrial wastewater contamination in the dry-weather flow, and also promote our understandings of the "fingerprint" of various domestic activities.

Hence, the objective of our study was to find the chemical markers to indicate sewage contamination on the basis of three typical industrial activities in an urbanized area (i.e., food, beverage, and pharmaceutical processing or manufacturing). To meet this objective, a large number of candidate marker species associated with these industries were considered according to production process survey and through laboratory testing of onsite collected water samples. We aimed to identify several suitable indicators that would be useful in tracking and quantifying potential sewage sources. Finally, the findings were applied in an urban drainage system of downstream Taihu catchment area, China to demonstrate the use of these new markers in detecting inappropriate sewage connections to storm drains.

2. Materials and methods

2.1. Sampling sites

To identify new markers indicating sewage contamination in dry-weather flow, a total of 14 sampling sites were chosen. These sites are located in the wastewater treatment facilities of industrial enterprises in Shanghai, China. The facilities cover 3 industrial trades and are associated with 12 sub-industrial trade types, which are related to agricultural food processing and manufacturing, alcohol, beverages, and refined tea manufacturing, and pharmaceutical manufacturing. A description of the sampling sites is given in the Supporting Information (Section S1). Additionally, one sampling site in a residential community was also chosen to establish the data library of screened chemical markers in sanitary sewage. Specifically, the sampling site is a residential community close to our university (i.e., No. 8 Anshan Village, which was built in the late 1970s).

2.2. Candidate markers and samples collection

It is necessary to select candidate marker species before conducting sample collection and analyses. For each chosen industrial trade type, candidate markers were determined based on the raw materials used and their production processes, as explained in the Supporting Information (Section S2). In total, 52 candidate markers were selected, including 14 markers for agricultural and sideline food processing and manufacturing, 33 markers for alcohol/beverage products manufacturing (4 markers overlapped with those for agricultural products manufacturing), and 13 markers for pharmaceutical products manufacturing (4 markers overlapped with those for agricultural products manufacturing, as well).

Ideal markers of contaminated dry-weather flow should also exhibit conservative behavior (i.e., no significant concentration change due to biological or chemical processes), besides their obvious differences in concentrations between polluted and relatively clear sources. To determine if the candidate markers exhibit conservative or reluctant behavior, sewage samples were measured at the influent and effluent of wastewater treatment plants (WWTP) for each enterprise. Specifically, two sampling activities were performed. The first sampling activity was conducted in May, 2015, and the second was conducted in Nov., 2015. For each sampling activity, the sewage samples were collected during the period of 9 a.m. to 6 p.m. with a sampling interval of 1 h, resulting in total 20 samples for each enterprise. The sewage samples (1.5 L each) were kept at approximately 4 °C and transported back to the laboratory for immediate processing.

As mentioned above, water samples were also collected in one residential community. Sanitary sewage from this community flows through two parallel indoor and outdoor sewer pipes: one is for blackwater from the toilet and another is for greywater from bathing, laundry, dish washing, and cooking. The sampling locations were located at the sewer network outlets of this residential community. Sample