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#### Review

## Fecal pollution source tracking toolbox for identification, evaluation and characterization of fecal contamination in receiving urban surface waters and groundwater

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#### HIGHLIGHTS

• E. coli and enterococci are poor for fecal pollution source tracking (FST).

• Human-specific HF183 Bacteroides 16S rRNA genetic marker is a good marker for FST.

• PPCPs and artificial sweeteners can be used as chemical markers for human FST.

• The use of a single microbial or chemical marker is challenging for FST.

• The use of both chemical and microbial markers is recommended as a toolbox for FST.

#### A R T I C L E I N F O

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#### ABSTRACT

The quality of surface waters/groundwater of a geographical region can be affected by anthropogenic activities, land use patterns and fecal pollution sources from humans and animals. Therefore, the development of an efficient fecal pollution source tracking toolbox for identifying the origin of the fecal pollution sources in surface waters/groundwater is especially helpful for improving management efforts and remediation actions of water resources in a more cost-effective and efficient manner. This review summarizes the updated knowledge on the use of fecal pollution source tracking markers for detecting, evaluating and characterizing fecal pollution sources in receiving surface waters and groundwater. The suitability of using chemical markers (i.e. fecal sterols, fluorescent whitening agents, pharmaceuticals and personal care products, and artificial sweeteners) and/or microbial markers (e.g. F + RNA coliphages, enteric viruses, and hostspecific anaerobic bacterial 16S rDNA genetic markers) for tracking fecal pollution sources in receiving water bodies is discussed. In addition, this review also provides a comprehensive approach, which is based on the detection ratios (DR), detection frequencies (DF), and fate of potential microbial and chemical markers. DR and DF are considered as the key criteria for selecting appropriate markers for identifying and evaluating the impacts of fecal contamination in surface waters/groundwater.

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*Abbreviations*: ACE, Acesulfame; ACT, Acetaminophen; AdVs, Adenoviruses; AHTN, Tonalide; aONE, α-Chlolestanone; ASs, Artificial sweeteners; bONE, β-cholestanone; bSTIG, 5β-stigmasterol; CBZ, Carbamazepine; CF, Caffeine; CFU, Colony forming unit; CHOA, Cholestanone; CHOE, Cholesterol; COP, Coprostanol; CTMT, Crotamiton; CYC, Cyclamate; DAS1, Diaminostilbene; DEET, N,N-diethyl meta toluamide; DF, Detection frequency; DR, Detection ratio; DSBP, Distyrylbiphenyl; DTZ, Diatrizoate; EC, *Escherichia coli*; ENT, Enterococci; FIB, Fecal indicator bacteria; FIM, Fecal indicator microbes; FWAs, Fluorescent whitening agents; HepVs, Hepatitis viruses; HHCB, Galaxolide; HPyVs, Human polyomaviruses; IOM, Iomeprol; IOP, Iopromide; NoVs, Noroviruses; PCR, Polymerase chain reaction; PFU, Plaque forming unit; PMD, Promidone; PMMoVs, Pepper mild mottle viruses; PPCPs, Pharmaceutical and personal care products; RVs, Rotaviruses; SA, Salicylic acid; EOCs, Emerging organic contaminants; EVs, Enteroviruses; SAC, Saccharin; SUC, Sucralose; TCC, Triclocarban; TCS, Triclosan; WWTPs, Wastewater treatment plants.

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#### Contents

1.	Introduction
2.	The use of microbial markers as a fecal pollution source tracking toolbox
	2.1. Traditional fecal indicator bacteria for fecal pollution source tracking
	2.2. The use of bacteriophages as fecal pollution source markers
	2.3. The use of enteric viruses as fecal pollution source tracking markers
	2.4. The use of host-specific anaerobic bacteria genes as fecal pollution source tracking markers
3.	The use of organic contaminants as a fecal pollution source tracking toolbox
	3.1. The use of fecal sterols/stanols as fecal pollution source tracking markers
	3.2. The use of fluorescent whitening agents as human fecal pollution source tracking markers
	3.3. The use of pharmaceuticals and personal care products as human fecal pollution source tracking markers
	3.3.1. Persistent PPCPs used as chemical markers of treated wastewater
	3.3.2. Biodegradable PPCPs used as chemical markers of untreated wastewater
	3.3.3. Hydrophilic PPCPs used as chemical markers of wastewater in surface water/groundwater
	3.3.4. Hydrophobic PPCPs used as chemical markers of microbial risks
	3.4. The use of artificial sweeteners as human fecal pollution source tracking markers
4.	Approaches for development of an efficient fecal pollution source tracking toolbox for the study area
	4.1. Selection of potential fecal pollution source tracking markers at a study site
	4.2. Evaluation of the suitability of potential fecal pollution markers via monitoring campaigns
	4.3. Application of statistical analyses in evaluating the suitability of fecal pollution markers
	4.4. Evaluation on environmental fate and mobility of fecal pollution source tracking markers in the environment
5.	Conclusions
Ack	knowledgment
Ref	ferences

#### 1. Introduction

With rapidly increasing population and economic growth, the demand for food and water resources has grown considerably. In particular, the scarcity of water is of great concern to society, especially for countries where natural water resources are limited. For this reason, urban stormwater harvesting, groundwater exploitation, or the use of reclaimed water are considered to be alternative strategies to meet the demand for potable and non-potable water supply (Fletcher et al., 2008). However, it is evident that stormwater, reclaimed water, surface waters and groundwater sources often contain a large variety of microbial and chemical pollutants, such as microorganisms and organic pollutants associated with anthropogenic activities, land uses, and fecal pollution sources that pose a serious threat to public health. Many studies have reported that pathogenic bacteria and viruses from infected animals and humans could enter the aquatic environment through the waste or feces of animals and humans. Subsequently, the occurrence of these microorganisms in the water may cause health risk and water impairment, particularly for tropical waters where the ambient conditions tend to be favorable for the growth of these microbes (Savichtcheva and Okabe, 2006; Tallon et al., 2005).

The presence of microbial and chemical pollutants in urban surface waters and groundwater is caused by both point sources of pollution (i.e. discharge of wastewater effluents into receiving water bodies) and non-point sources of pollution (e.g. sewer leakages, sewer overflow discharges, illegal discharges, wildlife animal wastes and runoff from urban areas or agricultural fields) (Hillebrand et al., 2012; Kuroda et al., 2012; Nakada et al., 2008). As a consequence, urban surface water quality is significantly variable and is largely dependent on the following factors: (i) weather conditions, such as rainfall intensity, antecedent dry period between storm events, and evaporation; (ii) catchment characteristics (including sewered catchment or nonsewered catchment, catchment size, land use, population density of the catchment, and atmospheric deposition); (iii) drainage infrastructure, such as separate or combined, open channels/streams or pipes, age, cross-connection, sewer overflows, connection to surrounding groundwater or existing septic tanks. Therefore, an in-depth understanding of the fecal pollution sources affecting the urban stormwater and groundwater quality is of critical importance to improve management efforts of water resources for preserving water quality, while allowing remediation action for contaminated sites to be operated in a more-cost effective and efficient manner.

Hitherto, numerous efforts have been made to detect and evaluate the effects of point- and non-point sources of pollution contributing to receiving surface waters and groundwater through using either specific types of microorganisms or organic tracers like pharmaceuticals and personal care products (PPCPs) that may be associated with a certain possible pollution. For instance, many previous studies have used fecal indicator microbes (FIM), such as fecal coliforms (FC), Escherichia coli (EC), enterococci (ENT), or F-specific coliphages (F + RNA coliphages) to identify and evaluate the microbiological quality of water bodies (Borrego et al., 1987; Griffin et al., 2000; Cole et al., 2003; Ibarluzea et al., 2007; McQuaig et al., 2012; Savichtcheva and Okabe, 2006; Tallon et al., 2005). Several studies have also recommended using trace organic contaminants such as PPCPs, artificial sweeteners (ASs), fluorescent whitening agents (FWAs) and fecal sterols as chemical markers to detect and evaluate the effects of human feces or human sewage on receiving water bodies (Hagedorn and Weisberg, 2009; Kuroda et al., 2012; Managaki et al., 2006; Nakada et al., 2008; Tran et al., 2014c; Van Stempvoort et al., 2011; Wolf et al., 2012). However, it has been widely acknowledged that the use of a single source tracking chemical or microbial marker can be insufficient to identify the origin of contamination sources in surface waters and groundwater, particularly in terms of areas where receiving water bodies are impacted by different non-point sources of pollution due to geographical characteristics.

Although the use of microbial and chemical markers for tracking fecal pollution sources in receiving surface waters and groundwater has been documented in several earlier publications (Armon and Kott, 1996; Scott et al., 2002; Hagedorn et al., 2011; Harwood, 2014; Stoeckel and Harwood, 2007; Wong et al., 2012), most of these studies have provided only qualitative information with a limited discussion on the advantages or disadvantages of microbial and chemical markers. To fill these gaps, this review provides a more in-depth discussion on the advantages and disadvantages of chemical and microbial markers based on quantitative data on the occurrence and fate of these markers in both engineered and natural systems. In particular, this review

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