

## Source Tracking of Inappropriate Discharges to Storm Drainage Systems

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### Abstract

This paper describes procedures that have been used to identify sources of inappropriate (“illicit”) discharges in storm drainage systems. Also included is a review of emerging techniques that may also be useful, especially in future years as they become more accessible and become proven technologies. This paper also describes a series of tests where the original methods developed previously for EPA (Pitt, *et al.* 1993), along with selected new procedures, were examined using almost 700 stormwater samples collected from telecommunication manholes from throughout the U.S. About ten percent of the samples were estimated to be contaminated with sanitary sewage using these methods, similar to what is expected for most stormwater systems. The original methods are still recommended as the most useful procedure for identifying contamination of storm drainage systems, with the possible addition of specific tests for *E. coli* and enterococci and UV absorbance at 228 nm. Most newly emerging methods require exotic equipment and unusual expertise and are therefore not very available, especially at low cost and with fast turn-around times for the analyses. These emerging methods may therefore be more useful for special research projects than for routine screening of storm drainage systems.

The Center for Watershed Protection (CWP) and Dr. Robert Pitt with the University of Alabama are currently being funded by EPA to complete a technical assessment of techniques and methods for identifying and correcting illicit and inappropriate discharges geared towards NPDES Phase II communities. The project has a two year duration. In the first year, most of our effort will be directed to collecting data. The most cost effective and efficient techniques will also be identified during this initial project period. In the second project year, the project team will develop draft guidance on methods and techniques to identify and correct illicit connections, test the efficacy of the draft guidance in four communities, complete a final “User’s Manual for Identifying and Correcting Illicit and Inappropriate Discharges,” and conduct training and dissemination. This project started in late summer, 2001.

### Introduction

Urban stormwater runoff includes waters from many other sources which find their way into storm drainage systems, besides from precipitation. There are cases where pollutant levels in storm drainage are much higher than they would otherwise be because of excessive amounts of contaminants that are introduced into the storm drainage system by various non-stormwater discharges. Additionally, baseflows (during dry weather) are also common in storm drainage systems. Dry-weather flows and wet-weather flows have been monitored during numerous urban runoff studies. These studies have found that discharges observed at outfalls during dry weather were significantly different from wet-weather discharges and may account for the majority of the annual discharges for some pollutants of concern from the storm drainage system.

There have been numerous methods used to investigate inappropriate discharges to storm drainage systems. Pitt, *et al.* (1993) and Lalor (1994) reviewed many of these procedures and developed a system that municipalities could use for screening outfalls in residential and commercial areas. In these areas, sewage contamination, along with low rate discharges from small businesses (especially laundries, vehicle repair shops, plating shops, etc.) are of primary concern. One of the earliest methods used to identify sewage contamination utilized the ratio of fecal coliform to fecal strep. bacteria. This method is still in use, but unfortunately has proven

inaccurate in most urban stormwater applications. The following discussion reviews the methodology developed by Pitt, *et al.* (1993) and Lalor (1994), and some new approaches that were investigated.

### **Use of Tracers to Identify Sources of Contamination in Urban Drainage Systems**

Investigations designed to determine the contribution of urban stormwater runoff to receiving water quality problems have led to a continuing interest in inappropriate connections to storm drainage systems. Urban stormwater runoff is traditionally defined as that portion of precipitation which drains from city surfaces and flows via natural or man-made drainage systems into receiving waters. In fact, urban stormwater runoff also includes waters from many other sources which find their way into storm drainage systems. Sources of some of this water can be identified and accounted for by examining current National Pollutant Discharge Elimination System (NPDES) permit records for permitted industrial wastewaters that can be legally discharged to the storm drainage system. However, most of the water comes from other sources, including illicit and/or inappropriate entries to the storm drainage system. These entries can account for a significant amount of the pollutants discharged from storm sewerage systems (Pitt and McLean 1986).

Permits for municipal separate storm sewers include a requirement to effectively prohibit problematic non-stormwater discharges, thereby placing emphasis on the elimination of inappropriate connections to urban storm drains. Section 122.26 (d)(1)(iv)(D) of the rule specifically requires an initial screening program to provide means for detecting high levels of pollutants in dry weather flows which should serve as indicators of illicit connections to the storm sewers. To facilitate the application of this rule, the EPA's Office of Research and Development's Storm and Combined Sewer Pollution Control Program and the Environmental Engineering & Technology Demonstration Branch, along with the Office of Water's Nonpoint Source Branch, supported research for the investigation of inappropriate entries to storm drainage systems (Pitt, *et al.* 1993). The approach presented in this research was based on the identification and quantification of clean baseflow and the contaminated components during dry weather. If the relative amounts of potential components are known, then the importance of the dry weather discharge can be determined.

The ideal tracer to identify major flow sources should have the following characteristics:

- Significant difference in concentrations between possible pollutant sources;
- Small variations in concentrations within each likely pollutant source category;
- A conservative behavior (i.e., no significant concentration change due to physical, chemical or biological processes); and,
- Ease of measurement with adequate detection limits, good sensitivity and repeatability.

In order to identify tracers meeting the above criteria, literature characterizing potential inappropriate entries into storm drainage systems was examined. Several case studies which identified procedures used by individual municipalities or regional agencies were also examined.

***Selection of Parameters for Identifying Inappropriate Discharge Sources.*** Table 1 is an assessment of the usefulness of candidate field survey parameters in identifying different potential non-stormwater flow sources. Natural and domestic waters should be uncontaminated (except in the presence of contaminated groundwaters entering the drainage system, for example). Sanitary sewage, septage, and industrial waters can produce toxic or pathogenic conditions. The other source flows (wash and rinse waters and irrigation return flows) may cause nuisance conditions, or degrade the ecosystem. The parameters marked with a plus sign can probably be used to identify the specific source flows by their presence. Negative signs indicate that the potential source flow probably does not contain the listed parameter in adverse or obvious amounts, and may help confirm the presence of the source by its absence. Parameters with both positive and negative signs for a specific source category would not likely be very helpful due to likely wide variations expected.

### **Parameters Suitable for Indicators of Contamination by Sanitary Sewage**

***Tracer Characteristics of Local Source Flows.*** Table 2 is a summary of tracer parameter measurements for Birmingham, AL. This table is a summary of the "library" that describes the tracer conditions for each potential source category. The important information shown on this table includes the median and coefficient of variation

(COV) values for each tracer parameter for each source category. Appropriate tracers are characterized by having significantly different concentrations in flow categories that need to be distinguished. In addition, effective tracers also need low COV values within each flow category. The study indicated that the COV values were quite low for each category, with the exception of chlorine, which had much greater COV values. Chlorine is therefore not recommended as a quantitative tracer to estimate the flow components. Similar data must be collected in each community where these procedures are to be used. Recommended field observations include color, odor, clarity, presence of floatables and deposits, and rate of flow, in addition to the selected chemical measurements.

**TABLE 1. Candidate Field Survey Parameters and Associated Non-Stormwater Flow Sources**

Parameter	Natural Water	Potable Water	Sanitary Sewage	Septage Water	Indus. Water	Wash Water	Rinse Water	Irrig. Water
Fluoride	-	+	+	+	+/-	+	+	+
Hardness change	-	+/-	+	+	+/-	+	+	-
Surfactants	-	-	+	-	-	+	+	-
Florescence	-	-	+	+	-	+	+	-
Potassium	-	-	+	+	-	-	-	-
Ammonia	-	-	+	+	-	-	-	+/-
Odor	-	-	+	+	+	+/-	-	-
Color	-	-	-	-	+	-	-	-
Clarity	-	-	+	+	+	+	+/-	-
Floatables	-	-	+	-	+	+/-	+/-	-
Deposits and stains	-	-	+	-	+	+/-	+/-	-
Vegetation change	-	-	+	+	+	+/-	-	+
Structural damage	-	-	-	-	+	-	-	-
Conductivity	-	-	+	+	+	+/-	+	+
Temperature change	-	-	+/-	-	+	+/-	+/-	-
pH	-	-	-	-	+	-	-	-

Note: - implies relatively low concentration  
 + implies relatively high concentration  
 +/- implies variable conditions

**Simple Data Evaluation Methods to Indicate Sources of Contamination**

**Negative Indicators Implying Contamination.** Indicators of contamination (negative indicators) are clearly apparent visual or physical parameters indicating obvious problems and are readily observable at the outfall during the field screening activities. These observations are very important during the field survey because they are the simplest method of identifying grossly contaminated dry-weather outfall flows. The direct examination of outfall characteristics for unusual conditions of flow, odor, color, turbidity, floatables, deposits/s tains, vegetation conditions, and damage to drainage structures is therefore an important part of these investigations. Table 3 presents a summary of these indicators, along with narratives of the descriptors to be selected in the field.

Correlation tests were conducted to identify relationships between outfalls that were known to have severe contamination problems and the negative indicators (Lalor 1994). Pearson correlation tests indicated that high turbidity and obvious odors appeared to be the most useful physical indicators of contamination when contamination was defined by toxicity and the presence of detergents. High turbidity was noted in 74% of the contaminated source flow samples. This represented a 26% false negative rate (indication of no contamination when contamination actually exists), if one relied on turbidity alone as an indicator of contamination. High turbidity was noted in only 5% of the uncontaminated source flow samples. This represents the rate of false positives (indication of contamination when none actually exists) when relying on turbidity alone. Noticeable odor was indicated in 67% of flow samples from contaminated sources, but in none of the flow samples from uncontaminated sources. This translates to 37% false negatives, but no false positives. Obvious odors identified included gasoline, oil, sewage, industrial chemicals or detergents, decomposing organic wastes, etc.

False negatives are more of a concern than a reasonable number of false positives when working with a screening methodology. Screening methodologies are used to direct further, more detailed investigations. False positives would be discarded after further investigation. However, a false negative during a screening investigation results in the dismissal of a problem outfall for at least the near future. Missed contributors to stream contamination may result in unsatisfactory in-stream results following the application of costly corrective measures elsewhere.

The method of using physical characteristics to indicate contamination in outfall flows does not allow quantifiable estimates of the flow components and, if used alone, will likely result in many incorrect determinations, especially false negatives. These simple characteristics are most useful for identifying gross contamination: only the most significantly contaminated outfalls and drainage areas would therefore be recognized using this method.

***Detergents as Indicators of Contamination.*** Results from the Mann-Whitney U tests (Lalor 1994) indicated that samples from any of the dry-weather flow sources could be correctly classified as clean or contaminated based only on the measured value of any one of the following parameters: detergents, color, or conductivity. Color and high conductivity were present in samples from clean sources as well as contaminated sources, but their levels of occurrence were significantly different between the two groups. If samples from only one source were expected to make up outfall flows, the level of color or conductivity could be used to distinguish contaminated outfalls from clean outfalls. However, since multi-source flows occur, measured levels of color or conductivity could fall within acceptable levels because of dilution, even though a contaminating source was contributing to the flow. Detergents, on the other hand, can be used to distinguish between clean and contaminated outfalls simply by their presence or absence, using a detection limit of 0.06 mg/L. All samples analyzed from contaminated sources contained detergents in excess of this amount (with the exception of three septage samples collected from homes discharging only toilet flushing water). No clean source samples were found to contain detergents. Contaminated sources would be detected in mixtures with uncontaminated waters if they made up at least 10% of the mixture.

***Flow Chart for Most Significant Flow Component Identification.*** A further refinement is the flow chart shown on Figure 1. This flow chart describes an analysis strategy which may be used to identify the major component of dry-weather flow samples in residential and commercial areas. This method does not attempt to distinguish among all potential sources of dry-weather flows identified earlier, but rather the following four major groups of flow are identified: (1) tap waters (including domestic tap water, irrigation water and rinse water), (2) natural waters (spring water and shallow ground water), (3) sanitary wastewaters (sanitary sewage and septic tank discharge), and (4) wash waters (commercial laundry waters, commercial car wash waters, radiator flushing wastes, and plating bath wastewaters). The use of this method would not only allow outfall flows to be categorized as contaminated or uncontaminated, but would allow outfalls carrying sanitary wastewaters to be identified. These outfalls could then receive highest priority for further investigation leading to source control. This flow chart (Lalor 1994) was designed for use in residential and/or commercial areas only.

In residential and/or commercial areas, all outfalls should be located and examined. The first indicator is the presence or absence of dry-weather flow. If no dry-weather flow exists at an outfall, then indications of intermittent flows must be investigated. Specifically, stains, deposits, odors, unusual stream-side vegetation conditions, and damage to outfall structures can all indicate intermittent non-stormwater flows. However, frequent visits to outfalls over long time periods, or the use of other monitoring techniques, may be needed to confirm that only stormwater flows occur. If intermittent flow is not indicated, then the outfall probably does not have a contaminated non-stormwater source. The other points on the flow chart serve to indicate if a major contaminating source is present, or if the water is uncontaminated. Component contributions cannot be quantified using this method, and only the "most contaminated" type of source present will be identified.

If dry-weather flow exists at an outfall, then the flow should be sampled and tested for detergents. If detergents are not present, the flow is probably from a non-contaminated non-stormwater source. The lower limit of detection for detergent should be about 0.06 mg/L.

If detergents are not present, fluoride levels can be used to distinguish between flows with treated water sources and flows with natural sources in communities where water supplies are fluoridated and natural fluoride levels

are low. In the absence of detergents, high fluoride levels would indicate a potable water line leak, irrigation water, or wash/rinse water. Low fluoride levels would indicate waters originating from springs or shallow groundwater. Based on the flow source samples tested in this research (Table 2), fluoride levels above 0.13 mg/L would most likely indicate that a tap water source was contributing to the dry-weather flow in the Birmingham, Alabama, study area.

**Table 2. Tracer Concentrations found in Birmingham, AL, Waters (mean, standard deviation, and coefficient of Variation, COV) (Pitt, *et al.* 1993 and Lalor 1994)**

	Spring water	Treated potable water	Laundry wastewater	Sanitary wastewater	Septic tank effluent	Car wash water	Radiator flush water
Fluorescence (% scale)	6.8 2.9 0.43	4.6 0.35 0.08	1020 125 0.12	250 50 0.20	430 100 0.23	1200 130 0.11	22,000 950 0.04
Potassium (mg/L)	0.73 0.070 0.10	1.6 0.059 0.04	3.5 0.38 0.11	6.0 1.4 0.23	20 9.5 0.47	43 16 0.37	2800 375 0.13
Ammonia (mg/L)	0.009 0.016 1.7	0.028 0.006 0.23	0.82 0.12 0.14	10 3.3 0.34	90 40 0.44	0.24 0.066 0.28	0.03 0.01 0.3
Ammonia/Potassium (ratio)	0.011 0.022 2.0	0.018 0.006 0.35	0.24 0.050 0.21	1.7 0.52 0.31	5.2 3.7 0.71	0.006 0.005 0.86	0.011 0.011 1.0
Fluoride (mg/L)	0.031 0.027 0.87	0.97 0.014 0.02	33 13 0.38	0.77 0.17 0.23	0.99 0.33 0.33	12 2.4 0.20	150 24 0.16
Toxicity (% light decrease after 25 minutes, $I_{25}$ )	<5 n/a n/a	47 20 0.44	99.9 <1 n/a	43 26 0.59	99.9 <1 n/a	99.9 <1 n/a	99.9 <1 n/a
Surfactants (mg/L as MBAS)	<0.5 n/a n/a	<0.5 n/a n/a	27 6.7 0.25	1.5 1.2 0.82	3.1 4.8 1.5	49 5.1 0.11	15 1.6 0.11
Hardness (mg/L)	240 7.8 0.03	49 1.4 0.03	14 8.0 0.57	140 15 0.11	235 150 0.64	160 9.2 0.06	50 1.5 0.03
pH (pH units)	7.0 0.05 0.01	6.9 0.29 0.04	9.1 0.35 0.04	7.1 0.13 0.02	6.8 0.34 0.05	6.7 0.22 0.03	7.0 0.39 0.06
Color (color units)	<1 n/a n/a	<1 n/a n/a	47 12 0.27	38 21 0.55	59 25 0.41	220 78 0.35	3000 44 0.02
Chlorine (mg/L)	0.003 0.005 1.6	0.88 0.60 0.68	0.40 0.10 0.26	0.014 0.020 1.4	0.013 0.013 1.0	0.070 0.080 1.1	0.03 0.016 0.52
Specific conductivity ( $\mu$ S/cm)	300 12 0.04	110 1.1 0.01	560 120 0.21	420 55 0.13	430 311 0.72	485 29 0.06	3300 700 0.22
Number of samples	10	10	10	36	9	10	10

**Table 3. Interpretations of Physical Observation Parameters and Likely Associated Flow Sources (Pitt, et al. 1993)**

**Odor** - Most strong odors, especially gasoline, oils, and solvents, are likely associated with high responses on the toxicity screening test. Typical obvious odors include: gasoline, oil, sanitary wastewater, industrial chemicals, decomposing organic wastes, etc.

*sewage*: smell associated with stale sanitary wastewater, especially in pools near outfall.

*sulfur* ("rotten eggs"): industries that discharge sulfide compounds or organics (meat packers, canneries, dairies, etc.).

*oil and gas*: petroleum refineries or many facilities associated with vehicle maintenance or petroleum product storage.

*rancid-sour*: food preparation facilities (restaurants, hotels, etc.).

**Color** - Important indicator of inappropriate industrial sources. Industrial dry-weather discharges may be of any color, but dark colors, such as brown, gray, or black, are most common.

*yellow*: chemical plants, textile and tanning plants.

*brown*: meat packers, printing plants, metal works, stone and concrete, fertilizers, and petroleum refining facilities.

*green*: chemical plants, textile facilities.

*red*: meat packers.

*gray*: dairies, sewage.

**Turbidity** - Often affected by the degree of gross contamination. Dry-weather industrial flows with moderate turbidity can be cloudy, while highly turbid flows can be opaque. High turbidity is often a characteristic of undiluted dry-weather industrial discharges.

*cloudy*: sanitary wastewater, concrete or stone operations, fertilizer facilities, automotive dealers.

*opaque*: food processors, lumber mills, metal operations, pigment plants.

**Floatable Matter** - A contaminated flow may contain floating solids or liquids directly related to industrial or sanitary wastewater pollution. Floatables of industrial origin may include animal fats, spoiled food, oils, solvents, sawdust, foams, packing materials, or fuel.

*oil sheen*: petroleum refineries or storage facilities and vehicle service facilities.

*sewage*: sanitary wastewater.

**Deposits and Stains** - Refers to any type of coating near the outfall and are usually of a dark color. Deposits and stains often will contain fragments of floatable substances. These situations are illustrated by the grayish-black deposits that contain fragments of animal flesh and hair which often are produced by leather tanneries, or the white crystalline powder which commonly coats outfalls due to nitrogenous fertilizer wastes.

*sediment*: construction site erosion.

*oily*: petroleum refineries or storage facilities and vehicle service facilities.

**Vegetation** - Vegetation surrounding an outfall may show the effects of industrial pollutants. Decaying organic materials coming from various food product wastes would cause an increase in plant life, while the discharge of chemical dyes and inorganic pigments from textile mills could noticeably decrease vegetation. It is important not to confuse the adverse effects of high stormwater flows on vegetation with highly toxic dry-weather intermittent flows.

*excessive growth*: food product facilities.

*inhibited growth*: high stormwater flows, beverage facilities, printing plants, metal product facilities, drug manufacturing, petroleum facilities, vehicle service facilities and automobile dealers.

**Damage to Outfall Structures** - Another readily visible indication of industrial contamination. Cracking, deterioration, and spalling of concrete or peeling of surface paint, occurring at an outfall are usually caused by severely contaminated discharges, usually of industrial origin. These contaminants are usually very acidic or basic in nature. Primary metal industries have a strong potential for causing outfall structural damage because their batch dumps are highly acidic. Poor construction, hydraulic scour, and old age may also adversely affect the condition of the outfall structure.

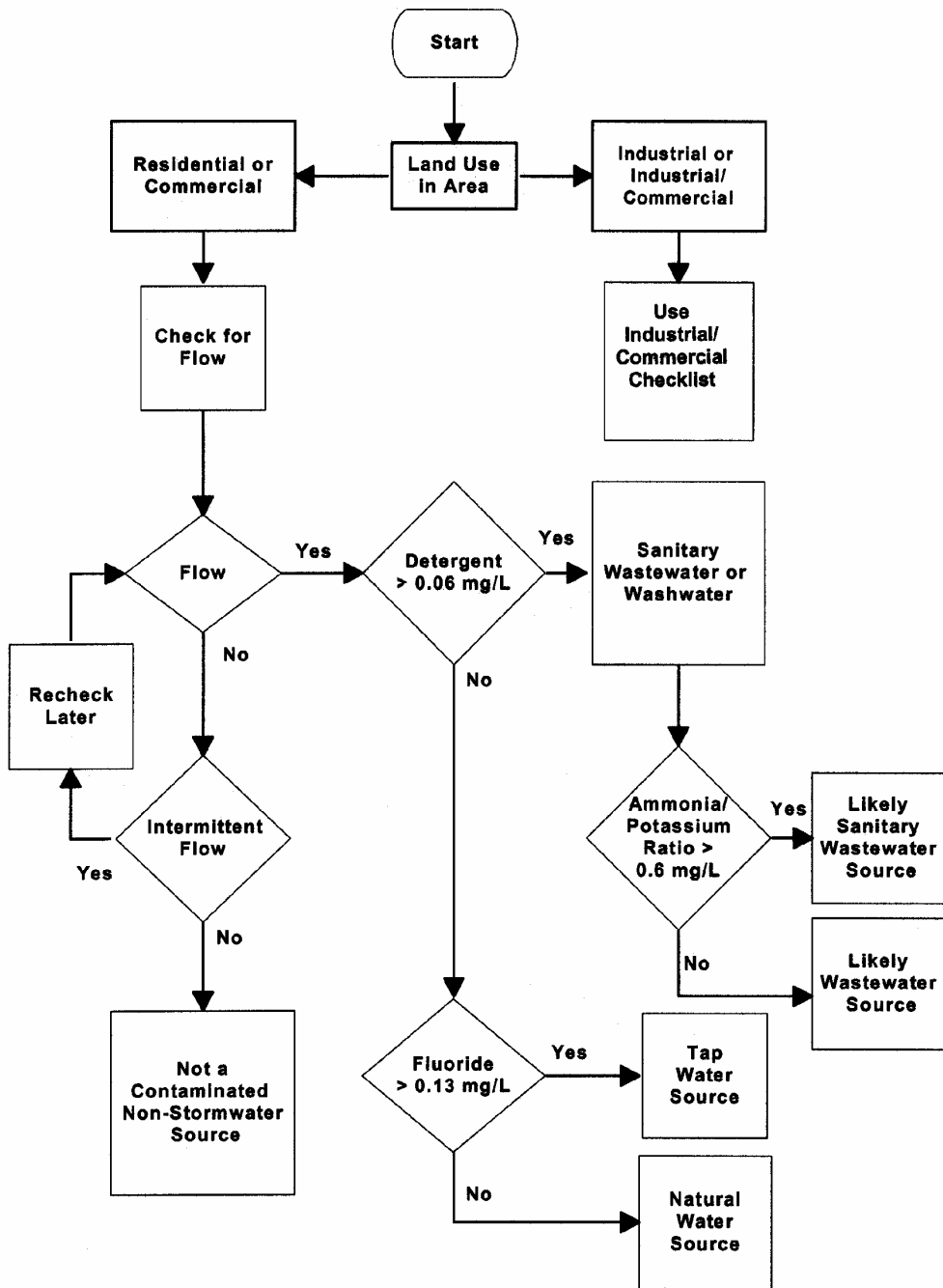
*concrete cracking*: industrial flows

*concrete spalling*: industrial flows

*peeling paint*: industrial flows

*metal corrosion*: industrial flows

Figure 1. Simple flow chart method to identify significant contaminating sources (Lalor 1994).



If detergents are present, the flow is probably from a contaminated non-stormwater source, as indicated on Table 2. The ratio of ammonia to potassium can be used to indicate whether or not the source is sanitary wastewater. Ammonia/potassium ratios greater than 0.60 would indicate likely sanitary wastewater contamination. Ammonia/potassium ratios were above 0.9 for all septage and sewage samples collected in Birmingham (values ranged from 0.97 to 15.37, averaging 2.55). Ammonia/potassium ratios for all other samples containing detergents were below 0.7, ranging from 0.00 to 0.65, averaging 0.11. One radiator waste sample had an ammonia/potassium ratio of 0.65.

Non-contaminated samples collected in Birmingham had ammonia/potassium ratios ranging from 0.00 to 0.41, with a mean value of 0.06 and a median value of 0.03. Using the mean values for non-contaminated samples (0.06) and sanitary wastewaters (2.55), flows comprised of mixtures containing at least 25% sanitary wastes with the remainder of the flow from uncontaminated sources would likely be identified as sanitary wastewaters using this method. Flows containing smaller percent contributions from sanitary wastewaters might be identified as having a wash water source, but would not be identified as uncontaminated.

### General Matrix Algebra Methods to Indicate Sources of Contamination Through Fingerprinting

Other approaches can also be used to calculate the source components of mixed outfall flows. One approach is the use of matrix algebra to simultaneously solve a series of chemical mass balance equations. This method can be used to predict the most likely flow source, or sources, making up an outfall sample. It is possible to estimate the outfall source flow components using a set of simultaneous equations. The number of unknowns should equal the number of equations available, resulting in a square matrix. If there are seven likely source categories, then there should be seven tracer parameters used. If there are only four possible sources, then only the four most efficient tracer parameters should be used. Only tracers that are linearly related to mixture components can be used. As an example, pH cannot be used in these equations, because it is not additive.

This method estimates flow contributions from various sources using a “receptor model”, based on a set of chemical mass balance equations. Such models, which assess the contributions from various sources based on observations at sampling sites (the receptors), have been applied to the investigation of air pollutant sources for many years (Scheff and Wadden 1993; Cooper and Watson 1980). The characteristic “signatures” of the different types of sources, as identified in the library of source flow data, allows the development of a set of mass balance equations. These equations describe the measured concentrations in an outfall’s flow as a linear combination of the contributions from the different potential sources. A major requirement for this method is the physical and chemical characterization of waters collected directly from potential sources of dry-weather flows (the “library”). This allows concentration patterns (fingerprints) for the parameters of interest to be established for each type of source. Theoretically, if these patterns are different for each source, the observed concentrations at the outfall would be a linear combination of the concentration patterns from the different component sources, each weighted by a source strength term ( $m_n$ ). This source strength term would indicate the fraction of outfall flow originating from each likely source. By measuring a number of parameters equal to, or greater than, the number of potential source types, the source strength term could be obtained by solving a set of chemical mass balance equations of the type:

$$C_p = \sum_n m_n x_{pn}$$

where  $C_p$  is the concentration of parameter  $p$  in the outfall flow and  $x_{pn}$  is the concentration of parameter  $p$  in source type  $n$ .

As an example of this method, consider 8 possible flow sources and 8 parameters, as presented in Table 4. The number of parameters evaluated for each outfall must equal the number of probable dry-weather flow sources in the drainage area. Mathematical methods are available which provide for the solution of over specified sets of equations (more equations than unknowns) but these are not addressed here.



The selection of parameters for measurement should reflect evaluated parameter usefulness. Evaluation of the Mann-Whitney U Test results (Lalor 1994) suggested the following groupings of parameters, ranked by their usefulness for distinguishing between all the types of flow sources sampled in Birmingham, AL:

- First set (most useful): potassium and hardness
- Second set: fluorescence, conductivity, fluoride, ammonia, detergents, and color
- Third set (least useful): chlorine

### **Emerging Tools for Identifying Sources of Discharges**

#### ***Coprostanol and Other Fecal Sterol Compounds Utilized as Tracers of Contamination by Sanitary Sewage.***

A more likely indicator of human wastes than fecal coliforms and other “indicator” bacteria may be the use of certain molecular markers, specifically the fecal sterols, such as coprostanol and epicoprostanol (Eaganhouse, *et al.* 1988). However, these compounds are also discharged by other carnivores in a drainage (especially dogs). A number of research projects have used these compounds to investigate the presence of sanitary sewage contamination. The most successful application may be associated with sediment analyses instead of water analyses. As an example, water analyses of coprostanol are difficult due to the typically very low concentrations found, although the concentrations in many sediments are quite high and much easier to quantify. Unfortunately, the long persistence of these compounds in the environment easily confuses recent contamination with historical or intermittent contamination.

Particulates and sediments collected from coastal areas in Spain and Cuba receiving municipal sewage loads were analyzed by Grimalt, *et al.* (1990) to determine the utility of coprostanol as a chemical marker of sewage contamination. Coprostanol can not by itself be attributed to fecal matter inputs. However, relative contributions of steroid components can be a useful indicator. When the relative concentrations of coprostanol and coprostanone are higher than their 5 $\alpha$  epimers, or more realistically, other sterol components of background or natural occurrence, it can provide useful information.

Sediment cores from Santa Monica Basin, CA, and effluent from two local municipal wastewater discharges were analyzed by Venkatesan and Kaplan (1990) for coprostanol to determine the degree of sewage addition to sediment. Coprostanols were distributed throughout the basin sediments in association with fine particles. Some stations contained elevated levels, either due to their proximity to outfalls or because of preferential advection of fine-grained sediments. A noted decline of coprostanols relative to total sterols from outfalls seaward indicated dilution of sewage by biogenic sterols.

Other chemical compounds have been utilized for sewage tracer work. Saturated hydrocarbons with 16-18 carbons, and saturated hydrocarbons with 16-21 carbons, in addition to coprostanol, were chosen as markers for sewage in water, particulate, and sediment samples near the Cocoa, FL, domestic wastewater treatment plant (Holm, *et al.* 1990). The concentration of the markers was highest at points close to the outfall pipe and diminished with distance. However the concentration of C16-C21 compounds was high at a site 800 m from the outfall indicating that these compounds were unsuitable markers for locating areas exposed to the sewage plume. The concentrations for the other markers were very low at this station.

The range of concentrations of coprostanol found in sediments and mussels of Venice, Italy, were reported by Sherwin, *et al.* (1993). Raw sewage is still discharged directly into the Venice lagoon. Coprostanol concentrations were determined in sediment and mussel samples from the lagoon using gas chromatography/mass spectroscopy. Samples were collected in interior canals and compared to open-bay concentrations. Sediment concentrations ranged from 0.2-41.0  $\mu\text{g/g}$  (dry weight). Interior canal sediment samples averaged 16  $\mu\text{g/g}$  compared to 2  $\mu\text{g/g}$  found in open bay sediment samples. Total coprostanol concentrations in mussels ranged from 80 to 620  $\text{ng/g}$  (wet weight). No mussels were found in the four most polluted interior canal sites.

**TABLE 4. Set of Chemical Mass Balance Equations**

	Source 1	Source 2	Source 3	Source 4	Source 5	Source 6	Source 7	Source 8	Outfall								
Parameter 1:	(m1)(x11)	+	(m2)(x12)	+	(m3)(x13)	+	(m4)(x14)	+	(m5)(x15)	+	(m6)(x16)	+	(m7)(x17)	+	(m8)(x18)	=	C1
Parameter 2:	(m1)(x21)	+	(m2)(x22)	+	(m3)(x23)	+	(m4)(x24)	+	(m5)(x25)	+	(m6)(x26)	+	(m7)(x27)	+	(m8)(x28)	=	C2
Parameter 3:	(m1)(x31)	+	(m2)(x32)	+	(m3)(x33)	+	(m4)(x34)	+	(m5)(x35)	+	(m6)(x36)	+	(m7)(x37)	+	(m8)(x38)	=	C3
Parameter 4:	(m1)(x41)	+	(m2)(x42)	+	(m3)(x43)	+	(m4)(x44)	+	(m5)(x45)	+	(m6)(x46)	+	(m7)(x47)	+	(m8)(x48)	=	C4
Parameter 5:	(m1)(x51)	+	(m2)(x52)	+	(m3)(x53)	+	(m4)(x54)	+	(m5)(x55)	+	(m6)(x56)	+	(m7)(x57)	+	(m8)(x58)	=	C5
Parameter 6:	(m1)(x61)	+	(m2)(x62)	+	(m3)(x63)	+	(m4)(x64)	+	(m5)(x65)	+	(m6)(x66)	+	(m7)(x67)	+	(m8)(x68)	=	C6
Parameter 7:	(m1)(x71)	+	(m2)(x72)	+	(m3)(x73)	+	(m4)(x74)	+	(m5)(x75)	+	(m6)(x76)	+	(m7)(x77)	+	(m8)(x78)	=	C7
Parameter 8:	(m1)(x81)	+	(m2)(x82)	+	(m3)(x83)	+	(m4)(x84)	+	(m5)(x85)	+	(m6)(x86)	+	(m7)(x87)	+	(m8)(x88)	=	C8

Equations of the Form 
$$C_p = \sum_n m_n x_{pn}$$

where:  $C_p$  = the concentration of parameter  $p$  in the outfall flow

$m_n$  = the fraction of flow from source type  $n$

$x_{pn}$  = the mean concentration of parameter  $p$  in source type  $n$

Nichols, *et al.* (1996) also examined coprostanol in stormwater and the sea-surface microlayer to distinguish human versus nonhuman sources of contamination. Other steroid compounds in sewage effluent were investigated by Routledge, *et al.* (1998) and Desbrow, *et al.* (1998) who both examined estrogenic chemicals. The most common found were 17 $\beta$ -Estradiol and estrone which were detected at concentrations in the tens of nanograms per liter range. These were identified as estrogenic through a toxicity identification and evaluation approach, where sequential separations and analyses identified the sample fractions causing estrogenic activity using a yeast-based estrogen screen. GC/MS was then used to identify the specific compounds.

***Estimating Potential Sanitary Sewage Discharges into Storm Drainage and Receiving Waters using Detergent Tracer Compounds.*** As described above, detergent measurements (using methylene blue active substance, MBAS, test methods) were the most successful individual tracer to indicate contaminated water in storm sewerage dry-weather flows. Unfortunately, the MBAS method uses hazardous chloroform for an extraction step. Different detergent components, especially linear alkylbenzene sulphonates (LAS) and linear alkylbenzenes (LAB), have also been tried to indicate sewage dispersal patterns in receiving waters. Boron, a major historical ingredient of laundry chemicals, can also potentially be used. Boron has the great advantage of being relatively easy to analyze using portable field test kits, while LAS requires chromatographic equipment. LAS can be measured using HPLC with fluorescent detection, after solid phase extraction, to very low levels. Fujita, *et al.* (1998) developed an efficient enzyme-linked immunosorbent assay (ELISA) for detecting LAS at levels from 20 to 500  $\mu\text{g/L}$ .

LAS from synthetic surfactants (Terzic and Ahel 1993) which degrade rapidly, as well as nonionic detergents (Terzic and Ahel 1993) which do not degrade rapidly, have been utilized as sanitary sewage markers. LAS was quickly dispersed from wastewater outfalls except in areas where wind was calm. In these areas LAS concentrations increased in freshwater but were unaffected in saline water. After time, the lower alkyl groups were mostly found, possibly as a result of degradation or settling of longer alkyl chain compounds with sediments. Chung, *et al.* (1995) also describe the distribution and fate of LAS in an urban stream in Korea. They examined different LAS compounds having carbon ratios of C12 and C13 compared to C10 and C11, plus ratios of phosphates to MBAS and the internal to external isomer ratio (I/E) as part of their research. González-Mazo, *et al.* (1998) examined LAS in the Bay of Cádiz off the southwest of Spain. They found that LAS degrades rapidly (Fujita, *et al.*, 1998, found that complete biodegradation of LAS requires several days), and is also strongly sorbed to particulates. In areas close to shore and near the untreated wastewater discharges, there is significant vertical stratification of LAS: the top 3 to 5 mm of water had LAS concentrations about 100 times greater than found at 0.5 m.

Zeng and Vista (1997) and Zeng, *et al.* (1997) describe a study off of San Diego where LAB was measured, along with polycyclic aromatic hydrocarbons (PAHs) and aliphatic hydrocarbons (AHs) to indicate the relative pollutant contributions of wastewater from sanitary sewage, nonpoint sources, and hydrocarbon combustion sources. They developed and tested several indicator ratios (alkyl homologue distributions and parent compound distributions) and examined the ratio of various PAHs (such as phenanthrene to anthracene, methylphenanthrene to phenanthrene, fluoranthene to pyrene, and benzo(a)anthracene to chrysene) as tools for distinguishing these sources. They concluded that LABs are useful tracers of domestic waste inputs to the environment due to their limited sources. They also describe the use of the internal to external isomer ratio (I/E) to indicate the amount of biodegradation that may have occurred to the LABs. They observed concentrations of total LABs in sewage effluent of about 3  $\mu\text{g/L}$ , although previous researchers have seen concentrations of about 150  $\mu\text{g/L}$  in sewage effluent from the same area.

The fluorescent properties of detergents have also been used as a tracer by investigating the fluorescent whitening agents (FWAs), as described by Poiger, *et al.* (1996) and Kramer, *et al.* (1996). HPLC with fluorescence detection was used in these studies to quantify very low concentrations of FWAs. The two most frequently used FWAs in household detergents (DSBP and DAS 1) were found at 7 to 21  $\mu\text{g/L}$  in primary sewage effluent and at 3 to 9  $\mu\text{g/L}$  in secondary effluent. Raw sewage contains about 10 to 20  $\mu\text{g/L}$  FWAs. The removal mechanisms in sewage treatment processes is by adsorption to activated sludge. The type of FWAs varies from laundry applications to textile finishing and paper production, making it possible to identify sewage sources. The FWAs were found in river water at 0.04 to 0.6  $\mu\text{g/L}$ . The FWAs are not easily biodegradable but they are readily photodegraded. Photodegradation rates have been reported to be about 7% for DSBP and 71% for DAS 1 in river water exposed to natural sunlight, after one hour exposure. Subsequent photodegradation is quite slow.

***Other Compounds Found in Sanitary Sewage that may be used for Identifying Contamination by Sewage.***

Halling-Sørensen, *et al.* (1998) detected numerous pharmaceutical substances in sewage effluents and in receiving waters. Their work addressed human health concerns of these low level compounds that can enter downstream drinking water supplies. However, the information can also be possibly used to help identify sewage contamination. Most of the research has focused on clofibric acid, a chemical used in cholesterol lowering drugs. It has been found in concentrations ranging from 10 to 165 ng/L in Berlin drinking water sampler. Other drugs commonly found include aspirin, caffeine, and ibuprofen. Current FDA guidance mandates that the maximum concentration of a substance or its active metabolites at the point of entry into the aquatic environment be less than 1 µg/L (Hun 1998).

Caffeine has been used as an indicator of sewage contamination by several investigators (Shuman and Strand 1996). The King County, WA, Water Quality Assessment Project is examining the impacts of CSOs on the Duwamish River and Elliott Bay. They are using both caffeine (representing dissolved CSO constituents) and coprostanol (representing particulate bound CSO constituents), in conjunction with heavy metals and conventional analyses, to help determine the contribution of CSOs to the river. The caffeine is unique to sewage, while coprostanol is from both humans and carnivorous animals and is therefore also in stormwater. They sampled upstream of all CSOs, but with some stormwater influences, 100 m upstream of the primary CSO discharge (but downstream of other CSOs), within the primary CSO discharge line, and 100 m downriver of the CSO discharge location. The relationship between caffeine and coprostanol was fairly consistent for the four sites (coprostanol was about 0.5 to 1.5 µg/L higher than caffeine). Similar patterns were found between the three metals, chromium was always the lowest and zinc the highest. King Co. is also using clean transported mussels placed in the Duwamish River to measure the bioconcentration potential of metal and organic toxicants and the effects of the CSOs on mussel growth rates (after 6 week exposure periods). Paired reference locations are available near the areas of deployment, but outside the areas of immediate CSO influence. *US Water News* (1998) also described a study in Boston Harbor that found caffeine at levels of about 7 µg/L in the harbor water. The caffeine content of regular coffee is about 700 mg/L, in contrast.

***DNA Profiling to Measure Impacts on Receiving Water Organisms and to Identify Sources of Microorganisms in Stormwater.*** This rapidly emerging technique seems to have great promise in addressing a number of nonpoint source water pollution issues. Kratch (1997) summarized several investigations on cataloging the DNA of *E. coli* to identify their source in water. This rapidly emerging technique seems to have great promise in addressing a number of nonpoint source water pollution issues. The procedure, developed at the Virginia Polytechnic Institute and State University, has been used in Chesapeake Bay. In one example, it was possible to identify a large wild animal population as the source of fecal coliform contamination of a shellfish bed, instead of suspected failing septic tanks. DNA patterns in fecal coliforms vary among animals and birds, and it is relatively easy to distinguish between human and non-human sources of the bacteria. However, some wild animals have DNA patterns that are not easily distinguishable. Some researchers question the value of *E. coli* DNA fingerprinting believing that there is little direct relationship between *E. coli* and human pathogens. However, this method should be useful to identify the presence of sewage contamination in stormwater or in a receiving water.

One application of the technique, as described by Krane, *et al.* (1999) of Wright State University, used randomly amplified polymorphic DNA polymerase chain reaction (RAPD-PCR) generated profiles of naturally occurring crayfish. They found that changes in the underlying genetic diversity of these populations were significantly correlated with the extent to which they have been exposed to anthropogenic stressors. They concluded that this rapid and relatively simple technique can be used to develop a sensitive means of directly assessing the impact of stressors upon ecosystems. These Wright State University researchers have also used the RAPD-PCR techniques on populations of snails, pill bugs, violets, spiders, earthworms, herring, and some benthic macroinvertebrates, finding relatively few obstacles in its use for different organisms. As noted above, other researchers have used DNA profiling techniques to identify sources of *E. coli* bacteria found in coastal waterways. It is possible that these techniques can be expanded to enable rapid detection of many different types of pathogens in receiving waters, and the most likely sources of these pathogens.

***Stable Isotope Methods for Identifying Sources of Water.*** Stable isotopes had been recommended as an efficient method to identify illicit connections to storm sewerage. A demonstration was conducted in Detroit as part of the Rouge River project to identify sources of dry weather flows in storm sewerage (Sangal, *et al.* 1996). Naturally occurring stable isotopes of oxygen and hydrogen can be used to identify waters originating from different

geographical sources (especially along a north-south gradient). Ma and Spalding (1996) discuss this approach by using stable isotopes to investigate recharge of groundwaters by surface waters. During water vapor transport from equatorial source regions to higher latitudes, depletion of heavy isotopes occurs with rain. Deviation from a standard relationship between deuterium and  $^{18}\text{O}$  for a specific area indicates that the water has undergone additional evaporation. The ratio is also affected by seasonal changes. As discussed by Ma and Spalding (1996), the Platte River water is normally derived in part from snowmelt from the Rocky Mountains, while the groundwater in parts of Nebraska is mainly contributed from the Gulf air stream. The origins of these waters are sufficiently different and allow good measurements of the recharge rate of the surface water to the groundwater. In Detroit, Sangal, *et al.* (1996) used differences in origin between the domestic water supply, local surface waters, and the local groundwater to identify potential sanitary sewage contributions to the separate storm sewerage. Rieley, *et al.* (1997) used stable isotopes of carbon in marine organisms to distinguish the primary source of carbon being consumed (sewage sludge vs. natural carbon sources) in two deep sea sewage sludge disposal areas.

Stable isotope analyses would not be able to distinguish between sanitary sewage, industrial discharges, washwaters, and domestic water, as they all have the same origin, nor would it be possible to distinguish sewage from local groundwaters if the domestic water supply was from the same local aquifer. This method works best for situations where the water supply is from a distant source and where separation of waters into separate flow components is not needed. It may be an excellent tool to study the effects of deep well injection of stormwater on deep aquifers having distant recharge sources (such as in the Phoenix area). Few laboratories can analyze for these stable isotopes, requiring shipping and a long wait for the analytical results. Sangal, *et al.* (1995) used Geochron Laboratories, in Cambridge, Massachusetts.

Dating of sediments using  $^{137}\text{Cs}$  was described by Ma and Spalding (1996). Arsenic contaminated sediments in the Hylebos Waterway in Tacoma, WA, could have originated from numerous sources, including a pesticide manufacturing facility, a rock-wool plant, steel slags, powdered metal plant, shipbuilding facilities, marinas and arsenic boat paints, and the Tacoma Smelter. Dating the sediments, combined with knowing the history of potential discharges and conducting optical and electron microscopic studies of the sediments, was found to be a powerful tool to differentiate between the different metal sources to the sediments.

## Conclusions

In almost all cases, a suite of analyses is most suitable for effective identification of inappropriate discharges. A recent example was reported by Standley, *et al.* (2000), where fecal steroids (including coprostanol), caffeine, consumer product fragrance materials, and petroleum and combustion byproducts were used to identify wastewater treatment plant effluent, agricultural and feedlot runoff, urban runoff, and wildlife sources. They studied numerous individual sources of these wastes from throughout the US. A research grade mass spectrophotometer was used for the majority of the analyses in order to achieve the needed sensitivities, although much variability was found when using the methods in actual receiving waters affected by wastewater effluent. This sophisticated suite of analyses did yield much useful information, but the analyses are difficult to conduct and costly and may be suitable for special situations, but not for routine survey work.

Another recent series of tests examined several of these potential emerging tracer parameters, in conjunction with the previously identified parameters, during a project characterizing stormwater that had collected in telecommunication manholes, funded by Tecordia (previously Bellcore), AT&T, and eight regional telephone companies throughout the country (Pitt and Clark 1999). Numerous conventional constituents, plus major ions, and toxicants were measured, along with candidate tracers to indicate sewage contamination of this water. Boron, caffeine, coprostanol, *E. coli*, enterococci, fluorescence (using specific wavelengths for detergents), and a simpler test for detergents were evaluated, along with the use of fluoride, ammonia, potassium, and obvious odors and color. About 700 water samples were evaluated for all of these parameters, with the exception of bacteria and boron (about 250 samples), and only infrequent samples were analyzed for fluorescence. Coprostanol was found in about 25 percent of the water samples (and in about 75% of the 350 sediment samples analyzed). Caffeine was only found in very few samples, while elevated *E. coli* and enterococci (using IDEXX tests) were observed in about 10% of the samples. Strong sewage odors in water and sediment samples were also detected in about 10% of the samples. Detergents and fluoride (at  $>0.3$  mg/L) were found in about 40% of the samples and are expected to have been contaminated with industrial activities (lubricants and cleansers) and not sewerage. Overall, about 10% of the

samples were therefore expected to have been contaminated with sanitary sewage, about the same rate previously estimated for stormwater systems.

Additional related laboratory tests, funded by the University of New Orleans and the EPA (Barbe', *et al.* 2000), were conducted using many sewage and laundry detergent samples and found that the boron test was a poor indicator of sewage, possibly due to changes in formulations in modern laundry detergents. Laboratory tests did find that fluorescence was an excellent indicator of sewage, especially when using specialized "detergent whitener" filter sets, but was not very repeatable. We also examined several UV absorbance wavelengths as sewage indicators and found excellent correlations with 228 nm, a wavelength having very little background absorbance in local spring waters, but with a strong response factor with increasing strengths of sewage.

Table 5 summarizes the different measurement parameters discussed above. We recommend that our originally developed and tested protocol, as reported by Pitt, *et al.* (1993), still be used as the most efficient routine indicator of sewage contamination of stormwater drainage systems, with the possible addition of specific *E. coli* and enterococci measurements and UV absorbance at 228 nm. The numerous exotic tests requiring specialized instrumentation and expertise do not appear to warrant their expense and long analytical turn-around times, except in specialized research situations, or when special confirmation is economically justified (such as when examining sewer replacement or major repair options).

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**Table 5. Comparison of Measurement Parameters used for Identifying Inappropriate Discharges into Storm Drainage**

Parameter Group	Comments	Recommendation
Fecal coliform bacteria and/or use of fecal coliform to fecal strep. ratio	Commonly used to indicate presence of sanitary sewage.	Not very useful as many other sources of fecal coliforms are present, and ratio not accurate for old or mixed wastes.
Physical observations (odor, color, turbidity, floatables, deposits, stains, vegetation changes, damage to outfalls)	Commonly used to indicate presence of sanitary and industrial wastewater.	Recommended due to easy public understanding and easy to evaluate, but only indicative of gross contamination, with excessive false negatives (and some false positives). Use in conjunction with chemical tracers for greater sensitivity and accuracy.
Detergents presence (anionic surfactant extractions)	Used to indicate presence of wash waters and sanitary sewage.	Recommended, but care needed during hazardous analyses (only for well-trained personnel). Accurate indicator of contamination during field tests.
Fluoride, ammonia and potassium measurements	Used to identify and distinguish between wash waters and sanitary sewage.	Recommended, especially in conjunction with detergent analyses. Accurate indicator of major contamination sources and their relative contributions.
TV surveys and source investigations	Used to identify specific locations of inappropriate discharges, especially in industrial areas.	Recommended after outfall surveys indicate contamination in drainage system.
Coprostanol and other fecal sterol compounds	Used to indicate presence of sanitary sewage.	Possibly useful. Expensive analysis with GC/MSD. Not specific to human wastes or recent contamination. Most useful when analyzing particulate fractions of wastewaters or sediments.
Specific detergent compounds (LAS, fabric whiteners, and perfumes)	Used to indicate presence of sanitary sewage.	Possibly useful. Expensive analyses with HPLC. A good and sensitive confirmatory method.
Fluorescence	Used to indicate presence of sanitary sewage and wash waters.	Likely useful, but expensive instrumentation. Rapid and easy analysis. Very sensitive.
Boron	Used to indicate presence of sanitary sewage and wash waters.	Not very useful. Easy and inexpensive analysis, but recent laundry formulations in US have minimal boron components.
Pharmaceuticals (colibric acid, aspirin, ibuprofen, steroids, illegal drugs, etc.)	Used to indicate presence of sanitary sewage.	Possibly useful. Expensive analyses with HPLC. A good and sensitive confirmatory method.
Caffeine	Used to indicate presence of sanitary sewage.	Not very useful. Expensive analyses with GC/MSD. Numerous false negatives, as typical analytical methods not suitably sensitive.
DNA profiling of microorganisms	Used to identify sources of microorganisms	Likely useful, but currently requires extensive background information on likely sources in drainage. Could be very useful if method can be simplified, but with less specific results.
UV absorbance at 228 nm	Used to identify presence of sanitary sewage.	Possibly useful, if UV spectrophotometer available. Simple and direct analyses. Sensitive to varying levels of sanitary sewage, but may not be useful with dilute solutions. Further testing needed to investigate sensitivity in field trials.
Stable isotopes of oxygen	Used to identify major sources of water.	May be useful in area having distant domestic water sources and distant groundwater recharge areas. Expensive and time consuming procedure. Can not distinguish between wastewaters if all have common source.
<i>E. coli</i> and enterococci bacteria	More specific indicators of sanitary sewage than coliform tests.	Recommended in conjunction with chemical tests. Relatively inexpensive and easy analyses, especially if using the simple IDEXX methods.

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