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Tracing Sewage Contamination in Urban Tropical Coastal Waters

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Abstrak

Umumnya masyarakat kurang memberikan perhatian terhadap kontaminasi limbah (cair) domestik di perairan pantai perkotaan. Hal ini karena terutama karena kontaminasi limbah domestik umumnya dilaporkan tidak terdeteksi. Pemanfaatan bio-indikator untuk mendeteksi kontaminasi limbah di perairan pantai perkotaan mempunyai permasalahan karena tekanan lingkungan. Untuk dapat memahami dengan lebih baik kontaminasi limbah domestik di perairan pantai perkotaan tropis, 48 sampel sedimen permukaan dasar perairan dan sampel air diambil pada musim Timur (Juli 1997) dan musim Barat (Pebruari 1998). Koprostanol, fecal sterol, telah dianalisis untuk mengetahui kontaminasi limbah domestik di perairan pantai perkotaan tropis. Analisis mencakup: a) kandungan koprostanol, b) karakteristik sedimen, dan c) coliform bacteria. Hasil analisis menunjukan bahwa pemanfaatan koprostanol dapat dengan jelas menunjukkan bahwa perairan pantai Semarng terkontaminasi limbah domestik, dimana bila menggunakan bio-indikator fecal coliform tidak terdeteksi.

Kata kunci: *pantai, kontaminasi, koprostanol, dan limbah domestik.*

Abstract

Most people give less attention on sewage contamination in urban coastal waters. It is mainly because sewage contamination in coastal waters mostly was reported not detected. Using bio-indicator for detecting sewage contamination in urban coastal waters has problems because of the environmental stress. To better understand of sewage contamination in urban tropical coastal waters, 48 surface bottom sediment and water samples of Semarang coastal waters were collected during East monsoon (July 1997) and West monsoon (February 1998). Coprostanol, a fecal sterol, had been analyzed to understand the sewage contamination in urban tropical coastal waters. The analyses included: a) coprostanol contain, b) sediment characteristics, and c) coliform bacteria. The results show that by using coprostanol it was clearly defined that Semarang coastal waters was contaminated by sewage, which by using bio-indicator, fecal coliform, could not be detected.

Key words: *coastal, contamination, coprostanol, and sewage.*

Introduction

Urban runoff and domestic/industrial waste effluents represent the most important contaminant sources in nearshore aquatic systems. In addition, most nearshore aquatic systems adjacent to urban centers are characterized by contaminated fine sediment that are brought in from a variety of point sources, such as streams and storm/sanitary sewer outfalls (Coakley, *et al.* 1992). The detection of sewage pollution in the environment is of considerable importance for health, aesthetic, and ecological reasons. Sewage contamination is usually determined by enumeration of *fecal coliform* bacteria. In the last two decades, various workers have questioned the reliability of the coliform test as an adequate indicator of sewage contamination, especially in urban coastal waters. This is mainly due to the lack of knowledge concerning bacterial die-off rates particularly in saline waters (Bartlett 1987). It is mostly because of osmotic affect (Manahan 1994). In urban coastal waters, environmental stress is increase because affected by several factors: a) the increase of salinity from freshwater to seawater, b) increase of industrial wastes that are toxic and heated, c) low dissolved oxygen, and d) chlorination of wastewater (Walker *et al.* 1982, Bartlett 1987).

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To solve those problems, various alternative indicators were proposed as indicator of sewage contamination. Several researchers proposed coprostanol as a chemical indicator of sewage pollution (Hatcher *et al.* 1977, Hatcher and McGillivary 1979, Brown and Wade 1984, Düreth *et al.* 1986, Holm and Windsor 1990, Coakley and Poulton 1991, Coakley *et al.* 1992, Bachtiar 1993, Bachtiar *et al.* 1996, Jeng and Han 1994, Jeng *et al.* 1996, Chan *et al.* 1998, Bachtiar 2002). Coprostanol (5*b*-Cholestan-3*b*ol) is produced by specific sources. It is the major fecal sterol of human, comprising 40 to 60 % of the total neutral sterols excreted (Walker *et al.* 1982). Coprostanol is also detected in feces of mammals and chicken, but was not found in intestinal content of marine animals. Therefore it is very specific indicators whose presence in marine environment uniquely confirms recent or continuing sewage contamination.

Beside the source of indicator have to be specific, the indicator have to be able be determined quantitatively and relatively conservative. As an organic material, coprostanol is degradable in the environment. Many studies show that coprostanol is promise indicator of sewage contamination (Kirchmer 1971, Dutka *et al.* 1974, Hatcher *et al.* 1977, Hatcher and McGillivary 1979, Brown and Wade 1984, Düreth *et al.* 1986, Holm and Windsor 1990, Coakley and Poulton 1991, Coakley *et al.* 1992, Bachtiar 1993, Bachtiar *et al.* 1996, Jeng and Han 1994). Most of the studies have been done at temperate region in fresh water environment. Using coprostanol as a sewage contamination indicator in urban tropical coastal waters, especially in Indonesia, is still poorly understood. Semarang coastal waters were used as a study area of typical urban coastal waters of Indonesia.

Material and Methods

Sample collection

In July 1997, during East monsoon (dry season), 19 surface bottom sediments and water samples were collected in Semarang coastal waters using van Veen grab sampler and van Dron water sampler a long a radial grid centered on the mouth of Banjir Kanal Timur, Semarang (Figure 1). In February 1998, during West monsoon (rainy season), sample collection was repeated at the same stations. In addition, three riverine bottom sediments (S-20, S-21, and S-22) were collected for identifying the sources. The position of sampling positions were located using hand held GPS and an Echo Sounder was used to determine bottom topography of study area. The data of sampling station are given in Table 1. Two surface bottom sediments as control were collected from Moro Demak (control-1), a small town, about 45 km East of Semarang, and from Karimunjawa Island (control-2), National Marine Park, about 60 km Northeast of Semarang.

To ensure that only the modern sediment layer was sampled, the sediment collected was limited from the top to 3-4 cm of surface sediment. The grab sampler was carefully used to ensure that the surface was as undisturbed as possible. Before sub-sampling, careful descriptions of the sample were made. Only if the samples showed indication of non-disturbed condition, such as a surface brown oxidized skin, were the top 3-4 cm collected. Sample were put in dark bottles and stored in a cooler on the boat during fieldwork. On return to the laboratory, the samples were kept at 5 ^{0}C before further preparation for analysis.

To better understand the sewage contamination, five samples of water and sediments were collected in Banjir Kanal Timur (BKT), Kali Tenggang (KT), Kali Tambak Lorok (KTL), and river's mouth for bacteria coliform analysis using Most Probable Number method.

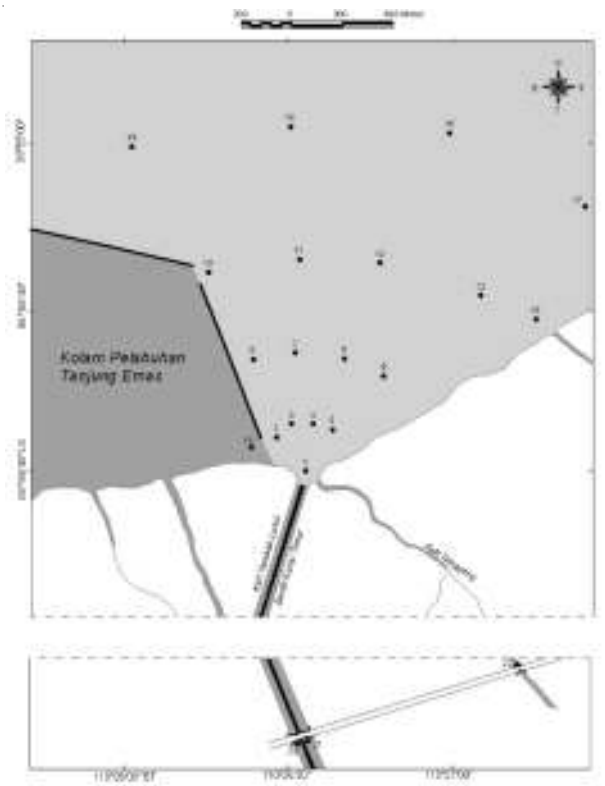

Figure 1. Banjir Kanal Timur Semarang coastal waters and the positions of sampling stations.

Table 1. Sampling site positions

No	Sample ID	Sampling Site Position		Distance from the Center Point (m)	Depth (m)
		Latitude	Longitude		
1	S-1	6° 56′ 24.5"	110° 26′ 36,5"	0	1.3
2	S-2	6° 56′ 12.5"	110° 26′ 31,1"	315	1.8
3	S-3	6° 56′ 9.3"	110° 26′ 35,5"	360	1.8
4	S-4	6° 56′ 8,4"	110° 26′ 42,3"	405	1.9
5	S-5	6° 56′ 12,5"	110° 26′ 48,0"	495	1.7
6	S-6	6° 56′ 2,2"	110° 26′ 26,8"	863	2.6
7	S-7	6° 55′ 58,2"	110° 26′ 35,0"	885	2.7
8	S-8	6° 55′ 58,0"	110° 26′ 46,2"	975	2.8
9	S-9	6° 56′ 3,0"	110° 26′ 56,8"	1,073	2.1
10	S-10	6° 55′ 35,9"	110° 26′ 15,8"	1,515	4.6
11	S-11	6° 55′ 29,5"	110° 26′ 33,5"	1,643	4.3
12	S-12	6° 55′ 29,5"	110° 26′ 56,7"	1,178	3.6
13	S-13	6° 55′ 42,5"	110° 27′ 16,5"	1,875	2.2
14	S-14	6° 54′ 46,6"	110° 25′ 54,5"	2,528	6.6
15	S-15	6° 54′ 40,0"	110° 26′ 30,5"	1,673	6.2
16	S-16	6° 54′ 45,8"	110° 27′ 12,5"	2,745	5.2
17	S-17	6° 55′ 6,5"	110° 27′ 51,5"	3,188	3.3
18	S-18	6° 55′ 42,5"	110° 27′ 38,0"	2,693	0.7
19	S-19	6° 56′ 19,0"	110° 26′ 23,3"	465	1.5
20	S-20	6° 57′ 50,5"	110° 26′ 29,4"	2,880	1.1
21	S-21	6° 57′ 49,8"	110° 26′ 33,0"	2,880	0.8
22	S-22	6° 57′ 35,0"	110° 27′ 14,5"	2,610	0.9
23	Control-1		Moro Demak	-	1.1
24	Control-2		Karimunjawa Island	-	0.9

Table 2. The Results of Coprostanol Analysis

No	Sample ID	Coprostanol concentration ($mg.g^{-1}$)	
		East Monsoon (July 1997) (Dry Season)	West Monsoon (February 1998) (Rainy Season)
1	S-1	1.150	1.213
2	S-2	1.470	*lost*
3	S-3	0.712	1.901
4	S-4	0.820	0.957
5	S-5	0.840	0.951
6	S-6	1.631	1.914
7	S-7	6.150	4.916
8	S-8	7.119	1.764
9	S-9	2.342	1.688
10	S-10	1.160	1.180
11	S-11	9.368	8.260
12	S-12	9.440	4.299
13	S-13	3.809	4.732
14	S-14	15.870	16.544
15	S-15	12.710	8.501
16	S-16	6.038	5.316
17	S-17	4.130	1.158
18	S-18	1.870	1.290
19	S-19	2.750	2.814
20	S-20	15.050	2.921
21	S-21	20.699	3.567
22	S-22	17.079	3.927
23	M D	1.400	0.700
24	KRJ	0.201	*na*

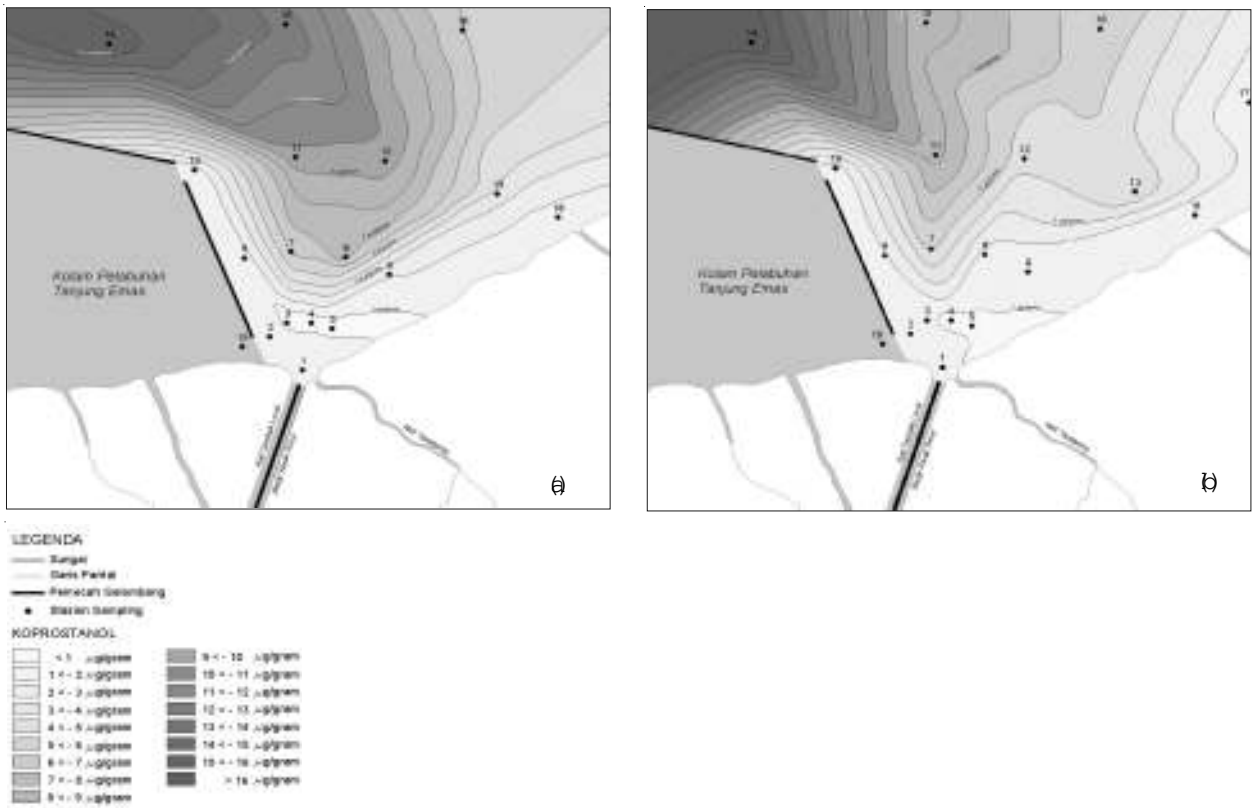

Figure 2. (a) Pattern distribution of coprostanol in the sediments of Semarang Coastal waters in July 1997 (East monsoon), and (b) in February 1998 (West monsoon).

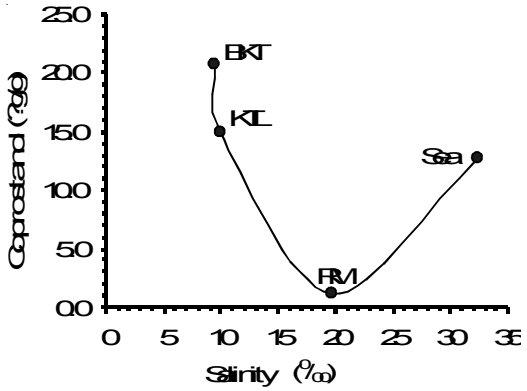

Figure 3. Coprostanol concentration in rivers (BKT and KTL), river's mouth (RM), and the sea.

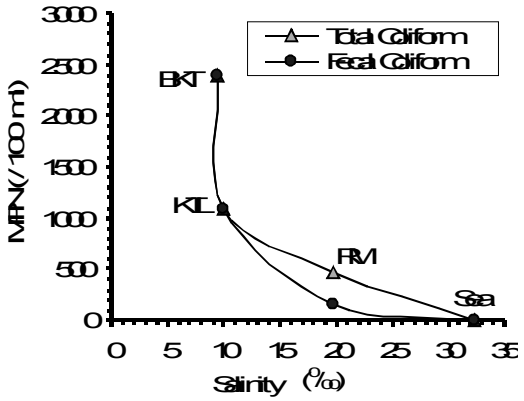

Figure 4. *Coliform* concentration in rivers (BKT and KTL), river's mouth (RM), and the sea.

Table 3. The Results of Coprostanol and *Coliform* Bacteria Analyses

No.	Station	Coprost. conct. in sediment (mg/g)	Coprost. conct. in water (mg/g)	MPN Total *Coliform* (/100 ml)	MPN Fecal *Coliform* (/100 ml)	Temp. (°C)	Sal. (‰)	DO (mg/l)	pH
1.	BKT	20.70	-	>2,400	>2,400	28.6	10.0	3.7	8.0
2.	KTL	15.05	-	1,100	1,100	28.2	9.4	4.6	7.8
3.	KT	n.a	n.a	n.a	n.a	28.7	9,6	3.9	8.2
4.	RM	1.15	-	460	150	29.7	19.7	6.3	7.9
5.	Sea	12.71	-	-	-	30.8	32.4	6.7	8.1

Note: BKT = Banjir Kanal Timur KT = Kali Tenggang
KTL = Kali Tambak Lorok RM = River's Mouth
n.a = not available

Coprostanol analysis

The analytical method of coprostanol analysis in sediment samples was combined between the method that had been used by Bachtiar *et al.* (1996) and Jeng & Han (1994). All solvent used were HPLC grade. All reagents employed were reagent grade, and all glassware was distilled. The analytical procedure is as follow:

- *Soxhlet extraction*

  About 20 to 30 g of dry sediment (depend on the grain size) were extracted with a mixture of benzene and methanol (1:1, v/v) in a Soxhlet apparatus for 24 h. Heptadecanol (C17:OH) was added to the extract as an internal standard.

- *Saponification*

  The spiked extract was concentrated and saponified with 0.5 N methanolic KOH (0.5 N KOH in 95:5 methanol/H_2O (%)). The neutral lipid was extracted with n-hexane four times.

- *Column chromatography*

  The extracted lipid was fractionated by silica gel (deactivated with 5% water) column chromatography. The less polar lipids were removed by elution with 40% hexane in chloroform, and the sterol-containing fraction was isolated using 10% methanol in chloroform. The fractions were collected in 9.5-dram vials. All samples were stored refrigerated until gas chromatography (GC) analysis.

- *Sample preparation for GC*

  The isolated sterol were concentrated to near dryness, and latter were transferred to HP septum-capped vial using 2 x 0.5 ml heptane. BSTFA (bis(trimethylsilyl)-trifluoro-acetamide) (100 :1), was added, and the samples were heated at 130°C for 15 minutes to make them more responsive on the GC capillary column. After cooling, the samples were ready for GC analysis.

- *Gas Chromatograph*

  Analysis of coprostanol was carried on a Hitachi 263-50 gas chromatography equipped with SE-30 column and Flame Ionization Detector (FID). Nitrogen was used as the carrier gas. The injector

and detector were set at 300°C. Oven temperature was programmed as follow 150°C – 280°C at 5°C min^{-1}. Coprostanol concentration was calculated based on relative response factor (RRF) from a reference solution containing 6 mg coprostanol and 6.9 mg reference standard (C18:OH). RRF was determined using the following formula:

$$RRF = \frac{mg\ copros.(std)}{area\ copros.\ (std)} \times \frac{area\ C18:OH\ (std)}{mg\ C18:OH\ (std)}$$

From (1), coprostanol concentration in samples can be determined using the following formula:

$$\mu g\ coprostanol = \frac{RRF \times area\ copros. \times \mu g\ IS}{area\ IS}$$

The precision of the method was determined by five replicate analyses of the same samples that gave a mean coprostanol value of 2.28 $mg.g^{-1}$ with the relative standard deviation was 6.4%. In addition to coprostanol, total organic contain (TOC) and grain size of sediment were also determined. The existence of coliform bacteria in four stations was analyzed to compare with coprostanol.

Results and Discussion

The results of coprostanol analysis were listed on Table 2. The data show that coprostanol was detected in all of the sediment samples, but not detected in all of water samples. There are two possibilities that coprostanol was not detected in water samples. First, the amount of suspended material was less than 5 g (1.5-2.7 g), the minimum amount required. Second, the accuracy of the equipment used was only in $mg.g^{-1}$ scale, therefore low concentration could not be detected. Afterward, the results were plotted in the base map of study area to understand the pattern distribution of contaminated sediments (Figure 2a and 2b).

Based on the pattern distribution of coprostanol in the sediments of Semarang coastal waters, it is clearly defined that Semarang coastal waters was contaminated by sewage, with low concentration in the area which close to the shoreline, and increased with increasing the distance from shoreline. Both in East and West monsoon, the contaminated sediments were distributed to the Northwest.

Bachtiar *et. al.* (1996) found different pattern distibution of coprostanol in the sediment of Hamilton Harbour. They found that the concentration of coprostanol decrease with increasing the distance from the outfall of Burlington Skyway STP. Right on the out fall, coprostanol was 934.75 $mg.g^{-1}$, and decrease to 4.78 $mg.g^{-1}$ at 2.512 m from the outfall. They also found that high coprostanol concentration could be found in coarser sediments if the distance was relatively close to the source. Those conditions in Hamilton Harbour are totally different with the conditions of Semarang coastal waters. Hamilton Harbour area is relatively closed area with only connected to Lake Ontario by ship canal, and the source of coprostanol is point source (outfall of Skyway STP). Semarang coastal water is open waters with non point source of coprostanol.

The data of field measurement of water quality parameters and the results of coprostanol and *coliform* analyses of four stations were listed in Table 3. and plotted in Figure 3 and Figure 4. The data show that coprostanol could not be detected in water samples, but in all of sediment samples coprostanol was detected, with relatively high concentration in river environment, and decrease in river mouth, and return to high concentration in sea environment. Based on characteristic of the sediments, the high concentration of coprostanol was found in fine sediments, and low concentration was found in more coarse sediments. Based on that facts, it seems to be clear that coprostanol come from the rivers.

Conclusions

Based on the results of this study, coliform bacteria show some disadvantage as an indicator in the environment with high environmental stress. It is necessary to use alternative indicator to understand sewage contamination in the urban tropical coastal waters, where usually as the environments with high environmental stress. Coprostanol has performance as a promise alternative indicator of sewage contamination in the environments with high environmental stress. It is very important to do extended research to develop coprostanol not only as an indicator of sewage contamination, but also to be the indicator of sewage pollution.

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