AN ABSTRACT OF THE THESIS OF Joanne Margaret Collins for the Masters of Science in Biology presented April 4, 1995.

Title: Occurrence and Fate of Fecal Pollution Indicator Bacteria in the Sediments of Tumon Bay, Guam.

Approved: _____
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Storm water runoff in the Tumon Bay area is routed into ponding basins and percolation fields to avoid direct drainage into coastal waters. Runoff was found to contain high numbers of fecal indicator bacteria (6.1 Log 10 fecal coliforms or enterococci 100 mL^{-1}). This study was conducted to investigate the occurrence of fecal indicator bacteria in Tumon Bay, to enumerate them over a one year period and determine if their occurrence was due to runoff contaminated with fecal matter leaching into underlying porus bedrock and then being transported by ground water out into coastal sediments.

Water and sediments were sampled monthly during 1993 from holes dug in the supra-tidal beach down to brackish water, and from the bay 5 m out from the low tide mark. Sediments were sampled for the presence of fecal indicators and potential pathogens. Salinity, silica and nitrate+nitrite data were also collected.

Bacterial enumerations on mEnterococcus, mFC, EMB, SS, and TCBS showed that indicators occurred more frequently during the rainy season (June - December). Water quality parameters indicated that ground water entering Tumon Bay was often diluted with storm water runoff. Increases in the frequency of occurrence and densities of bacteria enumerated from the sediments coincided with greater than predicted levels of Si.

In vitro laboratory experiments showed that the fecal bacteria *Streptococcus faecalis, Escherichia coli, Klebsiella pneumoniae*, and *Enterobactera cloacae* survived longer when they were not exposed to sunlight. The bacteria were capable of extended survival (2-7 days) in sediments compared to the overlying water column where they are exposed to sunlight.

TO THE OFFICE OF GRADUATE SCHOOL AND RESEARCH

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OCCURRENCE AND FATE OF FECAL POLLUTION INDICATOR BACTERIA IN THE SEDIMENTS OF TUMON BAY, GUAM.

BY
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INTRODUCTION

Both total coliform (TC) and fecal coliform (FC) bacteria have been traditionally used as indicators of fecal pollution and by inference, the potential health risk of swimming in polluted fresh and marine waters. Recently, another indicator group the enterococci (Ent) have been adopted by some government agencies in the United States, New Zealand, Australia, and Canada as a result of epidemiological studies conducted by the United States Environmental Protection Agency (USEPA) (Cabelli 1983, Cabelli et al. 1983). These studies suggest that enterococci are better correlated with disease risk than either total or fecal coliforms. Presently in the United States and U. S. territories, if the enterococci plate counts from marine waters are greater than 35 colony forming units (CFU) 100 mL^{-1}, the waters are not recommended for recreational use.

However, gastroenteritis and similar ailments may still occur due to swimming in and eating fish from waters that may still harbor true pathogens, although the waters contain fewer than the maximum allowable level of indicator contamination. The converse is also true; one does not always become ill from recreational use of waters that are above the recommended bacterial contamination limit. Further, indicator and pathogenic bacteria may also occur in locations other than the routinely tested surface waters, such as sediments and therefore can be an undetected health risk.

Indicator and potentially pathogenic bacteria can occur in aquatic sediments at densities several times higher than in overlying waters (Collins

1992, Grimes 1975, Matson et al. 1978, Shiaris et al. 1987). Matson (1993a) reported that high numbers of indictor bacteria were commonly found in 46% of the cases in the sediments underlying Guam's recreational waters that were indicator-free. The densities in the sediments at some locations were often sufficiently high (12% to 94% of the time) to pose a potential public health risk if they were resuspended into the water column. In many of these areas the source of fecal contamination was easily determined; polluted rivers, storm drains emptying directly into the bay, and a broken sewer line that ran along the beach. However, in Tumon Bay (one of Guam's busiest recreational areas) there was no obvious source for the high densities of indicator bacteria found in the sediments. Matson (1993a) found that at three sites along Tumon bay the sediments had sufficient densities of indicator bacteria so as to cause over-limit densities to occur in the overlying waters 12, 25 and 24% of the time if the sediments had been resuspended. Water quality monitoring strategies, however, presently do not include the enumeration of indicators in the sediment. This study was conducted in order to systematically investigate over a full year the occurrence of these indicator bacteria in the sediments of Tumon Bay, and to test whether their source was storm water contaminated with fecal matter.

Guam's street runoff contains pollutants such as oil, grease, detergents, and the traditional indicators (i.e., total and fecal coliforms) of potentially pathogenic bacteria (Zolan et al. 1978a, b). Collins (1992) reported average

densities of FC (19 x 10^6 100 mL^{-1}) and Ent (14 x 10^6 100 mL^{-1}) in street runoff (*n* = 6). These levels are equivalent to those found in Guam's sewage plant effluents.

Recently however, direct drainage of storm water into Tumon Bay has been minimized in order to improve the aesthetic value of the bay. Most storm water from the area is now being collected in ponding basins and percolation fields. These basins are large holes (~100 m^2 x 2-10 m deep) in the ground that are periodically dredged to remove the buildup of vegetation and mud. Percolation fields which resemble large leaching fields, consist of a series of interconnected perforated pipes buried in gravel below the ground, and are used to dispose of street runoff. Ponding basins and percolation fields have been constructed throughout the northern limestone plateau of Guam.

Runoff collected in ponding basins and percolation fields, and that which makes its way onto unpaved ground, leaches down into the porous limestone, then mixes with ground water and is transported by the ground water as it makes its way out into the bay. I speculate that the underground transport of contaminated runoff by ground water is largely responsible for the occurrence of the indicators and potential pathogens in the bay's waters and sediments.

Ground water from Guam's northern aquifer discharges continuously into the periphery of northern Guam, a process that is obvious on shore at low tides when the brackish water can be seen running down the intertidal sand to the ocean. Strong negative correlation of NO*x* (i.e., NO_3^- + NO_2^-) and Si with

salinity indicates that, in many places, the mixing of aquifer water with seawater occurs in the coastal transition zone rapidly, thoroughly and without any biogeochemical modification (Matson 1993b). In northeast Tumon Bay between 1987-1990, aquifer water was high in both NO*x* and Si (average 114 µM and 28 µM respectively) (Matson 1993b), whereas at salinities of > 34 ‰ Tumon Bay seawater has very little of either (NO*x* ≈ 0.05 - 5 µM, Si ≈ 2 - 8 µM) (Matson 1991). Matson (1993b) calculates that aquifer water leaks out around the 57 km perimeter of northern Guam at rates of 2.2 - 110 m^3 (m of shoreline)$^{-1}$ d^{-1}.

Because the carbonate soils lack organic exchange sites that bind microorganisms (Zolan et al. 1978a, b) and there is a continuous flow of ground water, it is possible that indicator and other fecal bacteria are rapidly transported out into the bay where they may survive and reproduce in the nutrient rich waters and sediments (Burton et al. 1987, Carrillo et al. 1985, Hardina and Fujioka 1991, Hood and Ness 1982, Kaysner et al. 1987, LaLiberte and Grimes 1982, Lopez et al. 1987, Pettibone et al. 1987, Rhodes and Kator 1988, Sinton et al. 1993, 1994, Vasconcelos and Swartz 1976). However, studies of the persistence of fecal bacteria in the aquatic environment often produce contradictory results because research is conducted in different geographical locations and under various environmental conditions. Factors such as water temperature (Evison 1988, Rhodes and Kator 1983, Solić and Krstulović 1992), insolation (Chamberlin and Mitchell 1978, Kapuscinski and Mitchell 1983, Sinton et al. 1994), pH (Curtis et al 1992, Solić and Krstulović 1992), salinity (Anderson

et al. 1979, Evison 1988, Hanes and Fragala 1967, Solić and Krstulović 1992), and nutrient levels (Evison 1988) all effect the survival of indicators and fecal bacteria, and these factors vary seasonally and geographically.

The proposed underground transport of fecal bacteria out into the near shore sediments may protect them from exposure to harmful UV light (Borja and Wood 1986, Curtis et al. 1992, Davies and Evison 1991) and thus further extend their survival and ability to reproduce. Thus, studies were also undertaken to investigate the potential survival of indicator and fecal bacteria in Tumon Bay sediments.

MATERIALS AND METHODS

Study Area

The northern half of Guam is comprised mainly of upraised limestone and lacks the rivers of the largely volcanic southern province. This bedrock structure provides for the occurrence of an extensive aquifer system, from which water seeps out along the northern coasts (Matson 1993b, Mink 1976, Zolan et al. 1978a).

Tumon Bay is in the west central part of Guam, Mariana Islands (Fig. 1). The village of Tumon has a commercially developed coastal area, with numerous large hotels, shops, commercial and residential buildings. The brackish water "seeps" and springs where ground water leaks through cracks and fissures are evident year round along the entire 3.2 km of Tumon Bay shoreline (Matson 1993b, Zolan et al. 1978a). Storm water runoff from roads, including Marine Drive (Australian Cable to Ypao Rd.) and Sanvitores Blvd. (Fig. 1), are collected in one of three ponding basins and two percolation fields along the Tumon Bay coast.

From January to December 1993, four sites were studied, two of which, YP1 and YP2, were located at the southwestern end of Tumon Bay at Ypao Beach Park. This area has two ponding basins that are situated approximately 300 m inland from the mean high water (MHW) and which receive runoff from the surrounding roadways. The two other sites, TR1 and TR2, were located at

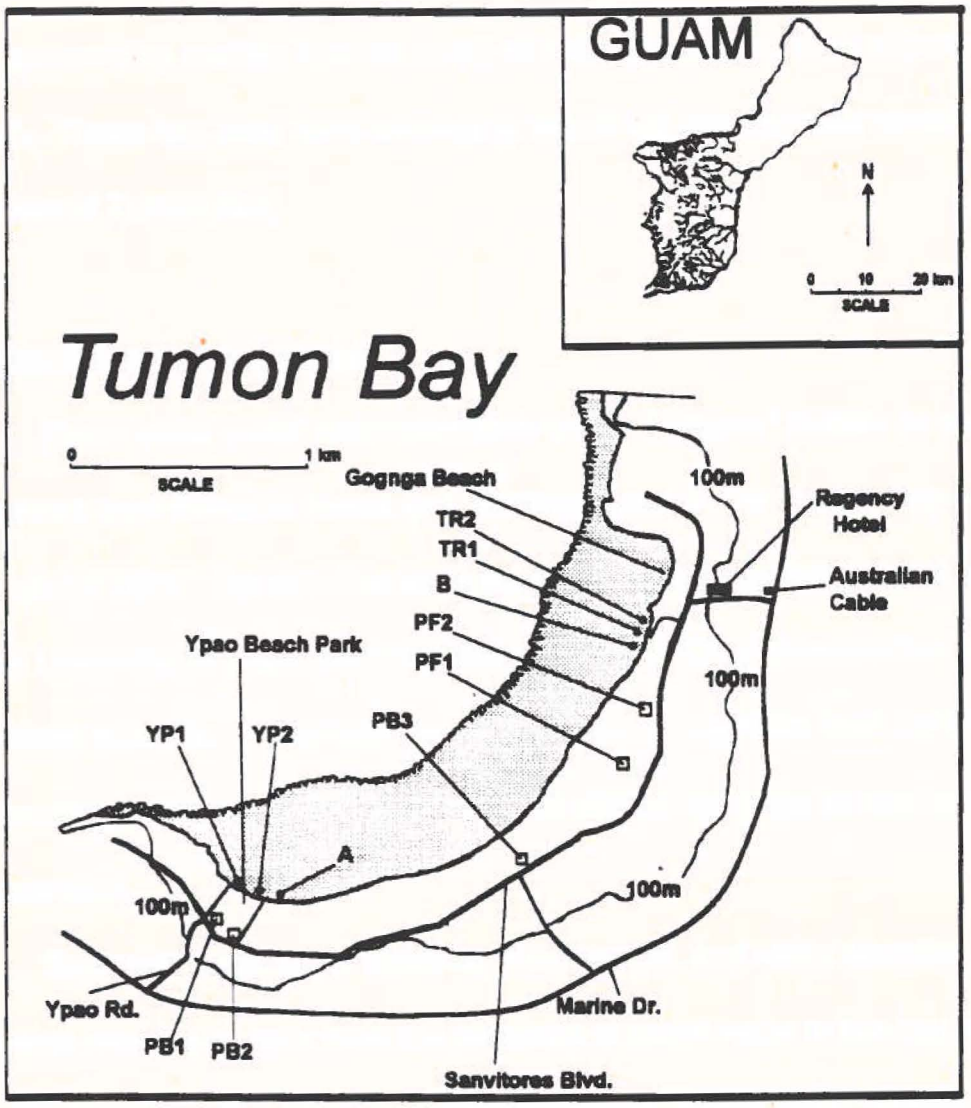

Figure 1. Map of Guam with Tumon Bay study area enlarged. Sites of ponding basins (PB), percolation fields (PF), the four sample sites (YP1, YP2, TR1, TR2), and site of flourescein studies (A, B) are shown.

the opposite northeastern end of Tumon Bay, 100 m and 70 m south of Gognga Beach (Fig. 1). The TR site area did not have either a ponding basin or percolation field. However, during this study, storm waters from Sanvitores Blvd. and surrounding parking lots ran freely off onto an unpaved area adjacent to the beach. These four sites were situated at two of the most popular recreational beaches of Tumon Bay.

Sediment and Water Sampling

Storm water runoff was collected in sterile Nalgene® bottles on several occasions from roadside drains and ponding basins in the Tumon Bay area throughout 1993. The samples were kept at ambient temperature and returned to the University of Guam Marine Laboratory for bacterial enumeration and water chemistry analysis. On several occasions sediment samples from the bottom of the ponding basins were collected in sterile Whirl-Pak® bags for bacteria enumeration.

Sediment samples for bacterial enumeration, and water samples for chemical analysis to determine the presence of storm water in the bay, were collected from the four sites at least once a month during 1993. At each site samples were collected from holes dug in the supra-tidal beach sand down to brackish water, and from the sub-tidal area 5 m out from the mean low tide level (Fig. 2). Sediment samples were collected from the bottom of the beach holes and stored in sterile Whirl-Pak® bags. Sub-tidal bay sediments were collected

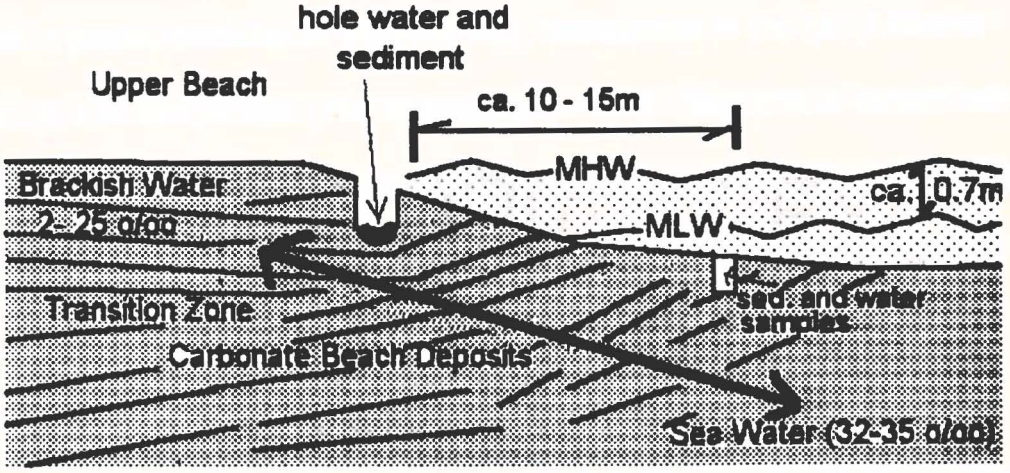

Figure 2. Idealized cross section through the beach at Tumon Bay showing ground water flow through the transition zone and out into the bay. Positions of hole and bay sample sites are also depicted.

with an 8 cm diameter and 40 cm long PVC core liner. The corer was pushed into the bay sediment, a hole was then dug by hand next to the core, and the ends were capped with rubber stoppers. The cores were then sectioned vertically at measured 3-5 cm intervals directly into sterile Whirl-Pak® bags in the field, and were returned to the University of Guam (UOG) microbiology laboratory at ambient temperature, where bacterial enumerations were processed within 6 h.

Samples for chemical analysis of beach hole and bay waters were collected in plastic scintillation vials (20 mL), transported to the UOG Marine Laboratory, and refrigerated prior to analysis (< 72 h). Beach hole waters were collected directly and pore waters were collected with a "spear" similar in design to that described by Corredor and Morell (1985). An 80 cm length of 3 cm diameter polyvinyl chloride (PVC) pipe closed at the top and covered at the bottom (obliquely angled) end with perforated plexiglass was pushed into the sediment to the required depths. Water samples were withdrawn from the spear using a 50 cm^3 syringe attached to aquarium tubing that was inserted into the side of the spear near the top and which ran to the plexiglass at the bottom.

Enumeration of Fecal Bacteria

Sediments from each site were diluted (10 g of wet sediment added to 90 mL of sterile seawater), shaken, and left to settle for 30 sec. Serial ten-fold dilutions were made with autoclaved seawater where necessary.

Fecal coliforms and enterococci were enumerated in duplicate by membrane filtration in accordance with accepted procedures (A.P.H.A. et al. 1992). Samples were filtered through sterile 0.45 μm HAWG white, gridded, Millipore® filters. For fecal coliform enumeration the filters were then placed in 60 mm culture dishes on sterile media pads saturated with 1.9 mL of mFC broth (Difco). Plates were incubated in a water bath at 44.5 °C for 24 h, and all blue colonies were counted. Enterococci were enumerated by placing filters on mEnterococcus agar (Difco), and incubated at 35 °C for 48 h. All pink and red colonies were counted.

Spread plates (0.1 mL) were used to isolate and enumerate suspected potential pathogens using three selective and differential media including: EMB agar for the detection of gram negative lactose fermenters, SS agar (Salmonella-Shigella, BBL) for the isolation of *Salmonella* and *Shigella* spp., and TCBS agar (Thiosulfate-citrate-bile-sucrose, Difco), to select for Vibrios, including *Vibrio cholera,* and other enteropathogenic vibrios. On several occasions, colonies were picked from spread plates, cultured and identified using the API 20E biochemical test strips and the analytical profiles for the identification of *Enterobacteriaceae* and other gram negative bacteria.

Evidence of Ground Water Transport

Salinity, Si, and NO*x* content in samples of beach hole and bay waters were measured to ascertain the amount of mixing of storm water with ground

water. Linear (least-squares) regression of NO*x* and Si levels (μM) with Cl^- levels (mM) in Tumon Bay water studied between 1987 and 1990 provided models for the dilution of these solutes due to conservative mixing of aquifer waters with coastal waters (Matson 1993b), where

$$\mu M\ NOx = -0.24 \times (mM\ Cl^{-1}) + 130,\ (R^2 = 0.93,\ n = 123),\ \text{and}$$

$$\mu M\ Si = -0.03 \times (mM\ Cl^{-1}) + 18,\ (R^2 = 0.76,\ n = 86)$$

Compared with ground water, storm water and surface runoff are much lower in NO*x* and higher in Si, while rain water is low in both. Thus, if the ground water entering Tumon Bay was diluted with storm water runoff, we would expect lower NO*x* and higher Si concentrations in the beach seeps than what was predicted from the model. Also, we would expect the simultaneous occurrence of indicators and other fecal bacteria.

Cl^- was measured with a Haake Buchler Chloridometer, model 442-5000, (relative precision ± 0.9%, or ± 0.31‰). Salinity was calculated from Cl^- using the following relationship,

$$\text{Salinity (‰)} = (mM\ Cl^- \div 550) \times 35$$

Full strength seawater (35‰) is defined as having 550 mM Cl^- (Stumm and Morgan 1974), $NO_3^- + NO_2^-$ (NO*x*) were analyzed with the spongy Cd-shaking method (Jones 1983), and Si with molybdate (Stainton et al. 1974).

To measure groundwater flow, fluorescein dye was placed in the water contained in the Ypao Beach ponding basin (PB1, Fig. 1), and the bay waters adjacent to the area were monitored. Water samples were taken at 1 h intervals

for 8 h, and twice daily for the next 4 days, to establish if the dye had flowed out into the bay along the ground water seeps. Water samples were scanned from 460 - 495 nm with a Beckman DU-65® spectrophotometer to detect fluorescein. In addition, two beach hole experiments were conducted at site A, 50 m north of Ypao Beach Park and at site B, 100 m south of the TR sites (Fig. 1) on separate occasions. Fluorescein was placed in beach holes dug 5 and 10 m above the MHW mark and down into the brackish water. This was done during an outgoing tide, and the adjacent bay waters were monitored. Sub-tidal bay waters were collected at 2 and 5 m intervals out from low tide hourly for the first 8 h and twice daily thereafter (morning and afternoon) for the following 3 days. Only visual monitoring was used at site B.

Survival of Indicators

Survival studies were conducted outside in the Marine Laboratory lanai in order to investigate survival of fecal indicator bacteria with and without sunlight and sediment. Microcosms, open boxes measuring 15 cm square (3375 cm^3) constructed of plexiglass, were placed in a flow-through seawater tank so that the water in the boxes was at ambient seawater temperature. The experimental design follows that described in Fujioka et al. (1981) and Sinton et al. (1994). Four commonly-occurring field isolates, *Enterobacter cloacae, Escherichia coli, Klebsiella pnuemoniae*, and *Streptococcus faecalis* were added at initial densities of ~10^8 100 mL^{-1} to separate tanks. The tanks were exposed to various

conditions including: (1) an enclosed tank that blocked sunlight, filled with sterile water (control), (2) sterile water and exposed to sunlight, (3) sterile water and sterile Tumon Bay sediment exposed to sunlight, to test for death by bactivory, and (4) non-sterile water and non-sterile Tumon Bay sediment, exposed to sunlight. Three liters of sterile filtered seawater were added to each tank, and, for those requiring sediment, 4 cm of Tumon Bay sediment were placed in the bottom. Sediment was sterilized by autoclaving.

Water and sediment samples were taken daily for 7 days and bacteria were enumerated using the spread plate technique on mEnterococcus and EMB agar. The spread plate technique can detect 1 CFU 0.1 mL^{-1} or 1000 CFU 100 mL^{-1}. Only *S. faecalis* and *E. coli* were studied in the non-sterile sediment and water tanks.

RESULTS

Enumeration of Fecal Bacteria

High numbers of indicator and potentially pathogenic bacteria were enumerated from storm water runoff (Table 1). Potential pathogens including *Escherichia coli*, *Enterobacter aerogenes*, *Klebsiella pneumonia*, *Salmonella* spp., *Pseudomonas aeruginosa* and *Serratia marcescens* were isolated from storm water runoff samples. Bacterial isolates identified from streak plates of storm water runoff, and beach hole and bay sediment suspensions, are given in Table 2. *E. coli*, *En. aerogenes*, *K. pneumonia*, and *Pseudomonas* spp. were the most common isolates from beach hole and bay sediments at all sites. During the rainy season, numbers of *Shigella* spp. and *Salmonella* spp. increased, but *Pseudomonas aeruginosa* was not isolated until after the rainy season had started. The only potentially pathogenic vibrios isolated and identified from the sediments were *Vibrio alginolyticus* and, on one occasion *Vibrio parahaemolyticus*, both were from the TR sites. All other *Vibrio* spp. isolated on TCBS that were also oxidase positive, gram negative rods, and several lactose fermenters on EMB were not able to be identified to species using the limited number of tests included in API 20E.

Table 1. Average Log 10 numbers of CFU per 100 mL of runoff water, or per gram of sediment, from street runoff and ponding basins of Tumon Bay, Guam.

Site	mEnt.	mFC	EMB	SS	TCBS
beach side sand volley ball	5.3	5.7	6.1	4.3	0.0
courts in front of TR1 site	5.3	5.7	6.1	4.3	0.0
	5.4	5.7	6.1	4.4	0.0
Ypao Park Rd. (east)	4.6	5.0	6.2	4.6	0.0
	4.6	5.0	6.1	4.6	0.0
	4.7	5.0	6.2	4.6	0.0
stormwater drain crn.	6.1	6.2	5.4	0.0	0.0
Sanvitores Blvd. and the	6.0	6.2	5.5	0.0	0.0
Regency Hill Rd.	6.1	6.3	5.4	3.0	0.0
	5.4	5.5			
	5.6	5.7			
beach bar car park (paved)	6.5	6.4	5.5	4.5	0.0
in front of TR sites	6.5	6.5	5.5	4.5	0.0
	6.4	6.5	5.3	4.4	0.0
Ypao Beach Rd. (west)	5.6	5.7	5.4	4.8	4.0
	5.6	5.7	5.4	4.9	4.0
	5.7	5.7	5.4	4.9	4.2
Sanvitores Rd. in front of	6.0	6.2	5.7	4.7	3.8
Ypao park	6.1	6.1	5.7	4.7	3.8
	6.0	6.0	5.7	4.8	0.0
beach bar car park (paved)	1.7		5.7	5.6	5.2
50 m south of TR	1.4		5.7	5.3	5.2
			4.5		
			3.8		
			3.5		
			3.5		

Table 1. (continued)

Site	mEnt	mFC	EMB	SS	TCBS
stormwater drain flowing into PB1	6.0 6.1	5.1 6.2			
Sanvitores Rd. in front of PB2	5.9 6.2 6.5	5.9 6.2 6.1			
Sanvitores Rd. in front of Signature Pub	5.5 5.3	5.4 6.2			
beach bar sand volley ball court 10 m south of TR1	5.3 7.0	5.2 7.0			
Sanvitores Rd. in front of the Sand Castle	5.9 6.2	5.9 6.2			
$\bar{x}$	6.1	6.1	5.7	4.8	4.3
n =	99	93	72	60	60
± 1 SD	6.3	6.3	5.7	5.0	4.7
Ypao ponding basin water	8.0 6.2 6.1	6.2 6.4 7.1	6.8 7.0 7.4	4.9 5.3 4.9	4.0 4.3 4.3
Ypao ponding basin sediment	4.0 5.3 3.8	3.5 4.4 3.4			

Table 2. Identification of bacteria isolated on EMB, SS, and TCBS agar spread plates.

media	identification	HOLE	BAY	RUNOFF	API ID.
EMB					
	Citrobacter spp.	0	0	1	good
	Enterobacter spp.	0	0	1	poor
	Enterobacter cloacea	2	5	3	acceptable - excellent
	En. agglom.	1	0	1	poor
	En. sakazakii	0	0	1	excellent
	Escherichia coli	12	7	3	good - excellent
	Klebsiella pnuemoniae	3	4	4	very good - excellent
	Past. aerogens	0	1	0	poor
	Pres. Ps. pseudomollei	1	0	1	poor
	Psuedomonas spp.	3	2	1	acceptable - excellent
	Pseudomonas aeurginosa	2	3	1	good - excellent
	Ps. pytrefaciens	1	1	0	excellent
	Seratia marcescens	1	0	0	very good
	no identification	17	21	6	
SS					
	Chrom. freundii	0	0	1	excellent
	K. pnuemoniae	0	0	1	very good
	Pseudomonas aeurginosa	0	0	1	good
	Seratia marcescens	1	0	0	very good
	Salmonella spp.	1	1	1	excellent
	Shigella spp.	2	0	0	very good
	no identification	0	0	0	
TCBS					
	Vibrio alginolyticus	2	0	0	excellent
	V. parahaemolyticus	0	1	0	good
	non pathogenic, no ID:				
	non-marine vibrios (grew 0 % NaCl only)	3	2	3	
	marine vibrios (grew in >3%)	9	16	0	
	grew 3% NaCl only	3	2	0	
	total tested	64	66	30	

The average daily rainfall for Guam 1993 was calculated from a 7 day running average of three sites and is shown in Figure 3. The rainy season started in early June. The results from bacterial enumerations from Tumon Bay beach hole sediments and in the top 0-5 cm of bay sediments, performed throughout the year on 5 differential and selective media, including; mEnterococcus, mFC, EMB, SS, and TCBS, show a general increase in density in the later part of the year which coincides with the rainy season (Figs. 4, 5, 6, 7, 8). Data from bacterial enumerations of Tumon Bay sediment and water chemistry analysis are given in the Appendix.

Paired linear regressions were performed between the bacterial numbers in sediment enumerated on mEnterococcus, mFC, EMB, SS, and TCBS, and the cumulative rainfall between 0-14 days prior to each collection date. Results showed no significant correlation between bacterial numbers and rainfall. There was also no significant correlation between cumulative rainfall and water chemistry (NO*x* and Si). However, correlations between bacterial numbers and daily rainfall, (calculated from 2-7, 14 and 21 day running average of daily rainfall prior to collection) were significant. The running average of the previous 4 days rainfall and bacteria enumerated on mFC, EMB and SS were positively correlated at $p = 0.01$. Enterococci and numbers on TCBS showed no significant correlation with average rainfall. There were positive correlations between NO*x* and running average rainfall of 2 days ($p = 0.05$) and 3 to 5 days

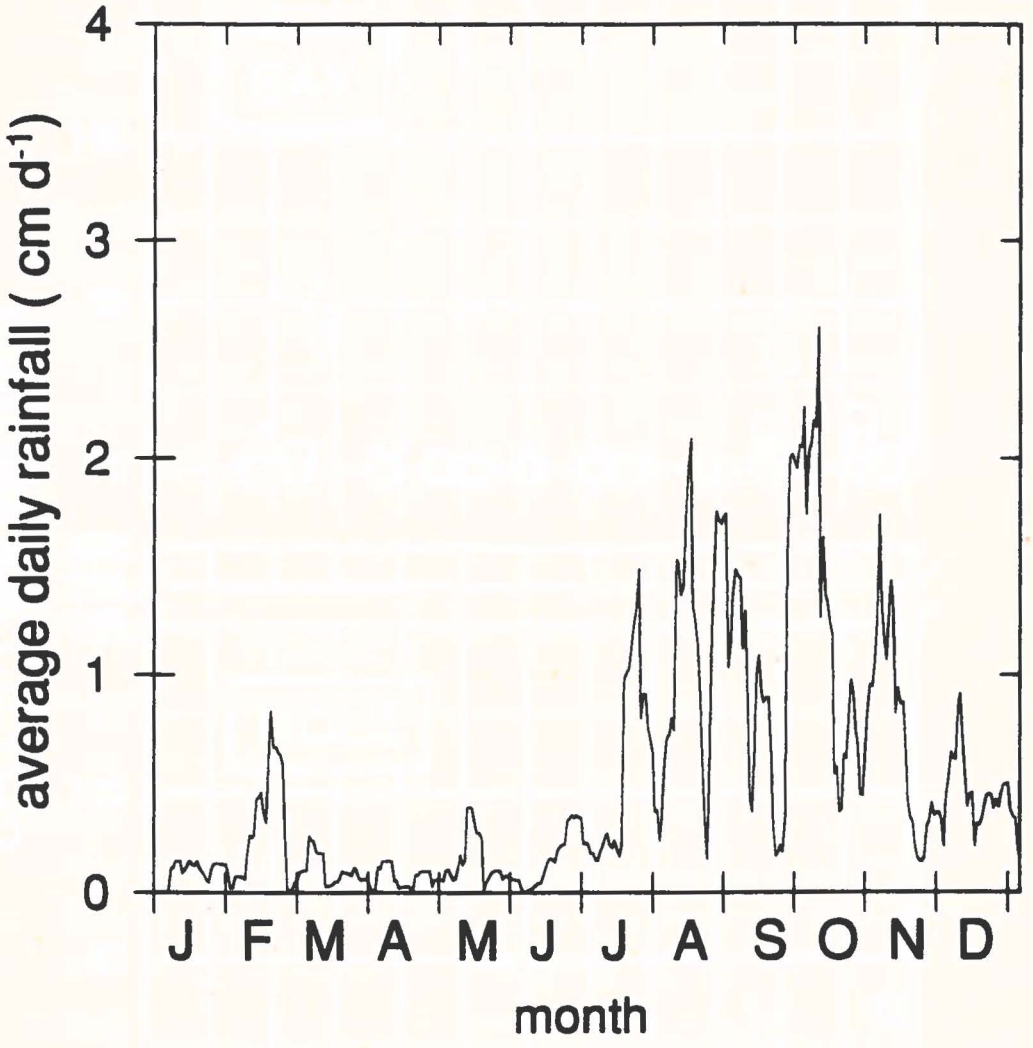

Figure 3. Seven day running average of daily rainfall taken from 3 sites (Naval Air Station, Nimitz Hil, and Anderson Air Force Base) between January and December 1993.

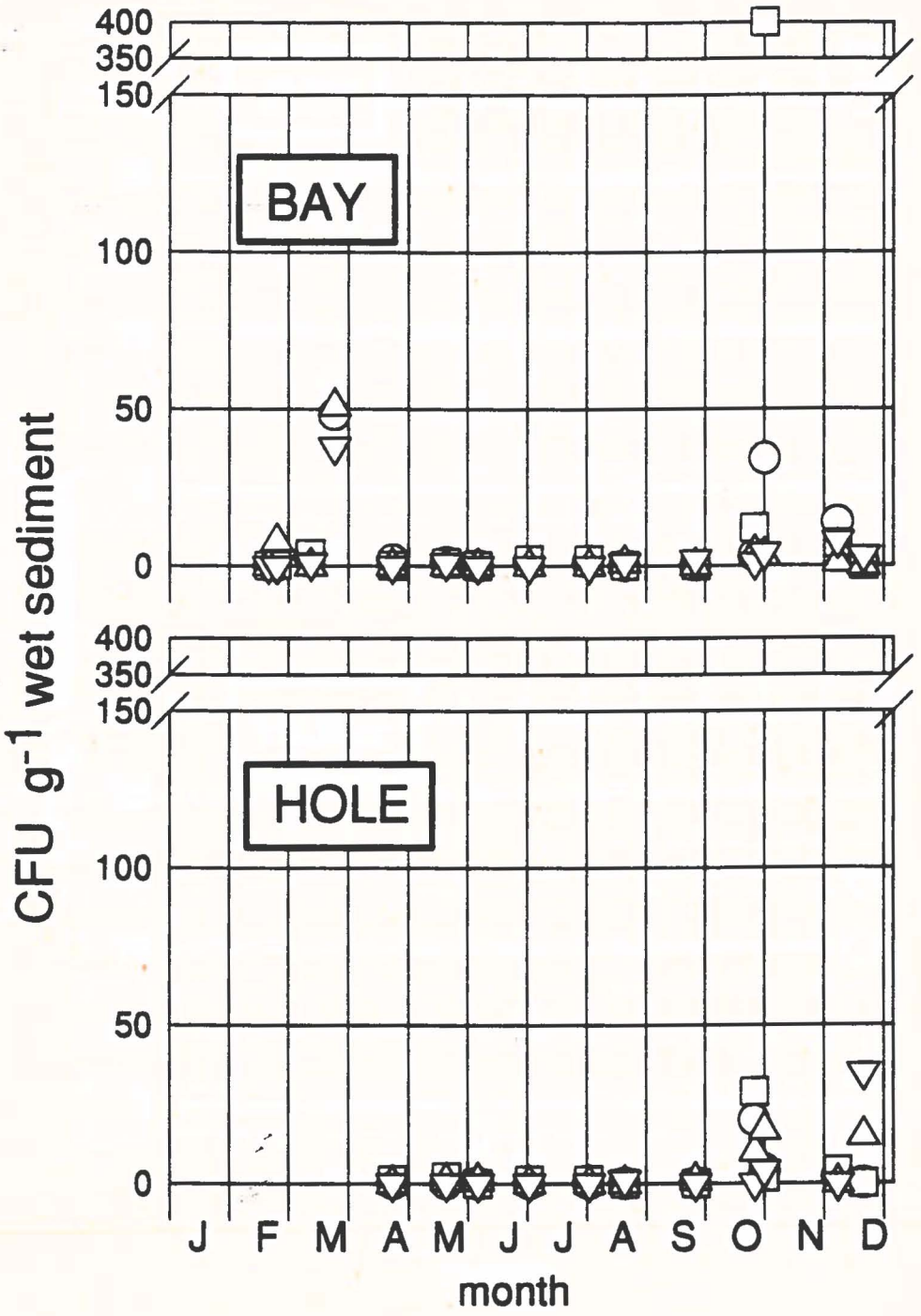

Figure 4. Enterococci per gram of bay and beach hole sediment. ○ YP1, □ YP2, △ TR1, ▽ TR2.

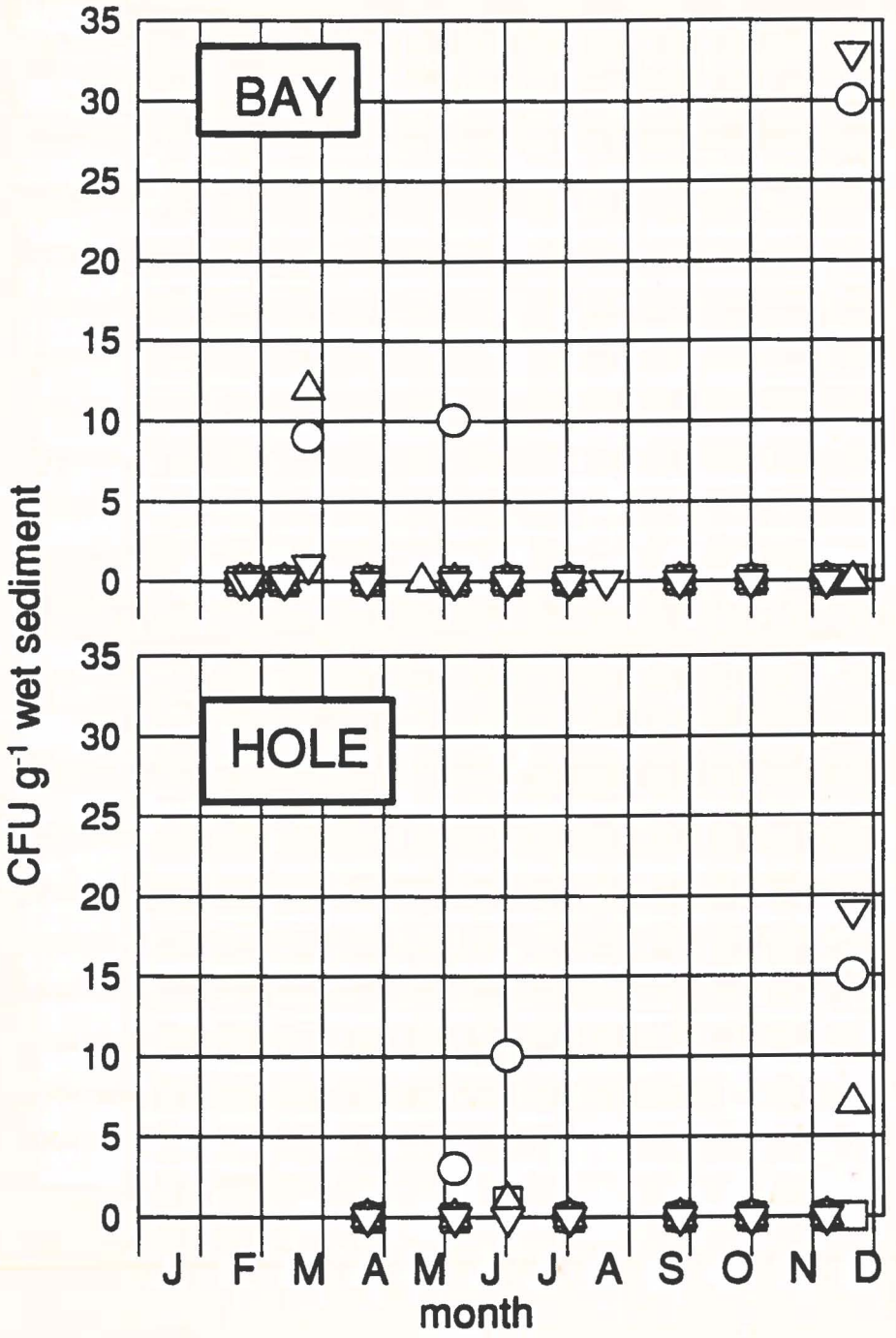

Figure 5. Fecal coliforms per gram of bay and beach hole sediment. ○ YP1, □ YP2, △ TR1, ▽ TR2.

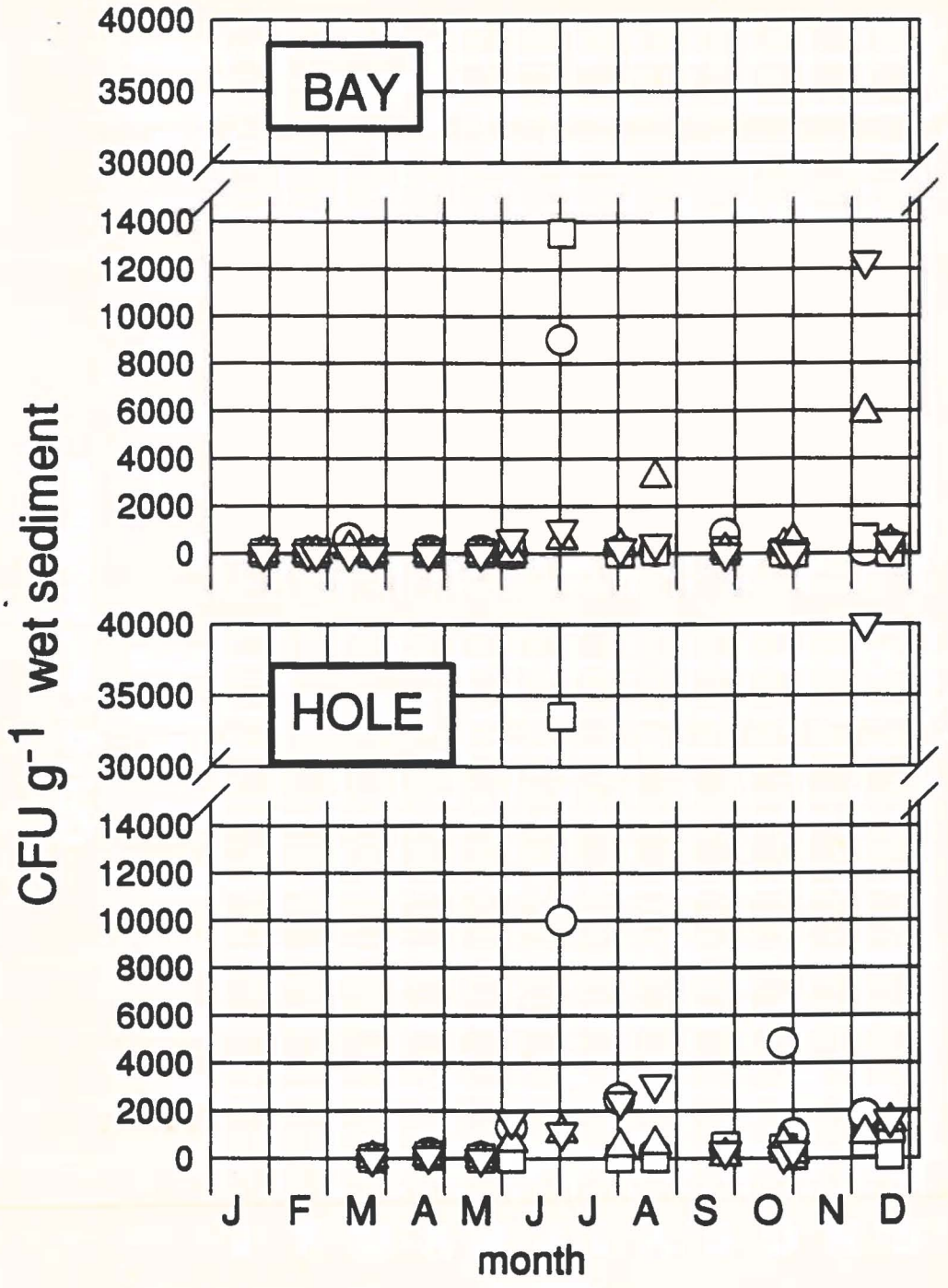

Figure 6. Lactose fermenters on EMB (coliforms) per gram of bay and beach hole sediment. ○ YP1, □ YP2, △ TR1, ▽ TR2.

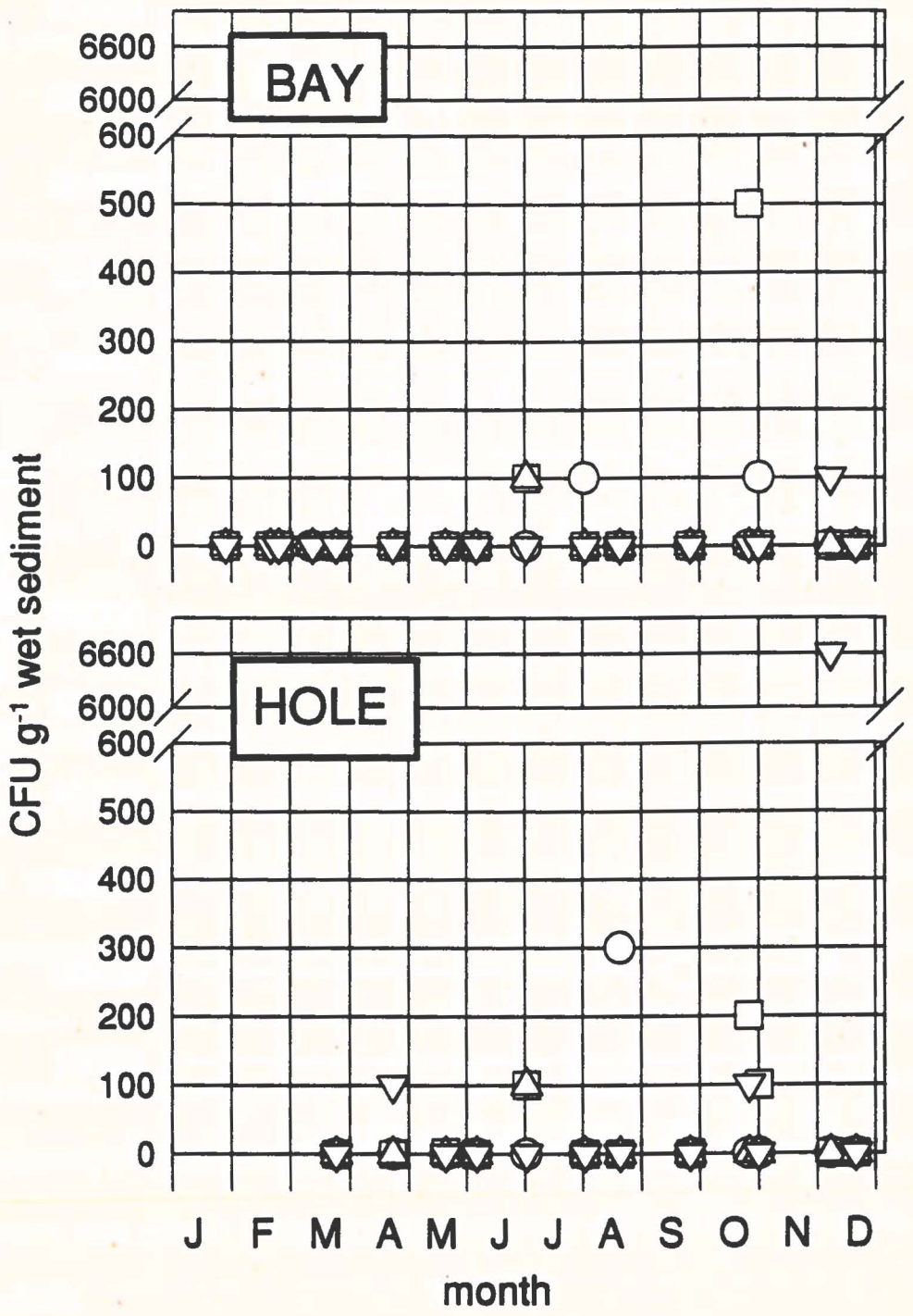

Figure 7. *Salmonella* and *Shigella* spp. per gram of bay and beach hole sediment. ○ YP1, □ YP2, △ TR1, ▽ TR2.

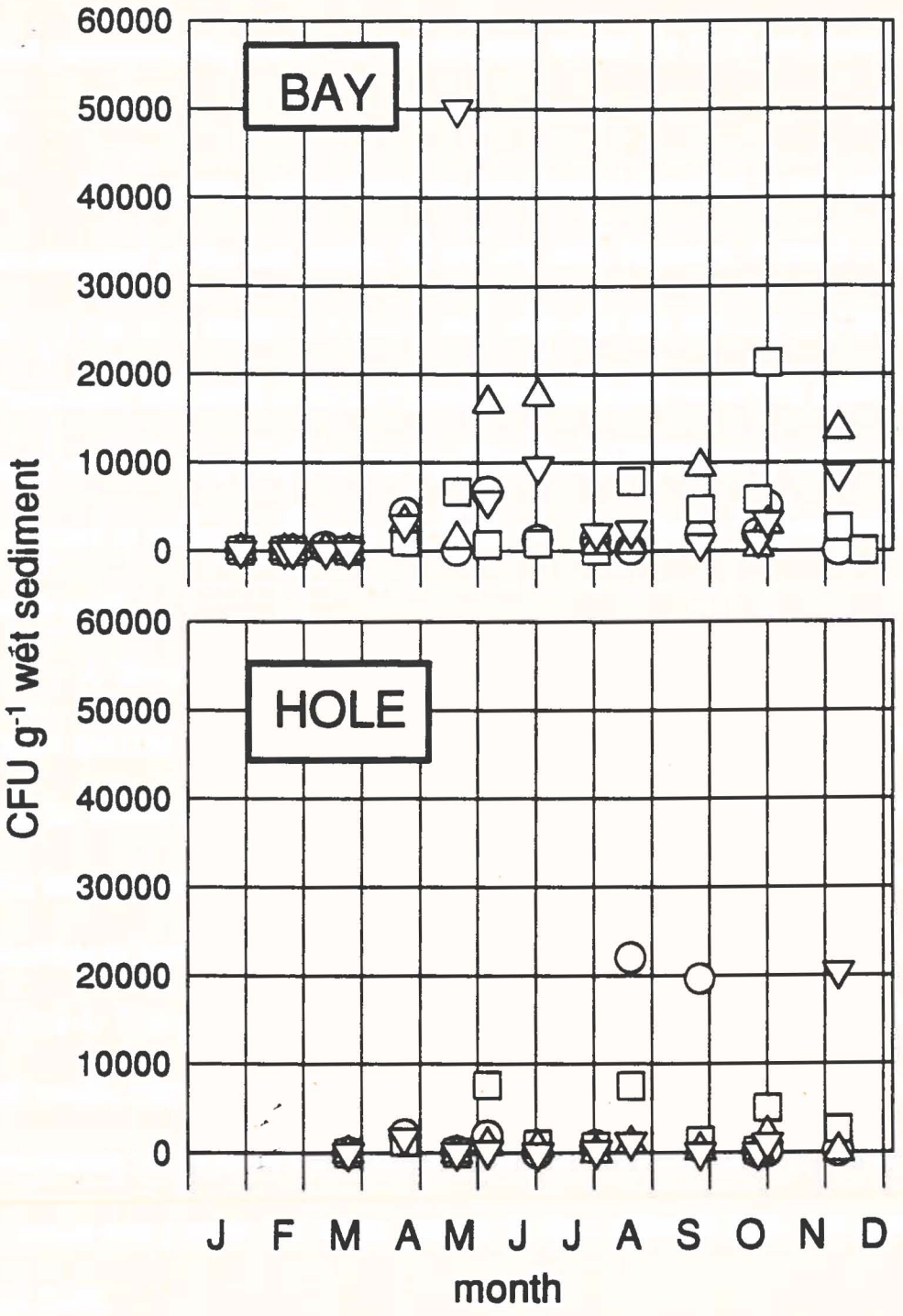

Figure 8. *Vibrio* spp. per gram of bay and beach hole sediment. ○ YP1, □ YP2, △ TR1, ▽ TR2.

($p = 0.01$). The densities of enterococci and fecal coliforms in the sediments and corresponding 4 day running average rainfall for 1993 are shown in Figure 9.

Comparisons of the seasonal averages of densities in sediments, enumerated on the five media, for dry and rainy seasons, showed an increase during the rainy season for both beach hole and bay sediments (Table 3). Parametric statistical analysis for multiple comparisons among pairs of means such as the Tukey-Krammer method (Sokal and Rohlf 1981) could not be used because of the large difference in variance. Therefore, non-parametric statistics were used.

A binomial sign test (+ or - frequency) resulted in a probability of $p = 0.001$ that the greater frequency of bacteria in the rainy season, isolated on all 5 media, would have occurred by chance, indicating that the increase density in the rainy season is significant for both hole and bay samples. A summary of the number of times sediments were sampled and bacteria were found is given in Table 4. The percentage of times bacteria were isolated from the sediments, on all media, increased during the rainy season for all cases. The increase in the frequency of bacterial occurrence, from that in the dry season to that in the rainy season was greater in the bay sediments than in beach hole sediments, except on EMB.

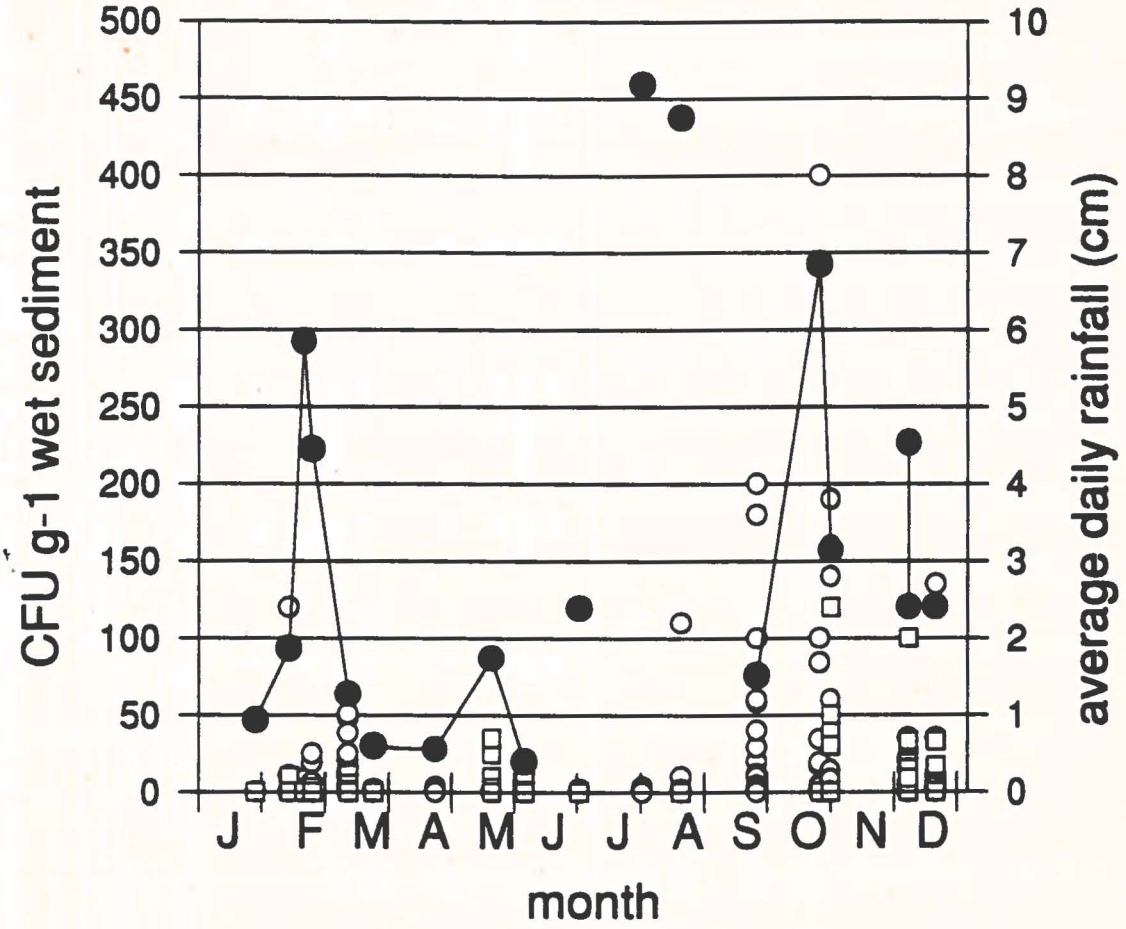

Figure 9. Fecal indicator densities in Tumon Bay sediments and 4 day running average of daily rainfall. ○ Enterococci, □ fecal coliforms, and ● rain.

Table 3. A comparison of seasonal averages of colony forming units (CFU) per gram of whole wet sediment from all Tumon Bay Sites

	Enumeration Media									
	m Entercoccus		m FC		EMB		SS		TCBS	
	dry	rainy	dry	rainy	dry	rainy	dry	rainy	dry	rainy
bay sediment										
$\bar{x}$	6	15	1	3	36	1400	0	28	2500	4400
± 1 SD	15	66	3	8	120	3300	0	88	9600	5600
variance	200	4300	12	69	14000	460000000	0	7600	89000000	30000000
n =	27	36	24	29	27	36	27	36	27	33
beach hole sediment										
$\bar{x}$	0	10	0	3	20	4100	4	210	2000	2700
± 1 SD	1	24	1	8	65	13000	29	1100	14000	6000
variance	0	550	1	62	3900	170000000	760	1200000	190000000	36000000
n =	12	36	8	28	12	36	12	36	12	32

Table 4. Occurrence of indicator bacteria in the sediment.

	enumeration media									
	m Enterococcus		m FC		EMB		SS		TCBS	
	dry	rainy	dry	rainy	dry	rainy	dry	rainy	dry	rainy
bay sediment										
# times present	12	25	4	6	4	25	0	6	10	30
# times sampled	27	36	24	29	27	36	27	36	27	36
% of time present	44	69	17	21	15	69	0	17	37	83
beach hole sediment										
# times present	4	19	1	10	6	33	2	7	8	26
# times sampled	12	36	8	28	12	36	12	36	12	32
% of time present	33	53	19	36	50	92	17	19	67	81

Ground Water Transport Evidence

Observed concentrations of NO*x* and Si in waters from beach holes and bay surface and pore waters are plotted relative to their salinity, along with the regression line (± 99% confidence intervals, C.I.) for the concentration values predicted by the conservative mixing model (Figs. 10 and 11). For the NO*x* data, 46% were below and 29% were greater than the 99% C.I. of what was predicted, and the remaining 25% were within the 99% C.I. (Fig. 10). For all Si data a similar trend was observed, 46% were below predicted, 25% of the samples were greater, and 29% were within the 99% C.I. of the model (Fig. 11). We expected the percentages of Si above and below those predicted to be opposite to what we found for NO*x*. Because this was not found beach hole water data alone were looked at, where, because of the constant water flow it was expected that conservative mixing of ground water and seawater would occur.

When beach hole NO*x* data and salinity data were plotted (Fig. 12) approximately one half (56%) of the water NO*x* concentrations were below the 99% C.I. of the model, while one third (34%) were above the predicted concentrations. The other 10% fell within the 99% CI. For the same beach hole water samples, the opposite trend was observed for Si, 60% had concentrations greater than predicted and approximately one third (36%) were below (Fig. 13).

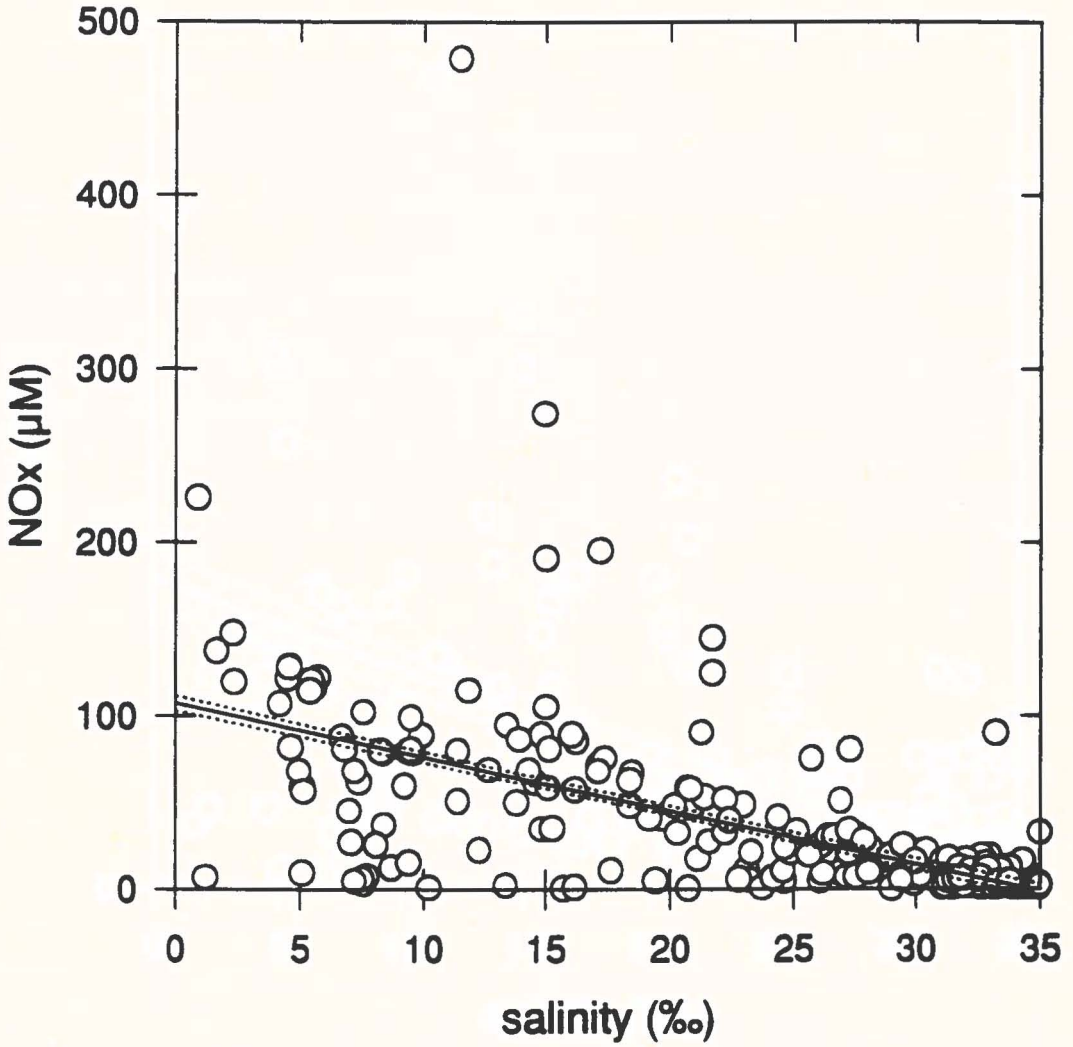

Figure 10. Beach hole and bay water NO*x* concentrations *vs.* salinity. Regression line (± 99% confidence intervals) of model shown.

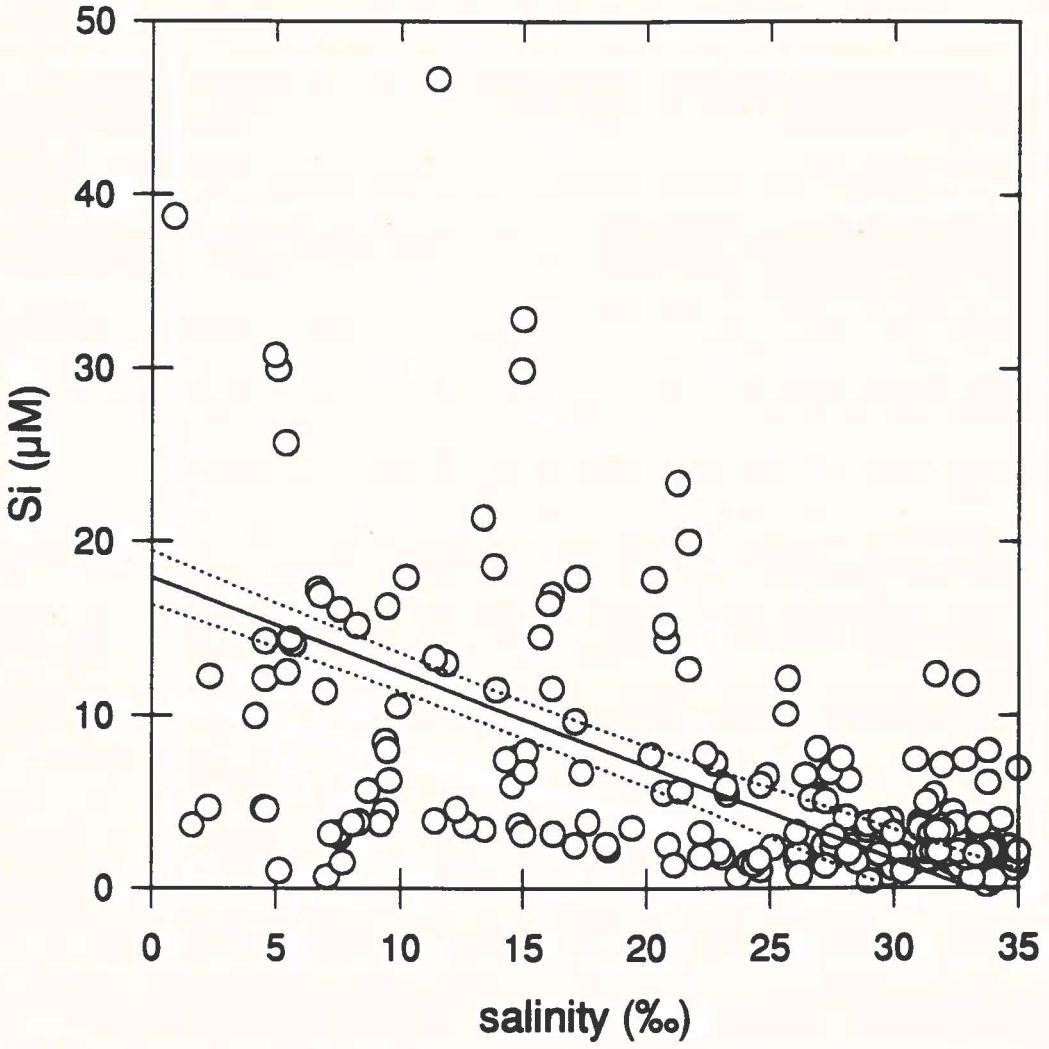

Figure 11. Beach hole and bay water Si concentrations *vs.* salinity. Regression line (± 99% confidence intervals) of model shown.

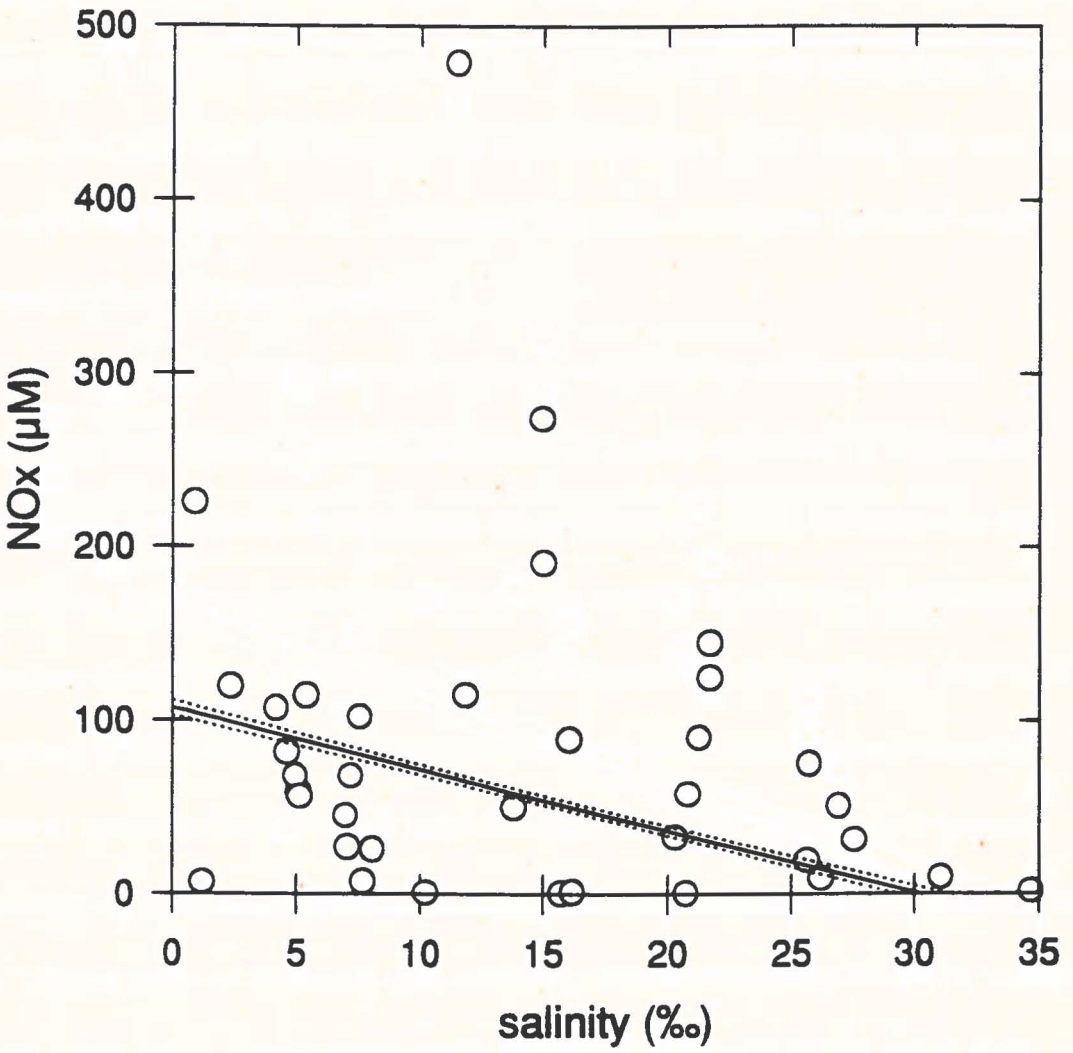

Figure 12. Beach hole NO*x* concentrations *vs.* salinity of beach hole water. Regression line (± 99% confidence intervals) of model shown.

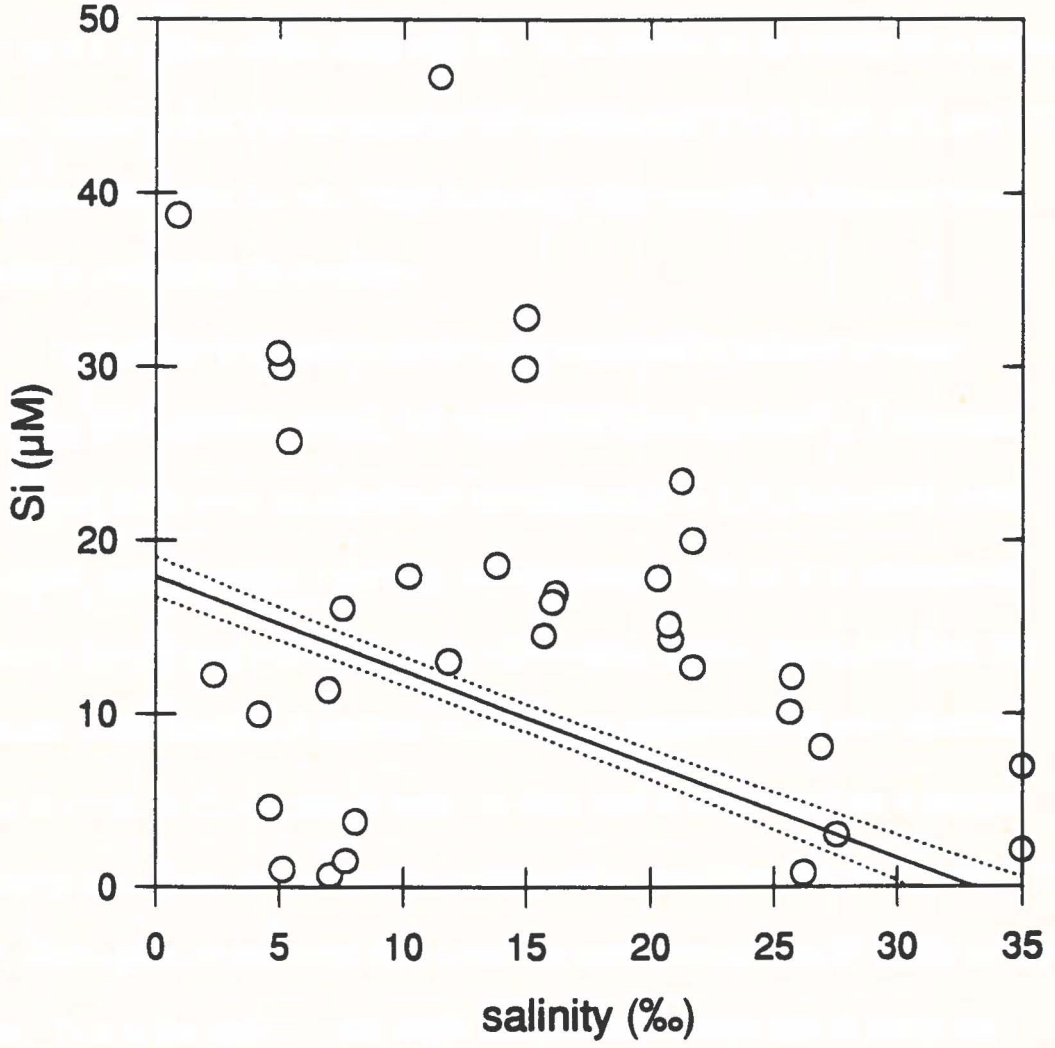

Figure 13. Beach hole Si concentrations *vs.* salinity of beach hole water. Regression line (± 99% confidence intervals) of model shown.

It was also expected that there would be some relationship between bacterial densities and water chemistry; a negative correlation with salinity and NO*x*, and a positive relationship with Si. Summaries of the relationship between water chemistry and the densities on mEnterococcus, mFC, EMB, SS, and TCBS are given in Figures 14 -18. There was very little correlation between water chemistry and bacterial densities.

Correlation analyses were strongly weighted by outliers of water chemistry and bacterial density, but when they were removed (± 3 standard deviations) there were no significant correlations ($p = 0.05$) between the two. Therefore, non-parametric techniques were used to test for any relationship between density and water chemistry (Chi-square G test, Sokal and Rolf 1981) because of the number of outliers and frequent occurrence of zero densities. In order to conduct Chi-square G tests the data had to be sorted into 4 groups. Bacterial data were transformed to represent the densities that would occur 100 mL^{-1}, if one gram of sediment was uniformly resuspended into 50 cm of overlying water. This is the approximate depth of water from which GEPA takes their water samples. Bacteria were then recorded as being above or below the maximum recommended contamination levels: < or > 33 CFU 100 mL^{-1} for mEnterococcus, and < or > 200 CFU 100 mL^{-1} for mFC, EMB, SS and TCBS. Counts were then taken of the number of times the data would fall into one of four groups: (1) bacterial numbers < the recommended contamination levels when water Si levels were < predicted, (2) < recommended bacterial

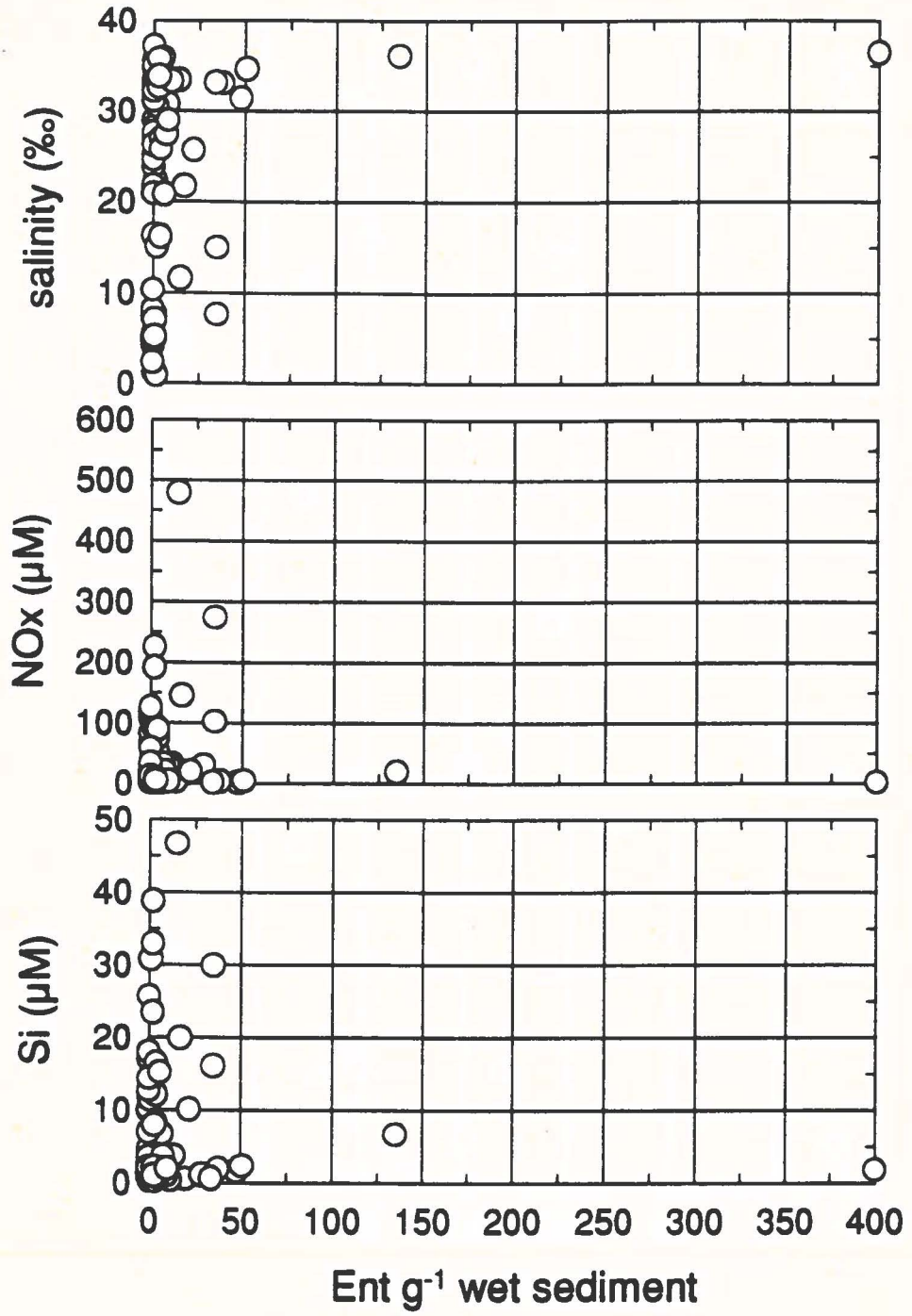

Figure 14. Salinity, nitrate + nitrite, and silica *vs.* enterococci per gram wet sediment.

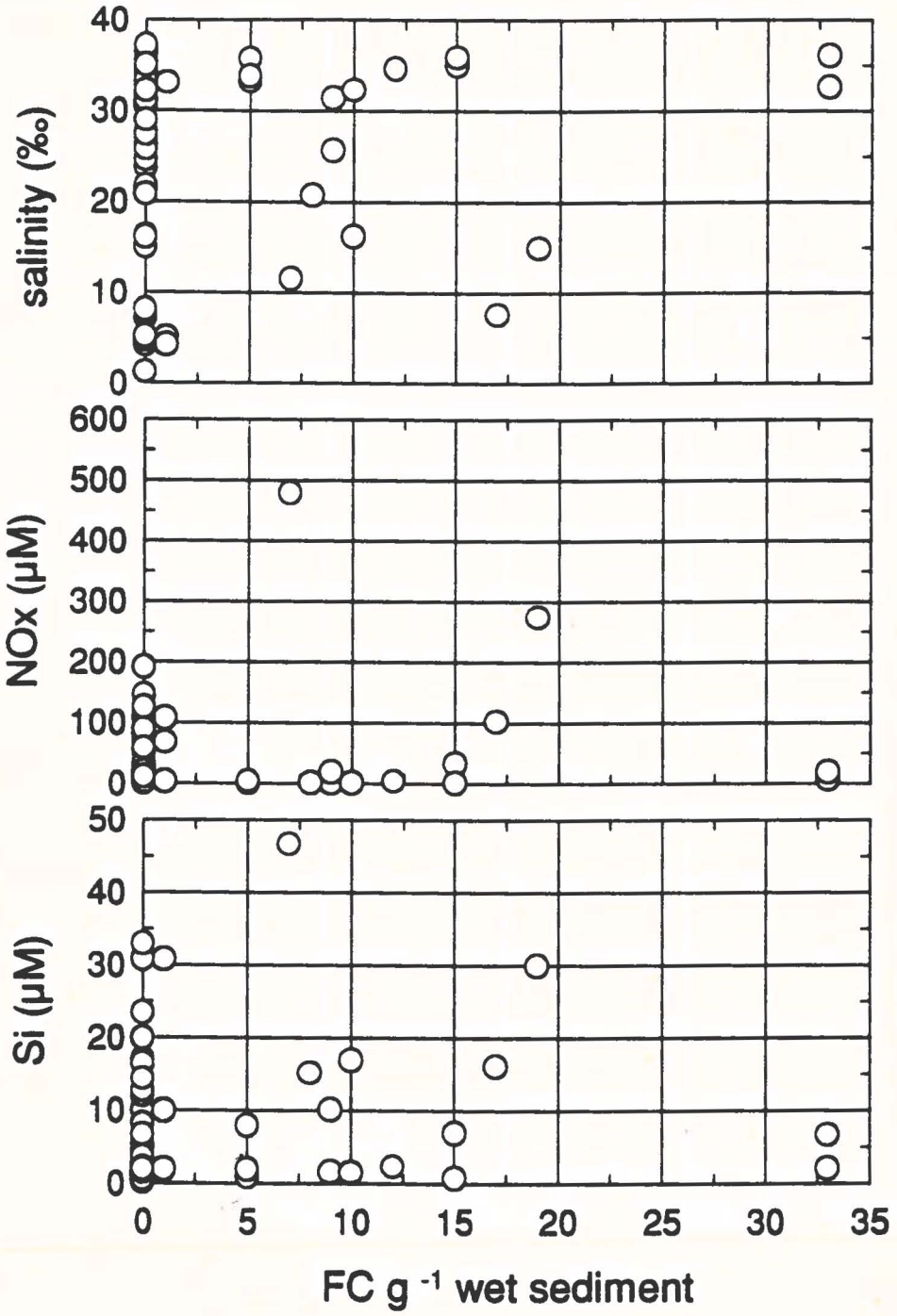

Figure 15. Salinity, nitrate + nitrite, and silica *vs.* fecal coliforms per gram wet sediment.

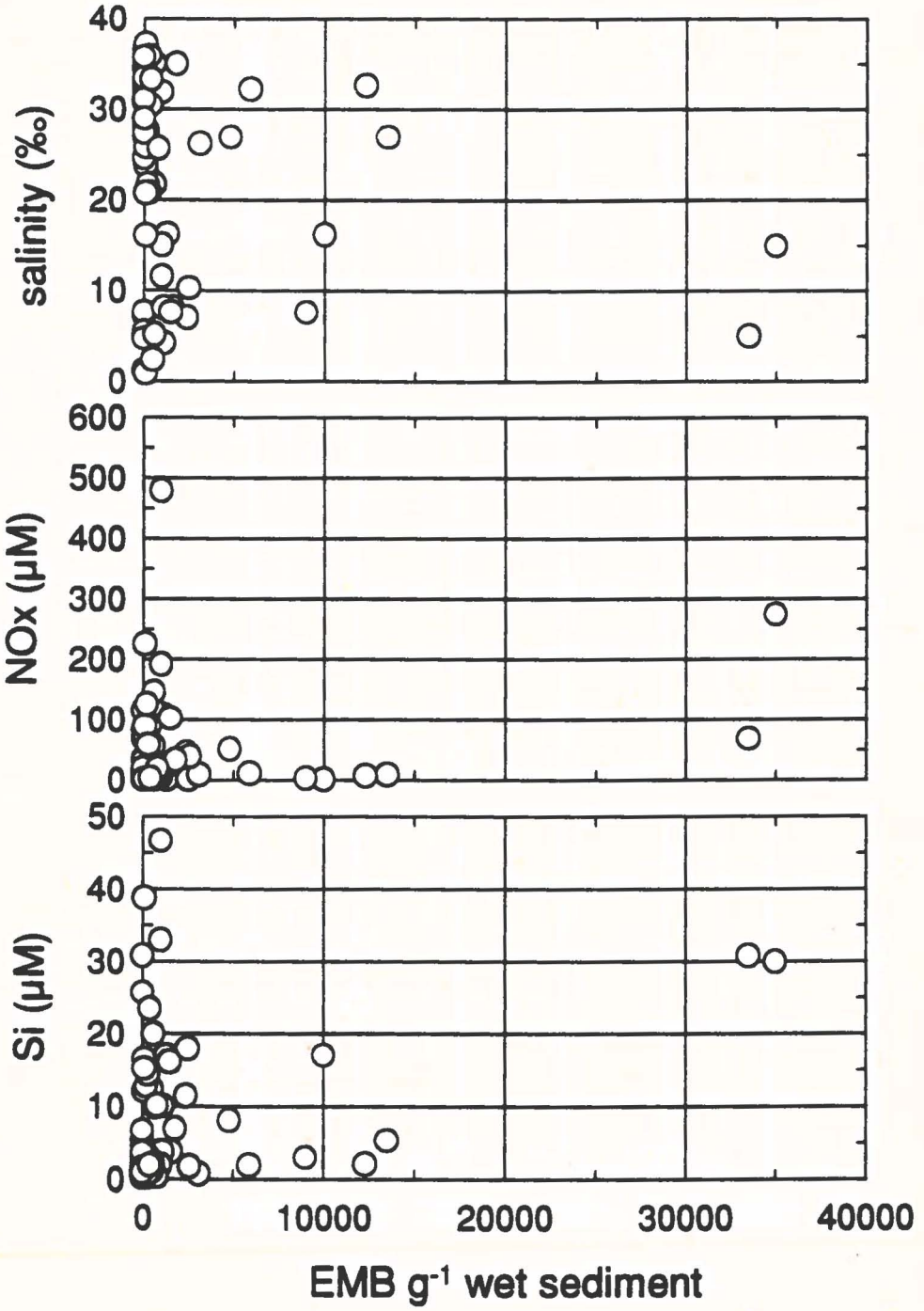

Figure 16. Salinity, nitrate + nitrite, and silica *vs.* lactose fermenters (on EMB) per gram wet sediment.

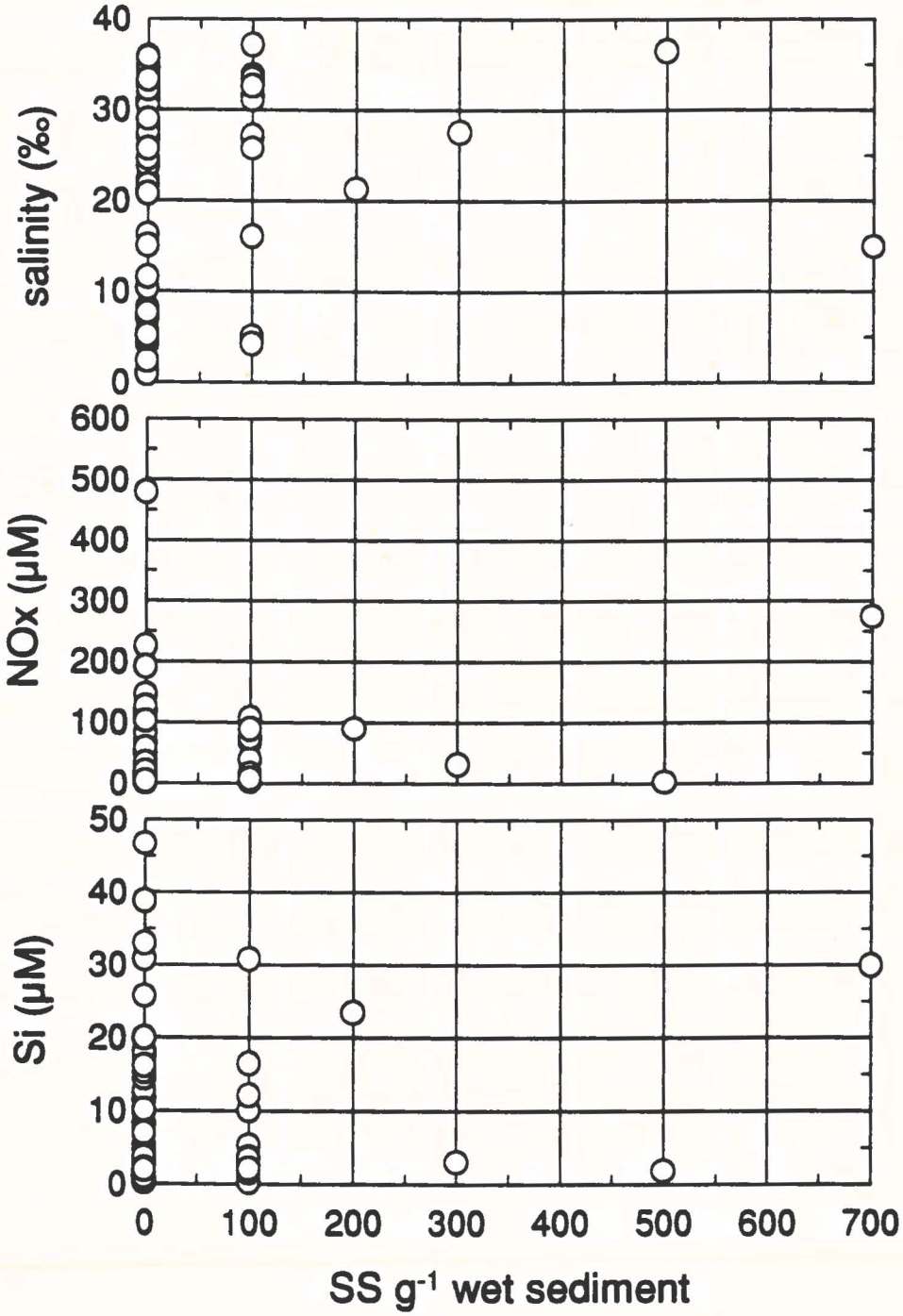

Figure 17. Salinity, nitrate + nitrite, and silica *vs. Salmonella* and *Shigella* spp. per gram wet sediment.

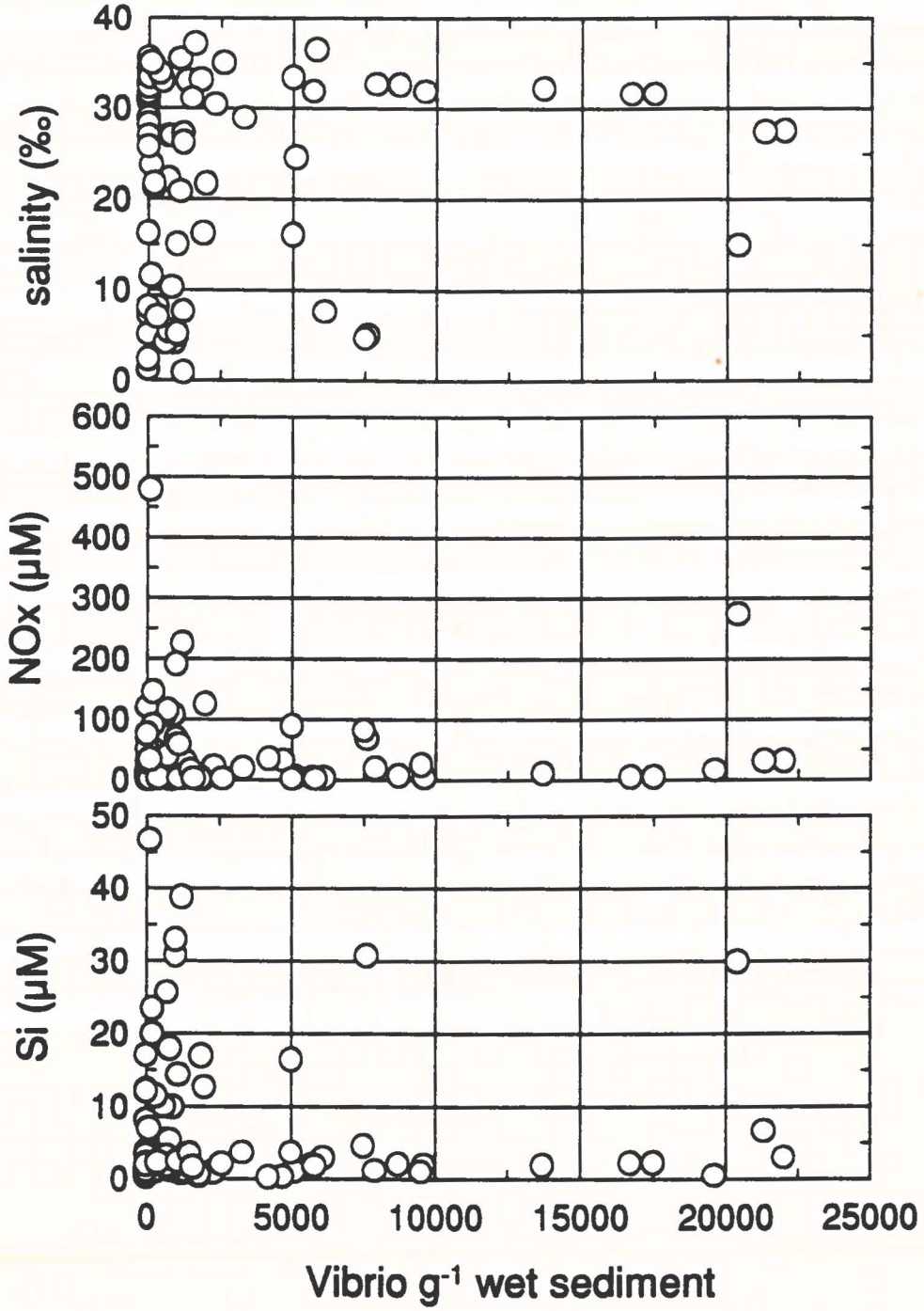

Figure 18. Salinity, nitrate + nitrite, and silica *vs. Vibrio* spp. per gram wet sediment.

contamination levels when water Si levels were > predicted, (3) > recommended bacterial contamination levels when water Si levels were < predicted,and (4) > recommended bacterial contamination levels when water Si levels were > predicted. This was done for NO*x* in the same way. The only significant relationship ($p = 0.05$) was between enterococci and Si.

Si levels were greater than predicted only 12% of the time when sampled during the dry season, this percentage increased to 60% of the time during the rainy season, as was expected if ground water was diluted with runoff. However, NO*x* data did not reflect the presence of runoff diluted with ground water. In fact, NO*x* levels were lower than predicted by the model more often in the dry season (64%) than in the rainy season (42%). It was expected that there would be a simultaneous occurrence of bacteria when there was evidence of storm water runoff in the ground water. There was a higher occurrence of bacteria when there were higher than expected levels of Si regardless of whether the NO*x* levels were greater or less than expected (Table 5).

Table 5. Bacterial occurrence and density in relation to deviations from predicted water chemistry.

medium		level compared to that expected			
		> Si < NOx	> Si > NOx	< Si < NOx	< Si > NOx
mEnterococcus					
	% of time present	63	73	35	47
	sample *n* =	8	37	40	15
	density $\bar{x}$	6	23	4	1
mFC					
	% of time present	57	30	18	13
	sample *n* =	7	23	33	15
	density $\bar{x}$	4	4.6	2	1
EMB					
	% of time present	75	79	50	40
	sample *n* =	8	34	38	15
	density $\bar{x}$	6025	1723	953	473
SS					
	% of time present	25	24	11	13
	sample *n* =	8	34	38	15
	density $\bar{x}$	25	65	11	27
TCBS					
	% of time present	83	85	59	46
	sample *n* =	6	33	37	15
	density $\bar{x}$	2717	3351	2455	3574

When fluorescein was added to the storm water runoff entering the ponding basin (PB1), there was no evidence of fluorescein in the bay during the following 48 h by which time the water in the ponding basin had drained. Because of this the beach hole studies were conducted. Fluorescein dye studies at sites A and B showed no flow of the dye from the beach holes (5 m and 10m above MLT) out into the bay. There was no visual or analytical evidence of fluorescein in the bay water during the hourly, or twice daily samples at site A. At site B, when the holes were excavated after 24 h, fluorescein was still evident in the holes. However, fluorescein did appear in the bay at site B 12 days later, when a storm had caused waves to wash above the height of the two holes.

Survival

Results of survival studies in tanks of sterile Tumon Bay seawater exposed to sunlight and in darkness are given in Figure 19. For all bacteria, the decline in numbers was more rapid in the tanks exposed to sunlight. When exposed to sunlight *Streptococcus faecalis* (Ent) showed a steady decrease in numbers for at least 3 days, then decreased more rapidly until there were less than the detection limit of 1 in 0.1 mL (log10 = 3) after 5 days, whereas *Streptococcus faecalis* in sterile seawater in darkness were still (~3.5 $\log_{10}$ 100 mL^{-1}) after 7 days. *Escherichia coli* and *Klebsiella pnuemoniae* exposed to sunlight were below the detection limit after 3-5 days and in darkness remained

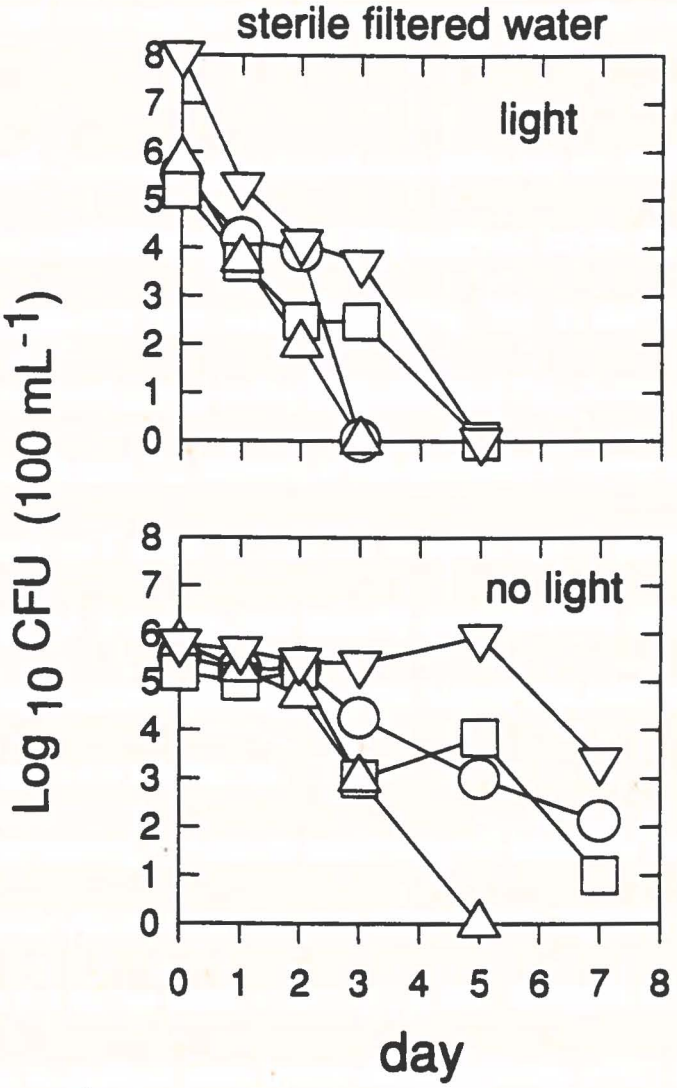

Figure 19. Survival of four bacterial field isolates from Tumon Bay sediment, in sterile seawater with no sediment exposed to sunlight, and no light. *Escherichia coli* (○), *Klebsiella pneumonia* (□), *Enterobacter cloacae* (△), and *Streptococcus faecalis* (▽).

at high numbers for at least 7 days. *Enterobacter cloacae* also had a higher die-off rates when exposed to light. Bacteria in darkness were still detectable at least two days after the bacteria that were exposed to sunlight were no longer detectable.

When sterile Tumon Bay sediment was added to sterile Tumon Bay water, bacteria exposed to sunlight (Fig. 20) survived longer than their counter parts in tanks of seawater without sediment (Fig. 19). In sterile sediment and water, bacteria survived longer in the sediment and than in the water column (Fig. 20): *S. faecalis* and *En. cloacae* in the sediment showed only a small decline in density over 7 days. *E. coli, En. cloacae,* and *K. pnuemoniae* had very little die-off in the first 2-3 days, and actually showed some regrowth during this time. Numbers decreased more rapidly after 3 days and decreased faster in the water column than in the sediments.

In tanks containing non-sterile sediment and seawater that were exposed to sunlight, a slower die-off rate was also observed for bacteria in the sediment than for those in the above water column (Fig. 20). Bacteria in the water column of tanks exposed to sunlight were below detection limits within 2 days. The die-off rate of *E. coli* was greater in the non-sterile sediment than in sterile sediment. However, *S. faecalis* survived equally well in sterile and non-sterile sediment.

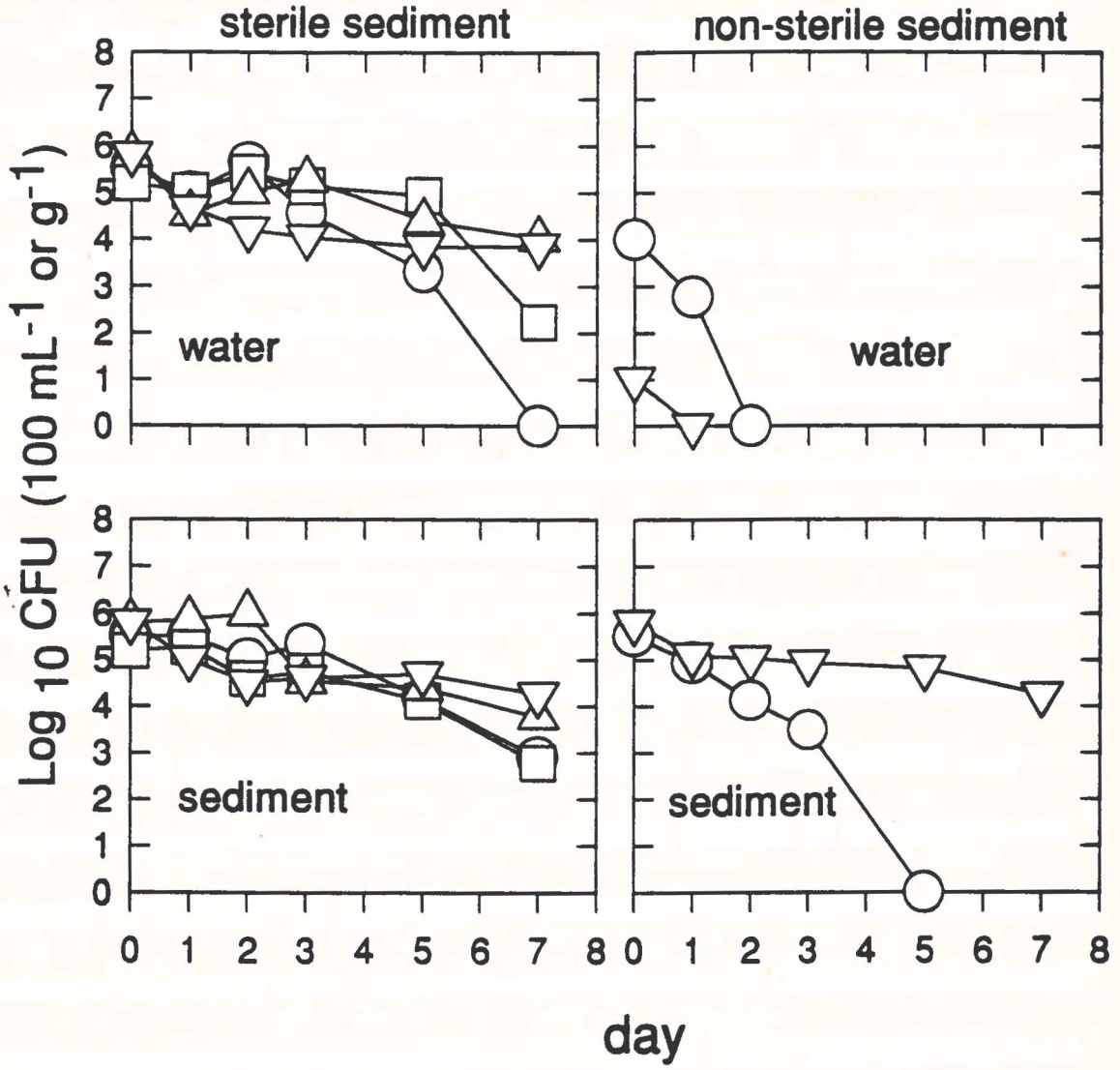

Figure 20. Survival of four bacterial field isolates from Tumon Bay sediment, in the water and sediments of microcosms containing sterile seawater and sediment, and non-sterile seawater and sediment, exposed to sunlight. *Escherichia coli* (○), *Klebsiella pneumonia* (□), *Enterobacter cloacae* (△), and *Streptococcus faecalis* (▽).

DISCUSSION

Storm water and roadside runoff in the Tumon Bay area was found to be polluted with large numbers of fecal indicators and potential pathogens. Numbers were as high as those of sewage treatment plant effluent (i.e., 6.3×10^5 to 2.5×10^6 FC 100 mL^{-1}, Matson 1993a). During heavy rains, sewers in the low lying areas of Tumon often overflow into the streets where they contaminate storm water that is disposed of into ponding basins, percolation fields, or which runs off onto unpaved lots, where it later percolates into the ground. There is the potential for fecal bacteria from polluted runoff to be transported out into the near shore waters and sediments due to the continuous ground water flow and lack of exchange sites (i.e., clay minerals and organic matter) in the carbonate soils that may allow for rapid delivery to the coast. Other mechanisms may affect transport to the coast as well. For example, Gannon et al. (1991) found that 60 - 77% of bacteria suspended in deionized water added passed through a column of saturated sandy aquifer material, but only 1.5 - 3.9% passed through when the bacteria were suspended in 10 mM NaCl. Thus, when bacteria reach near shore sediments with higher salinity pore waters it is possible that cell sorption to surfaces may occur and prevent the cells from being transported further out into the bay. This may explain the higher densities found in the beach hole sediments.

This study showed an increase in the occurrence and densities of fecal pollution indicators and potential pathogens in Tumon Bay sediments during the rainy season. These increases were often associated with departures from predicted chemical signals in the ground water. Guam EPA monitoring of several of the recreational waters also showed an increase in bacterial counts during the rainy season of the same year (GEPA, unpublished data 1993). In fact, there was a significant correlation between 4 day running average of daily rainfall and the densities recorded on mFC, EMB, and SS from the sediments. The overall lack of correlation however, between average daily and cumulative rainfall in the 0-14 days prior to sampling and the bacterial densities in the sediments, as well as with water chemistry is probably due to the infrequent sampling regime. Because of the long periods (ca. 4 weeks) between samples, sporadic events may have been missed.

Contaminated waters from ponding basins, percolation fields and runoff may be transported through limestone to the bay in pulses rather than being continuously diluted and mixed with the aquifer water as originally hypothesized. In this respect, Mates and Scheinberg (1991) found that monitoring Israeli beaches twice a week would approve them by all international standards. However, intensive monitoring (5 days a week, 3 times a day) indicated that these otherwise approved beaches were quite often polluted, and unsafe for bathing. At this sampling resolution, they found a strong correlation between

the amount of rain and the degree of pollution. They concluded that surveys once a day, 5 days a week were needed to adequately monitor pollution of the beaches.

Water chemistry samples did show deviations in NO*x* and Si levels from those predicted by the model generated by data collected between 1986-1989 (Matson 1993b). These deviations, however, were not always consistent with the model of ground water being diluted with runoff. Concentration deviations were both greater and less than predicted for both NO*x* and Si. This may indicate that ground waters are no longer conservatively mixed with Tumon Bay seawater, as they were when the data were collected to generate the conservative mixing models, but that biogeochemical processes such as denitrification and silica diagenesis now occur at a greater rate than in previous years. In this respect beach hole waters often did show evidence of the dilution of ground water with Si-rich runoff, as Si was higher (possibly due to diagenetic solubilization) than expected and NO*x* lower (possibly due to denitrification) than expected around 60% of the time.

Concomitant with the occurrence of these deviations we expected a simultaneous increase of indicator bacteria in the sediments. Plots of bacteria densities against water chemistry however, did not show obvious correlations between the two, and often no bacteria were present when Si was high and NO*x* was low. Using Chi-square G tests to assess any association between bacterial numbers (< or > threshold levels 100 mL^{-1}) and the deviation of water chemistry

from that predicted only yielded significant results between Si and bacterial densities. In fact, Si levels were greater than predicted far more often in the rainy season (60%) than the dry (12%), and this corresponded with higher occurrences and average densities of bacteria in the sediments. Unlike Si, corresponding low levels of NO*x* did not correlate with the occurrence or densities of bacteria and may not have been a good indication of the presence of NO*x* poor surface runoff. NO*x* levels were in fact lower than predicted more often during the dry season than they were during the rainy season, a phenomenon recently observed (Matson, unpubl.).

Other evidence of runoff mixing with ground water is the deposits of red Fe-rich dirt found on rocks in the sediments out in the bay. Iron crusts and layers were also commonly found in several strata, in beach holes due to tidally driven fluctuations in the water table, also, anoxic conditions were frequently observed in beach hole sediments and in the core samples of the TR sites, the sediments of which were black and smelled of sulfide. Anoxic conditions in sediments and pore waters reduce iron to its soluble form, precipitate it with S^{2-}, and as new oxidized water is introduced via groundwater and seawater, the pore waters then become oxidized and iron oxides (e.g., FeOOH, $FeOH_3$) are deposited at the reductocline in the sediments and on rocks.

The fluorescein dye added to the runoff water in the ponding basin at Ypao Beach Park was never detected in the adjacent bay waters, even though water chemistry data indicated that there was runoff present in the ground water.

Therefore, we were unable to establish the lag time, duration and pattern that runoff from the ponding basin would take to reach the coastal sediments. It may have been that the dye was too dilute, or that it precipitated out by the time it reached the shore and was therefore was not detectable in the coastal waters. However, in the beach hole studies, there was only a short distance (ca. 5-10 m) for the dye to flow out into the bay, yet the fluorescein did not appear in the bay until several days later. When holes were excavated 24 hours after adding the dye it was evident the dye had precipitated. The dye later became soluble when the holes were inundated with high salinity seawater during high tides, and fluorescein was seen in the bay waters 12 days after being added to the hole. This delay in the release of fluorescein from the holes out into the bay may be due in part to old brackish water being held in the supra- and inter- tidal sediments due to recent compaction of these sediments by vehicles traveling along the beach, causing the water and dye to remain there until they were washed out by high salinity water after extreme high tides. Repeating these ground water flow experiments with other dye tracers (e.g., food coloring) may give better results.

It was evident from the survival studies that fecal bacteria can survive for up to a week, and that underground transport would protect them from sunlight. Results of this work showed that bacteria in sterile seawater that were not exposed to sunlight had much greater survival than those exposed. The effects of sunlight in the inactivation of bacteria in receiving waters are well

documented. Fujioka et al. (1981) found that fecal coliforms and fecal streptococci (enterococci) would survive for 1 - 3 days in seawater in the absence of sunlight. But in the presence of sunlight, 90% of FCs would be inactivated in 30 - 90 min and 90% of FSs in 69 - 180 min. Fujioka and Narikawa (1982) showed that sunlight initially stresses the bacteria, and eventually causes cell death. After reviewing all the published data, Chamberlin and Mitchell (978) concluded that under field conditions sunlight is probably the most effective bactericidal factor in nature. Solić and Krstulović (1992) also found that sunlight had a greater effect on fecal coliform survival than either temperature or salinity, and that temperature and salinity were more detrimental in the presence of sunlight. Curtis et al. (1992) found that sunlight inactivated fecal coliforms in waste stabilization ponds, and that humic substances, pH and dissolved oxygen were important variables in the process by which light damages microorganisms. Bacteria in the Tumon Bay sediments had a higher survival than both those in the overlying water column and those in seawater with no sediment present. Results of these studies are supported by those of Gerba and McLeod (1976). They found that *E. coli* survived for longer periods of time in unsterile natural seawater when sediments were present than in seawater alone. They attribute the longer survival of bacteria in the sediments to a greater content of organic matter and nutrients. Also, *E. coli* numbers increased rapidly in autoclaved natural sediment and seawater, growth and final densities were less when sediment was not present. However, they found that

nutrients were eluted from the sediments after autoclaving and addition of sterile seawater. Therefore, it was interesting that bacteria in the water column above sterile sediments had a longer survival than did bacteria in both tanks of seawater without sediment and in the water column above non-sterile sediment. However, the seawater in the tanks of sterile autoclaved sediments were murkier than the seawater in tanks with no sediments or non-sterile sediment. Milne et al. (1991) found that suspended solids resulted in increased in survival. The sediment may therefore both shield bacteria from sunlight as well as provide a sufficiently rich medium to grow.

Further, bacteria in the non-sterile sediments showed a greater decline in numbers than those of sterile sediments. Hood and Ness (1982), and Sørensen (1991) attributed longer survival of bacteria in sterile sediment and seawater to the absence of bacterial predators. Rhodes and Kator (1988) found that significant reductions in the number of *E. coli* and *Salmonella* spp. were correlated with increased numbers of microbiota and phage forming microorganisms.

Together, these studies have revealed that higher numbers of indicator and potentially pathogenic bacteria in the sediments are due to a combination of sedimentation, sorption (which provides protection from bactivory), protection from inactivation from sunlight, and greater organic content and nutrients. Therefore, in Tumon Bay if fecal bacteria are transported via upwardly

percolating nutrient rich ground water into nutrient rich sediments, they are fed, and protected from sunlight and predators, thus allowing for extended survival (> 7 days) and possibly growth.

Considering all of the above evidence, it seems likely that contaminated storm water runoff is a major source of fecal pollution in Tumon Bay. Aquifer water itself does not contain high enough levels of fecal indicator bacteria and potential pathogens to be the source of fecal contamination found in the sediments (Roman Lizama, Laboratory Support Services, Public Utilities Agency of Guam, personal communication). However, fecal bacteria are also found to occur in the sediments during the dry season when there is little runoff. Further investigation needs to be conducted to establish if this is not a resident population that increases in numbers during the rainy season due to an increase in various nutrients carried in the ground water and runoff, or whether an additional source is leaky sewer pipes. Further survival studies would need to be conducted to see if the indicator bacteria are capable of reproduction and growth during the dry season so as to support the densities found in the rainy season. If these populations are not capable of growth then it could be sumized that polluted runoff cause bacterial densities to increase to above those present in the dry season.

Regardless of the source of this fecal pollution it, is of concern that the sediments can contain high densities of fecal bacteria when the overlying waters are indicator free. GEPA's results do show an increase in indicator numbers for

Tumon Bay during the rainy season, but rarely were the densities above the maximum recommended safe limit of 35 enterococci or 200 fecal coliforms 100 mL^{-1}. Matson 1993a, notes that in stormy and windy weather the Guam EPA records "TMTC" ("too many to count") for indicator densities in many otherwise "pristine" recreational waters. This is presumably due to the resuspension of contaminated bottom sediments. The sediments of Tumon Bay are an unacknowledged and a potentially hazardous reservoir of fecal bacteria.

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APPENDIX

Coastal Sediment and Water Quality Data for 1993.

Appendix. Coastal Sediments and Water Quality Data from 1993.

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			per gram							uM			
1/27													
(27)	YP1	0-3	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	YP1	3-5	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	YP1	5-7	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	YP1	7-9	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	YP2	0-3	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR1	0-3	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR1	3-6	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR1	6-8	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR1	8-10	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR1	10-13	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR2	0-3	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR2	3-6	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR2	6-9	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR2	9-12	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR2	12-15	0	0	nd	nd	nd		nd	nd	nd	nd	nd
	TR2	15-20	0	0	nd	nd	nd		nd	nd	nd	nd	nd
2/12													
(43)	TR1	0-5	1	0	200	100	0	0	33.9	6.3	2.3	0.27	2
	TR1	5-10	11	0	200	0	0	10	17.6	11	64	3.8	9.7
	TR1	10-15	0	0	0	0	0	15	13.4	95	79	3.4	12
	TR1	15-18	0	0	0	0	0	20	9.4	80	94	4.4	14
	TR1	-	-	-	-	-	-	25	11.4	51	87	3.9	13
	TR1	-	-	-	-	-	-	30	20.9	48	51	2.5	8.2
	TR2	5-10	0	0	0	0	0	10	14.8	35	74	3.6	11

nd = not done

- = no data

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	TR2	10-15	0	0	0	0	0	15	9.2	60	95	3.7	14
	TR2	-	-	-	-	-	-	20	8.3	79	99	3.8	14
2/19													
(50)	YP1	0-3	0.8	0	0	0	0	0	34.0	3.1	1.6	0.47	2
	YP1	3-7	0.6	0	0	0	0	-	-	-	-	-	-
	YP1	7-10	0.8	0	0	0	0	10	36.1	1.8	-6.3	0.8	1
	YP1	-	-	-	-	-	-	15	34.9	1.9	-1.5	1.1	1.6
	YP1	-	-	-	-	-	-	20	35.4	0.77	-3.4	0.67	1.3
	YP1	-	-	-	-	-	-	24	34.9	0.9	-1.8	1.5	1.5
	YP2	0-3	0.4	0	0	0	0	0	30.8	8	14	1.4	3.5
	TR1	0-4	8	0	0	0	0	0	30.7	10	14	1.5	3.5
	TR1	4-9	1.2	0	0	0	0	10	29.8	8.6	18	0.73	4
	TR1	9-13	0	0	0	0	0	-	-	-	-	-	-
	TR1	13-17	0.2	0	0	0	0	15	30.0	11	17	1.1	3.9
	TR1	17-20	0.2	0	0	0	0	20	27.2	14	28	1.5	5
	TR1	-	-	-	-	-	-	25	28.5	16	22	1.5	4.6
	TR1	-	-	-	-	-	-	30	27.9	12	25	2.2	4.8
	TR1	-						34	30.4	12	16	1.9	3.7
	TR2	0-3	0	*nd*	*nd*	*nd*	*nd*	0	*nd*	*nd*	*nd*	*nd*	*nd*
	TR2	3-8	0.2	*nd*	*nd*	*nd*	*nd*	-	-	-	-	-	-
	TR2	8-12	0	*nd*	*nd*	*nd*	*nd*	10	25.1	33	35	2.3	6.2
	TR2	12-17	0.4	*nd*	*nd*	*nd*	*nd*	15	12.7	69	82	3.7	12
	TR2	17-22	0.2	*nd*	*nd*	*nd*	*nd*	20	9.5	79	94	6.2	14
	TR2	-	-	-	-	-	-	25	15.0	105	73	3.1	11
	TR2	-	-	-	-	-	-	30	26.2	25	31	2	5.7
2/23													
(54)	YP1	0-3	3	0	0	0	0	0	34.7	1.8	-0.8	*nd*	1.7

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	YP1	3-6	1	0	0	0	0	-	-	-	-	-	-
	YP1	6-9	19	2	0	0	0	10	32.9	2.3	5.9	nd	2.5
	YP1	-	-	-	-	-	-	15	34.6	4.6	-0.56	nd	1.7
	YP1	-	-	-	-	-	-	20	34.7	1.7	-0.8	nd	1.7
	YP1	-	-	-	-	-	-	24	34.6	1.3	-0.32	nd	1.7
	YP2	0-3	4	0	0	0	0	0	34.5	4.8	-0.08	nd	1.7
	YP2	-	-	-	-	-	-	10	34.5	8.8	-0.08	nd	1.7
	YP2	-	-	-	-	-	-	13	34.3	6.8	0.64	nd	1.8
	TR1	0-4	0	0	0	0	0	0	32.6	4.6	6.88	nd	2.6
	TR1	4-8	0	0	0	0	0	-	-	-	-	-	-
	TR1	8-12	1	0	0	0	0	10	19.7	41	56	nd	8.7
	TR1	12-16	5	0	0	0	0	15	8.4	37	98	nd	14
	TR1	16-20	25	3	0	0	0	20	7.4	62	100	nd	15
	TR1	-	-	-	-	-	-	25	15.3	35	72	nd	11
	TR2	0-3	1	0	0	0	0	0	32.4	4.9	7.8	nd	2.7
	TR2	3-7	6	0	0	0	0	-	-	-	-	-	-
	TR2	7-12	2	0	0	0	0	10	21.6	27	49	nd	7.8
	TR2	12-17	0	0	0	0	0	15	19.2	41	58	nd	9
	TR2	17-22	1	0	0	0	0	20	18.4	48	61	nd	9.3
3/12 (71)	YP1	0-4	48	9	600	0	0	0	31.4	0.77	11	1.6	3.2
	YP1	4-8	12	5	700	0	0	10	34.6	0.77	-0.32	2.2	1.7
	YP1	-	-	-	-	-	-	15	33.9	1.5	2.1	1.8	2
	YP1	-	-	-	-	-	-	20	32.6	0.77	6.8	2.8	2.6
	YP1	-	-	-	-	-	-	22	31.1	1.1	13	3.8	3.4
	TR1	0-3	51	12	200	0	200	0	34.6	3.6	-0.56	2.3	1.7
	TR1	3-7	21	0	0	0	500	-	-	-	-	-	-

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	TR1	7-11	14	5	200	0	700	10	32.9	2.5	5.9	11	2.5
	TR1	11-17	14	0	100	0	500	15	31.7	3.4	10	12	3.1
	TR1	17-21	25	0	0	100	500	20	32.8	2.7	6.2	7.4	2.5
	TR1	-	-	-	-	-	-	25	33.4	3.1	4	3.5	2.3
	TR1	-	-	-	-	-	-	30	33.5	2.5	3.5		2.2
	TR2	0-4	38	1	0	0	0	0	33.1	2.6	5.2	1.9	2.4
	TR2	4-9	0	0	0	0	900	10	31.6	3.2	11	5.4	3.1
	TR2	9-14	4	0	0	0	900	15	28.2	11	24	6.2	4.7
	TR2	14-18	10	0	0	0	600	20	29.7	6.1	18		4
	TR2	-	-	-	-	-	-	28	30.9	4.8	14	7.4	3.5
3/24 (83)	YP1	hole sed	0	0	0	0	0	hole	34.7	1.4	-0.8	*nd*	1.7
	YP1	0-4	2	0	0	0	0	0	35.5	1.5	-3.9	*nd*	1.3
	YP1	4-8	0	0	0	0	0	10	34.1	0.91	1.3	*nd*	1.9
	YP1	-	-	-	-	-	-	15	34.3	2.2	0.64	*nd*	1.8
	YP1	-	-	-	-	-	-	20	34.9	2.7	-1.8	*nd*	1.5
	YP1	-	-	-	-	-	-	24	35.1	11	-2.2	*nd*	1.5
	YP2	hole sed	1	0	0	0	0	hole	1.2	6.6	130	*nd*	17
	YP2	0-3	0	0	0	0	0	0	35.5	15	-3.9	*nd*	1.3
	YP2	-	-	-	-	-	-	10	33.2	90	4.7	*nd*	2.3
	TR1	hole sed	0	0	0	0	0	hole	7.2	68	100	*nd*	15
	TR1	0-3cm	0	0	0	0	0	0	5.1	9.2	110	*nd*	16
	TR1	3-6	0	0	0	0	0	-	-	-	-	-	-
	TR1	6-11	0	0	0	0	0	-	-	-	-	-	-
	TR1	11-15	0	0	0	0	0	-	-	-	-	-	-
	TR1	15--19	0	0	0	0	0	-	-	-	-	-	-
	TR2	hole sed	0	0	0	0	0	hole	31.1	9.4	13	*nd*	3.4

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	TR2	0-5	0	0	0	0	0	0	31.6	nd	11	nd	3.1
	TR2	5-8	0	0	0	0	0	-	-	-	-	-	-
	TR2	8-13	0	0	0	0	0	-	-	-	-	-	-
	TR2	13-18	0	0	0	0	0	-	-	-	-	-	-
	TR2	18-23	0	0	0	0	0	-	-	-	-	-	-
4/22													
(112)	YP1	hole sed	0	nd	200	0	2000	hole	20.3	33	53	18	8.4
	YP1	hole H2O	2	nd	400	0	4400	hole	20.3	33	53	18	8.4
	YP1	0-5	1	nd	100	0	4200	0	nd	nd	nd	nd	nd
	YP2	hole sed	2	nd	100	0	1200	hole	0.9	220	120	39	18
	YP2	0-5	1	nd	0	0	1000	0	nd	nd	nd	nd	nd
	TR1	0-3	0	nd	0	0	3200	0	nd	nd	nd	nd	nd
	TR1	3-6	0	nd	0	0	1600	-	-	-	-	-	-
	TR1	6-11	0	nd	0	0	600	-	-	-	-	-	-
	TR1	11-15	0	nd	0	0	100	-	-	-	-	-	-
	TR1	15-21	0	nd	0	0	0						
	TR1	hole H2O	0	nd	100	0	43600	hole	11.8	115	85	13	12
	TR1	hole sed	1	nd	100	0	50000	hole					
	TR2	0-5	1	nd	0	0	3000	0	31.4	8.6	12	1.6	3.2
	TR2	5-10	0	nd	100	0	4000	10	29.6	25	18	3.6	4.1
	TR2	10-13	3	nd	0	0	4000	-	-	-	-	-	-
	TR2	13-18	0	nd	0	0	800	-	-	-	-	-	-
	TR2	18-23	1	nd	0	0	200	-	-	-	-	-	-
	TR2	hole H2O	1	nd	200	0	2800	hole	13.8	50	78	18	12
	TR2	hole sed	0	nd	100	100	1700	hole	13.8	50	78	18	12
5/19													
(139)	YP1	0-5	0	10	100	0	10	0	32.3	1.7	8.3	1.5	2.8

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			per gram							uM			
	YP1	5-10	0	0	2	0	1	5	31.7	3.1	10	2.3	3.1
	YP1	10-15	0	0	1	0	100	10	31.6	1.7	140	2.2	3.1
	YP1	-	-	-	-	-	-	15	33.8	1.8	2.5	2.8	2.1
	YP1	-	-	-	-	-	-	20	34.0	1.2	1.8	2.3	2
	YP1	hole sed	0	3	3	37	0	hole	15.7	0.39	71	15	11
	YP1	hole H2O	0	25	25	530	0	hole	15.7	0.39	71	15	11
	YP2	0-5	0	0	0	0	67	0	33.4	8.5	4	2.2	2.3
	YP2	5-10	0	0	0	0	400	5	34.3	16	0.64	3.9	1.8
	YP2	-	-	-	-	-	-	10	32.4	15	7.8	4.3	2.7
	YP2	-	-	-	-	-	-	15	33.8	12	2.6	6.1	2.1
	YP2	hole sed	0	0	0	10	2	hole	5.1	59	110	30	16
	YP2	hole H2O	0	35	35	17	4	hole	5.1	59	110	30	16
	TR1	0-5	0	0	0	0	1342	0	33.0	4.5	5.7	1.5	2.5
	TR1	5-10	0	0	0	0	348	5	32.5	4.4	7.6	3.0	2.7
	TR1	10-16	0	0	0	0	98	10	32.5	2.2	7.4	3.0	2.7
	TR1	16-22	0	0	0	0	74	15	31.9	1.4	9.5	3.5	2.9
	TR1	-	-	-	-	-	-	20	31.2	1.8	12	3.5	3.3
	TR1	-	-	-	-	-	-	25	29.9	2.8	17	3.9	3.9
	TR1	hole sed	1	0	0	0	11	hole	7.1	27	100	0.66	15
	TR1	hole H2O	0	0	0	0	17	hole	7.1	27	100	0.66	15
	TR2	0-4	0	0	0	0	500	0	33.0	19	5.7	1.4	2.5
	TR2	4-8	0	0	0	0	200	5	28.1	23	24	3.7	4.7
	TR2	8-11	1	0	0	0	0	10	23.3	21	42	5.4	7
	TR2	-	-	-	-	-	-	15	26.1	25	32	3.2	5.7
	TR2	-	-	-	-	-	-	20	20.7	58	52	5.5	8.3
	TR2	-	-	-	-	-	-	25	17.4	76	65	6.7	9.8
	TR2	-	-	-	-	-	-	28	14.8	89	74	7.6	11

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	TR2	hole sed	0	0	0	0	100	hole	7.7	7	100	1.5	14
	TR2	hole H2O	0	0	1	2	76	hole	7.7	7	100	1.5	14
6/4													
(155)	YP1	hole sed	0	10	1300	0	1900	hole	16.2	1.2	69	17	10
	YP1	0-6	0	0	0	0	6100	0	7.6	3.4	100	2.9	14
	YP1	6-12	1	0	0	0	6700	10	7.5	5.9	100	3.1	15
	YP1	-	-	-	-	-	-	15	7.2	4.4	100	3.1	15
	YP2	hole sed	1	1	0	0	7600	hole	5.0	68	110	31	16
	YP2	0-4 cm	2	0	0	0	700	0	26.9	9.2	28	5.3	5.3
	YP2	-	-	-	-	-	-	5	8.7	12	97	5.6	14
	TR1	hole sed	0	1	700	0	900	hole	4.2	107	110	9.9	16
	TR1	0-5	0	0	0	0	16700	0	31.6	4.9	11	2.2	3.1
	TR1	5-10	0	0	300	0	500	10	23.2	5	42	5.8	7.1
	TR1	10-15	0	0	200	100	8400	15	22.8	5.7	44	7.2	7.3
	TR1	-	-	-	-	-	-	20	9.4	15	94	8.4	14
	TR2	hole sed	0	0	1600	0	300	hole	8.1	26	100	3.7	14
	TR2	0-5	0	0	600	0	5700	0	31.9	3.9	9.7	1.9	3
	TR2	5-10	0	0	0	0	700	10	15.1	81	73	7.9	11
	TR2	10-14	0	0	200	0	400	15	5.7	122	110	14	15
	TR2	14-19	0	0	100	0	100	20	4.6	128	110	14	16
	TR2	-	-	-	-	-	-	25	5.6	117	110	14	15
	TR2	-	-	-	-	-	-	30	5.5	121	110	12	15
6/30													
(181)	YP1	hole sed	0	0	10000	0	0	hole	16.2	1.01	69.04	17	11
	YP1	0-4	1	0	9000	0	1200	0	7.6	3.2	101.44	3	14
	YP1	4-8	0	0	10000	0	0	10	7.5	5.8	102	3.2	15
	YP1	-	-	-	-	-	-	15	7.2	4.3	102	3.1	15

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	YP2	hole sed	1	0	33500	100	1000	hole	5.0	68	110	31	15.7
	YP2	0-4	2	0	13500	100	800	0	26.9	9.1	28	5.3	5.3
	YP2	-	-	-	-	-	-	5	8.7	12	97	5.6	14
	TR1	hole sed	0	0	1100	100	600	hole	4.2	106	110	10.0	16
	TR1	0-5	0	0	600	100	17500	0	31.6	4.8	11	2.2	3.1
	TR1	5-10	0	0	1000	100	68900	10	23.2	4.9	42	5.8	7.1
	TR1	10-16	0	0	300	0	23100	15	22.8	5.5	44	7.3	7.3
	TR1	-	-	-	-	-	-	20	9.4	15	94	8.5	14
	TR2	hole sed	0	0	1100	0	0	hole	8.1	25	100	3.8	14
	TR2	0-6	0	0	1000	0	9600	0	31.9	3.7	9.8	1.9	3
	TR2	6-12	0	0	200	0	1100	10	15.1	81	73	7.9	11
	TR2	12-17	0	0	1300	0	1400	15	5.7	120	110	14	15
	TR2	17-22	0	0	300	0	0	20	4.6	130	110	12	16
	TR2	-	-	-	-	-	-	25	5.6	120	110	14	15
	TR2	-	-	-	-	-	-	30	5.5	120	110	12	15
7/23 (211)	YP1	hole sed	0	*nd*	2500	0	800	hole	10.2	0.64	91	18	13
	YP1	hole H2O	1	*nd*	2700	0	600	hole	10.2	0.64	91	18	13
	YP1	0-5	0	*nd*	100	100	1200	0	27.2	6.6	28	2	5.2
	YP1	5-10	0	*nd*	600	0	1200	10	27.6	6.8	26	2	5
	YP1	-	-	-	-	-	-	15	32.5	4.9	7.4	4	2.7
	YP2	hole sed	0	*nd*	0	0	700	hole	5.4	110	110	26	16
	YP2	hole H2O	0	*nd*	0	0	800	hole	5.4	110	110	26	16
	YP2	0-4	0	*nd*	0	0	0	0	28.9	1	21	3.3	4.4
	YP2	-	-	-	-	-	-	8	29.5	6	19	3.5	4.1
	TR1	hole sed	0	*nd*	500	0	0	hole	2.4	120	120	12	17
	TR1	hole H2O	2	*nd*	900	0	0	hole	2.4	120	120	12	17

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	TR1	0-3	1	nd	300	0	700	0	22.2	32	46	3.2	7.5
	TR1	3-6	0	nd	300	0	200	-	-	-	-	-	-
	TR1	6-9	0	nd	1500	0	100	-	-	-	-	-	-
	TR1	9-12	0	nd	2400	0	100	10	14.6	62	75	5.9	11
	TR1	12-15	0	nd	1300	0	0	15	9.9	89	93	11	13
	TR1	15-20	0	nd	900	0	0	20	15.1	58	73	6.8	11
	TR2	hole sed	1	nd	2400	0	300	hole	7.0	45	104	11	15
	TR2	hole H2O	3	nd	3600	0	500	hole	7.0	45	104	11	15
	TR2	0-5	1	nd	0	0	0	0	28.2	11	24	2.1	4.7
	TR2	5-10	1	nd	200	0	2000	10	31.9	8.6	9.8	2.5	3
	TR2	10-15	0	nd	100	0	0	15	28.1	9.5	24	4	4.8
	TR2	15-20	0	nd	300	0	0	20	21.4	53	49	5.6	7.9
	TR2	-	-	-	-	-	-	25	14.3	68	76	7.5	11
8/17 (229)	YP1	hole sed	0	0	200	300	22000	hole	27.6	31	26	2.9	5
	YP1	hole H2O	0	0	100	0	7100	hole	27.6	31	26	2.9	5
	YP1	0-5	0	0	100	0	100	0	23.7	0.52	40	0.67	6.8
	YP1	5-11	0	0	200	0	300	10	30.2	7.9	16	1.9	3.8
	YP1	-	-	-	-	-	-	15	33.3	10	4.2	1.7	2.3
	YP1	-	-	-	-	-	-	20	29.0	0.26	21	0.38	4.3
	YP2	hole sed	0	0	0	0	7500	hole	4.6	82	110	4.5	16
	YP2	hole H2O	0	0	500	500	5500	hole	4.6	82	110	4.5	16
	YP2	0-4	0	0	100	0	7900	0	32.6	19	6.8	1.2	2.6
	TR1	hole sed	1	0	600	0	1000	hole	5.2	56	110	1	16
	TR1	hole H2O	110	0	800	0	600	hole	5.2	56	110	1	16
	TR1	0-5	0	0	0	0	5100	0	24.6	4.8	37	1	6.4
	TR1	5-10	0	0	3200	0	800	10	24.2	6.1	39	1	6.6

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	TR1	10-16	1	0	5100	200	2500	15	24.4	41	38	1.4	6.5
	TR1	-	-	-	-	-	-	20	24.6	10	37	1.7	6.4
	TR2	hole sed	0	0	3100	0	1200	hole	26.2	8.9	31	0.75	5.6
	TR2	hole H2O	10	0	400	0	3200	hole	26.2	8.9	31	0.75	5.6
	TR2	0-5	2	0	400	0	2300	0	30.4	22	15	0.92	3.7
	TR2	5-10	0	0	1200	0	0	10	27.2	20	27	1.3	5.2
	TR2	10-15	0	0	100	0	900	15	23.0	48	43	2.1	7.2
	TR2	15-20	0	0	200	0	600	20	18.5	67	60	2.2	9.3
	TR2	-	-	-	-	-	-	25	18.4	62	61	2.5	9.3
	TR2	-	-	-	-	-	-	30	16.2	86	68	3.1	10
9/22 (265)	YP1	hole sed	20	*nd*	400	0	19600	hole	*nd*	16	*nd*	0.46	*nd*
	YP1	hole H2O	200	*nd*	3000	100	20000	hole	*nd*	16	*nd*	0.46	*nd*
	YP1	0-5	3	*nd*	800	0	1800	0	*nd*	4.5	*nd*	0.33	*nd*
	YP2	hole sed	29	*nd*	500	0	1300	hole	*nd*	30	*nd*	1.1	*nd*
	YP2	hole H2O	7500	*nd*	8600	100	8200	hole	*nd*	30	*nd*	1.1	*nd*
	YP2	0-4	12	*nd*	100	0	4700	0	*nd*	32	*nd*	0.29	*nd*
	YP3	hole sed	58	*nd*	300	300	1200	hole	*nd*	nd	*nd*	*nd*	*nd*
	YP3	hole H2O	100	*nd*	7100	4200	2700	hole	*nd*	nd	*nd*	*nd*	*nd*
	YP3	0-5	3	*nd*	0	0	4200	0	*nd*	35	*nd*	0.21	*nd*
	TR1	hole sed	10	*nd*	100	0	300	hole	*nd*	29	*nd*	0.96	*nd*
	TR1	hole H2O	40	*nd*	2700	200	3000	hole	*nd*	29	*nd*	0.96	*nd*
	TR1	0-5	4	*nd*	100	0	9500	0	*nd*	25	*nd*	0.96	*nd*
	TR2	hole sed	0	*nd*	300	0	0	hole	*nd*	29	*nd*	1.1	*nd*
	TR2	hole H2O	60	*nd*	2100	0	5800	hole	*nd*	29	*nd*	1.1	*nd*
	TR2	0-5	1	*nd*	0	0	800	0	*nd*	9.4	*nd*	1.1	*nd*
	TR3	hole sed	2	*nd*	2600	100	700	hole	*nd*	39	*nd*	1.8	*nd*

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------						----------uM----------				
	TR3	hole H2O	180	nd	4700	600	5400	hole	nd	39	nd	1.8	nd
	TR3	0-5	0	nd	200	0	0	0	nd	31	nd	1.9	nd
10/22													
(295)	YP1	hole sed	4	0	4800	0	0	hole	26.9	51	28	8.05	5.3
	YP1	hole H2O	84	0	200	0	1500	hole	26.9	51	28	8.05	5.3
	YP1	0-3	34	0	0	0	1800	0	33.2	0.52	4.9	0.55	2.4
	YP1	3-5	1	0	100	100	500	10	31.9	1.5	9.5	7.1	2.9
	YP1	-	-	-	-	-	-	15	17.2	200	65	18	9.9
	YP1	-	-	-	-	-	-	18	13.4	2.1	80	21	12
	YP2	hole sed	2	0	400	200	200	hole	21.3	90	49	23	8
	YP2	hole H2O	100	0	3300	200	11200	hole	21.3	90	49	23	8
	YP2	0-5	400	0	0	500	5800	0	36.5	1.6	-7.5	1.8	0.81
	YP2	-	-	-	-	-	-	5	35.3	4.6	-2.9	2.9	1.4
	TR1	hole sed	17	0	600	0	200	hole	21.7	140	48	20	7.8
	TR1	hole H2O	19	0	4900	100	800	hole	21.7	140	48	20	7.8
	TR1	0-5	3	0	300	0	400	0	33.9	2.5	2.3	2.2	2
	TR1	5-10	0	0	100	0	0	10	22.2	52	46	1.8	7.5
	TR1	10-16	1	0	100	0	0	15	20.2	48	54	7.7	8.5
	TR1	-	-	-	-	-	-	20	17.1	68	65	9.7	9.9
	TR1	-	-	-	-	-	-	24	13.9	87	77	11.5	11
	TR2	hole sed	4	0	100	100	0	hole	25.7	75	33	12	5.9
	TR2	hole H2O	656	0	2400	0	0	hole	25.7	75	33	12	5.9
	TR2	0-5	4	0	100	0	1100	0	35.4	1.9	-3.7	2.4	1.3
	TR2	5-10	1	0	100	0	0	10	24.9	21	36	6.5	6.3
	TR2	10-15	3	0	200	0	0	15	24.6	24	37	6	6.4
10/27													
(300)	YP1	hole sed	2	0	1000	0	1000	hole	15.0	190	73	32	11

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	YP1	hole H2O	140	40	28000	0	72800	hole	15.0	190	73	32	11
	YP1	0-4	14	0	0	100	5000	0	33.4	1.5	4	3.6	2.3
	YP1	4-8	1	0	100	0	1300	-	-	-	-	-	-
	YP2	hole sed	4	0	100	100	5000	hole	16.0	89	69	17	10
	YP2	hole H2O	160	50	1100	400	100000	hole	16.0	89	69	17	10
	YP2	0-4 cm	7	0	0	0	21300	0	27.4	30	27	6.7	5.1
	TR1	hole sed	0	0	200	0	2000	hole	21.7	120	48	13	7.8
	TR1	hole H2O	190	120	500	0	13400	hole	21.7	120	48	13	7.8
	TR1	0-5	0	0	0	100	1500	0	31.1	16	13	3.5	3.4
	TR1	5-10	1	0	500	0	3000	10	26.4	30	30	6.5	5.6
	TR1	10-16	1	0	300	0	2200	15	12.3	23	84	4.6	12
	TR1	-	-	-	-	-	-	20	31.5	13	11	3.1	3.2
	TR1	-	-	-	-	-	-	25	32.0	17	9.3	3.3	2.9
	TR1	-	-	-	-	-	-	30	31.3	17	12	5	3.2
	TR1	-	-	-	-	-	-	35	31.8	13	10	3.3	3
	TR2	hole sed	0	0	300	0	1100	hole	20.8	58	52	14	8.2
	TR2	hole H2O	60	30	1000	0	8400	hole	20.8	58	52	14	8.2
	TR2	0-5	8	0	0	0	3300	0	29.0	19	21	3.7	4.4
	TR2	5-10	0	0	0	0	1300	10	26.7	30	29	5.2	5.4
	TR2	10-15	0	0	100	0	600	15	27.2	34	27	5	5.2
	TR2	-	-	-	-	-	-	20	27.9	28	25	7.5	4.9
	TR2	-	-	-	-	-	-	25	29.5	25	19	3.9	4.1
	TR2	-	-	-	-	-	-	28	30.0	16	17	3.2	3.9
12/3 (337)	YP1	hole sed	0	15	1800	0	100	hole	35.0	33	-2	6.9	1.5
	YP1	0-5	0	0	100	100	1600	0	37.1	1.8	-9.9	1.6	0.51
	YP1	5-10	1	30	100	0	0	-	-	-	-	-	-

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	YP2	0-5	0	0	600	0	2600	0	35.0	2.1	-2	2.1	1.5
	TR1	hole sed	15	7	1000	0	100	hole	11.5	480	87	47	13
	TR1	hole H2O	20	100	900	0	5000	hole	11.5	480	87	47	13
	TR1	0-5	0	0	5900	0	13700	0	32.2	11	8.6	1.9	2.8
	TR1	5-10	0	0	100	0	200	10	29.4	5	19	1.9	4.1
	TR1	10-15	0	3	100	0	0	15	32.8	15	6.2	1.8	2.5
	TR1	15-20	5	0	1000	100	300	20	32.8	9.4	6.4	2	2.6
	TR1	-	-	-	-	-	-	25	33.4	11	4	1.8	2.3
	TR1	-	-	-	-	-	-	29	32.9	12	5.9	1.8	2.5
	TR2	hole sed	35	19	40000	700	20400	hole	15.0	270	74	30	11
	TR2	hole H2O	768	600	6900	100	2100	hole	15.0	270	74	30	11
	TR2	0-5	3	33	12300	100	8700	0	32.6	6.6	7.1	2.1	2.6
	TR2	5-10	0	1	100	0	200	10	31.3	4.8	12	2.1	3.2
	TR2	10-16	5	0	1400	700	18800	15	31.6	5.4	11	2.1	3.1
	TR2	-	-	-	-	-	-	20	31.8	6.5	10	2.1	3
12/16 (350)	YP1	0	5	15	300	0	*nd*	0	35.9	0.45	-5.4	0.73	1.1
	YP1	hole sed	22	9	800	0	*nd*	hole	25.6	19	33	10	5.9
	YP2	0	3	5	0	0	*nd*	0	35.7	0.19	-4.6	0.91	1.2
	YP2	hole sed	6	8	100	0	*nd*	hole	20.7	0.39	52	15	8.2
	TR1	hole sed	35	17	1500	0	*nd*	hole	7.6	102	101	16	14
	TR1	0	10	5	400	0	*nd*	0	33.3	3.5	4.5	1.9	2.3
	TR1	10	11	1	0	0	*nd*	10	22.4	40	46	7.7	7.4
	TR1	15	5	0	0	0	*nd*	15	11.4	80	87	13	13
	TR1	20	2	1	0	0	*nd*	20	8.3	80	99	15	14
	TR1	25	0	0	100	0	*nd*	25	9.5	81	94	8	14
	TR2	hole sed	135	33	*nd*	*nd*	*nd*	hole	36.1	18	-6.08	6.7	1

Appendix. (continued)

Date d of yr	Site	Depth cm	Ent.	F.C.	EMB	SS	TCBS	Depth cm	Cl ‰	NOx	model NOx	Si	model Si
			----------per gram----------							----------uM----------			
	TR2	0	3	5	*nd*	*nd*	*nd*	0	33.8	3.9	2.6	7.9	2.1
	TR2	10	9	0	*nd*	*nd*	*nd*	10	9.5	99	94	16	14
	TR2	15	1	0	*nd*	*nd*	*nd*	15	16.2	58	69	12	11
	TR2	20	0	3	*nd*	*nd*	*nd*	20	6.7	88	104	17	15
	TR2	25	0	0	*nd*	*nd*	*nd*	25	6.8	81	104	17	15