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FLUVIAL SUSPENDED AGGREGATES AND CONTAMINANT ASSOCIATION

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MANAGEMENT PERSPECTIVE

Many contaminants in river systems are associated with suspended sediment. Modelling sediment transport is an important predictive technique for pollutant control. Sediment modelling conventionally uses, or presumes, dispersed size categories. Our work shows that a large proportion of the suspended sediment is flocculated and does not exist as dispersed particles. Also, the bacterial content of sediment flocs appears to be a controlling process for flocculation in freshwater. Bacteria absorb significant amounts of dissolved contaminants.

ABSTRACT

The relationship between suspended particles and contaminant transport has long been recognized. The significance of suspended flocs or aggregates to contaminant transport is, however, not clearly understood. In this study, suspended aggregates from the Sixteen-Mile Creek in Ontario and the Yamaska River in Quebec are used to understand and evaluate the significance of flocculated material to total suspended solids load, contaminant adsorption and transport process. Samples from the Sixteen-Mile Creek and the Yamaska River were sized by digitization and laser particle sizer analysis respectively. Yamaska River samples were also fractionated using a modified cascade filtration technique for bacteriological analysis and for determination of the nature of contaminant interactions with different aggregate size ranges. Suspended aggregates represent a significant proportion of the total suspended solid volume, and contain a large population of bacteria. This work indicates that a relationship may exist between contaminant binding, aggregate size and bacterial density.

KEY WORDS: contaminants, suspended aggregates, flocculation, rivers, bacteria

INTRODUCTION

Suspended particles play a strong role in the biological and chemical balance of the aquatic environment; they are now well known as important vectors for the transport of contaminants in river systems (Ongley et al., 1988 and Allan, 1986). The sorption of contaminants to suspended solids has been shown to be influenced by factors such as mineralogy, surface area, chemical coatings, organic content, and bacterial colonization (Rao et al., 1988 and 1989; Ackermann et al., 1983; Ongley et al., 1981; Hargrave and Kranck, 1976; Pfister et al., 1969; Lotse et al., 1968).

The phenomenon of flocculation and its role in contaminant transport within the fluvial system are, however, largely unknown. The common presumption that fine-grained sediment moves as primary particles reflects traditional sediment sizing techniques that characterize fluvial sediment as dispersed, mineral particles. Recent findings have illustrated that such primary particles may travel and behave as larger particles due to flocculation (Droppo and Ongley, 1989; Partheniades, 1986; Kranck, 1981, 1984; and Krone, 1978). Flocculation seems to be influenced by a number of factors such as sediment and water chemistry, particulate and dissolved organic carbon, fluid shear, temperature, pH and bacterial densities (Rao et al. 1990; Droppo and Ongley, 1989; Tsai et al., 1987; Kranck, 1984; Schubel and Kana, 1972).

Many researchers have focused on the physico-chemical factors which affect flocculation (Luckham and Vincent, 1983; Hunt, 1980; Sholkovitz, 1976). This research, however, reflects the marine environment where electrochemical flocculation is enhanced by the

suppression of the electrical double layer by the salt content of the water. In a freshwater environment, bacteria and their extracellular polymeric exudates may be the predominant factor in promoting and stabilizing flocs as well as playing an important role in contaminant adsorption (Paerl, 1974).

To better understand the role aggregates and associated bacteria play in contaminant transport processes, we studied aggregate characteristics in two fluvial systems; Sixteen-Mile Creek in Ontario (Figure 1) and the Yamaska River in Quebec (Figure 2). Our objectives were to understand and establish: (1) the relationship between bacteria and suspended solids; (2) the particle-size distributions and the bacterial content of different particle-size ranges; and (3) the association of contaminants with different particle-size ranges.

Sampling Sites

Two sample sites were sampled in Sixteen-Mile Creek which drains into Lake Ontario, between Toronto and Hamilton (Figure 1). Site 1 is a harbour which receives sewage effluent and stormwater runoff from urban, agricultural, and forested areas, and has extensive pleasure craft traffic during open water periods. Site 2 is upstream and receives only stormwater runoff from forested and agricultural lands. Samples were from both sites during the period of June 1988 to April 1989.

The Yamaska River is situated in the Eastern townships of Quebec (Figure 2), and receives discharges from various industries, domestic and farm wastes, and runoff of agricultural chemicals. Samples were taken from Station 5 in June and August of 1988.

MATERIALS AND METHODS

Sampling Procedure

The fragility of the aggregates is thought to pose the greatest difficulty in obtaining a represent of floc sample (Kranck, 1984; Gibbs and Konwar, 1982; Kranck and Milligan, 1980). Aggregates are assumed to be in equilibrium with the turbulent forces of the water column. The act of sampling alters the turbulence conditions and may have some disruptive effect on aggregates (Kranck, 1979).

The methods of aggregate sampling and sample transport for Sixteen Mile Creek are those of Droppo and Ongley (1989). Inverted microscope settling chambers were held under the surface of the water parallel to the direction of flow to minimize shearing. The chambers were filled to capacity, capped under water and transported in an upright position to the laboratory for settling and analysis the same day. This method of transport was found by Cleary et al. (1987) to be the least destructive to soil aggregates suspended in water. Droppo and Ongley (1989), following this procedure in a series of controlled experiments, also found that there was no significant aggregate breakage during transport in this manner.

Bacterial content of suspended solids for Sixteen-Mile Creek was sampled in a similar manner. A 1.3 ml vial was held in the direction of flow and capped under water. Vials were transported in an upright position on ice and analyzed for bacterial densities within 3 hours.

Because of distance to our laboratory, sampling at the Yamaska River site required the filling of a 200 L container with zero headspace to dampen turbulence during transport. This large volume of water was later gently agitated in the laboratory to resuspend the aggregates for sub-sampling and bacterial and particle size analysis.

Particle Size Analysis

The two study areas were originally selected for quite different purposes. Consequently, two types of aggregate-sizing techniques were used in this study. This method was used because of the lack of any convention for sizing of particle aggregates and the inherent limitations of existing sizing apparatus for flocculated sediments. Commercial sizing apparatuses are inherently destructive to flocs due to agitation and/or pumping required to circulate the suspension through the apparatus.

Size distribution for suspended aggregates from the Yamaska River was performed by a laser particle size analyzer (Malvern Instruments Ltd. Model 2600C) without sonication or chemical dispersion. This system comprises a light source (3 mV laser), and a receiving optic assembly interfaced with a microcomputer (Bale and Morris, 1987). Particle size distribution is derived from measurements of the near-forward Fraunhofer diffraction spectrum produced by a particle group randomly distributed in a sample cell mounted in the beam path between the laser source and the detector array. Although the Malvern analyzer does not require pumping of the sample suspension, some degree of aggregate breakage may result from turbulence caused by the magnetic stirrer. This method has the advantage that it measures the size distribution of thousands to millions of particles depending on the size of the particles and concentration of the suspension. It does not, however, permit analysis of individual flocs.

Sizing of Sixteen-Mile Creek particle aggregates used the method described in Droppo and Ongley (1989) for examining individual flocs.

The method comprised a combination of microscopy, photography and digitization. The settling column slide was placed on an inverted microscope, and transparencies were taken of the aggregates at 100x magnification at numerous stage positions. The transparencies were projected onto a translucent digitizer where the floc perimeters were digitized by hand. Although flocs tend to be irregular in shape, the digital output was transformed into equivalent spherical diameters (ESD) using the equation;

$$ESD = \sqrt{\frac{4.Ai}{\pi}}$$

Where Ai = the area of the flocs to provide a basis for size comparison. The numerical results were then subdivided into size classes. This technique permits analysis of individual flocs, but is limited in the number of flocs that can be measured (generally 500) due to the labour-intensive nature of the analysis.

Fractionation of Aggregate-Size Classes

A 5 L subsample of the Yamaska River water was fractionated using a modified cascade filtration procedure (Rao & Kwan, 1990) to estimate bacterial density and contaminant binding affinity of respective fractions. Cascade filters were Nitex nylon sheet type filters with sieve sizes of 88 µm, 64 µm, 40 µm, 20 µm. The particle size classes of 8-20 µm and 3-8 µm were obtained using 8 and 3 µm polycarbonate Nuclepore filters respectively and membrane filtration procedure. The classical cascade filtration procedure is widely used by researchers

for the purpose of fractionating biological materials (De Vitre et al., 1988; Leppard et al., 1988 and 1989; Perret et al., 1988 and Munawar et al., 1983). This technique has, however, some inherent limitation for aggregate sizing. As the aggregates collect on the filter surface, the filtering efficiency changes due to pore size clogging. Thus the filtrate may not contain all the particles that are expected to pass through. Also, the particles in the filtrate may have a tendency to flocculate and/or disintegrate, resulting in a spectrum of particles ranging from above and below the targeted size class. We make the assumption that the aggregates are stable within the limits of the cascade filtration method. Tests by Droppo and Ongley (1989) of aggregate stability during transport and our more recent microscopic observations of aggregates under moderate agitation on a microscopic slide lend support to this assumption; however, there is no independent measure of aggregate behaviour during filtration.

Microbiological Analysis

Bacterial content of each size fraction was determined using the acridine orange direct microscopic counting procedure (Rao et al., 1984). Suspended aggregates were resuspended from each filter; 1 ml of 1/10 dilution of each of the fractions was thoroughly homogenized (Marxsen, 1988) using a vortex mixer at #10 speed setting for 1 min. This procedure was used to obtain as uniform a dispersion as possible of bacteria from the aggregates. The homogenized mixture was immediately subjected to membrane filtration (0.1 µm Nuclepore membrane) and staining with acridine orange for 3 min. Total microscopic counts were determined using phase-contrast illumination (Rao et al., 1984).

A more general method of bacterial counts was used on Sixteen-Mile Creek suspended aggregates. The size of bacteria in freshwater and on suspended particles is generally small, ranging from 0.3 to 0.7 µm in diameter (Hobbie et al., 1977). The particle aggregates measured by digitization are far larger than this range. Therefore, free-floating bacteria were separated from aggregate-bound bacteria (in a low response water suspension) by filtration through a 1.0 µm Nuclepore filter. This process retains the aggregates and aggregate-bound bacteria on the filter while free-floating bacteria pass through the filter. While we do not believe the procedure is totally effective, it seems to reduce interference in the subsequent counts that might be induced by large amounts of free-floating bacteria that would otherwise be incorporated into the aggregate matrix during filtration. Total microscopic counts of bacteria were determined following the procedure of Rao et al. (1984).

Contaminant Binding

Using low response water a 200 ml suspension was made of each of the different size fractions from the Yamaska River. Two water soluble organic dyes, Acid Orange 60 and Basic Violet 1 (log octanol-water partition coefficients of 0.66 $\pm$ 0.24 and -0.17 $\pm$ 0.05 respectively (Rao et al. 1989)), representative of some dyes used in the Yamaska River basin, were tested for binding to suspended aggregates. Each suspension was treated with a 5 ppm solution of one or both of the contaminant dyes in order to establish the nature and extent of binding efficiency of these different size fractions. The sorption experiments were performed in triplicate in the dark in order to exclude photodegradation interferences of these photosensitive dyes. The concentrations of the dyes in the reaction mixtures were

determined spectrophotometrically at 24 h intervals after centrifugation (500xg for 10 min.). Throughout the experiment a control series was run using dye in distilled water. The absorbence maxima for Basic Violet 1 and Acid Orange 60 at 586 nm and 273 nm respectively, were used to determine the dye concentration. The results were calculated and expressed as mg dye removed by mg (dry wt.) aggregate fractions in each of these fractions.

Results and Discussion

Data on suspended sediments are typically analyzed and reported as primary particles. However, recent findings (Droppo and Ongley, 1989) indicate that flocculation is occuring in the freshwater fluvial system and that the naturally-occurring particle aggregates can represent a significant proportion of suspended sediment in river systems. Aggregate sizes in Sixteen-Mile Creek were far larger than the existing primary particle and ranged from 3 to 60 microns with a median size approximately 9 microns. The aggregates represent only 10 to 27% of the total number of suspended particles, but comprised 92 to 98% of the total suspended sediment volume (Droppo and Ongley, 1989). Schubel and Kana (1972) also found in an estuarine sample that aggregates made up only 11% of the total number of suspended particles but nearly 97% of the total suspended sediment volume. Thus, aggregates represent a significant portion of the suspended sediment and should therefore be considered as an important component involved in the transport of contaminants.

Because aggregates are comprised of inorganic and organic material in various stages of decomposition, they provide a ready nutrient base for microbes (Marshall, 1976). This nutrient

availability (generally carbon, nitrogen and phosphorus) supports elevated bacterial biomass associated with suspended sediments (Palmateer et al. 1989; Rao et al. 1990). Biddanda (1985) has demonstrated that active bacteria are necessary for aggregate formation. Total bacterial counts as related to the Sixteen-Mile Creek range from 0.76 to 15.7 x 10^7 and 1.74 to 11.24 x 10^7 /mg suspended solids for site 1 and site 2 respectively over the sampling period. These ranges are similar to those reported by Cammen and Walker (1982) for Bay of Fundy suspended solids. Total bacteria counts in the suspended aggregates are also comparable to or higher than bacteria reported from lake bottom sediment studies (Dutka and Kwan, 1983, Rao and Jurkovic, 1977, Dutka et al. 1974). The bacterial populations in the suspended aggregates may have similar functional significance to bacteria in bottom sediments for biochemical processes such as aggregate formation, biodegradation, biotransformation and physico-chemical binding of extraneous substances (Biddanda, 1985, Paerl, 1974, Sorokin and Kadota, 1972).

Results from the preliminary analysis of the Yamaska River water indicate that the predominant aggregate size fraction is 20 to 40 microns (Table 1). (Comparison of the cascade fractionation size classes with the digitizing size classes is not possible due to their methodological and theoretical differences). Relative to the other size fractions, the 20 - 40 µ fraction possesses the largest bacterial densities for the June and August samples (8.3 and 7.9 x 10^5 respectively - Table 1). Experiments performed on the June sample also indicated that the 20 to 40 µm fraction had the largest contaminant binding property (Figure 3). Paerl (1974) illustrated

using tritiated acetate and glucose that aggregates do take up dissolved organic compounds through bacterial mediation. He found that bacteria utilize the organic compounds and excreted them as bacterial exudates. These bacterial exudates serve as binding material for extracellular substances. The larger contaminant absorption of the predominant 20 - 40 µm fraction of the Yamaska River may therefore be related to its larger bacterial concentration and their associated exudates. The degree of absorption is also dependent on the solubility of the contaminant. The less soluble an organic contaminant is, the greater will be its absorption to the particulate phase. It should be noted that the contaminants (Acid orange 60 and basic violet 1) used in this study are highly soluble as illustrated by their low partition coefficients (Rao et al., 1990).

CONCLUSION

These preliminary findings suggest that the development of freshwater particulate aggregates and their apparent high affinity for contaminant binding may be associated with bacteria and their polymeric exudates. We found that there appears to be a predominant aggregate size of 20 - 40 µm in the Yamaska River which possesses the largest number of bacteria and the largest contaminant binding affinity for two representative organic industrial dyes. If the controlling factor in adsorption were surface area considerations, then the finest fraction (3-8 µm) should show the greatest contaminant binding association. More in-depth research is currently underway to further elucidate these findings.

Because aggregates can represent a significant volume of suspended material, it is perhaps essential that attention be focused on the transport characteristics and composition of the suspended organic-rich aggregates when developing contaminant transport models. If this research establishes that there is a consistent predominant size fraction which demonstrates the greatest contaminant binding affinity for other river systems, then this will have significant implications for the modelling of contaminants transported in association with suspended fine-grained particulate matter.

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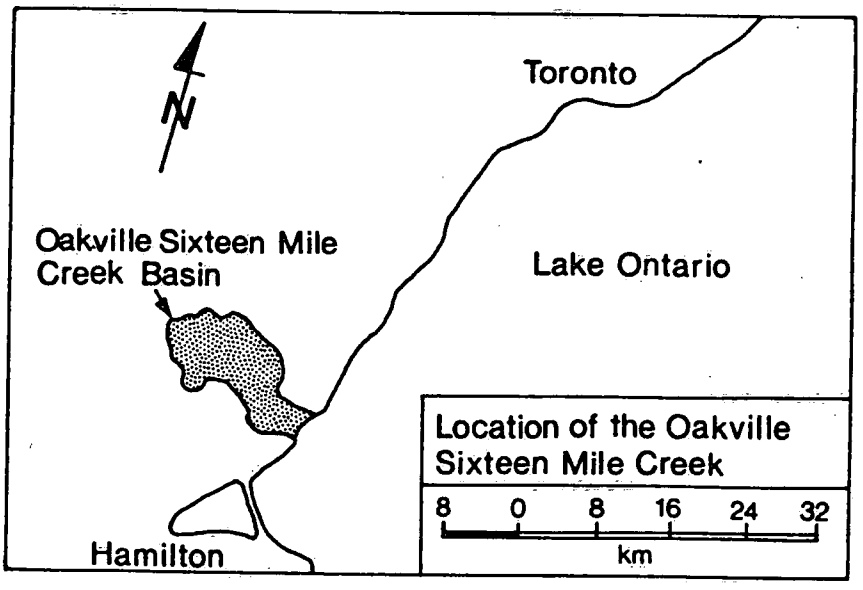
FIGURES

Table 1. Particle size distribution as measured by Malvern particle size analyzer and associated bacterial densities for the Yamaska River.

Particle Size (Range µm)	% by volume in the river water		Bacterial Densities ($x10^5$/ml)	Bacterial Densities ($x10^5$/ml)
	June	August	June	August
3 - 8	24.7	12.4	3.5	-
8 - 20	21.9	18.5	3.7	3.4
20 - 40*	29.9	26.8	8.3	7.9
40 - 64	11.0	16.4	6.8	5.2
64 - 88	3.5	9.8	3.6	6.0
>88	3.3	8.1	-	3.1
<3	6.5	8.0	-	-

* denotes predominant size fraction



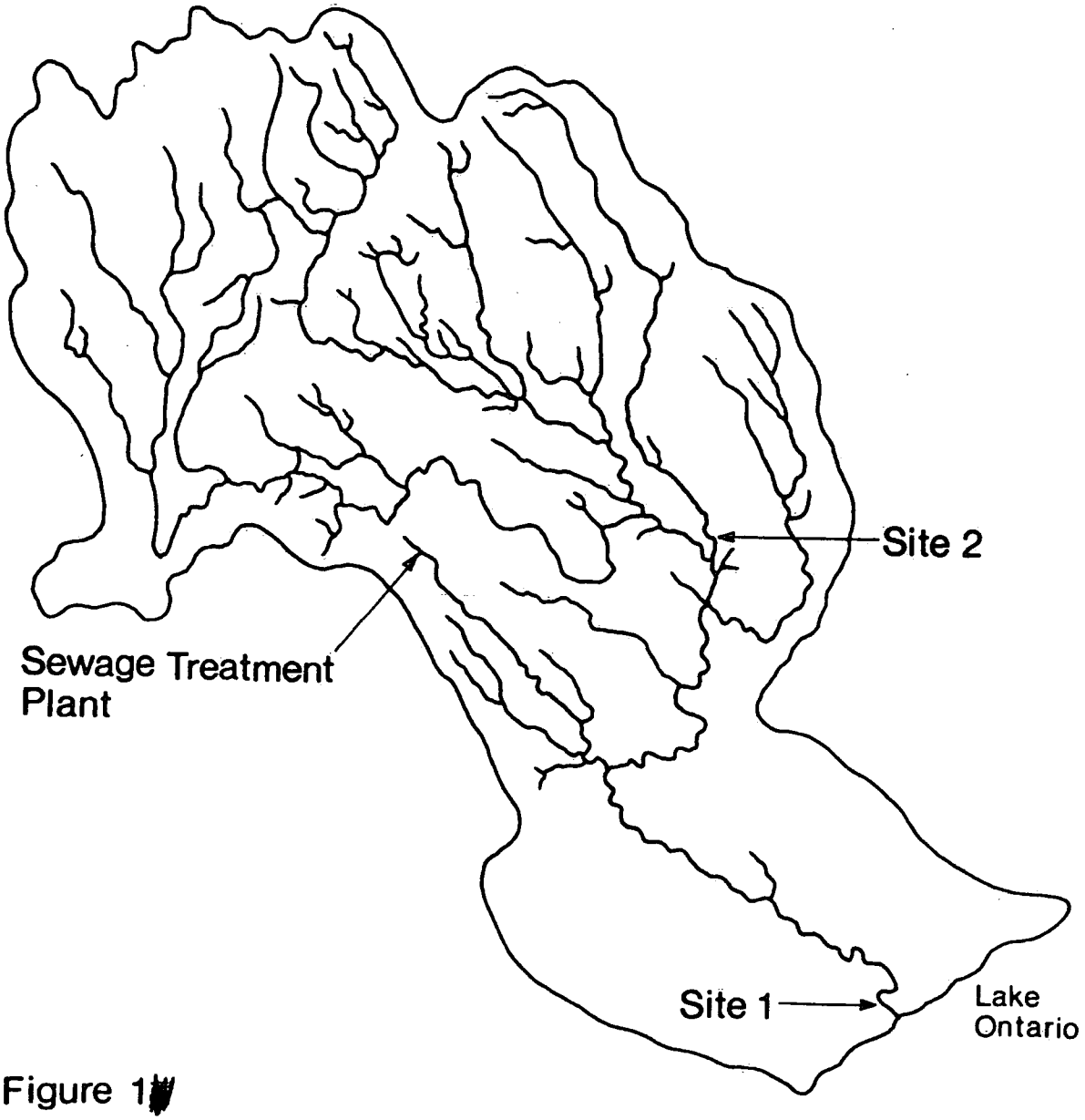
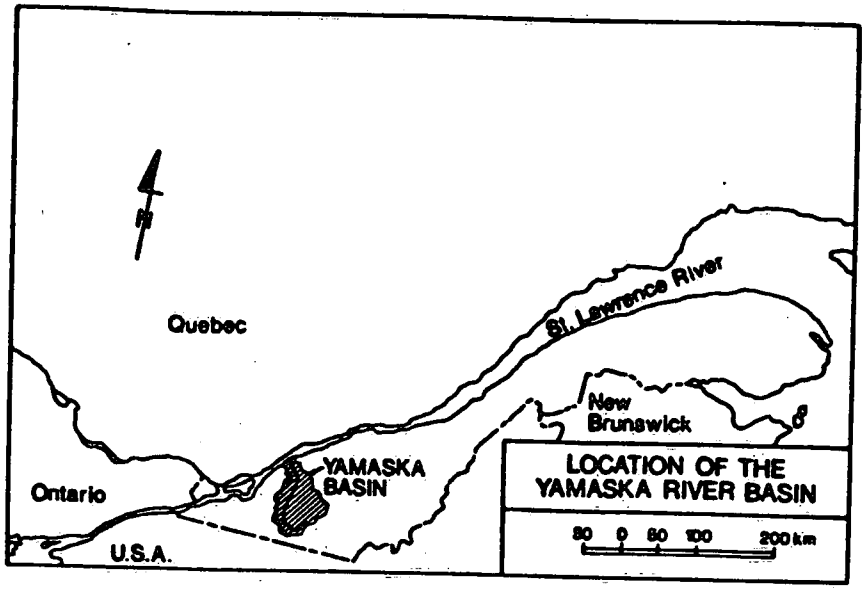


Figure 1

Oakville Sixteen Mile Creek Basin and Sample Sites.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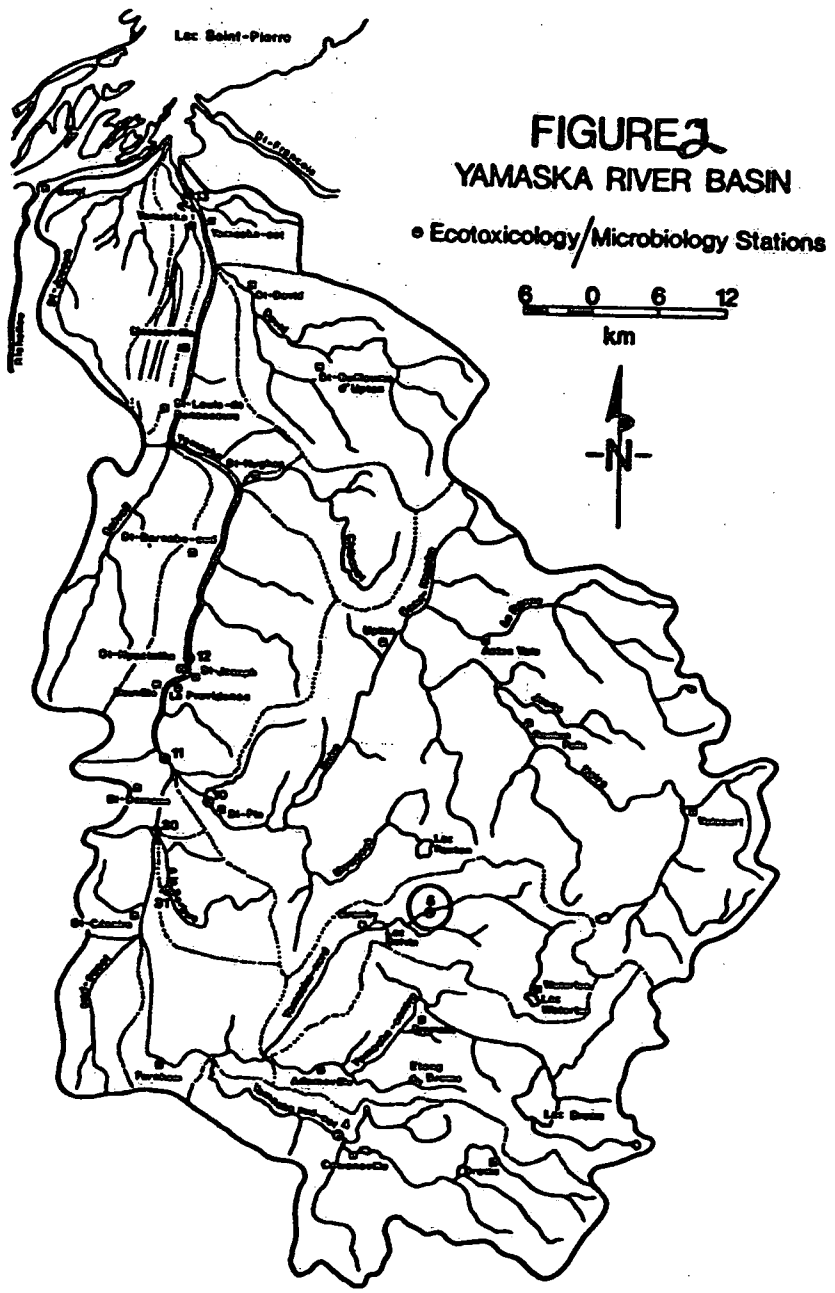
New Brunswick
Ontario
YAMASKA BASIN
U.S.A.
LOCATION OF THE YAMASKA RIVER BASIN
80 0 80 100 200 km


Lac Saint-Pierre
FIGURE 1
YAMASKA RIVER BASIN
• Ecotoxicology/Microbiology Stations
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km
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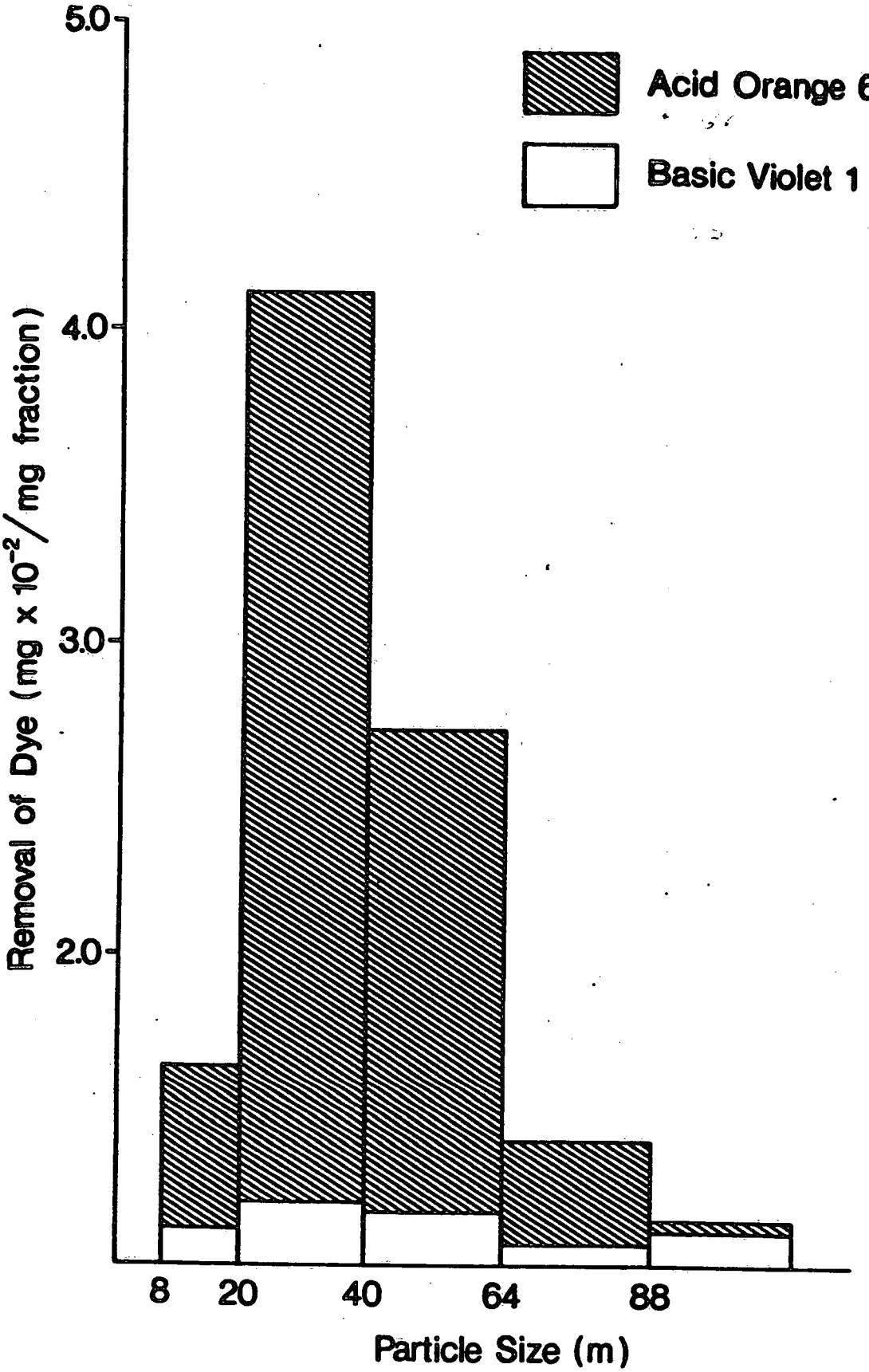

Fig. 3 Effect of Particle size on the contaminant dye removal in the Yamaska River (24 hr)