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Flocculation of fine-grained suspended solids in the river continuum

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Abstract Knowledge of factors that control flocculation along a river gradient is necessary to develop more realistic models of fine-grained sediment and associated pollutant transport in the river continuum. A direct image analysis procedure is used to characterize the number and size distribution of both primary and flocculated particles suspended in the water column of the Nith River. Sample locations in this southern Ontario river were determined according to the Strahler stream ordering system. Suspended solids concentration, water chemistry and river flow characteristics are examined to elucidate their effect on flocculation. In the Nith River, flocs represent a small proportion of the total number of particles transported but most ($> 90\%$) of the total volume of suspended solids. An increase in the floc median diameter (D_{50}) and floc number was observed with decreasing suspended solids concentration. Variables such as DO, pH, SS, DOC and TDS influence floc size (D_{50}) and floc number (floc-primary particle ratio). While floc number was variable, floc size distributions were similar irrespective of stream order.

INTRODUCTION

The term flocculation refers to the process whereby larger particles in the water column are formed from the aggregation of smaller particles through physical, chemical and biological means (Droppo & Ongley, 1994). Flocculation affects the size, surface area, density, settling velocity and deposition rate of suspended solids (Tsai *et al.*, 1987) and can affect the fate of sediment associated contaminants in rivers (Stone & Droppo, 1994). Much of the information on flocculation has evolved from marine (Kranck, 1975) and wastewater technology research (Bartlett, 1971). Fine-grained fluvial sediment is transported in a flocculated state (Sherman, 1953; Guy, 1969; Wall *et al.*, 1978). However, only recently has research focused on the complex interactions of physical, chemical and biological processes that control the phenomena of flocculation in freshwater fluvial systems.

Recent advances in flocculation research are largely due to the development and use of *in situ* sediment sizing instruments such as the modified Malvern Particle Size Analyzer, the Plankton Camera and the Floc Camera Assembly (Syvitski, 1991). A direct observation image analysis system has also been used to examine flocculation of suspended solids in rivers of southeastern Canada (Droppo & Ongley, 1992, 1994) and measure size characteristics of fine-grained surficial deposits in rivers (Droppo & Stone, 1994). These studies and others (Tsai *et al.*, 1987; Kranck, 1979; Krone, 1978) show that the concentration of particulate organic carbon, suspended solids and bacterial activity are important potential controlling factors of flocculation. To our knowledge, no research has examined how changing physical and chemical conditions in the river continuum, from headwaters to mouth, influence the particle dynamics of floc formation. Similarly, little is known about the significance of flocs relative to the total suspended solids load and their role in sediment associated chemical transport.

In this study, we report preliminary results of an ongoing study to investigate spatial aspects of flocculation in freshwater river environments. The objectives of this research are; (a) to examine cross-sectional and longitudinal variability in the size distribution and number of both primary and flocculated suspended particles of the Nith River and (b) to investigate potential physical and chemical factors controlling flocculation at each study site. These factors include stream gradient, stream width, flow velocity, discharge, temperature, dissolved oxygen (DO), pH, total dissolved and suspended solids, dissolved organic carbon (DOC), dissolved inorganic carbon (DIC), total organic carbon (TOC), particulate organic carbon (POC), soluble reactive phosphate (SRP), total phosphorus (TP) and the concentration of dissolved ions (Na, K, Mg, Ca, Al, Fe and Mn).

METHODOLOGY

The Nith River drainage basin is located in southern Ontario (Fig. 1). Physiographic and land use characteristics of this predominantly agricultural basin have previously been described (So & Singer, 1982). The drainage pattern of the Nith River was classified according to the Strahler stream ordering technique (Strahler, 1957).

A study site near Ayr, Ontario was used to examine cross sectional variability in floc size and distribution during conditions of low flow. The transect was divided into five panels of equal discharge. Suspended solids, associated flocculated materials and water chemistry were sampled 30 cm from the surface at the centroid of each panel. The size distribution of flocs was determined with a direct observation image analysis procedure (Droppo & Ongley, 1992).

Longitudinal variability in floc size and distribution was examined at five study sites (Fig. 1). Each study site reflects changing physical, chemical and biological gradients of the river associated with increasing stream order, from 1 to 5. Flocs were sampled in the centroid of flow at each site and sediment data (flocs and primary particles) are reported as number of particles per millilitre. Ratios of floc to primary particles and the percent by number for floc distributions are characterized by their median diameter (D_{50}). The lower resolution limit of the direct observation image analysis method is approximately 2 μm and size distributions reported in this paper do not represent the actual distribution of particles below this limit. Water and suspended sediment samples were collected on 13 July 1993, three days after a moderate summer rainfall. Suspended solids concentration was determined by filtering a known sample volume onto a tared 0.45 μm filter. Dissolved oxygen, temperature and pH were determined in the field with calibrated portable meters. Major ions (Na, K, Mg, Ca, Al, Fe, Mn), SRP, TP, TOC and DOC were analysed according to standard methods (Environment Canada, 1979).

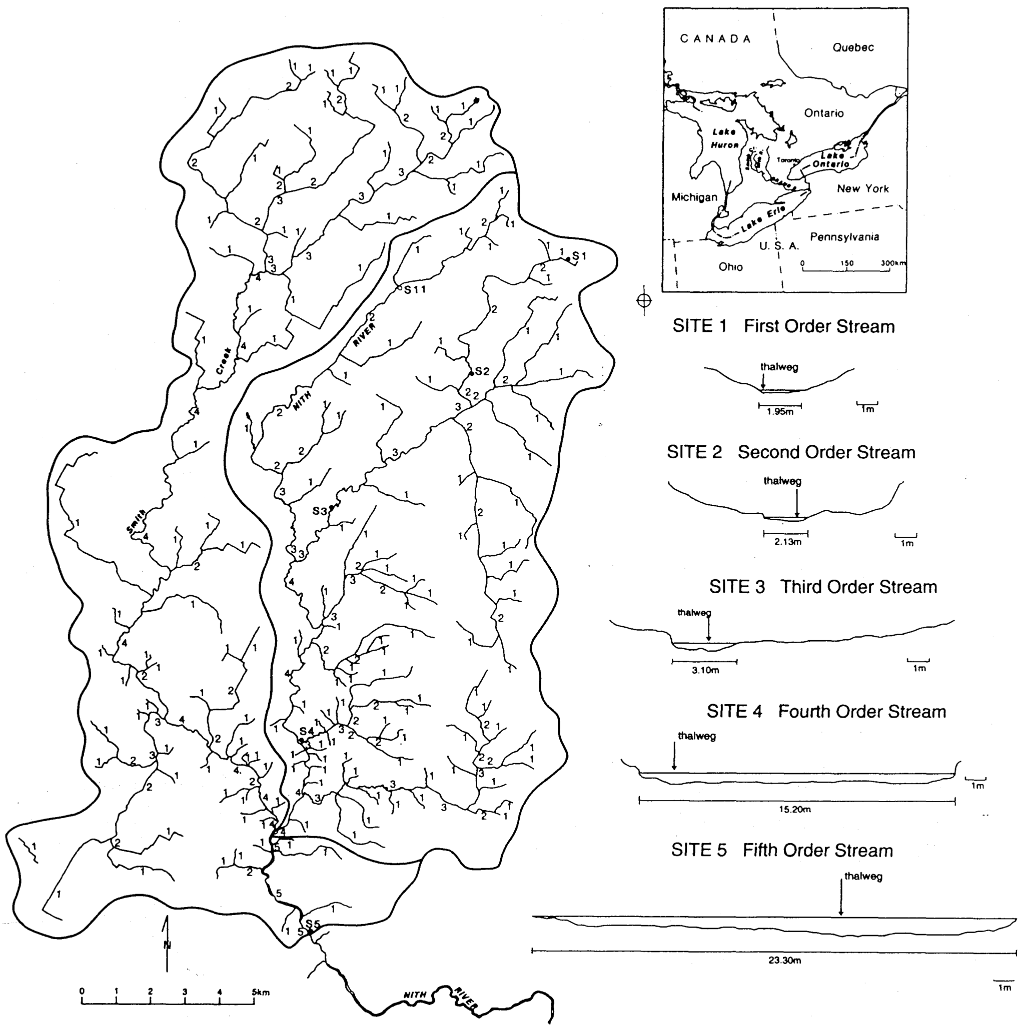

Fig 1. Study area.

RESULTS AND DISCUSSION

Cross-sectional variability in flocs

Previous work at the Ayr site has shown that the suspended solids load consists primarily of materials <63 μm and the cross-sectional concentration of suspended solids is relatively homogeneous for a full range of flows (Droppo, unpublished data). There is little cross-sectional variability in the physical and chemical variables of the five flow weighted panels at the Ayr site (Table 1). The median diameter (D_{50}) of these particles ranged from 3.3 to 3.6 μm and the largest number of flocculated particles was concentrated in the 2-3.7 μm range (Fig. 2). The distribution of flocs at this site is bimodal with an apparent lack of particles in the 3.7-4.9 μm range. Reasons for this previously observed 4-5 μm particle deficiency have been advanced by Stone & Saunderson (1992)

Table 1 Sediment, water chemistry and flow characteristics of the Ayr site.

	Flow weighted panel:				
	1	2	3	4	5
Depth (m)	1.22	1.12	1.02	0.89	0.62
Velocity (m s^{-1})	0.263	0.308	0.267	0.258	0.198
Q (m^3 s^{-1})	0.75	0.75	0.75	0.75	0.75
Temperature (°C)	23	23	23	23	23
SS (mg l^{-1})	4.81 ± (1.79)	4.54 ± (0.45)	4.40 ± (0.17)	5.09 ± (0.02)	7.17 ± (4.43)
POC (mg l^{-1})	0.728	0.765	0.832	0.834	0.755
DOC (mg l^{-1})	4.8	4.8	4.7	4.8	4.8
DIC (mg l^{-1})	44.7	44.7	45.6	45.9	46.1
TP (μgP l^{-1})	31.8	31.7	30.8	30.4	57.2
SRP (μgP l^{-1})	11.9	11.8	11.6	11.8	11.7
Floc D_{50}	3.59	3.28	3.43	3.61	3.31
Total flocs per ml	32	37	32	36	33

but recent evidence suggests that this size range may represent a possible preferential particle size for the process of flocculation (Droppo & Stone, 1994).

Longitudinal variability in flocs

The stream width, wetted perimeter and instantaneous discharge at study sites 1 to 5 increased with stream order (Table 2). Suspended sediment load increased downstream and the floc D_{50} ranged from 8.2-13.2 μm. Except for site 2, the number (Table 3) and size distribution (Fig. 3) of flocs were similar irrespective of stream order. In contrast to the Ayr site data, the frequency floc grain size data for sites 1 to 5 more closely approximated a normal distribution. However, the combined size distribution of flocs and primary particles was bimodal with a particle deficiency in the 3.5-5.3 μm range.

The majority (71-82%) of primary particles in the Nith River are found in the >1-5 μm size fraction (Table 4). Flocculated particles represent a small proportion of the total number of particles transported but most (>90%) of the total volume of suspended solids. As a proportion of the total number of particles, flocs ranged from a maximum of 21% (site 2) to a minimum of 7% (site 3) and the ratio of flocculated to primary particles was 0.27 and 0.07 at these sites, respectively. Site 2 was characterized by extensive in-channel and bank vegetation. The reduction of suspended solids and total number of flocs at this site (Fig. 3) is attributed to the sediment trapping efficiency of the abundant in-channel vegetation and reduced sediment availability from the river bank and bed. At this site, the channel bottom was characterized by coarse gravel and sand compared to finer silt-clay materials at the other four sites.

Results of our preliminary investigation suggest that decreasing suspended solids concentration is associated with increasing floc number and D_{50}. In laboratory

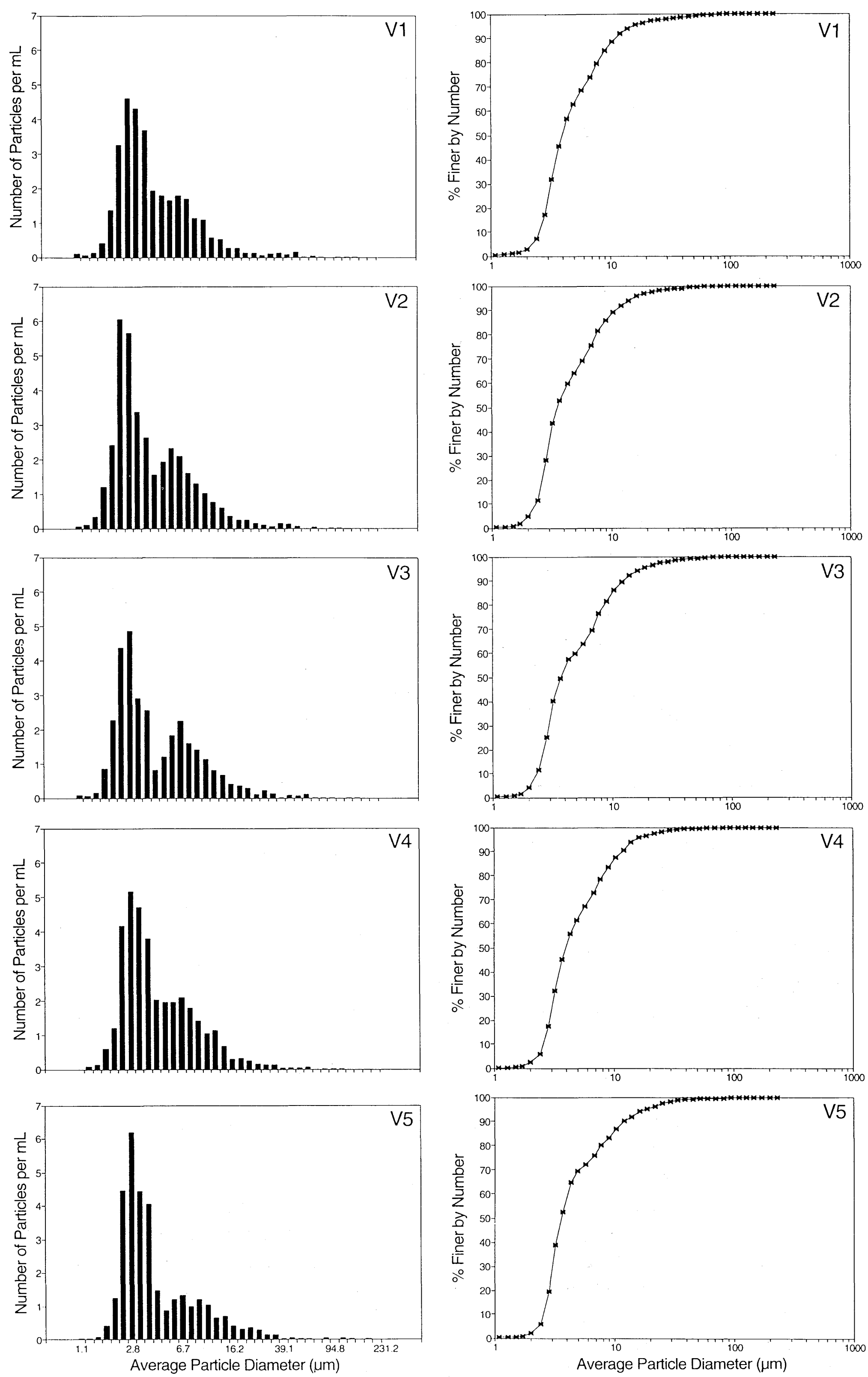

Fig 2. Cross-sectional distribution of floc number by size class.

Table 2 Hydraulic geometry of study sites 1 to 5.

	Study site: 1	2	3	4	5
Stream order	1	2	3	4	5
Contributing basin area (km^2)	1.68	9.58	57.53	150.03	310.60
Stream width (m)	1.95	2.13	3.10	15.20	23.30
Stream depth at thalweg (m)	0.12	0.09	0.32	0.46	0.77
Wet perimeter area (m^2)	0.16	0.22	0.62	5.86	12.80
Stream velocity at thalweg (m s^{-1})	0.01	0.17	0.47	0.50	0.26
Instantaneous discharge (m^3 s^{-1})	0.00	0.04	0.29	1.17	1.70
Stream gradient (%)	0.26	0.28	0.11	0.15	0.10

Table 3 Sediment and floc size characteristics.

	Study site: 1	2	3	4	5
Total number of particles ml^{-1}	13 067	76	16 542	12 615	9926
Total number of flocs ml^{-1}	1039	13	1145	1543	990
Total number of primary particles ml^{-1}	12 028	60	15 397	11 072	8936
Floc D_{50} (μm)	13.2	13.0	8.2	10.1	11.4
Floc primary particle ratio	0.09	0.27	0.07	0.14	0.11
Percentage flocculated particles	8.0	21.2	6.9	12.2	10.0
Suspended sediment (mg l^{-1})	18.1	5.9	56.1	50.1	57.8
Suspended sediment load (kg h^{-1})	0.1	0.8	59.2	211.2	353.6

experiments conducted at constant shear stress, a decrease in floc size was reported as sediment concentration increased (Tsai *et al.*, 1987). Conversely, results of a field investigation suggest that increasing sediment concentration leads to an increase in particle collisions and increasing floc size (Kranck, 1979). The latter relationship was also observed by Droppo & Ongley (1994) in southern Ontario rivers. Opposing views of the relationship between suspended solids and floc size/number indicate the complexity of the flocculation process in different aquatic environments and demonstrate the need to investigate other important contributing variables. The simultaneous interplay of changing velocity, turbulence, bed shear and solution chemistry as well as variable inputs of dissolved and particulate materials to the river from surface runoff, tile drainage and groundwater will ultimately affect the timing and amount of flocculation in the river continuum.

At site 3, the observed increased number of primary particles, decreased floc D_{50} and low floc-primary particle ratio may in part be related to increased velocity and

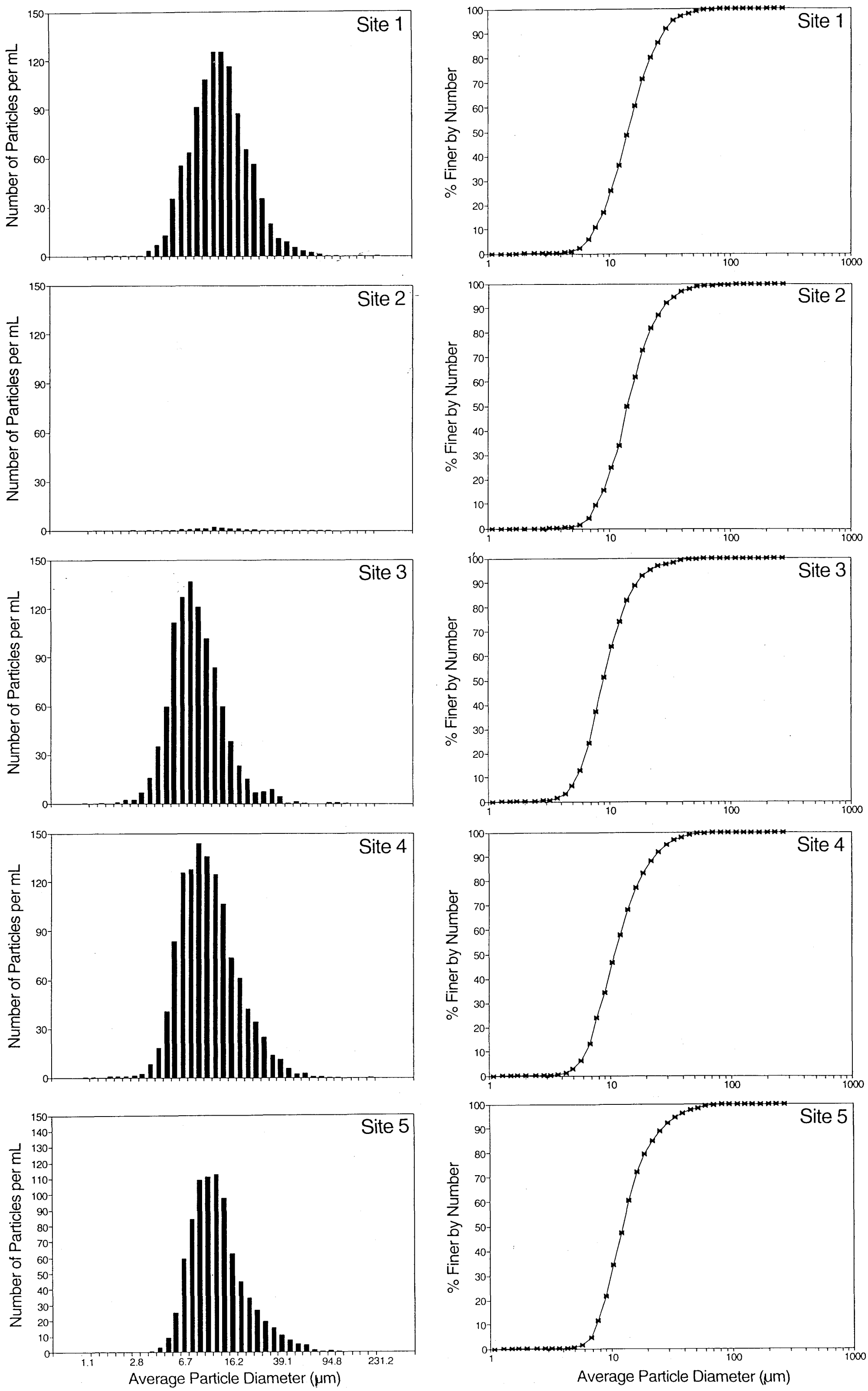

Fig 3. Longitudinal distribution of floc number by size class.

Table 4 Primary particle size characteristics (# of particles per size class).

Particle size range (μm)	Study site: 1	2	3	4	5
0-1	0	1 (1.7)	0	0	0
>1-5	9588 (79.7)	45 (75)	12 615 (81.9)	8290 (74.9)	6312 (70.6)
>5-20	2127 (17.9)	14 (23.3)	2205 (14.3)	2419 (21.8)	2310 (25.9)
>20-65	305 (2.5)	0	563 (3.7)	363 (3.3)	314 (3.5)
>65	8 (0.1)	0	14 (0.1)	0	0

( ) = % of total primary particles.

associated turbulence (Krone, 1978). The increased bed shear in the shallow water column and subsequent increased probability for particle collisions may have resulted in floc breakage. The increased number of primary particles at this site could also be attributed to both agricultural tile drainage inputs of fine-grained materials and re-suspension of river bottom sediment in areas of unrestricted cattle access above this site.

Table 5 Water chemistry.

	Study site: 1	2	3	4	5
pH	7.6	8.0	7.5	7.7	7.8
Dissolved oxygen (mg l^{-1})	6.2	10.4	4.2	6.8	6.9
Water temperature (°C)	17.8	21.5	22.6	23.5	24.2
NTU	132.0	3.6	72.0	76.0	72.0
TDS (mg l^{-1})	610.7	479.7	495.2	463.0	443.4
TDS load (kg h^{-1})	3.6	18.3	145.1	543.3	754.8
TOC (mg l^{-1})	7.68	12.05	13.01	12.46	10.53
DOC (mg l^{-1})	5.23	9.44	9.60	9.82	9.60
SRP (mg l^{-1})	0.079	0.011	0.350	0.315	0.154
Total phosphorus (mg l^{-1})	0.113	0.030	0.438	0.415	0.253
Na (mg l^{-1})	16.17	19.29	10.68	10.19	11.03
Mg (mg l^{-1})	24.75	17.47	17.43	15.23	17.22
K (mg l^{-1})	2.86	4.13	7.55	8.68	6.70
Ca (mg l^{-1})	97.37	60.38	80.78	68.78	68.88
Fe (mg l^{-1})	0.01	0.01	0.09	0.12	0.16
Al (mg l^{-1})	4.10	7.15	9.55	12.77	16.98
Mn (mg l^{-1})	nd	nd	0.02	0.02	0.03

Table 6 Correlation coefficients between flocs, water chemistry and hydraulic geometry ($n = 5$, $p \leq 0.05$).

	% Flocs	Floc/primary ratio	Floc D_{50}	Stream order
Stream order			−0.49	
Stream width				0.91
Stream gradient			−0.66	−0.82
SS	−0.63	−0.65	−0.80	0.82
Instantaneous Q				0.95
pH	0.92	0.92	0.64	
Temperature			−0.58	0.93
Dissolved oxygen	0.95	0.95	0.64	
TDS			−0.70	
TOC			−0.57	0.73
DOC	−0.58	−0.59	−0.94	0.58
Total P	−0.57	−0.58	−0.95	
SRP	0.66	0.68	0.80	−0.76
Na			−0.85	0.80
Ca	−0.74	−0.73		−0.53
Mg			0.56	−0.75
Fe			−0.57	0.97
Mn			−0.61	0.96
Al				0.99

Downstream variability in Nith River chemistry reflects in part, varying hydrological flowpathways and land use characteristics of each study site. Only temperature and concentrations of some cations (Mn, Al, Fe) increased downstream (Table 5). Correlation coefficients between floc characteristics (% flocs, floc-primary particle ratio, floc D_{50}), water chemistry and hydraulic factors shown in Table 6 are consistent with the observation of some previous laboratory and field investigations. However, due to the small data set, associations between these variables should only be used to make general inferences about potentially important variables affecting flocculation in the river continuum and to suggest areas of future research.

Nevertheless, Table 6 suggests that DO, pH, SS, DOC and TDS influence floc size (D_{50}) and floc number (floc-primary particle ratio). The link between oxygen content and flocculation in fluvial systems is indirect. Oxygen is required for the metabolism of some bacterial species (Morel & Hering, 1993) and the presence of bacteria can promote flocculation (Muschenheim *et al.*, 1989). The increased flocculation (floc-primary particle ratio) at sites in the Nith River with high DO may result from the presence of cyanobacterial species which have been found to produce extracelluar polyanions that are effective in flocculation and sedimenting clay particles (Levy *et al.*, 1992). Humic substances are pH-dependent and can affect the flocculation behaviour of clays. Tarchitzky *et al.* (1993) report that flocculation of clays increases with increasing humic substance concentration and with the increase negative charges as water pH is raised. Increasing sodium adsorption ratio and increasing pH may also promote flocculation of clay mixtures (Goldberg *et al.*, 1991).

Another factor that affects the degree of flocculation may be related to the source of particulate and dissolved materials to the river system which can vary considerably in a watershed. Headwaters are typically dominated by inputs from the adjacent landscape while middle-order streams are generally dominated by upstream inputs. Groundwater is a major contributor of DOC to stream water (Rutherford & Hynes, 1987) and variable flow pathways in the hyporheic zone alter the solute transport characteristics in rivers (Bencala, 1993). The physical, chemical and biological characteristics of tile drain and other terrestrial surface inputs in the headwaters of the Nith River affect the quality and quantity of dissolved and particulate materials transported to the water column. Organic inputs from these sources to the river will increase the DOC content and biological oxygen demand (BOD) while reducing the DO content and pH of the water. Such a shift in redox potential could explain the formation of smaller inorganic and organic particles through preferential adsorption of metal-organic complexes and result in a lower floc D_{50} (Ward *et al.*, 1990).

A drainage basin can be viewed as a gradient of physical, chemical and biological conditions that influence the source, availability and distribution of suspended solids within a river system (Vannote *et al.*, 1980). The gradient is based primarily on energy conditions of the system, where sediment erosion, transport and deposition are regulated to a large extent by fluvial geomorphological processes, from headwaters to the river mouth. However, biological and chemical gradients play an important role in the formational processes that affect both the nature and significance of fine-grained solids in freshwater fluvial systems. Considerable research is required to evaluate the timing and importance of changing downstream physical, chemical and biological variables that affect flocculation and associated-chemical transport in the river continuum.

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