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Structural Controls on Floc Strength and Transport

By:

Ian G. Droppo

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NWRI RESEARCH SUMMARY

Plain language title
Internal and external structure of composite particles influences sediment and pollutant transport behaviour.

What is the problem and what do sicentists already know about it?
Composite flocculated particles are ubiquitous within aquatic environments. A significant amount of research exists to explain how shear influences floc development and transport. We have a relatively poor understanding, however, on how the actual structure and composition of the composite particles influences the transport of sediment and therefore associated contaminants. This paper serves to provide some perspective on this issue through the use of optical image analysis techniques performed on flocs sampled from two distinctly different environments (rivers and combined sewer overflows).

Why did NWRI do this study?
This work is part of an ongoing study to help improve our predictive abilities of contaminated sediment transport, erosion and deposition. The information gained from this study will be used to improve numerical models that will in turn assist in the identification and evaluation of remedial measures for the improved management of our water resources.

What were the results?
Results suggest that the biological community (primarily the bacteria) is the dominant component that mediates the internal and external structural characteristics of the floc, regardless of the environment. The bacterial community not only influences floc development through its sticky nature (including its secretion of polymeric fibrils) promoting adhesion, but also through its influence over particle density, porosity and strength. All of these factors influence how a floc and associated contaminants are transported within the aquatic environment.

How will these results be used?
Results will be used to assist our understanding of particle and contaminant movement within the aquatic environment and to improve numerical models for such predictions.

Who were our main partners in the study?
This study is done in partnership with McMaster University, Ryerson University and the University of Exeter, UK.

Influence de la structure sur la force et le transport des flocs

Ian G. Droppo

Résumé : Le présent document examine en quoi la structure externe et interne des flocs influe sur leur force et leur transport dans divers milieux aquatiques. Nous avons mené l'étude dans des réseaux de collecte d'eaux usées et des réseaux fluviaux pour établir une corrélation entre la structure du floc, d'une part, sa force et son transport, d'autre part. La matrice interne du floc étudié semblait être régie par la présence d'une communauté biologique active au sein du floc. La principale influence de cette communauté est la production bactérienne de substances polymériques extracellulaires (SPE) de nature fibrillaire qui donnent sa structure au floc. Cette structure en évolution constante (en raison de l'activité physique, chimique et biologique) influe considérablement sur le transport du floc en modifiant sa taille, sa stabilité, sa densité (par le piégeage de l'eau), sa porosité et ses processus biochimiques. Une corrélation linéaire positive entre la taille du floc et la vitesse de précipitation a été établie. Les différences observées entre les milieux ont été attribuées à la diversité compositionnelle et structurale du floc. Généralement, les flocs précipitent avec leur axe le plus long parallèle à la direction de la précipitation dans un milieu à faible hydrodynamisme. La densité de tous les flocs était particulièrement faible (presque égale à celle de l'eau) en raison de la forte porosité et de la teneur élevée en matière organique. L'étude fait ressortir que les flocs ont été indûment caractérisés comme universellement instables à cause des changements observés dans les distributions du volume, lesquelles sont biaisées par la rupture de quelques grosses particules. Le modèle conceptuel présenté établit un lien entre la force des flocs et le transport des sédiments (dépôt et érosion) dans la colonne d'eau et dans l'interface eau-sédiments. Il est important d'observer et d'analyser les sédiments en suspension dans leur forme floculée naturelle, car les particules des flocs se comportent différemment (transport/précipitation) des particules sphériques primaires et théoriquement solides.

Sommaire des recherches de l'INRE

Titre en langage clair

La structure interne et externe de la structure des particules composites influe sur le transport des sédiments et des polluants.

Quel est le problème et que savent les chercheurs à ce sujet?

Les particules composites floculées sont très répandues dans les milieux aquatiques. Un grand nombre d'études expliquent comment le cisaillement influe sur la formation et le transport des flocs. Toutefois, nous comprenons mal en quoi la structure et la composition des particules composites influent sur le transport des sédiments et des contaminants associés. Dans le présent document, nous présentons quelques constats en nous fondant sur des techniques d'analyse par imagerie optique appliquées à des flocs prélevés dans deux milieux distincts (cours d'eau/trop-pleins d'égouts unitaires).

Pourquoi l'INRE a-t-il effectué cette étude?

Les travaux sont menés dans le cadre d'une étude visant à améliorer nos capacités de prédire le transport, l'érosion et le dépôt des sédiments contaminés. Les résultats de l'étude serviront à améliorer les modèles numériques qui, à leur tour, aideront à définir et à évaluer les mesures correctrices à mettre en œuvre pour améliorer la gestion de nos ressources en eau.

Quels sont les résultats?
Les résultats permettent de penser que la communauté biologique (principalement les bactéries) est la composante dominante qui régit les propriétés structurales internes et externes des flocs, quel que soit le milieu. La communauté bactérienne influe sur la formation des flocs par sa nature collante (notamment par la sécrétion de substances polymériques de nature fibrillaire) qui favorise l'adhérence, mais aussi par son influence sur la densité des particules, la porosité et la force des flocs. Tous ces facteurs influent sur la manière dont les flocs et les contaminants qui leur sont associés sont transportés au sein du milieu aquatique.

Comment ces résultats seront-ils utilisés?
Les résultats nous aideront à mieux comprendre le mouvement des particules et des contaminants en milieu aquatique et à améliorer les modèles numériques permettant de prédire ce mouvement.

Quels étaient nos principaux partenaires dans cette étude?
L'étude a été réalisée conjointement par l'Université McMaster, l'Université Ryerson et l'Université d'Exeter (Royaume-Uni).

Structural Controls on Floc Strength and Transport

Ian G. Droppo

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Abstract: This paper examines how the external and internal structure of a floc will influence its strength and transport within diverse aquatic environments. The paper uses examples from riverine and urban drainage systems to correlate floc structure to floc strength and transport. The internal matrix of the floc was observed to be mediated by an active biological community within the floc. The primary influence of this community is the bacterial production of fibrilar extracellular polymeric substances (EPS) that acts as a framework of the floc. This continually changing framework (due to floc physical, chemical and biological activity) has extending influences over floc transport by modifying floc size, stability, density (through the trapping of water), porosity and biochemical processes. A positive linear relationship of floc size to settling velocity was found, with differences between environments attributed to floc compositional and structural diversity. Generally, flocs settle with their long axes parallel to the direction of quiescent settling. The density of all flocs was characteristically low (approaching that of water) due to high porosity and organic content. The paper suggests that flocs have been unduly characterized as universally unstable due to observed shifts in volume distributions that are biased by the breakup of a few large particles. A conceptual model is provided which links floc strength to sediment transport (deposition and erosion) in the water column and at the sediment water interface. It is important that suspended sediment

be observed and analyzed in its natural flocculated form, as floc particles behave differently (transport/settling) from primary and theoretical solid spherical particles.

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Key words: Flocculation, bacteria, EPS, stability, transport, structure

INTRODUCTION

Flocculated particles are ubiquitous in aquatic environments, with flocculation now accepted as an important process in the transport of sediments and associated contaminants. The transport of cohesive sediment is dictated in part by the structure and settling velocity of the sediment. Flocculation significantly alters the transport characteristics of sediment in suspension by modifying the effective grain size, shape, density, porosity and water content (Droppo and Ongley, 1994; Droppo *et al.*, 1997; Phillips and Walling, 1995; Petticrew, 1996). Modelling of flocculation and particle transport is required to better understanding and predict sediment related issues focused around the source, fate and effect of sediments and associated contaminants. While the effect of shear on floc formation has been studied and modelled ostensibly around collision theory (e.g. Lee *et al.*, 2000; Winterwerp, 1998; Lick, *et al.*, 1993; van Leussen, 1988; Tsai *et al.*, 1987; Partheniades, 1986), model predictions would benefit from a better understanding of how floc structure (internal and external) influences floc transport. Milligan and Hill (1998) have shown that although turbulence plays a role in floc formation and size, it cannot be considered in isolation when predicting floc transport.

Because flocs are not solid homogeneous structures, but rather matrices of both organic and inorganic particles linked together by various physical, chemical and/or biological means, they are generally presumed inherently unstable and prone to break-up during sampling and analysis (Fennessy *et al.*, 1994, van Leussen and Cornelisse, 1993). While bottom shear stresses may be enough to break up flocs during the process of floc recycling (Partheniades, 1986; Droppo *et al.*, 2001), it is questionable if gentle sampling will modify floc size significantly. This is suggested primarily because the classical percent by volume distribution, often used to assess floc stability, is strongly biased by the breakup of a few large particles (Droppo, *et al.*, 1998). The biological community of flocs and their production of extracellular polymeric substances (EPS) are often believed to be a dominant controlling factor of floc stability in natural systems (Liss *et al.*, 1996; Dade, 1996; Droppo *et al.*, 1997). High resolution techniques such as transmission electron microscopy (TEM) and scanning confocal laser microscopy (SCLM) have revealed a presence of EPS fibrils which bridges and binds primary particles (organic and inorganic) together within the floc matrix (Liss *et al.*, 1996). These colloidal particles give the floc a pseudo plastic nature (Droppo and Ongley, 1992). It is believed that this internal structure (consisting of inorganic particles, organic particles and water) exerts significant influence over the chemical, physicochemical and biological processes operating within the floc (Liss *et al.*, 1996; Heissenberger *et al.*, 1996; Leppard, 1995, 1993, 1985; Decho, 1990). The significance of the internal floc structure on the outward gross behaviour of the floc is, however, still poorly understood.

To evaluate the structure (internal and external) of freshwater flocs and its potential impact on floc transport, correlative microscopy (Liss *et al.*, 1996; Leppard,

1992) is used as a tool to view the floc over a large range of resolutions. Correlative microscopy is a strategy that uses different microscope types, and their accessory techniques, in a multi-method context to derive multiple levels of information from a given specimen. By observing flocculated material over a full range of magnifications (>1.0 mm to sub μm) in conjunction with settling experiments, it is the purpose of this paper to provide a better understanding of how floc structure in both its gross and fine scale can affect floc transport and strength. This will help improve our knowledge and models of natural sediment dynamics and the associated contaminant source, fate and effect relationships.

MATERIALS AND METHODS

Study site

Sample sites were chosen to provide information on floc structure and behaviour in general and not for the characterization or spatial comparison of specific sites and their floc structure. Floc samples were collected from 16-Mile Creek, Milton, Ontario, from 14-Mile Creek, Oakville, Ontario and from a combined sewer overflow (CSO) entering Hamilton Harbour, Ontario. The 16-Mile Creek has a drainage basin of approximately 276 km^2. The sample site was located 20 km upstream of Lake Ontario and receives only surface and subsurface flow from forested and low density agricultural land. The site is more fully described in Ongley (1974). Fourteen Mile Creek has a drainage area of approximately 30 km^2. The sample site was located 5 km upstream of Lake Ontario and drains primarily residential and low density agricultural land. More detailed information on the 14-Mile Creek may be found in Ongley (1974). CSO samples were collected from the sewer outfall servicing the Kenilworth sewershed that has a contributing area of 265.5 ha. The sewershed is made up of a variety of pervious and impervious surfaces with residential, commercial and industrial land uses. It is estimated that the total overflow volume from the sewershed for a typical rainfall year (considered to be 1986) is 311,000 m^3 with a total sediment yield of greater than 60 tonnes of sediment (Irvine *et al.*, 1998). The sewershed and collection site is fully described in Droppo *et al.* (2002).

Conventional Optical Microscopy for Gross Structural Determination

Samples were collected following the method of Droppo and Ongley (1992). This method allows for the non-destructive direct sampling and observation/measurement of flocculated material within a settling column (plankton chamber). The flocs are imaged (sized) down to a lower resolution of approximately 2 μm (10x objective) using a Zeiss Axiovert 100 microscope interfaced with an image analysis system (Northern Exposure™ - Empix Imaging, Inc.). Flocs were characterized with both a percent by number and volume distribution.

Scanning Confocal Laser Microscopy for Internal Floc Matrix Determination

Samples stabilized in agarose (Droppo *et al.*, 1996) were stained with a solution of wheat germ agglutinin conjugated to Texas Red™ (WGA-TR, Molecular Probes, Inc., USA). This molecular probe is specific to many of the specific polysaccharides present in the fibrillar material (N-acetyl-D-glucosamine). The agarose stabilized flocs were then washed three times with 0.1 M phosphate buffer (pH 7.0). The flocs were then imaged using a Zeiss Micro System LSM (Model LSM 10 BioMed). The SCLM was equipped with an argon laser (emission lines at 418 and 514 nm) and a 63x (1.4 na) objective with an electronic zoom of 30X. Image slices were collected at 1 μm vertical intervals and reassembled to view the whole floc volume specific to the stained component.

Transmission Electron Microscopy for Ultrastructure Determination

More detailed ultrastructural observations of sediment samples were made by preparing the samples for TEM following a four-fold multi-preparatory technique (Liss *et al.*, 1996). This technique allows for the enhanced observation of specific components of the floc such as cells and polymeric material. Ultrathin sections were imaged in transmission mode (TEM) at an accelerated voltage of 80 kV using a JEOL 1200 Ex II TEMSCAN scanning transmission electron microscope.

Particle settling velocity, settling orientation, density and porosity determination

Settling experiments were performed following the methods of Droppo *et al.* (1997) that provide settling velocity for individual flocs. A drop of sediment collected with a wide mouth pipette (3.74 mm) from a gently homogenized sample bottle was introduced into an insulated 2.5 L capacity settling column. Settling flocs pass through the field of view of the stereoscopic microscope where they are video taped on a SVHS VCR through a CCD camera interface. Using Northern Exposure™, the settling velocity was derived by digitally overlaying two video frames containing identified flocs separated by a known time interval. Particle size and settling orientation are also attained from these images.

The density of a floc [expressed as excess density (1-wet floc density)] was estimated using Stokes' Law. As Stokes' Law is based on the settling of single impermeable spherical particles in a laminar region (Reynolds Number < 0.5), it is not ideal for the determination of floc density due to the heterogeneous structure and irregular shape of flocs (Hawley, 1982). Nevertheless Stokes' Law or a modification thereof has often been used to determine the wet density of singular flocs (Li and Ganczarczyk, 1987; Droppo *et al.*, 1997), and does provide an indication of how floc settling velocity, density, and porosity are related to aggregate size. The floc porosity can be expressed by a mass balance equation (Equation 1) assuming a typical density of dried silt and clay of 1.65 gm/cm^3.

$$\varepsilon=(\rho_s-\rho_f)/(\rho_s-\rho_w) \qquad (1)$$

Where ε = floc porosity, ρ_s = density of the dried solid material, ρ_f = wet density of the floc and ρ_w = density of the water] (Li and Ganczarczyk, 1987).

RESULTS AND DISCUSSON

It is well known that cohesive suspended sediment in any aquatic environment is preferentially transported as flocculated particles (Walling and Woodward, 1993; Milligan and Hill, 1998; Petticrew and Droppo, 2000; Droppo, 2001; Clifford, 2002; Krishnappan and Marsalek, 2002). The most significant impact of flocculation in terms of sediment and contaminant transport is that it alters the downward flux of sediment by changing its hydrodynamic properties (Partheniades, 1986; Li and Ganczarczyk, 1987; Ongley *et al.* 1992; Krishnappan, 2000). This is brought about by flocculation increasing the effective particle size by orders of magnitude over the absolute particle sizes and, as such, also changes the effective particle shape, density, porosity and composition of the characteristic particle (suspended or bed sediment) within a system (Li and Ganczarczyk, 1987; Nicholas and Walling, 1996; Droppo *et al.*, 1997, 2000; Phillips and Walling, 1999).

The significance of flocculation on the effective grain size distribution can be seen in Figure 1 and 2. Figure 1 shows the natural floc particles (a) and the absolute primary particles following sonication (b), while Figure 2 demonstrates the shift in distributions following deflocculation (sonication). The dispersed inorganic particles in Figure 1b and the absolute distribution (by volume) in Figure 2 are close to what traditional sediment sizing techniques (e.g. Sedigraph and Coulter Counter) would measure for grain size distributions (i.e. the classical chemically and physically dispersed inorganic grain distribution). As flocculated particles have significantly different transport characteristics compared to absolute primary particles (Krishnappan, 1990; Ongley *et al.*, 1992; Droppo *et al.*, 1997), the use of such a disaggregated distribution to characterize sediment for sediment and contaminant transport models would result in erroneous predictions. This is demonstrated by Ongley *et al.* (1992), who found significant differences in sediment flux estimates when modelling sediment transport with and without accounting for flocculation. They incorporated the flocculation model of Krishnappan (1990) and the dispersion model of Krishnappan and Lau (1982) in order to predict the sediment flux. The model showed that when flocculation was not taken into account, essentially there was minimal change in sediment flux, whereas, when flocculation was accounted for, virtually all of the sediment was deposited in a relatively short distance. While their model was overly simplistic (only flocculation and not floc breakage was assumed and only sedimentation occured and not resuspension), the effect of flocculation on sediment transport and transport models is obviously dramatic. More recent modeling efforts (e.g.

Perigault *et al.*, 2000; Krishnappan 2000; Krishnappan and Marsalek, 2002; Winterwerp, 1998 and 2002) are refining flocculation models for the prediction of sediment and contaminant transport/erosion in aquatic environments.

Floc Stability

Floc strength is a critical characteristic that will dictate its transport history within aquatic systems. A difficulty in characterizing flocculated particles is the maintenance of particle integrity during sampling and sizing. Evaluation of floc stability is difficult due to the current inability to assess individual flocs for strength and therefore the need to assess whole floc population response to stress. Generally such assessments are done in the laboratory, where often, unrealistic shears are used (Milligan and Hill, 1998) or in the field using sizing instruments with classical percent by volume distribution outputs (e.g. laser particle sizer, Bale and Morris, 1987).

Flocs are often assumed to be unstable and prone to breakup during sampling and analysis. This was the conclusion of Bale and Morris (1987), by observing the shifts in volume distributions following measurements made with a laser particle sizer 1) *in-situ*, 2) after sampling in a sample bottle and 3) following sonication. Volume distributions can be misleading however, as they are strongly influenced by the breakup of a few large flocs. This is shown in Figure 2 where a significant shift in the volume distribution (significant difference at α= 0.5, Modified Kolmogorov-Smirnov test) has occurred which is due primarily to the break up of 6 large flocs representing 90% of the volume of the total sample. While the extreme case is presented in Figure 2, it is worth noting the differences in the effect of sonication on the two distribution types. While there is a significant shift in the volume distribution, there is no significant difference in the percent by number distribution (no significant difference at $\alpha = 0.5$, Modified Kolmogorov-Smirnov test) even though the total particle counts increased significantly. The above observation, which is typical of most distributions examined, has significant implications as to the presumed stability of flocculated particles. It is evident that to characterize all flocs as being unstable, based on volume distribution shifts, may be erroneous as the break-up of a few large flocs via various sampling methods can change the volume distribution substantially. As such, it is more likely that the volume distribution is "unstable" than the majority of flocs themselves. It is therefore important that both the percent by number and by volume distributions be evaluated in order to better understand and model the transport of sediment and contaminants. Which distribution is considered of primary importance will depend on the nature of the research objective.

From the above extreme case of sonicating flocs, it is assumed that when using gentle sampling methods (such as the plankton chamber method used in this study), flocs are in fact relatively stable (although some breakage of large flocs is possible). This is supported by Phillips and Walling (1995) who demonstrated that sampling riverine suspended sediment in a bottle does not significantly affect the floc size distribution (by number and volume) provided that size measurements are made immediately after sampling.

The effect of shear on floc stability has been studied extensively (e.g. Boller and Blaser, 1998; Alldredge, *et al.*, 1990; Bache and Al-Ani, 1988; Hannah *et al.*, 1967), however, a poor understanding of compositional effects exit. Walker and Bob (2001) have shown that for a consistent shear, the stability of aggregates depends on the type of organic matter present (polysaccharide and humic acid were studied) as well as floc size. They found that under no circumstance did the addition of either polymer result in deflocculation due to strong interparticle forces and that humic acids increased stability more than polysaccharides for large flocs. These are of course concentration dependent (Muhle, 1993). Floc stability in natural systems is largely controlled by the floc biological community's production of extracellular polymeric substances (EPS) (Liss *et al.*, 1996; Muschenheim *et al.*, 1989). The EPS fibrils serve as a means of attachment and nutrient assimilation for the many cells that colonize the flocculated material (Costerton *et al.*, 1987). This material is made up of numerous components (e.g. DNA, carbohydrates, proteins, uronic acids) that comprise a sticky material that "glues" the inorganic and organic components of the floc together. This material gives the floc a pseudo plastic nature and hence a believed relatively stable structure (Droppo and Ongley, 1992). Within bed sediments, EPS has been shown to be a primary mechanism influencing bed sediment stability (Lau and Droppo, 2000; Dade *et. al.*, 1996). While difficult to quantify individual floc shear strength, it is evident that the EPS is an important component contributing to floc stability and essentially represents the framework of the floc structure (Leppard, 1997; Droppo, 2001). Figure 3 shows the typical interconnections of EPS fibrils with the inorganic clay particles forming a three dimensional matrix with high internal surface area. Figure 4 illustrates the EPS distribution and density within a floc. Figure 4a illustrates the EPS distribution within an individual slice through the X-Y plane of a riverine floc, while Figure 4b illustrates a slice through a stack of 52 images (1.0 μm intervals) in the Z direction of the same floc. The decreasing density gradient of EPS from the core of the floc to its edges (Figure 4b) is suggestive of a very stable floc core. This also gives some indication of floc building mechanisms and transformations as this density gradient will also be synonymous with age, compaction, and diffusion and advection potential. While not evaluated here, the surface tension between fibrils is likely very high due to their small size (fibrils diameter in Figure 3 are in the range of 4-6 nm) resulting in further stabilization (Figure 3). This will also result in the impediment to flow and the trapping of water within the floc (bound water) with concomitant effects on effective density and settling. This trapped water may have a significant effect on the chemical characteristics (Lee, 1994) and processing behaviour of the floc (similar to biofilms) in terms of contaminant transformation and electrochemical and diffusion gradients of such factors as redox potential and pH (Liss *et al.*, 1996; Costerton *et al.*, 1987; Karl, 1982). In addition, the extensive networks of fibrils support an enormous surface area for contaminant and nutrient adsorption and subsequent transport. As such, the biological community of the floc is a key component that influences the physical and biogeochemical characteristics of the floc. Work in laboratory flumes has rarely been able to document the stabilizing nature of EPS due to the relatively short time scale of experiments (i.e. minimal time for EPS development and integration). Generally, only samples collected in the field or wastewater reactor experiments have shown the floc stabilizing nature of EPS (Santschi *et al.*, 1998; Leppard, 1995, 1997). Further, work by Liao *et al* (2002) have shown that van der Waals and/or hydrophobic interactions are primarily important at the development phase of bioflocculation by overcoming the repulsive interactions and bringing particles close enough to form specific interactions. Once initiated the microbial binding (floc building/stabilizing) takes over in conjunction with further strengthening with ionic interactions and hydrogen bonding.

The Influence of Floc Structure on Floc Settling/Transport

Assuming a stable floc, settling experiments within a quiescent settling chamber have consistently shown that as floc size increases, settling velocity increases in a linear fashion regardless of the environment (Figure 5). This is counter to the Stokes' equation that states that settling is proportional to the square of the particle diameter. This difference is related to the floc structure being different from the solid spherical unit assumed in the Stokes' equation. As seen in Figure 1a, flocs deviate strongly from this assumption by being heterogeneous irregular shaped porous particles. It is also this morphological (shape, porosity) and compositional (organic/inorganic composition and water content) irregularity between flocs of similar sizes that give rise to a general high variability in the measurement of floc size to settling velocity (Figure 5). (The data in Figure 5 have statistically significant regression lines ($p=0.05$), however, the r^2 is characteristically low). Flocs will also settle orders of magnitude slower than those predicted by Stokes' equation with equivalent diameters. This too is related to floc morphology being very different from the solid spherical particles assumed by Stokes' Law and because of significant density differences between the two particle types (Nicholas and Walling, 1996) [solid spherical particle ~ 2.65 g cm^3; floc ~ 1.001 to 1.03 g cm^3 (Figure 6)].

The different settling characteristics in Figure 5 are related to the compositional differences in the floc structure for the two different environments. The CSO flocs possess a much higher organic content and therefore a lower density than the riverine flocs (Figure 6). The CSO flocs also possess a higher porosity that will increase their water content and promote a lower density (Figure 7). As a result, settling velocities for equivalent size flocs are lower for the CSO (Figure 5). The higher porosity and lower density of flocs compared to the riverine flocs may also suggest that the transport of flocs in the sewers is less turbulent resulting in a more open matrix floc as compared to the river where flocs are more compact. The above observations that as floc size increases, density decreases (approaching the density of water) and porosity increases are consistent with observations made from a variety of environments (Klimpel and Hogg, 1986; Li and Ganczarczyk, 1987; Logan and Hunt, 1988; Boller and Blaser, 1998; Droppo *et al.*, 2000). Floc density and porosity and their influence on transport are largely controlled by the biological community of the floc as discussed above and conceptually modelled by Droppo (2001).

The shape of a floc is also known to affect settling due to resistance effects against flow (fluid drag forces) (Li and Ganczarczyk, 1987). The shape of a floc is generally influenced by its origin/source and composition and by the flow field in which it is transported. Models will often use a characteristic floc shape(s) or fractal mathematics to account for the influence of shape on floc transport (e.g. Lee *et al.*, 2000; Winterwerp, 1998; Kranenburg, 1994). Flocs in the quiescent settling column experiments were found to generally settle with their long axes parallel or close to the direction of settling. However, very poor correlation was found between floc shape and settling velocity and any weak trends observed tended to contradict traditional theory [i.e. in quiescent environments, spherical flocs generally settled faster than cylindrical or disk shape flocs with similar mass and density (Li and Ganczarczyk; 1987)]. Given the weak non traditional trends observed and the fact that most flocs are being transported in a turbulent environment (floc will be tumbling during transport), the overall shape of a floc is likely irrelevant for settling with regards to orientation and less important than floc size or density in influencing settling velocity. Its influence will be its ostensible effect on drag in a flowing medium that is difficult to quantify given the structural complexity of flocs.

Conceptualizing Floc Transport in a Riverine Environment

Given that flocs settle out of suspension faster than smaller primary particles, one may expect a reduction in river turbidity if there were no active sources of sediment input. This is, however, generally not the case due to the mechanism of floc recycling. Floc recycling is the process whereby larger, less dense flocs are formed within the water column (zone of lower shear) and settle towards the bottom of the river. Once the flocs reach the high shear zone of the sediment water interface they may break up and be lifted back into suspension as primary particles or smaller flocs (Partheniades, 1986). At this point these particles may once again go through the floc building and break up cycle.

If the critical shear stress for floc break up is larger than the bed shear stress then the flocculated particles may settle on the bed (Partheniades, 1986) forming a surficial fine-grained laminae (SFGL) (Droppo and Stone, 1994). SFGL is characterized as a high water content, "fluffy", "buoyant" layer with substantial inter-particle/inter-floc spaces/pores with a density of approximately 1.1 g cm^{-1} (Droppo and Stone, 1994). Because of these structural characteristics, the layer is easily eroded (prior to significant biostabilization – Droppo *et al*, 2001) by the next storm event that increases the bed shear stress beyond the critical shear stress for erosion. As such, SFGL is a transient depositional feature that is highly related to flocculation within the water column, critical bed shear stress and floc strength. The mechanism of floc deposition and erosion has been described by Droppo *et al.* (2001) and Droppo & Amos (2001) to be a combination of the cyclic linkage between floc recycling and surficial fine grained lamina (SFGL) recycling (Figure 8). As discussed earlier, floc strength is a critical characteristic that will dictate its transport history within a riverine system.

CONCLUSION

Flocs are ubiquitous in all aquatic environments, including urban drainage systems, which transport cohesive sediment. Their external and internal structure is strongly mediated by an active biological component that influences overall floc stability and transport. Flocs behave hydrodynamically differently from individual grain particles due to differences in their effective size, shape, porosity, water content and density. A positive linear relationship was observed between floc size and settling velocity for two diverse environments (river and CSO), with differences in the regression coefficients attributed to compositional and structural diversity. Although flocs were observed to have densities as low as water due to high porosity and organic content, they settle relatively fast compared to their constituent primary particles due to their larger size. Flocs generally settle with their long axes parallel to the direction of quiescent settling, however, the importance of this factor in turbulent flows is poorly understood. The biological community's production of fibrilar EPS acts as a framework of the floc with concomitant effects on stability, density (through the trapping of water), porosity and biochemical processes. This paper suggests that flocs have been unduly characterized as universally unstable due to observed shifts in volume distributions that are biased by the breakup of a few large particles. Further work is required to quantify natural floc stability in order to better understand the transport dynamics of sediments within a variety of environments. The linkage of floc strength to sediment transport (deposition and erosion) is provided in a conceptual model which links the cycling of flocs between the water column and the sediment water interface. As flocculated sediment behaves (transports/settles) differently from the traditionally sized and modelled disaggregated particles, it is critical that predictive models for sediment and contaminant transport take account of the phenomenon of flocculation.

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FIGURE CAPTIONS

Fig 1. Example micrographs showing 16-Mile Creek sediment in a) natural flocculated form and b) disaggregated inorganic constituent particles.

Fig. 2. Example natural and disaggregated distributions by volume and number for 16-Mile Creek.

Fig. 3. Example TEM micrograph of floc illustrating the complex internal matrix of EPS fibrils and inorganic particles.

Fig. 4. Example SCLM micrographs illustrating the distribution of EPS material within a 14-Mile Creek floc a) across a single slice (X-Y plain) and b) through 52 images (slices) in the Z plain.

Fig. 5. Comparison of settling velocity to floc size for a riverine and CSO sample.

Fig. 6. Comparison of excess density (density of the floc minus the density of the water) to floc size for a riverine and CSO sample.

Fig. 7. Comparison of porosity to floc size for a riverine and CSO sample.

Fig. 8. Conceptual model of SFGL and floc recycling (reproduced with permission from Droppo *et al.*, 2001).

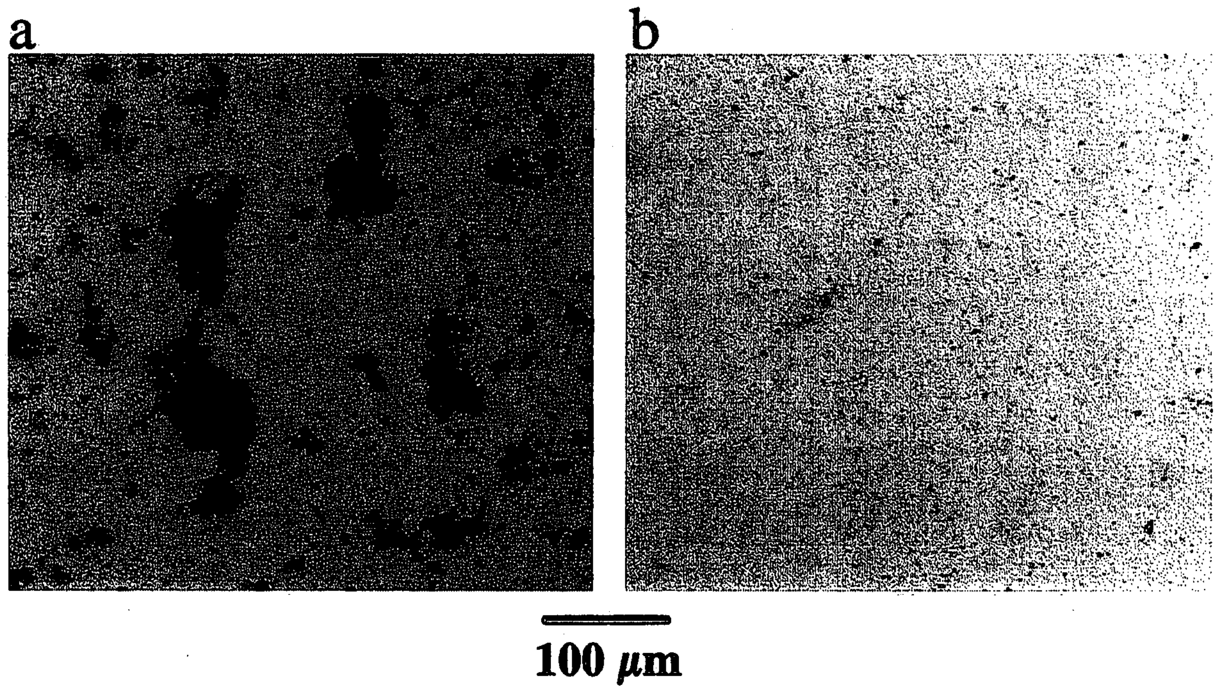

a
b
100 μm

Fig.1

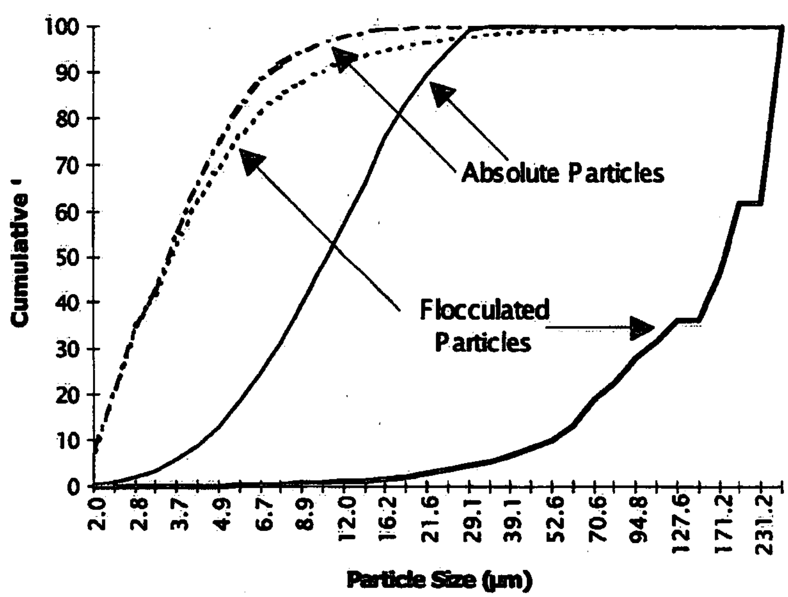

Cumulative
Particle Size (μm)
Absolute Particles
Flocculated Particles
100
90
80
70
60
50
40
30
20
10
0
2.0
2.8
3.7
4.9
6.7
8.9
12.0
16.2
21.6
29.1
39.1
52.6
70.6
94.8
127.6
171.2
231.2

Fig. 2

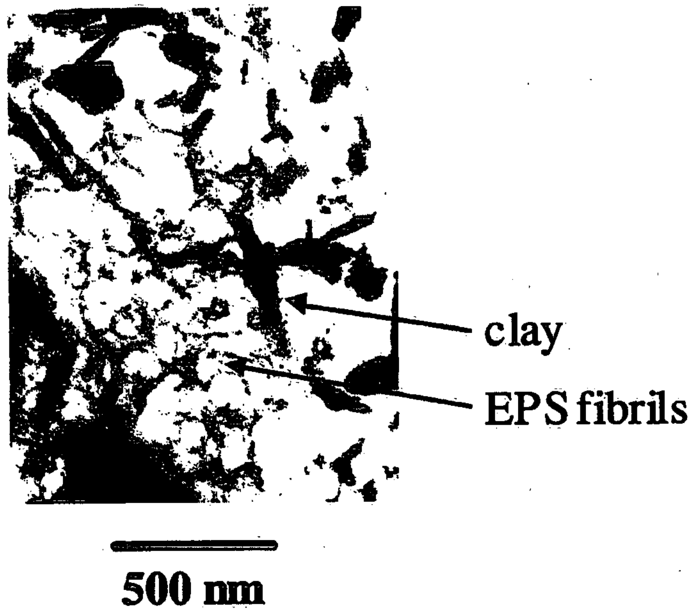

clay
EPS fibrils
500 nm

Fig. 3

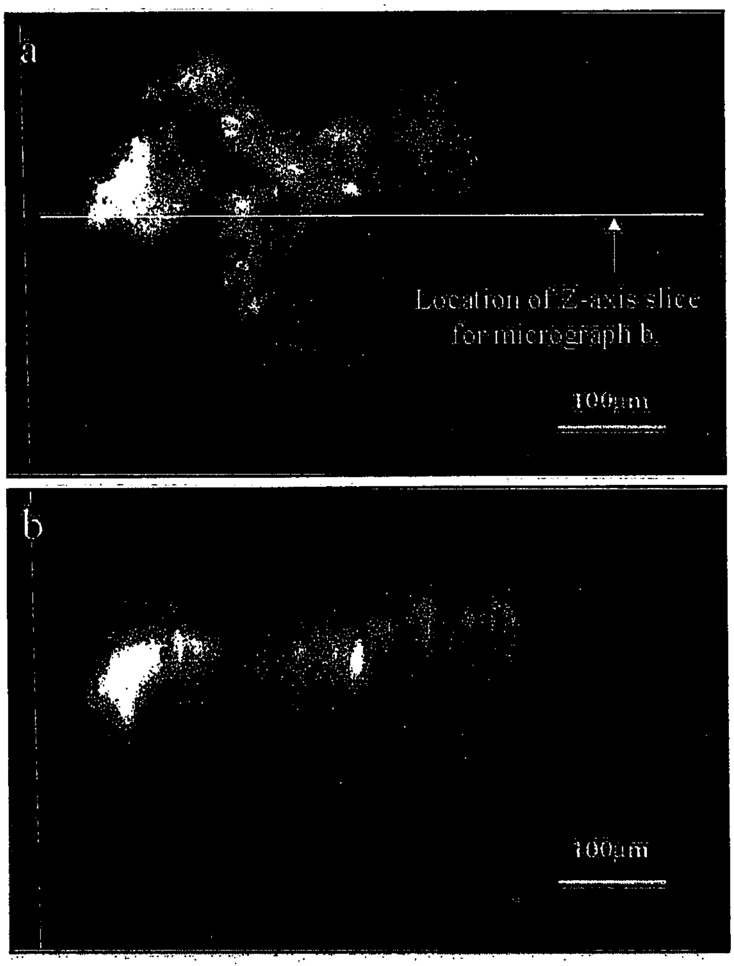
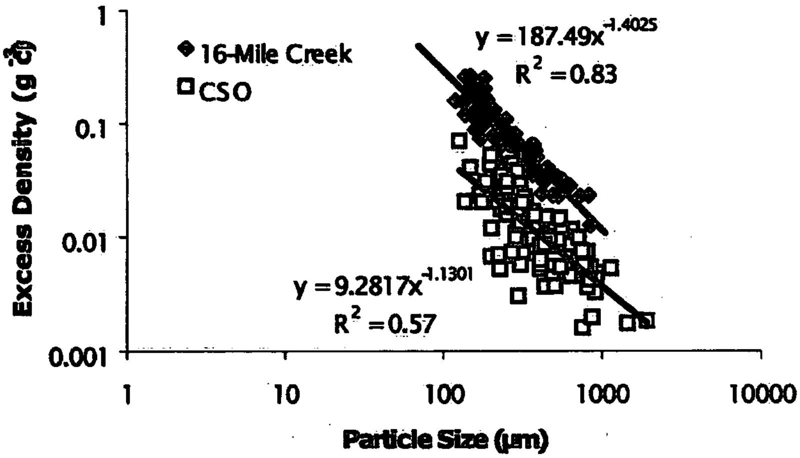
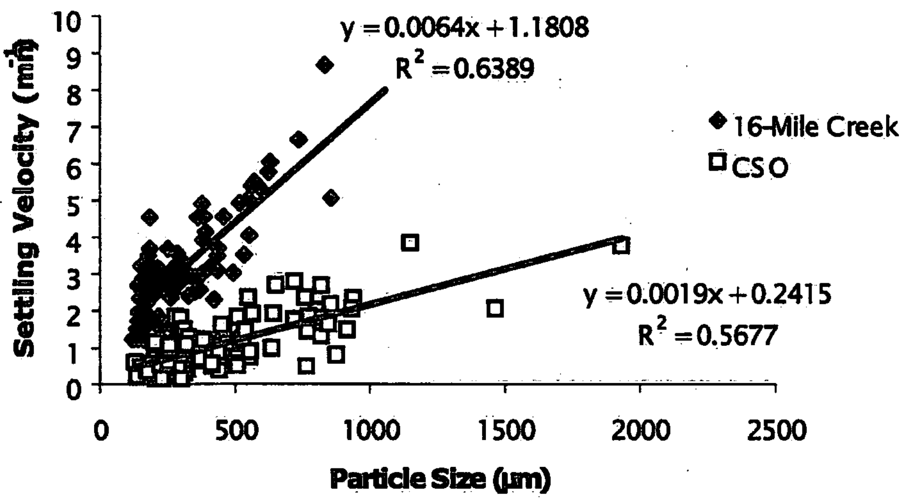


Fig. 4



1
0.1
0.01
0.001
Excess Density (g cm⁻³)
16-Mile Creek
CSO
y = 187.49x^-1.4025
R^2 = 0.83
y = 9.2817x^-1.1301
R^2 = 0.57
1
10
100
1000
10000
Particle Size (μm)

Fig. 5



10
9
8
7
6
5
4
3
2
1
0
Settling Velocity (mm s⁻¹)
y = 0.0064x + 1.1808
R^2 = 0.6389
16-Mile Creek
CSO
y = 0.0019x + 0.2415
R^2 = 0.5677
0
500
1000
1500
2000
2500
Particle Size (μm)

Fig. 6

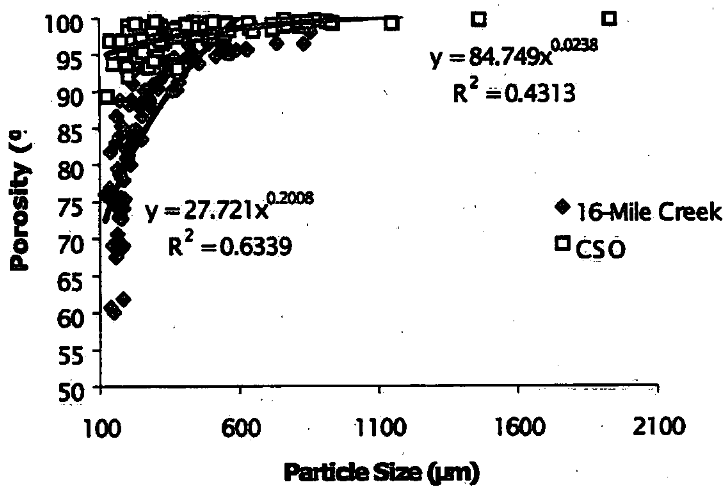

y = 84.749x^0.0238
R² = 0.4313
y = 27.721x^0.2008
R² = 0.6339
16-Mile Creek
CSO
Porosity (%
Particle Size (µm)
100
95
90
85
80
75
70
65
60
55
50
100
600
1100
1600
2100

Fig. 7.

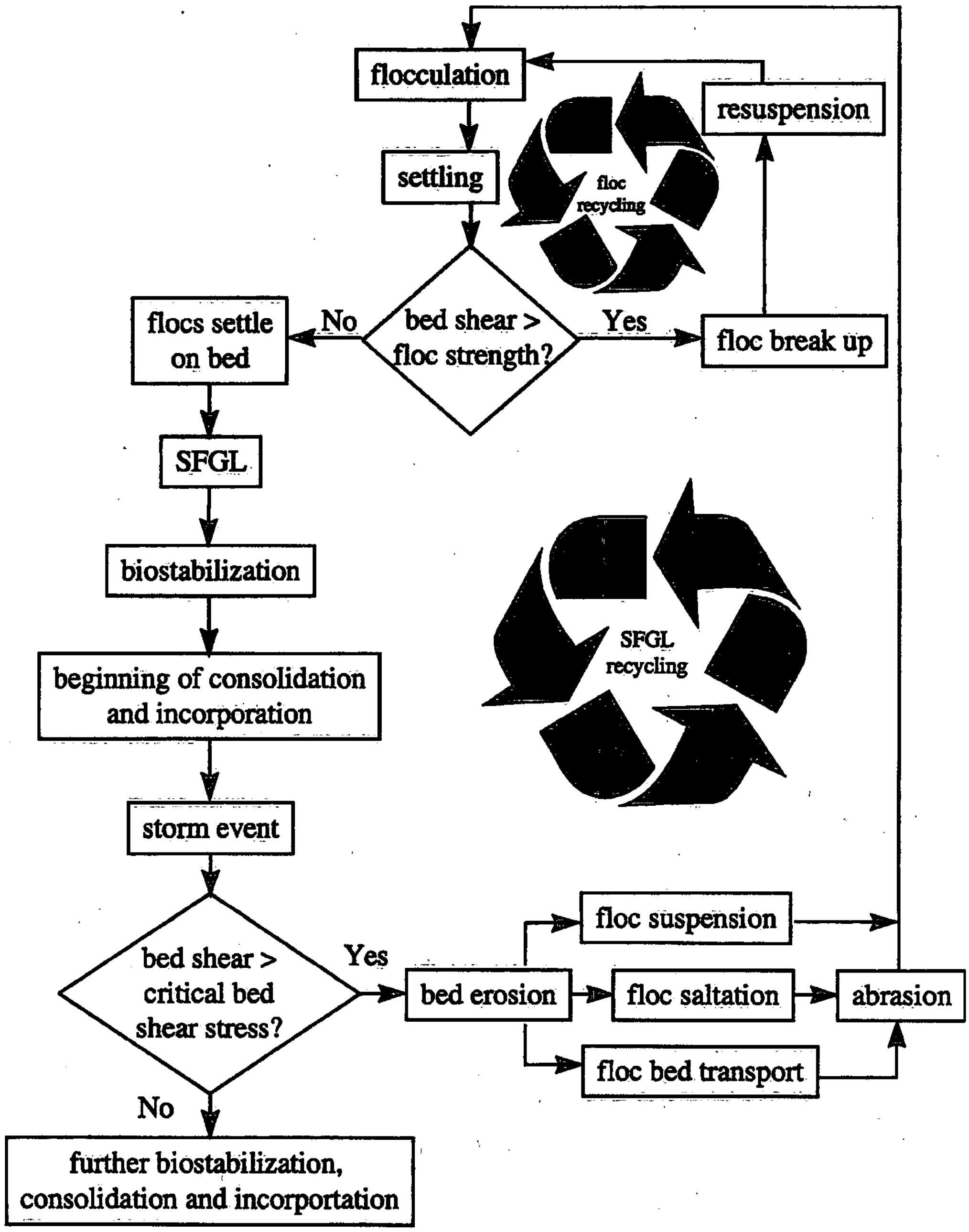

flocculation
resuspension
settling
floc recycling
flocs settle on bed
No
bed shear > floc strength?
Yes
floc break up
SFGL
biostabilization
SFGL recycling
beginning of consolidation and incorporation
storm event
floc suspension
bed shear > critical bed shear stress?
Yes
bed erosion
floc saltation
abrasion
floc bed transport
No
further biostabilization, consolidation and incorportation

Fig. 8

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