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Review of the action of organic matter on mineral sediment flocculation

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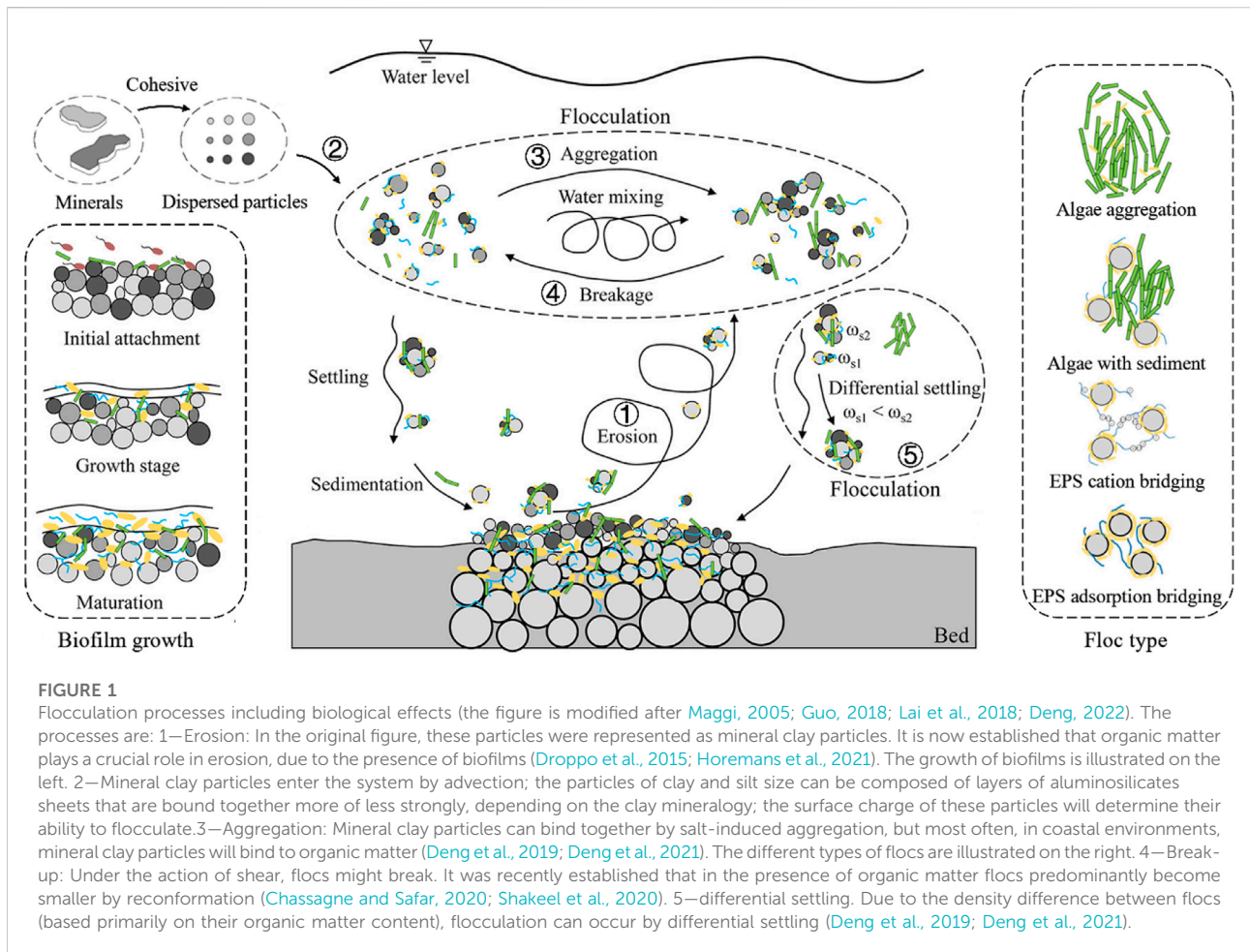
Sediment is found throughout the world's alluvial plain rivers, estuarine coasts and adjacent seas and is thereby a key factor in major ecosystems. Suspended mineral sediment can affect the biological activity of microorganisms and plants, by reducing light penetration in the water column or by binding to organic matter. Biological processes can, in turn, affect the physical and chemical properties of the sediment particles and influence the adhesion between particles. They can facilitate the sediment aggregation (flocculation) through bridging, patching and sweep, while biological decay will mainly help to disintegrate organic matter rich flocs. Biological activity also affects the properties of flocs (structure, density, sedimentation rate and composition). This activity is itself influenced by environmental conditions (like temperature, light and nutrient fluxes). Sediment flocculation thus involves complex relationships between several physical, chemical and biological factors. The role of biology in particular needs to be better integrated in sediment transport models, through the interaction between mineral clay particles, microorganisms and their excreted polymers (Extra Polymeric Substances, i.e., EPS). In this article, a summary of the state-of-the-art research regarding sediment flocculation is given. In particular, the action of organic matter on fine-grained sediment flocculation is discussed. The aim of the article is to provide a more comprehensive understanding of bio-sediment dynamics and give an outlook on remaining research questions.

KEYWORDS

sediment flocculation, bioflocculation, algae, extracellular polymeric substances (EPS), aggregation

1 Introduction

Sediment is fundamental to the evolution of natural aquatic environments such as rivers, estuaries and lakes (van Leussen, 1988; Eisma and Irion, 1993; van Leussen, 1994; Manning et al., 2010b). Suspended sediment dynamics play an important role in geomorphological evolution, biogeochemical cycling, transport of pollutants, and aquatic biological activity (McAnally and Mehta, 2001). Cohesive sediment refers to a mixture of clay- and silt-size particles with a small amount of fine sand-size particles. This



cohesive sediment is composed of mineral clay, organic matter and water in various amounts. Even though the proportion of clay-size particles and organic matter is only 5%–10% in most cohesive sediment, their presence results in remarkable rheological properties (yield stress, viscosity) of the overall sediment and can lead, in the water column, to a process called flocculation (the aggregation of mineral clay particles with themselves and/or with organic matter) through electrochemical and biochemical interactions (Dyer, 1989; Winterwerp and Kesteren, 2004; Chassagne, 2019; Chassagne et al., 2021).

Over 90% of the total volume of fine suspended sediment in an aquatic environment exists in the form of aggregates, i.e., flocs (Droppo and Ongley, 1994). Figure 1 is adapted from a classical picture illustrating the behavior of cohesive sediment in aquatic systems. It was adapted so as to show the effects of some organic parameters (EPS, short for Extra Polymeric Substances, and living microorganisms such as microalgae) on suspension, flocculation/break-up, settling and deposition. Mineral clay particles come in the water column either by hydrodynamic transport or resuspension by erosion from the bed (labels 1 and

2 in Figure 1). Under the action of microscopic forces which are linked to the particles' surface charge (van Leussen, 1988; Gregory, 2005), particles will flocculate, and form flocs, which can be large in volume and size. Flocs are usually large when they are made of a high content of organic matter, as organic matter has a low density and therefore relatively large organic matter particles can be found in the whole water column and thus aggregate with mineral sediment. To give an order of magnitude, flocs are usually found in the size range 20–200 μm , and flocs of highest size in this range (including flocs of even higher sizes) are organic-matter rich (Safar, 2022). This organic matter comes either from the water column (microalgae and their EPS) or from the bed, where biofilms can be created (Droppo, 2009; Lai et al., 2018; Ho et al., 2022). Pure mineral clay flocs (flocs composed of mineral clay aggregated through electrostatic interactions) remain limited in size by the Kolmogorov microscale (Mietta, 2010). Flocs can be divided into smaller or primary flocs under the action of shear stress or grow larger by collision with other particles (labels 3–5 in Figure 1). As the environmental factors are changing, it is argued that flocs stay in a dynamic process of aggregation and break-up (Burban et al., 1989; Eisma, 1991;

Lick et al., 1993). This classical picture should however be reconsidered in the light of organic matter properties: when organic matter consists of polymeric substances (such as proteins, sugars or DNA), flocs display elastic properties and show a high resistance to shear (Shakeel et al., 2020). Increasing shear rate then does not lead to break-up of flocs but rather to a decrease in volume, for a constant mass (the flocs become denser).

Flocculation is affected by three main types of factors: physical factors, chemical factors, and biological factors. There is quite some overlap between these different types, and one could well speak of physico-chemical factors or bio-chemical factors. Physical factors include hydrodynamic conditions (such as shear stress and vertical mixing) and sediment particle characteristics (such as particle size distribution and sediment concentration); chemical factors consist of environmental conditions (such as salinity, temperature, pH), mineral composition and surface charge of sediment particles; biological factors mainly refer to the action of microorganisms and their excreted biopolymers (Gibbs, 1985; Dyer and Manning, 1999; Winterwerp and Kesteren, 2004; Mietta et al., 2009a; Droppo et al., 2015; Fettweis and Lee, 2017). Many factors depend on each other (the role of biopolymers in flocculation is for instance related to salinity and temperature) which makes parametrizing of flocculation models extremely complex.

In the past 30 years, a lot of research has been performed on flocculation but the effect of biological processes was seldom taken into account (Thomas et al., 1999; Winterwerp, 1999; Maggi, 2005; Mietta, 2010). As there is a significant biological activity in aquatic environments, it has long been recognized that organic matter has a great impact on flocculation (Avnimelech et al., 1982). It is therefore not surprising that research on the effect of organic matter on sediment flocculation is gradually becoming more extensive. Most of the studies, however, are focused on the qualitative description of such flocculation. The distinction between different bio-flocculation mechanisms, the different floc types and their link to the factors cited above are still lacking. In this review, those correlations are discussed. The purpose of the review article is to summarize the state-of-the-art regarding sediment flocculation and presenting some of the open questions to be answered in the future.

2 The population balance model

In the standard model used to describe flocculation (the Population Balance Equation, PBE in short), a size class i is defined as a collection of particles with concentration n_i , all particles in a same class having the same size (diameter) d_i .

Aggregation and break-up processes of particles are expected to occur simultaneously and dynamically in the PBE model

(Winterwerp and Van Kesteren, 2004; Chassagne, 2019; Chassagne et al., 2021).

Two parameters are related to the particles' aggregation ability: the collision frequency and the collision efficiency. The aggregation rate of particles depends on both these parameters.

2.1 Collision frequency

There are three main expressions for the collision frequency, based on the different approach mechanisms between particles. Particles can approach one another by Brownian motion, mixing by shear or differential settling (McCave, 1984; van Leussen, 1994; Tsai and Hwang, 1995). So, if a collision occurs between a particle of diameter d_i and a particle of diameter d_j , the collision frequency $f_{i,j}$ can be expressed as follows (Hunt, 1980):

Brownian motion (BM):

$$f_{i,j}(BM) = \frac{2}{3} \frac{k \cdot T}{\mu} \cdot \frac{(d_i + d_j)^2}{d_i \cdot d_j} \quad (1)$$

Mixing by shear (SH):

$$f_{i,j}(SH) = \frac{G}{6} (d_i + d_j)^3 \quad (2)$$

Differential settling (DS):

$$f_{i,j}(DS) = \frac{\pi}{4} (d_i + d_j)^2 |\omega_{s,i} - \omega_{s,j}| \quad (3)$$

The parameter k is the Boltzmann constant ($1.38 \times 10^{-23} \text{ J K}^{-1}$), T is the absolute temperature (293 K), μ is the dynamic viscosity of the suspending medium—water ($1.005 \times 10^{-3} \text{ Pas}$ when the temperature is 20°C), G is the shear rate (s^{-1}), g is the gravitational acceleration (9.8 m s^{-2}), and $\omega_{s,i(j)}$ represents the settling velocity of the flocs.

Brownian motion is caused by the random thermal motion of liquid molecules, so in general it is only effective for sediment particles under the colloidal size of 1–2 μm (Dronkers et al., 1988; Lick et al., 1993; Partheniades, 1993). Brownian-induced flocculation is a slow process and in estuarine systems, where the water column experiences hydrodynamic mixing, it is not expected to be the dominant cause for flocculation.

The effect of shear rate is considered to be important for flocculation in estuarine systems. Many studies (Mehta and Partheniades, 1975; Eisma, 1991; Partheniades, 1991, 1993; Manning and Dyer, 1999; Winterwerp et al., 2006; Mietta et al., 2009a) have provided a better understanding about the relations between shear rate and flocculation. As is shown in Eq. 2, the collision frequency increases with increasing shear rate, thus promoting flocculation and the growth in particle size. However, when the particle size of flocs reaches the Kolmogorov microscale η , the increase upon shear rate will be limited. Eisma (1986) and van Leussen (1997) in particular argued that the maximum diameter of flocs should be

equivalent to the Kolmogorov microscale (minimum turbulent vortex scale) and this was also experimentally verified for salt-induced flocculation of mineral clay particles (Mietta et al., 2009a; 2009b). The Kolmogorov microscale is defined in Eq. 6.

2.2 Collision efficiency

The collision efficiency is mainly driven by the interaction forces between particles upon approach. These forces are linked to physical, chemical and biological processes, and include electrostatic forces, Van der Waals force (VDW), hydrophobic interactions and entropic forces. In the past decades, researchers have put forward a number of theories and models for sediment flocculation, among which there is the standard electrokinetic theory (named DLVO, for Derjaguin, Landau, Verwey and Overbeek), polymer bridging, cation bridging, sweep flocculation and others (Kruyt, 1949; van Leussen, 1988, 1994; Higgins and Novak, 1997; Bolto and Gregory, 2007; Lee et al., 2011). When studying the aggregation of similar primary particles (the primary particles being similar is one of the underlying hypothesis of the PBE model), the DLVO theory can provide a useful parameter (the ζ -potential) which helps to predict whether aggregation will or will not occur.

2.2.1 DLVO theory

The DLVO theory models the interaction potential between two approaching colloidal particles in a solvent (usually water). The interaction is made of two terms: a Coulombic repulsion and a van der Waals' attraction (Kruyt et al., 1952; Chassagne, 2019). The two particles have a surface charge of same sign and are surrounded by a so-called double layer which is composed, in majority, of counterions. The double layer is the result of the attractive interaction between the surface charge of sediment particles (usually negatively charged for mineral sediment) and the positively charged ions (cations) found in water. The ζ -potential is used to quantify the surface charge of a particle. It is defined as the electric potential at the surface of shear of the particle (the potential of reference is taken at infinity, where it is defined as zero). When this surface of shear is located at the surface of the particle, the ζ -potential is identical to the electric surface potential Ψ_0 (Chassagne and Ibanez, 2012). The potential Ψ_0 depends on the number of charged sites and the number of ions adsorbed on the particle surface. In most cases, the shear plane is located a few ångström away from the surface, in the layer called the Stern layer (Chassagne et al., 2009), which is the layer between the surface of the particle and the plane of shear. When the concentration of cations increases in the water, there will be a reduction in ζ -potential, as more ions will be located between the particle's surface and the shear plane, leading to better screening of the surface charge.

As is seen in Figure 2A, the electric potential drops from ζ_1 to ζ_2 when the salt concentration is increased from C_1 to C_2 . The

interaction energy curve displays a maximum which indicates that a repulsion force is preventing particles to aggregate when Coulombic repulsion is dominant. This barrier can be overcome by increasing ionic strength or lowering pH but can also be overcome by "pushing" the particles together. This happens *in-situ*, when the water flow enables particles to collide (see Section 2.1).

For more details about DLVO theory, the reader is referred to Chassagne (2019). There exists a lot of studies about the ζ -potential and the surface charge of clays (Rand and Melton, 1977; Sondi et al., 1996; Kosmulski and Dahlsten, 2006; Mietta et al., 2009a; Chassagne et al., 2009; Mietta, 2010; Tsujimoto et al., 2013), and the results show that either a decrease in pH or an increase in ionic strength usually decreases the absolute value of the ζ -potential. A low ζ -potential is synonymous for aggregation ability: particles with a low ζ -potential have a weak Coulombic repulsion. For these particles, van der Waals attraction will dominate and lead to flocculation Figure 2B. It was shown in (Mietta et al., 2009a; 2009b) that the ζ -potential could be related to the collision efficiency.

2.3 Break-up mechanisms

When shear forces are stronger than the bonding force between particles, it is expected that flocs will break-up.

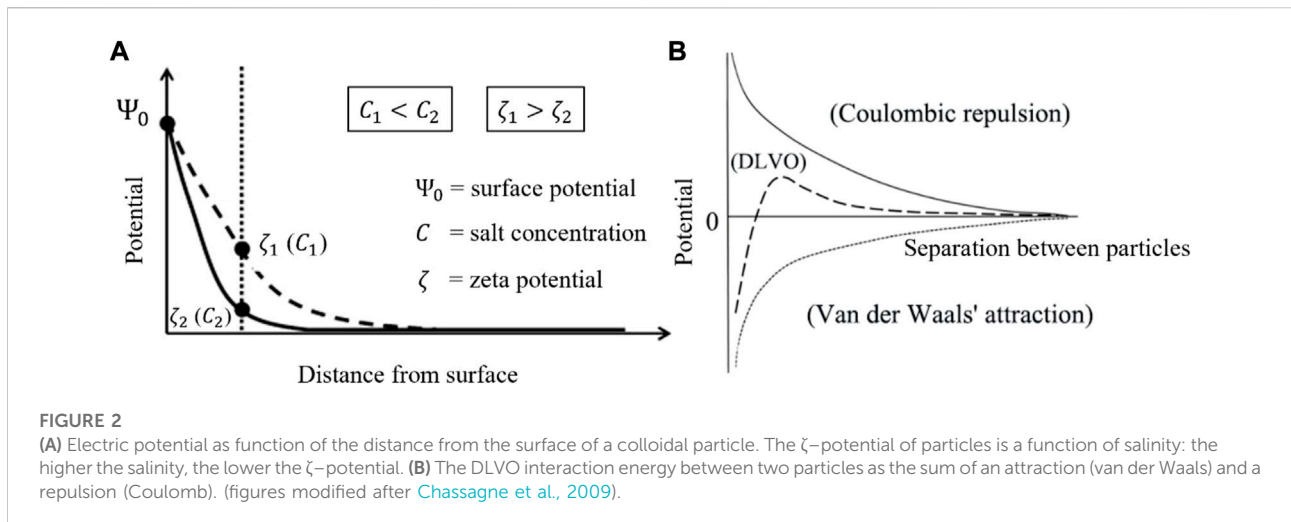
The break-up rate B_i is usually used to describe the break-up of flocs and it is traditionally a function of shear and size of particles (Lick and Lick, 1988). The break-up term corresponding to the breaking of particles is given by $B_i(TS)$. From a modeling point of view, particles can leave a size class either by break-up (flocs of smaller sizes are then created), through $B_i(TS)$, or by aggregation (a floc of larger size is then created), through the term $B_i(C)$ (Spicer and Pratsinis, 1996). The term $B_i(C)$ is also depending on turbulent shear, as this shear enables particles to collide and subsequently aggregate (Serra and Casamitjana, 1998). The expression for $B_i(TS)$ is:

$$B_i(TS) = EG^b d_i^p \quad (4)$$

where d_i is the size of floc in class i . The parameter p is usually taken equal to 1 and E and b are fitted to data. The term $B_i(C)$ is given by:

$$B_i(C) = \sum_{j=1}^N a_{i,j} f_{i,j} n_j \quad (5)$$

where $a_{i,j}$ is the collision efficiency, $f_{i,j}$ represents the frequency of particle collision, and n_j is the number of flocs of a given size d_j . Usually $a_{i,j}$, which should be between 0 (no aggregation) and 1 (always aggregation), is taken to be constant and arbitrarily equal to $a = 0.5$ (Mietta, 2010). It can be seen from Eq. 1 that the increase of either shear rate or the floc size will increase the break-up rate of particles. The term $B_i(C)$ depends on shear and particle size, through the



term $f_{i,j}$, see Eq. 2: the larger the particles, the larger the collision frequency.

In general, the floc break-up process is poorly understood (Thill et al., 2001). The empirical parameters should account for the bonding forces between particles as the stronger the bonds, the more resistant the floc is to any exterior force (Jarvis et al., 2005) or inner tensile stress (Yeung and Pelton, 1996), see Figure 5. The ability of flocs to resist damage (floc strength) is closely related to flocculation and the size and structure of flocs (Son and Hsu, 2009). Floc strength is a function of the electrochemical attraction between clay minerals and the agglutination of organisms and organic matter on particle surface (Passow, 2002; Bainbridge et al., 2012).

2.3.1 The Kolmogorov microscale

Shear stress is one of the most important external factors affecting the flocculation process and has been widely studied (e.g., Dyer and Manning, 1999; Mietta et al., 2009b; Partheniades, 1993; Winterwerp, 1998).

The turbulent shear rate G is often used to quantitatively describe the turbulent strength:

$$G = \sqrt{\epsilon/\nu} = \nu/\eta^2 \quad (6)$$

where ϵ is the turbulent energy dissipation rate, ν is the kinematic viscosity of water body, and η is the Kolmogorov microscale. The Kolmogorov microscale is the smallest scale of a turbulent eddy.

3 Flocculation by organic matter

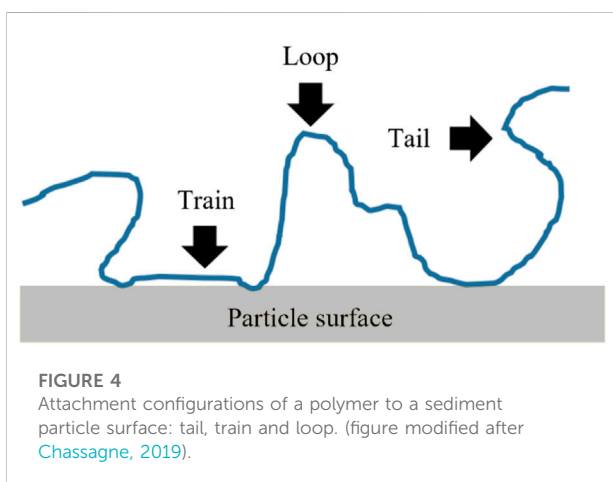
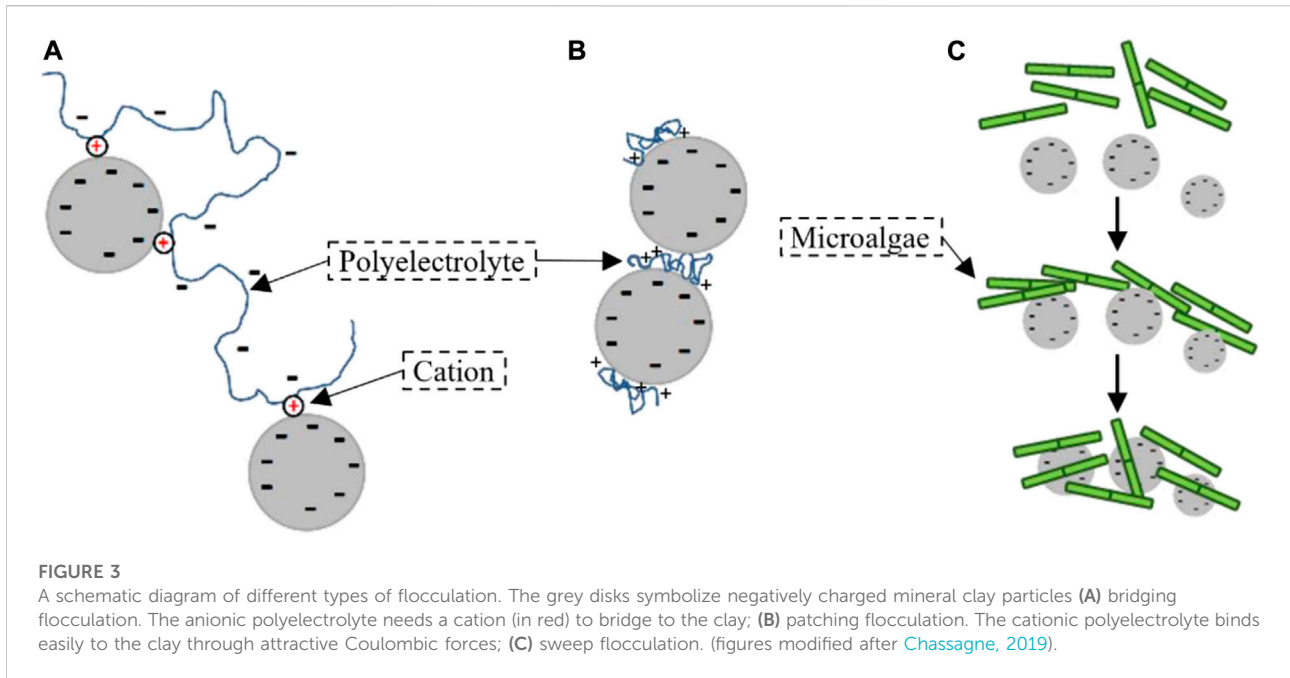
Organic matter, and more specifically the polyelectrolytes produced by microorganisms bind to sediment particles in different ways, depending on their polymeric chain length (defined by the polymeric molecular weight), and charged

groups. Their way to bind to sediment particles is also depending on salinity, pH and shear stresses. Polyelectrolyte-induced flocs can achieve sizes much larger than the Kolmogorov microscale, owing to the polyelectrolyte shape (chain-like) and elastic nature (Ibanez Sanz, 2018; Shakeel et al., 2020).

Depending on the polyelectrolyte, flocculation between mineral sediment and polyelectrolyte can occur through bridging (Riley, 1963; Nabzar et al., 1988; Hicks, 1988; Winterwerp and Kesteren, 2004), patching mechanism (Bergaya and Lagaly, 2013) or sweep, as shown in Figure 3.

A polyelectrolyte can stick to certain points of the sediment's surface as trains, separated from one another by loops and for much of its length it is able to extend into the solvent as tails (Figure 4). The attraction between the interacting molecular chains should be strong enough to overcome the entropy repulsive force caused by the decrease in freedom of the chains (Rosen, 2004).

Under suitable conditions, microbial organisms, such as diatoms, can produce polyelectrolytes (polysaccharides), which is a gelatinous organic matter (OM). These polyelectrolytes are called Transparent Exopolymer Particles (TEP) or Extracellular Polymeric Substances (EPS). They are polymeric chains which include a large amount of anionic polysaccharides like galacturonic acid that is the main component of pectin (Plude et al., 1991). The organic polymers can form biofilms on the surface of sediment particles and act as flocculating agent (Bar-Or and Shilo, 1988b; Bar-Or and Shilo, 1988a). Because of the ionization of their functional groups (such as carboxylic acid and phosphate), microbial cells and EPS have a high density of negative charges (Sheng et al., 2010), and anionic polysaccharides aggregate with negatively charged sediment particles through cationic bonding. These cations are salt ions found in the water, and their concentration is a function of salinity. The EPS secreted from microorganisms can also be a cationic polymer (Plude et al., 1991). Aly and Letey (1988) have



found that flocculation by cationic polymers is different from flocculation by anionic polymers. Flocculation with cationic polymer is done by charge neutralization, while flocculation with anionic polymer is mainly done by bridging mechanism (Shakeel et al., 2020). Studies have shown that adding divalent cation can enhance flocculation (Yeh, 1988; Park et al., 2010).

3.1 Bridging aggregation

Bridging aggregation occurs when a free polyelectrolyte in solution captures suspended mineral particles (see Figure 3A). Usually, one finds that the optimum polymer concentration to

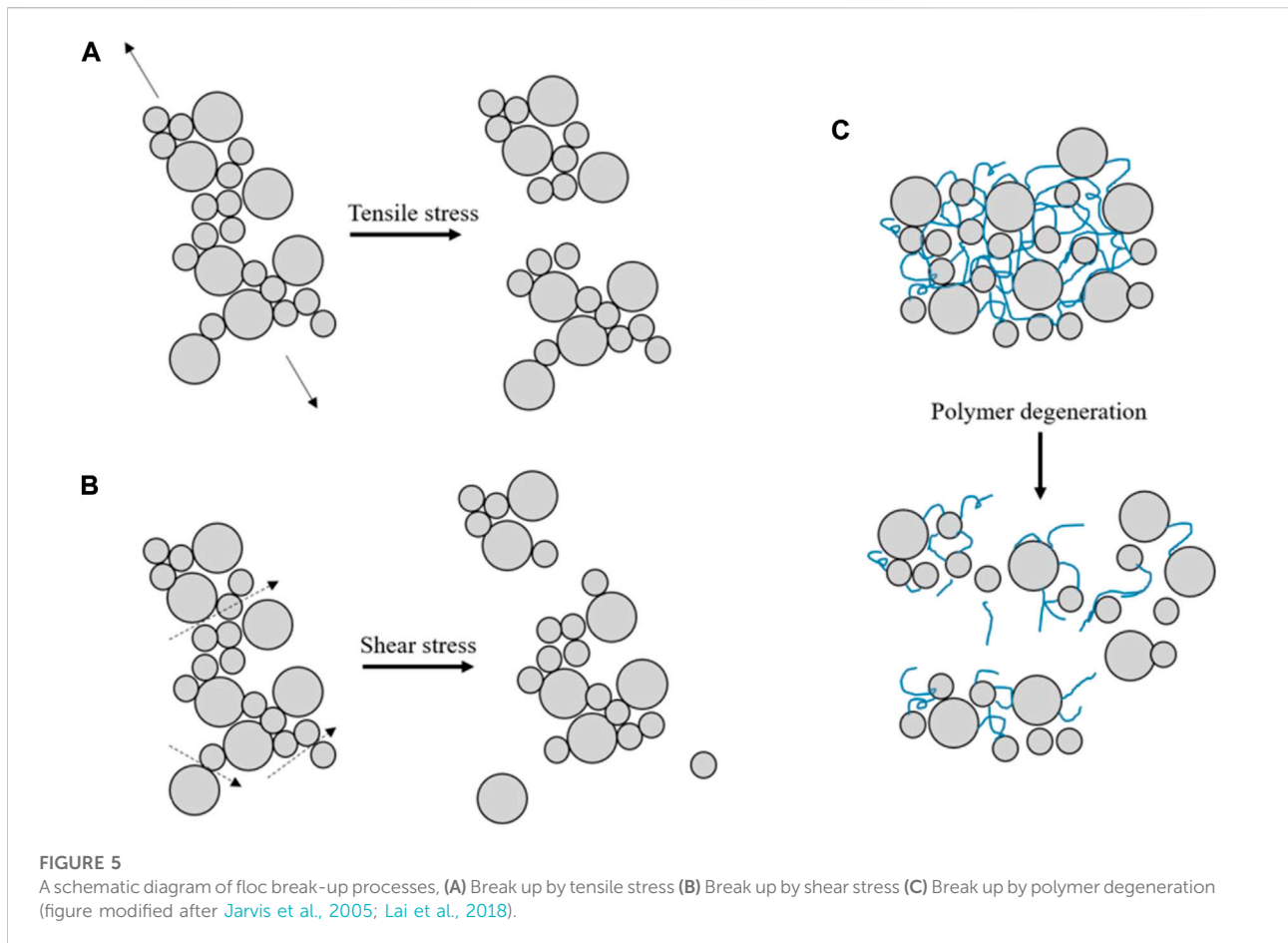
achieve flocculation corresponds to half surface coverage for the polymer. Polymeric bridges are changing as function of shear. Bridging aggregation can even occur with polyelectrolytes having surface charges of same sign as the ones of the particles. In that case, aggregation is enabled by the presence of oppositely charged ions in the water. When polymer bridging flocculation happens, the bridging particles should have an available surface to connect polymer chain segments (Biggs et al., 2000). Flocs formed by bridging through polymer chains are flexible and stretchable (Otsubo, 1992; Gregory and Barany, 2011).

3.2 Patching aggregation

Patching aggregation occurs when polyelectrolytes have a charge that is opposite in sign to the one of the sediment particles (see Figure 3B). The polyelectrolyte then strongly binds to the sediment particle, and its tails do not extend much into the solvent. Aggregation is then made possible between one polymer patch of one particle and the bare surface area of another particle. Flocs formed through patching aggregation have generally a higher strength than those formed through other ways. The force is dependent on the number of bound segments (Swenson et al., 1998).

3.3 Sweep flocculation

As is visible under a microscope, microorganisms can form a polymeric network with holes and channels (Li and



Ganczarzyk, 1990) through the bridging of their produced Extracellular Polymeric Substances (EPS) (Leppard, 1992; Jorand et al., 1995). This network has a large surface area and can absorb pollutants, nutrients and minerals (Costerton et al., 1987). At the same time, there are large and small interspaces inside the network. When sedimentation of this network occurs, some fine particles can be trapped by it and be embedded (see Figure 3C). This process is called sweep flocculation. Although sweep flocculation is usually studied in sewage treatment, a similar process has been observed in estuarine environments (Gregory, 2005; Lee et al., 2011). Sweep flocculation is a function of electrochemistry and other characteristics of the network. As a result, the network floc gets increasingly larger (Deng et al., 2019).

To summarize, the flocculation of fine sediment under the influence of organic matter is a complicated dynamic process involving chemistry, physics and biology. The time-dependence of flocs composed (in part) of organic matter is poorly understood, especially in relation with the living microorganisms that are known to bind to mineral sediment thanks to EPS (Deng et al., 2019; Deng et al., 2021; Safar et al., 2022).

Recent studies have found that organic matter-induced flocculation is also affected by biological decay and that the strength of bonds is time-dependent, see Figure 5. Flocs can therefore break without the influence of shear, simply by biodegradation (Mikutta et al., 2007; Jeldres et al., 2018).

The rate of microbial degradation depends on the environment and leads to catabolism (Foree and McCarty, 1970). The degradation is different for different types of EPS (Zinkevich et al., 1996; Comte et al., 2006). Living microorganisms (that produce EPS and other types of polyelectrolytes) are function of grazing (Porter, 1973) and water nutrients (Chai et al., 2006; Li et al., 2014). Further study on the degradation of EPS in combination with its interaction with sediment is required.

4 Discussion on flocculation models

4.1 The PBE vs. the logistic growth model

Using the PBE model explained in Section 2, it is found that an increase in turbulent shear stress increases the collision

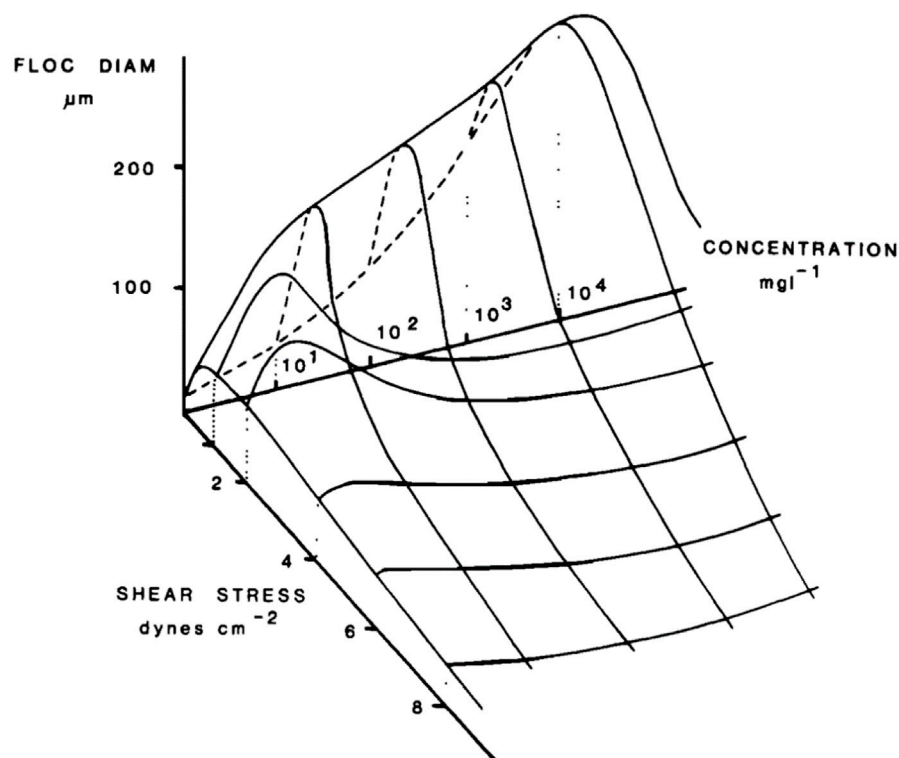


FIGURE 6

A conceptual diagram of the relationship between floc size, turbulent shear and suspended sediment concentration (figure following after Dyer, 1989).

frequency between sediment particles hereby promoting sediment particle aggregation. On the other hand, high shear stresses reduce the Kolmogorov microscale η and increases the break-up frequency of flocs.

Using the Population Balance Model, the steady-state average size of a floc will be the Kolmogorov microscale (Mietta, 2010). When a floc (in the model) has a size larger than the Kolmogorov microscale, it is expected to break-up into smaller flocs with a denser structure (Dyer, 1989; Winterwerp, 2002).

Previous studies suggest that the maximum size of flocs formed in turbulent flow is controlled by η (Eisma, 1991; van Leussen, 1997; Droppo et al., 2000; Mikkelsen, 2002a; Mietta et al., 2009a; Mietta, 2010). There is indeed a positive correlation between the floc size and Kolmogorov microscale, which are usually of the same order of magnitude (van Leussen, 1997; Mietta et al., 2009a; Kumar et al., 2010).

Dyer (1989) first suggested that there exists a critical value of shear stress, below which the floc size increases and after which decreases and conceptualized this in a figure (Figure 6). The optimum in particle size as function of shear stress is only occurring if all experiments (for each shear stress) are done for a fixed flocculation time, and that the equilibrium floc size has

not yet been reached. Indeed, if the equilibrium floc size would have been reached for all experiments, for fully suspended particles, flocs formed at the lowest shear would be the largest and the floc size as function of shear rate would be a monotonic decrease, as has been shown by Mietta et al. (2009b). For a fixed (short) flocculation time, it is expected that below a critical shear rate the flocculation rate is limited by the amount of collisions and above this critical shear by the Kolmogorov microscale. This is however not true for organic matter flocs—they can grow larger than the Kolmogorov microscale and their flocculation rate is extremely fast (Shakeel et al., 2020).

Note that on Figure 6 there is also a maximum in floc size as function of concentration for a given shear rate. The reason for this optimum is unclear, as one would expect, for any flocculation time, either an increase in floc size as function of concentration (related to an increase in collision frequencies) or that the floc size remains constant (equal to the Kolmogorov microscale η).

The fact that flocs follow the Kolmogorov microscale is true for salt-induced flocs (Mietta, 2010), under the condition that the flocs have reached their equilibrium size and that they remain suspended. At low shear, it can be shown that for dense particles, not all particles remain in suspension and hence the average mean diameter decreases with the increasing number of particles

that are not suspended (Mietta et al., 2009b). It should be noted that in the case of low turbulent shear stress, the flocculation process may change from being dominated by turbulent shear stress to being dominated by differential settling, thus affecting the flocculation process and the characteristics of the final flocculation (Lick et al., 1993; Chen et al., 1994; Fugate and Friedrichs, 2003).

The characteristic values of shear stress between these two regimes have been obtained in many studies, including laboratory experiments (e.g., Manning and Dyer, 1999; Mietta et al., 2009a; Kumar et al., 2010) and field observation studies (e.g., Markussen and Andersen, 2014; Sahin, 2014; Guo et al., 2017). Markussen and Andersen (2014) and Guo et al. (2017) showed that the characteristic turbulent shear rate in their *in-situ* studies was about $3\text{--}4\text{ s}^{-1}$, whereas the critical range obtained in other studies (Manning and Dyer, 1999; Serra et al., 2008; Kumar et al., 2010; Zhang et al., 2013; Sahin, 2014) was about $15\text{--}40\text{ s}^{-1}$, indicating that the flocs properties might be different in different estuaries.

In estuarine and coastal areas, the largest shear gradient is close to the channel bed (10%–20% of the water column above bed is affected), where about 80% of turbulence energy is present. This produces a powerful uplift force for particles and the local shear stress controls the maximum size of suspended flocs (Mehta and Partheniades, 1975). From a PBE modelling perspective, in an area with constant shear stress, the strength, size and density of flocs will be in a dynamic equilibrium. When the shear stress is varied from low to high, flocs will become smaller. When the shear rate is varied from high to low, flocs will grow larger. These processes are completely reversible when the PBE model is used, implying that the structure of a floc of a given size will always be the same, independently of its history.

One can question the validity of the PBE model in the case of *in-situ* flocculation, for several reasons. First of all, it is not proven, as the PBE assumes, that the size of a floc is governed by a dynamic balance between aggregation and break-up. As was shown in Chassagne and Safar (2020), it is well possible to model flocculation data by using a logistic growth model. This model, in contrast to the PBE model does not require break-up to ensure a realistic steady-state floc size. Indeed, without the break-up term, the PBE model would give an infinitely large equilibrium floc size. As discussed in Chassagne and Safar (2020), Chassagne et al., (2021), an equilibrium floc size can, for example, be reached because of a depletion of flocculating agent (like EPS). Over time, mineral sediment and EPS bind together until all available EPS is covered with mineral sediment and the floc size reaches a constant value. The amount of EPS is then the limiting factor for the equilibrium floc size (Shakeel et al., 2020). Using the PBE model, a steady-state floc size (not an equilibrium size) can be reached only when there is a balance between the aggregation and break-up terms, which then keeps the floc size constant over time. This type of balance is for instance found in thermodynamics, when one studies the

adsorption/desorption of molecules from a surface (Langmuir, 1918), but is not representative for EPS-induced flocculation.

Secondly, it has been observed that flocs do not necessarily break under shear, but rather change shape as the organic matter they contain is rather elastic. This implies that, even at constant shear, sediment and organic matter concentration, floc sizes can decrease over time due to the fact that flocs become denser (less volume, same mass) (Shakeel et al., 2020). Using the PBE model, it is only possible to decrease a particle size by break-up, therefore this process cannot be modelled. Using a logistic growth model, on the other hand, it is possible to model this decrease in floc size and parameterized it (Safar et al., 2022).

The logistic growth model is therefore a promising new tool for flocculation studies, but it requires further investigation. In particular some work should be devoted to the parametrization of the model and its calibration using *in-situ* data.

4.2 The DLVO model and prediction of flocculation

In estuarine areas, the suspended sediment particle will, according to DLVO theory, be destabilized and flocculate because of the increase in salinity between the river fresh water and the sea. In the presence of organic matter however, DLVO theory cannot always be applied, as the flocculation mechanisms will be driven by the presence of polyelectrolytes and microorganisms which are not accounted for in the DLVO theory.

Hunter and Liss (1979) already demonstrated in 1979, by analyzing the surface properties of suspended particles in four rivers in the United Kingdom by electrophoresis (a technique to assess the ζ -potential) that because of the organic coatings of the particles no major difference could be found in the particles' surface characteristics.

Different clay minerals with different particle sizes and surface charges are found in flocs. Goldberg (1991) found that the optimum flocculation salinity for different clay minerals is different. The optimum flocculating salinity of illite and kaolinite is 9–13 PSU, while that of montmorillonite is 20–24 PSU. This result is in line with DLVO theory, as montmorillonite clay, being delaminated, has a larger surface area than kaolinite or illite particles and hence the particles experience a larger Coulombic repulsion.

The general trend that was found is that the electrophoretic mobility (and hence the ζ -potential) is decreasing (in absolute values) with increasing salinity. This behavior is in line with the description given in Figure 2A and reflects the screening of the surface charge of the coated particles.

On the other hand, the DLVO approach, which predicts that the increase of ion concentration is beneficial for flocculation, cannot explain why an increase in sodium concentration will slow-down flocculation in some situations (Sobeck and Higgins, 2002).

Physico-chemical models also do not take into consideration the uneven charge distribution on sediment surfaces and the charge distribution of EPS, which most probably have a great influence on bio-sediment flocculation. Moreover, the colony formation and growth of microorganisms is a dynamic process that will influence bio-sediment flocculation. Microbial activity is a complex process that is hard to quantitatively study, especially in field observations. So far, the biological activity is studied by quantifying the number of microorganisms present in the water, understanding their growth cycle, analyzing their EPS, etc. The link between biological activity and sediment flocculation remains a key open question for the study of sediment transport dynamics.

5 Characterization of floc properties

Properties of flocs include size, shape, effective density and composition. These parameters are important to understand the cohesion of flocs and their settling velocity. The settling velocity of flocs is an important parameter for sediment transport modeling (Winterwerp, 1998; Manning et al., 2011; Verney et al., 2011; Chassagne et al., 2021).

5.1 Floc sizes and shapes

Generally, floc size is assessed by laser diffraction techniques (with equipment like Malvern Mastersizer and LISST) (Dyer and Manning, 1999; Agrawal and Pottsmith, 2000; Mikkelsen and Pejrup, 2001; Mietta, 2010; Filippa et al., 2011; Guo et al., 2017). These techniques convert the raw data (diffracted light) into particle size by making use of the assumption that particles are spherical and represented by their equivalent mean diameter. The particle sizes are given in logarithmically-spaced size bins, and the particle size distribution (PSD) is given as a volume-% (volume occupied by particles of a sizes within a given bin compared to the volume occupied by all particles). It is usually assumed that the D_{50} of the distribution is representative for the mean particle size of the distribution (Maggi, 2005). Nonetheless, it often occurs that a PSD displays multiple size peaks (van Leussen, 1994), which reflects the fact that different types of particles are present in the sample, such as mineral sediment, flocs and microorganisms like algae (Simon et al., 2002; Grossart et al., 2003; Tang, 2007; Deng et al., 2019; Safar, 2022). Microorganisms generally develop extensive web-like networks on flocs, with a larger size and lower fractal dimension than those without microbial colonization (Kjørboe et al., 1990; Azetsu-Scott and Passow, 2004).

Multiple peaks in PSD also occur because of the shape of particles (Agrawal and Pottsmith, 2000; Safar, 2022). For example, Liu et al. (2007) found by scanning electron microscope technique that the sediment flocs of the

Changjiang estuary at different salinities and organic matter have various shapes. Safar (2022) found, from underwater camera investigations, that flocs composed of a large amount or exclusively of organic matter are highly anisotropic, when this organic matter is composed of chain-like microalgae. Mhashhash et al. (2018) showed that at different suspended sediment concentrations (SSC) flocs of different shape are found. In high shear stress areas such as estuaries and coasts, flocs are usually small and spherical or ellipsoidal (van Leussen, 1994; Craig et al., 2020). Some typical flocs are given in Figure 7. In low shear stress environment, the flocs are usually elongated in a chain-like fashion due to their algal composition (Manning et al., 2007; Safar, 2022). The drag force on these flocs will be different and this will thus affect the aggregation between particles leading to larger flocs of various shapes (Maggi, 2005; Adachi et al., 2012). Anisotropic flocs containing a substantial amount of organic matter can withstand a certain extent of deformation, leading over to the reformation of these flocs into fairly spherical flocs (Jeldres et al., 2018; Safar, 2022). Degradation of polymers can also lead to this phenomenon (Heath et al., 2006).

5.2 Floc density and settling velocity

Figure 8 shows the relationship between effective density (floc particle density minus water density) and floc size obtained from several published studies. The results are for many different rivers and estuaries, such as the upper Tamar River in the United Kingdom (Fennessy et al., 1994), the Po River Estuary in Italy (Fox et al., 2004) and the Changjiang River Estuary (e.g., Wang et al., 2013; Guo et al., 2017) and laboratory experiments with natural sediment (e.g., Gibbs, 1985; Manning and Dyer, 1999). The effective density of flocs displays a significant decrease as function of particle size which can be of several orders of magnitude. However (not shown) in all these experiments there were also quite some large density differences for a given particle size. These differences arise from the fact that flocs of a given size can have very heterogeneous composition (Chassagne and Safar, 2020).

To describe quantitatively the relation between floc size and floc effective density, Kranenburg (1994) proposed that the flocs should be treated as fractals. A fractal is defined as a self-similar object (Mandelbrot, 1982). In existing studies, there are two main descriptions of fractals: one is the two-dimensional fractal dimension N_p based on perimeter and area, and the other is the three-dimensional fractal dimension N_f based on the volume of particles contained inside a floc. It is the latter that is of interest to determine the density of flocs. Kranenburg (1994) derived the following relationship between effective density and particle size:

$$\Delta\rho = \rho_f - \rho_w \propto (\rho_p - \rho_w) \left(\frac{d_f}{d}\right)^{N_f-3} \quad (7)$$

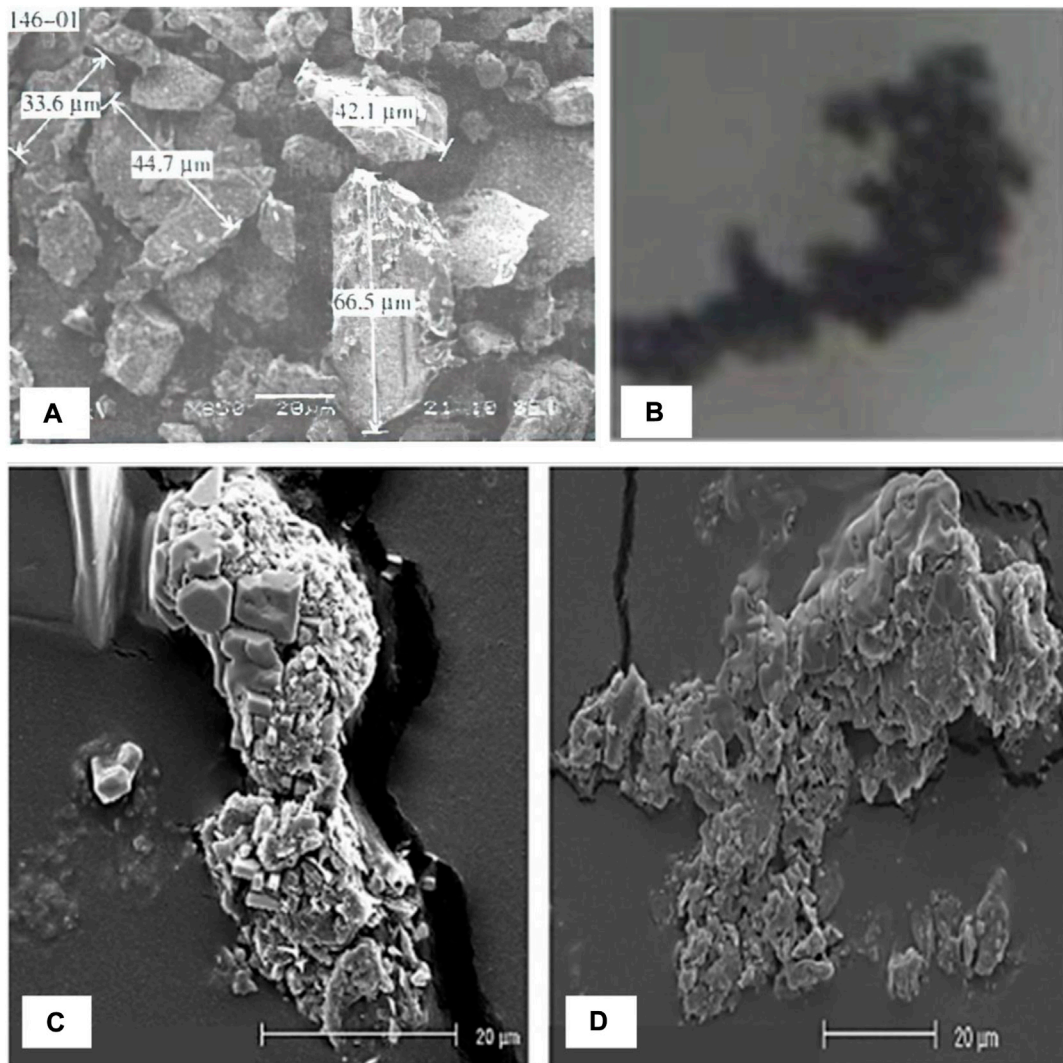


FIGURE 7

Photomicrography of flocs, (A) electron microscope photographs of flocs in the freshwater with humus, figure from [Liu et al. \(2007\)](#) (B) Camera photographs of flocs in the Rhine Region Of Freshwater Influence (RHine ROFI), figure from [Safar, 2022](#) (C,D) SEM photographs of flocs at salinity 20, different SSC, c = 100 and d = 200 mg L⁻¹, figures from: [Mhashhash et al. \(2018\)](#).

where $\Delta\rho$ is the effective density of floc, ρ_f and ρ_w are the density of floc and the density of water, respectively. ρ_p is the density of mineral sediment particles, d_f and d represent the floc size (D_{50}) and the mean particle size of the constitutive particles. It can be seen that for a same size of constitutive particles and flocs, different fractal dimensions lead to different effective densities. By tuning the fractal dimension, it is therefore possible to fit the different effective densities as function of floc sizes found in [Figure 8](#). Note, however, that the found fractal dimension is a purely empirical parameter, as, in contrast to theoretical flocs, flocs observed *in-situ* are usually composed of different primary constituents (mineral sediment, EPS, etc.).

Usually the density of flocs is obtained from settling velocities experiments from which, using Stokes' settling velocity, the density can be deduced. In general the settling velocity of a sinking object is expressed as:

$$\omega_s = \sqrt{\frac{4}{3} \frac{\alpha}{\beta} \frac{\Delta\rho \cdot g \cdot D_{50}}{C_D \cdot \rho_w}} \quad (8)$$

in which α and β are coefficients depending on the sphericity of the particles as defined in ([Winterwerp, 1998](#)). C_D is the drag coefficient and ω_s is the settling velocity of flocs. The drag coefficient is a function of particle Reynolds number ([Winterwerp, 1998](#)):

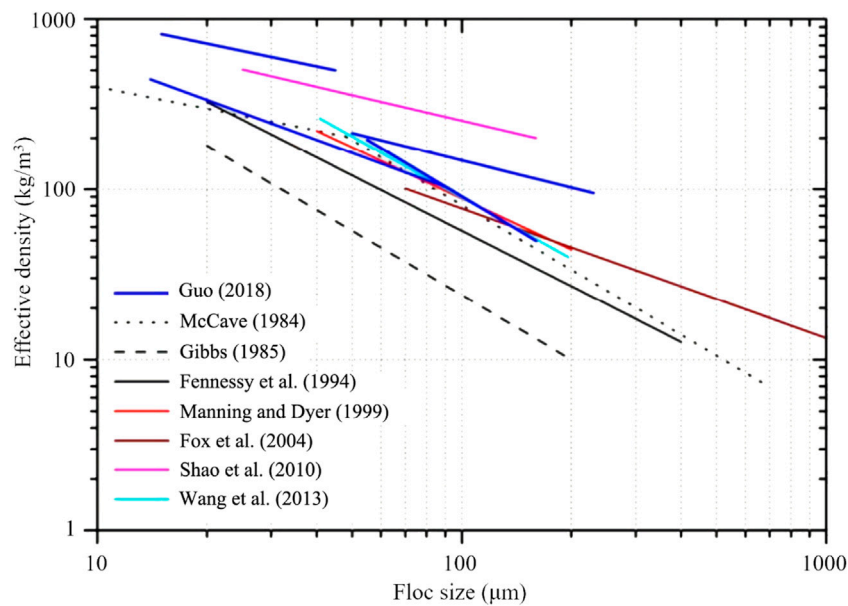


FIGURE 8

The change of effective density of flocs with particle size (McCave, 1984; Fennessy et al., 1994; Manning and Dyer, 1999; Fox et al., 2004; Shao et al., 2011; Wang et al., 2013; Guo, 2018).

$$C_D = \frac{24}{Re(1 + 0.15Re^{0.687})} \quad (9)$$

The particle Reynolds number is given by $Re = \omega_s D_{50}/\nu$, where ν is the kinematic viscosity of water.

For spherical particles, $\alpha = \beta = 1$. When the particle Reynolds number $Re \ll 1$, the above expression can be simplified to Stokes' settling velocity:

$$\omega_s = \frac{\Delta\rho \cdot g \cdot D_{50}^2}{18\mu} \quad (10)$$

In fine sediment transport models, the settling velocity is usually taken to be constant, and its value is about $0.05\text{--}0.1 \text{ mm s}^{-1}$ (e.g., Geyer et al., 2004; Hu et al., 2009). This assumption of a constant settling velocity is an approximation, as it was discussed above that flocs can change shape and density as function of shear (and time).

Experiments in controlled conditions and field observations have given evidence that the organic matter (especially the EPS) has substantial effects on floc density, porosity and settling velocity that leads to a larger size but lower density and settling velocity as compared to the pure (without EPS) sediment particles (Droppo, 2001; Guenther and Bozelli, 2004; Passow and De La Rocha, 2006; Bowers et al., 2007; Droppo et al., 2015). On the other hand, some studies found that organic matter can increase floc density under certain conditions (Fall et al., 2021). Low density and large size flocs are predominantly formed in the water column where large particles of organic

matter (like microalgae) are advected. Dense and small flocs with comparable settling velocity can be formed in regions where organic matter is less abundant, or where the shear is high, as by shearing flocs become denser and get a more spherical shape (by coiling) (Deng et al., 2019; Deng et al., 2021; Chassagne et al., 2021).

5.3 Floc composition and relation to flocculation and settling

It is usually assumed that flocs are composed of fine mineral sediment (clay and silt fraction) with some amount of organic matter (van Leussen, 1988, 1994). However, sand can also sometimes be entrapped in flocs (Deng et al., 2021).

5.3.1 Mineral sediment

The content of clay minerals in the suspended sediment of the Changjiang estuary is about 26%, of which 65%–70% are illite, and the rest are chlorite, montmorillonite and kaolinite. The settling velocity of illite is 9 times higher than that of montmorillonite when the salinity is 10 PSU. The main types of clay minerals vary as function of the tidal periods because the sediment transport and deposition mechanisms are different (Zhang, 1996). The laboratory experiments and field observations of the flocculation characteristics of fine sediment in the Changjiang Estuary show that the optimum flocculation salinity ranges from 4 to 16 PSU (Guan et al., 1996;

TABLE 1 Some parameters reported in *in-situ* observation of flocs.

Authors	Location	Sp.	Se.	Parameters				
				Size	ω_s	SSC	Sal.	OM
Eisma and Li (1993)	Dollard	○	—	○	—	○	○	○
van der Lee (2000)	Dollard	—	○	○	○	○	—	○
Thill et al. (2001)	Rhone	○	○	○	○	○	○	○
Mikkelsen (2002a)	Danish coast	○	○	○	○	○	○	○
Fugate and Friedrichs (2003)	Chesapeake Bay	○	—	○	○	○	○	○
Fox et al. (2004)	Po	○	—	○	○	○	○	○
Xia et al. (2004)	Pearl river	○	—	○	○	○	○	○

^aSp., Spatial variation; Se., Seasonal variation; Sal., Salinity; OM, Organic Matter.

^b“○” means the factor has been measured or taken into account, and “—” means it has not been measured or taken into account.

Jiang et al., 2002; Wan et al., 2015). Manning et al. (2010a) studied the flocculation process with different mud/sand mixtures and demonstrated that different ratios of mud and sand can influence the flocs settling velocity.

5.3.2 Extracellular polymeric substances and microbial communities

Flocs can be regarded as individual microecosystems with autonomous and interactive chemical, physical, and biological reactions and processes within the floc matrices (Droppo et al., 1997; Evans et al., 1997; Brinkmann et al., 2019). EPS and other polymeric substances are produced by microorganisms. These microorganisms can be microscopic algae (such as diatoms), which can form large flocs by themselves, and afterwards capture sediment particles (Deng et al., 2019).

The presence of organic matter like EPS promotes the flocculation of fine sediment (e.g., Droppo and Ongley, 1992; Gratiot and Manning, 2004; Mietta et al., 2009b; Fettweis and Baeye, 2015). It was found that organic matter play a major role in the floc collision rate and floc strength in the observation of seasonal variation of flocs. Large flocs usually appeared at algae bloom seasons due to biologically induced aggregation (Mikkelsen, 2002b; Uncles et al., 2010; Lee et al., 2014; Fettweis and Baeye, 2015). Besides, the biological effects on sediment flocculation also are found in fresh water environments (Guo and He, 2011).

Therefore, the organic matter composition of flocs is not only sensitive to environmental conditions but also varies with spatiotemporal changes. An overview of different studies is given in Table 1.

Organic matter composition can only be assessed in the laboratory. *In-situ* observations do however nowadays include Chlorophyll a measurements, which gives some indication of the microalgae content of flocs (Fettweis and Lee, 2017; Deng et al., 2019, 2021; Ho et al., 2022).

5.3.3 Type of microalgae studied

Diatoms are the largest component of microalgae which goes by the name *phytoplankton*. Therefore, most of the research on the effects of microorganisms on sediment flocculation in estuaries are done with diatoms. Diatoms in healthy state will remain suspended due to turbulence and water current.

Phytoplankton is a large community with strong adaptability. These microorganisms can live under very low nutrient concentration, very weak light intensity and quite low temperature. They can not only grow in rivers, streams, lakes and oceans, but also in short-term ponding or humid places (Blum, 1956; Round, 1981).

In 1968, Walsby (1968) described the aggregation of *N. nutans*, by the EPS this microorganism is secreting. Avnimelech et al. (1982) studied the aggregation process of *Chlamydomonas* and collar algae with clay by electron microscope in 1982 (Figure 9). This is the earliest image of algae and sediment flocculation. Besides, Avnimelech et al. (1982) compared the effect of four different forms of algae on clay flocculation and showed that algae can significantly promote the flocculation of clay in the presence of electrolytes. Therefore, the reason why the growth of algae is restricted in high turbidity areas may not only be due to the limited light supply but also because of the fact that algae have flocculated with sediment particles. In addition, some algae species are prevented to flocculate due to steric effects linked to the spatial conformation of their filamentous strains (de Lucas Pardo, 2014).

5.3.4 Effect of temperature and light

Temperature is the main factor affecting the geographical distribution of algae. Therefore, in the sea area where the water temperature changes greatly, the species changes greatly through the year. In winter, there are cold water algae (the optimum temperature for growth and reproduction is less than 4°C). When the temperature drops to 4°C, both the number of algae and the EPS will however usually decrease (Domozych, 2007; Kiemle

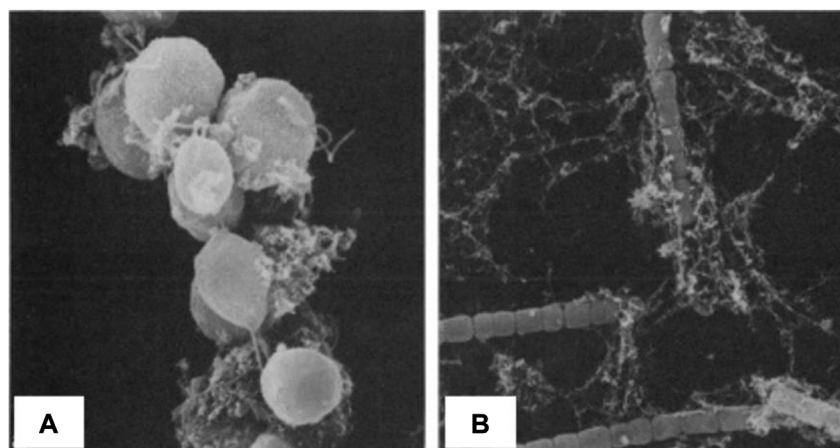


FIGURE 9

Flocs under scanning electron microscope (A) *Chlamydomonas* clay cluster, (B) *Anabaena* clay cluster, 1,200 times, 1,470 times; figures following after Avnimelech et al. (1982).

et al., 2007), and the production of EPS will also decrease, thus reducing the effect on flocculation (Wilén et al., 2000).

There are warm water algae (the optimum temperature for growth and reproduction is about 20°C) in summer, which can complete their life cycle in a short time. The highest value of photosynthesis is between 20 and 25°C, when the algae growth reaches the maximum value (Colijn and van Buurt, 1975; Blanchard et al., 1996), and algae then produce EPS with a high viscosity (Lupi et al., 1991).

Most of freshwater microalgae appear in spring and autumn. Some cyanobacteria only appear when the water temperature is high in summer. Light is the decisive factor to determine the vertical distribution of algae. The light absorption capacity of a water body is very strong, so the light intensity at 10 m depth is only 10% of that of water surface, and the light intensity at 100 m deep is only 1% that of water surface. Moreover, because seawater can easily absorb long-wave light, it also causes a spectral difference in light at various water depths. Different algae have different requirements for light intensity and spectrum. Green algae generally live at the surface of water, while red algae and brown algae can use short wave light such as green, yellow and orange to live in deep water (Reynolds, 2006; Wood, 2014; Castro and Huber, 2016).

5.3.5 Effect of water chemistry

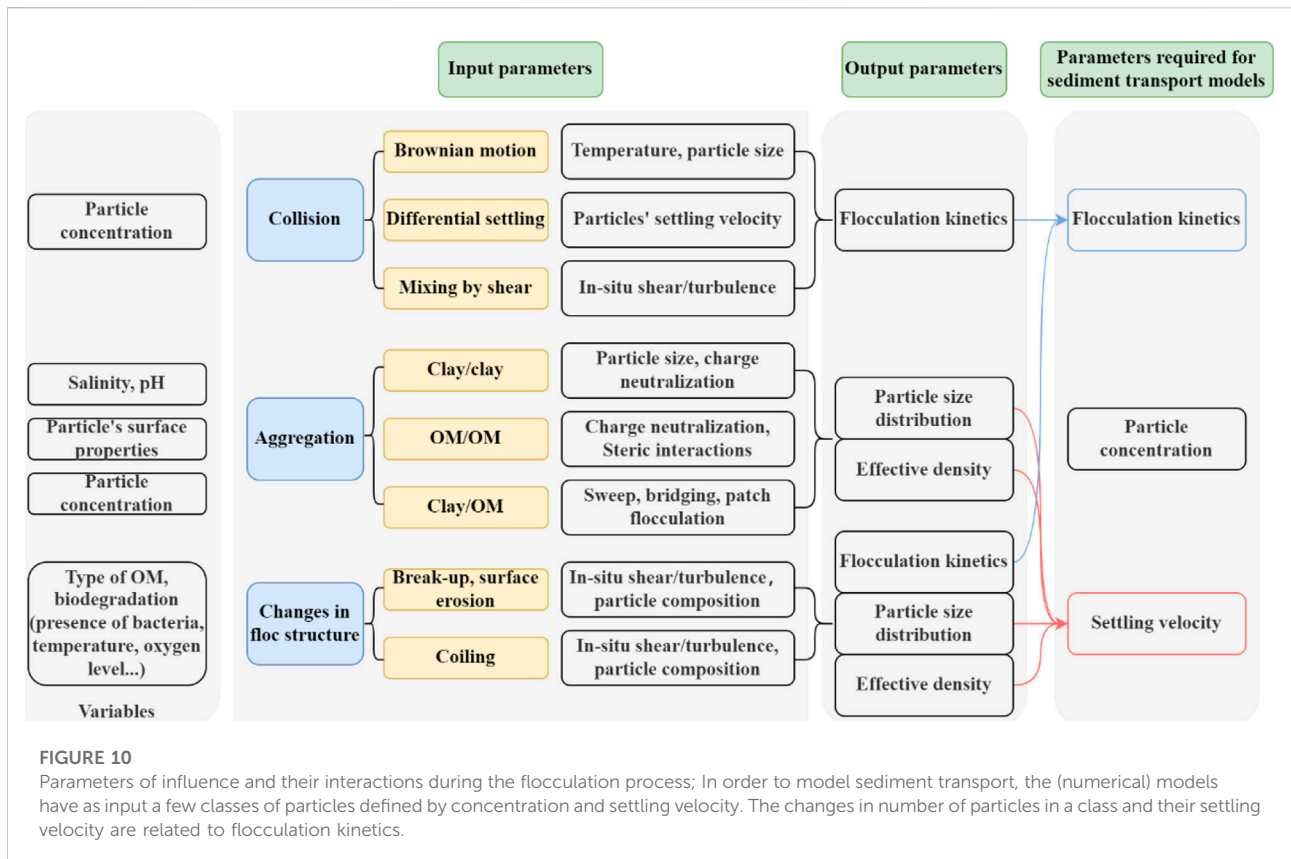
The chemical properties of the water body are also important factors for the appearance and species composition of algae (Pearson et al., 1987; Zou et al., 2011). For example, *Cyanobacteria* and *Gymnophyta* can live in eutrophic water and often form blooms; *Bacillariophyta* and *Chrysophyta* often exist in lakes with poor nutrition in mountainous areas; *Chlorophyta* and *Cryptophyta* often occur in small ponds. In

addition, the interaction between algae living in the same water area also plays an important role in their emergence and growth, as some algae can inhibit the growth of other algae by secretions.

The pH value of water has a direct impact on the physiological activities of bacteria and microorganisms on the surface charge of mineral sediment. pH can therefore significantly affect (bio)floculation by changing the EPS activity (Yokoi et al., 1995; Yokoi, 2002). With the increase of pH in a water body, the functional groups on the mineral sediment (and EPS) such as carboxyl and amino groups will dissociate or not leading to an increase or decrease in surface charge. Different bacterial species have different adaptability to pH and therefore, the effect of pH on sediment flocculation needs further study.

5.3.6 Other compositions

Other components of flocs are heavy metals, Humic substances (HS), pore water, and xenobiotic particulate matter (e.g., Microplastics (MPs), Engineered nanoparticles (ENPs), oil droplets). These elements have a unique function in floc formation (Droppo, 2001; Maggi, 2013; Ho et al., 2022). For instance, metal-rich precipitates are often found in flocs with a heterogeneous composition (Jonas and Millward, 2010). HS have strong negative charges, and, when adsorbed on suspended particles, will increase their negative charge. This leads to an enhanced stability of sediment particles (Bob and Walker, 2001; Fettweis and Lee, 2017). Pore water is present in the pore space in the floc matrices when particles aggregate to form large flocs (Droppo, 2001) which affects floc structure and density (Sherman, 1953; Logan and Hunt, 1987). MPs can become associated with SPM and become incorporated into floc matrices (Michels et al., 2018) and provide habitats and



substrates for microorganisms, which can create biofilms and scavenge various dissolved and particulate matter (Petrova and Sauer, 2012; Taylor et al., 2016). Oil droplets can easily attach to sediment particles, becoming a part of flocs and affect transport behavior (Droppo et al., 2016). With the intensification of human activities, more and more heterogeneous substances can be absorbed to part of flocs, which makes the study of floc composition and its degradation over time a worthwhile study.

6 Concluding remarks

In conclusion, flocculation of cohesive sediment mainly includes two processes: aggregation and decay [break-up by biodegradation or shear—whereby shear can also lead to a densification of flocs (without breakage)]. In these two processes, physical, chemical and biological factors will have a role and also interact with each other. These factors can either promote aggregation or decay. For example, turbulent shear stress and suspended sediment concentration can promote particle collision frequency and facilitate flocculation, but a very high shear will limit flocs growth. The effect of salinity (cation concentration) is important as well, as it enables in most cases to bind (negatively charged) sediment to (negatively charged) organic matter. Organic matter (such as EPS) will

usually enhance the stability of sediment particles, but can also degrade over time and lead to decay (break-up) of flocs.

Although many studies have been performed in the field of flocculation, floc characteristics and factors affecting sediment flocculation, there are relatively few studies on biological effects on sediment flocculation. Many authors have begun to pay attention to the study of biological factors in recent years, however, in most studies, the effects of organic matter and microbial communities are not differentiated, whereas the effects of these two factors should be distinguished in the context of sediment flocculation. Even though it is the microbial community that produces polymeric organic matter (such as EPS), two types of flocculation can be found: EPS acts on the surface properties of sediment while living microalgae aggregate themselves to form large flocs first, and then catch sediment particles within their network. A summary of the different processes involved in flocculation, and their link to the parameters required for sediment transport models are given in Figure 10.

Based on the discussions in the present article, the following points need to be studied in the future regarding the influence of organic matter on flocculation processes:

In terms of flocculation mechanisms, research has demonstrated that organic matter influences mineral sediment flocculation and flocs. Organic matter can promote flocculation but also inhibit the flocculation process. These different effects

are often related to different species of algae or different nature of microbial secretions, but a clear overview is still lacking. Most studies have focused on the exopolymers produced by microbial secretions, but overlooked the heterogeneous composition of flocs. It has been shown that algae and microorganisms themselves can form flocs and participate in the sediment flocculation process, and hence more research is needed to understand the dynamic link between microorganisms, exopolymers and mineral sediment. Finally, most of the research on the mechanisms of flocculation is still qualitative, and although some studies have been able to quantify the effects of organic matter, a fully parameterized and calibrated model is still lacking. A promising tool might be the model based on logistic growth that has recently been developed and shown to be working for laboratory studies on algae and EPS induced aggregation of mineral sediment.

In terms of floc properties, most of the current research has focused on floc size variation and less on properties such as floc composition, effective density and structural property variation. The study of these properties is necessary to understand the role of organic matter on flocculation and the time behavior of flocs (change in structure, degradation, etc.).

In terms of flocculation influencing factors, there is a large amount of research on the physical factors influencing sediment flocculation, such as the grain size distribution of sediment particles, hydrodynamics, etc., but there is still relatively little research on the combined physical and biochemical factors. Unlike parameters such as hydrodynamics, sand content and salinity, biochemical parameters, such as microalgae type and concentration are more difficult to control and quantify. Therefore their study, especially in laboratory experiments, require a multidisciplinary approach and the development of new equipment and techniques.

Finally, the large-scale transport of flocs in the overall water ecosystem are still poorly studied. Most quantitative studies are done in the laboratory but there is a need to connect these studies to numerical large-scale models. The flocculation model parametrization can be done in the lab and its implementation in large-scale transport model will help to better study the effect of microscopic, small-scale processes on large-scale sediment transport. To this end, it is also important to further develop *in-situ* monitoring tools, so as to integrate the monitoring of organic matter in the water column.

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Ecological issues are becoming increasingly important in present days societies. Sediment transport cannot anymore be seen as a purely physical process and the role of biology should be accounted for, as organic matter changes the properties of mineral sediment through flocculation. Importantly, through the interactions between mineral sediment, organic and xenobiotic particulate matter, the transport of microorganisms, nutrients and pollutants is a dynamic process in the water column that ought to be better studied. This can only be achieved through a disciplinary collaboration between researchers of the different relevant fields of expertise.

Author contributions

ZD: Writing—Original Draft, Conceptualization, Data Curation, Visualization; DH: Supervision, Writing—Review and Editing; QH: Supervision, Writing—Review and Editing; CC: Supervision, Writing—Review and Editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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