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Key Points:

- Salinity as low as 0.5 ppt facilitated aggregation of suspended mud particles into floc structures; the effect was minimal past 3–5 ppt
- The response of flocs to increased salinity was stronger in mud with organic matter; organic matter was needed for flocs larger than 100 μm
- Changes in floc growth rate and equilibrium size were modeled with a salinity-dependent aggregation efficiency parameter

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Salinity Impacts on Floc Size and Growth Rate With and Without Natural Organic Matter

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Abstract Due to the flocculation process, suspended mud aggregates carried by rivers to the coastal ocean are thought to undergo changes in size and shape in response to environmental drivers such as turbulence, sediment concentration, organic matter (OM), and salinity. Some have assumed that salt is necessary for floc formation, and that mud, therefore, reaches the estuary unflocculated. Yet mud flocs exist in freshwater systems long before the estuarine zone, likely due to the presence of OM acting as a floc-promoting binder. Therefore, it is important to consider how salinity affects flocculation, if at all, in the presence of OM. Here, we used experiments to examine the flocculation of a natural mud with and without OM. Results showed that the rate of floc growth and equilibrium size both increase with salinity regardless of the presence or absence of OM. However, the response of both to salinity was stronger when OM was present. In deionized water, natural sediment with OM was seen to produce large flocs. However, the size distribution of the suspension tended to be bimodal. With the addition of salt, increasing amounts of unflocculated material became bound within flocs, producing a more unimodal size distribution. Here, the enhancing effects of salt were noticeable at even 0.5 ppt, and increases in salinity past 3–5 ppt only marginally increased the floc growth rate and final size. Data from the experiment were used to develop a salinity-dependent model to account for changes in floc growth rate and equilibrium size.

Plain Language Summary Rivers bring a substantial amount of mud to coastal regions. Where this mud deposits is important in shaping the coastal land and nutrient dynamics. Mud particles are different from sand in that they can form aggregates known as flocs that constantly change shape and size under different conditions. As they change size, they change how fast they sink, and this influences where they deposit. Historically, salt has been assumed to create flocs and increase the mud settling rate as river sediments meet salty seawater. However, there is ample evidence that flocs exist in rivers before reaching the sea. It is possible, therefore, that flocs in estuaries are due to biological matter acting as a glue to bind mud particles together and may not be influenced by salt at all. We looked at the effects of saltwater on mud flocculation when biological matter is present and when it is absent. Our findings show that salinity increased the size of mud flocs, even more so than when organic matter was absent. However, organic matter was needed for flocs to reach sizes often found in nature. We also provided an equation to aid in the prediction of floc size under different salinities.

1. Introduction

1.1. Overview

Rivers undergo changes in flow physics, water chemistry, and organic constituents as they approach the end of their freshwater journey and transition to estuarine, and eventually, marine environments. The exact nature of the estuarine environment varies from river to river, but two key changes pertaining to the sediment transport dynamics of the system generally occur. The first is that the hydrodynamics of the system goes from being dominated by the gravitational body force and wall-bounded shear flow turbulence to a system that is also driven by tides, waves, density differences, and the initial inertia of the river upon entering the coastally influenced zone. This causes a general decrease in the turbulent energy of the fluid and its ability to keep sediment in suspension. Another generality is that the salinity in the system increases from near 0 PSU upstream of the estuarine zone to approximately 35 PSU in fully marine conditions. The spatial-temporal nature of the salt transition varies depending on the fluvial, tidal, and bathymetry boundary conditions of the estuary, but an increase in salt level must take place.

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Rivers deliver mixtures of sand and mud (clay and silts size particulate mineral and organic matter less than 63 microns in size that exhibit cohesive behavior) to estuarine zones, and the change in hydrodynamic, chemical, and organic quantities and constituents can all impact how sediment moves through the system and where sediment of different size fractions erodes or accumulates. Sand is primarily affected by hydrodynamics. Like with sand, a reduction in turbulent energy promotes the deposition of mud through the reduction in upward mixing forces on mud particles. However, mud can be influenced by the changes in hydrodynamics, salt levels, and biological processes through flocculation, which refers to continuous aggregation and breakup of muddy aggregates (Winterwerp, 1998). The reduction in turbulence can decrease the shear stress exerted on flocs and contribute to their growth. Changes in salt levels and the quantity and type of organic matter present in the water can further influence the size and density of mud flocs. Therefore, the physical, chemical, and biological conditions can all exert an influence on the settling speed of mud through the flocculation process. And while it has long been acknowledged that all three of these factors influence floc size, our ability to model floc size and settling velocity in dynamic conditions remains limited.

In this study, we examine the effects of salinity change within a turbulent suspension of varying intensity on the flocculation of mud both with and without natural organic matter. We then use the floc size and growth rate data obtained from the experiments to propose a simple model to account for the variations in salinity on a growth rate factor for flocs.

1.2. Background

Water salinity has long been deemed an influential factor in the flocculation of fine sediment (e.g., Odd, 1988). Fundamental to this understanding is the small mass of the fine sediment particles (which allows them to aggregate when close enough to each other) and the fact that the particles are often negatively charged due to the isomorphous substitution and preferential adsorption in their crystalline structure (Partheniades, 2009) or adsorption of low molecular weight (LMW) organic compounds (Tipping & Cooke, 1982; Tipping & Higgins, 1982). The negative charge causes a repulsion force that prevents the particles from getting close enough to aggregate. Increased salinity can promote flocculation by shrinking the electric double layer (EDL) that surrounds the charged clay minerals (Gregory & O'Melia, 1989). The EDL consists of a Stern layer that contains ions with a charge opposite to the clay mineral surface charge and a diffuse layer that contains free ions with a higher counterion concentration. When the ion concentration of water increases, both the Stern layer and the diffuse layer shrink, making it easier for particles to get close enough to each other for van der Waals forces to dominate and allow particles to aggregate and form flocs (Gregory & O'Melia, 1989). An extensive line of research has provided evidence for the growth-enhancing effects of salinity on the flocculation of cohesive sediment both in laboratory experiments and in field surveys.

Laboratory studies have generally reported that increased ion concentration of water leads to a growth in floc size and an associated increase in mud settling velocity. Edzwald et al. (1974) was one of the earliest attempts to quantify the effects of ion concentration in the water on suspensions of clay minerals such as kaolinite, illite, and montmorillonite. Based on their experiments, they reported enhanced aggregation for all clay suspensions with increased salinity. Further research has shown increase in floc size (e.g., Abolfazli & Strom, 2022; Guo et al., 2021; Mietta et al., 2009; Mikeš & Manning, 2010) and settling velocity (e.g., Li et al., 2021; Portela et al., 2013). While added salt is generally acknowledged to increase the size and settling velocity of clay-based flocs in the laboratory, the effects of salinity increases have also been found to have a threshold past which further increases in salt concentration have little effect on floc properties. For instance, $S = 15$ ppt was reported by Mietta et al. (2009), $S = 10$ ppt by Mikeš and Manning (2010), and $S = 7$ ppt by Guo et al. (2021) to be the concentration threshold for salinity effects.

It is difficult to isolate the effects of increased salinity on flocs in natural settings since other flocculation drivers such as turbulence, suspended sediment concentration, and the organic fraction of suspended sediment tend to co-vary along with salinity. Nevertheless, field studies have also generally described a positive, though less pronounced, relationship between salinity and floc size. In the Yangtze estuary, the largest flocs were observed in high slack water when salinity peaked and shear was lowest, while smaller flocs were found during flood and ebb tides when turbulence was stronger (Guo et al., 2017); whether the peak in floc sizes was due to changes in salinity or turbulence or was a result of larger material being advected into the sampling region is unknown. In the Ems Estuary, the largest flocs were observed in the seaward direction, where the suspended sediment concentration was not necessarily at its maximum (van Leussen, 2011). The presence of a halocline was suggested as the main driver for vertical variation in floc size in the Pearl River estuary by Y. Zhang et al. (2020). However, the authors later argued that, although increased salinity is important in enhanced fluctuation, the density stratification due to saltwater intrusion played a stronger role in modulation of the floc size distribution in the Pearl River estuary (Y. Zhang et al., 2021).

Another driver of cohesive sediment flocculation is organic matter (OM). OM is known to be a significant factor in aggregation dynamics of sediments and has been suggested to affect flocculation in freshwater (Droppo et al., 1997), estuarine (Eisma, 1986), and coastal (Fettweis et al., 2022) environments. In fact, Eisma (1986) proposed that OM was the first-order influence on floc size and that increases in salt concentration with an estuary have a minor to negligible effect. And, the laboratory experiments of Mietta et al. (2009) showed that flocs formed from mud devoid of OM were smaller compared to those formed from mud with natural OM. Due to their structural complexity, and relatively larger size compared to clay crystals, OM can modify the characteristics and behavior of clay particles in a myriad of ways. Microorganisms or a part of their structure can be bound in and become a part of floc structure (Droppo, 2001; Droppo et al., 1997). They can change the surface charge of clay particles (Beckett & Le, 1990), act as a binder for clay particles due to their complex structure (Labille et al., 2005; Theng, 2012), or grow on the particles (Shen et al., 2019; Tang & Maggi, 2018). The OM present in freshwater can have terrestrial or autochthonous origin and thereby vary seasonally (Lee et al., 2019). Additionally, despite the historical assumption that cohesive sediment is unflocculated when transported in fluvial systems, an increasing body of observational (e.g., Droppo et al., 1997; Fox et al., 2014; MacDonald & Mullarney, 2015; Osborn et al., 2021), experimental (e.g., Abolfazli & Strom, 2022), and analytical (e.g., Lamb et al., 2020; Nghiem et al., 2022) evidence support the ubiquitous presence of flocs in rivers and streams. Without the presence of measurable salinity in most cases, the explanation for the presence of flocs in freshwater systems has been the presence of OM serving as a binder for the inorganic and organic components of the flocs.

To account for the influence of drivers such as OM and salinity on mud transport, one must have a mathematical model for floc size or settling velocity that can be integrated into a larger hydrodynamic and sediment transport framework (e.g., Sherwood et al., 2018; Tarpley et al., 2019; Teeter, 2000; Verney et al., 2011; Winterwerp, 1998). Recently, there has been a push to include the impact of OM on floc population models (e.g., Shen et al., 2019). However, models that account for alterations in the floc sizes or settling velocity brought on by changes in salinity are more limited, perhaps due to the lack of data suitable for model development. The two models that include a salinity effect are the approaches by Delft3D (Deltras, 2021) and Horemans et al. (2020). In Delft3D, the salinity effect is modeled directly on the settling velocity of the mud (i.e., floc size is not modeled). The Delft3D salinity-driven model assumes that settling velocity, w_s, approaches a maximum value as salinity increases and approaches a value past which increases in salt no longer influence floc size and settling velocity, $S_{\max}$. Horemans et al. (2020) (hereafter, H20) takes a similar approach in that salinity effects are limited to a range of values below a cutoff or threshold salinity. But, rather than mapping these effects directly to the mud settling velocity, they propose a floc aggregation efficiency parameter, usually taken as a constant in floc size models, as a function of salinity. The equation for the efficiency parameter they developed was based on three data points from Edzwald et al. (1974) (hereafter referred to as E74), and using it allowed them to predict changes in floc size, and hence settling velocity, due to changes in estuarine salinity.

1.3. Study Purpose

The studies discussed above point to the significance of both ion concentration of the water and OM in the flocculation of cohesive sediments. Either can likely independently influence flocculation, and it is unclear if the independent effects are linearly additive or if there are interactions between the two that override the other or provide a joint effect. Answering this question is particularly important for modeling of mud in natural environments because both cations and OM are always present in some form. In addition, while changes in salinity are thought to drive changes in flocculation rate and equilibrium size, efforts to parameterize and model its behavior in natural environments are scarce and not well tested or calibrated.

In this study, our first goal is to test the hypothesis that the positive correlation between increased aggregation rate and equilibrium floc size and increased salinity is more pronounced when organic matter is present. That is, we expect the effect of salinity change on mud flocs to be positively modulated by organic matter. The rationale for this hypothesis is that the presence of both salt and OM affects flocculation by changing the electric charges that

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Table 1
Experiments Were Conducted Using Three Types of Sediment at Various Salinity Levels

Experiment set	Sediment type	Salinities tested [ppt]
1	Kaolinite clay	0, 2, 10
2	Natural mud (natural OM)	0, 0.5, 1, 2, 3, 5, 10
3	Treated mud (devoid of OM)	0, 2, 10

are present on the ions, clay particles, and OM molecules and the alterations they make to the inter-particle interactions. This suggests that there exists a potential interaction between the different components of flocculation, that is, sediment particles, ions, and OM. The specific questions we aim to answer in this study are: (a) is there a fundamental difference in the way natural mud flocs and those devoid of OM respond to an increase in salinity; and (b) is there a particular range of salt levels past which increases in salt in the suspension of either type of sediment no longer influence the flocculation properties? A broader question of consideration associated with our study is whether or not salt has any effect on a mud suspension if organic binders are already present and causing flocculation in freshwater rivers before they enter estuarine environments. The second goal of our study is to use data from the experiments to develop a method for including the impacts of changes in salinity within a Winterwerp (1998) type formulation for dynamic or equilibrium median floc size.

2. Materials and Methods

2.1. Approach Overview

To test our hypothesis, we measure the size distribution, equilibrium median size, and aggregation rate of mud flocs in a laboratory mixing chamber for three sets of experiments using OM-free kaolinite clay, natural sediment containing OM, and that same natural sediment treated to remove its organic content. Salinity and turbulent shear rate are varied across each of the three sets of experiments (Table 1). Data are collected with a floc camera to measure the size distribution of the particles in suspension and an optical backscatter sensor (OBS) is used to measure the turbidity. Comparisons between experiments with and without OM and across increasing salinities are used to test the hypothesis and answer the specific research questions. The turbulent shear rate during the experiments proceeds through step-downs in mixing rate to mimic the weakened turbulence conditions in a river plume, delta, or estuary. Below we present a detailed description of the mixing chamber, the camera system, the salt and sediment used in the experiments, and the experimental procedure.

2.2. Mixing Chamber and Data Acquisition Setup

The experiments were carried out in a 13 L mixing chamber (27.5 × 27.5 × 25 cm) equipped with an overhead stirrer motor connected to a paddle that allows the mixing rate and hence turbulent shear rate (G) to be adjusted within the chamber. G is a measure of the dissipation rate of turbulent kinetic energy, ϵ, and is defined as $G = \sqrt{\epsilon/\nu}$, where ν is the kinematic viscosity of water. G is a widely used parameter in modeling the flocculation process because turbulence drives both aggregation (due to increasing the likelihood of particle collision) and breakup of flocs (due to increased mechanical stress exerted on flocs) (e.g., Lee et al., 2011; Winterwerp, 1998). G was estimated using a relationship proposed by Logan (2012) based on tank and paddle geometry and paddle speed:

$$G = \left(\frac{52.3 C_D A s^3 R^3}{\nu V_T}\right)^{1/2} \tag{1}$$

where C_D is the drag coefficient, A and R are the paddle area and radius, respectively, s is the paddle speed, ν is the fluid kinematic viscosity of water, and V_T is the volume of water in the chamber. Our tank and paddle setup inevitably results in inhomogeneous turbulence with higher shear near the paddle and lower shear moving out away from the paddle. The G we report and link to the overall size distribution of flocs and particles in the tank via Equation 1 is a tank or volume-averaged G value. Therefore, in these experiments, we take the resulting size distribution of particles observed in the tank to be the integrated outcome of the various shears experienced by the particles as they are advected throughout the well-mixed tank. We use this approach partly for pragmatic reasons (it is difficult to create truly homogenous turbulence in a vessel in which the same set of particles can be tracked), and because no spatial gradients in suspended sediment concentration or suspended particle size have been observed within the tank. We speculate that floc size distributions measured in our tank at a particular G may be slightly smaller than those in a homogeneous turbulence field all else being equal because of the faster time scales associated with floc breakup in zones of higher shear.

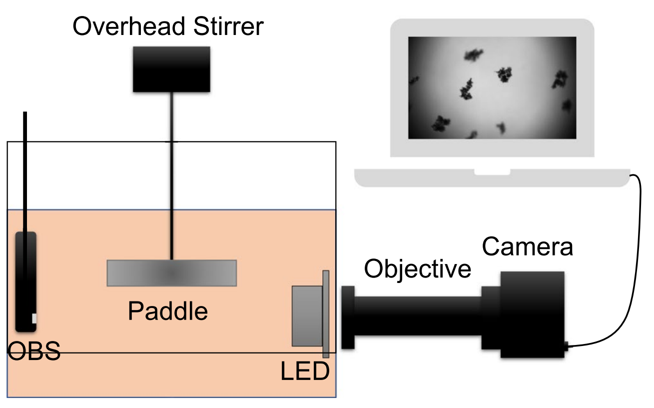

Figure 1. Schematic of the experimental setup.

The mixing tank was also equipped with a camera system that consisted of a waterproof LED light source placed inside the tank and a camera placed outside of the tank. The camera records images of particles passing through the slit created by the light source and the wall of the mixing chamber (Figure 1). The system is capable of capturing images of flocs in the size range of approximately 5–1,500 μm. Details on the mixing chamber and the camera system can be found in Tran and Strom (2017). The images captured by the camera were then organized using a Python script that was based on the procedure by Keyvani and Strom (2013) and processed in ImageJ to identify the particles present in each image. The particle, or floc, sizes were then extracted using the measured projected area of each particle as $d_f = \sqrt{4A_f/\pi}$, where d_f is the floc diameter or size, and A_f is the measured area of each floc. Associated floc volumes were calculated as: $V_f = \pi d_f^3/6$, where V_f is the floc volume. The identified particles or flocs were then sorted into log sized bins to produce floc size distributions based on floc volume and size statistics such as d_{50} (the median floc size by volume). Defining d_{50} by volume eliminates the need to assume a fractal dimension for flocs. Additionally, obtaining actual and real-time images of flocs with the camera system enabled us to visually compare the flocs formed in different conditions in addition to measuring their size (compared to the particle size and volume distribution estimation methods that are based on the principles of laser diffraction).

2.3. Salt

Previous experimental flocculation studies have used different types of salt to represent sea salt; for example, salts used include table salt (sodium chloride) (e.g., Nasser & James, 2006), commercial aquarium salts (e.g., Tan et al., 2014), and a mixture of multiple salts (e.g., Edzwald et al., 1974). While all of these salt mixtures increase the ion concentration and salinity of water, it has been shown that in addition to the concentration of salt (i.e., ion concentration), the type of salt can be quite important in setting the flocculation behavior of mud (e.g., Abolfazli & Strom, 2022). Divalent and polyvalent cations (compared to monovalent ions), for instance, are more efficient in facilitating the bonds between the sediment particles mostly due to cation bridging effects (Lai et al., 2018; Theng, 2012). To recreate saline water as close as possible to seawater, we used an American Society for Testing and Materials (ASTM) grade sea salt substitute (Lake Products Company LLC, Florissant, MO, USA) in our experiments. This salt contains nine other constituents in addition to sodium chloride including magnesium chloride and sodium sulfate. Deionized (DI) water was used as the background water in all experiments to eliminate the effects of the ions that are already present in tap water and their potential variations over the period of the study.

2.4. Sediments

Kaolinite clay, natural mud, and natural mud devoid of OM were all used in the experiments. The first set of runs was conducted using kaolinite clay (ActiveMinerals, Maryland, USA). Kaolinite has less total negative charge compared to other common minerals found in freshwater sediment (e.g., montmorillonite and illite) due to smaller isomorphous substitution that takes place in its tetrahedral or octahedral sheets. Because of this, and also due to the presence of positive charges on its edges and negative charges on its surfaces, kaolinite can create flocs even in DI water (Partheniades, 2009). Natural sediments typically consist of various inorganic and organic components. The base sediment used in the second and third sets of experiments was fine bed sediment collected from Stroubles Creek, which is a stream in southwest Virginia, USA. The sediment was wet sieved to include only the material in the clay and silt size range (<62.5 μm). During the study, the natural mud was kept in a dark fridge at 4°C. The organic matter content of the mud was measured as 11.7% based on loss on ignition. The sediment used for the third set of experiments was the same as the one used for the second set except it was treated with sodium hypochlorite following Siregar et al. (2005) to remove OM content. Experiments with kaolinite clay, which is devoid of OM, enabled us to draw a comparison between the naturally OM-free flocs and the flocs formed from the treated fine sediment. Figure 2 shows the disaggregated size distribution of the kaolinite clay and bed sediment used in the experiments. All flocculation

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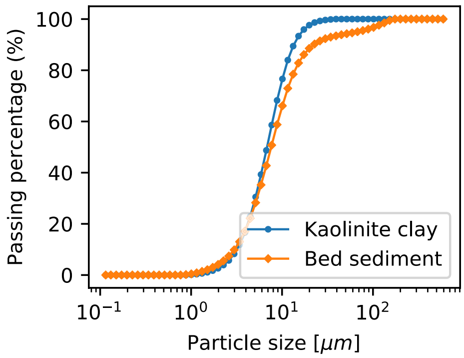

Figure 2. Disaggregated particle size distributions of kaolinite clay and bed sediment measured by the laser diffraction method.

experiments were conducted at a sediment concentration of 100 mgL^{-1}. Pre-weighed dry kaolinite clay was used for the first set of experiments. For the second and third sets, we used wet sediments, the OBS, and a calibration between NTU and the mass concentration to set the experiments to the 100 mgL^{-1}. The calibration curve was produced by filtering, drying, and weighing disaggregated particles. For both the treated and untreated natural sediments, 55 NTU was found to produce a concentration of 100 mgL^{-1}.

2.5. Experimental Procedure

In each experiment, the tank was first filled with 13 L of DI water, followed by the addition of salt (or no salt in the DI water experiments). In all three sets of experiments, the dry or wet sediment was added to 40 mL of DI water, and the mixture was sonicated for 15 min to break up any aggregates that may be present. Once the sonicated sediment was added to the tank, the experiments commenced with a 1-hr period of low turbulent shear ($G = 35\ s^{-1}$) where flocs were allowed to grow from a sonicated state. We referred to this first hour as the initial growth (IG) phase. Flocs were then broken up during a high shear (HS) phase at $G = 550\ s^{-1}$ over a period of 15 min to allow the process of flocculation to start from a more natural state (from turbulence-generated and not sonicated particles). After the HS phase, the suspension was put under five periods of descending turbulent shear rates (i.e., $G = 95$, 70, 50, 35, and 20 s^{-1}, respectively) to mimic the range of shear a river could experience as it makes its way to the sea. For the kaolinite clay, each period lasted 90 min. For the natural sediment, these periods were 150-min long as we had previously observed that natural sediment required more time compared to kaolinite clay to reach equilibrium in size after each step-down phase.

3. Results

3.1. Overview

The floc size population was extracted from the images that were captured at a frequency of 1 Hz and grouped into 1-min samples. The size distribution of each minute was then used to calculate the characteristic floc size statistics such as the d_{50} (median floc size by volume) as a function of time (Figure 3a). The d_{50} time series also provides the rate of change of the characteristic floc size as a function of G and S. Generally, flocs rapidly grew as soon as the sonicated sediment was added to the mixing chamber (Figure 3b). Flocs that form during this stage were then broken down during the high shear phase. From this point, they then increased in size again with each step-down in shear (Figure 3b). d_{50} was found to reach an equilibrium size faster at higher turbulence levels, and kaolinite reached equilibrium for a given shear rate faster than the natural mud. At the lowest shear rate ($G = 20\ s^{-1}$) using natural mud, some of the flocs grew large enough to settle out of suspension. This led

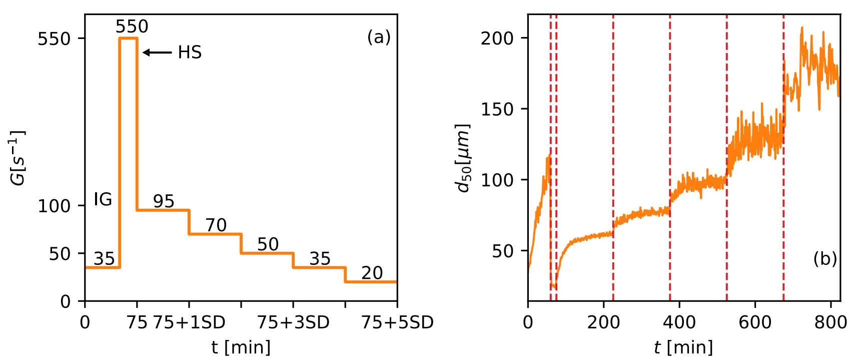

Figure 3. (a) Time series of turbulent shear rate (G) for all the experiments. The step-down (SD) duration of each phase was 90 min in the kaolinite experiments and 150 min in the natural sediment experiments; (b) floc d_{50} time series in the experiment with natural bed sediment at $S = 2$ ppt in response to variations in (G).

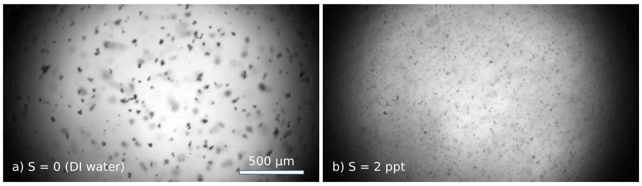

Figure 4. Snapshots of kaolinite flocs at equilibrium for a shear rate of $G = 35\ \mathrm{s}^{-1}$ in (a) deionized water and (b) $S = 2$ ppt experiments.

to a negative slope in the d_{50} time series in roughly the second half of the $G = 20\ \mathrm{s}^{-1}$ stage (Figure 3). We present the results by sediment type in the following sections.

3.2. Kaolinite

In the DI water experiment ($S = 0$ ppt), the sonicated kaolinite clay readily flocculated once it was added to the mixing chamber (Figure 4), reaching a d_{50} of about 40 μm within the 1-hr initial growth phase. Flocs were found to be weak enough to notably break up during the high shear (HS) phase of the experiment but ultimately grew back up to about 45 μm in diameter at $G = 20\ \mathrm{s}^{-1}$ by the end of the experiment (Figure 5).

The next two kaolinite experiments were conducted with added salt (Table 1). The presence of salt in the water caused the flocculation to be inhibited relative to the DI water experiment, leading to a decrease in the equilibrium floc size; in the experiment at $S = 2$ ppt, floc size increased to only 20 μm during the initial growth phase, and the d_{50} did not exceed 20 μm even at $G = 20\ \mathrm{s}^{-1}$. Although flocs were slightly larger in the experiment at $S = 10$ ppt compared to those at $S = 2$ ppt, they were still smaller compared to those in the DI water experiment at all turbulence levels (Figure 5).

The flocs' response to reduced turbulence during each step-down phase occurred quickly. New equilibrium sizes were reached at each new shear level within 20 min of the change in shear. To quantify their growth rate, we calculated $\Delta d_{50}/\Delta t$, which is the change of d_{50} over a specified duration of time, Δt (15 min for the kaolinite experiments), immediately after G was reduced at each step-down phase. The response was stronger in the DI water experiment, in which d_{50} increased at a rate of 0.2–0.4 μm min^{-1} at $G > 35\ \mathrm{s}^{-1}$ and more than 0.5 μm min^{-1} at $G = 20\ \mathrm{s}^{-1}$ (Figure 5). In comparison, in the salt experiments, d_{50} increased by only a few μm at each step-down with $\Delta d_{50}/\Delta t$ never exceeding 0.2 μm min^{-1} at any turbulence level.

3.3. Bed Sediment With Natural OM

Similar to the kaolinite experiments, we first examined the flocculation behavior of the natural bed sediment without the addition of salt using DI water ($S = 0$ ppt). In the absence of added salt, two distinct groups of suspended particles were observed across all shear rates. The larger of these two groups consisted of flocs in the >200 μm size range. The number of particles and total sediment volume in this larger group was, however, strongly outnumbered by particles in the second group, that is, the smaller un- or less-flocculated particles (Figure 6a).

Six salt experiments were conducted at salinities ranging from $S = 0.5$–10 ppt (Table 1). A key takeaway from these experiments is that it only took a very small increase in salinity to strongly enhanced the flocculation of natural mud such that at $S = 0.5$ ppt, the number of flocs were noticeably greater at all of the turbulence levels compared to the DI water experiments (Figure 6b).

The increase in d_{50} in response to increased salinity can be traced back to the aggregation of unaggregated particles and smaller flocs mostly in the 25–75 μm size range. That is, as salinity increased, the unaggregated particles became more and more integrated into the floc structure. The loss of fine material is evident in the images as the

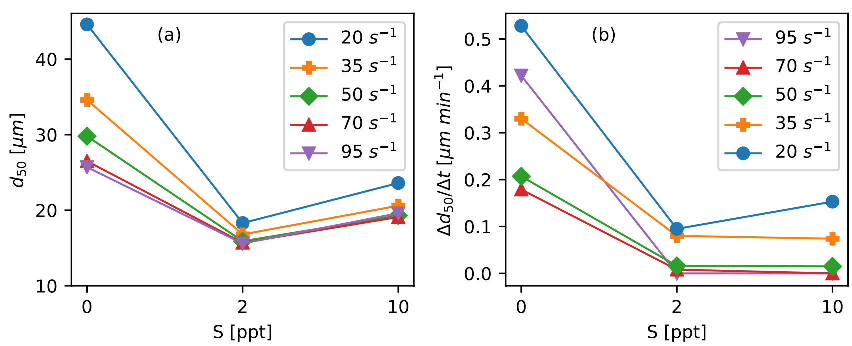

Figure 5. Kaolinite experiments: (a) floc size at equilibrium (d_{50}) by salinity at different turbulent shear rates; (b) rate of change of d_{50} in the first 15 min following each step-down phase by salinity at different turbulent shear rates.

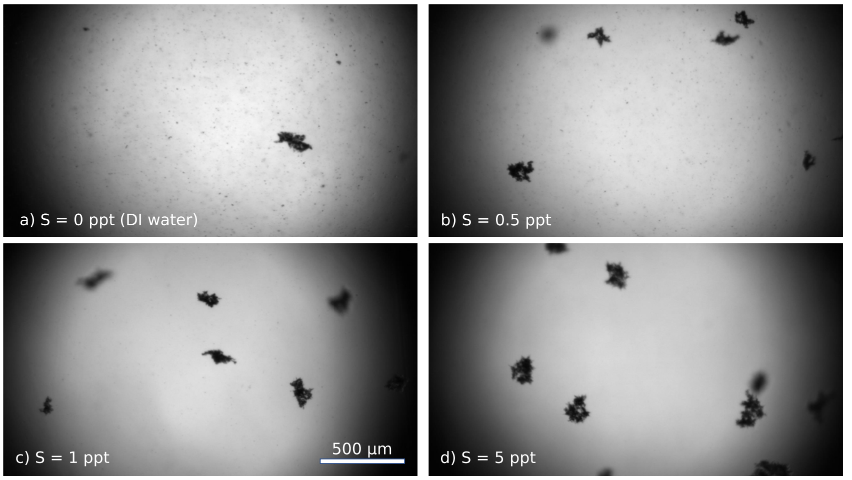

Figure 6. Snapshots of bed sediment flocs at equilibrium for a shear rate of $G = 35\ \mathrm{s}^{-1}$ at different salinities.

background of small dispersed particles in DI water reduces with salinity (Figure 6). The change in floc population in response to increased salinity is also evident in the particle size distribution (PSD) (Figure 7). While some relatively large flocs with the size of >200 μm did form in DI water, the fraction of discrete unaggregated particles was notably larger than those in the experiment at $S = 0.5$ ppt. The PSD moved toward larger particle sizes as salinity increased, leading to a larger d_{50}.

The equilibrium floc size (d_{50}) increased notably from the DI water experiment to $S = 0.5$ ppt, and then to 1, 2, and 3 ppt. These enhancing effects of increased salinity, however, were found to be most evident at $S \leq 3$ ppt, and a further increase in salinity from 3 to 5 ppt and then to 10 ppt did not result in notable increases in d_{50} (Figure 8). The stronger effects of increased salinity at lower salinity levels were observed at all shear levels.

Salinity affected not only the equilibrium floc sizes but also the growth rate of flocs in response to each reduction in turbulent shear rate (Figure 8b). While in DI water the equilibrium floc size increased at a very slow pace (or decreased as some larger flocs with diameters of roughly >250 μm settled out of the suspension), the presence of salt at the level of only 0.5 ppt in water increased the flocculation rate by as much as 2 μm min^{-1} at $G = 20\ \mathrm{s}^{-1}$. Similar to the equilibrium floc size, the enhancing effects of salinity on flocculation rate had a threshold, and the strongest effects were found at $S \leq 5$ ppt.

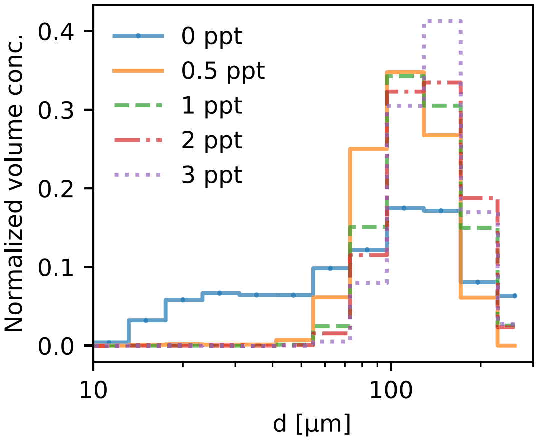

Figure 7. Normalized volume concentration distributions at equilibrium for a shear rate of $G = 35\ \mathrm{s}^{-1}$.

3.4. Treated Bed Sediment With OM Removed

Mud floc size and structure were clearly different when the treated sediment was used in the flocculation experiments. Interestingly, in the experiment with no added salt (DI water), the treated bed sediment devoid of OM did not form any visible flocs, and the suspended particles retained their sonicated dispersed form (Figure 9a).

It was only in the presence of salt that treated sediment started to aggregate and form flocs. In the experiment at $S = 2$ ppt, small flocs with the size of <20 μm formed immediately after the sonicated sediment was added to the mixing tank. However, d_{50} of the flocs never exceeded 30 μm during any of the shear step-downs (Figure 10). Similar behavior was observed in the experiment with $S = 10$ ppt. Although the size and growth rate of flocs were slightly greater compared to those in the experiment with $S = 2$ ppt, their d_{50} hardly reached 35 μm even at $G = 20\ \mathrm{s}^{-1}$.

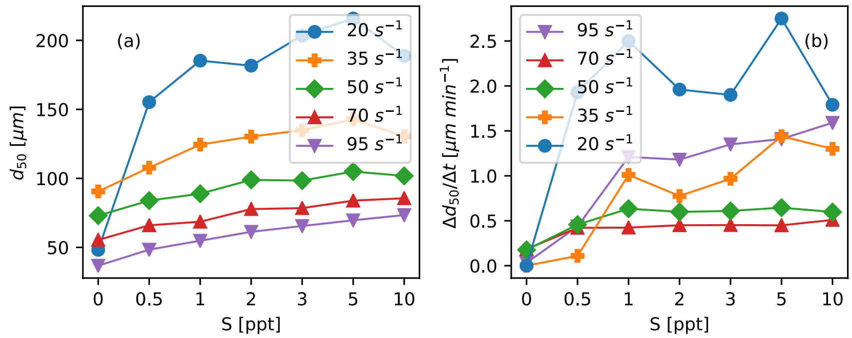

Figure 8. Mud experiments: (a) floc size at equilibrium (d_{50}) by salinity at different turbulent shear rates; (b) rate of change of d_{50} in the first 20 min following each step-down phase by salinity at different turbulent shear rates.

4. Discussion

4.1. Organic Matter and Flocculation of Natural Mud

Gums such as xanthan gum, guar gum, and chitosan are often used in laboratory experiments looking to study the role of organic material on flocculation (e.g., Furukawa et al., 2014; G. Zhang et al., 2013; Zeichner et al., 2021). In this study, we used an alternative approach by removing OM from natural sediment rather than adding gum to the suspension.

Our experiments revealed a stark difference between flocs formed from natural unaltered bed sediment (which contains OM) and flocs that form from suspensions of the same sediment after it has been treated to remove the organics (OM-free). Figure 12 visually highlights this distinction. In the images, salinity, shear rate, and base inorganic sediment components are identical in panels (a) and (b). The only item that is different is the OM content. Untreated bed sediment formed larger flocs, where most mud particles were incorporated into the floc structure. The incorporation of discrete particles caused the background to be clearer, resulting in more defined and darker flocs in the images due to their higher contrast with the clearer background. In comparison, flocs were smaller in the treated sediment experiment. The difference we observed in floc size is greater than that reported by Mietta et al. (2009), where the increase in floc size from OM-devoid mud to OM-containing mud was limited to <30 μm at $G = 35$ s^{-1}. The flocs formed from the OM-free mud were also more transparent and fragile than the OM-containing flocs. These differences between the sediment that contained OM and that devoid of OM were present at all salinity and turbulence levels that were tested.

The strong flocculation-enhancing role of the OM that is naturally present in freshwater sediment has been attributed to the structure and weight of OM molecules. Organic biopolymers, such as the extracellular polymeric substances (EPSs), contain numerous charged or uncharged groups in their structure, all contributing to the forces between the organic and inorganic components of the mud (Lai et al., 2018). The uncharged groups of biopolymers can interact with the negatively charged surface of clay minerals through the van der Waals forces. Hydrogen bonds can occur between the basal hydroxyl surface of silicates such as kaolinite and polar groups of biopolymers (Theng, 2012). Cationic groups of the polymers can further interact with clay mineral surfaces through electrostatic forces, while the anionic groups can attach to negatively charged surfaces of clay minerals through polyvalent cations acting as a bridge between the two (Philippe & Schaumann, 2014). All of these interactions can lead to a complex looped structure of OM-sediment bonds in the floc structure (Figure 11). The role of OM in flocculation is not limited to forming the general size and shape of flocs as a whole. The presence of OM can also alter the nature of the primary particles, that is, those particles that are the base-level building blocks of flocs. At this level, sediment particles and OM can firmly bind, thereby increasing the size of these building blocks (Fall et al., 2021). When OM is absent from the sediment, flocs form by absorption of differently charged sections of mineral structure (such as positively charged edges and negatively charged faces of kaolinite) (Partheniades, 2009) or due to polyvalent cation bridging between

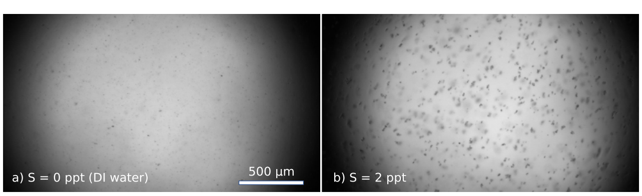

Figure 9. Snapshots of treated sediment flocs at equilibrium for a shear rate of $G = 35$ s^{-1} in (a) deionized water and (b) $S = 2$ ppt experiments.

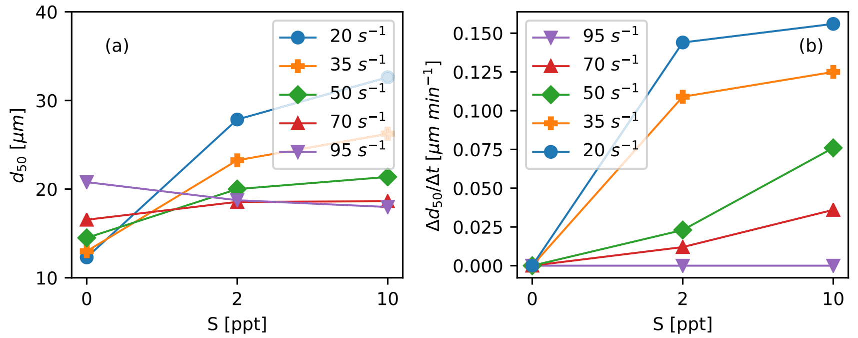

Figure 10. Treated mud experiments: (a) floc size at equilibrium (d_{50}) by salinity at different turbulent shear rates; (b) rate of change of d_{50} in the first 15 min following each step-down phase by salinity at different turbulent shear rates.

two negatively charged clay particles (Mietta et al., 2009) or shrinking of the EDL and the resulting net attractive van der Waals forces (Spielman, 1978). In all of these cases, the structure of the flocs is different from the flocs that contain OM (Figure 12).

4.2. Salinity and Flocculation

Salinity has historically been considered a driving factor in the flocculation of mud deposition in estuarine zones (e.g., Odd, 1988). Our results support the idea that the presence of salt increases both the rate of aggregation and the equilibrium size of mud flocs formed from fluvial bed sediment. While this general trend or principle is in line with historic understanding, the experiments presented here offer three more refined points of consideration to this general trend and observation regarding salt and its role in the flocculation of natural mud.

First, it appears to us that the primary role of salt is to make it easier for unaggregated small particles to flocculate with existing flocs or each other. Our experiments did not show a simple relationship whereby flocs do not form in freshwater and then do form when salt is added. For the untreated natural bed sediment, we still observed the formation of flocs even without the addition of any salt, that is, flocs were present in the pure DI water experiment (Figure 6a). However, a notable fraction of the suspended matter did remain in a discrete, unaggregated state, and the presence of flocs and unaggregated material resulted in a bimodal PSD (Figure 7). We expect that the existence of flocs with a diameter of >200 μm was most likely driven by OM, particularly macromolecules that contain multiple charges that act as a binder for sediment particles because identical experiments with the treated bed sediment (OM removed) in DI water did not produce any flocs (Figure 9). The bimodal size distribution of the suspended material for the $S = 0$ ppt experiment was likely the reason for the drop in equilibrium d_{50} at $G = 20\ \mathrm{s}^{-1}$ due to settling of a fraction of the largest particles (Figure 8a), which also resulted in negligible growth rate at this shear rate (Figure 8b). Considering the role of salt on the PSD then, the primary role of the salt in the natural sediment case was to change the PSD from a bimodal (unflocculated and flocculated) distribution to one that is more uni-modal (flocculated); or to change the fraction of material in suspension that resides in larger flocs.

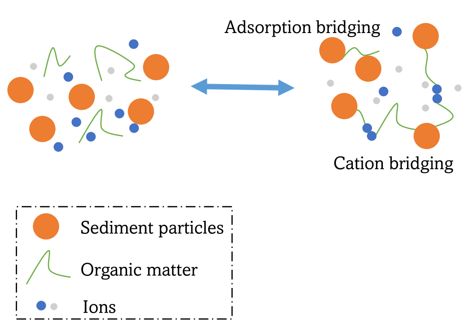

Figure 11. A schematic of the sediment-organic matter interaction mechanisms. Adsorption bridging occurs when free biopolymers capture suspended clay particles. Cation bridging refers to the aggregation of organic/inorganic particles through free cations.

Second, the PSD for the natural bed sediment with OM had a stronger response to increases in salt than the PSD for the bed sediment without OM. That is, the presence of OM did not override or remove a flocculation response to changes in salinity. Instead, it enhanced them. Without OM present, floc sizes increased in response to an increase in salinity. But the change in size, as represented by d_{50}, was limited to a change of approximately 10–15 μm from $S = 0$ ppt to $S = 10$ ppt (Figure 10); translated to settling velocity of approximately 0.03 mms^{-1} difference. In contrast, flocs formed with the untreated bed sediment experienced a change in d_{50} of 50–150+ μm with the addition of salt depending on shear rate (Figure 8); a change of almost 0.5 mms^{-1} in terms of settling velocity for $G = 35\ \mathrm{s}^{-1}$. We suspect that increasing salinity had less of an effect on the treated sediment because when OM is removed, the electrically charged points of contact that can cause

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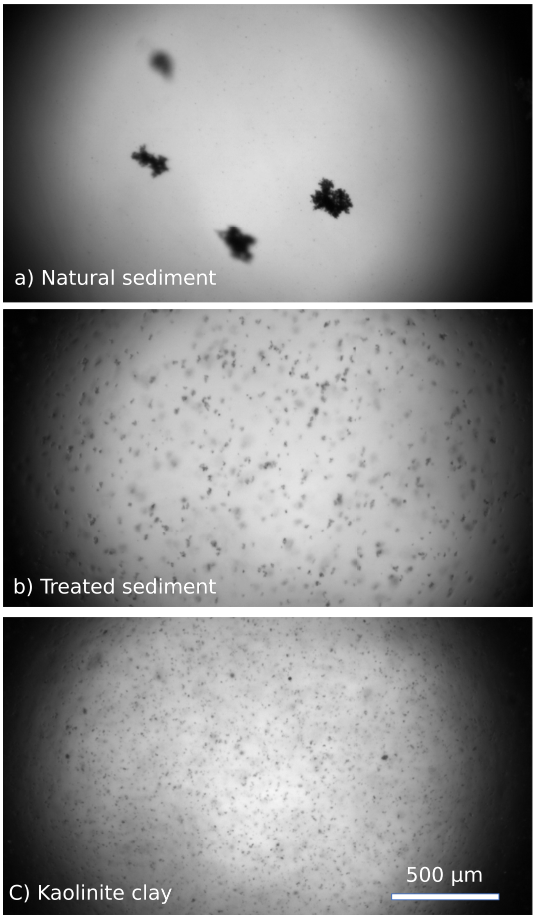

Figure 12. Snapshots of flocs formed from (a) natural bed sediment, (b) treated bed sediment, and (c) kaolinite clay all at equilibrium and $S = 2$ ppt, $G = 35\ \mathrm{s}^{-1}$, and $C = 100\ \mathrm{mgL}^{-1}$.

aggregation of particles are limited to the charged sediment surfaces, with the complex structures of biopolymers being no longer present (Theng, 2012).

Third, very small changes in initial salinity, going from 0 to 0.5 ppt, exerted a marked influence on both the equilibrium PSD and rate of growth of the suspended natural bed sediment (Figures 7 and 8). Relative increases past $S = 3$ ppt had little influence on either the equilibrium PSD or growth rate. The lack of response in the growth rate or equilibrium PSD after $S = 3$ ppt suggests that at this salinity, there are already enough ions to effectively shrink the EDL and promote flocculation. A limit to the flocculation-enhancing effects of increased ion concentration of water have also been reported previously in the literature, for instance $S = 10$ ppt (Mikeš & Manning, 2010), 15 ppt (Mikeš & Manning, 2010), or 7 ppt (Guo et al., 2021). This threshold concept has also been implemented in computational models. For instance, Delft3D assumes settling velocity, w_s, approaches $w_{s,\max}$ as salinity approaches $S_{\max}$ (Deltras, 2021), while Horemans et al. (2020) assumes that aggregation parameter of flocs, k'_A reaches a maximum as S approaches a prescribed maximum.

While the above points are about the natural mud that contained OM, we have to make two comments about the sediment samples that did not contain OM and behaved in some aspect differently than natural mud. With kaolinite, the smaller floc sizes in the salt experiments compared to the DI experiment are likely due to the differences in floc structures in these two cases. In DI water, kaolinite particles form random particle-to-particle bonds via the van der Waals forces. In the absence of ions, on the other hand, the positively charged edges and the negatively charged faces bond through electrostatic forces (Partheniades, 2009). A relatively high floc growth rate was observed at $G = 95\ \mathrm{s}^{-1}$ as the weak kalonite flocs transitioned from a purely disaggregated form (at $G = 550\ \mathrm{s}^{-1}$) to flocculated at $G = 95\ \mathrm{s}^{-1}$ (Figure 5b). With treated mud, the reversal of effect of G on d_{50} between $S = 0$ and 2 ppt is likely due to the absence of flocculation at $S = 0$ ppt (Figure 10). At $S = 0$ ppt, instead of breaking up the flocs, greater G kept the silt particles larger than 20 microns in suspension. On the other hand, at $S = 2$ ppt, at which flocculation did occur, higher shear led to break-up of flocs and, consequently, smaller particle size. This was most likely also the case with the untreated mud but was obscured by flocs larger than 100 µm.

One of the broader questions we wanted to consider with our study was whether or not salt has any effect on a mud suspension if organic binders are already present and causing flocculation in freshwater rivers. While the experiments do not definitively answer this question, we do believe that the experiments offer a few points of consideration that we then use to speculate on flocculation dynamics in natural rivers.

The experiments show that the presence of organics increases the response of the mud suspension PSD to the presence of salt. They also show that flocs can form without the presence of added salt and that the flocculation-promoting elements of added salt are strongest at very low salinities. Clearly, rivers carry different types and amounts of organic matter. And it has been amply shown that mud in fluvial settings is flocculated (e.g., Droppo, 2001; Droppo et al., 1997; Osborn et al., 2021). Our experiments in DI water showed that it is possible for flocs to form in pure DI water in the presence of natural organic matter. However, the fraction of the mud that was contained in flocs was low until some salt ions were added. For at least the case of Osborn et al. (2021) (and other unpublished data we have in different rivers in the USA), the flocculation state of the mud upstream of any measurable saltwater intrusion has tended to be larger than what we observed in the DI water case in the experiments presented in Section 3.3. In fact, using similar natural bed sediment, Abolfazli and Strom (2022) found a great degree of flocculation when using natural stream water compared to the DI water used for the $S = 0$ ppt experiment presented in this study. Together these observations suggest that the ions present in natural stream water, upstream of any saltwater intrusion, can sufficiently aid in promoting flocculation when organic matter

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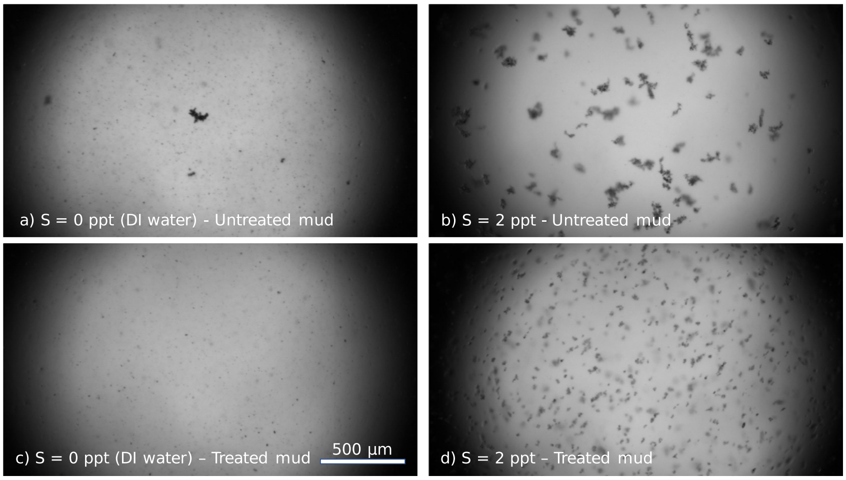

Figure 13. Snapshots of flocs formed from Mississippi River mud at equilibrium for a shear rate of $G = 35\ s^{-1}$ and concentration of $C = 100\ mgL^{-1}$.

is present, even when the overall conductivity of the water is still low relative to marine or estuarine standards. We, therefore, expect that flocs in rivers are governed not just by the organic matter, but also by other ions, salt or otherwise, that may be present. As a result, one can imagine a case where the organic matter and background ions, that is, non-saltwater-intrusion-related ions, are sufficient to promote the flocculation of a large fraction of the mud load in freshwater systems. Additionally, our findings suggest the effects of salinity on modulating the fraction of mud flocculated or the overall floc PSD to be limited to the head of a well-mixed estuary or to the fresh-saltwater interface in a vertically stratified salt wedge system.

As a final note, given the complex interaction between clay minerals, OM, and ions, we speculate that the order in which OM and ions are added to the clay affects the flocculation process. In the present study, we added salt to mud that already contained OM to mimic the increase in ion concentration as freshwater mud experiences higher levels of salinity as it approaches estuaries and oceans or experiences a spike in ion concentration due to deicing salt runoff (Abolfazli & Strom, 2022).

4.3. How Applicable Are These Findings to Coastal Mud?

Our experiments were conducted using natural mud gathered from a local stream in the Valley and Ridge province of Virginia, that is, Stroubles Creek. Stroubles Creek is a tributary to the New River, which in turn is a tributary to the Mississippi River. Sediment found in the creek can ultimately make its way to the Gulf Coast region, and spikes in salinity do occur in the creek following runoff of roadway deicing salts during winter (Abolfazli & Strom, 2022; Lakoba et al., 2021). Therefore exploring the role of organic matter and salinity on the flocculation of Stroubles Creek mud specifically has utility for understanding its transport dynamics. Nevertheless, it is reasonable to question how broadly applicable the experimental results are to other muds—and in particular, muds that are more proximally located to coastal zones where changes in salinity are more eminent.

As one step toward more broadly testing the response of different muds to changes in salinity both with and without natural organic material, we conducted experiments similar to those outlined in the methods and results for the Stroubles Creek mud using mud obtained from the bed of the main channel of the Mississippi River near Venice, LA. The mud was collected as part of a larger project in January 2021 using a Shipek grab sampler. Tests were run on the treated (OM removed) and natural (no removal of OM) muds in DI water and DI water with enough salt added to bring the salinity to 2 ppt. Images of the suspension are shown in Figure 13. Images and floc size measurements indicate that the general behavior concerning salts and organic matter for the Mississippi River mud was the same as that of the Stroubles Creek mud. The unaltered mud in DI water produced a bimodal distribution with a few larger flocs and many smaller aggregates and unflocculated particles (Figure 13a). Adding salt resulted in a much higher degree of flocculation, a more unimodal distribution, and an overall larger median size (Figure 13b). Without OM (i.e., for the treated case) no large flocs formed in DI water at $S = 0$ ppt (Figure 13c). The addition of salt led to a more flocculated state, but floc sizes were smaller than those produced at the same shear and salinity level but with the presence of OM (Figure 13d). The primary

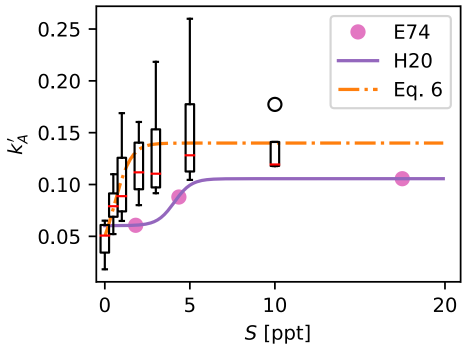

Figure 14. A comparison between our model for k'_A (Equation 6 with $k'_{A0} = 0.05$) and that of Horemans et al. (2020) (H20). Boxplots show the spread of our back-calculated k'_A over G values at each salinity. E74 depicts the data from Edzwald et al. (1974).

difference that we observed between the Mississippi River mud flocs and the Stroubles Creek mud flocs was that the Stroubles Creek mud flocs were, on average, larger and optically denser than the Mississippi River mud flocs for the OM tests at 2 ppt. We expect that this is due to higher levels of organic matter present in the fresh Stroubles Creek mud relative to the older Mississippi River sample.

4.4. Modeling the Influence of Salinity on Floc Size

Efforts to include salinity as a driver of mud flocculation using simple models have been made before. However, these models have been developed with sparse data in the less than 5 ppt salinity range (e.g., Deltras, 2021; Horemans et al., 2020). Here we examine the model of Horemans et al. (2020) using data from the natural mud runs in our experiment, and we also propose our own model for a salinity-dependent aggregation efficiency parameter within the Winterwerp (1998) modeling framework.

At equilibrium, the Lagrangian form of the Winterwerp (1998) floc size equation yields the following expression:

$$d_{50} = d_p + \frac{k_A C}{k_B \sqrt{G}} \tag{2}$$

where d_p is the disaggregated primary or constituent particle size, C is the mass concentration of sediment, and k_A and k_B are the aggregation and breakup coefficients defined as:

$$k_A = \frac{k'_A}{n_f} \frac{d_p^{n_f - 3}}{\rho_s} \tag{3}$$

and,

$$k_B = \frac{k'_B}{n_f} d_p^{-p} \left(\frac{\mu}{F_y} \right)^q \tag{4}$$

In Equations 3 and 4, n_f is the fractal dimension of the flocs, ρ_s density of the dry unflocculated sediment, μ is the dynamic viscosity of the water, F_y is the yield strength of the flocs, k'_A and k'_B are dimensionless aggregation and breakup efficiency coefficients, and p and q are model parameters. Through a scaling argument, p is typically taken to be $p = 3 - n_f$ (Kuprenas et al., 2018; Winterwerp, 1998). Following further scaling arguments, based on settling tests in stagnant water columns, Winterwerp (1998) took $q = 0.5$. Here we use the reasoning of Kuprenas et al. (2018) and set q to be a simple function of the size of the flocs relative to the Kolmogorov microscale, $\eta = \sqrt{G/\nu}$:

$$q = c_1 + c_2 \frac{d_{50}}{\eta} \tag{5}$$

where c_1 and c_2 are constant coefficients. The proposed formulation ensures k_B increases as d_{50} approaches η. In calculations, we used meters, kilograms, and seconds.

Similar to Horemans et al. (2020), we expect that one way to capture the influence of salt on the growth rate and equilibrium size of mud flocs is to seek out a relationship for the aggregation efficiency parameter, k'_A, expressed as a function of S in ppt. To do this, we back-calculated values of k'_A in Equation 3 using Equations 2, 4, and 5 and measured or known floc sizes, d_{50}, primary particles, $d_p = 8$ μm, shear rates, G, and sediment concentration, $C = 0.1$ gL^{-1}. For the calculations, we also used the following set of model parameters: $n_f = 2$, $\rho_s = 2{,}500$ kgm^{-3} (slightly smaller than the typical 2,650 kgm^{-3} for clay to account for the organic matter present in the mud), $\mu = 1 \times 10^{-3}$ NSm^{-2}, $F_y = 10^{-10}$ N, $c_1 = c_2 = 0.5$, and $k'_B = 1.16 \times 10^{-6}$ (Kuprenas et al., 2018).

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Our back-calculated k'_A increased with S (the values obtained are represented by boxplots in Figure 14). Following Horemans et al. (2020), we averaged all calculated k'_A for each salinity and then fit a hyperbolic tangent function through the average. The result of this process yielded:

$$\frac{k'_A}{k'_{A0}} = 0.4 + 1.2[1 + \tanh(S - 0.55)] \quad (6)$$

where $k'_{A0} = 0.05$ denotes k'_A at $S = 0$ ppt Equation 6 has a total residual sum of squares (RSS) of 0.064 and a $R^2 = 0.73$ when using the average k'_A for each S.

Overall, our data suggest a similar functional shape in the relationship between k'_A and S as that Horemans et al. (2020) and the Delft3D models. However, our data show a slight increase in the response of k'_A to increasing S and one that occurs at a lower salinity. For example, the change in k'_A with S in our case all occurs before a change in k'_A is predicted by the Horemans et al. (2020) model (Figure 14).

While our aim was to provide a functionality between k'_A and S, the data did show that k'_A was also affected by G. In the analysis, we present here we have grouped all of the variations with G at a single salinity value (reflective in the boxplots in Figure 14) into an average. However, we did observe a systematic increase in k'_A with a decrease in G. It should also be noted that it is possible that the equilibrium floc sizes are underestimated at the lowest shear rate in our experiments (i.e., $G = 20\ s^{-1}$) due to the settling of a small fraction of the flocs. The model presented in Equation 6 is based on the results of our experiments. How generic the equation is remains unknown. We expect that the form of the equation will capture the general behavior of mud flocs in the presence of increasing salinity and that scaling the equation by the zero-salinity aggregation efficiency will aid in making it extendable to other suspensions of very sediment and organic content. However, it is possible that the constant coefficients (i.e., 0.4, 1.2, and 0.55) may need to be adjusted to capture a given mud's behavior most accurately.

With the natural mud used in our experiment, most of the change in floc size and growth rate occurred at salinities below 5 ppt with the positive effects of salinity on enhancing k'_A being evident even at salinities as low as 0.5 ppt. While the model predicts a reversible response with S, one would expect the physical process to not be perfectly reversible. Additional data are needed with other natural sediment mixtures to better understand the level of generality that our data and Equation 6 reflect.

5. Conclusion

Our experiments reveal a strong interaction between organic matter and salinity in driving the flocculation dynamic of natural mud. Moving from DI water to salinities of 10 ppt increased the equilibrium floc size and growth rate of flocs formed in natural sediment both with and without natural organic matter. Without organic matter, flocs were limited in size to 20–30 μm even at 10 ppt. With natural OM present, a limited number of large flocs formed even in pure DI water. However, the fraction of material flocculated remained low, resulting in a bimodal suspended particle size distribution. Adding just a small amount of salt to bring salinity to 0.5 ppt notably increased the fraction of flocculated material and changed the particle size distribution from bimodal to unimodal. Increases in salt up to 3–5 ppt decreased the number of unflocculated particles further and increased floc sizes such that the average reached between 100 and 200 μm depending on the shear level. Increases in salinity past this level led to only marginal increases in floc size for a given shear rate. The data suggest that both organic material and ions associated with river or estuarine water are likely present if high degrees of flocculation, with flocs on the order of 100 μm and larger, are observed. The data also suggest that very low levels of salinity (e.g., 0.5 ppt) are needed to enhance floc size. It is therefore likely that large salinity-driven changes to the floc size distribution in nature are limited to the head of a well-mixed estuary or to the fresh-saltwater interface in a vertically stratified salt wedge system in rivers with low background freshwater ion concentration. Change in floc growth and equilibrium size brought on by salinity changes in the presence of natural mud and OM can be captured with a salinity-dependent aggregation efficiency parameter in a Winterwerp (1998) type formulation.

Data Availability Statement

Data associated with the paper are available in Abolfazli and Strom (2023).

Acknowledgments
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