Influence of Heavy Metals and Non-Cohesive Sediments on the Flocculation Behavior of Kaolin

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ABSTRACT

Flocculation of cohesive sediments are affected by various physical, chemical and biological factors. The present study investigated the effect of heavy metals on the flocculation of cohesive sediments and its resistance to breakage under the shear imparted by sand. Experimental investigations were conducted in a flocculator with varying speed agitator for various concentrations of Fe (II) (0mg/L, 0.1mg/L, 0.5mg/L, 1mg/L, 5mg/L, 10mg/L and 20mg/L) and sand (0%,10%,30%,40% and 50%) under different salinity conditions (0g/L, 15g/L and 30g/L). The floc characteristics were obtained using an image capturing and processing technique. The experimental results indicated that the addition of Fe (II) contributes to the floc formation as a result of aggregation of flocs, whereas the addition of sand to the Fe-kaolin suspension contributed to the breakup of flocs. The floc volume fraction of macroflocs were observed to increase with the increase in metal concentration. The increase in sand concentration led to an increase in the floc volume fraction of microflocs. Further the behavior of breakage coefficient of flocs on the addition of sand were analysed. It was observed that the breakage coefficient decreased with the increase in sand concentration. Further it was noticed that the flocs formed at higher Fe (II) concentration at high salinities possess higher breakage coefficient.

Keywords: Flocculation, Heavy metals, Non-cohesive sediments

1 Introduction

Estuaries are the zones where the sea water constantly mixes with the fresh water. Various phenomena of transportation, flocculation, settling and resuspension takes place within estuaries. Flocculation of cohesive sediments is aided by various physical, chemical and biological factors. Turbulence, gravitational circulation, suspended sediment concentration are some of the physical factors, while the chemical factors include toxic heavy metals, pH, salinity etc. Organic matter, aquatic flora, and fauna are among the biological factors that contribute to the cohesive sediment flocculation. Heavy metals are one of the major toxic constituent in the estuarine environment which affects the aquatic ecosystem [1]–[3]. The heavy metals get adsorbed to the sediment surface at suitable environmental conditions. The adsorption behaviour of heavy metals is greatly affected by their phase change [4], [5]. In estuaries, flocculation may facilitate the removal of heavy metals through adsorption and thereby settling [6]. Removal of heavy metals were more prominent at high tide and during estuarine mixing. More removal rate of heavy metals was also observed at high salinity reaches. Higher salinity conditions provide suitable environment for the metals to get adsorbed to the sediment and thereby enhancing settling. Even though the metals are said to be effective coagulants, how they affect the flocculation of cohesive sediments in estuarine mixing is unknown. Recent studies have investigated the effects of non-cohesive sediments on the flocculation of cohesive sediments [7]–[10]. It has been reported that presence of sand fraction retards the initial floc formation due to aggregation, [10]. Sand shear contributes to the breakup of cohesive sediment flocs. The resistance of flocs to breakage is generally quantified by breakage coefficient. The general practice adopted in population balance modelling is to assume binary breakage wherein the flocs are assumed to break into two equal parts thereby adopting

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a value of 0.5 for breakage coefficient. Previous studies indicated that the value of breakage coefficient can vary from 0 to 1 depending on the conditions at which flocculation takes place [11]. However, the breakage of kaolin flocs under the influence of Fe (II) is not known. Thus, the present study mainly aims at investigating the effect of sand fraction on the breakage coefficient of metal-kaolin flocs.

2 Materials and Methods

The mixing experiments were carried out in a jar test apparatus with six varying speed agitators. Samples were prepared in 1000 mL beakers with 1 g/L kaolin and different concentrations of Fe (II) (0 mg/L, 0.1 mg/L, 0.5 mg/L, 1 mg/L, 5 mg/L, 10 mg/L and 20 mg/L) and sand (0 %,10 %,30 %,40 % and 50 %) at different salinities (0 g/L, 15 g/L and 30 g/L). Initially the samples were mixed at 100 rpm for two minutes to ensure uniform mixing. Further the samples were flocculated at 10-20 rpm to promote floc formation for 10 minutes. Further 2mL of the liquid was pipetted out and transferred to a cuvette for the purpose of capturing and processing images. The samples were then agitated for five minutes at the resuspension rpm of sand ($52s^{-1}$), followed by addition of sand and agitation for 5 minutes. Another 2mL sample was pipetted out and transferred to the cuvette for further image analysis. The process was repeated with different sand concentrations from 0% to 50%. All the experiments were done in triplicates. The images of the flocs were captured using a DSLR camera provided with a reverse ring to obtain the macroscopic images of the flocs [12] under an additional light source of 48W. The captured images were then processed using an image processing software namely, ImageJ to obtain the floc characteristics. The image processing method involves (a) setting the scale of the image (b) converting the image type from RGB scale to the 8bit grey scale (c) image filtering by passing through band pass filter (d) Applying otsu thresholding to separate the particles into foreground and background (e) Setting the measurements, which include defining the required parameters (f) Analysing the parameters to obtain the result in excel. Some of the floc properties provided by the software are area, perimeter, major axis, minor axis, circularity, and roundness. From these, the diameter, fractal dimension, floc density, and floc volume fraction (FVF) were determined. Diameter of flocs are calculated from the area of the flocs. Based on the floc size obtained, the flocs are classified into three size classes, class I (0μm-50μm), class II (50μm-100μm), class III (> 100μm). Breakage coefficient of the flocs is determined as the cubic root of ratio of floc volume fraction of a particular size class after breaking to that before breaking. Determination of fractal dimension and floc density are done with the equations (1) and (2) given below.

$$n_f = \frac{\log A}{\log a} \tag{1}$$

where A = area of the floc

a = major axis of the ellipse containing the floc

$$\rho_f = \rho_w + (\rho_s - \rho_w)\left(\frac{D_p}{D_f}\right)^{3-n_f} \tag{2}$$

where, ρ_w = density of water

ρ_s = density of particles

D_p = diameter of primary particle

D_f = equivalent diameter of floc

3 Results and Discussion

3.1 Variation of floc size with metal concentration

Figure 1 shows the influence of metal concentration on the floc size. The diameter of the flocs was observed to increase with the metal concentration. At high concentrations of metal, larger flocs are observed. The diameter of the flocs varied from 78.6 µm for kaolin to 259 µm for highest metal concentration of 20 mg/L. The phenomenon of aggregation contributed to the increase in floc size.

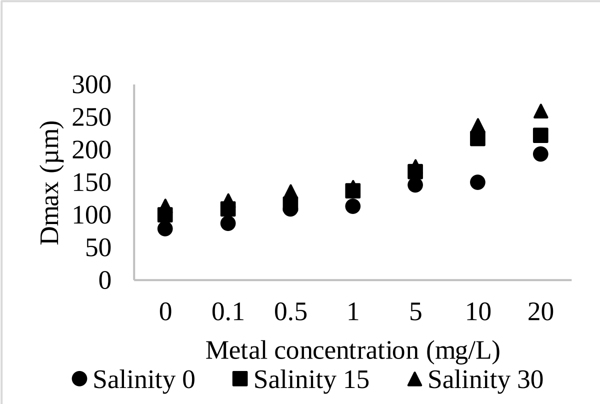

Figure 1: *Variation of maximum floc diameter with metal concentration*

Variation of floc size for different salinities indicates that increase in salinity also contributes to the floc size increment. Lower sized flocs were observed for salinity of 0 g/l whereas a higher floc size was observed for high salinity value of 30 g/L. Combined effect of metal concentration and salinity produces larger flocs than that at lower metal concentration and salinity.

3.2 Variation of floc size with sand concentration

Variations of the floc sizes after the addition of sand particles are shown in figure 2.

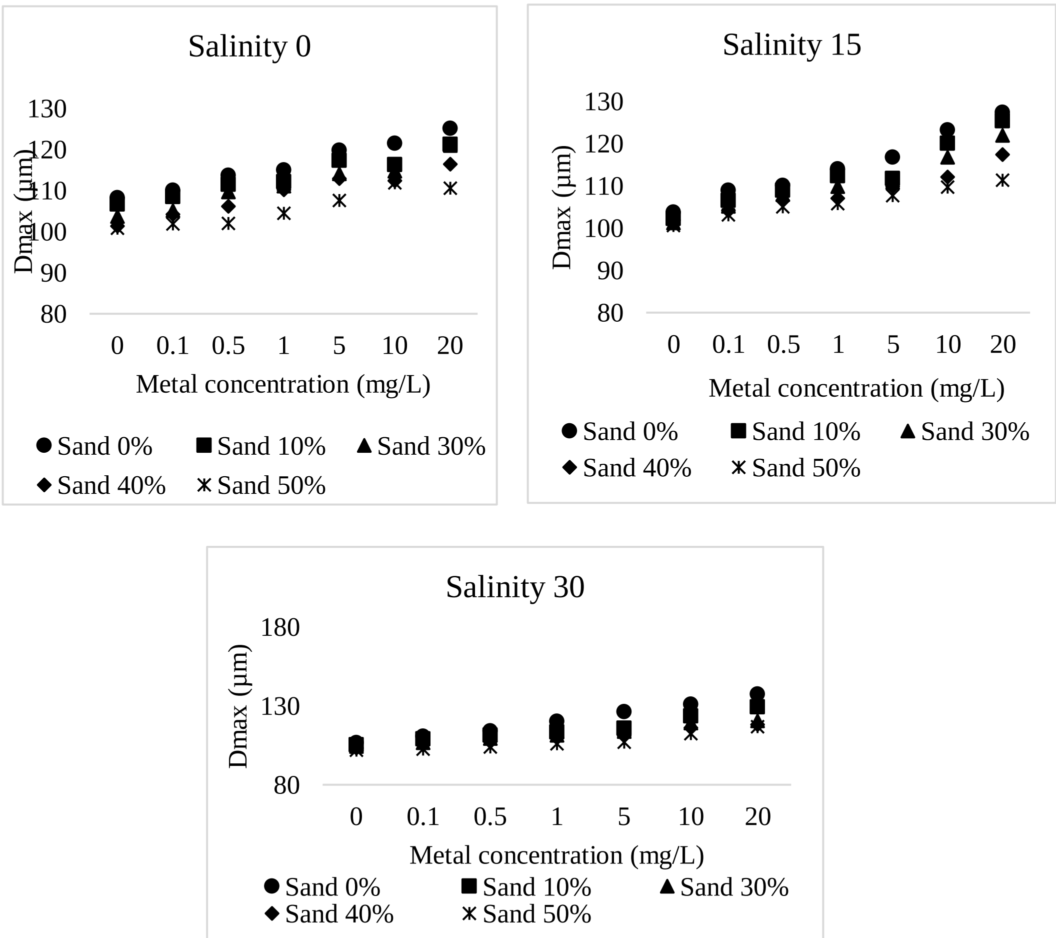

Figure 2: *Variation of maximum floc diameter with sand concentration*

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The incorporation of sand particles to the sample suspension of kaolin and metal resulted breakage of flocs forming microflocs. Microflocs were formed as a result of the breakup of macroflocs under the shearing action of sand. Increase in sand concentration from 0 % to 50 % decreased the floc sizes for all salinities. For salinity 30 g/L and 50 % sand content, the maximum floc size for kaolin (0mg/L metal concentration) reduced from 112µm (at 0% sand) to 79µm (at 50% sand). For a metal concentration of 20 mg/L at 30g/L salinity, the variation in D_{max} ranged from 122µm (0% sand) to 111.3µm (50% sand).

3.3 Effect of metal concentration on floc volume fraction

Figure 3 shows the variation of floc volume fraction of the three size classes (class I, class II and class III) with the metal concentration for each salinity.

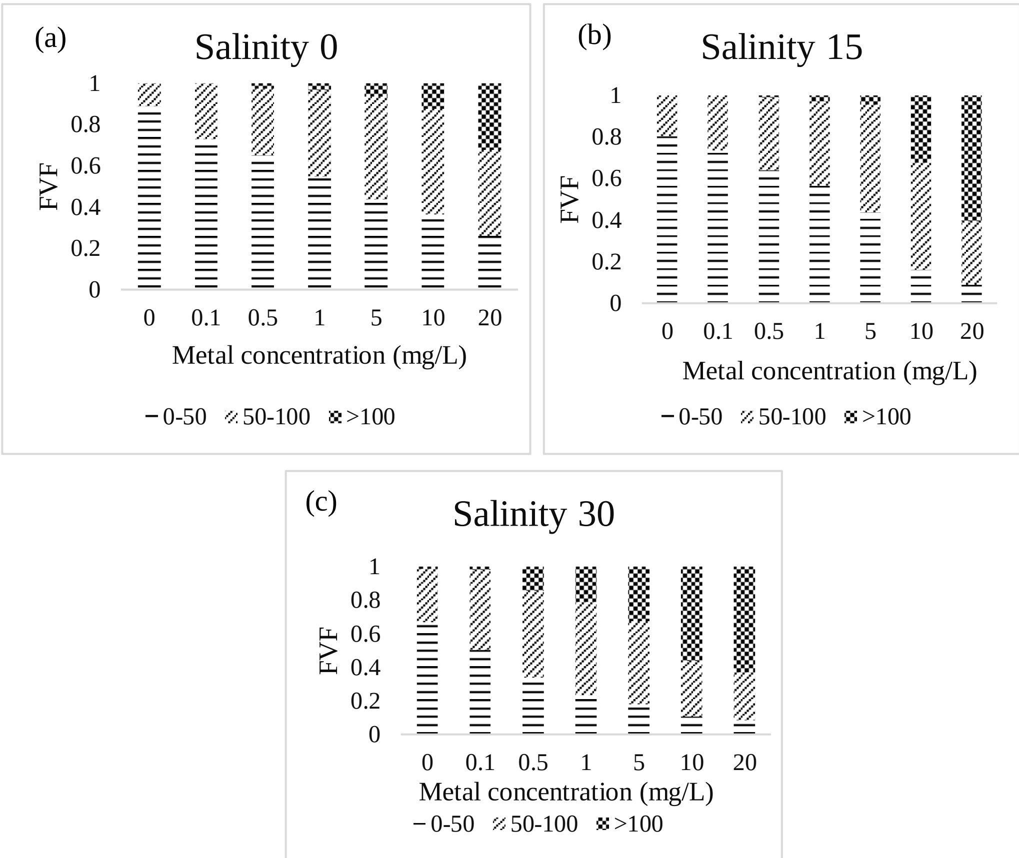

Figure 3: *Variation of floc volume fraction with metal concentration for (a) Salinity 0g/L (b) Salinity 15 g/L (c) Salinity 30 g/L*

The presence of metal enabled the formation of macroflocs by means of aggregation phenomenon. With increase in metal concentration, a considerable increase in the macroflocs were observed in class III. The presence of metal cations provided more active sites for the formation of flocs leading to the production of larger flocs. Salinity also contributed to the aggregation of particles to form macroflocs. Larger flocs were observed at high salinity of 30 g/L. Thus, it can be ascertained that the combined effect of metal concentration and salinity promotes floc growth providing environment for aggregation.

3.4 Effect of sand concentration on the floc volume fraction

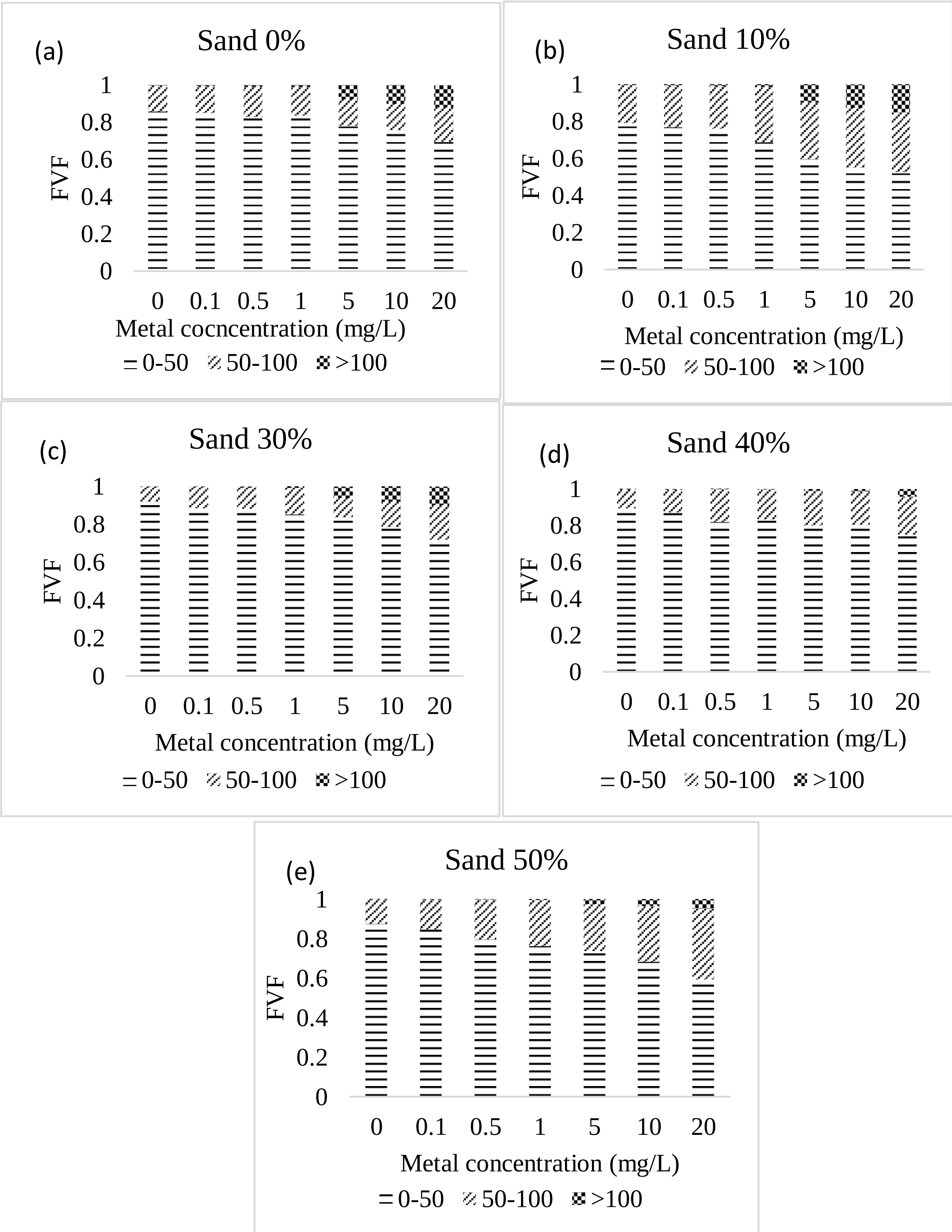

Figure 4: *Variation of FVF with metal concentration for (a) Sand 0% (b) Sand 10% (c) Sand 30% (d) Sand 40% (e) Sand 50%*

The influence of sand addition on the flocculation of metal-kaolin flocs are shown in figure 4. A shearing action of sand was observed for the metal-kaolin flocs wherein the sand imparts an additional shear that causes the fragmentation of flocs. The amount of macroflocs decreased due to their breakup under sand shear which simultaneously contributed to the microfloc population in the lower size classes (class I and class II). As the sand concentration increased, the microfloc population increased thereby causing a reduction in the macroflocs. At higher sand addition, more breakup of flocs was observed.

Figure 4 shows the variation of floc volume fraction with sand concentration for 30g/L salinities. More floc breakup was observed for lower metal concentration and lower salinity range. High metal concentration and salinity conditions reduced the effectiveness of sand shear relative to conditions of lower metal

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concentration and salinities. It was found that the presence of salt and metal made the kaolin flocs more resistant to breakage due to sand shear.

For additions of 50% sand for all salinities, a significant decrease in the macroflocs was seen. At 40% and 50% sand addition, no macroflocs for kaolin (0mg/L metal concentration) remained in suspension. When sand was introduced into kaolin suspension, floc breakage occurred at increased sand concentrations leading to microfloc formation, but in the presence of Fe (II), the kaolin flocs were more stable and resistant to breakage.

3.5 Variation of floc density with Sand concentration

Figure 5 shows the variation of floc density with the addition of sand. The increase in sand concentration produces more number of microflocs with high floc density. The kaolin flocs attain higher floc density due to their breakup into small fragments under sand shear whereas a lower floc density was exhibited by kaolin flocs at maximum metal concentration and salinity due to their larger size.

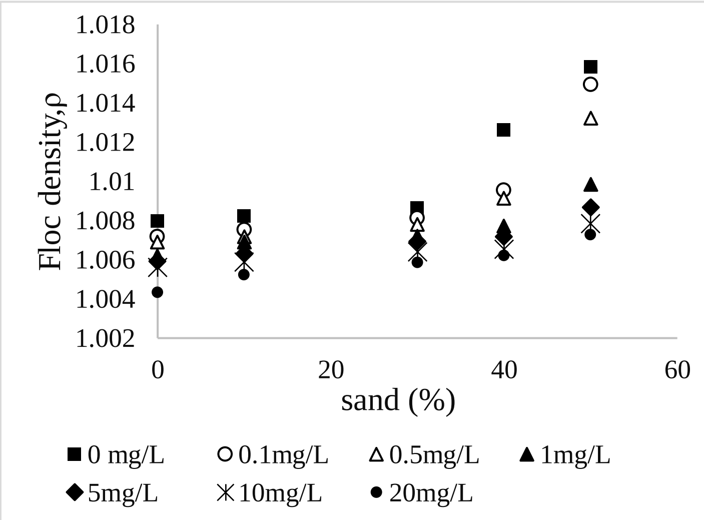

Figure 5: *Variation of floc density with sand concentration*

3.6 Breakage coefficient

Breakage coefficient is the ratio of cubic root of ratio of floc volume fraction of a particular size class after breaking to that before breaking. The breakage coefficient was normally adopted as ½ in the population balance modelling. But in the recent studies done by Priya *et al.* [11], it was observed that the value of breakage coefficient can vary from 0 to 1 depending on the conditions provided.

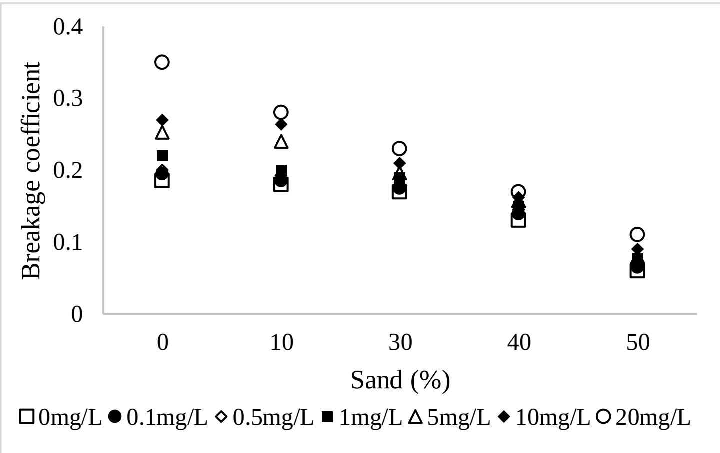

Figure 6: *Variation of breakage coefficient with sand concentration*

The addition of sand creates an additional turbulence that causes the flocs to break into small fragments. The action of sand breaks up the macroflocs of class III and the flocs fall into the lower size classes I and II. Figure 6 shows the variation of breakage coefficient with sand addition for different concentrations of metal. It was observed that with increase in sand concentration, the flocs are broken into microflocs of higher density. The presence of metal contributes to the strength of the flocs which is represented by the higher breakage coefficient at highest metal concentration. Higher values of breakage coefficient were observed for lower sand concentration and higher metal concentration of 20 mg/L.

4 Conclusions

The study analysed the effect of Fe (II) on the kaolin flocs and its resistance to breakage due to the shear created by sand. The results indicates that the presence of Fe (II) can enhance aggregation and promote floc growth. The increase in metal concentration increases the macrofloc formation. The presence of salinity also contributes to the floc growth. Maximum floc sizes are observed at high metal concentration and high salinity. Further the behaviour of metal-kaolin flocs in the presence of non-cohesive sediments were studied and shows that the presence of sand can provide an additional shear that causes floc breakage. The increase in sand concentration contributed to floc disruption, thereby increasing the microfloc population. Higher breakage is observed for lower metal concentration and low salinity. The breakage coefficient decreases with increase in sand concentration. Higher breakage coefficient occurs at low sand concentration whereas the value of breakage coefficient decreases at high sand conditions. The breakage coefficient is higher for high metal concentrations indicating the resistance of metal induced kaolin flocs to breakage.

5 Declarations

5.1 Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that cold have appeared to influence the work reported in this paper.

5.2 Publisher's Note

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