



1 **Abstract**

2           The Floc Size Distributions (FSDs) of suspended fine-grained sediment flocs play a prime  
3 role to estimate their own fate and the transport of contaminants attached to the flocs. However,  
4 developing an efficient flocculation model that is capable of simulating continuous and multimodal  
5 FSDs is still a challenge. Recently, the population balance equation solved by the Quadrature-  
6 Based Method of Moments (QBMM) with lognormal kernel density functions has been developed  
7 to investigate the aggregation and breakage processes. It coincides with some recent observations  
8 which describe a measured FSD in coastal waters with a set of constituted lognormal distributions.  
9 The newly developed lognormal QBMM was tested with several ideal flocculation kinetic kernels,  
10 none of which, however, was used for interpreting cohesive sediment dynamics. Therefore, it  
11 raised our interest to evaluate the model performance for fine-grained sediments in shear  
12 turbulence dominated environments. In this study, additional validations against two kaolinite  
13 laboratory experiments were tested in the framework of the extended QBMM. It is hypothesized  
14 that these subordinate lognormal distributions share the same value of standard deviation. Different  
15 from the previous methods, the common standard deviation is determined empirically to reduce  
16 the number of tracers and better represent the FSDs. With sediment flocculation kinetics, the  
17 predicted FSDs reasonably reproduce the FSDs observed in both the mixing chamber and the  
18 settling column experiments. Despite the lacking of explicit descriptions of microbial effects at the  
19 current stage, this model has the potential to be implemented into large-scale particle transport  
20 models and deserves a more in-depth study in the future.

21

22 *Keywords:* population balance equation; cohesive sediments; floc size distribution; subordinate  
23 lognormal distributions; mixing jar; settling column

## 1 **1. Introduction**

2           Large amounts of suspended particles, such as minerals from physico-chemical and  
3 biogenic origin (e.g., clays, quartz and carbonates), living and non-living organic matters (e.g.,  
4 plankton and detritus) and anthropogenic cohesive particles (e.g., microplastics), are trapped on  
5 mudflats, in navigational channels and on continental shelves each year. For example, about 480  
6 million ton suspended sediments are annually loaded from the Yangtze (Changjiang) River in  
7 China (Song et al., 2013) of which 40% may be deposited in the estuary (Milliman et al., 1985).  
8 The reason why such small particles can deposit in these high-energy turbulent environments is  
9 obviously flocculation. That is, the component microparticles, often covered by biofilm, collide  
10 and combine with each other, and as a consequence are aggregated into clusters having larger size,  
11 decreased density and higher settling velocities than their individual constituents. The flocculation  
12 kinetics can also modulate sediment bed exchanges and determine concentration dynamics (Letter  
13 and Mehta, 2011). Thus, it is critical to understand how these processes respond to various  
14 environmental factors in natural (both freshwater and saltwater) systems, since it controls the fate  
15 of the particles themselves, and further to the toxic substances attached to the flocs. Freshwater  
16 flocs in the river are not necessarily smaller than flocs in saline water environments, since stronger  
17 shear stresses in the estuary may be the dominant effect to control the floc size (e.g., along the  
18 Yangtze River, see Guo and He, 2011; He et al., 2015). Pronounced differences of floc sizes  
19 within a tidal cycle, between spring-neap cycles, or under stormy and calm weather conditions,  
20 also highlight the role of turbulent shear in the flocculation process (Guo et al., 2017, 2018). On  
21 the other hand, Cartwright et al. (2009) point out that in the York River estuary the flocs in the  
22 biologically-dominated mud site (where bioturbation is more prevalent, see Schaffner et al., 2001)  
23 have higher settling velocities than that in the physically-dominated site (where physically-induced

1 layering is more commonly observed within the bed), which emphasizes the effects of bioactivities  
2 on flocculation.

3         Flocs may range from microscopic (micron) size up to visible particles in units of  
4 millimeters, with a total size range of four orders of magnitude. Thanks to recently developed non-  
5 intrusive instruments such as the LISST (Laser In Situ Scattering and Transmissometry, see  
6 Agrawal and Pottsmith, 2000) and various digital camera systems (Benson and French, 2007;  
7 Cartwright et al., 2013; Eisma et al., 1990; Graham and Nimmo Smith, 2010; Keyvani and Strom,  
8 2014; Manning, 2004; Shen and Maa, 2016a; Tang and Maggi, 2015), the Flocc Size Distributions  
9 (FSDs) by particle volume or number can be better observed. Nevertheless, although large-scale  
10 field observations are the preferred way to study the sediment transport for engineering and  
11 environmental issues, they are hampered by high cost and geographical spreading. A coupled  
12 hydrodynamic and sediment transport model can complement measurements to cover the entire  
13 study domain. However, currently available three-dimensional (3-D) models are still not able to  
14 make satisfying quantitative predictions of sediment properties. This raises the following question:  
15 Can we do any better for the large-scale modeling of fine-grained sediment transport (Toorman,  
16 2012)? Actually, implementing a flocculation model (with the ability to predict the temporal and  
17 spatial varying FSDs) in sediment transport models, despite the challenge, could be a solution.  
18 One difficulty is that the typical issues we focus on have cell sizes in the order of 0.1 – 10 m in the  
19 vertical and 10 – 1000 m in the horizontal, and have time steps in the order of 0.1 – 10 minutes,  
20 whereas the particle micro-behaviors such as aggregation and breakage occur at smaller scales  
21 which are resolved at a sub-grid scale with a refined spatial resolution and a reduced time step.  
22 This limits the performance of some mesoscale simulations (e.g., Lattice Boltzmann model, see  
23 Zhang et al., 2016, 2017) applicable in large study domains. In reality, it requires that the

1 flocculation model is at reasonable cost, and that it has the potential to be extended to include other  
2 flocculation parameters besides the flocculation size, such as flocculation density (or fractal dimension), flocculation shape and  
3 flocculation composition that also influence the settling velocities of in-situ biomineral aggregates  
4 (Fettweis and Lee, 2017).

5 One candidate of such flocculation model is the Population Balance Equation (PBE) that  
6 is capable of tracking the number density  $n(L, \mathbf{x}, t)$  for particles with size  $L$  at location  $\mathbf{x} = \mathbf{x}(x, y,$   
7  $z)$  and time  $t$ . Flocculation-growth based single-group PBEs (Winterwerp, 1998, 2002), class-based two-  
8 or three-group PBEs (Lee et al., 2011, 2014; Shen et al., 2018a, 2018b) and quadrature-based  
9 multi-group PBEs (Prat and Ducoste, 2006; Shen and Maa, 2015, 2016b, 2017) are among a few  
10 applications that are successfully coupled with hydrodynamics at least in one-dimensional (1-D)  
11 applications. The flocculation-growth based PBEs only track the representative size of all the particles,  
12 and thus is unable to address multimodal FSDs which nevertheless are commonly observed in  
13 estuaries and coastal waters (e.g., Manning and Dyer, 2002; Benson and French, 2007; Verney et  
14 al., 2011). By comparisons with several analytical cases, Marchisio et al. (2002) indicates that  
15 quadrature-based PBEs are more desirable than class-based multi-class PBEs after investigating  
16 both the computational time and the accuracy of the representative sizes. Quadrature-based PBEs  
17 also have the advantages of including any measured initial FSD (e.g., Shen and Maa, 2016b) or  
18 including more particle properties (e.g., Vale and McKenna, 2005). Nevertheless, standard  
19 quadrature-based PBEs still lack thorough comparison with field data, especially for multimodal  
20 FSDs.

21 Lee et al. (2012) analyzed a large number of in-situ FSDs by flocculation volumes collected from  
22 the Belgian coastal zone and concluded that the observed FSDs can be approximated best by four  
23 lognormal distributions for primary particles (0.1 - 4  $\mu\text{m}$ ), microflocs (4 - 20  $\mu\text{m}$ ), macroflocs (50

1 - 200  $\mu\text{m}$ ) and megaflocs ( $> 200 \mu\text{m}$ ). The two-class PBE can efficiently and reasonably simulate  
2 the representative sizes of microflocs (with primary particles merged) and macroflocs (with  
3 megaflocs merged) (Lee et al., 2011, 2014). Later, Shen et al. (2018a, 2018b) extended the two-  
4 class model into three-class by adding an additional group to better address the appearance of  
5 megaflocs especially during algae bloom period. The simplified two-class or three-class models  
6 have also been successfully coupled with the open TELEMAC modeling system. In their model,  
7 however, only the representative sizes of the component lognormal FSDs are involved. That is,  
8 the simulated FSDs are not continuous, since they do not contain the prediction of the standard  
9 deviation of each lognormal FSD. This leads to the missing of a direct representation of the entire  
10 FSD shape.

11 This weakness is a challenge to be dealt with until Nguyen et al. (2016) solved the FSDs  
12 using the Extended Quadrature-Based Method of Moments (Extended QBMM or E-QBMM)  
13 (Yuan et al., 2012) with lognormal kernel density functions (Madadi-Kandjani and Passalacqua,  
14 2015). However, only simple conceptual aggregation and/or breakage kernels within zero-  
15 dimensional (0-D) frameworks are tested and compared with the reference work by Vanni (2000).  
16 The tested kernels are not realistic for describing collision and breakage processes of cohesive  
17 aggregates in natural aquatic environments. It is not clear whether the prediction of FSDs of  
18 estuarine mud will benefit from their contributions. Therefore, the objective of this study is to  
19 conduct additional validations for the flocculation processes of cohesive sediments. The common  
20 standard deviation of the constituted FSDs is determined empirically. Two experiments are  
21 employed to evaluate the model performance: (1) a kaolinite mixing jar test by Shen and Maa  
22 (2016a) as a 0-D validation case, and (2) a kaolinite settling column experiment by Maggi et al.  
23 (2007) as a vertical 1-D validation case. The remainder of the paper is structured as follows:

1 Section 2 briefly introduces the PBE, the E-QBMM and the solution methods. Section 3 mainly  
 2 describes two experiments for model validations. The results and discussion on model predictions  
 3 are carried out in Section 4, with some thoughts on the development of a coupled hydrodynamic,  
 4 turbulence and flocculation model. The conclusions are presented in Section 5.

5

## 6 **2. Model description and solution methods**

### 7 2.1 Moment transport equations

8 Consider the following PBE in a vertical 1-D format for describing the evolution of number  
 9 density of suspended particles in a carrier fluid (Shen and Maa, 2015, 2016b, 2017):

$$\begin{aligned}
 10 \quad & \frac{\partial n(L, z, t)}{\partial t} - \frac{\partial [n(L, z, t) \cdot w_s(z, t)]}{\partial z} - \frac{\partial}{\partial z} \left( D(z) \cdot \frac{\partial n(L, z, t)}{\partial z} \right) \\
 11 \quad & = \frac{L^2}{2} \int_0^L \left[ \frac{\alpha \cdot \beta((L^3 - \lambda^3)^{1/3}, \lambda)}{(L^3 - \lambda^3)^{2/3}} \cdot n((L^3 - \lambda^3)^{1/3}, z, t) \cdot n(\lambda, z, t) \right] d\lambda \\
 12 \quad & - n(L, z, t) \int_0^\infty \alpha \cdot \beta(L, \lambda) \cdot n(\lambda, z, t) d\lambda + \int_L^\infty a(\lambda) \cdot b(L | \lambda) \cdot n(\lambda, z, t) d\lambda - a(L) \cdot n(L, z, t) \\
 13 \quad & \hspace{20em} (1)
 \end{aligned}$$

14 where  $n(L, z, t)$  is the number density of the particles with size  $L$  at vertical coordinate  $z$  and time  
 15  $t$ ,  $w_s$  is the mass-weighted settling velocity,  $D$  is the eddy diffusivity,  $\lambda$  is the variable of integration  
 16 with dimension of size,  $\alpha$  is the collision efficiency,  $\beta(L, \lambda)$  is the collision frequency between  
 17 particles with size  $L$  and  $\lambda$ ,  $a(L)$  is the breakage frequency of particles with size  $L$ , and  $b(L | \lambda)$  is  
 18 the fragmentation distribution function describing the created number of particles with size  $L$  after  
 19 the breakage of a parent particle with size  $\lambda$ . The left hand side of Eq. 1 comprise the unsteady  
 20 term, the settling term and the diffusion term, respectively. The right hand side consists of the

1 flocculation sources and sinks (Shen and Maa, 2015). The advection term may be added in Eq. 1  
 2 for strong bulk transport of sediments.

3 To avoid tracking a large number of size classes, the FSDs can instead be stored in their  
 4 moments by the moment transformation:

$$5 \quad m_k(t) = \int_0^\infty L^k n(L,t) dL \quad (k = 0, 1, \dots) \quad (2)$$

6 in which  $m_k$  is the  $k$ th order moment of the FSD. Note that  $m_0$ ,  $m_2$  and  $m_3$  are proportional to the  
 7 total number, total surface area and total volume of the solid particles.

8 Substituting Eq. 2 into Eq. 1, the governing equation can be rewritten as a set of moment  
 9 transport equations (Marchisio et al., 2003a):

$$10 \quad \frac{\partial m_k(L, z, t)}{\partial t} - \frac{\partial [m_k(L, z, t) \cdot w_s(z, t)]}{\partial z} - \frac{\partial}{\partial z} \left( D(z) \cdot \frac{\partial m_k(L, z, t)}{\partial z} \right)$$

$$11 \quad = \frac{1}{2} \int_0^\infty n(\lambda, z, t) \int_0^\infty \alpha \cdot \beta(L, \lambda) \cdot (L^3 + \lambda^3)^{k/3} \cdot n(L, z, t) dL d\lambda$$

$$12 \quad - \int_0^\infty L^k n(L, z, t) \int_0^\infty \alpha \cdot \beta(L, \lambda) \cdot n(\lambda, z, t) d\lambda dL$$

$$13 \quad + \int_0^\infty L^k \int_0^\infty a(\lambda) \cdot b(L | \lambda) \cdot n(\lambda, z, t) d\lambda dL - \int_0^\infty L^k a(L) \cdot n(L, z, t) dL$$

$$14 \quad (k = 0, 1, \dots) \quad (3)$$

15 This equation is unclosed since the FSD should be resolved from its moments to make sure  
 16 that the source and sink terms are physical. The moments have to stay in the moment space, which  
 17 is the reliability condition of these quadrature-based methods (Wright, 2007; Nguyen et al., 2016;  
 18 Laurent and Nguyen, 2017). In fact, even a suitable closure does not necessarily guarantee a



1 realizable FSD, especially at the boundary. Nevertheless, this issue is not always considered, since  
2 approaches that determine if a vector belongs to the moment space (characterized by the Hankel  
3 determinants) are generally unsatisfactory and computationally heavy.

4

## 5 2.2 Solution method and sediment flocculation kinetics

6 In order to close the moment transport equation (Eq. 3), Yuan et al. (2012) proposed that  
7 the number density function can be reconstructed by a weighted superposition of non-negative  
8 functions:

$$9 \quad n(L) = \sum_{i=1}^N w_i \cdot \delta_{\sigma}(L, L_i) \quad (4)$$

10 where  $\delta_{\sigma}(L, L_i)$  is the kernel density function (i.e., the subordinate FSDs),  $w_i$  and  $L_i$  are the non-  
11 negative weights and corresponding representative sizes (also referred to as “abscissas”, “nodes”  
12 and “pivots”) of the constituted FSDs,  $N$  is the number of component FSDs, and  $\sigma$  is a unique  
13 nonnegative parameter shared by all subordinate FSDs. Hereafter we drop symbol  $z$  and  $t$  in the  
14 number density function  $n(L)$  to focus on the FSD at a specific time and location.

15 Lee et al. (2012) concluded that an observed FSD of estuarine mud can be decomposed  
16 into two to four lognormal distributions to estimate the settling flux with 3% - 10% errors. Thus,  
17 it is straightforward to select the kernel density function  $\delta_{\sigma}(L, L_i)$  as a lognormal distribution at  
18 first stage in our applications. In this sense, the parameter  $\sigma$  in Eq. 4 becomes the standard deviation  
19  $\sigma$  of the lognormal distributions (Madadi-Kandjani and Passalacqua, 2015):

$$20 \quad \delta_{\sigma}(L, L_i) = \frac{1}{L\sigma\sqrt{2\pi}} \exp\left[-\frac{(\ln(L/L_i))^2}{2\sigma^2}\right] \quad (5)$$

1 If  $\sigma \rightarrow 0$ ,  $\lim_{\sigma \rightarrow 0} \delta_\sigma(L, L_i) = \delta(L - L_i)$ , and thus Eq. 4 is reduced to:

$$2 \quad n(L) = \sum_{i=1}^N w_i \cdot \delta(L - L_i) \quad (6)$$

3 which can be solved by the standard QBMM (McGraw, 1997; Shen and Maa, 2015). Note that  
 4 although a continuous FSD can be approximated by a large number of delta functions in QBMM  
 5 (Eq. 6), it is numerically difficult to track the FSD with high order moments. That is, strictly  
 6 speaking, the FSDs predicted by the QBMM are usually limited by size classes less than eight.  
 7 Selecting lognormal distributions as the kernel density functions, nevertheless, will make the entire  
 8 FSD predictions truly continuous.

9 For an arbitrary function  $g(L)$ , the integral with the number density function  $n(L)$  as the  
 10 weight function can be expressed as (Madadi-Kandjani and Passalacqua, 2015):

$$11 \quad \int_0^\infty g(L) \cdot n(L) \cdot dL = \int_0^\infty g(L) \cdot \sum_{i=1}^N w_i \cdot \delta_\sigma(L, L_i) dL = \sum_{i=1}^N \sum_{j=1}^{N_i} w_i w_{ij} g(L_{ij}) \quad (7)$$

12 where  $L_i$  and  $w_i$  are the primary abscissas and weights ( $i = 1, 2, \dots, N$ ), and  $N$  is the number of  
 13 subordinate lognormal functions. For each lognormal function  $\delta_\sigma(L, L_i)$  (Eq. 5),  $L_{ij}$  and  $w_{ij}$  ( $j = 1,$   
 14  $2, \dots, N_i$ ) are the secondary abscissas and weights. In general,  $N_i \geq N$  is required in order not to  
 15 lose the accuracy of introducing the second abscissas and weights.

16 Selecting  $g(L) = L^k$ , and substituting Eq. 7 into Eq. 3, the governing equation can be  
 17 expressed as:

$$18 \quad \frac{\partial m_k}{\partial t} - \frac{\partial(m_k \cdot w_s)}{\partial z} - \frac{\partial}{\partial z} \left( D \cdot \frac{\partial m_k}{\partial z} \right)$$

$$19 \quad = \frac{1}{2} \sum_{i_1=1}^N \sum_{j_1=1}^{N_{i_1}} w_{i_1} w_{i_1 j_1} \sum_{i_2=1}^N \sum_{j_2=1}^{N_{i_2}} w_{i_2} w_{i_2 j_2} \alpha \cdot \beta(L_{i_1 j_1}, L_{i_2 j_2}) \cdot (L_{i_1 j_1}^3 + L_{i_2 j_2}^3)^{\frac{k}{3}}$$

$$\begin{aligned}
& - \sum_{i_1=1}^N \sum_{j_1=1}^{N_{i_1}} L_{i_1 j_1}^k w_{i_1} w_{i_1 j_1} \sum_{i_2=1}^N \sum_{j_2=1}^{N_{i_2}} w_{i_2} w_{i_2 j_2} \alpha \cdot \beta(L_{i_1 j_1}, L_{i_2 j_2}) \\
& + \sum_{i=1}^N \sum_{j=1}^{N_i} w_i \cdot a_{ij}(L_{ij}) \cdot \bar{b}_{ij}^{(k)} \cdot w_{ij} - \sum_{i=1}^N \sum_{j=1}^{N_i} w_i \cdot L_{ij}^k \cdot a_{ij}(L_{ij}) \cdot w_{ij} \quad (k = 0, 1, \dots) \quad (8)
\end{aligned}$$

$$\text{with } \bar{b}_{ij}^{(k)} = \int_0^\infty L_{ij}^k b(L | \lambda) dL \quad (9)$$

At each location and time step, the FSD, i.e., the  $L_i$  ( $i = 1, \dots, N$ ) and  $w_i$  ( $i = 1, \dots, N$ ) are reconstructed from the moments  $m_k$ , which is referred to as “moment inversion” (e.g., Passalacqua et al., 2018). It is important to note that in Lee et al. (2012)’s study, the parameter  $\sigma$  varies for each constituted distribution for a better match of the fitted and measured FSDs; in this study, however,  $\sigma$  is shared for all subordinate lognormal distributions. With the parameter  $\sigma$  properly selected, the predictions of the entire FSDs are not significantly deviated (Fig. 1). A common  $\sigma$  can also reduce the number of tracers. For example, as shown in Fig. 1, if using number of  $N$  lognormal FSDs to approximate the measurement, an unshared  $\sigma$  requires  $3N$  variables while a common  $\sigma$  requires only  $2N+1$ . If the common  $\sigma$  is further determined empirically (Eq. 10) rather than treated as an independent tracer, the number of tracers is reduced to  $2N$ . It is hypothesized that the standard deviation ( $\sigma$ ) of the subordinate distribution is enlarged with the increment of the characteristic mean size  $d_{32}$  until an equilibrium or quasi-equilibrium state. At  $t = 0$ , all particles are assumed concentrated on single size primary particles with  $\sigma = 0$ . Thus, the common  $\sigma$  is given by:

$$\sigma = \alpha_0 \cdot \ln \left( \frac{d_{32}}{l_p} \right) \quad (10)$$

where  $d_{32}$  ( $d_{32} = m_3/m_2$ ) is the Sauter mean size (Mugele and Evans, 1951; Sowa, 1992),  $l_p$  is the size of primary particles and  $\alpha_0$  is a fitting constant.

The  $k$ th order moments of the lognormal distribution  $\delta_\sigma(L, L_i)$  are (Magnus et al., 1996):

1  $m_k(L_i, \sigma) = \exp(k \cdot \ln L_i + k^2 \cdot \sigma^2 / 2)$  (11)

2 Let  $Z = \exp(\sigma^2 / 2)$ , Eq. 2 becomes:

3 
$$\begin{cases} m_0 = w_1 + w_2 + \dots + w_N = m_0^* \\ m_1 = Z(w_1 L_1 + w_2 L_2 + \dots + w_N L_N) = Z \cdot m_1^* \\ m_2 = Z^4(w_1 L_1^2 + w_2 L_2^2 + \dots + w_N L_N^2) = Z^4 \cdot m_2^* \\ \dots \\ m_k = Z^{k^2}(w_1 L_1^k + w_2 L_2^k + \dots + w_N L_N^k) = Z^{k^2} \cdot m_k^* \end{cases}$$
 (12)

4  $(k = 0, 1, \dots)$

5 After the common standard deviation  $\sigma$  being found in Eq. 10, the first  $2N$  terms in Eq. 12

6 can be employed to solve  $L_i$  and  $w_i (i = 1, \dots, N)$ :

7 
$$\sum_{i=1}^N w_i L_i^k = m_k^* \quad (k = 0, 1, \dots, 2N - 1)$$
 (13)

8 Eq. 13 can be solved by the Chebyshev algorithm given by Wheeler (Wheeler, 1974; Su et

9 al., 2007; Yuan and Fox, 2011; Shen and Maa, 2015). The main point of Eq. 13 is to find the

10 eigenvalues and eigenvectors of a real, symmetric, tri-diagonal Jacobi matrix, with the elements

11 of the matrix computed by the moments from a three term recurrence relation, which is based on

12 the theory of orthogonal polynomials. Although Eq. 13 is numerically ill-posed, it is practical to

13 apply standard QBMM to solve it for low order moments ( $N \leq 4$ ). An adjustable factor may be

14 included only when more subordinate lognormal distributions are required.

15 Therefore, the solution of governing equation (Eq. 8) can be summarized in the following:

16 (a) At  $t = 0$ , compute the initial  $2N$  moments  $m_0, m_1, \dots, m_{2N-1}$  for a given initial particle size

17 distribution (Eq. 2), assuming using number of  $N$  subordinate lognormal distributions to

18 approximate the entire FSD.

- 1 (b) Update the moments, by solving the advection (if necessary), diffusion and settling terms in  
2 Eq. 8.
- 3 (c) Estimate the common standard deviation  $\sigma$  (Eq. 10), and extract the primary abscissas  $L_i$  and  
4 weights  $w_i (i = 1, \dots, N)$  from the first  $2N$  moments  $m_k (k = 0, 1, \dots, 2N-1)$  (Eq. 13), using Wheeler's  
5 algorithm (for unknown kernel density functions).
- 6 (d) Find the secondary abscissas  $L_{ij}$  and  $w_{ij} (j = 1, 2, \dots, N_i)$  for each constituted lognormal FSD  $\delta_\sigma$   
7  $(L, L_i)$ , using Gauss-Stieltjes-Wigert quadrature (equivalent to use Wheeler's algorithm for  
8 lognormal kernel density functions).
- 9 (e) Calculates the source and sink terms in Eq. 8, updates the moments, and go to (b) to solve the  
10 transport equation for the next time step.

11 In order to address the cohesive sediment properties by solving Eq. 8, the flocculation  
12 kinetics are selected based on previous studies (e.g., Smoluchowski, 1917; Winterwerp, 1998;  
13 Marchisio et al., 2003b; Maggi et al., 2007; Shen and Maa, 2015, 2016b):

14 Collision frequency:  $\beta(L_{i_1 j_1}, L_{i_2 j_2}) = \frac{G}{6} (L_{i_1 j_1} + L_{i_2 j_2})^3$  (14)

15 Breakage frequency:  $a(L_{ij}) = E_b \cdot \left(\frac{\mu}{F_y}\right)^{1/2} \cdot G^{3/2} \cdot L_{ij} \cdot \left(\frac{L_{ij}}{l_p} - 1\right)^{3-nf}$  (15)

16 Fragmentation distribution function:

17  $\overline{b_{ij}^{(k)}} = L_i^k \cdot K^{(3-k)/3}$  (16)

18 where  $G$  is the shear rate,  $E_b$  is the breakage fitting parameter,  $F_y$  is the floc strength, and  $K$   
19 describes the number of daughter flocs created after a parent floc breaks up.

20 The settling velocity for each size group is represented as (Winterwerp, 1998):

$$w_{s,i} = \Phi_{HS} \frac{1}{18} \frac{(\rho_s - \rho_w)g}{\mu} l_p^{3-n_f} \frac{L_i^{n_f-1}}{1+0.15 \cdot Re_f^{0.687}} \quad (17)$$

where  $\rho_s$  and  $\rho_w$  are the densities of sediment and water,  $n_f$  is the fractal dimension of flocs,  $\mu$  is the fluid dynamic viscosity,  $g$  is the gravitational acceleration, and  $Re_f = w_{s,i} \cdot L_i \cdot \rho_w / \mu$  is the floc particle Reynolds number. The parameter  $\Phi_{HS} = \left(1 - \frac{c}{c_{gel}}\right)^A$  is the correction factor accounting for hindered settling (Cuthbertson et al., 2008), where  $c \propto L^3$  is the Suspended Sediment Concentration (SSC),  $c_{gel}$  is the gelling concentration, and  $A$  is a constant between 2.5 – 5.5 and set as 4.7 (Richardson and Zaki, 1954).

### 3. Case studies

#### 3.1 Case 1: Mixing jar experiments

Recently a laboratory experiment was carried out by Shen and Maa (2016a) to investigate the equilibrium FSDs of suspended kaolinite in a five liter cubic mixing chamber for different sediment concentrations, shear rates, salinities and guar gum dosages. The LIGHTNIN A310 propeller was used to generate the turbulence. The tank averaged shear rate  $G$  was controlled by the rotational speed of the propeller ( $N_r$ ) which was driven by a DC gear motor, and  $G$  was also influenced by the properties of the propeller as well as the volume of the chamber  $V_c$  (Ducoste and Clark, 1998):

$$G = \sqrt{\frac{\rho_w N_r D_r N_{rp}}{\mu V_c}} \quad (18)$$

in which  $D_r$  is the diameter of the propeller and  $N_{rp}$  is a propeller specific constant. Sediment samples were extracted by syringe and weighted after oven-dried overnight to determine the

1 sediment concentration ( $c$ ). The FSDs were observed by a camera and image processing system,  
2 with a resolution of  $2 \times 2$  pixels to identify a floc with minimum size of  $5 \mu\text{m}$ . The camera  
3 continuously took pictures every 2 s, with the camera and light source trigger controlled by a  
4 programmed microcontroller Teensy 2.0. The FSDs at steady state are reported by an average of a  
5 continuous one hundred images. The image processing procedure was supported by comparison  
6 with commercially available particles (Shen and Maa, 2016a), and later was also reported as “well  
7 correlated” with other image capturing systems (Ramalingam and Chandra, 2017). Such a mixing  
8 chamber and image processing system was placed at Virginia Institute of Marine Science (USA)  
9 from 2013 to 2015.

10 Shen and Maa (2016a) have successfully modeled the FSDs in most cases using the  
11 standard QBMM. However, for cases of sediments with guar gums, a strong bimodal FSD were  
12 observed. Their method cannot address the second peak of FSD with a maximum of eight nodes.  
13 In this study, the FSDs were instead estimated by two subordinate lognormal distributions ( $N = 2$ ).  
14 For each subordinate distributions, a three-node quadrature was used to estimate its moments ( $N_i$   
15  $= 3$ ). The settling and diffusion terms in Eq. 8 are neglected to address the average property in the  
16 mixing chamber. The initial kaolinite concentration ( $c$ ) was  $0.52 \text{ g/L}$ , the tank-averaged shear rate  
17 ( $G$ , calculated by Eq. 18) was  $55 \text{ s}^{-1}$ , and the guar gum concentrations  $c_{\text{guar}} = \{0, 5, 10, 15, 20, 30\}$   
18  $\text{mg/L}$ . For each case, the jar test was carried out independently, and the solution was discarded  
19 before the next test. The propeller rotated at its maximum speed for half an hour to destroy the  
20 flocs, and then was set to the proposed rotational speed to promote flocculation. Thus, all the  
21 particles are assumed in the form of primary particles at  $t = 0$  with  $l_p = 6 \mu\text{m}$ . The steady state  
22 FSDs were used to calibrate this model.

23

## 1 3.2 Case 2: Settling column experiments

2           The second validation is to compare the model results with FSD measurements in a settling  
3 column experiment by Maggi et al. (2007). The settling column was designed and manufactured  
4 at Delft University of Technology (Netherlands), with height 5 m (totally 4 m, from surface to 1  
5 m above bottom) and diameter 30 cm. A homogeneous and isotropic turbulent field was generated  
6 by an oscillating grid over the settling section of the column height (e.g., Cuthbertson et al., 2010,  
7 2018; Tang and Maggi, 2015). By comparison with several grid configurations, Maggi (2005)  
8 proposed that the rectangular grid was the optimal selection to compromise both the flow  
9 hydrodynamic behavior and the manufacturing costs. Driving systems controlled the movement of  
10 the grid with a maximum stroke ( $A_0$ ) of 8.4 cm. The shear rate  $G$  was linearly correlated and  
11 controlled by the grid frequency, with shear rate  $G = \{5, 10, 20, 40\} \text{ s}^{-1}$  corresponding to grid  
12 frequency  $f_g = \{0.05, 0.1, 0.25, 0.5\} \text{ Hz}$ , respectively. The aggregates passed through the settling  
13 section and reach the measuring section which was located at 0.5–1 m above bottom. A small  
14 fraction of the flocs were transferred to a collector, with the FSDs measured by a digital camera  
15 without being affected by large-scale water circulation in the settling column. The aggregates are  
16 illuminated by a laser source and captured in the images with a resolution of 6  $\mu\text{m}$  per pixel. The  
17 bottom 0.5 m was the bed section, with particles passing through the settling and measuring  
18 sections deposited on the bottom. In the experiment, kaolinite suspensions with concentrations  $c$   
19  $\pm 0.015 \text{ g/L}$  were maintained by a buffer tank on the top of the settling column. By assuming the  
20 downward flux above and below the measuring section are close, this flocculation process of  
21 suspended kaolinite can be simulated using a 0-D model (Maggi et al., 2007; Mietta et al., 2008;  
22 Shen and Maa, 2015). However, this assumption cannot be verified or argued without additional  
23 measurements. In this section, a vertical 1-D model (Eq. 8) was employed to reproduce the



1 experiment, with sediment mass concentration 0.5 g/L at the top boundary and shear rates 5, 10,  
2 20 and 40 s<sup>-1</sup> respectively. Although the vertical eddy diffusivity  $D$  was not reported in their  
3 experiment, van Leussen (1994) pointed out that its value in such experiment can be determined  
4 by the following relationship:

$$5 \quad D = \alpha_g \cdot f_g \cdot A_0^2 \quad (19)$$

6 in which  $\alpha_g$  is a constant that should be determined from the experiment. Here, it hypothesized that  
7 the energy from the grid bars is dissipated by the fluid of the whole cross section. Therefore,  $\alpha_g$   
8 can be estimated by the ratio of grid bar areas to the cross section of the settling column. It results  
9 in  $\alpha_g = 0.19$ . This value is similar to van Leussen's experiment with the settling column having  
10 similar column diameter and grid configuration. It is understandable that the energy will be  
11 dissipated rapidly when flocs settled out of the turbulent section, and thus it also assumes that the  
12 eddy diffusivity and the energy dissipation rate rapidly decreases to their minimum value from the  
13 settling-measuring section interface (i.e., 1 m above the bottom) to the bed. The initial and  
14 boundary conditions are in accordance with the following assumptions: (1) at  $t = 0$ , all particles  
15 are primary particles with size of 8  $\mu\text{m}$ ; (2) at the upper boundary, the sediments released from the  
16 buffer tank are all primary particles with a fixed concentration of 0.5 g/L; (3) at the lower boundary,  
17 sediments are freely deposited without erosion and resuspension; (4) the standard deviation  $\sigma$  in  
18 the bottom cell is set to 0 to avoid numerical instabilities. Finally, the observed FSDs at the  
19 measuring section are used to calibrate and validate the model.

20

## 21 4. Results and discussion

### 22 4.1 Comparison with mixing jar experiments: 0-D test

1           The parameters for different guar gum concentrations  $c_{\text{guar}} = \{0, 5, 10, 15, 20, 30\}$  mg/L  
 2 are summarized in Table 1. A value of  $K = 5$  is selected in the fragmentation distribution function  
 3 (Eq. 16) for all cases. The errors ( $E$ ) of predicted and observed FSDs are evaluated using the  
 4 following equation (Maggi et al., 2007; Shen and Maa, 2017):

$$5 \quad E = \frac{1}{2} \left( \sum_i |w_{M,i} - w_{E,i}| \right) \quad (20)$$

6 In this experiment, the measured number frequencies are given at  $L_E = \{5.27, 7.33, 10.2, 14.2,$   
 7  $19.8, 27.6, 38.4, 53.5, 74.5, 104, 144, 201, 280, 390, 543, 756, 1052\}$   $\mu\text{m}$ . Thus, the differences of  
 8 modeled  $w_M$  and experimental  $w_E$  at diameter  $L_E$  are evaluated using Eq. 20, with  $\sum_i w_{M,i} = 1$  and

$$9 \quad \sum_i w_{E,i} = 1.$$

10           The simulated FSDs agree well with the measurements for all guar gum dosages (Fig. 2),  
 11 with a maximum error of 0.07 (Table 1). Enhanced model predictions are achieved in this study,  
 12 compared with the previous study by Shen and Maa (2016a) that failed to mimic the bimodal FSDs  
 13 using an eight-node QBMM. Two subordinate lognormal distributions ( $N = 2$ ) of microflocs and  
 14 macroflocs are employed to represent the entire FSD. This selection is not only for reducing the  
 15 number of tracers. Notice that a better prediction is not always guaranteed by increasing the  
 16 number of subordinate FSDs. It is influenced by the properties of the flocculation kinetics. Not all  
 17 the target FSDs are decomposable into lognormal distributions. Also, it is not suitable to use a  
 18 large number of component FSDs to approximate a simple distribution. For example, when no  
 19 guar gum is added, it seems that the target FSD may be better predicted by one lognormal  
 20 distribution instead of two (Fig. 2(a)). Based on the above concerns,  $N = 2$  for  $L_1$  represents  
 21 microflocs and  $L_2$  represents macroflocs are employed for all cases in this study.

1           The breakage coefficient  $E_b$  is in the order of  $10^{-6}$  in this case, about one order of magnitude  
2 lower than clean sediment for a reduced breakage because of organic matter (Table 1). The ratio  
3 of  $\alpha/E_b$  is a critical parameter to exhibit floc growth (Mietta et al., 2011; Furukawa and Watkins,  
4 2012; Shen and Maa, 2016b). A maximum ratio of  $\alpha/E_b$  are occurred for guar gum concentration  
5 15 mg/L (Fig. 3(a)), with maximum size of macroflocs ( $L_2 = 151 \mu\text{m}$ , Fig. 3(c)) and highest weights  
6 ( $w_2 = 0.69$ , Fig. 3(d)) also at this dosage. It confirms the existence of an optimal  $c_{\text{guar}}$  dosage (Table  
7 1) for settling flocs in natural environment or waste water treatment. The sizes of microflocs  $L_1$   
8 are close to each other ( $\sim 32 \mu\text{m}$ , Fig. 3(c)), with its minimum weight ( $w_1$ ) also at  $c_{\text{guar}} = 15 \text{ mg/L}$   
9 (Fig. 3(d)). The fractal dimension  $n_f$  has the smallest value also at this optimal  $c_{\text{guar}}$  dosage (Fig.  
10 3(b)). An average fractal dimension of 2.7 (Table 1) indicates that the flocs in the mixing chamber,  
11 although with size enlarged, are still relatively compact. In our sensitivity test, a fractal dimension  
12  $n_f$  from 2.0 to 2.7 will not significantly alter the size of the major peak; however, the common  
13 standard deviation  $\sigma$  is reduced as  $n_f$  decreases (Fig. 4(a)). A high  $n_f$  will decrease the breakup  
14 frequency (Eq. 15), which lead to higher mean size and larger  $\sigma$ . It is essential to note that a  
15 common  $\sigma$  derived from the  $(2N)$ th moment (Madadi-Kandjani and Passalacqua, 2015) will lead  
16 to numerical instability and physically to a narrow spread of FSD in our applications, and thus is  
17 not used. With an empirical  $\sigma$  (Eq. 10) related to the mean size of FSD, a better estimation of the  
18 spread of FSD are reached. Although the selected  $\alpha_0$  (Eq. 10) and thus  $\sigma$  are close in all cases (Fig.  
19 3(b)(d)), the predicted FSDs are sensitive for the selection of  $\alpha_0$  (Fig. 4(b)). Increase of the value  
20 of  $\alpha_0$  will increase  $\sigma$  straightforwardly, and therefore make the spread of FSD wider and decrease  
21 the size of main peak (Fig. 4(b)).

22           Taking  $c_{\text{guar}} = 15 \text{ mg/L}$  as an example, the steady state is achieved after 2 hrs according to  
23 the model (Fig. 5(a)). The total particle number  $m_0$  reduced to 0.015 % of initial  $m_0$  at steady state.

1 The total particle volume  $m_3$  was unchanged for volume conservation in 0-D model. The moments  
2  $m_1$  and  $m_2$  decrease with time until arriving at steady state, since aggregation dominates at the  
3 beginning and breakage becomes comparable with aggregation after large floppy flocs occurring.  
4 The fourth moment  $m_4$  is integrated from the simulated FSDs at each time step. For spherical  
5 particles,  $m_4$  is proportional to the total surface area of particles settling per unit time (Mehta, 2013).  
6 Therefore, they are increased due to flocculation until a steady state. The time evolutions of FSDs  
7 are displayed every 15 minutes until steady state (Fig. 5(b)). It shows how a bimodal FSD  
8 generated from point distributed primary particles. Firstly, the major peak remains close while the  
9 standard deviation rapidly increases to yield a wider distribution. After that, the major peak swifts  
10 to the right side and microflocs dominated because of aggregation. Then, a second peak becomes  
11 obvious with time, indicating the occurrence of large macroflocs. The effect of breakage become  
12 important, since larger flocs with more constitute primary particles are easier to destroy. Finally,  
13 the aggregation and breakage arrive at equilibrium to form a FSD with two major peaks.

14

#### 15 4.2 Comparison with settling column experiment: 1-D test

16 By trial-and-error, the coefficients  $\alpha = 0.95$  and  $E_b = 1.05 \times 10^{-5}$  were selected to best fit  
17 the FSDs observed at the measuring section ( $Z = 0.75$  m). The parameter  $\alpha_0$  which controls the  
18 spread of simulated FSDs (Eq. 10) was set as 0.195. Other parameters for the best-quality  
19 simulations are summarized in Table 2. The case of shear rate  $G = 10 \text{ s}^{-1}$  was used for calibration,  
20 while  $G = \{20, 40\} \text{ s}^{-1}$  were adopted for validation. It can be shown that the predicted and observed  
21 FSDs at the measuring section reasonably match for  $G = \{10, 20, 40\} \text{ s}^{-1}$  (Fig. 6). It shows a  
22 significant improvement in comparison with that given by Maggi et al. (2007) or Shen and Maa  
23 (2015). The fitted FSDs in Fig. 6 are based on a superposition of two lognormal constituted FSDs

1 with different variances by the software DistFit (Chimera Technologies, USA) to better represent  
2 the observations. Notice that for  $G = 5 \text{ s}^{-1}$ , the low oscillating frequency ( $\sigma_g < 0.1 \text{ Hz}$ ) might lead  
3 to a non-homogenous mixing (Maggi, 2005). Therefore, a reasonable agreement was only attained  
4 by recalibrating the model by a larger collision efficiency  $\alpha = 1.1$  whereas a smaller variance with  
5  $\alpha_0 = 0.175$  (Fig. 6(d)). Note that by definition the  $\alpha$  should be less than unity; however,  $\alpha > 1$  is  
6 possible for flocs with low fractal dimensions (Lee et al., 2000; Shen and Maa, 2015). For all the  
7 cases, the major peaks of the FSD are properly addressed against measured value, while the  
8 secondary peaks for the macroflocs are more or less deviated from the observations. This is  
9 because of the assumption of a common standard deviation  $\sigma$ . The common  $\sigma$  should compromise  
10 a large  $\sigma$  of microflocs and a small  $\sigma$  of macroflocs in this application.

11 For  $G = 10 \text{ s}^{-1}$ , the vertical profiles of SSC, weighted settling velocity  $w_s$ , total particle  
12 number  $m_0$  and the arithmetic mean diameter  $d_{1,0}$  ( $d_{1,0} = m_1/m_0$ ) along the column are examined.  
13 The SSC in the upper column decreases as particles are aggregated and settled to the lower section  
14 (Fig. 7(a)). Although sediments at the top are operated by a buffer tank to maintain a constant SSC  
15 around 0.5 g/L, a minimum SSC occurs close to the surface ( $Z = 4.5 \text{ m}$ ) with the SSC decreasing  
16 to half of the initial value. The concentration at the measuring section does not vary significantly,  
17 while the SSC at the bed section largely increases ( $> 1 \text{ g/L}$ ) since the settled large flocs pass  
18 through the measuring section and are concentrated on the bottom. At the first few minutes, the  
19 settling velocities  $w_s$  at the bed section ( $Z < 1 \text{ m}$ ) are smaller than  $w_s$  at the upper section ( $Z = 1 -$   
20  $4.5 \text{ m}$ ) (Fig. 7(b)). This is because the oscillating grids only lay in the settling section, and thus  
21 shear rate is largely reduced at the bed section so that it cannot promote flocculation at the  
22 beginning. With time, however, the  $w_s$  at the bottom section increases since larger flocs are settled  
23 to the bottom, which also leads to an increase of particle diameter  $d_{1,0}$  at the bed section. The

1 deviation of settling velocity at the measuring section is small, and the  $w_s$  gradually decreases until  
2 the surface where primary particles are released from the buffer tank. The relative total particle  
3 number  $m_0(t)/m_0(t=0)$  decreases to 1‰ (Fig. 7(c)) in the measuring section, while the mean size  
4  $d_{1,0}$  increases to 45  $\mu\text{m}$  at that location (Fig. 7(d)).

5 Also for  $G = 10 \text{ s}^{-1}$ , the variation of SSC is merely 5 % at the measuring section ( $Z = 0.75$   
6 m) (Fig. 8(a)), which more or less confirms the assumption of a “constant” sediment concentration.  
7 At column heights at the settling section, however, the sediment concentrations obviously alter  
8 with time. During the simulating period, the concentration at  $Z = \{1.75, 2.75, 3.75\}$  m continuously  
9 decreases. Close to the surface ( $Z = 4.75$  m), the SSC decreases to 0.32 g/L during the first half an  
10 hour and becomes steady thereafter. At all column heights, the mean size  $d_{1,0}$  increases at the  
11 beginning as particle growing, and then arrives at steady state after half an hour (Fig. 8(b)). The  
12 settling velocities  $w_s$  (Fig. 8(c)) and standard deviation  $\sigma$  follow the same trend of  $d_{1,0}$  (Fig. 8(d)).  
13 In less flocculated areas ( $Z = 4.75$  m) close to the surface, the  $d_{1,0}$ ,  $w_s$  and  $\sigma$  reach equilibrium much  
14 faster than in well flocculated areas ( $Z = 0.75$  m).

15 The FSDs against different SSCs are also predicted in Fig. 9. A higher SSC of 0.75 g/L  
16 may increase the mean diameter  $d_{1,0}$  up to 57.0  $\mu\text{m}$ , while a SSC of 0.25 g/L will lower  $d_{1,0}$  down  
17 to 32.5  $\mu\text{m}$  (Fig. 9). This is because the flocculation rate is proportional to the particle  
18 concentrations, with higher concentrations resulting in high possibilities of collision. For  $G = 10$   
19  $\text{s}^{-1}$  with different concentrations  $c = \{0.25, 0.5, 0.75\}$  g/L, the sizes of microflocs  $L_1$  are close ( $L_1$   
20  $= 21.1 \pm 2.6 \mu\text{m}$ ). Larger standard deviations ( $\sigma = \{0.42, 0.49, 0.54\}$ ) are expected for higher SSCs.  
21 Both the sizes and weights of macroflocs increase as  $c$  increases ( $L_2 = \{53.1, 59.2, 62.7\} \mu\text{m}$  and  
22  $w_2 = \{0.27, 0.48, 0.7\}$ ). These predictions can be tested in future experiments.

23

### 1 4.3 Model evaluation

2           In this study, the applications of the E-QBMM are based on the observations that the in-  
3 situ FSD can be decomposed into a limited number of lognormal distributions. In general, four  
4 component lognormal FSDs are sufficient to represent a measured FSD (Lee et al., 2012) to  
5 identify groups of primary particles, microflocs, macroflocs and megaflocs. This means, only a  
6 maximum of eight tracers (i.e., their weights and representative sizes) with a common standard  
7 deviation are required in a large scale model. To further reduce the number of tracers for exhibiting  
8 a multimodal FSD, as low as four tracers can be employed as used in this study. Compared with  
9 previous two- or three-class PBEs, this model provides a straightforward percept of the entire  
10 continuous FSD instead of only discrete representative size groups. It avoids reformulation of the  
11 flocculation source and sink terms for different number of size groups, but adopts comparable  
12 number of tracers which will not significantly increase the computational demands. After validated  
13 by more field data, this model has the potential to simulate the spatially and temporally varied,  
14 continuous, and multimodal FSDs within the framework of large-scale simulations.

15           Although this method is efficient and powerful to deal with the entire FSD, it has a few  
16 weaknesses based on the assumptions that all the subordinate FSDs share the same standard  
17 deviation  $\sigma$ . The purpose of this selection is to reduce the number of tracer, and to make it possible  
18 to use the Wheeler's method to solve the "well-studied" low order non-linear equation system (Eq.  
19 13). However, from Lee et al. (2012) we know that the standard deviations of the constituted FSDs  
20 sometimes are largely different. A single  $\sigma$  is merely chosen by matching the simulated and  
21 observed FSD, without exploring its physical meaning by investigating the change of  $\sigma$  with  
22 environmental parameters (such as shear rate, salinity, temperature and bioactivities). In fact, the  
23 standard deviation  $\sigma$  used to characterize the width of the distribution in the model (or in the in

1 situ FSD) should also include non-spherical characters of the particles and is thus not only  
2 indicating different sizes of particles. Furthermore, a standard deviation  $\sigma$  does not always exist.  
3 Even treating  $\sigma$  as an additional tracer, none of the iteration methods (Madadi-Kandjani and  
4 Passalacqua, 2015), the Ridders' method (Press et al., 1992) or the Brent's method (Press et al.,  
5 1992) proposed by recent studies could guarantee a realistic  $\sigma$ . Physically it means that the code  
6 fails to find a single common  $\sigma$  to compromise all the component FSDs when more subordinates  
7 are included. Thus,  $N = 2$  is a practical selection on the first stage. A standard deviation  $\sigma$  derived  
8 from the  $(2N)$ th order moment of the FSD (Madadi-Kandjani and Passalacqua, 2015; Nguyen et  
9 al., 2016; Passalacqua et al., 2018), if applicable, results in a narrow spread of FSD in our tests.  
10 This may be because  $m_{2N}$  highlights larger particles and underestimates the value of  $\sigma$  due to  
11 rounding-off errors. This part can be improved if a better method to find the optimal  $\sigma$  to minimize  
12 the simulated and measured FSDs is available. It is also noticeable that the subordinate lognormal  
13 FSD assumption is based on the data collected in well-mixed Belgian coastal zones (Lee et al.,  
14 2012; Fettweis and Lee, 2017; Fettweis et al., 2016). Few measured data from other areas,  
15 especially stratified regions, are available to evaluate this decomposition. Another issue is that the  
16 current flocculation kinetic equations are decoupled with transport terms, and are always solved  
17 explicitly in each time step. That is, at time  $t$ , the tracer source and sink terms are estimated using  
18 the values at time  $t-dt$ . Therefore, better numerical schemes should be developed to accommodate  
19 both explicit and implicit time stepping.

20         Moreover, although flocculation dynamics in rivers and estuaries is strongly dependent on  
21 microbial activities (Wolanski and Elliott, 2016), this model does not explicitly include these  
22 effects, because it may result in an endless chain of processes at the current stage. Actually, even  
23 the most recently published papers still mainly focus on clean sediments without biology (e.g.,



1 Cuthbertson et al., 2018; Mhashhash et al., 2018; Tran et al., 2018; Zhang et al., 2018; Zhu et al.,  
2 2018). In fact, even easier parts of ecosystems, such as organic matter, phytoplankton and biofilms,  
3 are still not fully understood and quantifiable. It is possible to include an ecohydraulics library,  
4 such as AED2 (the Aquatic EcoDynamics modeling library,  
5 <http://aed.see.uwa.edu.au/research/models/AED/>), which can be used by the TELEMAC system  
6 (<http://www.opentelemac.org/>), but the chance for gain in accuracy is only achieved by decreasing  
7 the involved processes, due to their high degree of empiricism and uncertainty on the model  
8 parameters. Indeed, the accumulation of errors due to parameter uncertainty may outweigh the  
9 hoped-for increase in accuracy by including more processes. This problem of overparameterization  
10 is well documented in other fields (Reichert et al., 1996; Schoups et al., 2008)

11 It is also critical to note that there are different methods to measure in situ FSDs but they  
12 may not give the same results. The size is only well defined if the particle is a sphere. However,  
13 natural particles are seldom spheres and have irregular shapes and this introduces - when measured  
14 - intrinsically a size distribution. If we measure the size of a large amount of the similar but  
15 irregular particles (e.g., ellipsoids), the result is a distribution. A model that has been calibrated for  
16 a LISST derived FSD will not be able to reproduce the same flocs measured by a digital camera  
17 (e.g., Mikkelsen et al., 2005), meaning that the outcome of even the best flocculation model is only  
18 as good as the measuring system that is used to collect the FSDs. The measuring technique has  
19 weaknesses that are reflected in the FSDs. Generally camera systems cannot resolve the fine  
20 particles smaller than 10  $\mu\text{m}$ , while LISST has a limited size range for the fine and the very large  
21 particles. Out of range particles are influencing the size distribution. Nowadays, there is still no  
22 good way for correcting FSDs for these spurious data, but it should be aware that the very large  
23 (megaflocs) and the very small particles (primary particles) maybe under-represented or over-

1 represented in the in situ LISST derived FSDs. In reality, even if the size distributions in situ and  
2 in the model are well resolved, there are still uncertainties involved in the estimation of the density  
3 and the settling velocity (the ultimate parameter for the model). For example, when using fractal  
4 theory to estimate the floc density, small changes in fractal dimension may induce large changes  
5 in the settling velocity. The calculation of the settling velocity is still subject to calibration (e.g.,  
6 by using SSC values). Given the uncertainty of the measured FSDs, further studies are required to  
7 better represent particles from measuring points.

8

## 9 **5. Conclusions**

10 In this study, the multimodal FSDs of suspended cohesive sediments are successfully  
11 predicted by using the E-QBMM with sediment flocculation kinetics, assuming the target FSDs  
12 consisting of a set of subordinate lognormal distributions. The main conclusions are:

13 (1) Earlier studies usually fail to simulate the entire FSDs of the cohesive sediments in aquatic  
14 environments even with a large number of size classes. In this study, however, the FSDs are  
15 reasonably represented by the weight ( $w_i$ ) and the representative size ( $L_i$ ) of each component  
16 lognormal FSD, with a common standard deviation ( $\sigma$ ).

17 (2) In our validations against two laboratory experiments, two subordinate lognormal distributions  
18 for microflocs and macroflocs are employed to reproduce the observed FSDs in the mixing  
19 chamber and the settling column. This selection only introduces four tracers (i.e.,  $L_1$ ,  $L_2$ ,  $w_1$  and  
20  $w_2$ ), makes this method efficient, and has the potential to be implemented into large-scale sediment  
21 transport models.

22 (3) The common standard deviation  $\sigma$  is modeled empirically as a function of the mean size of the  
23 entire FSD, the elementary particle size and a constant. In our applications, better agreements of

1 the modeled and measured FSDs are achieved by using an empirical  $\sigma$  rather than extracting  $\sigma$   
2 from the  $(2N)$ th moment of FSD.

3 (4) High accuracy and robustness is not necessarily guaranteed if representing a simple FSD with  
4 excessive subordinate FSDs. It depends

5 on whether the shape of FSD is truly suitable to decompose into sub-components with the same  $\sigma$ .

6 (5) Future studies are needed to investigate the subordinate FSDs in stratified estuaries. Microbial  
7 processes or a second internal property (such as the floc density) in the PBE might be helpful to

8 better address the properties of biomineral suspended particulate matters. Besides, numerical  
9 schemes of both explicit and implicit treatment of the flocculation kinetic terms are expected to

10 coincide with the transport terms of the fluid and carrying particles.

11

## 12 **Acknowledgement**

13 We would like to thank three anonymous reviewers for their constructive comments. This

14 research was funded by the BelSPO (Belgian Science Policy Office) within the framework of the

15 BRAIN-BE (Belgian Research Action through Interdisciplinary Networks) INDI67 project

16 (Developments of tools and methods to support the monitoring of EU Marine Strategy Framework

17 Directive indicators 6 ‘Sea-floor integrity’ and 7 ‘Hydrographical conditions’ - Grant no.

18 BR/143/A2/INDI67) and the JPI-OCEANS (The Joint Programming Initiative Healthy and

19 Productive Seas and Oceans) WEATHER-MIC (How microplastic weathering changes its

20 transport, fate and toxicity in the marine environment – Grant no. BR/154/A1/WEATHER-MIC). )

21 project. The first author was also supported by the Open Research Fund of State Key Laboratory

22 of Estuarine and Coastal Research of China (Grant No. SKLEC-KF201811).

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1 **Table 1** Summary of model parameters and results for different guar gum dosages in the mixing  
 2 jar tests, with initial suspended sediment concentration  $c = 0.52$  g/L and tank averaged shear rate  
 3  $G = 55$  s<sup>-1</sup>.

Case	$C_{\text{guar}}$ (mg/L)	$\alpha$	$E_b$	$\alpha_0$	$n_f$	$L_1$ ( $\mu\text{m}$ )	$L_2$ ( $\mu\text{m}$ )	$w_1$	$w_2$	$\sigma$	$D_{50}$ ( $\mu\text{m}$ )	FSD Error
1	0	0.8	4.8E-6	0.162	2.8	32.9	110.0	0.73	0.27	0.5874	21.6	0.0714
2	5	0.8	3.5E-6	0.162	2.7	34.2	104.3	0.65	0.35	0.5794	23.7	0.0552
3	10	0.8	2.4E-6	0.166	2.65	31.9	100.5	0.45	0.55	0.6168	24.4	0.0490
4	15	0.8	1.0E-6	0.150	2.6	37.1	151.2	0.31	0.69	0.6297	32.6	0.0587
5	20	0.8	2.0E-6	0.165	2.7	29.8	113.3	0.34	0.66	0.6605	24.4	0.0367
6	30	0.8	3.9E-6	0.181	2.8	25.1	85.8	0.43	0.57	0.6872	18.3	0.0285

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1 **Table 2** Parameters used in the best-quality simulation for the settling column experiment.

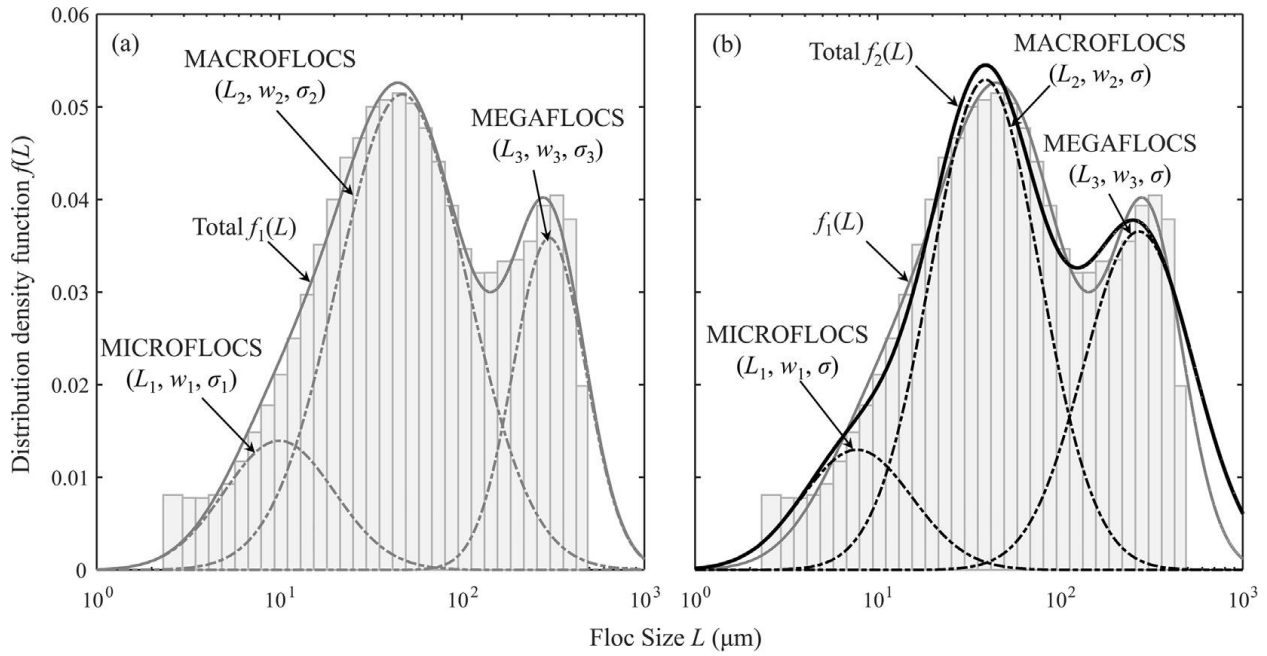
Symbol	Value	Description
$c$	0.5	Initial suspended sediment concentration (g/L)
$G$	5, 10, 20, 40	Shear rate generated by the oscillating grids in the settling section ( $s^{-1}$ )
$N_d$	2	Number of lognormal distributions
$N_{di}$	3	Number of pivots for each subordinate lognormal distribution
$\alpha$	$0.95^\dagger$	Collision efficiency
$E_b$	$1.05E-5$	Breakage fitting parameter
$\alpha_0$	$0.195^\dagger$	Coefficient for computing the shared standard deviation for all subordinate lognormal distributions
$n_f$	2.3	Fractal dimension
$K$	5	Coefficient in the fragmentation distribution function
$l_p$	8	Size of primary particles ( $\mu\text{m}$ )
$c_{gel}$	40	Gelling concentration (g/L)
$F_y$	$1.0E-10$	Floc strength (Pa)
$\Delta t$	0.1	Time step (s)
$\Delta z$	0.1	Vertical resolution (m)
$n_z$	50	Number of cells in the vertical direction.

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3  $^\dagger$  For shear rate  $G = 5 \text{ s}^{-1}$ , model results are recalibrated by setting  $\alpha = 1.1$  and  $\alpha_0 = 0.175$ .

1 **Figures**

2

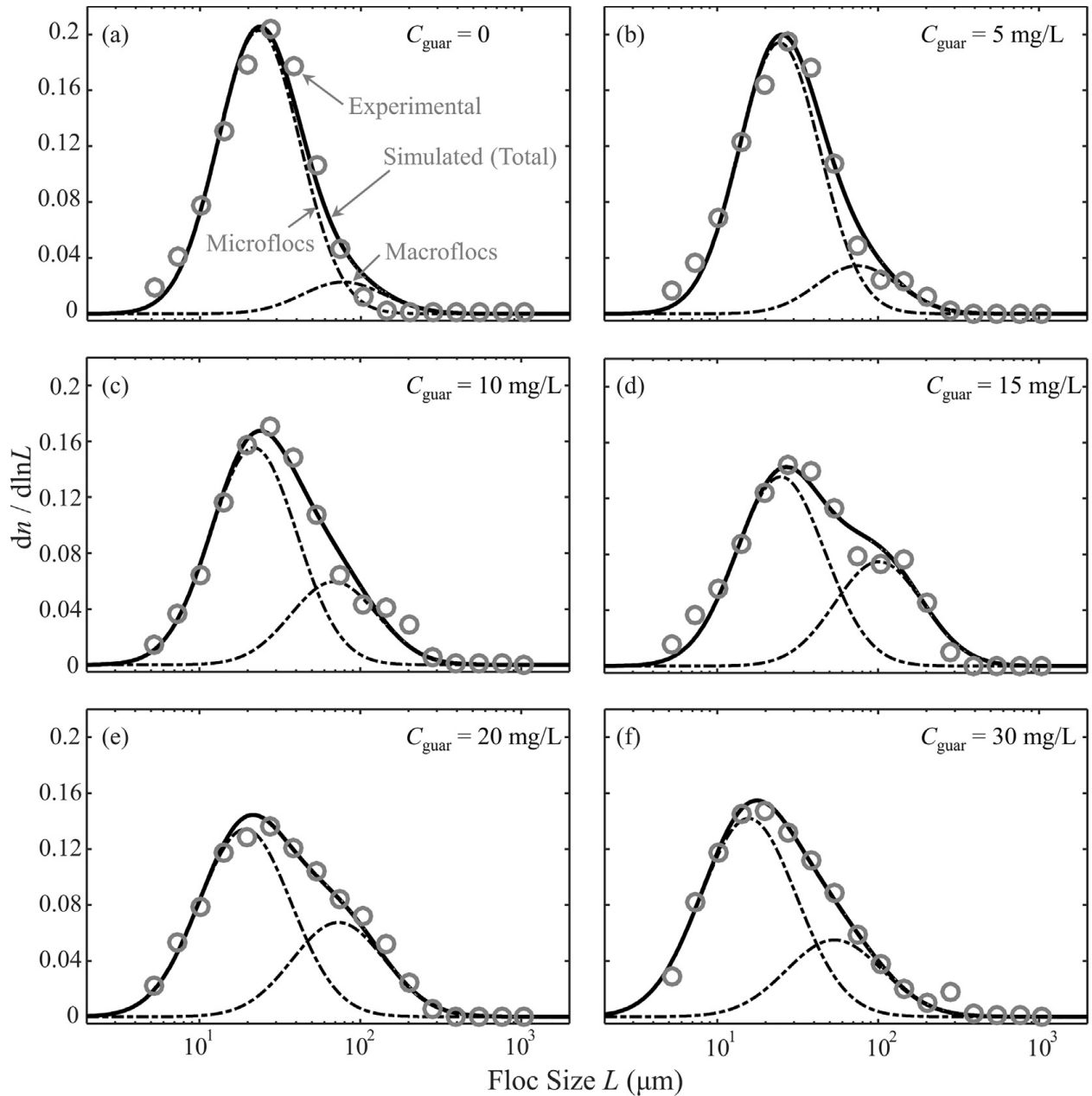


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4 Fig. 1 An example of decomposing a measured FSD into three lognormal distributions for  
 5 microflocs  $L_1$ , macroflocs  $L_2$  and megaflocs  $L_3$ . Subordinate FSDs (a) with varying standard  
 6 deviations  $(\sigma_1, \sigma_2, \sigma_3)$  and (b) with a common standard deviation  $\sigma$  are compared. The bars represent  
 7 a typical FSD collected by LISST at site MOW1 at Belgian coast. Dashed lines are subordinate  
 8 FSDs and the solid line is the superposed FSD. The entire FSD  $f_1(L)$  in subfigure (a) is also plotted  
 9 in subfigure (b) for comparison.

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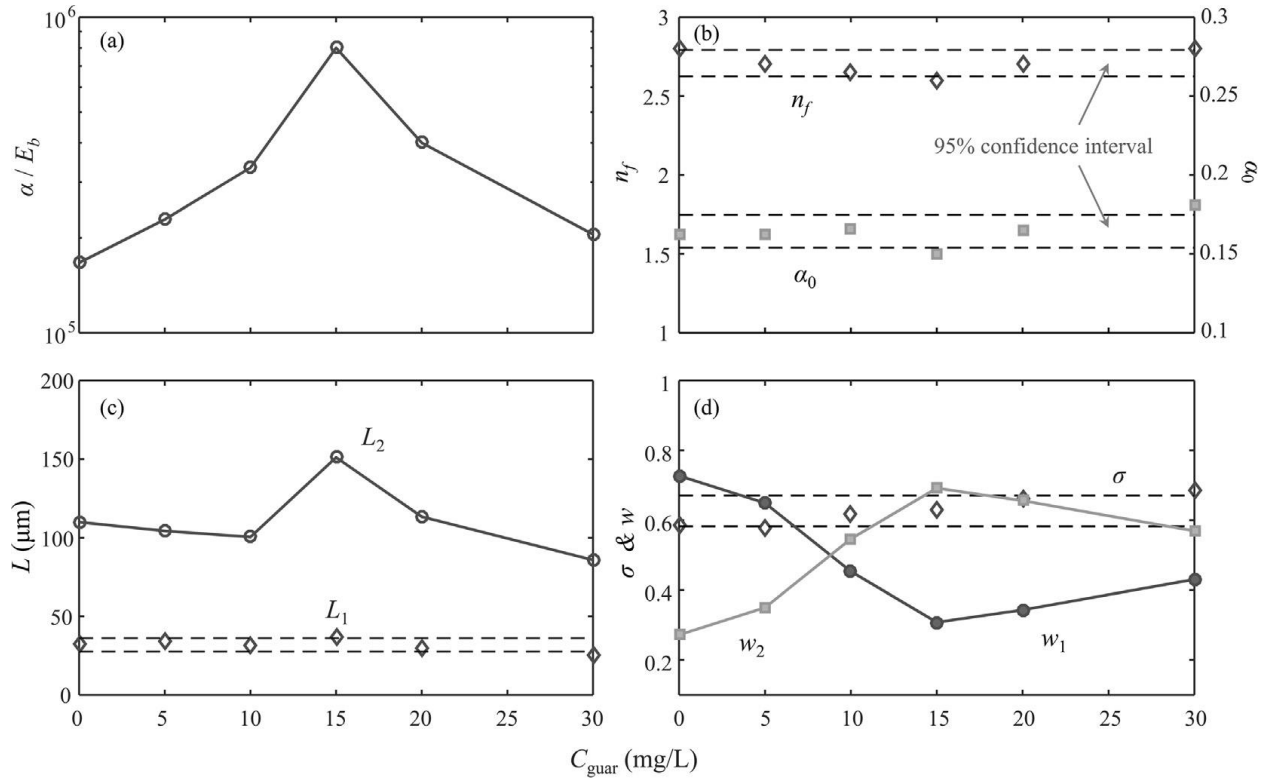
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2 Fig. 2 Comparison of simulated and observed FSDs for guar gum concentration  $c_{\text{guar}} = \{0, 5, 10,$   
 3  $15, 20, 30\}$  mg/L in the mixing jar experiments. Symbols are measurements and dark solid lines  
 4 are the modeled FSDs, with two subordinate lognormal distributions marked with dashdotted lines.

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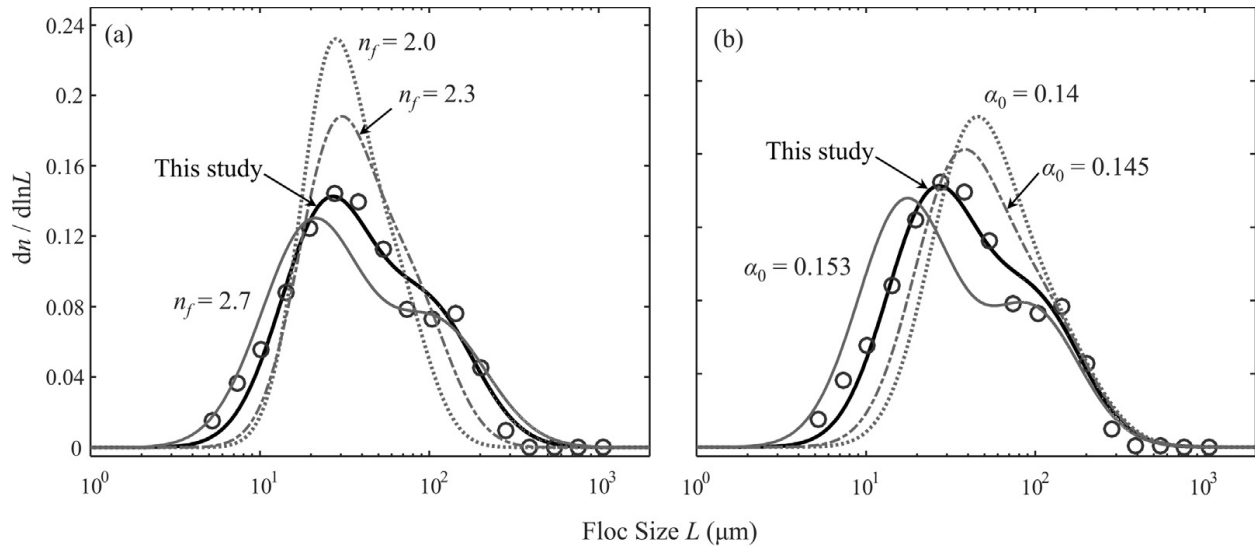
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Fig. 3 Model parameters of (a) the ratio  $\alpha/E_b$ , (b) the fractal dimension  $n_f$  and the standard deviation computing coefficient  $\alpha_0$ , (c) the predicted representative sizes  $L_i$ , and (d) the corresponding weights  $w_i$  and the common standard deviation  $\sigma$ , for  $c_{\text{guar}} = \{0, 5, 10, 15, 20, 30\}$  mg/L in the mixing jar experiments.



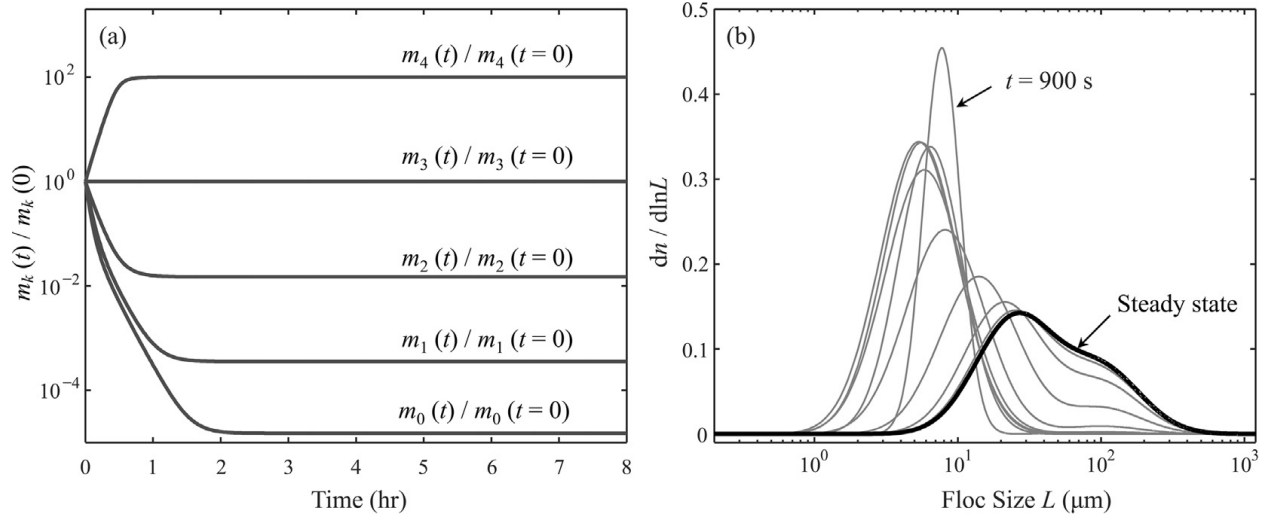
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2 Fig. 4 Sensitivity tests of (a) fractal dimension  $n_f$  and (b) standard deviation computing coefficient

3  $\alpha_0$ , for  $c_{\text{guar}} = 15 \text{ mg/L}$  in the mixing jar experiment.

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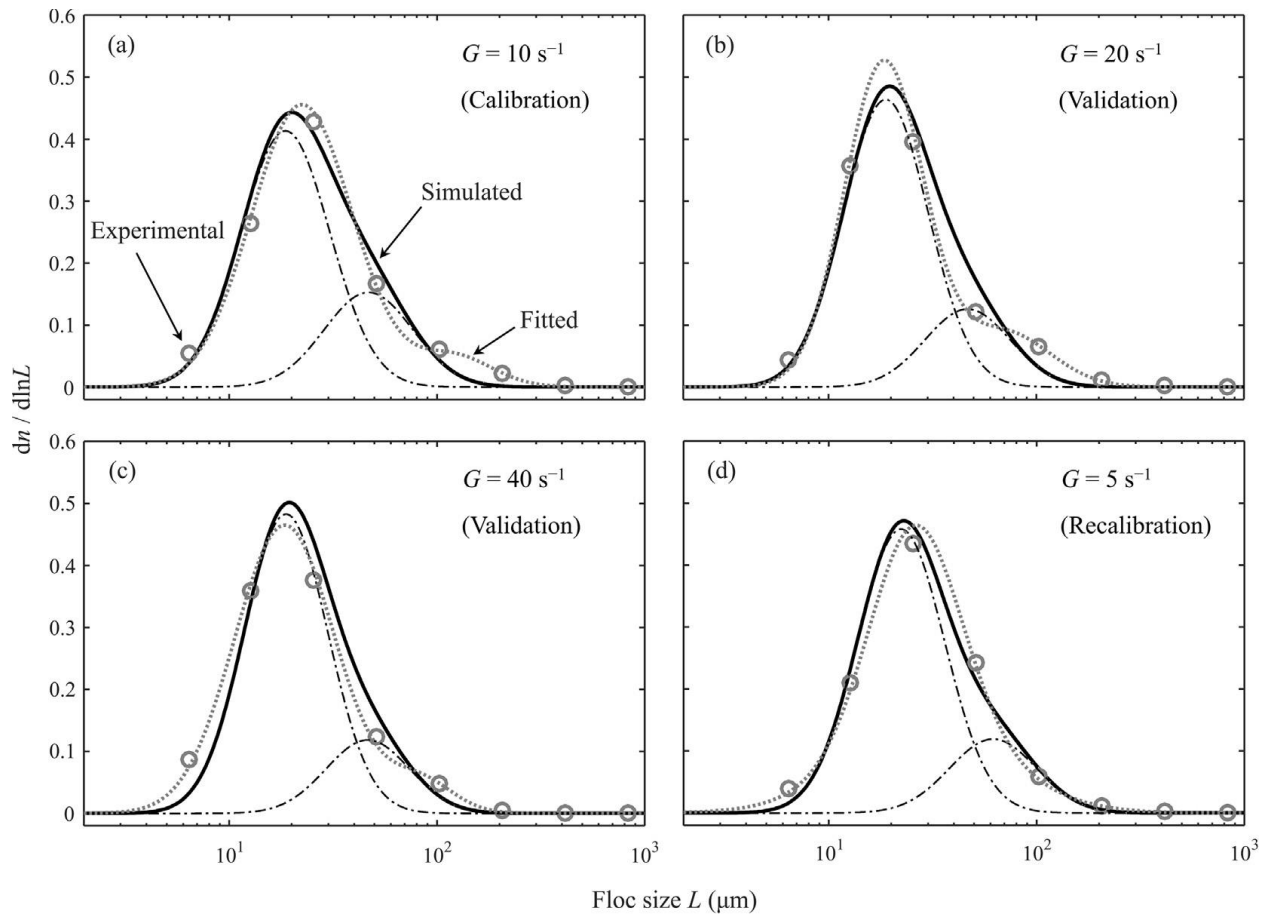


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2 Fig. 5 (a) Time evolutions of the predicted moments  $m_k$  ( $k = 0, 1, 2, 3, 4$ ) and (b) the predicted

3 FSDs shown every 15 mins, for  $c_{\text{guar}} = 15$  mg/L in the mixing jar experiment.

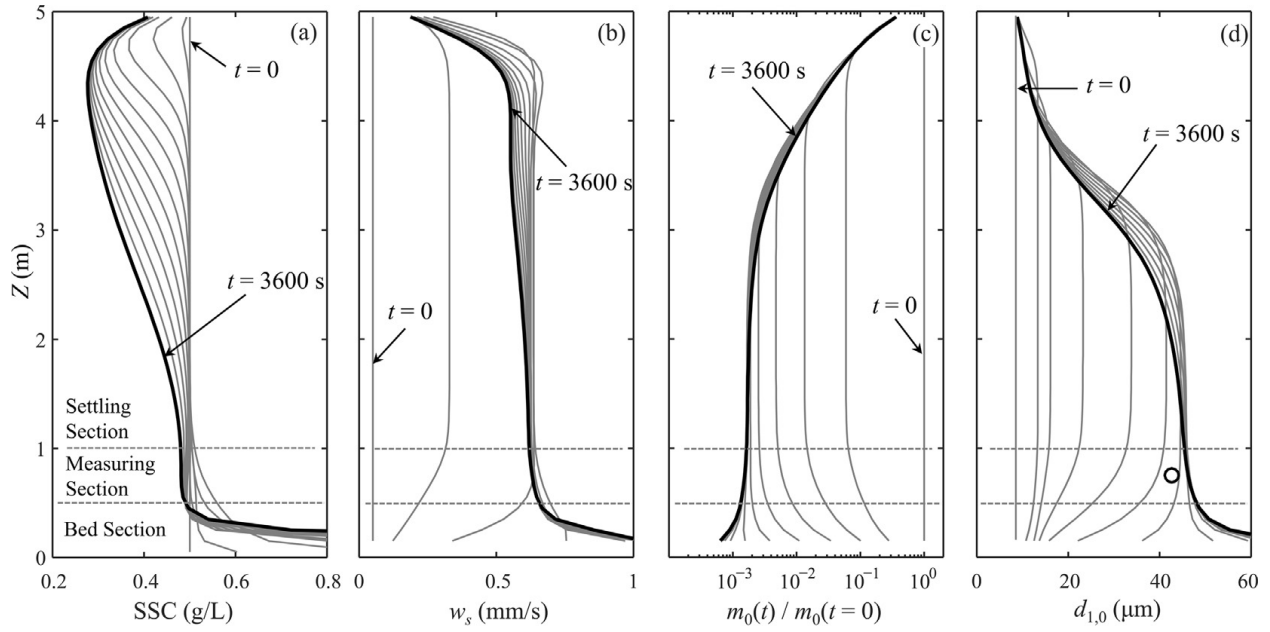
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2 Fig. 6 Comparison of simulated and measured FSDs in the settling column tests for initial  
 3 sediment concentration  $c = 0.5 \text{ g/L}$  and shear rate  $G = \{5, 10, 20, 40\} \text{ s}^{-1}$ . Symbols are  
 4 measurements, dotted lines are fitted FSDs by software DistFit (treated as reference FSDs), dash-  
 5 dotted lines are two modeled subordinate FSDs and the dark solid lines are the superposed  
 6 predictions.

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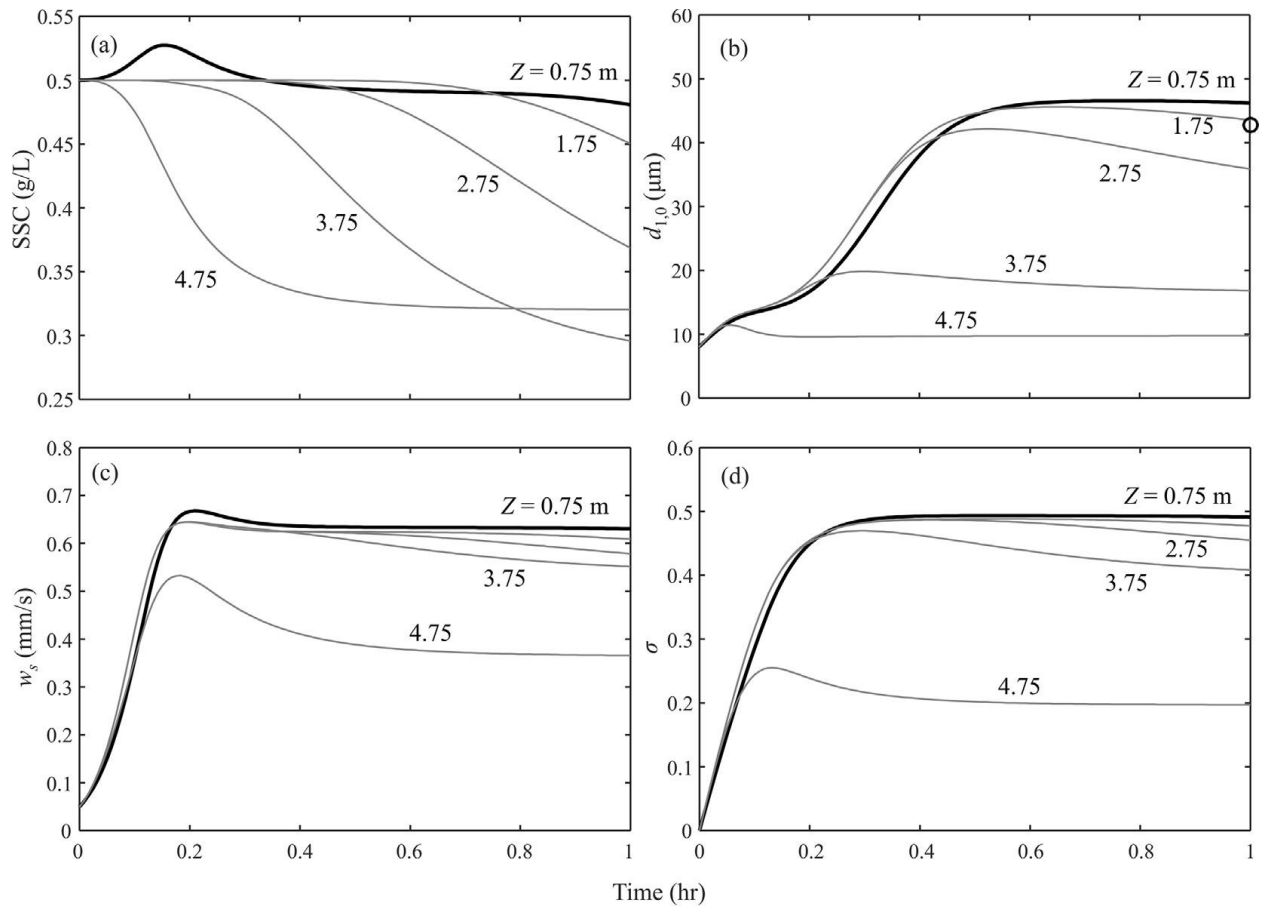


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2 Fig. 7 Vertical profiles of (a) SSC, (b) settling velocity ( $w_s$ ), (c) total particle number ( $m_0$ ) and (d)

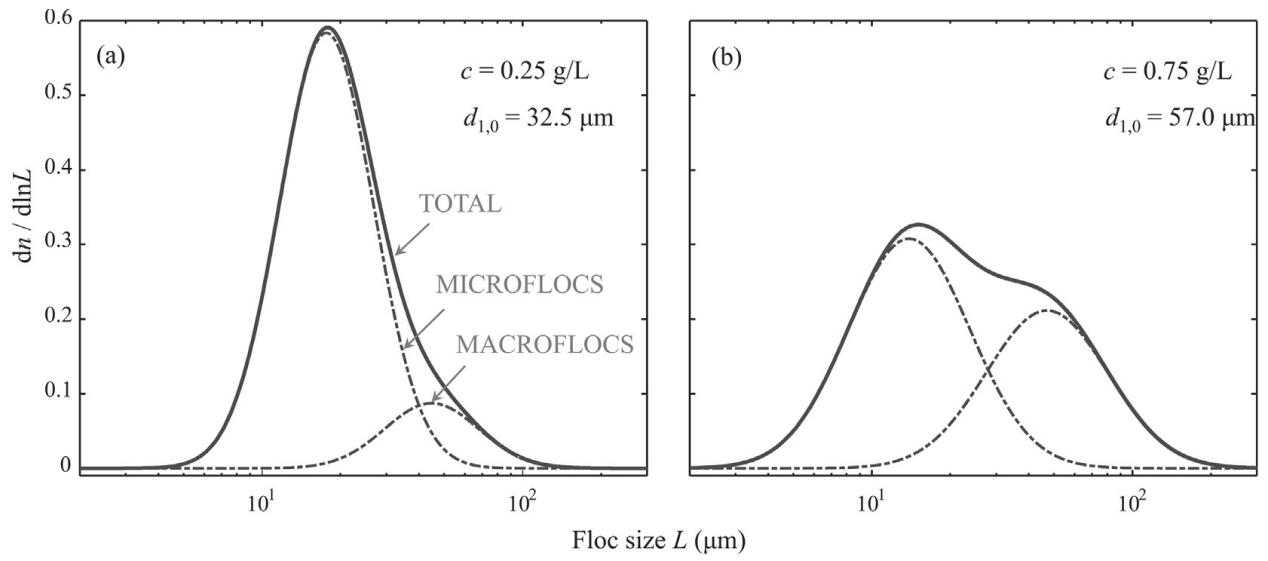
3 mean size ( $d_{1,0}$ ), for shear rate  $G = 10 \text{ s}^{-1}$  in the settling column experiment.

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Fig. 8 Time evolutions of (a) SSC, (b) means size ( $d_{1,0}$ ), (c) settling velocity ( $w_s$ ) and (d) standard deviation ( $\sigma$ ) at column height  $Z = \{0.75, 1.75, 2.75, 3.75, 4.75\}$  m, for shear rate  $G = 10 \text{ s}^{-1}$  in the settling column experiment. The symbol in subfigure (b) is the observed  $d_{1,0}$  at the measuring section.



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2 Fig. 9 Predicted FSDs for initial sediment concentrations (a)  $c = 0.25 \text{ g/L}$  and (b)  $c = 0.75 \text{ g/L}$ .

3 The dark solid line is the modeled FSDs, with two subordinate FSDs marked with dash-dotted  
 4 lines.

5