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Kinematic assessment of floc formation using a Monte Carlo model

Évaluation cinématique de la brisure de flocons avec un modèle de Monte Carlo

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ABSTRACT
A simple stochastic model is proposed to simulate floc formation due to simultaneous aggregation and breakage processes. The model is based on the constant-number Monte Carlo method where the number of flocs is kept constant during simulations. To produce equilibrium floc-size distributions, it uses established models of flocculation and new simple formulations of breakage probability and of the probability of producing fragments of a given size from broken flocs. The concept of fractal geometry is used to describe the geometry of flocs. The maximum size of flocs allowed, the median size of component particles, and their density are the main inputs needed to simulate floc formation. Simulated steady-state floc-size distributions were compared with field data observed at different locations, and good agreement was obtained. Dimensional analysis applied to measured and simulated data revealed that floc-size distributions are self-similar and can be described by the same function, regardless of the conditions of their formation.

RÉSUMÉ
On propose un modèle stochastique simple pour simuler une formation de floculation due à des processus simultanés d'agrégation et de rupture. Le modèle est basé sur la méthode de Monte Carlo à nombre constant où le nombre de flocons est maintenu constant pendant les simulations. Pour obtenir des distributions de taille de flocons à l'équilibre, on utilise des modèles établis de floculation et de nouvelles formulations simples pour la probabilité de rupture et la probabilité de produire des fragments d'une taille donnée à partir flocons brisés. Un concept de géométrie fractale est employé pour décrire la géométrie des flocons. La taille maximum admise pour les flocons, la taille médiane et la densité des particules qui les composent sont les principales données requises pour simuler leur formation. Les distributions simulées de taille de flocons à l'état stationnaire ont été comparées à des données en nature observées à différents endroits, et on a obtenu une bonne concordance. L'analyse dimensionnelle appliquée aux données mesurées et les simulées a montré que les distributions de taille de flocons sont auto-semblables et peuvent être décrites par la même fonction, indépendamment des conditions de leur formation.

Keywords: Monte Carlo simulation, flocs, aggregation, breakage, fractal dimension, size distribution, sediment transport, cohesive sediment.

1 Introduction

Fine particles in freshwater and marine environments aggregate and adhere to form fragile, porous agglomerations called flocs. Flocs have been observed in many settings, including stormwater ponds (Krishnappan *et al.*, 1999), water treatment facilities (Spicer and Pratsanis, 1996), lakes (Tsai *et al.*, 1987; O'Melia, 1990), rivers (Droppo and Ongley, 1994; Phillips and Walling, 1999; Krishnappan, 2000; Geyer *et al.*, 2004), deltas (Fox *et al.*, 2004), estuaries (Burban *et al.*, 1989; Eisma *et al.*, 1991; van Leussen, 1999; McAnally and Mehta, 2000; Milligan *et al.*, 2001; Manning and Dyer, 2002), continental shelves (Sternberg *et al.*, 1999; Hill *et al.*, 2001), and the open ocean (Lampitt *et al.*, 1993; Dierks and Asper, 1997). Incorporation of particles into flocs affects the optical and acoustical properties of the aqueous sediment suspensions in which flocs are found (Campbell and Spinrad, 1987; Hatcher *et al.*, 2001; Fugate and Friedrichs, 2002; Flory *et al.*, 2004), and it influences the transport and fate of particles and the substances attached to them (Kranck, 1973; Milligan and Loring, 1997; Hill *et al.*, 2000). Despite the importance of flocs to a variety of disciplines, models for predicting the size distribution of flocs are plagued by uncertainty regarding key rate processes. Specifically, an equilibrium floc size distribution is defined in large part by the competition between aggregation and breakage, but the rates at which these processes occur are poorly known (e.g., Hill and Nowell, 1995; Winterwerp, 1998).

The physical processes that lead to collision of particles in suspension are well understood quantitatively, but the fraction of total collisions that result in contact and sticking is not well known (Hill and McCave, 2001). Particles are brought into close

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proximity with other particles by Brownian motion, turbulent shear, and differential settling, and expressions for the rate at which collisions occur by these mechanisms have been available for decades (McCave, 1984). Once particles are in close proximity, however, hydrodynamical and electrochemical interactions become complex, severely limiting the predictive power of models of aggregation rate (Hill, 1992; Stolzenbach and Elimalech, 1994; Li and Logan, 1997a,b; Thomas *et al.*, 1999).

Floc breakage is the disruption of flocs by stresses created by relative floc-fluid motion generated by turbulent fluctuations or by particle settling (e.g., Pandya and Spielman, 1982; Adler and Mills, 1979; Hunt, 1986). The rates and the dominant mechanisms of floc breakage are not well understood, nor even agreed upon (Hunt, 1986; Dyer, 1989; Alldredge *et al.*, 1990; Milligan and Hill, 1998; Hill *et al.*, 2001). Models for floc breakage are complex, contain many adjustable parameters, and have not always been tested systematically against observations (e.g., Pandya and Spielman, 1982; Lu and Spielman, 1985; Ray and Hogg, 1987; Tsao and Hsu, 1989; Hsu and Tsao, 1992; Jackson, 1995; Hill, 1996; Spicer and Pratsinis, 1996; Kramer and Clark, 1999; McAnally and Mehta, 2000; Burd and Jackson, 2002; Ducoste, 2002; Lasheras *et al.*, 2002; Nopens *et al.*, 2002; Odriozola *et al.*, 2002; Kostoglou, 2003).

Given the uncertainty surrounding aggregation and breakage rates, a dynamic model of equilibrium floc size distribution that is free of adjustable parameters and their attendant uncertainties is not available at present. Furthermore, the complexity of these processes weakens the prospects for the timely development of such a model. This situation does not preclude progress in understanding of the factors that define floc size distributions, but it does demand alternative approaches to the problem.

One issue amenable to progress with alternative approaches is the size distribution of daughter fragments produced by disruption of a floc. The probability of producing fragments of a given size is a key parameter in mechanistic models of floc breakage (Pandya and Spielman, 1982; Hill, 1996), but little is known about it. Information regarding the form of this probability function may be gained, however, through comparison of observed floc size distributions with modeled floc size distributions based on a kinematic definition of breakage.

A typical kinematic treatment of floc breakage does not specify the forces responsible for breakage, but rather defines a maximal stable floc size. With this approach, rules are needed to determine the fate of flocs that exceed the maximal stable size. A common approach stipulates that whenever two particles aggregate to form a floc larger than the maximal allowed size, any particle mass involved in the collision simply is assigned to the size class that includes the maximal floc size (e.g., Hill and Nowell, 1995). This approach results in the eventual accumulation of all particle mass in maximally sized flocs. This result is unsatisfactory because observations from numerous environments show a range of floc sizes in suspension (Eisma *et al.*, 1991; Syvitski *et al.*, 1991; Kranck and Milligan, 1992; Jackson *et al.*, 1997; Syvitski and Hutton, 1997; Hill, 1998). Another approach is to stipulate that flocs larger than the maximal size are prone to breakage and to define either deterministically or probabilistically the size and number of the fragments produced by breakage (Pandya and Spielman, 1982; Hill, 1996). This strategy can prevent the accumulation of mass in the largest floc-size class.

This latter kinematic approach to modeling floc breakage trades one set of uncertainties for another. Maximal floc size, probability of breakup, and the size distribution of floc fragments are not known. Numerous observations of floc-size distributions, however, have been gathered in recent years that can provide inputs to a kinematic model. In this study, floc size data are used to define maximal floc size in a kinematic model. The model then uses established models of aggregation and new simple formulations of breakage probability and of the probability of producing fragments of a given size from broken flocs to produce equilibrium floc-size distributions. These distributions are compared to the measured size distributions. The favorable comparisons suggest that the simple probabilistic formulations of floc breakage and daughter fragment size distributions are reasonable and should be considered in future efforts to build dynamic models of equilibrium floc size distributions.

2 Monte Carlo model

Several methods have been developed to solve the population balance equation (pbe), which represents the mathematical interpretation of the aggregation/breakage problem (Drake, 1972; Elimelech *et al.*, 1995; Ramkrishna, 2000; Lee, 2001 for a review). Our interest is in the Monte Carlo (MC) method because of its well established advantage to solving multivariate aggregation/breakage problems (Tandon and Rosner, 1999). Discretization problems that result from the direct integration of the pbe in the case of polydispersed population, as occur with the finite difference method (Elimelech *et al.*, 1995), are not an issue in MC simulation. Because of its discrete nature, the MC method adapts itself to growth processes (Lin *et al.*, 2002).

The MC method, which derives its name from extensive use of random numbers, relies on sampling a given statistical ensemble and simulates physical processes (e.g., aggregation or breakage) by means of probabilistic tools. For instance, aggregation of two particles to form a third particle, or breakup of a particle to an ensemble of daughter particles are considered as discrete events. At each step of the simulation, a specific event is selected with a probability that is proportional to the rate of its occurrence. The selected event is then applied to one (in breakage) or two (in aggregation) particles selected stochastically from a predefined array of particles that represents the population. The simulation continues until the equilibrium conditions are reached. This method is sometimes referred to as "event driven" MC (Smith and Matsoukas, 1998; Lin *et al.*, 2002).

A major disadvantage of MC approach has been for a long time the rapid amplification of the computational time with the number of particles considered in the simulation. However, it is worthwhile to mention that computational time of this method varies with the complexity of the simulations. Advanced MC simulations where particles are tracked individually in their random movement due to Brownian motion and turbulence or deterministic displacement due to settling or stream flow require large

computational resources, especially when orientation of particles becomes an issue in simulations. Other disadvantages of traditional MC method have been for decades related to the fact that the size of the simulation array may increase, as in breakage, decrease, as in aggregation. In the first case the accuracy of the simulation becomes no longer satisfactory when substantial fraction of the original population has aggregated, and in the second the particle array becomes full. In both cases the simulation should be stopped. This type of MC simulation tracks a constant volume of the reacting system and is called constant-volume MC (Lin *et al.*, 2002). To overcome this size modification of the simulation array, a new method called constant-number MC has been proposed recently (Tang and Matsoukas, 1997; Smith and Matsoukas, 1998; Lee and Matsoukas, 2000; Lin *et al.*, 2002). In this method, the size of the simulation array is kept constant. A position in the array vacated due to an aggregation event is refilled by a particle selected randomly from the array, while a new particle(s) created by a breakage event replaces a particle (s) randomly selected from the array. This method has the advantage of maintaining constant statistical accuracy even for arbitrarily long simulation time with a finite number of particles. The advantages of this method compared to the constant-volume method were discussed by Smith and Matsoukas (1998) and Lin *et al.* (2002).

The proposed model to calculate the steady-state size distribution of flocs is based on the constant-number MC method. The new contributions in the present model are the simulation of the breakage of flocs and the integration of the concept of fractal geometry into the model using a recently proposed relationship between fractal dimension and the floc size (Khelifa and Hill, 2006). The idea of conservation of component particles is employed to track the evolution of the population (suspension). This method is more realistic than the approach used by Tandon and Rosner (1999) where volume and area of flocs were conserved after aggregation. The great advantage of the proposed method is that, after an aggregation or fragmentation event, the mass is conserved and tracked in the model. The amount of fluid trapped within the floc is not tracked, but is related to particle mass through the equation we proposed recently for floc density (Khelifa and Hill, 2006). Also, in the present study, the constant-number approach is applied to simulate aggregation process due to Brownian motion, shear and differential settling simultaneously.

A schematic description of the model is shown in Fig. 1. The inputs of the simulation are size distribution of the component particles (disaggregated particles), number N of the initial population, maximum size of flocs allowed in the simulation $D_{\max}$, densities of component particles ρ_s and the fluid ρ_w, and of course parameters defining environmental conditions such as turbulent kinetic energy dissipation rate and temperature. The simulation begins with an array of N component particles. Initial particles are placed in the array considering their initial size distribution and random selection of their position in the array. The maximum size $D_{\max}$ is transformed to the maximum number $N_{\max}$ of component particles allowed per floc with a model proposed recently (Khelifa and Hill, 2006). This model is based on the concept of fractal dimension. It relates the number of component particles forming a floc to its size, as described by the following three equations.

$$N_f = \left(\frac{D_f}{d_{50}}\right)^F \tag{1}$$

where d_{50} is the median diameter of the component particles. The coefficient F is the three-dimensional fractal dimension given by

$$F = 3\left(\frac{D_f}{d_{50}}\right)^{\alpha} \tag{2}$$

with α estimated by

$$\alpha = \frac{\log(F_c/3)}{\log(D_{fc}/d_{50})} \tag{3}$$

In Eq. (3), D_{fc} is a characteristic floc size at which the fractal dimension attains a characteristic value F_c. The values we have recommended for these two constants are 2000 μm and 2, respectively. It is important to note that we are still calling F "fractal dimension" even though it is in fact a function of the floc size, as shown by Eqs (2) and (3). Variation of F with floc size is supported strongly by data and by consideration of the heterogeneous composition and size of component particles within flocs (Khelifa and Hill, 2006).

2.1 Selection of main event

The second step in the simulation is to select the main event: aggregation or breakage. The null event is not included in the simulation. The method proposed by Lee and Matsoukas (2000) is based on calculation of probabilities of aggregation and breakage events using their rates of occurrence. The authors showed the performance of this method considering theoretical kernels for both aggregation and breakage. Little is known about the rate of real breakage of flocs (real kernels). As discussed in the previous section, existing models include unknown coefficients. This is why it is difficult to apply the method proposed by Lee and Matsoukas (2000) to real aggregation/breakage reactions. In the method we propose, the probability P_{brk} of occurrence of floc breakage event is based on the number of large flocs present in the simulation. After an extensive calibration study, we found that the following expression of P_{brk} improves considerably the prediction of the observations.

$$P_{brk} = \begin{cases} 0, & n_b = 0 \\ 0.5, & n_b = 1 \\ 1, & n_b > 1 \end{cases} \tag{4}$$

where n_b is the number of flocs larger than $D_{\max}$. This probability function shows that when flocs larger than $D_{\max}$ are absent in the population, breakage does not occur, and an aggregation event is always chosen. The opposite scenario occurs when 2 or more flocs larger than $D_{\max}$ are present. A breakage event is always chosen. This formulation represents a conceptual interpretation of the fact that flocs are fragile and have a short lifetime (Zimmermann-Timm, 2002 for a review).

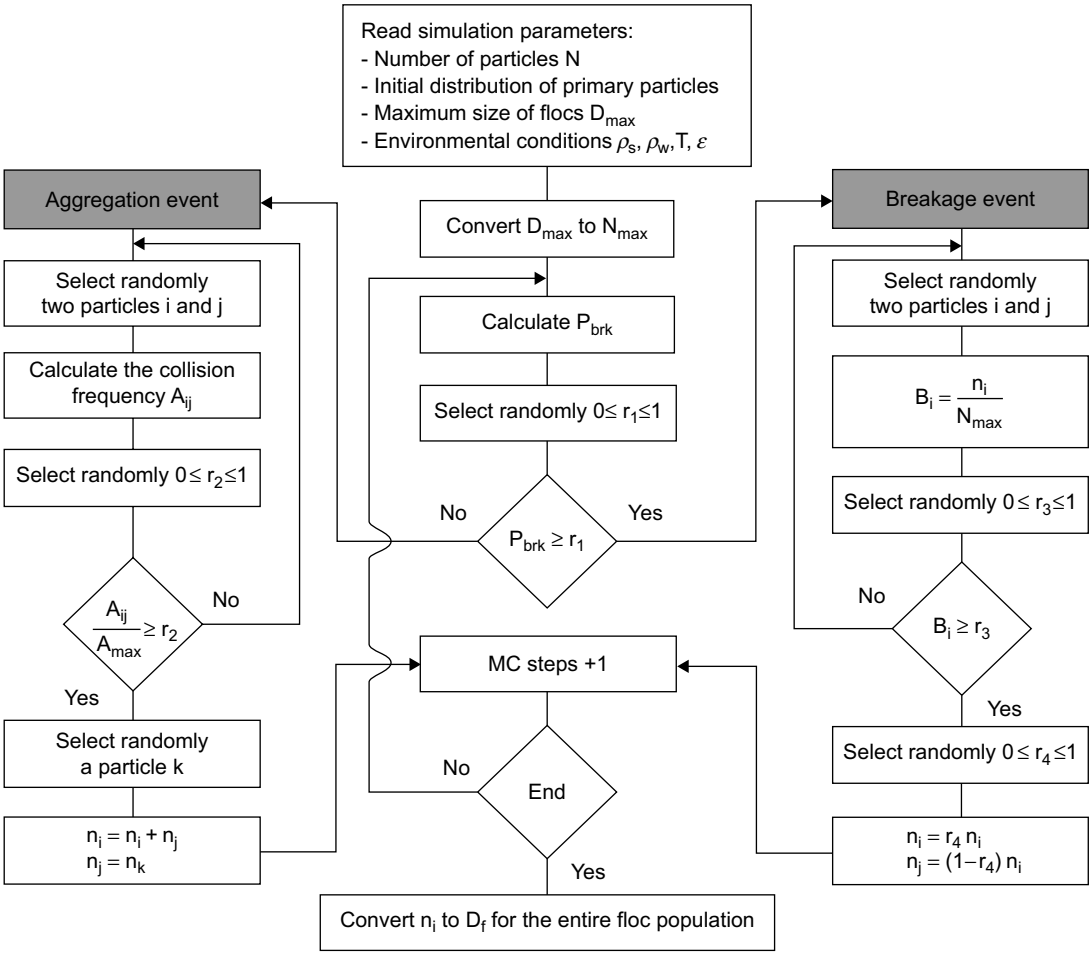


Figure 1 Flowchart for the proposed Monte Carlo (MC) model to simulate floc formation due to aggregation and breakage processes.

Because the null event is not considered in the simulation, the probability of the aggregation event is given by

$$P_{\text{agg}} = 1 - P_{\text{brk}} \tag{5}$$

A random number, r_1, is selected from a uniform distribution between 0 and 1. A breakage event is selected if $P_{\text{brk}} \geq r_1$, otherwise an aggregation event is selected (Fig. 1).

2.2 *Aggregation event*

In the case where the main event is aggregation, two particles, i and j, are selected randomly from the array and the corresponding collision frequency A_{ij} is calculated using the following equation (Jackson, 1995, 1998; Burd and Jackson, 1997, 2002)

$$A_{ij} = A_{ij}^{\text{diff}} + A_{ij}^{\text{turb}} + A_{ij}^{\text{sett}} \tag{6}$$

In Eq. (6), A_{ij}^{diff}, A_{ij}^{turb}, and A_{ij}^{sett} are the aggregation frequencies due to Brownian diffusion, turbulence and differential settling, respectively, which are assumed to act independently. As mentioned earlier, theoretical expressions of these rates are relatively well known (Hill, 1992; Elimelech *et al.*, 1995; Hill and Nowell, 1995; Thomsen and McCave, 2000) and are as follows:

$$A_{ij}^{\text{diff}} = \frac{2\kappa T}{3\mu}\frac{(d_i + d_i)^2}{d_i d_j} \tag{7}$$

$$A_{ij}^{\text{turb}} = \begin{cases} \frac{1}{6}\left(d_i + d_j\right)^3 \left(\frac{\varepsilon}{\nu}\right)^{1/2} & \text{for } d_i, d_j \le \eta \\ 1.08\left(d_i + d_j\right)^{7/3} (\varepsilon)^{1/3} & \text{for } d_i,\ d_j > \eta \end{cases} \tag{8}$$

$$A_{ij}^{\text{sett}} = \frac{\pi}{4}(d_i + d_j)^2 |W_i - W_j| \tag{9}$$

where d_i and d_j represent equivalent spherical diameters of particles (flocs) i and j, respectively, κ is the Boltzman constant, T is absolute temperature, μ and ν are dynamic and kinematic viscosities of the fluid, respectively, ε is turbulent kinetic energy dissipation rate, η is the Kolmogoroff length scale, and W_i and W_j settling velocities of particles i and j, respectively. Note that with these equations, collision efficiency is assumed to be unity. This assumption will increase aggregation rate, but the form of size distribution will not be affected unless collision efficiency is strongly diameter dependent. Past work suggests that it is not (e.g., Hill, 1992).

The next step in the aggregation simulation consists of selecting a random number, r_2, from a uniform distribution between 0 and 1 (Fig. 1). If $A_{ij}/A_{\max} \geq r_2$, the two particles are combined to form a floc containing $n_i + n_j$ component particles, where n_i and n_j represent the numbers of component particles in flocs i and j, respectively. $A_{\max}$ is the maximum value of the aggregation kernel among all particles. The new particle is stored in the position of the old particle i while a randomly selected particle is copied into the position vacated by particle j. If the aggregation

try was not successful, i.e., $A_{ij}/A_{\max} < r_2$, a new pair of particles is selected, and this step is repeated until a pair leads to a successful aggregation.

Calculation of $A_{\max}$ can slow MC simulations because it requires computation of A_{ij} for all particle pairs in the model. For each step in the simulation, the kernel A_{ij} is calculated about $N^2/2$ times. To reduce computation time, researchers often assume a constant kernel in the simulation (Smith and Matsoukas, 1998). This approach results in acceptance of any aggregation event. In the present study, $A_{\max}$ is calculated using the mean size of flocs and is corrected with an automatically adjusted correction factor C_F at each simulation step. The adjustment of C_F is performed such that the ratio between rejected and the accepted tries for aggregation remains close to unity (Elimelech *et al.*, 1995). Assuming a linear variation of C_F with this ratio, the following expression was used to calculate it:

$$C_F = 4\left(1 - \frac{n_r}{n_r + n_a}\right) \tag{10}$$

where n_r and n_a represent the number of rejected and accepted tries during the simulation of an aggregation event, respectively.

2.3 Breakage event

When the main event is breakage, two particles, i and j, are selected randomly from the array (Fig. 1). A random number, r_3, is selected from a uniform distribution between 0 and 1. If $n_i/N_{\max} = B_i \geq r_3$, particle i is split into two daughter particles k and l (binary fragmentation) such that

$$\begin{cases} n_k = r_4 n_i \\ n_l = n_i - n_k \end{cases} \tag{11}$$

where r_4 is a randomly selected number between 0 and 1 and n_k and n_l the number of component particles forming the daughter particles k and l, respectively. Daughter particle k is stored in the position vacated by particle i and the second daughter particle l is placed into the discarded particle j. If the test is not successful, a new pair of particles is selected. This step is repeated until a particle i leads to a successful breakage. One notes that this modeling procedure does not address the dynamic of floc breakage. It assumes that breakage results from splitting, erosion or both processes (Parker and Kaufman, 1972; Pandya and Spielman, 1982; Liem *et al.*, 1999; Thomas *et al.*, 1999). This is shown by the random selection of the size of the daughter particles following a breakage event (Eq. (11)).”

At the end of the simulation when an equilibrium floc size distribution is established, the diameter D_f of each floc composed of N_f component particles is calculated using Eqs (1)–(3).

3 Simulation conditions

All simulations discussed in this paper were performed with an array of $N = 15{,}000$ particles, as suggested by Smith and Matsoukas (1998). The size distribution of component particles was represented by the median size d_{50} (monosized distribution). Simulations were run for 5×10^5 MC steps. Size distribution of flocs was monitored every 10^5 steps. Mean size of flocs was stored during the simulation to monitor the evolution of floc-size distributions. Temperature and turbulent kinetic energy dissipation rate were kept constant during the simulation at 20° C and $5 \times 10^{-4}\,\mathrm{m^2/s^3}$ for typical estuarine conditions, respectively. Under these conditions, the Boltzmann's constant was set to $1.38 \times 10^{-23}\,\mathrm{J\,K^{-1}}$ and the dynamic viscosity of the water to 0.001 Pa s. Water density was kept constant at $1020\,\mathrm{kg/m^3}$ for seawater. The density and the diameter d of component particles were set to $2300\,\mathrm{kg/m^3}$ (Manning and Dyer, 1999) and 1 μm (Khelifa and Hill, 2006, for details), respectively.

4 Results

4.1 Steady-state size distribution

Examples of time evolution of mean size of flocs (expressed in mean number of component particles forming flocs) are shown in Fig. 2 and the corresponding steady-state size distributions are presented in Fig. 3 for simulations with $D_{\max}$ equal to 503 and 1007 μm. Both curves shown in Fig. 2 have an S-shaped form characterized by three main regimes: weak, rapid, and negligible (steady-state conditions) increases of the mean floc-size at MC steps between 0 and about 10^5, 10^5 to about 2×10^5 and larger than 2×10^5, respectively. This S-shaped form of the variations of the mean size of flocs with reaction time is well supported by previous findings reported in the literature (Tsai *et al.*, 1987; Lick and Lick, 1988; Burban *et al.*, 1989; Spicer and Pratsinis, 1996; Gonzalez and Hill, 1998; McAnally and Mehta, 2000). The first part of the evolution represents a growth period of flocs where

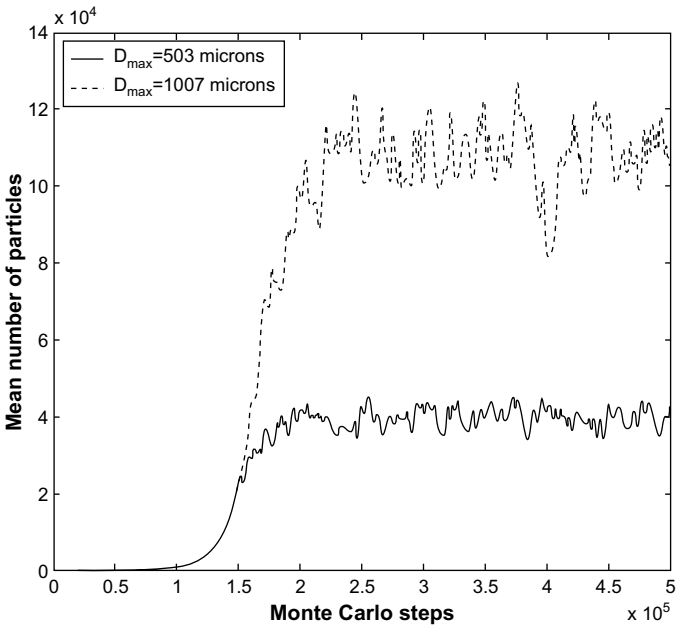
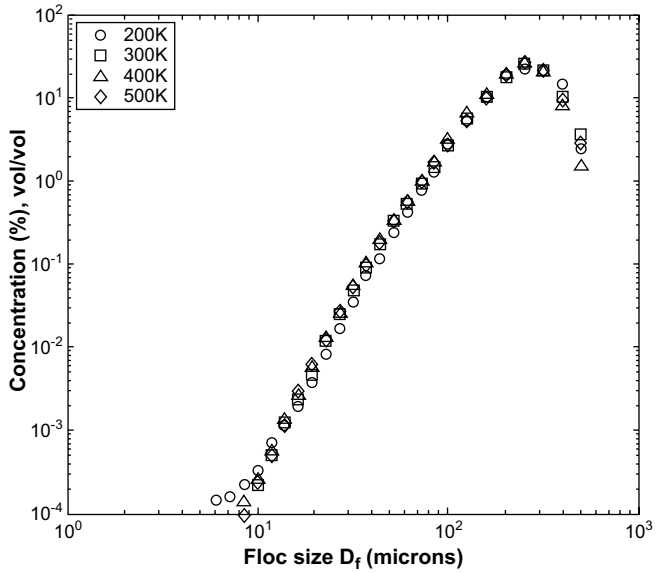
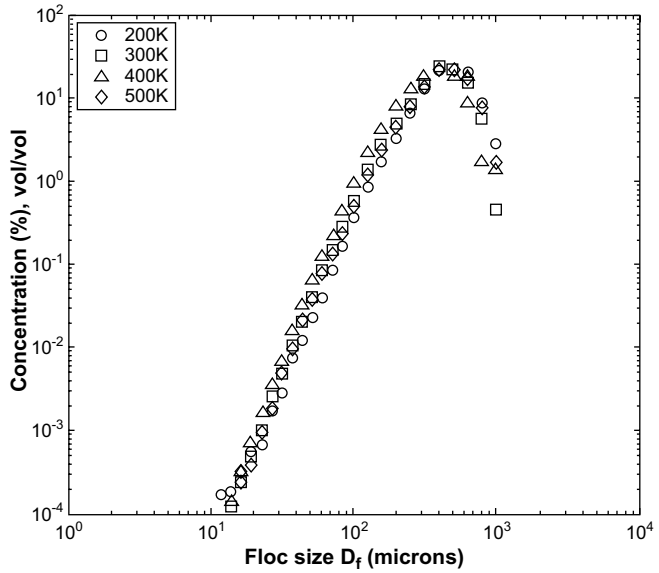


Figure 2 Example of time evolution of the mean floc size (evaluated by the mean number of component particles forming flocs) during Monte Carlo simulations with $D_{\max} = 503$ and 1007 μm (corresponding $N_{\max} = 5.16 \times 10^5$ and 14.63×10^5, respectively). Simulations were run with an initial population of 15,000 monosized particles.



(a)



(b)

Figure 3 Example of steady-state floc-size distributions obtained from Monte Carlo simulations after 2×10^5 (200 K), 3×10^5 (300 K), 4×10^5 (400 K) and 5×10^5 (500 K) Monte Carlo steps. Simulations were run with initial population of 15,000 particles of size $d = 1\,\mu$m. (a) $D_{max} = 503\,\mu$m; (b) $D_{max} = 1007\,\mu$m.

aggregation is the dominant process, but the rate of increase is small because the flocs are still small. The second phase occurs when flocs are large and encounter rate is high. Aggregation is still the dominant process during this second period, but breakage events may occur occasionally and more frequently at the end of this period. During the third phase, equilibrium conditions between aggregation and breakage processes are established and the mean size of flocs reaches its maximum value, around which it oscillates. This oscillation is one of the characteristics of MC simulations. Elimination of such "noise" requires multiple simulations and ensemble averaging of the results to smooth the solution. Nevertheless, even with one simulation only, size distributions monitored at 2×10^5, 3×10^5, 4×10^5, and 5×10^5 MC steps were similar, indicating stability of the steady-state conditions (Fig. 3).

4.2 Comparison with observations

Field observations from two different locations were compared with the simulated size distributions of flocs. The first site is the delta of the Po River in Italy (Fox *et al.*, 2004, for details) and the second one is the Eel River continental margin in northern California in USA (Hill *et al.*, 2000; Curran *et al.*, 2002, for details). In both studies, in situ size distribution of flocs was measured using a silhouette floc camera.

For each measured size distribution, D_{max} was determined and used in the corresponding simulation. Comparison between simulated and measured floc-size distributions for D_{max} equal to 503, 634, 799, and 1007 μm are shown in Figs 4–7. Each simulated size distribution was obtained after one simulation and by averaging computed distributions after 3×10^5, 4×10^5, and 5×10^5 MC steps. The same size bins were used to plot both measured and simulated size distributions. The agreement between measurements and simulations is good (Figs 4–7). This agreement suggests that effect of breakage on floc-size distribution can be reproduced with a simple kinematic parameterization of the process. In this parameterization a binary fragmentation is considered with equal probability of producing a daughter fragment of any size between d and the broken floc. Previous modeling approaches of floc breakage are generally complex and depend on tuning empirical parameters difficult to measure (see Lasheras

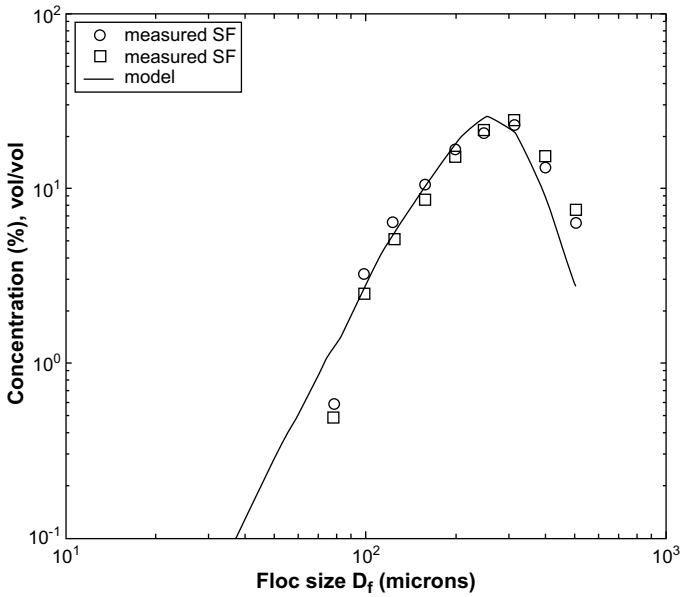


Figure 4 Comparison between measured and computed steady-state size distributions of flocs with $D_{max} = 503\,\mu$m and $d = 1\,\mu$m for typical estuarine turbulence ($\varepsilon = 0.0005\,m^2/s^3$). The data on size distribution indicated by SF are from the Strataform study (Hill *et al.*, 2000). The simulated size distribution is the average of size distributions obtained at 3×10^5, 4×10^5, and 5×10^5 Monte Carlo steps. The simulation was performed with 15,000 monosized particles.

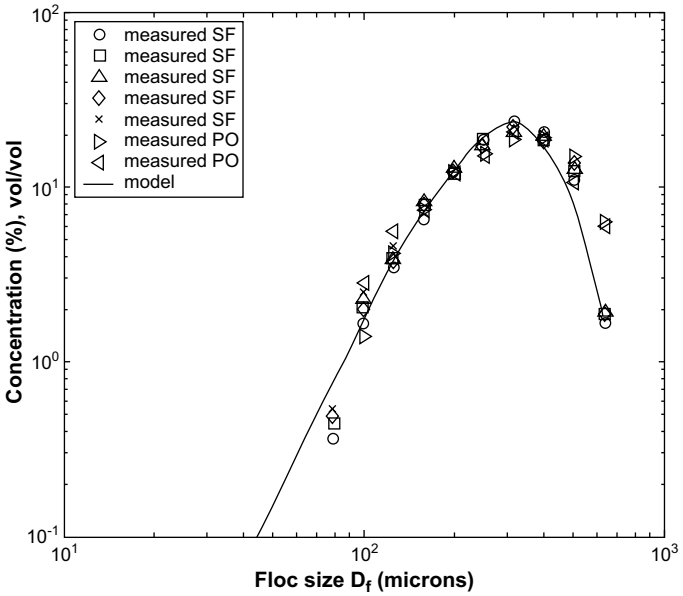


Figure 5 Comparison between measured and computed steady-state size distributions of flocs with $D_{max} = 634\,\mu m$ and $d = 1\,\mu m$ for typical estuarine turbulence ($\varepsilon = 0.0005\,m^2/s^3$). The data on size distribution indicated by SF and PO are from the Strataform (Hill *et al.*, 2000) and PO River (Fox *et al.*, 2004) studies, respectively. The simulated size distribution is the average of size distributions obtained at 3×10^5, 4×10^5, and 5×10^5 Monte Carlo steps. The simulation was performed with 15,000 monosized particles.

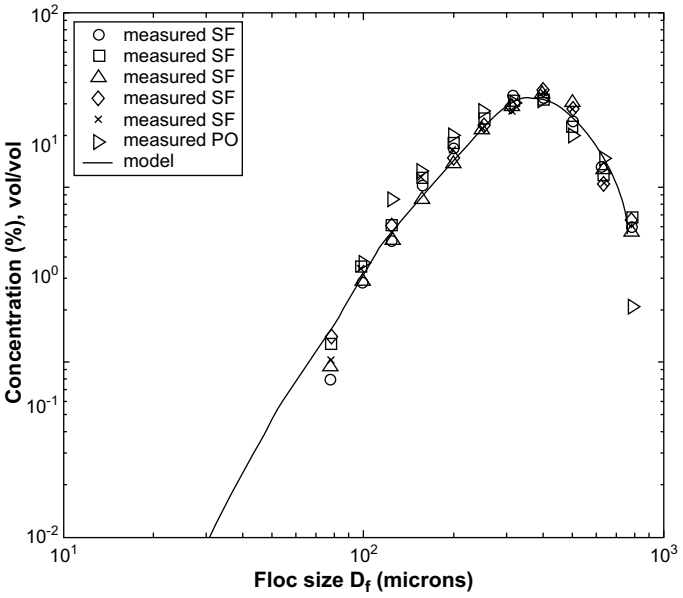


Figure 6 Comparison between measured and computed steady-state size distributions of flocs with $D_{max} = 799\,\mu m$ and $d = 1\,\mu m$ for typical estuarine turbulence ($\varepsilon = 0.0005\,m^2/s^3$). The data on size distribution indicated by SF and PO are from the Strataform (Hill *et al.*, 2000) and PO River (Fox *et al.*, 2004) studies, respectively. The simulated size distribution is the average of size distributions obtained at 3×10^5, 4×10^5, and 5×10^5 Monte Carlo steps. The simulation was performed with 15,000 monosized particles.

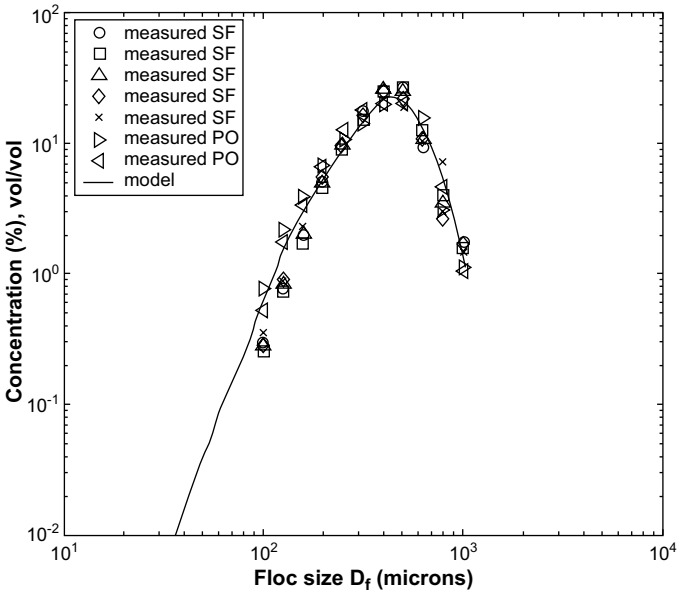


Figure 7 Comparison between measured and computed steady-state size distributions of flocs with $D_{max} = 1007\,\mu m$ and $d = 1\,\mu m$ for typical estuarine turbulence ($\varepsilon = 0.0005\,m^2/s^3$). The data on size distribution indicated by SF and PO are from the Strataform (Hill *et al.*, 2000) and PO River (Fox *et al.*, 2004) studies, respectively. The simulated size distribution is the average of size distributions obtained at 3×10^5, 4×10^5, and 5×10^5 Monte Carlo steps. The simulation was performed with 15,000 monosized particles.

et al., 2002 for a review). Examples of such approaches are methods proposed by Kramer and Clark (1999), Ducoste (2002), and Odriozola *et al.* (2002).

4.3 Comparison with Kranck's model

Parametrization of floc-size distributions has been investigated intensively by Kranck (1986, 1993), Kranck and Milligan (1985, 1991, 1992), and Kranck *et al.* (1993). They proposed that size distributions of flocs as well as disaggregated particles can be represented by the following parametric equation:

$$C = Q D^m \mathrm{e}^{-K\gamma D^2} \tag{11}$$

where C is the concentration of particles of size D; Q, m, and K are fitting parameters and the coefficient γ includes all terms from Stokes' law other than D^2, so that γD^2 defines a settling rate. The fitting (or free) parameters Q, m, and K are determined by fitting Eq. (11) to observed data.

Comparison between Eq. (11) and the proposed model is illustrated in Fig. 8 using an example of measured floc-size distribution presented by Kranck and Milligan (1992). The distribution was measured at San Pablo Strait at San Francisco Bay in California in USA. According to the authors, the best fit of Eq. (11) to the data is obtained with the values 0.0001, 2.72, and 0.081 for Q, m, and K, respectively. The simulated size distribution obtained with the MC model was obtained from one simulation after 4×10^5 steps considering 15,000 monosized particles of size 1 μm and a maximum floc size allowed of 1046 μm (the maximum value in the measured size distribution).

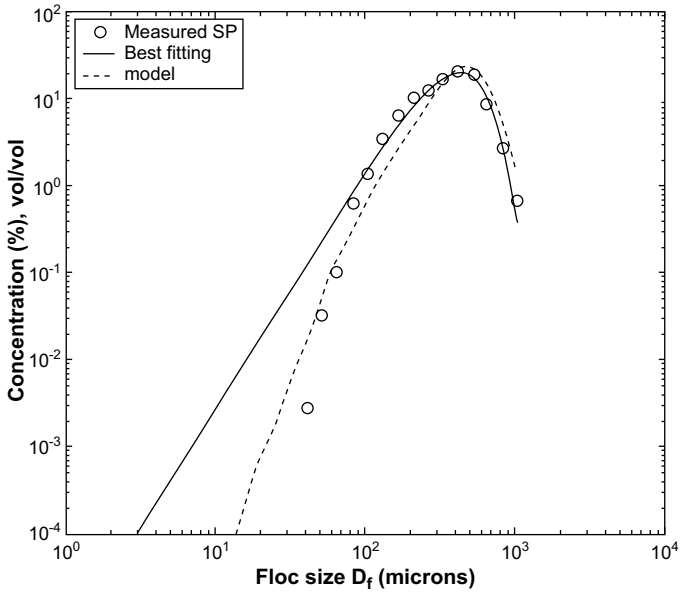


Figure 8 Comparison between measured, best fit of Kranck and Milligan model and computed steady-state size distribution of flocs with $D_{\max} = 1046\,\mu\text{m}$ and $d = 1\,\mu\text{m}$ for typical estuarine turbulence ($\varepsilon = 0.0005\,\text{m}^2/\text{s}^3$). The size distribution data indicated by SP are from Kranck and Milligan (1992). The simulated size distribution is obtained after 4×10^5 Monte Carlo steps and considering an initial population of 15,000 monosized particles.

Good agreement between simulated and measured distributions was observed (Fig. 8), especially for small sizes of flocs where Eq. (11) overestimates the distribution. For lager floc sizes, Eq. (11) fits the data well, while the simulated one slightly underestimates the data for intermediate sizes and overestimates the observations for large floc sizes. Overall, the trend of the measured size distribution is represented better by the simulated one than by the fitted curve. Again, what is important to keep in mind is that the simulated size distribution is predicted without using any tuning parameter, while the distribution calculated from Eq. (11) is a fitted curve using three free parameters.

5 Discussion

5.1 The model

This study has revealed that the effect of breakage (or fragmentation) on size distribution of flocs can be simulated using simple kinematic rules that do not specify whether breakage occurs by erosion or splitting (Pandya and Spielman, 1982; Lu and Spielman, 1985). The probability of occurrence of a breakage event is just a function of the number of large flocs (n_b) present in the simulation. Good agreement with observations was obtained when this probability is represented by Eq. (4). Conceptually, this equation shows that aggregation and breakage will have the same chance to occur when $n_b = 1$, and occurrence of a breakage event becomes certain when more than one large floc is present in the system. This formulation is consistent with the concept of a maximal equilibrium floc size.

The model preferentially destroys large flocs by specifying that the selection of a floc for breakup is a stochastic process, simulated by the simple test shown by $B_i \geq r_3$ (Fig. 1). This method results in preferential breakage of large flocs, but it also allows large flocs to persist for some finite period. Methods based on selection of the largest floc in the system for breakup performed poorly, especially in simulating the size distribution of large flocs. A distribution of daughter flocs following a breakage event is an important issue in solving the population balance equation (Lasheras *et al.*, 2002 for a review). Results of the present study suggest that combination of binary fragmentation and random distribution of daughter-floc size performs well. This approach is based on the argument that at an instant in time, the probability of multiple fragmentations occurring simultaneously is vanishingly low (Kramer and Clark, 1999).

The simulations discussed in this paper were performed for different maximum size of flocs. However, the turbulent kinetic energy dissipation rate (ε) was kept constant at $5 \times 10^{-4}\,\text{m}^2/\text{s}^3$ in the study. This may be considered as unrealistic, because existing theories as well as some observations indicate that maximum floc size decreases with turbulence (Parker and Kaufman, 1972; Tambo and Hozumi, 1979; Tambo and Watanabe, 1979; Hunt, 1982; Leentvaar and Rebhun, 1983; Lick and Lick, 1988). However, recent field studies have also shown no evident link between the two parameters (Hill *et al.*, 2000; Curran *et al.*, 2002). Further investigations at both laboratory and field scale are required to clarify this issue. Otherwise, application of the proposed model will continue to require a priori specification of maximum floc size. Nevertheless, the purpose of this study is to explore kinematic rather than dynamic control on steady-state size distribution of flocs. Thus, keeping ε constant does not affect equilibrium size distributions.

5.2 Self-similarity in floc-size distributions

Close examination of measured as well as simulated floc-size distributions shows that the distributions are similar, at least in term of shapes of the curves describing them. Similar observations have been reported by Hunt (1982), Droppo and Ongley (1994), Spicer and Pratsinis (1996), Tandon and Rosner (1999), and Kostoglou (2003). When floc size is non-dimensionlized by maximum floc size, simulated and observed size distributions collapse into a single set of similar distributions (Fig. 9). One notes that the variable shown on the vertical axis (concentration) was made dimensionless with the total concentration of flocs. Despite some scatter, Fig. 9 shows that floc-size distributions are self-similar and can be described by a single function. A polynomial fit to the data shows that this function can be described by the following relationship

$$C_r = -0.14S_r^4 - 1.45S_r^3 - 5.8S_r^2 - 6.7S_r + 0.67 \qquad (12)$$

In Eq. (12), C_r is the ratio between the concentration of flocs of size D_f and the total concentration of flocs, and $S_r = D_f/D_{\max}$. This function is shown in Fig. 9 in solid line. With this relationship, any dimensional floc-size distribution can be calculated if maximum size and total concentration of flocs are known.

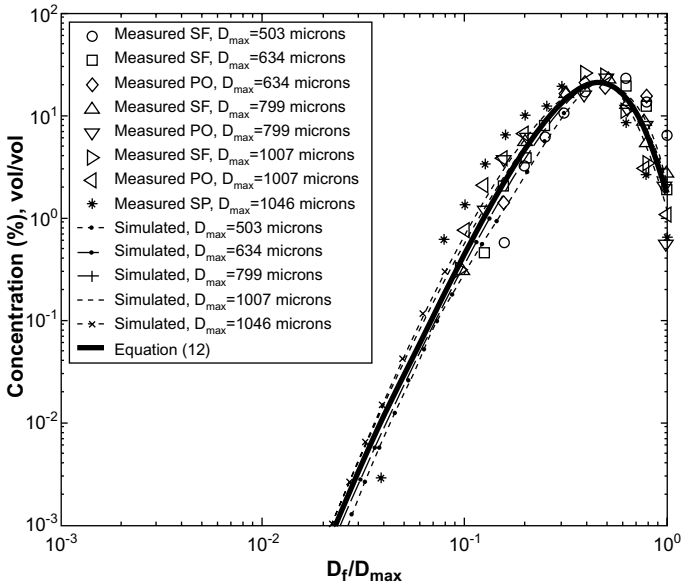


Figure 9 Evidence of self-similarity in floc size distribution: comparison between measured and computed steady-state size distributions of flocs using dimensionless variables. Measured size distributions are examples of the data shown in previous figures and simulated ones are the averaged size distributions shown in previous figures for different maximum size of flocs.

The roots of such self-preserving form of floc-size distribution reside, perhaps, in the existence of similarity in factors controlling the formation of flocs. Essentially, these controlling factors are the Brownian diffusion, turbulence, settling, and surface properties of particles forming flocs. Hunt (1982) has effectively shown that Brownian diffusion and turbulence produced self-similar size distribution of flocs formed by kaolinite and illite clay particles. Differential settling is expected to produce self-similar size distributions, as it is described by a power law of floc size. Surface properties of particles are more likely to affect maximum size of flocs, because they affect their strength (Kranenburg, 1999). Moreover, this interpretation seems to be supported also by our recent findings (Khelifa *et al.*, 2002) regarding self-similarity of size distributions of oil droplets formed under different turbulent flow conditions. The factors discussed above control also formation of oil droplets.

6 Conclusion

A new breakage model of flocs was integrated into a constant-number MC simulation to predict floc-size distribution due to Brownian motion, turbulence and differential settling at equilibrium. The success of the MC model in reproducing observed in situ size distributions suggests that integration of the new fractal model to describe floc geometry is appropriate. It suggests also that floc breakage can be simulated with simple rules. Essentially, these rules consist of relating the probability of breakage occurrence to the number of large flocs in the simulation, selecting the floc to breakup stochastically with preferential breakage of large flocs, and application of binary fragmentation and random distribution of daughter-floc size after breakup. At this stage, maximum size of flocs permissible in the simulation and the size of component particles are the key physical inputs of the model. Future upgrade of the model consists of integrating an empirical–theoretical model to predict the maximum size of flocs if the environmental conditions such as turbulence, salinity, sediment concentration, and particle composition are known. This study has also shown that measured and simulated floc-size distributions are self-similar. An empirical relationship is proposed to calculate size distribution of flocs if their maximum size and concentration are known.

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Notation

A_{ij} = Total collision frequency
$A_{\max}$ = Maximum collision frequency
A_{ij}^{diff} = Aggregation frequencies due to Brownian diffusion
A_{ij}^{turb} = Aggregation frequencies due to turbulence
A_{ij}^{sett} = Aggregation frequencies due to differential settling
C = Concentration of particles of size D_{f}
C_{F} = Correction factor
C_{r} = Concentration ratio
D_{f}, D_{fc}, d_i, d_j = Equivalent spherical diameter of flocs
$D_{\max}$ = Maximum diameter of flocs
d = Diameter of component particles
d_{50} = Median diameter of component particles
F, $F_{\max}$, $F_{\min}$ = Three-dimensional fractal dimension of flocs
K = Fitting parameter for Kranck's model
m = Fitting parameter for Kranck's model
N = Size of the simulation array
$N_{\max}$ = Maximum number of component particles per floc of size $D_{\max}$
n_b = Number of flocs larger than $D_{\max}$
n_i, n_j, n_k, n_l = Number of component particles in flocs
n_{r}, n_{a} = Number of rejected and accepted tries during the simulation of Aggregation event
P_{agg} = Probability of occurrence of aggregation event
P_{brk} = Probability of occurrence of breakage event
Q = Fitting parameter for Kranck's model
Re = Particle Reynolds number
r_1, r_2, r_3, r_4 = Random numbers
S_{r} = Size ratio equals $D_{\text{f}}/D_{\max}$
T = Absolute temperature
W_i, W_j = Settling velocities of flocs

α = Constant used to calculate the fractal dimension
ε = Kinetic energy dissipation rate
η = Kolmogoroff length scale
κ = Boltzman constant
μ = Dynamic viscosity of the water
ν = Water kinematic viscosity
ρ_f = Floc density
ρ_s = Sediment density
ρ_w = Water density

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