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Models for effective density and settling velocity of flocs

Models pour la densité effective et la vitesse de sédimentation de flocs

ALI KHELIFA, *Emergencies Science and Technology Division, Environmental Technology Centre, Environment Canada, 335 River Road, Ottawa, Ontario, K1A 0H3, Canada. Tel.: (613) 991-9455; fax: (613) 991-9485; e-mail: ali.khelifa@ec.gc.ca (author for correspondence)*

PAUL S. HILL, *Department of Oceanography, Dalhousie University, Halifax, NS, B3H 4J1, Canada. E-mail: phill@phys.ocean.dal.ca*

ABSTRACT
New models to predict settling velocity and effective density of flocs are proposed. The models are based on the concept of fractal geometry, but with the assumption of variable fractal dimension with the floc size. The best results are obtained when the fractal dimension is estimated by a power law function of the floc diameter. The models are compared with observations from 26 published data sets relating floc size to settling velocity measured under various conditions and at different locations. The floc size covered by the data varies between 1.4 and about 25,500 μm. Five commonly used models are also compared to these data and found to reproduce inadequately the full range of the observations. Sensitivity analysis shows that, with the proposed models, the spread in the data may be reproduced by varying the size of primary particles from about 0.05 to 20 μm. The new models are proposed for integration into numerical models to simulate sediment transport of cohesive sediments, contaminants, and biological microorganisms such as phytoplankton.

RÉSUMÉ
Des modèles nouveaux sont proposés pour prédire la vitesse de sédimentation et la densité effective des flocs d'argile. Les modèles sont basés sur le concept de géométrie fractale, mais avec l'hypothèse de dimension fractale variable avec la taille des flocs. Les meilleurs résultats sont obtenus lorsque la dimension fractale est estimée par une fonction puissance du diamètre des flocs. Les modèles proposés sont comparés avec les observations provenant de vingt-six séries de données publiées. Ces dernières relient la taille des flocs à la vitesse de sédimentation mesurée dans des conditions variées et à des endroits différents. La taille des flocs concernés par les données varie entre 1.4 et 25500 μm. Cinq modèles utilisés fréquemment sont aussi comparés à ces données et sont apparus inadéquats de reproduire les observations. L'analyse de sensitivité montre qu'avec les modèles proposés, les données peuvent être reproduites en variant la taille des particules primaires composant les flocs d'environ 0.05 à 20 μm. Les nouveaux modèles sont proposés pour intégration dans les modèles numériques pour simuler le transport de sédiments cohésifs, de contaminants et de microorganismes biologiques comme le phytoplancton.

Keywords: Flocs, flocculation, fractal dimension, settling velocity, effective density, sediment transport, cohesive sediment, marine snow.

1 Introduction

Gravitational settling is widely acknowledged as a key mechanism behind removal of suspended particles from the water column in aquatic environments such as stormwater ponds, water treatment installations, lakes, river deltas, estuaries and marine environments (e.g. Krishnappan *et al.*, 1999; McAnally and Mehta 2000; Fox *et al.*, 2003). Settling is also recognized as playing a key role in transferring contaminants from the water column to the bottom (e.g. Droppo *et al.*, 2000). Sediment particles, organic matter and contaminants aggregate to form large flocs that are hundreds to thousands of micrometers in diameter. This aggregation mechanism alters the settling process by repackaging small, slowly sinking particles into large, rapidly sinking flocs (see Geyer *et al.*, 2003 for a review).

Numerous attempts to model settling velocity as a function of floc size, shape and density have been undertaken. Early attempts demonstrated that the assumption of size-invariant density was inappropriate and should be modified using empirical factors to predict the settling velocity of natural flocs (Graf, 1971; Hawley, 1982; van Leussen, 1988, for a review). The effects of floc size on the flow regime near the settling floc and the consequent alteration of the drag force were discovered also in the 1960s and 1970s (Graf, 1971; Raudkivi, 1976). From these controlling parameters of the settling process, the density of flocs is now arguably considered as the one most in need of further research, because it

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is not well known and difficult to measure directly (Azetsu-Scott and Johnson, 1992). Modeling of the settling process depends on a good understanding of the variability of floc density with environmental conditions such as floc size, sediment concentration, composition, microbiological activity, salinity and temperature.

In the recent decades substantial effort has been devoted by many investigators to elucidation of the relationships between effective density (excess density) and floc size. In most of the studies, the effective density of flocs was estimated from settling velocity measurements and using established settling laws (e.g. Stokes law) that express settling velocity as a function of, among other variables, size, shape, viscosity and excess density. Moreover, much attention has been devoted to obtaining undisturbed settling velocities of flocs, which has led to development of various devices for measuring settling velocity of flocs at both laboratory and field scales. Most of these devices are based on imaging techniques (Li and Ganczarczyk, 1987; Burban *et al*., 1990; Eisma *et al*., 1990; van Leussen and Cornelisse, 1993; Fennessy *et al*., 1994; Hill *et al*., 1994, 1998, 2000; Syvitski *et al*., 1995; Puls and Kühl, 1996; Spicer and Pratsinis, 1996; Sternberg *et al*., 1996, 1999; Diercks and Asper, 1997; van Leussen, 1999; Manning and Dyer, 2002; Fox *et al*., 2003). A comparison of *in situ* techniques developed during the 1980s and early 1990s for measuring settling velocity was presented by Dyer *et al*. (1996). As a result of this relatively long history of investigation, a large number of data series of settling velocity are now available for use in attempts to develop a general relationship between floc size, settling velocity and excess density.

In this paper, data series of settling velocity from different sources are combined. A modified Stokes law is applied to transform the data to a series of effective density data. The data are then compared with the models proposed by Tambo and Watanabe (1979), Hawley (1982), McCave (1984), Kranenburg (1994), Lau and Krishnappan (1997) and Winterwerp (1998). A comparative analysis of these models is discussed and a new modeling approach, based on the concept of fractal geometry (Meakin, 1988), is presented. Two models to calculate effective density and settling velocity of flocs under almost any realistic size range are finally proposed and compared with the observations.

2 Experimental data

Twenty-six series of field/laboratory settling velocity data from various sources were considered in this study (Table 1). The data cover a wide range of floc size from 1.4 to about 25,500 μm. They were measured in various environments, which include fresh water and sea water conditions.

Calculation of the floc settling velocity, V_f, is usually performed considering the balance between the drag and the gravitational forces exerted on the floc (equilibrium settling conditions). This leads to the following expression of V_f (see for instance Batchelor, 1991; Winterwerp, 1998):

$$V_\mathrm{f} = \left(\frac{4}{3}\,\theta g C_\mathrm{d}^{-1}\,\frac{\Delta\rho}{\rho_\mathrm{w}}\,D_\mathrm{f}\right)^{0.5} \tag{1}$$

where θ is a dimensionless particle-shape factor, g is the gravitational acceleration (ms^{-2}), C_d is the dimensionless drag coefficient, $\Delta\rho = \rho_\mathrm{f} - \rho_\mathrm{w}$ is the effective density (excess density) of the floc (kg m^{-3}), D_f is the equivalent spherical diameter of the floc (m) and ρ_f and ρ_w are floc and water densities, respectively (kg m^{-3}).

The drag coefficient shown in Eq. (1) can be estimated using the following commonly used empirical relationship (Raudkivi, 1976):

$$C_\mathrm{d} = \frac{24}{Re}\left(1 + 0.15\,Re^{0.687}\right), \tag{2}$$

where $Re = V_\mathrm{s} D_\mathrm{f}/\nu$ is the particle Reynolds number in which ν represents the kinematic viscosity of the water (m^2 s^{-1}).

Other formulations of the drag coefficient have been proposed (e.g. White, 1974), but for the range of Reynolds numbers characteristic of flocs, these formulations give similar results. For $\theta = 1$ (spherical floc), $C_\mathrm{d} = 24/Re$ and $\Delta\rho = \rho_\mathrm{s} - \rho_\mathrm{w}$, in which ρ_s is the density of the primary particles forming the flocs, Eq. (1) becomes the well known Stokes law. Equation (1) combined with Eq. (2) is referred as "modified Stokes law" in this paper. Assuming $\theta = 1$, this modified Stokes law is used to generate a series of effective density from the settling velocity data shown in Table 1. The results are shown in Fig. 1, while the original series of settling velocity are shown in Fig. 2.

3 Existing models

Equation (1) shows that good estimation of V_f is closely related to an accurate approximation of the effective density $\Delta\rho$ of flocs. This is why various models have been proposed to calculate the latter. Most of them are regression functions of observations, which generally cover a narrow range of floc size and specific conditions of floc formation (Table 2). A comparison between effective density data and the predictions obtained with these models shows that the various models are successful at predicting excess density only over limited size ranges (Fig. 1). The corresponding comparison for settling velocity data reveals similarly limited ranges of agreement between models and data (Fig. 2). From the five considered models, the model proposed by McCave (1984) provides, perhaps, the best representation of the data, except for flocs larger than about 1000 μm. Nevertheless, none of the models describes properly the entire range of the data. In general, they overpredict settling velocity and excess density for both large and small particles.

4 Proposed models

The objective of this study is to develop a new model to calculate the effective density of flocs for almost any realistic size, as covered by the data shown in Fig. 1. This model is then combined with the modified Stokes law to establish a new general model for the settling velocity of flocs under the same size range. The approach used to develop the effective density model is based on the concept of fractal geometry, with one key modification that

Table 1 Information about considered laboratory/field data on floc settling velocity obtained from the literature

Reference	Site of observation	Measurement method	Size (μm)
Fox *et al.* (2003)	Po River Prodelta, Italy	Image analysis	65–1380
Droppo (2002)	—	—	120–855
Manning and Dyer (2002)	Tamar Estuary, UK	Image analysis	30–455
Dyer and Manning (1999)	Elbe Estuary, Germany	Image analysis	52–810
Dyer and Manning (1999)	Tamar Estuary, UK	Image analysis	61–572
Manning and Dyer (1999)	Annular flume, laboratory	Image analysis	41–191
Sternberg *et al.* (1999)	Eureka, California, USA	Image analysis	131–765
Hill *et al.* (1998)	Trr Inlet, Glacier Bay, Alaska, USA	Image analysis	661–2176
Diercks and Asper (1997)	Black Sea, Turkey	Image analysis	953–5451
Diercks and Asper (1997)	Gulf of Mexico, USA	Image analysis	827–7416
Syvitski and Hutton (1996)	Bedford Basin, Halifax, Nova Scotia, Canada	Image analysis	651–3665
Dyer *et al.* (1996)	Ems Estuary, Germany	Image analysis	107–638
Dyer *et al.* (1996)	Elbe Estuary, Germany	Image analysis	174–441
Sternberg *et al.* (1996)	Northen California continental margin, USA	Image analysis	127–431
Syvitski *et al.* (1995)	Bedford Basin, Halifax, Nova Scotia, Canada, deployment of May 5, 1991	Image analysis	149–3201
Syvitski *et al.* (1995)	Bedford Basin, Halifax, Nova Scotia, Canada, deployment of May 8, 1991	Image analysis	125–3539
Fennessy *et al.* (1994)	Tamar Estuary, UK	Image analysis	21–569
ten Brinke (1994)	Oosterschelde Tidal Basin, The Netherlands	Image analysis	50–694
Azetsu-Scott and Johnson (1992)	Linear density gradient settling column, laboratory	Image analysis	470–1350
Burban *et al.* (1990)	Horizontal Couette type flocculator and insulated settling tube, laboratory	Image analysis	11–266
Burban *et al.* (1990)	Horizontal Couette type flocculator and insulated settling tube, laboratory	Image analysis	10–214
Alldredge and Gotschalk (1988)	San Pedro and Santa Barbara Basins, California, USA	Technique base on dye injection 3 cm below the floc	350–25,532
Li and Ganczarczyk (1987)	Settling column and laboratory flocs	Image analysis	37–746
Klimpel and Hogg (1986)	Stirred tank and settling column, laboratory	Image analysis	20–1596
Gibbs (1985)	Settling column and field flocs from Chesapeake Bay, Maryland, USA	Microscopic observation of the floc motion	19–230
McCave (1975)	Coulter Counter measurements performed by Sheldon *et al.* (1972), as quoted by the author	Approximated using Stokes law and estimated effective density	1.4–362

relaxes the assumption that fractal dimension does not vary as a function of floc size. Although relaxation of this assumption may appear at first glance to be antithetical to self-similarity inherent in a fractal model, it in fact is not. As discussed below, it is a pragmatic recognition of the fact that natural flocs comprise a range of finite component particle sizes.

4.1 *Effective density*

The effective density of flocs formed by polysized particles can be expressed as

$$\rho_{\rm f} = \rho_{\rm s}\,\frac{\sum_{i=1}^{k} d_i^3}{{D_{\rm f}}^3} \tag{3}$$

where d_i represents the diameter of the ith primary particle.

Under the assumption that the structure of flocs is self-similar, the concept of fractal geometry can be used to describe the geometrical characteristics of this structure. This concept has been applied widely to description of floc geometry (see Vicsek, 1992, for a review). According to a fractal model the equivalent spherical diameter, $D_{\rm f}$, of an aggregate composed of k primary monosized particles of diameter d can be approximated, considering the properties of fractal objects (Meakin, 1988; Wiesner, 1992; Kramer and Clark, 1999), by

$$D_{\rm f} = d\;k^{1/F} \tag{4}$$

where F is the three-dimensional fractal dimension of flocs.

Extension of Eq. (4) to general conditions of polysized particles forming natural flocs (Jackson, 1998; Thomas *et al.*, 1999) results in the expression

$$D_{\rm f} = \left(\sum_{i=1}^{k} d_i^F\right)^{1/F} \tag{5}$$

It is easy, considering the case of monosized particles for instance, to show that when Eq. (3) is combined with Eq. (5),

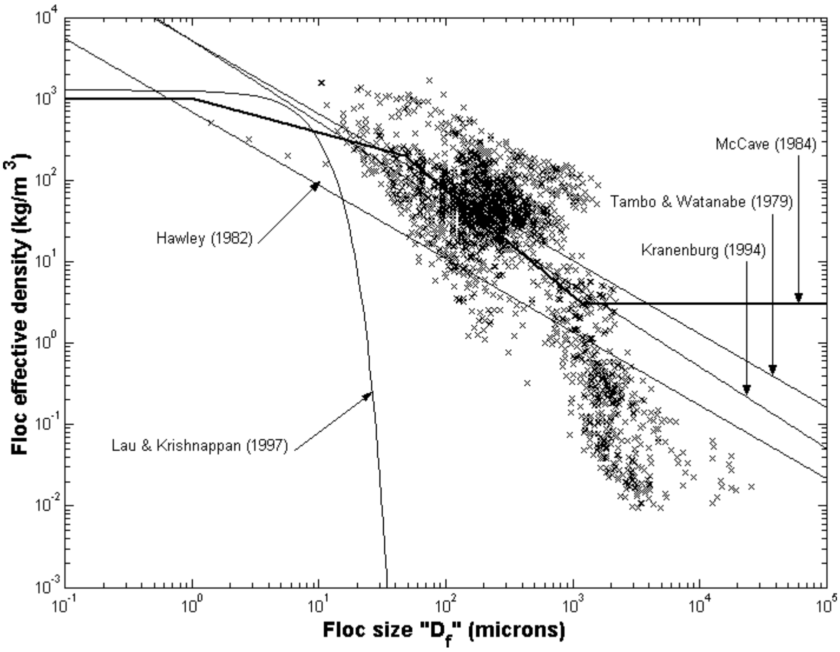

Figure 1 Comparison between data of effective density (cross symbol) and predictive models proposed by Tambo and Watanabe (1979), Hawley (1982), McCave (1984), Kranenburg (1994) with $d = 4\,\mu$m and Lau and Krishnappan (1997).

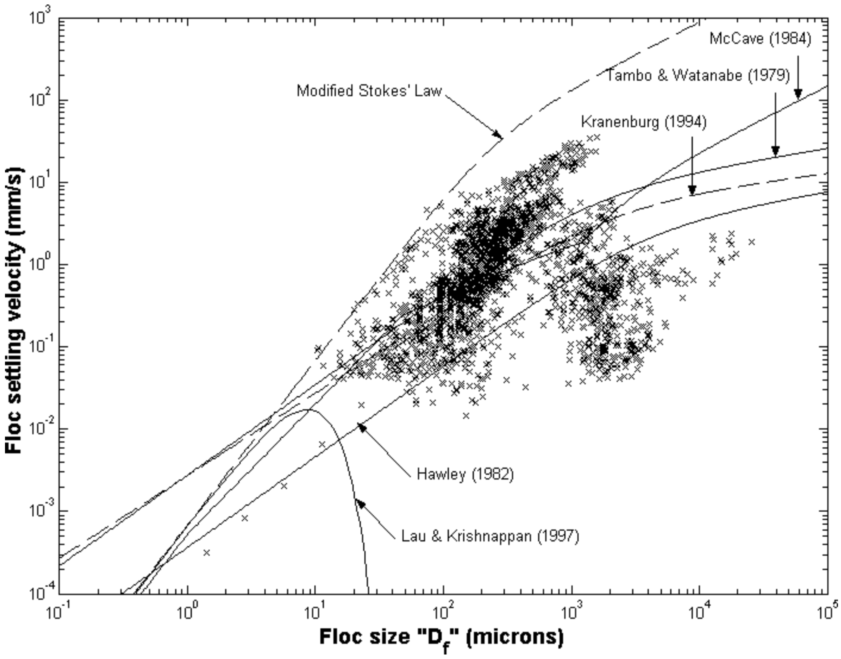

Figure 2 Comparison between data of floc settling density (cross symbol) and predictions using the modified Stokes law and the models for floc effective density proposed by Tambo and Watanabe (1979), Hawley (1982), McCave (1984), Kranenburg (1994) with $d = 4\,\mu$m (combined with modified Stokes law, this model correspond exactly to the model proposed by Winterwerp (1998) for settling velocity) and Lau and Krishnappan (1997). Prediction using the modified Stokes' law is shown by the dashed line.

the density of flocs goes to zero when the number of monomers k becomes large, because F is < 3, its maximum value. This is not realistic. Equation (3) with D_f expressed by Eq. (5) should satisfy appropriate "boundary" conditions, as suggested below

$$\rho_f = \begin{cases} \rho_s & \text{at } k = 1 \\ \rho_w & \text{at } k = \infty \end{cases} \tag{6}$$

If one considers a simple linear variation of ρ_f such as

$$\rho_f = \rho_s \left[C_1 \frac{\sum_{i=1}^{k} d_i^3}{\left(\sum_{i=1}^{k} d_i^F\right)^{3/F}} + C_2 \right] \tag{7}$$

where C_1 and C_2 are two constants calculated considering Eq. (6), then

$$\rho_f - \rho_w = (\rho_s - \rho_w) \frac{\sum_{i=1}^{k} d_i^3}{\left(\sum_{i=1}^{k} d_i^F\right)^{3/F}} \tag{8}$$

Table 2 Information about models of effective density/settling velocity of flocs considered in this study

Reference	Model for	Expression	Comments
Winterwerp (1998)	Settling velocity	$V_\mathrm{f} = \frac{1}{18}\frac{\alpha}{\beta} g \frac{\rho_\mathrm{s}-\rho_\mathrm{w}}{\mu} d^{3-F} \frac{D_\mathrm{a}^{F-1}}{1+0.15\,Re^{0.687}}$	α and β shape-related coefficients and equal 1 for spherical particles
Lau and Krishnappan (1997)	Effective density	$\rho_\mathrm{a} - \rho_\mathrm{w} = \rho_\mathrm{s}\, \exp\left(-0.02\, D_\mathrm{a}^{1.85}\right)$	The floc size D_a is in μm and the densities in g cm^{-3}
Kranenburg (1994)	Effective density	$\rho_\mathrm{a} - \rho_\mathrm{w} \propto (\rho_\mathrm{s} - \rho_\mathrm{w}) \left(\frac{D_\mathrm{a}}{d}\right)^{F-3}$	—
McCave (1984)	Effective density	$\rho_\mathrm{a} - \rho_\mathrm{w} = \begin{cases} 1 & \text{for } D_\mathrm{a} \le 1\,\mu\text{m} \\ \propto D_\mathrm{a}^{-0.42} & \text{for } 1 \le D_\mathrm{a} \le 50\,\mu\text{m} \\ \propto D_\mathrm{a}^{-1.3} & \text{for } 50 \le D_\mathrm{a} \le 1200\,\mu\text{m} \\ 0.003 & \text{for } D_\mathrm{a} \ge 1200\,\mu\text{m} \end{cases}$	The floc size D_a is in μm and the densities in g cm^{-3}
Hawley (1982)	Effective density	$\rho_\mathrm{a} - \rho_\mathrm{w} = (\rho_\mathrm{s} - \rho_\mathrm{w}) \left(\frac{D_\mathrm{a}}{d}\right)^{-0.9}$	—
Tambo and Watanabe (1979)	Effective density	$\rho_\mathrm{a} - \rho_\mathrm{w} = 0.0013 \left(\frac{D_\mathrm{a}}{1}\right)^{-0.9}$	The floc size D_a is in cm and the densities in g cm^{-3}

Introducing the following mean variables:

$$m_3 = \frac{\sum_{i=1}^{k} d_i^3}{k}, \quad \text{and} \quad m_F = \frac{\sum_{i=1}^{k} d_i^F}{k} \tag{9}$$

Equation (8) becomes

$$\rho_\mathrm{f} - \rho_\mathrm{w} = (\rho_\mathrm{s} - \rho_\mathrm{w})\, k^{(F-3)/F}\, \phi \tag{10}$$

where $\phi = m_3/m_F^{3/F}$. This term represents the effect of size distribution of primary particles forming flocs, while the term $k^{(F-3)/F}$ shows the effect of the fractal dimension and the floc size. For instance, for the case of monosized particles Eq. (10) simplifies to

$$\rho_\mathrm{f} - \rho_\mathrm{w} = (\rho_\mathrm{s} - \rho_\mathrm{w})\, k^{(F-3)/F} \tag{11}$$

Equation (11) is similar to the model proposed by Kranenburg (1994) for the effective density of flocs, because for monosized particles of size d and considering Eq. (4), the last term in the right side of this equation transforms to (Meakin, 1988)

$$k^{(F-3)/F} = \left(k^{1/F}\right)^{F-3} = \left(\frac{dk^{1/F}}{d}\right)^{F-3} = \left(\frac{D_\mathrm{f}}{d}\right)^{F-3} \tag{12}$$

and Eq. (11) becomes:

$$\rho_\mathrm{f} - \rho_\mathrm{w} = (\rho_\mathrm{s} - \rho_\mathrm{w}) \left(\frac{D_\mathrm{f}}{d}\right)^{F-3} \tag{13}$$

which represents exactly the model proposed by Kranenburg (1994) for the effective density of flocs.

Relationships deduced from measurements of settling velocity by Hawley (1982) and Gibbs (1985) are also similar to Eq. (13) except that the exponents of the size ratios they proposed were constant and equaled −0.9 and −0.97, respectively. Similarly, Lick and Lick (1988) established from experimental measurements a relationship similar to Eq. (13). However, the exponent they proposed for the size ratio was a function of the shear stress.

The agreement between the observed data and the model shown by Eq. (13), assuming a fractal dimension of 2 as considered by Winterwerp (1998), is poor (Fig. 1). Of course, the conditions under which the flocs shown in this figure were formed are varied. Mineral composition, particle size distribution, sediment concentration, salinity, organic-matter content and microbiological activity are examples of parameters that may affect the structure of flocs. Variability in component particles within flocs arguably causes floc structure to deviate from the ideal, self-similar geometry assumed in the beginning of this section and by Kranenburg (1994) and others. Assumption of constant fractal dimension, if in fact it is a decreasing function of size, would cause the observed overprediction of density and settling velocity at large and small floc sizes.

Variation of fractal dimension with the floc size has been reported by many investigators. For instance, in their experimental investigation of fractal dimension of suspended particles in seawater using light-scattering, Martinis and Risovic (1998) reported that the fractal dimension of aggregates decreases from the value 3 for small particles with diameters $<0.02\,\mu$m to about 1.2 for large aggregates with diameters $>200\,\mu$m. Previous measurements of the authors (Risovic and Martinis, 1996) also revealed a decrease of the fractal dimension from 2.5 to 1.68 when the size of marine particles increased from 2 to 200 μm.

To take into consideration the possible variability in the structure of flocs, a variable fractal dimension with the size of flocs is proposed. The fractal dimension of a floc with diameter closer to the size of the primary particles should approach the value of 3, which applies to solid particles. Large flocs should possess fractal dimensions of about 2 as is commonly observed (e.g. Meakin, 1988; Winterwerp, 1998; Dyer and Manning, 1999; Kramer and Clark, 1999). It is then legitimate to consider a continuous decrease of the fractal dimension as the size of flocs increases. A power law such as

$$F = \alpha \left(\frac{D_\mathrm{f}}{d}\right)^{\beta} \tag{14}$$

represents a reasonable proposed approximation for F, where the coefficient α and the exponent β can be calculated using the following boundary conditions

$$F = \begin{cases} 3 & \text{at } D_f = d \\ F_c & \text{at } D_f = D_{fc} \end{cases} \tag{15}$$

In Eq. (15), the fractal dimension takes its maximum value 3 when the size of the floc approaches the size d of primary particles, and it reaches a lower value F_c when the size of the floc becomes D_{fc}, a characteristic size of flocs as discussed further in the next sections. Combination of Eqs (14) and (15) gives

$$\alpha = 3 \quad \text{and} \quad \beta = \frac{\log(F_c/3)}{\log(D_{fc}/d)} \tag{16}$$

Under more general conditions of polydisperse particle-size distributions, the median size d_{50} of component particles within flocs could be used to represent the size of primary particles instead of d. Therefore, the general form of the proposed model for the effective density of flocs becomes

$$\rho_f - \rho_w = (\rho_s - \rho_w) \left(\frac{D_f}{d_{50}}\right)^{F-3} \phi \tag{17}$$

where F is described by Eqs (14)–(16) with d replaced by d_{50}.

4.2 Settling velocity

The proposed model to calculate the floc settling velocity is obtained considering the modified Stokes law (Eqs (1) and (2)) and the proposed model for effective density of flocs (Eq. (17)). Combination of these equations leads to the following expression for the settling velocity of flocs:

$$V_f = \frac{1}{18} \theta g \frac{\rho_s - \rho_w}{\mu} d_{50}^{3-F} \frac{D_f^{F-1}}{1 + 0.15\, Re^{0.687}} \phi \tag{18}$$

in which μ is the dynamic viscosity of the water.

Note that if $Re \ll 1$, flocs are spherical ($\theta = 1$) and non-porous ($F = 3$), and component particles are monosized ($\phi = 1$), Eq. (18) simplifies to the well-known Stokes formula. Moreover, the proposed model shown by Eq. (18) becomes identical to the model proposed by Winterwerp (1998) for monosized particles and with constant fractal dimension F.

4.3 Comparison with data

As shown by Eqs (14)–(17), the model proposed in this study for effective density of flocs depends on four key particle parameters: the size of primary particles d (or d_{50}), D_{fc}, F_c, and the density of particles ρ_s. Sensitivity analysis showed that the model is more sensitive to d and D_{fc} (Figs 3–6) than to D_{fc} and ρ_s (data not shown). The central curve (solid and tick line) corresponds to $d = 1.0\,\mu$m, $D_{fc} = 2000\,\mu$m, $F_c = 2.0$, and $\rho_s = 2300$ (kg/m^3). With these values, the proposed model reproduces the general trend shown by the data. The choice of these values as well as the ranges chosen for the sensitivity analysis were based on values commonly reported in the literature. For instance, it is often reported that the average fractal dimension of relatively large flocs is around 2 (e.g. Meakin, 1988; Winterwerp, 1998; Dyer and Manning, 1999; Kramer and Clark, 1999). The characteristic floc size (D_{fc}) at which this value is expectedly reached was set to 2000 μm, after conducting a series of calibration tests. The density of primary particles forming real flocs was set to

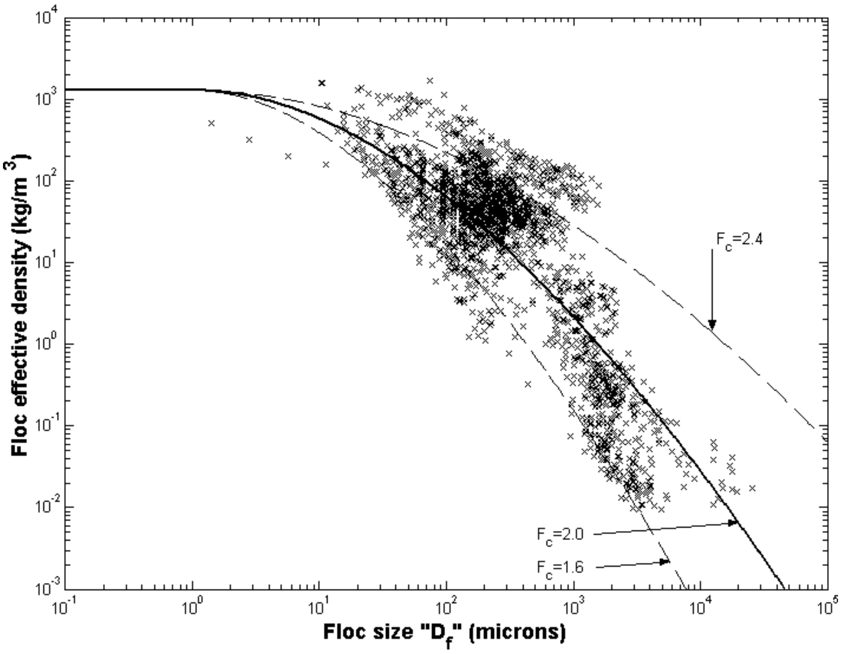

Figure 3 Comparison between data of floc effective density and the proposed model for three different F_c (1.6, 2.0 and 2.4) of flocs, $d = 1.0\,\mu$m, $D_{fc} = 2000\,\mu$m, and $\rho_s = 2300\,\text{kg m}^{-3}$.

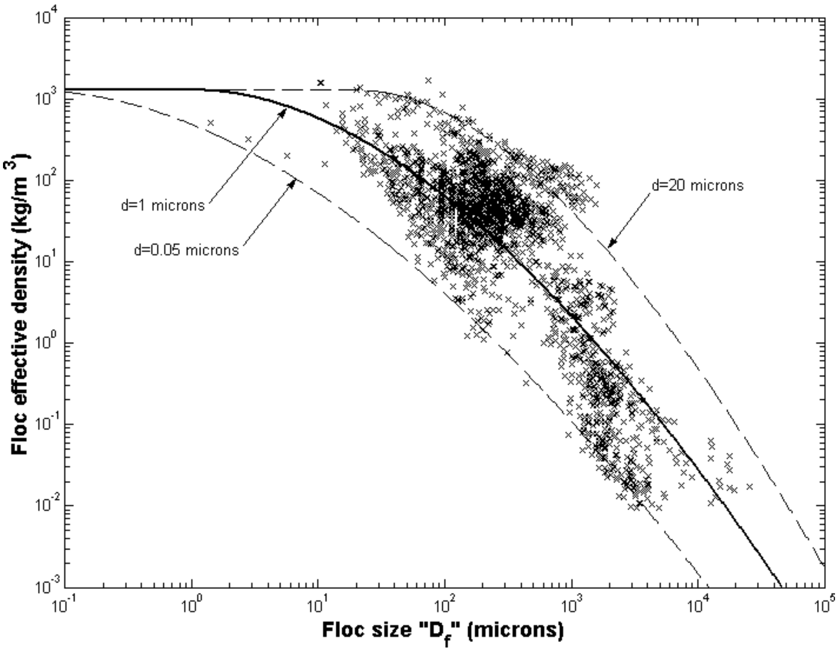

Figure 4 Comparison between data of effective density (cross symbol) and the proposed model for three different diameters d (0.05, 1 and 20 μm) of primary particles, $D_{fc} = 2000\,\mu$m, $F_c = 2.0$ and $\rho_s = 2300$ kg m^{-3}.

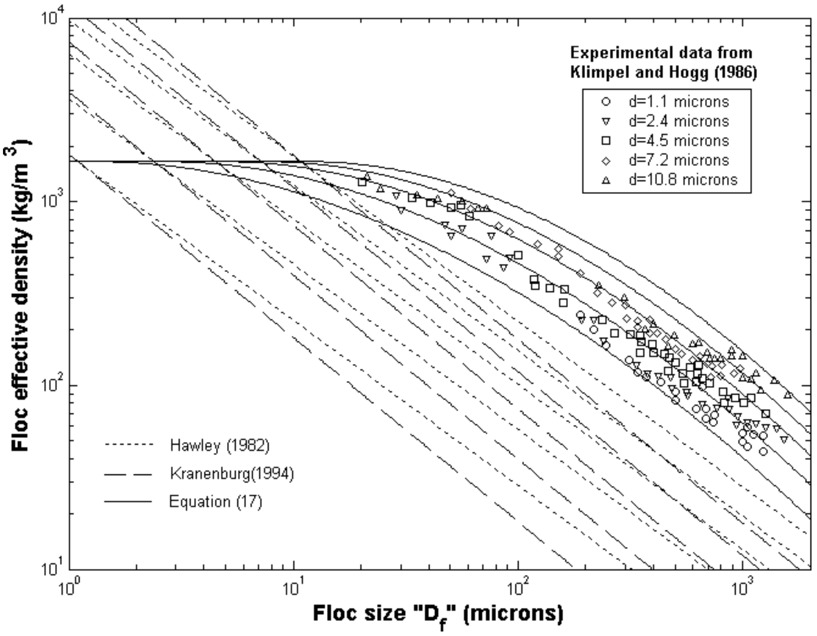

Figure 5 Illustration of effects of the size of primary particles on the effective density of flocs: comparison between experimental data from Klimpel and Hogg (1986, symbols) and proposed model (solid lines) with $D_{fc} = 2000\,\mu$m, $\rho_s = 2650$ kg m^{-3} for quartz, F_c equals 2.38 and d (mass mean diameter) equals 1.1, 2.4, 4.5, 7.2, 10.8 μm (solid lines from left to right). Corresponding results using models proposed by Hawley (1982) and Kranenburg (1994) are also shown in doted and dashed lines, respectively.

the average value of 2300 kg m^{-3} typical of clay minerals that are common in flocs. Manning and Dyer (1999) have proposed a value of 2256 kg m^{-3}.

From the four parameters discussed above, d is one to which predicted effective density shows the greatest sensitivity. As shown in Fig. 4, the effective density of flocs increases with d. The data are well represented by the model, at least in terms of trends, when d varies from 0.05 to 20 μm. The relatively large values of effective density apparent in the data at floc size around 1000 μm derive from laboratory data from Klimpel and Hogg (1986). They used pure quartz (density 2650 kg m^{-3}) instead of natural clay. This size range of the primary particles is not unrealistic, as flocs may contain silt and clay particles with sizes up to 63 μm (Hill *et al.*, 1998), or smaller particles (marine particles) of 0.02 μm for which Martinis and Risovic (1998) have associated a fractal dimension of 3. Moreover, the significant effect

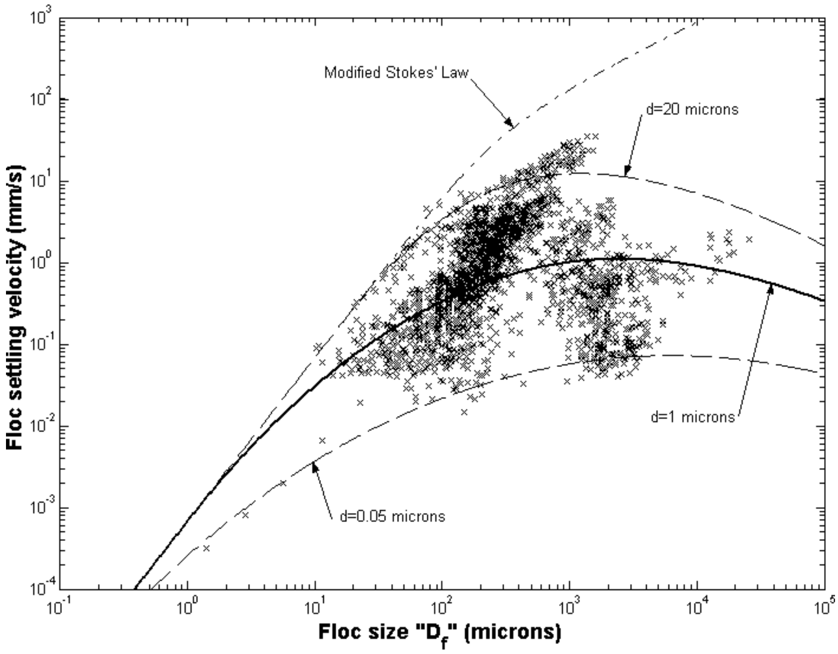

Figure 6 Comparison between data of floc settling velocity (cross symbol) and predictions using the modified Stokes law and the proposed model for floc effective density (Eq. (17)) for three different diameters d (0.05, 1 and 20 μm) of primary particles, $D_{fc} = 2000$ μm, $F_c = 2.0$ and $\rho_s = 2300\,\mathrm{kg\,m^{-3}}$. Prediction using the modified Stokes law is also shown.

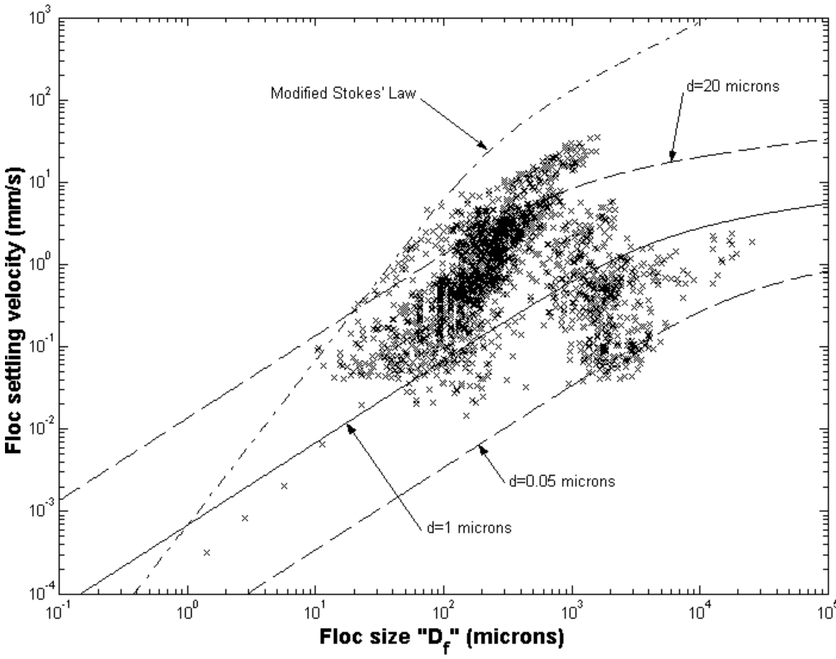

Figure 7 Comparison between data of floc settling density (cross symbol) and the model proposed by Winterwerp (1998) for three different diameters d (0.05, 1 and 20 μm) of primary particles. Prediction using the modified Stokes law is also shown.

of size of primary particles on effective density of flocs found in this study is in agreement with experimental data obtained by Klimpel and Hogg (1986) and with what Hill *et al.* (1998) have argued more recently as the underlying cause of marked variability in size versus settling velocity data. From system variables such as mixing intensity, mixing time, flocculent concentration, sediment (pure quartz) concentration, and size of primary particles, Klimpel and Hogg (1986) found in their experimental study that the size of primary particles had the most significant effect on the effective density of flocs. The latter increases with the size of primary particles for any given floc size. The data compare favorably with the proposed model using Eq. (17) (Fig. 5). The factor ϕ in this equation was evaluated from the size distributions of the primary particles available in the paper of Klimpel and Hogg. For each series of data, i.e. each size of primary particle, Eq. (17) was eye fitted to the data considering various values of the F_c, the second important parameter affecting the effective density of flocs. The density of primary particles ρ_s and D_{fc} were kept constant at 2650 (density of pure quartz) and 2000 μm, respectively. The fitting shown in Fig. 5 was obtained with F_c equals to 2.38 for d (mass mean diameter) equal to 1.1, 2.4, 4.5, 7.2 and 10.8 μm. This value of F_c (2.38) is close to the value 2.4 of the fractal dimension obtained by Ray and Hogg (1987) for flocs formed under similar conditions as those considered by Klimpel and Hogg (1986). For comparison, results obtained using models proposed by Hawley (1982) and Kranenburg (1994) are also shown in Fig. 7. These are the only models in Table 2 that include the size of primary particles as a variable in their expressions.

The predicted effective density also shows important sensitivity to the parameter F_c (Fig. 3). This behavior of the model is supported by observations, as for instance Martinis and Risovic (1998) reported that the magnitude of the fractal dimension is related to the mechanism of aggregate growth. Aggregates formed through particle–cluster aggregation have fractal dimensions larger than those obtained from cluster–cluster aggregation even if they have the same size. The range of F_c considered in this study (Fig. 3) reflects also what is commonly reported. Fractal dimensions for clay aggregates ranging from 1.4 to 2.4 have been reported in the literature (see for instance Meakin, 1988; Kranenburg, 1994; Li and Logan, 1995; Kramer and Clark, 1999).

The proposed model for settling velocity (Eq. (18)), considering monosized primary particles, compares well with the observations (Fig. 6). Compared to the models shown in Figs 1 and 2, the proposed models reproduce much better the data of effective density (Fig. 4) and of settling velocity (Fig. 6). The scatter in the data and their trend are well captured by the present models when different values of d or F_c are considered to represent more precisely the observations. For small floc size, depending on the size of the primary particles considered in the simulations, the model converges to Stokes law as do the data. When floc size increases, the model shows an increase of the settling velocity weaker than the increase shown by the modified Stokes law. The increase depends on the size of the primary particles and vanishes at a floc size of 1000 to about 5000 μm, depending on the size of the primary particles. Beyond this range of floc size, at which a maximum settling velocity is reached, the model shows a decrease of the settling velocity when the floc size increases further. This trend is very well supported by the data in Fig. 6. As shown in Fig. 4, this is due to the decrease of the effective density of flocs when their size becomes large and, of course, to the increase of the drag coefficient due to the increase of the floc size. This observation provides a sound explanation for the relative constancy of settling velocities of large flocs despite observed variability in size (Hill *et al.*, 1998).

5 Discussion

The fact that the structure of real flocs is in general not self-similar, which is conceptually interpreted by the variability of the fractal dimension with the scale in this paper, is not new. It has been reported by various investigators. For instance, fractal dimensions ranging from 1.0 to 3.0 have been reported for aggregates of different types of primary particles formed by various aggregation processes (see Li and Logan, 1995, for a review). Kranenburg (1999) recognized that because of the large variability of properties such as mineral composition, particle size distribution and organic-matter content, flocs may not have self-similar structure. In their field study on fractal dimension of sediment flocs, De Boer *et al.* (2000) have observed an increase of the fractal dimension with a decrease in floc size. Also, microbiological effects often produce floc structures that cannot be described properly with the assumption of self-similarity (Fennessy *et al.*, 1994). In their experimental investigation of floc structure using image analysis, Spicer and Pratsinis (1996) found that application of the concept of fractal geometry to describe the structure of flocs revealed a decrease of fractal dimension as the flocs grew. This observation was also reported by Tambo and Watanabe (1979) and Oles (1992).

One of the important consequences of the variability of the fractal dimension with floc size is that the assumption made on the invariance with changing floc size of the number of particle bonds in a critical plane (Bremer *et al.*, 1989, Kranenburg, 1999) becomes questionable. The decrease of fractal dimension with floc size suggests that this number will decrease when flocs grow and, accordingly, floc strength will decrease with increasing size. In other words, flocs become more fragile as they grow, which is widely corroborated by observations reported by many investigators.

True fractal structures are an idealization. No curve, surface or volume in the real world is a true fractal. Real flocs are produced by processes that act over a finite range of scales only. This is why, perhaps, estimation of their fractal dimension, if the terminology is still applicable, varies with the scale. Even if this suggestion seems to contradict the hypothesis of self-similarity as applied to flocs, it is well supported by the developer of this fractal theory, Mandelbrot (1975), when he explained this theory using the example of the wire ball observed at various scales in his book at page 13. More interestingly, results of this study show that it is, perhaps, appropriate to consider the fractal dimension for flocs just as the parameter "D" (exponent) found by Richardson earlier (quoted by Mandelbrot, 1975) when he established the dependence of measured length of the coastline of Great Britain on the measuring scale used, as discussed by Mandelbrot (1975) in page 24. Of course, further studies are required to establish the appropriate relationship between the fractal dimension, or more properly the exponent "D", and the size of flocs. The power law proposed in this paper (Eq. (14)) improves the fit to data, as compared to the existing models.

This study has shown also that the size d of the primary particle has a dominant effect on prediction of effective density of flocs and, hence, on predictions of settling process. In fact, two of the five models considered in this study, models proposed by Hawley (1982) and Kranenburg (1994), include the effects of this parameter in their expressions. A sensitivity study on these two models for the effective density of flocs and the model of Winterwerp (1998) for settling velocity was conducted to test if these models are able to reproduce the data when d is varied. Only results for the settling velocity obtained with the model of Winterwerp are shown (Fig. 7). The settling velocity models of Hawley provide very similar results as the Winterwerp model. Results related to effective density showed the same behaviors. All the models are sensitive to the parameter d. The model proposed by Winterwerp covers a wider range of the data than the model of Hawley when d varies between 0.05 and 20 μm. Nevertheless, the trends shown by the data at small and large floc sizes are poorly simulated by both models. For instance, the convergence to the Stokes law at small sizes and the decrease of the settling velocity at large floc sizes are not captured by the models. It is worthwhile to mention that recently Tang *et al.* (2002) proposed a fractal-based method to describe the settling behavior of flocs. The method is based on a model to calculate the density of flocs (their Eq. (9)) as a function of the size of primary particles. Assuming the equivalent spherical diameter of flocs as the diameter of collision, we found that their density model transforms to the model proposed by Kranenburg (1994) times a "structure prefactor" k_c. Tang *et al.* estimated the latter by $k_c = 0.414F - 0.211$. Under the assumption of constant fractal dimension, the expression proposed by Tang *et al.* becomes similar to the model of Kranenburg.

Regarding the factor ϕ related to the size distribution of primary particles (Eqs (17) and (18)), a relatively weak effect on both the effective density and the settling velocity of flocs was shown when simulations were compared to the experimental data of Klimpel and Hogg (1986). Nevertheless, further sensitivity analysis should be conducted considering data with real size distributions of primary particles. At this stage we recommend application of the model using the median size d_{50} of the size distribution. Using median size, this factor becomes unity. In addition, the model presents good flexibility for introducing the controlling parameters and for integrating accurate characterization of settling velocity into numerical models designed to simulate transport of cohesive sediments and other type of marine particles. The model is also well adapted for non-linear fitting to settling velocity data.

Finally, it is worthwhile to mention that the scatter shown in the data is not necessarily completely related to the natural processes controlling the formation of flocs, but could be related also to the relative uncertainty of different measurement techniques used by the community. For instance, in methods based on image analysis the resolution of the cameras used in the observations varies widely and can affect the estimation of floc size (Milligan, 1996). Moreover, the preconditioning of images can certainly vary from one study to another. Operations such as image erosion and dilation are very important during any image analysis dealing with edge detection and estimation of particle size. All of these image-analysis-related factors may have important effects on the scatter shown by data of settling velocity.

6 Conclusion

A series of 26 data sets of floc settling velocity was collected from the literature. The data derived from laboratory and field observations performed under various conditions and at different locations. Using a modified Stokes law, this series was transformed to a series of effective density data. A consistent decrease of the effective density of flocs with increasing size was observed. The floc size covered by the data varies between 1.4 and 25,500 μm. Five commonly used models, including those based on strict application of the concept of fractal geometry, fail to reproduce the data over the full size range. New models of settling velocity and effective density of flocs propose relaxation of assumption that fractal dimension is invariant with floc size. This proposal is supported by observations and is consistent with limitations of fractal model when scales of flocs approach scales of component particles. The new models are useful for future modeling work because they best describe observed variability over a large range of floc size. Recommended values of the parameters needed for future applications of the models, if they are not measured, are $d = 1.0\,\mu$m, $D_{fc} = 2000\,\mu$m, $F_c = 2.0$, and $\rho_s = 2300\,\text{kg m}^3$.

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Notation

C_1, C_2 = Constants
C_d = Drag coefficient
D_f, D_{fc} = Equivalent spherical diameter of flocs
d_{50} = Median diameter of primary particles
d_i = Diameter of the ith primary particle
F, F_c = Three-dimensional fractal dimension of flocs
k = Number of primary particles forming a floc
g = Gravitational acceleration
V_f = Floc settling velocity
Re = Particle Reynolds number
α, β = Coefficients relating fractal dimension to floc size
μ = Dynamic viscosity of water
ν = Kinematic viscosity of water
ρ_f = Floc density
ρ_s = Density of component particles within flocs
ρ_w = Water density
θ = Particle-shape factor

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