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Seasonal variation of floc population influenced by the presence of algae in the Changjiang (Yangtze River) Estuary

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Abstract

The variation of the floc population in the Changjiang Estuary has been studied for both winter and summer season as a function of the presence of living (micro)algae. The influence of algae has been characterized through the use of the chlorophyll-a concentration to suspended sediment concentration (CC/SSC) ratio. Two whole tidal cycle sampling campaigns were carried out and a full set of parameters (particle size distribution, particle concentration, salinity, velocities, chlorophyll-a concentration) was recorded as function of time for 6 vertical depths. It is found that the floc population can be described by three particle classes. The two most dynamic classes (microflocs and macroflocs) co-exist in the water column. It was nonetheless found, due to the correlation between CC/SSC and particle sizes that the system is at steady state, both in summer and in winter. This can be explained by the limited flocculation ability between the classes due to

their segregation in the water column. In winter, macroflocs are found at the top of the water column but their amount and size are very reduced with a mean CC/SSC value of $13 \pm 11 \mu\text{g g}^{-1}$. In summer, algae-rich macroflocs are abundant at the top of the water column with a mean CC/SSC value of $21 \pm 18 \mu\text{g g}^{-1}$, especially at flood tide. Microflocs, on the other hand, have a higher density and are generally found deeper in the water column. At high water slack, both macroflocs and microflocs will settle but will never catch-up. The fact that the flocs are at steady-state in terms of flocculation is of importance for sediment transport modelling.

Key words: Flocculation; Algae; Changjiang Estuary; Yangtze Estuary; Particle size distribution; Floc size; Tidal variation; Seasonal variation

1. Introduction

Flocculation of fine sediment particles, resulting in time and space-dependent settling velocities, plays an important role in the fine sediment dynamics in estuaries (van Leussen, 1988). A large number of studies have been devoted to study flocculation, by field observations (Alldredge and Gotschalk, 1989; Braithwaite et al., 2010; Fennessy and Dyer, 1996; Guo et al., 2017; Li et al., 2017), laboratory measurements (Guan et al., 1996; Manning and Dyer, 1999; Wan et al., 2015) and modelling (Khelifa and Hill, 2006; Mietta et al., 2011; Winterwerp, 1998).

Many factors can influence sediment flocculation and studies have concentrated on the influence of shear rate (Keyvani and Strom, 2014; Manning and Dyer, 1999), SSC and salinity (Eisma and Irion, 1993; Fettweis et al., 2010, 2007; Karbassi et al., 2014; Manning et al., 2010a, 2010b; Mietta et al., 2009a; Pérez et al., 2016; van Kessel et al., 2011). It has long been recognized

that in natural environments, organic matter is always part of flocs (Eisma et al., 1991). In that review article, the authors confirm that the kinetics for flocculation are related to the tidal cycle, and that microscope observations have shown that flocs consist of mineral particles held together by organic matter (organic matter with or without hard parts of organisms). They state that in west-European estuaries there is no evidence for an influence of salinity on in situ floc size distributions. A same result was found for our site of interest, i.e. the Changjiang Estuary (Guo et al., 2017) and the Seine estuary (Verney et al., 2009). As salt-induced flocculation is a slow process it will only be dominant in the case that no other flocculating agent is present (like polyelectrolytes, stemming from industry or produced by microorganisms) (Mietta et al., 2009a, 2009b). Salinity can however influence the binding of organic matter to mineral clay (Wilkinson et al., 2017).

Polyelectrolytes like Extracellular Polymeric Substances (EPS) have been shown to drive flocculation (Droppo, 2001; Furukawa et al., 2014; Lee et al., 2017; Paterson and Hagerthey, 2001; Tolhurst et al., 2002; Uncles et al., 2010). EPS primarily consists of carbohydrates, proteins, nucleic acids, and polymers, which can absorb fine sediment particles and change their surface property (Ni et al., 2009; Sheng and Yu, 2006). When we refer to algae-induced flocculation in this article, it is implied that EPS-induced flocculation (whereby the algae can produce EPS) is also accounted for. There is a strong correlation between algae bloom season and flocculation, when higher temperature and higher light intensity promote algae activity and EPS secretion enhance sediment particles flocculation (Chen et al., 2005; Fettweis and Baeye, 2015; Lee et al., 2017; Shen et al., 2018; van der Lee, 2000). Most research associated to algae effects usually focus on the algae bloom season (spring season, from March to June). We wanted to investigate the role of algae in different seasons (summer and winter). One of the purposes of the present article is to compare floc size distribution

in summer and winter, related to microalgae content of the flocs.

Most studies also rely on observations made using instrument at a fixed position above bed. In our study, we performed measurements as function of both depth and time in order to highlight the vertical variation in particle distributions and their composition in terms of algae content.

In our previous study, realized in summer (Deng et al., 2019), we already found that the living microorganisms (algae) have a specific influence on flocculation in summer, outside the algae bloom season, as function of different hydrodynamics conditions and position in the water column. It was shown that the algae particles could participate in the flocculation process and a key parameter—the sediment to chlorophyll concentrations ratio (CC/SSC) —was introduced that reflected the dependence of flocculation on algae concentration. In particular, it was concluded from the study that large algae flocs (> 100 microns) were formed in a region devoid of sediment particles whereas the lowest size fraction (< 5 microns) is composed of sediment particles eroded from the bed at high shear. The particle size distribution (PSD) but also composition in the water column is therefore dynamic and changes with depth and hydrodynamic conditions. As sediment transport models are relying on estimation of the particle density (to assess their settling velocity), it is important to study the variations of floc properties in space and time.

In the present study, we compare the algae influence on flocculation in different seasonal conditions (summer and winter), and show how the floc properties such as effect density and settling velocity are changing with the seasons. Most of sediment accretion occurs in the flood season (summer) while erosion occurs in the dry season (winter) in the Estuary Turbidity Maximum (ETM) area of Changjiang Estuary (Li et al., 2018). It will therefore be interesting to compare the dependence of CC/SSC ratio on shear, particle size and position in the water column in winter and

in summer. Typical questions we like to answer are:

– How is the floc size evolving as function of season and tidal cycle in the whole water column?

– How are floc properties (density, settling velocity, size) linked with sediment/algae ratio?

2. Methodology

2.1 Site description

Two full neap tidal cycle (13 h) sampling programs were carried out from 10th to 11th of January 2014 and 25th to 26th of July 2016 in the South Passage of Changjiang Estuary (Figure 1). River discharges are about 11,000 m³ s⁻¹ during winter survey and 65,000 m³ s⁻¹ during summer survey, respectively. There were no storms in the period before the observations, the tidal current that varied with tidal phases was the main driver of turbulent shear.

In the Changjiang Estuary, the annual mean tidal range is about 2.6 m. The depth-averaged velocity can reach up to 1 m s⁻¹ in surface water (Chen, 1995), and the depth-average velocity is higher in summer (0.76–0.96 m s⁻¹) than winter (0.43–0.78 m s⁻¹) (Yun, 2004). The concentration of chlorophyll-a (which is a proxy for algae concentration) in the South Passage and near the study site has been given in Chen et al. (1999) and Wang et al. (2015) and the values are found to be in the range 3–30 mg m⁻³ (= 3 to 30 µg l⁻¹) in surface water. The suspended sediments in the Changjiang Estuary are mostly fine-grained particles (their amount can reach up to 95%). The average D₅₀ of the suspended sediment is 7–11 µm, whereas the D₅₀ range of the top bed sediment is 5.9–182.8 µm in the ETM zone. The average D₅₀ is 14.2 µm in South Passage area (although there are spatial variations). In particular, most of sediment in the study site (located in South Passage) has a D₅₀ of about 8.2 µm, and consists of clayed silt (about 20% clay, 70% silt and 10% sand) (Liu

et al., 2007). The Suspended Sediment Concentration (SSC) in the Changjiang Estuary varies greatly over time and space, ranging from 0.1 to 20 g l⁻¹. The prevailing wind is in South-East direction in summer with wind speeds of about 9.4 m s⁻¹. The prevailing wind in winter is in North-West direction with wind speeds of about 7.4 m s⁻¹ (Yun, 2004).

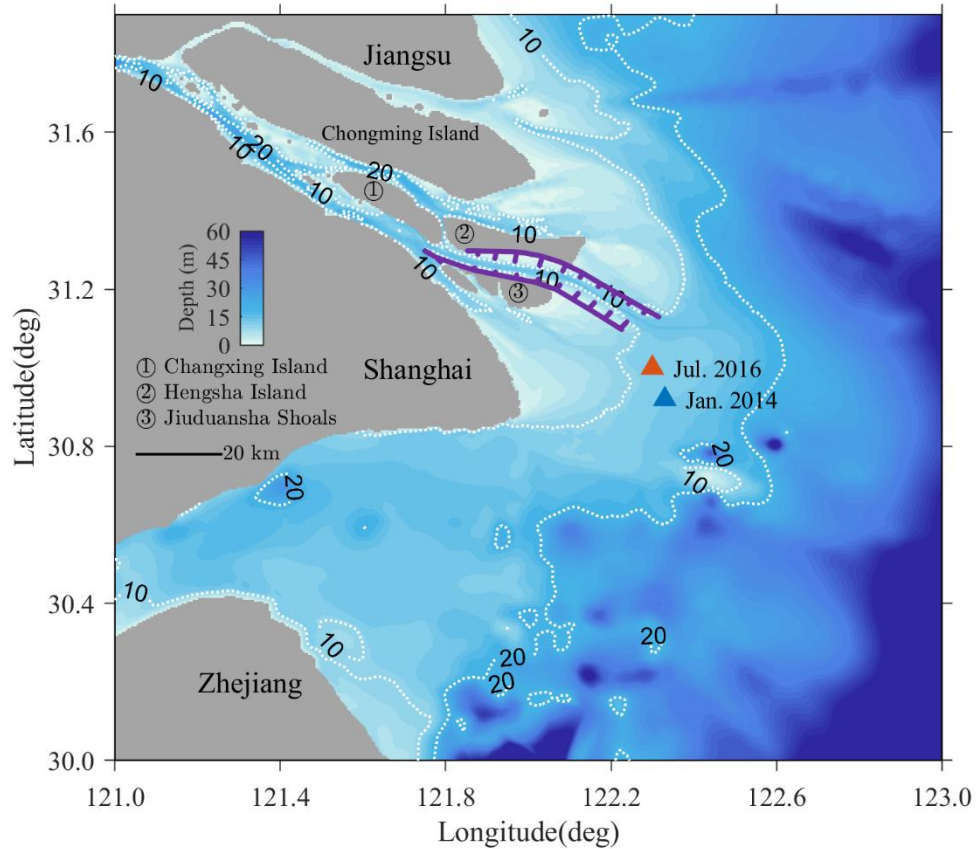


Figure 1 Map of the morphology of the Changjiang estuary and the study sites in summer (July in 2016) and winter (January in 2014).

2.2 Data acquisition

Each field survey was carried out with a multi-instruments device and covered a whole tidal cycle. The device included the LISST-100C to measure particle size distribution and volume concentration, OBS-3A for water turbidity, salinity, a multi-parameter water quality meter (Manta

2) for chlorophyll-a concentration and ADCP (300 kHz) for hydrodynamics. All the instruments were attached together to make sure that all the detectors would measure at the same position. All the instruments were set to record at an interval of 1 second. A full vertical profile was measured every hour and the instruments were pulled slowly from bottom to surface at the speed of 0.05 m s⁻¹. At specific locations in the water column, corresponding to the positions where the water samples are taken (see underneath), the instruments were left for 2 min at the same position in order to acquire statistically significant data. The data presented in the article is the average over the 2 min period. The hydrodynamic parameters were measured by ADCP (Acoustic Doppler Current Profiler, 300 kHz), which was set up 0.5 m under the ship with a 1.71 m blanking distance and a vertical resolution (bin size) of 0.5 m. Records of the water current direction and velocity were done in real time, with an accuracy of $\pm 0.5\%$ / ± 5 mm/s (RD Instruments, 1994). Temperature and salinity were also recorded.

In addition, water samples were collected by 1.2 L water sampler (horizontal trap sampler), then divided into two glass bottles, one for SSC analysis (even hours) and another for chlorophyll-a analysis or particle size analysis (the odd hours). We took these double water samples at 6 vertical depths (0H, 0.2H, 0.4H, 0.6H, 0.8H, 1H) per hour. In our definition, 0 H represents the surface and 1 H the bottom. The height corresponding to 1 H was sampled 0.5 m above the bed. A water sample for each height was filtered through a 0.45 μ m cellulose acetate pre-weight filter paper, dried and weighted to estimate the sediment concentration distribution through the water column.

2.2.1 Particle Size Distribution and concentration

The particle size distribution and volume concentration were recorded by LISST-100X with

the path length reduction module (PRM) of 90%. The LISST-100X is a submersible multi-parameter system for in-situ measurements (Agrawal and Pottsmith, 2000; Pottsmith and Keir, 2013). A particle size distribution and volume concentration were obtained by small-angle light scattering (670 nm diode laser). The silicon detector has 32 specific log-spaced angle ranges. The raw data was post-processed to obtain particle size distribution and volume concentration. The validity of the data was assessed by checking the transmission value (of the raw data) which should be between 0.3 to 0.9 (Pottsmith, 2015). Furthermore we have thoroughly checked the data for a possible schlieren effect (Mikkelsen et al., 2008), by comparing LISST and OBS data, and discarded the data affected by it.

The OBS-3A (Optical Backscatter Sensor) is an optical sensor that measures turbidity and was used, after calibration, to estimate the Suspended Sediment Concentration (SSC). The calibration regression coefficients were: $R^2 = 0.9194$ in winter and $R^2 = 0.9439$ in summer, respectively.

2.2.2 Chlorophyll-a concentration

Chlorophyll-a is used as a proxy for determining the presence of algae (Knap et al., 1996). The concentration of chlorophyll-a found in a water sample was used to estimate the concentration of phytoplankton (algae) and was recorded with a Manta 2 instrument (Water Quality Multiprobe, Eureka Environmental Engineering Company). Fluorescence sensors were used to induce chlorophyll-a fluorescence by shining a beam of light of corresponding wavelength (435–470 nm) into the water and then measuring the higher wavelength light which was emitted (Eureka Environmental Engineering, 2016; Lamb et al., 2012). The laboratory chlorophyll-a analysis water sample was filtered through a 0.45 μm ultra-fine glass fiber filter paper and stored at $-20\text{ }^{\circ}\text{C}$ for

chlorophyll-a measurements in laboratory. The calibration regression coefficients were: $R^2 = 0.4960$ in winter and $R^2 = 0.8337$ in summer, respectively. More details can be found in a previous study (Deng et al., 2019).

2.2.3 Complementary laboratory experiments

The primary particles analysis was measured by Malvern Mastersizer 2000 laser granularity analyzer (measurement ranges from 0.02–2000 μm , the repetition error is within 3%). Before the instrument measurements, the water sample was handled with hydrogen peroxide (H_2O_2) and hydrochloric acid (HCl) to remove the organic matter and carbonate. Hexametaphosphate ((NaPO_3)₆) was then added as a dispersant and the sample was ultrasonically shaken (for 15 minutes).

2.3 Data processing

2.3.1 Floc properties

In this article, we will refer to “particles” or “flocs” but it should be understood that either particles or flocs can be composed of (a) entirely mineral clay, (b) mineral clay – organic matter aggregates (organic matter being algae and their produced EPS), (c) algae and algae aggregates.

The density $\Delta\rho$ of flocs was estimated from the LISST and OBS data, using equation (2) (Fettweis, 2008; Verney et al., 2009):

$$\Delta\rho = \rho_F - \rho_W = \left(1 - \frac{\rho_W}{\rho_P}\right) \frac{SSC}{V_F} \quad (2)$$

Where ρ_F is the floc density, ρ_W is the water density which is calculated by one-atmosphere equation of state of seawater (Millero and Poisson, 1981), ρ_P is the sediment particle density which

is estimated to 2650 g l⁻¹, SSC is the mass suspended sediment concentration obtained from the OBS and V_F is the floc volume concentration from LISST.

The settling velocity was calculated according to Stokes' law:

$$\omega_s = \frac{D_{50}^2 \Delta \rho g}{18\mu} \quad (3)$$

Where μ is the viscosity of water, D_{50} is the mean particle diameter and g is the gravity constant.

2.3.2 Shear rate

To estimate the shear stress in the water column, the velocity data should be converted into shear rate G (Guo et al., 2017; Pejrup and Mikkelsen, 2010):

$$G(z, H, u_*) = \sqrt{\frac{u_*^3 \times (1 - z/H)}{\nu \kappa z}} \quad (4)$$

Where ν is the kinematic viscosity of the water [m² s⁻¹], H is the total water depth [m], z is the height above bed, κ is Von Karman's constant (approximately 0.4) (Chien and Wan, 1999). The friction velocity, u_* [m s⁻¹] is given by:

$$u_* = \frac{u(z) \times \kappa}{\ln\left(\frac{z}{z_0}\right)} \quad (5)$$

Where $u(z)$ is the current velocity amplitude [m s⁻¹] at position z . z_0 is assumed to be constant and equal to 3 mm. This value has been used in other research of sediment transport in the turbidity maxima of the Changjiang Estuary (e.g. Ge et al., 2012; Guo et al., 2017).

2.3.3 Algae-Sediment Ratio

Algae particles populate the whole water column and biomineral flocs have such a density that these flocs settle to the bottom of the water column. This results in having a higher chlorophyll-a

concentration at the bottom than in the upper layer. Although some species of algae are adapted to low light condition, most of algae would primarily be expected to be found in the upper water layer as they rely on photosynthesis for their growth. This is especially the case for *Skeletonema* which is the dominant algae species in Changjiang estuary, and which has an affinity with surface waters. In order to scale the results with SSC, we defined a CC/SSC ratio to represent the algae relative concentration (Deng et al., 2019):

$$Ratio = CC/SSC \quad (7)$$

where CC is the chlorophyll-a concentration [$\mu\text{g l}^{-1}$], SSC is the suspended sediment concentration [g l^{-1}]. In Deng et al. (2019) it was in particular found that a high chlorophyll-a concentration usually corresponds with high SSC in summer.

3. Results and discussions

Figure 2 and Figure 3 show the time series of the water hydrodynamics and flocculation parameters during the winter and summer surveys, suspended sediment concentration (SSC), salinity, chlorophyll-a concentration (CC) and floc size (D_{50}) were recorded. shear rate (G), floc effective density ($\Delta\rho$), floc settling velocity (ω_s) were estimated using Eqs. (1,2,3).

3.1 Seasonal variations

Each of the two surveys started from flood tide to ebb tide, lasted for 13 hours, and included a whole tidal cycle with four periods: Low Water Slack (LWS), Maximum Flood Velocity (MFV), High Water Slack (HWS) and Maximum Ebb Velocity (MEV).

The water depths as function as time demonstrated the tidal asymmetry with an ebb-dominance

that the flood periods are about 4 hours while the ebb periods are about 6 hours both in winter and summer surveys. Because the observation station was further away from the coast in winter, the water depth in winter (11–12.5 m) was deeper than in summer (7.5–11 m). The water depth variation amplitudes in winter were smaller than in summer. The tidal range in winter was smaller than in summer, which means that the hydrodynamics in winter are less energetic. This was also reflected in the shear rate (Figure 2a, b). The shear rate ranged from 0.1–20 s⁻¹ in winter, and from 0.4–50 s⁻¹ in summer. The shear rate in summer was always higher than in winter for the same tidal period. In addition, the shear rates remained at a high value at flood period and ebb period. The SSC correlated with shear rate both in winter and summer surveys. The maximum SSC usually appeared at the high shear rate periods (MFV and MEV). Although the SSC in bottom water was lower during the summer survey than during the winter survey, the SSC in upper water was higher than during the winter survey (> 0.05 g l⁻¹).

The salinity in winter was higher than in summer because of a smaller river discharge in winter (Figure 2c, d). The salinity in winter ranged from 18.5 to 27.3 PSU (it increased with rising tide), which means that the water column was mainly affected by sea water. The salinity ranged in summer from 4.8 to 22.0 PSU meaning that the water column was always affected by both fresh and sea water. The vertical gradient of salinity indicated different stratifications in winter and summer: water is better mixed in winter than in summer. The stratification corresponded with high CC/SSC in summer at the top of the water column, probably due to favorable algae growth conditions during that period. These flocs are mainly advected from the sea at flood tide (Zhao and Gao, 2019).

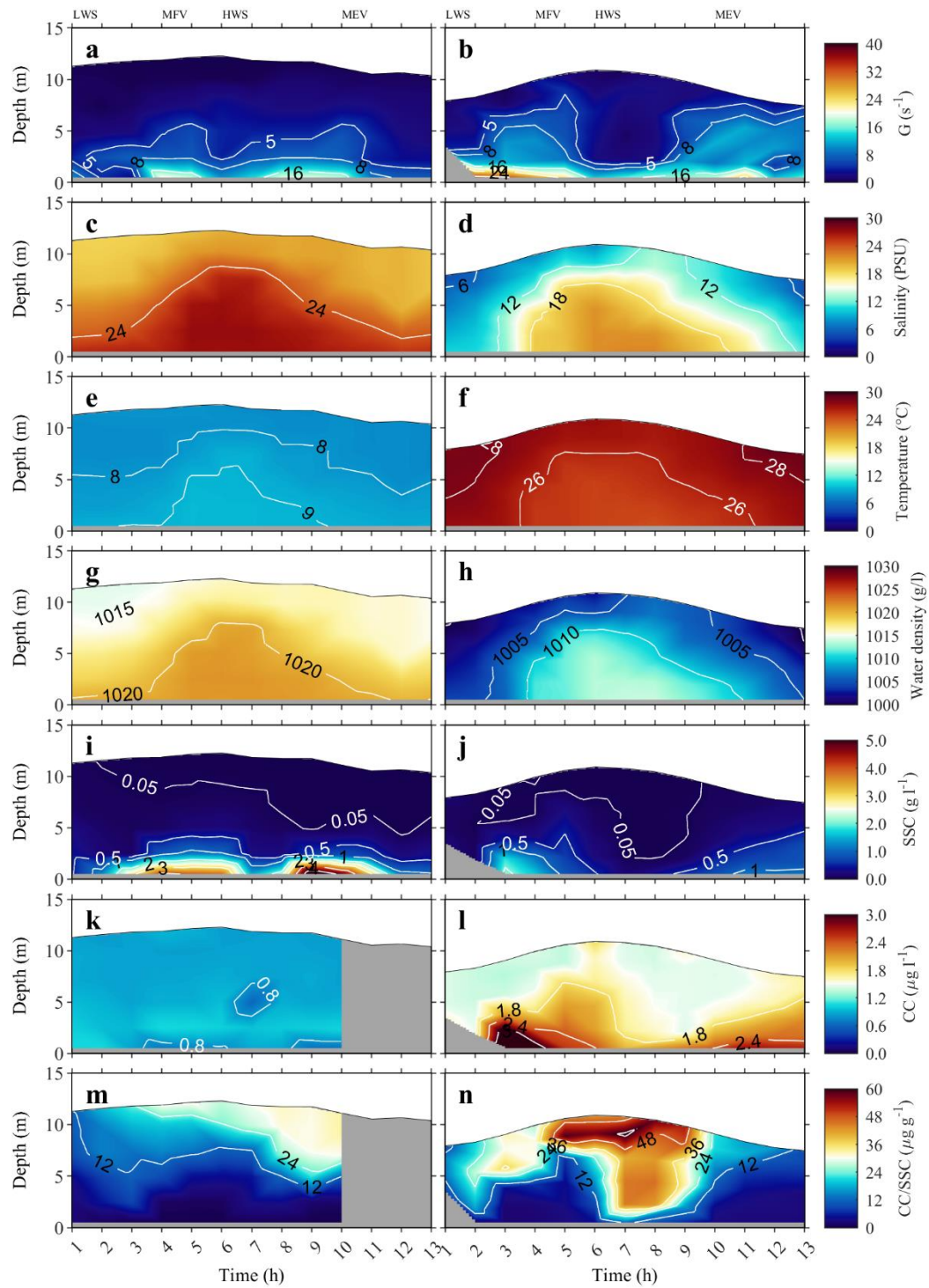


Figure 2 Vertical distribution of shear stress (a, b), salinity (c, d), Temperature (e, f), water density (g, h),

sediment concentration (i, j), chlorophyll-a concentration (k, l) and algae-sediment ratio (m, n).

(Left: 2014-01 winter season, Right: 2016-07 summer.

LWS: 1–2 h, MFV: 4–5 h, HWS: 6–7 h, MEV: 10–11 h).

The temperature in winter (7–10 °C) was lower than in summer (25–29 °C) (Figure 2e, f). The salinity is shown to correlate with the water density for both the winter and the summer survey period: a high salinity corresponds to a high water density and vice-versa. There is no correlation between SSC and water density as there is no correlation between SSC and salinity, a feature that was already observed in diverse estuaries worldwide and this estuary in particular (Eisma et al., 1991; Guo et al., 2017).

3.2 Mean floc properties in winter and in summer

In Figure 3 it is shown that the median floc size (D_{50}) in winter (13–65 μm) is smaller than in summer (13–359 μm). Studies in the same area have shown that Particulate Organic Carbon (POC) correlates positively with chlorophyll-a, and that the amount of POC is reduced in winter as compared to summer (Zhao and Gao, 2019). This is in line with the results presented in Figure 2k, l, where the CC is lower in winter as compared to summer. Flocculation induced by particulate organic matter (as represented by POC) is therefore reduced in winter. Flocs of higher size can only be observed if their effective density is low enough so that their residence time in the water column is significant—this can only be achieved if flocs contain a substantial amount of algae.

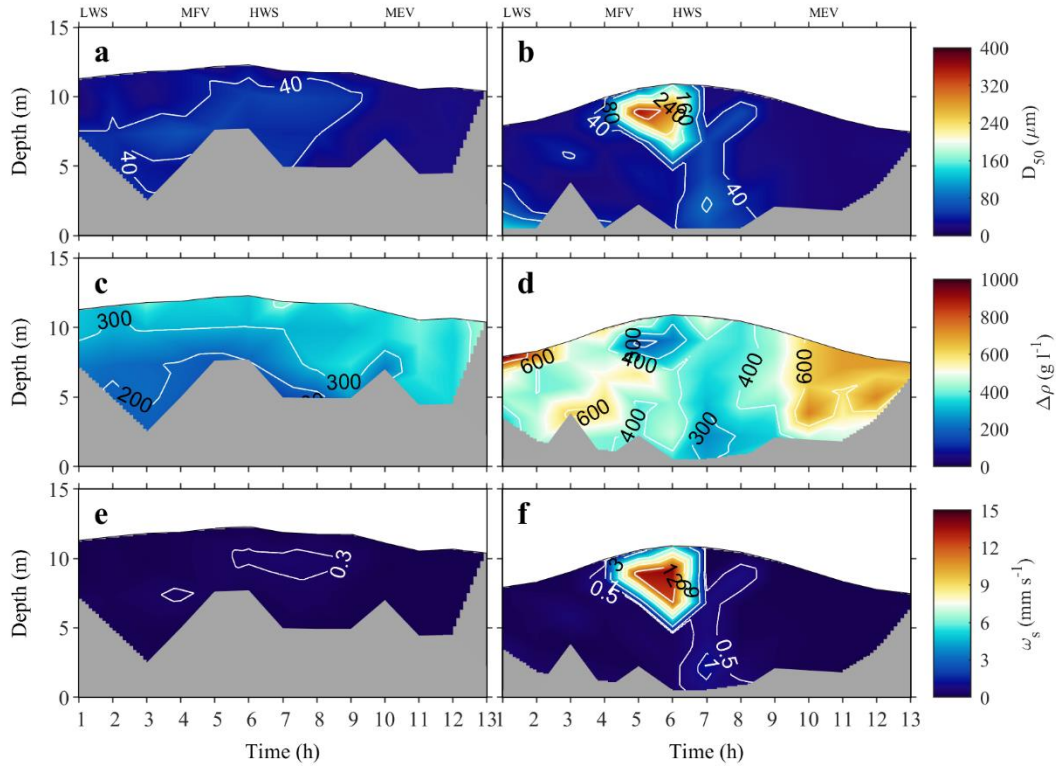


Figure 3 Vertical distribution of mean floc size (a, b), effective density (c, d) and settling (e, f) velocity. (Left: 2014-01 winter season, Right: 2016-07 summer, LWS: 1–2 h, MFV: 4–5 h, HWS: 6–7 h, MEV: 10–11 h)

The effective floc density map shows that large particles are found in the upper layer, especially at HWS. These particles have a higher settling velocity than other particles found at the same time in the water column, but have a similar density. Particles of higher density are found (in all layers) when the shear rate is higher than in other periods (both around 8–12 h in winter and in summer). The mean floc size has a positive correlation with effective density in summer: large flocs usually have a smaller density, and vice versa. This is better represented in Figure 4. This result is consistent with many other studies where the relation between density and size is an exponentially decreasing function (Manning et al., 2006; Manning and Dyer, 1999; Mikkelsen et al., 2007). The mean effective density of flocs in winter appears to be a little smaller in winter than that in summer. This is counter-intuitive if one assumes that flocs in summer are composed of more algae than in winter

(which is confirmed by comparing the CC/SSC ratio in both seasons). However, as discussed in the previous section, the shear rates were overall higher in summer than in winter. This has two effects:

1—the high shear rate in summer leads to more sediment resuspension during MFV, whereby this sediment is flocculated by biological agents (EPS secreted by algae). Even silt-size particles could be trapped in these flocculated structures, leading to flocs with higher density. These flocs even reach the surface water (except for HWS periods), see section 3.3.2. As is shown in Figure 8, the sediment that is in surface water in winter contains more clay-size particles than in summer, which is due to both lower shear rates and lower organic material content in that season compared to summer.

2—There can be a reconfiguration of flocs under shear (a reduction in volume while sediment mass is preserved) resulting in a higher density in summer (Guo, 2018; Sterling et al., 2004; Verney et al., 2009; Winterwerp, 1998). This hypothesis is debatable as reconfiguration is depending strongly on residence time and the nature of the organic matter that composes the floc. The composition of organic matter is known to be different in summer and winter (Hart et al., 1990; Morrissey et al., 2014). The organic matter in summer is usually composed of living algae and their EPS, which can capture effectively mineral sediment particles, leading to higher particle density than in winter. In winter the organic matter is scarce and usually composed of debris and dead organic matter or EPS (Craig et al., 1989; Grey et al., 2001).

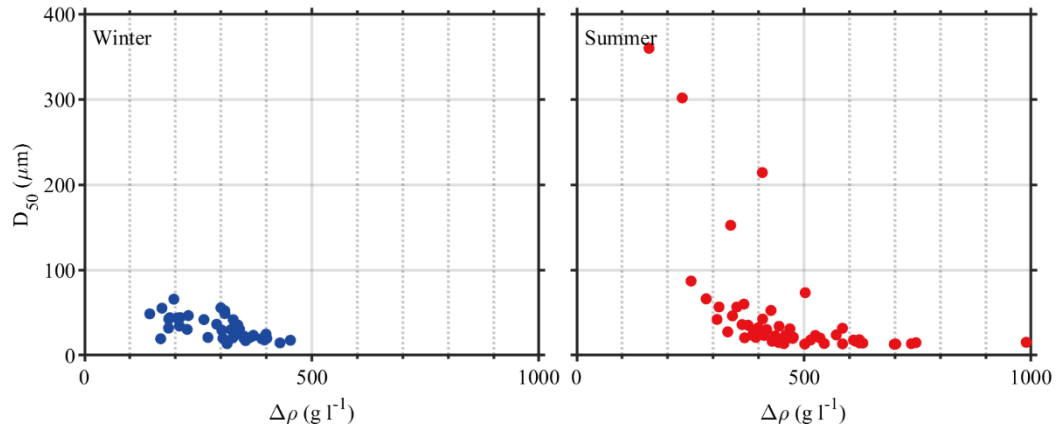


Figure 4 Correlation between effective density with mean floc size

The settling velocity is positively correlated with D_{50} and large flocs have a larger settling velocity than small flocs; this is to be expected since the settling velocity is depending on the particle size squared, whereas it is only linearly correlated with density, see Eq. (2) and that density is within the limited range 140–990 g l^{-1} .

3.2.1 Variations with shear rate, salinity and SSC

There are many studies about the influence of shear rate (Keyvani and Strom, 2014; Mietta et al., 2009a), of salinity (Pérez et al., 2016; Verney et al., 2009), and of SSC (Eisma and Li, 1993; Milligan and Hill, 1998) on sediment flocculation. In the Changjiang Estuary, the correlations between D_{50} and salinity/SSC are poor. In addition, the shear rate displays an overall negative relationship with D_{50} , that is, large flocs usually appear at low shear rate periods (HWS), but the coefficient of determination is poor ($R^2 < 0.5$) both in winter and summer. These poor correlations are in line with previous studies (Deng et al., 2019; Guo et al., 2017).

Table 1 shows the variations of floc properties and velocity, salinity and SSC in winter and summer, and comparisons to other results of North Passage (NP) in Changjiang Estuary in summer season (Guo et al., 2017). The main difference was that there were larger flocs ($> 100 \mu\text{m}$) in summer

in the South Passage compared to the North Passage, especially in surface water, this can be seen by comparing Figure 3 with fig.2 in Guo et al.(2017). As the study area chosen in the South Passage was closer to the sea, the salinity was overall higher and the shear rate lower than in Guo et al.(2017).

Table 1 Characteristics of flocs and velocity, salinity, SSC, CC and CC/SSC in the South Passage (winter and summer) and North Passage (summer)

Parameters	Winter	Summer	North Passage*
D ₅₀ (μm)	32±13	45±64	43±10
Density (g l ⁻¹)	143–453	159–989	60–450
Settling (mm s ⁻¹)	0.02–0.4	0.05–14	0.08–0.3
Current Velocity (m s ⁻¹)	0.5±0.2	0.9±0.4	1.4
Salinity (PSU)	18.5–27.3	4.8–22.0	2–11
SSC (g l ⁻¹)	0.6±1.2	0.3±0.4	0.24±0.12
CC (μg l ⁻¹)	0.9±0.1	1.8±0.5	-
CC/SSC (μg g ⁻¹)	13±11	21±18	-

* Data from Guo et al. (2017).

3.2.2 Variation with CC and CC/SSC in the tidal cycle

The chlorophyll-a concentration (CC) was relatively uniform over the whole water depth with a mean value of 0.9±0.1 μg l⁻¹ (0.6–1.0 μg l⁻¹) in winter and it was more stratified with higher concentrations towards the bed with a mean value of 1.5±0.5 μg l⁻¹ in summer (1.3–3.7 μg l⁻¹). It was observed that the CC in the bottom water is always higher than at the surface (also in winter). We will show (see Figure 11 and section 3.4.2) that a large amount of algae are binding to mineral clay by differential settling during the HWS period. This leads to an accumulation of algae and algae debris at the bed.

The CC/SSC ratio is shown in Figure 2m, n. The ranges of CC/SSC were around 0.1–33.2 μg g⁻¹ in winter and 1.5–61.6 μg g⁻¹ in summer, the higher values in upper water layer can reflect the vertical variation of algae activities. The correlation of CC with SSC in summer was already

discussed in (Deng et al., 2019). Even though the dataset presented here is of another summer (2015 in Deng et al. (2019) and 2016 in the present article), we found that the same conclusions hold. The CC/SSC ratio plot illustrates that the algae percentage is higher in surface water, reflect that the algae (CC) is not always distribute with sediment (SSC), they tend to stay in surface water, due to their buoyancy or phototaxis. From Figure 2m, n, one finds that a threshold value for CC/SSC is around 10–20 $\mu\text{g g}^{-1}$. Above this value, it is expected that flocs will be predominantly governed by the organic matter (algae), as bimodal PSD are found. These type of flocs are found at the top of the water column where algae can benefit from photosynthesis (Reynolds, 2006). During the summer survey the algae-dominated flocs start to populate the whole water column in the period HWS to ebb tide. At low hydrodynamic activity the buoyant algae particles in the water column are less likely to flocculate with mineral sediment particles which will be predominantly found at the bottom of the water column.

The trends for D_{50} as function of CC/SSC are shown in Figure 5. As was already observed in a previous study (Deng et al., 2019), there is no correlation between D_{50} and CC/SSC in the summer period (in 2015). It is confirmed that no overall trend is observed between D_{50} and CC/SSC in summer (in 2016), nor in winter. There is however a marked increase in D_{50} with CC/SSC ratio corresponding with low density particles at 5–6h in summer 2016, which indicates that the algae play a significant role in large flocs formation during flood tide, when a lot of algae particles are imported from the seaside. The details of algae effects on floc PSDs will be discussed in section 3.3 and 3.4.

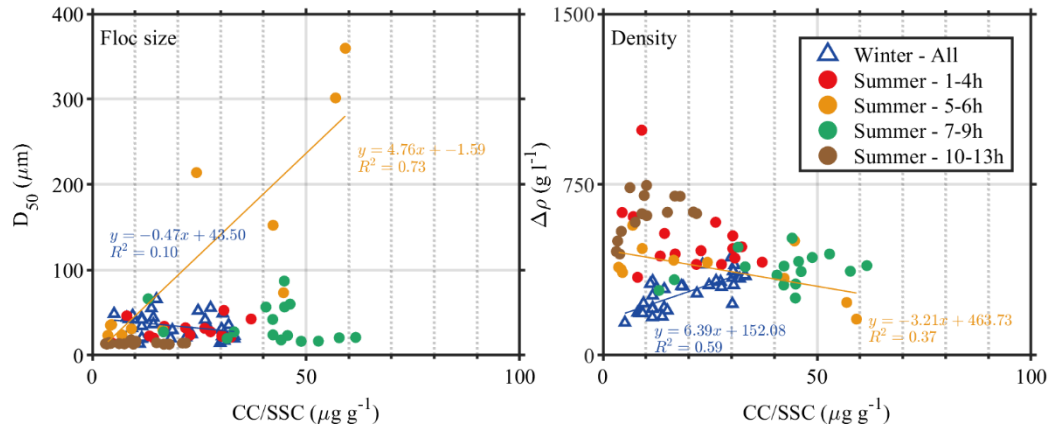


Figure 5 D_{50} and effective density as function of CC/SSC for winter and summer conditions

The density is better correlated with CC/SSC as can be seen in the Figure 5. The decreasing trend observed in summer can be understood as a higher CC/SSC is then linked to a higher CC and therefore a lower density (as in summer, SSC is relatively constant over the whole water column, see Figure 2j). A higher CC/SSC ratio in winter corresponds to a lower SSC (as in winter, CC is relatively constant over the whole water column, see Figure 2k), but as in winter there is a very low CC and SSC, the correlation between density and CC/SSC is not to be trusted. The density is nonetheless much lower than in summer for the same CC/SSC. We will come back to this point in section 3.3.2.

3.3 Particle Size Distributions (PSD) during winter and summer surveys

Large flocs usually form at the upper part of the water column between MFV and HWS both in winter and summer (Figure 3a, b). The vertical variation of flocs PSDs is however different in winter and summer when one takes a closer look at the PSDs at different layers, see Figure 6. Flocs are usually smaller in surface water (0 H) than in middle water (0.4 H) in winter at LWS (2 h) and MFV (4 h) whereas they are small in summer for these periods. This implies that the flocculation processes are different in winter and summer. Large flocs are hardly formed in summer in LWS (2

h), due to higher shear rates. Large flocs are formed at low shear rate both in winter and summer at HWS (6 h). In particular, large flocs can be form in the middle and surface water in summer HWS. This phenomenon will be discussed in the following section.

3.3.1 Variation within the tidal cycle

The PSD in winter at MFV is different from the other periods: at MFV the larger peak of the PSD is found at 150 μm , whereas at the other periods it is around 20 μm . This behavior can be ascribed to small sand particles that are eroded during maximum currents. In comparison, during summer, the bed is more stabilized because of the presence of EPS and thus no large peak in MFV is observed. The largest peak in summer is always around 20 μm at LWS, MFV and MEV, however a second peak, around 100 μm , is distinguishable in three figures indicating the presence of particles in that size range. This confirms the fact that in summer more organic particles, at all depths, are present. At HWS (6 h), two PSDs are observed. Between [0 H–0.6 H], the PSD peaks at about 20 μm (with the peak having an asymmetric shape towards the highest sizes– again indicating the presence of larger particles). Between [0.6 H–1 H] the PSD peaks at 30–50 μm and does not display any large PSD asymmetry, but a significant amount particles in the range 100–400 μm .. This transition seems to be in line with the change in salinity at HWS: clay-algae particles are probably trapped under the pycnocline (see Figure 3j) whereas algae-rich particles are located above (resulting in a large CC/SSC ratio, see Figure 3l). This trapping mechanism has also been reported by other authors (Lee et al., 2016; Ren and Wu, 2014; Yao et al., 2016). The evolution of the PSD around the changes in salinity gradients is shown in more details in Figure 7, reflecting the very dynamic PSD's around HWS.

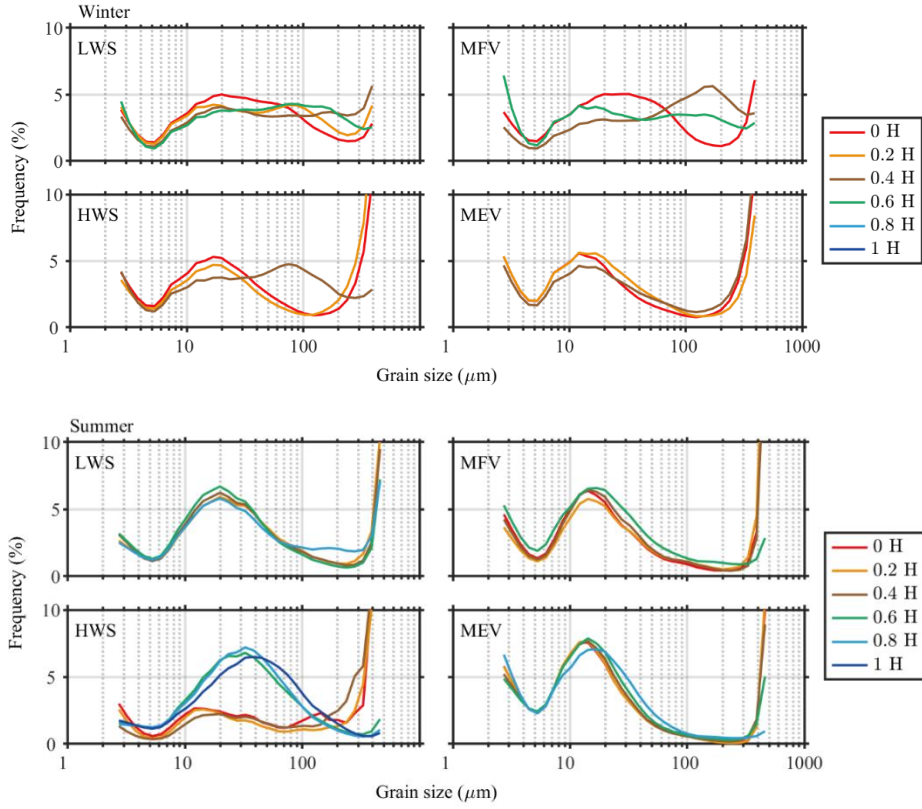


Figure 6 Vertical variations of floc size distribution measured by LISST in different tidal phases, at neap tide.

The measurements are presented for LWS (2 h), MFV (4 h), HWS (6 h) and MEV (10 h).

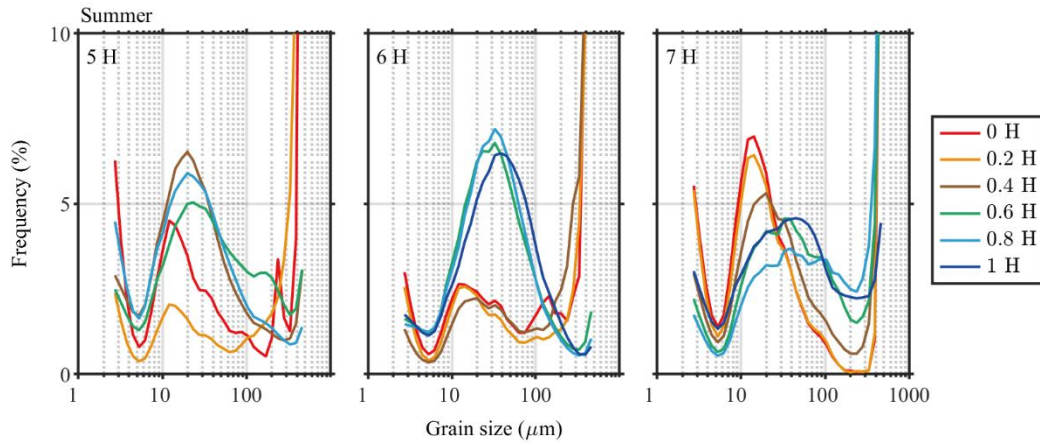


Figure 7 Vertical variations of flocs PSDs in summer during the period 5–7 hours.

3.3.2 Particle characteristics at the top of the water column

The PSD of the particles collected at the top of the water column (in the surface water) was further investigated by comparing the PSD measured in-situ and the PSD obtained in the laboratory

on samples treated so as to remove organic matter. These laboratory samples are labelled “primary particles” (pp in short). The results are presented in Figure 8. It is found that primary particles in summer ($D_{50} = 6 \mu\text{m}$) have the same mean size than in winter ($D_{50} = 5 \mu\text{m}$).

The PSD of the treated samples from the winter survey displays the presence of more fine clay-size particles ($< 1 \mu\text{m}$) than in the samples from the summer survey. The difference can be explained by the fact that in summer fine particles are more likely to be captured by organic matter and hence be found at deeper depths. In winter fine particles (devoid of organic material) can populate the whole water column. This very fine material cannot properly be recorded by LISST, as the lower size range of the LISST is $2 \mu\text{m}$. A remarkable amount of large particles are found in the surface water in-situ, both in summer and winter. These particles are usually not expected to be found in surface waters if they are pure mineral particles (with a density close to 2600 g l^{-1}). We nonetheless found, after treating the summer samples so as to remove organic matter, that large particles can still be found (see Figure 8, summer, dashed lines). The nature of these particles is not known, they might be skeletons of diatoms. Particles were monitored by LISST and OBS, from which particle densities were deduced using equation (2). As LISST cannot properly measured the fine material in the size range ($< 2 \mu\text{m}$) and that a significant amount of fine particles in that size range is found in suspension in winter, the estimation of the density is biased. (see Supplementary information, where the scattering in calibration lines for SSC in winter is shown). This results in the fact that flocs in summer appear to have a higher density than flocs formed in winter as discussed at the end of section 3.2.2.

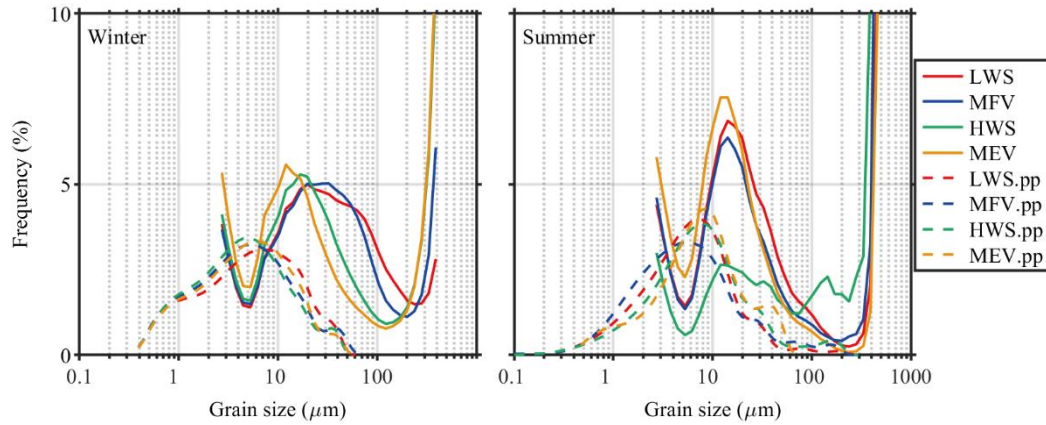


Figure 8 Surface water size distribution of both dispersed (“pp”, i.e. measured in the lab after deflocculation) and flocculated particles (as measured in-situ) in different tidal phases.

3.4 Floc classes

Following what other authors do, we subdivided the PSD into 3 classes: single-grain : particles of size $< 5 \mu\text{m}$, microflocs: particles of size $> 5 \mu\text{m}$ and size $< 200 \mu\text{m}$ and macroflocs of size $> 200 \mu\text{m}$ (Mikkelsen et al., 2006). It is expected, in light of the discussion in the previous sections, that single-grain particles are predominantly mineral based, macroflocs are composed in majority of organic material and microflocs are a combination of mineral sediment and algae.

3.4.1 Variation with CC/SSC

No correlation was found between CC/SSC and the D_{50} estimated from the whole PSD's. We here study the dependence of the D_{50} of each size class. The D_{50} of each class D_{50} as function of CC/SSC is given in Figure 9. From the figure, it can be seen that there is a correlation between D_{50} and CC/SSC: D_{50} of microflocs is decreasing with CC/SSC whereas the D_{50} of macroflocs is increasing. The volume-% of microflocs/macroflocs also display a clear trend with CC/SSC: the higher the CC/SSC, the more macroflocs are present in the water column. It was furthermore

verified (not shown) that the relative volume-% of macroflocs is decreasing with SSC. This is in line with the laboratory tests performed in Deng et al. (2019), where it was shown that the CC/SSC ratio is driving the steady-state size of the flocs: at higher CC/SSC ratio, larger flocs are obtained, irrespective of the amount of sediment or flocculant in presence. Algae sediment flocculation reaches a steady-state within 30 min in jar test experiments (Deng et al., 2019), for a large range of SSC ($0.07\text{--}0.7\text{ g l}^{-1}$) and a high shear rate (90 s^{-1}). This would hint to the fact that micro and macroflocs are (on average) at proper conditions that large flocs can be formed in HWS of summer, due to the HWS periods last about 2 hours with low shear rate.

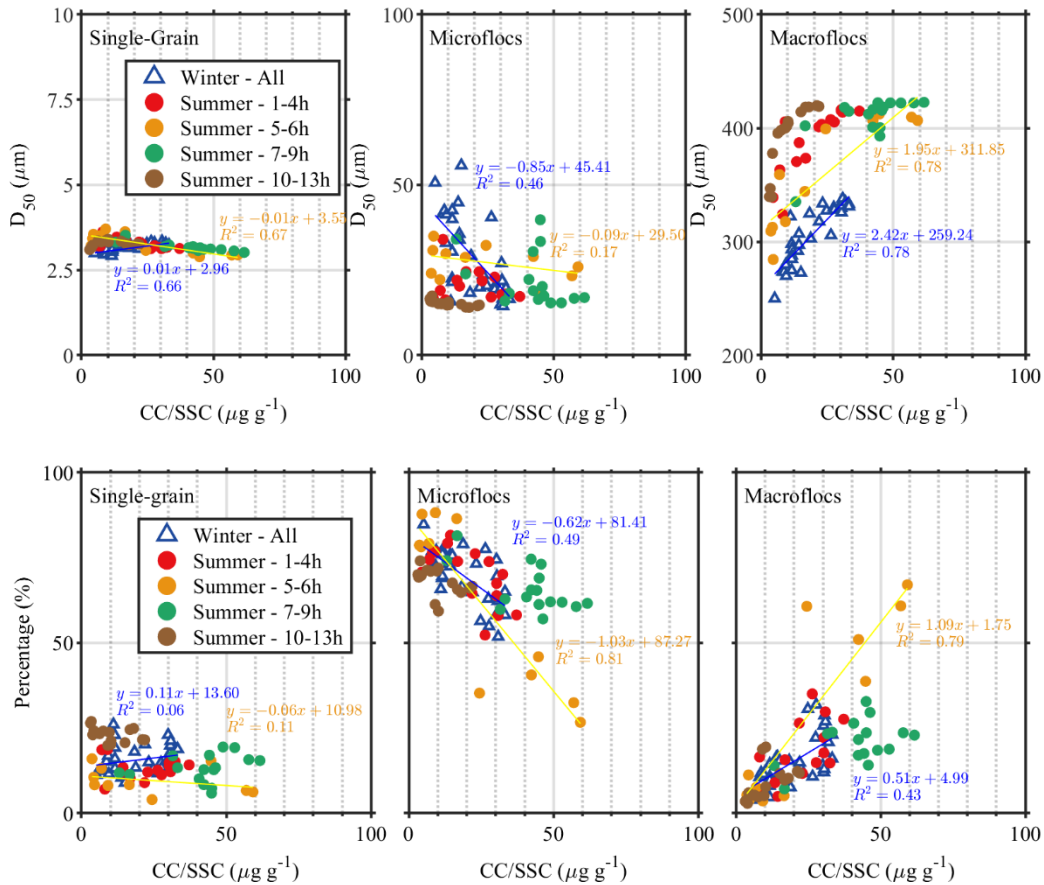


Figure 9 Relationship between the mean size D_{50} and volume % of each class with CC/SSC

3.4.2 Transfer between classes

In sediment transport modelling, like in numerical programs such as TELEMAC (Blumberg et al., 1996; Blumberg and Mellor, 1987), DELFT3D (Lesser et al., 2000, 2004) or ECOMSED (Sherwood et al., 2018), two or three classes of particles are usually defined that are associated with a concentration and a settling velocity. In the present article, we like to discuss the spatial and temporal variation of the concentrations of the classes in terms of the relative volume fraction of these classes that can be obtained from LISST data.

In section 3.4.1, it was shown that the CC/SSC ratio is correlated to the size of the micro and macroflocs, which, as we discussed is an indication that the system is at steady-state.

Despite the fact that the system is at steady-state (in terms of flocculation), the PSD is very dynamic and shifts between unimodal and bimodal depending on hydrodynamic conditions. This shift is related to the relative volume-% of micro and macroflocs. A bimodal peak appears when the two classes (microflocs and macroflocs) have a comparable relative volume fraction (volume-%). The evolution of the % volume of the three classes and CC/SSC are given in Figure 10 as function of time. The water column is divided in three: surface (0 H–0.2 H), middle (0.4 H–0.6 H) and bottom (0.8 H–1 H). One can see that there is a slight increasing trend in CC/SSC from 11.6 to 33.2 $\mu\text{g g}^{-1}$ in surface water while no significant trend is found in other water layers during the tidal period in winter. The CC/SSC ratio increases at flood tide in summer, at all depths (from 9 to 57.8 $\mu\text{g g}^{-1}$), where a relative increase of the volume-% of macroflocs compared to the volume-% microflocs can also be observed, which indicates that algae-rich particles are brought in the water column from the

sea. This is coherent with the observation made by Wu (2015) who observed that the dominant species to be found at the observation station is *Skeletonema* which is found predominantly in sea water.

It is well-known that algae can aggregate in large flocs at the top of the water column where they use sunlight to perform photosynthesis (Maggi and Tang, 2015; Takabayashi et al., 2006). Subsequently, the algae-dominated flocs populate the whole water column in the period HWS to ebb tide (see Figure 7 and Figure 11), slowly sinking to the bottom of the water column. As the biomineral flocs are forming and settling, the CC/SSC ratio, which is about $50 \mu\text{g g}^{-1}$ at depths > 7 m increases below 7 m from 0 to $50 \mu\text{g g}^{-1}$ over time. This is to be linked with the observations shown in Figure 2, where it was found that CC iso-lines are decreasing towards the bed over time in that time period. This implies that a significant amount of algae is reaching the seafloor, and that macroflocs are algae-dominated. It is known that algae flocs have a dynamic settling velocity, as algae can adapt their buoyancy by photosynthetically-produced oxygen, therefore the CC/SSC ratio remained high both in surface (0 H) and middle water (0.4 H), despite the settling of algae-based flocs (Fernández-Méndez et al., 2014).

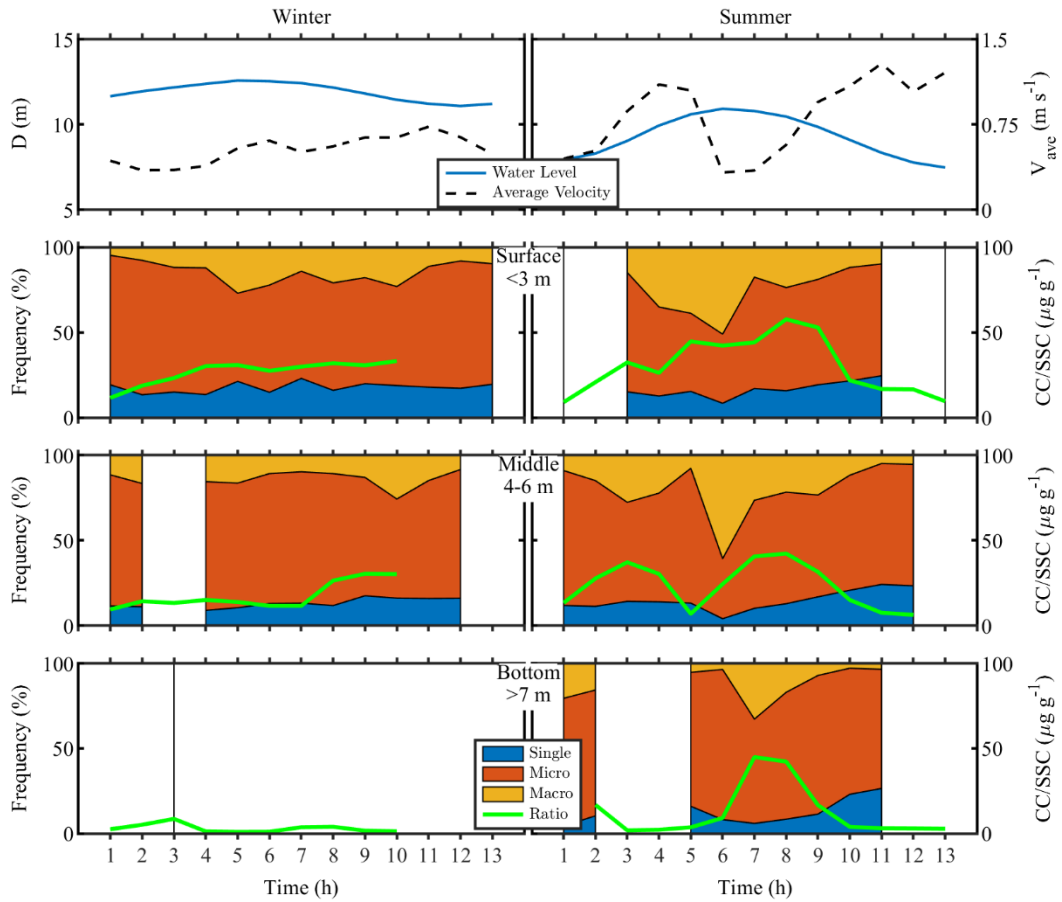


Figure 10 Time series of three floc fractions with CC/SSC ratio in winter and summer season in surface, middle and bottom water layers

The overall D_{50} is also changing over depth in time. At 6 h, during HWS, the largest D_{50} is shifting towards the middle depth, but its value is lower than at 5 h. At 7 h, the D_{50} is small everywhere, despite the fact that macroflocs are observed in the bottom layer: as the SSC is high in the bottom layer microflocs are the most abundant type of floc present, hereby reducing the overall D_{50} . As was observed in Figure 6, Figure 7 and Figure 8, the PSDs remain however bimodal, confirming the presence of large particles at any depth.

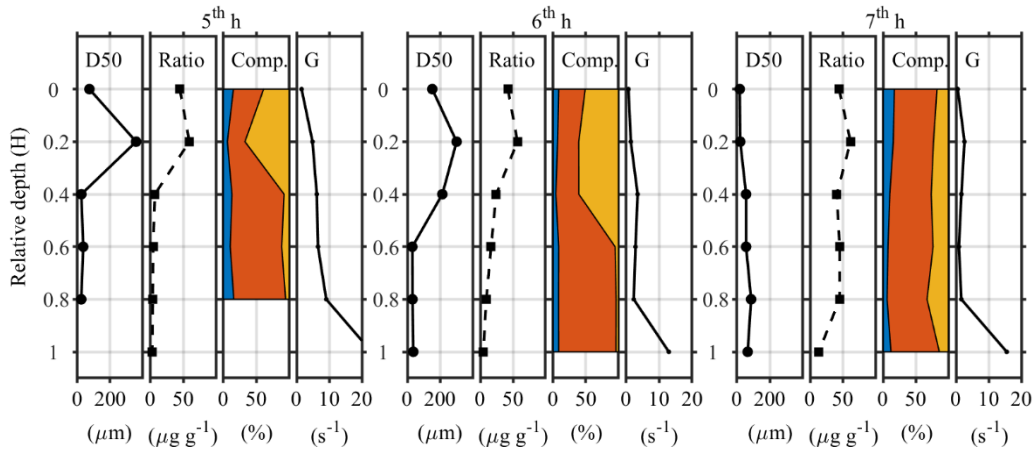


Figure 11 Vertical variations of D_{50} , CC/SSC ratio, floc size fractions and shear rate in summer during the period 5–7 hours.

It has been shown in Deng et al. (2019) that if the water column is well-mixed bimodal PSD's shift towards unimodal PSD's, as microflocs flocculate with macroflocs. This raises the question of residence in the water column, and the collision probability between micro and macroflocs. The variations in PSDs with depth in the period 5–7 hours (see Figure 11) was discussed above, where the clear difference between the PSDs found in the water column at [0 H–0.4 H] and [0.6 H–1 H] was attributed to the presence of a pycnocline: the residence time of algae flocs at specific depths are not only dependent on hydrodynamics but also on density—this is especially the case for algae-rich flocs which have smaller density than sediment dominated flocs. During HWS the algae flocs settle down, as do the mineral sediment particles and both have then no chance to meet as the slow settling algae flocs leaving the pycnocline do not catch-up with the faster settling sediment flocs that are found at lower depth. This can in part explain why multimodal PSDs are quite often observed in the system. Therefore, despite finding bimodal distributions, the flocculation ability between classes is very reduced and it can be concluded that the system is in good approximation at steady-state.

4. Conclusions

In this article, the seasonal variation of particle size distributions (PSD) were studied in relation with the presence of living organic matter (algae). The presence of algae lead to both unimodal and bimodal PSDs. When the PSD is bimodal two floc fractions (microflocs and macroflocs) can clearly be distinguished. These two classes of flocs have different properties that can be studied as function of relevant parameters (shear, CC/SSC ratio, seasonality). The existence of these two classes of particles makes it therefore problematic to reduce the study of the PSD to the study of its D_{50} and it was shown that no correlation could be found between the relevant parameters (CC/SSC in particular) and D_{50} .

A clear difference is found between the winter and the summer surveys. Both the algae production and the shear are low in winter, leading to the suspension of smaller flocs with lower density than in summer (the average floc size is $32 \pm 13 \mu\text{m}$ in winter and $45 \pm 64 \mu\text{m}$ in summer, the range density of flocs is $143\text{--}453 \text{ g l}^{-1}$ in winter and $159\text{--}989 \text{ g l}^{-1}$ in summer). The flocs are composed of small amount of clay and organic particles in winter. In winter, more fine mineral sediment (< 2 microns) are found at the top of the water column than in summer. In summer, the flocs are larger by two orders of magnitude and can contain both clay and silt particles, which makes them having a higher density than in winter, as in-situ monitoring techniques cannot properly assess the very fine (< 2 microns) clay fraction suspended in winter. Moreover, as the shear rates were higher in summer also, close to the bottom, larger silt and sand particles could be suspended compared to winter.

A good correlation is found between the presence of algae (through the measurement of chlorophyll-a concentration) and the presence of large, buoyant flocs at the top of the water column

in summer, where algae perform their photosynthesis. The algae appear predominantly during the flood tide period. Large flocs, i.e. macroflocs, (with high CC/SSC ratio) are then formed during the early flood period in the surface water. These flocs then settle to the middle water layer and finally reach the bottom water layer during the HWS period. At the same time, microflocs with a higher density than macroflocs are found deeper in the water column. During HWS both macroflocs and microflocs settle down but cannot flocculate as they never catch-up due to the difference in settling rates. This is the reason why, in line with the work of Soulsby et al. (2013), who studied Northern European estuaries, it can be concluded that the floc population can be treated as if at steady state in the Changjiang Estuary. This finding is of importance for the modelling of sediment transport in this estuary, as the relative concentration of the microflocs/macroflocs populations at a given depth is then solely governed by their settling rates and local hydrodynamic conditions (Manning et al., 2010; Manning et al., 2011; Spearman et al., 2011; Spencer et al., 2010).

The algae-sediment interaction in the estuarine system is shown in Figure 12. The key factor to study the changes in size, density and relative abundance of flocs are the CC/SSC ratio, which is related to the volume-% fraction of micro and macroflocs. The full PSD's are changing with tidal conditions and position in the water column (Figure 12), in line with our previous study (Deng et al., 2019). In addition, due to the sensitivity of algae to their living environment, the floc characteristics may vary from one location to another in the estuary. For example, in our previous study, where the observation position was closer to the channel, the biological effect was weaker, and combined with a strong estuarine tidal action, the distribution of floc sizes and settling velocities were comparable during flood tide and ebb tide. In the present study the observation location was closer to the open sea, the algae activity was relatively strong, and hence a strong tidal asymmetry

in floc size was observed, that is, flocs properties were clearly different at flood and ebb tide.

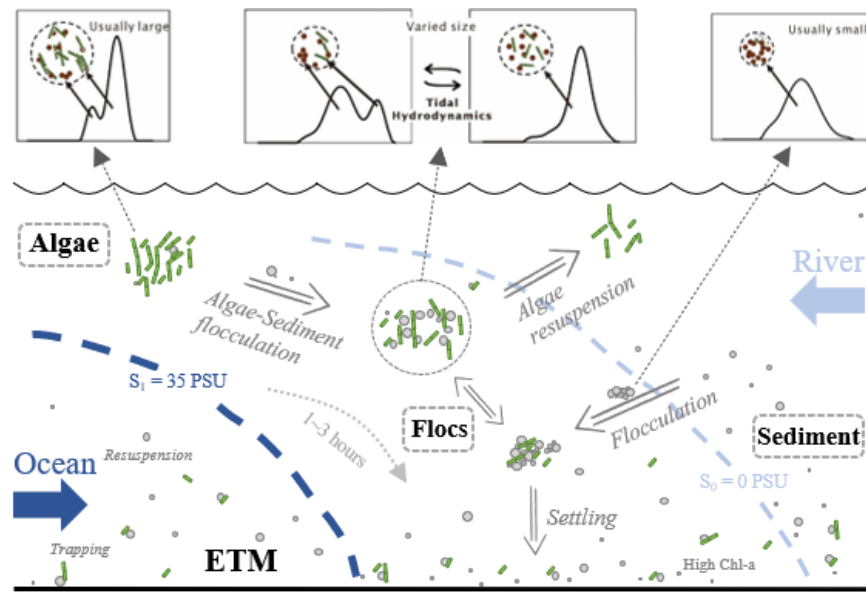


Figure 12 Algae-Sediment flocculation processes in estuary

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Data Availability

Part of datasets related to this article can be found at <http://dx.doi.org/10.17632/pdfxj2mkp3>, an

open-source online data repository hosted at Mendeley Data (Deng, Zhirui, “Seasonal variation of
floc population influenced by the presence of algae in the Changjiang Estuary: Datasets and
Supplementary Materials”, 2021, Mendeley Data). Due to the confidentiality agreement of relevant
projects, more data that support the findings of this study are available from the corresponding
author, [Q.], upon reasonable request.

Conflict of interest

The authors declared that they have no conflicts of interest to this work.

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