Single-grain, microfloc and macrofloc volume variations observed with a LISST-100 and a digital floc camera

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Abstract

Fine-grained (<63 μm) particles in the aquatic environment flocculate into larger, porous entities (flocs). Flocculation models have been proposed wherein the particles in the water exist either as single grains or as part of flocs. This approach, however, contradicts the idea put forward by Eisma [Eisma, D., 1986. Flocculation and de-flocculation of suspended matter in estuaries. Neth. J. Sea Res. 20, 183–199.] that a flocculated suspension would consist of large porous, fragile flocs, macroflocs (>~125 μm), made up of smaller, more sturdy flocs (microflocs, <~125 μm) and single grains. This paper combines results from a LISST-100 laser diffraction particle sizer and a digital floc camera in order to produce full-size spectra covering in situ particle sizes >2.5 μm, the smallest size resolvable by the LISST-100. For the first time, this allows a detailed investigation of changes in floc volume during break-up and flocculation. In accordance with Eisma [Eisma, D., 1986. Flocculation and de-flocculation of suspended matter in estuaries. Neth. J. Sea Res. 20, 183–199.], flocs are divided into macroflocs (here >133 μm) and microflocs (36–133 μm). In addition, a single-grain fraction (<36 μm) is introduced as particles of this size are of importance for the optical properties of the water column. The variation of these three fractions over a range of forcing conditions is examined using a particle volume size range from 2.5 to 9900 μm. When stress increases, the volume occupied by macroflocs decreases while the volume occupied by microflocs increases. It is also demonstrated that when stress decreases, the volume occupied by macroflocs first increases and then decreases as flocs settle out during quiescent conditions. However, in general no overall relationship between stress and floc size was found because parameters other than stress influence floc size, most notably resuspension, settling, advection, and biological activity. On average, macroflocs make up 40–65% of the total suspended volume, whereas single grains and microflocs each make up 15–34% of the total suspended volume. Because an inverse relationship between floc size and density generally exists, this means that occasionally the majority of the suspended mass can be found in single grains and microflocs. This has implications for the optical properties of the water column as scattering is dominated by the smallest particles in suspension. The findings also have implications for existing flocculation models that would probably benefit from inclusion of the microfloc fraction.

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Keywords: Flocculation; Aggregation; Turbulence; Single grains; Microflocs; Macroflocs; Italy; Adriatic Sea; Apennine margin; 42°28′N; 14°16′E; 44°48′N; 12°28′E

1. Introduction

In the aquatic environment fine-grained (<63 μm) inorganic sediment particles and organic matter such as
algal cells aggregate into larger, porous aggregates known as flocs (Eisma, 1986). The flocculation process influences the optical properties of the water column as the fine-grained particles become packed into flocs (Mikkelsen and Pejrup, 2000; Flory et al., 2004). The reason for this is that the surface area of the suspended particles controls the scattering of light from a suspension of particles (Bale et al., 1994). Packing of fine-grained particles as part of flocs in effect diminishes the total surface area, even though the mass concentration of total suspended matter (TSM) may remain constant (Hatcher et al., 2001; Mikkelsen, 2002; Flory et al., 2004). In addition, the sediment dynamics of a flocculated suspension differs from that of a single-grain suspension. When single grains become packed into flocs, their settling velocities increase by orders of magnitude (Van Leussen, 1986).

The in situ floc size can be orders of magnitude larger than the component grains making up the flocs (Milligan, 1996; Mikkelsen and Pejrup, 2000). This was probably first recognized by Krone (1962), who later suggested an ordered floc structure (Krone, 1963, 1978, 1986; Van Leussen, 1988). According to Krone, the smallest agglomeration of individual mineral particles ‘glued’ together in clusters, all with the same porosity, were called particle aggregates (pa). All particle aggregates glued together in larger clusters, all with the same porosity, were called particle aggregate aggregates (paa). Particle aggregate aggregates further agglomerated into particle aggregate aggregate aggregates (paaa) and so on. These orders of aggregation, in which all flocs of a given order ideally had the same porosity, were denoted pna with (n-1) indicating the order of aggregation. Based on field observations from Dutch estuaries, Eisma (1986) suggested a similar, but simpler classification of flocs in which single grains flocculated into relatively small and sturdy flocs of the order of ~100 μm called microflocs, which further flocculated into much larger, porous flocs called macroflocs during periods of relatively low stress. Despite the appeal of Eisma’s simple model, the proportion of particles in suspension bound in single grains, microflocs, and macroflocs at a given time, and the factors controlling it, remain largely unknown. While low levels of turbulence can promote flocculation, increasingly high levels will cause flocs to break-up (Van Leussen, 1988; Dyer, 1989). Whether the flocs break up into microflocs or down to their constituent single grains is unclear.

Hill et al. (2001) used a floc camera to investigate the influence of turbulence on floc size. They were limited in their observations by the fact that their camera could only resolve particles >250 μm. Thus, while Hill et al. (2001) were able to conclude that millimeter-sized flocs disappeared (i.e. became smaller than 250 μm) at stresses of approximately 0.1 Pa, they were not able to observe whether the flocs broke down to single grains or microflocs. Agrawal and Traykovski (2001) used a LISST-100 laser particle sizer (Agrawal and Pottsmith, 2000) to observe variations in floc size during the same deployment as Hill et al. (2001). Agrawal and Traykovski (2001) observed a decrease in the mean floc size detected by their LISST during the periods when Hill et al. (2001) were unable to detect flocs. They inferred that this was due to the break-up of flocs during high stress, but were hampered in their investigation by the lack of information on flocs >500 μm, the largest size that could be resolved by their LISST. The present paper further investigates floc formation and break-up by using a novel approach whereby the in situ particle (floc) size spectra from a LISST-100 type C (2.5–500 μm) and a digital floc camera (DFC; 135–9900 μm) are merged into full size spectra covering particle sizes from 2.5–9900 μm (Mikkelsen et al., 2005). For the first time, the present study makes it possible to investigate how floc volume changes in a given size class affect changes across virtually the entire size range of particulate material in the sea.

2. Study site and methods

The field work was carried out as part of the ONR-supported EuroSTRATAFORM project, which was concerned with sedimentary processes and the development of sedimentary strata proximal to the Po River and on the Apennine margin, Western Adriatic Sea. A newly developed particle tripod named INSSECT (IN situ Size and SEttling Column Tripod; Mikkelsen et al., 2004) was deployed on four occasions during a research cruise in May/June 2003. The study site was a 250-km-long stretch of the Apennine Margin, in the Adriatic Sea off the east coast of Italy. Measurements took place off the mouth of three rivers discharging into the Adriatic Sea: the Po, the Chienti and the Pescara (Fig. 1, Table 1). INSSECT is a rotating tripod equipped with an array of instruments for measuring particle characteristics (Mikkelsen et al., 2004). The purpose of the INSSECT deployments was to investigate the temporal and spatial variations in suspended particle characteristics such as size and settling velocity along the margin; in this paper focus is on the combined results from the Digital Floc Camera (DFC), the LISST-100 particle sizer (Agrawal and Pottsmith, 2000), and...
the Modular Acoustic Velocity Sensor (MAVS; Thwaites and Williams, 1996). The INSSECT was deployed in 10–12 m of water a few hundred metres off the coast, and the duration of each deployment varied between 1 and 3 d (Table 1).

2.1. DFC and LISST-100

The DFC is described in detail in Mikkelsen et al. (2004). It takes still pictures of the suspended particles by means of silhouette photography whereby the particles are lit from behind using a LED flash. The particles thus appear dark on a bright background. The field of view (FOV) is a $4 \times 4 \times 2.5$ cm slab of water, and the DFC is focused on the middle part of the slab. A small aperture setting and a bright light source ensure that all particles in the FOV are in focus. The LED flash strobes the DFC with a 20 $\mu$s pulse. This ensures that the particles in the image are ‘frozen’ and not blurred due to particle movement. The images are stored on an

Table 1

<table>
<thead>
<tr>
<th>Location</th>
<th>Date and time (UTC), 2003</th>
<th>Lat, Long</th>
<th>Depth (m)</th>
<th>DFC images</th>
<th>LISST samples</th>
</tr>
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<td>1240</td>
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<td>1315</td>
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<td>5 June 15:00–7 June 11:35</td>
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<td>1330</td>
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</tbody>
</table>

Fig. 1. Map of the study area in the Adriatic Sea with the three deployment sites indicated.
internal hard drive, capable of storing up to 1000 grey scale images with a size of 1024 × 1024 pixels in 256 grey scale values. The pixel size of the camera is 45 μm. In this study, nine coherent pixels were chosen as the minimum number of pixels to define a particle in an image, giving a minimum resolvable particle size of approximately 135 μm. During deployments the DFC was programmed to take a picture every 5 or 10 min depending on the length of the deployment. Upon recovery the images from the DFC were off-loaded to a PC and analysed using a set of MATLAB image processing scripts yielding the volume distribution in logarithmically spaced size bins (Table 2).

The LISST-100 was programmed to measure at 4 Hz in a burst mode where every 10 measurements were averaged into one sample. Each sample was thus obtained at 0.4 Hz. For each burst 10 samples were saved with an interval between samples of 5 s. The burst length was thus 50 s. The interval between individual bursts was 5 min. Upon recovery the data were offloaded and analysed according to standard LISST data analysis procedures (cf. Agrawal and Pottsmith, 2000; Mikkelsen and Pejrup, 2000), yielding the volume distribution in logarithmically spaced size bins from 2.5–500 μm (Table 2).

### Table 2
Lower limit, mid point and upper limit in microns for size bins 1–32 (bold, covered by the LISST-100) and size bins 25–50 (italic, covered by the DFC)

<table>
<thead>
<tr>
<th>Size bin #</th>
<th>Lower limit (μm)</th>
<th>Mid point (μm)</th>
<th>Upper limit (μm)</th>
<th>Size bin #</th>
<th>Lower limit (μm)</th>
<th>Mid point (μm)</th>
<th>Upper limit (μm)</th>
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<td>3.78</td>
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<td>5.27</td>
<td>5.72</td>
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<td>63.12</td>
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<td>87.89</td>
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<td>24</td>
<td>112.67</td>
<td>122.39</td>
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<td>49</td>
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<td>7681.4</td>
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<tr>
<td>25</td>
<td>132.96</td>
<td>144.43</td>
<td>156.9</td>
<td>50</td>
<td>8344.3</td>
<td>9064.5</td>
<td>9846.9</td>
</tr>
</tbody>
</table>

The two instruments overlap in eight size bins; 25–32.

### 2.2. Full size spectra

All LISST-100 volume distributions from each burst were averaged into one distribution, which was then merged with its corresponding DFC particle volume distribution according to the procedure described by Mikkelsen et al. (2005). This procedure takes advantage of the fact that the LISST-100 and the DFC overlaps in eight size classes; 133–500 μm (cf. Table 2). The shape of the size spectra from the two instruments in these overlapping bins is compared and usually they are found to be offset but with the same shape (Mikkelsen et al., 2005). Based on the offset in the eight overlapping size classes, a conversion factor is found that will collapse the overlapping part of the LISST-100 spectrum onto the overlapping part of the DFC spectrum. The entire LISST-100 size spectrum is then multiplied by this conversion factor, and the two spectra can simply be overlaid. This produces an in situ size spectrum covering particles from 2.5–9900 μm, with the upper size limit being defined by the camera design. Mikkelsen et al. (2005) demonstrated that this is a robust method of obtaining full in situ particle size spectra, even when ‘rising tails’ (Mikkelsen et al., 2005) exist in the LISST-100 spectra. From the full in situ volume distribution,
the particle diameters \( D_{50} \) and \( D_{25} \) were then computed where \( D_{50} \) is the median particle diameter, and \( D_{25} \) is the upper quartile diameter for the full volume distribution (see Hill et al., 2000, for details).

For each full size spectrum, the volume concentrations in the size bins were also summed into larger size classes and divided by the total volume concentration. This yielded the relative contribution of particle volume in six classes: size bins 1–16, 17–20, 21–24, 25–28, 29–32, and 32–50 (cf. Table 2). The purpose of this division was to investigate how the various volume fractions varied as the floc size varied, and it was found that division into classes composed of less than four bins made any graphical interpretation and presentation of results impractical. Finally, an even coarser division, composed of the three size fractions 2.5–36 \( \mu \text{m} \), 36–133 \( \mu \text{m} \) and > 133 \( \mu \text{m} \), was performed. The two largest size fractions roughly correspond to the microfloc and macrofloc groupings introduced by Eisma (1986), whereas the finest size fraction is considered to denote single grains.

### 2.3. MAVS

The MAVS computes current velocity and direction by measuring the differential travel-time of sound along four paths defined by the geometrical arrangement of four transducer pairs (Thwaites and Williams, 1996). It was programmed to sample at 4 Hz in a burst mode, with a burst length of 12.5 s (50 measurements) every 5 min. Ideally, in order to analyse the influence of turbulence on floc break-up in a quantitative manner, the Kolmogorov microscale of turbulence should be computed from the velocity measurements (Soulsby, 1983). However, due to the relatively low sampling frequency and burst period length of 12.5 s, this was deemed inappropriate. This paper does not attempt to quantify the stress levels at which turbulence causes floc break-up and re-floculation, but aims to investigate how floc volume is redistributed during break-up. Therefore the absolute stress level is not as important as the general trend of the temporal variation in stress and its variability within and between deployments. The squared current velocity (\( U^2 \)) was used as a proxy for current stress (Soulsby, 1983). This strategy does not yield information on near-bed stress due to waves. An estimate of the distance the water was advected during each deployment was obtained by vector addition of the average current velocities for each burst multiplied by the time interval between bursts.

### 3. Results

When averaged over entire deployments, \( U^2 \), \( D_{50} \), \( D_{25} \), volume concentration (VC) and single-grain:macrofloc proportions show little correlation (Table 3). \( U^2 \) was 3–8 times higher during the Chienti deployment because of the occurrence of a brief (12 h) wind event. Despite the higher current speeds, floc sizes were similar to floc sizes during the Po deployment and the Pescara II deployment. Floc sizes were markedly smaller for the Pescara I deployment, even though its averaged \( U^2 \) was second highest. Averaged VC for these deployments differed as well, with the Po value several times higher than the Chienti value, which was in turn several times larger than the Pescara II values. The Pescara I value was similar to the Chienti value. Size distributions showed no obvious relationships to other parameters.

These observations demonstrate the complexity of linking mean particle parameters to mean environmental conditions. The time histories of resuspension, aggregation, disaggregation, advection and settling all affect the particle size distribution, yet these processes are often not adequately represented by simple averages of variables such as the square of the mean current. To explore in more detail how current stress and in situ particle size distribution are linked, time series were plotted for each deployment (Figs. 2–5). Figs. 2–5 show 2-h moving averages of the temporal variation

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Chienti</th>
<th>Po</th>
<th>Pescara I</th>
<th>Pescara II</th>
</tr>
</thead>
<tbody>
<tr>
<td>( U^2 ) (( \times 10^{-3} ) m(^2) s(^{-1}))</td>
<td>9.2 ± 7.3</td>
<td>2.0 ± 2.4</td>
<td>3.5 ± 2.1</td>
<td>1.2 ± 1.3</td>
</tr>
<tr>
<td>( D_{50} ) (( \mu \text{m} ))</td>
<td>280 ± 177 (62–1645)</td>
<td>291 ± 487 (17–2809)</td>
<td>112 ± 144 (38–1891)</td>
<td>277 ± 265 (8–1630)</td>
</tr>
<tr>
<td>( D_{25} ) (( \mu \text{m} ))</td>
<td>499 ± 251 (212–1759)</td>
<td>505 ± 565 (61–2946)</td>
<td>301 ± 242 (111–2049)</td>
<td>490 ± 309 (29–1750)</td>
</tr>
<tr>
<td>VC (( \text{mm}^3 \text{L}^{-1} ))</td>
<td>95 ± 69 (22–438)</td>
<td>259 ± 517 (4–4870)</td>
<td>90 ± 53 (20–425)</td>
<td>39 ± 49 (3–337)</td>
</tr>
<tr>
<td>Single grain fraction (%)</td>
<td>20 ± 7 (7–43)</td>
<td>34 ± 16 (0–65)</td>
<td>34 ± 7 (6–49)</td>
<td>23 ± 17 (0–78)</td>
</tr>
<tr>
<td>Microfloc fraction (%)</td>
<td>15 ± 7 (3–36)</td>
<td>20 ± 10 (0–43)</td>
<td>26 ± 5 (7–39)</td>
<td>18 ± 8 (0–39)</td>
</tr>
<tr>
<td>Macrofloc fraction (%)</td>
<td>65 ± 10 (34–90)</td>
<td>46 ± 23 (10–100)</td>
<td>40 ± 11 (20–88)</td>
<td>58 ± 20 (7–100)</td>
</tr>
</tbody>
</table>

Values in parentheses show the range.
in $U^2$, water depth and net advection (A), D$_{50}$ and D$_{25}$ (B), volume ratios for the 6 combined size bins (C), absolute VC for the 6 combined size bins (D), and the single grain: microfloc: macrofloc ratio (E) for the four deployments. The lowercase letters on top of panel A in all figures represent interpretation of the temporal variation in floc size and volume, denoted by the vertical lines.

Fig. 2. Temporal variation during the deployment off the Chienti River of (A) $U^2$, water depth and net advection; (B) floc size (expressed as D$_{50}$ and D$_{25}$); (C) volume ratio for 6 combined size bins; (D) absolute volume concentration for the size ranges in C; (E) volume percent of single grains, microflocs, and macroflocs. All data are presented as 2-h moving averages.
3.1. Chienti

Initially, stress decreased and reached a local minimum (Fig. 2A; a) at which time it increased and reached a local maximum (c) followed by a period of generally elevated, but varying stress with two local minima (d,e) and an overall maximum for the deployment (f). This was followed by a rapid decline (f–g), and levelling out (g-h). Towards the end of the deployment (i), stress increased slight-
Net advection was almost 11000 m during the deployment (46 h).

Floc size, represented by D_{50} and D_{25}, initially increased as stress decreased (Fig. 2B; a), decreased as stress increased (a–c), and stayed low when stress was high and fluctuating (c–f). When stress started to decrease after reaching its maximum value (f), floc size increased rapidly. Towards the end of the deployment floc size reached a second minimum, when stress was at its global minimum (f). Note that floc sizes reached minima at the second highest stress level (c) as well as at the lowest stress (g).
The volume ratio of flocs in the 4 smallest size classes was almost constant and equal for the 4 size classes throughout the deployment (Fig. 2C). Also the volume ratio of particles in the 258–500 \( \mu \)m size range was almost constant throughout the deployment. During the period of elevated currents (b–f), the volume ratio of particles in the 36–258 \( \mu \)m size classes increased while it decreased for particles >500 \( \mu \)m. After the storm the volume ratio in all size classes returned to the pre-storm levels.

Fig. 5. As Fig. 2, but for the second deployment off the Pescara River.
With respect to absolute volume concentrations (VC), it is seen from Fig. 2D that during the onset of the storm (b–d) the concentration of particles in the 36–500 μm size classes increased. The VC in these size classes then started a general decline (d–f), while the VC for particles >500 μm increased. As for the volume ratios, the VC in all size classes after the storm returned to pre-storm levels.

The single-grain pool did not vary much during the deployment (Fig. 2E). The volume of microflocs started to increase, at the expense of macroflocs, at b, during increasing stress. As stress reached its second highest level (c), macroflocs began to form, and the volume ratios of microflocs as well as single grains decreased. Towards the end of the deployment the volume of macroflocs decreased (f).

3.2. Po

Stresses off the Po River (Fig. 3A) were a factor of 3–5 lower than off the Chienti River. Net advection during the deployment (22 h) was 935 m. Zero net advection occurred ~6 h into the deployment (c). Floc sizes were initially large, around 750–1100 μm, but decreased rapidly to 100–250 μm with some variation (Fig. 3B; b–h). This is mirrored in the large standard deviation of the average sizes with a D50 of 291 ± 487 μm and a D25 of 505 ± 565 μm (Table 3). Examination of the DFC images showed that the large floc sizes at the beginning of the deployment were caused by the frequent occurrence of one or two very large flocs, 2–5 mm in diameter, in the field-of-view. This is demonstrated by the variation in the volume ratio, where flocs >500 μm make up the majority of the particle volume at the beginning of the deployment (Fig. 3C). From b, however, all ratios generally varied around 0.1.

Particles >500 μm also initially dominated the total VC (Fig. 3D; b). Upon the disappearance of these large flocs from the DFC imagery the total volume did not show much variation, except for an event (d) where VC increased by almost an order of magnitude. The event manifests itself as a doubling of floc sizes and an almost 30% increase in the macrofloc ratio (Fig. 3B, E; d).

Last, considering the variation in only single grains, micro and macroflocs (Fig. 3E) it is seen that the macroflocs make up ~70% of the volume at the beginning of the deployment (a–b). After the disappearance of the macroflocs the most dominant fraction is the single grains, which makes up 40–50% of the total volume for the remainder of the deployment, while microflocs and macroflocs each make up ~30%. The macrofloc volume increased briefly during the period when there was a sudden increase in VC (Fig. 3C, D, E; d).

3.3. Pescara I

Stresses during the first Pescara deployment (Fig. 4A) were comparable to stresses at the Po site, and generally decreased over the sample period. Net advection during the deployment (22 h) was ~4000 m. The flocs were generally the smallest observed for all surveys with D50 varying around 110 μm (Table 3, Fig. 4B). The volume ratios generally varied around 0.1 for particles >36 μm and around 0.35 for particles <36 μm (Fig. 4C). The total VC variation (Fig. 4D) shows an event where almost all particle classes increased in roughly the same proportions. As the VC fell back to its ‘background’ level (c–d), the ratio of macroflocs increased slightly (Fig. 4C, E). The overall trend during the deployment was a slight decrease in the volume of single grains and a slight increase in the volume of macroflocs (Fig. 4E).

3.4. Pescara II

Stresses were the lowest observed for all surveys (Table 3), with net advection ~1500 m (Fig. 5A). Zero net advection occurred a few hours into the deployment (b). Floc sizes were comparable to those observed off the Chienti River, but varied throughout the deployment with no clear trend (Table 3, Fig. 5B). Initially the suspension was made up mainly of single grains and macroflocs (Fig. 5E; a–c). Subsequently the fraction >69 μm started to increase (Fig. 5C; c). This caused the volume of the microflocs to increase to approximately 20% (Fig. 5E; b–c). The volume of single grains started to decrease as the volume of microflocs and especially macroflocs continued to increase (d). The macrofloc volume towards the end of the deployment was the same as at the beginning, whereas the single-grain volume had decreased and macrofloc volume increased. Throughout the deployment there was no obvious relationship between the variation in stress and variation in the various size range fractions (Fig. 5C, D).

4. Discussion

4.1. Variation at individual deployment sites

During the Chienti deployment floc sizes varied by a factor of 2–4 with a minimum size during the storm and a corresponding increase in VC. Such a decrease in floc
size and increase in VC could have been brought about in different ways: (1) resuspension of sand would lower the in situ size and increase VC; (2) floc break-up would lower in situ size but not increase VC; and (3) floc break-up together with resuspension of either sand or mud, or a combination of mud and sand would lower in situ size and increase VC.

It is possible to infer the size distribution of the bed sediment under the assumption that the changes brought about in the size spectra and VC were solely related to resuspension of sand from the bed. Fig. 6 shows in situ size spectra from two periods with low and high stress (Fig. 2A; a,c). If the change in in situ size was solely due to resuspension of sand, then the sand size distribution could be found by subtracting the two spectra (Fig. 6). In this particular case, the sand spectrum would have a median diameter of 265 μm with lower and upper quartile diameters of 142 and 422 μm. This estimate of the bed sediment texture can then be compared with the actual texture of the bed sediment. It has been found that for water depths around 8–12 m, the bed sediment along the Apennine margin is mixed with a mean size of approximately 100 μm (Passega et al., 1967; George et al., subm. ms.). Thus it is unlikely that the observed changes in the in situ size spectra are due to resuspension of sand from the bed, since this should have caused the in situ grain size to decrease even more than was observed.

The temporal variation in VC also supports the conclusion that the changes in in situ size were related to floc break-up and reformation. From the variation in VC it is seen that the 133–258 and 258–500 μm size classes peaked at the same time and at the same concentration ~70 mm$^3$ L$^{-1}$ (Fig. 2C, d). Immediately following the peak, however, the VC of the 133–258 μm particles dropped below the VC of the 258–500 μm particles. Since fine-grained sand particles cannot settle out prior to coarser-grained sand particles, it follows that the particles in the 133–500 μm size range must have been flocculated to some degree; hence the most likely cause of change in volume in the two size classes was ongoing flocculation.

From Fig. 2D it can be seen that particles in the 69–133 μm range initially followed the increase in particles in the 133–500 μm range (b–c). However, as stress peaked the finer fraction separated (c) and increased with a VC that was generally 10 mm$^3$ L$^{-1}$ lower than the VC of the 133–258 and 258–500 μm ranges (c–d). The 69–133 μm range peaked simultaneously with the 133–258 and 258–500 μm classes (d), but then started to decrease faster than these two size classes. Halfway between e and f, at the second peak in VC, the difference between the size ranges was almost 40 mm$^3$ L$^{-1}$ for the 69–133 and 258–500 μm sizes, whereas it was only ~20 mm$^3$ L$^{-1}$ at the first peak in VC (at d). Again, such a difference in the decrease of absolute volume cannot be brought about by single-grain (sand) settling, but must be influenced by flocculation and floc break-up. Furthermore, it can be seen that the VC of the 258–500 μm size class increased while the 69–133 μm ranges were constant (between e and f). If the increase in the 258–500 μm size class had been due to increased resuspension of sand, then the 69–133 μm size class should also have increased due to resuspension, as the bed sediment is mixed (Passega et al., 1967; George et al., subm. ms.). However, since the 69–133 μm size class was constant it follows that the second increase and peak of the 258–500 μm size class must have been due to flocculation. Thus, it seems likely that the majority of the changes in the 69–500 μm sizes were due to flocculation dynamics. It was shown above that if the changes were due to resuspended sand, this sand would have a median diameter of 265 μm, and a D$_{50}$ of 422 μm.

In review, Fig. 2E can then be interpreted as follows: initially floc sizes increased slightly, due to an increase in macrofloc volume from ~65% to ~75% of total volume by scavenging on microflocs (as the volume of single grains remained constant). The volume of single grains and microflocs then started to increase due to macrofloc break-up (a–b). At b the single-grain volume reached a local maximum, and then started to decrease while the microfloc volume continued to grow, by ‘feeding’ on single grains and broken macroflocs. At the second-highest level of stress (c) the

![Fig. 6. Full in situ particle size spectra from the deployment of the Chienti River. Spectra denoted a and c are from periods with low and high stress, respectively (Fig. 2A; a,c). The spectrum denoted c-a is the difference of the two spectra. This spectrum indicates the size distribution of the bottom sediment under the assumption that the change in the shape of the size spectra between a and c was due to resuspension of bottom sediment.]
macrofloc volume reached a minimum volume (47%), while microflocs reached a maximum (30%). The volume of microflocs stayed at this level until d and then started to decrease as the increase in macrofloc volume was faster than the decrease in single-grain volume. The volume of the macroflocs continued to increase even though stress was high and variable (c–f), and reached a local maximum of 75% between f and g. As stress was decreasing (f–g), the ratio of the three fractions again approached the ratio prior to the storm. Towards the end of the deployment the volume of single grains and microflocs increased, but at a time when stress was at a minimum (h). During this time the absolute VC of the largest size classes was decreasing (Fig. 2D), so the increase in single grains and microflocs is most likely due to the settling out of large flocs at low stress. Between h and i stress increased, and the increase was accompanied by an increase in macrofloc volume. This could be due to the beneficial influence of turbulence at low stresses, stimulating single-grain and microfloc aggregation (Van Leussen, 1988) or it could record the resuspension of intact macroflocs from the bed.

Stresses were much lower during the other three deployments. Additionally, the sea was calm and wave-induced resuspension can therefore be ruled out as a cause of change in floc size, volume concentration and volume ratios during these deployments. The main causes of change in floc size during these deployments would thus be settling or advection. Changes in the floc size or volume ratios that occurred at two consecutive zero net advection times must primarily be related to settling, since the water was then advected back to its starting position. On two occasions zero net advection was observed: during the Po and the second Pescara deployment (Fig. 3A; a,c, Fig. 5A; a,b). During the deployment off the Po zero net advection occurred approximately 6 h into the deployment (Fig. 3A; c). During this period floc sizes decreased and the volume ratio of single grains and microflocs increased (Fig. 3B, E; a–c). The decrease in floc size and the change in volume ratios were mainly related to settling and not the advection of water containing smaller flocs and a lower volume of macroflocs. The same pattern was observed between the periods of net zero advection during the second Pescara deployment (Fig. 5E, a,b), though not as pronounced as during the Po deployment.

Off the Po River a sudden increase in VC was observed mid-way through the deployment (Fig. 3D; d). This increase, which was also observed with an OBS and in the beam attenuation record from the LISST-100, is unlikely to have been caused by resus-
single grains the temporal variation of the two fractions would be unchanged. Obviously, the absolute volume \% of the two fractions would change.

Considering all four deployments together, it is interesting to note the widespread presence (in volume) of single grains. To our knowledge this has not been observed previously, due to the lack of instrumentation capable of covering the full size spectrum of particles in the aquatic environment. Since the density of flocculated particles generally decreases with increasing size (Fennessy et al., 1994; Sternberg et al., 1999; Mikkelsen and Pejrup, 2001), the mass ratios will be even more skewed towards the single grains and microflocs and away from the macroflocs. This division of mass is generally consistent with what has been reported from other studies that use floc fraction to describe flocculated sediment dynamics (e.g., Curran et al., 2002a, 2004; Fox et al., 2004a). Floc fraction is the fraction of mass in suspension that is incorporated into flocs. Fox et al. (2004a) investigated floc fractions off the Po River and found floc fractions close to unity at the river mouth for flocs > 135 μm. At the 8-m isobath, the floc fraction had decreased to ~0.08. Fox et al. (2004a,b) concluded that the suspended sediment was highly flocculated in the river, and settled out at water depths shallower than 8 m. This removal of highly flocculated sediment left behind a population of suspended sediment further offshore that was only slightly flocculated, with a floc fraction of ~0.08 (Fox et al., 2004a). In our study, the minimum volume ratio of macroflocs off the Po River was found to be around 0.25 when the 2-h moving average was applied to the data (Fig. 3E), but as low as 0.10 in the individual measurements (Table 3). This figure is reconcilable with the floc fraction estimates of Fox et al. (2004a,b), and thus supports the idea of a ‘stranded’ population of microflocs and single grains in the surface plume off the Po River, slowly sinking into the bottom boundary layer where they were detected by the instrumentation on the INSECT.

The fact that the single grains constitute up to 78% of the suspended volume has implications for the optical properties of the water column, as light scattering and light attenuation responds strongly to small particles on a mass basis (Bale et al., 1994; Mikkelsen, 2002). The Po River plume is incorporated into the Western Adriatic Coastal Current (WACC) upon discharging into the Adriatic Sea. The WACC is highly visible on satellite imagery and easily observable from ships as a front. However, from CTD casts and water samples obtained simultaneously with the INSECT deployments, the suspended sediment concentration in the visible part of the WACC was only found to be a few mg L⁻¹. Similar findings were reported by Mertes and Warrick (2001), working in river plumes off the coast of California. They found that SSC determined from SeaWiFS imagery of the river plumes could only account for up to 2% of the sediment discharge, estimated from rating curves. The observations presented here suggest that high optical visibility despite low sediment mass concentration can be explained by the presence of a high volume ratio of the single grains and microflocs and a low volume ratio of the macrofloc fractions.

The findings presented here also shed light on other sedimentological issues. It was demonstrated by Van Leusen (1994) and Fennessy et al. (1997) that macroflocs were the dominant carriers of mass in estuarine environments and that they were responsible for the mass flux of sediment to the bed in tidal channels and on intertidal flats. The results presented here, when viewed in conjunction with the findings of Fox et al. (2004a,b), suggest a somewhat different picture for the sediment dynamics of coastal currents, at least during low-energy conditions. The flocculated sediment that settles out close to the discharge point (e.g. a river mouth, cf. Fox et al., 2004b) is composed primarily of macroflocs, leaving mostly microflocs and single grains behind. Provided no sediment is added to the plume the interaction of the microfloc and single-grain fractions then determines the particle dynamics of the suspended matter, with macroflocs being of little importance. If the aggregation timescale to reach macrofloc size is longer than the timescale for settling of the microfloc and/or single-grain fraction, then macroflocs are unlikely to dominate the particle dynamics of the plume/current after the initial settling out. Thus, future studies concerned with the sediment dynamics of coastal currents and plumes could benefit by focusing some attention on the dynamics of the microflocs and single-grain fractions, after the initial settling out of sediment. An exception to the importance of these two smallest fractions would be if sediment was added by resuspension to the plume/current in the down current direction (cf. Curran et al., 2002b).

In terms of the flocculation dynamics of particulate suspensions, the results call for a re-evaluation of existing flocculation models such as the one presented by Curran et al. (2002a). In this model the suspension is assumed to be composed of a macrofloc fraction and a single-grain fraction. The single-grain fraction of the model increases due to macrofloc break-up. While it has been observed here that such a suspension is occasionally possible (cf. Figs. 3E, 5E), results from the Chienti deployment (Fig. 2E) suggest that macroflocs break up into microflocs, and that it is the interaction...
between the macroflocs and microflocs that determines the flocculation dynamics during high-energy events.

During the Chienti deployment the single-grain fraction did not change much, either in relative or absolute terms (Fig. 2C, D, E). However, the second Pescara deployment (Fig. 5) showed a different pattern. Both the volume ratio and the absolute VC of the single grains decreased during the deployment. The microflocs increased their ratio of the total volume during the deployment while the volume ratio of the macroflocs remained constant; this suggests that the single grains flocculated into microflocs during this low-energy deployment. It could be hypothesised that during energetic events there is a dynamic equilibrium between the single grains and the microflocs, so that microflocs are constantly breaking up, and single grains are constantly flocculating into microflocs (Fig. 2E). At the same time, when stresses are high, there will be a flux of volume and mass from the macrofloc fraction into the microfloc fraction, thereby increasing the microfloc volume. When stresses relax, this flux is reversed and macroflocs reform. During low-energy periods the single grains flocculate into microflocs and, provided no single grains are added to the suspension, this will cause a decrease in the single-grain fraction (Fig. 5E; d–h). The microflocs then flocculate into macroflocs, which settle out due to the increase in settling speed with increased size (Hill et al., 1998; Mikkelsen and Pejrup, 2001).

The proposal that suspended sediment should be treated as a three-phase system is in agreement with results from deep-sea sediment trap flux studies using $^{234}$Th. In order to reconcile observations of $^{234}$Th profiles and sediment fluxes with models of vertical flux, several authors have found it necessary to include three size fractions of suspended matter in the models (Tsunogai and Minagawa, 1978; Murnane et al., 1990). Typically, these models have had a dissolved, a single-grain, and a floc component, with the floc component being the only component assigned a settling velocity. Using such a model, Murnane et al. (1990) obtained particle sinking rates that were an order of magnitude smaller than rates obtained from sediment traps. Murnane et al. (1990) concluded that inclusion of two classes of sinking particles in the model probably could reduce the difference between the observed and modelled particle fluxes. Candidates for these two classes could be microflocs and macroflocs.

The results presented here raise several questions and suggest a number of directions for future work in studies on fine sediment dynamics: First, the data presented here only cover the coastal current along the Apennine margin. Little is known about the single-grain: microfloc: macrofloc ratio in rivers, estuarine environments, or the deep sea, all areas where flocculation and fine-grained sediment dynamics are of great importance. Second, the composition of the particles observed in this study is not well known. Estimates of in situ density using video analysis suggest that floc densities along the Apennine margin vary from 8 to 28 kg m$^{-3}$ (Mikkelsen et al., in press). This indicates that the flocs have a high content of organic matter. It is well established that the flocculation efficiency is dependent on biologically related factors, such as the ‘stickiness’ of the particles (Van der Lee, 2000). Under identical forcing conditions flocs with a high organic content may be hypothesised to flocculate and/or deflocculate differently from flocs with a high inorganic content; thus the rate of change in single grain: microfloc: macrofloc proportions may differ between different systems and environments. Third, with the three-phase system presented here, the question remains if the flocculation efficiency for single grains and microflocs and break-up coefficients for microflocs and macroflocs (Van Leusen, 1988) is the same. Fourth, the variation of the single-grain fraction documented here has implications for the optical properties of the water column, as it is the smallest particles that dominate the scattering of light in water. Future studies concerned with the optical properties of turbid coastal waters would likely benefit from the inclusion of modelling and/or measurement of the mass and surface area transfer between the single-grain, microfloc and macrofloc fractions due to flocculation and floc break-up (Flory et al., 2004). For example, it could be hypothesised that during spring tides the macrofloc ratio would be smaller than during neap tides, due to floc break-up during the more energetic spring tide conditions. In turn, this could affect the optical properties of the water column in a periodic manner. Finally, the results presented here were obtained with all instrumentation mounted on a tripod, thus within an Eulerian framework. Therefore the results are influenced by advection as well as by settling of suspended sediment. Measurements of the variation of the single-grain: microfloc: macrofloc ratios in a Lagrangian framework would benefit our understanding and interpretation of the results, as the influence of advection would be minimised.

5. Conclusions

In situ particle (floc) size is affected by the time history of resuspension, aggregation, disaggregation, settling and advection. Therefore relationships between
mean parameters of the particle size distribution and other environmental parameters often do not exist. Full size spectra obtained by combining data from a LISST-100 and a digital floc camera on the Apennine margin off Italy were used to investigate how volume fractions of suspended matter varied. Floc volume was divided into single grains (<36 μm), microflocs (36–133 μm) and macroflocs (>133 μm). During energetic events (storms) the total volume concentration increased due to resuspension, and macroflocs were observed to break up, decreasing the macrofloc volume. Simultaneously, the microfloc volume increased while the single-grain volume was relatively constant, suggesting a flux of volume from the macroflocs to the microflocs and a dynamic equilibrium between the microfloc and the single-grain fractions. Increases in volume concentrations believed to be due to trawler resuspension were also observed. This caused a different response in floc volume variation compared to the storm event as floc size and macrofloc volume increased simultaneously. This suggests that the method of resuspension influences floc size differently: Mechanical shocks (such as trawling) ‘scoop’ sediment from the bottom and to a large extent do not break down the flocs, whereas high levels of sustained turbulence erode the bed sediment and cause the flocs to break down into microflocs. Single grains and microflocs accounted for up to 60% of the suspended volume. Due to the inverse relationship between floc size and density this means that in terms of mass this ratio would be even higher. This is in accordance with previous estimates of floc fractions of the order of ~0.1 in this area. The relatively high proportion of single grains also explains why the coastal waters along the Apennine margin are highly visible on satellite imagery, while the SSC is only a few mg L⁻¹. Present flocculation and sedimentation models may benefit from inclusion of the microfloc and single-grain fraction in their description of the suspended sediment.

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