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Review

Mini-review: Assessment of size distribution of suspended particles in coastal and estuarine environments using *in situ* instruments

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Abstract—Size distribution of suspended particles is crucial to understand their dynamics in coastal and estuary environments, since size of particles is one of the key factors controlling behavior of particle in water column, including coagulation/disaggregation processes and sinking rates from upper waters. Although the size distribution has been measured widely by electric resistance method using a coulter counter after sample water collection, fragile nature of large sized suspended particles, such as aggregates or flocs, hindered evaluation of *in situ* particle size distribution. Application of optical technique to measure *in situ* distribution of marine particles (2–500 μm), therefore, has been developed, i.e., small-forward angle scattering of lease beam with interaction of marine particles which follows the Mie theory. Among the instruments based on the above principle, commercially available LISST-100 has been widely used for the studies of sediment transportation near the bottom, dynamics of phytoplankton, and dynamics of suspended particles in the upper ocean. In this review, several examples of LISST-100 applications to coastal waters are introduced, together with some methodological problems associated with types and compositions of suspended particles. For the future studies of suspended particles dynamics in the field, information of structural and biochemical characteristics of marine particles is also crucial in addition to *in situ* assessment of their size distribution.

Key words: Particulate organic matter (POM), Marine snow, Settling particles, LISST-100, Optical particle sizing technique, Laser diffraction, Light scattering, Material cycle

Introduction

To understand the dynamics of bio-elements in coastal and estuarine environments, studies on suspended particles in water column are indispensable because of many reasons. Firstly, various portions of suspended particles are composed of organic materials such as phytoplankton or organic detritus associated with high metabolic activities, which play a central role in biogeochemical cycles in these environments. Secondly, suspended particles absorb and concentrate many forms of dissolved materials including trace metals from surrounding water column due to their large surface to volume ratio and physico-chemical interaction. Thirdly, suspended organic particles in upper water column are generally tended to sink out toward sediment and these settling processes are important to support life of various benthic populations as well as interaction of bio-elements through sediment-water column interface. However, in near shore environments, several sources of particulate materials of organic or inorganic forms are identified, i.e., riverine inputs, *in situ* algal and other biological production, sediment re-suspension and airbone deposition, and function of these particles in dynamics of bio-elements are largely depended on their physical and chemical characters as well as their bio-availability, which are still difficult to identify (Ward et al., 1994).

Organic form of suspended particles in coastal environments are defined conventionally as the size of particle larger than 0.4–1 μm using various filtration techniques, and all the materials passing through the filter are called dissolved organic materials (Sheldon, 1972; Wangersky, 1994). Upper size limit of suspended particles is not defined, but, separation of suspended materials from settling materials was commonly made based on behavior of particles in the water column, i.e., the particulate materials collected by sediment trap are methodologically defined settling materials. However, settling processes of particles in coastal and estuary environments are complicated as discussed in the following session, because of various origin and physico-chemical characteristic of particles as well as temporal and spatial variability of

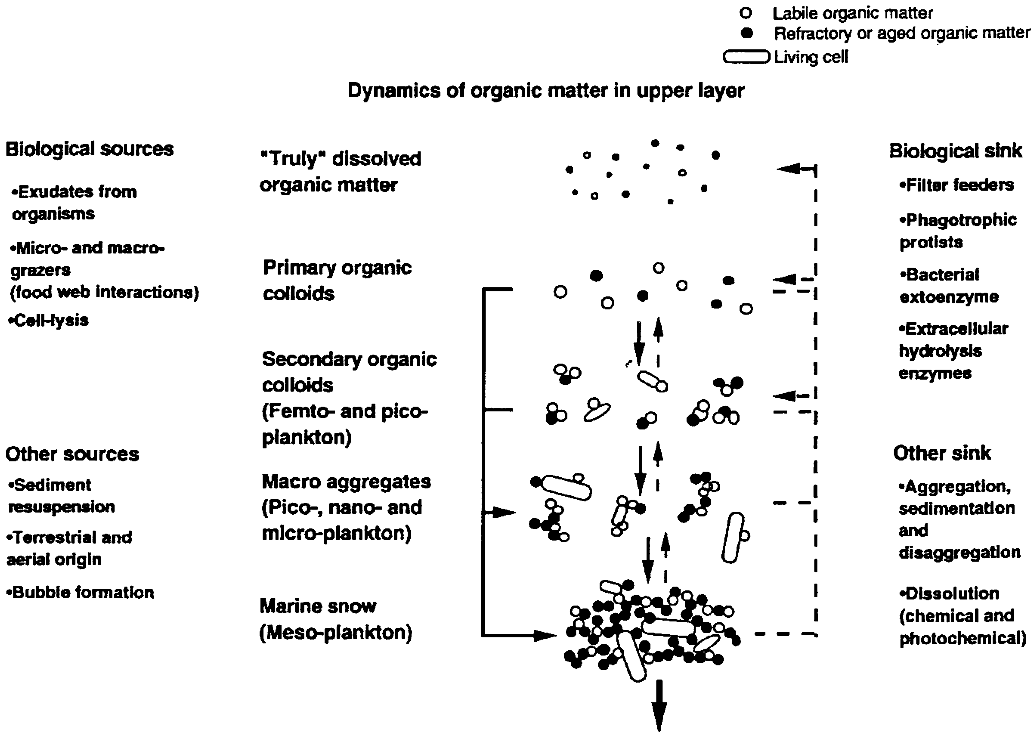

Fig. 1. Conceptual model of coagulation/disaggregation processes in the upper layer of marine environments (Hara and Koike, 2000).

physical environmental conditions such as salinity and currents.

Information of size distribution of suspended particles are crucial to understand their dynamics in these environments, since size of particles is one of the key factors controlling behavior of particle in fluid, including coagulation/disaggregation processes and sinking rates from upper waters. Various biological and non-biological sources and sinks of different size ranges are linked with coagulation/disaggregation processes in the water column (Hara and Koike, 2000: Fig 1), and, in the processes of coagulation/disaggregation, physical forcing controlling collision of particles are different according to their particle size, i.e., Brownian motion for less than 2 μm particles, laminar or turbulent shear for the 2–40 μm ones, and differential sedimentation rate between particles for larger than 40 μm ones (Johnson et al., 1994).

Observation of large size of suspended particles such as marine snow (>0.5 mm) in coastal environments has long history, and back to 50 th using submersible (Suzuki and Kato, 1953) and scuba divers (Shank and Trent, 1980). Also, for the size range of some 1–50 μm marine particles, use of coulter-counter method on board or land based laboratory after bottle sampling has been introduced in middle 60 th by Sheldon et al. (1972), and widely applied not only for field measurements, but also experimental studies such as grazing experiments of phytoplankton populations (Parsons, et al., 1967; Lampert, et al., 1986). Based on the early measurements of size distribution of suspended particles by using a coulter-counter, coagulation process of suspended particles in the ocean has been discussed (McCave, 1984). However, critical drawback of the coulter method is that this instrument needs sample water collection using pump or conventional sampling bottle before analysis, the procedure of which often destroys large sized suspended particles especially in the forms of non-living aggregates or flocs (Eisma et al., 1983). Using a under water camera and image analysis technique, Eisma et al. (1983) showed that size of aggregates in Ems estuary was roughly one order magnitude larger than that obtained by the coulter-counter method. Therefore, several instruments has been developed and applied for *in situ* assessment of particles size distribution in marine environments.

In this review, instrumental approaches to obtain *in situ* size distribution of suspended particles in coastal and estuary waters are outlined, but special focus is LISST-100 instruments (Agrawal and Pottsmith, 2000), which has been developed originally to assess sediment re-suspension in estuarine waters, but extended their application to upper water column, where the biological particulate formation is predominant compared to the bottom water environments. First, comparison of LISST-100 methodology to other *in situ* instruments of particle sizing with different methodology is summarized. Then, application of LISST-100 to various coastal and estuary environments is reviewed together with arguments on

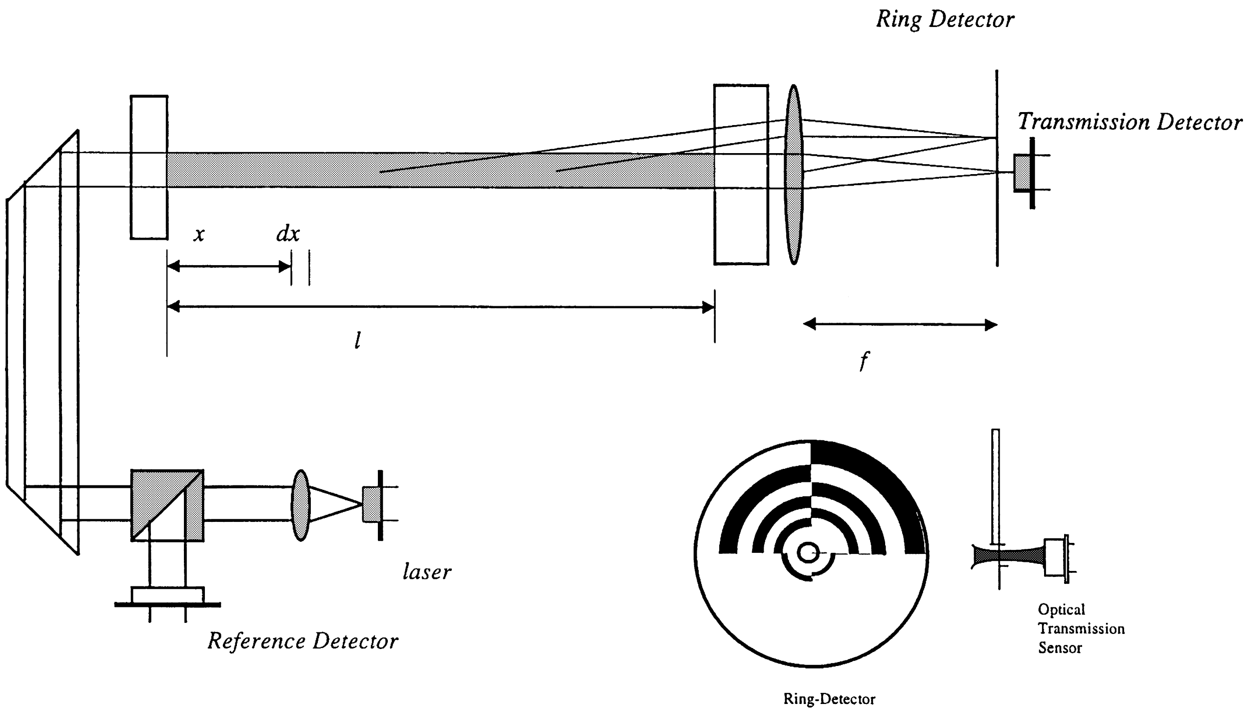

Fig. 2. Basic optical arrangement of LISST-100 and details of ring-detector (Agrawal and Pottsmith, H.C. 2000).

their advantages and some drawbacks. Finally, future aspect of marine particle sizing is discussed.

In situ sizing of particles using optical techniques

Methodology for *in situ* particle size analysis in marine environments has been categorized into several groups, which includes direct observation by scuba divers (Shanks and Trent, 1980, Alldredge and Gotschalk, 1988), image analysis of particles in water after *in situ* camera or video systems deployed from a research ship, and application of laser diffraction techniques. History of underwater camera application for particle analysis has been back to the early 80 th, when development of underwater camera has been stimulated by biological application such as counting zooplankton in the field (Ortner et al., 1981) as well as requirements of settling particle identification for sediment trap studies (Honjo et al., 1984; Asper, 1987). For example, Ortner et al., (1981) used Benthos 373 camera at the cod end of plankton net and obtained species composition of zooplankton in the Atlantic Ocean. Also, Honjo et al. (1984) applied an underwater still camera system to monitor abundance and size of marine snow during sediment trap experiments of deep waters. Since then, many type camera and video systems have been developed and applied in the field to observe *in situ* size distribution of marine particles, which provides valuable information regarding *in situ* their numbers, size and morphology of relatively large suspended particles such as marine snow in the ocean.

Application of optical technique to measure *in situ* distribution of marine particles has been independently developed by two groups using a same principle (Gentien et al., 1995; Agrawal and Pottsmith, 1994)., i.e., small-forward angle scattering of lease beam with interaction of marine particles following the Mie theory (Born and Wolf, 1975). Before the above *in situ* instruments, Bale and Morris (1987) used a commercial laboratory instrument of particle sizing manufactured with a same principle and adapted underwater use.

Principle of laser diffraction is based on the fact that scattering angles of collimated laser beam by particles in water are related to particle size, i.e., smaller particles have larger angle of scattering. Basic optical geometry used for laser diffraction method, developed by Sequoia Scientific, Inc. USA, is shown in Figure 2 (Agrawal and Pottsmith, 2000). Scattering beam by the presence of particle is detected by a ring detector, which locates at the focal plane of receiving lens of focal length of f. The radii of the detector rings increase logarithmically, and each ring on the detector represents a small range of logarithmically increasing scattering angles. The ring detector can detect scattered laser beam of 32 different scattering angles (LISST-100), and main beam without scattering can detected by photodiode placed behind the ring detector, giving the function of optical transmissometer (Fig. 2).

Correlation between ring detector output of two size class particles (10 μm and 86 μm) and detector ring numbers is shown in Figure 3 (Agrawal and Pottsmith, 2000). The scattered laser beam due to large particles senses at small angles (inner ring of small number), and vice versa. As the magnitude of scattering optical power at each ring is propor-

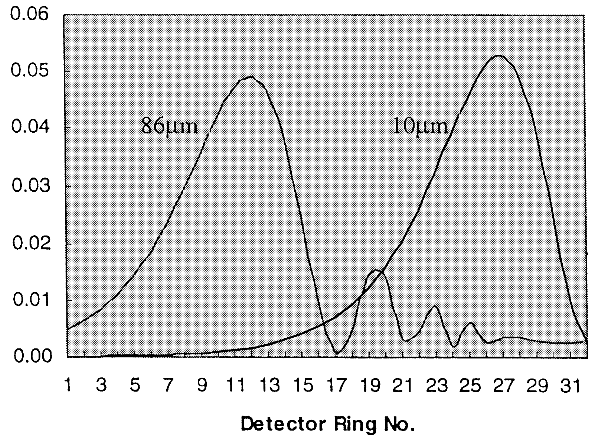

Fig. 3. Scattering signature of two particle sizes (10 μm and 86 μm) and its relation to detector ring number (Agrawal and Pottsmith, H.C. 2000).

tional to the number of particles of similar size class, optical power distribution obtained by ring detector contains basic information on size distribution of particles. Mathematical inversion of this data obtained by ring detector leads to a size distribution, i.e. concentration of sediments in 32 size classes. The size classes span a 200 : 1 range of particles in several types of LISST-100 instruments (1.25 to 250; 2.5 to 500; or 7.5 to 1500 μm).

Application of LISST-100 to natural marine particles and some technical issues

Since Lisst-100 was originally developed to monitor sediment transport of bottom layer of coastal and estuary environments, calibration of the instrument was commonly conducted by standard solid spherical particles of known diameter such as latex micro-particles. Also, refractive index of particles using computation of the scattering matrix was assumed to be solid spherical particles (Agrawal and Pottsmith, 2000). Therefore, if the refractive index of targeted natural particles is different from that of solid spherical particles, some error should occur in the final size distribution.

Regarding the shape effect of particles on LISST-100 distribution measurements, Agrawal et al. (in press) recently reported qualitative and quantitative differences of light scattering properties between homogeneous spherical standard particles and random shaped natural particles. Using AC spark plug dust as random particles, they found that random shaped particles, sieved through identical sieves as standard spheres, appeared 1/4 to1/2 Φ larger than standard particles in LISST-100 measurements. Also, size distribution of random particles became wider toward larger size range. Therefore, several pre-cautions are necessary to interpret the size distribution of natural particles obtained by the LISST-100 observations. Another type of irregular particles is the form of aggregates or flocs, although compact aggregates may behave like solid particles. In the case of larger loose aggregates, the laser beam would be scattered by many smaller and compact aggregates composing larger loose aggregates, resulting unreliable size distribution of suspended aggregates (Pedocchi and Garcia, 2006).

Regarding the application of LISST-100 for bio-particles such as phytoplankton populations, previous studies confirmed some reliability of the obtained size distribution and concentrations (Serra, et al., 2001; Arata, 2002). Serra et al., (2001) compared the particle-size distribution of phytoplankton, purple photosynthetic sulphur bacteria (*Chromatiaceae*), and suspended inorganic sediments measured *in situ* by LISST-100 to that obtained by laboratory instruments such as an optical microscope (OM), and a flow cytometer (FC). They concluded that, while all of these instruments gave reliable values of the particles concentration and size for the given populations, LISST-100 was the fastest and most reliable instruments since manipulation of the samples was not required for LISST-100. Even using the same laser refraction instruments, they noticed the difference of size distribution obtained with LISST-100 compared to those with a Galai laboratory laser size analyzer (GL) for populations with large and porous aggregates, such as phytoplankton cells. The difference would be attributed to the breakage of fragile algal aggregates during the measuring procedure used by GL.

Similar comparison was also made by Arata (2002) by using cultured morphologically different phytoplankton populations, i.e., *Prorocentrum minimum* as spherical phytoplankton and *Skeletonema costatum* and as chain forming ones. In the case of *P. minimum*, having spherical shape and rigid structure, both LISST-100 and optical microscope measurements gave similar peak size and volume concentrations. During the batch culture growth of S. *costatum*, microscopic counting of the cells as converted to their spherical diameter shifted their peak from ca. 15 to 30 μm and finally large aggregates of 150 μm size range. LISST-100 measurements also showed similar the above shift although peak is not sharp like microscopic counts. Arata (2002) concluded that LISST-100 gave size range of 20–100 μm for the chain forming *S. costatum* and *Chaetoceros spp* in exponential growth phase, and their aggregates during stationary growth phase observed more than 100 μm.

Assessment of phytoplankton dynamics and upper ocean biological processes

A Spanish group extensively used LISST-100 laser refraction method to analyze temporal and spatial distribution of phytoplankton and their relation to physical aquatic environments such as local turbulence and light condition. Serra et al. (2003a) observed daily vertical migration of purple photosynthetic sulphur bacteria, *Thiocystis minor* population in a lake. Since this sulfur bacteria migrates towards light (photo taxis) as well as towards lower redox condition rather rapidly (the fastest vertical displacement was 0.6 $m\,h^{-1}$), LISST-100 instrument, which can monitor vertical distribution of particles in every 1 min interval within 10 cm vertical accuracy, was suitable equipment for analyzing their behavior. LISST-100 was also applied to analyze the role of the physical flow field on the horizontal and vertical distributions of different phytoplankton populations in a shallow coastal ecosystem (Serra et al., 2003b). Using two study conditions of different energy field, they found that the mixing level was enough to homogenize vertical distribution of phytoplankton population at two sites, although the level is not enough to homogenize the temperature in the low-energy period. Mikkelsen (2002) observed occurrence of algal bloom at sheltered Danish coastal waters in September. Size spectra assessed LISST-100 showed increased aggregates size toward bottom, suggesting that the end of algal bloom induce aggregation of algae and settlement of large aggregates from upper water column.

Koike et al. (2002) improved sensitivity of the original LISST-100 instrument by modifying its light pass length from 5cm to 20 cm, and applied this instrument to observe fine scale vertical distribution of suspended particle size distribution in upper layers of Japanese coastal waters. Theoretically LISST-100 (Type C) can assess 32 size ranges from 2.7 to 460 μm, but we noticed some artifact at the upper margin. Vertical profile of 4 size ranges of particles volume together with some physical parameters (temperature and density gradient) obtained at the central station of Sagami Bay in summer was shown in Figure 4 (Koike et al., 2002). Half hour time difference between the casts of physical parameters by CTD and LISST-100 resulted some 5–7 m deeper mixing layer for the LISST-100 profile, but, total volume of particles (2.7–258 μm) in the mixing layer showed constant number of ca. 25 μl/L. Large increase of total particle volume (3 times higher compared to mixing layer) was observed within pycnocline. In addition, comparison of particle size distribution between mixing layer (1–4 m) and pycnocline (20–25 m) at the station clearly indicated accumulation of larger sized particles (size ranges between 10–100 μm and 100–150 μm) in pycnocline, possibly by aggregation process (Fig. 5). Accumulation of large suspended particles such as marine snow has been often reported below the mixing layer (MacIntyre et al., 1995), but accurate change of their size distribution can be achieved by using LISST-100 *in situ* assessment.

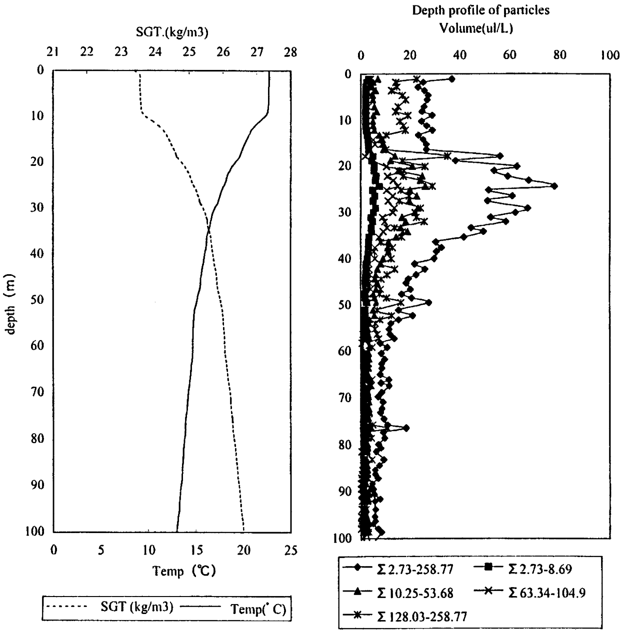

Fig. 4. Vertical distribution of suspended particles (4 size classes) observed with LISST-100 and physical parameters at central Sagami Bay in summer (Koike et al., 2002).

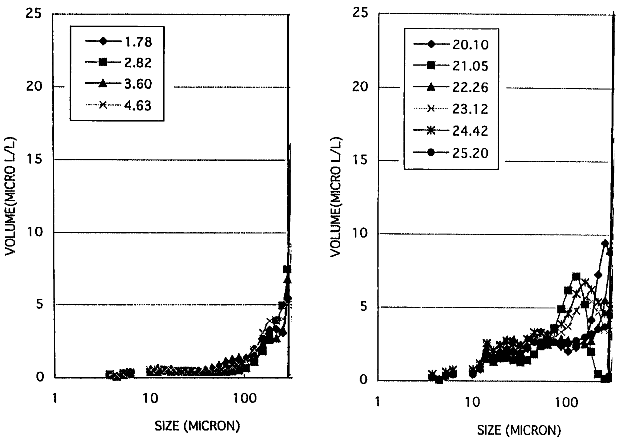

Fig. 5. Size distribution of suspended particles at mixing layer (1–4 m: left) and at pycnocline depth (20–25 m: right) at central Sagami Bay in summer (Koike et al., 2002).

Assessment of near bottom sediment transport

Historically, the mass concentrations of suspended particle matters in estuary and coastal waters have been measured by optical sensors such as transmissometers and optical backscatter sensors (OBS), which showed bulk and averaged information of total particles (Jones and Wills, 1956; Downing et al., 1981; Moody et al., 1987). Several studies, however, have shown that the above optical measurements depends not only total amounts of suspended particle concentrations (SSC), but also their size distributions (Baker and Lavelle, 1984; Gibbs and Wolanski, 1992). In addition, flocculation of fine sediments to coarse-sized aggregates in seawater changes their size distributions. To understand the sedimentary processes of suspended particles, assessment of *in situ* size distribution of suspended sediments is indispensable. For the above purpose, use of laser refractory instruments, typically LISST-100, has become popular because of its wide size range of size assessment and easily processing of large volume of data (Pedocchi, and Garcia, 2006).

Another important parameter for sediment transport in estuary and coastal environments is settling velocities of particles, since the latter is not evaluated simply by size of suspended sediments but also needs information of particle density. In this reason, independent measurements of the size and settling velocities of suspended sediments are also required. For this purpose, settling chambers of various designs have been used to measure the settling velocity of suspended sediments. Settling chamber adapted with a laser refractory instrument is also commercially available (Agrawal and Pottsmith, 2000), which can evaluate settling velocities of 8 different size classes of suspended particles (5–500 μm). This instrument was originally developed for settling velocities of suspended sediment, but it can apply for other particles such as phytoplankton population.

Since settling chamber method needs long time for one measurement, thus limits their information rather a spot measurement. Combining *in situ* size distribution of flocculated suspended sediments obtained by LISST-100, and the total concentrations of suspended particles (TSC) obtained from 2L bottle samples of the corresponding depth, mean *in situ* effective density and mean settling velocity of suspended sediments (flocs) can be approximately computed (Mikkelsen and Pejrup, 2001). For the calculation of effective density ($\Delta\rho$: floc density minus seawater density), they assumed large parts of suspended sediments existed as flocs, which composed mostly seawaters in terms of the floc volume. Stokes's law was applied to calculate mean settling velocity of suspended particles, using above obtained $\Delta\rho$ and mean size of particles from LISST-100.

This approach has one obvious advantage over settling chamber method, because spatial and temporal mapping of the above information in the study area, such as an estuary, can be possible by the combination of quick water sampling with LISST-measurements of less than a few minutes. Mikkelsen and Pejrup (2000) studied *in situ* particle size distribution and aggregates densities within a dredging plume of Donnish coast using LISST-100 deployment. From the spatial variation in size distribution and mean density of suspended sediments, which obtained from LISST-100 total volume measurements and SSC concentrations from bottle samples, they calculated settling velocity of sediments within the plume from time to time. They found that the difference in settling velocity of suspended particles within a plume remained only a factor of 1.7 times, since .increase of *in situ* particle size due to the flocculation processes within the plume (2–3 times) was compensated by decreasing mean density of suspended particles.

Future aspects of *in situ* measurements of particles size distribution in coastal environments

Since size range of suspended particles observed in coastal and estuary environments extends from 1 μm to 1 cm size range, which represents four orders of magnitude size range, no single *in situ* instrument can cover these large size ranges. Therefore, combination of multi-instruments is necessary to obtain to cover full size range of suspended particles. Recently, Mikkelsen et al. (2006) combined two *in situ* instruments to monitor full range of suspended particles in coastal waters, i.e., using LISST-100 from size range of 2.5–500 μm and digital floc camera (DFC; 135–9900 μm), to examine flocculation and de-flocculation processes of suspended particles at the near bottom depth of stations located off the mouth of rivers discharging Adriatic Sea. According to Eisma (1986), three size ranges of suspended particles, i.e., single grains ($<$36 μm), microflocs (36–125 μm) and macroflocs ($>$125 μm), were classified, and variation of these three fractions over range of forcing conditions was examined. It is demonstrated that when physical stress decreases, the volume occupied by macroflocs first increases and then decreases as flocs settles out during clam conditions. They concluded that no overall relationship between physical stress and floc size was found because parameters other than stress influence floc size, especially re-suspension, settling, advection and biological activity. Full size range of particle size spectra (1 μm to 1 cm) in California coastal water was also reported using different combinations of *in situ* and laboratory instruments (Jackson et al., 1997). To assess the particle aggregation processes in the upper coastal waters, they used Coulter particles counter and Elzone particle counter for the size range 4–100 μm and several types of *in situ* image analysis systems for $>$100–400 μm size range. In this study, most of the particle mass was in the size range of 0.1–3 mm, and volume distribution obtained is consistent with theories that assume particle sizes are controlled by simultaneous coagulation and disaggregation.

The maximum size range obtained by commercially available laser diffraction instrument is the LISST-FLOC (Sequoia Scientific Inc. USA), which covers the largest sizes, 7.5 μm to 1.5 mm (nominal), and is intended for measuring flocculated marine particles. Application of the laser diffraction instrument is desirable, because of fragile nature of larger sized particles in the coastal environments. Several problems are still remained unsolved, however, to apply this instrument for study of the large marine aggregates. So far, there is no study regarding determination of refractive index for marine organic aggregates, and, in the upper ocean, organic aggregates are one of the predominant forms of particles. In addition, possible complicated structure of loose organic aggregates of large size may give totally misleading information by laser diffraction as cited before (Pedocchi and Garcia, 2006). Extensive inter-calibration using a various forms of marine aggregates between laser diffraction instruments and other methods such as video-camera image is required both in laboratory and field.

The other important information to understand the dynamics of marine particles is their chemical and morphological characters, which can not be obtained by laser diffraction instruments. Sampling of marine snow of visible size in upper coastal environments has often conducted by scuba-divers (Alldredge, 1991). Also, for the chemical and biological analyses of smaller size range of suspended particles (ca. <10–20 μm), size fractionation technique using various pore sized filters has been applied after sampling of the targeted water (Volkman and Tanoue, 2002). Therefore, separation and collection of intact marine aggregates of size range between 20–500 μm from marine environments are remained as the most difficult challenge. Using a density difference of marine aggregates, some trial to this direction was already reported (Gustafsson, et al., 2000). Together with the information collected by laser diffraction instrument such as LISST-FLOC, structural and biochemical information of marine aggregates can extensively improve our understanding of dynamics of marine suspended particles, especially larger size aggregates.

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