A method to measure marine particle aggregate disruption in situ

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Abstract

Particle aggregation within aquatic environments is a primary factor controlling the vertical flux of suspended matter. The aggregation process is controlled by the rate of particle interaction, enhanced by turbulent motions and differential settling, and the probability that particles making contact will stick together. The larger the particle aggregate, the faster it will sink. We describe a new and novel method to measure the state of particle aggregation in situ as a function of turbulent energy using an inexpensive attachment, the in situ dis-aggregation system (iDAS), to a commercially available particle size instrument, a Sequoia Scientific LISS-100X. A small chamber is attached to the instrument tand sample is drawn into the chamber using an inexpensive, variable speed thruster designed for remotely operated underwater vehicles. Ambient water drawn into the sample chamber passes through a flexible tube of defined diameter and length. The flow velocity through the tube and tube dimensions are used to estimate the turbulent energy that particles experience. As turbulence increases, particle aggregations disrupt, shifting the size distribution towards smaller particles. The method was tested under controlled laboratory conditions using standard test clay material and within several coastal environments along the East Coast of the United States. In all instances, particle aggregations are shown to disrupt as turbulence increases, causing the particle size distribution to shift toward smaller fractured aggregates and component particles. The IDAS can be used to directly measure the aggregation state of suspended material and potentially to estimate the bonding strength between aggregated particles of different type.

Aggregation of suspended particles within aquatic environments controls the rate of sinking and subsequent removal of matter from the water column. Particle settling rate is required to accurately model sediment transport in coastal waters (Dryer 1995) and is a primary factor in estimating vertical carbon flux within the global ocean (Ducklow et al. 2001). The topic of marine particle aggregation, which is also referred to as coagulation, was thoroughly reviewed in both practice and theory by Burd and Jackson (2009).

Particle aggregation occurs when two or more particles come in contact and stick together, forming a larger aggregate (floc) that settles faster than the individual component particles. Particles stick together through a combination of electrostatic force that operates primarily on inorganic matter having high metal content and biological exudates that can serve as glue to hold particles together. Particle interaction rate is a function of sediment concentration and relative particle motion, either due to differential sinking rate or small-scale turbulent motions. The resulting flocs may be described as loose, heterogeneous associations of particulate matter held together by relatively weak bonds (Fig. 1).

Turbulence plays a dual role in the particle aggregation process. Small-scale motions serve to bring particles together and as flocs grow in size the particle size distribution (PSD) shifts toward larger sizes (Burd and Jackson 2009). We note that in this sense, we refer to a floc as an independent particle. However, turbulence can also limit floc growth and disrupt larger flocs due to shear forces induced by small-scale turbulent eddies. Shear at these scales can be parameterized using the dissipation rate of turbulent kinetic energy $\varepsilon \, m^2 s^{-3}$, which increases as the flow becomes more turbulent. Floc disruption, either as smaller fractured flocs or individual component particles, will shift the PSD toward smaller particles. Higher relative concentrations of aggregates within the particle population will result in more noticeable shifts in the PSD with increasing turbulence-induced disruption. Conversely, it follows that the absence of aggregates will render the PSD insensitive to increasing $\varepsilon$.

In the last two decades in situ instrumentation has been developed to measure the size distribution of suspended particulate matter nondestructively (Gartner et al. 2001; Montovanelli and Ridd 2006; Olson and Sosik 2007; Davies...
et al. 2015). For example, the Sequoia Scientific LISST-100X instrument (LISST) measures in situ light scatter in 32 near-forward directions, between 0.082° and 13.84° from the direction of propagation, from a small volume of ambient water (Agrawal and Pottsmith 2000). The signals are inverted for particle volume across logarithmically spaced bins of median size ranging from 1.09 to 184 μm based on assumptions of random particle shape and orientation (Agrawal et al. 2008). While these instruments can accurately resolve details of the PSD, they alone are incapable of definitively assessing the amount of material arranged within aggregations. To address this limitation, we constructed and tested under controlled laboratory conditions and within natural environments a simple attachment to a LISST instrument, the in situ dis-aggregation system (iDAS). The attachment is designed to systematically subject ambient particle populations to calibrated turbulent fluid motions while simultaneously measuring associated changes in the PSD resulting from floc disruption. Our method is a refinement of laboratory techniques reported by Rau et al. (2018) and builds on ideas and methods reported by Slade et al. 2011. The purposes of this communication are to describe the iDAS construction, outline operation procedures, present results from laboratory and field experiments, and discuss potential sources of uncertainty in the measurements.

Materials and procedures
iDAS design

The concept of subjecting particle suspensions to controlled levels of turbulence using flow through a narrow tube was tested and reported in an earlier communication (Rau et al. 2018). The researchers set up a hydrostatic pressure

\[
e = \frac{\tau_w u}{\rho d^2 / 4}
\]

where \(\tau_w\) is the tube wall shear stress (Schlichting and Gersten 2017), \(\bar{u}\) is the average sample flow velocity in the tube, and \(\rho\) is the fluid density. Standard clay material (bentonite, volumetric mean diameter of 3.7 μm, Alfa Aesar, stock no. A15795) was used to create suspensions within saline water in order to induce the formation of flocs. The study found the PSD of the clay particles, measured using a LISST Type B instrument and validated with microscopic imaging, to systematically shift with increasing \(e\) to smaller size due to aggregate disruption upon passage through the syphon and subsequent exposure to the flow-generated turbulence.

The iDAS was developed based on lessons learned from the DTM experience with the goal of simplifying the measurement procedures and enabling deployment within natural environments. The device consists of a sampling chamber fitted around the measurement end of a LISST and sealed tightly around the external housing of the instrument with an O-ring (Fig. 2). The chamber is essentially a repurposed Sequoia Scientific LISST-100X 5.4 L calibration chamber. With the LISST inserted in the chamber the sample volume, decreased by the immersed volume of the instrument (approximately 1 L), is approximately 4.4 L. Ambient water is drawn through the iDAS using a variable speed remotely operated vehicle thruster motor (Blue Robotics T200) inserted into a short length of 10.2 cm (4 in.) diameter plastic pipe and attached directly to side of the sample chamber. The T200 is a three-phase, brushless motor and is attached to an open-loop electronic speed controller (ESC, also available from Blue Robotics) and an ESC consistency tester (we used a HJ ESC Consistency Tester). The ESC provides pulses of current to the motor and the pulse frequency determines motor speed. In the current version, the thruster is controlled manually through a power cable that connects the iDAS to a 15-V power supply and the ESC, both of which are positioned above water on the deck of the deployment platform. Ambient water is drawn into the sample chamber through a flexible Tygon tube of diameter \(d = 1.27\) cm and length \(l = 2.15\) m (\(l/d = 169\)) that is attached to the sample chamber opposite the thruster motor. The iDAS design concept can be easily adapted to other instruments by using appropriate sample chambers. We chose the tubing length so that the portion of the tube where turbulence is fully developed exceeds a length of 100d, which is more than sufficient to ensure that all particles within the
flow experience the highest rates of shear at each flow condition (Bäbler et al. 2015). The total cost of the iDAS component parts as described is approximately $650 (see Supporting Information Table S1) and construction requirements are minimal using simple hand tools, plastic cement, and waterproof tape to seal the lid of the Sequoia Scientific sample chamber.

In order to compute $\epsilon$, $\bar{u}$ must first be calibrated against the thruster speed. The calibration must represent the entire iDAS system in order to account for all frictional effects. This is accomplished by immersing the iDAS attached to a LISST within a large water tank with the free end of the sample tube attached to the base of a volume-calibrated bucket. The bucket is positioned at the water surface and allowed to fill to approximately $\frac{3}{4}$ of the volume. In this configuration, iDAS draws water from the bucket at a rate proportional to the thruster speed. As iDAS extracts water the bucket is raised manually in order to minimize the water level and hydrostatic pressure difference between the bucket and the tank. For a given thruster speed ($S_{ESC}$), expressed as propeller revolutions per minute (calibration provided by Blue Robotics, www.bluerobotics.com), $\bar{u}$ is computed from the amount of time ($T_b$ s) required for iDAS to extract a prescribed volume of water ($V_b$ m$^3$) from the calibrated bucket;

$$\bar{u} = \frac{4}{\pi d^2} \frac{V_b}{T_b}. \quad (2)$$

The T200 is an open loop design in that motor speed is completely determined by current pulse frequency with no feedback from the motor. Consequently, there can be slight changes in the empirical relationship between motors independent of experimental error. To better understand the total calibration uncertainty, we conducted the procedure using two different motors (Fig. 3). The empirical relationship between $\bar{u}$ and $S_{ESC}$ for the combined data sets is expressed as a second-order, least-squares polynomial regression; $\bar{u} = -2.04 \times 10^{-8} S_{ESC}^2 + 2.61 \times 10^{-4} S_{ESC} + 0.0146$. The Pearson product moment correlation is $R = 0.97$ and the root mean-squared error and mean absolute error as defined by Willmott...
et al. (1985) are 0.0275 and 0.0258 m s$^{-1}$, respectively. The uncertainty equivalence in $\varepsilon$ is 0.001 m$^2$ s$^{-3}$ at the lowest turbulence range and 0.01 m$^2$ s$^{-3}$ at the highest range. Since a primary source of error in flow rate calibration is the ability to manually adjust the bucket height in order to minimize hydrostatic pressure, total uncertainty can be reduced with replicate calibration runs and operator practice. Uncertainty due to slight differences between motors can be reduced with periodic motor-specific calibration and monitoring of changes in motor performance.

The maximum flow velocity achieved with iDAS is approximately 0.7 m s$^{-1}$. At the low range of $\bar{u}$ flow through the tube is laminar, at which point the sampled particles are representative of ambient conditions without exposure to elevated turbulence. Turbulent flow does not occur until the Reynolds number, $Re = \bar{u}d/\nu$ and $\nu$ is the kinematic viscosity of the water ($= 8.175 \times 10^{-7}$ m$^2$ s$^{-1}$, where the water tank temperature was 29°C), exceeds 2300 (Schlichting and Gersten 2017). For the iDAS equipped with our 1.27-cm diameter tube, flow will transition from laminar to turbulent when $\bar{u}$ exceeds 0.23 m s$^{-1}$. The resulting range in $\varepsilon$ generated by the iDAS is, therefore, on the order of $10^{-3}$ to $10^{-4}$ m$^2$s$^{-3}$. At the lower iDAS range, $\varepsilon$ is characteristic of turbulence observed within the upper ocean, for example, $\varepsilon = 10^{-3}$ m$^2$ s$^{-3}$ (Thorpe 2005). At the upper end of the measurement range $\varepsilon$ exceeds reported oceanic values. We note that the sample tube diameter could be increased in order to establish a lower range of $\varepsilon$ in accordance with Eq. 1, as long as the sample flow rate and tubing length are adjusted accordingly to maintain fully developed turbulent flow.

The size of the smallest turbulent eddies is described as the Kolmogorov length scale, $\eta$,

$$\eta = \left(\frac{\nu^3}{\varepsilon}\right)^{\frac{1}{4}},$$

and $n = 0.25$. The range in $\eta$ for the corresponding range in iDAS turbulence is ~ 200 μm (low $\varepsilon$) to ~ 40 μm (high $\varepsilon$). The Kolmogorov length scale is useful for approximating maximum floc size and the onset of flocculation but does not necessarily serve as an absolute limit to floc size (Rau et al. 2018).

### Procedures

The iDAS data collection procedure involves setting the thruster motor at a defined speed calibrated to flow rate and $\varepsilon$, letting the sample chamber flush adequately, and then collecting a time series of LISST observations long enough to obtain a statistically significant sample. LISST observations are collected at a rate of 1 s$^{-1}$, allowing for adequate flushing of the instrument interrogation volume. iDAS is set to the lowest flow rate first (laminar flow), and then progressively stepped through the range of flow rates from low to high at constant time intervals (Fig. 4). The LISST data time stamps are used to identify the records that represent particles subjected to a given $\varepsilon$. Time intervals are typically between 5 and 10 min for each $\varepsilon$, which allows for adequate flushing of the sample chamber, and only the last 2 min of each interval are analyzed for PSD. While we maintained the same initial flushing time at each flow rate for procedural simplicity, we recognize that sampling time could be further optimized since the minimum flushing time will decrease with increasing flow rate.

Data analysis software supplied by Sequoia Scientific is used to invert the forward light scatter for PSD using the random shape approximation (Agrawal et al. 2008). For each record, forward light scatter is inverted for particle volume in each size bin. Average bin volume, $V_p$ μL L$^{-1}$, is then computed for the final 2-min time series at each flow rate setting. In addition to particle light scatter, the LISST measures the transmittance of the laser beam ($\lambda = 670$ nm) through the water mixture from which beam attenuation, $c$ cm$^{-1}$, is computed. Since beam attenuation varies linearly with suspended particulate mass (SPM), LISST $c$ can be used as an indication of changes in ambient suspended matter concentration when measured under laminar flow conditions. However, $c$ is also sensitive to changes in aggregate morphology (Slade et al. 2011) and, therefore, we advise that iDAS measurements be accompanied with independent measurements in ambient water conditions, for example, temperature, salinity, and light scatter, that can impact LISST light scatter measurements.

In order to indicate where in the PSD particle mass is removed or added due to floc disruption, the minimum average particle volume, $[V_p]_{\text{MIN}}$, observed across all flow rates throughout the course of each experiment is subtracted from $V_p$ for each size bin (Fig. 4);

$$V_p' = V_p - [V_p]_{\text{MIN}}$$

At low $\varepsilon$ and minimal disruption, $V_p'$ is zero within size bins representing individual particles and a maximum for bins representing flocculated material. As $\varepsilon$ increases and disaggregation shifts particle mass to smaller size, $V_p'$ will decrease within size bins where particle mass is removed by floc disruption and increase in smaller size bins where disrupted particle mass is added.

A complication in the interpretation of $V_p'$ arises when particle volume is added to the LISST size range as a result of disruption of flocs larger than the largest size bin. Upon disruption, particle mass associated with dislodged particles and floc fragments will add to the measured PSD if it falls within the LISST size range. The addition of particle mass from outside of the LISST size range will lead to an underestimation of floc disruption within larger LISST size bins and potentially an overestimation of the accumulation of disrupted particle mass in smaller size bins.
Assessment

Clay suspension tests

iDAS was tested under laboratory conditions using standard clay suspensions obtained from the Source Clay Repository of the Clay Minerals Society, Chantilly, Virginia. Two types of clay were used: Georgia kaolinite (KGa-1b) and Wyoming montmorillonite (SWy-2). The composition of these natural samples is dominated by aluminum oxide and other minerals and they readily aggregate due to electrostatic forces when suspended within saline water. The mean particle size of KGa-1b and SWy-2 is approximately 5 and 4 μm, respectively.

Experiments were conducted in a large laboratory tank filled with 450 L of clean, fresh water. The water was made brackish (≈10 psu) with the addition of Instant Ocean, a saltwater aquarium product designed to produce a saline mixture similar to natural seawater with respect to the concentrations of chloride, sodium, sulfate, magnesium, calcium, and potassium (www.instantocean.com). Normally, LISST measurements are automatically stored relative to a clear water calibration. However, for the tank experiments, the "clear water" used for reference was the tank water measured prior to the addition of clay material. Clear water was prepared by filling the tank with tap water, adding the required amount of Instant Ocean, and filtering the brackish water through 0.5 and 0.1-μm inline cartridge filters using a recirculation pump. The beam attenuation and PSD were monitored with the LISST instrument submerged within the tank without the iDAS attached and filtering was halted when c and V_p were minimized and stable.

In each experiment, a quantity of clay was weighed and added to the tank; 4 g of KGa-1b was added resulting in a mass concentration of 8.8 mg L⁻¹ and 5 g of SWy-2 was added resulting in a mass concentration of 11.1 mg L⁻¹. A drum fitted with small paddles and attached to a variable speed electrical motor (further described in Rau et al. 2018) was positioned vertically within one end of the tank and slowly rotated in order to keep the clay material suspended throughout the course of each experiment. Drum rotation speed was adjusted such that the added turbulence was enough to keep the clay material suspended, but not enough to prohibit the formation of larger flocs. For each suspension, iDAS was positioned at the bottom of the tank with the entrance to the sample tube positioned at approximately mid-depth.

Both clay materials were found to aggregate, although SWy-2 formed larger sized aggregations than KGa-1b by a factor of four. Aggregated particle volume for KGa-1b peaked within the 7.9-μm size bin while SWy-2 produced a peak volume at 41.5 μm (Fig. 5). For both clay materials, the distribution of particle volume appears to have been contained within the LISST size range. However, when the particles were subjected to the highest turbulence, volume increased slightly in the larger size bins > 100 μm, suggesting perhaps the presence of larger aggregates beyond the size range of the LISST. Higher turbulences could have disrupted such aggregates and shifted mass into the measurement range. A similar explanation might be inversion error related to the presence of larger particles outside the range of LISST measurements (Mikkelsen et al. 2005). Excluding these large size bins and limiting the
analysis to the size range < 100 μm, the largest particles, characterized as the size of the 95th percentile of the PSD population ($D_{95}$), was 11.8 μm for KGa-1b and 60.9 μm for SWy-2. In each experiment aggregations disrupted systematically with increasing turbulent energy, shifting the PSDs toward smaller particle size. Given the fragile nature of clay flocs held together by weak electrostatic forces (Glasgow and Ping Hsu 1982) it is reasonable to expect that the maximum floc size should not exceed the average size of the turbulent eddies, assuming that the associated shear stress is enough to disrupt larger aggregates. A comparison of $D_{95}$ with $\eta$ indicates that floc size for both clays is smaller than $\eta$ and that the slopes with respect to $1/\varepsilon$ are $n = 0.25$ for SWy-2 and 0.13 for KGa-1b (Fig. 6).

The particle concentration within the tank was found to decrease slightly during the course of the experiments and was likely due to a small degree of particle settling. As a result, one can expect the particle population within the tank to have shifted slightly toward smaller particles with time since larger particles will preferentially settle faster than smaller particles. It is, therefore, possible that particle settling may impact the observed changes in PSD in addition to aggregate disruption. To investigate this effect, we measured a 70-min time series of a bentonite test clay suspension (Alfa Aesar, stock No. A15795, mean diameter = 3.7 μm) where iDAS was switched between laminar flow and turbulent flow every 5 min (Fig. 7). Turbulent flow ranged from 0.008 m$^2$ s$^{-3}$ in the beginning portion of the time series to 0.313 m$^2$ s$^{-3}$ at the end of the time series. The data highlight several aspects of the tank measurements. First, both total particle volume summed across the 32 LISST size bins and $c$ measured with laminar flow gradually decreases with time as a result of particle settling. Second, superimposed on the long-term trends are changes in particle volume and attenuation in response to turbulent energy. When the flow is turbulent, $V_p$ decreases rapidly in response to aggregate disruption. Conversely, when turbulence is relaxed to laminar flow (shaded portions of the time series), $V_p$ rebounds toward ambient conditions. Beam attenuation reacts similarly; values are depressed with aggregate

![Fig. 5](image-url). Change in particle volume distribution for standard clay particles suspended in brackish water (salinity = 10.6 psu). Clay samples were purchased from the Mineral Clay Society; Kaolinite (KGa-1b, left panel) and Montmorillonite (SWy-2, right panel). The mass concentrations of clay suspension are 8.8 mg m$^{-3}$ (KGa-1b) and 11 mg m$^{-3}$ (SWy-2). Associated $D_{95}$ values are shown above each set of curves and identified with color and line format. The shaded regions within the PSDs indicate the approximate range in $D_{95}$.

![Fig. 6](image-url). The non-linear relationships between $D_{95}$ for kaolinite (KGa-1b, circles) and montmorillonite (SWy-2, squares) versus $1/\varepsilon$, given the formulation $D_{95} = (1/\varepsilon)^n$. The Kolmogorov length scale, $\eta$, is shown for reference (dashed line).
disruption and elevated with floc formation. The decrease in particle volume due to settling, estimated from the peak $V_p$ measured across adjacent laminar flow periods, is approximately $0.1 \, \mu L^{-1}$ while the turbulence-induced decrease in particle volume ranged between $1 \, \mu L^{-1}$ at the lowest turbulent flow rate and $1.5 \, \mu L^{-1}$ at the highest flow rate. Thus, while particle settling is clearly evident within the time series, the superimposed turbulence-induced changes in particle volume (and $c$) are an order of magnitude larger in amplitude compared with laminar flow conditions.

Another way to think about particle aggregation is apparent density; the average density of the loosely associated particles, including the interstitial water. An aggregate would have a lower apparent density than an individual clay particle since the density of clay is greater than that of water. Hence, apparent density will increase with aggregate disruption. Hurley et al. (2016) argued that since beam attenuation is proportional to and, therefore, a reasonable proxy for SPM, and since apparent density is defined as mass per unit volume, then the ratio of $c$ to $V_p$ is a reasonable proxy for particle density. Using attenuation measured at the laminar flow setting as an estimate of ambient conditions, our results indicate that $c/V_p$ increased by 90% (1.2–2.4) for KGa-1b across the range of $\varepsilon$ investigated while SWy-2 increased by a factor of 3 (0.6–2.8). Since $c$ and, therefore, SPM decreased only slightly throughout the experiments due to particle settling compared with floc formation and disruption, these results further illustrate the relative effects of particle settling and aggregation dynamics on the measured particle morphology.

For both clay samples, aggregate disruption is clearly evident within the $V_p'$ distribution in size bins larger than the maximum size of the primary particles; $D > 4.82 \, \mu m$ for KGa-1b and $D > 4.09 \, \mu m$ for SWy-2 (Fig. 8). The $V_p'$ distribution appears to pivot around the upper limit of the primary clay particle size with increasing $\varepsilon$, revealing progressively increasing primary particle volume and decreasing floc volume for each of the clay samples. The increase in primary particle volume is consistently less than the decrease in floc volume, indicating that the aggregated material was loosely packed with a significant amount of interstitial space. However, the increase in primary particle volume is different between the two clays. For KGa-1b, the increase in primary particle volume represented 25% of the decrease in aggregate volume at the highest $\varepsilon$ investigated. SWy-2 primary particle volume, on the other hand, only increased by approximately 3% relative to aggregate disruption. Thus, the kaolinite flocs were smaller and perhaps more compact relative to the montmorillonite flocs.

These results are similar to those reported for bentonite using the DTM (Rau et al. 2018). Bentonite aggregates disrupted with increased turbulent energy. $D_{95}$ determined from the LISST PSD was generally less than $\eta$ and the rate of decrease in $D_{95}$ with increasing $\varepsilon$ was less than or similar to the theoretical change in $\eta$.

In situ measurements of natural suspensions

The iDAS was tested in several estuarine and coastal environments along the East Coast of the United States (Table 1).
Measurements within the Severn River (SR) were collected in the boat basin of the U.S. Naval Academy, Annapolis, Maryland. Measurements in North Carolina coastal waters (NCC) were collected at the U.S. Army Corps of Engineers research pier located in Duck, NC. Finally, measurements were collected in western Long Island Sound (LIS) in waters adjacent to the University of Connecticut Avery Point campus, Groton, CT from the research vessel R/V Connecticut. For each experiment, iDAS was deployed at a fixed depth of approximately 1 m and a time series of LISST measurements was collected while manually stepping through the range of turbulent flow rates. Simultaneous measurements of water temperature, salinity, turbidity, expressed as either Formazin Nephelometric Units (FNU) or beam attenuation, and fluorometric chlorophyll a (Chl a) concentration were measured using a YSI EXO sonde (SR) or SeaBird sensors (NCC and LIS).

At each location, the environmental conditions were reasonably stable. The mean turbidity (FNU or c), standard deviation (σ), and coefficient of variance (CV = σ/ Mean) of turbidity, for example, at each study site was SR: FNU = 6.7 m⁻¹, σ = 0.09 m⁻¹, and CV = 0.013, NCC: c = 6.61 m⁻¹, σ = 0.35 m⁻¹, and CV = 0.052, and LIS: c = 2.23 m⁻¹, σ = 0.19 m⁻¹, and CV = 0.084. Similarly, the CV for temperature, salinity, and Chl a concentration ranged from 0.004 to 0.056, 0.001 to 0.005, and 0.008 to 0.099, respectively. As in the laboratory observations of clay, the PSD of the natural particle populations shifted toward smaller particles with increasing ε (Fig. 9). Also similar to the clay materials, the V_p distribution for the natural populations appeared to shift progressively toward smaller particles and to pivot around a size bin that clearly separates larger flocs that disrupted with increasing turbulence from a gradually evolving population of smaller primary particles. The pivot size bin is different for each environment; 6.7 μm for SR, 15.4 μm for NCC, and approximately 30 μm for LIS, perhaps indicating differences in primary particle size. In addition to the location of the pivot point, the shape of the PSD for particles smaller than the pivot size changes from one site to the next. Unlike the clay suspensions that indicate a single primary particle peak, the natural populations indicate more complicated PSDs, including bimodal (SR) and trimodal (LIS) examples. These results are not unexpected since natural particle populations are highly heterogeneous compared with standard test particles. The type and concentration of component material is a function of source, for example, erodible sediments and local microbial production, and environmental conditions. The shape of the disrupted PSD may reveal key aspects of the suspended material resulting from geographic or seasonal changes in the type.

Table 1. Ambient water properties.

| Site | Date          | Lat (°N) | Lon (°W) | T (°C) | S (psu) | NFU | c₅₆₇₀ (m⁻¹) | Chl σ (mg m⁻³) |
|------|---------------|----------|----------|--------|--------|-----|--------------|----------------|
|      |               |          |          | x      | σ      | x   | σ            | x              | σ             |
| SR   | 27 March 2019 | 38.9832  | 76.4792  | 7.99   | 0.45   | 4.42| 0.02         | 6.92           | 0.17          | 3.55          | 0.03          |
| NCC  | 10 October 2017| 36.1832  | 75.7470  | 22.40  | 0.10   | 30.25| 0.10         | —              | 2.22          | 0.31          | 2.40          | 0.10          |
| LIS  | 12 June 2018  | 41.2899  | 72.0384  | 14.25  | 0.08   | 30.20| 0.03         | 1.16           | 0.57          | 1.41          | 0.14          |

Fig. 8. Change in clay particle volume distribution referenced to the minimum mean volume observed in each size bin; Kaolinite (KGa-1b, left panel) and Montmorillonite (SWy-2, right panel).
and size of the primary particles and the propensity for aggregate formation. Unlike the clay experiments, $V_p$ within the natural environments spanned the entire LISST size range and beyond. This was most evident within the NCC and LIS data sets where $V_p$ measured with laminar flow, and therefore most representative of ambient conditions, generally increased with particle size throughout the LISST range. This suggests the presence of ambient particles larger than the LISST measurement range and, therefore, the possibility that particle mass could have been added to the LISST range from the disruption of larger aggregates as measurements at higher flow rate progressed. NCC particle volume, for example, increased within size bins $> 49 \mu m$ at the maximum dissipation rate. This would suggest that floccs larger than the LISST size range had disrupted and the fragmented particles shifted mass into the larger LISST size bins. An alternate or perhaps additional explanation may be that a different water mass with larger particles had advected past the iDAS during the end of the measurement sequence. The ambient attenuation measurements suggest a small but abrupt decrease at 22-min elapsed time followed by greater variability and a gradual increase throughout the remaining time series. Simultaneously, Chl $a$ concentration (not shown) increased slightly from approximately 1 to 1.5 mg m$^{-3}$ and followed by an increase in variability. At 65 min elapsed time, representing the highest iDAS turbulent energy and the appearance of the large particle tail, there was an abrupt increase in $c_p$ by approximately 0.5 m$^{-1}$ and simultaneously a decrease in Chl $a$ concentration by approximately 0.5 mg m$^{-3}$.

**Discussion**

The iDAS appears to measure the disruption of particle aggregations subjected to turbulent energy. Disruption increases with $\varepsilon$ and results in a shift in the PSD toward smaller particles. The LISST has been shown in laboratory experiments to accurately retrieve the size distribution of natural sediments when compared to traditional sieving, filtering, and weighing techniques (Traykovski et al. 1999) and has been used successfully to investigate aggregate formation and disruption of natural particle suspensions when subjected to qualitative changes in turbulent shear (Slade et al. 2011). The primary advantage of iDAS is that turbulent shear can be
controlled and quantified and, therefore, can be compared directly to observed particle dynamics.

Turbulence is generated by flow through the sample tube. We chose the iDAS tubing dimensions to ensure that the flow had sufficient length to become fully developed (~ 50 d), that all particles were exposed to the maximum amount of shear in the fully developed turbulent flow, and that the tubing was manageable for deployments. Maximum turbulent shear occurs near the pipe walls. Bäbler et al. (2015) numerically investigated aggregate disruption in turbulent channel flow by placing particles at different starting locations in the channel and determining the location of disruption. They found that an aggregate starting at the channel centerline was typically transported to the high-shear near-wall regions of the channel in approximately 100 Kolmogorov timescales ($\sqrt{\nu / \varepsilon}$). In the current iDAS design, the tube dimension ensures that all particles are exposed to fully developed turbulence for a minimum of 700 Kolmogorov timescales after the flow reaches fully developed conditions at ~ 50 d. This length is likely more than needed, but we chose to be conservative with this proof of concept. Regardless, we recognize that the current iDAS sample tube dimensions produce turbulence levels in the upper range of oceanic conditions and that further research directed at optimizing tube dimension and flow rate is necessary if lower turbulence ranges are desirable.

A potentially significant drawback to the iDAS approach when deployed in natural environments is instability in the ambient particle population during the measurement sequence. Measuring 4 different turbulence levels across the thruster range, each requiring 10 min of sample chamber flushing and PSD measurement spans 40 min. Stability can, as discussed, be monitored by the LISST c time series representing laminar flow and any additional in situ sensors deployed simultaneously with iDAS. In addition to monitoring the ambient particle population, instabilities could be more effectively avoided by decreasing the required measurement time. This could be achieved with a smaller sample chamber requiring less flushing time, adjusting the flushing time as a function of flow rate, and fewer turbulence steps within the range of interest.

In addition to required stability in the particle population under investigation, care must be taken to avoid large gradients in water density. Schlieren, for example, effects near-forward light scatter due to small-scale fluctuations in water density and has been shown to impact LISST-100X measurements, both beam attenuation and PSD inversions, made within strong density gradients, such as a pycnocline, where the buoyancy frequency is greater than 0.025 s$^{-1}$ (Mikkelsen et al. 2008). Thus, iDAS deployments should be conducted with knowledge of the vertical density structure of the water column and, if possible, co-located with simultaneous measurements of water temperature and salinity in order to avoid strong density gradients.

When compared with microscopy and in situ holographic imaging techniques, the LISST inversion for PSD appears to retrieve volume attributed to individual particles comprising aggregates in addition to the volume of aggregated particles (Graham et al. 2012; Rau et al. 2018). In essence, a portion of the volume of individual particles comprising aggregates may be double counted, appearing within the PSD as particle volume within the aggregated size range and additional volume within the size range representing the individual primary particles. Nonetheless, we observed a general increase in primary particle volume upon aggregate disruption indicating that at least a portion of the aggregated primary particle volume was not detected. The increase in primary particle volume upon floc disruption may, therefore, represent volume attributed to self-shading of aggregated primary particles that do not contribute to the forward scatter measured by the LISST. Comparing the volume decrease in large size bins to volume increase in smaller size bins may, therefore, have the effect of overestimating the apparent interstitial space within flocs since the volume of disrupted material added to the primary particle size range will be depressed. Given that interstitial fluid contributes to the computations of apparent floc density and sinking rate (Burd and Jackson 2009), an underestimate of primary particle volume change will result in an underestimate of floc apparent density and, consequently, sinking rate. On the other hand, it may be possible to interpret the relative change in primary particle volume with floc disruption as the degree of self-shading, yielding more accurate estimates of interstitial space. This will require future research based on more detailed interpretation of the PSD resulting from light diffraction inversion.

iDAS is uniquely capable of providing in situ observations of natural particles that can be used to identify the size range of aggregates prone to disruption at specified levels of $\varepsilon$. The imposed stresses on these aggregates by the turbulent flow field can be determined through turbulence theory and assumptions about aggregate disruption mechanisms, that is, how disrupted mass is redistributed within the PSD (e.g., Parker et al. 1972; Bäbler et al. 2008). With the development of these concepts, iDAS has the potential to quantify the bonding strength between particles within natural suspensions and advance our knowledge of particle dynamics within aquatic environments.

It should also be noted that the LISST-100X is no longer offered by Sequoia Scientific as a standard product and has been replaced with a slightly smaller version, the LISST-200X. As with the LISST-100X, the new instrument can be equipped with a calibration chamber that slips on and surrounds the measurement end of the housing. Thus, an iDAS design could be easily adapted to the LISST-200X using the associated calibration chamber. However, there is no reason why the iDAS approach could not be applied to other particle imaging...
approaches, including laser sheet (Davies et al. 2017; Mar-
Kussens et al. 2020) and holographic (Many et al. 2019) approaches.

**Comments and recommendations**

We have described a new in situ instrument, iDAS, for mea-
suring the disruption of naturally occurring marine aggregates. The greatest value of the instrument is the ability to determine through direct measurements the change in the PSD as a function of ϵ. We believe that such data will eventually lead to better estimates of floc strength and, therefore, more accurate marine particle dynamics models with application to problems in sediment transport and upper ocean carbon flux. While the approach shows promise in this regard, issues of environmental
stability during observations and the disruption of flocs larger than the LISST measurement size range can potentially confound accurate interpretation of the data. At a minimum, however, the iDAS should prove useful in determining the aggregation state of natural waters as a function of ecological condition, such as particle concentration and type and the biochemical state of microbial communities.

The current iDAS design is preliminary intended as a proof of concept. Deployment is limited by the time required to collect valid data, the need for manual operation, and the fact that the power source and controller are located above water. Future enhancements may include a smaller, more rapidly flushing sample chamber and automated thruster motor control based on signal stability to minimize sampling time at each flow rate and reduce the likelihood of encountering unacceptable environmental variability. Submersible batteries could replace the topside power source and a programmable controller, such as a Raspberry Pi, BeagleBone, or Arduino UNO, could monitor the LISST signals, define the optimum flushing time for each flow rate (i.e., turbulence level), and step the ESC controller through the range of pre-determined flow rates. Such improvements would allow deployment to deeper depths and potentially enable a profiling capability. The resulting observations would yield depth-dependent changes in PSD and aggregation state resulting in better estimates of vertical particle export efficiency. Additionally, a second, free-flowing LISST could be positioned alongside of the iDAS as an ambient PSD reference with which to compare the disrupted particle population.

iDAS should always be deployed with additional environ-
mental sensors to monitor ambient conditions during iDAS operation and aid in interpretation of the data. At a minimum, this should include temperature and salinity in order to avoid potential Schlieren effects and a characterization of the type of particles present, such as phytoplankton and inorganic particles. At a minimum, this would avoid having to rely upon laminar flow measurements to assess environmental stability.

Better knowledge of aggregate bonding strength, disruption and associated changes in PSD is expected to lead to more accurate models of marine particle dynamics. We believe that the iDAS is an important step in this direction. While this remains the ultimate goal of iDAS development, refinements to the instrument and more effective data interpretation await future research.

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Acknowledgment
We wish to thank the U.S. Naval Research Laboratory and the Pennsylvania State University for institutional funding to support this research, Dr. Joseph Smith, U.S. Naval Academy, for providing logistical support of observations within the Severn River, and Dr. James O’Donnell, University of Connecticut, and the crew of the R/V Connecticut for providing us the opportunity to collect observations in Long Island Sound.

Conflict of Interest
None declared.