Size distributions and fractal properties of particles during a simulated phytoplankton bloom in a mesocosm

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(Received 12 August 1994; in revised form 15 December 1994; accepted 5 January 1995)

Abstract—Marine snow aggregates (>500 μm) composed primarily of phytoplankton are known to have fractal dimensions of 1.52 to 1.72 and are assumed to form by the physical coagulation of smaller particles. In order to study the bulk fractal properties of these smaller particles during a coagulation event, concentrations of small particles (2 to 300 μm) were measured during a simulated phytoplankton bloom in a laboratory mesocosm. Particle concentrations were presented as size distributions in terms of either solid volume or average length. Both distributions indicated that particles were continuously coagulating throughout the bloom as evidenced by a greater increase in the concentration of larger particles (50 to 300 μm in average length) than smaller particles (2 to 50 μm), and decreases in fractal dimensions. Average fractal dimensions of all particles in the size range 20–200 μm were calculated using a new method called the particle concentration technique (PCT). The PCT required both solid volume and length size distributions on the same population of particles as input. As coagulation of phytoplankton and other particles in the tank proceeded during the phytoplankton bloom (7 to 11 days after inoculation), the average fractal dimensions of these particles decreased from \( D = 2.49 \pm 0.41 \), a value close to the Euclidean value of 3 for a sphere, to \( D = 1.68 \pm 0.08 \), a value typical of larger marine snow aggregates. This suggests that although marine snow-sized aggregates can appear to be dominated by non-fractal particles such as phytoplankton, they are primarily formed from many types of smaller aggregates present in the water column that, on average, can have low fractal dimensions.

INTRODUCTION

Coagulation is an important mechanism of particle removal in natural systems since this process can transform many small, slowly settling particles into larger, faster settling aggregates. Mathematical descriptions of aggregates and coagulation processes have been totally revised in the last decade since aggregates formed by coagulation have fractal structures (Witten and Cates, 1986; Meakin, 1988; Logan and Wilkinson, 1990). Aggregate properties such as mass and solid volume do not scale with size raised to an integer value of 3, but to fractional powers called fractal dimensions, that can be considerably less than 3.

Aggregates formed by Brownian motion have fractal dimensions that are solely a function of particle stickiness (Meakin, 1988; Lin et al., 1989; Torres et al., 1990).

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Highly destabilized (very sticky) particles that undergo fast cluster–cluster aggregation by Brownian motion yield fractal dimensions of approximately 1.8 (Jullien et al., 1984; Schaefer et al., 1984; Schonauer and Kreibig, 1985). Less destabilized particles that must collide many times before adhesion, produce aggregates with fractal dimensions between 1.9 and 2.2 (Jullien and Kolb, 1984; Weitz and Oliveria, 1984; Meakin and Family, 1987). No such universality in the magnitude of the fractal dimension, however, has been established by experimental observations for other coagulation mechanisms such as shear motion and differential sedimentation. Fractal dimensions ranging from 1.0 to 3.0 have been reported for aggregates of different types of particles formed by either shear motion or differential sedimentation (Li and Ganczarczyk, 1989a; Logan and Wilkinson, 1990, 1991; Jiang, 1993).

In marine systems, the fractal nature of marine snow has been established from measurements of aggregate properties such as settling velocity and porosity. Logan and Wilkinson (1990) calculated fractal dimensions of 1.39 ± 0.15 for all types of marine aggregates (0.4–20 mm) considered as a single class of aggregates, and 1.52 ± 0.19 for large diatom aggregates (7–20 mm). Using in-situ photographs, Kilps et al. (1994) calculated a fractal dimension of 1.72 for marine snow particles (1–60 mm) by assuming that two- and three-dimensional fractal dimensions were equal.

Although seawater contains significant concentrations of smaller aggregated particles, defined here as microaggregates (<0.5 mm), previous research has not directly considered the fractal dimensions of these microaggregates. Marine snow-like aggregates can be generated from smaller particles in rolling cylinders as described by Shanks and Edmondson (1989). Laboratory studies on fluorescent microsphere aggregates generated in this device demonstrated that fractal dimensions of small aggregates (4–20 μm in length) were in the neighborhood of 1.6 (Logan and Kilps, 1995). Fractal dimensions of smaller marine particles, such as phytoplankton-dominated microaggregates formed during phytoplankton blooms, however, have not been reported.

Particle size distributions have been used for more than three decades as tools to describe coagulation processes. The average shapes of particle size distributions in the ocean have been found to agree with predictions based on a coagulation model assuming steady state size distributions (Hunt, 1980; McCave, 1984). Data used to test these models have primarily been collected using electronic particle counters (Sheldon et al., 1972; McCave, 1983; Longhurst et al., 1992), although others have used microscopic analysis or camera systems (Harris, 1977; Lambert et al., 1981; Gardner et al., 1991). In most cases these observations provided basic information of size distributions in the ocean where the systems were at nearly steady conditions. However, these measurements have not considered particle size distributions under more dynamic conditions, for example during a phytoplankton bloom.

In this study, a phytoplankton bloom was simulated in a mesocosm under well controlled conditions. We investigated changes in concentrations of particles less than 300 μm in length by measuring size distributions using both a resistance particle counter and an image analysis system. Since aggregates formed by coagulation have fractal dimensions less than 3, we measured fractal dimensions of particles in the size range of 20 to 200 μm to see if they were within the range expected for aggregates formed by coagulation. Fractal dimensions were calculated using a new method, called the particle concentration technique (PCT), that required two measurements of particle sizes for the same population of particles. It was demonstrated that as phytoplankton increased in concentration,
average fractal dimension of all particles became lower, eventually attaining a value of \~1.7.

MATERIALS AND METHODS

Mesocosm

The coagulation experiment was conducted in a cylindrical fiberglass tank (1400 liters) in a temperature-controlled (12.5–13°C) environmental chamber as described by Alldredge et al. (1995). The tank was filled with 1150 liters of seawater from the Santa Barbara Channel, California, which had been filtered through a subsand filter. Fifty liters of unfiltered seawater were added as an inoculum for the tank. Artificial light was set at 14 h light: 10 h dark cycle, and illuminated at 1500 \( \mu \text{E} \text{m}^{-1} \text{s}^{-1} \) (surface), decreasing to 87 \( \mu \text{E} \text{m}^{-1} \text{s}^{-1} \) 10 cm above the bottom. Energy dissipation rates increased from 0.0137 to 0.0206 cm\(^2\) s\(^{-3}\) as water samples were withdrawn from the tank (Alldredge et al., 1995), resulting in an average shear rate of 1.3 s\(^{-1}\) (range of 1.21 to 1.50 s\(^{-1}\)). The tank experiment began on 6 March 1993 (Day 0). Bulk 20 liter samples were obtained from the tank everyday at 9.00 am through a spigot located 30 cm above the tank bottom.

Particle sizing and counting techniques

Particle size distributions. Particle sizes in natural waters can be described using a size distribution function \( n(L) \), defined by \( dN = n(L) dL \), where \( dN \) is the number of particles per unit water volume with size in the range \( L \) to \( L + dL \) (Twomey, 1977; Hunt, 1980). The characteristic length \( L \) can be any scale measure of size, e.g. volume, diameter, and maximum or average length. In practice, particle counts are often acquired in such a way that the cumulative size distribution, \( N(L) \), the number concentration of particles larger than a given size \( L \), is commonly used, where \( N(L) = \int_{L}^{\infty} n(L) dL \) (Twomey, 1977; McCave, 1984).

Previous research on seawater has shown that suspended particles follow a power law distribution function over a large portion of a size range (Sheldon et al., 1972; McCave, 1984), or

\[
N(L) = kL^b
\]  

where \( k \) and \( b \) are constants. The exponent \( b \), which is the slope of size distribution of log–log plot (log\((N(L))\) vs log\((L)\)), is frequently cited to describe the shape of the curve.

Volume distributions. An electronic particle counter (Coulter Multisizer II, Coulter Corp.) was used to measure particle size distributions, \( N(v_c) \), in terms of solid volume \( v_c \). These size distributions also were expressed as \( N(d_e) \), in terms of solid equivalent diameter \( d_e \), based on calibration with precise-sized latex microspheres. This instrument can be used to count and size porous particles on the basis of changes in resistance caused by the passage of particulate matter across a small aperture, because the signal pulse from a particle passing through the aperture has been found to be proportional to the solid volume of the porous particle (Trewack and Morgan, 1977; Jackson et al., 1995). In this study, samples were analyzed using two apertures, 50 and 100 \( \mu \text{m} \), providing size ranges of
2 < d_s < 30 \mu m and 4 < d_s < 60 \mu m. Sample volumes were 0.1 and 0.5 ml for the 50 and 100 
\mu m apertures, respectively. Seawater (\textasciitilde 33\% salinity) from the tank was filtered through
carbonate filters (0.2 \mu m; Poretics Corp.) and used as electrolyte. Data were
transferred from the Coulter counter to a 80486 personal computer and analyzed using
their software (AccuComp, Version 1.14, Coulter Corp.). Size distributions from the two
orifices were combined into a single size distribution by overlaying the distributions and
using data for 2 < d_s < 9.5 \mu m from the 50 \mu m orifice and 9.5 < d_s < 60 \mu m from the 100 \mu m
orifice. Reported results are the average of three measurements. Sample volume used for
analysis produced a minimum measurable particle concentration of 2 ml^{-1} at the largest
particle sizes for each orifice.

\textit{Length distributions.} An image analysis system was used to generate particle size
distributions, N(l), in terms of average length l. This system employs a computerized
optical particle sizing and counting technique capable of transforming a gray level
microscopic image obtained through a camera into a binary image. The image analyzer
CUE II (Olympus Corp.) used in this study consisted of four components: a microscope
(BH-2, Olympus Corp.), a CCD-video camera (XC-57, SONY) with a resolution of 510 ×
492 pixels, an image monitor (Trinitron, SONY), and a 80486 personal computer.
Computer software (Cue-2, Version 3.0, Olympus Corp.) was loaded into the computer
and used to analyze the image. The Ferret's average length of particles, obtained by
measuring the particle length at four different angles (0°, 45°, 90° and 135°) was calculated
by the system. All measurements were made at 400× and based on the analysis of 100
fields corresponding to a measurable size of 3–300 \mu m and a minimum detectable particle
concentration of 10 ml\(^{-1}\) for the largest particles.

To prepare samples for the image analysis system, tank samples (5 ml) were filtered onto
a 0.2 \mu m black polycarbonate membrane filter (25 mm diameter, Poretics Corp.) and
stained in the funnel with acridine orange at a high concentration (0.1\%) in order to stain
all particles in the sample. After 5 min of staining, the sample was filtered through the
membrane by vacuum at 2.5 psi (the lowest vacuum for our system). An additional backing
filter (25 mm diameter, 5 \mu m, Millipore Corp.) was placed behind the membrane to
distribute particles on the membrane evenly. The dry membrane filter was placed on a
glass slide with one drop of immersion oil on the top and covered by a cover glass. All
samples were prepared in duplicate. The slides were stored at 4°C and analyzed under blue
light.

\textit{Relationship between length and solid equivalent diameter.} Since solid spheres are used
in the Coulter counter calibration procedure to relate particle volume and size, porous
objects have a greater size (length) than implied by measurements of solid equivalent
diameter (d_s). As a result, the optical length l determined using the image analyzer is
greater than d_s produced by the Coulter counter.

By comparing the properties of fractal and Euclidean objects, it can be seen that for
fractal aggregates l varies exponentially with d_s. Assuming that primary particles have
similar size or volume, the total solid volume of primary particles within an aggregate, v_s,
can be related to the aggregate length scale l using the fractal geometry expression,

\[ v_s = a_f l^D \]  \hspace{1cm} (2)

where \( a_f \) is a constant (Jiang and Logan, 1991).
According to a Euclidean description of a porous aggregate, the solid volume of an aggregate is

$$v_s = a_e d_s^3$$  \( (3) \)

where $a_e$ is a constant ($a_e = \pi/6$ for a spherical object). The solid volume can be related to the encased volume by the porosity $p$, as

$$v_s = (1 - p) v_e$$  \( (4) \)

where $v_e = a_e l^3$ is the encased volume of the aggregate of actual size $l$ (Logan and Wilkinson, 1991). Combining equations (3) and (4), the relationship between $l$ and $d_s$ becomes

$$l = b_e d_s$$  \( (5) \)

where $b_e = (1 - p)^{-1/3}$. Since porosity does not vary with particle size for Euclidean objects, $b_e$ is a constant and particle length is a linear function of solid equivalent diameter.

For fractal objects, $D$ is less than 3 and porosity is no longer constant due to the heterogeneous porous structure of fractal aggregates. Combining equations (2) and (3), the correlation between $l$ and $d_s$ can be obtained as

$$l = b_f d_s^{3/D}$$  \( (6) \)

where $b_f = (a_e/a_f)^{3/D}$ is a constant that is a function of characteristics of aggregates including fractal dimension, packing factor, shape coefficient, and size of primary particles (Jiang and Logan, 1991). Since $l$ shows a power law relationship with $d_s$, where $3/D > 1$, the length of aggregates increase with $d_s$ much faster than predicted from the Euclidean-based relationship [equation (5)]. Therefore, the particle size represented by $d_s$ obtained from the Coulter counter will be much less than $l$ measured using the image analyzer as aggregates become larger. For example, assuming $b_e = 2$, $b_f = 1.5$ and $D = 2$, at $d_s = 20 \mu m$ the average length is $40 \mu m$ for a Euclidean object while it is $134 \mu m$ for fractal objects (Fig. 1). When $d_s$ is doubled to $40 \mu m$, the corresponding average length will increase by only
100% to 80 μm for Euclidean objects, but will increase by 184% to 380 μm for fractal objects.

**Calculation of fractal dimensions**

Jiang and Logan (1991) were able to calculate fractal dimensions from a single size distribution using dimensional analysis, but their method was dependent on the assumption that the size distribution was at steady-state. To overcome this limitation, they later proposed a new non-steady-state method that required the analysis of size distributions both in terms of \( v_s \) and \( l \) (Jiang and Logan, submitted). Their method, called the two-slope method, did not require an assumption of steady-state conditions but did require that both size distributions be able to be described by a single power law distribution over the size range of interest.

In the present study, a modified approach is proposed that does not require a single power law function for a particle size distribution. The technique, referred to here as particle concentration technique (PCT), defines the correlation between solid volume and length directly on the basis of cumulative particle concentrations from the two types of size distributions \( N(v_s) \) and \( N(l) \). An assumption is made that particles with solid volume greater than \( v_s \) are the same population of particles with length greater than \( l \), if the cumulative particle counts in terms of solid volume and length are equal, or

\[
N(v_s) = N(l)
\] (7)

In other words if particle concentrations produced by the Coulter counter and the image analyzer are the same, then that population of particles must have a relationship between the solid volume \( v_s \) and average length \( l \) defined by equation (2). The two unknowns \( a_f \) and \( D \) can be calculated from a linear regression of matched \( v_s \) and \( l \) data by linearizing equation (2) using a log–log transformation, or

\[
\log(v_s) = \log(a_f) + D \log(l)
\] (8)

A series of values of \( v_s \) versus \( l \) were obtained from the two size distributions, \( N(v_s) \) and \( N(l) \) at solid volume intervals of \( \Delta \log(d_f) = 0.05 \) (the bin sizes on the Coulter counter). The first value of \( v_s \) was defined by the first concentration of particles for which \( N(v_s) = N(l) \), and the last value was defined by the lowest detectable concentration for either instrument. The slope of the regression line of \( \log(v_s) \) vs \( \log(l) \) was the fractal dimension \( D \).

**RESULTS**

**Particle size distributions**

Particle size distributions, measured in terms of solid equivalent diameter (2–50 μm) with the Coulter counter, indicated that particle concentrations increased between Days 7 and 11 [Fig. 2(a)]. Concentrations of larger particles (\( d_i > 20 \mu m \)) increased faster than those of smaller particles. For example, the concentration of particles greater than 2 μm increased less than a factor of 3 (25,000 ml⁻¹, Day 7 to 65,000 ml⁻¹, Day 11), while the concentration of particles greater than 20 μm increased more than one magnitude from 56 ml⁻¹ to 846 ml⁻¹ (Table 1). After Day 11, size distributions did not show any appreciable
changes in shape, indicating particle size distributions in the tank system were in a stationary, if not steady, state.

Although particle concentrations based on solid equivalent diameter indicated that the size distributions could be fitted by a power law function [Fig. 2(a)], the slopes of size distributions gradually increased from −2.70 on Day 7 to −2.18 on Day 11 (Table 1). This change suggested that the percentage of larger particles increased with time and that the system was not at steady-state.

Size distribution slopes for smaller particles (<9.5 µm) measured with only the 100 µm orifice were different than slopes for the same particle sizes measured using the 50 µm orifice (data not shown). On Days 7 and 8, these slopes from 100 µm measurement were also inconsistent with the data obtained by others using a single orifice with an Elzone
Table 1. Summary of particle counting. Particle sizes represented by solid equivalent diameter are much smaller than their average length due to the fractal nature of aggregates

<table>
<thead>
<tr>
<th>Time (day)</th>
<th>Coulter Measurement</th>
<th>Image Analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Particle Counts (ml⁻¹)</td>
<td>Slope</td>
</tr>
<tr>
<td>d, &lt; 50 (µm)</td>
<td>&gt;2</td>
<td>&gt;4</td>
</tr>
<tr>
<td>7</td>
<td>25058</td>
<td>5792</td>
</tr>
<tr>
<td>8</td>
<td>34710</td>
<td>11866</td>
</tr>
<tr>
<td>9</td>
<td>44634</td>
<td>17087</td>
</tr>
<tr>
<td>10</td>
<td>53435</td>
<td>23379</td>
</tr>
<tr>
<td>11</td>
<td>64600</td>
<td>25538</td>
</tr>
<tr>
<td>12</td>
<td>64587</td>
<td>25570</td>
</tr>
<tr>
<td>13</td>
<td>60541</td>
<td>23812</td>
</tr>
</tbody>
</table>

As described in the Methods section, however, for each example we overlaid the two size distributions measured using the 50 and 100 µm orifices at 9.5 µm to produce a single daily size distribution, and consequently this combined size distribution, rather than the distribution from 100 µm orifice measurement, was reported in this study. As a result, the combined size distributions are consistent with other size spectrum collected for these days (Jackson et al., 1995).

The size distributions provided by the image analysis system [Fig. 2(b)] exhibited the same general pattern with time as those obtained from the Coulter counter. The concentrations of larger particles increased much faster than smaller ones, causing the slopes to increase from b = -1.77 to -1.20 (Table 1), and after Day 11 also indicated an essentially stationary size distribution. It should be noted that the size scale represented by average length given by the image analyzer did not directly match solid equivalent diameter given by the Coulter counter due to the fractal nature of the aggregates. As previously discussed, the average length of fractal aggregates can be much larger than their solid equivalent diameter as aggregates become bigger, since the length scale increases exponentially with the solid equivalent diameter. Consequently, the slopes of the size distributions from the two methods of measurement were found to be much different from each other.

Particle volume and mass

Although only a small fraction of the complete size distributions of all particles in the tank was measured, increases in the total solid particle volume measured using the Coulter counter reflected changes in biomass concentrations in the tank. Based on the data of Coulter particle counting in the d, range 2–50 µm, an exponential growth phase was found from Day 7 to Day 10 with maximum specific growth rate of 0.7 day⁻¹ (Fig. 3). By Day 11, the total particle volume in this size range was constant. A comparison with total dry mass analysis over this same period by Allredge et al. (1994) demonstrates that the two curves represented by different parameters were remarkably similar.
Fractal dimensions

From log–log regressions using equation (8), fractal dimensions were determined for particles between 20 and 200 μm based on average particle length. Fractal dimensions decreased with time, from $2.49 \pm 0.41$ on Day 7 to $1.68 \pm 0.08$ on Day 12 (Fig. 4), indicating the average porosity of all particles became higher. Because aggregates
normally have lower fractal dimensions than non-coagulated single particles. The change in fractal dimension also reflected that the proportion of aggregates increased with time in the tank. After Day 12, the fractal dimension did not demonstrate any significant change. The magnitude of the final value of $D = 1.68$ was close to the fractal dimensions of marine snow previously reported (Logan and Wilkinson, 1990).

**DISCUSSION**

Previous field investigations have suggested that aggregation commences at the termination of a phytoplankton bloom when nutrients are depleted (Krank and Milligan, 1988; Alldredge and Gotschalk, 1989). However, our measurements demonstrate that the formation of microaggregates is not closely linked to nutrient limitation. From Day 8 to Day 11, the concentration of particles $>100 \, \mu m$ (average length), many of which were phytoplankton aggregates, increased from undetectable levels ($<10 \, ml^{-1}$) to $487 \, ml^{-1}$, while nutrients were still relatively enriched (Alldredge et al., 1995). The decreasing values of $D$ measured throughout the study also support our observations on continuous aggregation in the tank. These results suggest that although large concentrations of marine snow-sized aggregates may be observed after loss of nutrients following a phytoplankton bloom, significant amounts of microaggregates can be produced prior to nutrient depletion. This finding in our mesocosm study agrees with previous observations of a natural diatom bloom in the North Sea where microaggregate ($<0.5 \, mm$) formation occurred prior to nutrient limitations (Riebesell, 1991).

Using a dimensional analysis, Hunt (1980) predicted that, under shear-dominated conditions, particle size distributions produced by coagulation would have a slope of $-3$, in terms of solid volume, at steady-state. His analysis required the assumption of a dynamic steady-state, implying that there would be a continuous source of primary particles that coagulated through the size distribution to form aggregates removed through sedimentation. This predication was supported by slopes in the range of $-3 (-2.7 \, to \, -3.4)$ of microaggregate-sized marine particles assumed to have been formed by shear coagulation (Hunt, 1982). However, slopes of $-2.2 \, to \, -2.7$ in our mesocosm suggest that Hunt’s approach is inappropriate for our study. The fractal nature of aggregates formed in the tank accounted for part of the difference between the measured values and the predicated slopes (Jiang and Logan, 1991). Nevertheless, the disagreement between this experiment and the prediction was more likely due to the fact that the experimental mesocosm never achieved steady-state, and that particles could not settle, two conditions that violated critical assumptions of the dimensional analysis.

Changes in particle size distributions measured during Days 7 to 13 in the mesocosm reflected the total production of phytoplankton and their aggregation in the mesocosm, but not all particles measured in the tank were associated with phytoplankton. According to either solid volume or dry mass, total biomass grew from Day 7 to Day 11 with a doubling time of approximately 1 day (Fig. 3). Although seawater particles introduced into the system were assumed to be primarily single particles mainly in the form of phytoplankton, by Day 7 many of the particles in the size range measured in this study were not phytoplankton or even associated with phytoplankton. The abundance of non-phytoplankton particles can be seen by comparing the size distribution of phytoplankton particles reported by Alldredge et al. (1995), which include individual cells, chains, and phytoplankton in aggregates, with total particle concentrations measured with our image
Fig. 5. Abundance of phytoplankton (data from Alldredge et al., 1995) and non-phytoplankton particles (image analyzer data from this study) in different size classes on Days 7 and 12.

analysis system (Fig. 5). Most of the larger particles (36 to 288 μm) were enriched with phytoplankton, but there was a large fraction of smaller particles (2 to 36 μm) that were not phytoplankton. By the end of the bloom on Day 12, all particle concentrations had increased but only 10 to 25% of particles in each of the 3 smaller size classes (<36 μm) were enriched in phytoplankton. This indicated a large fraction of particles in the tank were something other than phytoplankton. These non-phytoplankton particles likely consisted of bacteria and material that is usually unidentifiable and therefore classified as detritus. Non-phytoplankton particles also likely included TEP particles that have been found to be important in driving coagulation in the mesocosm and other systems (Logan et al., 1995; Passow and Alldredge, 1995). Since high concentrations of acridine orange were used to stain all particles, TEP particles were included in total image analysis particle counts due to the absorption of acridine orange by TEP and other particles enmeshed in TEP.

Fractal dimensions

Analysis of fractal dimensions demonstrated that microaggregate-sized particles (20–200 μm) in the mesocosm were fractal. However, the magnitude of fractal dimension decreased significantly from 2.49 on Day 7 to 1.68 on Day 12 (Fig. 4). Although several tank characteristics such as shear rate and particle concentrations changed with time, and this might have produced some variation in fractal dimensions, a more likely explanation for the decrease in D is that the fractal dimensions reported in this work describe the average properties of all particles in the tank and not just aggregates. Since it was impossible using our methods to distinguish aggregates from other non-coagulated particles in the water column, some particles analyzed in the size range were not fractal aggregates, but solid primary particles with a Euclidean structure (D = 3). The inoculum used for the mesocosm likely contained few aggregates and by Day 7 the average of
$D = 2.49$ was only slightly less than 3. As coagulation proceeded in the tank, the coagulation of particles in the tank increased the proportion of fractal aggregates causing the average fractal dimension of all particles to decrease. Therefore, the overall fractal dimension not only described the fractal nature of the particles, but also indirectly provided an indication of the extent of coagulation in the tank.

The PCT approach used here to calculate fractal dimensions does not directly measure a fractal dimension for a single particle. Instead, it is a fitting parameter that resolves observed properties (size and mass) of the whole ensemble of suspended particles. It is similar to the approach used by Jackson et al. (1995) in that both methods use $D$ to resolve differences between particle size and mass spectra in the tank at any given time. It is different, however, in that Jackson and co workers compared particle spectra of different populations (sizes) of particles, while in this study the same population of particles was studied using two different particle counting techniques. This difference in approaches probably accounts for differences in fractal dimensions between the two studies.

The final fractal dimension of 1.68 calculated in this study at the end of the phytoplankton bloom is close to values previously reported for much larger particles of marine snow. Logan and Wilkinson (1990) calculated $D = 1.39 \pm 0.15$ from size and porosity data and $D = 1.26 \pm 0.06$ from settling velocity data for general marine snow, and $D = 1.52 \pm 0.19$ from size and porosity data for diatom-typed marine snow. Several factors contribute to differences between fractal dimensions determined in this and previous studies. First, particle sizes ($<200 \, \mu m$) examined here are smaller than the size range of marine snow ($>500 \, \mu m$), and other investigators have noticed that fractal dimension of aggregates produced by shear coagulation may vary considerably with size (Li and Ganczarczyk, 1989b; Jiang and Logan, submitted). Second, previous calculations of fractal dimensions from settling velocity data may have underestimated fractal dimensions (Kilps et al., 1994). Logan and Wilkinson (1990) made an assumption in their calculations that aggregate settling velocities obeyed Stokes's Law. This assumption was recognized as a limitation, however, since drag coefficients could not be derived from in-situ settling velocities for marine snow aggregates (Alldredge and Gotschalk, 1988).

Another reason for differences in fractal dimensions in different studies is the definition of length scale. In this study average particle length was used, while some other studies have used maximum length. Due to the highly amorphous shapes of aggregates, fractal dimensions may have some variation with different types of length scales. The relationship between a fractal dimension based on average length $l_a$, defined as $D(l_a)$, and $D(l_m)$ based on maximum $l_m$, can be seen by taking the derivative of equation (8), either in terms of $l_a$ as

$$D(l_a) = \frac{d \log(v_s)}{d \log(l_a)}$$

or in terms of $l_m$ as

$$D(l_m) = \frac{d \log(v_s)}{d \log(l_m)}$$

From these two equations, we can write the equality

$$\frac{D(l_a)}{D(l_m)} = \frac{d \log(l_m)}{d \log(l_a)}$$
In this study, it was found that the slopes of log($l_m$) vs log($l_a$) for Days 8 to 13 varied from 1.01 to 1.17 with an average of 1.10, meaning $d \log(l_m)/d \log(l_a) = 1.1$. Therefore, fractal dimensions based on average length would be 10% lower if maximum length had been used.

In conclusion, the decrease of the average fractal dimension of particles 20 to 200 $\mu$m in length from 2.49 to 1.68 (between Days 7 and 12) indicates that, on average, particles in the tank became more fractal as coagulation proceeded in the tank. Although marine snow-sized aggregates can be classified as phytoplankton-marine snow as a result of a large number of phytoplankton cells in the aggregate, this study suggests that smaller particles are also coagulating and become progressively more fractal, eventually reaching values similar to that of marine snow. The inclusion of these smaller fractal particles into larger aggregates likely helps explain the low fractal dimensions of marine snow.

Acknowledgements—Funding was provided by ONR Grant N00014-91-J-1249.

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