



Fractal dimensions of small (15–200 μm) particles in Eastern Pacific coastal waters

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Abstract

Particles 3–300 μm (average length) in seawater include single cells, non-viable particles of identifiable origin (such as fecal pellets), aggregated particles formed from water column debris, and aggregated mixtures of all of these materials. While macroscopic marine snow-sized aggregates (>0.5 mm in average length) have been shown to be fractal, relatively less is known about the average characteristics of smaller particles. We calculated the fractal dimensions of microscopic particles 15–200 μm in length through simultaneous measurements of particle size distributions as a function of solid equivalent diameter (from solid volumes measured using a Coulter Counter) and average length (from image analysis of acridine-orange stained filtered particles). Particle size distributions were measured at two eastern Pacific coastal areas, one in Monterey Bay, CA, and the other in East Sound, WA. Average fractal dimensions of particles indicated that D was highest in East Sound ($D = 2.59 \pm 0.17$) during a phytoplankton bloom that did not appear to be aggregating, and lowest at one site in Monterey Bay ($D = 1.77 \pm 0.34$), where old diatom flocs and marine snow-size aggregates were observed. There was no direct relationship between D and total particle concentration, chlorophyll a , or transparent exopolymer particles (TEP) concentration, although the highest concentration of TEP was found at the site with the lowest fractal dimension. Particles with low fractal dimensions are produced through coagulation. Our subjective assessment of the importance of aggregate formation at these sites, based on diving and microscopic observations, indicated that aggregates were more abundant at sites where particles had lower fractal dimensions. Thus, we attribute the low fractal dimensions of these small particles to be the result of their formation through coagulation processes. © 1998 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Large aggregates (>0.5 mm) in marine systems, collectively known as marine snow, are classified according to the predominant particle type in the aggregate (such as diatoms and fecal pellets) or are considered as unidentifiable aggregates of water column debris (Aldredge and Silver, 1988). Numerous studies have demonstrated that marine snow-size particles are fractal. By measuring aggregate properties such as size and porosity, three dimensional fractal dimensions, D , of marine snow were found to range from 1.39 ± 0.15 (all types of marine snow) to 1.52 ± 0.19 (diatom aggregates) (Logan and Wilkinson, 1990). When $D < 2$ two dimensional images can be used to calculate D . This is possible because the fractal dimension of the three-dimensional object imbedded in two-dimensions, D_2 , is equal to D when $D < 2$, but when $D \geq 2$ then $D_2 = 2$ (Meakin, 1988). In situ photographs of marine snow aggregates 1–60 mm in length were calculated to range from 1.28 ± 0.11 for aggregates containing large amounts of miscellaneous debris to 1.86 ± 0.13 for large diatom aggregates; when combined into a single group, $D = 1.72 \pm 0.07$ (Kilps et al., 1994).

There is relatively little known about the fractal dimensions of particles smaller than marine snow, and therefore, there is no way to know whether the fractal dimensions of particles in marine systems are a function of particle size or not. Phytoplankton should have fractal dimensions close to three based on comparison to Euclidean objects such as cubes and spheres, but less than half of the particulate organic carbon in seawater may be associated with living materials (Sheldon et al., 1972). Average fractal dimensions of smaller ($< 300 \mu\text{m}$) particles in natural waters have been inferred, but not directly measured, from size distributions by different methods (Jiang and Logan, 1991; Li and Logan, 1995; Jackson et al., 1995). Fractal dimensions calculated using these methods vary over a relatively wide range of 1.5 to > 3 , and it is not clear if it is the method or characteristics of the particles that cause this wide variation in D . Early estimates of D made from size distributions reported in the literature for particles $> 2 \mu\text{m}$ produced estimates of D from 1.61 to 3.75 based on the assumption that marine systems could be analyzed as steady state size distributions (Jiang and Logan, 1991). D values larger than 3 are physically unrealistic, and values of $D < 3$ have been consistently obtained in more recent studies using different techniques. During a phytoplankton bloom in a laboratory mesocosm a fractal dimension of 2.3 was derived by matching the size spectra obtained with particle counters for smaller (20–300 μm) particles with those for larger (~ 0.1 –10 mm) particles obtained using in situ photographic and imaging systems (Jackson et al., 1995). An analysis of particles over a more restrictive size range of 20–200 μm in the mesocosm, however, suggested that the average fractal dimensions of particles changed over time. Fractal dimensions decreased from 2.49 at the start of the experiment to 1.68 at the end of the experiment, coincident with the appearance of a large concentration ($> 10^3 \text{ l}^{-1}$) of marine snow sized particles (> 0.5 mm) (Li and Logan, 1995). The different results in these two experiments suggests that either one of these techniques is not accurate, or that fractal dimensions of natural marine particles may not be constant over wide particle size ranges (i.e. D for 10 μm -size particles is different from that of 10 mm size particles).

In the present study, we calculated the average fractal dimensions of naturally occurring small particles (size range of 15–200 μm) from particle size distributions using a new method called the particle concentration technique (PCT) (Li and Logan, 1995). The fractal dimensions of particles were determined on samples taken at various depths over several days at two eastern pacific coastal areas, one in Monterey Bay, CA, and the other in East Sound, WA. We compared our calculated values of D with subjective assessments of large aggregate (by divers) and smaller aggregate (3–300 μm) abundances to determine whether lower values of D (more fractal particles) were consistent with higher concentrations of amorphous particles. Aggregates can form in proportion to phytoplankton concentration (Jackson, 1990), but it has been shown in several systems (freshwater, seawater and mesocosms) that the rapid formation of diatom aggregates can also be driven by the coagulation of nearly invisible polysaccharide-rich particles called transparent exopolymer particles (TEP) (Alldredge et al., 1993; Passow et al., 1994). In order to interpret our results within the context of factors potentially affecting particle aggregation, we compared the average fractal dimensions of small particles to the concentrations of TEP, chlorophyll a , and total particles at these sites. These field investigations were conducted as a part of a multi-investigator program called SIGMA (Significant Interactions Governing Marine Aggregation) set up to study the development and fate of natural phytoplankton blooms.

2. Materials and methods

2.1. Study sites and sampling

Field investigations were conducted in two Pacific coastal areas, one in Monterey Bay, CA, and the other in East Sound, WA. In Monterey Bay, samples were taken from two sites, one ~ 2 miles from land (MB1) that was subject to considerable near-shore wave processes, and one 8 miles from land (MB2), which was relatively more of an open-water site. In East Sound (ES), all samples were taken at a single location in the center of a small bay (approximately 2 miles wide and 8 miles long) where the water was generally calm. The locations and dates of our field investigations and the descriptions of the three sampling sites are summarized in Table 1. An assessment of site conditions relative to aggregate abundance at these sites is based on in-situ observations made by experienced divers (A.L. Alldredge and C. Gotschalk, University of California, Santa Barbara, personal communication).

Bulk water samples were collected each morning (7–9 a.m.) at various depths using 30 l Niskin bottles deployed from the ships. Monterey Bay samples were analyzed immediately in the laboratory on board the RV Sproul. East Sound samples were brought back to the laboratory located on Friday Harbor Island and were analyzed within 1 h.

Concentrations of chlorophyll a in seawater were measured by standard fluorometric methods (Parsons et al., 1992) using a Turner Model 111 fluorometer. TEP concentrations were measured using a new spectrophotometric technique (Passow and Alldredge, 1995). Briefly, seawater samples were gently filtered, stained with

Table 1

Locations, dates and site depths of the field investigations, and qualitative assessment of aggregation at three different study sites

Site	Cruise date	Depth (m)	Aggregation assessment based on diver observations ^a
Monterey Bay–MB1 (36° 55' N, 121° 56' W)	July 23–28, 31, 1993	~ 25	High concentration of large (up to several cm) diatom flocs; flocs appeared old
Monterey Bay–MB2 (36° 50' N, 121° 54' W)	July 29, 30, 1993	~ 55	Some marine snow-sized flocs, although relatively less than at MB1
East Sound (48° 40' N, 122° 54' W)	April 14–24, 1994	~ 25	Water appeared green due to a phytoplankton bloom, but few aggregates observed relative to both MB1 and MB2

^a A.L. Alldredge and C. Gotschalk (Personal communication). Old flocs are characterized on the basis of having a white (as opposed to green or brown) color, few identifiable viable particles, eroded fecal pellets, and a highly amorphous appearance (Alldredge and Gotschalk, 1990).

alcian blue, and the filters soaked in 80% sulfuric acid for 2–3 h to re-solubilize adsorbed dye. The amount of dye was measured by absorbance at 787 nm and the final concentration of dye calculated after correcting for the absorbance of a distilled water blank prepared in the same manner. Colloidal gum xanthan particles were used as a calibration standard. TEP have been found to range from 50 to 350 $\mu\text{g l}^{-1}$ (xanthan equivalents) in coastal Pacific surface waters, with higher concentrations ($> 271 \mu\text{g l}^{-1}$) associated with flocculating diatom blooms (Passow and Alldredge, 1995).

Particle distributions were measured either as solid volume distributions using a particle counter (Coulter Multisizer II, Coulter Corp.) or as length distributions using an image analyzer (Scanarray-2, Galai). Details of the procedures have been described elsewhere (Li and Logan, 1995). Briefly, particle solid volumes, v_s , are measured and converted to solid equivalent diameters, d_s , using $d_s \equiv (6v_s/\pi)^{1/3}$. Volume distributions were measured using orifices with two different diameters (100 and 400 μm) providing maximum size ranges of $2.4 < d_s < 60 \mu\text{m}$ and $12 < d_s < 240 \mu\text{m}$. Data from the two orifices (2–3 measurements per sample) were combined into a single size distribution by overlaying the distributions. This resulted in data being used from the 100 μm orifice for $2.9 < d_s < 19 \mu\text{m}$ and from the 400 μm orifice for $19 < d_s < 200 \mu\text{m}$. Sample volumes used for analysis produced minimum measurable particle concentrations of 0.01 ml^{-1} in Monterey Bay and 0.05 ml^{-1} in East Sound for the largest particles.

To prepare samples for analysis using the image analysis system, seawater samples (15 ml in Monterey Bay and 10 ml in East Sound) were filtered onto 1.0 μm (pore diameter) black polycarbonate membrane filters (25 mm diameter, Poretics Corp.) and stained in the funnel with acridine orange. Samples were illuminated with blue light (BH-2, Olympus Corp.) and analyzed using the system software for particle

number and area. Projected area A was converted to length l using $l = (4A/\pi)^{1/2}$. Fifty fields (equivalent to 1/5 sample filtration area) were analyzed at $100\times$ for each sample, resulting in a measurable size range of 7–300 μm and minimum particle concentrations for the largest particles of 0.3 ml^{-1} in Monterey Bay and 0.5 ml^{-1} in East Sound.

2.2. Fractal dimensions using the PCT

Fractal dimensions of particles were calculated using the particle concentration technique (PCT) as described in Li and Logan (1995). The PCT is more useful for calculating fractal dimensions for marine particles than the “two-slope” method (Logan and Kilps, 1995; Jiang and Logan, 1996) since the PCT can be applied to curved size distributions. Briefly, the PCT method requires that the same population of particles be described using two different size scales, the solid volume (or solid equivalent diameter) and aggregate length. These aggregate sizes are obtained using two different instruments, an aperture impedance particle counter, such as a Coulter counter, and an image analyzer. Since a Coulter counter is calibrated with solid spheres particle sizes are reported in terms of solid volumes, which can be related to a diameter by $v_s = \pi d_s^3/6$, where the subscript on d indicates that this diameter is a solid equivalent diameter, that is the diameter that the particle would have if all of the particle porosity were removed. The actual size of the aggregate, which does include aggregate porosity, is seen using a microscope and measured in terms of an equivalent length (based on projected surface area) using an image analyzer as described above. According to fractal geometry, the solid volume and aggregate length are related by

$$v_s = al^D \quad (1)$$

where a is a constant that can be a function of the fractal dimension (Jiang and Logan, 1991). Based on our definition of $v_s = \pi d_s^3/6$, we can similarly write

$$d_s = bl^{D/3} \quad (2)$$

where $b = (6a/\pi)^{1/3}$ is a constant.

Two typical size distributions from the same water sample are shown in Fig. 1A. These distributions are presented in terms of $N(d_s)$ and $N(l)$, where N is the total number concentration of particles greater than the indicated size and the parentheses indicate what sizing technique was used to specify N . Because the Coulter counter measures the solid volume of a particle, and d_s is therefore a measure of the size of the particle if all the water in the particle were removed (i.e. if the particle had a porosity of 0), $d_s < l$. Based on the PCT assumption that we are measuring the same population of particles, the cumulative particle concentrations in terms of solid volume and length are equal, or

$$N(d_s) = N(l) \quad (3)$$

At any particle concentration, N , equation (3) allows us identify what particle lengths, measured using the image analysis system, correspond to solid volumes (or diameters)

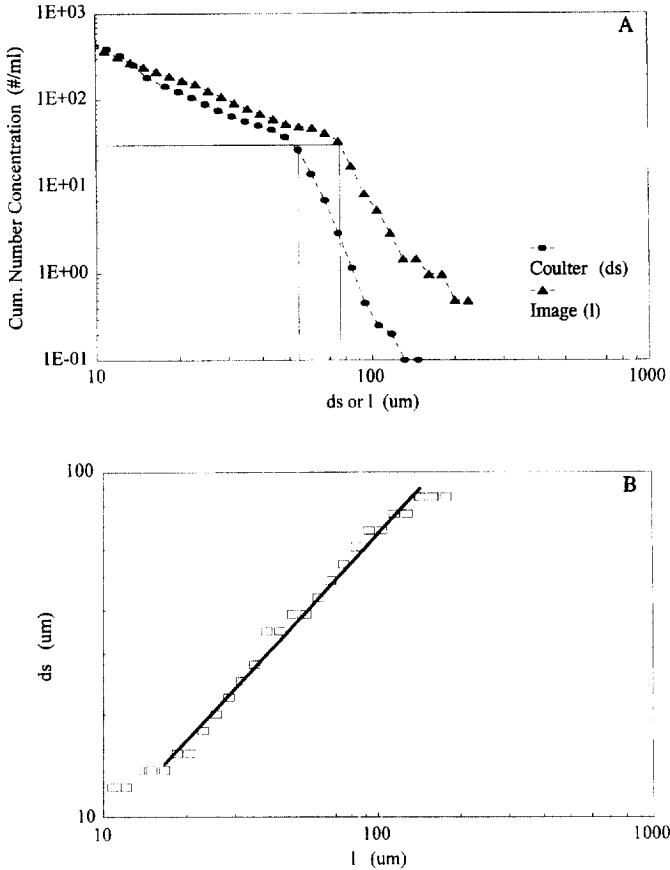


Fig. 1. (A) Cumulative particle size distributions in terms of solid equivalent diameter (d_s) measured using a Coulter counter and equivalent length (l) measured using an image analysis system; (B) The relationship between d_s and l established using the particle concentration technique (PCT). This sample was taken from the surface water in East Sound on 21 April, 1994.

measured with the particle counter. In other words, when the particle concentrations measured using the two different systems are equal, or $N(d_s) = N(l)$, we can identify a d_s that corresponds to l . For example, in Fig. 1A we see that at a particle concentration of $N = 30 \text{ ml}^{-1}$, a particle with a solid diameter of $d_s \approx 54 \text{ }\mu\text{m}$ will have an equivalent length of $l \approx 76 \text{ }\mu\text{m}$.

To calculate the D we take the logarithms of both sides of equation (2), producing the linear equation

$$\log d_s = \log b + \frac{D}{3} \log l \tag{4}$$

D can be calculated from the slope in equation (4) from a linear regression of a matched set of l versus d_s data produced over a wide range of l .

Due to the large number of data sets, calculation of D was performed using a spreadsheet (Quattro-Pro v.5.0) macro program. For each sample, the macro began by comparing the highest count for the $N(l)$ distribution with those from the $N(d_s)$ distribution to search for the number $N(d_s)$ that was closest to $N(l)$. On this basis, the first pair of d_s and l were identified. For this same data set, the macro then used successive particle number concentrations, $N(l)$, separated into length scale intervals of $\Delta \log(l) = 0.047$, each time identifying a particle concentration $N(d_s)$ closest to $N(l)$. This produced a series of d_s vs l data such as the one shown in Fig. 1B. In order to minimize the errors arising from the least accurate portions of the data sets (at the two extremes of the sizes), the first five and the last three data pairs were omitted from the regression analysis. Thus the fractal dimensions calculated from our data typically apply to particles 15–200 μm in length.

It should be emphasized here that distributions with changes in slopes can be used to calculate D with the PCT since the slopes of the size distributions are not used in the calculation. Instead, D is calculated from the slope after pairing d_s and l data based on the number of particles. As shown in Fig. 1 for a typical set of data, plots of d_s vs l were linear. Changes in the slopes of the size distributions had little effect on the final calculation of D . For example, at particle sizes l of 50–80 μm the sudden change in slope in Fig. 1A did not produce substantial deviations in the slope used to calculate D as shown in Fig. 1B.

2.3. Fractal dimensions using other techniques

We compared our results using the PCT to two other methods in order to show the usefulness of the PCT method. The first method required an assumption that the particle size distributions were at steady state, while the second method used the average slopes from the two size distributions (two-slope method). If a size distribution undergoing coagulation reaches a steady condition where the slope of the size distribution is constant, then the fractal dimension can be calculated from either a length distribution (Jiang and Logan, 1991) as

$$D_1(\text{SS}) = -2S_1 - 3 \quad (5)$$

or from the solid equivalent diameter distribution as

$$D_d(\text{SS}) = \frac{9}{2S_d + 3} \quad (6)$$

where S_1 and S_d are the slopes of cumulative particle concentration obtained from log–log plots of N as a function of l or d_s , and D_1 and D_d are the corresponding fractal dimensions. When size distributions for the same population of particles are measured in terms of length and solid equivalent diameter, the fractal dimension can be calculated from the two slopes (Logan and Kilps, 1995; Jiang and Logan, 1996) as

$$D(\text{TS}) = \frac{3S_1}{S_d} \quad (7)$$

All data used to calculate D by the PCT method were used to calculate D by the two-slope method. Therefore, when size distributions (such as those shown in Fig. 1A) had changes in the slope, an average slope, was calculated over the whole particle size range.

3. Results

3.1. Concentration profiles

Particle concentrations varied by orders-of-magnitude at some sites, but generally were highest at Monterey Bay Site 1 (MB1) and lowest at Monterey Bay Site 2 (MB2). Average particle concentrations (\pm SD) measured with the Coulter counter (binned into three size classes) are shown as a function of depth in Fig. 2 for the entire study period. Total particle concentration for all particles ($2.9 < d_s < 200 \mu\text{m}$) at MB1 varied widely from 4500 to 27 400 ml^{-1} (average of 15 900 ml^{-1}). At MB2 total particle counts were generally lower than those at MB1, ranging from 2500 to 7600 ml^{-1} (average of 5,100 ml^{-1}). While MB1 had more smaller particles ($> 2.9 \mu\text{m}$) than MB2, the opposite was true for the larger particles ($> 84.5 \mu\text{m}$).

Total particle concentrations in East Sound ranged from 4600 to 16 200 ml^{-1} with an average of 10 400 ml^{-1} . Particle counts in these three size classes decreased only slightly with depth. Compared to the particle profiles in Monterey Bay, concentrations of larger particles ($> 84.5 \mu\text{m}$) in East Sound were higher in surface waters. In

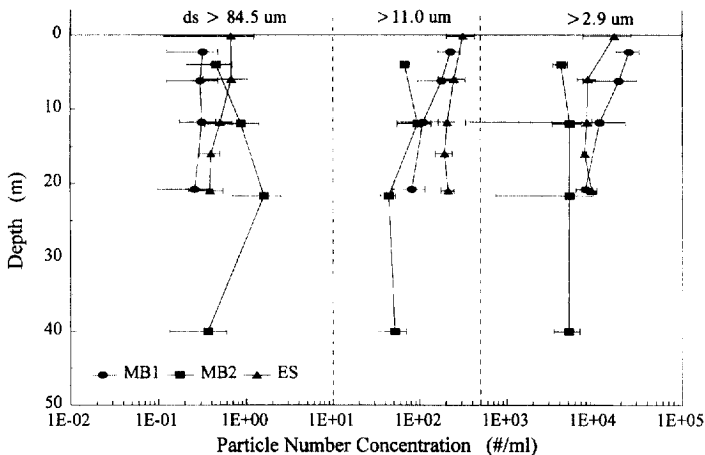


Fig. 2. Vertical profiles of particle concentrations in Monterey Bay, CA (●: Site 1; ■: Site 2) and East Sound, WA (▲) averaged over the entire sampling period. Particles were measured using a Coulter counter and particle size is represented in terms of solid equivalent diameter (d_s). Particles are grouped into 3 different size classes where the maximum d_s is 200 μm for all size classes (Error bars represent \pm SD for sampling period).

general, there appeared to be less variation in particle concentration in East Sound than in Monterey Bay despite the longer study period in East Bay.

3.2. Fractal dimensions

Slopes of the size distributions produced with the Coulter counter were not constant over time and were highest at the Monterey Bay sites. Slopes ranged from -2.8 to -3.8 (average of -3.33 ± 0.48) at MB1, and from -2.3 to -2.9 (average of -2.59 ± 0.26) at MB2 (Table 2). In East Sound, the slopes were lower and less variable, ranging from -2.2 to -2.8 (average of -2.49 ± 0.27). When combined with data from the image analysis system, the slopes of the two size distributions produced lower fractal dimensions of particles at the Monterey Bay sites than in East Bay (Table 2). Daily average fractal dimensions of all particles $15\text{--}200\ \mu\text{m}$ in length are shown in Fig. 3. We found no significant change in D with depth at the three sites.

Table 2

Fractal dimensions measured from the average slopes of particle size distributions using different techniques (SS = steady state-one slope. TS = two slope, and PCT = particle concentration technique)

Site	Average slopes		Fractal dimensions			
	$-S_1$	$-S_d$	D_1 (SS)	D_d (SS)	D (TS)	D (PCT)
Monterey Bay-1	1.83 ± 0.25	3.33 ± 0.48	0.66 ± 0.49	2.61 ± 0.58	1.67 ± 0.28	1.77 ± 0.34
Monterey Bay-2	1.49 ± 0.14	2.59 ± 0.26	-0.01 ± 0.27	4.02 ± 0.54	1.71 ± 0.22	2.12 ± 0.16
East Sound	1.96 ± 0.23	2.49 ± 0.27	0.93 ± 0.46	5.35 ± 2.33	2.42 ± 0.18	2.59 ± 0.17

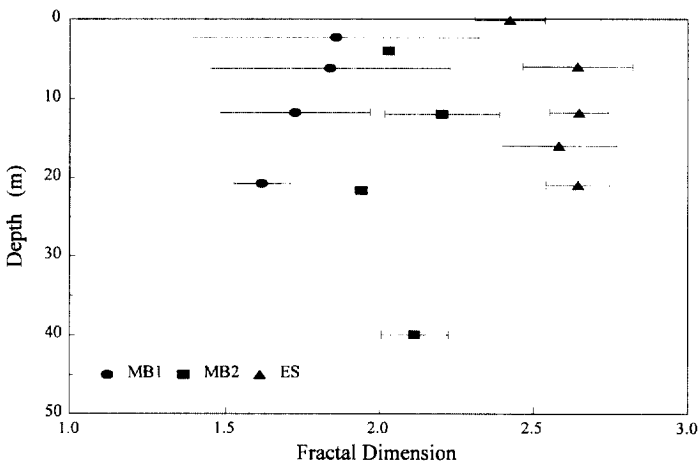


Fig. 3. Average fractal dimensions of all (aggregated and non-aggregated) particles in the size range $15\text{--}200\ \mu\text{m}$ (in length) as a function of depth and location, Monterey Bay (●: Site 1; ■: Site 2) and East Sound (▲) (Error bars are \pm SD).

Average fractal dimensions calculated using the PCT were lower at the two Monterey Bay sites with 1.77 ± 0.34 (range 1.1–2.5) at MB1 and 2.12 ± 0.16 (range 1.9–2.5) at MB2. Fractal dimensions were significantly higher in East Sound waters than in either MB1 or MB2 samples (t-test, $P < 0.001$), ranging from 2.2 to 2.9 with an average of 2.59 ± 0.13 .

On average, fractal dimensions calculated using the two-slope technique were similar to those calculated using the PCT at the East Sound and MB1 sites (Table 2). The fractal dimension of $D(\text{TS}) = 1.71 \pm 0.22$ at MB2, however, was smaller by more than one standard deviation than that of $D = 2.12 \pm 0.13$ calculated using the PCT. Fractal dimensions calculated assuming the size distributions represented a coagulating system that was at steady state varied over an impossible range of $D_1(\text{SS}) = -0.01$ to $D_d(\text{SS}) = 5.35$. Part of the reason for the unrealistic range of fractal dimensions calculated using this technique could be due to non-linear size distributions. However, it is more likely that meaningful fractal dimensions were not produced by the steady-state technique because the systems did not meet the necessary assumptions for applying this method (i.e. the systems were not coagulating systems at steady state).

3.3. TEP and chlorophyll *a* in surface waters (≤ 10 m)

Since only surface waters were analyzed for TEP and chlorophyll *a*, average fractal dimensions were recalculated based only on the surface water samples. D values for surface waters were not different (\pm SD) than those produced by averaging all samples over the whole water column (Table 3). In Monterey Bay TEP concentrations were nearly 70% higher on average at MB2 ($176 \pm 95 \mu\text{g l}^{-1}$), where divers observed a larger concentration of macroscopic aggregates, than they were at MB1 ($104 \pm 24 \mu\text{g l}^{-1}$), where there were relatively fewer aggregates observed. Similarly, chlorophyll *a* concentrations were higher at MB1 ($6.1 \pm 2.2 \mu\text{g l}^{-1}$) than at MB2 ($2.4 \pm 0.9 \mu\text{g l}^{-1}$).

Chlorophyll *a* concentrations in the East Sound ($8.2 \pm 6.0 \mu\text{g l}^{-1}$) were on average only slightly higher than those from the Monterey Bay sites, but they steadily increased during the last 9 days of our field study (16 to 24 April) in East Sound by nearly an order of magnitude from $2.3 \mu\text{g l}^{-1}$ to $22 \mu\text{g l}^{-1}$ (Fig. 4). This increase in chlorophyll *a* was attributed primarily to the bloom of the *Thalassiosira* sp. TEP concentrations in East Sound ($89 \pm 24 \mu\text{g l}^{-1}$) were the lowest among the three study sites despite having the highest concentrations of chlorophyll *a*. Since microscopic particles at this site also had the highest fractal dimension, this suggests that TEP concentration and D are inversely related.

4. Discussion

Fractal dimensions significantly less than three demonstrate the fractal nature of the total population of small marine particles (15–200 μm in length). Since the fractal dimension of an amorphous aggregate is lower than that of a solid particle, such as

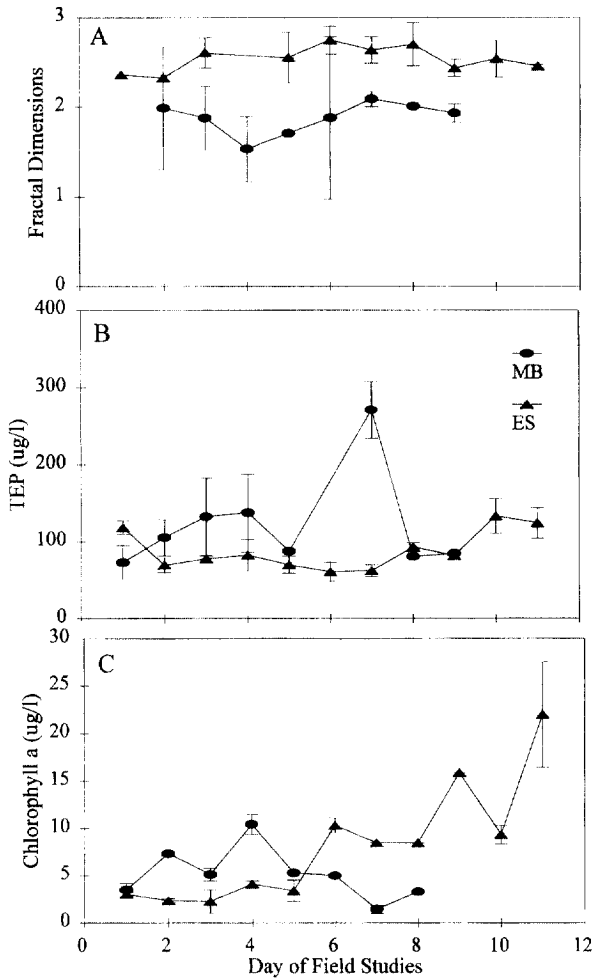


Fig. 4. Daily changes in fractal dimensions of particles (15–200 μm in length) (A), TEP (B) and chlorophyll *a* (C) in surface waters (≤ 10 m) in Monterey Bay (●; MB2 days 7 and 8, MB1 other days) and East Sound (▲). (Error bars are \pm SD).

a single diatom, the magnitude of the average fractal dimension for this size range of particles indicates whether or not a majority of the particles in this size range are amorphous particles formed by coagulation. Based on fractal dimensions of particles measured in a mesocosm study, we previously proposed that the average fractal dimension reflected the aggregation state of a phytoplankton bloom (Li and Logan, 1995). In the mesocosm experiment the average fractal dimension decreased from $D = 2.5$, at the beginning of the phytoplankton bloom, to $D = 1.7$ (typical of marine snow), when aggregates were highly abundant at the end of the bloom. The magnitude

of D was therefore consistent with the observed abundances of larger aggregates in water samples. A comparison of the mesocosm experiment with the East Sound data ($D = 2.59 \pm 0.13$) suggests that the sampling site in East Sound was either a non-coagulating system or was coagulating very slowly. It may be that the phytoplankton bloom in the East Sound eventually coagulated at a time after our study was concluded.

In contrast to East Sound results, in Monterey Bay the fractal dimensions of small particles at MB1 averaged 1.77 (range 1.1–2.5). This compares well to estimates of D from several other studies. Fractal dimensions of < 2 were reported by Risović and Martinis (1996) for suspended seawater particles larger than $\sim 0.5 \mu\text{m}$ analyzed by a light scattering technique. They found D for this size range of particles was 1.74 ± 0.09 ($n = 34$) at 8 sites for samples from depths of 10–800 m. At an unidentified coastal site off Southern California they measured $D = 1.8$, which compares well to $D = 1.77$ found at site MB1. The D measured at MB1 is also similar to $D = 1.7$ measured in the mesocosm experiment for similarly-sized particles after the phytoplankton bloom coagulated (Li and Logan, 1995), and are in the range of values reported for larger marine snow aggregates (Logan and Wilkinson, 1990; Kilps et al., 1994). The observation of low fractal dimensions for smaller particles at sites with higher concentrations of marine snow (Site MB1), and higher fractal dimensions at sites with few marine snow aggregates (Site EB) suggests that many of the particles present in the smaller size range of 15–200 μm at MB1, like those of marine snow, are formed by coagulation. Consistently low D values for the Southern California sites also suggests that there is no sudden shift in the fractal nature of particles between the sizes of 15 to 200 μm and marine snow size particles.

The average fractal dimension at the Monterey Bay site derived using the PCT, however, was higher than those reported by Jackson et al. (1997) for the same site calculated using a different technique. Jackson et al. used a fractal scaling technique (FST) to match the shapes of size distributions of small particles (the same Coulter counter data set reported here) and the size distributions of larger particles obtained using in-situ photographic and imaging systems. The shapes of the size distributions from the different instruments were found to be consistent with each other by Jackson et al. if the fractal dimensions of the particles were 2.24–2.3, values substantially higher than the 1.77 obtained here for site MB1. Both methods rely on aperture impedance counters, which have limitations reviewed by Jackson et al. (1997), and both methods require that D is constant over the size range examined. The main difference between the two approaches is the range of particle sizes over which it is assumed that D is constant: the FST is based on a comparison of different portions of the particle size distributions (particles vary widely in size) each measured using different machines, while the PCT is based on a comparison of the same portion of the size distribution (same size particles) measured using different instruments. In the current study we measured only D 's of small particles, and found that they were similar in magnitude to those typically measured for marine snow, suggesting that D is constant over large size ranges as required by the FST. However, we did not actually measure D for both large and small particles present in the system at the same time. Because we cannot be certain from our data that D was constant over large particle sizes, we cannot

conclude whether non-constant D 's contributed to different outcomes using the PCT and FST approaches.

The PCT method should be accurate even when there are changes in slopes in the size distributions, but procedures used to prepare and analyze samples could have affected the particle sizes. The transport of water samples from the site to the laboratory can induce particle breakup, but because fractal aggregates are self-similar the average fractal dimension of the aggregates will be preserved even if the sizes of the aggregates change. Filtering particles onto a surface could also affect the apparent length of some portions of amorphous particles, but we used an average length based on the whole cross sectional area of the particle, which minimizes the contribution of any portion of the aggregate to the overall size. Particle overlap on filters probably occurred, but we do not know to what extent this contributed to errors in our measured size distributions. Particle overlap (coincident particles) could also have been a problem in the Coulter Counter measurements although it was not expected to be a problem at the particle concentrations used in our experiments. It is also possible that v_s measured using the Coulter Counter could be inaccurate due to breakup in the orifice or inaccurate translation of measured resistance to solid volume. Breakup is generally thought to be minimized when particle sizes are less than 50% of the orifice size (Gibbs, 1982, 1983). We only used data for particle sizes with $d_s < 100 \mu\text{m}$ ($l < 200 \mu\text{m}$), a range well within the 50% criterion for the 400- μm orifice used here. In addition, D is calculated from many points of d_s and l , and therefore most of the data used to calculate D were for particles substantially smaller than the orifice. Jackson et al. (1995) reviewed several studies made with aperture impedance counters and concluded that particle volumes reported for porous particles should be accurate to within a factor of two compared to that for a solid sphere, indicating that d_s measured by resistance particle counters is accurate to within a factor of $2^{1/3} = 1.26$. Even if individual estimates of v_s or d_s made here were slightly inaccurate, the calculation of D is believed to be reliable since a linear relationship was obtained between d_s and l to calculate D as was shown in Fig. 1.

The best evidence that sampling preparation and analysis did not affect the magnitude of D is the consistent relationship between D and the relative abundances of large aggregates at the three sampling locations. Large aggregate concentrations, evaluated by divers with extensive experience in sampling marine snow, indicated that macroscopic aggregate concentrations decreased in the order MB1 > MB2 > ES. This ordering was supported by our observations of acridine orange-stained slides, because the fewest micro-aggregates were present in East Bay samples. As the fractal dimension increases particles become more spherical, less tenuous and amorphous in appearance (Witten and Cates, 1986; Kilps et al., 1995). This ordering suggested by increasing concentrations of micro and macro aggregates is consistent with the reverse ordering of fractal dimensions of MB1 < MB2 < ES.

The production of high concentrations of marine snow sized aggregates cannot be predicted just from high total particle concentrations at a site. While aggregate abundances decreased in the order MB1 > MB2 > ES, particle concentrations decreased in the order MB1 > ES > MB2 (15900 particles ml^{-1} > 10400 particles ml^{-1} > 5100 particles ml^{-1} , respectively) indicating aggregate abundances did not

covary with total particle concentrations. Chlorophyll *a* concentrations were similarly not consistent with our ranking of the importance of aggregation in the system according to large aggregate abundances. Although we measured higher chlorophyll *a* concentrations in East Sound than in Monterey Bay, there was little evidence of particle aggregation in East Sound as shown by low aggregate abundances and consistently high fractal dimensions of smaller particles during the 10 day study. Thus, neither chlorophyll *a* or total particle concentrations were predictors of active aggregation at these sites.

The concentration of TEP was measured at the three sites because recent studies indicate that aggregation of diatom blooms in Pacific waters proceeds at a rate dependent more on the concentration of TEP than on the concentrations of phytoplankton and other particles (Aldredge et al., 1993; Logan et al., 1995). TEP are formed from polysaccharide exopolymers excreted by phytoplankton and bacteria. These polysaccharide materials are referred to as biofloculants because they enhance particle aggregation kinetics (Bar-Or and Shilo, 1987; Bender, et al., 1994). TEP either makes particles at a site sticky (Jackson, 1995), or the TEP themselves coagulate and sweep other particles from the water column in a manner less dependent on the concentration of particles (Logan et al., 1995). Fractal dimensions of aggregates formed by Brownian motion (Witten and Cates, 1986) and shear coagulation (Li and Logan, 1997) are lower when they are formed from particles that are sticky (destabilized) than they are when formed more slowly with more stable (less sticky) particles. Thus, low fractal dimensions result from rapid coagulation which can only occur if particles readily stick to one another.

On a relative basis higher TEP concentrations should promote more rapid particle aggregation, and a greater proportion of aggregated particles in the water column with low fractal dimensions, than lower TEP concentrations. When TEP data are compared to the measured fractal dimensions we find that fractal dimensions at the two sites with the highest concentrations of TEP (MB1 and MB2) also had the lowest fractal dimensions. However, there was no direct correlation between TEP concentrations and *D* since the site with the highest average concentration of TEP (MB2) did not have the lowest fractal dimension (Table 3) although the concentrations of TEP at all these sites was quite variable. Thus, if there is a relationship between TEP and *D* it will need to be established by obtaining data from more sites than examined in the present study.

There are many factors affecting the absolute rates of aggregation, such as the shape and sizes of particles forming the aggregate, fluid shear rate, particle concentration, TEP sizes and concentration, the size of primary phytoplankton species, particle stickiness, and disaggregation (Jackson 1990, 1995; Logan et al., 1995). Despite the complex nature of aggregation kinetics and marine environments, our results suggest that aggregation is an important process in forming particles in the size range of phytoplankton in eastern Pacific coastal waters and that these particles can have fractal dimensions significantly less than three. The fractal nature of these particles has many important implications for the properties of these marine particles. Fractal particles are highly porous and therefore will have sizes much larger than those indicated by aperture impedance particle counters. Laboratory settling velocity

Table 3

Comparison of site characteristics of surface waters (<10 m) in terms of average fractal dimension and concentrations of TEP and chlorophyll *a*

Site	N(> 2.9 μm) (#/ml)	TEP ($\mu\text{g l}^{-1}$)	Chlorophyll <i>a</i> ($\mu\text{g l}^{-1}$)	Fractal dimension, <i>D</i>
Monterey Bay-1	15900 \pm 1300	104 \pm 24	6.1 \pm 2.2	1.82 \pm 0.15
Monterey Bay-2	5100 \pm 500	176 \pm 95	2.4 \pm 0.9	2.05 \pm 0.05
East Sound	10400 \pm 1600	89 \pm 24	8.2 \pm 6.0	2.54 \pm 0.13

experiments (using aggregates composed of fluorescent microspheres) have demonstrated that fractal particles will have lower drag coefficients, and therefore higher settling velocities, than non-porous particles of similar size and density (Johnson *et al.*, 1996). These laboratory results are consistent with measurements on marine particles since lower drag coefficients have been reported for re-coagulated marine sediments even after assuming an upper limit for a primary particle density of 2.65 g cm^{-3} (Kajihara, 1971; Gibbs, 1985). Fractal particles also have higher collision frequencies with other particles than predicted by coagulation models based on impermeable spheres when collision rates are corrected for fluid streamlines around spheres (Li and Logan, 1997). Thus, our finding that phytoplankton-sized particles are fractal will have important implications for predictive equations used in models of particle-particle interactions and settling velocities used to predict carbon fluxes to sediments.

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