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Physical and chemical effects of grain aggregates on the Palos Verdes margin, southern California

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Abstract

Large discharges of wastewater and particulate matter from the outfalls of the Los Angeles County Sanitation Districts onto the Palos Verdes shelf since 1937 have produced an effluent-affected sediment deposit characterized by low bulk density, elevated organic matter content, and a high percentage of fine silt and clay particles relative to underlying native sands and sandy silts. Comparison of the results of grain-size analyses using a gentle wet-sieving technique that preserves certain grain aggregates to the results of standard size analyses of disaggregated particles shows that high percentages (up to 50%) of the silt and clay fractions of the effluent-affected mud are incorporated in aggregates having intermediate diameters in the fine-to-medium sand size range (63–500 μm). Scanning electron microscope images of the aggregates show that they are predominantly oval fecal pellets or irregularly shaped fragments of pellets. Deposit-feeding polychaete worms such as *Capitella* sp. and *Mediomastus* sp., abundant in the mud-rich effluent-affected sediment on Palos Verdes shelf, are probably responsible for most of the grain aggregates through fecal pellet production.

Particle settling rates and densities, and the concentrations of organic carbon and *p,p′*-DDE, a metabolite of the hydrophobic pesticide DDT, were determined for seven grain-size fractions in the effluent-affected sediment. Fecal pellet grain densities ranged from about 1.2 to 1.5 g/cc, and their average settling rates were reduced to the equivalent of about one phi size relative to spherical quartz grains of the same diameter. However, repackaging of fine silt and clay grains into the sand-sized fecal pellets causes an effective settling rate increase of up to 3 orders of magnitude for the smallest particles incorporated in the pellets. Moreover, organic carbon and *p,p′*-DDE exhibit a bimodal distribution with relatively high concentrations in the finest size fraction (0–20 μm), as expected, and a second concentration peak associated with the sand-sized fecal pellets. The repackaging of fine-grained particles along with their adsorbed chemical compounds into relatively fast-settling pellets has important implications for the mobilization and transport of the sediment and the desorption of chemicals from grain surfaces. © 2002 Published by Elsevier Science Ltd.

1. Introduction

Recent research in marine sedimentology includes more multidisciplinary, process-oriented studies in which the characteristics of sediment particles and deposits are understood to be the result of interactions between complex physical, biological and chemical systems. To a significant degree, the present emphasis on multidisciplinary programs that seek a quantitative understanding

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of the processes that affect the production, transport, accumulation and reworking of shelf and slope sediment reflects a desire to refine our ability to describe and predict change. Change prediction requires detailed knowledge of forcing functions, system interactions, and especially the rates at which key processes operate.

This paper describes a study to determine the in situ physical and chemical characteristics of the particulate matter in an effluent-affected sediment layer (Lee et al., 2002) that has been created by accumulation of organic-rich sediment since about 1937 around and downcurrent (northwest) of the White's Point outfall system. The textural properties of the effluent-affected sediment are compared to pre-effluent, native sediments that surround and underlie the effluent-affected layer on Palos Verdes continental margin near Los Angeles, California (Fig. 1). The results provided inputs for bed mixing and bottom boundary layer models developed by Sherwood et al. (1996), and the pesticide desorption and sediment transport analysis of Wiberg and Harris (2002).

Most studies of the grain-size characteristics of unconsolidated sediments on continental margins have followed standardized methods that use one or more techniques to disaggregate flocs and other aggregates and keep them disaggregated during the size determinations. Because our research was aimed at defining the dynamical and chemical properties of the sediment grains, we chose to use a non-dispersive sediment analysis method, developed by C.A. Butman and her colleagues at Woods Hole Oceanographic Institution, that is designed to preserve in situ grain-size distributions by minimizing the destruction or production of multi-grain aggregates (Fuller and Butman, 1988; Wheatcroft and Butman, 1997). Standard disaggregated grain-size distributions also were determined for most samples in order to rapidly assess aggregation states and to provide a dataset comparable to previous studies in the area.

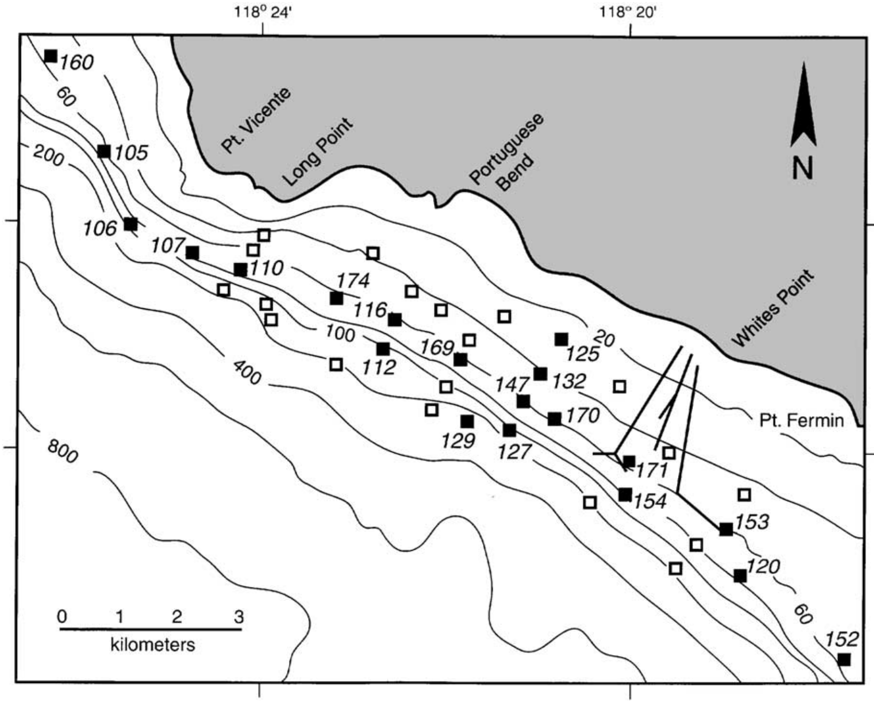

Fig. 1. Bathymetry of Palos Verdes shelf and upper slope showing the Whites Point effluent diffuser system operated by LACSD and the stations that were sampled in 1992. Data from labeled coring sites shown here by solid squares are presented in subsequent figures.

Sand-sized fecal pellets are shown to be abundant in the effluent-affected sediment and we present calculations to assess the potential importance of the pellets to sediment resuspension and transport rates in the bottom boundary layer. Finally, we show that chemical compounds adsorbed to fine-grained particles will be incorporated into pellets and aggregates by deposit-feeders. This can have a significant impact on the rates at which the compounds desorb to ocean water during episodes of sediment resuspension and transport (Wiberg and Harris, 2002).

2. Methods

In July 1992, box cores and gravity cores were collected at 38 stations between the 30 and 200 m isobaths on the continental margin adjacent to the Palos Verdes Peninsula (Fig. 1). The box corer obtained 20 cm × 30 cm rectangular samples of the bed up to 50 cm thick. The gravity cores obtained cylindrical samples, 10 cm in diameter and up to 3 m in length. Samples for grain-size analyses, chemical and physical property tests were obtained with a specially designed device that took relatively undisturbed 8.3 cm-diameter subcores from the box cores immediately after recovery (Lee, 1994). The subcoring device employed a fixed internal piston that was held just above the sediment/water interface while a plastic core tube was pushed into the boxcore with a constant-speed motorized driver. This device minimized core shortening which can be a problem if a piston is not used (Blomqvist, 1991).

Several subcores were obtained in each box core. One subcore was split and described on shipboard and the halves were sealed in separate airtight storage tubes and refrigerated. A second subcore was left unsplit, capped, sealed and frozen for future chemical tests. A third core was analyzed on shipboard by a non-destructive whole-core profiling device that measures sediment bulk density (see Lee et al., 2002). Most of our grain-size determinations were done on one half of the refrigerated subcores, while the frozen subcores were set aside for analyses of organic matter and the chlorinated hydrocarbons, DDT and PCB.

Our study focused on determining the characteristics of the sediment at three horizons: (1) the surface of the sea floor; (2) the subsurface sediment layer that contained the highest concentration of organic material; and (3) the native silty sand that underlies the effluent-affected layer. The upper two centimeters of each box core subsample was used for the surface sample, which we assume reflects the most recent conditions at each site. Many studies (Stull et al., 1986a; Lee, 1994; Drake, 1994 and references cited therein) have shown a strong correlation between organic carbon content, bulk sediment density and concentrations of the DDT metabolite p,p'-DDE in the effluent-affected layer on Palos Verdes margin. Accordingly, identification of the subsurface layer that was likely to contain the largest amounts of organic matter and p,p'-DDE was based on downcore profiles of bulk density derived from the USGS whole-core logger (Fig. 2). Two centimeters of sediment from within the zone of lowest bulk density was selected for the peak effluent discharge or effluent-peak horizon. This horizon is believed to have been deposited in the late 1960s and early 1970s when discharges of organic-rich particulate matter from the White's Point outfalls reached maximum values of more than 150,000 tons/yr. This is evidenced by a correlative peak in organic C and N concentrations in the subsurface at many shelf sites (Fig. 2; and Stull et al., 1996). The third horizon, in the native pre-effluent sediment, was also selected based on the bulk density profiles of the subcores (Fig. 2). The native sandy sediments are known to exhibit high bulk density (> 1.8 g/cc), a product of relatively low porosity and low content of organic matter (Lee, 1994).

2.1. Particle characteristics

Grain-size distributions were determined for both "aggregated" and "disaggregated" samples. The aggregated size tests involved gentle wet sieving of approximately 1–2 g of the moist refrigerated sediment through a stack of seven stainless steel sieves with the following mesh sizes in microns: 500, 250, 125, 90, 63, 45 and 20 (Fuller and Butman, 1988). The aggregated sediment

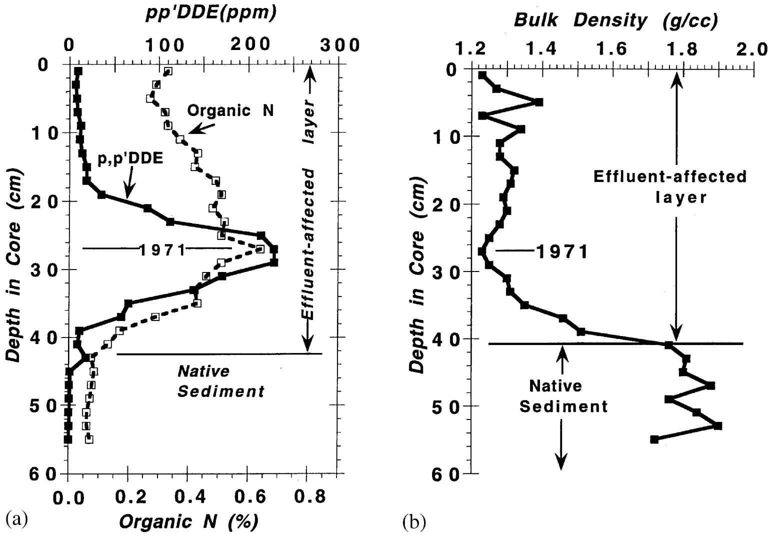

Fig. 2. (a) Organic C and *p,p′*-DDE profiles and (b) bulk sediment density in a core collected in 1989 at LACSD monitoring station 6C (near USGS box core site 147 see Fig. 1). The organic-rich "effluent peak" sediment accumulated in about 1971/1972 when the discharges of effluent were highest. Native sediment below the effluent-affected layer is readily identified by its relatively high bulk density and low organic C and contaminant content.

samples had never been allowed to dry out and were not pre-treated in any way; no organic solvents were added and no steps were taken to disperse any naturally occurring grain aggregates. Filtered artificial sea water (salinity = 34 ppt) was used for the wet sieving which was accomplished with a gentle stream of water from a Teflon wash bottle (Wheatcroft and Butman, 1997). Wet sieving continued until visual tests showed that no particles were passing the sieves. Each sieve fraction was then quantitatively transferred to a filtration funnel fitted with a pre-weighed 47 mm diameter *Nuclepore* polycarbonate membrane filter having a nominal pore size of 0.4 μm. The fractions were filtered onto the membranes, washed three times with a total of about 50 ml of distilled water, dried at 50°C for at least 6 h and reweighed using an electrobalance to determine the weight of the sediment plus filter. The finest grain-size fraction, 0–20 μm, was determined by membrane filtration of 50 ml aliquots from the collected wash water. Replicate and duplicate analyses show that the wet sieve sizing method has a reproducibility of about ±10% of the weight percentage n each size fraction. Replicate analysis of a well-mixed test sample of the effluent-affected mud by different laboratory technicians showed a similar amount of variability, in agreement with the findings of Wheatcroft and Butman (1997).

For the disaggregated sediment analyses, several grams of the moist sediment were treated with solvent to remove organic material and this was followed by ultrasonic dispersion. Sand and silt/clay fractions were separated by sieving and the sands were dried and sieved at whole phi intervals. Silt and clay fractions were separated into whole phi intervals by settling and pipette analysis. Replicate and duplicate analyses show that the sieving and pipette methods have a reproducibility of about ±10% of the percentage in each size class.

Standard light microscopy of the sand fractions from the effluent-affected layer revealed many oval grain aggregates. Because it seemed probable that the density of the aggregates would differ substantially from the density of quartz (2.65 g/cc) and that this could significantly impact sediment transport calculations, we decided to examine the settling characteristics of all sand fractions using a Rapid Sediment Analyzer (RSA). The RSA consists of a large-diameter (20 cm) clear plastic settling cylinder, 2 m in length, that is filled with distilled water and equipped with a computer-controlled system for introducing and weighing sediment as it settles and accumulates on a collection pan near the base of the cylinder. Normally, a few grams of dry sand containing a range of grain diameters (e.g., 63–1000 μm) is introduced at the top of the cylinder and the accumulation rate vs. time is used to separate the sample into quartz-equivalent size classes. In the present case, we wanted information on the "effective" grain densities of the various aggregates. To do this, we followed the same procedures as before to prepare a set of wet-sieved sand fractions of known size class (e.g., 63–125 μm) that we maintained in a wet condition in small vials until they were individually introduced to the top of the settling column; the wet introduction method required a simple modification of the standard RSA sand introduction system. The RSA graphically recorded weight as a function of time for each wet-sieved size class and these data were then used to estimate maximum, minimum and average grain densities using the settling velocity equation determined for discrete, spherical grains by Gibbs et al. (1971).

2.2. *Chemical analyses*

We analyzed two sets of samples to determine concentrations of *p,p′*-DDE and total organic carbon (TOC) in the grain-size classes. The first set of samples consisted of six bulk samples of the effluent-affected layer from the unopened frozen cores collected in July 1992. Relatively, large samples (2–4 g) were used for this analysis because of the detection requirements of the chemical procedure for pesticide testing. After slow thawing, two aliquots were wet sieved through carefully cleaned stainless steel screens to yield two sets of size-differentiated samples, one for grain-size analysis and the second for the chemical testing. Unfortunately, comparison of the grain-size distribution results for the frozen samples with results we had obtained earlier for samples that had been refrigerated *but never frozen* showed significant increases in the amount of sand-sized material in several of the frozen samples. This suggested that the core freezing process had artificially produced more large grain aggregates from normally dispersed fine-grained particles. Quick freezing of small (<1 g) samples has been used successfully by Wheatcroft and Butman (1997) for the preservation of marine sediment samples for grain-size analysis, so we suspect that the speed of sample freezing may be an important part of the explanation for the grain-size changes that we observed. The results are tentatively attributed to a core freezing process that was too slow, allowing water migration and local dehydration to occur in areas near the freezing front within the 10-cm diameter cores.

Because our frozen cores were rendered unreliable for size and chemical fractionation of aggregated samples, we obtained fresh samples of the effluent-affected sediment from the sealed, refrigerated subcores that had been taken from USGS box core 147 B3 (Fig. 1). Two samples were selected for fractionation analysis, one at the surface of the core (0–2 cm) and the second within the zone of low-density sediment at 32–34 cm. Size analyses of these new samples showed no evidence of artificially produced aggregates when compared to samples from this location that had been analyzed previously at USGS and at Woods Hole Oceanographic Institution by R. Wheatcroft.

The use of unfrozen sediment samples that were stored under refrigeration for about 1.8 yr prior to testing is not standard geochemical practice. To assess the possibility of chemical changes in the unfrozen core, we analyzed 11 bulk samples (2-cm sections) from the unfrozen archive core half, 147 B3 SS, for *p,p′*-DDE, *p,p′*-DDD, *p,p′*-DDT and *p,p′*-DDMU concentrations and compared the results with data that had been obtained in earlier tests on frozen samples from the subcore 147 B3 GC (Lee, 1994). Both subcores, 147 B3 SS and 147 B3 GC, were collected from the same box core (designated core 147 B3). All the chemical analyses were carried out by the analytical chemistry group at the Arthur D. Little Company, Cambridge, Massachusetts.

We selected 11 bulk samples to provide enough comparative data points spaced evenly along the core to reveal decreases in the concentrations of

Table 1
Chemical analyses of the bulk core section samples from the frozen subcore, 147 B3 GC, and the refrigerated subcore, 147 B3 SS

Depth in core (cm)	p,p'-DDT Fr[a]	p,p'-DDT Re[a]	p,p'-DDE Fr	p,p'-DDE Re	p,p'-DDD Fr	p,p'-DDD Re	DDMU Fr	DDMU Re
0–2	895	1650	11,350	13,500	605	877	1700	2140
4–6	485	465	6220	14,600	385	743	1180	2020
8–10	342	806	8190	13,000	583	931	1640	2360
12–14	1560	764	11,400	14,200	860	944	2110	2720
16–18	271	943	11,900	11,400	644	789	2490	1600
20–22	365	869	9340	14,600	708	977	1570	1720
24–26	380	1140	21,500	23,700	878	1430	3860	3990
24–26		1520		23,800		1460		4240
28–30	1200	1780	16,800	36,300	1790	1900	4800	6830
32–34	588	66.7	27,800	36,500	1700	1980	4000	7500
36–38	1600	3410	41,400	113,000	1940	2700	19,000	17,800
40–42	2340	1070	180,000	137,000	4640	7470	34,700	18,700

The values for the frozen core are labeled "Fr" and those from the refrigerated core are labeled "Re". A replicate analysis was performed on the sample at 24–26 cm in 147 B3 SS. All concentrations are in ng/g.

[a] p,p'-DDT values for the intervals below the surface layer (0–2 cm) are "detection limit" values; that is, the actual values are less then the limit given.

the major DDT components that may have occurred during the refrigerated storage period. We reasoned that any decrease in the total contents of DDT and, particularly, in the concentration of the principal component, p,p'-DDE, would signal chemical changes in the refrigerated core half that would cast some doubt on the fractionation results. The results show that, with few exceptions, the refrigerated subcore actually had somewhat greater concentrations of all of the various DDT metabolites (Table 1). Examination of metabolite ratios (not shown) also indicated that any loss or transformation of chemical compounds during refrigerated storage was not significant and, thus, we believe that our fractionation data can be used with confidence.

3. Results

3.1. Aggregated and disaggregated grain-size distributions

Sediment samples on Palos Verdes shelf and upper slope contain a sand-size aggregate fraction ranging from as little as 2% to as much as 54% by weight (Fig. 3). Aggregates comprise about 2–10% (mean = 4.6%) of the native sediment underlying the effluent-affected layer; 10–54% (mean = 22%) at the effluent-peak horizon of the early 1970s; and, 6–43% (mean = 13%) at the seafloor. Modal size of the aggregates generally is in the 63–125 μm size class in both the subsurface native sediment and the recently deposited sediment at the seafloor (Fig. 3). In contrast, a number of the effluent-peak samples display an aggregate mode in either the 125–250 or 250–500 μm size classes (e.g., samples from sites 112, 170 and 154 in Fig. 3).

With few exceptions, sample disaggregation greatly reduced the 125–250 and 250–500 μm fractions, shifting the bulk of that material into the 0–20 μm size class (Fig. 3). Reductions in the very fine sand (63–125 μm) aggregate mode typically were less dramatic but still important, particularly in the effluent-affected layer samples. Because the aggregate fractions tend to be relatively minor in the native sediment, disaggregated and aggregated size distributions are similar, exhibiting very fine sand or coarse silt modes and long tails into the 0–20 μm class (Fig. 3). Disaggregation of the effluent-peak sediment produced the largest changes in grain-size distributions. In

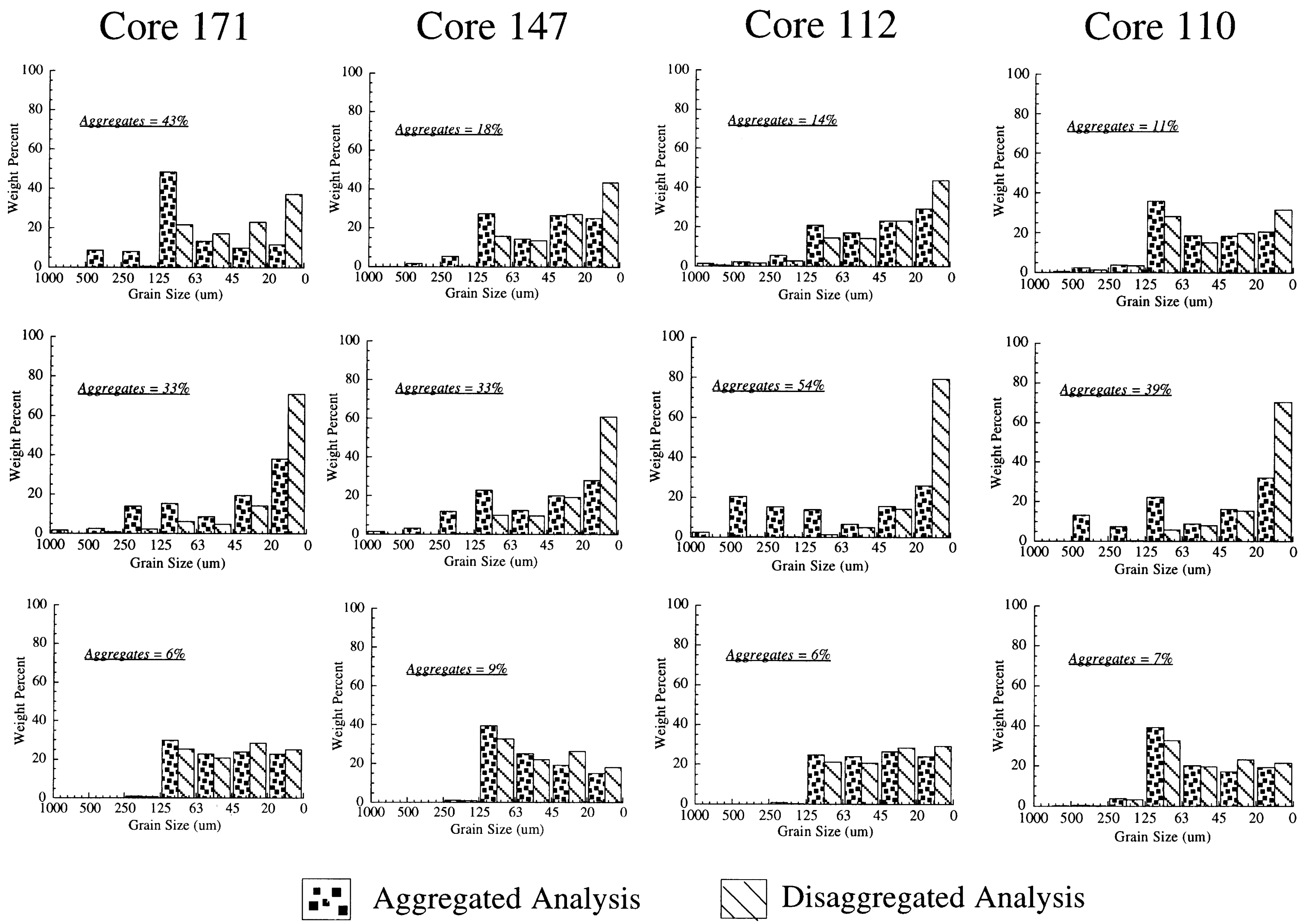

Fig. 3. Histograms of the aggregated and disaggregated grain-size distributions for the surface sediment (top row) effluent peak (middle row) and native sediment (bottom row) horizons at selected sites on Palos Verdes margin (refer to Fig. 1 for core locations). The total % aggregates in each sample is calculated as the sum of the differences between aggregated and disaggregated percentages in each size class whenever the aggregated value exceeded the disaggregated value.

several cases, the sand fractions in the effluent-affected sediment became negligible or minor (<10%) components of the disaggregated sediment (Fig. 3) and essentially all of the sand fraction (>63 μm) in a few effluent peak samples (e.g., 112, 174 and 170) consisted of aggregates of smaller particles.

The weight percentage of aggregated, sand-sized grains in the three sediment horizons in 13 box cores along the outer shelf and upper slope from south of Pt. Fermin to northwest of Pt. Vicente is presented in Fig. 4. As expected, aggregate content in the native sediment is small, shows relatively little spatial variation and no relationship to the location of the White's Point effluent diffusers. In contrast, percentages of aggregates in both the effluent peak and surface sediment horizons are clearly related to the location of the waste diffusers. Correlation between aggregate content and the diffusers is particularly evident for the surface sediment at sites 154, 171 and 170, all within 2 km of the diffusers (Figs. 1 and 4). Aggregate percentages also were very large near the diffusers at the time when the effluent-peak horizon was accumulating (most likely in 1970–1972; Fig. 2), but the aggregate fraction is also substantial in effluent-peak sediment up to 10 km alongshelf to the northwest.

3.2. Aggregates in scanning electron microscope images

Scanning electron microscopy (SEM) of selected samples shows that the bulk of the aggregates in all of the effluent-affected layer samples exhibit oval to oblong shapes that are characteristic of fecal pellets produced by benthic deposit-feeders. The pellets in Fig. 5 are representative of the various grain-size classes that were studied. In accordance with the aggregated and disaggregated grain-size results (Fig. 3), the pellets are composed primarily of grains with intermediate diameters <30 μm, and the bulk of the grains appear to be <10 μm, although we made no direct visual counts of the component grains. The SEM images show that the pellets contain a mixture of terrigenous and biogenic particles (Fig. 5). As shown later in this report, the pellets are rich in organic C suggesting that the component particles may be bound together by organic matter that was present in the sediment and/or was secreted by the pelletizing deposit-feeder.

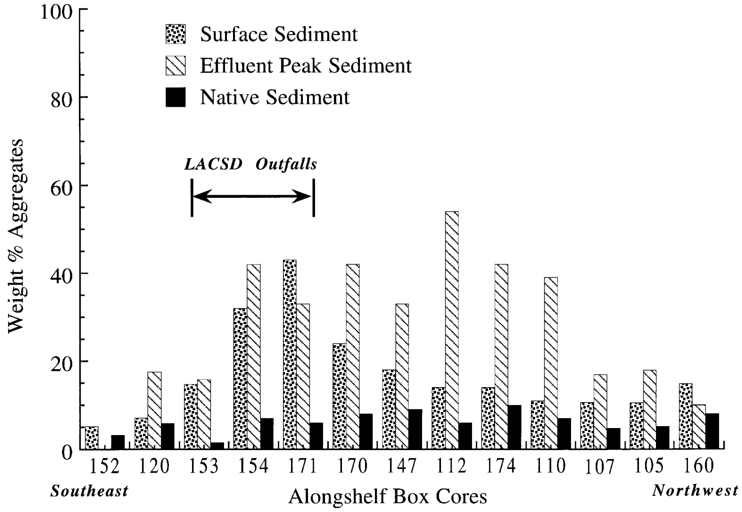

Fig. 4. Weight percent of sand-sized aggregates estimated as described in Fig. 3 in the three sediment horizons at stations located along the outer shelf/upper slope of Palos Verdes margin. The Whites Point (LACSD) effluent diffusers are indicated. Refer to Fig. 1 for core locations.

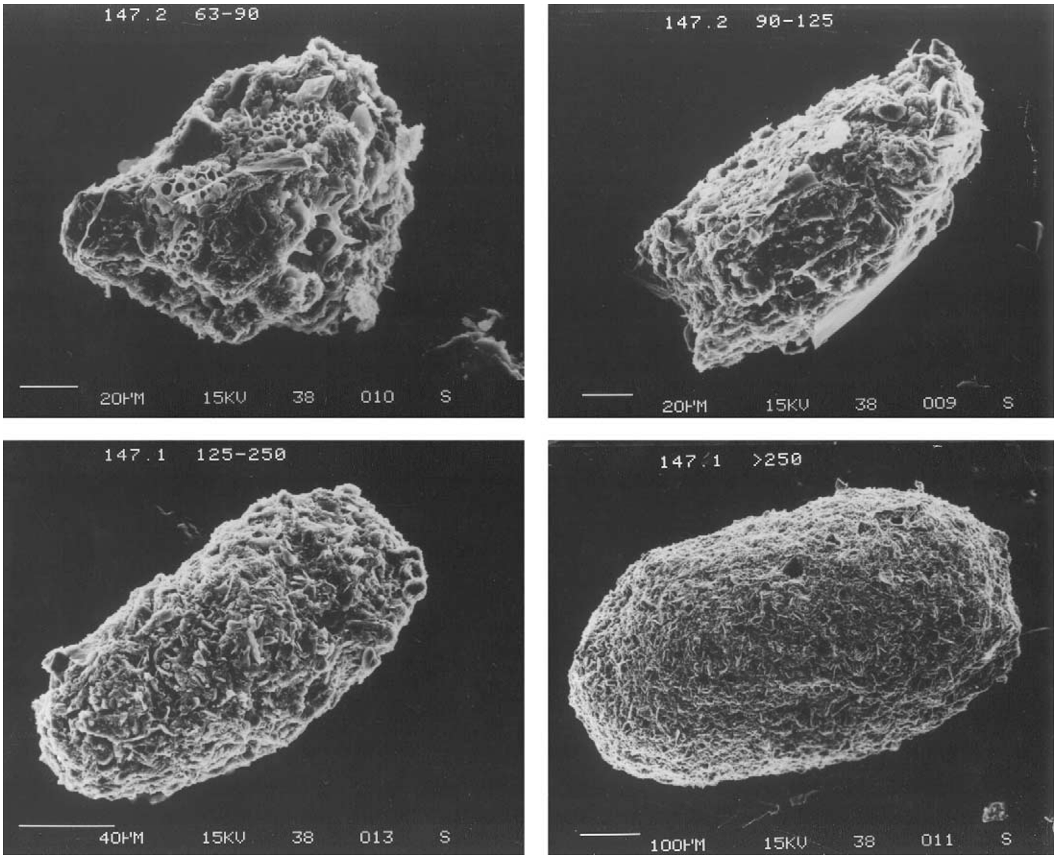

Fig. 5. Images of grain aggregates made with a SEM. The aggregates were selected from within the effluent-affected layer of Core 147 B3 and illustrate examples from each of the sand grain-size classes.

3.3. Settling rates and estimated particle densities

Settling tests on the silt and clay sized ($<63\,\mu m$) fractions of the Palos Verdes samples usually showed only minor deviations of settling velocity from expected values for discrete quartz grains as predicted by Stokes Law. We interpret this to mean that the fine-grained particles are mostly discrete grains or very compact pellet fragments with negligible void space. In contrast, the sand fraction in many of the samples exhibited significant departures from quartz-sphere settling velocity (Fig. 6a). Whenever the sand fractions were composed predominantly of aggregates, mean settling velocities were reduced by factors of 3–5 relative to quartz spheres (Figs. 6a and 7a); usually the decrease was most pronounced for the 125–250 µm size class.

Although the aggregates settle at rates well below those for quartz, the measured settling rates of about 0.5–4 cm/s (Fig. 6a) are much larger than the expected settling rates of the fine-grained material incorporated in the aggregates. For example, the Stokes Law settling velocity for quartz grains that are 20 µm and smaller is <0.03 cm/s. Accordingly, the aggregation process can cause an effective increase in settling rate of as much as two orders of magnitude or more for the finest particles. Changes of that magnitude would have significant effects on the transport of the Palos Verdes sediment during storm events.

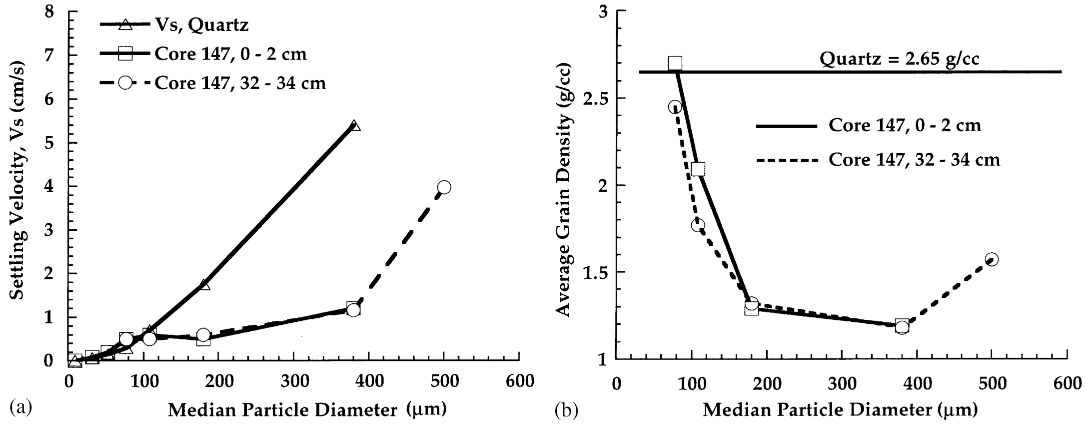

Fig. 6. (a) Average settling rates (cm/s) vs. the median grain diameter for the various size classes in surface and effluent peak samples at box core site 147 (Fig. 1). Settling rate for quartz spheres is shown for reference. (b) the estimated average bulk density (g/cc) for the median diameter particle in each size class with quartz particle density (2.65 g/cc) shown for reference.

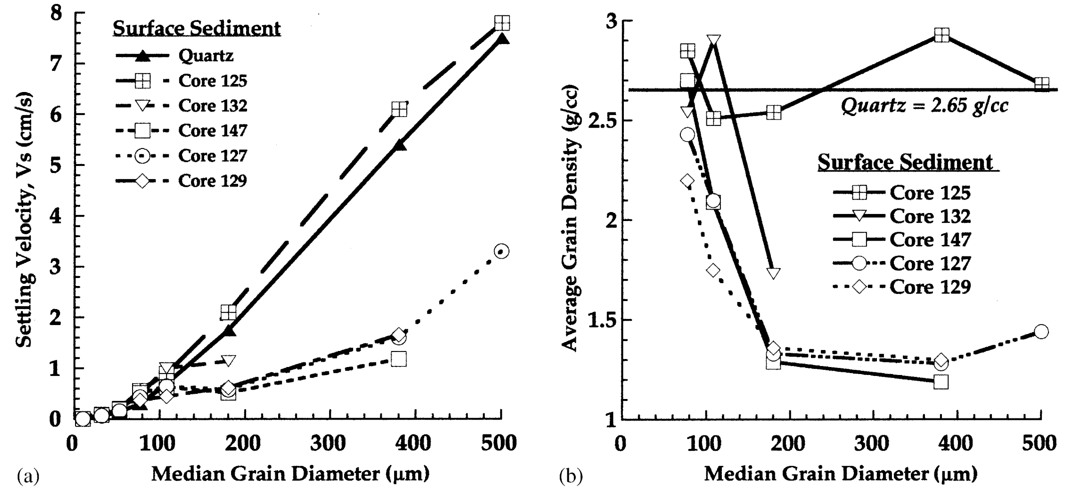

Fig. 7. (a) Average settling rates and (b) average bulk density for the median diameter grains in the sand-sized fractions of surface sediment samples collected at a series of stations that cross the margin from a water depth of about 30 m (site 125) to 300 m (site 129). Sediment at site 125 on the sandy inner shelf contains little effluent-affected material relative to sediment on deeper shelf areas.

Settling velocity is influenced by grain-size, density and shape (Gibbs et al., 1971). SEM images show that the larger (>125 μm) pellets in Palos Verdes sediment tend to be rounded ovoids with Corey shape factors of about 0.7 which is typical of an average mineral grain that exhibits moderately high sphericity according to the study of Baba and Komar (1981). We have noticed that the sphericity of the pellets in our samples tends to be about 10% greater in the 63–90 μm size class, because these smaller sand-sized grains are frequently more equidimensional fragments of pellets. The small fragments are not necessarily as well-rounded as the larger pellets (Fig. 5), but it is uncertain whether slight variations in degree of rounding of particle corners has much effect on settling rates of sand.

Bulk densities for the median diameter of the sand particles in each sieve-size fraction (e.g., 188 μm for the 125–250 μm class) were estimated from the settling velocities measured with the RSA (Fig. 6b) by rearranging the empirical equation of Gibbs et al. (1971) to solve for grain density. In general, the Palos Verdes sediments are composed of particles with quartz densities, or slightly higher, in the finest sand (63–125 μm) sizes (Fig. 6b). However, the coarser sand fractions typically contain more aggregates and are consistently of lower average bulk density then quartz. In samples that contain very high percentages (e.g., >90%) of pellets (Fig. 3 and 7b), the estimated mean grain densities were about 1.2–1.3 g/cc, essentially about 50% of the density of quartz.

The average grain density characteristics of the sand-sized material on Palos Verdes shelf are examined further in Fig. 8 which presents the relationship between estimated median particle density and the percentage of aggregates (pellets) in each size class. The data points include all samples from the native sediment and the effluent-affected layer. Median grain density in these samples ranges from about 1.2 to 2.9 g/cc and tends to vary in an expected manner (inversely) with the percent of aggregates in the size class (Fig. 8).

There are two interesting aspects of the grain density vs. percent aggregate scatter plots in Fig. 8: (1) the data for the very fine sand (63–125 μm) reveal a strong linear relationship, whereas there is an obvious nonlinearity (or "break") in the data for the two larger sand fractions; and, (2) the minimum grain density for the very fine sand size class is about 1.6 g/cc, significantly greater than the minimums in the larger size classes. Explanations for these observations may include the effect

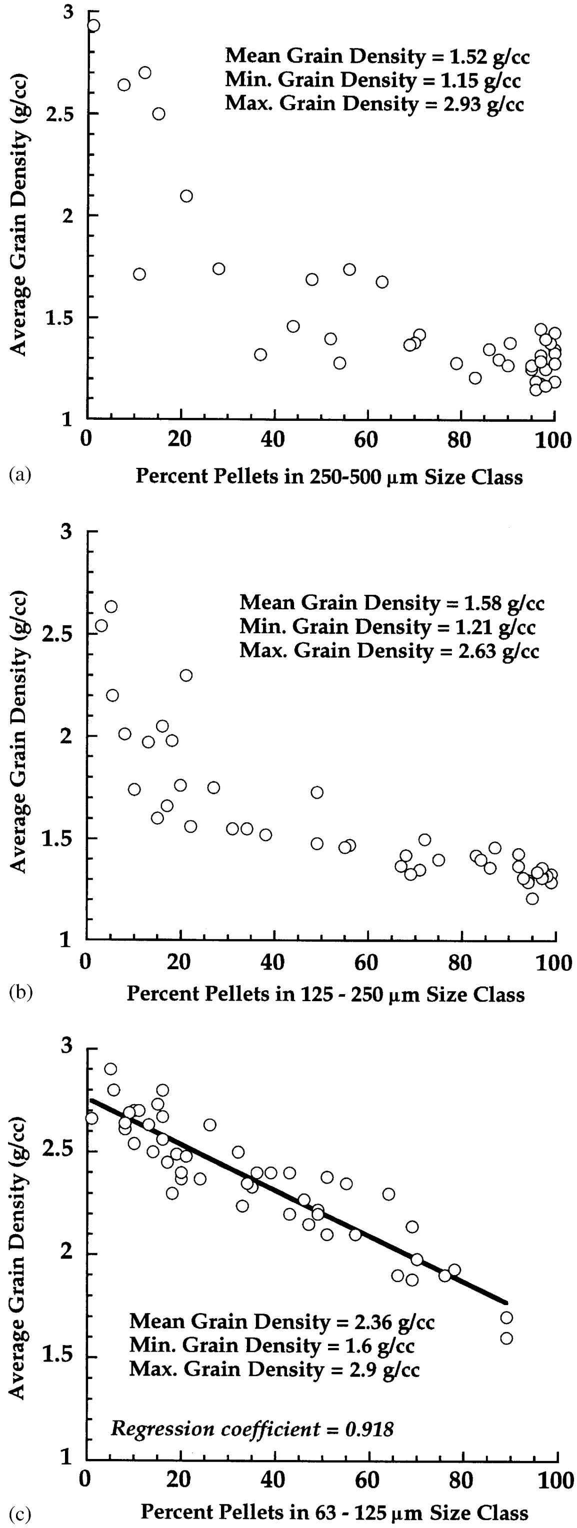

Fig. 8. Estimated wet grain densities as a function of the percentages of aggregates (i.e., pellets) in each of three grain-size classes; (a) 250–500 μm; (b) 125–250 μm; (c) 63–125 μm. All samples from each of the three sediment horizons are included in these graphs. Mean maximum and minimum grain density values for each set of data are shown. These data are derived from settling rate measurements made with the USGS rapid sediment analyzer.

of grain shape variations on settling rates, but we presently do not have enough data to draw firm conclusions.

3.4. Chemical distributions vs. grain-sizes

Distributions of organic C and *p,p′*-DDE in the two effluent-affected layer samples from Core 147 B3 are similar (Fig. 9). Both chemical variables, when presented as a percentage of their total amounts in the sample, exhibit a bimodal distribution with modes in the 125–250 and 0–20 µm size fractions. Organic C is similarly distributed in the two samples. Although we did not analyze disaggregated samples for organic C and *p,p′*-DDE distributions, it seems likely that the bulk of these chemicals are both primarily associated with organic matter coatings on the fine-grained particles. The fact that the organic C and *p,p′*-DDE distributions are similar supports the idea that they occur together in association with organic matter films, coatings and discrete fine-grained particles.

4. Discussion

4.1. Origin of the grain aggregates

Grain aggregates can form through both physical/chemical and biological mechanisms. Aggregates of physical/chemical origin are common in estuaries and near the mouths of rivers where waters containing large concentrations of clay particles mix with saline ocean waters causing flocculation (Whitehouse et al., 1960; Gibbs, 1983; Eisma, 1986; Kranck and Milligan, 1992). The flocs usually exhibit very low density (<1.2 g/cc) because much of the floc is void space. Nevertheless, many flocs can be quite large (mm's in diameter) and therefore they can be an important factor in the settling of sediment from turbid, surface water plumes and also within the bottom nepheloid layer (Owen, 1976; McCave, 1984; Sternberg et al., 1999; Hill et al., 1999). However, owing to the relatively weak forces that hold such flocs together, they can be disrupted easily in a turbulent shearing fluid and it is usual that floc size

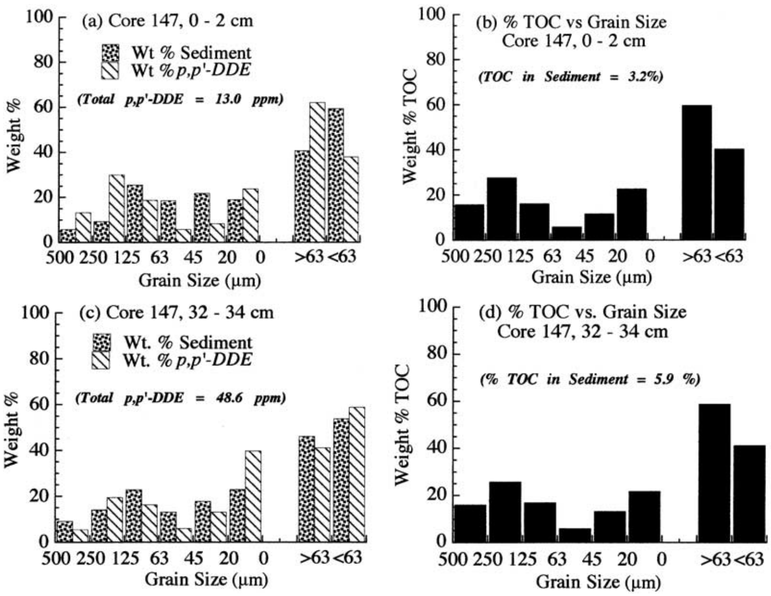

Fig. 9. Distributions of *p,p′*-DDE and organic C vs. aggregated grain-size in surface sediment and effluent peak sediment at core site 147. (a) and (c) show weight % sediment and *p,p′*-DDE vs. aggregated grain-size in surface and effluent-peak samples. The *p,p′*-DDE is presented as the percentage of the total amount of *p,p′*-DDE in each bulk sample. (b) and (d) show the weight % organic C vs. aggregated grain-size in each of the two samples at site 147.

tends to be limited by the turbulence of the fluid. These sorts of aggregates are exceedingly difficult to sample and analyze; in fact, it is unlikely that physical flocs would be preserved during our wet-sieving analysis. They are simply too easily dispersed even when care is taken to gently wet sieve the sediment.

Aggregates that have a better chance of surviving the gentle wet-sieving process are the relatively more compact fecal pellets that are produced by the feeding activities of planktonic and benthonic faunas. In the present case, microscopic examination of samples with large differences in the aggregated vs. disaggregated sand percentages has clearly shown that the aggregates on Palos Verdes margin are predominantly compact, oblong to sub-spherical pellets with dimensions (Fig. 5) that are known to be produced by many species of benthic deposit-feeders (R. Wheatcroft, pers. commun., 1993). We visually estimated the relative amounts of terrigenous and biogenic particles in selected pellets from each sand fraction using both standard light microscopy and electron photomicrographs (see examples in Fig. 5). In most cases, this qualitative test indicated that the Palos Verdes pellets are composed of large amounts of fine-grained terrigenous particles with lesser amounts of biogenic detritus. This suggests pellet production was predominantly controlled by the benthic infauna rather than by zooplankton feeding. The abundance of fine-grained sediment in the aggregates is also compatible with production of pellets by deposit-feeders that may select for organic-rich, fine-grained particles during feeding activities (Taghon, 1982; Wheatcroft, 1992).

Stull et al. (1986b, c, 1996) have summarized an extensive set of data based on biannual collections of benthic infaunas beginning in 1972 at a series of standard stations on the Palos Verdes margin. During the early years of the surveys, the middle and outer shelf areas from the White's Point diffuser system to Pt. Vicente and beyond were covered with organic-rich, effluent-affected mud inhabited by an opportunistic, pollution-tolerant fauna dominated by Polychaeta. Two species that are indicative of contaminated sediment, *Schistomeringos longicornis* and *Capitella* spp. were especially abundant (Fig. 10). Suspended solids discharged by the White's Point system were composed of about 70% organic matter and consistently exceeded 150,000 metric tons/yr during 1968–1971. Much of this material, along with adsorbed chemical compounds like *p*,*p*′-DDE, was deposited along the outer shelf beneath the persistent northwestward mean bottom current described by Noble (2002). The result was the rapid accumulation (at rates of several cm/yr near the diffusers) of organic-rich fine mud, development of anaerobic conditions with significant concentrations of hydrogen sulfide in the sediment, and populations of deposit-feeding Capitellid worms in excess of 1000 individuals per m^2 over large areas of the shelf and upper slope (Fig. 10).

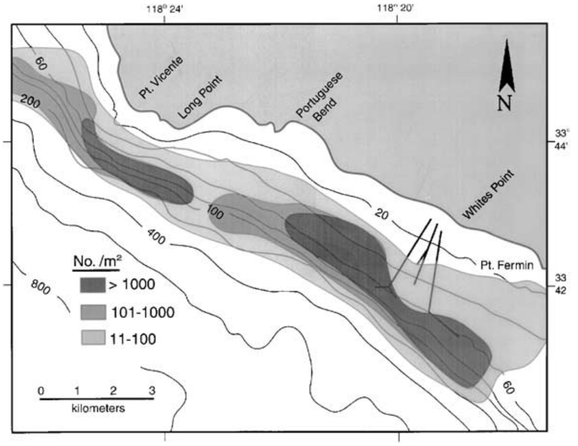

Fig. 10. Distribution of the opportunistic polychaete *Capitella* spp. in the surface sediment on Palos Verdes margin in 1972 as determined by Stull et al. (1986). Capitellid worms are deposit-feeders and prolific producers of sand-sized fecal pellets. They are most abundant in the upper 10 cm of the sediment bed.

The Capitellids are well-known pelletizers that produce relatively compact prolate pellets ranging up to about 1 mm in long dimension (Cuomo and Rhoads, 1987). *Capitella capitata*, one of the species known to have been especially abundant on Palos Verdes shelf in the early 1970s (Stull et al., 1986; B. Thompson, pers. commun., 1994), is a "head-down" deposit-feeder that produces compact and "durable" sand-sized pellets (G. Taghon, pers. commun., 1993). A generic link between the Capitellids and the pellets on Palos Verdes margin is strongly indicated by the strikingly similar distributions of pellet abundance in the effluent-peak samples and the numbers of *Capitella* spp. found by Stull et al. (1986b) in 1972 (compare Figs. 10 and 11b). It seems reasonable to suggest that a substantial part of the aggregates that we observe in the effluent-affected sediment were produced by the Capitellid polychaetes; this generic link is also compatible with the essentially random distribution of pellets in the native sediment below the effluent-affected layer (Fig. 11c).

This interpretation is also supported by recent data that reveals a continuing association between *Capitella* spp. abundance and pellet concentrations in the surface sediment. Presently, grain aggregates comprise up to 40% of the sediment at site 171 near the diffusers (Fig. 11a) and recent benthic community studies (J. Stull, pers. commun., 1994) show that Capitellid worms are present in large numbers near the diffusers. In parallel with the pellet percentages, Capitellid abundance decreases substantially with distance from the diffusers owing to the reductions in particulate effluent discharges that have been achieved by Los Angeles County during the last 20 years and the resultant decreases in organic carbon and nitrogen in the bottom sediment (Stull et al., 2002).

4.2. Pellets and p,p′-DDE

Discharges of DDT's through the Whites Point diffuser system are believed to have begun in the late 1940s and continued until 1971 when the main source, Montrose Chemical Corporation, was disconnected from the Los Angeles County Sanitation Districts (LACSD) wastewater system. Because there was a residue of contaminant still in the collection pipes in 1971, it took several years for DDT concentrations in the effluent to decline to near background values (Stull et al., 1996). Although reliable discharge records for the DDT wastes are not available prior to 1972, in some areas close to the diffusers the effluent-affected layer has recorded a detailed history of the accumulation of contaminated sediment that offers at least a qualitative proxy for actual DDT discharge data. Thus, it is seen that the principal

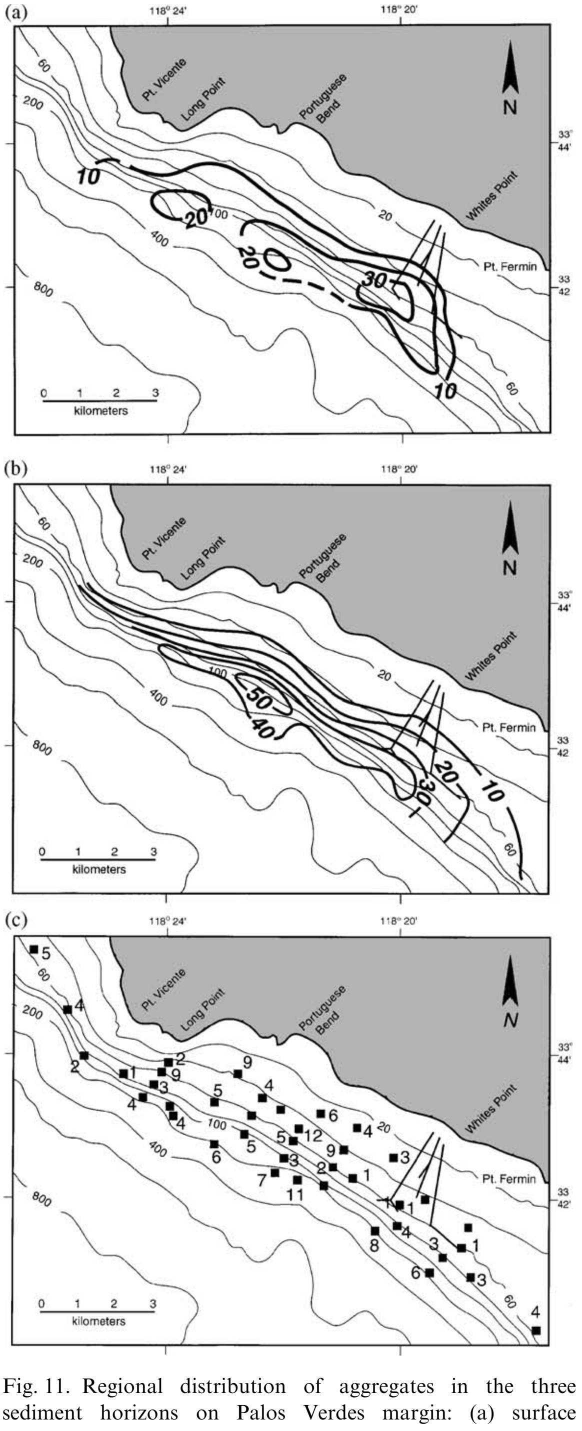

Fig. 11. Regional distribution of aggregates in the three sediment horizons on Palos Verdes margin: (a) surface sediment; (b) effluent peak; and (c) native sediment. Average values in the three layers are about 13%, 22% and 5%, respectively.

DDT degradation product, *p,p′*-DDE, has varied in concentration from undetectable in native sediment to as high as several hundred ppm in the low-density zone of the effluent-affected layer to values of a few ppm to tens of ppm near the sea floor (Fig. 2). Significantly, the core profiles show that the organic-rich sediment of the early 1970s also appears to have carried the highest historical concentrations of *p,p′*-DDE (Fig. 2). It also appears to have been a time when pelletization by deposit-feeders like *Capitella* spp. was especially important, as indicated by the alongshelf distribution of sand-sized aggregates and *p,p′*-DDE in the subsurface of the effluent-affected layer (Fig. 11b and 12). This correlation allows an estimate of the date of the pronounced DDT peak that is so common in Palos Verdes shelf cores, and which provides an important marker horizon for determination of variation in sedimentation rates after 1970/1971 (see Sherwood et al., 1996).

Sewage particles leaving the diffuser system were principally silt and clay-sized and, given the hydrophobic nature of chlorinated hydrocarbons and their tendency to adsorb to particle surfaces, it is likely that initially the DDT's were most concentrated in the 0–20 μm sediment size class. During the late 1960s and early 1970s, the concentration of particulate matter leaving the outfall diffusers was on the order of 250–300 mg/l and physical/chemical flocculation was probably important (Krone, 1962; Drake, 1976). Flocculation may have caused accelerated settling of both fine-grained sewage and ambient particles, explaining the observation that the effluent-affected layer tends to be thickest near the diffusers (Lee et al., 2002). After reaching the seafloor near and downstream (northwest) of the diffusers, pelletization of the sediment by the benthic infauna began. The result of the feeding and production of fecal pellets by the infauna is evident in the distribution of *p,p′*-DDE and organic C in the sediment in Core 147 B3, collected just a few km's northwest of the diffusers (Fig. 9). More than 60% of the total amount of *p,p′*-DDE in the surface sediment of Core 147 B3 now resides in the sand fraction which is composed of about 53% pellets, and more

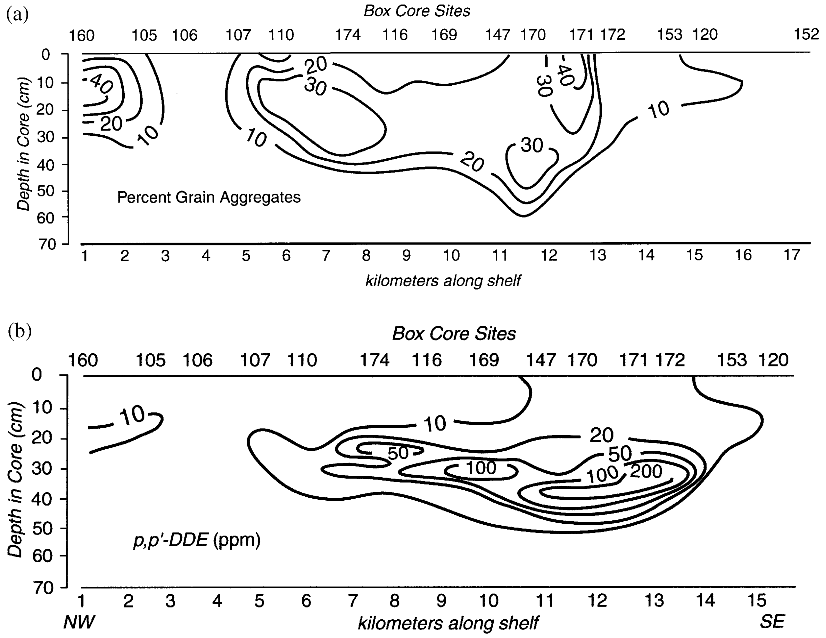

Fig. 12. (a) Percent aggregates in the effluent-affected layer and underlying native sediment as defined by a series of stations along the shelf/upper slope from site 152 to site 160 (see Fig. 1). (b) Concentration (in ppm) of *p,p′*-DDE in sediment along the 60-m isobath as determined in 1992 by the USGS (Lee, 1994; Lee et al., 2002).

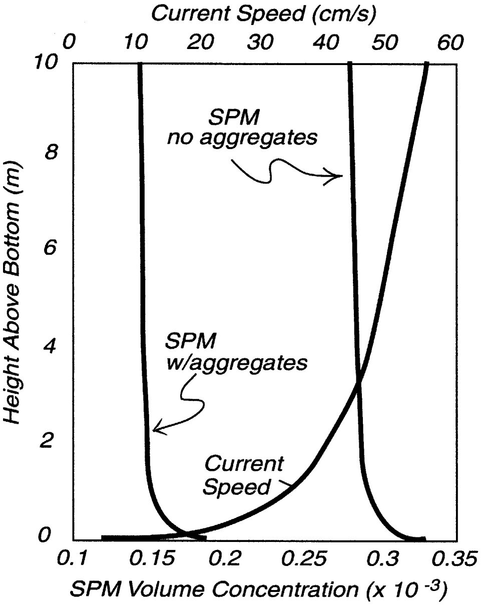

Fig. 13. Current speed (cm/s) and volume concentrations of suspended particulate matter (SPM) in the bottom boundary layer at a site near core site 147 calculated using the model of Glenn and Grant (1987). Current data for this model run were 35 cm/s for the current at 100 cm above the bottom and 25 cm/s for the wave orbital velocity. These conditions are representative of storms that were observed in winter 1992/1993 at about 60 m water depth (Wiberg et al., 2002). An increase of about 100% in suspended matter is predicted by the Glenn and Grant model when disaggregated grain-size data is used instead of the aggregated grain-size distribution observed at core site 147.

than 40% of the *p,p*′-DDE is in the highly pelletized (74%) sand fraction of the effluent-peak horizon (Fig. 9).

It is perhaps noteworthy that most of the *p,p*′-DDE in the effluent-peak sediment is in the 0–20 μm fraction, whereas more is in the 125–250 μm fraction in the sediment at the sea floor. Two closely related explanations for this are: (1) the discharge-induced sedimentation rate was so high in the early 1970s that the benthic fauna had less time to thoroughly pelletize the sediment before it was buried and, consequently, less of the chemical was transferred from the finest size class to the pellets; and/or (2) the difference is a function of the length of time that the sediment remains near or at the seafloor to be affected by physical processes of erosion and resuspension, with preferential chemical desorption from the finer grains followed by redeposition.

We suspect that the second hypothesis is more significant because of the known desorption properties of the chlorinated hydrocarbons (Wu and Gschwend, 1986; Wiberg and Harris, 2002). Specifically, Wiberg and Harris calculate that the grain-size control on the desorption rate for *p,p*′-DDE will produce a relative rapid loss of the chemical from the finest-grained particles when they are suspended in relatively large volumes of sea water (i.e., when the concentration of sediment in the water is small, as would be the case during storm resuspension events). In contrast, large fecal pellets with *p,p*′-DDE adsorbed to their constituent grains will desorb chemical relatively slowly even when suspended in large amounts of sea water. This is because the interior parts of pellets are not in direct contact with the water and therefore do not lose the chemical; only the *p,p*′-DDE near the surface of the pellet is available for desorption on the time scales (hours to 1 or 2 days) of storm-induced resuspension. Therefore, if the sediment remains at the seafloor for a longer time before burial beneath the physically mixed layer (about 1–2 cm thick according to Wiberg et al., 2002), it is likely that more of the *p,p*′-DDE adsorbed to the 0–20 μm particles will be lost to solution and the *p,p*′-DDE concentration mode will shift to the pellets, as is the case in Core 147 B3 in the surface sediment (Fig. 9). Conversely, the effluent-peak horizon, which was near the seafloor for less time owing to the higher rate of sedimentation in the early 1970s (see Sherwood et al., 1996, for a discussion of variations in historical sedimentation rates), would tend to have more of the *p,p*′-DDE in the fine-grained part of the size distribution.

Wiberg and Harris (2002) draw attention to other important chemical effects that are caused by the pelletization of the fine-grained sediment on Palos Verdes margin. For example, because the sand-sized pellets will be somewhat more difficult to lift from the seafloor during storms owing to their relatively large sizes and settling velocities, they will tend to remain close to the seafloor throughout the storm event. Since the concentration of sediment in the water is an important factor in the desorption process, the tendency of the pellets to move near the bed where particle concentrations are typically higher (Fig. 13) will reduce the loss of adsorbed compounds; this is in part because a chemical that desorbs has a greater chance to re-adsorb when particle concentrations are high. In addition, the pellets, which have measured settling rates of 1–6 cm/s (Figs. 6 and 7), will return to the seafloor rapidly as a storm wanes, and thus they will spend less time in suspension than the finer particles which are resuspended several meters above the seafloor and require many hours to settle back to the bed. Finally, we note that pelletization of fine-grained sediment will potentially exert a strong control on the absolute loss of contaminant from the sediment body. In a sense, the fecal pellets act as timed-release capsules giving up adsorbed chemicals at rates that are much lower than they are for the finer-grained individual particles.

4.3. Modeling sediment transport

Some of the dynamical consequences of pelletization have been alluded to in the previous discussion of chemical desorption. It is important to note the following effects that pelletization may have on sediment dynamics:

1. A pelletized sediment may be more readily set in motion by waves and currents because part of the finest size classes have been incorporated into the aggregates and are no longer available to produce cohesive bed effects. Cohesive beds have relatively high erosion thresholds because of the tendency for fine silt and clay particles to be bound together by interparticle forces; for example, grain cohesion is relied upon to limit erosion in clay-lined irrigation canals. Generally, sandy sediment exhibits little or no tendency to develop excess erosion resistance and, thus, pelletization may reduce cohesive bed effects by increasing the sand fraction at the expense of the fine particles.
2. The amount of sediment eroded and placed in suspension by waves and currents depends on the excess shear stress which is the amount by which the wave/current bed shear stress exceeds the threshold shear stress required to move a given particle (Smith, 1977; Grant and Madsen, 1979; Cacchione and Drake, 1991). For non-cohesive sediments, threshold shear stress varies directly with grain-size and density; the threshold for fine sand is about 1.0–1.3 dyn/cm^2 whereas the threshold for non-cohesive silt is of the order of 0.5–1.0 dyn/cm^2 (White, 1970; Miller et al., 1977; Drake and Cacchione, 1986). Consequently, pelletization can cause substantial decreases in the amount of fine-grained sediment placed into suspension during storms. An example that is relevant to the Palos Verdes shelf near the diffusers is shown in Fig. 13 where the results of the bottom boundary layer (BBL) model of Grant and Madsen, (1979, 1982), as amended by Glenn and Grant (1987), are presented. If the artificially disaggregated grain-size distribution is used in the model instead of the actual aggregated size distribution, the BBL model overestimates the amount of sediment in suspension by about 100% (Fig. 13). Flux of sediment by the mean current would be overestimated by a similar amount and calculations of regional flux divergence over a shelf with bed sediment grain-size variations would be degraded (see Sherwood et al., 1996).
3. Previous research on suspended sediment in the marine BBL has shown that concentrations of sediment in suspension commonly were smaller and also decreased vertically more rapidly away from the bed than was predicted by models (Wiberg and Smith, 1983; Cacchione and Drake, 1990; Wiberg et al., 1994). Part of the problem may have been that the bed sediment was partially pelletized and was responding to the dynamics of the wave/current turbulence as a coarser-grained sediment; if the pellet fraction was not accounted for, BBL model predictions would be in error. Wiberg et al. (2002) have used the aggregated grain-size data to successfully model the resuspension and vertical distribution of sediment in the BBL on Palos Verdes shelf.

Apart from the importance of the dynamics of the aggregates, we note, as did Wheatcroft and

Butman (1997), that it is important to assess the degree to which a given sediment is pelletized if field observations of suspended sediment concentrations are made with optical or acoustical instruments. For example, optical variables measured by beam transmissometers and optical backscattering (OBS) devices are strongly influenced by both the concentration and the grain-size distribution of a particle suspension (Baker and Lavelle, 1984; Moody et al., 1987). At the same volume concentration, a change in the grain-size of a suspension from clay to coarse silt will cause beam transmission to increase by a factor of 3–5 (i.e., small amounts of fine silt and clay in suspension have a disproportionately large impact on light transmission and scattering). Consequently, laboratory calibrations of these devices should be carried out with suspensions of known grain-size and interpretation of field data must consider the uncertainty introduced by grain-size variability in the BBL. Physical sampling of the suspended matter as a back-up for the optical measurements is highly recommended.

4.4. Comparison to other studies

The degree to which a sediment is pelletized will vary with the sediment type (e.g., sand is unlikely to undergo pelletization) and the kinds of organisms that inhabit the bed. A large number of organisms feed on detritus, either in suspension or in the bed, and they can rapidly produce great quantities of fecal matter (Haven and Morales-Alamo, 1972). The fecal pellets come in many sizes and shapes (R. Wheatcroft, written commun., 1993) and they vary significantly in their resistance to breakdown by physical and chemical action (Nowell et al., 1981; Taghon et al., 1984; Risk and Moffat, 1977).

It is too early to make general statements about the importance of fecal pellets in mud deposits on continental shelves because only a few recent studies have included both aggregated and disaggregated grain-size determinations. However, aggregates have been shown to be important components of the sediment off the coast of central California (Hyland et al., 1990; Drake et al., 1992), in the STRESS study area off northern California (Wheatcroft and Butman, 1997), and of course on the Palos Verdes shelf off Los Angeles, California (this report). Each of these shelves is characterized by bottom sediment that contains significant silt and clay components seaward of the wave-swept inner shelf.

Interestingly, recent research in an area off the Eel River in northern California appears to underscore the importance of the types of infaunal organisms. The Eel River is one of the largest sediment sources in California and the adjoining shelf is very muddy with new river sediment accumulating at rates of at least 1 cm/yr on large sections of the margin (Wheatcroft et al., 1996; Sommerfield and Nittrouer, 1999). However, in shelf areas where pelletization would seem likely, we are finding that the sediment contains only a small aggregate component (Drake, 1999). Preliminary data on the benthic infauna suggests that it does not include large numbers of deposit-feeders that are known pelletizers, or are known to produce compact, durable pellets (R. Wheatcroft, pers. commun., 1997). On the other hand, studies underway in the Eel shelf "STRATAFORM" project (sponsored by US Office of Naval Research) indicate that physical flocculation in turbid water plumes during river floods can be very significant in determining where the fine sediment is deposited (P. Hill, pers. commun., 1998).

Contaminated sediments like those on the Palos Verdes margin commonly are organic-rich and inhospitable to normal benthic faunas (Pearson and Rosenberg, 1978; Stull et al., 1986c; Wheatcroft and Martin, 1996). Pollution-tolerant, opportunistic species such as *Capitella capitata* can occupy such areas in large numbers as they did on the Palos Verdes shelf in the 1970s, producing great quantities of compact pellets and strongly affecting the physical and chemical properties of the sediment. Similarly, polluted sediments are probably most common in bays and estuaries that receive large industrial and sewage discharges from nearby population centers. If predictive modeling of sediment transport and dispersal of chemical contaminants is to be done successfully in such areas (Sherwood et al., 1996), it is important to determine the aggregation state of the sediment.

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