Published in Sedimentary Geology, v.130, pp. 269–281, (2000).

STREAM–SCANNING LASER SYSTEM, ELECTRIC SENSING COUNTER AND SETTLING GRAIN SIZE ANALYSIS: A COMPARISON USING REFERENCE MATERIALS AND MARINE SEDIMENTS

EMANUELA MOLINAROLI
Dip. Scienze Ambientali, Università di Venezia, Dorsoduro 2137, 30123 Venice, Italy
[E-mail: molinaro@helios.unive.it] **(Author for Correspondence)**

GIOVANNI DE FALCO
International Marine Centre, Loc. Sa Mardini, 09072 Torregrande (OR), Italy
[E-mail: defalco@barolo.icb.ge.cnr.it]

SANDRO RABITTI
Ist. Biologia del Mare, CNR, Riva Sette Martiri 1364/A, 30122 Venice, Italy
[E-mail: rabitti@vecnr8.ibm.ve.cnr.it]

ROSANA ASUNTA PORTARO
CNR, Istituto per la Geologia Marina, via Gobetti 101, 40137 Bologna, Italy

ABSTRACT

Surface and deep-sea core sediments and two sets of standards were measured by three different techniques - Galai Cis I laser system, Coulter Counter TAII, and Micromeritics SediGraph 5000D - in order to compare the Galai results with the other two.

The differences between the three types of measuring device turned out to be greater in sediments than in standards, and were attributed to the physical properties, shape, density and composition of the particles (complexity of the matrix).

Comparison between moment statistics showed that the Galai determines coarser grain sizes than the Coulter and finer than the SediGraph, particularly as regards analysis of surface sediments. The relationships between Galai and SediGraph were estimated using analysis of variation/residuals within individual intervals. The analysis showed a higher variability of residuals for the coarser fractions (8-16 µm and 16-32 µm) with respect to the finer (2-4 µm and 4-8 µm) fractions. The <2 µm SediGraph fraction, with a cut-off at 0.49 µm, showed good correspondence with the <2.5 µm Galai analysis.

Keywords: Marine sediments; Standard materials; Granulometry; Instruments; Comparisons

1. Introduction

Over the last twenty-five years, there has been increasing interest in the significance of grain size data as an indicator of sedimentary environment and processes (Reed et al., 1975; Taira and Scholle, 1979; McLaren and Bowles, 1985; Vandenberghe et al., 1997). Grain size analysis has recently been used in environmental studies, relating fine-grained samples to micro-pollutants in several environments, e.g., marine and transitional (Albertazzi et al., 1987; Menegazzo et al., 1989; Moore et al., 1989; Hieke Merlin et al., 1992; Hathaway et al., 1994; Zonta et al., 1994). Toxic metals are usually bound to marine pelitic sediments, including those found in estuaries and lagoons (Li, 1991; Yücesoy and Ergin, 1992). Marine sediments usually have high clay concentrations, which increase the particle-particle interface.

Concurrently, several electronic systems have been developed, permitting faster and more accurate analyses than the conventional pipette method, and various studies have focused on comparisons between instruments (Syvitski, 1991; Stein, 1985; Konert and Vandenberghe, 1997). Syvitski et al. (1991) described the principal techniques used in modern geological particle size analysis, the precision and accuracy of several methods also providing some recommendations.

Of laser systems for grain size analysis, the Galai Cis-1 is less frequently used than the Malvern Mastersizer, from which it differs in measurement technique. The literature reveals very few works comparing measurements obtained with the Galai and other more popular instruments (Syvitsky et al., 1991; Jantschik et al., 1992). None of these works examines the silty or clayey fractions of natural sediments. For this reason, in the present study the Galai Cis I™ laser system was compared with older systems, i.e., the Coulter Counter TA II™ and Micromeritics SediGraph 5000D™. Comparisons were

carried out using two sets of standards and two sets of samples (surface and deep-sea core sediments), in order to evaluate the differences between instrumental results with increasing matrix complexity.

Each system defines particle size in a different way and thus measures different characteristics of the same material.

Attention focused mainly on the silt-clay mixture fraction, which is the size posing the greatest analytical problems.

2. Instruments: Basic Principles and Operating Procedures

In the *Galai Cis-1* system a laser-based optical analysis channel (using a finely focused He-Ne laser beam, 1.2 µm) employs the theory of "time of transition" in a photo-defined measurement. A wedge prism (600 µm diameter), rotating at a constant speed, scans the incoming laser beam circularly on to a focusing objective, which then scans through the sample measurement volume. The device covers a range from 0.5 to 600 µm of particle diameters and collects signals in 300 discrete size intervals by means of a personal computer. Measurement does not depend on carrier fluid temperature or viscosity.

The Galai Cis-1 is based on a completely different technique from the diffraction laser systems (e.g., Malvern Mastersizer), which use the principle that a particle of a given diameter diffracts a beam of light to a certain angle which increases with decreasing particle size.

The *Coulter Counter model TA II* measures the number and volume of particles suspended in an electrically conductive liquid (e.g., NaCl solution, as used here).

The sampling unit and the signal transfer principle are identical to those of the Coulter Multisizer (Fontolan and Grenni, 1995). Results are related to spherical equivalents of the same electrical resistance (i.e., the same volume) as the measured particles. Suspended particles are sent through a small aperture in a glass tube. Electrodes are attached both inside and outside the aperture. The electrical resistance between the two electrodes changes as soon as the particles pass through the aperture. The amplitude of the voltage pulses thus induced is proportional to the volume of the particles. Measurement does not depend on carrier fluid temperature or viscosity. Pulses are assigned to one of 16 channels pre-calibrated with standards. In terms of diameter, the sequence of channels gives a one-third phi scale.

The *Micromeritics SediGraph 5000D* determines the size distribution of particles dispersed in a liquid, assuming Stoke's settling velocities of particles, by measuring the attenuation of a finely collimated X-ray beam as a function of time and height in a settling suspension. The transmittance of the suspension, which increases with time due to particle sedimentation, is electronically transformed into concentration values and indicated linearly as a cumulative mass percentage on the Y axis of an X-Y recorder. To minimise the time required for analysis, the sample cell is continually lowered with respect to the X-ray beam, so that the effective sedimentation depth decreases with time. The X axis of the recorder is synchronised with the sedimentation cell movement, so that the equivalent spherical diameter, corresponding to time-span and sedimentation depth, is indicated on the abscissa. The mechanism covers a range from 0.2 to 100 µm of particle diameters. The SediGraph resolves particle size to 0.2 µm and reports the unresolved component as a percentage of total sample weight. The SediGraph has a

considerable disadvantage, due to the high quantity of material required for analyses ($4x10^4$ mg l^{-1}).

The advantage of the Galai and Coulter methods is that they use small amounts of sample material (300 mg l^{-1} for Galai; negligible for Coulter). This is useful when the amount of sample material for analysis is low (e.g., aerosol, some cores, suspended sediments).

3. Methods

A total of 27 samples were used for assessment, of which 10 were from deep-sea core sediments, 10 from surface marine sediments, and 7 were standards. Although the small number of samples (20) does not allow us to make any kind of quantification or to generalise our results, we emphasise that this is a preliminary experiment with a significant outcome.

Sediments were examined with particular attention to the silt and clay size range. For this reason, most samples were dispersed by ultrasonic energy in water, and sand was removed on a 63 µm sieve. The deep-sea core sediment samples were mainly silty-clay (93-99% weight percent of samples were <63 µm), whereas the surface sediments were richer in the sandy fraction (20-80% weight percent of samples were <63 µm). The fraction less than 63 µm was treated with H_2O_2 to remove organic matter**.** The dry sample was then subdivided into three subsamples, which were treated in the following way:

Galai Cis-1: samples were dispersed in a 6‰ Na-hexametaphosphate solution for 24 hours and ultrasonically treated (bath) for a time not exceeding 10 minutes. Samples

were suspended in a 5 ml cuvette and analysed. Four analytical replicates were performed and $8x10^4$-$3x10^5$ counts were made.

Coulter Counter: samples were ultrasonically treated (bath) for a time not exceeding 10 minutes. They were then dispersed in a 3% NaCl electrolyte solution, surfactant dispersant (Coulter dispersant) was added, and the samples were immediately analysed. Tubes with apertures of 50 µm and 280 µm were used to cover the range from 0.63 to 128 µm. High-resolution size distributions were achieved with four analytical replicates ($5-7x10^4$ particles counted) and then averaged. The resulting data sets were combined by matching and recomputing the value fractions under the combined intervals to 100%. Data were acquired by an automatic acquisition system composed of an IBM-AT computer connected to the Coulter Counter main unit through an interface adapter (Boldrin et al., 1986).

SediGraph: the sample fraction <63 µm was dispersed in a 6‰ Na-hexametaphosphate solution and ultrasonically treated (bath) for a time not exceeding 10 minutes. The density of quartz was assumed for average particle density. Analysis temperature was 30°C, i.e., a density of 995.7 kg m^{-3} and a viscosity of 0.8007 Pa s for the liquid. Samples weighing 2.5 g were used.

The lower limits of the size range is different for the three devices (<0.23, 0.5 and 0.63 µm for SediGraph, Galai and Coulter respectively). Consequently, the <0.5 µm size fraction is below the detection limit for the Galai and Coulter systems. Therefore, the correct way to compare results from the three instruments for the overall set of samples is to consider the grain size intervals which they detect. For SediGraph results, it was consequently decided to set the cut-off at the lower limit of 0.49 µm and then to normalise the data to 100%.

Statistical parameters (mean, median and sorting) and frequency distribution differences were computed to compare results obtained with the three analysers: median and mean indicate the central tendency of frequency distribution; the sorting coefficient is a measure of the standard deviation. Detailed analysis of the differences for each pair of instruments was made using the difference of frequencies for each grain size interval. Frequencies were computed for 7 grain size intervals (< 1; 1-2; 2-4; 4-8; 8-16; 16-32; and 32-63 µm) and the mean differences of frequency for each interval and pair of instruments were computed.*

4. Results and Discussion

4.1 Natural and Synthetic Standards

The preliminary phase of this study consisted of a comparison of the Galai, Coulter and SediGraph systems using two sets of standards: (i) a natural reference material (garnet, median values of 2.5, 3.85, and 12 µm (8.64, 8.02 and 6.38 ϕ), and (ii) synthetic standard polystyrene divinyl benzene (P.D.V.B) and latex spheres, median values 5.22, 9.0, 13.9 and 19.1 µm (7.58, 6.80, 6.17 and 5.71ϕ).

The garnet standard has a density of 3.85 g cm^{-3}, is produced by Micromeritics, and is regularly used as a reference material (for calibration) for the SediGraph. A quantity of 0.5 g in 25 ml of deionized water containing 0.05 weight percent of sodium metaphosphate is usually adopted during testing.

The latex spheres standard is regularly used as a reference for the Coulter Counter. The spheres are durable and do not change in size in most of the electrolyte solutions used

* The raw data, which cannot be printed here for want of space, may be obtained by writing to the authors.

with the instrument. A number from 1 to 5 drops per 50 ml are usually adopted for both 50 µm and 280 µm aperture sizes.

Figure 1 shows scatter plots of median values for the two standards analysed using the three instruments and taking into account the fact that garnets and latex spheres are used as an internal test for SediGraph and Coulter respectively, to check if the instrument is working well. Comparison between median values must be carried out using the phi scale as it is linear.

The Coulter results for the 8.64, 8.02 and 6.38 ϕ garnets were shifted slightly, median values falling at 8.58 (-0.7%), 8.20 (2%) and 6.48 (2%) ϕ respectively. The Galai results for the same size garnets were also shifted towards coarser values, but with greater differences, median values falling at 8.07 (-7%), 7.80 (-3%), and 6.05 (-5%) ϕ. Figure 2 compares frequency histogram distributions for the three garnet standards analysed by Galai and Coulter. The Galai results for the 7.58, 6.80, 6.17, and 5.71 ϕ latex spheres were slightly shifted towards greater median values. The results plotted at 7.43 (-2%), 6.70 (-1%), 6.14 (-0.5%), and 5.67 (-0.7%) ϕ respectively. These particular standards cannot be analysed by the SediGraph, due to their synthetic composition.

4.2 Natural Sediments

The differences between the mean and sorting values of Galai, Coulter and SediGraph were compared using the linear phi scale (as above) (Tab. 1**).**

The mean values estimated by the Galai and Coulter instruments for core samples differed by a maximum of 0.61 ϕ, whereas those estimated by the SediGraph showed differences of up to 1.51 ϕ with respect to Galai and 1.57 ϕ with respect to Coulter. As regards values obtained by applying a cut-off of 0.49 µm to the SediGraph data, the

differences among the means produced by the various instruments fell greatly: up to 0.3 ϕ between Galai and SediGraph, and up to 0.91 ϕ between Coulter and SediGraph.

Greater variability was found between the means measured by the three instruments for surface samples. Those of Galai were slightly lower (coarser grain sizes) than those of Coulter, where higher values, up to 1.96 ϕ, were found with respect to SediGraph, using a cut-off value of 0.49 µm. The differences were accentuated when comparing Coulter and SediGraph (up to 2.34 ϕ). A qualitative check of surface samples was also carried out under the optical microscope, and revealed the presence of particles between 20 and 30 µm. From a mineralogical viewpoint, the presence of opaques and heavy minerals was shown.

The sorting estimates of Galai were lower than those of Coulter in almost all samples with larger deviations in the deep-sea core sediments (Tab.1).

The analysis of moment statistics showed the good fit between the measurements recorded by the three instruments on core samples (finer fraction), whereas there were obvious differences in surface samples (coarser) for which Galai determined larger grain sizes than Coulter and finer than SediGraph. These differences were probably due to the lack of precision of Coulter in measuring high-density coarse particles, which do not long remain suspended in the aqueous suspension used in this work.

4.3 Comparison of standards with natural sediments

Figure 3 compares the median values of standards and sediments obtained using Galai, Coulter and SediGraph. The dissimilarity between the results increased with increasing sample heterogeneity. In the Galai *vs.* Coulter comparison (Fig. 3A), the median values of the latex spheres plotted very close to the bisectrix, those of garnets were close to it,

and those of sediments were highly scattered. It must be borne in mind that latex spheres represent a perfect suspension, because the particles are truly spherical. Comparisons of SediGraph *vs.* Galai and SediGraph *vs.* Coulter showed the same trend (Fig. 3B, C): garnet median values were close to the bisectrix and sediment median values were scattered along the plane, with an increase in differences for coarser surface sediments. Since the three methods measure different physical properties, these deviations may be explained by irregularly shaped grains and different particle densities. The SediGraph defines a particle diameter as equivalent to that of a sphere settling in the same liquid at the same speed as the particle of unknown size. The density of quartz is assigned to the sphere.

The presence of heavy minerals observed under the optical microscope indicates that, although their sedimentation rate is higher than that of quartz, they may be the cause of the shift towards coarser values shown by SediGraph on surface samples.

Comparisons of the difference in frequency distributions between Galai and Coulter showed that the dissimilarities between the instruments were in the 1-2 µm (Galai lower ~ 10%) and 4-8 µm size intervals, with higher values for Galai in the coarser interval (Galai higher ~ 15% for deep-sea core; ~ 8% for surface sediments). The Galai instrument therefore shifted towards coarser fractions in both surface and deep-sea sediments (Fig. 4 A and B).

Comparisons of the difference in frequency distributions of SediGraph *vs.* Galai and Coulter showed differences between deep-sea core and surface sediments. SediGraph detected more particles in the <1 µm size interval in deep-sea core samples (Fig. 4 C and E), and in the >32 µm in surface samples (Fig. 4D and F). Galai *vs.* SediGraph frequencies were on average higher in the 2-4 µm and 4-8 µm intervals, by ~ 8% and

10-13% in the overall sets of samples respectively; in contrast they were ~ 8% less in the <1 µm in deep-sea core samples and ~ 20% less in the 32-63 µm interval in surface sediments.

A mineralogical study (Guerzoni et al., 1996) showed that our deep-sea core sediments included calcite, dolomite and clay minerals (illite, chlorite, smectite, kaolinite) in the <2 µm fraction. Analysis of the samples by SEM also showed that some of the samples were biogenic, mostly coccoliths and diatom frustules, with a density much lower than that of quartz grains (2.65 g cm^{-3}). In particular, one deep-sea core sediment was classified as a tephra, i.e., rich in volcanic glass. Particle shape and density are likely to be the most important factors in measuring grain size.

In the deep-sea core sediments the low density of biogenic components and the shape of clay minerals may explain the dissimilarities in the finer grain size interval between the SediGraph and the other devices. In the surface sediments, the higher density of heavy minerals may explain the SediGraph shift towards more coarser values, with respect to Coulter and Galai.

Therefore, all the techniques that employ Stoke settling (pipette analysis, SediGraph etc.) are likely to be inaccurate for determining particle size in samples which contain mixtures of materials.

4.4 Relation between Galai and SediGraph analysis

The relationships between the data obtained by the two instruments were estimated using variation/residuals analysis within individual intervals. Although the low number of samples (20) did not allow us to make a generalised statement of our results, we emphasise that this is a preliminary attempt with a significant outcome.

The residual values between Galai and SediGraph data were derived using the following formulae:

Residual values = (% Galai – % SediGraph) / % Galai

Residual values were plotted *vs.* SediGraph data in Figure 5.

The residuals are normalised factors which are not dependent on the magnitude of the frequency in the considered grain size interval. The residual values tend to zero in comparison to coincidental data series and tend to constant values for linearly correlated data series. Variable residual values indicate that there is no a linear correlation between the series.

In the plots of Fig. 5, residual values were linearly related to Sedigraph data in the 2-4 µm ($r=-0.82$, $p<0.001$; slope=-0.022) and 4-8 µm ($r=-0.88$, $p<0.001$; slope=-0.022) intervals. The correlation coefficient decreased in the 8-16 µm interval, and there was a corresponding increase in the slope ($r=-0.73$, $p<0.001$; slope=-0.054), which indicated an increase in the variability of the residuals. Significant correlation was not found in the 16-32 µm ($r= 0.39$, $p=0.082$) interval.

The increase in the variability of the residuals in the coarser grain size intervals (8-16 µm and 16-32 µm) may indicate a greater heterogenity of physical properties (shape and density) of the particles.

The <2 µm SediGraph values were compared with the <2 µm, <2.5 µm, <4 µm and <8 µm Galai values. Table 2 lists comparable Galai and SediGraph fractions.

Deep-sea core sediments showed that the SediGraph <2 µm and the Galai <4 µm fractions plot in the ranges 45.5-53.8% and 47.1-53.6% respectively. Surface sediments showed greater differences (SediGraph: 21.0-47.5%; Galai: 21.3-49.4%). Comparison

between the <2 µm and <8 µm fractions showed very large differences (Galai <8 µm core: 77.4-85.7%; surface sediments: 47.0-84.1%).

Similar comparisons are given for SediGraph results with cut-off at 0.49 µm and Galai results (Tab.2). The deep-sea core sediments showed that the SediGraph <2 µm and the Galai <2.5 µm fractions plotted in the ranges 25.4-34.5% and 27.9-31.6% respectively. In short, the SediGraph <2 µm fraction matches better with the Galai <4 µm fraction.

Our results showed that the <2 µm SediGraph values without cut-off correspond to the <4 µm Galai analysis ($r = 0.79$, $p < 0.01$), while those of the <2 µm SediGraph with cut-off correspond to the < 2.5 µm Galai analysis ($r = 0.76$, $p < 0.01$).

In Figure 6, residuals derived from comparison of <2 µm SediGraph values, and <2 µm and <2.5 µm Galai values were plotted *vs.* <2 µm SediGraph values. The <2.5 µm Galai corresponded better with <2 µm SediGraph than with the <2 µm Galai. Indeed, with the exception of three surface sediments, the residual values computed between <2 µm Sedigraph and <2.5 µm Galai were close to zero (±0.2). In contrast, residuals computed between <2 µm Sedigraph and <2 µm Galai were shifted towards negative values (from 0 to –0.6).

We conclude that the size yielded by SediGraph will always differ from that determined by Galai. Nevertheless, the method of measuring grain size intervals in the two devices, using the lower SediGraph cut-off limit of 0.49 µm, allows better comparison of data.

The same type of comparison between SediGraph and Coulter was attempted, but the results were very poor (Tab. 2).

5. Conclusions

1. Discrepancies in results among the three instruments increased from standards to sediments due to greater matrix complexity. Differences were attributed to the variability of physical properties (shape, density etc.) due to the heterogeneity of particle composition, confirmed by microscope analysis.
2. The comparison between moment statistics demonstrated that Galai determined coarser grain sizes than Coulter and finer than SediGraph. The differences were more marked in analyses of surface sediments, which were coarser, for which Galai provided mean values which are up to 1.12 ϕ lower than those of Coulter and up to 1.96 ϕ higher than those of SediGraph. The comparison between moment statistics may be carried out only considering the analytical range common to all three instruments, with a SediGraph data cut-off value of 0.49 µm.
3. Comparisons among the frequencies calculated for each grain size intervals (<1; 1-2; 2-4; 4-8; 8-16; 16-32; and 32-63 µm) revealed differences among the measurements supplied by the instruments. In Galai *vs.* Coulter analysis frequency values were ~10% lower in the 1-2 µm interval, and ~15%, (deep-sea core) and ~8%, (surface sediments) higher in the 4-8 µm interval.
4. The analysis of residuals within individual intervals between SediGraph and Galai showed a higher variability of residuals for the coarser (8-16 µm and 16-32 µm) compared to the finer fraction. This may indicate a greater heterogenity in physical properties (shape and density) of the particles in these coarser grain size intervals.

   The <2 µm SediGraph analysis, with cut-off at 0.49 µm, corresponded well with the <2.5 µm Galai analysis.

Acknowledgements

This work was partly supported by the Regione Autonoma della Sardegna (STRIDE Programme). The authors would like to thank Ms G. Walton for revision of the English text.

References

Albertazzi, S., Hieke Merlin, O., Menegazzo Vitturi, L., Molinaroli, E., Tassi Pelati, L., 1987. Distribution and behaviour of ^{137}Cs in nearshore sediments of the northern Adriatic Sea and at the Adige River estuary, northern Italy. Applied Geochemistry, 2, 357-366.

Boldrin, A., Cesca, A., Rabitti, S., 1986. An automatic data acquisition system for Coulter Counter Model TA II. Bollettino di Oceanologia Teorica a Applicata, IV, (3), 159-164.

Guerzoni, S., Portaro, R., Trincardi, F., Molinaroli, E., Langone, L., Correggiari, A., Vigliotti, L., Pistolato, M., De Falco, G., Boccini, V., 1996. Statistical analyses of grain-size, geochemical and mineralogical data in core CM92-43, Central Adriatic basin. In: Guilizzoni, P., Oldfield, F. (Editors), Paleoenvironmental Analysis of Italian Crater Lake and Adriatic Sediments, Memorie Istituto Italiano Idrobiologia, 55, 231-245.

Hathaway, J.C., Buchholz, ten Brink, M.R., Manheim, F.T., Bothner, M.H., 1994. Contaminated sediments in Boston Harbor and Massachusetts Bay: spatial and temporal trends. EOS, Trans. Am. Geophys. Union ,75, (3), 144.

Hieke Merlin, O., Menegazzo Vitturi, L., Molinaroli, E., Morani, A., Tassi Pelati, L., 1992. Chernobyl fallout and radionuclide activity depth-profiles in sediments at the River Adige estuary (Northern Adriatic Sea, Italy). Terra Nova, 4, 434-442.

Konert, M., Vandenberghe, J., 1997. Comparison of laser grain size analysis with pipette and sieve analysis: a solution for the underestimation of the clay fraction. Sedimentology, 44, 523-535.

Jantschik, R., Nyffler, F., Donard, O.F.X., 1992. Marine particle size measurement with a stream – scanning laser system. Marine Geology, 106, 239-250.

Li, Y.H., 1991. Distribution pattern of the elements in the ocean: A synthesis. Geochim. Cosmochim. Acta, 55, 3223-3240.

McLaren, P., Bowles, D., 1985. The effects of sediment transport on grain-size distributions. J. Sed. Petrology, 55, 457-470.

Menegazzo Vitturi, L., Molinaroli, E., Pistolato, M., Rampazzo, G., 1989. Sediment properties and their influence on the geochemical composition in the Lagoon of Venice. Boll. Oceanogr. Teor. Appl., 7, 91-205.

Moore, J.N., Brook, E.J., Johns, C., 1989. Grain size partitioning of metals in contaminated coarse-grained river floodplain sediments: Clark Fork River, Montana U.S.A.. Environ. Geol. Water Sci., 14, (2), 107-115.

Reed, W.E., leFever, R., Moir, G.J., 1975. Depositional environment interpretation from settling velocity (psi) distribution. Geol. Soc. America Bull., 86, 1321-1328.

Stein, R., 1985. Rapid grain-size analyses of clay and silt fractions by SediGraph 5000D: comparison with Coulter Counter and Atterberg methods. J. Sed. Petrology, 55, 590-593.

Syvitski, J.P.M., 1991. Principles, methods, and application of particle size analysis. Cambridge University Press, New York, p. 368.

Syvitski, J.P.M., LeBlanc W., Asprey, D.W., 1991. Interlaboratory instrument calibration experiment. In: Syvitski, J.P.M. (editor), Principles, Methods, and Application of Particle Size Analysis. Cambridge University Press, New York, pp. 174-193.

Taira, A., Scholle, P.A., 1979. Discrimination of depositional environments using settling tube data. J. Sed. Petrology, 49, 787-800.

Vandenberghe, J., Zhisheng, A., Nugteren, G., Huayu, L., Van Huissteden, K., 1997. New absolute time scale for the Quaternary climate in the Chinese loess region by grain-size analysis. Geology, 25,1, 35-38.

Yücesoy, F., Ergin, M., 1992. Heavy-metal geochemistry of surface sediments from the southern Black Sea shelf and upper slope. Chem. Geol., 99, 265-287.

Zonta, R., Zaggia, L., Argese, E., 1994. Heavy metals and grain-size distributions in estuarine shallow water sediments of the Cona marsh (Venice Lagoon, Italy). Sci. Total Environ., 151, 19-28.

Figure Legends

Fig. 1 Comparison of median values for reference standards: (A) garnets (SediGraph standard). Galai and Coulter results shown in X axis, standard reference values in Y axis; (B) latex spheres (Coulter Counter standard) analysed by Galai. Galai results shown in X axis, standard reference values in Y axis.

Fig. 2 Comparison of frequency histogram distributions of three garnet standards analysed with Coulter and Galai.

Fig. 3 Median values for sediments and standards (latex spheres and garnets). (A) Galai vs. Coulter; (B) SediGraph vs. Galai; (C) SediGraph vs. Coulter.

Fig. 4 Mean differences between frequencies for grain size intervals reported in X axis. A, C, E: deep-sea core sediments. B, D, F: surface sediments. Standard deviations are indicated by bars.

Fig. 5 Residual values (see text for explanation) plotted *vs.* SediGraph 2-4 µm, 4-8 µm, 8-16 µm and 16-32 µm fractions.

Fig. 6 Residual values derived from comparison of <2 µm Sedigraph values and <2 µm and <2.5 µm Galai values plotted *vs.* <2 µm SediGraph values.

Table 1. Galai, Coutler and SediGraph mean, median and sorting statistics for deep-sea core (C) and surface sediments (S).

Samples	Galai Mean		Coulter Mean		SediGraph Mean		SediGraph Mean*		Galai Median		Coulter Median		SediGraph Median		SediGraph Median*		Galai Sorting	Coulter Sorting	SediGrap Sorting	SediGrap Sorting*
N.	ϕ	µm	ϕ	µm	ϕ	µm	ϕ	µm	ϕ	µm	ϕ	µm	ϕ	µm	ϕ	µm	ϕ	ϕ	ϕ	ϕ
C-525	8.2	3.5	8.2	3.3	9.7	1.2	8.5	2.7	8.0	4.0	8.5	2.7	9.3	1.6	8.2	3.4	1.2	1.5	2.7	1.6
C-534	8.1	3.8	8.5	2.8	9.4	1.5	8.5	2.7	8.0	4.0	8.8	2.3	9.1	1.8	8.2	3.5	1.2	1.4	2.5	1.7
C-545	8.1	3.8	8.2	3.4	9.6	1.3	8.4	2.9	8.0	4.0	8.5	2.7	9.2	1.7	8.1	3.8	1.3	1.7	2.7	1.7
C-554	7.9	4.1	7.8	4.5	9.6	1.3	8.4	3.0	7.9	4.2	8.1	3.6	9.1	1.8	8.0	3.9	1.3	1.7	2.7	1.7
C-564	7.9	4.2	8.3	3.3	9.7	1.2	8.5	2.8	7.9	4.2	8.6	2.5	9.3	1.6	8.1	3.7	1.3	1.5	2.7	1.6
C-574	8.1	3.5	7.9	4.3	9.2	1.7	8.3	3.1	8.0	3.9	8.0	4.0	8.8	2.2	7.9	4.1	1.2	1.5	2.6	1.7
C-584	8.1	3.6	8.0	4.0	9.0	1.9	8.3	3.1	8.0	3.9	8.0	3.8	8.7	2.5	7.9	4.3	1.3	1.6	2.7	1.7
C-594	8.1	3.5	7.8	4.4	9.4	1.5	8.3	3.1	8.0	3.9	8.0	4.0	8.9	2.1	8.0	4.0	1.2	1.3	2.8	1.6
C-604	8.1	3.6	8.8	2.3	8.8	2.2	8.1	3.7	8.0	3.9	8.8	2.3	8.3	3.2	7.6	5.1	1.2	0.7	3.1	1.7
C-614	7.9	4.1	8.2	3.3	9.5	1.4	8.3	3.1	7.9	4.2	8.5	2.7	9.0	2.0	7.9	4.2	1.3	1.2	2.9	1.6
S-89	7.3	6.4	7.5	5.5	8.7	2.4	7.6	5.2	7.5	5.4	7.9	4.3	8.4	3.0	7.2	6.9	1.6	1.7	3.0	2.1
S-26	6.7	9.9	7.8	4.6	6.7	9.4	5.9	17.1	6.1	14.6	8.0	3.9	5.3	24.8	5.0	30.6	1.4	1.7	2.8	1.7
S-B1	7.2	6.9	7.4	6.0	8.1	3.7	7.3	6.2	7.2	6.6	7.1	7.2	7.5	5.7	6.9	8.6	1.4	1.6	2.7	1.9
S-B2	7.1	7.5	7.5	5.7	8.0	4.0	7.1	7.1	7.2	6.9	7.3	6.4	7.2	6.6	6.6	10.1	1.5	1.7	2.7	1.9
S-B7	7.4	6.0	7.7	4.7	8.2	3.3	7.1	7.2	7.5	5.6	7.8	4.5	7.5	5.6	6.4	11.5	1.5	1.6	3.0	2.0
S-B8	6.9	8.6	7.0	7.7	5.7	18.6	5.6	20.8	6.8	9.2	7.0	7.7	5.4	24.0	5.3	25.0	1.4	1.6	1.9	1.3
S-F381	7.8	4.6	7.9	4.3	6.0	15.8	5.2	26.8	7.8	4.5	8.5	2.7	4.9	32.8	4.8	35.0	1.4	1.6	2.3	1.2
S-F382	6.7	9.8	7.4	6.0	6.3	12.9	5.5	22.1	6.7	9.7	7.5	5.4	5.0	30.3	4.9	33.1	1.7	1.7	2.5	1.4
S-F383	6.9	8.5	7.6	5.2	6.8	8.7	5.8	18.5	6.7	9.4	7.8	4.5	5.3	25.5	5.0	30.6	1.6	1.9	3.2	1.6
S-F44	8.1	3.6	8.2	3.5	8.7	2.4	7.7	4.7	8.0	4.0	8.4	3.0	8.7	2.4	7.2	6.8	1.3	1.3	3.3	2.2

* SediGraph results with the cut off at 0.49 µm

Table 2
Comparison between SediGraph and Galai systems of different size fractions for deep-sea core (C) and surface sediments (S)

Samples N.	SediGraph < 2µm (>9ϕ)	Galai < 4µm (>8ϕ)	Galai < 8µm (>7ϕ)	SediGraph < 2µm* (>9ϕ)	Galai < 2.5 µm (>8.6ϕ)	Coulter < 4µm (>8ϕ)	Coulter < 2µm (>9ϕ)
C-525	53.8	53.6	85.7	34.0	30.6	63.7	34.3
C-534	52.0	49.9	83.3	34.5	28.0	67.79	40.9
C-545	52.3	50.4	81.1	31.4	29.6	62.11	36.5
C-554	52.0	47.1	77.4	30.9	28.2	52.27	30.0
C-564	53.5	47.9	78.6	31.6	27.9	66.18	37.2
C-574	48.0	52.0	84.6	29.3	30.2	48.48	26.7
C-584	46.3	51.5	82.0	27.1	31.4	52.67	29.3
C-594	50.0	52.2	85.1	30.1	29.6	49.92	21.1
C-604	45.5	51.2	84.3	25.4	30.1	88.14	38.8
C-614	51.7	47.4	77.6	28.8	26.4	65.77	32.8
S-89	42.5	34.1	62.4	21.2	18.1	47.57	21.6
S-26	21.2	21.3	39.5	8.1	11.6	51.36	31.0
S-B1	34.3	28.4	55.3	19.2	15.2	34.59	20.5
S-B2	31.2	28.1	53.3	15.8	15.3	36.98	21.8
S-B7	36.6	35.1	60.6	17.7	20.1	46.33	27.4
S-B8	9.8	22.9	47.0	3.6	10.8	27.74	14.2
S-F381	14.7	44.0	71.8	4.4	26.5	65.31	35.9
S-F382	17.2	25.5	46.8	5.6	13.3	41.06	21.6
S-F383	21.6	28.0	47.7	7.5	15.8	46.33	28.7
S-F44	47.5	49.4	84.1	27.2	28.2	62.09	25.4

* SediGraph results with the cut off at 0.49 µm

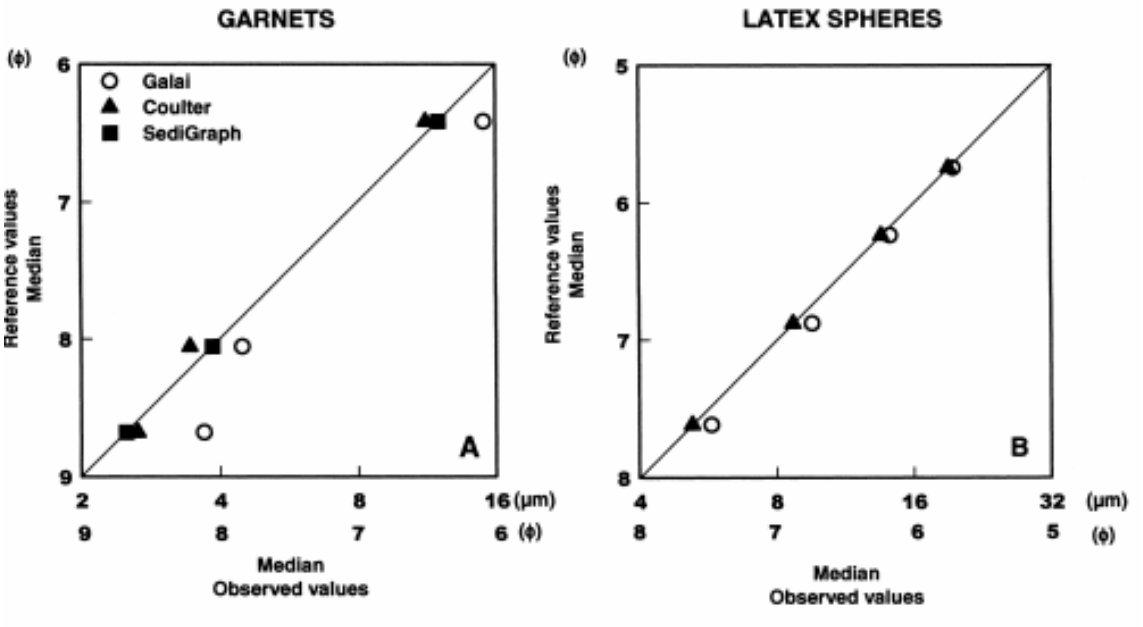
GARNETS
(ϕ)
6
7
8
9
Galai
Coulter
SediGraph
Reference values
Median
A
2
4
8
16 (μm)
9
8
7
6 (ϕ)
Median
Observed values
LATEX SPHERES
(ϕ)
5
6
7
8
Reference values
Median
B
4
8
16
32 (μm)
8
7
6
5 (ϕ)
Median
Observed values

FIG. 1

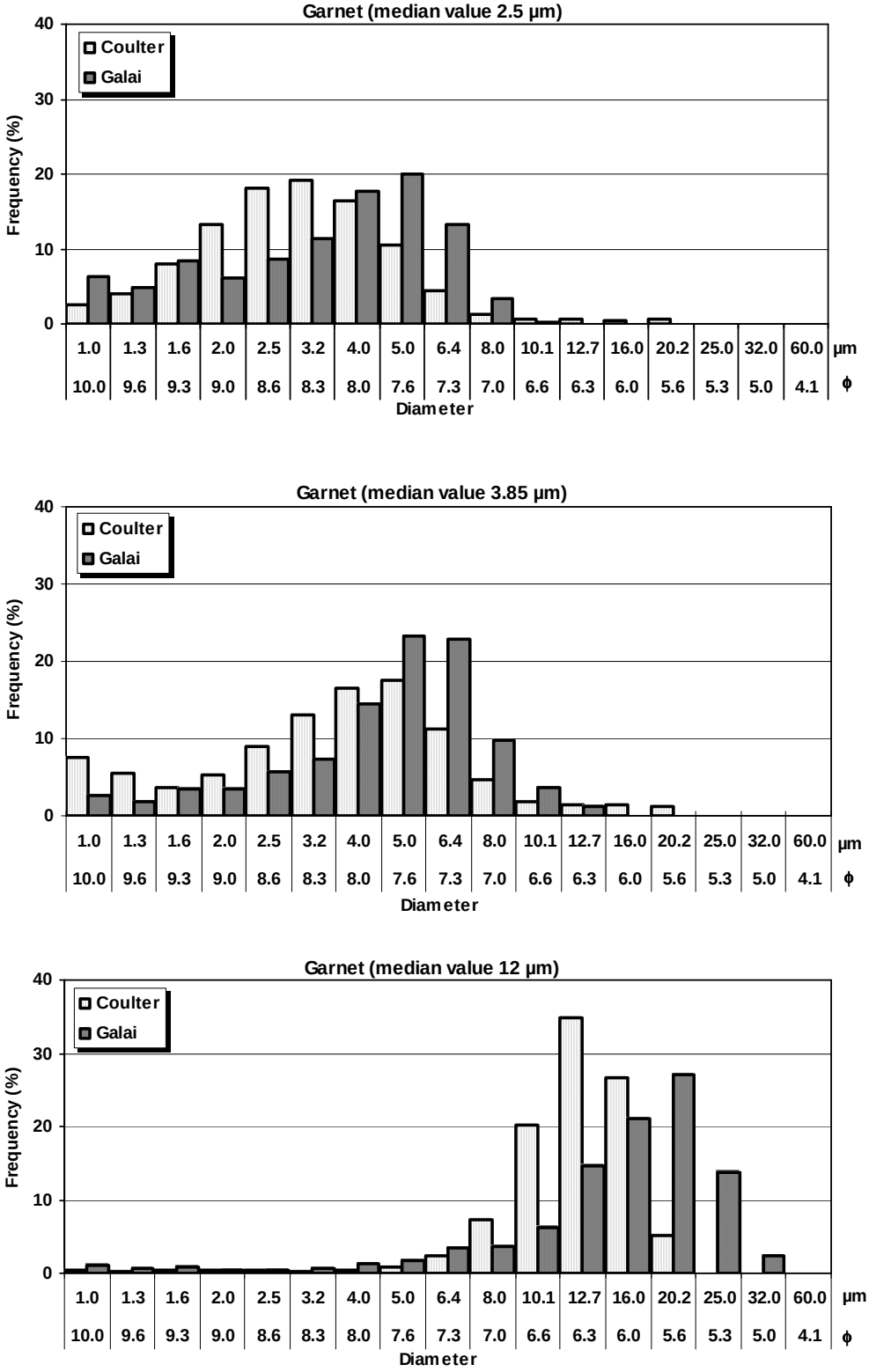
Garnet (median value 2.5 µm)
Coulter
Galai
Frequency (%)
0
10
20
30
40
1.0 1.3 1.6 2.0 2.5 3.2 4.0 5.0 6.4 8.0 10.1 12.7 16.0 20.2 25.0 32.0 60.0 µm
10.0 9.6 9.3 9.0 8.6 8.3 8.0 7.6 7.3 7.0 6.6 6.3 6.0 5.6 5.3 5.0 4.1 ϕ
Diameter
Garnet (median value 3.85 µm)
Coulter
Galai
Frequency (%)
0
10
20
30
40
1.0 1.3 1.6 2.0 2.5 3.2 4.0 5.0 6.4 8.0 10.1 12.7 16.0 20.2 25.0 32.0 60.0 µm
10.0 9.6 9.3 9.0 8.6 8.3 8.0 7.6 7.3 7.0 6.6 6.3 6.0 5.6 5.3 5.0 4.1 ϕ
Diameter
Garnet (median value 12 µm)
Coulter
Galai
Frequency (%)
0
10
20
30
40
1.0 1.3 1.6 2.0 2.5 3.2 4.0 5.0 6.4 8.0 10.1 12.7 16.0 20.2 25.0 32.0 60.0 µm
10.0 9.6 9.3 9.0 8.6 8.3 8.0 7.6 7.3 7.0 6.6 6.3 6.0 5.6 5.3 5.0 4.1 ϕ
Diameter

FIG. 2

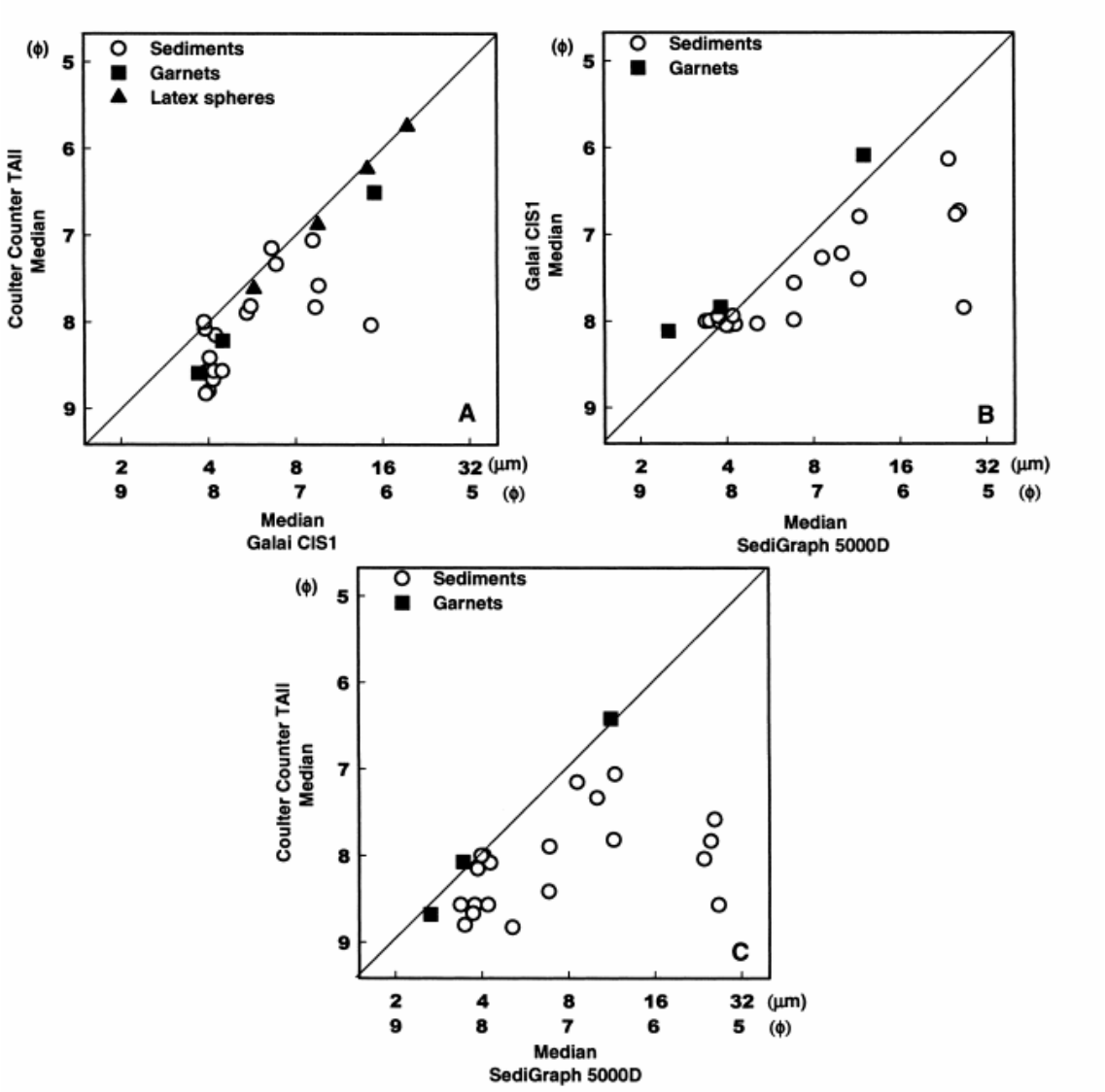
(ϕ)
Sediments
Garnets
Latex spheres
Coulter Counter TAll Median
A
Median Galai CIS1
(μm)
(ϕ)
Galai CIS1 Median
B
Median SediGraph 5000D
C

FIG. 3

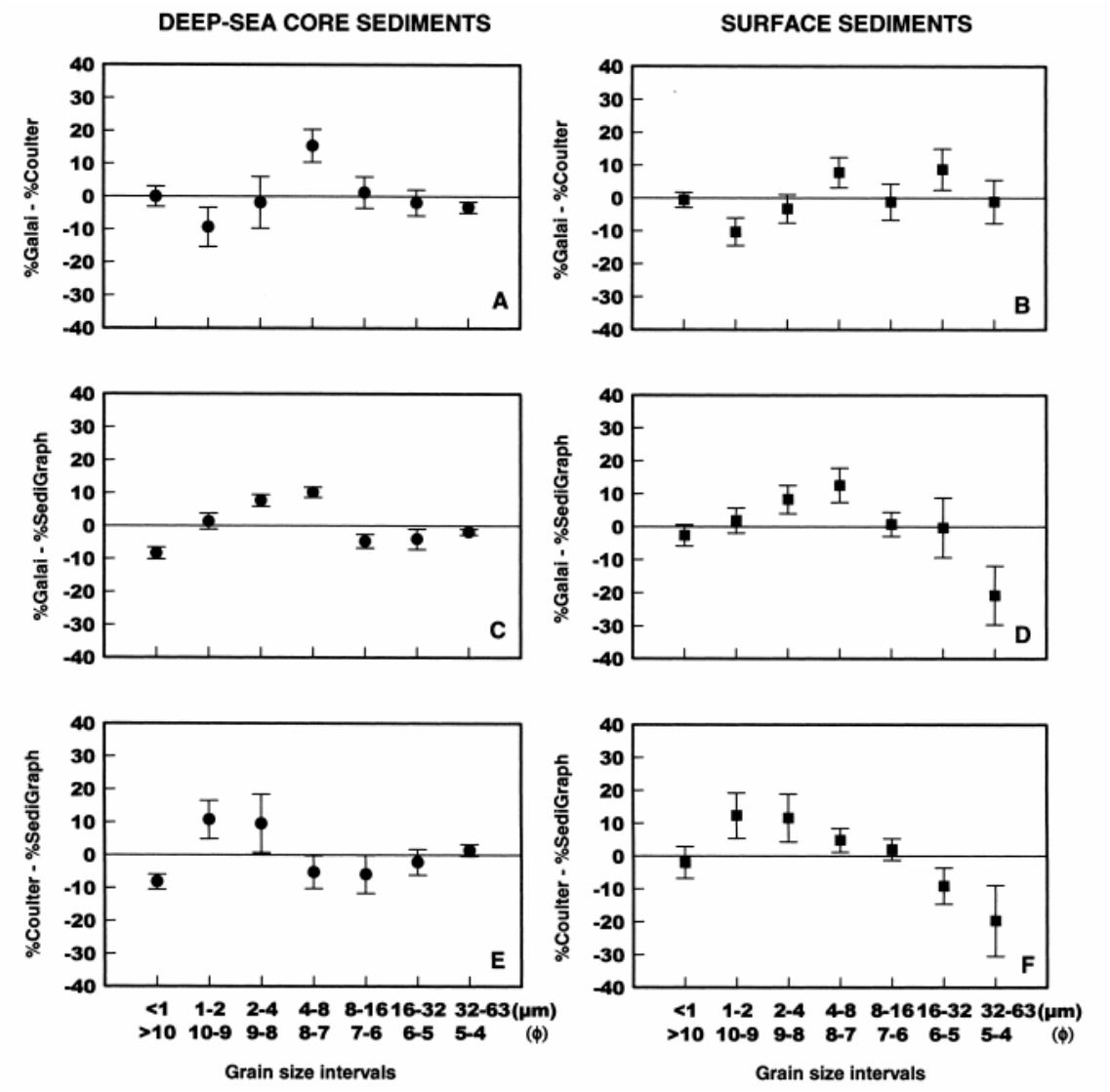
DEEP-SEA CORE SEDIMENTS
SURFACE SEDIMENTS
%Galai - %Coulter
%Galai - %SediGraph
%Coulter - %SediGraph
A
B
C
D
E
F
<1 1-2 2-4 4-8 8-16 16-32 32-63(μm)
>10 10-9 9-8 8-7 7-6 6-5 5-4 (ϕ)
Grain size intervals

FIG. 4

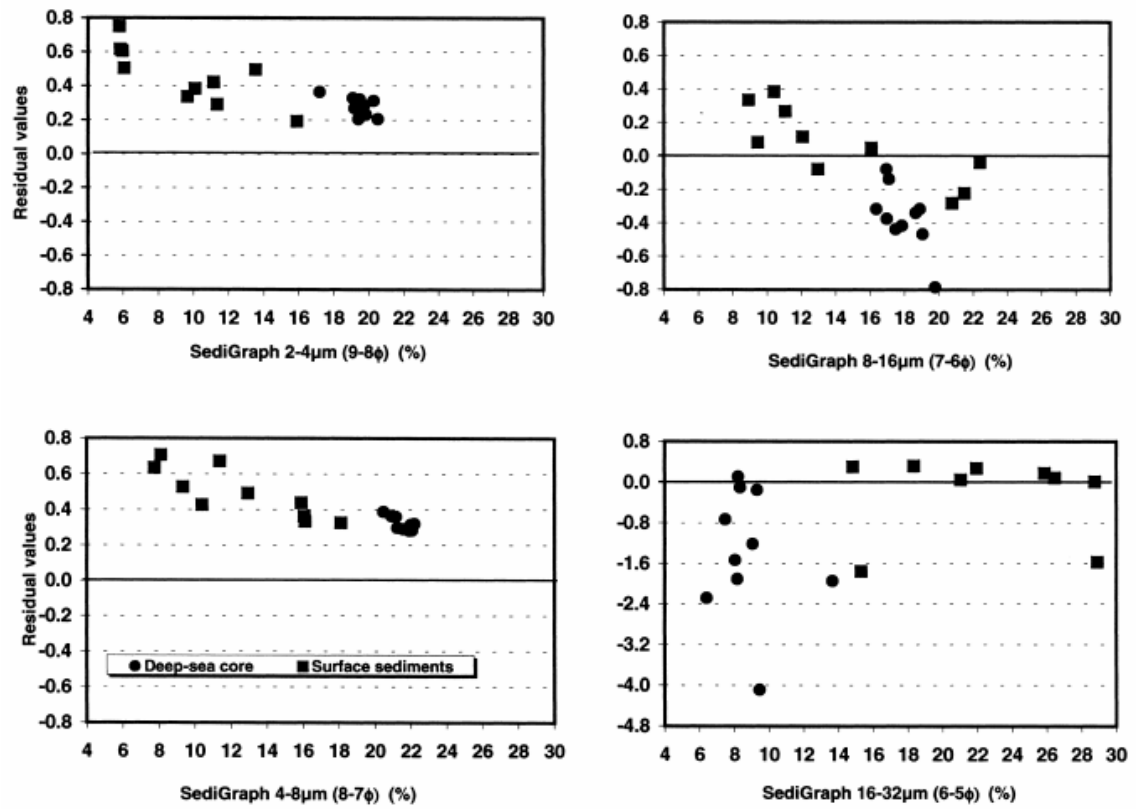
Residual values
SediGraph 2-4µm (9-8ϕ) (%)
SediGraph 8-16µm (7-6ϕ) (%)
SediGraph 4-8µm (8-7ϕ) (%)
SediGraph 16-32µm (6-5ϕ) (%)
Deep-sea core
Surface sediments

FIG. 5

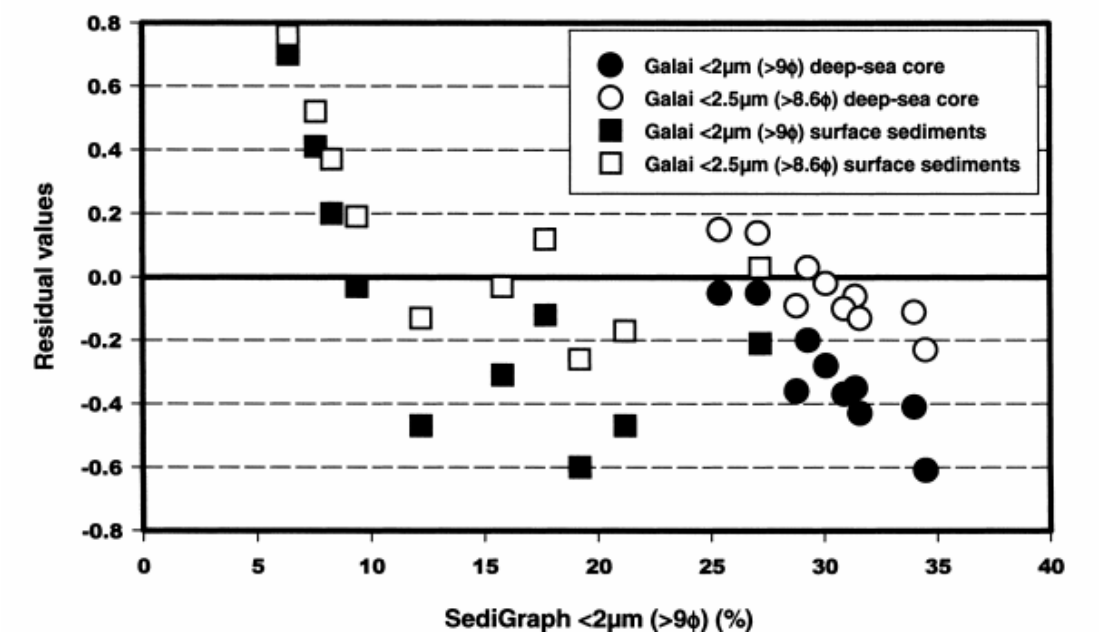
Galai <2µm (>9ϕ) deep-sea core
Galai <2.5µm (>8.6ϕ) deep-sea core
Galai <2µm (>9ϕ) surface sediments
Galai <2.5µm (>8.6ϕ) surface sediments
Residual values
SediGraph <2µm (>9ϕ) (%)

FIG. 6