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Use of SEM/EDS and X-ray Diffraction Analyses for Sand Transport Studies, Lake Erie, Ohio

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ABSTRACT



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Natural tracers offer advantages over artificial tracers because larger volumes can be studied over longer time intervals. However, natural tracers present their own set of problems, including non-uniform starting conditions due to variation within the tracer population, dilution by mixing, and abrasional destruction of the grain characteristic.

A beach nourishment project in Lake Erie used about 7000 m^3 of sand that was found to have a distinctive ironrich clay coating on quartz sand grains. The sand was uniquely introduced, because it came from a quarry 15 km inland and had not been used for other beach nourishment projects in the region. Experiments were undertaken to identify the coating, assess the hydraulic equivalence of the tracer sand to native beach sand, and to determine the resistance of the grain coating to abrasion.

Results indicate the sand behaved as a natural tracer that was detectable for approximately 2.5 years after introduction. Plume centroid migration rates averaged 2.45 mday⁻¹ westward during the first 20 months of the study. Eventual dissipation of the tracer plume (due to mixing and abrasional loss of the coating) resulted in an apparent reversal of the plume migration direction.

ADDITIONAL INDEX WORDS: Tracers, sand transport, SEM/EDS, X-ray diffraction.

INTRODUCTION

Natural Tracers

The utility of tracers to calculate sand transport rates in the coastal environment has been extensively discussed (e.g., INMAN and CHAMBERLAIN, 1959; INGLE, 1966; CRICKMORE, 1967; Komar and Inman, 1970; International Atomic ENERGY AGENCY, 1973; MADSEN, 1989; WALKER and INMAN, 1989; WHITE and INMAN, 1989; WHITE 1998). Artificial tracers involve marking a certain amount of native beach sand with paint (WHITE and INMAN, 1989), fluorescent dye (SHER-MAN et al., 1994; VOULGARIS et al., 1998; CIAVOLA et al., 1997, 1998; BADR and LOTFY, 1999; MICHEL and HOWA, 1999), or radioactivity (INMAN and CHAMBERLAIN, 1959; CHEONG et al., 1993). Natural tracers utilize some other sand that differs from native beach sand in mineral composition or surface texture features (BLACKWELDER and PILKEY, 1972; ABDALLA, 1991; ALMAGOR and KARNIELI, 1996; REED and WELLS, in press). In both instances, the requirements are that: (1) tracer grains must be hydraulically equivalent to native beach sand, (2) the introduction of the tracer must not change the characteristics of the transport system, and (3) in the transport system, advection must dominate over dispersion (MAD-SEN, 1989).

Although the literature suggests that artificial tracers have been used more frequently than natural tracers have, each offers certain advantages. Artificial tracers are easily and distinctively tagged, thus presenting fewer problems with identification and rapid compilation of results. On the other hand, smaller volumes are typically used in artificial tracer studies, which limits the duration and scale of the study. The opposite conditions apply to natural tracers, although problems related to recognition of natural tracers have been partly addressed by use of new technology, such as scanning electron microscope/energy dispersive spectrometry (SEM/EDS) methods (*e.g.*, DEBOER and CROSBY, 1995).

Tracers and Beach Nourishment Studies

Beach nourishment is a commonly accepted tool of coastal management (*e.g.* VANDEGRAAFF *et al.*, 1991). In the Great Lakes, since 1955 there have been at least 416 individual beach nourishment projects at 60 sites, involving a total volume of about 19 million m^3 (O'BRIEN *et al.*, 1999). DOBROWS-

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Figure 1. Location of the study area along the Lake Erie shoreline of Ohio (insert map), showing the location of the beach nourishment project (fill sand pile) on the east side of Heidelberg Beach, and sample locations. The fill sand came from a quarry near Collins, Ohio (insert map).

KI (1998) found that inland mined sand is cost effective for projects with volumes less than about $38,000 \text{ m}^3$ provided that the quarries are within about 20 km of the project site, otherwise dredged sand is more cost effective.

Beach nourishment projects have been used as the opportunity to apply both artificial and natural tracers (*e.g.*, EIT-NER and RAGUTZKI, 1994; VERHAGEN, 1996; FOSTER *et al.*, 1996; GUILLEN and HOEKSTRA, 1997; COOPER, 1998). Findings have indicated that some key aspects of the longevity of the beach fill are: grain size relationships between the fill and native beach sand (SWART, 1991), grain density (EITNER, 1996), and carbonate content (EITNER and RAGUTZKI, 1994). Monitoring studies have shown that beaches return to their pre-nourishment grain-size distributions within months to about five years (PSUTY and MOREIRA, 1992; GUILLEN and HOEKSTRA, 1997).

History of the Tracer

The Ohio shoreline of Lake Erie has been extensively affected by coastal zone development and modification of sediment budgets (CARTER, 1977; CARTER and GUY, 1980; GUY *et al.*, 1994; MACKEY, 1996). Recession rates range upwards to 3 myr⁻¹ (CARTER and GUY, 1980; MACKEY and GUY, 1994). As a consequence, when a private marina and jetty were proposed on the east side of Heidelberg Beach, located approximately 20 km east of Sandusky, Ohio (Figure 1), the Ohio Department of Natural Resources required mitigation in the form of a beach nourishment project.

The marina owner purchased approximately 7000 m³ of fill sand from Mesenburg Quarry, located 15 km inland near Collins, Ohio. The source sand body is a tabular glacial-outwash deposit (Don Guy, Ohio Geological Survey, *personal communication*). The sands are brown probably due to groundwater discharge and precipitation of iron-rich clay coatings around grains. In contrast, native Lake Erie beach sands are gray due to dominance of rounded and abraded quartz grains.

The fill sand was emplaced as a transient point source on

Heidelberg Beach in September, 1991. The sand pile gradually diminished, until finally disappearing in May 1993. Although it had not been anticipated, the introduction of this distinctive sand presented the opportunity for evaluating its use as a natural tracer.

METHODOLOGY

There were seven sample locations used in this study (Figure 1), the specific location determined by accessibility and presence of beach (much of the intervening areas consist of seawalls and other hard stabilization structures). At each sample location, a beach transect was established which consisted of 1–5 stations approximately 100 m apart (the number of stations was determined by beach width). There were a total of 18 stations used in this study. At each station, a shore-normal transect involved 3 samples: one in the surf zone (about 1 m water depth), one in the swash zone, and the third about 5 m inland from the swash zone, typically within the berm (if present). The berm sample would be collected from 2 cm below the surface, to eliminate eolian sand contamination (which is relatively minor in this particular study).

A time series of samples was collected from the seven locations described above and also from the pile of fill sand at approximately three month intervals over a 2.5 year period, starting in September 1991. Samples from the pile of fill sand were used to determine the suitability of the natural tracer (composition of the grain and the grain coating, grain size, and abrasion resistance). Samples from the seven study locations were used to calculate concentrations of tracer grains. Tracer grain concentrations were determined from 132 samples by point counting methods (identifying > 400 grains per slide). Tracer concentrations in the individual samples ranged from 0% to 8% (TOKAR, 1993; EVANS, unpublished data).

Petrography studies used standard practices for making grain mounts. Heavy minerals were separated using a Franz



Figure 2. A: SEM photomicrograph split image of native beach sand, showing lack of grain coatings on a quartz sand grains (left, at $50\times$), and an enlargement of one grain (right, at $250\times$). B: Photomicrograph of tracer grains, taken from the fill sand pile on the east side of Heidelberg Beach at the start of the study, showing a grain coating obscuring the surface of individual quartz sand grains ($200\times$). C: split screen SEM photomicrograph showing the iron-rich, clay coating on quartz sand grains (left, at $200\times$) and an enlargement of indicated target area (right, at $400\times$). D: high magnification ($3,500\times$) SEM photomicrograph showing the morphology of clay minerals that constitute the coating on quartz sand grain.

Isodynamic Magnetic Separator (HESS, 1966), and identified using optical techniques (KRUMBEIN and PETTIJOHN, 1938) or SEM/EDS methods (PORTER, 1962).

Scanning electron microscope (SEM) samples were mounted on carbon stubs using the methods of WALKER (1978), carbon-coated, and examined on a Hitachi S-2700 SEM with a LaB₆ filament. Elemental determinations used an EDAX SW 9100–60 energy dispersive spectrometer (EDS) with a bombardment time of 100 seconds, beam intensity of 20 kV, and sample tilt of 20 degrees. Semi-quantitative EDS analyses were performed using the method of BEAMAN and ISARI (1972).

X-ray diffraction (XRD) analyses of the clay coatings on the sand grains were performed by abrading the coating in a rock tumbler, concentrating the clay-size fraction by settling, and preparing standard clay mineral XRD slides using the methods of CARVER (1971). Each slide was analyzed using a Philips APD 3250 X-ray diffraction unit with Cu K-alpha radiation at a scanning speed of 0.02 degrees two-theta. Following qualitative analysis to determine which clay minerals were present, semi-quantitative determinations were made using the Deep Sea Drilling Project method (COOK *et al.*, 1975). The DSDP semi-quantitative method involves comparing field data (known minerals, but unknown relative abundances) to a standard. The standard is prepared by using equal weight percentages of the identified minerals, and then calibrating the intensities of the primary diffractogram peaks for each mineral. The calibration constant for each mineral is then used to calculate the relative abundance of the respective minerals in the field data.

Grain size analysis was conducted on several sand samples collected directly from the fill sand pile to demonstrate that



Figure 3. Results of semi-quantitative XRD analysis of the fill sand grain coatings. The upper curve shows peaks identified from oriented clay mineral samples obtained by abrading the fill sand grains. The lower curve was created in the laboratory using a standard, which consisted of 94% illite, 3% chlorite, and 3% quartz. See text for discussion of method.

the fill sand had similar grain size characteristics to native beach sand. Native beach sand samples were collected from a variety of places for the purposes of comparison. All samples were treated identically in preparation for sieve analysis, including methods of washing, drying, and splitting samples. The results of each sieve analysis was plotted on cumulative frequency diagrams, and statistical parameters (the mode, mean, standard deviation, skewness, and kurtosis) were calculated using the methods of FOLK and WARD (1957).

Laboratory abrasion tests were performed on fill sand grains to demonstrate the persistence of the grain coating. The accepted practice with artificial tracers (dyed grains) is to abrade the grains in a rock tumbler for 24 hours (WALKER and INMAN, 1989; MADSEN, 1989). In this study, individual tests in a rock tumbler lasted up to one week. One set of tests involved dried sand, and another set involved sand saturated with distilled water.

RESULTS

Petrography

Native beach sands are moderately sorted and rounded, and consist of >95% gray quartz, with minor amounts of heavy minerals (mostly garnet and magnetite), metamorphic and sedimentary rock fragments, and shell fragments. Individual grain surfaces are clean and polished (Figure 2A). Trenching shows that the beach deposits are planar laminated, with alternating beds of coarse-grained and fine-grained sand and occasional layers of heavy mineral placers (TOKAR, 1993). Flat-pebble clasts of Ohio Shale are concentrated in some stratigraphic layers.

The fill sand is also moderately sorted and rounded, and consists of 90% quartz, 3% feldspar, 3% heavy minerals (mostly magnetite, garnet, and ilmenite), 2% rock fragments, and 2% unknowns (obscured by the grain coating). There is a minor amount of matrix (silt- and clay-sized fraction) within these samples, which we interpret as naturally abraded grain coatings. Grain coatings are visible, even under low power microscopy (Figure 2B), and give the deposit an overall tan to light brown color.

Identification of the Grain Coating

SEM photomicrographs demonstrate the presence of clay mineral coatings based upon characteristic morphology (Figure 2C and 2D). Semi-quantitative XRD analyses determined that the coatings consist of approximately 94% illite, 3% chlorite, and 3% silica (Figure 3). Semi-quantitative EDS measurements indicate that the mean weight % Fe_2O_3 present is $8.3\% \pm 4.4\%$. This hematite is present within one or several of the minerals comprising the grain coating. Geochemical fingerprinting was accomplished by using SEM/EDS to show that tracer grains recovered from down drift locations (after the experiment started) were geochemically identical to tracer grains taken directly from the fill sand pile (prior to the start of the experiment). The results (Figure 4) are interpreted to show that individual fill sand grains can be treated as natural tracers due to the geochemical distinctiveness of their grain coatings.

Hydraulic Equivalence

Grain size analyses demonstrated that the fill sand has grain size characteristics that are intermediate within the range of values observed for native beach sand (Figure 5). The mean grain size for the fill sand is 1.33 ± 1.07 mm versus the mean grain size for native beach sand in the area of 1.41 ± 0.92 mm (TOKAR, 1993). Other properties (grain shape, sorting, mineral composition) also support hydraulic equivalence of the tracer sand and native beach sand (TOKAR, 1993).

Abrasion Resistance

The results of lengthy (one week long) tumbling experiments indicate that about half of the grains lost their coat-



Figure 4. Energy dispersive spectrometry plot showing oxide percentages from two samples: a natural tracer grain recovered from the Lake Erie nearshore after start of the study, and a grain taken directly from the pile of fill sand at the start of the study. The ability to geochemically "fingerprint" the fill sand enhances the reliability of its use as a natural tracer.

ings entirely, while the remainder of the grains retained a portion (generally 5% to 20% by surface area) of the coatings. The coatings were easily identifiable on those grains that retained any portion of the coating (Figure 6A). Our field results tend to show that the laboratory tests were more rigorous than natural abrasion processes, because large quantities of tracer grains are still identifiable 2.5 years after their introduction to the Lake Erie coastline (Figure 6B).

Field Behavior of the Tracer

The results of periodic sampling at eight locations are summarized in Table 1. Two general trends can be observed, in chronological order. The first trend consists of westward migration of the tracer during the first 20 months after its introduction. This westward migration is in accord with the prevailing longshore drift direction for this region of the Lake Erie coast (METTER, 1953; WORTHY, 1980; CARTER and GUY, 1980). The plume centroid, as defined by the highest tracer concentrations, reached Mitiwanga Beach (1.5 km down drift) by May, 1993 (20 months after introduction), thus the plume centroid had a mean migration rate of approximately 2.45 mday⁻¹. The maximum sediment velocity observed in this study was approximately 20.55 mday⁻¹ over a 12 month interval, based upon the first appearance of tracer grains at individual sample locations (TOKAR, 1993).



Figure 5. Cumulative frequency diagram for grain size distributions of fill sand (taken directly from the fill sand pile prior to eventual dispersion) and native beach sands from various locations. Cumulative frequency plots for the fill sand are within the range of variation observed in plots for the native beach sands, suggesting hydraulic equivalence of the natural tracer and native beach sand.



Figure 6. A: SEM photomicrograph of a fill sand grain that has been subjected to laboratory abrasion tests (tumbled in a dry rock tumbler for 48 hours), showing partial retention of the original grain coating ($800 \times$). B: SEM photomicrograph of a natural tracer grain recovered from the Lake Erie nearshore 12 months after the start of the study, showing similar partial loss of the grain coating due to abrasion ($70 \times$).

The second trend occurs after the source of the tracer (the fill pile of sand on Heidelberg Beach) essentially disappeared in May 1993. After this time, the plume centroid stagnated at Mitiwanga Beach, and then eventually dissipated (concentration values fell below 3%). The "retrograde" position of the plume centroid during these later sampling intervals is interpreted as an artifact of the plume dissipation phase. In these cases, the concentration values are significantly less, making recognition of the plume centroid more problematical.

DISCUSSION

Tracer Validity

Our results show that the sand used in this beach nourishment project contained quartz grains with distinctive Fe-rich clay coatings that could be used as natural tracers. The tracer grains are hydraulically equivalent to native beach sands, are relatively resistant to abrasion, and can be geochemically "fingerprinted" using SEM/EDS methods.

The concept of source area fingerprinting has been applied

elsewhere to soils and sediments utilizing grain size, shape, and composition (e.g., OLDFIELD et al., 1979; WALLING et al., 1979; GRIMSHAW and LEWIN, 1980; HSIEH, 1984; BROWN, 1985; PEART and WALLING, 1986; LESCHAK and FERRELL, 1988; BUI et al., 1989; JOHNSON et al., 1991). The utility of applying SEM/EDS methods to study fine-grained suspended sediments in rivers and estuaries has been demonstrated before (e.g., PAERL, 1973; YIN and JOHNSON, 1984; WEIDEMANN et al., 1985; BERNARD et al., 1986; EISMA, 1986; DEBOER and CROSBY, 1995). Nevertheless, we believe this study is the first documented use of SEM/EDS fingerprinting techniques for sand populations in nearshore sediment transport studies.

Problems Encountered

After validating the tracer (identifying the grain coating, demonstrating hydraulic equivalence to native sand populations, and assessing abrasion resistance of the coating), the chief difficulties encountered were: (1) non-uniform starting conditions due to variation in grain coating characteristics within the population of tracer grains, (2) difficulty in tracer

Location (E to W)	Sample Collection (month/year)							
	09/91	09/92	12/92	03/93	05/93	08/93	12/93	03/94
Beulah Beach	NM	P2	NA	P2	P2	P3	P2	NP
Fill Pile	P3	P3	$\mathbf{P3}$	P3	P3	P2	P2	P1
Heidelberg Beach	$P2^*$	$P3^*$	$P3^*$	P3	P3	P3	P3	$P2^*$
Island View Beach	NM	NM	NA	$P3^*$	P3	P3	$P3^*$	P2
Mitiwanga Beach	NM	P2	NA	$\mathbf{P3}$	$P3^*$	$P3^*$	P2	P2
Ruggles Beach	NM	P2	NA	$\mathbf{P3}$	P3	P2	NP	NP
Oberlin Beach	NM	P2	NA	P2	P2	P2	NP	NP
Old Woman Creek Beach	NM	NM	NA	P2	NM	NP	NP	NP

Table 1. Summary of tracer concentration data.

Explanation: P = Tracer present; $P3 \ge 3\%$ concentrations; P2 = 1-3% concentrations; P1 = 0-1% concentrations; NP = Tracer not present; NM = Not measured; NA = Site not accessible due to ice conditions; * = PLUME centroid (highest measured concentrations).

recognition due to inevitable mixing with native beach sand, and (3) related difficulties in recognition due to inevitable abrasion loss of the grain coating itself. There are no simple solutions to these problems, but the weak magnetic susceptibility of the iron-rich clay grain coating did facilitate determining the presence or absence of tracer grains, by using a Franz Isodynamic Separator. Such work could only be qualitative, however, because abrasion loss of the grain coating also obviously affects magnetic susceptibility.

CONCLUSIONS

This study has demonstrated that source area fingerprinting can be applied to sand transport studies in nearshore environments, using SEM/EDS and XRD methods. In Lake Erie, a natural tracer was created by the unique introduction of quartz sand grains with distinctive grain coatings. The advantages of using such a natural tracer include the larger volume (in this study, approximately 7000 m³) and longer duration (in this study, 2.5 years) of the tracer experiment. The disadvantages include the problems with variation within the tracer population, dilution by mixing, and abrasion loss of the measured characteristic (the grain coating). Also, such a large volume tracer study is probably more costly than most small volume, artificial tracer studies.

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