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Tracing the Movement of Fine-Grained Sediment in Aquatic Systems: A Literature Review

J.P. Coakley and B.F.N. Long

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Abstract

A comprehensive literature search of major computer-maintained data bases was made for all references dealing with tracing fine-grained sediment movement. A total of 103 references on the topic were compiled and reviewed. The report summarizes the state of the art in tracing fine sediments and evaluates the various techniques for use in studies of contaminated sediment dynamics in the Great Lakes-St. Lawrence aquatic system.

Résumé

On a effectué, dans les principales bases de données gérées par ordinateur, une recherche documentaire détaillée de tous les ouvrages traitant de l'étude à grain fin. Au total, 103 ouvrages traitant de la question ont été compilés et examinés. Le rapport résume les connaissances actuelles en matière d'étude des sédiments fins à l'aide de traceurs et il évalue les différentes techniques applicables à l'étude de la dynamique des sédiments contaminés dans le système aquatique des Grands Lacs et du fleuve Saint-Laurent.

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INTRODUCTION

Since the early 1950s, the technique of estimating the sediment transport rate in rivers and other bodies of water through the use of sediment tracers has become more and more common. In essence, the technique relies on the ability to recognize a distinguishing property of the existing sediment or to deliberately mark a volume of sediment in such a way that its subsequent history in the natural environment can be monitored. In brief, a good sediment tracer should have the following properties:

1. Its hydraulic behaviour should be similar to that of the unmarked local sediment.
2. Its presence should be easily quantifiable.
3. The point source of the tracer material or property should be precisely known.

The transport rate of the actual sediment can be estimated from the measured transport rate of the tracer. A thorough description of the concept and techniques involved is presented in Courtois (Appendix, reference no. [25]), Nelson and Coakley [64], and Tola [91]. The simplicity and directness of the approach is obvious and accounts, in large measure, for its appeal in sediment transport studies. Furthermore, in cases where sediment transport is nonuniform, episodic, or otherwise complex (such as in estuaries, for example), tracer techniques may provide the only feasible means of quantifying such transport process.

Problems in Tracing Fine Sediment

Until recently, the development of sediment tracer technology in aquatic systems has been driven largely by coastal engineering concerns, such as quantifying longshore drift and beach sand move-

ment. For that reason, most of the techniques are related to sand and coarser sediments. The large grain-size of these materials allows them to be more readily labelled and detected. However, in ecological investigations, where the material studied consists primarily of fine sediments (silts and clays), important complications arise, some of which are listed below.

1. Fine sediments may be carried in suspension and rapidly dispersed over large areas, thus increasing both the amount (or activity) of tracer and the number of survey samples required for tracer plume definition. This wide dispersal potential can also create problems insofar as the safe containment of radioactive tracers is concerned, especially in urbanized freshwater ecosystems.
2. Fine sediments in suspension are affected by inter-grain electro-chemical attractions, which can cause partial or complete flocculation. Marking natural fines or simulating them would lead to unpredictable changes in flocculation processes and could thus influence the hydraulic behaviour of such sediments.
3. Affixing a stable surface tag onto fine particles of irregular surface properties is difficult and may result in changes in the effective particle diameter, shape, and density, thus also affecting hydraulic behaviour. In addition, because of their large surface/mass ratio, interpretation of surface-labelled tracer data for transport rates can be problematic.
4. Physical simulation of fine sediment particles (e.g., by incorporating the tracer in glass and grinding it to size) is difficult because of problems associated with matching the grain-size and density of the simulated grains with those of the natural material.

The above difficulties might explain the relative scarcity of long-term tracer studies on fine sediments in the literature. Most of the experiments recorded are aimed at short-term problems related to dredged spoil dispersion patterns and harbour siltation (Tola [92]). Techniques for tracing fine sediment have also been developed in the Gironde Estuary (Sauzay et al. [78]) and the Rhine River (de Groot et al. [28]). The need for more research into tracing fine sediments is becoming more and more pressing given the important role such sediments play in the dispersal of adsorbed contaminants and toxic chemicals in the Great Lakes system. This report is viewed as an initial step in assessing the utility of tracer techniques for studying fine sediment dynamics in the Great Lakes and the St. Lawrence River.

ments (Appendix) compiled from computerized data bases (WATDOC, FLUIDEX, DELFT, GEOREF), as well as from the personal reference files of the authors. The 103 references making up the bibliography were organized into two major cross-referenced groupings: tracer type and tagging procedure (Table 1). In addition, references published prior to 1970 were identified separately, as these often have been superseded by improved technologies and should therefore be downgraded to some degree. Furthermore, these references are also more difficult to obtain and evaluate. The list of references cited at the end of the text itself is associated with aspects of tracing coarse-grained sediments and so is presented separately from the appendix.

REVIEW OF TECHNIQUES FOR TRACING FINE SEDIMENTS

This report features a comprehensive listing of the existing literature relevant to tracing fine sedi-

Tracer Types

Two main tracer types are identified in the literature: artificial and natural.

Table 1. Classification of References Related to Fine-Grained Sediment Tracers

| Tracer types | Tagging procedure | | | Total |
|-------------------------------------|--|--|--|-------|
| | 1. Artificial particle | 2. Surface label | 3. Adsorption | |
| Artificial | | | | |
| A: Radioactive | 21,25,26,45,66,71, 72,73,75,76,77,78, 79,80,81,83,84,90, 91,96 (20)* | 1,2,3,4,7,15,21,25, 26,30,35,36,37,38, 39,40,45,46,47,49, 50,51,52,53,58,60, 66,68,69,71,72,73, 74,75,77,78,79,80, 81,83,84,85,86,87, 88,89,90,91,92,93, 94,96,97 (53) | 6,11,12,15,16,17, 18,19,21,22,25, 26,27,33,37,38, 44,61,63,90,91 95 (22) | 95 |
| B: Nonradioactive | | | | |
| 1. Neutron-activable | | | 5,9,28,31a,41, 47a,100 (7) | 7 |
| 2. Incidentally introduced | | | 8,10,22,23,29,34, 42,43,59,62,65, 70,75,82 (14) | 14 |
| 3. Fluorescent | 57a (1) | | | 1 |
| 4. Chemical tracer | | 55,61 (2) | 55,56,61 (3) | 5 |
| Natural | | | | |
| C: Naturally occurring substance | 20,54,63 (3) | | 32,41 (2) | 5 |
| D: Naturally occurring radioisotope | | | 13,14,62,63 (4) | 4 |
| E: Unclassified general | 24,30,31,48,57,60,64,67,88,98,99 (11) | | | 11 |
| References pre-1970 | 18,19,20,33,35,36,42,45,46,47,48,49,50,51,52,53,61, 63,68,69,70,71,72,73,74,75,76,84,85,86,87,98 (32) | | | 32 |

*Totals for individual tracer types are shown in brackets.

Each is further subdivided into various categories, e.g., radioactive, nonradioactive, naturally occurring, incidentally introduced. The various subdivisions have been coded in a straightforward way (Table 1), and these codes have been assigned to the individual references in the appendix for cross-reference purposes.

Artificial Tracers

Artificial tracers are those tracer materials that are tagged or simulated by the experimenter. They are preferred in some cases because they give the researcher complete control over the conditions of the experiment, especially insofar as timing and background levels are concerned. They include the following:

- radioactive tracers
- neutron-activable tracers
- incidentally introduced chemical elements
- fluorescent tracers

In choosing an artificial tracer, attention must be paid to the environment into which it will be injected. The selection must take into account the local hydrodynamic conditions as well as the geochemistry and physical properties of the local sediment. Some of the more important criteria governing the choice of an artificial tracer may be listed as follows (Nelson and Coakley [64]):

- It must be conservative, i.e., sufficiently durable and stable to survive unchanged over the duration of the experiment, otherwise its reduction, decay, or consumption rate must be predictable and fully known.
- It must be detectable at low concentrations in order to reduce as much as possible the amount of material to be used.
- It should be temporary in order to avoid long-term contamination of the site.
- Its injection or sampling should not change or disturb the natural transport processes.

- It must be inexpensive.
- It must be nontoxic to humans and to the aquatic environment in the concentrations used.

For fine sediment studies, the main artificial tracers used are radioactive (created by surface coating, incorporation in glass, or adsorption) and nonradioactive (including neutron-activable elements incorporated in glass or natural ores, fluorescent tracers, and incidentally introduced chemical tracers).

Radioactive Tracers

The use of radioactive tracers is by far the most commonly used technique in past studies of fine sediment transport (Table 1). The bibliography lists 95 references on radioactive tracers out of a total of 142. The technique involves affixing a small quantity of a radioisotope to the sediment or simulated sediment and injecting the resulting tracer into the natural sediment system. Because of the relatively high activity of such tracer isotopes, detection and counting can be carried out in real time in the field. This is one of the prime advantages of radioactive tracers, and it allows the experimenter to follow the tracer plume as it moves. Another advantage is that such tracers naturally decay to background levels, so they leave no permanent contamination at the site. The overriding disadvantage, however, is the inherent health and environmental hazards in their use. Because of these hazards, trained personnel are required to handle the tracers, and special permits must be obtained for their use.

Choosing the best radionuclide to use in a tracer experiment depends more on the duration and detectability of the isotope and safety-related aspects of the experiment conditions than on the grain-size of the sediment material. For instance, Sauzay et al. [78] used Scandium-45 (half-life of 84 days) in their study of fluid mud pools in the Gironde Estuary. The Sc-45 was readily adsorbed onto the clay and its γ -activity allowed measurements over a relatively long period (6 mo). Au-198, however, with its half-life of 65 h, is best used for short-term studies. Table 2 lists some of the more common tags used in the references cited in the appendix, their half-lives, and other relevant properties.

Table 2. Radionuclides Used in Sediment Tracer Studies*

| Radionuclide | Decay mode† | Efficiency | γ -Ray energy (MeV) | Half-life |
|-----------------------|------------------|------------|----------------------------|-----------|
| ⁴⁶ Sc | β^- | 100% | 0.89-2.01 | 83.9 d |
| | | 100% | 0.89 | |
| ⁵¹ Cr | Electron capture | 9% | 0.32 | 27.8 d |
| ⁵⁹ Fe | β^- | 45% | 1.29 | 45 d |
| | | 53% | 1.10 | |
| ⁶⁰ Co | β^- | 100% | 1.17-1.33 | 5.26 y |
| | | 100% | 1.17 | |
| ⁶⁵ Zn | β^+ | 49% | 1.12 | 245 d |
| ⁹⁵ (Zr+Nb) | β^- | 49% | 0.76 | 65 d |
| | | 49% | 0.72 | |
| | | 100% | 0.75 | |
| ⁹⁹ Mo | β^- | 17% | 0.74, 0.78 | 67 h |
| | | 82% | 0.14 | |
| ¹¹⁰ Ag | β^- | 100% | 0.66 | 253 d |
| | | 74% | 0.89 | |
| | | 33% | 0.94 | |
| ¹¹³ In | | | 0.36 | 103 m |
| ¹²⁴ Sb | β^- | 100% | 0.60 | 60 d |
| | | 51% | 1.69 | |
| ¹⁴⁰ La | β^- | 38% | 0.33 | 40.2 h |
| | | 48% | 0.49 | |
| | | 44% | 0.82 | |
| | | 96% | 1.59 | |
| ¹⁶⁹ Tb | β^- | 30% | 0.30 | 72.1 d |
| | | 31% | 0.88 | |
| | | | others | |
| ¹⁸¹ Hf | - | - | - | 42.4 d |
| ¹⁷⁵ Hf | - | - | 0.1-0.5 | 70 d |
| ¹⁹² Ir | β^- | 100% | 0.32-0.6 | 74.2 d |
| | | 60% | 0.49 | |
| | | | others | |
| ¹⁹⁸ Au | β^- | 99% | 0.41 | 65 h |

*Modified after Nelson and Coakley (1974) and Tola (1982).

† β^- —Electron emission.

Neutron-Activable Tracers

According to Table 1, neutron-activable tracers have been used a number of times for fine sediment studies. An excellent description of the technique is found in de Groot et al. [28]. The major advantage they hold over the radioactive tracers is their relative safety of use and lower cost. The tracer elements chosen are not radioactive when injected, but radioactivity is induced when the sediment sample is brought to the laboratory and exposed to a neutron source (reactor) prior to analysis. Although initial equipment costs are lower than for radioactive tracers, the costs of repeated neutron-activation

analysis (NAA) in outside laboratories, i.e., between \$15 and \$50 per sample depending on the laboratory used and the analysis specifications, could be considerable. The main disadvantage is the lack of good resolution of transport directions and rates due to the limitations caused by point-sampling in the field and NAA some time afterward. This also means that response to the results is relatively slow, and it is not possible to follow the tracer movement in the field. In some studies of long-term transport, the slowness of neutron-activable tracer techniques does not pose a serious problem, and they can still be considered as viable alternatives. The choice of such a tracer ultimately depends on the distribution

Table 3. Elements Potentially Useful as Neutron-Activable Tracers*

| Element | Coincidence method detection limit (ppm) | γ -Ray spectrometry (Ge-Li) method detection limit in sediment (ppm) | Natural content (ppm) † | Price/kg (\$US) |
|----------------|--|---|-------------------------|-----------------|
| Tantalum (Ta) | — | 1 | 1 | 125 |
| Antimony (Sb) | 0.2 | 0.2 | 2 | 5 |
| Cobalt (Co) | 1 | 1 | 20 | 30 |
| Indium (In) | — | 4 | < 4 | 4 000 |
| Iridium (Ir) | — | 0.05 | 0.05 | 4 000 |
| Terbium (Tb) | 3 | 3 | (2) | 15 000 |
| Europium (Eu) | — | 0.2 | 1 | 30 000 |
| Cesium (Cs) | — | 0.5 | 2‡ | |
| Lanthanum (La) | — | 0.2 | 35‡ | |
| Barium (Ba) | — | 10 | 700‡ | |

* Modified after de Groot et al. (1970).

† Natural content in Rhine River sediments.

‡ Natural content in Humber Bay sediment, Lake Ontario.

and predictability of background levels of potential tracer elements. Elements that have potential for use as neutron-activable tracers are listed in Table 3. Most likely candidates are found among the elements on the sixth row of the periodic table, such as cesium, barium, tantalum, and the lanthanum series, all of which have high neutron-capture "cross-sections" and, thus, favourably low detection limits in NAA (Attas [9], Krezoski [47a]). Rare earth elements also have an affinity for fine sediments and thus are relatively easy to use as sediment tags (see Surface Adsorption).

Incidentally Introduced Chemical Elements

In areas where a readily identified chemical source exists, such as at a sewage treatment plant or nuclear generating station outfall, or at the outlet of a polluted river, the sediments in contact with that source might become contaminated or labelled by a characteristic contaminant to the extent that they may be used as incidental tracers. Fine sediments, especially clays, are usually good ion-exchange and sorption media; insoluble trace metals and radionuclides are readily adsorbed onto the large surfaces they offer (Eichholz et al. [33]; Mundschenk [62]; Auffret et al. [10]).

One advantage of such incidental tracers is that, unlike the two previously mentioned techniques, they do not require the purchase of expen-

sive tracers or the injection of any foreign or hazardous material into the natural system, since they are already there. Like neutron-activable tracers, however, they suffer from the inability to allow following the tracer movements in real time. Once again, this is not a problem if long-term, integrated transport patterns are being studied. Sediment samples also must be taken back to the laboratory for analysis and counting — a procedure that could take days or weeks. Also, analysis for some chemical elements or species could be expensive (up to several hundred dollars per sample). Another disadvantage of such techniques is that they provide only a qualitative estimate of the transport since the "tracer" addition times and rates are uncontrolled by the experimenter and are often highly variable. Furthermore, partitioning of the chemicals between solid and liquid phases is often not well known, and thus creates a problem for quantitative interpretation.

Good examples of the application of such a technique are found in Clifton and Hamilton [23] and Olsen et al. [65]. Both references describe the use of anthropogenic radionuclides introduced into the aqueous environment either by global fallout or in the effluent of a nuclear facility (the Windscale fuel reprocessing plant in England and the Oyster Creek Nuclear Generating Station in New Jersey, respectively) to trace sediment dynamics. Of the two studies, the latter deals more specifically with sediment transport, while the former is aimed more specifically at determining sediment accumulation rates.

Other studies that deal with tracing sediments using incidentally introduced chemicals (radionuclides) are by Perkins et al. [70], Gross and Nelson [42], and Biscayne et al. [14].

In the case of a heavily polluted river (such as the Humber River), it is also conceivable that surficial adsorption of characteristic pollutants onto its fine sediment discharge might render the sediment recognizable after it has entered the general sediment population of the receiving body of water. Initial examination of trace metal contents of sediment samples from Humber Bay (Table 3) indicates that qualitative dispersal patterns of sediments from the Humber River may be thus defined. Examples of potential incidental tracers for Humber River sediments are copper, zinc, chromium, and lead. However, some loss of resolution occurs because precise shoreline locations of these inputs cannot be pinpointed, and atmospheric inputs are often significant.

Attempts have been made to identify sources of sediments in a depositional basin using statistical refinements such as cluster analysis. Poulton (1989) used this technique to examine ratios of contaminant concentrations from various Great Lakes sites and was able to identify zones where characteristic ratios, relatable to various putative sources, predominated. By observing the spatial relationship of these zones to their sources, it is possible to interpret integrated, long-term sediment transport patterns. This technique can thus be added to the list of potential tracer techniques.

Similarly, investigations have been made into using conservative organic components associated with sewage treatment plant effluents as incidental tracers of sediments originating at these sources. Three such organic compounds studied were Coprostanol (Hatcher and McGillivray [43]), linear alkylbenzenesulphonates or LAS (McEvoy and Giger [59]), and vitamin E acetate (Eganhouse and Kaplan [34]). All of these substances can be quantitatively evaluated in sediments using gas chromatography or mass spectrometry, and are believed to be conservative enough to be used as tracers.

Fluorescent Tracers

Fluorescent tracers for fine sediment are very rare in the literature; only one was found (Louisse et al. [57a]). Part of the reason for this is that they are difficult to affix to natural particles without changing

their hydraulic behaviour. Also they are difficult to quantify accurately. Nevertheless, because these tracers offer the advantage of being relatively inexpensive and also detectable in real time in the field, some work on such tracers is still being carried out (Louisse et al. [57a]). Tests, both in the laboratory and in the field, were conducted using a commercial synthetic particle in powder form (finer than 10 μm) that contained a fluorescent pigment, such as Day-glo.

Natural Tracers

Natural tracers include all natural sediment particles or distinctive sediment properties that can be related directly to a certain source and are sufficiently conservative and easily quantified to be used as sediment tracers. Examples of such tracers are heavy mineral suites (Byrne and Kulm [20]), trends in grain-size parameters (Pettijohn and Ridge, 1932; McLaren and Bowles, 1985; McLaren, 1986), distinctive clay mineral assemblages (Lafond and Martin [54]; Neiheisel and Weaver [63]), and anthropogenic particles (Dell and Booth [29]).

Such techniques are useful only for qualitative descriptions of sediment transport patterns integrated over relatively long periods. They do not provide any quantitative idea of transport rates, and often the resolution of a transport pattern is impossible due to analytical limitations. However, because they can utilize data usually collected for other purposes (such as grain-size data, for instance), which are relatively inexpensive to obtain, they can be useful as a crude, initial indicator of transport patterns.

Tagging and Tracer Preparation

The tagging technique for particles destined to be released as tracers into the aquatic environment must be selected carefully in terms of the detection method planned. The tracer must be easily distinguished from the natural sediment. However, tagging procedures should not alter the mechanical, geochemical, and hydrodynamic properties of the sediment.

Artificial tracers may be prepared by several different techniques. Three approaches are presently used:

- surface coating of sediment collected from the test site;

- manufacture or processing of an artificial sediment or natural ore that incorporates the tracer in the matrix;
- surface adsorption of a chemical substance.

Surface Coating

Surface coating consists of coating sediment particles collected at the release site with a thin, durable film of tracer material. This procedure preserves the morphology of the natural grain, but it alters very slightly the mean diameter by the thickness of the coating. Though feasible for coarse particles such as sand, this technique is generally not suitable for fine material because it changes the grain-size and surface physical properties too much. Also, because of the larger surface-to-volume ratio of the finer particles, this procedure attributes much more weight to them than to the larger particles in the size distribution, and, since fine particles tend to migrate faster, the method usually leads to an overestimation of the total sediment transport.

Artificial Sediment Particles

Glass

The predominant medium for use as a synthetic fine sediment is glass. Glass has the advantage of being of roughly similar density to silicate minerals (quartz and feldspar), which constitute the bulk of detrital fine sediments, especially in glaciated terrains such as the Great Lakes area. It is also able to incorporate a number of distinctive tracer elements within its internal structure, thus providing almost ideal "mass labelling," i.e., tracer concentration proportional to mass, rather than surface area. Both of these considerations result in realistic and accurate transport estimates. The main drawback in the use of glass as a tracer medium is that clay-sized particles cannot be easily simulated. That probably explains the relatively small number of references dealing with glass simulation of very fine sediments (Table 1). Synthetic sediments made from glass can be used for both radioactive and neutron-activable tracer studies.

Neutron-activable tracer glasses, which are used in large quantities, can best be prepared by commercial glass manufacturers. In contrast, because of the small quantities needed and their specialized nature, radioactive glasses can be obtained only from government atomic energy

research agencies (such as Atomic Energy of Canada [AECL], Whiteshell Nuclear Research Establishment in Pinawa, Manitoba, or the Commissariat d'Énergie Atomique [CEA] in Saclay, France). The glass is manufactured commercially (e.g., by Saint-Gobains in France) and is then activated by the government agency. Before activation, the glass is ground mechanically to fit the size distribution of the local sediment at the injection site. For practical reasons, the fit is usually made to the modal size fraction, although for more precise work, the entire size distribution curve may be reconstructed (Coakley et al., 1974).

A variety of tracer elements have been incorporated into glass (Table 3), however, one of the main restrictions is the amount of element that can be accommodated within the glass structure. The proportion by weight of the tracer element is usually 5% or less (Coakley et al., 1973).

Natural Ores and Commercial Powders

Instead of using an artificial glass containing the distinctive tracer element, a commercial, naturally occurring ore of the tracer element might sometimes be less expensive and more conveniently obtained. Such materials would have all the advantages of glass, in addition to allowing the content of the tracer element to be even higher than that allowed in the glass. The use of finely ground polucite (an alumino-silicate ore containing approximately 24% of the distinctive element, cesium [Cs]) has been suggested as a tracer in areas where the background conditions are suitable (Michael Attas, 1986, AECL, pers. comm.).

Fluorescent tracer particles in the form of a commercial powder have been used to trace fine sediments (Louisse et al. [57a]). Also, it is conceivable that naturally fluorescent ores, such as scheelite (CaWO_4) or calcite (CaCO_3), could be used if they were finely ground and if their densities were suitable. However, no report of such usage was found in the literature search.

Surface Adsorption

Table 1 shows that surface adsorption has been used the most in tagging fine sediments for transport tracing, especially for radioactive elements. Its most obvious advantage is that the tracer element is deposited physico-chemically onto the surface of the natural sediment itself. Thus the major

hydraulic properties remain virtually unchanged, even for silts and clays. Another important advantage is that the technique can be carried out in the field, so it is ideal for such applications as studying the movement of dumped dredged spoil (Tola et al. [93]). Like all surface tags, however, the technique leads to a slight overestimation of the transport rate, as finer particles carry proportionally more tracer than coarser ones.

Successful labelling techniques have been carried out using radioactive Hf-181, 175 (Etcheber et al. [38]), Sc-46 (Sauzay et al. [78]), Au-198, and Cr-51 (Bougault [16], Bougault et al. [19], and Tola et al. [93]). In the case of Au-198, the gold tracer is first prepared in the form of a chlorauric (gold/hydrochloric acid) solution. When this solution is brought into contact with the silt suspension, the Au-198 is reduced to metallic gold and is fixed onto the particle surface. Cr-51 is precipitated onto the surface of the particle from a chromium hydroxide solution.

De Groot et al. [28] described the use of stable isotopes of cobalt and tantalum as neutron-activable tracers for fine sediments, mainly clay. Relevant details of these elements and other elements are given in Table 3. Cobalt may be attached to the sediment particles as intercalated Co-ions between clay lattice layers, exchangeable ions in the electric double layers of clay minerals, precipitated $\text{Co}(\text{OH})_3$, or chelated Co fixed to the organic matter in the sediments.

Of these modes, only the latter two provide the relative tightness of bonding necessary for the use of cobalt as a tracer. On the other hand, tantalum, like most rare earth elements, adheres readily to sediment. TaCl_5 is first dissolved in dilute HF, and the sediment is boiled in the solution.

Ecker et al. [31a] experimented with numerous elements (e.g., gold, rhenium) as neutron-activable tracers for fine-grained dredged spoil before choosing the element iridium (Table 3). Ten kilograms of iridium metal in powdered form were first converted to the soluble iridium salt. The salt was then surface-adsorbed onto approximately 10^4 kg of dredged sediment.

Injection of Fine-Grained Tracer Materials

Injection of tracer materials is carried out in such a way that disturbance of the local transport

environment by the tracer is minimized. Large amounts of tracer dumped on the bottom, for instance, would form a pile and alter the natural equilibrium that thereto existed on the lake bed. In such a case, the tracer movement measured initially would be due primarily to the erosion of this unnatural obstacle. Alternatively, releasing the tracer too high in the water column would cause an initial direct advection with the local currents, thus distorting the resulting estimates of actual bottom sediment transport rates.

The key to accurate tracer measurement of sediment movement is the rapid mixing and incorporation of the tracer into the local sediment system. For tracing fine sediment moving in suspension, it is definitely best that the tracer be injected directly into the suspension, rather than placed on the bottom. In such a case, good mixing occurs immediately during the release operation. Such an injection procedure is feasible for tracing instantaneous discharges from a plant or a polluted river. However, in cases where movement takes place some time after injection, by resuspension from the bottom during storms, for example, careful direct placement on the bottom is the most feasible approach.

Injection System For Radioactive Glass Tracers

Radioactive glass tracers are usually injected in relatively small amounts (5 kg or less) so they can be easily isolated in a small container prior to and during injection. The techniques most commonly used are

1. time-delayed release from soluble plastic bags placed on the bottom (Schulz and Pillot, 1965; Duane, 1970);
2. direct release from containers emptied by a diver (Vernon, 1964), with due attention given to personal safety;
3. release from specially designed injection systems opened on or above the seabed.

The advantage of the last technique is that it creates a dispersion cloud directly during the immersion. The larger diameter of the tracer origin point thus produced on the bottom greatly enhances tracking of the subsequent dispersion.

Injection of Adsorbed Tracers

When used for tracing dredged spoil movement, surface-adsorbed tracers are injected from the barge as part of the dumping procedure (Tola et al. [93], Ecker et al. [31a], Louisse et al. [57a]). In the case of large-scale marking and re-injection of natural fine sediments from the site, injection can be carried out from a boat other than that used in the detection phase in order to avoid contamination of the detectors. A secondary circuit including a high flow pump (50 L/min) ensures the dilution of the labelled sediment to a concentration of 18 g/L in order to avoid the formation of a density current and delivers the suspension at a depth of about 1 m below the surface.

Injection lasts less than a minute and marker floats are placed at the beginning and end to indicate approximately the extremities of the sediment suspension, thus aiding the subsequent dynamic detection process.

Injection of Neutron-Activable Tracers

One of the important differences between neutron-activable tracers and radioactive tracers is the large quantities of tracer needed to achieve a similar level of detection. For instance, assuming a uniform dispersal of a fine-grained tracer over an area of 5 km², a detection limit of 1 ppm for the traced element, and a 10 cm active sediment transport layer, the amount of tracer element needed is approximately 250 kg.

Injecting such large quantities of tracer poses some difficulty. One of the approaches considered for use in the Great Lakes area involves a boat equipped with apparatus for mixing local water with the tracer material to form a thin slurry and for pumping the slurry over the side. This technique is intended to disperse the tracer rapidly into the local suspended sediment flow.

Detection and Survey of Tracer Distribution

Sampling of the subsequent tracer distribution (or plume) constitutes the most delicate part of an experiment, as it determines the quality of the data collected. Excellent position-fixing is mandatory for reliable results, and samples are usually collected along a predetermined grid or survey pattern. In a lacustrine environment, a number of hydrodynamic factors (currents, waves, intermittent resuspension)

affect the transport. Thus, the probable direction of tracer transport is difficult to determine beforehand. For this reason, the sampling grid can be complex and the operation might require a large number of samples. In addition, cores must be taken in the field and vertically scanned in the laboratory for tracer concentration to determine the depth of the sediment layer actively being transported. This step is required in order to keep track of tracer losses in the system.

The amount of tracer injected and the sampling grid size are interrelated. They depend on the expected spatial dispersion of the tracer, the thickness of the mobile sediment layer, and the sampling mode. In a theoretical calculation of the number of samples required in a fluorescent tracer study, De Vries (1966) showed that the relative error of the concentration measurements is inversely proportional to the square root of the number of marked grains in the sample. A minimum of 100 tagged particles are required per sample collected in order to have an error of 10%.

Sampling modes commonly used include:

- point-by-point sampling (point sampling)
- static detection
- dynamic detection on the bottom
- dynamic detection in suspension

Courtois and Sauzay (1970) demonstrated that the quantity of tracer injected depended on the type of detection. The relative quantity of the tracer used thus varies from 1 (dynamic detection) to 10 (static detection) and 100 (point sampling).

Point Sampling

Point sampling mode is often used in connection with neutron-activable or chemical tracers. It consists of collecting sediment samples in the field

- by centrifuging the water column (suspended sediment)
- on submerged grease-coated plates (surficial bottom sediment)
- by grab samples (bottom sediment)
- by coring (bottom sediment).

The sampling pattern is designed according to the assumed sediment transport direction and dispersion. The pattern can be a radial or rectangular grid of suitable size.

Static Detection

Static detection is used primarily in association with radioactive tracers. The method involves the fixed positioning of a detector array so as to intersect and measure the sediment flow past that position (Courtois and Sauzay, 1970). Due to the small area of coverage analyzed per sensor at each point (1 m^2), this method requires the use of a much larger amount of tracer than that required for dynamic detection (ratio 1 to 10). This type of detection is best used when transport directions are somewhat limited, as in a river or estuary. It is also promising for measurement of the temporal evolution of the tracer at a single point for specialized studies such as those on the role played by sedimentary bedforms in bedload transport.

Dynamic Detection on the Bottom

For dynamic detection of the tracer distribution on the bottom, the detector may be mounted on a sled or on a remotely operated vehicle (ROV), taking measurements at points along predetermined traverse lines. The detector normally covers a strip of the bottom approximately 1 m wide, so it has the opportunity of detecting a large number of tagged grains. The availability of compact data acquisition systems has made this procedure the most commonly used for detecting radioactive tracers on the bottom. Case et al. (1971) estimate that 2% of all tracers deposited on the bottom can be detected within one hour over an area of 300 m x 600 m, which would correspond to the equivalent of 50 000 samples collected with a Shipek grab sampler (Nelson and Coakley, 1974).

The accuracy of this method is directly proportional to the number of cross sections through the tracer plume. The sample rate is generally 1 Hz and the data are recorded continuously on magnetic tape or analog (strip-chart) recorder. Results are available in real time, so the detection plan may be actively modified in the field to follow the movement of the tracer plume.

Dynamic Detection in Suspension

Dynamic detection in suspension is specifically used to determine the dispersion of the radioactive

tracer plume in suspension. It is well documented by Tola [92] and Tola et al. [93].

Detection begins immediately after injection. Two boats are used simultaneously. One boat is used for injecting the tracer, and, afterward, both boats carry out the transverse and longitudinal surveying of the suspended tracer plume. Two detectors per boat are needed in order to obtain sufficient and reliable results.

Survey of the tracer dispersion takes place at constant velocity passing as close as possible to the centre of gravity (or maximum concentration) of the plume as it is advected by the flow. The velocity of the detection boats must be such that the concentration variation due to the movement of the plume during a traverse across it is negligible. The survey is considered instantaneous.

Analysis and Interpretation of Tracer Experiment Results

Qualitative results may be readily obtained by plotting the tracer concentration isolines and determining visually the transport pathway by the rate of change of the resulting pattern with respect to the injection point. If the quantity of tracer detected is low, or if natural or incidentally introduced tracers are being used, the analysis obtained can give, at best, only semi-quantitative results. For quantitative results, however, analysis of tracer results is more complicated. The principal techniques found in the literature are briefly discussed below. It should be borne in mind that these techniques were developed mainly for coarser materials, and so might not be suitable for fine sediments.

Count-Rate Balance Method

The count-rate balance method is described in Courtois [25, 26]. This method is designed to obtain the transport rate, or flux (q) passing through a cross section orthogonal to the resultant direction. This quantity is defined as follows:

$$q = \rho \cdot L_t \cdot V_m \cdot E_m \quad (1)$$

where q = transport rate (m^3/day)

ρ = density of sediment

L_t = the width of transport (m)

V_m = the mean transport velocity (m/day)

E_m = the transport thickness (m)

The velocity of transport of the plume (V_m) is determined by the displacement, per unit time, of its centre of gravity. The position of the centre of gravity is obtained from successive surveys. The transport thickness (E_m) is best estimated by coring and interpreting profiles of activity versus depth in the cores.

Lavelle Method

Other authors have developed different methods for determining rates of sediment transport from tracer data. Lavelle et al. (1978) used an advection-diffusion model that described the movement of the sediment cloud in two dimensions.

$$\frac{\partial C}{\partial t} + V_x \frac{\partial C}{\partial x} - D_x \frac{\partial^2 C}{\partial x^2} - D_y \frac{\partial^2 C}{\partial y^2} = 0 \quad (2)$$

where $C(x,y,t)$ represents concentration of the tracer at points x,y , at time t
 V_x is the sediment advection velocity
 D_x and D_y are the sedimentary diffusion coefficients in the x and y directions, respectively.

Inman Method

Inman et al. (1980, p. 1225) used two different approaches to measure the transport in the littoral zone: temporal and spatial.

Temporal Approach

The sampling is made by coring at fixed intervals along a fixed line across the tracer path at varying times. The tracer is injected on one line (L_i) at time (t_o) and is sampled along line (L_e) at time, t_i (t_i varies from 0 to ∞).

If y_i is the distance between L_i and L_e , then

$$M = \int_0^{x_b} \int_{t_o}^{\infty} \frac{y}{t} Z_o \bar{N} dx dt \quad (3)$$

where M is the total mass of tracer injected
 $Z_o(x)$ is the thickness of the mobile layer
 $\bar{N}(x,y,t)$ average mass concentration (mass/volume); (i) refers to a particular sampling event

The characteristic longshore velocity or advection rate of the tracer-marked sand can then be expressed as

$$V_L = \frac{1}{\sum N_i} \sum N_i \frac{y_i}{t_i} \quad (4)$$

and the volume of the sediment flux (Q_L) is

$$Q_L = \int_0^{x_b} V_L Z_o(x) dx \quad (5)$$

The flux can be presented also as the immersed weight per unit time (I_L):

$$I_L = Q_L (\rho_s - \rho) g N_o \quad (6)$$

where Q_L = volume flux of sediment
 N_o = mass concentration (mass/volume) of the local beach sand
 g = acceleration due to gravity
 ρ = density of water
 ρ_s = density of solids

Spatial Approach

Grab samples are collected synoptically over a fixed grid at a fixed time after the injection. The injection line is defined inside this grid as y_o , and the sampling lines (y_i) extend outward on both sides of y_o .

The equation of conservation of the tracer in this case is slightly different from (3):

$$M = \int_0^{x_b} \int_{-\infty}^{+\infty} Z_o \bar{N} dx dy \quad (7)$$

but the volume and the immersed sediment flux are the same as in the temporal approach (5, 6).

Tola Method for Suspended Load

In a more complex treatment, but one more directly related to fine sediment, Tola [91] used an analytical model for the dispersion of fine sediment in a semi-infinite medium of finite and constant depth to determine the horizontal dispersion parameters of a tracer injection. Assuming that the horizontal dispersion follows a Gaussian law, he defined a longitudinal dispersion equation:

$$\sigma_x^2 = \int_{-\infty}^{+\infty} X^2 \bar{C}_m dx / \int_{-\infty}^{+\infty} \bar{C}_m dx \quad (8)$$

where σ_x is the variance or dispersion of the longitudinal Gaussian distribution relative to the GXY axis

x is the abscissa relative to GXY axis, moving with the centre of gravity of the tracer distribution

\bar{C}_m is the maximum average concentration

The denominator in equation (9) represents the total mass along the longitudinal axis (x). A similar expression can be defined for the transversal dispersion with respect to the horizontal axis (y). Further manipulation of these diffusion relationships enables the researcher to track the evolution of the tracer plume with time.

SUMMARY AND CONCLUSIONS

The variety of tracer techniques described in the literature for tracing fine sediments suggests strongly that the choice of technique for tracing fine sediments depends primarily on the nature of the application. The best technique for one site or application is not necessarily the best for another. Most of the investigations were associated with short-term studies of dispersal plumes from dumped dredged spoil. It is clear that, for such studies, where the ability to follow the fine sediment plume in real time is important, the radioactive tracer techniques are preferable. An excellent example is the technique developed by Tola and his colleagues, involving the surficial attachment to short-lived radioisotope Au-198 to natural clayey sediments by means of a special chemical process. The sediment hydrodynamic properties remain virtually unchanged, and the transport of the tracer-marked sediment plume can be followed using dynamic detection procedures. Furthermore, this technique permits the natural sediment to be tagged concurrently with its injection into the system.

Longer-term fine sediment studies must deal with intermittent resuspension and transport events over periods of weeks or months, so longer-lived radioisotopes would be necessary and detection would be confined to the bottom deposits. Use has been made of isotopes such as Iridium-192 (75 days) and Scandium-46 (84 days), with bottom surveys being carried out using a sled-mounted detector or a remotely operated vehicle. However, the use of long-lived radionuclides requires much attention to the safety-related factors, especially if the study is located in multiple-use areas, such as the Toronto waterfront, where, in addition to bathing beaches, there are three drinking water intakes within potential reach of the tracer plume.

Given these safety considerations and the stated goal of focusing on longer-term transport trends, nonradioisotopic tracers might offer definite advantages. Although the literature here is much sparser than for radioactive tracers, the most

attractive among this type are the neutron-activable tracers. These tracers have relatively low environmental impact, but at the same time, they give adequate resolution for most time-integrated transport applications. Their major problem is their inability to provide information on real-time transport patterns.

For studies of long-term fine sediment transport integrated over periods of years, use might also be made of incidentally introduced chemicals adsorbed onto the existing fine sediments being transported. These include both organic chemicals and trace metals. However, because the rate of their introduction into, and removal from, the system is not known, and their respective sources can often only be speculated upon, the level of resolution is less than those described above. As a result, they probably may best be regarded as preliminary indicators of transport, providing only qualitative or directional information on contaminated sediment transport.

Finally, natural tracers, including grain-size properties, heavy minerals, and clay mineral suites, also offer limited use in tracing long-term transport patterns. The major disadvantage of these is that because the shoreline and watershed sediments in the Great Lakes area, for instance, are a complex of glacial and glaciolacustrine sediments, the source inputs of the properties can only be inferred. In any event, grain-size data are important for other aspects of contaminant investigations, so this information is supplied "free" to imply overall transport patterns, especially in the case of the fine sands and silts, where flocculation does not alter the original sediment texture.

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Appendix
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APPENDIX

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