A method is described of tracing the movement of particles conveyed by a fluid medium by determining selected properties of the particles to be traced, selecting or forming tracer particles the corresponding properties of which are substantially similar and having the property that they emit or can be stimulated to emit electromagnetic radiation within a limited range of wavelengths. A quantity of the tracer particles is introduced into the fluid medium and the intensity of electromagnetic radiation within the said limited range from a sample of the fluid medium at or taken from a detection location remote from that at which the tracer particles were introduced is detected. Apparatus for performing the method in situ at the site from which samples are taken is also described.
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METHOD AND APPARATUS FOR TRACING FLUID-BORNE PARTICLES

The present invention relates generally to a method for tracing fluid-borne particles, and to apparatus for use in the method. The invention also extends to and includes a system for tracing such fluid-borne particles and the use of natural and artificial tracer particles, to trace fluid-borne particles.

Urbanisation, industrialisation and construction has upset the natural balance of many environments with man made changes leading to an alteration of the natural hydrodynamics and sediment transport pathways. One of the most significant and often costly effects of these changes is the movement and accumulation of sediment. Alteration of the sediment pathways can cause problems for harbour design, maintenance and management, and require dredge disposal of sediment for clearing navigational channels. In order to determine whether a port expansion or development is viable it is necessary accurately to predict the short and long term effects of sediment movement. If a new port could silt up in months it would no longer be viable due to the large maintenance dredging costs likely to be incurred.

In addition the ability to predict particle movement is important because fine sediment (silts and clays) are highly adsorptive and soak up large quantities of
pollutants including persistent polychlorinebiphenyls, polyaromatic hydrocarbons, radioactive materials, heavy metals, chemicals and nutrients. Therefore anyone involved in sediment movement must know the environmental, ecological and economical effects.

Several ways have been tried to predict the movement of particles. Various particle tracers have been tried with various degrees of success: these include, for example radioactive particles, spores, and glass particles. As a means to predict sediment movement, these are costly, ineffective and distrusted by the public. Water tracers (fluorescent dyes) have also been used in an attempt to infer sediment movement, but the problem with these is that the majority of sedimentary and other particles do not move in the same way as the fluid in which they are borne. At present the standard procedure is to use laboratory studies to develop numerical or mathematical models. These are useful, but have several limitations because the end user does not always know what assumptions have been made to develop the model. Alternatively data may be collected from the field, using water current data, turbidity/sediment concentration readings and salinity. Again, assumptions must be made based on laboratory measurements concerning the current required to move sediment.

There has long been a need for a safe, cheap, simple and
effective means of tracing particles, especially sedimentary particles. If turbidity alone is monitored inaccuracy can result: for example it is very difficult to know whether sediment detected by a turbidity meter is merely circulating around it or whether it is previously deposited sediment which is washing back from a remote location. A tracer would be ideal in many situations similar to this where a high turbidity reading does not necessarily denote local sediment disturbance.

The present invention seeks to provide a new technique for tracing fluid-borne particles, which will avoid many of the problems and inaccuracies encountered with prior art techniques. Embodiments of the invention may enable movement of fluid-borne particles, such as contaminants, to be followed accurately. From accurate data it should then be possible to make predictions, which are important for pollution, environmental and economic reasons.

According to the present invention there is provided a method of tracing the movement of particles conveyed by a fluid medium by determining selected properties of the particles to be traced, selecting or forming tracer particles the corresponding properties of which are substantially similar to those of the said particles and having the property that they emit or can be stimulated to emit electromagnetic radiation within a limited range of wavelengths, introducing a quantity of the tracer
particles into the fluid medium and detecting the emission of electromagnetic radiation within the said limited range from a sample of the fluid medium at or taken from a detection location remote from that at which the tracer particles were introduced, whereby to determine the presence of tracer particles in the sample.

The tracer particle may be made of an artificial material or a natural material. It has been found that certain plastics materials, such as polyethylene and polypropylene are suitable. The particles may be fluorescent or luminescent, and if the latter may be chemiluminescent, bioluminescent or thermoluminescent.

The tracer particles used are preferably environmentally benign and preferably have particle size below 1000 microns. The particle material preferably has a density of from less than 0.5 to 10g/cm³. The detection system used to detect the presence of particles in the fluid may include a laser as the light source for stimulating fluorescent emissions.

The invention also extends to the use of fluorescent tracer particles, and a detection system for example a laser optics system, to trace the presence of fluid-borne tracer particles.

The method may include a preliminary step of determining,
before introducing the tracer particles, the background radiation within the said ranges of wavelengths be used in many types of fluids, including aqueous solutions or suspensions such as sea water, sewage or sludge; a gaseous medium, an oil based medium, eg. petroleum and crude oil; and non-aqueous solvents and suspensions. The detection of tracer particles may involve returning samples to the laboratory where measurements are taken. This however causes delay, and it may be a matter of days before readings can be made on the samples taken, which may be unacceptable. Checking or verification of results is difficult and repeat testing is virtually impossible.

Apparatus capable of testing samples of the fluid at the site from which they were taken would overcome this problem, but of course, since the fluid of interest may be sea or the atmosphere there are considerable difficulties in achieving this.

Another aspect of the present invention therefore seeks to provide an improved technique for tracing fluid-borne particles which avoids many of the problems and inaccuracies encountered when the measuring site is some distance from the place where the samples are taken. The invention enables movement of fluid-borne contaminants to be accurately followed and predicted by in-situ assessments.
It is preferable that the method of the invention includes the further step of passing a sample of fluid medium, at the said detection location, through a flow cell of known cross sectional area, determining the flow rate of fluid medium through the said flow cell, directing stimulating radiation within a first wavelength range into the said flow cell, detecting stimulated (fluorescent) radiation within a second wavelength range and determining from the intensity of detected radiation within the said second wavelength range, the determined flow rate, the known cross sectional area and a previous calibration of the flow cell with a known sample, the concentration of tracer particles per unit volume of the sample under test.

In another embodiment of the invention the flow cell is drawn in a transit through the fluid across a region of interest and the instantaneous output of the said detector is recorded to provide an indication of the variation in concentration of the particles across the said transit.

The present invention also comprehends a method of detecting cracks or fractures in a pipeline conveying a fluid, comprising steps of introducing a plurality of tracer particles the dimensions and physical properties of which permit them to remain in suspension in the said fluid at least in the fluid-dynamic conditions existing
within the length of the pipeline under test, the said tracer particles having the further property of emitting or being capable of being stimulated to emit electromagnetic radiation at or within a known range of wavelengths, and determining the presence of the said tracer particles within a fluid surrounding the pipeline by passing the said surrounding fluid through a flow cell equipped with means for stimulating and detecting electromagnetic radiation within the said known range of wavelengths.

In another aspect the invention provides apparatus for use in tracing fluid-borne particles, comprising means defining a fluid transit chamber having an inlet an outlet and substantially constant cross section to allow fluid to pass therethrough without creating additional turbulence or changes to pressure therein, means for determining the rate of flow of fluid through the said chamber, and means for detecting electromagnetic radiation within a predetermined wavelength range within a fluid in the chamber.

The apparatus of the invention includes means for directing stimulating radiation of a given wavelength or wavelength range into the said chamber. This may be achieved by locating the source close to the chamber or, if circumstances dictate that the source should be remote, an optical fibre or other waveguide may be used.
to convey the radiation to the chamber

The invention also comprehends the use of particles capable of emitting electromagnetic radiation within a restricted range of wavelengths to trace the movement of fluid-borne particles.

The tracer particles may be artificial or natural, environmentally benign fluorescent or luminescent.

Certain particles having acute oral toxicities of more than 16.0 grams per kg have been used and no deaths were recorded in test animals. The physical properties of the tracer particles can be selected to match the target particle in terms of:

- size, with less than 1000 microns being the preferred range;
- shape, the normal shape used is angular and platey;
- density, the range is preferably from less than 0.5 to 10g/cm³
- fluorescence; tracer particles of several different spectra are available.

The duration of detectability for the tracer particle varies from hours to years and this is important for construction and dredging studies in ports where readings can be taken without increasing background. In addition by altering the duration of detectability (lightfastness) a specific organism with a short and finite life span can
be mimicked and traced closely for a normal life-span; this is particularly important when tracing bacterial, planktonic and larvae/egg organisms. It is also possible to look at the interaction from two different sewage-sediment outfalls at the same time.

The surface charge of the tracer particle is preferably negative after introduction to natural waters and the flexibility of the tracer ensures close correlation with the target particle.

The tracer can be used for low energy environments where there exists slow and long-term settling rates, and erosion/resuspension activity. This factor is particularly important for use in industrial/commercial settling tanks, reservoirs, drying areas, estuarine mudflats and beaches. Tracers are also ideal to study high energy environments where it may be impossible to safely install conventional equipment due to damage or loss. Tracers, and especially this type, could be used to study many high energy environments including storms, floods, flashfloods, tidal waves etc.

Possible tracer targets could include fine cohesive and coarse grained sediments, polluted sediment, drill cuttings and other pipe line emissions from oil platforms, sewage, buoyancy plumes, sludge, spores, fish eggs, larvae, pathogens and particle tracing in
atmospheric air quality modelling.

Detection of the tracer particles may be achieved in the laboratory by using an analytical flow cytometer. Use of an analytical flow cytometer allows detection of many different types of tracer particles having different physical, optical or chemical properties. The physical properties of the tracer particles, including size, shape, density, duration of detectability and biodegradability, may be adapted to match as closely as possible those of the target particle. The size may be determined by grinding and/or sieving (for an appropriate size range) or by settling in fluid (for a finer size range) and density may be raised by the introduction of different fillers before extrusion and grinding of the plastics material. This allows particles having many different properties to be made and used to trace almost any target particle, whether in an hydraulic or an aeolian environment. Particles having a wide size range may be needed for some cases.

The technique according to the invention enables a tracer particle to be designed to suit exactly the waterborne, liquid borne, gas or atmospheric borne target particle, which can then be traced accurately, simply and cheaply through any fluid medium, the tracer particles being inert and environmentally benign. For example the tracer technique can be used for tracing dredge sediment,
disposal and dispersal work, polluted sediment movement, capping effectiveness of disposal sites, the construction industry, for monitoring silting on coral reefs to prevent burial, port and harbour management, for navigational channel investigations to monitor sediment returning (both for wavelength and quantity), in beach rejuvenation, land reclamation, lake and reservoir siltation studies, for water and sediment residence times, in dam studies (erosion and silting), lake studies to investigate nutrient build up and eutrophication, in flash flood events, tidal waves and other high energy events, for pollution in gaseous emissions eg. from motor vehicles or from factory outfalls such as in chimneys and leaks from pipe lines and other areas, in sewage to monitor existing outfalls and proposed new outfalls (to investigate the interaction of two or more outfalls), mimic T90 die off rates for E.coli when investigating the breakdown of sewage, for the investigation of seepage from drilling platforms and leaks from pipe lines including pollution discharges. When used for freshwater and aquaculture purposes, eg. tracing fish eggs, plankton, spores, pathogens, the traces can be used to gain insight on feeding and spawning grounds to enable protection, farming and aquaculture.

The tracing techniques are also ideal for tracing atmospheric emission including tracing of chimney stack particulate material, particles and contaminants in gases.
including methane, air quality monitoring, many air-borne pollutants being carried and transported as very fine particles over large distances.

Other applications include rain water run off and flushing rates in urban sewage works, in industrial pipe line systems, and for tracing particles and contaminants in other liquid and gaseous mediums for example oil, chemicals, food stuffs, blood and medical uses.

Specific embodiments of the invention will now be more particularly described, by way of example, with reference to the accompanying drawings, in which:

Figure 1 is a flow chart showing the major steps in conducting an assessment of sediment movement;

Figure 2 is a partial flow chart showing an alternative assessment technique;

Figure 3 is an overall view of the manner in which the latter stages of the assessment of figure 2 are conducted; and

Figure 4 is a schematic sectional view of apparatus for performing the method of Figure 2.

Referring now to the drawings, the main method steps illustrated in Figure 1 comprise a first step 10 in which a sample of the actual sediment to be assessed is taken to determine the physical properties thereof, namely the particle shape, average size, density and, if
appropriate, electronic charge.

Step 2 illustrated in Figure 1 is the step of creating a tracer particle to substantially the same physical properties as the specimen sample taken in step 1. In creating the tracer particle a selected plastics material such as polystyrene, polyethylene or polypropylene may be used which has been formed with the incorporation of an appropriate fluorescent material, and suitable fillers to vary its density. The plastics material is then extruded and ground down to the required particle size. Prolonged grinding makes the particles more rounded if this is required.

Step 3 comprises introduction of a quantity of tracer particles formed in step 2 into the fluid medium bearing the actual sediment to be assessed; step 4 comprises taking samples of the fluid, after having given the tracer particles sufficient time to disperse into the area of interest, the samples being returned to the laboratory where, in step 5, the concentration of fluorescent tracer particles in the samples taken is counted using, for example, a flow cytometer.

Because of the time delay involved in returning samples to a laboratory for testing the alternative and preferred process illustrated in Figure 2 may be utilised. This comprises the same steps 1, 2 and 3 as illustrated in the
method described in relation to Figure 1, but step 4 is performed by conducting a transit across the area of interest and, instead of taking individual samples for subsequent assessment, immediate assessment is made utilising an in-situ detector which will be described in more detail in relation to Figures 3 and 4.

Step 5 is then to record or log the tracer concentration values determined across the transit by the detector.

As can be seen in Figure 3, the in-situ detector generally indicated 12 is adapted to be towed behind a boat 13 by a towing line 14, and to transmit electrical signals representative of the detection taking place along a connection cable 15 to on-board apparatus 16. This apparatus may include a computer with appropriate memory and data recording and storage means. The computer may store data relating to the known fluid and other characteristics of the location. For example, this may be the geographical and navigational details of an area such as a harbour or other navigation system, known flow patterns, tides or the like, which can be of use in setting up a framework within which the data detected at the time from the tracer particles can be assessed.

The sensor 12 is illustrated in more detail in Figure 4, and comprises a casing generally indicated 17 within which is formed an elongate chamber 18 having an inlet
end 19 and an outlet end 20. The inlet 19 and outlet 20 are rounded so that, when the sensor 12 is towed through a fluid medium, the fluid can enter the chamber 18 without substantial disturbance, flow through it without creation of turbulence or pressure changes, and exit through outlet 20 substantially undisturbed. The exterior shape of the casing 17 is chosen so as to pass through the fluid medium without creating significant turbulence, and for this purpose may be provided with means (not shown) for retaining it in a given orientation so that the open end 19 is always aligned with the flow direction.

Within the chamber 18 is located a turbine 21 of a flow meter operable to measure the rate of flow of fluid through the chamber 18 so that, with a knowledge of the cross sectional area of the chamber, it is possible to determine the volume of fluid passing through the chamber in unit time.

Within the casing 17 is located a light source 22 which directs light towards an array of filters 23, 24 which filter out all light except that which will excite fluorescence in the tracer particles used. The light source may be a laser, and the light may be determined to the chamber via a waveguide of optical fibre. A photo sensor 25, sensitive to the fluorescent radiation wavelength, which is within a known narrow range of
wavelengths, is located to receive fluorescent radiation from the particles as they are swept along the chamber 18 having passed the area irradiated by light from the source 22. The photodetector 25 may, in other embodiments, be positioned differently. For example it may be positioned directly in line with the light source, at 90° to the line of the incoming radiation, or even at some other angle between 90° and 180° as illustrated. In whatever orientation there may be provided reflectors in the chamber to concentrate or divert light in its path from the source to the photodetector. The electrical output from the photodetector 25 is passed, together with the output from the flow meter 21, along cable 15 to the processing equipment 16 for determination of the tracer particle concentration.

In addition a standard suite of oceanography data probes could be added in or outside the flow cell 18 to record, for example temperature, salinity, depth and turbidity.

In practice, for work in water, the detector would be towed through the water to monitor dispersion of the tracer particles injected into the fluid and the dispersal of the plume with the result being plotted on board the towing boat to allow re-surveying or tracking of the plume. In a further refinement two or more stimulating light sources, and detectors sensitive to different respective wavelengths may be provided to
detect two or more particles having different fluorescent wavelengths so that the effects of, for example, two or more outfalls can be monitored.

The light sources may be of any known type, including white light sources, or may be lasers having characteristic emission wavelengths, or one of the known lasers capable or producing a range of wavelengths, suitably filtered, if necessary to transmit the light to that which stimulates the required fluorescence. The detector is a robust portable piece of equipment and for work in water is used below the water surface. It may be considered preferable, especially when using particularly delicate or expensive instruments, to minimise the amount of equipment at the sensor head under water. For this reason if a laser is used as the light source in a probe intended to be towed under water the light source may be carried on board the vessel and the light transmitted by optical fibre to the sensor head. In other situations, for example if the equipment is fixed to a secure and immovable submarine structure such as a pier leg or the like, where no buffetting movement is anticipated, all of the equipment may be incorporated in a single self-contained housing. If the water has an excessively high turbidity with a large quantity of disturbed sediment the fluorescent light may be scattered and adsorbed thereby causing diminished emission responses. There is thus an upper limit on background turbidity above which the
possibility of reading error arises. Although as so far described the detector has been envisaged for in-situ testing and measurement in water, it can also be used to measure particles in air, provided the adverse influence of sunlight is removed. There are various ways in which this can be achieved for example using a pump to pull air containing the dust particles through a long pipe. The pipe must be opaque to prevent light entering the cell. The detector may be mounted at the exit of a chimney stack and gas drawn in from a chimney stack into which the tracer has been injected. The tracer concentration is monitored in the issuing gas to give a measure of particle concentration. The fluorometer could also for example, be mounted on a motor vehicle to measure particle emission.

Similarly the detector may be used to measure particles in an oily medium, changes being made to the cross sectional area of the flow cell to compensate for the viscosity of the oil.

The present in-situ method of measuring pollutants in air avoids many of the problems experienced with existing systems such as adhesive pads used to measure the weight increase to determine pollutants and tracer particles, filtering the air to determine total pollutant, gravimetric monitoring to monitor the total weight of pollutants on filter paper, or environmental probes which
filter a known quantity of air and measure the total weight per cubic meter of air.

For use in air rather than water the detector could be housed with an infra-red or similar air particle counter. This would correlate the air-borne particulate material with the air-borne fluorescent tracer material. The laser and the fibre optic system could be housed in a sufficiently small unit to be attached to a remote controlled helicopter which would then be flown around the source of the pollutant stack which would give a precise insitu measurement both temporally and spatially. The same is possible for the water based insitu detector having the capability of housing the laser detection system in a remote controlled boat, submarine or water vehicle. This would reduce costs and increase the number, wavelength and accuracy of sampling points.

The air based system would monitor the fluorescent tracer particles introduced into the air to measure air pollutants both large and small, respirable and non-respirable, fumes, gases and smoke. The air borne system is suitable for use to monitor aerially a surface slick of fluorescent particles mimicking a sewage outfall, release of run off, leak or sediment disturbance. The water based insitu laser system would provide a surface slick monitoring when linked with a remote control boat. The water based laser system could also be used to
investigate suspended layers of pollutant, neutrally buoyant plumes or pollutants trapped on a thermocline, density gradient or similar situations for example soil slicks, sewage plumes and immiscible pollutants. Such plumes, slicks or emissions may be labelled using fluorescent tracer particles with specific physical properties to label the target particles and ensure good detection.

The laser system as described could not be used for sediment analysis unless a "plough and filter" system was also used. For example, a plough device churns up the sediment which is filtered through a flow cell containing a fluorescent detector.

The detector system may be operated by remote control, manual operation or through a remote operating vehicle. For example a sample of sediment would be drawn up through a filter for a known period of time and volume. The intake tube is closed, the filter jetted backwards with clean water to clean it. The sample drawn up would be analysed and although not a rapid analysis time - approximately 1 minute - the system offers extremely rapid response when several lasers/flow detectors are used.

In order to be able to conduct successive tests with accuracy it is preferable that the rate at which the
detectability of the fluorescent particles decays be a known value. Although it is possible to compensate for residual fluorescence by making preliminary background tests, it is more accurate if the residual fluorescence from any previous tracer tests is minimised.
CLAIMS

1. A method of tracing the movement of particles conveyed by a fluid medium by determining selected properties of the particles to be traced, selecting or forming tracer particles the corresponding properties of which are substantially similar and having the property that they emit or can be stimulated to emit electromagnetic radiation within a limited range of wavelengths, introducing a quantity of the tracer particles into the fluid medium and detecting the intensity of electromagnetic radiation within the said limited range from a sample of the fluid medium at or taken from a detection location remote from that at which the tracer particles were introduced.

2. A method according to Claim 1, characterised in that it includes the further step of passing a sample of fluid medium, at the said detection location, through a flow cell of known cross sectional area, determining the flow rate of fluid medium through the said flow cell, directing stimulating radiation within a first wavelength range into the said flow cell, detecting stimulated (fluorescent) radiation within a second wavelength range and determining from the intensity of detected radiation within the said second wavelength range, the determined flow rate, the known cross sectional area and a previous calibration of the flow cell with a known sample, the concentration of tracer particles per unit volume of the fluid medium.
sample under test.

3. A method of tracing fluid-borne particles according to Claim 2, characterised in that the flow cell is drawn in a transit through the fluids across a region of interest and the instantaneous output of the said detector is recorded to provide an indication of the variation in concentration of the particles across the said transit.

4. A method of detecting cracks or fractures in a pipeline conveying a fluid, comprising steps of introducing a plurality of tracer particles the dimensions and physical properties of which permit them to remain in suspension in the said fluid at least in to fluid-dynamic conditions existing within the length of the pipeline under test, the said tracer particles having the further property of emitting or being capable of being stimulated to emit electromagnetic radiation at or within a known range of wavelengths, and determining the presence of the said tracer particles within a fluid surrounding the pipeline by passing the said surrounding fluid through a flow equipped with means for stimulating and detecting electromagnetic radiation within the said known range of wavelengths.
5. Apparatus for use in tracing fluid-borne particles, comprising means defining a fluid transit chamber having an inlet an outlet and substantially constant cross section to allow fluid to pass therethrough without creating additional turbulence or changes to pressure therein, means for determining the rate of flow of fluid through the said chamber, and means for detecting electromagnetic radiation within a predetermined wavelength range within a fluid in the chamber.

6. Apparatus according to Claim 5, characterised in that it includes means for directing stimulating radiation of a given wavelength or wavelength range into the said chamber.

7. The use of particles capable of emitting electromagnetic radiation within a restricted range of wavelengths to trace the movement of particles having similar physical properties.
FIG 1

1. TAKE SAMPLE OF SEDIMENT
   DETERMINE PHYSICAL PROPERTIES

2. CREATE TRACER PARTICLE HAVING
   SAME PROPERTIES PLUS FLOURESCENCE

3. INTRODUCE TRACER PARTICLE
   SAMPLES TO FLUID

4. SAMPLE FLUID
   RETURN TO LABORATORY

5. ASSESS TRACER CONCENTRATION
   IN SAMPLES

FIG 2

4. CONDUCT TRANSIT WITH
   IN SITU SENSOR

5. LOG TRACER CONCENTRATION
   VALUES ACROSS TRANSIT

SUBSTITUTE SHEET
A. CLASSIFICATION OF SUBJECT MATTER
IPC 5 GO1P5/20 GO1M3/22

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 5 GO1P GO1M GO1F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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<td>JOURNAL OF PHYSICS E. SCIENTIFIC INSTRUMENTS vol. 15, no. 11, November 1982, BRISTOL GB pages 1131-1138 R.THORN ET AL. 'Non-intrusive methods of velocity measurement in pneumatic conveying' see page 1134, right column, paragraph 5 - page 1135, right column, paragraph 3; figure 7</td>
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Date of the actual completion of the international search
18 January 1994

Date of mailing of the international search report
11.02.94

Name and mailing address of the ISA
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