Tracer dispersal in sandy sediment porewater under enhanced physical forcing

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Abstract

This research (1) integrated a fluorescent dye injection and monitoring system for measuring the mixing of a fluorescent dye tracer (fluorescein) in permeable (sandy) sediments with a cabled ocean observatory, Kilo Nalu, Oahu, Hawaii, and (2) used this system to conduct remotely controlled in situ measurements of wave-enhanced porewater mixing in a physically well-characterized wave-dominated setting. Laboratory results indicated that the fiber-optic sensor is effective at measuring fluorescence-traced enhanced mixing in sandy sediments. Observed dye mixing, driven by paddle-generated surface waves in a laboratory wave channel was 2–3 orders of magnitude greater than molecular diffusion, and decreased with depth in the sediments. Similarly, field experiments used a remotely controlled submersible syringe pump for fluorescent tracer injection into sediment that was monitored with a fiber-optic sensor. These experiments were carried out at 10 m water depth, with mean wave heights of 0.3–1.5 m and periods of ~15 s. At 15 cm sediment depth, transport rates of 0–5 cm h\(^{-1}\) were measured, with maximum dispersion coefficients 2–3 orders of magnitude faster than sedimentary molecular diffusion. Hydrodynamic measurements collected simultaneously via Kilo Nalu correlated with porewater transport, with significant wave height and diffusion having the strongest covariation.

Keywords: Permeable sediments; Sandy sediments; Fiber-optic sensor; Diffusion; Porewater transport

1. Introduction

Hydrodynamic processes in permeable sediments (permeabilities \(>10^{-12}\) m\(^2\)) can enhance solute transport by several orders of magnitude (Huettel and Webster, 2001). Although there are numerous mathematical treatments of wave-driven solute exchange between permeable sediment porewater and the overlying seawater (e.g., Harrison et al., 1983; Shum, 1992; Webster and Taylor, 1992; Webster, 2003), there have been few attempts to measure this exchange in the field, notably Webb and Theodor (1968), Precht and Huettel (2004), and Reimers et al. (2004).

Determining solute fluxes in permeable sediments is difficult. For example, fluxes determined by benthic chambers may be significantly underestimated because of the attenuation of the effects of important hydrodynamic processes such as surface gravity waves and bottom currents (Jahnke et al., 2004).
Similarly, laboratory experiments using wave channels or flumes may not simultaneously mimic all of the hydrodynamic processes that occur in the field, including varying bottom currents (magnitude and direction), varying surface waves (amplitude, period, direction), and sand ripple formation and migration. Thus, there is a need for \textit{in situ} methodologies that can provide estimates of solute fluxes with minimal hydrodynamic or sedimentological disturbance.

Enhanced mixing of solutes through permeable sediments is important to consider for many reasons. For example, sandy sediments are effective at trapping and transporting labile particulate organic carbon from the water column to the sediment where organic carbon becomes mineralized by microbial processes. Wave-forced nutrient input from the sediments to the overlying water may stimulate the production of phytoplankton or coral species (e.g., Haberstroh, 1994; Falter and Sansone, 2000; Jahnke et al., 2000). Also, sandy sediments are important habitats for bottom fish, invertebrates, and other commercially important species, and the proper management of these environments requires knowledge of nutrient fluxes from the sediments.

The objectives for this research were to develop an observatory-usable tracer injection and fiber-optic monitoring system for sandy sediments, using commercially available components, and to interface the system with a cabled ocean observatory, Kilo Nalu (Oahu, Hawaii). This system was used to conduct remotely controlled \textit{in situ} measurements of wave-enhanced porewater mixing in a physically well-characterized, wave-dominated setting.

2. Methods

2.1. Laboratory experiments

A visible-light spectrometer (USB2000-FL, Ocean Optics) and a blue LED excitation source (USB-LS-450, Ocean Optics) were used in conjunction with a 15-cm long stainless steel fiber-optic sensor (F100-angle, Ocean Optics). A 600-μm diameter bifurcated fiber-optic cable assembly (BIF600-vis–NIR, Ocean Optics) was used to connect the sensor to the source and the spectrometer.

Fluorescein, C_{20}H_{10}O_{5}Na_{2}, has a maximum excitation wavelength of 490 nm. The peak of fluorescein emission in well-sorted commercial silica sand (porosity = 0.45, mean grain size = 0.35 mm, and permeability = \(2.5 \times 10^{-10} \text{ m}^2\)), as used in our laboratory studies, occurred at 512 nm (Fig. 1). A separate peak emission occurred at 490 nm due to the reflection of the blue excitation light off of sand grains (Fig. 1).

Laboratory dye migration experiments were performed in a 7.5 m × 0.13 m × 0.37 m (length × width × depth) wave channel (Fig. 2). A motor-driven paddle at one end of the channel generated progressive surface waves that dissipated by breaking on an artificial beach at the opposite end of the channel. Shredded PVC was used as a diffuser behind the paddle to dampen the backwashing of water from the paddle return with the progressive surface wave. A 2-m section of the middle of the wave channel was filled with silica sand (see above) to a mean depth of 10 cm, and was filled with tap water to 15 cm above the flattened water-sand interface. A fiber-optic sensor was buried horizontally at 2-cm sediment depth for one set of experiments and 5-cm sediment depths for another set of experiments, at a distance of 4 m from the paddle.

A computer-controllable syringe pump (NE-515 OEM, New Era Pump Systems), 1/16 in. stainless steel tubing, and a dye reservoir (1L, Cubitainer®) was used to inject 2 mL of 10 mg L⁻¹ aqueous fluorescein at 10 mL min⁻¹ into the sand adjacent to the sensing end of the probe so that the sensor tip was in the center of the resulting dye sphere. To prevent any additional dye from diffusing out of the tubing, a small cap with a hole made from a needle was used so that dye could only exit the tubing when
force was applied to the syringe plunger. In preparing the dye solution, small amounts of methanol were added successively to the solution until neutral buoyancy in freshwater was obtained. Temperature-driven density differences between the wave-channel water and the dye were avoided by allowing the dye and the water to equilibrate to ambient temperatures before dye injection. The fiber-optic sensors were calibrated using silica sand homogenized with known concentrations of fluorescein in 50 mL beakers. During experiments, measurements were logged ca. every 10 s using Matlab software (Mathworks). The surface waves were maintained at a constant amplitude of 4 cm, a period of 0.46 s, and a wavelength of 0.4 m, as determined from digitized video.

Data from these enhanced mixing experiments were modeled as the radial diffusion of a dissolved substance from a sphere. Thus, the concentration, \( C \), at a distance \( r \) and time \( t \) for release of a substance with initial concentration \( C_0 \) and diffusion coefficient \( D \) from a sphere of initial radius \( a \) is (Crank 1957)

\[
C = \frac{1}{2} C_0 \left[ \text{erf} \left( \frac{a + r}{2\sqrt{Dt}} \right) + \text{erf} \left( \frac{a - r}{2\sqrt{Dt}} \right) \right]
- \frac{C_0}{r} \sqrt{\frac{Dt}{\pi}} \left[ e^{-(a-r)^2/4Dt} - e^{-(a+r)^2/4Dt} \right].
\]  

(1)

When \( r = 0 \) (as in these experiments, in which the concentration observed by the sensor is at the center of the sphere), the equation reduces to

\[
C = C_0 \text{erf} \left( \frac{a}{2\sqrt{Dt}} \right) + \frac{a C_0}{\sqrt{Dt\pi}} e^{-a^2/4Dt}.
\]  

(2)

Although Eqs. (1) and (2) are diffusive models, advective transport rates are inferred from the diffusive model by varying \( r \) in Eq. (1) as a function of time, \( t \). Enhanced tracer mixing was compared with molecular diffusion of fluorescein for these sediments. The free-solution molecular diffusion coefficient, \( D_o \), is 1.7 m² h⁻¹ (Kaiser and Maurice, 1964). Sedimentary molecular diffusion of fluorescein, \( D_S \), is equal to \( D_o \) divided by the tortuosity, which was estimated empirically from inverse porosity, \( 1/\varphi \) (Boudreau, 1997; Ullman and Aller, 1982). Table 4.11 in Boudreau (1997) lists several empirical estimations of tortuosity from porosity. Based on orders-of-magnitude differences in measured diffusion rates in our experiments, tortuosity differences between the empirical relationships presented in Boudreau (1997) did not play an important role in our model calculations. The inverse of porosity has proven to be a useful estimate for tortuosity when porosity values are less than 0.7 (Ullman and Aller, 1982), and the value obtained from this relationship was approximately the median of all other empirical tortuosity values calculated using the equations presented in Boudreau (1997).

2.2. Field experiments

The experimental setup of the laboratory experiments was integrated into the Kilo Nalu cabled ocean observatory (Fig. 3) on the south shore of Oahu, Hawaii in order to monitor in situ porewater mixing (Pawlak et al., 2006). The Kilo Nalu Observatory supplies data (TCP/IP (Ethernet)) and power (24 V DC) connections to a suite of instruments that monitor surface waves, tides, currents and water quality (http://www.soest.hawaii.edu/OE/KiloNalu).

The spectrometer and syringe pump used in the laboratory experiments were installed in custom-fabricated PVC 10.2-cm (i.d.) and a 15.2-cm (i.d.)
PVC housings, respectively, and mounted on a ~4–5 × 10^3 m^2 area of flat sandy seafloor situated among extensive reef development at 10 m water depth.

Grain size distribution was homogenous with sediment depth, and was determined from intact cores (7.5 cm diameter) down to ~20 cm, sectioned every 2 cm (sediment porosity = 0.49; ~75% of sediment grains within 125–250 μm). No obvious infaunal burrows were observed within the experimental sediments. Buckets of sand obtained from the site in bulk (down to about 25 cm sediment depth) were taken to the laboratory and used to produce a 1-m long column of sediment; the permeability was determined to be ~3.9 × 10^{-11} m^2 using the falling head method (e.g., Wit, 1967; data courtesy of A. Hannides). This was comparable to the permeability of 8.8 × 10^{-11} m^2 estimated from the Carman–Kozeny equation (Boudreau, 1997). It was not determined whether or not permeability in these sediments changed with sediment depth, but we conclude that the likely minor changes in permeability with depth were inconsequential.

Reagent grade NaCl (Aldrich) was used to make the dye neutrally buoyant in seawater. Porewater obtained from wells within 2 m of our sampling site at a range of sediment depths (0, 7.5, 15, 20, 30, 40, 50 cm depth-data not shown) on May 16, 2006, and June 9, 2006, showed that salinity did not vary with depth. In addition, long-term water column observations of temperature and conductivity do not indicate evidence of submarine groundwater discharge at this particular site. Additionally, during the March experiments, a multi-parameter sonde (YSI Inc.) showed that salinity in the water column, 2 m above the sediment–water interface, during the course of individual experiments (2–20 h) did not vary substantially (at most 0.2). Buoyancy-driven mixing may have been responsible for some of the dye movement, but is considered negligible compared to the rate of wave- or current-induced mixing at this site.

Fig. 4 illustrates how the in situ experiments were configured on the seabed. A wire mesh grid (0.2 m × 0.4 m) with 1.27 cm spacing was used to ensure proper sediment depth placement of the fiber-optic sensor and tubing, with the sensor placed 15 cm below the top of the grid, and the top of the grid level with the sediment–water interface. The fiber-optic cable was secured to the wire mesh with cable ties and buried in the sediments. Burial eliminated flow disturbances and vibrations from currents in the water column. After the wire mesh was buried, the overlying sediment surface was smoothed by hand. Based on visual observations of sand movement under stronger physical conditions, the sensor depth was uncertain to ±2 cm.

The serial data ports of the spectrometer and the syringe pump were connected to the Kilo Nalu network using a serial-to-ethernet portserver.
(TS 4 MEI, Digi International Inc.), which enabled network communication with computers on shore. Instruments were powered using a 24–12 V and a 24–5 V DC/DC converter (VHB75-D24 Series, V-Infinity LCC) for the syringe pump and spectrometer, respectively.

Dye migration experiments were conducted on successive days on February 4–9, 2006 and March 1–10, 2006. Surface-wave characteristics and near-bottom currents were measured using an acoustic Doppler current profiler (ADCP; Workhorse Sentinel, Teledyne RD Instruments) connected to the Kilo Nalu Observatory and stationed 10 m from the porewater tracer experiments. The ADCP measured velocities over the 10-m water depth in 0.5-m bins, with pressure and velocity sampled simultaneously at 1 Hz.

3. Results and discussion

3.1. Fiber-optic fluorescence sensors in sandy sediments

Calibration curves were determined for fluorescein standards prepared in silica-sand sediments (Fig. 5). One difficulty encountered was that fiber-optic sensor response, although extremely rapid (< 2 s), varied over repeated insertions into the same standard, despite the same emersion time and depth of the sensor in the sand. The fluorescence variability was attributed to irregular packing of sand grains around the tip of the sensor after each insertion, which changed the effective volume of porewater observed by the spectrometer. This fluorescence variability was as high as 25%, but was usually around 10–12%.

Attempts to correct the variability included adding a modified flow-through tip, and adding clear tubing to the sensing end of the probe to standardize the porewater volume that was illuminated by the LED. However, these sensor tip modifications were not effective, as calibration regressions were poor and detection limits were substantially increased. We chose to accept the variability and to normalize fluorescence measurements to initial starting conditions for each insertion. In addition, preventing movement of the probe in the sand, once emplaced, largely eliminated changes in observed fluorescence, presumably by eliminating changes in the volume of illuminated porewater. Fortunately, once the sensor is placed at depth, sand grain repositioning is unlikely below the top few centimeters of the sediment.

3.2. Enhanced tracer dispersal

Laboratory wave-channel experiments exhibited enhanced porewater transport of fluorescein at both the 2- and 5-cm sediment depths, as compared to sedimentary molecular diffusion \((D_s = 7.8 \times 10^{-7} \, \text{m}^2 \, \text{h}^{-1})\), and dye transport was slower with increasing depth in the sand (Fig. 6). The shape of the curves differed from a purely diffusive profile, which suggests that advective processes also played a role. A weak backflow within the bed was observed towards the paddle, likely driven by a Lagrangian mass transport (Stokes drift) associated with finite amplitude shallow-water waves (see Kantha and Clayson, 2000).

Similarly, the field tracer experiments (Figs. 7 and 8) revealed enhanced diffusivities that were 2–3 orders of magnitude greater than molecular diffusion under the existing wave conditions (i.e., the tracer decay corresponded to an apparent diffusion coefficient of \(\sim 10^{-2}–10^{-3} \, \text{m}^2 \, \text{h}^{-1}\), compared to the value of \(10^{-7} \, \text{m}^2 \, \text{h}^{-1}\) for molecular diffusion). Physical conditions during dye migration experiments conducted on successive days on February 4–9, 2006 are shown in Fig. 9(a) and (b), which shows significant wave height and peak wave-period calculated from spectral analysis of the ADCP data over 1-h intervals. There is little
variation in significant wave height, although changes in period associated with changing swell sources are evident. Near-bed wave-induced velocities, calculated using linear wave theory, are shown in Fig. 9(c), along with steady current velocities averaged hourly over the bottom 4 m of the water column (Fig. 9(d)). The wave-induced velocities are relatively constant, reflecting the steady wave forcing. Mean near-bed currents, which show more significant variability, respond to the barotropic surface tide, but are also strongly influenced by local baroclinic forcing (Alford et al., 2006).
The physical conditions during the February tracer experiments were relatively weak, and direct relationships could not be determined between porewater decay experiments and physical forcing. Significant wave heights tripled in March compared to the conditions in February, and additional tracer injections were performed (Fig. 8). Physical conditions also changed both throughout individual experiments and between experiments (Fig. 10). Similarly, there was considerably more variability between successive tracer injections in the March field experiments compared to the February experiments.

The data from the one-dimensional mixing experiments (Figs. 7 and 8) cannot be explained by diffusion alone (Eq. (1)), suggesting that advective processes are also contributing to tracer dispersal. A closer examination of the tracer decay raw data and enhanced diffusion rates of the model in Eq. (1) can provide information regarding the upper and lower bounds of both diffusion and advection. We can use Eq. (1) to estimate advective velocities when diffusion fails to explain the rate of tracer decay; this is done by calculating the distance the dye must have moved \( r \) given the observed concentration of the dye at a given rate of diffusion. An advective velocity is then calculated from the temporal rate of change of \( r \). When the measured concentration is above the maximum concentration predicted for a particular diffusivity, no solution is obtained for \( r \). While it is unlikely that the diffusion rate would be invariable over the course of individual experiments, this analysis can provide some guidance on the role of advective processes in the observed dispersion. The analysis additionally requires that the initial injection is spherical, with constant concentration throughout. Potential mixing upon injection and dilution of dye at the edges violates this assumption, so we have discarded the first and last 15% of the data sets in our analyses.

The calculated advective velocities were averaged for each experiment so that they could be correlated with average physical processes measured by the ADCP. Fig. 11 shows the relationship between

![Fig. 11. Daily experiment-averaged characteristics of advective velocity (m h\(^{-1}\)) versus physical parameters during the in situ experiments (\(\square\): February, \(\circ\): March). Open symbols indicate a westward direction of currents (predominant), while closed symbols are eastward flowing currents. Bars on plots are ±1 standard deviation of the mean. The March point identified by the square is noted for its different physical regime compared to the other points. Panels (a) and (b) show near-bed current velocity (\(V_c\)) and the wave velocity (\(V_w\)), respectively. Panels (c) and (d) show significant wave height (\(H_{1/3}\)) and wave orbital length (\(D_{o,bo}\)), respectively, derived from spectral analysis of field measurements.](image)
various physical processes and porewater advection for February and March data. There was a positive relationship between porewater advective velocity and current velocity for March, but not February. The March 10 experiment appears as a notable outlier, as identified by the boxed data points. For this experiment, there may have been a shift in the mechanism responsible for the dominant physical forcing, given that the surface-wave height was the highest (with correspondingly larger wave velocities and wave orbitals) and bottom currents were slightly attenuated. The March 10 experiment displayed the fastest tracer decay among all the experiments, and came 2 and 3 days after the slowest decay rates observed, thus demonstrating the dynamic nature of sediment porewater mixing rates.

The 3–5 cm h\(^{-1}\) wave-induced porewater advective velocities inferred in this study compare well with the results of the few previous studies of wave-induced porewater circulation (Table 1). Notably, porewater advective velocities measured by Riedl et al. (1972) and Shum (1993) are both similar in magnitude, and appear to be typical of finer grained sandy sediments.

Average wave velocity and wave height did not exhibit any obvious relationship with porewater advection, however, March wave orbitals demonstrated a slightly negative relationship (excluding the March 10 experiment; Fig. 11). This might be accounted for if the role of bedforms is considered. If sand ripples are effective in driving advective flows at 15 cm sediment depth, as suggested by Shum (1992), and sand ripple wavelengths are dependent upon wave orbital diameters (Wiberg and Harris, 1994), then a longer sand ripple wavelength would decrease advective velocities generated due to the corresponding decrease in pressure gradient within the bed. As ripple wavelength increases, the pressure differential between crest and trough is largely invariable while the distance over which it acts increases. However, we might expect that this relationship would break down for high orbital amplitudes as bedforms transition from an orbital ripple regime to an anorbital regime where wavelength scales with particle size (Wiberg and Harris, 1994).

Although our experiments to date cannot discriminate between individual physical processes, they nevertheless provide information on the overall rate of porewater movement. We can calculate the maximum diffusion coefficient for each tracer decay curve using Eq. (1) and observe how the diffusion coefficient changes throughout any given experiment if there were no advection (Fig. 12). Experiment-averaged diffusion coefficients and their corresponding standard deviations are largely constrained within a maximum diffusion coefficient between \(10^{-2}\) and \(10^{-3}\) m\(^2\) h\(^{-1}\), 3–4 orders of magnitude faster than sedimentary molecular diffusion (order of \(10^{-7}\) m\(^2\) h\(^{-1}\)). The maximum diffusion coefficient corresponds to significant wave height for the March experiments, which might be expected since wave action can increase porewater motion and dispersion independent of an advective response.

Based on the variability in physical forcing and the weak correlations with advective velocity, it may be that one or more physical characteristics may not be solely responsible for the transport and movement of sediment porewater. Additionally, the sum of physical forces combined may not necessarily work in conjunction, but may perhaps interfere with one another, thus confounding interpretation of porewater

<table>
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<tr>
<th>Transport process</th>
<th>Velocity (cm h(^{-1}))</th>
<th>Method (laboratory/field modeled)</th>
<th>Reference</th>
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<tr>
<td>Advection, waves</td>
<td>130</td>
<td>Field</td>
<td>Webb and Theodor (1968)</td>
</tr>
<tr>
<td>Advection, waves</td>
<td>26</td>
<td>Field</td>
<td>Precht and Huettel (2004)</td>
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<td>3.6</td>
<td>Modeled</td>
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<td>Reimers et al. (2004)</td>
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<td>3–5</td>
<td>Field</td>
<td>This study</td>
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<td>Laboratory</td>
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<tr>
<td>Diffusive (for O(_2))</td>
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<td>Field</td>
<td>Precht and Huettel (2004)</td>
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</table>
mixing results. Because of these multiple physical interactions, more elaborate experimental setups are required. Our current research addresses these issues with the development of an array of fiber-optic sensors (up to 7) to capture multi-dimensional tracer movement. A camera system, scanning laser altimeter, and subsurface pressure transducers are also being added to the observatory to monitor changes in bedforms, including sand deposition/resuspension and sand ripple formation and migration, which may significantly affect to advective porewater transport (see discussion of the latter by Huettel et al., 1996; Huettel and Webster, 2001).

4. Conclusions

The fiber-optic system worked well in both laboratory and field applications, providing useful data on the hydrodynamic processes occurring in permeable sediments. These processes include oscillating flows from passing surface waves and advection due to sand ripples and bottom currents, which can increase porewater transport by several orders of magnitude over molecular diffusion. This increased transport can induce biogeochemically important rates of particulate matter and solute exchange between the overlying water and permeable sediments.

When making geochemical measurements in these types of environments, it is critical that simultaneous physical oceanographic data be recorded, the simplest being measurements of surface-wave direction, amplitude and period, and bottom current speed. Additionally, observations of bedforms, such as migrating sand ripples, must be taken into consideration due to the advective flows they create in sandy sediments (Webb and Theodor, 1968; Shum, 1992; Huettel and Webster, 2001).

Our research efforts have led us to in situ data acquisition via a cabled ocean observatory, the benefits being (1) simultaneous data acquisition of physical conditions; (2) real-time, long-term deployments; and (3) adaptive experimentation. The integration of our system to the observatory is an important step in evaluating hydraulically driven solute exchange between seawater and underlying sandy sediments because it allows measurement of enhanced solute exchange rates in a natural setting under a range of well-monitored hydraulic conditions.

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