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Transport of Gas and Solutes in Permeable Estuarine Sediments

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LONG-TERM GOALS

The long-term goals of this project are to 1) quantify gas bubbles and their composition in shallow nearshore marine sand and 2) to assess the role of gas bubbles in shallow sandy coastal sediment for the transport of solutes through the sand and sediment-water exchange of matter. Due to their compressibility, gas bubbles embedded in shallow water sediments cause interstitial water oscillations under passing surface gravity waves, and these oscillations provide a mechanism for enhanced solute dispersion and flux.

OBJECTIVES

1) To detect gas bubbles and in coastal and estuarine sand deposits and to assess temporal and spatial distribution of sedimentary bubbles in sublittoral beds including sands inhabited by microphytobenthos and seagrass.
2) To quantify the size range and composition of the gas bubbles in the sediment and the overlying water.
3) To determine the volume change and migration velocities of interstitial bubbles and the links to pressure oscillations
4) To assess dispersion and transport of solutes caused by bubble volume change and migration under different pressure conditions.

APPROACH

The project combines instrument development with laboratory and field measurements. The instrument development includes testing of a hand-held ultrasound device for the detection of small gas bubbles embedded in sandy sediment and the construction of a laboratory column reactor that allows application of realistic pressure oscillations to incubated sediment cores with gas enclosures. These approaches are used for the

- *Measurement of gas release volumes from the sediments using benthic chambers or bubble traps.* While the chambers allow changing the advective transport component and thereby also gas ebullition, the bubble traps collect bubbles under the natural flow conditions.
- *Determination of gas composition.* The composition of the sampled gas volumes is analyzed using a Gas Chromatograph (GC).
- *Measurement of gas bubble dimensions and distribution.* Bubble size analyses are performed using ultrasonic and optical methods on sediment cores maintained at in-situ pressure, light and temperature and without changing the orientation of the core.
- *Mapping of the spatial and temporal distribution of high sedimentary photosynthetic production and oxygen supersaturation and depletion as potential sites for free gas development.* Measurements with an in-situ fluorescence detector and an oxygen microprofiler are used to map areas of benthic photosynthetic oxygen production.
- *Determination of gas content, distribution and migration in the surface sediment.* Content, distribution and migration of free gas in the surface layers of the sand sediment is investigated with a tunable ultrasound square wave pulser, with measurement rate adjustable from 10 Hz to 1000 Hz in 10 Hz increments connected to one sending and one receiving high-frequency transducer (1 MHz).
- *Measurement of solute transport caused by bubble compression and migration.* This process is investigated in a laboratory column setup which allows measurement of the migration behavior and velocities of gas bubbles in permeable sandy sediments under the influence of sinusoidal pressure oscillations and determination of transport rates, dispersion and interfacial flux of solutes and colloidal material.

For a more detailed description of the methods and technologies used in this project in the previous years we refer to the first and second annual reports. Below a summary of the work completed within the reported project year 2009/2010.

WORK COMPLETED

Assessing spatial and temporal distribution and characteristics of bubble ebullition in a shallow coastal environment with strong benthic photosynthesis (May 26-28).
The goal was to determine the spatial and temporal distribution of the gas release and the volume and composition of gas released. The working hypothesis was that maximal volumes are released during mid day when light intensity is highest, and that the composition of gas is dominated by oxygen. Six 50-cm diameter transparent PEVA plastic domes were anchored 10 cm above the sediment surface to trap gas bubbles released from the sand at the study site at St. Joes Bay.

Measurement of the effect of gas release for nutrient release from sandy sediments measured in St. Joe Bay sediments using benthic chamber (two field trips: 3/2010 and 5/2010)
The goal was to quantify fluid exchange between sediment and overlying water caused by a known ebullition rate and to assess the potential effects on nutrient exchange. The working hypothesis was that nitrogen ebullition enhances the interfacial fluxes and water and solutes. Two chambers (19 cm inner diameter, 30 cm height) were equipped with a gas line that released nitrogen bubbles within the sediment, two control chambers had no gas lines. Nitrogen Gas was pumped at 4 ml min^{-1} through the

sediment into each chamber. Samples collected at hourly time intervals were analyzed for Bromide tracer content, dissolved and total phosphate.

Assessing the detection limit for small bubbles (50-200 µL) with our new acoustic detection method. The goal of this experiment was to assess the minimum detectable bubble size in submerged saturated sand. The working hypothesis was that the high acoustic frequency used (1 MHz) permits detection of bubbles in the millimeter size range. Single bubbles were injected at 16, 27, 40, or 56 mm below the sediment-water interface into sand enclosed in an acrylic cylinder, and bubble volume was increased at 50 µL increments until ebullition occurred. An A303S transducer, positioned in the overlying water, recorded pulse-echo signals before any air was injected and then after each 50 µL air injection.

Detection of small bubbles produced by benthic photosynthesis
The goal was to assess whether the small bubbles generated by microalgae in the surface layer of submerged sand can be detected non-invasively with our acoustic method. The working hypothesis was that our acoustic method can distinguish between sediments with and without embedded photosynthetic gas bubbles. Natural marine sand was placed in a trough and covered with seawater. The sand was illuminated by a 400-W metal halide lamp facilitating photosynthesis by the sedimentary diatoms and cyanobacteria and bubble formation in the sand surface layer. Acoustic readings measured before and after bubble production in the sand were compared using pulse-echo mode and through-transmission mode.

Field experiment comparing pulse-echo readings in natural sands with and without bubbles (6/21010). The goal was to assess whether our hand-held acoustic method is suitable for non-invasive detection of small bubbles embedded in sand in-situ. The working hypothesis was that our method can follow the production of sedimentary bubbles in sands during the daily light cycle. The measurements, carried out in St. Joseph Bay, FL (sand with diatoms) compared areas with and without embedded bubbles over the diurnal cycle following the evolution of the bubbles. Control measurements were taken at the end of the day after releasing the bubbles from the sand.

Investigation of the sand volume and sand surface area affected by ebullition from a single source. The goal was to assess the pore water flow pattern produced by the ebullition and to assess the vertical and horizontal components of the pore water transport. The working hypothesis was that a substantial lateral component of the pore water flows expands the sediment volume affected by the ebullition. Sand was placed in a 57-cm diameter drum, and the pore water was stained with fluorsceine dye. Gas was released at 2.4 cm sediment depth in the center of the core at a gas flow rate of 27 mL min^{-1}. After 10 to 40 h, the dye wash out patterns in the sand core were measured.

Generation of controlled pressure oscillations in a laboratory column and investigation of the effect of sedimentary bubbles of solute transport.
The goal was to produce sinusoidal pressure oscillations as caused by passing surface gravity waves that compress gas bubbles thereby causing transport and to assess the effect of ascending bubbles. The working hypothesis was that the defined pressure oscillations cause bubble release. We designed a set up consisting of a linear motor acting on the diaphragm of a pressure transducer that transferred the programmed pressure oscillations to a laboratory pressure tank. Flow caused by ascending bubbles was characterized using particle image velocimetry (PIV).

Participating Scientists and students. Scientists and students participating in this work are Dr. Markus Huettel (PI), Dr. Richard Wildman (Postdoc), Chiu Cheng (graduate student), Lee Russell (graduate student), and Stacia Dudley (undergraduate student).

RESULTS

Assessing spatial and temporal distribution and characteristics of bubble ebullition in a shallow coastal environment with strong benthic photosynthesis (May 26-28).
Photosynthetic bubble ebullition followed a diurnal cycle with maximal volumes released during the late afternoon. This result is caused by the gradual build up of the oxygen concentrations in the pore water throughout the day, and only when supersaturation concentrations are exceeded oxygen bubbles start forming in the pore space. Bubbles have to grow to approximately 1 – 2 mm in diameter before their buoyancy can overwhelm the viscous forces and ebullition starts. Bubbles collected before 16:30 are mostly released by the surface of the sediment, while the large increase in ebullition rate in the late afternoon is supported by bubbles released from the upper sediment layer (Fig. 1). A rough approximation suggests that about 1 L of gas was released per m^2 sand on a sunny day. Because the bubbles are very small, they equilibrate within minutes with the partial gas pressures in the shallow water and, thus, their composition is very similar to air.

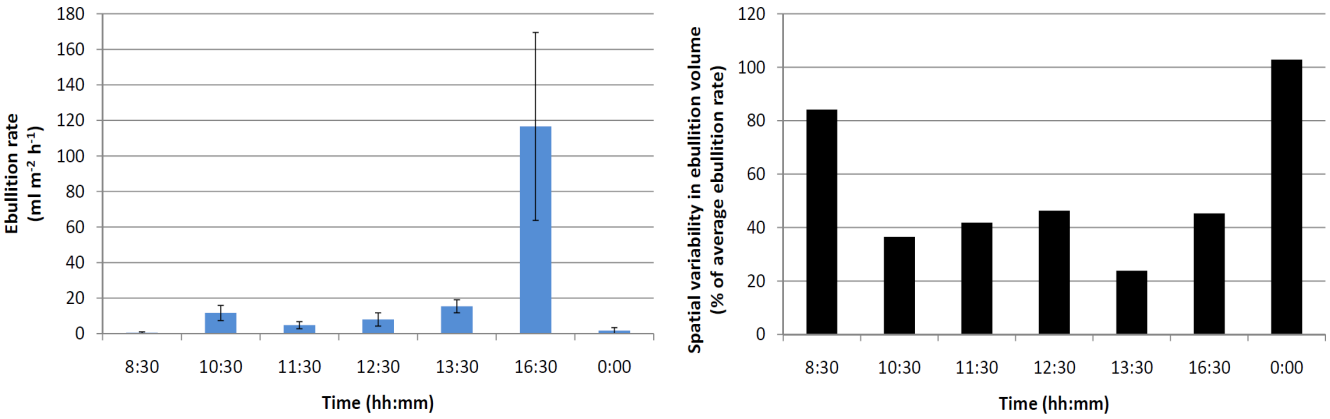

Figure 1. Results from the field bubble ebullition measurements at St. Joseph Bay. Left: The ebullition rate increased throughout the day from near zero values in the morning to maximum values in the late afternoon. Right: The spatial variability of the ebullition remained relatively constant. The high variability recorded at 8:30 and 0:00 were caused by the very low gas volumes collected at those times.

Measurement of the effect of gas release for nutrient release from sandy sediments measured in St. Joe Bay sediments using benthic chamber (two field trips: 3/2010 and 5/2010)
In March, we measured an increase of pore water exchange in the chamber with ebullition (one ebullition chamber failed), while in the reference chambers no such increase was observed (Fig. 2). Phosphate concentrations were below detection limit in all chambers. In May, pore water exchange in the surface layer of the chambers with ebullition was lower than in the chambers without ebullition due to higher pore water flushing caused by the stirring device in these chambers. Despite higher bromide fluxes in the chambers without ebullition, phosphate concentrations in these chambers tended to increase, while in the chambers without ebullition phosphate tended to remain constant or decrease.

This suggests that bubbles may transport phosphate from deeper sand layers to the surface. However, these trends were not significant and further experiments need to confirm this hypothesis.

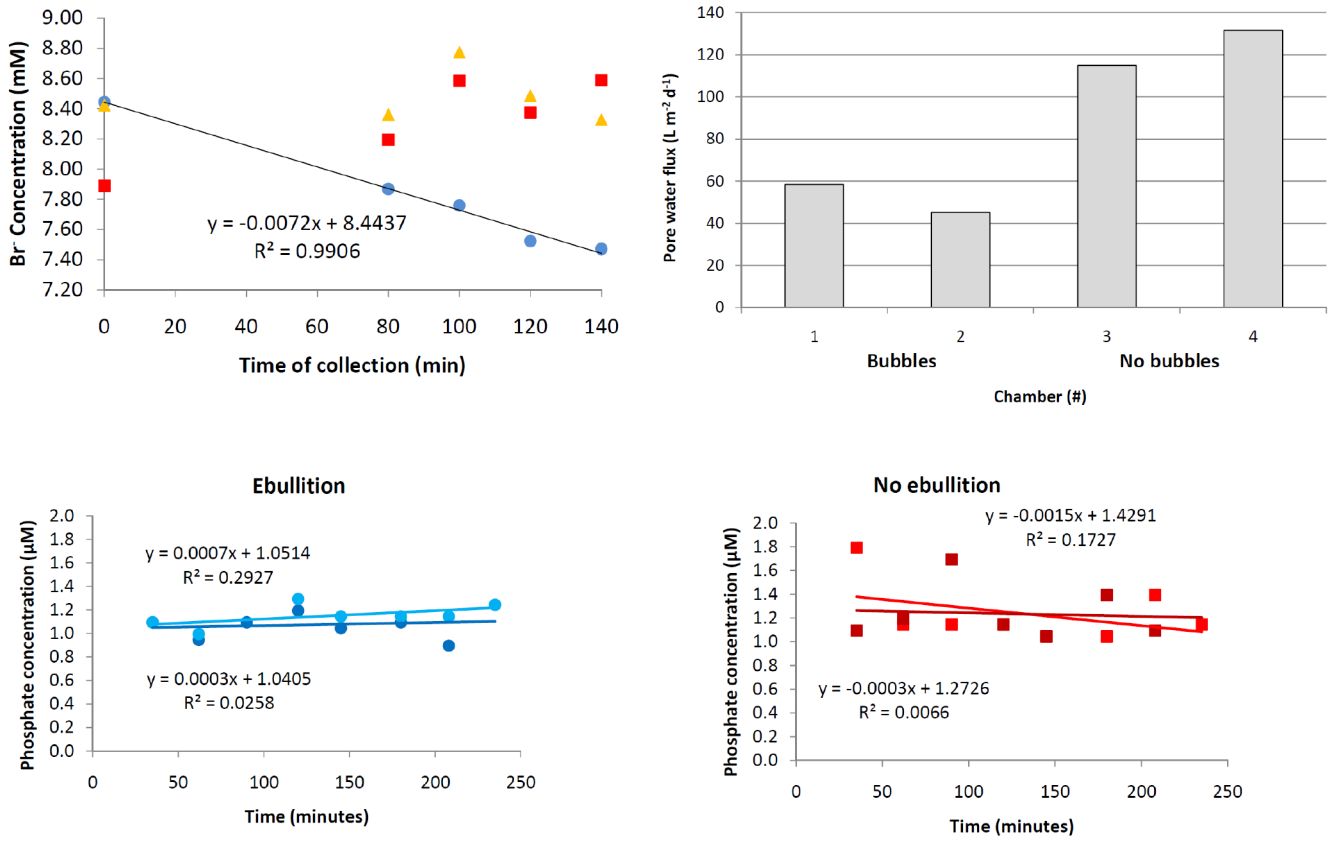

Figure 2. Results from the chamber ebullition experiment at St. Joseph Bay. Upper Left: Changes of bromide tracer concentrations in chambers with (blue circles) and without ebullition (orange and red symbols) over time in March 2010. Ebullition increased pore water exchange relative to the chambers without ebullition. Phosphate concentrations were below detection limit. Upper Right: Bromide flux in chambers with and without ebullition. Fluxes where higher in chambers without ebullition due to flow-enhanced exchange. Lower Left: Phosphate concentration changes in chambers with ebullition seemed to increase but the trend was insignificant. Lower Right: Phosphate concentrations in chambers without ebullition. Phosphate seemed to stay constant or to decrease slightly but the trends were not significant.

Assessing the detection limit for small bubbles (50-200 µL) for the EPOCH XT-based new acoustic method.

Our new method can detect single gas bubbles as small as 1 mm in diameter embedded within the upper 6 cm of saturated sand. The recorded pulse-echo changed shape in the region where bubbles were injected into the sand. Differences between sound records from sediment with a gas inclusion and without gas were distinguishable after an average gas injection of 143 µL (SD = 68 µL), corresponding to a spherical bubble of 1.0 mm diameter (Fig. 3). The echo waveform changed as the gas cavity grew, because the shape of the bubble changed. Gas bubbles were equally detectable at 16,

27, 40, or 56 mm sand depth. Bubbles embedded deeper within the sediment can be detected by lowering two small transducers into the sediment and operating the transducers in through-transmission mode, or similarly by lowering one transducer operating in pulse-echo mode and one reflection plate horizontally into the sediment.

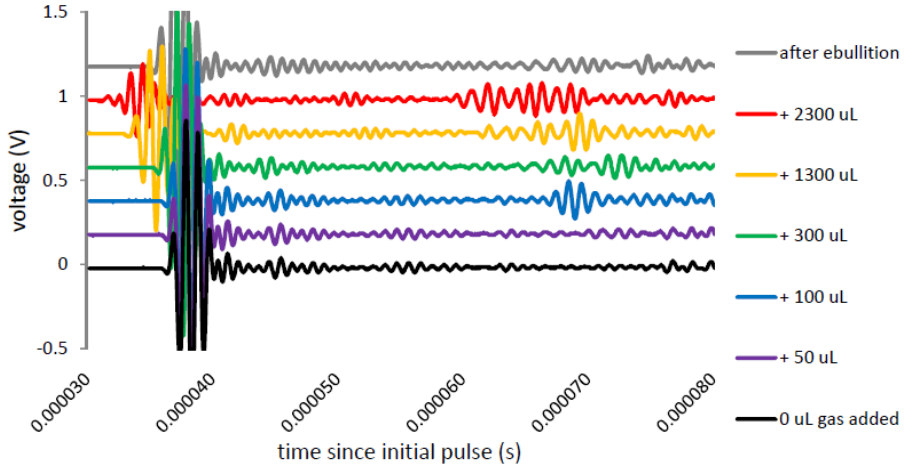

Figure 3. Pulse-echo recordings of gas bubbles of different volumes injected at 27 mm sediment depth. Waveforms are staggered vertically to allow for visual clarity. Sets of peaks at 35-40 µs and 60-75 µs represent the SWI and a gas cavity in the sand, respectively.

Acoustic detection of small bubbles produced by benthic photosynthesis

Laboratory tests demonstrate that our acoustic method can be used for the non-invasive detection of gas bubbles generated by photosynthesis in the upper layers of marine sands. The amplitude of the reflection grew as much as 5.6, 6.0, and 7.4 dB over time up to 4.5 hours into the period with illumination. Two layers of bubbles were identified, one between 1 - 3 mm, the second between 4 and 5 mm sand depth (Fig. 4). When the EPOCH XT was operated in through transmission mode embedded bubbles caused decreased in amplitude in the zone were bubbles were present and peaks moved slightly earlier in successive waveforms. Since embedded bubbles reflect some of the sound pulse, the amplitude of the sound waves reaching a receiving transducer are diminished.

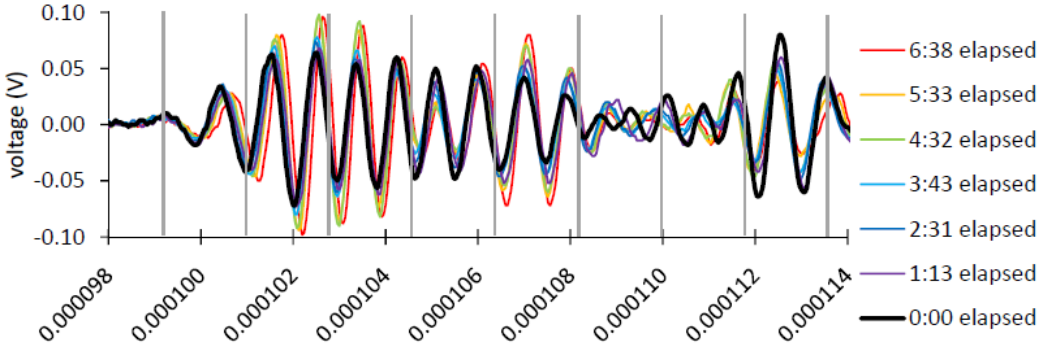

Figure 4. Pulse-echo recordings of sand sediment exposed to light starting at 0:00 h. Data were collected with the A303S transducer in through transmission mode. Gray vertical gridlines are spaced 1.8 µs apart to represent the best estimate of the time needed for sound to travel 1 mm. Waveform amplitudes changed due to bubble formation at 1-3mm and 4-5 mm depth.

Field measurements comparing acoustic pulse-echo readings in natural sands with and without bubbles (6/21010).

These measurements in St. Josephs Bay sands demonstrated that our hand-held acoustic method can non-invasively detect gas bubbles embedded in the surface layer of sublittoral sandy sediment. The locations of the center of sound reflection (V_c), showed significant differences between sand areas with embedded bubbles and sand areas without bubbles (Fig. 5). The strength of the pulse-echo reflection signal (V_s) supported the findings for V_c, and showed differences between sand with and without bubbles. The differences were significant ($p<0.05$, ANOVA). These measurements showed that the hand-held acoustic method can detect bubbles in marine sands with natural sediment heterogeneities and topography. Embedded bubbles reached 0.5 to 1 mm in diameter revealing that the acoustic method is capable in detecting small gas bubbles that typically cannot be seen on standard echo records of sediments working at in the kHz range.

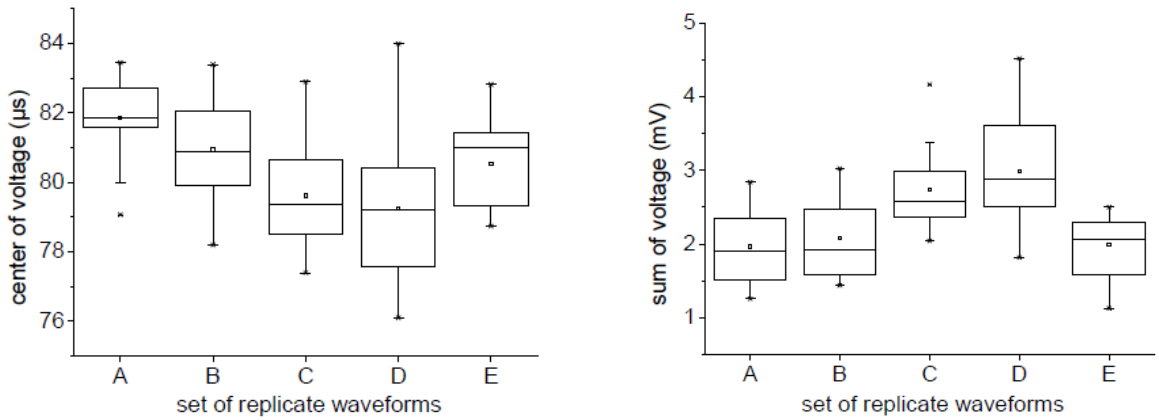

Figure 5. Results from the acoustic field measurements with the EPOCH XT detector. Left: Center of the pulse-echo signal (V_c expressed in µs travel time, 1 µs corresponds to 1.8 mm distance). Occurrence of bubbles shifted the center of reflection 3-5 mm into the sand bed. Right: Strength of the pulse-echo signal (V_s, expressed in mV). Bubbles increased reflection signal strength by approximately factor 1.5. Sets C and D represent sand that contained bubbles that formed during the midday sun, sets A, B and E represent sand without bubbles collected during periods with low or no light.

Investigation of the sand volume and sand surface area affected by ebullition from a single source. The pore water flows caused by gas bubbles moving towards the sediment surface and out of the sediment produce a three-dimensional pore water flow field including horizontal flow components that extend the influence of the single bubble source to a large area and sand volume (Fig. 6). In this test series, the bubble source located at 2.4 cm sediment depth affected an area of 154 cm^2 (14 cm diameter) at the sediment surface and a funnel shaped sediment volume of 205 cm^3 volume that extended below the bubble infusion depth. This demonstrates that a few spaced bubble sources can affect the entire sediment surface and cause deep pore water circulation in sand beds.

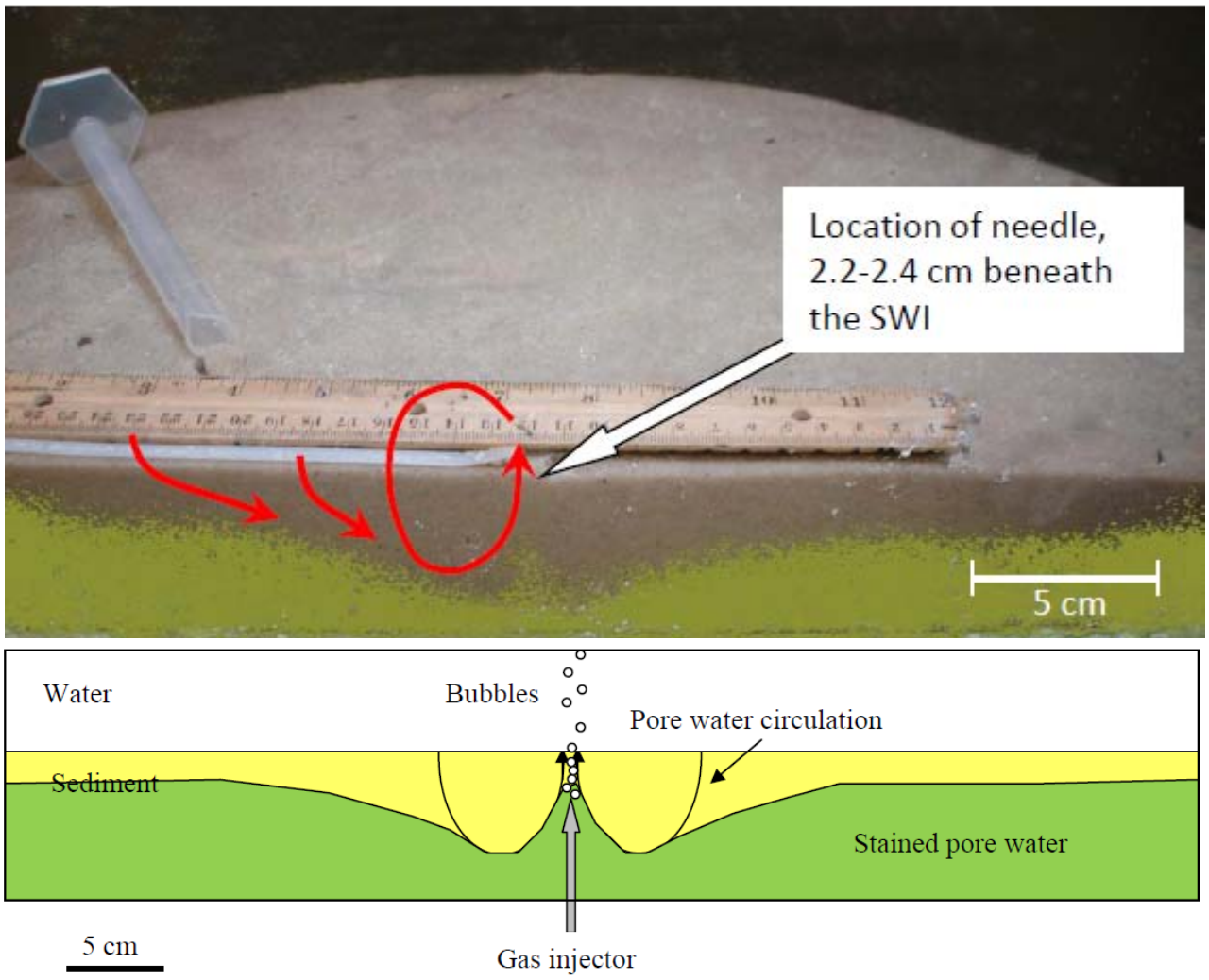

Figure 6. Bubbles released at 2.4 cm sediment depth cause a pore water circulation pattern that reaches deeper than the bubble injection point, affecting a funnel-shaped sediment volume and a relatively large sediment surface area.

Generation of controlled pressure oscillations in a laboratory column for the investigation of the effect of sedimentary bubbles of solute transport.

We build an instrument that produces pressure oscillations as caused by passing surface gravity waves that compress sedimentary gas bubbles thereby causing pore water transport (Fig. 7). The set up uses programmable linear motor acting on the diaphragm of a pressure transducer that transfers defined pressure oscillations to a laboratory pressure tank. A pressure sensor in the column records the resulting pressure oscillations, providing feedback to the pressure generator and real time data. With a particle image velocimetry system we could quantify the flow field caused by bubbles emerging the sediment and the solute transport caused by ebullition.

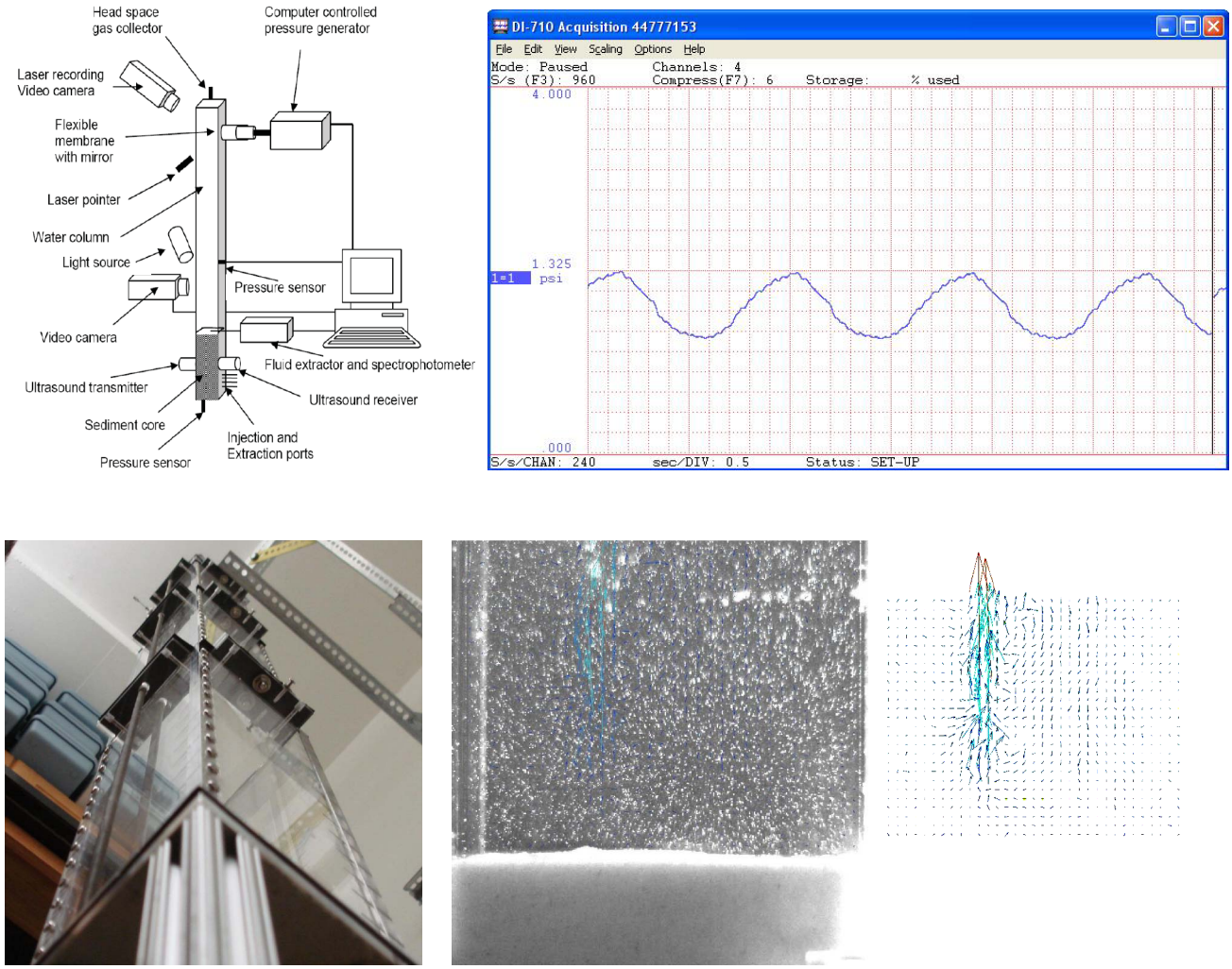

Figure 7. Upper Left: Schematic of the pressure tank and the instruments that are used to produce and quantify pressure oscillations in the device. Upper Right: Sinusoidal pressure oscillations produced by the linear motor and recorded by the pressure sensor in the pressure tank. Lower Left: Image of the pressure tank. The sides of the tank are 10 cm wide, the total height is 2 m. Lower Middle: PIV image with vector field caused by a bubble emerging from the sediment. Lower Right: Vector field extracted from the image on the lower left showing the velocity distribution and associated microturbulence caused by the emerging bubble.

IMPACT/APPLICATIONS

Our measurements in the northern Gulf of Mexico indicate that the shallow sands release approximately 1 L gas bubbles m^{-2} d^{-1}. As these bubbles start as oxygen bubbles, this corresponds to an oxygen flux of 44 mmol m^{-2} d^{-1} that could cover half of the sediments oxygen demand. The ebullition of the small bubbles from the upper, biogeochemically very active sediment layer causes transport of solutes from the sediment to the water as demonstrated using bromide tracer. Laboratory experiments with dye tracer show that the ebullition causes pore water circulation reaching deeper than the depth of bubble formation and affecting a relatively large area. A few evenly-spaced bubble sources thus can affect interfacial fluid exchange of the entire sand surface. With the acoustic instrument system we

developed, single gas bubbles as small as 1 mm (~150 μL) can be detected in sands down to at least 6 cm sediment depth. We show that this allows scanning sediments for embedded oxygen bubbles or following the fate of embedded bubbles non-invasively. To our knowledge, this is the first method that allows such small bubble detection in sediments and also in sands that are a difficult medium from acoustic methods due to their high reflectivity. Our acoustic detection method may have applications in medical sciences and industrial production processes, and permits detailed investigations of bubble dynamics within saturated sands. Our newly developed pressure tank system with computer-controlled pressure oscillations allows laboratory testing of the effects of pressure changes on bubbles embedded in the sediment. We show with Particle Image Velocimetry the influence of bubbles released from the sand on the turbulence and transport vectors in the diffusive boundary layer (DBL) that controls transport across the sediment water interface. Each bubble released from the sediment carries pore water across the DBL interface and thereby enhances interfacial flux. The hypoxic zones developing in many coastal areas show dramatically the increase of sedimentary gas production, when organic matter processing is completed entirely by microbial processes. Gas bubble research thus will gain momentum and our research support these future investigations.

TRANSITIONS

The project results on benthic oxygen production and bubble transport are relevant for benthic ecologist and oceanographers who seek a better understanding of the cycles of carbon and nutrients. The highly sensitive bubble detection method may be applicable in medical sciences and industrial production processes where bubbles can cause problems (e.g. paint application on cars). When operated by an underwater vehicle, this detection method may also be useful in detecting reflecting objects buried in the sand.

RELATED PROJECTS

NSF project "Further Development of the Eddy Correlation Technique; NSF-OCE-0536431; 01/01/06-12/31/10; $742,717; P. Berg, (PI), M. Huettel (co-PI). This ongoing project was awarded as a follow-up of our first eddy correlation project (OCE-0221159) and supported continuation of our research and instrument development related to the eddy correlation technique for the measurement of oxygen flux in permeable coastal sediment. In addition to intensive field campaigns at the two main field sites, the Virginia Coast Reserve LTER site and the Apalachicola NERR site, we have visited several other sites that appeared particularly challenging with respect to flux measurements. These include the rocky bottoms in The Great Lakes with dense colonies of the invasive Quagga mussel overgrown with filamentous algae and the deep ocean floor off the coast of Japan. Data from these sites have provided both new insights on benthic oxygen metabolism and a demonstration of the advantages of the eddy correlation technique in diverse environments.

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