Transport of Gas and Solutes in Permeable Estuarine Sediments

Markus Huettel
Department of Earth, Ocean and Atmospheric Science
Florida State University
117 N Woodward Ave.
Tallahassee, Florida 32306-4320, USA
phone: (850) 645-1394 fax: (850) 644-2581 email: mhuettel@fsu.edu

Award Number: N00014-08-1-0360
http://myweb.fsu.edu/mhuettel/

LONG-TERM GOALS

The long-term goals are 1) to assess the role of small gas bubbles in shallow sandy coastal sediment for sediment geotechnical properties, the transport of solutes across the sediment-water interface and the biogeochemical processes in the sediment surface layer 2) to develop and apply an acoustic technique for the detection and quantification of small gas bubbles in sandy sediment at a high spatial resolution.

OBJECTIVES

1) Quantification of small gas bubbles in the surface layer of coastal sand sediment and assessment of their temporal and spatial distribution.
2) Quantification of the volumes and composition of the gas bubbles in the sediment and the overlying water and the changes of volume and composition over time.
3) Measure the dispersion and transport of solutes caused by bubble volume change and migration under different pressure conditions.
4) Develop and apply an acoustic technique with high spatial and temporal resolution for the detection and quantification of embedded gas bubbles

APPROACH

The project combines instrument development with laboratory and field measurements.

- We developed a **hand-held ultrasound device for the detection of small gas bubbles** embedded in sandy sediment. This device was tested in the laboratory and in the field, and its functionality is demonstrated by measuring the spatial and temporal distribution of small bubbles produced by photosynthesis in sublittoral sands.
- We developed an **instrument for the application of realistic pressure oscillations to incubated sediment cores** with gas enclosures. This instrument is used to quantify the effect of buried bubbles on sediment properties and interfacial solute exchange.
- We measured of **gas release volumes** from sandy sediments using benthic chambers and bubble traps. While the chambers allowed changing the advective transport component and thereby also

gas ebullition, the bubble traps collected bubbles under the natural flow conditions. The composition of the sampled gas volumes is analyzed using a Gas Chromatograph (GC).

- We conducted **gas-stripping experiments** in laboratory column reactors filled with natural sands and in the field using gas injection techniques that test the gas stripping caused by nitrogen and oxygen ebullition.
- **Content, distribution and migration of free gas in the surface layers of the sand** sediment was 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 was investigated in the field using benthic chambers and a laboratory column setup which allowed 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 and results in the previous years we refer to the first three annual reports. Below a summary of the work completed within the reported project year 2012/2013.

WORK COMPLETED

Compilation of thesis and manuscripts

We are at the end of our project and we therefore emphasized data analysis and compilation of findings. Graduate student Chiu Cheng successfully defended and submitted his Master's thesis addressing bubble ebullition in permeable marine sands. Two publications were prepared, one was submitted the other manuscript will be submitted before end of 2013. Graduate student Lee Russell presently is writing his Ph.D. dissertation based on this ONR reserach.

Evaluation of ebullition caused by sedimentary photosynthesis and methanogenesis

For these experiments photosynthetic gas bubbles released from the sediment surface were quantified using six 500-mm diameter cone-shaped flexible, transparent bubble traps, anchored 100 mm above the sediment surface. Gas accumulating at the apex of the bubble trap were collected with a gastight glass syringe. Each of the daily set of 4 time points of collected gas from both sediment and gas trap samples were added to obtain the total gas volume per day. The 3 individually calculated daily volumes were then averaged, and normalized for area to obtain an average rate expressed in ml m^{-2} d^{-1}. More details of these experiments is available in our previous reports. During this project phase, we analyzed and compiled the results of these experiments.

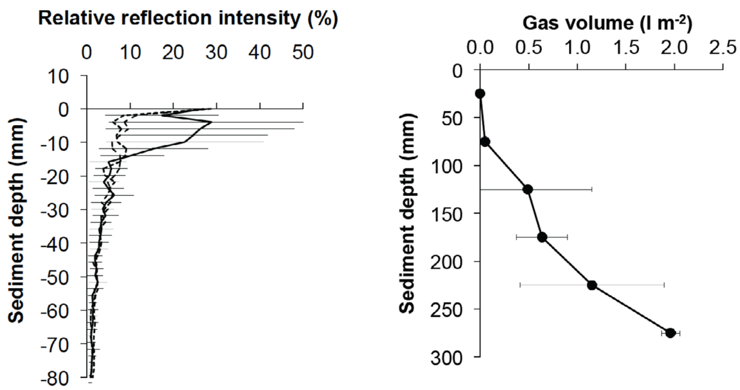

Fig. 1. Left: Distribution of oxygen bubbles in St. Joseph Bay sediment, Florida. Dotted lines present distribution under dark conditions. Right: Methane profiles in sediment affected by groundwater seepage. Free methane gas builds up below 10 cm depth.

The average gas production rate recorded at St Joes Bay in August was 285 ± 92 ml m^{-2} d^{-1} , which included 225 ± 90 ml m^{-2} d^{-1} recovered from gas traps and 60 ± 17 ml m^{-2} d^{-1} extracted from sediment cores. During peak gas production period, the gas flux reached 270 ± 26.7 ml m^{-2} d^{-1} in the traps. At a measured average bubble diameter of 1 ± 0.3 mm, the volumes accumulating in our gas traps suggested that up to 516000 bubbles m^{-2} d^{-1} were released by ebullition in August 2011. Gas extracted from sediment cores on average contained increased oxygen (13-63% O_2, average 44 ± 21.5%), and less nitrogen than air (37-87% N_2, (average 52 ± 21.6%). Gas trap samples contained less oxygen (18-57% O_2, average 33 ± 20.7%) and more nitrogen (43-82% N_2, average 59 ± 14.6%) than the sedimentary gas. No other gases were detected at amounts ≥ 0.5% in the samples. The average CH_4 gas ebullition measured with the seepage meter was 226.7 ± 69.5 ml m^{-2} d^{-1}. The extraction of the 24 sediment cores revealed an average gas content of 6.0 ± 5.5 ml per dm^{-3} sediment (Fig. 1).

Evaluation of gas retention capacity

Free gas generated in the sediment does not immediately rise to the surface but has to accumulate to a bubble that is large enough to overcome the pressure and interlocking of the sand grains enclosing the bubble. The sand thus has a gas retention capacity that was determined by quantitatively injecting air with a steel capillary at selected depths (3, 5, 10, 15, 20, 25 mm) of sand sediment until ebullition occurred. Average diameters of photosynthetic bubbles released from the sediment were measured using underwater photographs of the rising bubbles and subsequent image analysis using the Image J software. Gas samples were analyzed with a gas chromatograph with thermal conductivity detector and a molsieve 5A column. More details of these experiments is available in our previous reports. During this project phase, we analyzed and compiled the results of these experiments.

Bubbles injected at a gas flow rate of 1.23 ± 0.07 ml min^{-1} at 3 mm sediment depth could not grow larger than ~3 mm in diameter (0.02 ml) before ebullition, injection at ~25 mm depth limited bubble size to 10 mm diameter (4.2 ml), and at 200 mm sediment depth bubbles had to reach 17 mm diameter (20 ml) before ebullition. As a consequence of the depth-related bubble size limitation, the frequency of ebullition at a constant gas injection rate decreased with increasing gas injection depth. The experiment showed that ebullition from a shallow single point at a relatively low gas flow rate of 1.2 ml min^{-1} affected a sediment surface area of 380 cm^2 and pore water circulation in a sediment volume of 1330 cm^3.

Evaluation of the influence of ebullition depth on interfacial solute flux

Sedimentary CH_4 gas was collected with a seepage meter as well as with gas extraction from sand sediment. Methane bubbles contained in sandy sediment were quantified by extracting gas from large sediment cores (~ 300 mm height, 190 mm diameter) that were sampled using an acrylic core liner. This core liner (400 mm height, 190 mm inner diameter) was pushed ca. 300 mm into the sediment at 24 random locations in the sublittoral zone (1.5 to 1.8 m depth). The two openings of the core liner were closed with gastight lids (a hole had to be excavated next to the core liner to apply the lid to the lower opening), then the sediment core was removed from the seabed.

After measuring the height of the retrieved core, the sediment was gently mixed by putting the core liner into a horizontal position and rotating it slowly, allowing burried gas bubbles to escape the sand. Gas released from the sand was collected through a port in the core liner lid with a gastight syringe and subsequently quantified volumetrically in an inverted water-filled graduated cylinder. Gas-retention potential at greater sediment depths was measured by quantitatively injecting gas into sandy sediment at 100, 200 or 300 mm depth until bubbles surfaced (Fig. 3). As a consequence of the depth-related bubble size limitation, the frequency of ebullition at a constant gas injection rate decreased with increasing gas injection depth, and the lag time between initiation of gas injection and ebullition reached up to 20 min when gas was injected at 30 mm sediment depth. Ebullition of larger bubbles released more pore water per bubble, but the small bubbles were more effective in releasing pore water, e.g. for the 3 mm bubbles, the volume of pore water released corresponded to 7% of the bubble volume, while for the 25 mm bubbles, the release volume corresponded only to 2% of the bubble volume. Interfacial flux was at least 4 times higher in sand cores with ebullition compared to the control cores with diffusive-only transport (t-test, $p<0.05$, n = 13).

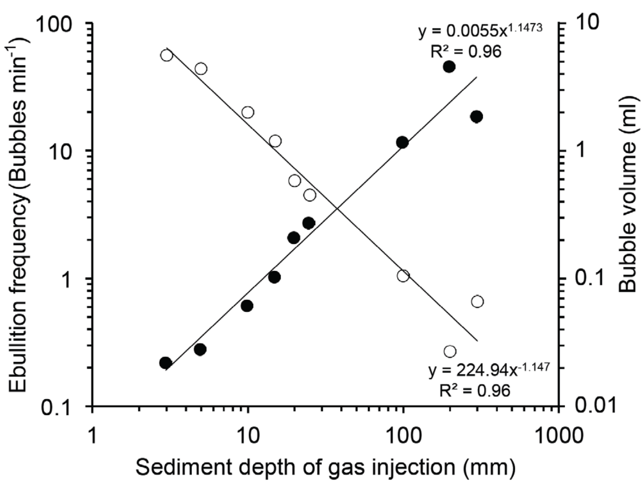

Fig. 2. The frequency (open circles) and volume (solid circles) of bubbles released after injection of gas at 1.24 ml min^{-1} at different sediment of nearshore marine sand.

Evaluation of mechanisms of sediment-water solute exchange by ebullition

In these experiments, gas was injected in the center of sand cores with stained pore water and the dye release caused by ebullition was followed in the water column and by disecting the sediment. More details of these experiments are available in our earlier reports. In addition, we investigated within the last year the effects of ebullition on circulation of water above the sediment using particle image velocimetry (PIV) in a transparent container (185 x 295 x 170 mm) filled with a 20 mm layer of black sand (median grain size 0.506 µm) and 100 mm of overlying water. Air was injected with a syringe at 2 mm sediment depth through a thin tube mounted to the bottom of the sediment core, allowing controlled release of bubbles ~2 mm diameter. From the two-dimensional velocity data, contour maps were created using Surfer 11 software. The volume of overlying water that was affected was calculated by measuring the 2-D area of each flow field and converting it to volume, assuming axial symmetry. For a given bubble size - in our PIV experiments 2 mm diameter - , the frequency of ebullition determined the velocity of this vertical fluid transport and the shape of the water volume that was affected by the bubble stream. After ~1 second, the maximum average flow velocities caused by this ebullition reached 80 mm s^{-1} at 50 mm above the sand and continuing ebullition did not increase this velocity significantly. Using a conservative flow velocity of 2 mm s^{-1} and a diameter of the upward directed flow of 10 mm, the ebullition pumped approximately 10 ml min^{-1} from the laminar sublayer into the turbulent section of the boundary layer. Experiments with slow (2.2 bubbles s^{-1}) and rapid (11 bubbles s^{-1}) ebullition showed that the relationship between vertical distance above the sediment surface and the width of the resulting turbulent cone (delineated by flow < 2 mm s^{-1}) for both release rates extended to approximately 60 mm above the sediment surface before this linear relationship ended (Fig. 3). The volume of water affected by the ebullition increased with increasing bubble flow. The increase in vertical flow velocity with distance from the sediment surface was proportional to the ebullition rate. Likewise the total volume of water affected by the ebullition increased with increasing velocity.

Evaluation of interfacial solute flux caused by ebullition
In these experiments, in-situ tracer experiments were conducted, in which nitrogen gas was released at approximately 80 mm depth in sand sediment enclosed in benthic chambers. The interfacial fluxes were calculated from the Br^- tracer concentration changes in the enclosed water column over time. More details of these experiments is available in our previous reports. During this project phase, we further analyzed and compiled the results of these experiments. In the 5 stirred chambers with ebullition, the pore water flux, calculated from the Br^- decrease, averaged 9.6 ± 4.0 L m^{-2} h^{-1}, while in the 4 controls, flux averaged 3.3 ± 1.7 L m^{-2} h^{-1}. This difference was statistically significant (t-test, $p<0.05$, n = 9). At a constant gas release rate, interfacial fluid flux was highest when bubbles were released from the sediment surface layer (3 mm depth). The largest decrease in flux was recorded when gas injection was increased from 5 to 15 mm sediment depth. Decrease in bubble frequency with increasing depth, relative to the highest frequency (at 3 mm release depth) ranged from a factor of 1.3 (at 5 mm release depth) to 15 (25 mm release depth), while the decrease in pore water flux per bubble relative to the smallest bubble volume (3 mm release depth) ranged only from 1.3 to 2.9-fold (Fig. 4).

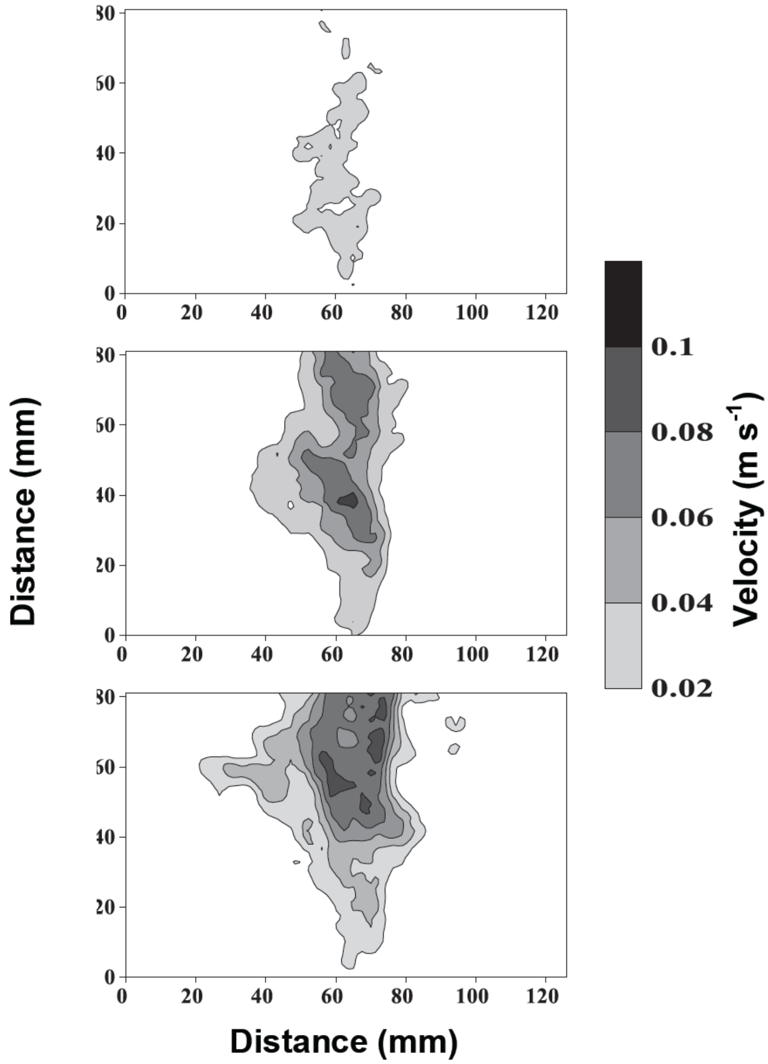

Fig. 3. Increase of the fluid velocity and turbulence caused by ebullition. PIV measurements were done at 0.5 s time intervals. The fluid velocity and turbulence increased rapidly over time, expanding the funnel-shaped zone that was affected by the ebullition.

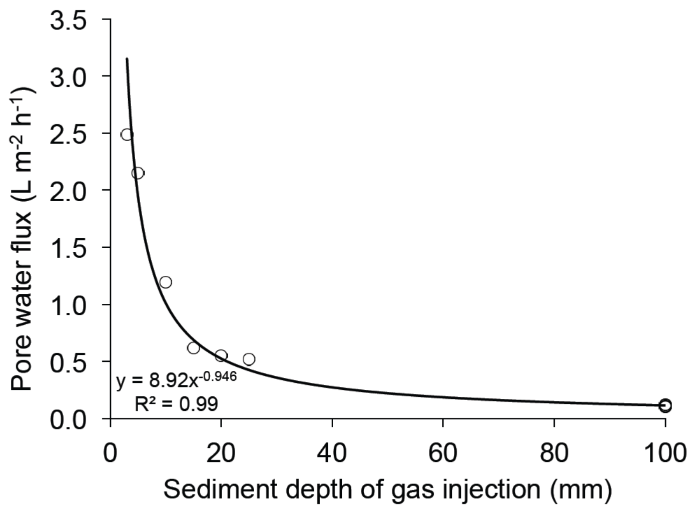

Fig. 4. Pore water flux resulting from gas injection at different sediment depth at a constant rate of 1.2 ml min^{-1}. At shallow injection depth, the ebullition was more effective for increasing interfacial solute flux compared to the deeper injection.

IMPACT/APPLICATIONS

Our study addressed processes in the shallow part of the nearshore zone (< 2 m water depth), which according to the National Geophysical data center (ETOPO data set, http://www.ngdc.noaa.gov) includes about 780,000 km^2, or 3%, of the global shelf. Approximately one third of the global shelf, the region shallower than 30 m water depth, receives sufficient light for benthic primary production, and O_2 bubbles produced by aquatic plant photosynthesis has been reported from water as deep as 25 m (Gattuso et al. 2006; Hermand 2004; Hermand et al. 1998). Thus, up to 25% of the global shelf area could be affected by O_2 bubble production. However, in many shelf areas the depth, sufficient light reaching the sea floor will likely be less than 25 m due to water turbidity. The water in our protected St Josepths Bay study area is usually relatively clear, and with a total area of 178 km^2, about 47% (83.5 km^2) of SJB has a water depth of ≤ 6 m where permeable sandy sediments dominate. If ebullition occurred entirely at ≤ 6 m depth, we calculate based on our measurements, an estimated sedimentary gas formation volume in August of 24,000 ± 7,600 m^3 d^{-1}, from which ebullition removed 19,000 ± 7,500 m^3 d^{-1} (79%). Using the measured pore water flux per bubble values, we can estimate the pore water release volume for SJB in areas ≤ 6 m water depth. Based on the measured diameter of the released bubbles (1 to 2 mm), the volume of pore water flux caused by ebullition can reach 2,900 to 8,600 m^3 d^{-1} in SJB under sunny conditions during our August measuring campaign. Bubble sizes in our laboratory PIV experiments were similar to those observed at our study sites. Assuming that small bubble ebullition from one point mixes a water column of ~40 mm diameter as indicated in our experiments, about 130,000 ebullition points could result in a complete mixing of the water near the sediment surface assuming continuous ebullition. At our SJB study site, up to 430,000 bubbles m^{-2} d^{-1} were released, suggesting that ebullition of gas bubbles contributed to mixing of released pore water into the water column. At high benthic primary production rates, O_2 is released from the sediment as gas bubbles and is lost for sedimentary oxidation reactions. The loss of O_2 through ebullition changes the distribution and availability of this preferred electron acceptor and can thus have an important role in the biogeochemistry and metabolic processes in permeable sediments. On the other hand, the bubbles rising through the water column can provide O_2 to the water column, which may be a factor during periods of O_2-consuming decaying phytoplankton blooms in the water column. Similar to the O_2 bubbles, the CH_4 bubbles that ascended through the sediment and the water column can influence biogeochemical process in sand and water. CH_4 is a substrate of CH_4 oxidizing bacteria, and CH4 bubbles rising through sediment layers containing sulfate or O2 can fuel CH4 oxidation and microbial growth. Likewise, CH4-containing bubbles ascending through the water column can support methanotrophs, as was shown after the Deepwater Horizon accident where only small amounts of the massive volume of CH4 that was released from the well head in the deep sea reached the surface because it was apparently consumed by water column bacteria (Reddy et al. 2012). Although the observed vertical fluid transport driven by the fewer CH_4 bubbles may have been less compared to the transport by the numerous O_2 bubbles at SJB, the larger CH_4 bubbles can transport pore water and associated solutes from deeper sediment layers to the surface. CH_4 ebullition thus establishes a pathway for substances including ammonium, ferrous iron, dissolved phosphate and sulfide into and through the oxic upper layers of coastal sands, where these solutes may provide important nutrients or reduced substrates for microbial oxidation reactions.

TRANSITIONS

The results of our studies show that the small bubbles generated by photosynthesis in the surface layer of shallow sediments have broad implications for sediment geotechnical, geochemical and biological properties. Through changing sediment structure and pore water circulation, the bubbles influence the sediment erosion threshold, biogeochemical zonations, living space for organisms and thereby the role of the sediments in the cycles of matter. The results are relevant to sedimentologists, benthic ecologist and oceanographers who seek a better understanding of the ecological functioning of the shallow nearshor environment and the possible implications of sea level rise and global warming. Our results on gas stripping and gas exchange introduce new insights in a process that so far has not been included in our modeling of coastal cycles of elements. These results thus can open a new field of research addressing the role of bubble ebullition for benthic-pelagic coupling and element cycling. Our highly sensitive bubble detection method may be applicable in medical sciences and industrial production processes where bubbles can cause problems. 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 "Collaborative Research: Eddy Correlation and chamber measurements of benthic oxygen fluxes in permeable sediments," $343,385 (Huettel funds), P. Berg, (PI), M. Huettel (co-PI). This project was awarded as a follow-up of our second eddy correlation project (OCE-536431) 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. Within this project, we deployed the eddy flux instrument at our ONR project field sites in order to produce data on oxygen flux that can be compared with the fluxes produced by bubble ebullition. 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. Within the GRI-funded project: “The Deep-C Consortium: Deep Sea to Coast Connectivity in the Eastern Gulf of Mexico” $203,471 (Huettel funds), E. Chassignet (PI), M. Huettel (one of several Co-PIs), we investigate the effect of the input of old (i.e. crude oil) and new (i.e. phytoplankton detritus) organic matter on sediment properties and biogeochemical reactions. As crude oil from the recent oil spill in the Gulf and phytoplankton affect the sediment in the shallow nearshore zone, this project is closely linked to our research within the ONR project. Specifically, hydrocarbons alter sediment cohesiveness and permeability, thereby altering the effect of the oxygen bubbles on sediment water exchange. Bubble ebullition is restricted while crude oil pollution can limit benthic primary production through toxic components but at lower concentration also enhance the production by increasing microbial growth and associated mobilization of nutrients.

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PUBLICATIONS

Wildman, R. A. J., and Huettel, M. (2012) Acoustic detection of gas bubbles in saturated sands at high spatial and temporal resolution, *Limnology and Oceanography: Methods* 10, 2012, 129–141.

Cheng, C., Huettel, M. and Wildman, R.A. (submitted). Ebullition-enhanced solute transport in coarse-grained sediments