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Physical Controls on Methane Ebullition from Reservoirs and Lakes

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ABSTRACT

Understanding the nature and extent of methane production and flux in aquatic sediments has important geochemical, geotechnical, and global climate change implications. Quantifying these processes is difficult, because much of the methane flux in shallow sediments occurs via ebullition (bubbling). Direct observation of bubble formation is not possible, and bubbling is episodic and dependent upon a number of factors. Whereas previous studies have correlated methane flux with surface wind intensity, detailed study of Lake Gatun in Panama and Lago Loiza in Puerto Rico suggest that methane flux is more closely correlated with the shear stress in sediments caused by bottom currents. Bottom currents in turn are a complex function of wind, internal pressure gradients, and lake bathymetry. A simple physical model of bottom currents and sediments in these lakes suggests that most methane ebullition originated from the upper 10–20 cm of the sediment column. Our data reaffirm previous studies showing that ebullitive methane flux is minor in water deeper than ~5 m.

INTRODUCTION

Methane forms in sediments when the decomposition of organic matter exhausts all other available oxidants (dissolved oxygen, nitrate, and sulfate). When methane concentrations become sufficiently high, a separate gas phase forms that eventually coalesces into bubbles below the sediment–water boundary. Methane is particularly common in terrestrial aquatic environments, where sulfate concentrations are generally small.

The ebullitive (bubbling) form of methane plays an important role in both the geomechanical properties and the geochemical behavior of fine-grained, organic-rich sediments. Large concentrations of methane gas have the potential to cause blowouts during drilling operations as well as decreasing the geotechnical stability of engineered structures on aquatic substrates (Wheeler, 1990; Sills et al., 1991; and Silva and Brandes, 1998). Methane bubbles are well documented as acoustic turbidity that inhibits interpretation of seismic records (Judd and Hovland, 1992; Anderson et al., 1998). In terrestrial environments, methane bubbles can effectively scavenge volatile organic compounds from the sediment and transmit them into the atmosphere (Vroblesky and Lorah, 1991) as well as playing an important role in the hydraulic conductivity of aquifers (Ronen et al., 1989). In all of these examples, the rates of formation and triggering mechanisms of methane flux remain poorly understood.

The concentration of atmospheric methane, a greenhouse gas, has more than doubled over the past 200 years (Blake and Rowland, 1988). Methane constitutes approximately 22 percent of total greenhouse forcing (Lelieveld et al., 1998). An estimated 25 to 50 percent of this methane is derived from wetland environments in tropical and subtropical regions (Matthews and Fung, 1987; Anselmann and Crutzen, 1989; and Reeburgh et al., 1993). Because methane has the potential to contribute significantly to global warming, understanding causes for this increase is important. Two environments that may be providing additional methane flux in the tropics are rice paddies and artificial lakes (Schlesinger, 1991).

Bubbling, also termed ebullition, is the primary release mechanism from lakes and other shallow water environments. Quantifying methane generation and flux to the atmosphere from lakes via ebullition is difficult, because bubbling is episodic and depends on many factors (Keller and Stallard, 1994). Methane generation is strongly temperature dependent, and temperate-latitude environments show much greater seasonal variation in methane flux than tropical environments (e.g., Walter and Heimann, 2000). In natural and man-made wetlands, methane flux can also be strongly dependent on depth of the water table (Bogner et al., 2000).

In an effort to quantify methane flux in shallow, tropical aquatic environments with minimal seasonal temperature change—Lago Loiza in east central Puerto Rico and Gatun Lake in central Panama—were studied in detail during the summers of 1994 and 1995 (Joyce, 1996). These studies focused on the relationship of bottom shear stress and winds to methane bubbling. The goal was to gain a better understanding of why bubbling is episodic, the physical factors controlling its rate of occurrence, and the temporal and spatial constraints of ebullition. Improved understanding of the specific factors that trigger bubbling may lead to a better understanding of the mechanical and acoustic behavior of shallow sediments as well as the contribution of tropical reservoirs to the global methane budget, and therefore to possible causes of global warming.

Methane escapes aquatic environments via three primary mechanisms: diffusion, ebullition (bubbling), and advection through rooted water plants. Ebullition produces the largest emissions and dominates flux in environments where methane production rates are high and water is shallow (Rudd and Hamilton, 1978; Crill et al., 1988; Devol et al., 1988; Bartlett et al., 1990; and Keller and Stallard, 1994). Diffusion is predominant in deeper, colder, less productive settings (Jannasch, 1975; Strayer and Tiedje, 1978; Molongoski and Klug, 1980; and Kelly et al., 1994). Methane gas in the form of bubbles is more likely to escape oxidation, because bubble transport is orders of magnitude more rapid than diffusive transport. Thus, shallow environments, which are often ebullitive, usually emit more methane than deeper environments. The third mechanism of methane emission, uptake and advective transport through plants, may also transport significant quantities of methane in places where rooted plants, including rice and water lilies, are present (Dacey and Klug, 1980; Cicerone and Shetter, 1981).

Current-induced bottom shear stress and reduced hydrostatic pressure are thought to trigger increased gas release from lake environments. The effect of wind-induced surface turbulence on diffusive gas flux has long been known to enhance air–water gas exchange, thus facilitating gas release from the water surface (Kanwisher, 1963; Sebacher et al., 1983; and Wanninkhof et al., 1985). However, until studies by Miller and Oremland (1988) and Keller and Stallard (1994), previous work had not associated increased wind velocities with increased ebullition. Keller and Stallard (1994) have suggested that the strong correlation observed between wind velocities and bubbling episodes in Gatun Lake, Panama, may be the result of wind-induced benthic shear stress on lake-bottom sediments. The relationship between benthic shear stress and gas flux, however, has not yet been quantified.

Winds impose a frictional stress on the surface of the water, and energy is propagated through the water column in the form of currents and waves. Currents, if strong enough, can apply a significant shear stress to the sediments, triggering the release of bubbles that are unable to escape by their buoyancy alone. It is therefore reasonable to expect that current velocities would show a better correlation with bubbling events than wind velocities, because they would record not only wind-induced currents but also disturbances that are a result of other influences such as seiches, internal waves and pressure gradients, and man-made disturbances such as barges and boats. It should be noted, however, that high current velocities that act consistently over long periods of time would cease to generate ebullition once the bubble supply is depleted.

Site Descriptions

Lago Loiza in Puerto Rico and Gatun Lake in Panama were chosen as sites for the gas measurement and bottom-current experiments. Both lakes are easily accessible and have been studied in detail because of their great local importance: Lago Loiza as the primary potable water source for San Juan and Gatun Lake as a portion of the Panama Canal Waterway. Keller and Stallard (1994) studied Gatun Lake extensively from 1986 to 1988. With the aid of these two authors, we returned to Gatun Lake in order to: (1) determine whether gas fluxes had changed in 7 years time; and (2) document the direct relationship between bottom shear and gas flux, rather than wind and gas flux (observed in 1988). From a scientific perspective, the two lakes provide an interesting contrast, because both are man-made reservoirs in tropical regions, yet they have markedly different physical and chemical characteristics (Table 1). Lago Loiza, located in east central Puerto Rico 12 km south of San Juan, was created by damming of the Rio Grande de Loiza in 1953 and provides approximately two thirds of the drinking water for the capital city. Eastern Puerto Rico's climate is characterized by intense rainfall during the wet seasons from August to November and April to June. Significant changes in water level follow the seasonal and annual variations in rainfall. Surface-water elevation ranges from 41 m (134.5 ft) above mean sea level at maximum capacity to 35 m (114 ft) above mean sea level during drought. Study sites were located within a kilometer of the dam, at the northern end of the reservoir at depths of 1 m to 6 m.

Gatun Lake, located in central Panama, is a man-made, freshwater lake, formed by the damming of the Chagres River in 1907 during the construction of the Panama Canal. Water level in the lake is carefully regulated by 14 gates of the Gatun Dam Spillway. An average water elevation of 26 m above sea level is maintained with only a 2 m per year fluctuation (Zaret, 1984). Studies were undertaken in Laboratory Cove, adjacent to the Smithsonian Tropical Research Institute field station at Barro Colorado Island. Water depths at the sampling stations ranged from 1 m to 10 m. Data collection consisted of

Table 1. *General attributes of Lakes Loiza and Gatun.*

	Lake Loiza (Puerto Rico)	Lake Gatun (Panama)
Surface area (km^2)	2.4	431
Drainage basin size (km^2)	538	2,313
Average depth (m)	6.1	12.7
Maximum depth (m)	17.2	29.0
Maximum depth fluctuation (m)	5.1	2.0
Surface temperature (°C)	24–31[1]	27–31[1]
pH	6.5–8.9[1]	7.4–7.9[1]
Average rainfall (cm/y)	160	260
Secchi depth (m)	0.8	3.3–7.0
Average phosphorus concentration (mg/L)	0.33	0.05
Average nitrogen concentration (mg/L)	1.70	0.02
Primary production	0.31–0.41 g $C/m^3/h$	0.003 g $C/m^2/h$ [2]
Retention time (y)	0.053	1
Phytoplankton bloom frequency	Constant	Rare
Trophic status	Eutrophic to hypereutrophic	Mesotrophic

Note: All Lake Loiza data from Quinones-Marquez (1980) unless otherwise indicated. All Lake Gatun data from Zaret (1984) unless otherwise indicated.

[1] Values obtained from 1994 and 1995 field experiments.

[2] Calculated from oxygen values assuming a Redfield Ratio of 106:138 (Stumm and Morgan, 1996).

three separate field experiments: Lago Loiza 1994, Gatun Lake 1994, and Lago Loiza 1995.

METHODS

Bubbles emanating from lake-bottom sediments were collected in partially submerged inverted 26-cm-diameter funnels, previously used and described by Keller and Stallard (1994). Bubbles entered the funnels through the funnel mouths and displaced water within the plugged necks of the funnels. Funnels were deployed at the water surface in groups of three to five, spaced 2 m to 3 m apart, and connected to loops on polyline with plastic clips. Clusters of funnels were placed above particular water depths ranging from 1 m to 10 m, and care was taken to maintain equivalent bottom distances below each funnel in the group. Funnels were deployed at water depths of 1 m, 3 m, and 10 m in the Gatun study; 1 m, 3 m, and 6 m in the Loiza 1994 study; and 3 m and 6 m in the Loiza 1995 study. The largest depth represents the maximum depth within the study area at the time of the experiment. A depth of 1 m was not sampled in the Loiza 1995 study because the very low water levels that existed several days prior to the experiment had abruptly risen almost 3 m, causing exposed sediments to be re-wetted. It was unknown whether methanogenesis in re-wetted sediments would be representative.

At 2-hour intervals, gas volumes were removed from the funnels with 20-ml glass syringes. The 2-hour interval was chosen to constrain timing of bubbling episodes and to limit dissolution into the water or consumption of gases within the collection devices by microbial organisms. Keller and Stallard (1994) note that during their study of Gatun Lake, less than 3 percent of methane in the collectors was lost within 2 hours after injecting known volumes of gas.

Methane fluxes from the lake surface to the atmosphere were calculated according to:

$$f = \frac{VC}{A\Delta t} \quad \text{Eq. 1}$$

where V is sample volume, C is the methane concentration of the sample, A is the collector area, and Δt is the time interval of collection.

Gas volumes were accurately and carefully measured, but because of problems with equipment availability, methane concentrations were not measured in this experiment. Consequently, methane concentrations are based on previous work at Gatun Lake and similar settings (Keller and Stallard, 1994), with appropriate accounting for error involved in these assumptions. These researchers noted that methane concentration varied little at each sampling site (±10 percent methane), but that methane concentrations were generally inversely related to depth. The higher methane concentrations observed in bubbles from shallow environments were probably due to rapid stripping of dissolved gases other than methane as a result of more frequent bubbling in shallow sites (Keller and Stallard, 1994). In order to obtain concentration estimates for the Gatun 1994 experiment, a linear regression was fit to Keller and Stallard's concentration data.

Methane studies had not been performed previously at Lago Loiza, and bubble-composition data were unavailable. A range of concentrations based on data from similar environments was incorporated into flux calculations. Methane concentrations in bubbles from freshwater, non-vegetated regions have been shown to range from 43 to 100 percent methane (Strayer and Tiedje, 1978; Chanton et al., 1989; and Keller and Stallard, 1994). From depths less than or equal to 6 m, the minimum methane bubble concentration recorded was 50 percent methane (Chanton et al., 1989). In order to consider a likely range of methane flux from Lago Loiza, minimum and maximum concentrations of 50 percent and 100 percent were assumed by using a methane concentration of 75 percent and incorporating a standard deviation of 25 percent methane into flux calculations.

Error for flux measurements was calculated as the propagation of error for each of the components of flux (A, Δt, C, and V). The area of the gas collector, time interval, and gas volume could be measured very accurately, making concentration estimates the primary source of error. The flux error, calculated as a standard deviation, and neglecting area and time terms, is (Bevington, 1969):

$$\sigma_f = \left(\frac{\sigma_v^2}{V^2} + \frac{\sigma_c^2}{C^2} + 2\frac{\sigma_{vc}^2}{VC} \right)^{1/2} \qquad \text{Eq. 2}$$

where σ_v is the uncertainty in volume measurements, V is the average volume per funnel per sampling interval, σ_c is the uncertainty in concentration estimates, and C is the average methane concentration (Keller and Stallard, 1994). Because there is no correlation between concentration and sample volume (Keller and Stallard, 1994), the covariance term is neglected.

Potential sources of error for volume estimates result from measurement error (±0.5 ml), dissolution into the water column (–3 percent), and barometric changes (±2 percent). Atmospheric pressure was not measured during the experiments. However, Atkinson (1973) reports that barometric changes of 10 to 20 mm Hg (corresponding to a 1 to 2 percent change in overall atmospheric pressure), which are common, are capable of causing bubbles to move in or out of solution. For the Gatun study, which based concentration estimates on a 1988 dataset, concentration error was calculated to be a function of the standard deviation in concentration per site (approximately ±10 percent methane) plus the analytical error in gas chromatograph measurements (±3 percent). For the Loiza studies, concentration error was assigned a value equal to the maximum deviation (±25 percent) in concentration values, assuming an "average" concentration of 75 percent methane.

A self-logging InterOcean Systems S4 current meter recorded current velocities throughout the field experiments. The S4 current meter measures water velocity as a potential gradient through an electromagnetic field, making it capable of detecting very small velocities. Currents were measured continuously and vector averaged within the internal microprocessor over a programmed time period. The accuracy of the current meter is 2 percent of the reading ±1 cm/s, and the resolution is 0.2 cm/s. The meter was deployed 0.5 m from the sediment–water interface at the intermediate depth site for the duration of the experiments.

An anemometer-driven bicycle odometer was used to measure wind speed, and measurements were recorded at approximately 2-hour intervals.

Methane Flux Data

Lago Loiza

The Loiza 1994 experiment began July 26 at 0630 h and ended July 27 at 1700 h (local time). The intended sampling interval was 2 hours, but floating water hyacinth mats frequently interfered with sampling. Average methane flux was greatest from the 3-m site, followed by the 1-m and 6-m sites. In constructing flux histograms for the 6-m site, 2-hour weighted averages were calculated, and measurements following a delay in sampling were distributed across the un-sampled interval using weighted averages. The largest methane emissions occurred during early-afternoon hours (Figure 1A). Contribution to average methane bubble flux from the 3-m site was 75 percent and from the 6-m site was 25 percent. As in the Gatun study, the deepest site had lower fluxes than the shallower sites.

Gas samples were also measured at Lago Loiza in 1995, but the current meter malfunctioned, resulting in very little recorded velocity data. Consequently, the relationship between current velocities and methane flux was not evaluated for this experiment. However, it is interesting to note that ebullitive flux from the Loiza 1995 experiment was one to two orders of magnitude lower than the previous year's experiment (8 ± 13 (mg/m^2)/day to 24 ± 22 (mg/m^2)/day), likely the result of a rapid water depth increase (3 m in 4 days) that occurred just prior to the experiment.

Gatun Lake

The Gatun study began September 18 at 1000 h and ended September 21 at 1100 h (Figure 2). Gas measurements were taken every 2 hours. Methane fluxes from Gatun Lake were calculated using methane concentrations of 80 percent, 72 percent, and 43 percent for the 1-m, 3-m, and 10-m sites, respectively, as calculated by the Keller and Stallard (1994) study. Average methane fluxes from sites in Gatun Lake were 5 ± 16 (mg/m^2)/day at the 10-m site, 1,088 ± 240 (mg/m^2)/day at the 3-m site, and

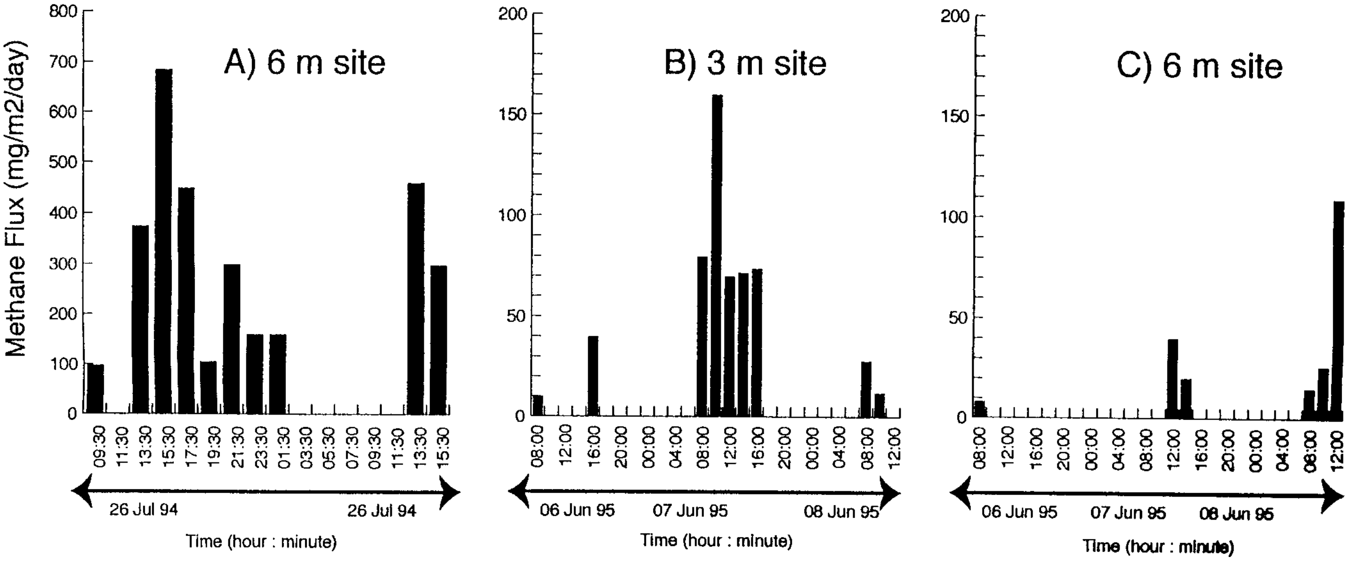

Figure 1. Histograms of methane flux for Lago Loiza. (A) 6-m site, 1994 experiment. (B) 3-m site, 1994 experiment. (C) 6-m site, 1995 experiment.

884 ± 212 (mg/m^2)/day at the 1-m site. Methane flux from the 1-m and 3-m sites at Gatun was consistent with values measured previously at Gatun Lake (Keller and Stallard, 1994), but ebullitive flux from the 10-m site was comparatively small. At Gatun Lake, ebullitive methane fluxes from the two shallowest sites were similar in magnitude. Of the total ebullition, 55 percent was from the 3-m site, and 45 percent was from the 1-m site. Bubbling from the 10-m site was negligible, contributing only 0.2 percent of the total ebullitive emissions.

The contrast between ebullition from the two shallowest sites and ebullition from the deep site supports previous observations that methane bubbling from depths greater than 5 m is often suppressed (Keller and Stallard, 1994; Galy-Lacaux et al., 1999). Keller and Stallard (1994) measured methane flux from several coves in Gatun Lake during a series of experiments and noted that in many locations methane flux associated with bubbling was "strongly anticorrelated with depth." In contrast to this relationship, however, they found that ebullition from the shallow site in Gatun was less than ebullition from intermediate sites, probably as a result of wave activity's inhibiting fine-grained organic sediment accumulation near the shore of the lake. Similar results were evident in the 1994 Gatun experiment.

Most gas release occurred during short-lived bubbling events. The largest bubbling events occurred during morning hours between times of 0600 h and 1200 h (Figure 2). Between 55 and 58 percent of the total bubbling at the 1-m and 3-m sites occurred between these hours. Bubbling generally subsided at night, generating only 9 to 10 percent of the total gas flux between midnight and 0600 h. Minor bubbling at the 1-m and 3-m sites, averaging 4 ml to 6 ml of gas per funnel, was interrupted periodically by major bubbling, averaging 25 ml to 27 ml of gas per funnel. During the 73-hour experiment, 60 percent of the total gas emissions at the 1-m and 3-m sites occurred over a total time of only 14 hours.

The large error estimate for the 10-m site is a function of small volumes measured and the estimated concentration (43 percent methane), which is small relative to the standard deviation (±10 percent methane) for concentration measurements.

Wind and Water Current Analysis

Average wind and current velocities were calculated over gas sampling intervals using weighted averages. Wind velocities were usually greatest during daytime hours and subsided at night. Magnitude of current velocity, however, did not correspond to any particular time of day. The influence of wind on current velocity appears to be overprinted by flow toward an outlet in Lago Loiza and by boat/barge effects in Lake Gatun.

Bottom shear stress is a function of current velocity gradients. A model designed to relate these two parameters has been developed that incorporates the empirical current data. Bottom shear stress is dependent upon the water column velocity gradient as defined by the logarithmic law of the wall (Middleton and Wilcock, 1994, p. 389):

$$\frac{V_o}{U_*} = \frac{1}{\kappa} \ln z + C \qquad \text{Eq. 3}$$

where U_* is the bottom friction velocity (defined as $U_* = (\tau/\rho)^{1/2}$), κ is Von Karmen's constant, z_b is the height of the current meter above the lake bed, V_b is current

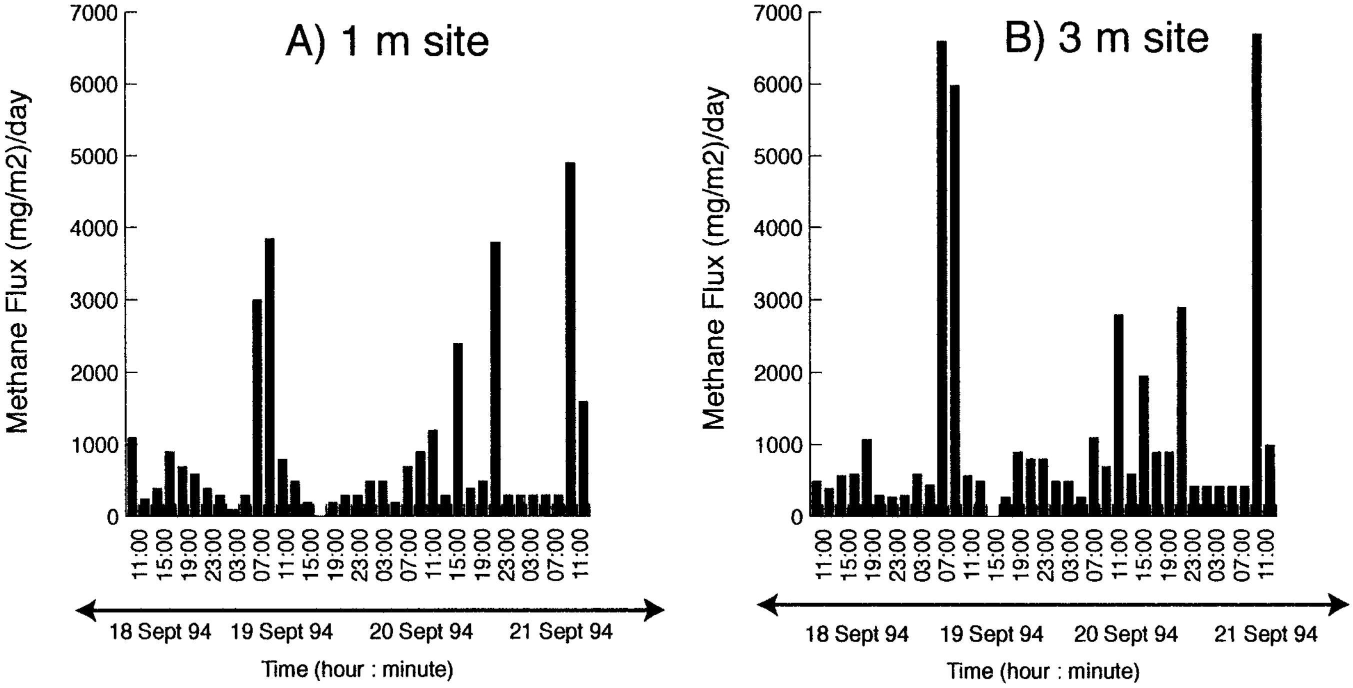

Figure 2. Histograms of 1994 methane flux for Lake Gatun. (A) 1-m site. (B) 3-m site. Minimal methane flux was recorded at the 10-m site and is not shown.

velocity at z_b, and C is the constant of integration. κ, which has been shown empirically to range from 0.38 to 0.41, was assumed to equal 0.40 for these calculations. The integration constant in Equation 3 can be determined by assuming that the velocity is zero at a finite distance (known as the sediment roughness length, z_o) above the sediment–water interface. The sediment roughness length is dependent upon sediment structure and grain size. It was estimated to be 0.02 cm for muddy sediments, based on the data of Soulsby (1983). Under these conditions, the law of the wall can be rewritten as:

$$\tau = \left[\frac{1}{\kappa}\ln\left(\frac{z_b}{z_o}\right)\right]^{-2} {}^*V_b{}^*|V_b|^*\rho \qquad \text{Eq. 4}$$

The current velocity, V_b, was obtained from current meter data measured from a known elevation above the lake bed (0.5 m), and fluid density was assumed to be 1,000 kg/m^3, the approximate density of fresh water. Using these parameters, current velocities of the magnitude observed in the lakes would produce bottom shear stresses up to 0.026 Pa for a velocity of 10 cm/s (approximately the maximum observed in these two lakes).

Lake-bottom sediments, as well as many other geologic materials, behave as Bingham fluids. Bingham fluids are similar to Newtonian fluids in that, when subjected to shear stresses, the magnitude of shearing is directly related to their viscosity. Bingham fluids differ, however, in that they have a finite cohesive strength. Sediments can be sheared only by bottom currents that are able to overcome this cohesive strength. Therefore, only current and wind velocities above 'threshold' values will be able to trigger bubble release. The shear stress imparted to a Bingham fluid is given by

$$\tau = \tau_o - \mu\frac{\partial V_x}{\partial z} \qquad \text{Eq. 5}$$

where τ is shear stress, τ_o is the cohesive strength of the sediments, μ is fluid viscosity, V_x is current velocity, and z is depth.

When $\tau > \tau_o$, the shear stress is great enough to move the sediments and induce bubbling (Figure 3). The cohesive strength of the sediments in Lakes Gatun and Loiza is unknown and may vary according to changes in organic content, pore pressure, and hydrostatic pressure (Dyer, 1986; Randkivi, 1998). High concentrations of gas bubbles lower the threshold yield strength by increasing pore pressure, and increases in organic content and in hydrostatic pressure raise the yield strength by increasing sediment cohesiveness and promoting compaction. In quiet regions, where the sedimentation rate exceeds the consolidation rate, layers of 'liquid mud' may form. These fluid sediments respond to small shear stresses. The sediments in Lakes Gatun and Loiza, which were deposited from quiet productive waters, are very fluid, both visually and to the touch. Reported sediment density

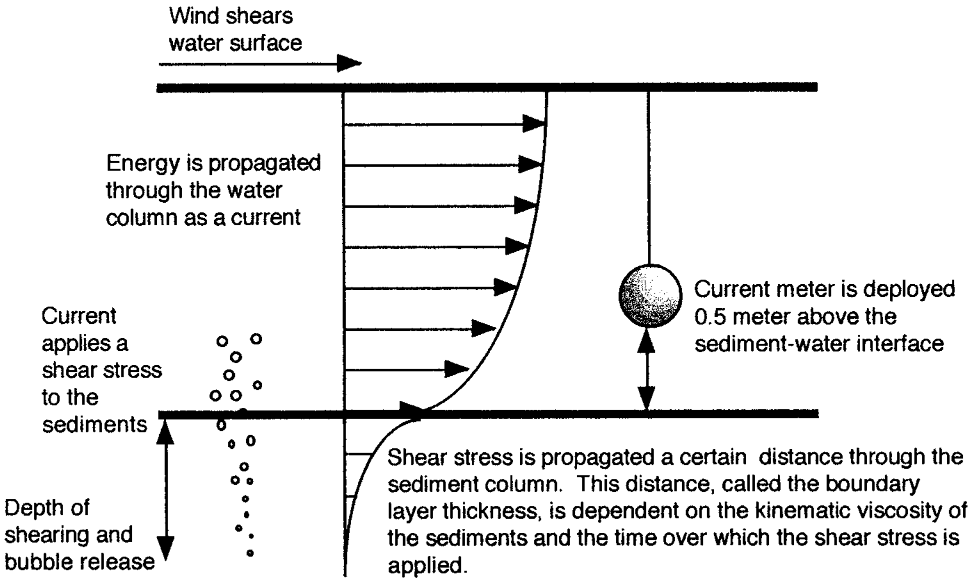

Figure 3. Graphical representation of wind- and current-induced bottom shearing, which is thought to trigger episodes of bubble release. Bottom shear stresses are calculated using current data collected by the current meter deployed a known distance above the sediment–water interface.

in Lago Loiza is not much greater than water (Webb and Soler-Lopez, 1997). Cohesive strengths for low-density, muddy sediments such as these are on the order of 0.01 to 0.1 Pa (Krone, 1974). The measured currents in Lakes Loiza and Gatun are therefore capable of producing shear stresses large enough to overcome the cohesive strength of the sediments.

When the sediment shear strength is exceeded, the sediment column begins to move. In this case, the boundary condition for Equation 3 changes from zero to a finite value of velocity, V_o, at the sediment roughness length, z_o.

$$\frac{V - V_o}{U_*} = \frac{1}{\kappa} \ln\left(\frac{z}{z_o}\right) \qquad \text{Eq. 6}$$

Under these conditions, the shear stress from the water column to the sediment column remains constant, and an expression for the upper boundary of the sediment can be rewritten as:

$$V = V_o + \left(\frac{\tau_o}{\rho}\right)^{1/2} \frac{1}{\kappa} \ln\left(\frac{z}{z_o}\right) \qquad \text{Eq. 6a}$$

The time-dependent velocity structure of the sediment column can be described with a simple diffusion equation (Bird et al., 1960):

$$\frac{\partial V}{\partial t} = \gamma \frac{\partial^2 V}{\partial z^2} \qquad \text{Eq. 7}$$

where V is the velocity within the sediment and γ is the kinematic viscosity of the sediments. Equation 7 was solved with an explicit finite difference method using real-time velocity measurements from the current meter (assuming a surface sediment velocity of $[V - V_o]$ as shown in Equation 6) and a range of typical kinematic viscosities for soft muddy sediments (10^{-8} m^2/s to 3×10^{-7} m^2/s; Randkivi, 1998). The shear stress within the sediment was then calculated as the product of the velocity gradient and the kinematic viscosity. Under these conditions, significant sediment velocity, and hence bubble release, would not be present below approximately 20 cm below the sediment–water interface at both Lago Loiza and Lake Gatun (Figure 4).

Methane Flux, Current and Wind Data, and the Shear Stress Model

For Gatun Lake data, flux measurements collected after periods of excessively large fluxes (>2,000 $mg/m^2/day$) were not included in the correlation calculations, because depletion of bubbles in the sediments presumably would make subsequent shearing irrelevant. Threshold values of wind and current velocity yielding shear stress greater than 0.01 Pa were chosen in order to eliminate low shear stress values, which would be ineffective in moving sediments (Equation 4). Wind velocities below threshold values of 4.5 km/h and current velocities below values of 6.5 cm/s (1-m data) and 7.5 cm/s (3-m data) were excluded from the dataset. These current velocities, corresponding to shear stresses of

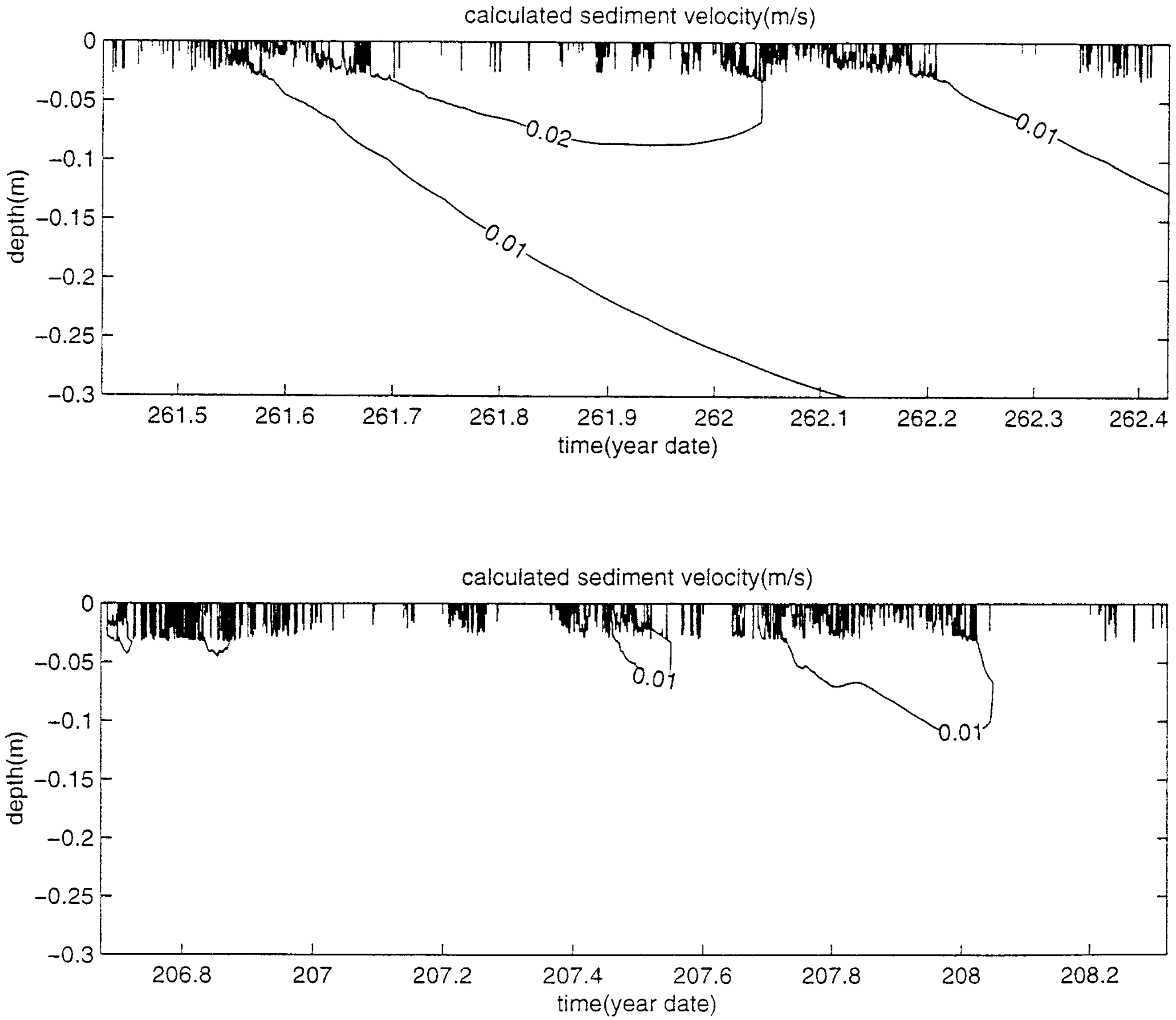

Figure 4. Time-dependent velocity of sediment calculated using Equation 7 and simplified assumptions discussed in the text for (top) Lake Gatun and (bottom) Lago Loiza.

0.011 Pa and 0.015 Pa, respectively (Equation 3), are within the shear strengths believed to be appropriate for fluid sediments of Gatun and Loiza.

A correlation was observed between wind velocity and methane flux ($r = 0.63$ and 0.52) at both the 1-m and 3-m Gatun sites (Figure 5A and B). Current velocity and methane flux were better correlated at the 1-m site ($r = 0.84$) than at the 3-m site ($r = 0.56$) (Figure 5C and D). Current velocity and methane flux demonstrate a stronger correlation than wind velocity and methane flux at both sites. During the experiment, several episodes of wave generation and water oscillation, resulting from ship traffic, were observed. The fact that currents are often generated by sources other than wind shearing may explain the poorer correlation between wind velocity and gas flux. All of the correlations were stronger at the shallower site, but whether the difference is physically significant, or why it might be so, is unclear.

Volumetric and temporal patterns of ebullition from the 1-m and 3-m sites were strongly correlated (Figure 6; $r = 0.92$), indicating that the mechanism that triggers bubbling acts over wide horizontal distances (tens of meters in this case). The shearing model is supported by this observation, because it demonstrates that bubbling episodes are not random occurrences but are instead triggered by the physics of water motion.

Methane flux data from the Loiza 1994 study were not as complete as the 1994 Gatun data because of water hyacinth–induced problems in accessing and maintaining the gas collection equipment. The majority

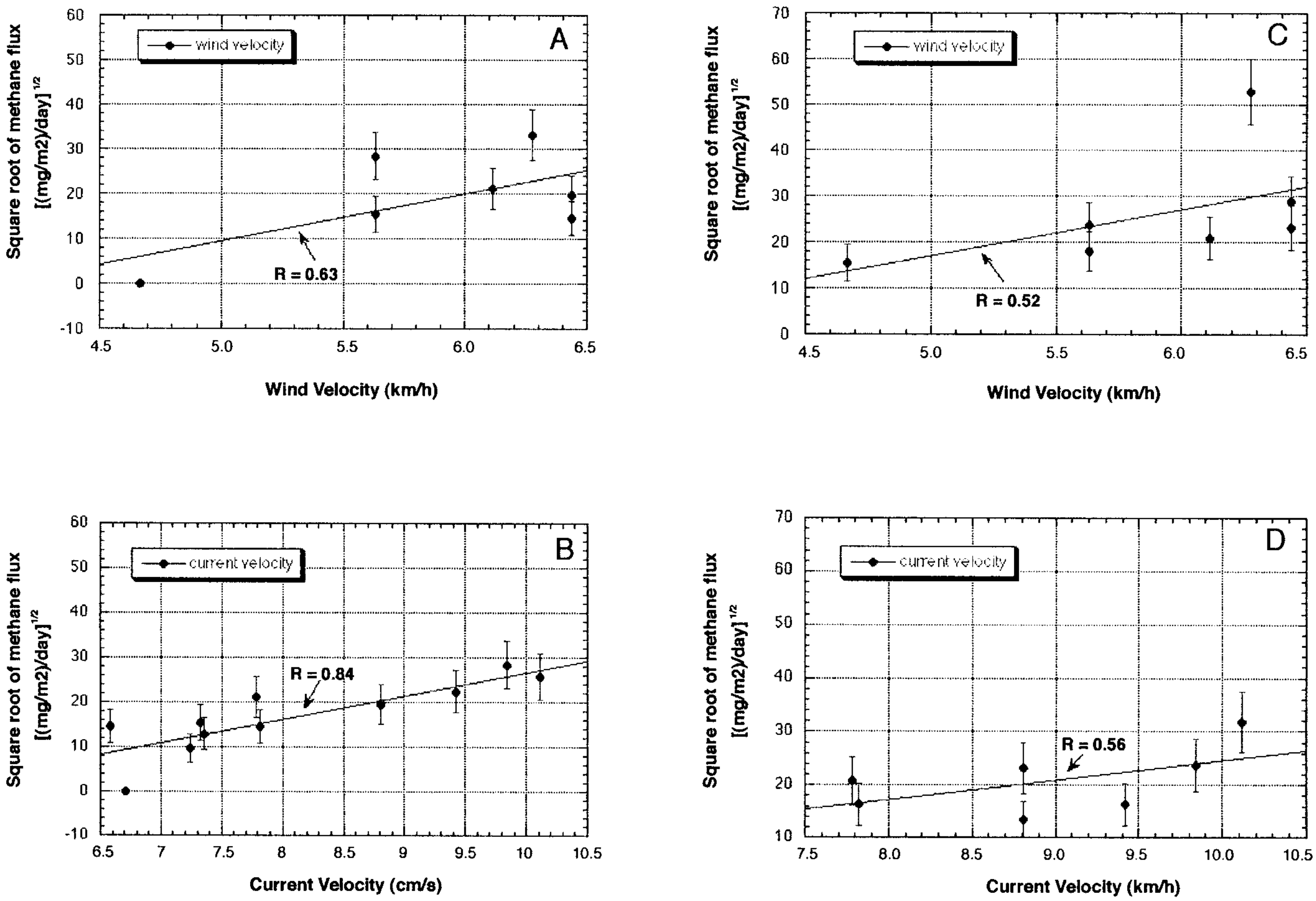

Figure 5. Relationship of methane flux to (A) wind velocity and (B) current velocity per sampling interval for 1-m site data from Lake Gatun. Independent variables include only data greater than or equal to threshold values of 4.5 km/h and 6.5 cm/s for wind velocity and current velocity, respectively. Relationship of methane flux to (C) wind velocity and (D) current velocity for 3-m site data from Lake Gatun. Independent variables include only data greater than or equal to threshold values of 4.5 km/h and 7.5 cm/s for wind velocity and current velocity, respectively. The plots do not include values that were collected after periods of flux greater than 2,000 (mg/m^2)/day. Because shear stress is proportional to the square of the current velocity in Equation 4, the square root of methane flux should therefore be proportional to the bottom-current velocity.

of the data were collected from the 6-m site. The following analysis describes data from 12 sampling intervals at the 6-m location. Flux measurements, collected after periods of excessively large fluxes (>600 mg/m^2/day), wind velocities below threshold values of 8.5 km/h, and current velocities below values of 7.0 cm/s were not included in correlation calculations. It should be noted that the value for "excessively large fluxes" at Lago Loiza is less than that at Gatun Lake because overall flux values at Lago Loiza were much smaller. Threshold parameters were higher because wind and current velocity were generally greater at Lago Loiza than at Gatun Lake. A poor correlation ($r = 0.32$) was found between wind velocity and methane flux (Figure 7A). Current velocity ($r = 0.81$) was better correlated (Figure 7B and C). The poor relationship between wind velocity and methane flux may be the result of shielding of the water surface against wind shear by water hyacinths (similar phenomenon noted by Barber et al., 1988) and the existence of currents related to flow of water toward the lake outlet.

CONCLUSIONS

This study highlights several features of methane production and flux in shallow terrestrial sediments with significant geotechnical and global climatic change implications. Temporal correlation of bubbling across sites in Gatun Lake and positive correlations between current velocity and gas flux support the bottom-shearing model for bubble release (Figures 5 and 6). Variability in these relationships is observed because the magnitude of methane flux depends not only on threshold shear velocity but also on the quantity of bubbles in the sediments available for release. Large bubbling events may occur with no apparent trigger because during prolonged quiescent periods, substantial accumulations of

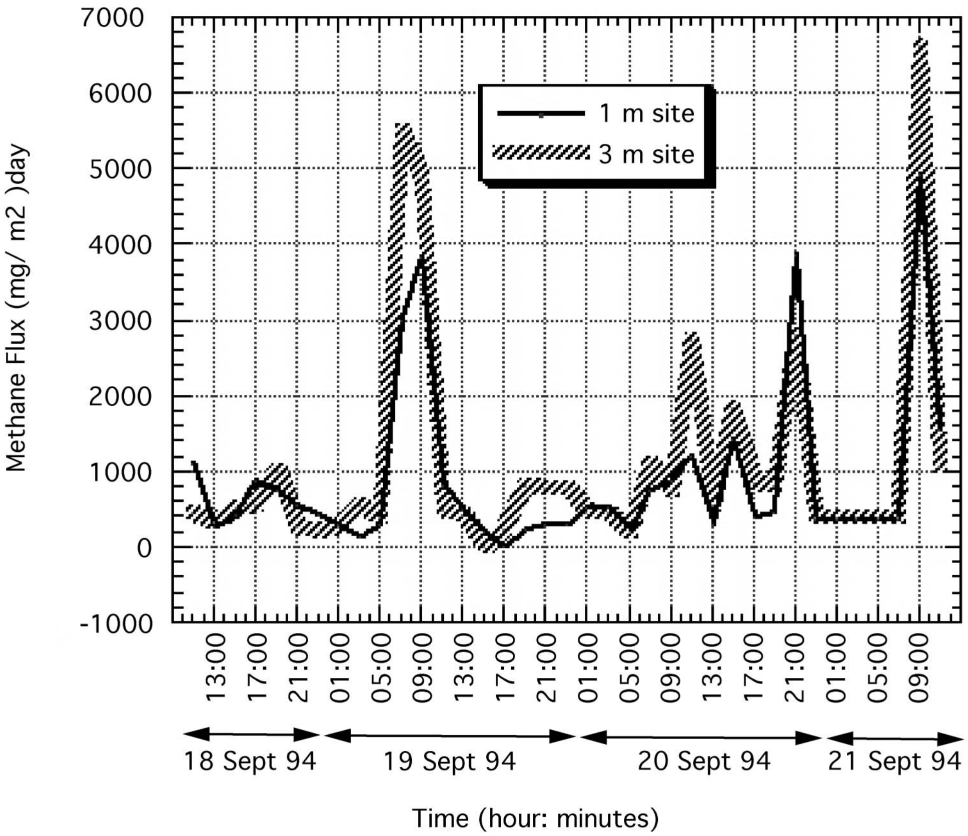

Figure 6. Comparison of methane emissions from the Gatun study (1-m and 3-m sites). The temporal correlation between sites is strong ($r = 0.92$, N = 37), indicating that the process that induces ebullition acts over tens to hundreds of meters.

bubbles escape by their buoyancy alone. Conversely, significant current velocities may trigger no gas release because sediments have been depleted of gas bubbles by recent, prior events.

Current velocity generally correlates well with bubbling when data that follow large flux events or are below threshold values are systematically excluded (Figures 5 and 7). Wind velocity does not correlate as well as current velocity with ebullitive methane flux because currents that are capable of producing bottom shearing may be caused by phenomena such as seiches, density-induced currents, or flow toward a dam.

The bottom-shearing model may explain why large concentrations of methane bubbles have been documented as acoustic anomalies in sheltered, shallow marine sediments such as Ekernforde Bay in Germany (Anderson et al., 1998; Jackson et al., 1998), whereas in other organic carbon–rich marine settings such anomalies are not common. If bottom currents produce sufficiently high shear stress that the cohesive strength of the unconsolidated sediments is exceeded, methane tends to be released. Conversely, weak bottom currents may allow accumulation of methane at shallow depths in the sediments.

This study of Lago Loiza and Lake Gatun is similar to previous studies in showing that depth exerts an important influence on methane ebullition (Keller and Stallard, 1994; Galy-Lacaux et al., 1999). Water depth can influence methane flux by changing the solubility of methane through pressure and temperature variations, attenuating shear stress at depth, and providing a greater opportunity for oxidation of methane in the sediments and water column. For these reasons, ebullition typically is not important in deep water but is generally the primary mode of gas transport in shallow lakes. Fluxes generated via ebullition, often orders of magnitude greater than fluxes generated by diffusion alone (Strayer and Tiedje, 1978; Keller and Stallard, 1994), usually escape oxidation because of their rapid transport. In each of the field experiments conducted, methane flux from the deepest site was much smaller than flux from shallower sites. The 10-m site in Gatun Lake produced almost no ebullition, although bubbles could be generated by intentionally disturbing lake-bottom sediments. In the two 1994 experiments, the 3-m site produced more bubbling than the 1-m site. This discrepancy is probably a result of heterogeneous production rates, possibly related to the coarser, less organic character of near-shore sediments or the spatially heterogeneous deposition of organic material by floating organisms.

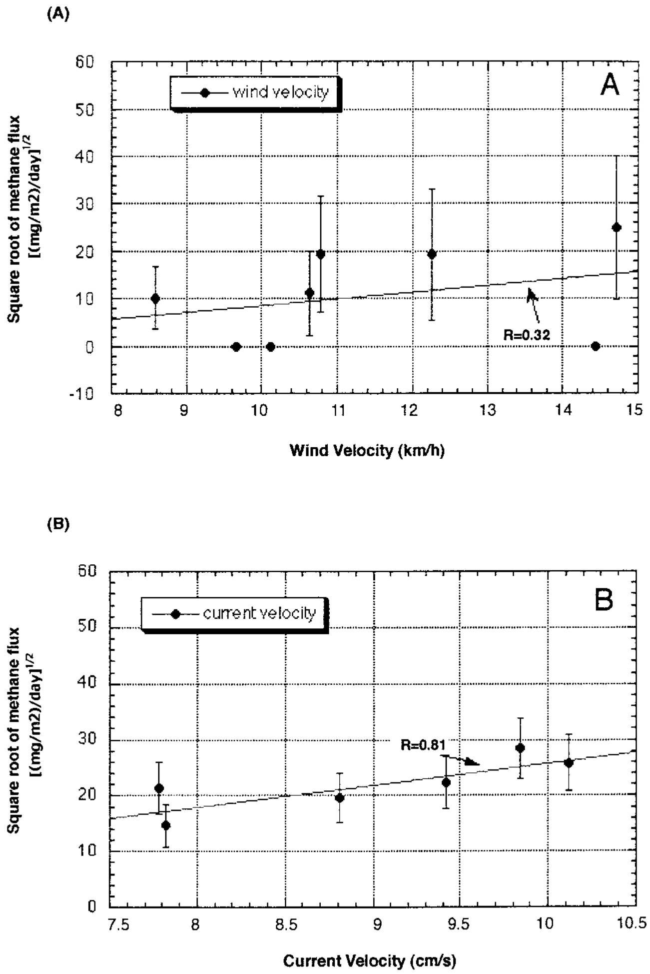

Figure 7. Relationship of methane flux to (A) wind velocity and (B) current velocity for 6-m site data from Lago Loiza. Independent variables include only data greater than or equal to threshold values of 8.5 km/h and 7 cm/s for wind velocity and current velocity, respectively. The plots do not include values that were collected after periods of flux greater than 600 (mg/m^2)/day.

The contrast between the 1994 and 1995 Loiza field experiments demonstrates how sudden rises in water elevation may temporarily halt ebullition. These results, consistent with other gas-flux studies involving rising or falling water (Martens and Klump, 1984; Chanton et al., 1989; Bartlett et al., 1990), have direct implications for modeling methane flux from reservoirs with widely fluctuating water levels, because abrupt rises in water level may create a significant 'lag time' in ebullition, whereas falling water elevations may induce periods of above average rates of ebullition.

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