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Release of multiple bubbles from cohesive sediments

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[1] Methane is a strong greenhouse gas, and marine and wetland sediments constitute significant sources to the atmosphere. This flux is dominated by the release of bubbles, and quantitative prediction of this bubble flux has been elusive because of the lack of a mechanistic model. Our previous work has shown that sediments behave as elastic fracturing solids during bubble growth and rise. We now further argue that bubbles can open previously formed, partially annealed, rise tracts (fractures) and that this mechanism can account for the observed preferential release at low tides in marine settings. When this mechanical model is applied to data from Cape Lookout Bight, NC (USA), the results indicate that methanogenic bubbles released at this site do indeed follow previously formed rise tracts and that the calculated release rates are entirely consistent with the rise of multiple bubbles on tidal time scales. Our model forms a basis for making predictions of future bubble fluxes from warming sediments under the influence of climate change. **Citation:** Algar, C. K., B. P. Boudreau, and M. A. Barry (2011), Release of multiple bubbles from cohesive sediments, *Geophys. Res. Lett.*, *38*, L08606, doi:10.1029/2011GL046870.

1. Introduction

[2] Methane is generated in sediments during the later stages of anoxic microbial organic matter decay (methanogenesis), the melting of shallow gas hydrates, or the thermal decomposition (catagenesis) of organic compounds. Near-surface methane profiles in sediment porewaters indicate that the dissolved methane flux is effectively intercepted by aerobic and anaerobic oxidation [*Boetius et al.*, 2000; *Dale et al.*, 2006; *Caldwell et al.*, 2008]. Bubble formation and rise (ebullition) constitutes, therefore, the primary pathway for gas release from sediments [*Hovland et al.*, 1993; *Joyce and Jewell*, 2003]. While the flux of methane as bubbles from sediments is probably modest, i.e., <10 Tg yr^{-1} [*Chappellaz et al.*, 1993], the potency of methane as a greenhouse gas makes any flux of interest to climate modellers. Furthermore, as the temperature of mid- and high-latitude sediments increase with global warming, methane fluxes will also increase, due to thawing of sediments [*Shakhova et al.*, 2010], thermal effects on diagenetic sources, and melting of shallow gas hydrates.

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[3] Prediction of future fluxes of methane from sediments has been stymied by a lack of a mechanistic model for bubble release. Recent studies [e.g., *Johnson et al.*, 2002; *van Kessel and van Kesteren*, 2002; *Barry et al.*, 2010], show that bubbles grow in sediments via elastic expansion and fracture of this medium. X-ray imaging [e.g., *Boudreau et al.*, 2005] shows that bubbles in cohesive sediments are thin, cornflake-like bodies, as a result of non-fluid sediment mechanics. *Gardiner et al.* [2003], *Algar and Boudreau* [2009, 2010], *Jain and Juanes* [2009], and *Algar et al.* [2011] have used elastic fracture mechanics and an idealized oblate spheroidal geometry to create an overall model of the initial growth and rise of bubbles in cohesive sediments. Such a 3D elastic-fracture process is extremely difficult to include in standard diagenetic transport-reaction models [*Berner*, 1980; *Boudreau*, 1997; *Burdige*, 2006], which are typically used to calculate fluxes from sediments to the overlaying waters or the atmosphere. The present paper offers a model for the calculation of bubble release rates.

[4] In addition, the release of bubbles in marine sediments is often tied to decreases in overlying pressure, e.g., low tides for shallow coastal sediments [*Martens and Klump*, 1980; *Chanton et al.*, 1989]. Preferential release of bubbles at low tide has been linked to expansion of the gas when the hydrostatic pressure drops [e.g., *Chanton et al.*, 1989]. Such statements are undoubtedly true, but at the same time, they do not explain quantitatively the mechanics of the observed release.

[5] *Algar and Boudreau* [2009] calculated the effects of tidal pressure variations on the growth rate of the first bubble to form in sediment similar to that at Cape Lookout Bight [*Martens and Klump*, 1980]. Figure 1 illustrates an expansion of those calculations for an initial bubble in the same sediment. We assume 8-m mean water depth and impose semi-diurnal tides of ±1 m, ±3 m, ±5 m, and ±7 m, the last to produce a tidal environment similar to the Bay of Fundy, Canada. Notice that while the volume of a bubble changes in proportion to the tidal amplitude, for tides ≤ ±3 m, the growth curve always returns to the no-tide growth curve. This means that tides have no net effect on the mass of gas within a bubble, and Figure 1 illustrates perfect gas expansion and contraction. Tides greater than ±5 m in amplitude do produce a net increase in growth; however, the effect is apparent only as the bubbles grow large (>500 mm^3), and it is not clear what natural environments would have such large tides coupled to strong sediment methanogenesis.

[6] This prediction of a modest effect of tidal variations on initial growth rates appears to fly in the face of the observed tide-bubble flux correlation at various locations, including Cape Lookout Bight and White Oak River [*Martens and Klump*, 1980; *Chanton et al.*, 1989]. The present communication offers a simple solution to this

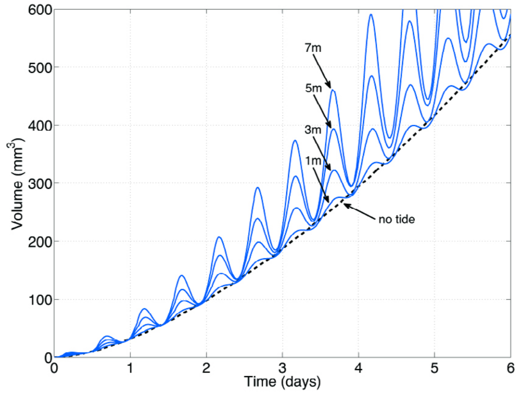

Figure 1. Model simulations that illustrate the effects of tidal height on the volume of a growing bubble started at high tide. The model was run with tidal amplitudes of 1, 3, 5 and 7 m. The dashed line shows bubble growth in the absence of tidal forcing. The influence of tidal height on bubble growth is not evident until tides exceed three meters in height.

apparent contradiction and reveals more details of the unfamiliar physics of bubbles in sediments.

2. Model

[7] The elastic-fracture model of bubble growth and rise is detailed by *Johnson et al.* [2002], *Gardiner et al.* [2003], *Algar and Boudreau* [2009, 2010], *Barry et al.* [2010], and *Algar et al.* [2011]. The gas exerts a pressure on the bubble walls, which opposes the total external pressure (load) and prevents the bubble from closing. When gas diffuses into a bubble, the internal pressure mounts and the bubble grows elastically in thickness $2b$ (Figure S1 of the auxiliary material).[1] If the gas source is sufficiently strong, then the internal force can grow to exceed the sum of the external load and the fracture toughness of the sediment. The bubble length, $2a$ in Figure S1, then grows by opening a crack, thus reducing the stresses in the surrounding sediment as the width $2b$ decreases elastically. These processes can be repeated many times as a bubble grows. (Note bubble, fracture and crack are interchangeable terms in this paper.)

[8] Growing bubbles begin to rise in sediments once the crack half-length, a, reaches a critical value, a_r, where [*Algar et al.*, 2011]

$$a_r = \left(\frac{3K_{IC}\sqrt{\pi}}{10\rho_s g}\right)^{2/3} \quad (1)$$

in which K_{1C} is the tensile fracture toughness, ρ_s is the bulk density of the sediment, and g is the acceleration due to gravity.

[9] The volume of a bubble, V_b, when $a = a_r$ is determined by the amount of linear elastic expansion [*Algar et al.*, 2011]:

$$V_b = \frac{16(1-\nu^2)\rho_s g a_r^4}{3E} \quad (2)$$

where ν is Poisson's ratio and E is Young's modulus.

[1]Auxiliary materials are available in the HTML. doi:10.1029/2011GL046870.

[10] We can now examine how a bubble reforms at an initial rise point and what are the effects of tides. To this end, we employ the finite-element model of bubble growth, LEFM-RD, of *Algar and Boudreau* [2009, 2010]. This model couples a reaction-diffusion equation, which describes gas production and diffusional transport through sediment porewater, to a linear elastic fracture mechanical model (LEFM) for the physics of the sediment.

[11] Fracture occurs when the stress intensity factor at the upper tip of the bubble, $K_1^{(+)}$, exceeds the fracture toughness of the sediment, K_{1C}, at which point the crack length is extended upward, and the internal bubble pressure drops. In a linear depth-dependent pressure field, $K_1^{(+)}$ is [*Algar et al.*, 2011]

$$K_1^{(+)} = \frac{10}{3\pi}\rho_s g a\sqrt{\pi a} + \frac{2}{\pi}\sigma_{-a}\sqrt{\pi a} \quad (3)$$

where σ_{-a} is the internal bubble loading at the crack tail (bottom of the bubble) in excess of the ambient total pressure. When a bubble rises $\sigma_{-a} = 0$. We need not consider fracture at the bottom of the bubble because the stress at the top of the bubble always exceeds K_{1C} before it can be exceeded at the bottom. This preference for growth at the top is illustrated in Animation S1 of the auxiliary material, which shows a growing bubble in gelatin, a mechanical analog for sediment [*Barry et al.*, 2010]. (Note we recognize that the stress field perturbation that is caused by the container bottom and sides affects bubble growth direction in this experiment.)

[12] If $a = a_r$, the bubble will release from its growth point and rise. When a bubble rises, the crack closes behind the bubble. If this crack now takes time to heal, i.e., to rebuild the broken bonds and the sediment strength to a pre-fracture level [e.g., see *Boudreau et al.*, 2005, Figure 4c], the partially annealed crack will have a lower K_{1C} than the surrounding sediment. As K_{1C} is lower in a partially annealed crack, equation (1) informs us that a_r will be smaller; therefore, subsequent bubbles will also be smaller than the initial bubble when the former rise. Assuming that the specific growth rate, i.e., mass delivered by diffusion to a new bubble per unit time, remains roughly constant, subsequent bubble release will be more frequent. It now remains to be seen if rise tracts with lower K_{IC} are preserved and if the frequency of release can correspond to tidal frequencies.

3. Application and Results

[13] Our hypothesis is that subsequent bubbles re-open pre-existing bubble-created fracture paths and that this re-opening is aided by tidal pressure variations. To test this conjecture we utilize bubble and methanogenesis data for Cape Lookout Bight, as an example, and apply equations (1)–(3) and LEFM-RD to calculate the apparent K_{IC} of the re-opened paths and the potential release rates. Parameter values for this site can be found in Table S1 of the auxiliary material.

[14] K_{1C} of the bubble paths can be estimated because *Martens and Klump* [1980] report sizes and numbers of bubbles released from sediments at Cape Lookout Bight during low tide (Table 1). Using these volumes and equation (2), the critical rise size, a_r, is calculated for each volume class, with

Table 1. Time Required to Grow a Bubble in a Previously Formed, but Partially Annealed Fracture, From an Initial Flaw Size of a_0 to the Size at Which It Will Leave the Sediment, a_r[a]

Percent of Total Bubbles	Mean Volume (mm^3)	K_{1C} ($Nm^{-3/2}$)	a_r (mm)	Minimum Growth Time $a_0 = a_r$	Maximum Growth Time $a_0 = 0.001$ m
6	1	13	7	52 min	3.6 hr
32	7	27	14	3.3 hr	11 hr
39	38	51	18	6.5 hr	31 hr
13	101	74	23	14 hr	56 hr
10	210	97	27	17 hr	88 hr

[a]Bubble data are for Cape Lookout Bight, NC, as reported by *Martens and Klump* [1980].

an assumed E value for these sediments (Table S1). Then, assuming that these bubbles all rose when $K_1^{(+)} = K_{1C}\,(\sigma_{-a} = 0)$, equation (3) provides K_{1C}. These K_{1C} values (Table 1) are 10–20% of the limited values measured *in situ* in other sediments, i.e., 500–1500 N $m^{-3/2}$ [*Johnson et al.*, 2002]. The escaping bubbles appear to have followed paths with significantly lower strength than undisturbed sediment.

[15] Next we need to estimate the time required to grow the bubbles in Table 1 using the LEFM-RD model. This model demands, however, an estimate of the initial flaw (crack) size, a_0, from which the bubble will grow before release. In the case at hand, we are unaware of the typical flaw size for the pre-existing rise path. Fortunately, we can set upper and lower bounds on a_0 and, thus, obtain upper and lower bounds on the formation times.

[16] As an upper bound, we use $a_0 = a_r$, as calculated by equation (2). This value means that subsequent bubbles do not go through a sequence of fracture events to reach their release volume. Instead, methanogenesis will supply gas to the crack and open it up purely by elastic expansion. When the volume becomes critical, i.e., V_b, the bubble rises. This produces the shortest possible growth time and a lower bound. The other possibility is that there is a minimum a_0 for sediments. We do not know this value, but our previous work indicates that initial growth times (i.e., minutes) for bubbles with $a_0 \leq 0.001$ m are negligible compared to the time for growth from a_0 to a_r. Thus, if we start with this value, we will obtain a maximum growth time for the bubbles. The minimum and maximum growth times are given as the last two columns of Table 1.

[17] The results in Table 1 show that observed bubbles of a volume of 38 mm^3 or smaller can be easily created on tidal time scales by re-opening a rise path. These bubbles account for 77% of the bubbles collected at Cape Lookout Bight.

[18] The larger bubbles in Table 1 cannot be grown on tidal scales, but these larger bubbles could be, in part, the result of the collision and union of smaller bubbles, both in the sediment and in the water column. An example of such collisions within sediment is provided in Animation 2; this movie shows a clear box filled with gelatin containing a pre-established bubble rise path. Each subsequent bubble follows the initial path, but there is a natural constriction, i.e., a point with a higher K_{1C}, about mid-way along the path. A rising bubble will stall at this point because it is not large enough to force the constriction open. The next bubble will rise to that point, merge with the previous bubble and continue to rise, as the larger bubble can force the gap open. The result is a bubble with twice the volume leaving the seabed.

[19] Figure 2 shows the growth history of several bubbles due to the reopening of a previously formed fracture with an initial length of 0.007 m. A particular bubble's history starts at a point on the abscissa, e.g., point A. Once each bubble reaches a critical size, it is removed by rise, e.g., point B, and another bubble starts to form in its place, e.g., point C.

[20] Figure 2 shows all the bubbles formed from a single site during a period of 24 hours. Between two and four bubbles form during each low tide. During high tide bubble growth ceases (point D) and reverses as the increase in ambient pressure compresses the bubble, which also causes it to dissolve. In this simulation, bubbles do not dissolve appreciably and growth restarts on the falling tide (point E). These results demonstrate multiple bubble release on tidal cycles.

4. Conclusions

[21] Once a rise path is formed, subsequent bubbles can easily form and rise by re-opening this fracture. Drops in pressure, e.g., low tides, promote this process. Results from our LEFM-RD model show that over 75% of the bubbles observed to release from the sediments at Cape Lookout

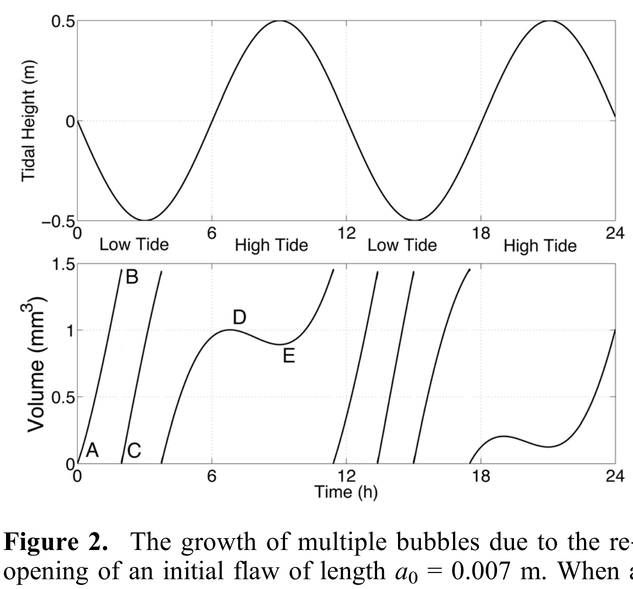

Figure 2. The growth of multiple bubbles due to the re-opening of an initial flaw of length a_0 = 0.007 m. When a bubble that starts at A reaches its critical size B, as determined by equation (1), it is removed and another one starts to grow in its place C. Bubbles only grow to critical sizes during low tidal periods and start to dissolve during high tide D, and then recommence growth on the falling tide E. Tidal height was taken to be 1 m.

Bight, NC, USA can easily be generated by this fracture-reopening mechanism.

[22] With our model, we are one step closer to predicting bubble fluxes from sediments. The missing elements are observations of the areal density of release points, a geographical and temporal tabulation of methane source strengths in relevant sediments, and better documentation of K_{IC} from these same sediments. If we can estimate the effects of warming on the source strengths, then we could predict bubble fluxes of the future.

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References

Algar, C., and B. P. Boudreau (2009), Transient growth of an isolated bubble in muddy, fine-grained sediments, *Geochim. Cosmochim. Acta*, *73*, 2581–2591, doi:10.1016/j.gca.2009.02.008.

Algar, C., and B. P. Boudreau (2010), Stability of bubbles in a linear elastic medium: Implications for bubble growth in marine sediments, *J. Geophys. Res.*, *115*, F03012, doi:10.1029/2009JF001312.

Algar, C. K., B. P. Boudreau, and M. A. Barry (2011), Initial rise of bubbles in cohesive sediments by a process of viscoelastic fracture, *J. Geophys. Res.*, doi:10.1029/2010JB008133, in press.

Barry, M. A., B. P. Boudreau, B. D. Johnson, and A. H. Reed (2010), First-order description of the mechanical fracture behavior of fine-grained surficial marine sediments during gas bubble growth, *J. Geophys. Res.*, *115*, F04029, doi:10.1029/2010JF001833.

Berner, R. A. (1980), *Early Diagenesis: A Theoretical Approach*, 241 pp., Princeton Univ. Press, Princeton, New Jersey.

Boetius, A., K. Ravenschlag, C. J. Schubert, D. Rickert, F. Widdel, A. Gieseke, R. Amann, B. B. Jøorgensen, U. Witte, and O. Pfannkuche (2000), A marine microbial consortium apparently mediating anaerobic oxidation of methane, *Nature*, *407*, 623–626, doi:10.1038/35036572.

Boudreau, B. P. (1997), *Diagenetic Models and their Implementation*, 417 pp., Elsevier, Berlin.

Boudreau, B. P., C. Algar, B. D. Johnson, I. Croudace, A. Reed, Y. Furukawa, K. M. Dorgan, P. A. Jumars, A. S. Grader, and B. S. Gardiner (2005), Bubble growth and rise in soft sediments, *Geology*, *33*, 517–520, doi:10.1130/G21259.1.

Burdige, D. J. (2006), *Geochemistry of Marine Sediments*, 630 pp., Princeton Univ. Press, Princeton, New Jersey.

Caldwell, S. L., J. R. Laidler, E. A. Brewer, J. O. Eberly, S. C. Sandborgh, and F. S. Colwell (2008), Anaerobic oxidation of methane: Bioenergetics, and the ecology of associated microorganisms, *Environ. Sci. Technol.*, *42*, 6791–6799, doi:10.1021/es800120b.

Chanton, J., C. Martens, and C. A. Kelley (1989), Gas transport from methane-saturated, tidal freshwater and wetland sediments, *Limnol. Oceanogr.*, *34*(5), 807–819, doi:10.4319/lo.1989.34.5.0807.

Chappellaz, J. A., I. Y. Fung, and A. M. Thompson (1993), The atmospheric CH_4 increase since the last glacial maximum: 1. Source estimates, *Tellus Ser. B*, *45*, 228–241.

Dale, A. W., P. Regnier, and P. van Cappellen (2006), Bioenergetic controls on anaerobic oxidation of methane (AOM) in coastal marine sediments: A theoretical analysis, *Am. J. Sci.*, *306*, 246–294, doi:10.2475/ajs.306.4.246.

Gardiner, B. S., B. P. Boudreau, and B. D. Johnson (2003), Growth of disk-shaped bubbles in sediments, *Geochim. Cosmochim. Acta*, *67*, 1485–1494, doi:10.1016/S0016-7037(02)01072-4.

Hovland, M., A. G. Judd, and R. A. Burke (1993), The global flux of methane from shallow submarine sediments, *Chemosphere*, *26*, 559–578, doi:10.1016/0045-6535(93)90442-8.

Jain, A., and R. Juanes (2009), Preferential mode of gas invasion in sediments: Grain-scale mechanistic model of coupled multiphase fluid flow and sediment mechanics, *J. Geophys. Res.*, *114*, B08101, doi:10.1029/2008JB006002.

Johnson, B. D., B. P. Boudreau, B. S. Gardiner, and R. Maass (2002), Mechanical response of sediments to bubble growth, *Mar. Geol.*, *187*, 347–363, doi:10.1016/S0025-3227(02)00383-3.

Joyce, J., and P. W. Jewell (2003), Physical controls on methane ebullition from reservoirs and lakes, *Environ. Eng. Geosci.*, *9*, 167–178, doi:10.2113/9.2.167.

Martens, C., and J. Klump (1980), Biogeochemical cycling in an organic-rich coastal marine basin: 1. Methane sediment-water exchange processes, *Geochim. Cosmochim. Acta*, *44*, 471–490, doi:10.1016/0016-7037(80)90045-9.

Shakhova, N., I. Semiletov, A. Salyuk, V. Yusupov, D. Kosmach, and O. Gustafsson (2010), Extensive methane venting to the atmosphere from sediments of the East Siberian Arctic Shelf, *Science*, *327*, 1246–1250, doi:10.1126/science.1182221.

van Kessel, T., and W. van Kesteren (2002), Gas production and transport in artificial sludge deposits, *Waste Manage.*, *22*, 19–28, doi:10.1016/S0956-053X(01)00021-6.

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