Formation of seep bubble plumes in the Coal Oil Point seep field

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Abstract The fate of marine seep gases (transport to the atmosphere or dissolution, and either bacterial oxidation or diffusion to the atmosphere) is intimately connected with bubble and bubble-plume processes, which are strongly size-dependent. Based on measurements with a video bubble measurement system in the Coal Oil Point seep field in the Santa Barbara Channel, California, which recorded the bubble-emission size distribution (Φ) for a range of seep vents, three distinct plume types were identified, termed minor, major, and mixed. Minor plumes generally emitted bubbles with a lower emission flux, Q, and had narrow, peaked Φ that were well described by a Gaussian function. Major plumes showed broad Φ spanning very small to very large bubbles, and were well described by a power law function. Mixed plumes showed characteristics of both major and minor plume classes, i.e., they were described by a combination of Gaussian and power law functions, albeit poorly. To understand the underlying formation mechanism, laboratory bubble plumes were created from fixed capillary tubes, and by percolating air through sediment beds of four different grain sizes for a range of Q. Capillary tubes produced a Φ that was Gaussian for low Q. The peak radius of the Gaussian function describing Φ increased with capillary diameter. At high Q, they produced a broad distribution, which was primarily described by a power law. Sediment-bed bubble plumes were mixed plumes for low Q, and major plumes for high Q. For low-Q sediment-bed Φ, the peak radius decreased with increasing grain size. For high Q, sediment-bed Φ exhibited a decreased sensitivity to grain size, and Φ tended toward a power law, similar to that for major seep plumes.

Introduction

Marine seeps emit the important greenhouse gas methane (CH₄), which is at least 20 times more potent than carbon dioxide (Khalil and Rasmussen 1995), to the hydrosphere and atmosphere. The marine seep contribution has been estimated at 10–30 Tg year⁻¹ (Kvenvolden et al. 2001), of a total natural source budget of 160–240 Tg year⁻¹ (IPCC 2001; Kvenvolden and Rogers 2005). Seep estimates are poorly constrained because few quantitative seep emission rates have been published (e.g., Hornafius et al. 1999), and dissolution (Leifer and Patro 2002) followed by microbial oxidation (Rehder et al. 1999) present a significant barrier to seafloor CH₄ transport to the atmosphere. Seep bubble dissolution and gas loss are strongly dependent on bubble size, water depth (Leifer and Patro 2002), and plume processes that can enhance significantly CH₄ transport (Leifer et al. 2006a). Thus, the bubble size distribution (used to initialize a numerical bubble model) is key to predicting the fate of seep CH₄. In this study, carried out in the Coal Oil Point seep field in the Santa Barbara Channel, California, we explore the relationship between gas flux, bubble size distribution, and sediment substrate to better understand the bubble-emission size distribution.

Published bubble-emission size distributions

Few seep bubble-emission size distributions, Φ, have been published, where Φ is the number of bubbles per radius increment emitted per second (also see the Appendix for definitions of all terms used in this study). Leifer and Boles (2005) proposed classifying seep bubbles into major and minor bubble plumes. In general, major plumes have greater flux (Q) than minor plumes, and their Φ are well described by a power law, i.e., ∼r⁻ˢ, where r is the radius, and s is the power law exponent. In contrast, minor plumes have narrow,
peaked \( \Phi \). Leifer (2010) identified additional modification of the size distribution based on oil contamination.

Bubble size distributions have been reported for the Gulf of Mexico (Leifer and MacDonald 2003) from exposed hydrate at 550 m, which had a \( \Phi \) for a minor vent with peak radius, \( R_p \), at 2,900 \( \mu \)m (\( R_p \) is the equivalent spherical peak radius where the distribution is at its maximum). Also, a major plume was observed with a weakly size-dependent \( \Phi \). Offshore Norway at the Håkon Mosby Mud Volcano, Sauter et al. (2006) reported a narrow \( \Phi \) with \( R_p \approx 2,600 \mu \)m in ~1,000 m of water. Likewise, Leifer and Judd (2002) identified a narrow, peaked sea-surface \( \Phi \) for North Sea seep bubbles. Several marine seep bubble size distributions have been reported for the Coal Oil Point (COP) seep field in the Santa Barbara Channel, including \( \Phi \) for three minor plumes that exhibited narrow, sharp \( \Phi \), and a major plume with a broad, shallow \( \Phi \) (Leifer and Boles 2005; Leifer and Tang 2007).

**Methods**

**Field measurement system**

Seep bubbles were observed with a video bubble measurement system (BMS; Fig. 1), which is reviewed in Leifer et al. (2003a), along with analysis approaches. The BMS measurement volume is backlit by two 300-W, wide-dispersion underwater lights (ML3010; DeepSea Power & Light, San Diego, CA), shining onto a translucent screen. Backlighting causes bubbles to appear (ideally) as dark rings surrounded by central bright spots, aiding computer analysis. Side-lighting produces half-moon shapes, which may require manual outlining (Leifer and MacDonald 2003). When bubbles are too close to the backlighting screen, off-axis rays obscure the bubbles’ edges, decreasing contrast and biasing \( r \) toward potentially significant underestimation (Leifer et al. 2003b).

To ensure the bubbles have a defined size calibration, clear screens with size scale markings delineate the BMS’s measurement volume in the axis along the camera view direction. Bubble blockers under the delineation screens in front and behind the measurement volume prevent bubbles from rising into the camera’s field of view at an unknown distance (i.e., not in the measurement volume). Parallax errors are minimized through long focal length, i.e., high zoom settings. The underwater video camera (SuperCam 6500; DeepSea Power & Light, San Diego, CA) allowed complete remote control, including shutter speed, which is set fast enough to prevent bubble blurring (Leifer et al. 2003a).

Although the ideal BMS would comprise long focal-length settings, and large distance between camera, measurement volume, and illumination screens, water turbidity, the need for a fast shutter speed, and illumination losses require trade-offs. Thus, the BMS components are mounted on an aluminum tubing framework with linear bearings to allow easy component repositioning based on water conditions and measurement requirements (i.e., spatial resolution, shutter speed, etc.).

All bolts are graphite lubricated to prevent seizing. Light and video cables are secured to the frame (and boat) with a stainless steel strain relief cable. The BMS is slightly negatively buoyant, with buoys to maintain a vertical orientation. Cables and a buoy line are taped together into a neutrally buoyant cable bundle. Video was recorded onboard with a mini digital video recorder (Sony Video Walkman; Sony, Tokyo).

**Laboratory measurement setup**

Bubbles were created from fixed, upright, 0.159-, 0.318-, and 0.635-cm-diameter, stainless-steel capillary tubes and from sediment beds. In the sediment laboratory studies, a range of airflows were passed through a 10-cm-thick sediment bed for a range of grain sizes, and the bubble
size distributions were measured. The sediment bed was a plastic, open-top cylinder, 13×10×12.5 cm deep, with a ceramic air stone fixed to the bottom in its center. It was filled with sediment of a particular grain size purchased from an aquarium supply store, creating a layer ~10 cm thick. The sediment bed was located in a glass, 40-L fish tank (15×30×67.5 cm deep) filled with deionized water to which salt was added to 35‰ salinity. The air stone was connected to a rotameter-controlled exterior air source (FL3804-ST; Omega, Stamford, CT) with three flows, $Q_{f1}=6.4$, $Q_{f2}=6.9$, and $Q_{f3}=8.1 \text{ cm}^3\text{s}^{-1}$ at a constant and measured pressure. $Q$ was determined from the fill time for a 1,000-mL inverted, submerged beaker, and was measured before and after each test series for each $Q$.

Illumination was by two 500-W light bulbs, which could be adjusted with a variable transformer. Video was recorded from the BMS video camera, which was positioned ~42.5 cm from the measurement volume. Image quality was improved significantly by preventing off-axis light rays (Leifer et al. 2003b). Specifically, the entire tank exterior was shrouded with flat-black plastic and particleboard with circular openings on the tank’s front and rear that allowed for camera viewing and illumination. Light passed through two translucent white plastic sheets, which were sandwiched between the particleboard and tank to provide homogeneous illumination. Short video sequences were digitized and processed in the same manner as for the field data.

Sample sediment grains were digitally photographed (Coolpix 5400, Nikon, 12 megapixel), and analyzed using the photo imaging processing and analysis software, ImageJ (W. Rasband, 1997–2008, U.S. National Institutes of Health, Bethesda, MD, http://rsb.info.nih.gov/ij/). Imaged particles were thresholded (i.e., made binary above and below an intensity value), a watershed filter (cf. segmentation filter that separates touching particles) was applied, and each particle’s area calculated. Then, a mean equivalent radius was calculated. Sediment dry density was determined by weighing a sediment-filled, 1-L beaker, both dry and filled with water. Based on the weight difference, the void volume or porosity, $\phi$, was calculated, with the remaining volume combined with the weight to yield the sediment grain density. The relative density or packing efficiency, $\zeta$, was defined as $\zeta = 1 - (\phi/1,000)$.

Analysis

Video clips (Fig. 2a) were acquired at 8-bit, 60 fields per second digital video resolution (720×240 pixels), and analyzed using macros written in ImageJ. The macros (Leifer and MacDonald 2003; Leifer et al. 2003a; Leifer 2010) extract the video to 60 frames per second by linearly interpolating the odd and even image rows on each frame (Fig. 2d). Macros also removed background intensity variations (Fig. 2c), decreased pixilation noise, filtered out blurry bubbles, and thresholded the image sequence (Fig. 2e).

Aided by macros that predict bubble location in subsequent frames, and to automatically track bubbles where possible, a significant fraction of bubbles were tracked between frames. For each bubble, the position, major and minor axes, angle, perimeter, area, circularity, and skew based on the thresholded outline, as well as frame number were recorded. Then, a convex-hull outline was calculated and the bubble re-measured. A convex hull is the smallest polygon that can contain the bubble outline (also a rubber-band outline), and has the effect of filling concavities where the bubble appears as a “half moon”, which can result from off-axis illumination (Fig. 2f). Further analysis uses routines written in MatLab (Mathworks, Natick, MA).

Two measurements are available for each bubble, one with and one without a convex hull. In cases where the bubble is half moon-shaped, the convex-hull outline is used; otherwise, the non-convex-hull outline is used. Discrimination is based on the circularity, $C$, defined as $C=4\pi \text{ area}/\text{perimeter}^2$. If $C(\text{convex-hull}) > C(\text{non-convex-hull}) + 15$, then the convex-hull measurement was used. For the bubbles labeled in Fig. 2f, $C=0.238$ and 0.781 for the non-convex-hull and convex-hull outlines, respectively.

Then, for each tracked bubble, the mean bubble speed and equivalent spherical radius, $r$, from all measurements during its passage across the field of view, and the standard deviations were determined. The bubble speed is corrected based on the mean bubble trajectory to a vertical velocity, $V_Z$. $V_Z$ allows conversion of the steady-state size distribution (layer concentration) to the emission size distribution (flux passing through a horizontal plane), by accounting for the number of repeat measurements of each bubble. The mean bubble rise trajectory is calculated from all the bubble trajectories using a least-squares linear regression analysis of the positions of the tracked bubbles. The trajectory angle results from camera tilt and currents. A polynomial fit of $V_Z$ to $r$ is used to derive a representative $V_Z(r)$. Then, outliers of more than one to two standard deviations are removed from the dataset (depending on the variability in $V_Z$), and $V_Z(r)$ recalculated. $V_Z(r)$ includes the effects of buoyant rise, turbulence, oil, and bubble-induced upwelling flows. The upwelling flow is the vertical fluid motion driven by momentum transfer from the rising bubbles.

All bubbles are $r$- and time $(t)$-segregated, and histograms $N(r,t)$ were calculated. The histogram $r$-bin widths are logarithmically spaced, and chosen so that a statistically significant number of bubbles are counted in bins near the distribution peak(s). The measurement uncertainty is $N^{0.5}$, where $N$ is the number of bubbles in each histogram bin.
Normalization of the histogram by the radius increment and depth interval yields the bubble-layer population size distribution, $\Psi$, which is the total number of bubbles per meter of depth per size increment (typically micrometers), and is what sonar observes. The bubble-emission size distribution $\Phi$ (cf. number of bubbles passing vertically through a plane per second per size increment) can be calculated from $\Psi$ in combination with $V_Z(r)$, the frame rate, and the measurement volume height. $\Phi$ is used to initialize bubble models, and to calculate the vertical gas flux (Leifer and MacDonald 2003). This multiple count correction between $\Psi$ and $\Phi$ includes effects from bubble trajectory tilt, bubbles touching the edge (not counted), and bubble eccentricity (major axis/minor axis) variations with $r$.

In the geophysics literature, bubble measurements typically are reported as concentration size distributions, defined as the number of bubbles per unit radius per size increment, and are characterized by a power law—for example (Johnson and Cooke 1979),

$$F(r, t) = k(t)r^{-s(t)}$$  \hspace{1cm} (1)

where $F$ is or $\Psi$, $k(t)$ a constant that may vary with time ($t$), and $s$ the power law exponent. The fit is determined from a least-squares linear regression analysis over a selected range of $r$. The parameters $k$ and $s$ can be derived from a least-squares linear regression analysis applied to the log of both sides of Eq. (1), i.e., to

$$\log(F(r)) = -s \log r + \log k$$  \hspace{1cm} (2)

For minor seep bubble plumes, $F(r)$ is poorly represented by a power law, being sharply peaked and narrow (Leifer and Boles 2005). Thus, $F(r)$ also was fitted with a parametric Gaussian model:

$$F(r) = A(t)e^{-[(r-R_P)/\tau]^2}$$  \hspace{1cm} (3)

where $A$ is the peak value of $F$, $R_P$ the peak radius, and $\tau$ the Gaussian half-width fitted to the model using a least-squares method (MatLab Curve Fitting Toolbox; Mathworks, Natick, MA). Uncertainties in $R_P$ and $\tau$ are given by 95% confidence intervals.

**Site description**

The COP seep field is probably the best-studied seep field in the world. Studies have quantified seep area (e.g., Allen et al. 1970; Fischer and Stevenson 1973; Fischer 1978) and emission fluxes (Hornafius et al. 1999; Clark et al. 2000) using sonar techniques, ocean geochemistry, and direct gas capture. Fischer and Stevenson (1973) noted changes in the seep field on decadal timescales, with significant areal decrease from 1946 to 1973. Based on a comparison of sonar data and oil company seep maps, they attributed this decrease to offshore production. During the last decade, the seep field has been mapped and flux determined from sonar data, and by a direct gas capture flux buoy (Washburn et al. 2001, 2005), suggesting that $\sim1.5 \times 10^3$ m$^3$ day$^{-1}$ ($4.5 \times 10^{10}$ g year$^{-1}$) seep gas is emitted to the atmosphere from $\sim3$ km$^2$ of seafloor (Hornafius et al. 1999), with roughly an
equal amount dissolved into the coastal ocean (Clark et al. 2000). The seep field releases ~80 barrels day$^{-1}$, or 5×10$^6$ L year$^{-1}$ (Clester et al. 1996), with oil slicks a common channel feature (Leifer et al. 2006b). Also, it has been noted that oil emissions vary with tides (Mikolaj and Ampaya 1973).

Geologic structures including anticlines, faults, and outcroppings are the dominant control of the seepage spatial distribution. Seeps generally follow several water depth trends (Fig. 3) related to controlling geologic structures (Leifer et al. 2010, this issue). The inner trend is ~20 m deep, and includes Shane Seep (34°24.370′N, 119°53.428′W). A second trend at ~40 m depth includes the Trilogy Seeps (Trilogy A: 34°23.634′N, 119°52.702′W) and Horseshoe Seep. The deepest trend (~70 m depth) includes the Patch (34°21.850′N, 119°49.755′W) and La Goleta Seeps.

**In situ bubble-stream classes**

Seep bubble plumes in the seep field are highly diverse, spanning a range of bubble sizes, fluxes, temporal variability, spatial distributions, and oil contamination. This wide diversity of seepage can exist for vents in close proximity—for example, most seepage classes occur in the SCUBA diver-accessible Shane Seep area, an extensively studied megaseep (Leifer et al. 2004). Megaseeps are so termed because they release >10$^6$ L day$^{-1}$ (Clark et al. 2010, this issue). Shane Seep covers ~10$^3$ m$^2$ and comprises thousands to tens of thousands of vents, as well as two to five main bubble plumes—the number has varied over the years (Leifer et al. 2004). Each main plume has a clearly defined surfacing footprint, and arises from one to several dominant plumes and tens to hundreds of smaller plumes at the seabed. At the sea surface, the large bubble plumes are 2–5 m in diameter and are associated with strong upwelling and outwelling flows. Surface expressions correspond to seabed hydrocarbon volcanoes and/or mounds, which are a few meters in diameter and are located in areas with a higher density of seep vents. The main water-column bubble plume arises from the most prolific seabed vents, which are situated in an area with the highest vent density and seabed features. Surrounding the central area is a peripheral area dominated by low-flow vents at lower spatial density. The vent spatial density generally decreases with distance from the central seep features, along two linear trends that are presumed related to rock fractures and faults.

Survey and BMS video (see Fig. 4) and analysis of bubble-plume $\Phi$ suggest three important seep vent classes with distinct $\Phi$, these being major, minor, and mixed (cf. below). In general, $Q$ for major plumes is larger than for minor plumes. $\Phi$ for mixed vents have characteristics of both major and minor plumes.

**Results**

In situ major and minor bubble size distributions

$\Phi$ were analyzed from BMS video (Fig. 5) of a minor and major vent near a major Trilogy Seep pockmark. For one minor vent, $\Phi$ was sharply peaked and well described by three Gaussian functions with peak radii, $R_p$, at 1,878±58, 2,979±331, and 4,149±707 µm and half-widths, $\tau$, of 344±81, 436±303, and 596±1,183 µm, respectively. Most likely, there were multiple seabed vent orifices within close proximity; bubbles were non-uniformly distributed with respect to lateral position and size in the BMS video. For this minor plume, the flux, $Q$, was 0.57 cm$^3$ s$^{-1}$. 

![Fig. 3 Map of sonar return from the Coal Oil Point (COP) seep field, Santa Barbara Channel, California. Contours and color map are logarithmically spaced for April 2005. Inset shows southwest US, red box seep field location. Seep names are informal. Sonar spatial resolution is 100×100 m for the deep seeps, and 50×50 m for the COP Seeps (from Leifer et al. 2010, this issue)](image-url)
The rise velocity function, $V_Z(r)$, was calculated (Fig. 5b) and used to estimate the upwelling velocity, $V_{UP}$, from comparison with rise parameterizations, $V_B$, for clean and dirty (contaminated) bubbles in stagnant fluid at a temperature of 13°C. There was a general decreasing trend in $V_Z$ with $r$, with the largest bubbles rising slower than the dirty $V_B$. This can be explained only by a loss of buoyancy due to oil, particularly because Patro et al. (2002) showed that bubbles in seawater larger than $r \sim 1,500$ µm should behave hydrodynamically clean. Because there are three distinct bubble-emission modes, it seems likely that the largest bubble population is the oiliest. $V_{UP}$ was estimated from the difference between $V_Z(r)$ and $V_B(r)$ dirty as $\sim 10$ cm s$^{-1}$.

The major plume that was analyzed from Trilogy Seep was located close to the minor vent shown in Fig. 5a. The vent pulsed strongly and produced large bubbles, although

Fig. 4 Seep bubble images showing different vent types. a Major plumes and minor plumes in a hydrocarbon volcano caldera, and b minor vents in peripheral area. Bubble measurement system images of c major vent plume, and d minor vent bubble plume in main caldera

Fig. 5 a Bubble-emission size distributions, $\Phi$, versus radius, $r$, and Gaussian fits for a minor vent plume at Trilogy Seep, and b vertical velocities, $V_Z$, for tracked bubbles versus $r$ for minor vent in a. Also shown are clean and dirty bubble rise velocity parameterizations in stagnant water (from Leifer and Patro 2002), and polynomial fits of $V_Z(r)$ to $r$ and $r=200$-µm binned data. Outliers in $V_Z$ are in red. c, d Same as a and b for major vent plume at Trilogy Seep
not as large as for the major plume at Shane Seep reported in Leifer and Boles (2005), which had $Q = 68 \text{ cm}^3 \text{s}^{-1}$. This Trilogy Seep major plume (Fig. 5c) had a significantly lower flux $Q$ of $10.4 \text{ cm}^3 \text{s}^{-1}$. Also, whereas bubble fragmentation was observed for the major plume at Shane Seep, fragmentation was not observed for this major Trilogy Seep vent. This difference likely results from the lower $Q$ and because the BMS was closer to the seabed for the SCUBA deployment at Shane Seep. $\Phi$ was well described by a three-part power law, which was very shallow for mid-sized bubbles of $1,000 < r < 3,000 \mu m$ with $\Phi \sim r^{-0.21}$, and significantly steeper for larger and smaller $r$, with power law exponents ($s$) of 2.83 and 2.22, respectively. Furthermore, there is some suggestion of a Gaussian shape at the small radius end of each power law fit (for example, see the Gaussian function centered at $3,262 \mu m$ in Fig. 5c). The vertical velocity, $V_Z$, for this major plume was highly variable due to turbulence, but with a trend in $V_Z$ that increased somewhat faster than the $V_B(r)$ parameterizations. This most likely resulted because larger bubbles tend to rise in a clump, i.e., in zones of locally higher buoyancy and thus transient increases in $V_{UP}$ (Leifer et al. 2009). Based on the $V_Z(r)$ trend more closely approximating $V_B(r)$ dirty, $V_{UP}=19.7 \text{ cm s}^{-1}$.

A longer time series was analyzed for a minor vent at Patch Seep (Fig. 6), and showed a $\Phi$ with three narrow Gaussian emission modes: $R_P = 2,062 \pm 21, 2,313 \pm 25$, and $2,735 \pm 76 \mu m$ with $\tau = 109 \pm 28, 137 \pm 36$, and $303 \pm 79 \mu m$ and correlation coefficients ($R^2$) of 0.978, 0.79, and 0.848, respectively. $\Phi(r,t)$ showed rapid shifts in the dominant $R_P$ with $t$ (Fig. 6b), which suggests several distinct vents in very close proximity that were active at different times. $R_P(t)$ was calculated for the strongest emission mode, and compared with the layer flux (for a $1-$cm layer), $Q_{layer}$, revealing two different bubble populations (Fig. 6c). One population (of size spanning the two smaller emission modes) had $Q$ independent of $r$ ($R^2=0.004$), while the other population, which arose from bubbles in the largest emission mode, showed $R_P Q^{0.43}$ ($R^2=0.54$). Thus, the greater $\tau$ for the largest emission mode is largely due to the dependency of $Q$ on $R_P$, as the shape oscillations of $r=2,735 \mu m$ bubbles are not markedly different from those of $r=2,313 \mu m$ bubbles (Leifer et al. 2000), which had much smaller $\tau$—note the similarity of $\tau$ for 2,313- and 2,062-$\mu m$ bubbles.

In situ mixed vent bubble size distributions

Although mixed plumes exhibited characteristics of both major and minor plumes, $Q$ values were not necessarily intermediate. Moreover, although mixed plumes were fitted with a combination of Gaussian distributions and power laws, the fits often were poorer than for the major and minor vents. $\Phi$ for a mixed vent on the east flank of the main hydrocarbon
volcano at Shane Seep (Fig. 7a) showed two Gaussian distributions with \( R_P = 969 ± 29 \mu m, \tau = 62 ± 34 \mu m \), and \( R_P = 889 ± 163 \mu m, \tau = 860 ± 227 \mu m \). There is a suggestion of another peak in \( \Phi \) at \( R_P \sim 1,400 \mu m \), of non-Gaussian shape that was not fitted. \( \Phi \) for \( 850 < r < 3,000 \mu m \) could be described by the power law \( \Phi \sim r^{-1.05} \). Further insight into the plume dynamics can be inferred from a time-dependent \( \Phi(t,r) \) showing that the plume pulsed (Fig. 8b). Moreover, the higher \( Q \) values in the second pulse (Fig. 8a) were accompanied by many smaller bubbles with \( \Phi \) that better described by a power law (Fig. 7a). Interestingly, \( V_Z(r) \) for this plume (Fig. 7b) closely followed the parameterization for clean bubbles, even down to \( r \sim 1,000 \mu m \). This suggests a value of \( V_{UP} \) of \( \sim 10 \text{ cm s}^{-1} \).

Laboratory sediment bubble formation

High-speed (7,000 frames per second), high-resolution (1,280×800 pixels) video (Phantom from Vision Research, Wayne, NJ) for gas bubbles rising through sediment in the laboratory \( (R_G = 0.43 ± 0.3 \text{ cm}) \) demonstrated the complexity of the mixed plume formation process (Fig. 9). Bubbles moved sediment grains (gray arrow), even lifting grains several centimeters. During formation, bubbles deformed dramatically, occasionally leading to pinching-off of one or two smaller bubbles (black arrow). Bubbles also were observed to become partially concave after formation, with their downstream hemisphere snapping up into the bubble (black double arrow). This created tiny internal droplets, and
in some cases led to the formation of very small bubbles. Finally, bubble coalescence was observed (white arrow).

Laboratory minor and major plumes

Airflow through capillary tubes for low $Q$ produced a single bubble chain. Gradually increasing $Q$, the bubble generation shifted to emission of two or three dominant sizes with sporadic small bubbles, and then for high $Q$ to a broad size spectrum. $\Phi$ for low $Q$ were sharply peaked and well described by a Gaussian function (Fig. 10a). A high-$Q$ bubble plume from a 0.16-cm-diameter capillary tube was analyzed and showed $\Phi$ spanning a broad range, $200<r<4,000 \mu$m (Fig. 10b), which could be described, albeit poorly, by the power law $\Phi \sim r^{-0.27}$. Finer structure in $\Phi$ suggested that several portions of $\Phi$ were well described by Gaussian functions; for example, there were two peaks at peak radii ($R_p$) of 600 and 1,030 $\mu$m and half-widths ($\tau$) of 128 and 164 $\mu$m, with good $R^2$ values of 0.925 and 0.936, respectively. The broad peak at $R_p=2,900 \mu$m, $\tau=549.2 \mu$m was reasonably described by a Gaussian function for $r>R_p$ but not for $r<R_p$ ($R^2=0.783$). Size segregation with $t$ was observed at the measurement volume, presumably because of the faster rise of larger bubbles—the tail of each bubble pulse comprised many smaller bubbles.

Laboratory mixed bubble plumes

Bubble plumes generated by percolating bubbles through a range of sediment sizes in the laboratory showed a dominant peak ($R_p$) that varied inversely with grain size ($R_G$)—for example, $R_p=4,777, 2,947, 2,300, and 1,239 \mu$m for $R_G=0.03, 0.43, 0.55, and 0.77$ cm, respectively (Table 1). In all
cases, Φ was described by a mixture of power law(s) and/or Gaussian function(s). For sediment with $R_G=0.55$ cm and for the low $Q_{F1}$ (Fig. 11a), Φ for $1,550<\tau<3,400 \, \mu m$ was well described by a broad Gaussian distribution with $R_P=2,157 \pm 179 \, \mu m$ and $\tau = 671.9 \pm 288 \, \mu m$ ($R^2=0.968$); for $R_P<1,550$ and $R_P>2,200 \, \mu m$, however, Φ was well described by steep power laws with $s=1.55$ and 3.12 for the small and large $r$-ranges, respectively.

With increasing $Q$ through a given sediment type, there was a shift from Φ being a mixture of Gaussian and power law functions to a single power law (Fig. 11c), with Φ for high $Q$ being similar to Φ for major plumes (Fig. 5c). For sediment with $R_G=0.55$ cm, Φ for the low $Q_{F1}$ was described largely by two non-contiguous power laws with $s=1.55$ and 3.12 for the small and large size populations, respectively (Fig. 11a). For the highest $Q_{F3}$ (Fig. 11c), Φ was best described by a single, broad (585<$r<5,020 \, \mu m$), relatively shallow ($s=1.29$) power law. The intermediate or transition $Q_{F2}$ produced a Φ similar to that for $Q_{F1}$, which was well described by bimodal non-contiguous power laws with power law exponents $s=2.33$ and 2.47 for the ranges $600<r<1,620$ and $2,320<r<5,200 \, \mu m$, respectively. Compared with Φ for $Q_{F1}$, the $r$ ranges were larger and the values of $s$ were intermediate (Fig. 11b), while the Gaussian trends in Φ for $Q_{F1}$ had largely disappeared. Because it appears that $Q_{F2}$ is a transition between low- and high-$Q$ behaviors, a power law spanning the range $550<r<4,750 \, \mu m$ was calculated for $Q_{F2}$, and found $s=2.33$, steeper than for $Q_{F3}$. To summarize, with increasing $Q$ the troughs in Φ that separated the two distribution modes were progressively filled, while the range of these modes increased between $Q_{F1}$ and $Q_{F2}$. Also, the $r$ dependency of Φ decreased, and the small Φ distribution was significantly filled from $Q_{F2}$ to $Q_{F3}$. Φ for $Q_{F3}$ was shallow and broad, consistent with the Φ associated with fragmentation.

Different behavior is expected for bubble formation from sediments where bubble size and sediment grain size are comparable—$R_P=R_G$ (e.g., Fig. 11), where sediment grain size is much larger than bubble size—$R_P<R_G$, and where sediment grain size is much smaller than bubble size—$R_P>R_G$. Where $R_P<R_G$, the sediment should have minimal impact on Φ; unfortunately, the sediment bed in the laboratory was insufficiently thick to adequately study this case. Where the two were comparable, high-speed video (Fig. 9) showed bubbles moving sediment during formation. For the studied flow range, sediments 2–4 (Table 1) all had $R_P<<R_G$ and exhibited decreased bimodality for higher $Q$ (e.g., Fig. 10c) than lower $Q$ (e.g., Fig. 10a).

For $R_P>R_G$ (sediment #1, $R_G=300 \, \mu m$), a different behavior is anticipated, because the bubbles cannot easily slip through the pathways of the sediment without moving many grains. Thus, elastic failure starts to become a key process in bubble migration and formation (Boudreau et al. 2005) for the smallest grain size that we studied, although this sediment was non-cohesive, unlike clays. One result is that bubble emissions tended to occur in transient pulses separated by times longer than the pulse length. For the low $Q_{F1}$ (Fig. 12), Φ contained a broad ($\sim 1,200<r<5,200 \, \mu m$)

### Table 1 Summary of laboratory sediment parameters

<table>
<thead>
<tr>
<th>Sediment</th>
<th>$D_G$ (mm)</th>
<th>$R_G$ (mm)</th>
<th>$dBD$ (g cm$^{-3}$)</th>
<th>$\phi$ (cm$^3$ m$^{-3}$)</th>
<th>$\zeta$</th>
<th>Grain density (g cm$^{-3}$)</th>
<th>$R_P$ (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sediment 1</td>
<td>0.6</td>
<td>0.3</td>
<td>1.414</td>
<td>0.255</td>
<td>0.745</td>
<td>1.90</td>
<td>4,777</td>
</tr>
<tr>
<td>Sediment 2</td>
<td>8.6±7.0</td>
<td>4.3±3.5</td>
<td>1.576</td>
<td>0.191</td>
<td>0.809</td>
<td>1.95</td>
<td>2,947</td>
</tr>
<tr>
<td>Sediment 3</td>
<td>11.0±10.0</td>
<td>5.5±5</td>
<td>1.425</td>
<td>0.339</td>
<td>0.661</td>
<td>2.16</td>
<td>2,300</td>
</tr>
<tr>
<td>Sediment 4</td>
<td>15.4±1.92</td>
<td>7.7±9.6</td>
<td>1.292</td>
<td>0.440</td>
<td>0.56</td>
<td>2.31</td>
<td>1,239</td>
</tr>
</tbody>
</table>

$D_G$ mean grain size (diameter); $R_G$ mean grain radius; $dBD$ dry bulk density; $\phi$ porosity; $\zeta$ relative density; $R_P$ peak bubble size distribution radius.
distribution of large bubbles, particularly compared to the other, coarser-grained sediment beds extending to greater than 5,000 µm. The dominant peak (by volume) was 4,777 µm, ~65% larger than for the next largest sediment where \( R_P = 2,947 \) µm and for which the value of \( R_G \) (4,300 µm) was comparable to \( R_P \) (Table 1). These larger bubbles arrived in pulses, supporting the idea that retention in the sediment facilitated coalescence into larger bubbles, which then migrated vertically and escaped the sediment transiently. There also was a small population of bubbles with \( R_P = 523.3 \) µm and \( \tau = 50.0 \) µm (fit not shown), a feature that was common in almost all sediment plumes for low \( Q \).

For higher \( Q \), \( \Phi \) changed dramatically (Fig. 12b). Although many large bubbles were still produced, the dominant peaks shifted to \( R_P = 462 \) and 641 µm, and were quite broad (relative to \( R_P \)) with \( \tau = 95 \) and 231 µm, respectively, comparable to \( R_G \). Also different from the other sediment beds, there was no indication of the formation of a power law in \( \Phi \), indicating bubble fragmentation was not dominant in the formation process. The small-bubble \( \Phi \) (\( R_P = 462 \) µm), though, was well described by a Gaussian function, extending to \( \tau \approx 250 \) µm, similar to coarser-grained sediment beds (e.g., Fig. 11a). Meanwhile, the large-bubble size distribution was significantly less important, whereas for the coarser sediment grains the large-bubble \( \Phi \) became more important (i.e., \( s \) became less or \( \Phi \) became shallower). Part of the explanation likely lies with increasing fluidization of the sediment bed—with increasing \( Q \) there was a dramatic increase in sediment grains lofted into the water column to above the measurement volume.

Discussion

Three distinct types of bubble plumes were observed in the field and laboratory—major, minor, and mixed. In the laboratory, minor and major plumes were produced from capillary tubes (fixed orifice) with the distinction being flux and orifice size. Mixed plumes were produced for unconsolidated sediment, although for high flux (\( Q \)) sediment bubble plumes were major.

Where bubbles escape individually, laboratory studies for bubbles escaping from a capillary tube have shown a narrow size distribution (Blanchard and Syzdek 1977). With increasing \( Q \), turbulence intensifies and smaller bubbles begin to appear. Finally, a broad bubble spectrum is produced for very high \( Q \) (Slauenwhite and Johnson 1999) that is weakly size-dependent. As a result, minor and major bubble plumes exhibit highly distinct \( \Phi \) due to different formation mechanisms. Note that \( Q \) for most major seep plumes is orders of magnitude greater than \( Q \) for most minor plumes.

Minor vents

Minor seep bubble formation is by pinch-off from vent orifices that are analogous to capillary tubes. Laboratory bubble size from an orifice is controlled by several factors such as orifice diameter, gas flux (Vazquez et al., unpublished data), orientation (Winkel et al. 2004), horizontal water velocity (Tsuge et al. 1981), and surfactant contamination (Hernandez-Aguilar et al. 2006) including bubble oiliness (Leifer and Boles 2005). \( \Phi \) for the range of capillary tubes used in the present laboratory studies was well described by a Gaussian function. For low \( Q \) in the laboratory, the broadness or half-width (\( \tau \)) of the Gaussian function is due to bubble-shape oscillations—passive acoustic observations of bubble size suggest a far narrower \( \tau \) (Leifer and Tang 2007).

For a vertically oriented orifice in stagnant water, \( r \) is related to the capillary tube orifice radius, \( r_{\text{cap}} \),

\[
r = 6r_{\text{cap}} \sigma_L / g (\rho_L - \rho_G)
\]

where \( \sigma_L \) is the liquid surface tension, \( g \) is gravity, and \( \rho_L \) and \( \rho_G \) are the liquid (seawater) and gas (methane) densities, respectively. However, numerical (Oguz and Prosperetti 1993) and experimental (Vazquez et al., unpub-
lished data) evidence suggests greater complexity with the bubble volume, varying as $Q^{6.5}$ above a critical gas flow rate, $Q_C$. For $Q<Q_C$, an increase in $Q$ causes the bubble formation rate to increase, but not $r$, i.e., $r$ is independent of $Q$. Both of these behaviors were observed in the Patch Seep minor vent bubble population (Fig. 8), with $r \sim Q^{0.43}$ for one population, in excellent agreement with theory. This suggests that there were three vents (or sediment pathways) with different characteristics that combined to produce the $\Phi$ shown in Fig. 8. This also demonstrates how $\tau$ increased for $Q>Q_C$, i.e., based on comparison with laboratory $\tau(R_P)$, vents can be classified as $Q>Q_C$ or $Q<Q_C$.

Multiple peaks were observed for a number of minor plumes and for laboratory sediment plumes. The most common secondary peak was of very small bubbles even in cases where a peak was not clearly resolved (Leifer 2010). These very small bubbles often were observed with $\Phi$ distinct from $\Phi$ for large bubbles (i.e., not fit with the same functional form). Based on the high-speed video, these bubbles likely were created during bubble pinch-off (Fig. 9). For some vents, bubble peaks were for bubbles with integer multiple volumes (Fig. 5a) of the bubble volume. A bubble with $R_P=2,979$ $\mu$m has 3.99 times the volume of a bubble with $R_P=1,878$ $\mu$m. This suggests bubble formation with two modes where either one or four bubbles are produced from the same $Q$. Bubbles from another, far lower probability peak with $R_P=4,149$ $\mu$m may have originated from a separate vent. For most minor vents with multiple peaks, however, there was no clear relationship.

**Major vents**

$\Phi$ for major bubble plumes were primarily well described by shallow power laws, suggesting bubble fragmentation as the dominant formation mechanism. For the laboratory studies, increasing $Q$ from a capillary tube caused bubbles in the dominant peak (a minor plume at low $Q$) to visibly fragment into smaller bubbles. For the laboratory capillary tube and $Q_{f3}$, the process was not complete, and a dominant peak remained; however, $R_P$ for $Q_{f3}$ was smaller than $R_P$ for low $Q$, suggesting that $R_P \sim Q^{2.5}$ is inappropriate once fragmentation becomes important. Apparently, $R_P \sim Q^k$ where $k$ is a constant less than 0.4. At higher $Q$, there no longer is evidence of a preferential emission $R_P$, i.e., Gaussian peak. Because fragmentation occurs after the gas jet exits the orifice, it is unlikely to depend directly on orifice characteristics.

**Mixed vents**

Mixed vent $\Phi$ exhibited characteristics of both minor and major plumes, as well as different characteristics similar to neither of these types. Having characteristics of both suggests incomplete fragmentation of a bubble size distribution $\Phi$, which has a preferred size (Gaussian). Mixed plumes were not observed for capillary tubes, but were created for sediment-bed plumes. Thus, we propose that sediment grain mobility plays a critical role in the mixed plume formation process.

Three sediment bubble formation regimes are proposed for typical seep vents based on the grain size ($R_C$) to peak radius ($R_P$) ratio, leading to distinct $\Phi$. Specifically, for $R_G>R_P$ bubble buoyancy is insufficient to move sediment grains, while pore sizes are large with low capillary forces; thus, we predict high permeability (rapid migration) and $\Phi$ similar to capillary tube $\Phi$. In this study, $\Phi$ measured for sediment grain size #5 ($R_G=2.35$ cm) were similar to $\Phi$ for the bubble fret—i.e., the sediment-bed layer was too thin, and no analysis was conducted (cf. not shown in Table 1). At the other extreme, $R_G<R_P$ (sediment grain size #1), all bubbles can easily shift and lift many grains; however, capillary forces are high and the bubbles are larger than the sediment voids; thus, permeability is low, enabling sediment retention of bubbles. Here, migration bears similarity to elastic failure, because grains must shift to allow passage of a bubble, a process that was observed to occur very unsteadily. For the sediments in this study, however, cohesion does not play a role. Thus, the regime where elastic failure is truly dominant lies in even finer-grained sediments than used in this study, like clays, which do exhibit cohesive forces. Buoyancy and pressure forces must overcome cohesive forces and sediment compressibility to allow bubble migration. There exists extensive literature on bubble migration through fine-grained sediments (e.g., Sills and Wheeler 1992; O’Hara et al. 1995; Boudreau et al. 2005; Judd and Hovland 2007; Leighton and Robb 2008). From fine-grained cohesive muddy sediments, like a freshwater marsh, pulses of bubbles sporadically and infrequently escape, spanning very large to small bubbles, a characteristic of only the finest sediments studied in the present laboratory work. Finally, bubbles with $R_G>R_P$ can move and lift sediment grains during formation (Fig. 8). This leads to mobile sediment “orifices” and thus complexity in $\Phi$, producing mixed bubble plumes. $Q$ affects these categories: for higher $Q$, sediment with $R_G>R_P$ could become mobilized. Also, sediment grain characteristics at the vent likely change as bubbles remove smaller grains from the vent orifice, increasing $R_G$ until the remaining grains are too large to lift. Finally, for fine-grained sediments, the elastic failure mechanism is circumvented if the time interval between bubbles in the migration pathway is short, such that the sediment has sufficiently low relative density (high void space) to provide easy migration. In such cases, the sediment acts more like a fluid, unable to retain bubbles. The high-$Q$ case for the finest sediment in this study likely was close to this behavior.
Unfortunately, the seabed vents were not characterized; thus, the relationship between grain size and $Q$ elucidated in the laboratory requires further field studies for validation. Because of their greater buoyancy force, larger bubbles (correlating to higher $Q$; Leifer 2010) can be assumed to lift and displace larger sediment grains than smaller bubbles. Larger bubbles also generate stronger upwelling flows, which should lift larger grains (assuming the same grain density) higher. Small grains were observed being upwelled (and then sinking) in some of the seep video. As a result, for constant vent flux, there should be a gradual removal of small grains, with only grains too large to be moved remaining. Thus, there likely is a pattern of a shift from mobile to fixed orifice. Specifically, tar deposition, settling of sand grains into locked positions, and bacterial mats all could play a role in the evolution of the initial mixed $\Phi$ vents to minor or major $\Phi$ vents. Reversion to a mixed vent would occur if the migration pathway shifts (due to blockage of the vent or subsurface pathway), or after a transient or long-term increase in $Q$ that remobilizes sediments, or through deposition of new sediment.

Unsteady $Q$ has been observed in many seep bubble plumes. On 10-s timescales, wave-induced surge modulates bubble formation (Leifer and Boles 2005); however, seep vents have been noted to exhibit unsteady flow on second and sub-second timescales. Leifer and Boles (2005) observed a major plume that pulsed on sub-second timescales; moreover, pulsing was observed for the major seep vent in Fig. 5c. This pulsing is not a measurement artifact; size segregation of the pulse due to different $V_Z$ of bubbles of different $r$ are identified easily in the time- and size-resolved $\Phi$, with smaller bubbles arriving at the measurement volume later than larger, faster rising bubbles (e.g., Fig. 8). In the laboratory studies, pulsing was observed for sediment-bed plumes. This likely results from flow instabilities in the sediment bed, a process documented in fluid dynamics studies using magnetic resonance imaging of two-phase trickle-bed flow (Sederman and Gladden 2005). Trickle beds are used in chemical catalysis reactions, and for $Q$ above a threshold exhibit rapid pulsing behavior. A different type of pulsing was observed in the laboratory for relatively fine-grained, non-cohesive sediment ($R_P\gg R_G$) related to elastic failure migration, as discussed above. Pulsing was not observed for high $Q$ through sediments where $R_P\sim R_G$, nor for low $Q$ for capillary tubes. Similarly, low-$Q$ minor seep vents did not significantly pulse. Finally, for major plumes from the capillary tube, rapid pulsing was observed at the measurement volume on quarter- to half-second timescales. Because the gas escapes the orifice continuously, turbulence processes must be responsible.

Although one would expect that with increasing grain size, and hence space between grains, there would be an increase in the dominant $R_P$ for low $Q$, laboratory observations showed the opposite. This trend held not only for sediments where $R_P\sim R_G$, but also for the relatively fine-grained sediment where $R_P<< R_G$. This suggests that the progressively decreasing ability of the sediment to retain bubbles is the underlying process, rather than pore size; finer sediments can more effectively trap bubbles prior to and during formation, because the capillary pressure required for a bubble to pass through pore spaces increases for smaller grain size. Sediment mobility also plays a role. High-speed video showed this process where during bubble formation, sediment grains surrounding the bubble are pushed aside and then settle after pinch-off. With increasing $R_G$, the sediment is less mobile because of packing leading to smaller pore throat and thus tending to decrease bubble size at formation. Also, the greater capillary force with decreasing grain size for a given buoyancy force ($Q$) will tend to decrease bubble size at formation.

Conclusions

The size-dependent flux distributions $\Phi$ for different seep vents were measured with a video bubble measurement system in the Santa Barbara Channel, California. Three distinct plume types were identified—major, minor, and mixed. Minor plumes generally were of lower emission flux ($Q$), with narrow, peaked $\Phi$ that were well described by Gaussian functions and were consistent with laboratory observations of capillary tube bubble formation at low $Q$. Major plumes showed a broad $\Phi$ spanning very small to very large bubbles, and were well described by a power law. Field observations of major plumes were consistent with laboratory bubble plumes produced from capillary tubes for higher flows, where bubble fragmentation created the broad, shallow $\Phi$. Examination of a long time record of a minor plume vent showed that some vent bubbles increased as $Q\sim (Q)^{0.43}$, in agreement with theory for $Q$ greater than a characteristic $Q$.

Mixed plumes showed characteristics of both minor and major plumes, which were very similar to those of laboratory sediment-bed bubble plumes. This suggests that unconsolidated sediment grains were key to mixed plume formation. High-speed video showed significant interactions between bubbles and mobile sediment grains, with grains being moved or lifted during the bubble formation and fragmentation process. Furthermore, the behavior of bubbles from sediments with grain sizes smaller than the bubble size was quite different than the behavior observed where grain and bubble sizes were similar. It was concluded that sediment retention ability was inversely related to sediment grain size over the range of (non-cohesive) sediment grain sizes studied here, and thus sediment grain size was inversely related to the size of the dominant peak
of \( \Phi \). For sediment with grains smaller than the bubble size, retention is efficient, creating pulsed emission separated by relatively long intervals. This emission mode was a poor model of field seep observations in the Coal Oil Point seep field. We propose that vents cycle between mobile sediment-bed mixed emission, and fixed capillary-like major and minor vent activity through a range of processes.

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**Appendix**

- \( A (-) \) Peak value of \( F \) in Gaussian functional fit
- \( C (-) \) Circularity of bubble outline. Used in image processing
- \( D_G (\text{mm}) \) Equivalent spherical diameter of sediment grains
- \( F \left( \# \text{~cm}^{-3} \text{~cm}^{-3} \right) \) Function that represents either \( \Phi \) or \( \Psi \)
- \( N (-) \) Number of bubbles analyzed
- \( Q (L \text{~min}^{-1}) \) Flow (corrected to STP)
- \( Q_{Layer} (L \text{~min}^{-1}) \) Layer flux (total volume of bubbles in a layer)
- \( R^2 (-) \) Correlation coefficient
- \( R_G (\text{mm}) \) Equivalent spherical radius of sediment grains
- \( R_P (\mu m) \) Radius of peak concentration in \( \Phi \)
- \( V_B (\text{cm} \text{~s}^{-1}) \) Bubble rise velocity in stagnant fluid
- \( V_{UP} (\text{cm} \text{~s}^{-1}) \) Upwelling velocity
- \( V_V (\text{cm}^3 \text{~s}^{-1}) \) Porosity or sediment void volume
- \( V_Z (\text{cm} \text{~s}^{-1}) \) Bubble vertical velocity
- \( dBD (g \text{~cm}^{-3}) \) Dry bulk density
- \( g (\text{m} \text{~s}^{-1} \text{~s}^{-1}) \) Gravity
- \( r (\mu m) \) Equivalent spherical bubble radius
- \( r_{cap} (\mu m) \) Capillary tube orifice radius opening
- \( s (-) \) Power law exponent in \( \Phi \)
- \( t (s) \) Time
- \( \phi \) Porosity
- \( \zeta (-) \) Relative density
- \( \rho_G (g \text{~cm}^{-3}) \) Bubble gas density
- \( \rho_L (g \text{~cm}^{-3}) \) Water density
- \( \tau (\mu m) \) Gaussian function half-width
- \( \Phi (\# \text{~cm}^{-3} \text{~cm}^{-3} \text{~s}^{-1}) \) Bubble flux size distribution
- \( \Psi (\# \text{~cm}^{-3} \text{~cm}^{-3} \text{~s}^{-1}) \) Bubble-layer population size distribution

**References**


