Supplementary Method

**Numerical bubble propagation model**

The numerical bubble propagation model employed for the clean bubbles at GC 185 is described in detail in Rehder et al. [1], and is an adaptation of the model described in Leifer and Patro [2] and Leifer et al. [3] for deep ocean applications. The model solves the coupled differential equations that describe the rate of change in the bubble molar content (n), equivalent spherical radius (r), pressure (P), and depth (z). Recently, the model has been modified to include pressure-dependent effects including compressibility, depth-dependent solubility, depth-dependent density effects on bubble rise, and hydrate skin effects within the hydrate stability field [1,3]. The driving force for bubble dissolution is the concentration difference between the bulk aqueous gas concentration and the bubble interfacial concentration, which is determined by the gas solubility at the in situ P-T conditions. The mass flux, $F_M$, from the bubble is described by:

$$F_M = 4\pi r^2 k_B(r,D)(C-HP_B)$$

where $k_B$ is the individual bubble gas transfer rate, $D$ is the gas diffusivity, $H$ is the Henry’s law constant, and $P_B$ is the bubble pressure. The density of CH$_4$ with depth deviates from the ideal gas law, as its compressibility ($\zeta$), decreases with depth. $\zeta$ is defined from $PV=nRT\zeta$, where $R$ is the ideal gas constant, $V$ is volume, and $T$ is temperature. In the model, a factor $q^*$, defined $q^*=RT\zeta/V$, describes the conversion between moles and atmospheres for a non-ideal gas, and compressibility enters the model by increasing the initial molar content of the bubble at a given $r$ and $z$, and is used to relate changes in $P_B$ and $n$. The change in size of the bubble is described by [1,3]:

$$\frac{\partial r}{\partial t} = \{q^*n/\partial t - \rho_wg\partial z/\partial t\}\{3(P_A + \rho_wgz + 4\alpha/r^2)\}^{-1}$$
where \( \sigma \) is the surface tension, \( \rho_w \) is the water density, \( g \) is the gravitational constant, and \( P_A \) is atmospheric pressure. Compressibility is then introduced by defining \( q^* \) [1,3], \( q^* = q/\zeta \). The change in bubble pressure is described by\(^1\):

\[
\partial P_B/\partial t = -3P_B/r\partial r/\partial t + q\partial n/\partial t
\]

The change in bubble depth is described by its stagnant fluid rise velocity, \( V_B \), and motions of the surrounding fluid including the upwelling flow, \( V_{up} \)\(^1\):

\[
\partial z/\partial t = V_B(r,T) + V_{up}
\]

For model initialization, the in situ physical, fluid dynamics, plume, and chemical parameters are set\(^1,3\). For plume simulations, the seafloor emission bubble size distribution is calculated, but in this study only single bubbles are simulated. The bubbles are simulated with an imposed upwelling flow (\( V_{up} \))\(^1,3\). The model solves the coupled differential equations with a third-fourth order Runge-Kutta scheme\(^2,3\). The tolerance is set to ensure numerical convergence. Integration is stopped if a bubble reaches the sea surface or dissolves to a set size limit.

**Model Run**

Based on detailed observations of the bubble plumes observed at GC 185\(^4,6\), the initial bubble size distribution is 0.2-10 mm, the bubble rise velocities (\( V_B \)) span ~5-30 cm s\(^{-1}\), the upwelling velocity (\( V_{up} \)) is 5-20 cm s\(^{-1}\), the volume flux is 21-61 cm\(^3\) s\(^{-1}\), and the mass flux at the main seep is 61-145 mmol s\(^{-1}\). The mean temperature from the profiles collected during the submersible transects (Supplementary Figures 5 & 6) were used in all simulations. Sensitivity to temperature is weak, thus changes of a few degrees Celsius has a negligible effect on the model predictions. The measured dissolved CH\(_4\) profiles were input as the bulk aqueous gas concentrations in the simulations, thus plume saturation effects were neglected. Although this is a conservative assumption, for the hydrostatic pressures over most of the bubbles lifetime, plume
concentrations are neglectable. For the simulation, air (O₂ and N₂) was in equilibrium with atmospheric values and the bubble release depth was 550 m. All simulations are for non-oily (have a mobile surface) single bubbles with a range in initial equivalent spherical radii of 1000-6000 μm. Three sets of simulations were run for different upwelling velocities; 0, 20, and 30 cm s⁻¹. The upwelling velocity of 30 cm s⁻¹ can be viewed as a maximum velocity, and is based on the lateral distance of the sea surface oil footprints from the seafloor vents⁵. The upwelling velocity of 20 cm s⁻¹ is the measured velocity at the main plume at GC 185⁴. The three sets of simulations are presented in Supplementary Figure 4, and show a range of dissolution depths for a given initial bubble radius with bubbles dissolving closer to the seafloor under no upwelling conditions and bubbles transiting farther distances through the water column with upwelling flow. The upwelling velocity of 20 cm s⁻¹ is the most representative of the plumes examined in this study⁴, and it was assumed that the upwelling flow extends through the water column based on the shoaled thermocline in the submersible dives in comparison to the background CTD casts (Supplementary Figures 5&6) and the relatively narrow plume spreading angle of ~5.2° based on sonar data (Fig. 1). Given the beam spread, the sonar data estimate of the plume-spreading angle is a high estimate.

The results of the simulations presented in Supplementary Figure 4 show that, for an upwelling flow of 20 cm s⁻¹, non-oily bubbles with a radius >2500 μm reach the sea surface, and bubbles with radii >2400 μm reach the summer mixed layer. Bubbles with initial radii less than 1000 μm dissolve at 400 m (150 m above the seafloor). The peak in the bubble size distribution at the main plume at GC 185 is ~2700 μm, and the majority of the emission mass flux is contained in bubbles >4000 μm. Non-oily bubbles of both sizes can reach the summer mixed layer, however the smaller bubbles (<3500 μm) will have lost most of their CH₄ before reaching
the mixed layer while the larger bubbles will have retained a significant fraction of their original methane (I. Leifer, pers. Comm.). Bubbles with an initial radius larger than ~6000 μm will likely break up into smaller bubbles during ascent; as the hydrostatic pressure decreases the bubbles expand. An example of the change in bubble radius during ascent is shown for a bubble with an initial radius of 4000 μm in Supplementary Figure 4b. These simulations are conservative in that they only consider a single bubble. In reality, we sampled a bubble plume that has properties that would act to enhance bubble survival and mediate bubble transfer greater distances through the water column. These new results for single non-oily bubbles are comparable to previous simulations of oily bubbles (immobile surface) that did not consider depth-dependent solubility and compressibility⁵, and show a greatly enhanced vertical methane transport in comparison to previous non-oily bubble models that did not take into account pressure-dependent bubble phenomena⁵. These new results indicate that bubbles between 2400-6000 μm from both non-oily and oily plumes have the potential to reach both the summer and winter mixed layer from deepwater seeps (>500 m) in the Gulf of Mexico.

**Supporting References**


Supplementary Discussion

Potential effects of mixing on water column δ^{13}C-CH_4 profiles

The bottom water δ^{13}C-CH_4 sampled by both submersible and CTDs above all three seep sites ranges from -55.4 to -47‰, indicating that the methane is primarily thermogenic in origin. In all but two of the profiles there is no significant increase in δ^{13}C-CH_4 from the seafloor to the mixed layer, including at background sites >400 m from the bubble plumes. An increase in δ^{13}C-CH_4 from -45.9‰ at the seafloor to -41.9‰ near the sea surface is observed in the submersible profile from the main plume at GC 185 (Fig. 3b). Likewise, an 18‰ enrichment from -44.7‰ at the seafloor to -27.9‰ near the mixed layer is observed in a CTD cast adjacent to the main bubble plume (Fig. 3a). There are two processes that can result in an increase in δ^{13}C-CH_4 from the site of bubble discharge to the mixed layer; mixing and aerobic methane oxidation in the water column.

In order to produce an enrichment in δ^{13}C-CH_4 from linear mixing, the other end member must have a δ^{13}C-CH_4 isotopic composition that is heavier than both the input δ^{13}C value at the vent site and the plume δ^{13}C value measured in the mixed layer. Assuming linear mixing of plume methane with ambient seawater in atmospheric equilibrium with a CH_4 concentration of ~1.7 nM and a δ^{13}C isotopic value of -47.5‰, consistent with the background CTD profiles (Fig. 2a, 3a), the values of -41.9‰ and -27.9‰ observed in the submersible and CTD profiles at GC 185 cannot be produced. Furthermore, the enrichment in δ^{13}C is likely not due to mixing with other thermogenic sources as other vent gases measured at GC184 and GC 234 range from -49‰ to -44‰ and values greater than these are not observed in the background profiles (Fig. 3a). Also, while some seep sites in the northern GOM emit thermogenic CH_4 into the water column (such as the seeps examined in this study), the majority of the seeps contain primarily
deep-sourced, microbially derived methane with δ^{13}C values average -65‰ to -80‰. Linear mixing of the thermogenic plume CH4 with background seep-derived biogenic CH4 averaging -73‰ would produce depleted values rather than the 4-18‰ enrichment observed. Even if 99.9% of the input flux of biogenic seep derived CH4 was oxidized, the δ^{13}C composition of the methane would be -54.5‰ (computed using model in the methods section). This value is more negative than the δ^{13}C-CH4 emitted to the water column from the vents sampled in this study. Based on these points, we conclude that mixing cannot produce the enriched δ^{13}C-CH4 values, up to -27.9‰, measured at GC 185, and that they can only be produced by microbial aerobic methane oxidation in the water column.

The δ^{13}C-CH4 data do not rule out mixing in the water column, but rather show that mixing cannot produce the isotopically enriched δ^{13}C-CH4 values observed in the main plume at GC 185 and that they are the result of oxidation. There is some evidence for mixing in the lower water column and within the mixed layer based on the δ^{13}C-CH4 profile collected by submersible in the secondary plume at GC 185 (Fig. 3b). The bottom water δ^{13}C-CH4 in the secondary plume is -55.4‰, significantly lighter than the other plumes, indicating a stronger signature of biogenic methane. This plume also has a slower input flux compared to the main plume at GC 185 based on repeated submersible dives at this site. In the first ~50 m above the seafloor, the δ^{13}C-CH4 in the secondary plume increases to values observed in the main plume, indicating some lateral advection of the site GC 185 bottom water and mixing between the two plumes. Above this depth, the δ^{13}C-CH4 in the secondary plume becomes lighter and more representative of the values immediately above the vent. Within the mixed layer, the δ^{13}C-CH4 becomes enriched as a result of mixing with thermogenic methane from the main plume (Fig. 3a). The increase in δ^{13}C-CH4 in the secondary plume cannot be produced by mixing with ambient seawater in
atmospheric equilibrium (1.7 nM and -47.5‰) as the mixed layer methane concentration above the secondary plume is ~500 nM or ~300 times that of ambient seawater. Thus, the δ^{13}C-CH₄ data from the secondary plume do suggest some mixing in the water column, however they indicate that it is mainly concentrated in the bottom 50 m of the water column and in the mixed layer, and that mixing of methane is primarily between plumes. Because mixing is only observed in the secondary plume and not in the main plume, it does not have a significant effect on our estimates of CH₄ oxidation in the main plume.

**Supporting References**


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Supporting References


Supplementary Table 1 | Estimates of CH$_4$ fluxes to the atmosphere from different marine environments

<table>
<thead>
<tr>
<th>Location</th>
<th>CH$_4$ Flux ($\mu$mol m$^{-2}$ d$^{-1}$)</th>
<th>Water Depth (m)</th>
<th>Type of Flux$^a$</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shallow water (&lt;200 m)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Paleo Dnepr Area, Black Sea</td>
<td>200</td>
<td>90</td>
<td>max. diffusive</td>
<td>1</td>
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<tr>
<td>Dnepr Shelf, Black Sea</td>
<td>66</td>
<td>&lt;200</td>
<td>max. diffusive</td>
<td>1</td>
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<tr>
<td>NW Black Sea</td>
<td>53</td>
<td>&lt;200</td>
<td>max. diffusive</td>
<td>2</td>
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<tr>
<td>Coal Oil Point$^b$</td>
<td>1296</td>
<td>&lt;70</td>
<td>diffusive</td>
<td>3</td>
</tr>
<tr>
<td>Coal Oil Point</td>
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<td>&lt;80</td>
<td>bubble</td>
<td>4</td>
</tr>
<tr>
<td>Coal Oil Point</td>
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<td>&lt;80</td>
<td>bubble</td>
<td>5</td>
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<tr>
<td>Deep Water (&gt;200 m)</td>
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<td></td>
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<tr>
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<td>0.9-3.5</td>
<td>&gt;1000</td>
<td>diffusive</td>
<td>6</td>
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<tr>
<td>Subtropical North Pacific</td>
<td>1.6</td>
<td>&gt;1000</td>
<td>diffusive</td>
<td>7</td>
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<tr>
<td>Sargasso Sea</td>
<td>4.4</td>
<td>&gt;1000</td>
<td>diffusive</td>
<td>7</td>
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<tr>
<td>Sea of Okhotsk$^c$</td>
<td>1.5-63</td>
<td>100-700</td>
<td>diffusive</td>
<td>8</td>
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<tr>
<td>Arabian Sea</td>
<td>4.6-13.9</td>
<td>&gt;1000</td>
<td>diffusive</td>
<td>9</td>
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<tr>
<td>Sorokin Trough, Black Sea</td>
<td>49</td>
<td>2080</td>
<td>max. diffusive</td>
<td>1</td>
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<tr>
<td>Black Sea</td>
<td>27</td>
<td>basin average</td>
<td>diffusive</td>
<td>10</td>
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<tr>
<td>Gulf of Mexico hydrocarbon plumes</td>
<td>200-10,500</td>
<td>500-600</td>
<td>diffusive</td>
<td>This Study</td>
</tr>
</tbody>
</table>

$^a$The type of flux is split into two categories. A bubble flux is an advective flux across the sea surface. A diffusive flux is that from the oceanic mixed layer to the atmosphere with max. indicating the maximum flux estimated during the respective study. $^b$The diffusive flux estimate is from an area 7 km downstream of the focused vents at Coal Oil Point. $^c$Range includes both deep and shallow water seep areas.

Supporting References


**Supplementary Table 2** Average daily wind speeds and calculated gas transfer coefficients, $k_w$

<table>
<thead>
<tr>
<th>Plume Experiments</th>
<th>Sampling Date (mm/dd/yy)</th>
<th>Avg. Wind Speed ($m s^{-1}$) ± 1σ</th>
<th>$k_w$ avg</th>
<th>$k_w$ max</th>
<th>$k_w$ min</th>
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<tbody>
<tr>
<td><strong>Submersible Sampling</strong></td>
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<td></td>
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<tr>
<td>GC 185 Main Plume</td>
<td>08/15/03</td>
<td>10.5 ± 2.7</td>
<td>42.6</td>
<td>67.3</td>
<td>23.5</td>
</tr>
<tr>
<td>GC 185 Main Plume</td>
<td>08/14/03</td>
<td>7.8 ± 0.8</td>
<td>23.5</td>
<td>28.6</td>
<td>18.9</td>
</tr>
<tr>
<td>GC 185 Secondary Plume</td>
<td>08/14/03</td>
<td>7.8 ± 0.8</td>
<td>23.5</td>
<td>28.6</td>
<td>18.9</td>
</tr>
<tr>
<td>GB 425 Mud Volcano</td>
<td>08/21/03</td>
<td>3.0 ± 1.3</td>
<td>3.5</td>
<td>7.1</td>
<td>1.1</td>
</tr>
<tr>
<td>GC 234 Main Plume</td>
<td>08/23/03</td>
<td>5.9 ± 1.6</td>
<td>4.2</td>
<td>8.5</td>
<td>1.4</td>
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<tr>
<td>GC 185 Main Plume 2002</td>
<td>06/11/02</td>
<td>4.9 ± 2.1</td>
<td>9.3</td>
<td>18.9</td>
<td>3.0</td>
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<td><strong>CTDs</strong></td>
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<tr>
<td>GC 185 Main Plume</td>
<td>08/14/03</td>
<td>7.8 ± 0.8</td>
<td>23.5</td>
<td>28.6</td>
<td>18.9</td>
</tr>
<tr>
<td>GC 185 Main Plume</td>
<td>08/17/03</td>
<td>2.1 ± 1.3</td>
<td>1.7</td>
<td>4.5</td>
<td>0.2</td>
</tr>
<tr>
<td>GC 185 Main Plume</td>
<td>08/15/03</td>
<td>10.5 ± 2.7</td>
<td>42.6</td>
<td>67.3</td>
<td>23.5</td>
</tr>
<tr>
<td>GC 185 Background</td>
<td>08/19/03</td>
<td>1.5 ± 1.1</td>
<td>0.9</td>
<td>2.6</td>
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<td>2.2 ± 1.3</td>
<td>1.9</td>
<td>4.7</td>
<td>0.3</td>
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<tr>
<td>GC 234 Main Plume</td>
<td>08/21/03</td>
<td>3.0 ± 1.3</td>
<td>3.5</td>
<td>7.1</td>
<td>1.1</td>
</tr>
<tr>
<td>GC 234 Background</td>
<td>08/22/03</td>
<td>2.0 ± 1.2</td>
<td>1.5</td>
<td>4.0</td>
<td>0.2</td>
</tr>
</tbody>
</table>
Supplementary Figure 1 | Manifestations of the prolific hydrocarbon flux through the seafloor in the northern Gulf of Mexico.  

a, Seafloor gas hydrate outcrop at GC 185. The gas hydrate is in direct contact with bottom water requiring a continuous supply of CH₄ from below. The gas hydrate is orange due to the presence of oil, and the outcrop is covered with a thin veneer of sediment. Perennial bubble plumes are emitted to the water column adjacent to the outcrop as shown in the right panel.  
b, Picture taken at the seafloor of the main bubble plume at GC 185. Bubbles are emitted to the water column from a seafloor vent ~1 m from the gas hydrate outcrop shown in a. The picture was taken from the Johnson Sea-Link research submersible. The submersible’s manipulator is holding a white panel behind the plume to aid in bubble emission descriptions. The diameter of the plume at the seafloor is ~60 cm.
Supplementary Figure 2 | Traces of perennial oil slicks at the sea surface above GC 185 and GC 234. Traced oil slicks from SAR images in Mineral Management Service Lease Blocks GC 185 (“Bush Hill”) and GC 234, as well as GC 233 (“Brine Pool”) from July 2001 and June-July 2002. The 2001 dates are in red and the 2002 dates are in black. Note that the oil slicks at all three sites are present in each acquired scene, thus are perennial. Figure is modified from Ref. 1.

Supporting References

Supplementary Figure 3 | Numerical non-pressure dependent model simulations of bubble radius changes and dissolution depths for the oily plumes at GC 185. a, Bubble dissolution depth. 500 m = the seafloor and 0 m = the sea surface. b, Normalized ratio of final to initial bubble radius at the sea surface for different initial bubble radius, r, for bubbles with oil as a surfactant. Figure is modified from Ref. 1., and the model is described in Ref. 2. In Case 1, an upwelling velocity of 10 cm s\(^{-1}\) was assumed to extend through the water column\(^1\), and the dissolved CH\(_4\) concentration was equivalent to 0.01 atm. Case 2 had no upwelling flow and the same dissolved CH\(_4\) concentration. Case 3 had no upwelling flow and a dissolved CH\(_4\) concentration of 0.25 atm\(^1\). In the model runs\(^1\), a broader range of initial bubble sizes made it to the sea surface in the oily plumes because the contaminated bubbles have an immobile surface, which causes them to exchange gas at a slower rate than the clean bubbles. The results of the model for Case 1, the most representative of the plumes examined in this study (Supp. Table 1), all bubbles with an initial radius >4.5 mm rise all the way through the water column to the sea surface (a). Bubbles with an initial radius between 4.5 and 6 mm have the greatest potential to make it to the mixed layer, because they surface without significantly changing size (e.g. b; \(r_{\text{final}}/r_{\text{initial}} \sim 0.8\) to 1.5). The model used for these simulations is a simplified version of the numerical model presented in the Supplementary Method. The model did not include pressure-dependent effects including compressibility, depth-dependent solubility, depth-dependent effects on bubble rise, and hydrate skins within the hydrate stability field. Thus, though this model was inappropriate for the deepwater plumes in this study, it is useful for demonstrating that excluding factors that enhance bubble-mediated methane transfer through the water column, oily bubbles >4.5 mm reach the sea surface. Taking into account depth-dependent effects, non-oily bubbles >4 mm are also predicted to reach the mixed layer and sea surface (Supplementary Method, Supplementary Figure 4).

Supporting References


Supplementary Figure 3| Numerical non-pressure dependent model simulations of bubble radius changes and dissolution depths for the oily plumes at GC 185. 

a, Bubble dissolution depth. 500 m = the seafloor and 0 m = the sea surface. 

b, Normalized ratio of final to initial bubble radius at the sea surface for different initial bubble radius, r, for bubbles with oil as a surfactant. Figure is modified from Ref. 1. The model is described in Ref. 2. In Case 1, an upwelling velocity of 10 cm s\(^{-1}\) was assumed to extend through the water column, and the dissolved CH\(_4\) concentration was equivalent to 0.01 atm. Case 2 had no upwelling flow and the same dissolved CH\(_4\) concentration. Case 3 had no upwelling flow and a dissolved CH\(_4\) concentration of 0.25 atm. In the model runs, a broader range of initial bubble sizes made it to the sea surface in the oily plumes because the contaminated bubbles have an immobile surface, which causes them to exchange gas at a slower rate than the clean bubbles. The results of the model for Case 1, the most representative of the plumes examined in this study (Supp. Table 1), all bubbles with an initial radius >4.5 mm rise all the way through the water column to the sea surface. Bubbles with an initial radius between 4.5 and 6 mm have the greatest potential to make it to the mixed layer, because they surface without significantly changing size (e.g. \(\text{radius}_{\text{final}} / \text{radius}_{\text{initial}} \approx 0.8\) to 1.5). The model used for these simulations is a simplified version of the numerical model presented in the Supplementary Method. The model did not include pressure-dependent effects including compressibility, depth-dependent solubility, depth-dependent effects on bubble rise, and hydrate skins within the hydrate stability field. Thus, though this model was inappropriate for the deepwater plumes in this study, it is useful for demonstrating that excluding factors that enhance bubble-mediated methane transfer through the water column, oily bubbles >4.5 mm reach the sea surface. Taking into account depth-dependent effects, non-oily bubbles >4 mm are also predicted to reach the mixed layer and sea surface (Supplementary Method, Supplementary Figure 4).

Supporting References


Supplementary Figure 4| Numerical pressure-dependent model simulations of dissolution depths and bubble radius change rates for non-oily bubbles emitted at 550 m water depth. 

a, Bubble dissolution depth versus initial equivalent spherical radius for upwelling velocities (\(V_{\text{up}}\)) of 0, 20, and 30 cm s\(^{-1}\). The upwelling velocity of 20 cm s\(^{-1}\) is the measured rate at the main gas vent at GC 185. 

b, Numerical simulation of the variation in bubble size of a non-oily bubble with an initial radius of 4000 \(\mu\)m over 1250 s (550 to 100 m water depth).
Supplementary Figure 5] Temperature and salinity profiles collected during three submersible transects from the seafloor to the sea surface immediately adjacent to the bubble plumes at GC 185 and from a “background” CTD cast. a, Main plume transect 1, b, Main plume transect 2, c, Secondary plume transect, and d, GC 185 CTD cast. The highest bottom water and mixed layer methane concentrations were observed in the Main Plume Transect 2 and the Secondary Plume Transect. In Main Plume Transect 2, bottom water is transported upwards with the bubbles as observed in the near constant temperature from 550-420 m (b). In the Main Plume Transect 2 and Secondary Plume Transect, the thermocline is locally shallower by ~20-30 m in comparison to the background CTD casts (d and Supplementary Figure 6), indicating vertical transport of water to the mixed layer (b,c).
Supplementary Figure 6 | Temperature and salinity profiles collected from background CTD casts during the August 2003 field season at a, GC 234 and b, GB 425. The temperature profiles are in blue and the salinity profiles are in red. These profiles are typical of mid-summer conditions at each site.
**Supplementary Figure 7**| Density and dissolved oxygen profiles collected by background CTD casts. The profiles were collected during August 2003 field season at a, GC 185, b, GC 234, and c, GB 425. These profiles are typical of the mid-summer conditions at each site.