



Three-dimensional acoustic monitoring and modeling of a deep-sea CO₂ droplet cloud

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[1] We show that release of 5 liters of liquid CO₂ at 1000 m depth can be readily detected acoustically, and tracked for over 30 minutes, and 150 m of ascent, with both surface ship (38 kHz) and ROV (675 kHz) sonars. The released liquid broke up into droplets covered with a hydrate film. The remarkably sensitive acoustic response of the droplets may be attributed to the high sound speed contrast between CO₂ (300 m/sec) and sea water (1500 m/sec), the near spherical shape of the droplets created by the hydrate shell, and the high compressibility of the liquid. The observed cloud conformed closely to models of CO₂ disposal, allowing for reasonable predictions of larger scale processes. This offers a remarkably sensitive technique for examination in real time of engineered releases of CO₂, volcanic sea floor liquid CO₂ plumes, or leakage from geologic CO₂ storage. **Citation:** Brewer, P. G., B. Chen, R. Warzinski, A. Baggeroer, E. T. Peltzer, R. M. Dunk, and P. Walz (2006), Three-dimensional acoustic monitoring and modeling of a deep-sea CO₂ droplet cloud, *Geophys. Res. Lett.*, 33, L23607, doi:10.1029/2006GL027181.

1. Introduction

[2] Scientific and public attitudes towards the role of the ocean in taking up fossil fuel CO₂ are confused. It has been recognized for decades that the ocean, through its alkalinity, “acts as a giant regulator of carbon dioxide” [Callendar, 1938]; the surface ocean now absorbs 30% of all fossil fuel CO₂ emissions [Sabine et al., 2004], and the accumulated fossil fuel CO₂ burden is now about 500 Gt CO₂. Without the benefit of massive absorption of this artifact of mankind’s energy use the world would face an insurmountable atmospheric CO₂ problem. Ocean uptake of fossil fuel CO₂ by the surface ocean has therefore typically been described as a beneficial natural process, quite distinct from the possible direct injection of CO₂ into oceanic deep waters as a means of CO₂ sequestration [Brewer et al., 1999; Intergovernmental Panel on Climate Change (IPCC), 2005], with concerns over environmental harm [Seibel and Walsh, 2001]. So strong has been the divide that while terrestrial CO₂ enrichment experiments releasing up to

10³ tons CO₂/year routinely take place [DeLucia et al., 1999], oceanic experiments with the release of only a few kilograms of CO₂ have been subject to enormous scrutiny, yet there are compelling needs for carrying out a wide variety of such experiments.

[3] With the belated recognition that the scale of surface ocean CO₂ invasion is now so large that changes in sea water pH of 0.3 or more are predicted to occur by mid-century [Brewer, 1997; Caldeira and Wickett, 2003], the lines between so called “passive” and possible “direct” introduction of CO₂ to the ocean have become blurred. Stabilization of atmospheric CO₂ levels at 550 ppmv is often mentioned as a desirable policy goal, and emission trajectories for achieving this have been described [Wigley et al., 1996]. Yet this implies at equilibrium the transfer of some 6 trillion tons of CO₂ to the ocean over the course of several centuries [IPCC, 2005]. The consequences of this for marine ecosystems are largely unknown [Cicerone et al., 2004; Royal Society, 2005], but are generally expected to be negative [Portner et al., 2004]. While simple aquarium studies can shed some light on potential impacts, open ecosystem perturbation studies will also be required.

[4] Here we describe advances in one such experimental technique, that of observing a freely released CO₂ cloud, such as might occur from engineered releases of CO₂ directly into the deep ocean, leakage of CO₂ from sub-sea floor geologic disposal sites, or from natural vents.

[5] Liquid CO₂ is a highly compressible fluid, with the property of being less dense than sea water at depths less than ~2500 m, and more dense at greater depths. Thus gravitationally stable pools of liquid CO₂ may be placed on the deep sea floor for experimental purposes [Brewer et al., 1999], and these have been used to investigate the perturbed pH field and biological responses around an experimental site [Barry et al., 2004]. Equivalent experiments in shallower waters, where CO₂ forms a rising cloud, have been considered very difficult to execute and plans for medium scale experiments have lead to controversy [Figueiredo et al., 2003]. Yet it is this scenario that is of great interest, for it simulates the release of liquid CO₂ from volcanic sites [Sakai et al., 1990; Lupton et al., 2006] and relates to possible leakage of CO₂ from sub-seafloor geological storage sites. The assessment of ocean ecosystem impacts at such sites has recently been identified by the International Energy Agency’s Greenhouse Gas Programme as requiring a critical study.

[6] The difficulty of observing a freely released rising CO₂ cloud is such that only very small scale observations of individual rising droplets [Brewer et al., 2002] have been reported so far. These few observations have been used as input for sophisticated models [Alendal and

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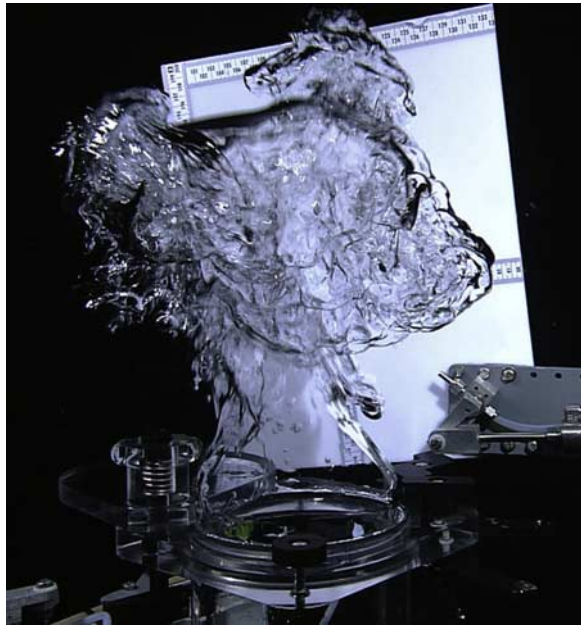


Figure 1. Image of the 1025 m \sim 5 L liquid CO₂ experiment at the moment of release. The container volume is 7.5 L, diameter is 15 cm. The white panel with the scale is for size estimation and to provide visual contrast for imaging the transparent fluid. Immediately after release the buoyant liquid begins to break down into small \sim cm size globules due to Taylor instabilities. The ensemble of droplets of varying sizes ascends as a dissolving complex cloud, with the rise rate of individual droplets depending upon size, and the changing density with the surrounding sea water.

Drange, 2001; Chen et al., 2003; Sato, 2004; Gangstø et al., 2005] of larger scale CO₂ clouds, but no observational constraints have yet been available to guide and test model development.

2. Experiments

[7] Our experiments took place in November 2005 at a location 55 miles due west of Moss Landing, CA. We contained and delivered the liquid CO₂ as described by *Peltzer et al. [2004]*, using the MBARI ROV Tiburon for experimental control and data acquisition with the ROV mounted Simrad 1000 675 kHz sonar. The ROV was positioned directly under the ship's hull for data acquisition with the downward looking Simrad EK 500 38 kHz sonar, which had a maximum signal acquisition depth of about 1,000 m. Liquid CO₂ was dispensed into an acrylic "detritus sampler" of about 7.5 liter volume, 15 cm diameter, with an open bottom for introduction of the buoyant fluid, and a closed lid that could be opened under hydraulic control (Figure 1). Care was taken to minimize hydrate formation by turbulent mixing during the CO₂ loading step. The rapid opening of the sampler lid allowed the CO₂ to escape (1,000 m depth, 3.9°C, 34.370 salinity) with minimal resistance. The fluid began at once to break up due to Taylor instabilities (Figure 1), and no attempt was made to control droplet size.

[8] The rapidly rising, divergent, transparent fluid cloud is impossible to track visually and invasion of the cloud by the vehicle disturbs the signal. Larger globules of liquid rise faster than small droplets giving the cloud a changing and unknown vertical ascent characteristic; and the cloud is undergoing horizontal forcing from ocean currents as it rises. The dissolution rate is slow enough [*Brewer et al., 2002*] that over the cloud lifetime it may experience quite varied physical forcing, and changing temperature, salinity, and thus density conditions. It is commonly assumed that pH sensing may be used to track a CO₂ plume, but for small releases the signal is undetectable, and slow dissolution and hydration kinetics [*Zeebe and Wolf-Gladrow, 2001*] greatly exacerbate the problem and thus some remote sensing technique is called for. In a small-scale pilot experiment carried out in late 2004 with the ROV Ventana we first recognized the remarkably sensitive acoustic detection of liquid CO₂ droplets but were unable at that time to track the cloud and constrain the experiment.

[9] The adopted strategy for tracking our moving invisible target was first to back-up the vehicle so that the rising cloud was several meters in front as detected by the scanning 675 kHz ROV sonar with a $\pm 15^\circ$ vertical cone. The ROV was then positioned about 10 m away from the cloud (Figure 2, top) and held stationary, thus giving the changing horizontal dimensions. Once the sonar signal indicated that the cloud had risen beyond view the ROV was piloted upwards, and positioned immediately above the cloud (the point at which no sonar signal was detected) and held stationary as the cloud moved up and out of view again. This yielded rise rate, and what are essentially a series of horizontal slices through the vertically moving cloud.

[10] At a depth of 1000 m both the CO₂ cloud, and the echo of the ROV, were detectable by the downward looking 38 kHz shipboard sonar (Figure 3a). This provided confirmatory response of the relative cloud-ROV positions, and also gave an indication of the cloud vertical extent at all

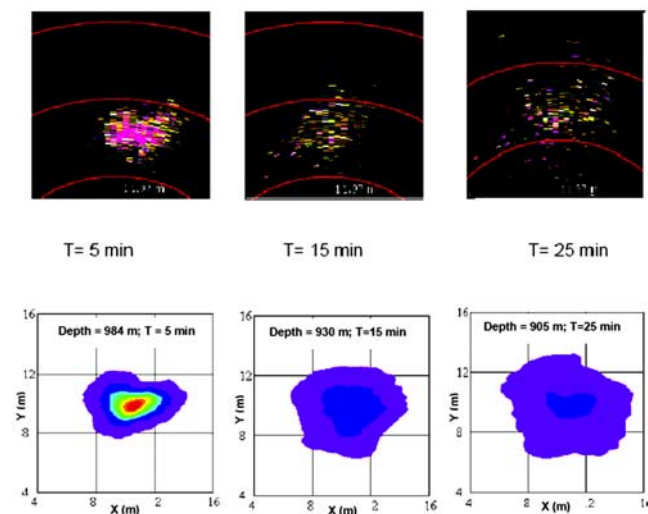


Figure 2. Comparison of the ROV borne 675 kHz sonar signal of horizontal slices through the rising cloud at 5, 15, and 25 minutes after release with a (top) numerical model [*Chen et al., 2003*] and (bottom) with grid scales in meters.

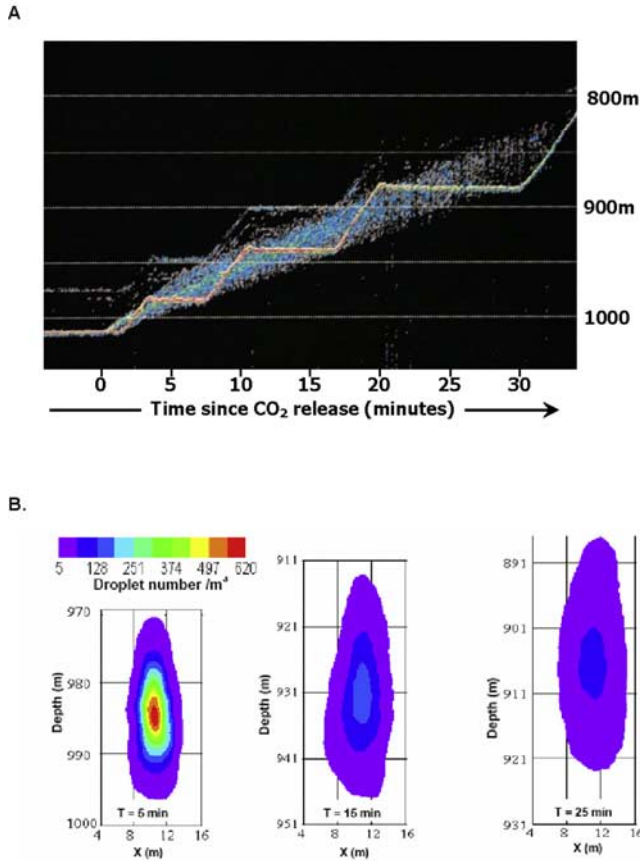


Figure 3. (a) Time series sonar image obtained from the downward looking Simrad EK 500 38 kHz system. The initial cloud release is at 1,000 m depth. The sharp, linear “stair case” return is from the ROV itself, the diffuse cloud is the return from the ascending CO₂ droplets. The vehicle itself is near-stationary in the horizontal, and the image is akin to a chart recording of the experiment. The observing strategy was to immediately back the vehicle away from the point of release so as to avoid disturbance of the cloud, and maintain 10–20 m horizontal separation. The vehicle was then flown above the cloud and then kept stationary while the cloud passed by; the sequence was then repeated. The cloud was detectable from the surface ship for over 30 minutes, and through 150 m of ascent. (b) Numerical model of the vertical extent of the cloud [Chen *et al.*, 2003] at 5, 15, and 25 minutes after release.

times. By combining both sonar data sets a three-dimensional picture in real time was obtained.

3. Results

[11] We were able to track the droplet cloud from the 1000 m CO₂ release for over 30 minutes and over 150 m of ascent (Figure 3a), giving an average ensemble rise rate for the cloud of about 5 m/min. This may be compared with the rise rate of from 6–9 m/min for the individual droplets measured by Brewer *et al.* [2002] using an imaging box technique and visual tracking. The difference is attributable to the different environmental conditions, release depths, and thus CO₂ physical properties. In the Brewer *et al.*

[2002] study the release point was 804 m depth ($T = 4.398^{\circ}\text{C}$, $\rho_{\text{CO}_2} = 0.9423 \text{ g/cc}$) and observation was lost at 341 m depth ($T = 7.291^{\circ}\text{C}$, $\rho_{\text{CO}_2} = 0.8632 \text{ g/cc}$); the greater depth, and thus ocean and liquid CO₂ densities account for the slower rise rate. Changes in external water CO₂ chemistry between the two experiments are not significant in controlling the result.

4. Discussion

4.1. Comparison With Laboratory Studies

[12] The behavior of the droplets formed from the 5-L release has similarities to that observed for individual droplets studied in the laboratory under similar conditions in a high pressure tunnel [Haljasmaa *et al.*, 2005]. In the water tunnel experiments droplets much larger than 10 mm diameter were observed to shed CO₂ until approximately this diameter was reached, and rise rates of about 0.13 m/s were observed. Formation of a hydrate shell was not observed in the water tunnel experiments, whereas this is ubiquitous in natural sea water. Based upon the water tunnel experiments a dissolution rate of $3.5 \mu\text{mol/cm}^2/\text{s}$ is predicted at 1000-m depth and 4.1°C .

4.2. Comparison With Numerical Models

[13] We performed two numerical simulations of the 5-L release, one by a single CO₂ droplet simulation incorporating dissolution rates and buoyancy forces calibrated by laboratory and field studies to determine the initial sizes of droplets, and another by a turbulent two-phase cloud model [Chen *et al.*, 2003] to predict droplet cloud dynamics.

[14] The early single droplet observations [Brewer *et al.*, 2002] were described by a very simple spherical buoyancy equation applicable for a drag coefficient of 1:

$$u = 8gr(\rho_{sw} - \rho_{CO_2})/3\rho_{CO_2} \quad (1)$$

Where u is the terminal rise velocity, g is gravity, r is the droplet radius, and ρ_{sw} and ρ_{CO_2} are the changing in situ densities of seawater and liquid CO₂ respectively. This approach was criticized [Zhang, 2005], but strongly supported in a rebuttal by Alendal *et al.* [2006]. The droplet sub-model used here for the slip velocity (u) and diameter-shrinking rate (\dot{d}) is of the form:

$$\dot{u}_c = \frac{\rho_s}{\rho_c} \left[\left(1 - \frac{\rho_c}{\rho_s} \right) g - \frac{3u_c^2}{4d_c} C_d \right] - u_c \frac{\dot{m}_c}{m_c} \quad (2)$$

$$\dot{d}_c = -\frac{1}{\rho_c} \left(\frac{d_c}{3} \dot{\rho}_c + \frac{2Sh_e D_f (C_{cs} - C_s)}{d_c} \right) \quad (3)$$

Where m , ρ , and C are the mass of the droplet, the density, and the CO₂ concentration respectively. The subscripts c , cs , and s indicate CO₂, CO₂ droplet surface, and seawater respectively. Sh_e and C_d are the effective Sherwood number and effective drag coefficient respectively. The dotted symbols are derivatives with respect to time. The last two parameters play a key role in the prediction of droplet dissolution and rising velocity. Here the experimental data based values of Sh_e and C_d given by Chen *et al.* [2003] are used in the simulations.

[15] Field and laboratory observations, and model predictions, show that the droplets formed rapidly after free liquid release range from 8 to 10 mm diameter. Over the observed CO₂ cloud ascent from the release depth of 1,000 m to 850 m (4.8°C) we calculate from the droplet sub-model that a droplet with an initial diameter of 10 mm will decrease in size to 4.7 mm, and the rise velocity will decrease from 0.11 m/s to 0.08 m/s. A droplet with an initial 8 mm diameter will decrease to 1 mm, and the velocity decrease is from 0.10 m/s to 0.02 m/s.

[16] We use droplet number density (number/m³) to visualize the CO₂ cloud dynamics from the two-phase modeling simulations. With initial diameters of 8–10 mm set randomly droplets disperse horizontally due to local water velocities, while rising vertically (Figure 3b). Our simulations of vertical sections are well matched to the sonar images for up to 15 minutes from release not only for the height of the CO₂ cloud but also for cloud width. The simulated cloud rises slowly with a slightly lower velocity in comparison with that detected by sonar at 25 minutes. This may be due to a downward flow predicted in the model by locally dense CO₂ enriched sea water [Chen *et al.*, 2005]. Because of a relatively smaller computation domain in the horizontal (20 m × 20 m) than the vertical (200 m), the open boundary conditions at top and bottom make the modeled flow field sensitive to the small negative buoyancy effect. This predicted downward flow developed to a detectable level in comparison with the droplet rise velocities after about 20 minutes, when CO₂ dissolution caused the small droplets to rise with lower velocity. In the real ocean no significant down-welling was observed, since the small size of the release produced negligible local density increases. We note that in earlier published work [Brewer *et al.*, 2002] a small programming error in the simple model used lead to a slight exaggeration of rise rate for small droplets late in the plume development.

[17] For modeled horizontal sections simulations agreed well with the ROV sonar record with modeled and observed scales of 12 × 12 m² in cloud/droplet areas. The cloud dilutions detected by the ROV sonar correspond to simulations with maximum droplet number densities of 620, 160, and 80 at each sampled time (5, 15, and 25 minutes) respectively.

5. Conclusions

[18] While direct comparisons with acoustic detection of the more commonly observed methane gas bubble clouds [Heeschen *et al.*, 2003] are difficult, there is every indication of extraordinarily sensitive acoustic detection for a liquid CO₂ release within the hydrate formation zone, and we may ask why this should be so. Although methane gas clouds form a hydrate skin, observations [Rehder *et al.*, 2002] suggest that the tensile strength of the film is not sufficient to completely overcome the distortion of the rising bubble shape due to buoyancy forces. The greater density and thus lower rise rate of liquid CO₂ droplets reduces the buoyancy, and the tensile strength of the hydrate film, ≈0.1 N/m under these conditions [Yamane *et al.*, 2000], is sufficient to overcome distortions and produce a near perfect spherical shape of high velocity contrast, creating a highly efficient acoustic scatterer. There is excellent agreement between

models and field work, suggesting that quite accurate predictions can be made of the fate of CO₂ clouds in the deep ocean. The results suggest that very sensitive detection of small amounts of liquid CO₂ leaking from the sea floor may be detected acoustically, but that rapid dissolution of the cloud results in signal detection only within about 150 m of vertical ascent from the source. The experiment reported here was of small enough scale that no significant seawater density changes resulted from CO₂ dissolution, and at the frequencies used here acoustic detection of the dissolved CO₂ is not possible. However even quite subtle density changes in sea water may be detected at lower frequencies [Holbrook *et al.*, 2003], and thus for larger releases detection of the near-field dissolved plume may be achievable. Since the use of acoustics for biological detection is well established, this tool also offers a unique way to detect water column biological responses to liquid CO₂ released from the sea floor. We were not able to measure directly the change in pH field resulting from such a small release, and that remains a challenge for future work.

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