Tests of Acoustic Target Strength and Bubble Dissolution Models Using a Synthetic Bubble Generator

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ABSTRACT

To test methods used for converting observations of acoustic backscatter to estimates of the volume and transport of free gas escaping the seabed, a bubble generator has been constructed and used at sea. The bubble generator creates individual bubbles of the sizes commonly associated with methane seeps, 1–5-mm radii, which can be released at preplanned rates. The bubble generator was deployed off the coast of New Hampshire at a depth of 55 m, and acoustic backscatter between 16 and 24 kHz was collected from a shipboard echo sounder while transiting over the rising bubbles. Bubble sizes and compositions (either Ar or N₂) were known at the source. A model for bubble evolution, accounting for changes in bubble size and composition due to hydrostatic pressure and gas diffusion across the gas–liquid boundary, was coupled with an acoustic target strength (TS) model to generate predictions of the acoustic backscatter from bubbles that had risen to different depths. These predictions were then compared with experimental observation. Good agreement between prediction and observation was found in most cases, with the exception of the largest (4 mm) gas bubbles at depths of 30 m or less. The exact cause of this bias is unknown, but may be due to incorrect assumptions in models for the bubble TS, rise velocity, or mass transfer rate.

1. Introduction

Methane gas bubbles have been found escaping from the seabed throughout the world's oceans (Judd 2004). Once in the water column, rising methane gas bubbles can lose methane to dissolution into the seawater, where it may become oxidized to CO₂ (Valentine et al. 2001), or may directly reach the atmosphere (Rehder et al. 2002; Mau et al. 2007). Understanding the fate of this seabed-sourced methane on atmospheric methane and the global carbon cycle in general, requires knowledge of the location and number of methane gas seeps, the size of the gas bubbles, and rate at which gas is transferred between gas bubbles and the surrounding ocean waters.

Acoustic methods are often used to detect, quantify, and monitor seeps of methane gas bubbles (Merewether et al. 1985; MacDonald et al. 2002; Heeschen et al. 2003; Greinert et al. 2006; Schneider von Deimling et al. 2011; Römer et al. 2012; Kannberg et al. 2013; Jerram et al. 2015). Ambiguities between the size and number of gas bubbles in narrowband acoustic observations are often resolved using direct capture techniques (Weber et al. 2014) or optical imaging techniques (Leifer et al. 2003; Wang et al. 2016) at or near the seabed. Low emission rates, high-resolution broadband acoustic techniques, or a combination of the two that makes it possible to identify individual bubbles offers the opportunity to invert observations of acoustic target strength (TS) for bubble size (Weidner et al. 2019). When the bubble density is too high to resolve individuals, broadband techniques that capture the bubble's natural frequencies can be used to invert for bubble size distribution and, ultimately, void fraction (Römer et al. 2012; Weber et al. 2014; Wang et al. 2016). In either inversion scenario, it is common to use acoustic scattering models that assume

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the bubble is spherical (Weber et al. 2014; Anderson 1950; Jech et al. 2015). Bubbles greater than approximately 1 mm in radius, however, take on irregular shapes that are more closely approximated by oblate spheroids than by spheres (Clift et al. 1978). One of the objectives of the present work is to develop and use an experimental technique where the error in the assumption of spherical bubbles can be assessed.

In addition to localizing and quantifying methane gas bubble seepage at the seafloor, it is important to understand the evolution and ultimate fate of the gas bubbles. As a bubble rises through the ocean, decreasing hydrostatic pressure acts to increase the bubble size, a tendency that is sometimes in competition with the exchange of the bubble's gas constituents with surrounding water. Gas transport across the gas–liquid boundary can occur in either direction (into or out of the bubble) according to Henry's law. Models describing the changing size of rising bubbles in response to the competing process of gas dissolution and reducing hydrostatic pressures generally specify the bubble as either “clean” or “dirty” to model the rate of gas transfer (Leifer and Patro 2002; McGinnis et al. 2006; Gros et al. 2016, 2017; Socolofsky et al. 2015). The gas transfer rate for clean bubbles (Levich 1962) acts as an upper bound, and this rate is reduced by surfactants and other material that immobilizes the gas–liquid boundary (Clift et al. 1978). Methane bubbles within the deep ocean (i.e., at depths and temperatures where methane hydrate can be formed) form a hydrate coating that immobilizes the bubble boundary (Rehder et al. 2002), suggesting that a “dirty” bubble gas transfer rate is appropriate. Above the hydrate stability zone, a “clean” bubble gas transfer rate may not always be appropriate, as evidenced by observations of bubbles composed of other gases and it is possible that even a “dirty” bubble prediction may overpredict the rate of bubble dissolution (Johnson and Cooke 1981; Weber et al. 2005). A second objective of the present work is to develop an experimental method by which gas bubble evolution models can be assessed in a variety of environments where the seawater may have different levels and types of surfactants and particulate matter.

To examine both bubble evolution and acoustic scattering from bubbles, a synthetic bubble generator has been designed and constructed. The synthetic bubble generator precisely controls the size and rate of bubbles generated per second, 0.01–10 Hz, and can be used with a variety of gases (e.g., air, Ar, N₂). The system creates individual bubbles at sizes between 1 and 5 mm, within the most common range of bubble sizes found at natural methane seeps (Römer et al. 2012; Weber et al. 2014; Wang et al. 2016; Leifer and MacDonald 2003).

The bubble generator is preconfigured to create bubbles at selected rates and sizes and is then deployed as an autonomous system. As built, the bubble generator can be deployed on the seabed at depths of up to 200 m, limited by the operational characteristics of a differential pressure sensor and a first-stage gas regulator used in the system.

The synthetic bubble generator was deployed to the seabed multiple times off the coast of New Hampshire, adjacent to the Isles of Shoals in a water depth of 55 m. Both N₂ and Ar gas bubbles were generated, at sizes ranging from 2.35- to 4.21-mm radius. Ar bubbles were chosen as a practical, safe proxy to CH₄ bubbles: both Ar and CH₄ have low aqueous concentrations in the ocean and similar diffusion coefficients and Henry's law constants (Hayduk and Laudie 1974; Sander 2015). By contrast, Ar and N₂ have very different aqueous concentrations and Henry's law constants (Sander 2015), and bubbles made from both these gases were anticipated to have observably different behaviors (i.e., sizes and TS). Using both Ar and N₂ gases provided a more complete test of both the bubble evolution and acoustic scattering models. For each deployment of the bubble generator, acoustic backscatter from the gas bubbles between 16 and 24 kHz was collected by transiting over the bubble generator multiple times with a broadband split-beam echo sounder. These data represent both a first controlled test of the bubble generator and a combined test of bubble evolution and acoustic bubble characterization.

The design of the bubble generator is described in the following section. Section 3 describes the field tests with the bubble generator, and the acoustic results are compared to a bubble evolution model in section 4. Conclusions from this study are described in section 5.

2. Bubble generator design

Gas is supplied to the bubble generator from a standard 100-ft³ (~2832 L) scuba tank (Fig. 1). Operating the system at a depth of 100 m, this tank of gas pressurized to its maximum value of 3000 psi (21 MPa) is large enough to generate approximately 9×10^8 bubbles of 2.5-mm radii. In general, the number of bubbles that can be generated depends on the deployment depth, and the size and rate of bubbles generation. Operationally, the gas supply and available battery power have been found to be large enough to create bubbles for at least 1 full day of operation without recharging or refilling. The system can be used with multiple gases; in the present work it is used with N₂ and Ar.

A schematic of the bubble generator is shown in Fig. 2. Pressurized gas from the scuba tank is supplied to a

FIG. 1. Final bubble maker assembly on board the R/V *Gulf Surveyor*.

precise pressure regulation system through a balanced first-stage gas regulator, which reduces the scuba tank pressure to 1 MPa (150 psi) over the ambient pressure (regardless of depth). A feed solenoid with a response time of 2 ms, placed between the first-stage gas regulator and an internal reservoir, is used to fill an interior gas reservoir. This gas reservoir serves to reduce the magnitude of the pressuring fluctuations during and after firing the exhaust solenoid, and allows for multiple firings of the feed solenoid while a prescribed reservoir pressure is reached in order to reduce pressure overshoot. Gas bubbles are created by using a 4-ms-long voltage pulse to energize a normally closed exhaust solenoid valve whose input is connected to the internal reservoir and whose output is open to the ocean. The rate of bubble generation is prescribed by the rate at which the exhaust solenoid is fired. The size of the bubble created depends on the pressure difference between the gas reservoir and the exhaust port (i.e., the local ambient pressure at the deployment depth), as well as the exhaust solenoid orifice size. For the configuration used in the present work, the orifice of the solenoid was 0.79 mm in diameter. Drop in replacement solenoids are available with a range of diameters from 0.04 to 0.99 mm. The difference between the internal gas reservoir and ambient pressures is monitored using a differential pressure transducer, which has an accuracy of 0.08% times its range of 0.34 MPa (50 psi); during bubble generation operations the feed solenoid is used to maintain a prescribed differential pressure. The range of differential pressures used to create bubbles is limited by

this sensor to between 7.0×10^{-4} and 0.34 MPa (between 0.1 and 50 psi). The allowable operating pressure on the differential pressure sensor limits the deployment depth for the system to 200 m.

Control of the feed and exhaust solenoids, and monitoring of the differential pressure sensor, is achieved using a Microchip ATmega328 microcontroller in an Arduino Pro Mini. The differential pressure sensor, which has an analog output of 0–5 V linearly corresponding to its range of 0–0.34 MPa, is read using a 10-bit successive approximation analog-to-digital converter on the microcontroller. Two general purpose I/O pins on the microcontroller are used to drive two N-channel metal-oxide-semiconductor field-effect transistors (MOSFETs) that drive the feed and exhaust solenoids at programmable rates and durations (both durations were fixed at 4 ms in the present work).

During bubble generation, a prescribed pressure threshold is compared to the differential pressure reading before and after firing the feed solenoid, to determine whether the internal pressure is at or above the threshold. If the system pressure remains below threshold, the feed solenoid is repeatedly fired until the threshold is reached. Prior to firing the exhaust threshold, the differential pressure is read to verify that the ambient pressure is lower than the internal reservoir pressure so that a backflow of seawater into the system will not occur; this scenario often occurs (temporarily) during deployment when the bubble generator is started on deck at atmospheric pressure and is deployed to a higher pressure. The repressurization of the internal reservoir between generating bubbles typically occurs within a few milliseconds, depending on the size of the bubble and the pressure difference being used. Multiple bubble sizes can be generated during a single deployment, using the microcontroller clock and a bubble generation schedule defined within a script.

The size of the generated bubble is a function of the output volume flow rate of gases through the orifice, and the duration that the solenoid is open. The flow rate is dependent on both the internal and external pressures, and the orifice size. A model of this type of system (pneumatic fluid flow for compressible gasses through an orifice of fixed size with sharp edges) exists for steady-state flow (Sanville 1971; Beater 2007; International Organization for Standardization 2014) and, although it does not perfectly reflect the transient nature of the 4-ms-duration exhaust solenoid firing used in the present system, provides some sense of the depth-dependent performance of the system. In general, the model predicts that for a given pressure and solenoid opening time, the size of the generated bubble should decrease with depth. The predicted rate of

FIG. 2. Schematic of synthetic bubble generation system.

change is higher at shallower depths, decreasing by approximately 25% in the first 50 m below the ocean surface.

Given the depth dependence in the bubble sizes created (for fixed values of differential pressure and exhaust solenoid opening durations), a calibration procedure was developed that can be performed at the operation depth of the bubble generator. The calibration employs an inverted graduated cylinder above the outlet orifice to capture a number of gas bubbles. The gas volume in the cylinder is monitored using an underwater video camera with audio, and with illumination provided by LED dive lights. A prescribed

bubble generation rate is verified using the sound (a broadband “click”) of the firing exhaust solenoid. The volume is calculated by measuring the change in the water level (the meniscus) inside the cylinder as it filled while counting the number of bubbles created, and then dividing the two in order to get the volume of an individual bubble and its effective radius, in similar fashion to the method used for monitoring natural bubble ebullition by Padilla et al. (2019). This calibration procedure was used in a 6-m-deep freshwater test tank and in the field at the depth of the experimental data (section 3) using air, N₂, and Ar, for several different bubble sizes, providing a sense of the

bubble sizes created for different selections of differential pressure and at two different depths (Fig. 3).

3. Field tests

The bubble generator was deployed over 2 days in October 2017, south of the Isles of Shoals (42.9455°N, 70.62403°W) off the coast of New Hampshire, at a depth of 55 m. Bubbles of three sizes of both Ar and N₂ were made over the course of the 2 days of deployments, using a two-stage purging procedure when switching between gases. Bubbles were generated at a rate of one every 4 s. Individual deployments of the bubble generator were used for each size and gas type.

The bubble maker was deployed using a float and weight mooring system, in which a tripod holding the bubble maker (see Fig. 1) was lowered to the seafloor, after which a length of positively buoyant line was slowly paid out as the vessel drifted away from the tripod location. After several tens of meters of drift away from the tripod location, a weight attached to the line was lowered to the seabed, and a second section of line was allowed to rise toward the surface where a float was attached. This line served to help recover the tripod at the end of the experiment, and this deployment method allowed the pickup line to be located far enough away from the tripod that it would not interfere with downward-looking acoustic backscatter measurements of the bubbles. The pickup line does appear, however, in the lower portion of the acoustic data collected during the experiment (Fig. 4).

Acoustic data were collected with a Simrad ES18 split-beam echo sounder operating over a bandwidth of 16–24 kHz using linear frequency modulated pulse. The ES18 has an 11° beamwidth (measured at 3 dB down from the peak of the main lobe) at 18 kHz. The echo sounder was calibrated both in an 18 m × 12 m × 6 m (length × width × depth) tank at the University of New Hampshire (UNH) and at sea using a 64 mm copper sphere, following the standard target calibration method often used for split-beam echo sounders (Demer et al. 2015). With a bandwidth of 8 kHz, the echo sounder has a range resolution of approximately 10 cm. For the bubble release rate of one bubble every four seconds, and with a nominal bubble rise velocity of 20 cm s⁻¹ for bubbles of the size used (>1 cm), the bubbles were spaced far enough apart in the water column to be individually observed by the echo sounder.

The individual bubbles from the bubble generator appear in the acoustic record as targets at near-constant spacing rising through the water column. The 2.4-mm radii N₂ bubbles are shown in Fig. 4 between 20- and 55-m depth. Fish and other scatterers are also visible

FIG. 3. Differential exhaust solenoid pressure vs bubble size calibration. Black denotes a tank calibration conducted at 6-m depth, while gray denotes field calibrations conducted at 55-m depth. Air uses a dot marker, N₂ uses a square, and Ar uses × marks. The N₂ and Ar curves were collected on the data of the acoustic data collection, and the air calibration as conducted at a different time and location (although similar water depth).

throughout the water column. The strong contiguous horizontal target at ~45 m is the floating pickup line attached to the bubble generator. While this echogram appears continuous, it is an amalgamation from four separate passes over the bubble generator.

The maximum acoustic backscatter corresponding to each bubble is found by searching the time series for each ping for local maxima. The local maxima are defined by a threshold value, a minimum separation from other local maxima candidates, and a maximum width of the portion of the peak that has risen above the threshold. A threshold value of -70 dB (corresponding to the color scale in Fig. 4), a minimum separation between local maxima of 24 data points (0.77 m at the echo sounder sample rate of 23 437.5 Hz), and a maximum peak width of 20 samples (0.64 m) were used for this work. The range over which the algorithm operates is manually limited in each ping to minimize erroneous detections from fish and other targets within the water column. The results were then manually scrutinized to remove obviously erroneous results such as fish or the bubble generator pickup line. An example of the final bubble-target selection is shown in Fig. 4 (right). The acoustic backscatter value, associated with each local maximum, is converted to TS using an offset derived from the standard sphere calibration. This applied offset accounts for the ES18 beam pattern using alongship and athwartship phase angles calculated using split-aperture correlation techniques (Burdic 1991; Demer et al. 2015).

FIG. 4. (left) Echogram of match filtered data from ES18 transducer from all pings containing 2.4-mm N_2 bubbles. (right) Echogram from (left) overlaid with picked targets shown as white $\times$ marks.

The results of the field tests are series of depth-dependent TS estimates for bubbles that originated at the bubble generator with different sizes and compositions (Ar and N_2). These estimates are binned in 5-m

increments, with the resulting distributions shown as boxplots in Fig. 5. The distributions vary depending on the bubble size and composition. Ar bubbles with a size at generation of 2.35-mm radius show a TS that steadily

FIG. 5. Estimated target strength vs depth. Boxplots are binned per angle, red lines are median values, boxes represent the 25th–75th percentiles, and red crosses are outliers. (top) Ar and (bottom) N_2 data with bubble sizes increasing from left to right. The Texas A&M Oilspill Calculator (TAMOC) model is overlaid as solid black line, and the number of bubbles in each bin is listed along the right vertical axis of each panel.

decreases from a median value of -53.0 dB at 55-m depth to -56.4 dB at 20-m depth, a reduction in scattering cross section of approximately a factor of 2. N_2 bubbles created with a similar size (2.45-mm radius at the bubble generator) exhibit a near-constant median TS with depth, ranging from -52.6 to -51.9 dB between depths of 25–55 m, where the majority of the observations lie, and -53.3 and -50.7 dB at 20 and 15 m, respectively, where there are a substantially smaller number of observations. In both cases, the distributions of the observations (represented by boxes in Fig. 5 defined by the 25th and 75th percentiles of the data) are narrow enough (often 1–2 dB) to provide confidence in TS trends: decreasing TS with decreasing depth for Ar bubbles, relatively constant with depth for N_2 bubbles.

With the exception of the 3.70-mm Ar bubbles, the results for the larger bubbles show significantly wider distributions of TS. At any given depth, the separation between the 25th and 75th percentiles ranging from 3 to 8 dB for 4.05-mm Ar bubbles and 6 to 10 dB for 4.21 N_2 bubbles. In either case, it is difficult to discern a consistent trend in the depth-dependent TS.

4. Data–model comparison

The acoustic observations were compared to predicted bubble responses using two models: a model for the evolution of a rising bubble from the Texas A&M Oilspill Calculator (TAMOC) as described by Gros et al. (2016, 2017) and an acoustic TS model from Clay and Medwin (1977). The bubble evolution model starts with an initial known bubble size and concentration, and predicts the changes in bubble size and composition as it rises through the water. Bubble size is affected both by gas diffusion across the gas–liquid boundary, according to Henry’s law, and by changes in hydrostatic pressure as the bubble rises. The initial gas concentration is either 100% Ar or 100% N_2 , and the initial bubble size is determined through the field calibration described in section 2. Aqueous concentrations of N_2 and Ar are calculated assuming equilibrium with atmospheric concentrations, using temperature and salinity profiles collected with a CTD during the experiment, and dissolved oxygen is estimated using *World Ocean Atlas* data. The variation between the minimum and maximum values from an average oxygen profile from the area resulted in less than 0.04-mm deviation in bubble radius when averaged over depth, suggesting a low sensitivity to dissolved oxygen.

For the six cases shown in Fig. 5 (three Ar bubbles and three N_2 bubbles), the predicted bubble radii as a function of depth is shown in Fig. 6. In each case there

is a net loss of mass from the bubbles as they rise: the largest increase in size is by a factor of 1.4 for the smallest N_2 bubbles, whereas the change due to pressure alone between 55- and 0-m depth corresponds to a change in volume by a factor of 6.5 according to the ideal gas law or a change in radius of nearly 2. Ar bubbles exhibit a higher net loss of gas than N_2 bubbles, at a rate high enough to cause the bubble to decrease in size in the lower portion of the water column despite the decreasing hydrostatic pressure as the bubbles rise. The increased rate of mass transfer out of the Ar bubbles is attributed to the relatively lower aqueous concentrations of Ar than N_2 ; Ar has a Henry’s law constant that is twice that of N_2 .

The modeled backscattering cross section σ_{bs} (m^2) of a single bubble in the radial direction follows that given by Clay and Medwin (1977):

$$\sigma_{bs} = \frac{a^2}{[(f_r/f)^2 - 1]^2 + \delta^2}, \quad (1)$$

where f_r is the resonant or natural frequency, f is the center frequency of the FM pulse, a is the bubble radius (m^2), and the damping factor δ incorporates losses due to reradiation, thermal conductivity, and shear viscosity. The calculation of (1) requires knowledge of the ratio of specific heats, which is calculated using assuming that the heat capacities can be calculated as the mole-fraction-weighted sums of the heat capacities of the individual gas constituents. The backscattering cross section is converted to TS using

$$TS = 10 \log_{10}(\sigma_{bs}), \quad (2)$$

where TS is the target strength of a single bubble with a backscattering cross section defined in (1). For the bubbles investigated here, at frequencies well above resonance, losses due to reradiation dominate δ , and the impact of δ grows with increasing bubble size. The factor δ acts to reduce the TS by up to approximately 1 dB under the conditions considered here.

The radii and gas compositions of the bubbles at all depths are input into the TS model through the resonance frequency and damping constants, to produce predicted TS curves that are overlaid on the empirical data in Fig. 5. The model predictions for the smallest bubble size for each gas align well with the median values for the data, particularly at depths where the number of observations are highest. For these smallest bubbles, the difference between the model prediction and the median TS observation at the source (i.e., the bubble generator) is less than 0.5 dB. The consistency between model prediction and median observation

FIG. 6. Simulated bubble radii from Texas A&M Oilspill Calculator (TAMOC) model for calibrated bubble sizes of Ar (solid) and N_2 (dashed) using calibrated bubble sizes and gas concentration upon creation and measured environmental parameters in the water column. (left to right) The starting radii for Ar bubbles are 2.35, 3.7, and 4.05 mm for Ar and 2.45, 3.76, and 4.21 mm for N_2 .

remains until the bubbles reach depths of 20–25 m or less, where the model overpredicts the observations by 1–2 dB although with a relatively low number of observations.

The medium sized Ar bubbles (3.70-mm radius at the source) are qualitatively similar to the smallest bubbles in that there is good agreement between the model prediction and the observation at the deeper depths, and an overprediction of the modeled TS at shallower depths (in this case, depth bins of 30 m or less) by 1–2 dB. The model predictions for the medium size N_2 bubbles (3.76-mm radius at the source) are within 1 dB of the median TS observation at all depths except where the number of observations is small (e.g., 16 observations at 15-m water depth; 6 observations at 45-m water depth). The spread of the data, as evidenced by the difference in TS values corresponding to the 25th and 75th percentiles, is higher for the medium sized N_2 bubbles than for the medium sized Ar bubbles, however, particularly for the 20- and 25-m-depth bins.

The large Ar bubbles (4.05-mm source radius) show good agreement at the deeper observation depths, except where the number of observations is low, with differences of less than 0.5 dB at 35 m and approximately 1 dB at 30 m. The model predictions begin to increasingly overpredict the median TS observations at shallower depths, predicting a TS that is approximately 4 dB higher in the

15-m-depth bin. The model overprediction is more pronounced for the large N_2 bubbles (4.21-mm source radius), with deviations from the median observed TS as small as 1 dB at the 35-m-depth bin to approximately 4 dB at depth bins between 5 and 25 m.

To further compare the data–model differences, the observed TS values have been subtracted from the model predictions at each depth bin and grouped by bubble size and initial gas composition. These differences are shown independently of depth in Fig. 7 as empirical probability density functions (i.e., histograms normalized so that they numerically integrate to 1) with a bin resolution of 0.5 dB. The 5th, 15th, 50th, 85th, and 95th percentiles of these same sets of data are shown in Table 1. Both the mode (Fig. 7) and the median values (Table 1) for Ar suggest that the predicted TS is approximately 1 dB higher (a 25% difference in σ_{bs}) than the observed TS for the medium sized and largest bubbles created, and little to no difference for the smallest bubbles created. The TS difference is also positively skewed, and there is an increasingly large number of model overpredictions as the source bubble size grows. Small and medium sized N_2 bubbles show similar results to those for Ar, with a difference in both mode and median values for the TS difference between predicted and

FIG. 7. Empirical probability density functions ρ calculated for the difference between the predicted and observed TS for (left) Ar and (right) N_2 . A positive value indicates that the predicted TS was higher than the observed TS. The density functions use a bin width of 0.5 dB.

observed that is less than 1 dB, and a positive skewness. The largest N_2 bubbles show the most significant deviations between model and predictions. The mode in the TS difference occurs at 0 dB, but the median value shows the model predicting a TS that is 2.5 dB (178%) higher than the observation, and 15% of the predictions are 10 dB (1000%) higher than the observation.

5. Discussion

The tests conducted here act as end-to-end tests of 1) the experimental method for measuring bubble TS, which includes uncertainties due to echo sounder calibration, bubble size calibration, and potential experimental error due to misclassification of marine organisms and other scatterers as bubbles; 2) the model for bubble evolution, which includes dissolution rates, rise velocities, and changes in hydrostatic pressure; and 3) the TS model for a bubble

of a given size and composition, which assumes that bubbles are spherical. The agreement between observed and predicted TS for the smallest bubbles examined suggests that, in these cases, all three (experimental method, bubble evolution model, and TS model) are valid. The agreement for the smallest bubbles is particularly compelling given the different behavior of both Ar and N_2 (Fig. 6). That is, the 2.35-mm Ar bubbles and 2.45-mm N_2 bubbles are not distinguishable at the source based on measurement of TS, but show observably different depth-dependent TS values that are well matched between prediction and observation.

For the medium and large Ar bubbles the prediction initially provides an accurate match to the median observed TS, at 35 m or greater except where the number of observations is low (<10), but then consistently overpredicts the median observed TS for shallower bubbles. These bubbles are predicted to initially decrease in size as

TABLE 1. Percentile values for the difference between observed and predicted TS values for the six types of bubbles investigated using the bubble generator. Positive values indicate the model prediction is greater than the observed TS. These data correspond to the empirical probability density functions shown in Fig. 7.

Source bubble composition and size	5th percentile	15th percentile	50th percentile	85th percentile	95th percentile
Ar (2.35 mm)	-1.6	-0.8	0.3	1.4	4.2
Ar (3.70 mm)	-0.9	-0.2	1.0	3.0	7.9
Ar (4.05 mm)	-3.1	-1.1	1.2	5.7	11.6
N_2 (2.45 mm)	-2.7	-1.4	0.2	2.9	7.0
N_2 (3.76 mm)	-3.4	-1.7	-0.2	4.9	12.7
N_2 (4.21 mm)	-2.8	-0.9	2.4	9.9	13.9

they rise at depths below 25 m, followed by a slight increase in size as the bubbles continue to rise to shallower depths, remaining above 3 mm in radius at all depths (Fig. 6). Bubbles of this size and at these depths scatter acoustic waves at frequencies well above the bubble resonance frequency, and have a predicted σ_{bs} that is proportional to the bubble's geometric cross section. That prediction assumes small values of $ka = 2\pi a/\lambda$, where λ is the wavelength at 18 kHz. At 18 kHz, ka ranges from 0.2 to 0.3 for bubble radii between 3 and 4 mm, making the small ka assumption somewhat weak and possibly causing a nonnegligible error in the model. This error is likely exacerbated by the non-spherical shape of the bubbles. Assuming a nominal bubble rise velocity of 20 cm s^{-1} , the Reynold's and Eotvos numbers for a 3-mm-radius bubble are 1200 and 5, respectively, which places the bubbles in the wobbling ellipsoidal regime (see Fig. 2.5 in Clift et al. 1978). A 4-mm gas bubble would have somewhat larger Reynold's and Eotvos numbers, acting to increase the ellipticity of the bubble. The size and random wobbling motion of the bubble and thus its orientation with respect to the incident acoustic wave likely act to further weaken the assumption of small ka .

That the modeled TS predictions match the observations for the medium and large bubbles at depths of 35 and 40 m, however, suggests there may be a nonacoustic cause for the bias between predicted TS and median observed TS at shallow depths. The models overpredict the observed TS values, which would suggest that the bubbles are either losing mass faster than the bubble evolution model predicts, or rising more slowly. Bubbles of the larger size studied here are expected to experience varying irregularities in shape and oscillations (wobbling) as they rise, which makes mass transfer rate predictions difficult to make. The TAMOC bubble evolution model uses Johnson et al.'s (1969) empirically adjusted parameterization for the mass transfer coefficient for ellipsoidal bubbles. Johnson et al.'s parameterization appears to be within 20% of the data used to derive it, which would correspond to a 20% variability in the rate of change of bubble radius. Bubble rise velocity observations exhibit a similar variability, for large bubbles, due to variations in surfactants at the gas-liquid boundary and/or to the manner in which bubbles are detached from their orifice [see Kulkarni and Joshi (2005) for a review]. Although any vertical component of turbulence is assumed small relative to the bubble velocity, this contribution is ignored in the modeled rise velocity. It is possible that some combination of errors associated with the mass transfer rate or rise velocity, for large bubbles, contributes to the mismatch between prediction and observation found in the present work.

In addition to the bias between prediction and observation, the spread of observed TS values was considerably larger for larger bubbles (Fig. 5). This may be a result of the combination of larger ka values and the wobbling nature of these ellipsoidal bubbles, causing a nonisotropic acoustic scattering pattern that is reflected in the data. It is also possible that bubble fragmentation occurred: subsequent to this field experiment, it was observed that bubbles of the largest size created by the bubble generator were splitting at the source, one slightly smaller bubble than desired and one very small bubble. This behavior was associated with fouling of the exhaust orifice and may have corrupted the results for the largest bubbles, although good agreement between observation and prediction at the deeper depths suggests that this experimental error may not have been present during the field experiment. Bubble fragmentation, where shear forces acting on the bubble overcome its surface tension, may also be an explanation for the variability and bias for the largest Ar and N_2 bubbles, although the medium-sized N_2 bubbles showed good agreement between model and prediction and were similar size to the largest Ar bubbles in the upper part of the water.

It is useful to examine the comparison between TS observations and model predictions by translating the TS residuals shown in Fig. 7 to uncertainties in bubble size. For example, statistics from these bubble size residuals provide some sense of how accurately and precisely the acoustic observations could be inverted for bubble size estimates under the assumption that the modeled bubble evolution were true. The uncertainty in bubble radius is described by some uncertainty in the observed σ_{bs} and is given by

$$s_a = \frac{da}{d\sigma_{bs}} \sigma_{\sigma_{bs}}, \quad (3)$$

where s_a is the standard deviation of the bubble radius, $da/d\sigma_{bs}$ is the change in that radius with respect to the change in backscatter cross section, and $\sigma_{\sigma_{bs}}$ is the standard deviation of the observed backscatter cross sections for a bubble. Using the assumption that the observations occur at frequencies well that are much larger than f_r ,

$$\sigma_{bs} \cong \frac{a^2}{1 + \delta^2} \quad (4)$$

and

$$\frac{d\sigma_{bs}}{da} \cong \frac{2a}{1 + \delta^2}. \quad (5)$$

If the damping coefficients are assumed to be very small, $da/d\sigma_{bs} \cong 1/(2a)$, and the expected uncertainty in a bubble radius estimate can be found from:(16)

$$s_a = \frac{da}{d\sigma_{bs}} s_{\sigma_{bs}} \cong \frac{s_{\sigma_{bs}}}{2a}. \quad (6)$$

As an example, the medium sized N_2 bubbles at 40 m had a TS of -48 dB ($\sigma_{bs} = 1.6 \times 10^{-5}$) and an effective radius of 3.9 mm. A 1-dB TS uncertainty equates to a factor of approximately 25% or 0.4×10^{-5} , which gives an uncertainty of 0.5 mm according to (6). A 4.6-mm N_2 bubble at 30 m depth with TS = -47 dB and a TS uncertainty of 2.5 dB would be nearly 4 mm.

6. Conclusions

A bubble generator system was designed and constructed for studying the methodologies associated with quantifying the flux of methane from gaseous seeps. Field trials with the system, in which both Ar and N_2 bubbles were created, were used to simultaneously test an acoustic scattering model and a model for the evolution of a bubble rising through the ocean. The agreement between the model and the experimental observations suggest that both models (acoustic and bubble evolution) are accurate, at least for the smaller bubbles used in this work, and neglecting the scenario where errors in both models cancel each other out. Larger systematic offsets and larger random fluctuations were observed for the larger bubbles. The larger random fluctuations are explained in part by the predicted size-dependent behavior of TS observations (the uncertainty in backscattering cross section is proportional to the radius of the bubble), although the observed TS fluctuations for the largest bubble sizes well exceeded this. Several possibilities exist for the bias between the observations and the predictions for the largest bubbles, including errors in the predictions of rise velocity and gas transfer rate, and in the acoustic model. Further study is required in order to isolate the specific cause(s) of this bias.

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