

Oil & Natural Gas Technology

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Quarterly Research Performance

Progress Report (Period Ending 3/31/2019)

Dynamic Behavior of Natural Seep Vents: Analysis of Field and Laboratory Observations and Modeling

Project Period (10/01/2016 to 09/30/2019)

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1 Accomplishments

1.1 Summary of Progress Toward Project Objectives

The *overarching goal* of this project is to develop a computer model to predict the trajectory and dissolution of hydrate-armored methane bubbles originating from natural seeps. The model is based on the Texas A&M Oilspill (Outfall) Calculator (TAMOC), developed by Dr. Socolofsky, and which has been refined and validated through this project to explain fundamental laboratory and field observation of methane bubbles within the gas hydrate stability zone of the ocean water column. *Our approach* is to synthesize fundamental observations from the National Energy Technology Laboratory's (NETL) High-Pressure Water Tunnel (HPWT) and field observations from the Gulf Integrated Spill Research (GISR) seep cruises (cruises G07 and G08), conducted by the PIs in the Gulf of Mexico, to determine the dissolution pathways and mass transfer rates of natural gas bubbles dissolving in the deep ocean water column. We will achieve these objectives by pursuing the *following specific objectives*:

1. Analyze existing data from the NETL HPWT.
2. Synthesize data from the GISR natural seep cruises.
3. Refine and validate the seep model to predict available data.
4. Demonstrate the capability of the seep model to interpret multibeam data.

Ultimately, the *main outcome and benefit* of this work will be to clarify the processes by which hydrate-coated methane bubbles rise and dissolve into the ocean water column, which is important to predict the fate of methane in the water column, to understand the global carbon cycle, and to understand how gas hydrate deposits are maintained and evolve within geologic and oceanic systems, both at present baselines and under climate-driven warming.

During this reporting period, we focused on Tasks 5 and 6 and continued working on Task 7. For Task 5, we conducted new experiments in the Offshore Technology Research Center (ORTC) for the acoustic calibration of the M3 multibeam sonar in gas bubble plumes outside the hydrate stability zone. These experiments included measurement of the bubble size distribution and gas flow rate together with M3 sonar imaging. For Task 6, we have used these data to validate our acoustic models and to quantify their performance. Aside from the experiments, we focused most of our effort this period on model validation and quantification of model errors, as summarized in

Task Name	Assigned Resources	Year 1 / Phase 1				Year 2 / Phase 2				Year 3 / Phase 3		
		Qtr 1	Qtr 2	Qtr 3	Qtr 4	Qtr 1	Qtr 2	Qtr 3	Qtr 4	Qtr 1	Qtr 2	
Task 1.0 - Project Management and Planning	Socolofsky	●										
Task 2.0 - Analyze NETL Water Tunnel Data Subtask 2.1 - Evaluate hydrate formation time Subtask 2.2 - Track hydrate crystals on bubble interface Subtask 2.3 - Validate bubble shrinkage rates Milestone: Obtain NETL HPWT Data Milestone: Adapt Matlab code to NETL data	Socolofsky Socolofsky Wang Wang											
Task 3.0 - Synthesize GISR Field Data Subtask 3.1 - Bubble characteristics from high-speed camera Subtask 3.2 - Synchronize acoustic and camera datasets Milestone: Develop Matlab code for M3 and EM-302 data	Wang Wang Wang											
Decision Point 1												
Task 4.0 - Refine and Validate Seep Model Subtask 4.1 - Validate to NETL Water Tunnel Data Subtask 4.2 - Validate to GISR Field Data Subtask 4.3 - Finalize and distribute seep model Milestone: Adapt seep model to NETL data Milestone: Quantify seep model performance	Socolofsky Socolofsky Socolofsky Socolofsky											
Decision Point 2												
Task 5.0 - Conduct No-Hydrate M3 Experiment Milestone: OTRC Experimental Report	Wang											
Task 6.0 - Apply Seep Model to GISR Multibeam Data Subtask 6.1 - Analyze M3 data to characterize hydrate shells Subtask 6.2 - Analyze EM-302 data for bubble concentration Milestone: Quantify performance of acoustic models	Socolofsky Socolofsky Wang											
Task 7.0 - Document Model Validation Milestone: Complete model validation	Socolofsky											
Task 8.0 - Data Distribution / Archiving	Socolofsky											

Figure 1: Project Timeline.

the completed Milestone report for Task 6 (Quantify performance of acoustic models). We also achieved the Milestone for Task 7 (Complete model validation). All actions for Task 4.0 were also completed. Finally, we continue to draft journal manuscripts that summarize our simulation results, a major element of Task 7 (Document model validation). A detailed report of our progress on each of these tasks for the present performance period is reported herein and in the Milestone report for Task 6.

1.2 Progress on Research Tasks

Figure 1 presents the project timeline, showing each of the project tasks, subtasks, and milestones as identified in the Project Management Plan (PMP), and updated to show Task 5.0 now in the second quarter of Phase 3. The present reporting period concludes the second quarter of FY 2019 (Phase 3 of the project). During this period, we completed work on Tasks 4 and 5 and continued effort on Tasks 6 and 7. We have also submitted documentation for completion of the Milestones

“OTRC Experimental Report,” “Quantify Performance of Acoustic Models,” and “Complete Model Validation.” The summary of the completed work together with work conducted on these ongoing tasks during the present reporting period is summarized in the following sections.

1.2.1 Task 1.0: Project Management Planning

The Project Management Plan was completed during the first quarter of Phase 1 and accepted in final form as of October 28, 2016.

1.2.2 Task 2.0: Analyze NETL Water Tunnel Data

In this project, we have analyzed the comprehensive data set of HPWT data collected by NETL. To do this, we have transferred a complete copy of all raw data (primarily image files and time history data of pressure and temperature in the HPWT during each experiment) to Texas A&M University and have installed this data on a secure internal server. Data transfer was completed on March 24, 2017, and achieved Milestone 1 for the project (Obtain NETL HPWT Data). Task 2 was completed as of June 30, 2018. The sections below summarize the key results obtained for each of the Subtasks of this Task.

Subtask 2.1 - Evaluate Hydrate Formation Time

This subtask was completed as of September 30, 2017, and all of the post-processed data has been submitted with the report for Decision Point 1 (see § 1.2.4). In this task, we identified the moment that hydrate skin coverage was completed for each bubble in the experiments as well as for key moments when the hydrate dynamics changed. For a complete description of the data analysis for this subtask and the post-processed results, see the full report for Decision Point 1.

Subtask 2.2 - Track Hydrate Crystals on Bubble Interface

This subtask was completed as of December 31, 2017, and a complete analysis of the results with conclusions was submitted with the first-quarter progress report of FY2018. For this task, we analyzed all of the high-speed camera data for gas bubbles with hydrate shells to track the motion of hydrate plates when the hydrate coverage was not 100%. We found two main types of behavior. First, when hydrate plates are large and their spacing is non-uniform, the plates are observed to translate across the leading edge of the bubbles. The mean speeds of this hydrate shell movement was 10 cm/s, with peak speeds close to the rise velocity of bubbles (20 cm/s). Second,

during hydrate dissociation, when many, small hydrate crystals cover the bubble surface in a quasi-uniform distribution, the hydrate particles are not observed to translate over the surface of the bubble. Instead, they remain knitted together, and the boundary condition at the bubble/water interface appears to be no-slip.

Based on these observations, we anticipate that mass transfer rates for the large hydrate shells that move across the leading edge of the bubble will be higher than for dirty bubbles; whereas, we expect the mass transfer rates for hydrate-coated bubbles and cases with small hydrate particles uniformly distributed over the bubble surface to be similar to dirty bubbles or slower. Because the system pressure inside the HPWT was not constant during these events, we will evaluate these mass transfer rates in the context of Task 4 as we compare the model results to these data.

Subtask 2.3 - Validate Bubble Shrinkage Rates

This subtask was completed as of April 30, 2018, and has been reported in several quarterly reports through the project performance period. We adapted our Matlab image analysis program for bubble size evaluation to the NETL HPWT dataset and compared our results for bubble size to those reported by NETL in their report by Levine et al. (2015). Although there were small differences in our computed sizes, these are attributable to different choices in the cut-off and cut-on criteria for identifying the bubble edge and were negligible in comparison to the inherent variability in the data due to bubble motion. This variability is primarily caused by two factors: 1.) rotation of the bubbles when they have non-spherical shape and 2.) changes in the image magnification as the bubbles move toward and away from the camera. Both factors lead to experimental error in the computed bubble sizes. We evaluated this error by analyzing long data sets in sequentially shorter sample periods. Our analysis concluded that bubble shrinkage rates are converged after a minimum of 500 s of sampling, as this is adequate time for the bubble to wander about the whole measurement volume and experience several rotations. These data will be used extensively in Task 4 as we validate the shrinkage rate predictions of the model to those measured in the HPWT.

Progress Toward Milestones

Milestone 1 (Obtain NETL HPWT Data) was completed on March 24, 2017, and Milestone 2 (Adapt Matlab Code to NETL Data) was completed on September 26, 2017. These Milestones conclude the Milestones associated with Task 2.

1.2.3 Task 3.0: Synthesize GISR Field Data

The project PIs conducted two research cruises to natural seeps in the Gulf of Mexico under funding to the GISR consortium. These were the G07 cruise in July 2014 to Mississippi Canyon (MC) block 118 and to Green Canyon (GC) block 600 and the G08 cruise in April 2015 to MC 118. Both cruises were on the *E/V Nautilus* and utilized the remotely operated vehicle (ROV) *Hercules*. This project utilizes two main datasets from these cruises: data from our stereoscopic high-speed camera system mounted on the ROV (Wang et al. 2015) and acoustic data collected by an M3 sonar mounted on the ROV and an EM-302 multibeam sonar mounted on the haul of the ship. The image data from the G07 cruise was analyzed previously and reported in Wang et al. (2016). This project analyzes all of the acoustic data and performs a complete analysis of the image data for the G08 cruise. This task was completed as of December 2017, and the outcomes of each subtask are reported below.

Subtask 3.1 - Bubble Characteristics from High-Speed Camera.

This subtask was completed as of September 30, 2017, and all of the post-processed data were submitted with the report for Decision Point 1 (see § 1.2.4). In this task, we have analyzed images from our high-speed, stereoscopic image system to compute bubble sizes and the rise velocities of individual bubbles. For a complete description of the data analysis for this subtask and the post-processed results, see the full report for Decision Point 1.

Subtask 3.2 - Synchronize Acoustic and Camera Datasets.

This subtask was completed as of March 31, 2018. Data from the cameras and acoustic measurements have been reported separately. The image data include bubble size distributions and rise velocity, and are reported in the report for Decision Point 1. The acoustic data have been analyzed to predict the *in situ* target strength, which is a measure of the acoustic backscatter from the bubbles within each sample volume. This work was reported in the report for Milestone 3. The final output of this subtask was a calibration curve relating the observed bubble characteristics to the target strength measured by the M3 and EM 302 multibeam sonars. The calibration curve for the EM 302 was reported in our report for Milestone 3, and the calibration curve for the M3 was included in our quarterly report for the second quarter of Phase 2. These data along with results of Task 5 (OTRC experiment) will be used in Task 6 to evaluate the seep model at the field scale.

Progress Toward Milestone

Milestone 3 (Develop Matlab Code for EM 302 and M3 Data) was completed on September 29, 2017. This Milestone concludes the Milestones associated with Task 3.

1.2.4 Decision Point 1

The report for Decision Point 1 was completed and submitted as of October 31, 2017. Based on successful completion of the go/no go success criteria for Decision Point 1 outlined in the PMP, we were granted permission to continue into project Phase 2 and begin work on Task 4.

1.2.5 Task 4.0: Refine and Validate Seep Model

Since the Deepwater Horizon accident, the project PIs have been developing a numerical model to predict the fate of petroleum bubbles and droplets in the ocean water column. This model is called the Texas A&M Oil spill Calculator (TAMOC), and is freely available through <https://github.com/socolofs/tamoc>. This model can compute the dissolution of a natural gas bubble in the ocean water column, and prior to this project, had been applied to study the fate of methane released from natural gas seeps along the continental slope of the Gulf of Mexico. In this project, we applied this numerical model to simulate the experiments in the NETL High-Pressure Water Tunnel (HPWT; see Task 2) and the field observations from the GISR expeditions (see Task 3). These simulations are used to validate our model for the formation time of hydrate skins of natural gas bubbles within the hydrate stability zone of the oceans and our equations for mass transfer from bubbles with and without a hydrate skin. This model is important to predict the distribution of methane in the ocean water column from natural seeps, accidental oil well blowouts, hydrate production, or from gas release caused by anthropogenic or changing climate forcing.

Subtask 4.1 - Validate to NETL Water Tunnel Data.

This subtask was completed as of January 31, 2019. In the NETL HPWT experiments, cameras observed the bubbles over time as they dissolved into the surrounding flow, and these experiments were conducted at different pressure and temperature conditions. Because the pressure and temperature in the HPWT is prescribed by the operator and independent of bubble position (the pressure is controlled by a set of piston pumps and the bubble is held at a constant depth in the water tunnel), we have adapted the TAMOC model to allow pressure and temperature to be prescribed functions of time so that we can model the exact conditions experienced by a bubble during an

experiment.

As the raw experimental observations are camera images of bubbles, the quantitative observations are obtained by additional image processing and calculations (completed during Task 2 of this project). We identified three sets of derived data values to use in model validation. These were hydrate transition time, mass transfer rate, and bubble evolution. For the hydrate transition time, our analysis concluded that the gas injection method used in the experiments (i.e., slowly collecting gas in a cap before release and pressurization) does not conform to the behavior in the ocean (i.e., instantaneous release from the seafloor), such that hydrate formation time is observed in the HPWT, but not predictable due to the highly variable injection times. Hence, we initiate simulations using dirty-bubble mass transfer rates immediately following hydrate formation. We then used the observed bubble shrinkage rates to infer the mass transfer rate. Our analysis showed that the mass transfer rates β_{obs} observed in the water tunnel for non-hydrate conditions were faster than empirical rates for dirty bubbles but still much slower than empirical rates for clean bubbles. We used these observations to establish a correlation between the empirical and observed mass transfer rates in the non-hydrate experiments. Our best-fit relationship was

$$\beta_{obs} = 1.6\beta \tag{1}$$

where β_{obs} was observed by NETL for non-hydrated bubbles and β is the empirical, dirty-bubble mass transfer rate predicted by TAMOC. Finally, we assessed the model performance and the performance of Equation (1) by simulating each NETL HPWT experiment using TAMOC, including experiments with a hydrate shell. Figure 2 shows the model performance using these methods. In this figure, the calibration data are the red curve, which shows a broader range of agreement than for the validation data in blue. This occurs because there are more experiments in a more diverse set of operational conditions in the calibration set than the validation set of data. Across all experiments, the model agrees on bubble shrinkage rate within $\pm 15\%$ relative error for over 93% of the experiments.

Subtask 4.2 - Validate to GISR Field Data.

This subtask was completed as of January 31, 2019. In the GISR field experiments, three observation platforms were used: *in situ* imaging from the stereoscopic imaging system at discrete points from the sea floor to about 250 m altitude and acoustic backscatter measurements from the EM 302 haul-mounted multibeam sonar and from the M3 multibeam sonar mounted on the ROV. In this Subtask, we validated the TAMOC model predictions at the seeps surveyed during the G07

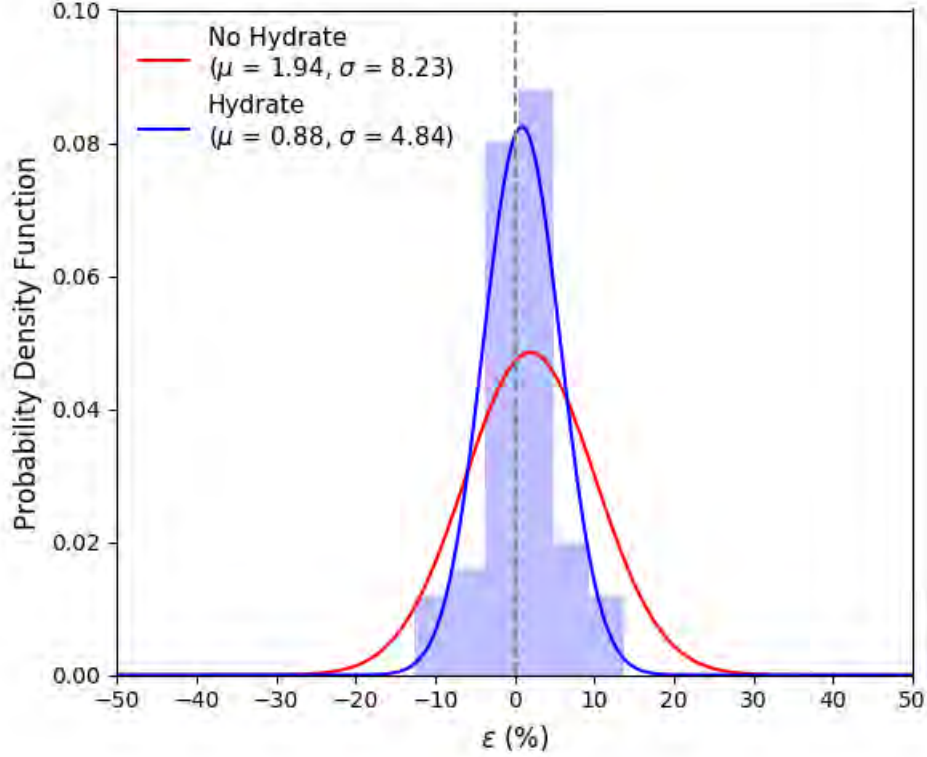


Figure 2: Relative percentage error for bubble shrinkage rate between the observed and modeled bubbles in the NETL HPWT. Red and blue curves are a Gaussian fit to the measured statistics; the histogram shows the measured PDF of the results with hydrate.

and G08 GISR expeditions to these measured data. We post-processed the raw camera images and acoustic backscatter to yield three derived datasets for model calibration and validation. These were the bubble size distribution and flow rate, which served as initial conditions to the model, the lateral spreading of bubbles in the M3 acoustic images, and the observed height of maximum bubble rise in EM-302 data. For all field-scale simulations, we used a correlation developed earlier to predict the hydrate formation time and we assumed the mass transfer coefficients were equal to those predicted by empirical formulas.

Both the camera and M3 data we used to generate initial conditions or input parameters for the model. The initial bubbles size distribution and flow rate are needed to initialize a simulation of a seep flare. From the bubble spreading in the M3 acoustic images we were also able to measure the lateral turbulent diffusivity that affects the bubble spreading. This is the main parameter in the random-walk model for the bubble tracking. The model performance was then assessed through its ability to predict the rise heights and trajectories of the natural seep flares observed by the

EM-302 (see a sample result in Figure 3). We used the target strength of acoustic backscatter

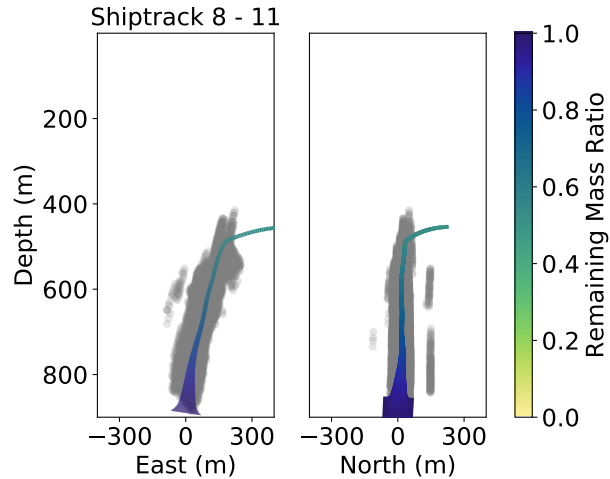


Figure 3: Comparison of model simulation for flare centerline trajectory and bubble mass flux (colorbar data) and flare detection by the EM 302 (gray circles) for one ship track during the G07 GISR cruise.

predicted by TAMOC to compare to instrument noise levels and showed that natural seep flares become acoustically transparent when the largest bubbles in the plume shrink to sizes that are no longer observable. In our validation exercise, we showed that using d_{98} of the initial bubble volume distribution as the bubble size for prediction of the flare height, we could have an r^2 value of 0.98 comparing our model-predicted flare rise heights to the measured data, a bias of 41 m absolute height (out of rise heights between 400 m and 1800 m), and an average mean percentage error of rise height of 4.7%. This performance is quite good and exceeds that of other models that are used in the literature to predict natural seep flares.

Subtask 4.3 - Finalize and Distribute Seep Model.

The above two sub-tasks completed the model validation; this subtask was completed as of January 31, 2019. We provided the source code of the model with the archive of NETL HPWT simulation results. The model is also maintained as publicly available through the Github code sharing website (see Section 2 Products, below). This concludes the major activity under Task 4.

Progress Toward Milestones

Milestone 4 (Adapt TAMOC model to NETL data) was completed on June 19, 2018. Milestone 5 (Quantify seep model performance) was completed with the quarterly report, submitted in January 31, 2019. These Milestones conclude those associated with Task 4.



Figure 4: CCD video camera image of an airstone bubble plume at 3.0 NL/min gas flow rate at 11.45 m above the source.

1.3 Decision Point 2

The report for Decision Point 2 was completed and submitted as of May 31, 2018. Based on successful completion of the go/no go success criteria for Decision Point 2 outlined in the PMP, we were granted permission to continue into Task 5 (OTRC Experiment).

1.3.1 Task 5.0: Conduct No-Hydrate M3 Calibration Experiment in OTRC

The main activities conducted during this reporting period were related to the OTRC experiment, which was conducted from February 25 to March 4, 2019. In this experiment, we simulated two different natural seep vents at five different flow rates in the 16.8 m deep, central pit of the OTRC's directional wave basin. We measured the bubble size distribution from an *in situ* CCD camera (see Figure 4 for a sample image), water velocity in the plume using a Vectrino II acoustic Doppler velocimeter (ADV), and observed the plumes using an M3 multibeam sonar (see Figure 5 for a sample acoustic image of a bubble plume). Using a tungsten carbide ball bearing, we calibrated the M3 acoustic response, and from the measurements of simulated natural seeps, we further validated our acoustic models for bubble dynamics in the M3 images.

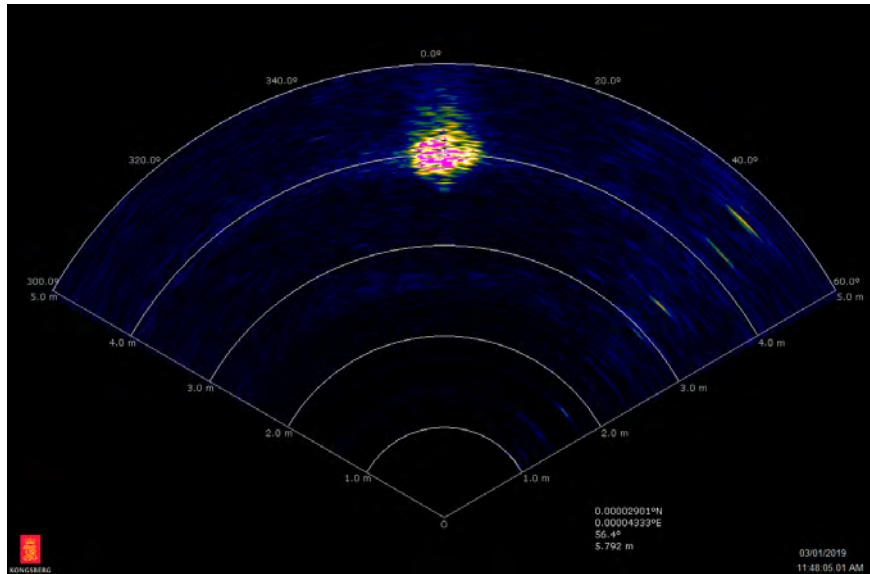


Figure 5: Acoustic image from M3 of an airstone bubble plume at 3.0 NI/min gas flow rate at 12.0 m above the source.

Progress Toward Milestone

Full details of the OTRC experiment set up and results were provided in the report for Milestone 6 (OTRC experiment report, submitted March 21, 2019). The validation of the acoustic models, which relies in part on the data collected in this experiment, were also reported in the report for Milestone 7 (Quantify, performance of acoustic models; associated with Task 6, below). These Milestones conclude those associated with Task 5.

1.3.2 Task 6.0: Apply Seep Model to GISR Multibeam Echosounder Data

In this Task, we use the seep model validated in Task 4 together with the acoustic data analyzed in Task 2 and refined in Task 5 to evaluate the characteristics of the natural seeps at MC 118 and GC 600. This includes an evaluation of the acoustic signature of hydrate shells that may be present in the M3 acoustic cross-sectional data obtained by the ROV and the water column trajectory and flow rate that may be extractable from the haul-mounted EM 302. Together, these activities will explore the role of hydrate shells on the fate of methane from natural seeps and predict the vertical distribution of methane in the water column originating from these seep sources.

Subtask 6.1 - Analyze M3 Data to Characterize Hydrate Shells.

During the present reporting period, we have focused on validation of our acoustic models. For the M3, this entailed calibration of the acoustic response of the instruments through the OTRC

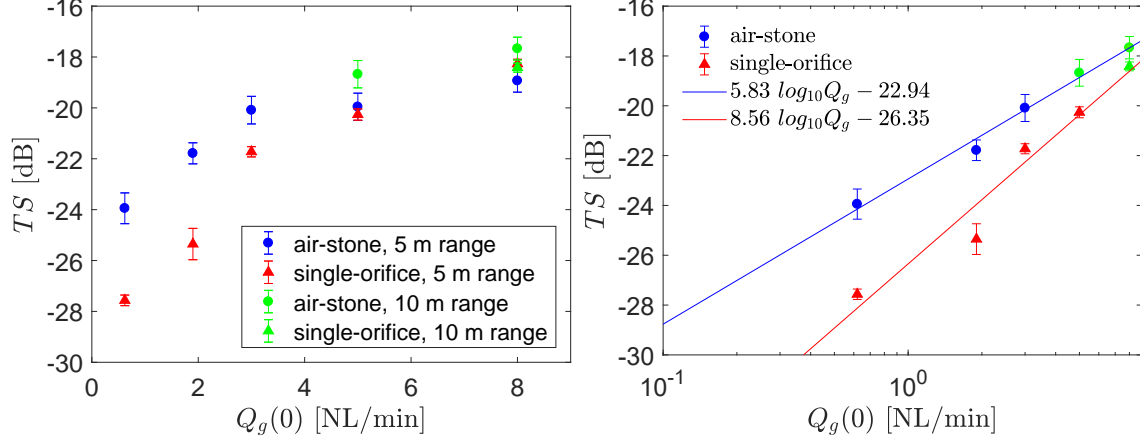


Figure 6: Target strength of bubble plumes as a function of gas flow rate. The symbols represent time-averaged data, and the errorbars represent the standard deviation of the instantaneous data. Left panel: data only in the linear scale; right panel: data with the fitted line on the log-linear scale, where data of 10 m range were used for large Q_g .

experiments. This work is reported in detail in the report for Milestone 7 submitted together with this quarterly report.

From the OTRC experiments, we obtained a calibration of the multibeam sonar acoustic response. Knowing this calibrated response, we can then relate the target strength of acoustic backscatter emitted by a source to that source’s properties. In this project, we want to relate the target strength of a bubble flare to the void fraction or volume flux in the flare. Figure 6 shows the correlation between the calibrated target strength TS measured by the M3 and the flow rate for each bubble plume modeled in the OTRC. Since we measured both the flow rate and gas bubble size distribution in the laboratory, we can also directly compute the bubble flare flow rate using the measured TS . To do this, we utilize the known acoustic behavior of the plume for the measured bubble size distribution. The result of the calculation is shown in Figure 7. In this figure, we show the measured gas bubble flow rate and the flow rate estimated from the target strength observed by the M3. For most cases, the variability in the prediction estimated from the statistical variation of the measurement signal (i.e., the error bars in the figure) falls within the range of the measured data (i.e., the known flow rates). Overall, the goodness-of-fit has an R^2 value of 0.89, which is quite good. Hence, this gives strong validation that our acoustic models are correct and that our calibrated M3 response can accurately estimate gas flow rate at the field scale using the bubble size distributions measured from the ROV.

In the coming reporting period, we will use this new acoustic model to analyze the M3 data

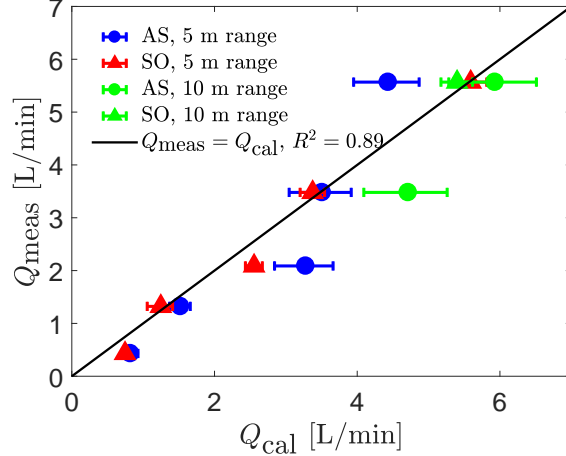


Figure 7: Direct comparison of gas fluxes from different diffusers.

from the GISR cruises with the goal to both estimate *in situ* gas flow rates and determine whether there are acoustic signatures of the hydrate shells in the field data from the M3.

Subtask 6.2 - Analyze EM-302 Data for Bubble Concentration.

We also analyzed the performance of our acoustic model for the EM-302 data. Like the M3, the EM-302 is an uncalibrated instrument, but because of the scales over which we took observations (several hundred meters), it was not possible to calibrate the EM-302 in the laboratory. Instead, we apply the manufacturer-reported post-processing algorithms and the sonar equation to obtain a computed target strength that is proportional to the true target strength (see the report for Milestone 7, Quantify performance of acoustic models).

To compute the target strength throughout each seep surveyed, we first extracted the water column data using the Fledermaus Watercolumn software package and used the position of the ship and seep site to identify each bubble flare in the acoustic data. Figure 9 shows an example seep capture for the Sleeping Dragon site during the G08 cruise. We then post-processed this data to compute the cross-sectional summed target strength along the trajectory of the plume. Because the EM-302 target strength data are relative, we shift each curve to a known value obtained from the measured data at the seafloor. These measured data included the bubble size distribution and the gas flux measured by the stereo camera system on the ROV during a dive closest in time to the EM-302 survey.

To evaluate the performance of our acoustic analysis of the EM-302 data and to validate the TAMOC model, we simulated each bubble flare with TAMOC. We initialized each simulation us-

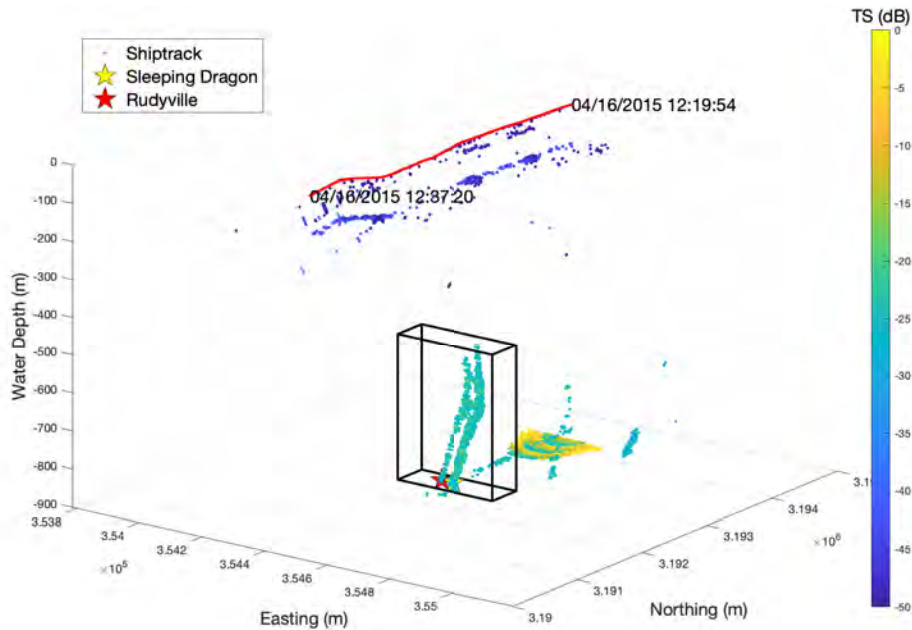


Figure 8: Watercolumn backscatter observed by the EM-302 during a survey transect of the Sleeping Dragon seep flare at MC 118.

ing the measured bubble composition, gas flux, and bubble size distribution at the seafloor. We then computed the total target strength from the simulated data as a function of height above the seafloor. Figure 9 presents a sample comparison at the two main seep sites in the GISR dataset (GC 600 and MC 118). The model generally tracks within the 95% confidence interval of the acoustic data quite well, and has an overall relative percentage error of -20%. Considering the fact that the EM-302 is uncalibrated and the gas flow rate and bubble size distribution is unsteady at the sea floor, this performance is considered very good.

Progress Toward Milestone

Milestone 7 (Quantify performance of acoustic models) was completed as of April 30, 2019, and submitted with the present quarterly report. This concludes all Milestones for Task 6.

1.3.3 Task 7.0: Document Model Validation

In this Task, we document the model validation through reporting to NETL, distribution of the model over Github, and reporting of our findings in journal articles in the peer-reviewed literature.

During the present reporting period, we focused on reporting for the Milestone reports for

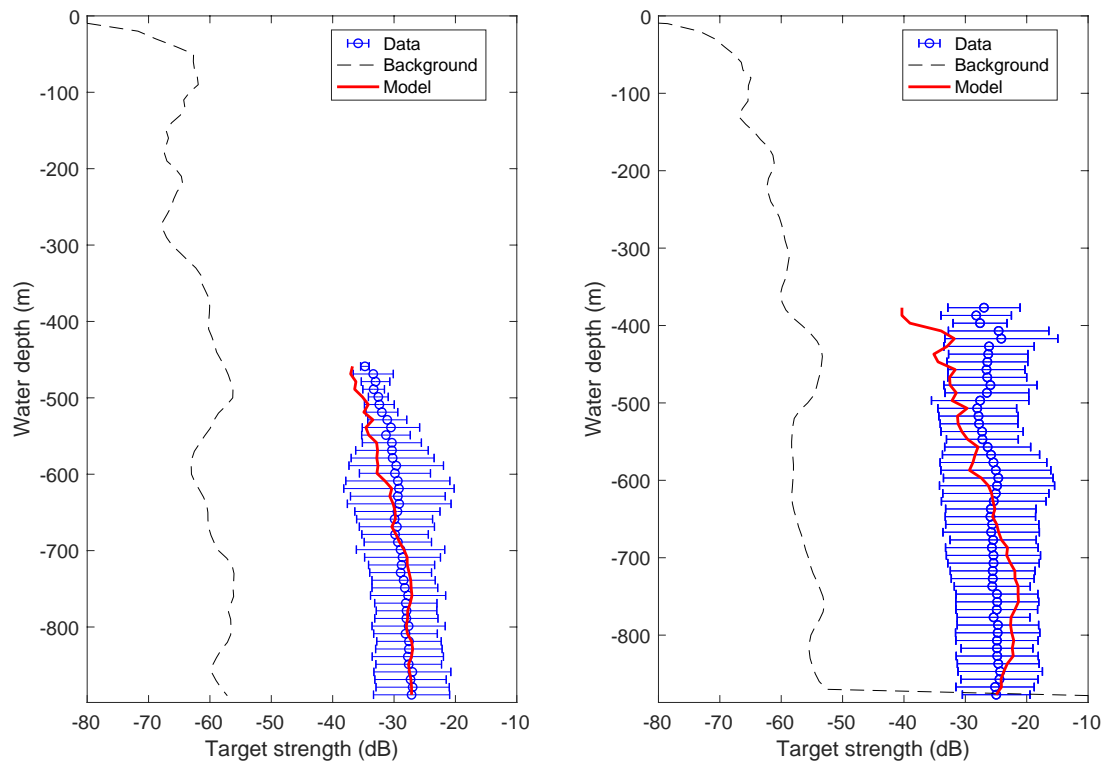


Figure 9: Comparison between the observed target strength (blue data) and the target strength predicted by TAMOC (red curve) for a seep survey at GC 600 (left panel) and MC 118 (right panel).

Tasks 5 and 6. We also continued work on journal manuscripts stemming from the body of work conducted through this project. These in-progress journal papers are summarized as follows:

- “Dynamics of deepwater natural gas seeps within the hydrate stability zone,” to be submitted to *Geophysical Research Letters*. This manuscript is in its final editing stage before submission to the journal and will focus on the results of Task 3.
- “Modeling the behavior of hydrate-affected bubbles rising in the deep ocean,” to be submitted to *Geochemistry, Geophysics, Geosystems*. This manuscript is the second chapter in the Ph.D. dissertation of Inok Jun, who graduated in December 2019. This chapter needs to be lightly edited before submission to a journal, and will focus on the results of Tasks 3 and 6.
- “Predicting natural seep flare heights in the deep ocean,” to be submitted to *Journal of Geophysical Research–Oceans*. This manuscript is the third chapter in the Ph.D. dissertation of Inok Jun, who graduated in December 2019. This chapter needs to be lightly edited, and will focus on the results of Tasks 4.2 and 6.
- “Mass transfer rates in high pressure water tunnel experiments for methane and natural gas with hydrate armoring.” This manuscript will report the data analysis from Task 2 and model validation of Task 6. We have started to write this paper, and this will be a major element of the Ph.D. dissertation for B. Kim.

Each of these manuscripts will be a major portion of our project effort continuing into the remainder of project Phase 3.

Progress Toward Milestone

During the present reporting period, we concluded Milestone 8 (Complete model validation). This Milestone is verified through the present Quarterly Report summarizing the conclusion of Task 4, through the completion of Milestone 5, Quantify seep model performance, and through the validation presented in Milestone 7, Quantify performance of acoustic models. While the model will continue to be improved and updated through the coming years, the major advances proposed for this project have been concluded.

1.4 Deliverables

To date, we have completed the following list of deliverables:

1. **Project Management Plan (PMP)**. The PMP was delivered in its accepted and final form on October 28, 2016.
2. **Data Management Plan (DMP)**. No revisions were requested by the Project Officer to the plan submitted with the proposal; hence, the original DMP is the present guiding document. Revisions will be updated as necessary throughout the project as required by the Project Officer.
3. **Task 2 NETL HPWT Analyzed Data**. The recipient shall provide time series of hydrate formation time, periods of crystal motion on the bubble/water interface, and bubble equivalent spherical diameter to NETL in the format of their choice (ASCII, Matlab, NetCDF, etc.) by the end of Task 2. We have provided these data through the reports for Milestone 2, Decision Point 1, and the quarterly reports.
4. **Task 3 GISR Seep Cruise Analyzed Data**. The recipient shall provide all post-processed analyses of the GISR high-speed camera data for the Gulf of Mexico seep cruises along with time series of corresponding M3 and EM-302 datasets. The camera data shall be provided to NETL in the format of their choice; M3 and EM-302 data shall be provided in the manufacturer raw format. The recipient shall submit these data to NETL by the end of Task 3. We have provided these data through the reports for Milestone 3, Decision Point 1, and the quarterly reports.
5. **Task 4 Validated Seep Model**. The recipient shall provide the refined and validated seep model to NETL. The recipient shall submit the model to NETL by the end of Task 4. We have provided the source code to the validated seep model in the data archive submitted for Milestone 5, Quantify seep model performance.

As of the present reporting period, we have concluded the deliverables for Tasks 2, 3, and 4. The next set of deliverables will be generated at the conclusion of Tasks 6, 7, and 8. Progress toward these deliverables is summarized above in the reporting for each Task.

1.5 Milestones Log

Table 1 presents the schedule of milestones with their verification methods for the duration of the project period. The Table reflects the change to the project schedule such that the OTRC

experiment (Task 5) was due in March 2019. Presently, all Milestones identified in the Project Management Plan have been completed.

1.6 Plans for the Next Reporting Period

During the next reporting period, we will be applying our numerical models to the GISR multibeam data (Task 6) to bring that work to a close. This work will apply the correlation obtained through the OTRC experiments (Task 5) to predict bubble concentration and flow rate for the natural seeps observed by the GISR cruises in the Gulf of Mexico. These activities will conclude the research activity of this project and demonstrate the utility of the insight gained through these research tasks. We will also work diligently on the journal publications stemming from this work.

References

- Clift, R., J. R. Grace, and M. E. Weber (1978), *Bubbles, drops, and particles*, Academic Press.
- Gros, J., C. M. Reddy, R. K. Nelson, S. A. Socolofsky, and J. S. Arey (2016), Simulating GasLiquidWater Partitioning and Fluid Properties of Petroleum under Pressure: Implications for Deep-Sea Blowouts, *Environ Sci Technol*, 50(14), 7397-7408.
- Medwin, H. and C. S. Clay (1998), *Fundamentals of Acoustical Oceanography*, Academic Press, Boston, 712 pp.
- Levine, J., I. Haljasmaa, R. Lynn, F. Shaffer, and R. P. Warzinski (2015), Detection of Hydrates on Gas Bubbles during a Subsea Oil/Gas Leak, NETL-TRS-6-2016, EPAAct Technical Report Series, U.S. Department of Energy, National Energy Technology Laboratory: Pittsburgh, PA.
- Rehder, G., I. Leifer, P. G. Brewer, G. Friederich, and E. T. Peltzer (2009), Controls on methane bubble dissolution inside and outside the hydrate stability field from open ocean field experiments and numerical modeling, *Marine Chemistry*, 114(1-2), 19–30.
- Römer, M., H. Sahling, T. Pape, G. Bohrmann, and V. Spiess (2012), Quantification of gas bubble emissions from submarine hydrocarbon seeps at the Makran continental margin (offshore Pakistan), *J Geophys Res-Oceans*, 117(19), 2011jc007424.
- Warzinski, R. P., F. Shaffer, R. Lynn, I. Haljasmaa, M. Schellhaas, B. J. Anderson, S. Velaga, I. Leifer, and J. Levine (2014a), The role of gas hydrates during the release and transport of well fluids in the deep ocean, DOI/BSEE Contract E12PG00051/M11PPG00053, Final Report, U.S. Department of Energy, National Energy Technology Laboratory.
- Warzinski, R. P., Lynn, R., Haljasmaa, I., Leifer, I., Shaffer, F., Anderson, B. J., and Levine, J. S.

Table 1: Milestones schedule and verification methods.

	Milestone	Comments
Title	Acquisition of NETL HPWT data	
Date Completed	March 24, 2017	
Verification Method	Email verification	
Title	Adapt Matlab code to NETL data	
Date Completed	September 28, 2017	
Verification Method	Report	
Title	Matlab code for M3 and EM-302 data	
Date Completed	September 29, 2017	
Verification Method	Report	
Title	Adapt seep model to NETL data	
Date Completed	June 19, 2018	
Verification Method	Report	
Title	Quantify seep model performance	
Date Completed	January 31, 2019	
Verification Method	Quarterly Reports and Data Archive	
Title	OTRC Experimental Report	
Date Completed	March 21, 2019	
Verification Method	Report	
Title	Quantify performance of acoustic models	
Date Completed	April 30, 2019	
Verification Method	Report	
Title	Complete model validation	Included in Project Timeline but not in Project Management Plan
Date Completed	April 30, 2019	
Verification Method	Quarterly Reports	

(2014b). Dynamic morphology of gas hydrate on a methane bubble in water: Observations and new insights for hydrate film models. *Geophys Res Lett*, *41*(19), 2014GL061665.

Weber, T. C., L. Mayer, K. Jerram, J. Beaudoin, Y. Rzhanov, and D. Lovalvo (2014), Acoustic estimates of methane gas flux from the seabed in a 6000 km² region in the northern gulf of mexico, *Geochemistry Geophysics Geosystems*, *15*(5), 1911–1925.

2 Products

2.1 Publications, Conference Papers, and Presentations

- Socolofsky, S. A., Kim, B., Kovalchuk, M., Levine, J., and Wang, B., “Mass transfer rates for hydrate-armored bubbles in the NETL High Pressure Water Tunnel,” Poster presented at the Gordon Research Conference on Natural Gas Hydrate Systems, Galveston, Texas, February 25 to March 2, 2018.
- Kim, B., Socolofsky, S. A., and Wang, B., “Hydrate formation time analyzed from data for NETL High Pressure Water Tunnel experiments,” Poster presented at the Gordon Research Conference on Natural Gas Hydrate Systems, Galveston, Texas, February 25 to March 2, 2018.

2.2 Websites or Other Internet Sites

The natural seep model used for this project, the Texas A&M Oilspill Calculator (TAMOC), is published via an open source code sharing service at:

<http://github.com/socolofs/tamoc>

2.3 Technologies or Techniques

Nothing to report.

2.4 Inventions, Patent Applications, and/or Licenses

Nothing to report.

2.5 Other Products

Nothing to report.

3 Participants and other collaborating organizations

3.1 Project Personnel

- 1. **Name:** Scott A. Socolofsky
- 2. **Project Role:** Principal Investigator

3. **Nearest person months worked during reporting period:** 1
 4. **Contribution to Project:** Overall project management and direction. Dr. Socolofsky has led the collection of the HPWT data, directed the data analysis methods, and completed all project reporting requirements.
 5. **Collaborated with individual in foreign country:** No
 6. **Travelled to foreign country:** No
- 1. **Name:** Binbin Wang
 - 2. **Project Role:** Co-Principal Investigator
 - 3. **Nearest person months worked during reporting period:** 2
 - 4. **Contribution to Project:** Analyzed the image data for the G08 cruise, created model for acoustic data from M3 sonar and EM-302 multibeam, and compared the measured data to model results from TAMOC. He also trained the Ph.D. student to begin analysis of the NETL HPWT data.
 - 5. **Collaborated with individual in foreign country:** No
 - 6. **Travelled to foreign country:** No
- 1. **Name:** Byungjin Kim
 - 2. **Project Role:** Ph.D. Student
 - 3. **Nearest person months worked during reporting period:** 3
 - 4. **Contribution to Project:** Organized the HPWT data, summarized the existing results from the NETL reports, and analyzed HPWT data for bubble size, hydrate formation time, and bubble interface mobility.
 - 5. **Collaborated with individual in foreign country:** No
 - 6. **Travelled to foreign country:** No
- 1. **Name:** Soobum Bae
 - 2. **Project Role:** Ph.D. Student
 - 3. **Nearest person months worked during reporting period:** 3
 - 4. **Contribution to Project:** Soobum Bae is working as an unfunded Ph.D. student to help analyze the HPWT data. He has helped to classify the video image data and to evaluate the hydrate equation of state.

5. **Collaborated with individual in foreign country:** No

6. **Travelled to foreign country:** No

1. **Name:** Inok Jun

2. **Project Role:** Post-doctoral Scholar

3. **Nearest person months worked during reporting period:** 1

4. **Contribution to Project:** Dr. Inok Jun has developed the correlation for hydrate formation time and the methods to identify the height of rise of natural seep flares in the oceans. Though funded from other sources, PI Socolofsky has directed her research to also benefit the present project.

5. **Collaborated with individual in foreign country:** No

6. **Travelled to foreign country:** No

3.2 Partner Organizations

None to report.

3.3 External Collaborators or Contacts

This project works in close collaboration with researchers in the DOE/NETL funded project “Fate of Methane in the Water Column,” led by the U.S. Geological Survey (USGS) in Woods Hole (Carolyn Ruppel), and with a new project led by the University of Rochester (John Kessler) to advance understanding of the environmental implications that methane leaking from dissociating gas hydrates could have on the ocean-atmosphere system. Dr. Socolofsky visits and communicates with researchers in these projects regularly and shares updates on work in progress. Accomplishments associated with these collaborations are detailed in Section 1.

4 Impact

None at this point.

5 Changes / Problems

Personnel. As reported in past quarterly reports, one adjustment from the proposed activities in the PMP is that a Ph.D. student (Byungjin Kim) was not hired to work on this project until the

second quarter of project Phase 1, instead of our original plan to hire a student in the first quarter. This occurred as it took time to complete contract negotiations and to effectively recruit a high-quality student to this project. Despite this delay in hiring, the project activities have remained on schedule. We anticipate that this hiring delay will result in the need for a short no-cost extension at the end of the project, which will allow B. Kim to complete his dissertation and the journal manuscripts stemming from the research conducted through this grant.

One other change is that we have had to delay the OTRC experiment (Task 5) both to benefit from a parallel effort of the Co-PI and because of closure of the lab during repairs to the filter pump in November through January. This was discussed in detail in past quarterly reports. The total delay for Task 5 was six months, which is similar to the delay outlined above with respect to the Ph.D. student hiring so that we expect to conclude all planned project tasks by the end of the short, no-cost extension expected above.

6 Special Reporting Requirements

None required.

7 Budgetary Information

Table 2 reports expenditures for Phase 1 of the project, and Table 3 for Phase 2. Table 4 summarizes expenditures for the current phase (Phase 3) of the project.

Table 2: Budget Report for Phase 1

Baseline Reporting Quarter	Budget Period 1							
	Q1		Q2		Q3		Q4	
	10/1/16 - 12/31/16		1/1/17 - 3/31/17		4/1/17 - 6/30/17		7/1/17 - 9/30/17	
DE-FE0028895	Q1	Cumulative Total	Q2	Cumulative Total	Q3	Cumulative Total	Q4	Cumulative Total
Baseline Cost Plan								
Federal Share	\$33,752	\$33,752	\$29,716	\$63,468	\$27,810	\$91,278	\$53,034	\$144,312
Non-Federal Share	\$12,029	\$12,029	\$12,029	\$24,058	\$8,019	\$32,077	\$4,009	\$36,086
Total Planned	\$45,781	\$45,781	\$41,745	\$87,526	\$35,829	\$123,355	\$57,043	\$180,398
Actual Incurred Cost								
Federal Share	\$11,037	\$11,037	\$22,617	\$33,654	\$25,957	\$ 59,610	\$ 69,499	\$129,110
Non-Federal Share	\$12,029	\$12,029	\$12,029	\$24,058	\$8,019	\$32,077	\$4,009	\$36,086
Total Incurred Costs	\$23,066	\$23,066	\$34,646	\$57,712	\$33,976	\$91,687	\$73,508	\$165,196
Variance								
Federal Share	\$-22,715	\$-22,715	\$-7,099	\$-29,814	\$-1,853	\$-31,668	\$16,465	\$-15,202
Non-Federal Share	\$0	\$0	\$0	\$0	\$0	\$0	\$0	\$0
Total Variance	\$-22,715	\$-22,715	\$-7,099	\$-29,814	\$-1,853	\$-31,668	\$16,465	\$-15,202

Table 3: Budget Report for Phase 2

Baseline Reporting Quarter	Budget Period 1							
	Q1		Q2		Q3		Q4	
	10/1/16 - 12/31/16		1/1/17 - 3/31/17		4/1/17 - 6/30/17		7/1/17 - 9/30/17	
DE-FE0028895	Q1	Cumulative Total	Q2	Cumulative Total	Q3	Cumulative Total	Q4	Cumulative Total
Baseline Cost Plan								
Federal Share	\$18,473	\$162,785	\$35,552	\$198,337	\$22,681	\$221,018	\$44,423	\$265,441
Non-Federal Share	\$10,125	\$46,221	\$10,125	\$56,336	\$6,750	\$ 63,086	\$ 3,374	\$66,460
Total Planned	\$28,598	\$208,996	\$45,677	\$254,673	\$29,431	\$ 284,104	\$47,797	\$331,901
Actual Incurred Cost								
Federal Share	\$29,427	\$158,537	\$29,427	\$187,964	\$28,798	\$216,762	\$16,441	\$233,204
Non-Federal Share	\$10,125	\$46,211	\$10,125	\$56,336	\$6,750	\$ 63,086	\$3,374	\$66,460
Total Incurred Costs	\$39,552	\$204,748	\$39,552	\$244,300	\$35,548	\$279,848	\$19,815	\$299,664
Variance								
Federal Share	\$10,954	\$-4,248.13	\$-6,125	\$-10,373	\$6,117	\$-4,256	\$-27,982	\$-32,238
Non-Federal Share	\$0	\$0	\$0	\$0	\$0	\$0	\$0	\$0
Total Variance	\$10,954	\$-4,248	\$-6,125,64	\$-10,373	\$6,117	\$-4,256	\$-27,982	\$-32,237

Table 4: Budget Report for Phase 3

Baseline Reporting Quarter	Budget Period 1							
	Q1		Q2		Q3		Q4	
	10/1/16 - 12/31/16		1/1/17 - 3/31/17		4/1/17 - 6/30/17		7/1/17 - 9/30/17	
DE-FE0028895	Q1	Cumulative Total	Q2	Cumulative Total	Q3	Cumulative Total	Q4	Cumulative Total
Baseline Cost Plan								
Federal Share	\$14,625	\$280,066	\$14,628	\$294,694	\$23,288	\$317,982	\$43,553	\$361,535
Non-Federal Share	\$8,012	\$74,472	\$8,012	\$82,484	\$5,342	\$87,826	\$2,671	\$90,497
Total Planned	\$22,637	\$354,538	\$22,640	\$377,178	\$28,630	\$405,808	\$46,224	\$452,032
Actual Incurred Cost								
Federal Share	\$13,668	\$246,872	\$28,289	\$275,161	\$	\$	\$	\$
Non-Federal Share	\$8,012	\$74,472	\$8,012	\$82,484	\$	\$	\$	\$
Total Incurred Costs	\$21,680	\$321,344	\$36,301	\$357,645	\$	\$	\$	\$
Variance								
Federal Share	\$-957	\$-33,194	\$13,661	\$-19,533	\$	\$	\$	\$
Non-Federal Share	\$0	\$0	\$0	\$0	\$	\$	\$	\$
Total Variance	\$-957	\$-33,194	\$13,661	\$-19,533	\$	\$	\$	\$

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