Mapping observations using AUV and numerical simulations of leaked CO₂ diffusion in sub-seabed CO₂ release experiment at Ardmucknish Bay

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Abstract

A controlled sub-seabed CO₂ release experiment (QICS: Quantifying and Monitoring Potential Impacts of Geological Carbon Storage) was conducted in Ardmucknish Bay, Scotland, in 2012, to quantify the effect of leaked CO₂ on marine environments. In this experiment, CO₂ was injected beneath the seafloor, and closely monitored as the CO₂ was allowed to leak into the overlying water column. We performed mapping observations using an autonomous underwater vehicle (AUV) equipped with sensors to monitor physical and chemical conditions in the vicinity of the CO₂ leakage. We also numerically simulated the behavior of the low-pH plume caused by the CO₂ leakage, within the calculated tidal current in the bay.

The results of AUV mapping observations showed that physical and chemical characteristics of the water mass in Ardmucknish bay are sensitive to tidal variations (low or high tide), and the ascent speed of the leaked CO₂ bubble and the pH decrease in the vicinity of the CO₂ leakage area are predominantly controlled by the tidal phase in the bay. The observations using an acoustic Doppler current profiler (ADCP) installed in the AUV indicated that the current in the bay flows westwards at high tide and eastwards at low tide. The simulated tidal current agreed well with the tidal current observed by ADCP. Numerical simulation suggested that the pH decrease due to the leaked CO₂ is restricted to the vicinity of the CO₂ leakage area, and fluctuates greatly with tidal variations. In flood tide periods, diffusion of the low-pH plume caused by the leaked CO₂ is barely detectable because low-pH seawater intrudes into the bay from the bay mouth.

1. Introduction

Carbon capture and storage (CCS) has gained attention in recent years as a method for curbing atmospheric emissions of CO₂. Initial experiments on CCS using aquifers were conducted in 1996 within the Sleipner region (Norway) of the North Sea oilfields (Baklid et al., 1996). Similarly, large-scale geological carbon storage was examined in the Gorgon Project (Australia; 2008 onwards) and the Midwest Geological Sequestration Consortium (United States; 2011 onwards). However, a common characteristic of these projects is that they focus on the verification of CO₂ capture and storage techniques (Singh et al., 2010).

There is some concern that CO₂ leakage from sub-seabed storage reservoirs may reduce the pH of seawater near the storage site. Few investigations or observations have been conducted to investigate the impact of a pH decrease on the surrounding marine environment. In Japan, CCS-related research on this issue has focused on the natural analogue provided the release of CO₂ into the ocean from hydrothermal areas of the seafloor (Shitashima et al., 2008; Shitashima et al., 2013).

In 2012, in the United Kingdom, the Quantifying and Monitoring Potential Impacts of Geological Carbon Storage (QICS) project was organized, representing the world's first attempt to examine the effects of CO₂ leakage on marine environments by artificially injecting CO₂ gas into the sub-seabed (<http://www.bgs.ac.uk/qics/home.html>). The QICS experiment, the first controlled CO₂ sub-seabed release experiment, was conducted in Ardmucknish Bay, Scotland, in May–September, 2012. In the QICS experiment, we performed mapping observations using an autonomous underwater vehicle (AUV), equipped with sensors, in the vicinity of the CO₂ leakage area. This paper focuses on understanding the environmental conditions of the overlying water column in the CO₂ leakage area, and predicting the behavior of the

low-pH plume caused by the leaked CO₂ by numerical simulation.

2. Methods

2.1. CO₂ leakage experiments

The QICS experiment site is located in Ardmucknish Bay, near Oban in western Scotland (Figure 1). CO₂ injections began on May 17, 2012, and ended on June 23, 2012. The injection rate of CO₂ gas into the sub-seabed during the CO₂ release period is given in the Table 1. Over the first 16 days, the injection rate was approximately 80 kg/day; over the next 10 days, 150 kg/day; and over the final 5 days, 200 kg/day. The total CO₂ injection amount over the experiment period was approximately 4.2 tons (Blackford et al., 2014).

2.2. Observation equipment

We used an AUV equipped with a variety of physical and chemical sensors to observe environmental parameters at the experiment site. Photographs of the research vessel and the AUV used in this series of experiments are shown in Figure 2. The following physical and chemical sensors were used: an acoustic Doppler current profiler (ADCP: Teledyne RD Instruments); a side-scan sonar (Marine Sonic Technology); a G-CTD (Neil Brown Ocean Sensors, Inc.) for salinity, temperature, and depth measurements; an oxygen optode (Aanderaa Data Instruments) for dissolved oxygen measurements; an FLNTU ECO Puck (WET Labs) for chlorophyll-a and turbidity measurements; and an ion-selective field-effect transistor-based in-situ pH/pCO₂/oxidation–reduction potential sensor (Shitashima et al., 2013).

The AUV mapping observations were carried out over a 300 × 700 m rectangle and a 190 × 270 m rectangle around the CO₂ leakage area (Figure 3). The larger (300 × 700

m) rectangle contains 11 grid lines (25 m spacing), which were traversed on May 23 by the AUV at a cruising speed of approximately 3 knots and at a depth of 3 m. The smaller (190 × 270 m) rectangle contains 20 grid lines (10 m spacing), which were traversed, at a cruising speed of approximately 1.5 knots, by the AUV on May 24 and 25, and on May 20, 21, and 26 at depths of 2m and 4 m above the seafloor, respectively.

The AUV mapping observation time for one layer is approximately 2 hours, and observations were set to start about 1 hour before high tide or low tide, and finish about 1 hour after low tide or high tide, respectively. The mapping observations in flood tide and ebb tide were held during roughly 2 hours between high tide and low tide. The tidal pattern of the bay and the AUV observation schedule are shown in Figure 4. From the AUV mapping observations, data on the bathymetry of the CO₂ leakage site, tidal currents at the surface and near the seafloor, temperature, and salinity were collected for this study.

3. Results and discussion

3.1. AUV mapping observation

Figure 5 shows horizontal distributions of temperature and salinity for the larger rectangle area (300×700 m) at a depth of 3 m during a low tide period on May 23. Horizontal distributions for the small rectangle area (190 × 270 m) at a depth of 2 m above the seafloor during a flood tide period and a high tide period on June 21 are indicated in Figure 6. During the flood tide period, water mass of low temperature and high salinity flows into the inner part of the bay from the offshore. During the subsequent high tide period, the intruded water mass mixes with seawater in the inner part of the bay, forming uniform water mass.

3.2. Tidal change and seawater flow in Ardmucknish Bay

As an example, the flow direction and velocity in the surface and bottom layers, observed at high and low tides on June 26 in Ardmucknish Bay, are displayed in Table 2 and Figure 7. The AUV mapping observation was carried out during four tidal phases; high tide, ebb tide, low tide, and flood tide.

A westwards flow was observed in both surface layer (average: 4.6 cm/s at 264.1°) and bottom layer (average: 4.1 cm/s at 252.7°) at high tide. At ebb tide, slow westwards flow was detected in both surface layer (average: 2.0 cm/s at 289.1°) and bottom layers (average: 1.9 cm/s at 297.6°). At low tide, relatively rapid flows running eastwards in the surface layer (average: 9.1 cm/s at 95.0°) and south-eastwards in the bottom layer (average: 10.4 cm/s at 123.9°) were observed. At flood tide, the flow was south-eastwards in the surface layer (average: 6.6 cm/s at 119.5°) and north-westwards in the bottom layer (average: 5.7 cm/s at 292.5°). At the CO₂ leakage site, the flow during low tide was rapid and flowed eastwards, but the flow during high tide was slow and flowed westwards.

Based on these results and the bathymetry of Ardmucknish Bay, we estimated the current patterns of the bay during flood and ebb tides (Figure 8). During flood tides, the seawater flows into the inside of the bay from the offshore area, and reaches the inner part of the bay. A part of this seawater advances toward Loch Etive. Conversely, during ebb tides, seawater from the inner part of the bay flows out towards the bay mouth with an inflow of the fresh water from Loch Etive. The current of the inner part of the bay (the area around the experiment site) is westward-flowing at high tide and eastward-flowing at low tide.

4. Flow model and low-pH plume model

The Princeton Ocean Model (POM; Blumberg and Mellor, 1983, 1987) was applied to create flow and low-pH plume models. The POM consists of a series of three-dimensional primitive equations, uses the Mellor-Yamada level 2.5 turbulent closure model (Mellor-Yamada, 1982; Galperin et al., 1988) for vertical eddy coefficients, and a Smagorinsky-type parameterization for horizontal mixing with the Boussinesq approximation and hydrostatic assumption. In general, the governing equations in POM are converted from three-dimensional coordinate into the orthogonal curvilinear coordinate (horizontal) and σ -coordinate (vertical) to smoothly follow complex terrain geometries. However, because σ -coordinate system is insufficient to the representation of the diffusion of scalar quantity in order to use the coordinate transformation, three-dimensional Cartesian coordinates was employed to simplify our model calculation in this study. Two-dimension data (the Z-axis and X-axis, the Z-axis and Y-axis) was calculated by using the MATLAB code of Benjamin (2008). This code is used for incompressible and viscous flows, and is simple numerical method for solving the Navier Stokes equations. In the Navier Stokes equations of Benjamin (2008), finite difference calculus is employed to solve the diffusion, continuity, and Navier Stokes equations (under the Boussinesq approximation, assuming hydrostatic equilibrium and a free surface). The calculated vertical flow is then re-calculated using the horizontal flow velocity in the POM model.

Shitashima et al. (2008) reported that the behavior of low-pH plume was similar to that of high-temperature plume in the deep-sea hydrothermal systems. Therefore, an advection-diffusion equation of temperature and salinity was applied to calculate the low-pH plume behavior in this study.

4.1. Initial conditions and boundary conditions

The computational domain and seafloor topography of the model are shown in Figure 9. As the non-dimensional numbers to completely determine the flow, Reynolds number= 1.0×10^3 and Prandtl number=9.2 are used. The seafloor is defined as a step-wise topography, and the computational grid consists of $143 \times 143 \times 50$. The external force used to drive the model is tidal only, and wind is not considered. Harmonic analysis of the tide is performed using hourly predicted values for the experimental period from the tide table for Oban Bay (Calet Fisheries Ltd., 2012). Harmonic analysis is based on the method of Boon (2007). The size and ascent velocity of the leaked CO₂ bubbles can be qualitatively predicted using the Allen method and/or the Newton method, and calculated sizes and ascent velocities are provided in Table 3.

Video camera observations during the experiment showed that the ascent speed of bubbles leaking from the experiment site ranged from 18.2 to 36.4 cm/s, and the diameter of the rising CO₂ bubbles was predominantly ≤ 0.5 cm (Figure.10). The calculated ascent speeds for a 0.5 cm diameter bubble were 18.5 cm/s by the Allen method, and 16.4 cm/s by the Newton method; these speeds are in close agreement with the estimated ascent speeds from video camera observations.

When the ascent speed of the leaked CO₂ bubbles was approximately 18 cm/s, bubbles escaped to the atmosphere from the seafloor (12 m deep) in approximately one minute. In our numerical calculation, we used 20 cm/s as the initial ascent speed of the low-pH plume. We estimated the initial seawater pH in the CO₂ leakage area from the fugacity of CO₂, calculated using the equation of state for CO₂, and the equation provided by Wiss (1974). Assuming the first dissociation constant of carbonic acid in seawater (K_s^+) to be 6.8×10^{-7} , the pressure correction coefficient at 1.2 atm to be 1.01, and X[HCO₃] to be the alkalinity of seawater, we calculated the seawater pH by

$K_s^+ = [H^+][HCO_3^-]/[H_2CO_3]$ with $[HCO_3^-] = 2.4 \times 10^{-3}$ (mol/l), $[H_2CO_3] = 0.27$ (mol/l), and $K_s^+ = 6.8 \times 10^{-7}$ (mol/l•atm), which leads the pH is 4.1 when $[H^+] = 0.79 \times 10^{-4}$.

The calculated result for seawater pH, at a depth of 12 m near the CO₂ leakage area, suggested that pH decreases to approximately 4.1. Field observation results of in-situ pH at the CO₂ leakage area by the in-situ pH sensor showed that the pH decreased to approximately 5 – 6 (Shitashima et al., this issue). Based on these results, we took a pH of 5 as the initial pH value in the CO₂ leakage area for our numerical simulation.

4.2. Simulation results

4.2.1. Comparison between simulated flow and observed flow

The simulated horizontal vector diagrams for the surface layer at low tide and high tide are shown in Figure 11. For comparison, the average flows in the surface layer observed by the ADCP are also indicated in Figure 11. An eastwards flow of several cm/s in the surface layer at low tide was simulated. The observed flow of the 2 and 4 m layers above the seafloor were 96° and 8 cm/s, and 11° and 6 cm/s, respectively. Additionally, a westwards flow of several cm/s in the surface layer at high tide was simulated. The observed flow of the 2 and 4 m layers above the seafloor were 264° and 4 cm/s, and 293° and 2 cm/s, respectively.

The simulated flow at low tide and high tide agree well with values observed by the ADCP, indicating that our simulations were suitable for this application. In addition, this numerical simulation demonstrated that the lags of flow velocity and flow direction were created in each layer owing to topography effects and the timing of tidal fluctuations.

4.2.2. Simulated distributions of the low-pH plume

We estimated the behavior of the low-pH plume due to the leaked CO₂ using calculated flows. Figure 12 shows contour maps of the low-pH plume produced by continual release of low-pH seawater (pH 5) from the leakage area, with a rising speed of 20 cm/s. Contour maps consist of six horizontal sections (water depths of 6, 8, 10, 12, and 16 m) simulated for 06:00 (local time) on June 21 (high tide). The low-pH plume does not appear in the 6 m layer, but appears in the layers deeper than 8 m. The low-pH plume is confined within a narrow area, limited to the vicinity of the CO₂ leakage point, and the maximum decrease in pH (i.e., ΔpH) is only about 0.2 pH units. Figure 13 demonstrates the vertical distribution of simulated pH under the same conditions as for the horizontal contour map. The distribution of the low-pH plume is restricted to near the CO₂ leakage point, and the plume has not reached the surface. Moreover, pH only decreased around the CO₂ leakage point, suggesting that the low-pH plume does not diffuse to a wide area.

In Ardmucknish Bay, the pH in the deep layer at the bay mouth is already low, and this low-pH water is transported into the inner part of the bay during flood tide periods. Therefore, even if CO₂ leaked continuously, the pH of the surrounding seawater is already low enough to cause difficulties in detecting additional low-pH water, particularly during high tide periods.

5. Conclusions

The temperature, salinity, and other characteristics of seawater in Ardmucknish Bay varied with tidal periodicity. From flood tide to high tide, offshore seawater characterized by high salinity, low temperatures, and low-pH flows into the bay. Our results suggest that a seawater pH change due to leakage of CO₂ in the bay is barely detectable, although this is dependent on the volume of the CO₂ leakage and the

contact time of the CO₂ bubbles with seawater. Nonetheless, a pH decrease caused by the leaked CO₂ was observed during the low tide period. This observation agrees with the result reported by Kita et al. (this issue); specifically, the strong CO₂ bubble ejection was observed by video camera during the low tide period, but not during the high tide period. In other words, CO₂ leakage was weak at high tide and strong at low tide, and this result is attributable to variations in pressure balance with tidal periodicity.

As estimated from video camera observations and calculation, the diameter of the majority of CO₂ bubbles produced in this experiment was 0.5 cm or less, and diameters over 1 cm were rarely observed. Bubble size is an important parameter because it affects the dissolution rate of bubbles in seawater. Kano et al. (2009) conducted calculations with a two-phase flow model (micro-scale domain) and the MEC Ocean Model (meso-scale domain), using an assumed average bubble diameter of 2 cm. Mori et al. (this issue) numerically predicted the fate of CO₂ bubbles for two potential bubble diameters of 1 and 2 cm.

We simulated a slight low-pH plume or pH decrease in the vicinity of the leakage area with a bubble diameter of 0.5 cm. Because the ascent speed of the CO₂ bubbles is roughly 13 – 36 cm/s, bubbles leaked from a 12 m deep seafloor are expelled into the atmosphere within about 60 s. The seawater contact time of the leaked CO₂ bubbles during ascent to surface is very short, and bubbles may escape into the atmosphere without dissolution of all the bubbles in seawater. However, some of the CO₂ injected into the sub-seabed potentially remains in sediments, since it is unlikely that all the injected CO₂ dissipated into the atmosphere. Mori et al. (this issue) estimated that some of the injected CO₂ escapes into the atmosphere in a gaseous phase, alongside some dissolution in seawater, and some (approximately 10% to 40%) remains within

the sediment at the injection area.

In this study, we focused on the behavior of the low-pH plume caused by the leaked CO₂, and simulated the diffusion of the plume. Some uncertainties remain, such as the volume of leaked CO₂ from the sub-seabed, the seawater contact time of the ascending CO₂ bubble, and the dissolution rate of the CO₂ bubble into seawater. Accordingly, it is important to combine assessments of both data collection, through on-site monitoring, and numerical analysis.

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References

- Baklid, A., Korbøl, R., Owen, G., 1996: Sleipner west CO₂ disposal, CO₂ injection into a shallow underground aquifer. SPE 36600, 269-277.
- Benjamin, S., 2008: A Compact and fast Matlab code solving the incompressible

301 Nervier Stokes equation on rectangular domain. www-math.mit.edu/, March 31,
 302 2008

303 Blackford, J.C., Stahl, H., Bull. J.M., Berges. B.J.P., Cevatoglu. M., Lichtschlag. A.,
 304 Connelly D., James R.H., Kita, J., Long, D., Naylor, M., Shitashima, K., Smith, D.,
 305 Taylor, P., Wright, I., Akhurst, M., Chen, B., Gernon, T.M., Hauton, C., Hayashi,
 306 M., Kaieda, H., Leighton, T.G., Sato, T., Sayer, M.D.J., Suzumura, M., Tait, K.,
 307 Vardy, M.E., White, P.R., Widdicombe S., 2014: Detection and impacts of leakage
 308 from sub-seafloor Carbon Dioxide Storage. Nature Climate Change.
 309 doi:10.1038/nclimate2381

310 Blumberg, A.F. and Mellor, G.L., 1983: Diagnostic and prognostic numerical
 311 circulation studies of the South Atlantic Bight. Journal of Geophysical Research, 88,
 312 4579-4592.

313 Blumberg, A.F. and Mellor, G.L., 1987: A description of a three-dimensional coastal
 314 ocean circulation model. In Three-Dimensional Coastal Ocean Model. Vol.4, edited
 315 by N Heaps. pp.208. American Geophysical Union, Washington, D.C., 1987.

316 Boon, J., 2007: World Tides User manual. Gloucester: John Boon Marine Consultant.
 317 [http://mathworks.com/matlabcentral/fileexchange/24217-world-tides/content/WOR](http://mathworks.com/matlabcentral/fileexchange/24217-world-tides/content/WORLD%20TIDES%20USER%20MANUAL.pdf)
 318 [LD%20TIDES%20USER%20MANUAL.pdf](http://mathworks.com/matlabcentral/fileexchange/24217-world-tides/content/WORLD%20TIDES%20USER%20MANUAL.pdf)

319 Galperin, B., Kantha, L.H., Hassid, S., Rosati, A., 1988: A quasi-equilibrium turbulent
 320 energy model for geophysical flows. J. Atmos. Sci., 45, 55-62.

321 Kano, Y., Sato, T., Kita, J., Hirabayashi, S., Tabeta, S., 2009: Model prediction on the
 322 rise of pCO₂ in uniform flows by leakage of CO₂ purposefully stored under the
 323 seabed. Int.J.Greenhouse Gas Control, 3, 617-625.

324 Kano, Y., Sato, T., Kita, J., Hirabayashi, S., Tabeta, S., 2010: Multi-scale modelling of
 325 CO₂ dispersion leaked from seafloor off Japanese coast. Marine Pollution Bulletin,

326 50, 215-224.

327 Kita, J., Watanabe, Y., Hayashi, M., Stahl, H., this issue: Benthic megafauna and CO₂

328 bubble dynamics observed by underwater photography during a controlled sub-seabed

329 release of CO₂. This issue.

330 Mellor, G.L. and Yamada, T., 1982: Development of a turbulence closure model for

331 geophysical fluid problems. Rev. Geophys. Space Phys., 20, 851-875.

332 Mori, C., Sato, T., Kano, Y., Oyama, H., Dmitry, A., Tsumune, D., Maeda, Y., :

333 Numerical study of the fate of CO₂ purposefully injected into the sediment and

334 seeping from seafloor in Ardmucknish Bay. This issue.

335 Shitashima, K., Maeda, Y., Koike, Y., Ohsumi, T., 2008: Natural analogue of the rise

336 and dissolution of liquid CO₂ in the ocean. International journal of greenhouse gas

337 control, 2, 95-104.

338 Shitashima, K., Maeda, Y., Ohsumi, T., 2013: Development of detection and

339 monitoring techniques of CO₂ leakage from seafloor in sub-seabed CO₂ storage.

340 Applied Geochemistry, 30, 114-124.

341 Shitashima, K., Maeda, Y., Sakamoto, A., : Detection and monitoring of leaked CO₂

342 through sediment, water column and atmosphere in sub-seabed CCS experiment.

343 This issue.

344 Singh, V., Cavanagh, A., Hansen, H., Nazarian, B., Iding, M., Ringrose, P., 2010:

345 Reservoir modeling of CO₂ plume behavior calibrated against monitoring data from

346 Sleipner, Norway. SPE paper 134891 presented at the SPE Annual Technical

347 Conference and Exhibition held in Florence, Italy, 19–22 September 2010

348 Weiss, R.F., 1974: Carbon dioxide in water and seawater. The solubility of a non-ideal

349 gas. Marine Chemistry, 2, 203-215.

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352 **Captions**

353

354 Figure 1: Location of the Quantifying and Monitoring Potential Impacts of Geological
355 Carbon Storage (QICS) CO₂ release experiment.

356 Figure 2: Photographs of the research vessel (R.V. *Seòl Mara*) and the autonomous
357 underwater vehicle (AUV).

358 Figure 3: Observation area (coarse grid and fine grid) of the AUV.

359 Figure 4: Schedule of AUV mapping observations in relation to tidal patterns.

360 Figure 5: Horizontal distributions of temperature and salinity during the CO₂ release
361 experiment (May 23).

362 Figure 6: Horizontal distributions of temperature and salinity at flood tide and high
363 tide. The observation depth is 2 m from the seafloor (June 20 and 21).

364 Figure 7: Distribution of the flow direction and velocity in the surface and bottom
365 layers observed at high and low tide. The average speed and direction of
366 flow observed in AUV mapping are depicted by white arrows (vectors).

367 Figure 8: The estimated flow pattern in Ardmucknish Bay during flood tide and ebb
368 tide.

369 Figure 9: The computational domain and seafloor topography of Ardmucknish Bay.

370 Figure 10: Estimation of ascent speed for the leaked CO₂ bubbles.

371 Figure 11: Calculated horizontal vector diagrams in the surface layer, at low tide and
372 high tide, in Ardmucknish Bay at 06:00 (LT) on June 21.

373 Figure 12: Contour maps of calculated pH for horizontal sections, at various depths, at
374 the CO₂ leakage area in Ardmucknish Bay at 06:00 (LT) on June 21.

375 Figure 13: Vertical distribution of calculated pH values at the CO₂ leakage area in

376 Ardmucknish Bay at 06:00 (LT) on June 21.

377

378 Table 1: Injection rate of CO₂ gas into the sub-seabed during the CO₂ release period.

379 Table 2: Average flow collected by an acoustic Doppler current profiler (ADCP) in the

380 AUV mapping observation.

381 Table 3: Calculated size and ascent velocity for CO₂ bubbles using the Allen and

382 Newton methods.

Table. 1

	D1-D2	D4-D19	D21-D31	D32-D36	mean
(Standard liter/min)	8	32	58	80	44.46
(kg/day)	19	80	148	200	112.21

Table. 2

	Surface Current						Bottom Current						
Date & Time(GMT)	E-W component		N-S component		Vector		E-W component		N-S component		Vector		Remark
	average	RMS	average	RMS	flow	mean	average	RMS	average	RMS	flow	mean	
	(cm/sec)	(cm/sec)	(cm/sec)	(cm/sec)	(deg)	(cm/sec)	(cm/sec)	(cm/sec)	(cm/sec)	(cm/sec)	(deg)	(cm/sec)	
2012/6/26 14:00-15:00	8.83	38.83	-0.82	8.84	95.28	8.87	9.44	37.10	-0.82	9.94	94.95	9.47	Low tide Altitude 2m
2012/6/26 14:00-15:00	5.81	6.93	-3.20	4.07	118.85	6.63	-4.99	6.07	-3.20	3.24	237.34	5.93	from Low tide to High tide(Flood tide) Altitude 4m
2012/6/26 14:00-15:00	-3.96	33.06	-0.47	8.59	263.28	3.99	-3.56	31.03	-0.47	7.61	262.53	3.59	High tide Altitude 2m
2012/6/26 14:00-15:00	-1.50	3.09	0.64	1.73	292.94	1.63	-1.46	2.91	0.64	1.45	293.47	1.59	from High tide to Low tide(Ebb tide) Altitude 4m

Table. 3

diameter	Alen's Law	Newton's Law
0.5cm	18.5cm/s	16.4cm/s
1.0cm	37.0cm/s	23.2cm/s
1.5cm	55.5cm/s	28.5cm/s

Figure.1

Figure.2

Figure.3

Figure.4

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Figure.6

Figure.7

Figure.8

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Figure.10

Figure.11

Figure.12

Figure.13