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**Libe Washburn and Jordan F. Clark**

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# **Measurement of Hydrocarbon Fluxes due to Natural Seepage in the Northern Santa Barbara Channel**

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## **Abstract**

We describe the design, calibration, and deployment of a buoy and gas capture assembly for measuring bubbling gas flux in oceans and lakes. The assembly collects gas in a chamber while continuously measuring the position of the gas-water interface that forms as gas accumulates. Interface position is determined from the differential pressure between the chamber and ambient seawater. A spar buoy provides flotation and stability to reduce vertical motions due to surface waves. The gas collection assembly and spar, referred to as a flux buoy, is suitable for deployment from small boats under conditions of light wind and small waves. We are using the flux buoy to determine the spatial distribution of natural hydrocarbon seepage off the south-central California coast. Hydrocarbon seepage from continental shelves may be an important source of atmospheric methane.

## 1. Introduction

In some water bodies bubbles originating at the sea floor contribute significantly to the total flux of methane and other gasses to the local atmosphere. For example, methane generated by thermogenic and bacterial processes beneath the sea floor (Hovland et al. 1993) can form plumes of rising bubbles extending to the sea surface. This occurs strongly offshore of Coal Oil Pt. near Santa Barbara, California where natural hydrocarbon seepage produces extensive, dense bubble plumes (Fischer 1977; Hornafius et al. 1999; Quigley 1997; Quigley et al. 1999). Natural hydrocarbon seeps occur elsewhere on continental shelves including the Gulf of Mexico (MacDonald 1998), the Gulf of California (Simoneit et al. 1990), and the North Sea (Hovland et al. 1993). Gaseous marine hydrocarbon seepage is potentially an important source of atmospheric methane (Hovland et al. 1993).

Bubbling gas from sediments also contributes to the atmospheric flux of methane and other gasses from lakes. Crill et al. (1988) found that methane flux in an Amazonian floodplain lake from bubbles was much greater than the diffusive flux and comprised most of the total flux. In Mono Lake, California natural gas seeps produces strong, localized methane flux, accounting for about 7% of the total flux over the lake surface (Oremland et al. 1987; Romero, 1996). An extreme example of bubbling gas flux occurred in Lake Nyos, Cameroon during a deadly out-gassing of CO<sub>2</sub> in August, 1986 (Freeth and Kay 1987; Kanari 1989). Woods and Phillips (1999) use numerical simulations to examine how turbulent bubble plumes might produce large CO<sub>2</sub> fluxes to trigger gas eruptions in lakes.

Quantifying the bubbling flux of methane and other gasses to the atmosphere from oceans and lakes is important for estimating global budgets. However, some sources such as natural marine seepage are poorly constrained due to a lack of measurements. To estimate the hydrocarbon seepage contribution to the global methane budget, it is assumed that the spatial distribution of seeps is lognormal, but this has not been verified by observation (Hovland et al. 1993). Few systems for direct measurement of bubbling gas flux have been reported. One example, developed by Sebacher and Harriss (1982), used a floating gas collection chamber and detector system for measuring the diffusive and bubbling flux of methane in

lakes and protected waters. This instrument was used in the study by Crill et al. (1988). Oremland et al. 1987 measured methane flux from natural gas seeps in Mono Lake by timing the capture of gas in a collection bottle. Our instrument works on a similar principal, but is more suitable for measuring flux over a large field of seepage because it is automated.

We describe the design, calibration, and deployment of an instrument for measuring gas flux to the ocean's surface due to rising bubble plumes such as arise from marine hydrocarbon seeps. The instrument (hereafter referred to as a flux buoy) measures flux using a gas capture technique. A primary goal is to quantify the spatial distribution of methane seepage over continental shelves. Because our study area is the region of very strong seepage off Coal Oil Pt., California, the instrument is configured to measure high fluxes. However, the basic design may be readily adapted for use in environments with much weaker gas flux.

The rest of the paper is organized as follows: Section 2 describes the flux buoy design and operation; field deployments are described in Section 2; laboratory calibrations in Section 3; and results from field trials in Section 4. Section 5 summarized the instrument characteristics and performance.

## 2. Buoy design, construction, and operation

The flux buoy measures gas flux by capturing gas bubbles a few m below the sea surface. Bubbles from rising plumes are directed into a collection chamber through a circular cone at the base of the flux buoy (Fig. 1). As gas accumulates in the collection chamber, a gas-water interface forms, then descends at a rate proportional to the volume flux of gas. The pressure difference  $\Delta p$  between the inside of the collection chamber and surrounding seawater (hereafter referred to as differential pressure) is,

$$\Delta p = (\rho - \rho_g)gh \approx \rho gh \quad (1)$$

where  $\rho$  is the seawater density,  $\rho_g$  is the density of the gas in the chamber ( $\rho_g \ll \rho$ ),  $g$  is the acceleration of gravity, and  $h$  is the distance between the gas-water interface measured and the top of the collection

chamber. The volume flux of gas per unit area of sea surface, adjusted to standard temperature  $T_a$  and pressure  $p_a$ , is,

$$\dot{q} = (\rho g)^{-1} \frac{\partial \Delta p}{\partial t} \frac{T_a}{T_c} \frac{p_c}{p_a} \frac{A_c}{A_{co}} \quad (2)$$

where  $\partial \Delta p / \partial t$  is the time rate of change of differential pressure,  $T_c$  and  $p_c$  are temperature and pressure of the gas in the collection chamber, and  $A_{co}$  is the area of the collecting cone ( $0.27 \text{ m}^2$ ). Ideal gas behavior has been assumed.

Gas accumulates in the chamber until an adjustable threshold  $\Delta p$  is reached. Then a microcomputer-controlled valve opens to release the accumulated gas and start a new measurement cycle. Gas vents through a port at the base of the electronics pressure case that also contains the sensors, electronics and microcomputer (Fig. 1). The buoyancy loss due to the small volume of gas vented from the chamber does not significantly affect the buoy's vertical position in the water. The threshold  $\Delta p$  is set to prevent filling the chamber and spilling gas out the open bottom. The valve can also be opened on command at any time from the research boat.

Data acquisition, valve operation, and data transmission are controlled by a Blue Earth Research Micro485 Micro-controller. This small computer includes four 12-bit A/D channels (0 to 5 volts range), battery backed-up static random access memory (RAM), clock, digital input/output, RS422 serial communications, and a BASIC interpreter. The control program can be uploaded to the computer via the serial communication link and stored in RAM. The program runs automatically upon power up, a useful characteristic because the electronics are sealed in the electronics pressure case.

Gas vents through a tube (Fig. 2) opened by a 12V solenoid valve (Automatic Switch Company, model 8030B-12V) with an opening large enough to empty the collection chamber in less than 1 s. The micro-controller opens the valve through a MOSFET switch when the vent  $\Delta p$  threshold is exceeded.



Finding a suitable differential pressure transducer proved difficult because of the requirements for corrosion resistance for use in seawater and high sensitivity for resolving small pressure differences. The first sensor tried used two gauge-type transducers with differential electronics. Tests in a swimming pool revealed that this sensor had poor stability and was unsuitable. A second sensor using a variable reluctance scheme performed successfully. Table 1 summarizes the characteristics of this sensor and the other pressure and temperature transducers used on the flux buoy; Fig. 2 shows their arrangement in the electronics pressure case.

The electronics pressure case is a polyvinyl chloride (PVC) cylinder (pipe section) 0.25 m in diameter and 0.26 long with a wall thickness of 7 mm. It is capped on both ends with PVC plates 24 mm thick. The lower plate (base plate, shown in Fig. 2) is square; the upper plate is octagonal; and both are 0.31 m across. O-rings seal the plates on the ends of cylinder. Sections of threaded rod 7 mm in diameter extend along the cylinder through the plates; nuts on the threaded rods compress the plates to seal the pressure case. A clear Plexiglas window on the upper plate permits inspection of the sensors and electronics without opening the pressure case. A bulkhead connector penetrates the upper plate and connects the data transmission cable to the electronics. Ports in the base plate expose the temperature sensor, absolute pressure sensor, and one side of the differential pressure sensor to gas and seawater at the top of the collection chamber. Another port through the base plate exposes the other side of the differential pressure sensor to seawater pressure outside the collection chamber.

The collection chamber, made of clear polycarbonate plastic, is cemented to a PVC flange; the flange attaches with bolts threaded into the bottom of the base plate. An O-ring seals the flange to the base plate to prevent gas leakage. The chamber can be changed easily in the field to accommodate different flux rates. Two chambers have been used in our field experiment with areas  $A_c = 1.6 \times 10^{-3}$  and  $7.1 \times 10^{-3} \text{ m}^2$ ; the larger chamber is used in areas with the highest volume flux. The cone for directing gas into the collection chamber is 0.58 m in diameter and made of high-density polyethylene plastic. It was cut from a liquid storage tank with a conical bottom. The cone diameter is much larger than seep bubble

diameters in the plumes (typically  $\leq 1$  cm; (Quigley 1997)). A small metal cone is attached to the apex of the plastic cone with its outlet at the base of the collection chamber. Four sections of threaded rod 0.97 m long and 12 mm in diameter join the cone to the gas capture assembly (pressure case, electronics, sensors, collection chamber). They extend from four brackets on the cone, through the base plate of the pressure case, to 0.11 m above the pressure case. Tensioned cables diagonally connect the threaded rods to stiffen the gas capture assembly. The spar buoy attaches to the upper ends of these rods.

The spar buoy is constructed from PVC pipe 0.09 m in diameter and 3.1 m long. The upper end of the pipe is sealed with a cap and the lower end with a PVC flange mounted on a fiberglass plate. Holes in the fiberglass plate engage the four sections of threaded rod extending above the pressure case. A collar of closed cell foam mounted on the spar buoy provides additional flotation (Fig. 1). Four dive weights totaling 18 kg are attached to the cone at the base of the buoy for ballast.

The long, narrow configuration of the flux buoy dampens motions due to small amplitude, high frequency surface waves. Its natural period of oscillation is 14 s as determined in a series of “bounce tests” in a swimming pool. This is longer than the periods of waves encountered during field deployments (5-8 s) conducted in conditions of light wind and low swell. During field deployment about 1.3 m of the buoy extends above the sea surface. The top of the collection chamber is about 2.6 m below the sea surface.

Buoy position is logged every 2 s with a differential Global Positioning System (GPS) receiver (GPS receiver, Garmin model 45; differential beacon receiver, Garmin model GBR21) mounted near the top of the buoy in a waterproof box (Fig. 1). Antennas for receiving the GPS and differential beacon signals attach to the top of the waterproof box. A plastic window in the cover of the waterproof box allows observation of the GPS receiver’s display. Data from the gas collector and GPS receiver are separately transmitted by cables via RS422 to the research boat and converted to RS232 before logging and display on a computer. During post-cruise processing, GPS position data are smoothed with a fifth order, low-pass Butterworth filter with a cutoff (half power) frequency of 0.033 hz (30 s period). Data

from the gas collector and GPS receiver are merged in software based on time. In field operations the buoy is either allowed to drift freely or is gently towed on a bridle (Fig. 1) through seepage areas as the research boat moves slowly forward.

### 3. Buoy Calibration

Pressure sensors were calibrated in the laboratory prior to the field deployments by connecting a manometer to the differential and absolute pressure ports. Hydrostatic pressure measured with the manometer was compared with digital counts from the A/D converters for  $\Delta p$  and  $p_a$  over the range 0-1900 pa and 102,000-132,000 pa, respectively (Egland 2000). The responses of both sensors were linear over the range of pressures encountered during field sampling.

To verify operation of the buoy as a flux-measuring instrument, tests were conducted in a swimming pool using a metered bubbling gas (nitrogen) source with adjustable volume flow rate. During the tests, the buoy freely floated in the pool on a slack tether. Nitrogen was fed through a ball-type flow meter and tubing to a porous air stone placed on the pool bottom below the buoy. Manual adjustment of a valve in the line to the flow meter produced a range of flow rates. After each adjustment the system was allowed to run for a few minutes before recording the rate from the ball flow meter. Egland (2000) discusses the pool calibration procedure in more detail.

In the flux range 0-20  $\text{m day}^{-1}$  ( $0 \sim 2 \times 10^{-4} \text{ m s}^{-1}$ ),  $\dot{q}$  estimated from (2) nearly equaled  $\dot{q}$  computed from the ball flow meter (Fig. 3); the slope of the best-fit line relating the two is 0.96. At greater flow rates the buoy underestimated  $\dot{q}$  because the collection chamber filled so rapidly that gas escaped out the chamber bottom before complete venting occurred. Fluxes above 20  $\text{m day}^{-1}$  were not encountered in our field experiments, but use of a larger collection chamber could prevent overflow in regions of very high flux.

#### 4. Field experiments

Several field trials of the buoy were conducted in the region of strong hydrocarbon seepage off Coal Oil Point near Santa Barbara, California during fall, 1998 and summer, 1999 (Fig. 4). Each field experiment consisted of a series of drifts of the buoy along tracks up to several hundred meters long. Most drift tracks crossed regions of strong hydrocarbon seepage visually identifiable by bubble plumes at the sea surface. A few tracks were obtained away from areas of seepage to evaluate the effects of surface waves and buoy motion. We present representative results from field experiments on 8 July and 19 August 1999.

Examples of time series of  $\Delta p$ ,  $p_c$ , and  $T_c$  obtained from the flux buoy on 8 July 1999 are shown in Fig. 5 from a region of strong bubbling gas flux emanating from the Coal Oil Point anticlines (Fischer 1977; Hornafius et al. 1999; Quigley et al. 1999). Steep increases in  $\Delta p$  occurred when the buoy was over the strongest bubble plumes and the collection chamber filled rapidly (Fig. 4A). Abrupt drops lasting less than 1 s in the  $\Delta p$  time series (i.e. less than the sampling interval) correspond to venting of the collection chamber. To permit digital filtering and other analysis, these pressure drops were removed in data processing to produce an approximately monotonic time series (Fig. 5B, upper curve). In the removal procedure, the value of  $\Delta p$  just before a drop was added to those after the drop; subsequent drops were similarly removed.

Temperature in the collection chamber also dropped abruptly during venting: at the first valve opening in Fig. 5D,  $T_c$  dropped 0.8 C from an initial value of 18.9 C. The source of the colder water was upwelling within the bubble plumes where “boils” and strong surface divergences were observed. Laboratory studies have shown that turbulent bubble plumes in stratified environments can entrain, mix, and upwell ambient waters from depth (Asaeda and Imberger 1993; McDougall 1978). After each venting,  $T_c$  increased as gas accumulated in the chamber. The gradual increase in temperature evident in Fig. 5D after the bubble plumes were crossed likely resulted from drift of the buoy out of the cool, upwelled plume. Steps in temperature are artifacts from the A/D conversion.

Ideally, surface waves and buoy motion would have no effect on  $\Delta p$  since they produce equal pressure fluctuations inside and outside the chamber. High frequency fluctuations are evident in the  $\Delta p$  time series, however, consistent with small surface waves (Fig. 5A). Fluctuations with periods of 4-6 s are prominent in the time series of  $p_c$  with standard deviation  $\sigma_{p_c} = 867$  pa ( $\sim 5$  cm wave amplitude) over the first 500 s (Fig. 5C). Because gas flux estimates are proportional to  $\partial\Delta p/\partial t$  by (2), they are sensitive to these pressure fluctuations. To investigate the effects of waves and buoy motion, time series of  $\Delta p$  and  $p_c$  were obtained on 19 August 1999 in a region without measurable seepage activity (Fig. 6 A and B). Temperature was nearly constant over the record (data not shown).  $p_c$  fluctuated about a mean of 127,170 pa with  $\sigma_{p_c} = 1080$  pa (Fig. 6B); fluctuations in  $\Delta p$  were much lower (Fig. 6A) with  $\sigma_{\Delta p} = 63$  pa. The spectrum of  $p_c$  for the record has a broad peak in the frequency range 0.12-0.28 hz (periods of  $\sim 4-8$  s) consistent with surface waves (Fig. 7A, upper curve). At lower frequencies the spectrum is approximately white. The  $\Delta p$  spectrum is also nearly white also with low, broad spectral peaks centered at 0.15 and 0.27 hz (Fig. 7A, lower curve). Spectral amplitudes of  $\Delta p$  are generally lower than of  $p_c$  by factors of 100 or more.

Squared coherence between  $\Delta p$  and  $p_c$  indicates that fluctuations in these quantities are significantly correlated in the surface wave frequency range 0.21-0.38 hz and around 0.14 hz (Fig. 7B). Thus, we conclude that surface waves and buoy motion are a significant source of noise in our measurements of  $\Delta p$ . Noise in the time series of  $\Delta p$  was reduced by applying a fifth order, low-pass Butterworth digital filter. After experimenting with a range of cutoff frequencies  $f_c$ , we determined that  $f_c$  in the range 0.02-.05 hz was a good compromise between smoothing and along-track resolution. Results presented here use  $f_c = 0.022$  hz (45 s period). The lower curve of Fig. 5A shows an example of a smoothed  $\Delta p$  time series.  $\dot{q}$  is computed from (2) where the partial derivative  $\partial\Delta p/\partial t$  is estimated from a first difference of the smoothed  $\Delta p$  time series.

Time series of  $\dot{q}$  for the data record of Fig. 5 clearly show areas of strong seepage flux with a maximum of about  $10 \text{ m day}^{-1}$  ( Fig. 5A, lower curve and Fig. 8A). The buoy track corresponding to this time series was 530 m long (Fig. 8B). The buoy was towed in tightly curving arcs through the areas of strongest seepage to maximize data collection there. The mean flux along the track is  $\bar{\dot{q}} = 0.56 \text{ m day}^{-1}$ . For comparison, Quigley (1997) estimated a mean flux in the range  $(3.7\text{-}6.2) \times 10^{-2} \text{ m day}^{-1}$  over the entire seep field area ( $\sim 3 \text{ km}^2$ ). A second flux time series (Fig. 9), also from 8 July 1999 and in the same seepage area (Fig. 1), shows three intervals of high flux with a maximum of almost  $7 \text{ m day}^{-1}$ . Peaks labeled 1 and 4 were encountered in about the same place where the buoy track doubled back on itself (Fig. 9B). The track length is 381 m and the along-track mean flux is  $\bar{\dot{q}} = 0.58 \text{ m day}^{-1}$ , similar to other estimate.

Flux noise levels  $\dot{q}_n$  due to buoy motion and surface waves are typically much smaller than  $\dot{q}$  in the bubble plumes. We estimate  $\dot{q}_n$  from a 1200 s record, part of which is shown in Fig. 6, where all fluctuations result from buoy motion and surface waves. This record is typical of those obtained under conditions of light winds and small waves off Coal Oil Point. For the record of Fig. 6,  $\dot{q}_n = (0.3 \pm 6) \times 10^{-2} \text{ m day}^{-1}$ . Over the first 500 s of the record of Fig. 5, before the bubble plumes were crossed,  $\dot{q}_n = (0.1 \pm 8) \times 10^{-2} \text{ m day}^{-1}$ . Occasional large negative values  $\dot{q}_n$  result from accidental tugs on the towing bridle as the research boat maneuvered the buoy through the bubble plumes (e.g. 850 s in Fig. 8A). Fluctuations in  $\dot{q}_n$  with periods in the range 50-70 s are evident in Fig. 8A before and after the bubble plumes were crossed. These probably resulted from low frequency vertical motions of the buoy.

## 5. Summary

We have developed a buoy and gas capture assembly, referred to as a flux buoy, for direct measurement of gas flux due to rising bubble plumes in oceans and lakes. We are using the flux buoy to

quantify gas flux to the atmosphere in regions of strong hydrocarbon seepage over the continental shelf. An extensive series of flux measurements been obtained in the region of hydrocarbon seepage off Coal Oil Point near Santa Barbara, California. The maximum flux observed for seeps in the Coal Oil Point anticline is of order  $10 \text{ m day}^{-1}$ . During calibration tests in a swimming pool, the buoy was capable of measuring fluxes up to  $20 \text{ m day}^{-1}$ ; use of larger collection chambers would permit measurement of higher fluxes. The lowest detectable fluxes depend on sea state and the resulting vertical motion of the buoy. For conditions of light wind and small swell off Coal Oil Point, typical mean noise levels are in the range  $(0.1 - 0.3) \times 10^{-2} \text{ m day}^{-1}$  with standard deviation  $(6 - 8) \times 10^{-2} \text{ m day}^{-1}$ .

The gas capture assembly can be operated independently of the buoy for applications such as deployment over gas vents on the seafloor or lakebeds. A smaller version of the buoy could also be made for deployments in small bodies of water where surface wave effects are minimal.

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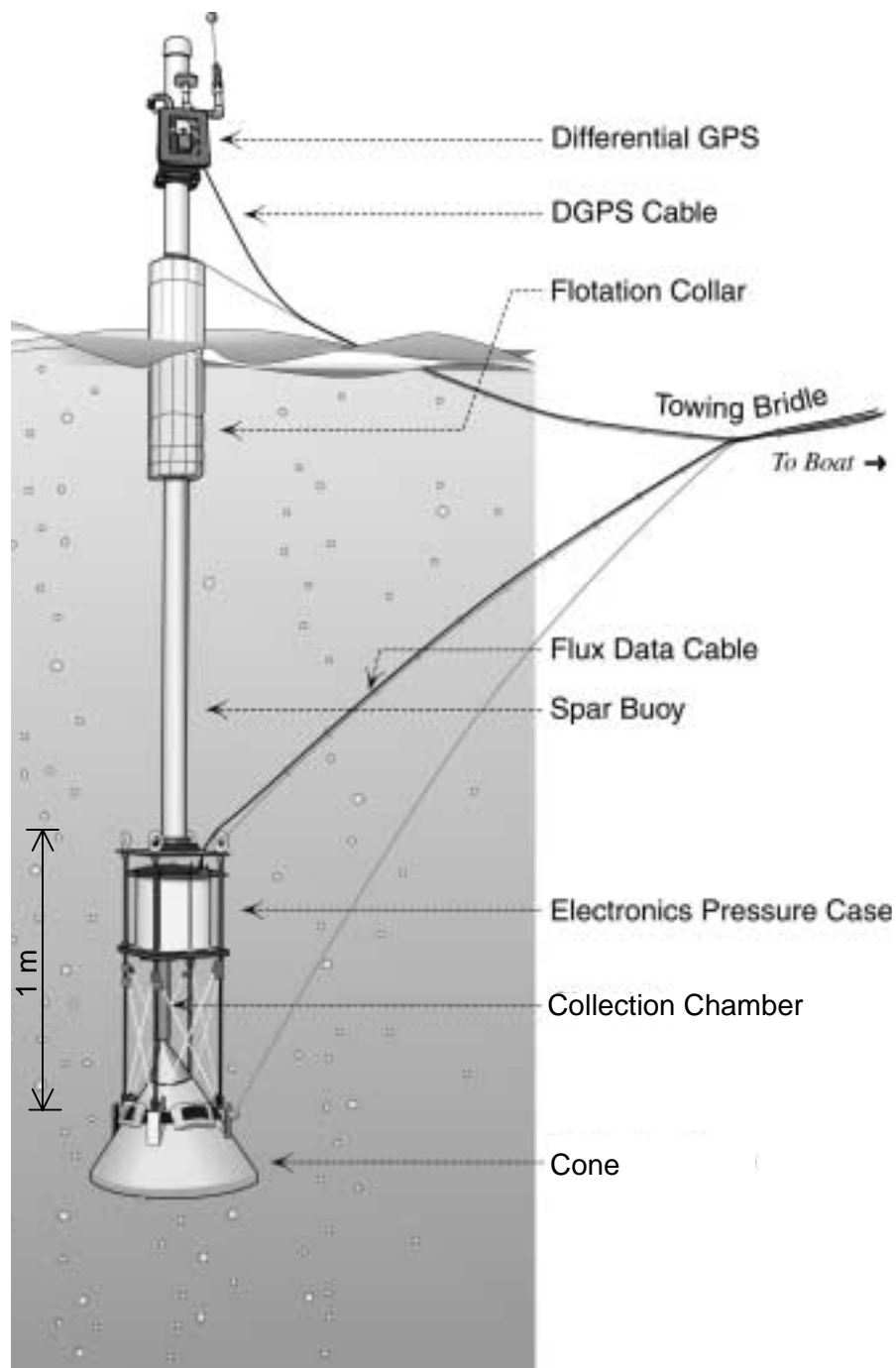
Table 1. Pressure and temperature sensor characteristics

Sensor	Manufacturer	Model	Range	Accuracy	Least Count Resolution
Differential pressure	Validyne	DP-15-26-N-1-5-4-A (signal conditioner: Validyne CD101-B-6-A-1-M)	0 – 3300 pa (0-5 psi)	± 0.25% full scale	0.83pa
Absolute pressure	Omega	PX303-050A 5V	0-345,000 pa (0-50 psi)	± 0.25% full scale	89.7pa
Temperature	Omega (signal conditioner: Analog Devices)	HYP4-16-1 1/2-100-E4-48-PR (signal conditioner: Analog Devices 5B350-02)	0-100 C	± 0.1% full scale	0.1 C

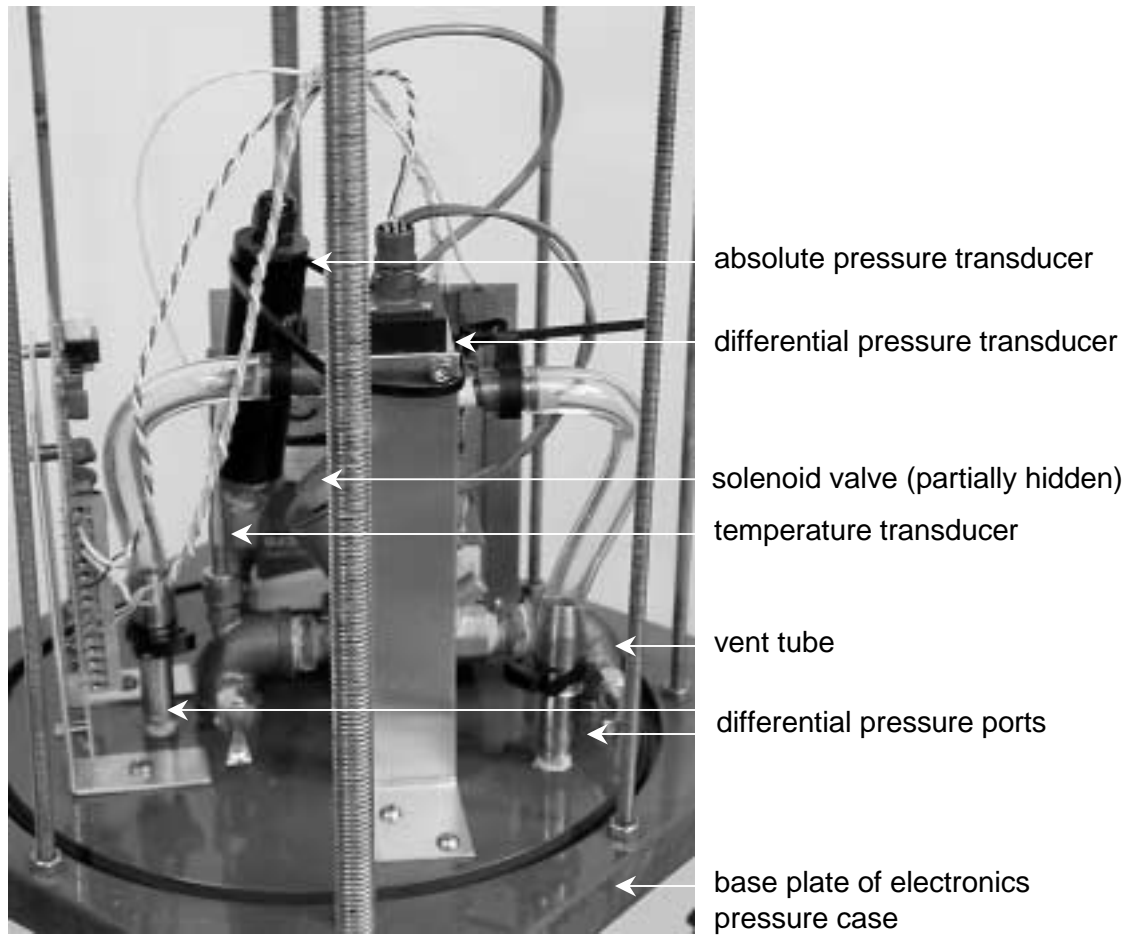
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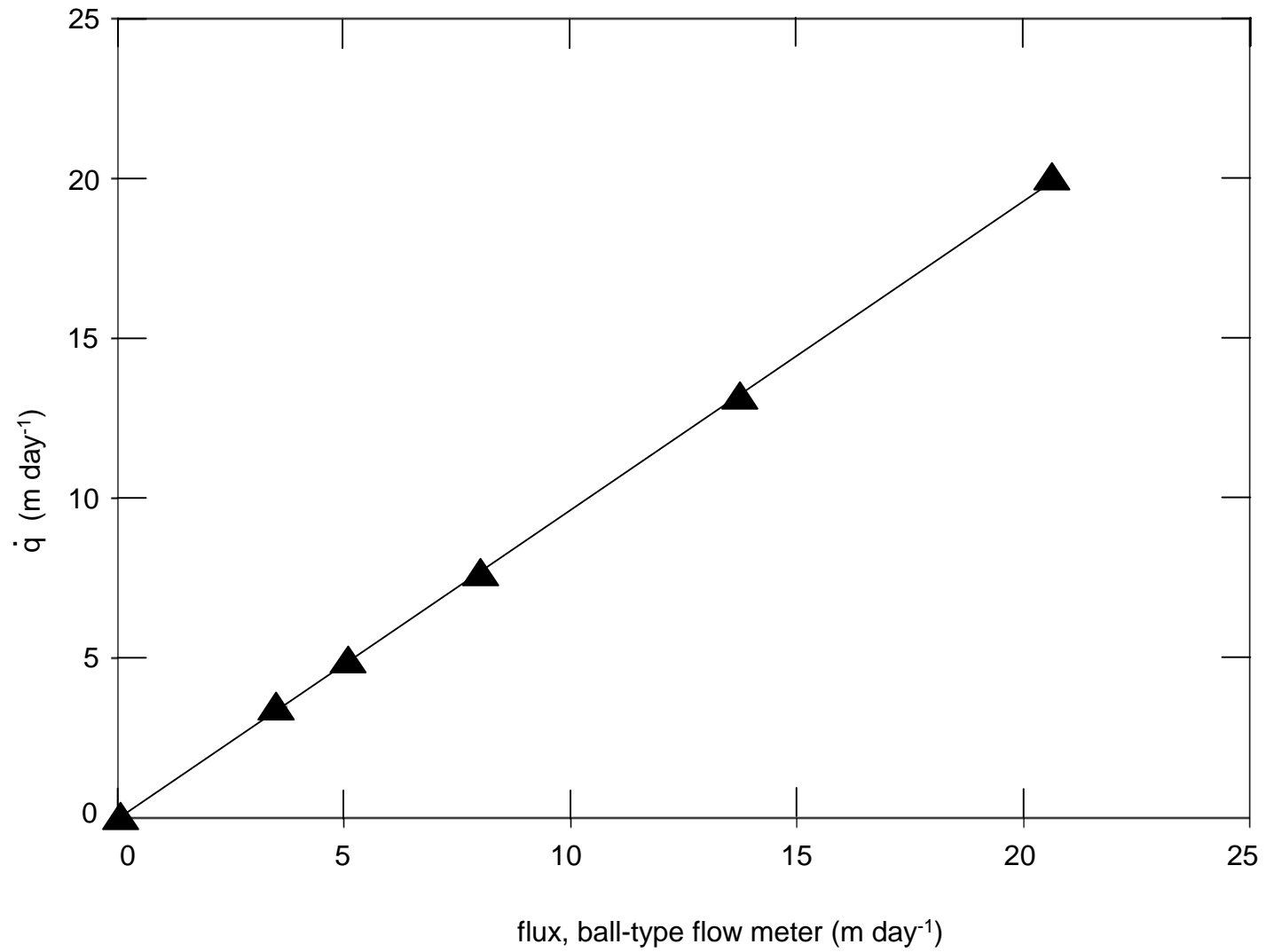
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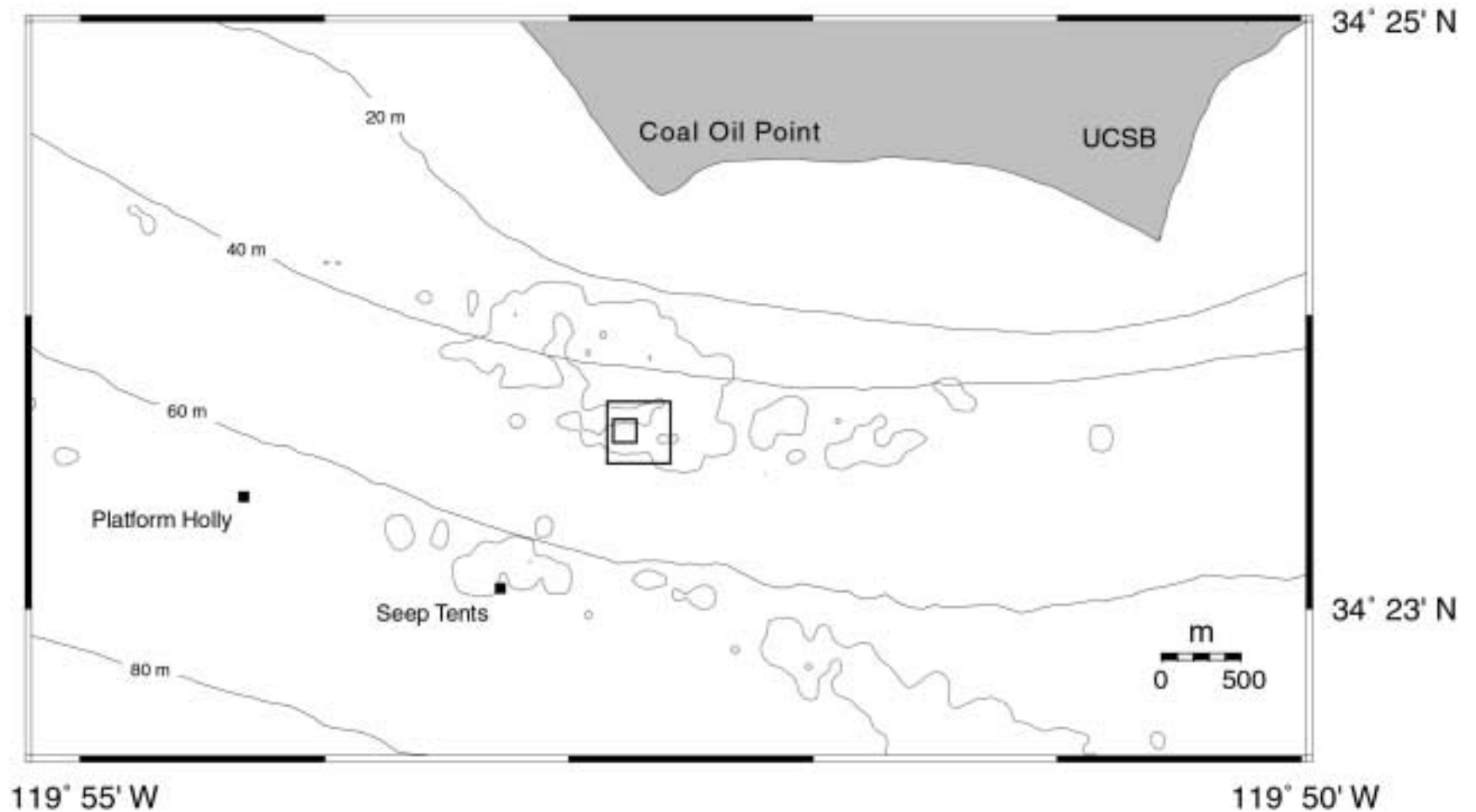
**Figure 1.** Schematic of flux buoy. Electronics pressure case contains sensors, electronics, gas release valve and microcomputer. Diagonal white lines around collection chamber are tensioned cables to provide stiffness. Scale at left indicates 1 m. (Bubble size is exaggerated.)



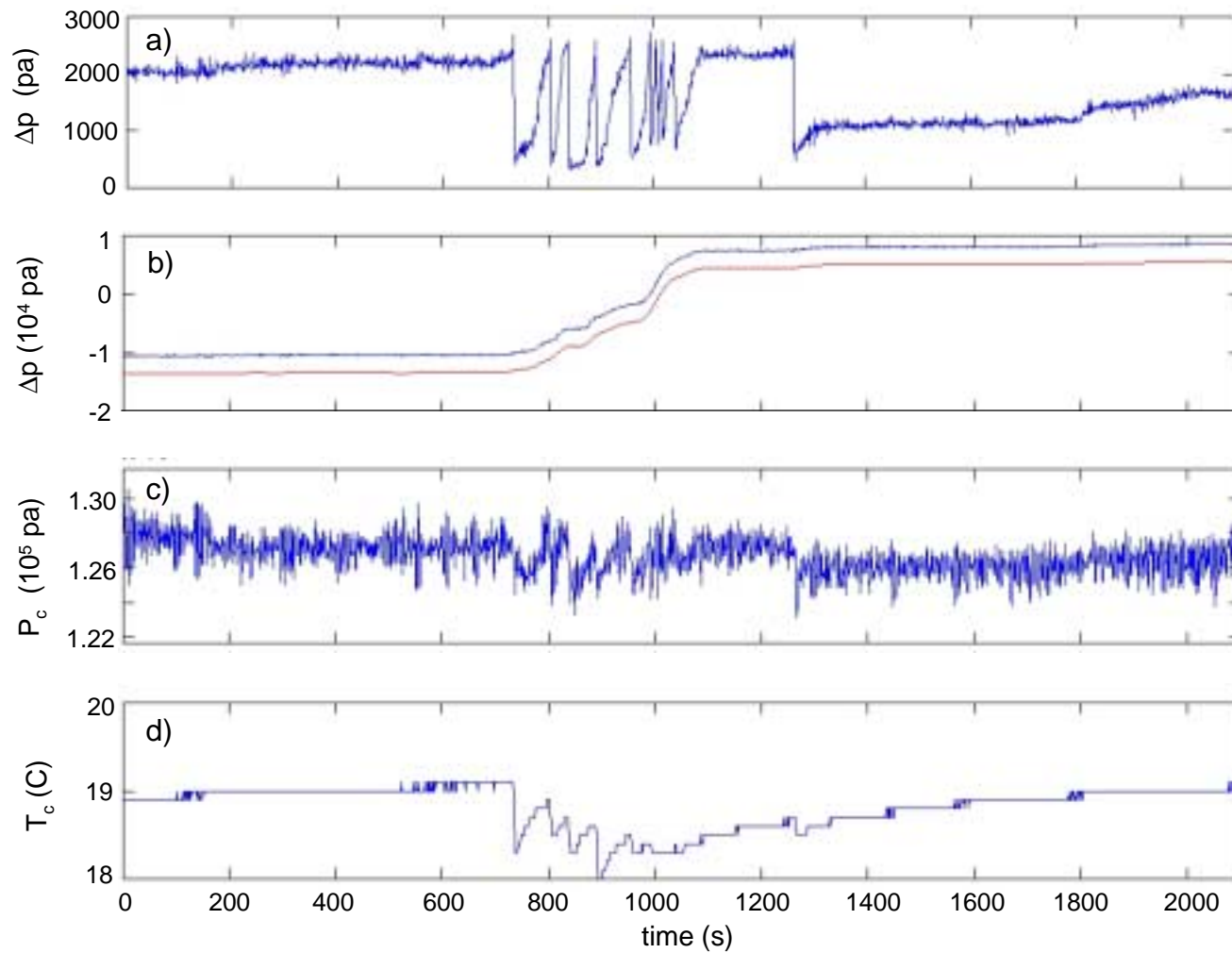
**Figure 2.** Interior of electronics pressure case identifying temperature and pressure transducers.



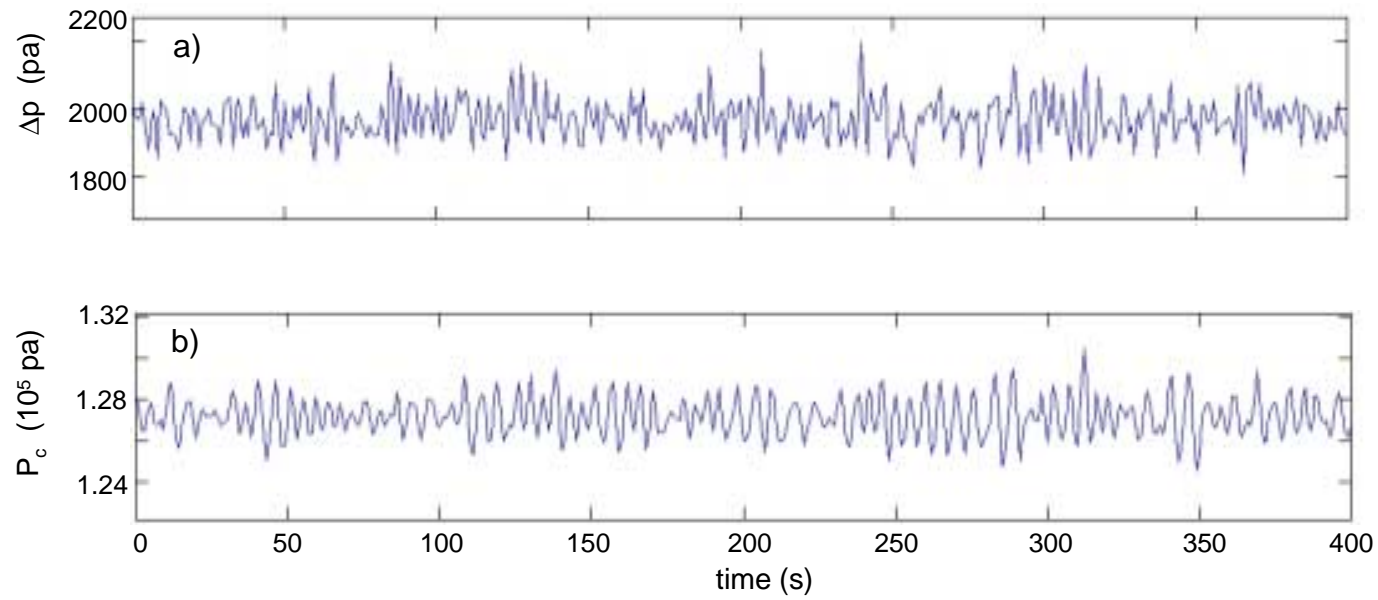
**Figure 3.** Calibration of flux buoy from swimming pool tests. x-axis axis shows flux measured with ball-type flow meter and y-axis shows flux  $\dot{q}$  derived from flux buoy using equation (2). Slope of fitted line is 0.96.



**Figure 4.** Study area off Coal Oil Point near Santa Barbara California. Closed contours indicate areas of hydrocarbon seepage activity obtained from sonar surveys described by (Quigley, 1997) and (Quigley et al., 1999). Bathymetric contours show depths of 20, 40, 60, and 80 m. Larger square shows area of Fig. 8, smaller square of Fig. 9. Platform Holly is an oil production platform. The seep tents are large pyramidal structures on the sea floor to capture natural gas.

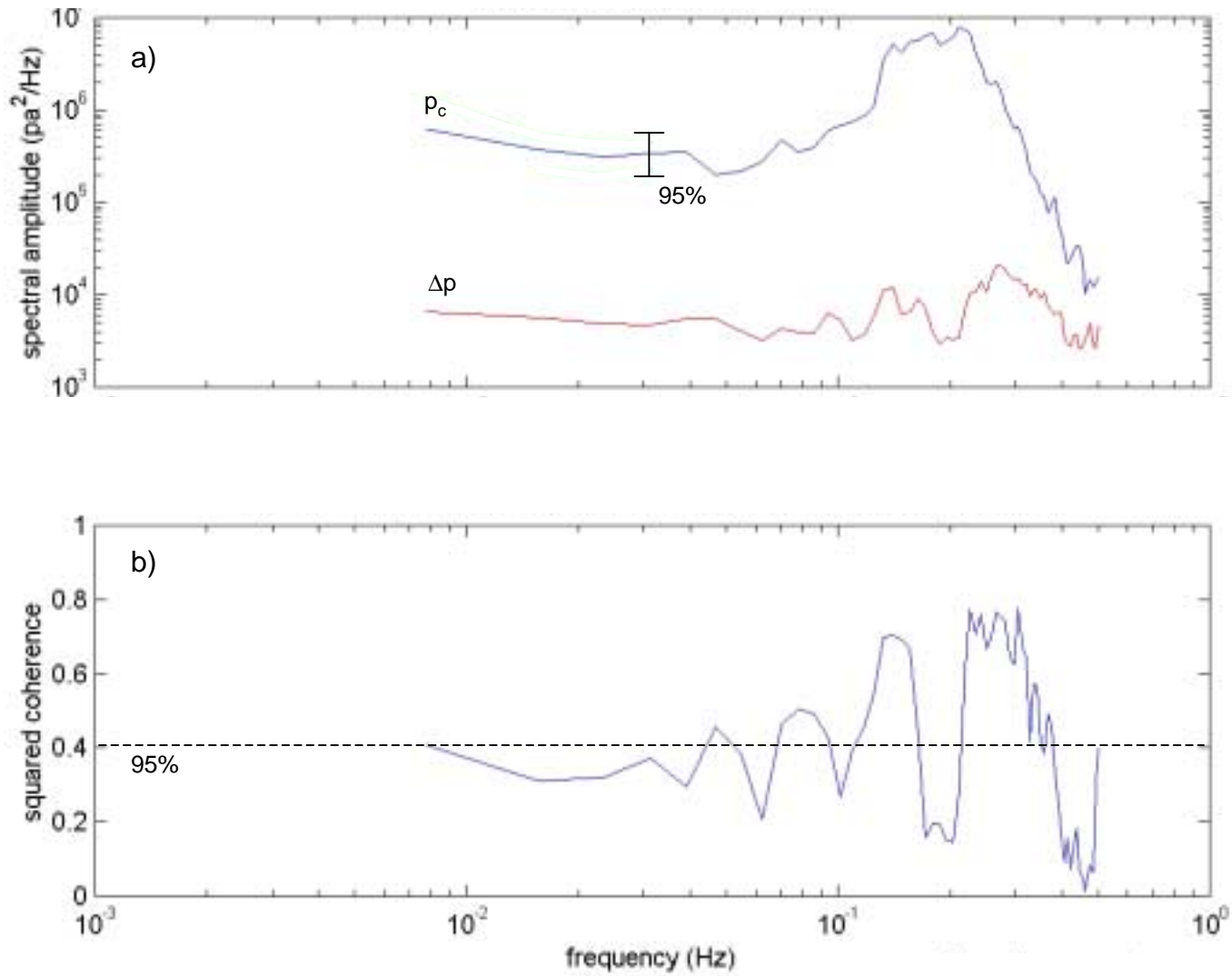


**Figure 5.** Times series for drift 2 on 8 July 1999 of: (a)  $\Delta p$ ; Discontinuities indicate venting of gas from collection chamber; (b)  $\Delta p$ , but with discontinuities removed and mean set to zero. Lower curve shows same time series offset by 3000 pa and smoothed with a 5-pole low pass Butterworth filter with cutoff frequency 0.022 hz (45 s period); (c)  $p_c$ ; and (d)  $T_c$ .

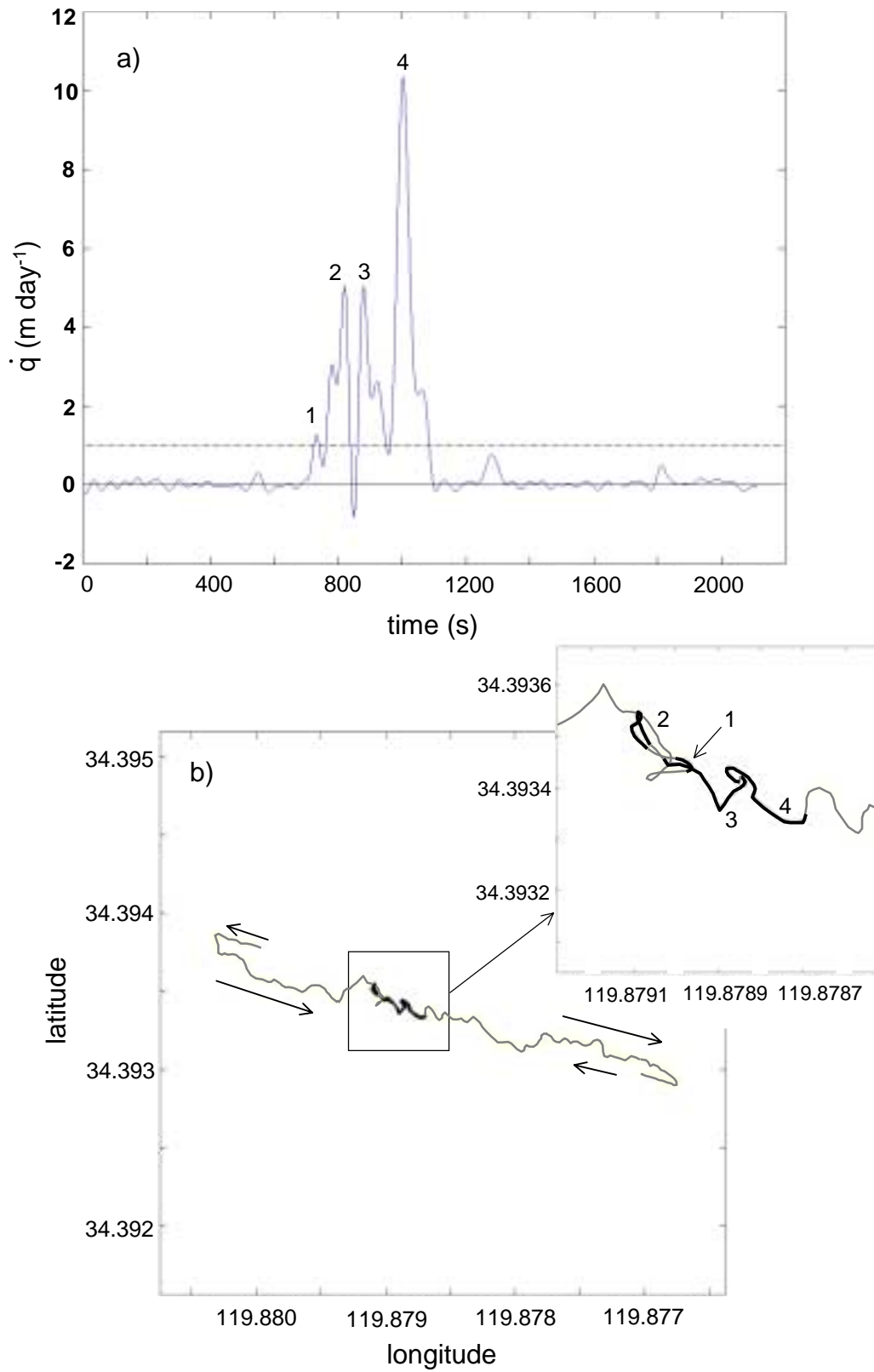


**Figure 6.** A portion of times series for drift 2 on 19 Aug 1999 of: (a)  $\Delta p$  and (b)  $p_c$  showing the effects of surface waves. No venting of gas from collection chamber occurred during this record and no seepage was encountered.

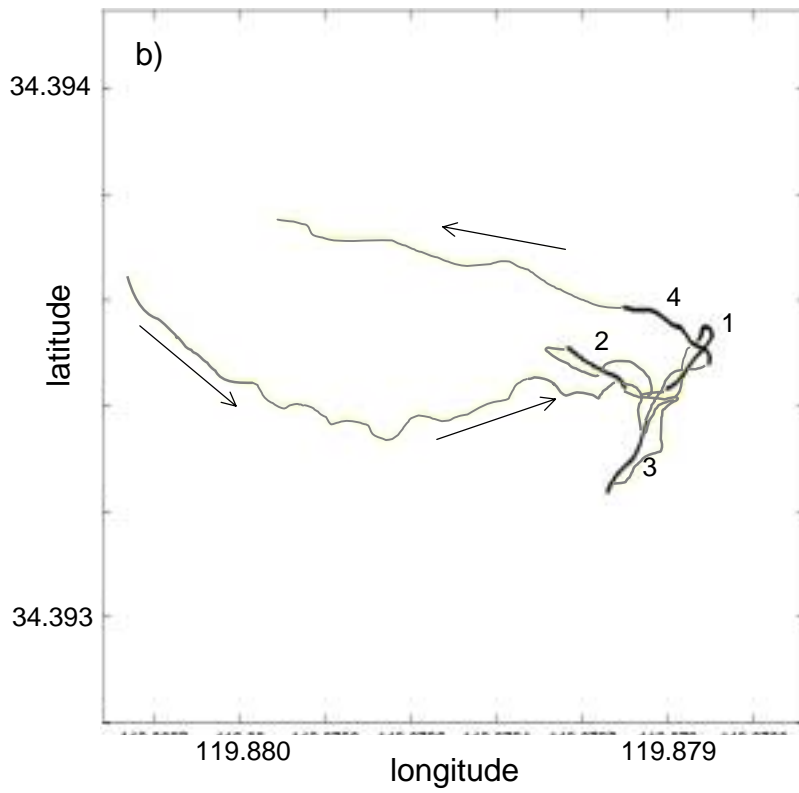
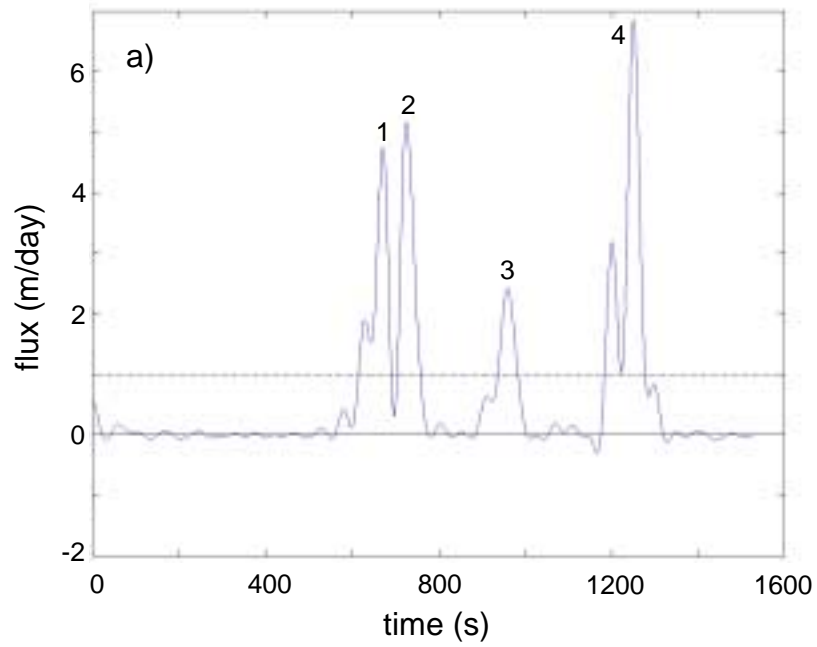




**Figure 7.** (a) Autospectra of  $p_c$  (upper curve) and  $\Delta p$  for time series of Figures 5Band 5A, respectively. Bar shows 95% confidence interval for spectral amplitudes. (b) Squared coherence between absolute and differential pressure time series. Coherence above dashed line is significant at 95% level.



**Figure 8.** (a)  $\dot{q}$  for drift 3 on 8 July 1999. (b) Drift track of flux buoy. Dark line indicates  $\dot{q}$  greater than  $1 \text{ m day}^{-1}$  (panel a, dashed line). Square is 400 m on a side. Inset shows close up view of buoy track. Smaller square is 70 m on a side.



**Figure 9.** As in Fig. 8 but for the drift 2 of the flux buoy on 8 July 1999. The square in panel b) is 150 m on a side.

