Methane Plume Emissions Associated with Puget Sound Faults in the Cascadia Forearc

Harlan Paul Johnson[1], Susan G Merle[2], Tor A Bjorklund[1], Susan L Hautala[1], Tamara Baumberger[3], Sharon L Walker[3], Junzhe Liu[1], Nicholas D Ward[1], and Chenyu Wang[1]

[1]University of Washington
[2]University of Oregon PMEL
[3]PMEL/NOAA

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Abstract

Key Points: * Extensive methane bubble plumes have been discovered on the Puget Sound seafloor. * The emission sites of these plumes are associated with major fault zones that penetrate the Cascadia forearc. * Dissolved methane arising from the plumes is mixed throughout the estuary by tides and local mixing. Abstract Methane gas plumes have been discovered to issue from the seafloor in the Puget Sound estuary. These gas emission sites are co-located over traces of three major fault zones that fracture the entire forearc crust of the Cascadia Subduction Zone. Multibeam and single-beam sonar data from cruises conducted in 2011, 2018, 2019, 2020 and 2021 identified the acoustic signature of over 330 individual bubble plumes. Dissolved gas from the plumes combines to elevate seawater methane concentrations of the entire Puget Sound estuary. Fluid samples from adjacent terrestrial hot springs and deep-water wells surrounding the estuary contain a helium-3 isotope signature, indicating a deep fluid source located near the underlying Cascadia Subduction Zone plate interface. However, limited data from this pilot study suggest that Puget Sound seawater emission sites lack either similar chemical isotope signatures or elevated thermal anomalies expected from association with a deep plate-interface reservoir. The existence of vigorous marine methane plumes arising from areas of thin sediment cover associated with deeply-penetrating forearc fault zones but presenting no thermal or chemical anomalies found in other similar forearc environments, remains an unresolved paradox. Plain Language Summary Puget Sound is a large inland sea located in western Washington State where seawater circulation is dominated by vigorous tidal forcing from the North Pacific Ocean. The deep Puget Sound is the largest estuary in North America measured by contained water volume and the second largest estuary in terms of area after Chesapeake Bay. Shipboard sonar images have revealed approximately 330 bubble plumes of methane gas and fluid rising from the seafloor of the estuary. Large clusters of bubble plume sites are concentrated over the major regional fault zones that penetrate the western North American plate beneath Puget Sound, including the South Whidbey Island Fault, the Seattle Fault and the Tacoma Fault Zones. Although the forearc Basin is surrounded by terrestrial hot springs and water wells that show a clear chemical signature of fluid arising from the underlying Cascadia Subduction Zone plate interface, based on our limited sampling there is currently no evidence for similar chemical or temperature anomalies in the Puget Sound plumes and the source of the methane bubble plumes is still unidentified.

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[1]School of Oceanography, University of Washington, Seattle, WA, 98105, USA. [2]Cooperative Institute for Marine Ecosystem Resources Studies - Oregon State University, and NOAA PMEL Earth Ocean Interactions Program, Newport, OR, 97365, USA. [3]NOAA/Pacific Marine Environmental Lab, 7600 Sand Point Way NE, Seattle, WA 98115, USA, [4]Coastal Sciences Division, Pacific Northwest National Laboratory, Sequim, WA, 98382, USA.

Corresponding author: H. Paul Johnson (paulj@uw.edu)

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- Extensive methane bubble plumes have been discovered on the Puget Sound seafloor.
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Methane gas plumes have been discovered to issue from the seafloor in the Puget Sound estuary. These gas emission sites are co-located over traces of three major fault zones that fracture the entire forearc crust of the Cascadia Subduction Zone. Multibeam and single-beam sonar data from cruises conducted in 2011, 2018, 2019, 2020 and 2021 identified the acoustic signature of over 330 individual bubble plumes. Dissolved gas from the plumes combines to elevate seawater methane concentrations of the entire Puget Sound estuary. Fluid samples from adjacent terrestrial hot springs and deep-water wells surrounding the estuary contain a helium-3 isotope signature, indicating a deep fluid source located near the underlying Cascadia Subduction Zone plate interface. However, limited data from this pilot study suggest that Puget Sound seawater emission sites lack either similar chemical isotope signatures or elevated thermal anomalies expected from association with a deep plate-interface reservoir. The existence of vigorous marine methane plumes arising from areas of thin sediment cover associated with deeply-penetrating forearc fault zones but presenting no thermal or chemical anomalies found in other similar forearc environments, remains an unresolved paradox.

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Puget Sound is a large inland sea located in western Washington State where seawater circulation is dominated by vigorous tidal forcing from the North Pacific Ocean. The deep Puget Sound is the largest estuary in North America measured by contained water volume and the second largest estuary in terms of area after Chesapeake Bay. Shipboard sonar images have revealed approximately 330 bubble plumes of methane gas and fluid rising from the seafloor of the estuary. Large clusters of bubble plume sites are concentrated over the major regional fault zones that penetrate the western North American plate beneath Puget Sound, including the South Whidbey Island Fault, the Seattle Fault and the Tacoma Fault Zones. Although the forearc Basin is surrounded by terrestrial hot springs and water wells that show a clear chemical signature of fluid arising from the underlying Cascadia Subduction Zone plate interface, based on our limited sampling there is currently no evidence for similar chemical or temperature anomalies in the Puget Sound plumes and the source of the methane bubble plumes is still unidentified.

1 Introduction

Natural marine seeps of methane are a small but still important source of a strong greenhouse gas that enters the global environment. The spatial distribution of the methane bubble streams issuing from crustal ruptures can provide insight into key geological and tectonic processes by illuminating the variations in permeability associated with fault zones. Methane emissions from the surface waters of estuaries into the atmosphere arise from a multitude of sources including rivers, benthic production, and simple diffusion from organic-rich sediments and these are poorly constrained on a global scale (Rosentreter et al., 2021). In this study we present the identification of 330 Puget Sound gas bubble plumes using single-beam sonar and traditional Niskin bottle sampling. We then describe the inconsistency presented by our observed normal emission seawater chemistry and temperature data that is in contrast to the emission site locations that are correlated to deeply-penetrating fault zone traces in the Cascadia forearc. The source region for the plume gas/fluid is not resolved or explained by our limited pilot-study data, and we present potential alternative hypotheses for the source of these abundant plumes. We also discuss potential future methods for resolving whether these fault zones are direct fluid pathways from a deep reservoir via the forearc fault zones or if instead the methane gas is biogenically-derived from organic material present within recently deposited sediments.

Active methane emissions from coastal margins have been known for over 50 years (Hovland and Judd, 1992 and references therein). Present methane emission estimates, compiled with large uncertainties, indicate that between 3 and 48 Tg/y of methane is released to the atmosphere from marine seeps, including sources that are both thermogenic and biogenic in origin (Hornafius et al, 1999; Etiope, 2012; Razaz et al, 2020). Globally, the largest known concentrations of marine methane emission sites lie on active continental margins that overlie subduction zones, and Cascadia is an unusually well-studied example (Salmi et al., 2011; Hautala et al, 2014; Embley et al, 2017; Baumberger, et al, 2018; Riedel et al, 2018; Johnson et al., 2019; Merle et al 2021). Gas emissions are also present in abundance on some passive coastal margins, although the mechanism for producing those methane seeps appears to differ from those along subduction zone margins (Skarke et al, 2014; Ruppel and Kessler, 2017; Mau et al, 2012, 2015, Plaza-Faverola, 2019, Garcia-Tigreros et al, 2020).

Multibeam and single-beam sonar data from recent cruises revealed extensive bubble plume fields present within the Puget Sound and Hood Canal estuaries. Sonar data from 18 cruises using two different data acquisition systems contributed to the plume identifications. In 2011, the *RV Thompson* returned to its Seattle home port at the end of cruise TN265. Atypically, the EM302 multibeam sonar was left in the data acquisition mode during transit through northern Puget Sound, and included sonar return data from acoustic reflectors within the water column. These data were archived in the Rolling-Deck-2-Repository database.

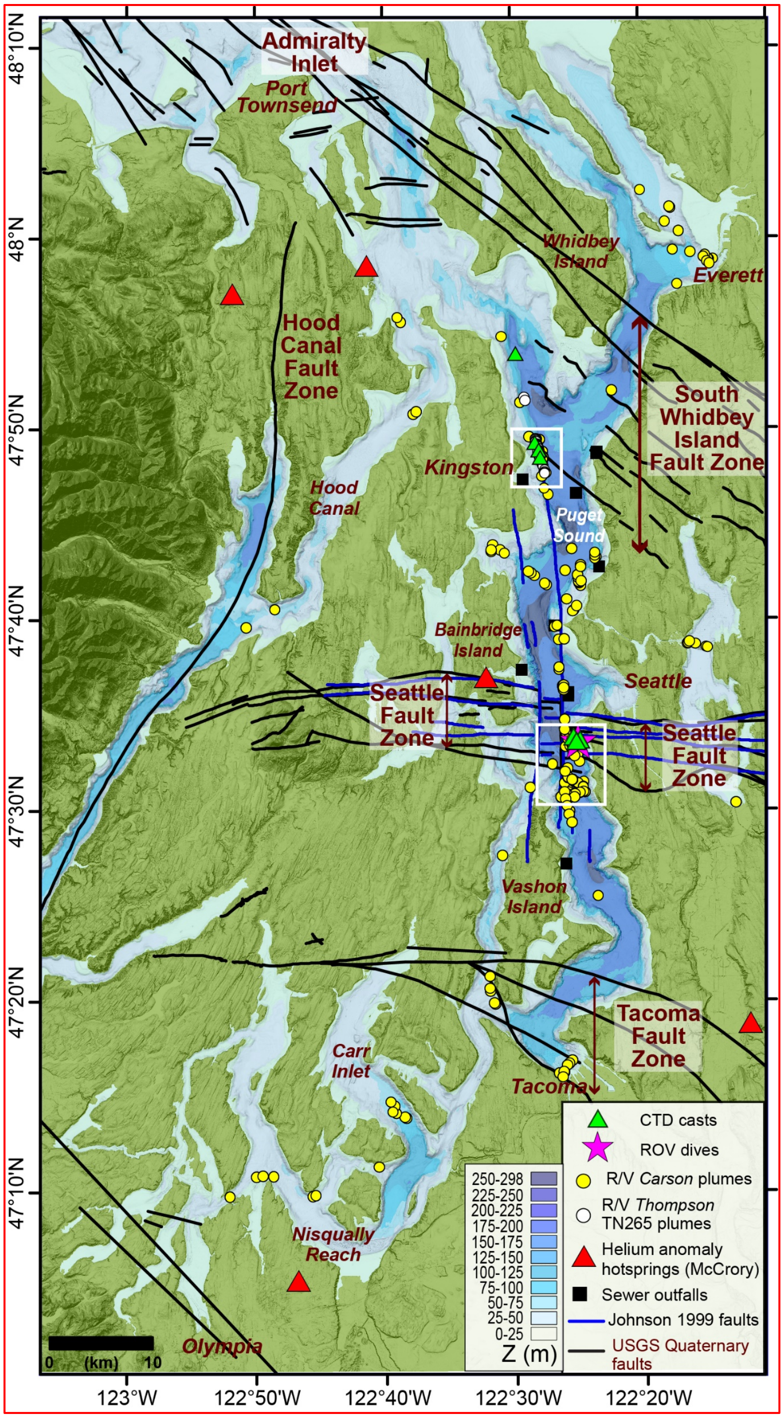

Figure 1: Map of Puget Sound and Hood Canal in north-central Washington State. Yellow circles are sites of individual bubble plume emissions based on CARSON single-beam 38 kHz sonar. Black lines are trace projections of the SWIF, SFZ and TFZ faults and the N-S Hood Canal and Puget Sound faults of Johnson et al., (1999). Black squares are urban sewer outfalls. Red triangles are locations of hot springs and water wells with helium isotope anomalies indicative of a deep subduction zone source (McCrory et al, 2016). Green triangles are CTD casts from the CARSON.

The archived *RV Thompson* multibeam water column data in Puget Sound were processed for on-going compilations of coastal methane emission sites on the outer Cascadia Margin and an unanticipated North-South oriented field of gas bubble plumes was discovered just east of the uplifted Kingston Arch anticline in northern Puget Sound (Figure 1). This new emission field was located where the edge of approximately East-West trending South Whidbey Island Fault (SWIF, Johnson et al, 1996; Sherrod et al, 2008) intersects a roughly North-South trending fault imaged by Johnson et al (1999) (Figure 2). The Kingston Arch has been described as a gentle anticline that forms the northern boundary of the Seattle Basin and has been topographically incised but does not appear to be bounded by any previously mapped faults (Johnson et al., 1994, 1996).

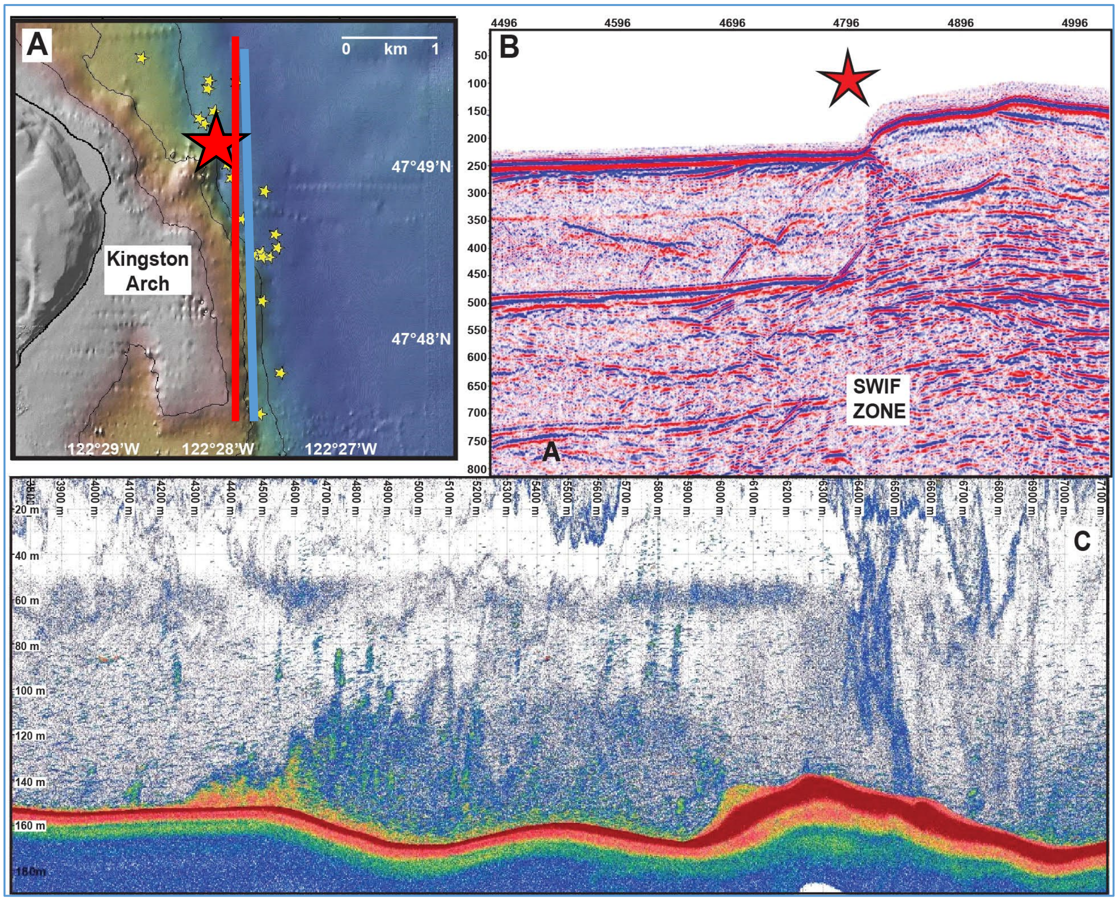

Figure 2: **Panel A**: Yellow circles show bubble plume locations from CARSON sonar acquired near Kingston Arch with red star indicating location of fault offset co-located in Panel B. Red line is MCS trackline for image in Panel B. **Panel B**: Offset sediment layers from the South Whidbey Island Fault Zone from archived MCS data (Johnson et al., 1996). Red stars in Panels A and B are co-located. **Panel C:** CARSON 38 kHz sonar image of bubble plumes at Kingston Arch, showing both gas bubbles (center) and fluid emissions (right) at position of red star in Panel A. Heavy red trace is the bottom reflection.

Following the 2011 *RV Thompson* cruise, an additional 17 cruises on the *RV Carson* were conducted in 2018, 2019, 2020 and 2021, with most of these cruises following tracklines determined by other academic and research objectives rather than searching for active bubble plume sites. Gas bubble plumes were identified in the sonar records from the 38 kHz single-beam Simrad ES38B with post-cruise data processing with the ESP3 program (Veloso et al, 2015; Ladroit et al, 2020). These cruise sonar records provided additional evidence of gas bubble plumes that are widely distributed throughout the water columns of Puget Sound, Hood Canal and even extend into Lake Washington (Figures 1, 2, 3 and 4).

The plume site locations appear to be stable in time, although repeat sonar imaging only exists for cruises TN265, RC0014, RC0019, RC0022, RC0038, and RC0047a. These repeat cruises show that plume sites can be re-identified at different times using slightly different sonar frequencies, including both 30 kHz multibeam sonar on the *RV Thompson* and 38 kHz single beam sonar on the *RV Carson*, and that emissions have remained active for at least the last 10 years. Although single isolated bubble plumes were found distributed throughout the estuary, the primary clusters of bubble streams are concentrated above three major fault zones (Figure 1).

Acoustic reflections in the sonar data indicate that a small portion of the gas bubbles plumes reach from the seafloor to the sea surface within the plume fields located at both Kingston Arch and the Alki Uplift, which have water depths of 180 meters and 160 meters respectively (Figures 2, 3 and 4). The acoustic observations of bubbles reaching the surface were confirmed by science party observations on the *RV Carson* deck that identified gas bubbles breaking the water surface at both sites. The presence of a frothy film on the water surface directly overlying the immediate area of the sites but absent from the adjacent waters was also observed, similar to observations at other methane emission regions. This suggests that the bubbles are likely 'organically coated', a phenomenon that impacts both bubble rise velocity and can slow methane gas diffusion into the water column (Leifer and Patro, 2002; Leifer and MacDonald, 2003; Leifer, 2010; Fu et al., 2020). The success of quasi-random tracklines of the *RV CARSON* in identifying the plumes, with a limited seafloor sonar footprint of approximately 25-meter diameter at 200-meter water depth, implies there are still many unidentified plumes within the estuary not included in the present inventory.

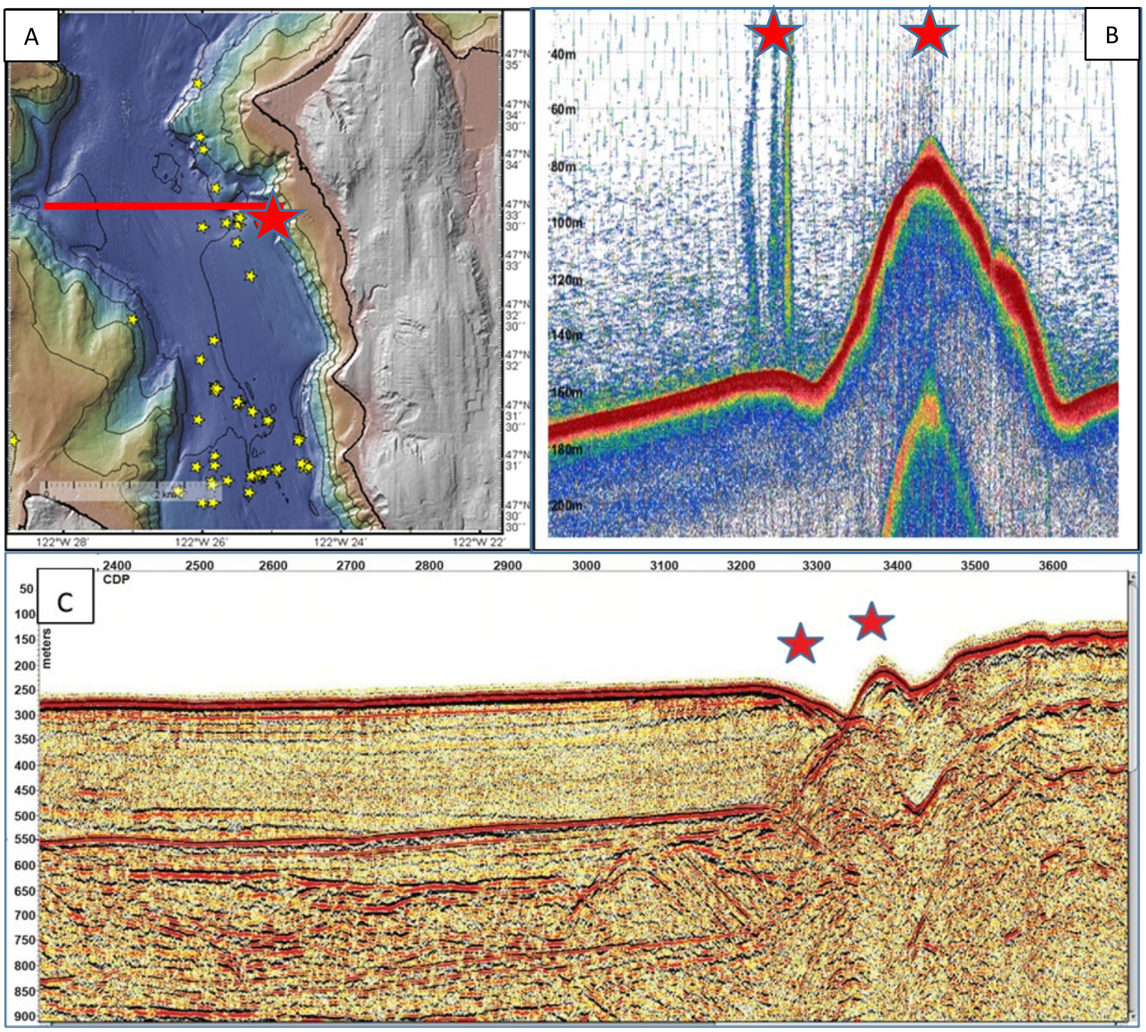

Figure 3: Panel A: Yellow circles are bubble plume locations near Alki Point within the projections of the Seattle Fault zone (Figure 1). Red line is approximate trackline for MCS image in Panel B. Red stars in all 3 panels are co-located**. Panel B**: CARSON sonar image of gas plumes shown in Panel A. **Panel C**: Archive MCS profile of sub-surface structures beneath the bubble plume site, showing basement rock uplift.

Introduction

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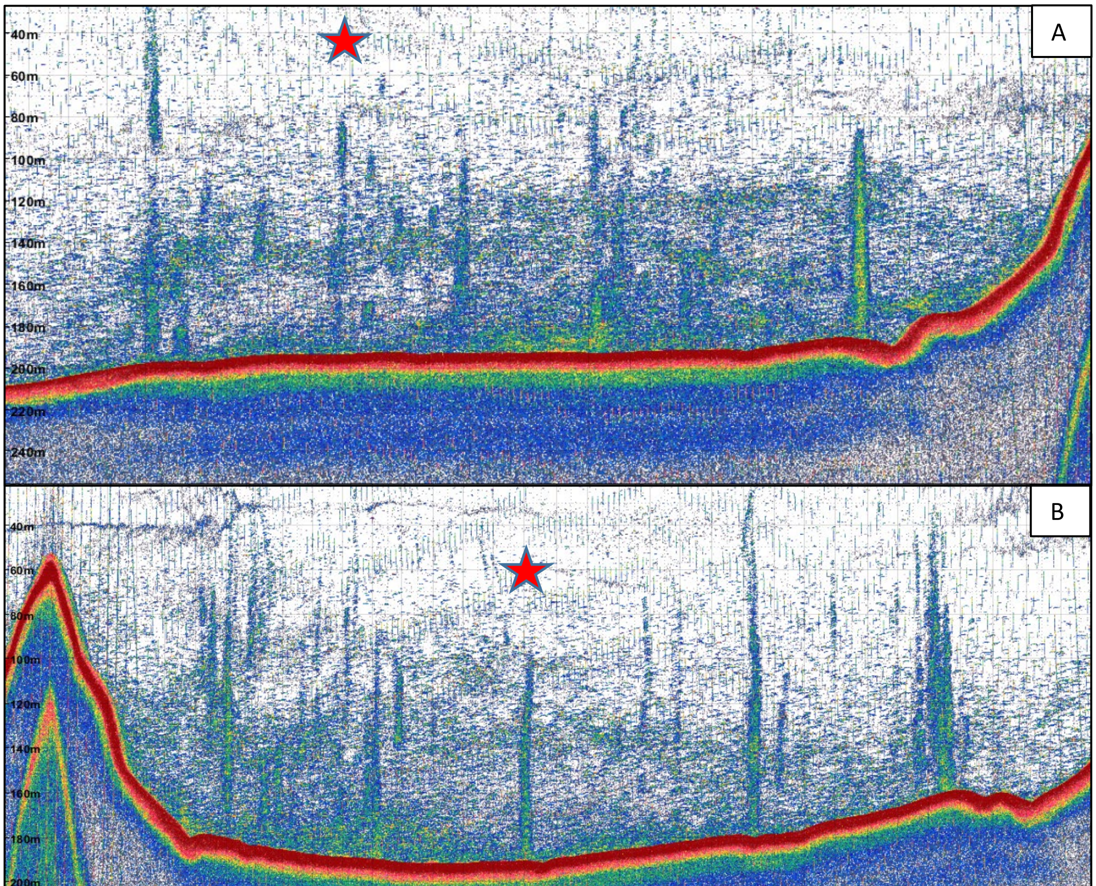
2. Methods

2.1 Separating Anthropogenic and Geological Sources

Understanding the source region of bubble plumes occurring within any urban estuary first requires separation of geological emissions from anthropogenic gas bubbles arising from sewer outfalls, storm drains and recent and historical dredge deposits. Storm drains in Puget Sound are clearly noted in NOAA navigational charts, are located near the shoreline in shallow water depths (<20 meters), and their freshwater discharge is unlikely to produce gas bubbles from the seafloor. Urban sewer outfalls are located in deeper water and their discharge can produce temporary gas plumes which are well-mapped (Figure 1). For example, a weak column of gas issuing from the Seattle West Point sewer outfall was observed during one *RV CARSON* cruise and was excluded from the plume inventory.

Dredge dump sites that were previously legally-permitted are also well-mapped although these dumps have been tightly restricted in Puget Sound over the last several decades. Mud slides and land-fill dumps from early urban development can also be recognized by detailed bathymetric mapping. However, a controlled photography and coring study in the Sound indicated that over 85% of the initial volume of dredge and slump material is removed by the vigorous tidal currents within a period of less than 4 months, and consolidation of any residual sediment matrix material is also completed within this short time interval (Nittrouer and Sternberg, 1975). As occurs with riverine input into the estuary, these potential anthropogenic sources of methane are associated with buoyant freshwater. Previous studies have shown that methane concentrations in low salinity river water decrease due to oxidation by methanotrophic bacteria almost immediately (hours) after exposure to saline water (de Angelis and Lilley, 1987; Upstill-Goddard et al., 2000; Schubert et al, 2006; Pack et al, 2015; Boetus et al, 2013).



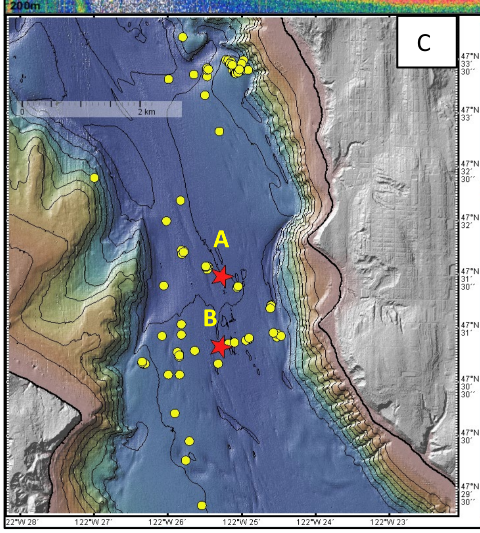


Figure 4: Panels A and B: RV Carson 38 khz sonar profiles across Puget Sound over the projected locations of the Seattle Fault Zone, showing plume fields extending entirely across the Seattle basin (Figure 1). Sonar image locations are shown on lower map. **Panel C:** Map showing locations of sonar profiles in Panels A and B. Red stars are the center of the profiles in Panels A and B shown above. Yellow circles are individual methane plume locations.

2.2 Chemistry of Fluid Samples

In an attempt to determine the origin of the plume gas emissions, we sampled near-vent seawater on two short pilot cruises (RC0019 and RC0038) using Niskin bottles mounted on the *RV CARSON* CTD cage. The CTD employed was a Seabird 9*plus*. For the RC0038 cruise, Niskin bottles were also attached to a small PHANTOM 2D2HD Remotely Operated Vehicle (ROV). Niskin bottle samples were analyzed for helium concentrations and isotopes and the traditional seawater nutrients of silica, ammonium, and nitrate/nitrite compounds. Data and Niskin bottle samples from the Kingston plume site were also taken by CTD cast on RC0014, but only limited nutrient chemistry was done on these samples (Supplemental Tables T1 and T2). On these three cruises, fluid samples obtained with Niskin Bottles on the CTD cage were obtained from within a few meters of the bubble plumes, as indicated by reflections from the metal cage present within the surface ship sonar images.

The three seafloor fluid samples obtained by the ROV were obtained adjacent (<1 m) to the seafloor bubble orifices using Niskin bottles mounted on the front of the ROV. However, the number of near-bottom seafloor samples was constrained by limited bottom time for the ROV, poor visibility and imprecise acoustic navigation, resulting in only 3 discrete fluid samples being obtained within a few tens of centimeters from two of the active Seattle Fault/Alki Point vent orifices (Figure 1).

2.3 Flow-Through Methane Measurements of Surface Seawater

In April 2021, a flow-through CH_4 analyzer was installed on the *RV CARSON* cruise RC0052 to measure surface water methane concentrations (nM) and the stable isotopic composition of methane ($\delta^{13}C\text{-}CH_4$) during a survey of the Alki Point bubble emission field (Figure 5). This analyzer was attached to the seawater intake that provided continuous flow-through surface water to the Sea-Bird thermosalinagraph instrument that is part of the normal *CARSON* scientific instrumentation. Ship speed for most of the survey was 8 to 9 knots but slowed to 2 knots when directly over the Alki Point field (Figure 5). The partial pressure of methane (pCH_4; atmospheric units) in surface waters was measured using a headspace equilibration chamber (Frankignoulle et al. 2001) interfaced to a Los Gatos Methane Carbon Isotope Analyzer cavity ringdown spectrometer, which also provided the carbon stable isotopic data. Methane concentration range for this instrument is 10-500 ppm; precision for ^{13}C is 1 per mil, and for pCH_4 is 0.2% across the range which for 100 ppm would be +/- 0.2 ppm. The maximum drift for isotope measurements is 2 per mil and not applicable for concentrations. Factory specifications and calibration data for this instrument are available in the Supplemental Text T-1.

Equilibration chamber headspace pCH_4 was converted to CH_4 concentrations based on the ideal gas law as follows:

$$[CH_4] = pCH_4 \, x \, \frac{1}{R \, x \, T} \qquad \text{(Equation 1)}$$

where, R is the universal gas constant (0.082057 L atm °K^{-1} mol^{-1}), and T is temperature in °K. The concentration of methane dissolved in the surface waters was then calculated based on the temperature-dependent Henry's Law constant (K_H) (Wiesenburg and Guinasso, 1979):

$$K_H = e^{\left[-68.8862 + \left(101.4956 \, x \frac{100}{T}\right) + \left(28.7314 \, x ln ln \left(\frac{T}{100}\right)\right)\right]} \qquad \text{(Equation 2)}$$

where T is temperature now in °C units.

2.4 Helium Sampling

Samples for helium analysis were taken at the Kingston (RC0019) and Alki Point (RC0038) plume sites using standard Niskin bottles attached to the CARSON CTD cage. At the Alki Point site, fluid samples were obtained by attaching two additional Niskin bottles to the front of a small commercial Phantom ROV. These Niskin bottles were the traditional 'non-gas-tight' sampling containers. Helium isotope measurements were performed by the NOAA laboratory at PMEL Newport, Oregon (Baumberger et al, 2018; Supplemental T-3). Immediately upon recovery of the CTD/Niskin package and the ROV onto the surface ship deck, air-free water samples were flushed through 24-inch-long sections of refrigeration grade Cu tubing with duplicate half-sections cold-weld sealed (RC0038) or flushed through 32-inch-long sections of refrigeration grade 3/8 inch OD Cu tubing with a clamp seal (RC0019) for later laboratory analysis (Young and Lupton, 1983). Isotope ratios and concentrations of helium were then determined at the NOAA/PMEL Helium Isotope Laboratory in Newport, OR, USA using a dual collector, 21-cm-radius mass spectrometer with 1σ precision of 0.1 to 0.3 % in δ^3He, where δ^3He is the percentage deviation of $^3He/^4He$ from the atmospheric ratio, and a concentration accuracy of 1% relative to a laboratory air standard.

2.5 Temperature of the Plume Fluid

A NOAA Miniature Autonomous Plume Recorder (MAPR) with temperature, pressure, turbidity, and negative ion sensors was also attached to the ROV frame for our Kingston and Alki

Point cruises (Baker et al., 1997; Walker et al., 2004; Figure 6). No temperature anomalies were detected with the surface ship CTD sensors and only extremely modest +0.005°C to +0.010°C temperature anomalies higher than the adjacent seawater values were observed when the MAPR was deployed directly overlying active vent orifices by the ROV at the Alki Uplift field (Figure 6). The temperature resolution of the MAPR is 0.001°C and the time interval for the temperature spikes shown in Figure 6 was approximately 35 seconds. With a geothermal gradient of approximately 25°C/km present in the Puget Sound forearc region (Blackwell et al, 1990; Salmi et al, 2017), the modest MAPR temperature anomalies indicate that the upwelling vent fluid and gas from the sub-surface must have been in thermal equilibrium with the low temperatures found only in very uppermost tens of centimeters of seafloor sediments.

2.6 Routine Chemical Nutrients

Chemical analyses were also performed on the CTD/Niskin samples taken over both the Kingston (RC0019) and Alki Point (RC0038) plume fields and included traditional nutrient data of ($Si(OH)_4$, NH_4, [NO_3] and [NO_2]. These data are available as a Supplement Tables T-1 and T2. These data show only normal Puget Sound seawater composed of North Pacific and riverine mixture from estuarine circulation, without any of the chemical concentration anomalies that are routinely observed in forearc plumes within similar tectonic settings (Nakamura, 2014; Kusuda et al, 2014; Morikawa et al, 2016; Giggenbach et al, 1993, Batista-Cruz et al, 2019). Although Niskin bottles deployed on a CTD wire are not an efficient tool for capturing rising plume fluid, they have been widely and successfully used in both deep-water hydrothermal and continental methane studies. In this study however, the fluid captured by Niskin bottles appears to be only normal Puget Sound seawater.

2.7 Archive Data

In May of 1980, the newly-established Pacific Marine Environmental Laboratory of the National Oceanic and Atmospheric Administration-PMEL began a comprehensive study of particulate and dissolved organic compounds in Puget Sound that included a full-depth profile of dissolved methane that extended from north of Admiralty Inlet to south of the Tacoma Narrows (Curl et al, 1983). This government document was retrieved from the archives (https://books.google.com.gi/books?id=C_UeAQAAIAAJ&source=gbs_navlinks_s) and Figure 11 of the NOAA report was digitally extracted and the units of dissolved methane obtained from these CTD samples converted from original microliter/liter STP to the standard units of nM for the contour labels. This extracted Figure 11 from the NOAA/PMEL report is reproduced here as Figure 5. Although the 1980 sampling track did not pass directly over the Alki Point plume field, the closest point on the 2-D plot is indicated by a red star. The near-surface methane values in this reproduced figure closest to the Alki Point location are substantially higher (134 nM) than those measured by our recent flow-through surface measurements (30 nM) shown in Figure 5 taken 40 years later. This substantial variation in methane concentrations is a relatively common phenomena associated with time-series measurements of methane vents, which can vary by a factor of 10 on daily, annual, or decadal time scales (Salmi et al, 2011, Römer et al, 2016; Lee and Hautala, 2021; Ola Dølven et al, 2021).

2.8 Geological Context

Puget Sound is a glacially carved fjord within the forearc plate of the Cascadia Subduction Zone. The estuary forms the southern part of the Salish Sea, which also includes Hood Canal and the Straits of Georgia, which are linked to the North Pacific through the Straits of Juan de Fuca. In the north, Puget Sound is separated from the Straits of Juan de Fuca by a shallow 60-meter-deep double sill at Admiralty Inlet, located just north of the Kingston Arch (Figure 1; Thomson et al, 2007; Moore et al 2008; MacCready, 2020). South of the formal estuary entrance at Admiralty Inlet, Puget Sound covers a surface area of 2,330 km^2, has a length of 182 km, a 16 km maximum width, and a maximum water depth of 280 meters. The estuary is geographically separated into individual basins; the North Basin which includes San Juan Islands, the Main Seattle Basin that extends southward from Admiralty Inlet to Tacoma Narrows, the South Sound located south of the Tacoma Narrows, the semi-isolated Whidbey Basin, which is a land-bounded region located east of Whidbey Island that receives much of the Skagit River inflow, and the adjacent Hood Canal (Figure 1; Supplemental Figure S1).

Bathymetrically, these hydrologically-interconnected basins are partitioned by prominent shallow sills. Tidal flow over these sills induces turbulence that periodically disrupts the normal estuarine circulation which consists of a layer of relatively cold low-salinity riverine-input water flowing northward that overlies the warmer, generally southward-flowing, high-salinity Pacific Ocean water beneath (Cannon et al. 1990, Thomson et al, 2007, Sutherland et al., 2011, Ianson et al, 2016).

The majority of the Puget Sound bubble plume fields are located associated with surface traces of the three major regional fault zones that fracture the northern Cascadia forearc plate. These are the South Whidbey Island (SWIF), the Seattle Fault (SFZ), and the Tacoma Fault Zones (TFZ), which have been well-studied and mapped in considerable detail (Johnson et al, 1994, 1996, 1999, 2004; Pratt et al, 1997, 2003, 2015; Nelson et al, 2003; Brocher et al, 2000, 2001; Blakely et al, 2002, 2011; Sherrod et al, 2000, 2004, 2008; Clement et al, 2010). Some of the non-clustered bubble streams also overlie the unnamed North-South trending Seattle Basin Fault described in Johnson et al (1999; Figures 1 and 8B).

These regional fault zones have been proposed to penetrate completely through the North American forearc plate down to the Cascadia Subduction Zone plate interface approximately 43 km below the surface (Blakely et al, 2002, 2011; Hyndman et al, 2015; Wells et al, 2017). The specific details of these fault zones have been extensively discussed in Johnson et al, (1994, 1999); Gomberg et al., (2010); Blakely et al, (2011), Sherrod et al, (2000, 2004, 2008), McCrory et al, (2016) and Hyndman et al (2015) and are only briefly summarized below (Figures 8A and 8B). It is important to note here that there is a significant diversity in interpretations of these fault zones. As an example, published studies supportive of East-West trending cross faults in the Puget Lowland are Johnson et al. (1999), Calvert et al. (2003), and Mace and Keranen (2012), while other interpretations that do not show the cross faults were published by Blakely et al. (2002), Pratt et al. (2015) and the USGS Active Faults Database (2021). The present study remains agnostic regarding these different interpretations.

The South Whidbey Island Fault (SWIF) extends from southern Vancouver Island in the northwest to Seattle in the southeast and has produced at least four Holocene earthquakes with the most recent 2700 years BP (Sherrod et al, 2008; Sherrod and Gomberg, 2014). Kingston Arch is located on the southern edge of the SWIF zone and is a broad thinly-sedimented anticline that forms the northern boundary of the Seattle Basin (Johnson et al, 1999; Sherrod et al, 2008; Sherrod and Gomberg, 2014). The eastern submerged portion of the Kingston Arch anticline is the site of an extended linear North-South marine plume field located where the N-S trending fault zone described by Johnson et al (1999) intersects the southern part of the E-W trending SWIF (Figures 1 and 8A). The Kingston Arch anticline is at the junction of these two obliquely intersecting faults and originated as an isolated basin-margin fold that began in the late Eocene and remains seismically active well into the Holocene (Johnson et al, 1994, 1996; Brocher et al, 2001; Mace and Keranen, 2012; Pratt et al, 1997, 2015).

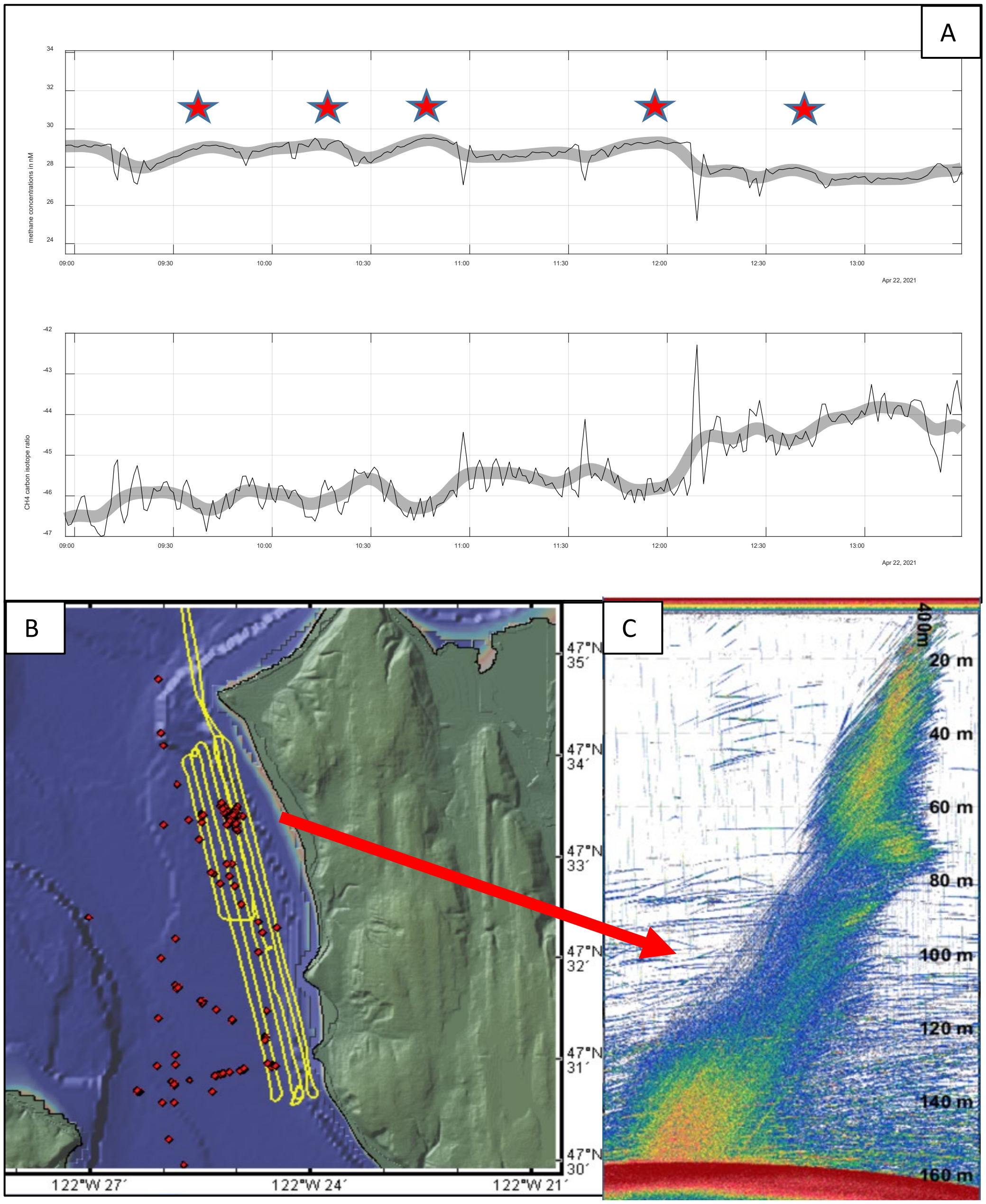


Figure 5: CARSON survey of the Alki Point plume field with the flow-through methane analyzer sampling of the surface waters. **Panel A:** Top curve shows methane concentrations (in nM) of the surface water with red stars indicating passes over the large cluster of plume emissions. Thin black lines are unsmoothed data showing sharp turns where the seawater intake was lifted above sea level and grey curve is 7-point smoothing of the data. Bottom curve shows co-registered CH_4 carbon isotope ratio with both unsmoothed (black) and smoothed (grey) data. **Panel B:** Yellow line of ship track for survey in Panel A, with red dots showing methane emission sites. **Panel C**: Image of methane bubble plume from Alki Point field from CARSON 38 khz sonar during survey, red arrow points to plume location.

The East-West trending Seattle Fault Zone (Figures 1 and 8B) is a south-dipping reverse fault that has over the course of the last 40 My lifted a hanging wall to overlie the Seattle Main Basin that lies immediately to the north. This central Main Seattle Basin is a 10 km deep depression filled with both Oligocene and younger Holocene/Quaternary sedimentary rocks, that can be up to 400 meters thick in a few regions (Johnson et al., 1994, 1996, 1999, 2004; Pratt et al., 1997; Blakely et al., 2002; Kelsey et al, 2008; Brocher et al., 2001, 2004; ten Brink et al., 2002; 2006). The Holocene and Quaternary sediment thickness is extremely variable both along- and across-strike, thinning over uplifts (i.e., Kingston, Alki Point) and thickening to several hundred meters in mid-basin. Relevant to this study, sediment is almost absent at Alki Point where the methane plume fields are dense and their emissions vigorous (Figure 5A and 5C), Johnson et al, 1999; Johnson, Samuel., pers. comm. 2021).

The most recent major earthquake on the SFZ was a magnitude M7 about 915 A.D. (Bucknam et al., 1992; Mace and Keranen, 2012). During this seismic event, shorelines on southern Bainbridge Island were uplifted by 4 meters (Sherrod et al., 2000; Kelsey et al., 2008), with uplift also occurring near Alki Point (Figures 8A, 8B and 9): ten Brink et al, 2006; Kelsey et al., 2008). The Alki uplift and the broader SFZ experienced approximately 8 meters in vertical motion during this earthquake and both the north and south boundaries of this uplifted block at Alki Point are active marine methane vent sites (Figure 8A, 8B and 9; ten Brink et al, 2006).

Within the Main Seattle Basin where the East-West trending Seattle Fault is located, Johnson et al (1999) used USGS Multi-Channel Seismic profiles to map a series of north-south trending faults that strike orthogonally across the east-west trending axis of the Seattle Uplift (Figure 8A and 8B). These north-south faults within the Main Seattle Basin are associated with some of the individual isolated plume emission sites that do not appear in clustered fields. Such spatial variability suggests that the active faulting in the Puget Basin may be both more complex and perhaps more abundant than has been previously recognized (Savage and Brodsky, 2011).

The Tacoma Fault Zone (TFZ, Figure 1) is a south-verging thrust fault that is the structural contact between the Main Seattle Basin in the north and the South Sound to the south (Johnson et al., 2004; Sherrod et al., 2004; Blakely et al 2011). Trenching of terrestrial fault exposures showed that TFZ active tectonic events occurred in A.D. 770, 900-930, and 1160 (Sherrod et al., 2004). These geological records of active faulting and lidar-illuminated terrestrial scarps demonstrate the

SWIF, the SFZ and the TFZ still pose a significant modern seismic hazard to the heavily-populated central and southern Puget Sound regions (Sherrod et al, 2004).

Puget Sound sediments have a complex depositional history and include several layers of glacial and inter-glacial sediments beneath the uppermost Quaternary layer, while below that are Eocene turbidites, which in turn rest on the volcanic Eocene Crescent basalts (Johnson et al.,1994, Rau and Johnson,1999). Although poorly sampled, post-glacial Holocene sediments in the uppermost part of the Seattle Basin seafloor consist primarily of clay and silt, with only minor sand (Crecelius et al, 1975; Grundmanis, 1990; Cochrane et al, 2015). Johnson et al. (1999, their Figure 3) show that the terrestrial portion of the Puget Basin includes several layers of glacial and inter-glacial sediments at elevations above sea level, which are the above sea level strata that hosts the water wells and warm springs that tap aquifers with the helium-3 signature (Booth, 1994; McCrory et al, 2016).

The location of the marine methane emission sites overlying deep-seated forearc fault zones initially suggested that gas and fluid could arise from the underlying plate interface of the Cascadia Subduction Zone, as has been noted in terrestrial aquifers that are immediately adjacent to the estuary (McCrory et al, 2016: Figures 1 and Supplemental Figure S1). This would be similar to fluid and gas vents located in other global forearcs, where the depth to the plate interface is similar to the 43 km depth beneath Puget Sound (Doğan, et al, 2006, Reynard, 2016). In support of this initial hypothesis, the regional faults that penetrate through the Cascadia forearc crust have been previously proposed as pathways for upward migration of the inter-plate fluid generated from heating along the subduction zone below Puget Sound (Hyndman et al, 2015, McCrory et al, 2016, Wells et al, 2017; Delph et al, 2018).

Wells et al. (2017) suggested that over-pressured fluids within the subduction zone interface that are in contact with the underlying oceanic plate are incompletely drained by the regional faults in the forearc and that this partial and episodic fluid drainage controls the temporal segmentation of the 14-month cycle of Episodic Tremor and Slip present in northern Cascadia (Chapman and Melbourne, 2009; Gao and Wang, 2017; Nakajima et al, 2018). The permeability and transport properties of these deep fault conduits in the Cascadia forearc have also been suggested to be episodically enhanced by incident seismic waves from either local or distant earthquakes (Hyndman, et al. 2015; Audet and Burgmann 2014; Vidale et al, 2011).

The offsets of the East-West Seattle Fault traces produced by the North-South along-strike faults of Johnson et al (1999) could provide the high crustal fluid permeability that has been previously observed in both estuarine faults (Naudts et al, 2012) and terrestrial fault zones (Evans et al, 1997; Brixel et al, 2020, Sibson, 1996, Bense et al, 2013: Figures 8A and 8B). The intersection between two faults with different directional trends provides wide fault-damage zones with high permeability that are transport pathways for upwelling crustal fluid (Savage and Brodsky, 2011; Sun et al, 2020).

3 Results

Abundant multi-channel profiles have examined the sub-seafloor of Puget Sound in both across- and along-strike surveys of the North-South trending topography of the estuary. Correlation of these tectonic features with the locations of bubble plume clusters detected by the *RV CARSON* single-beam sonar data shows that most of the large multi-plume vent fields are underlain by these deep regional fault zones (Figure 9), and this is particularly evident at the Kingston (SWIF) and Seattle Fault Uplift (SF) fields. As a caveat, the SWIF and SF areas are both located where the MCS profile coverage is extensive, and the bubble plume fields are also well-surveyed by the *RV CARSON* sonar tracklines, suggesting a probable sampling bias to the emission site distribution.

Some terrestrial-bounded estuaries with very high sedimentation rates have been shown to host methane bubble streams which are not associated with underlying methane reservoirs or permeable fault zones (Judd and Hovland, 2009; Römer et al, 2014). However, gas emissions from sediment pores from these recent estuary deposits require sediment deposition rates that are an order of magnitude higher than the 0.5 cm/year of Puget Sound measurements in the Main Seattle Basin (Judd and Hovland, 2009; Lavelle et al, 1986). Further, limited sampling shows the uppermost layer of Puget Sound sediments has a well-defined sulfate-methane-transition zone which is depleted in CH_4 and extends for at least a meter in depth in the estuary (Sultan et al, 2016; Grundmanis, V., 1990).

Puget Sound bottom water is well-flushed tidally and the bottom waters of all three basins are very well-oxygenated (Deppe et al, 2018). Due to methanotrophic bacteria and a well-developed sulfate methane transition zone, the upper sediment layers of the estuary do not appear to be able to produce free-methane bubble streams without accumulation within a sub-surface reservoir. Any vertical gas transfer likely requires transport by way of a pre-existing, perhaps faulted pathway,

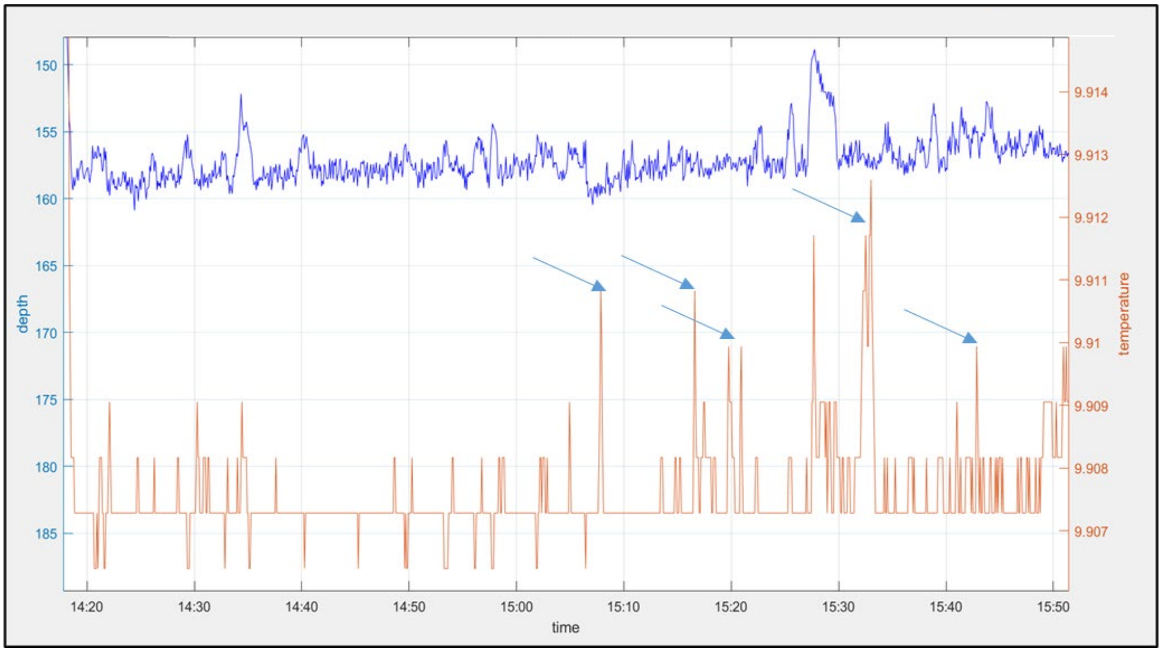


Figure 6: Top blue curve shows water depth above the bubble plume vent orifices at the Alki Point plume field, showing changes in elevation above the seafloor; shown to illustrate that the small temperature anomalies are not due to changes in altitude above seafloor. Bottom red curve shows the temperature data from the NOAA MAPR instrument. Data have abrupt changes due the sensor A2D converter resolving weak temperature variations. Arrows point to temperature anomalies associated with passage of the ROV directly over the vent orifices.

similar to other methane gas emission sites (Prouty et al, 2020). While several areas in Puget Sound show single isolated bubble streams, most identified gas emission sites are tightly clustered in areas that either overlie the boundaries of the well-mapped regional fault zones (Figure 9) or are distributed along the linear fault traces and are particularly abundant in clusters where fault zones with different geographical orientations intersect.

In dramatic contrast to our expectations derived from other global forearc vents and from the terrestrial aquifer data from water wells and warm springs that permeate the Cascadia forearc (McCrory et al 2016), our limited Niskin fluid data from the marine bubble plume field showed only normal Puget Sound seawater. Specifically, these samples were composed of Northeast Pacific Ocean seawater mixed with a variable amount of freshwater riverine input (Moore et al., 2008). Also contrary to our expectations from other subduction-zone forearc vents which exhibit elevated temperatures, no thermal anomalies were identified within our limited sampling of Puget

Sound plume fields. However, given the resource constraints of this pilot program, it remains possible that more well-positioned samples from the plume vent orifices might yield different and more satisfying results.

The one unequivocal result is that there is a high abundance of methane bubble plumes within Puget Sound. These plume emissions are spatially correlated with the traces of major fault zones that penetrate deeply into the Cascadia forearc (Figures 1 and 9). However, this limited field program observed (a) extremely small temperature anomalies near the bubble emission sites which indicates that gas provides the prime buoyancy driving the plume flow, (b) normal North Pacific seawater chemistry and salinity for the water samples taken in close proximity to bubble plumes, (c) based on the 1980 and 2021 surveys shown in Figures 5 and 7, methane is the most probable plume gas, and (d) an absence of any helium-3 isotope anomalies indicative of a deep igneous or a mantle-related source region for the plume fluids.

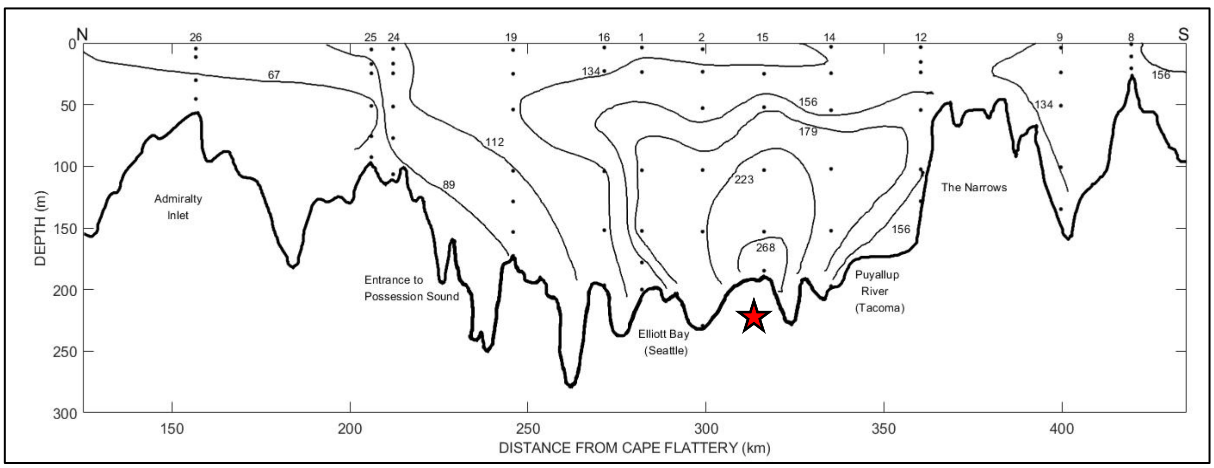


Figure 7; Figure extracted from a 1983 NOAA/PMEL Report showing contoured data from a series of CTD casts taken from Admiralty Inlet to South of the Tacoma Narrows (for locations, see Figure 1). Dissolved methane concentrations are in units of nM. Small black dots are the positions of the Niskin bottle samples. Red star is approximate position of the Alki Point plume field shown in Figure 1.

A flow-through seawater analyzer measuring methane concentrations and carbon isotopes in CH_4 show that fluids at the sea surface above the Alki Point plume site have concentrations of methane that are orders of magnitude higher than both North Pacific water and atmospheric abundances (Figure 5). The estuary methane also appears to have carbon isotope ratios weakly consistent with a thermogenic rather than biogenic origin, although oxidation by methanotrophic

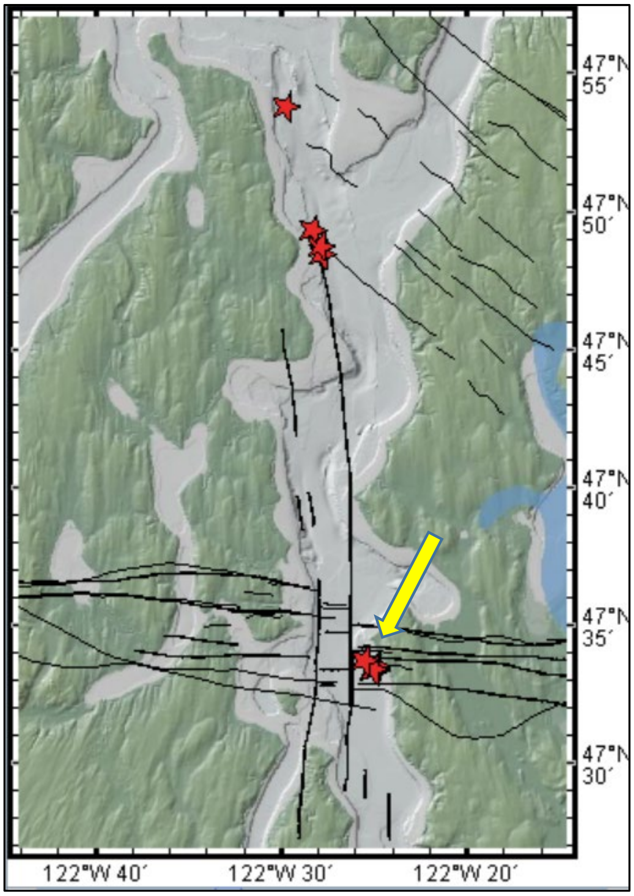
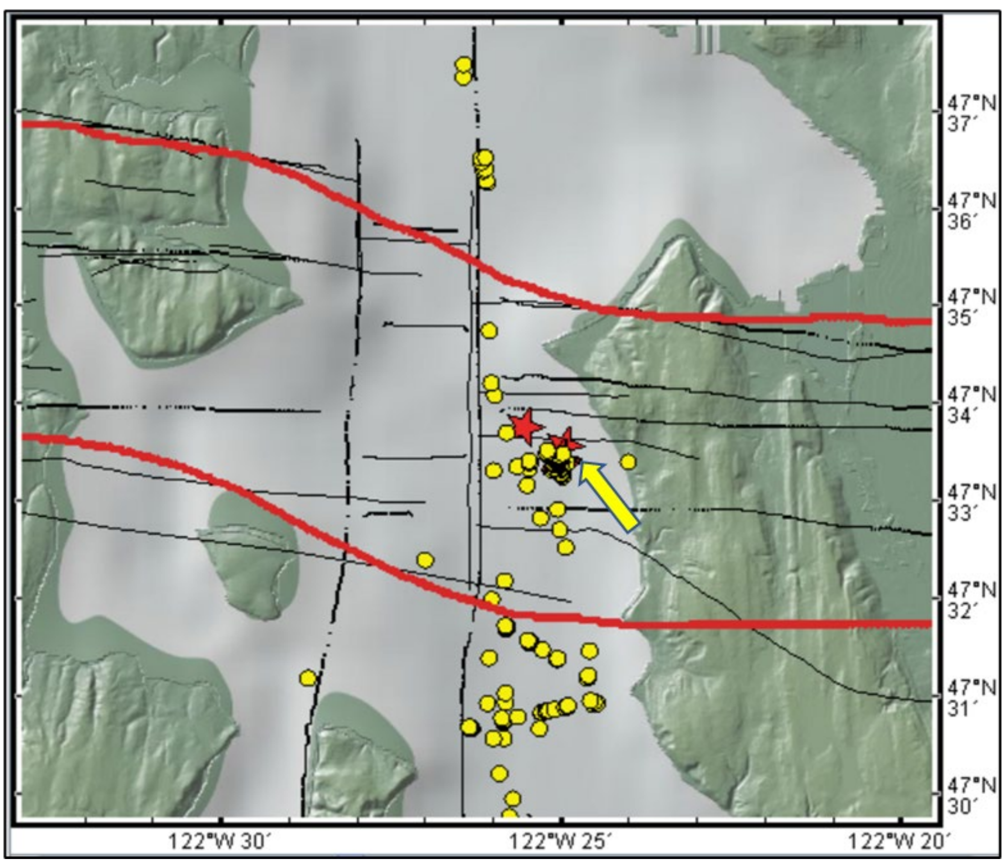


Figure 8A: Top Left – Red stars show locations of CTD casts and yellow arrow points to ROV dive site. Black lines are USGS fault trace compilations with the addition of the N-S Johnson et al (1999) fault lines from multi-channel seismic surveys. Gray regions are seawater and green areas terrestrial.

Figure 8B: Bottom: Expanded view of Alki Uplift site, red stars are CTD casts, yellow arrow points to ROV dive sites. Heavy red lines denote boundaries of the Alki Uplift of ten Brink et al (2006). Yellow circles are methane plume sites, showing locations that are both overlying the primary fault traces and also at the fault edges.



bacteria within the water column can confuse this bimodal interpretation (Figure 5). Isotopic analysis of both carbon and hydrogen in methane from fluid issuing directly from the seafloor orifice rather than the sea surface would be a far more definitive test, but these samples are not yet available and logistically challenging to obtain.

The correlation of the Puget Sound plume emission sites with the locations of major regional fault zone traces strongly suggests that relatively recent tectonic activity may provide vertical pathways through the Holocene sediments deposited on the estuary seafloor, which varies widely in thickness within the Basin (Johnson et al, 1999). This hypothesis may be valid for plumes located in the middle of the basins, where bubble streams are active - even though the recent sediments can reach several hundreds of meters in thickness in these areas. All three of the regional Puget Basin fault zones are still considered tectonically active (Sherrod et al, 2004, 2008). This tectonic activity may disturb the recent seafloor sediments and generate pathways for an initially deep-seated source of gas of unknown initial composition that is substituted by methane during upward transit through the sediment column (Algar and Boudreau, 2010; Algar et al, 2011; Boudreau, 2012). This model of a deep reservoir for the methane, while initially appealing, is not supported by the temperature or chemistry data of the plume fluid that we sampled and have described above.

The recent limited methane data from surface waters shown in Figure 5 is corroborated by 1980 archive NOAA/PMEL data (Figure 7) that shows that Puget Sound has high methane concentrations throughout the entire estuary and within the entire water column. These concentrations were extremely high in the Main Seattle Basin near Alki Point, where the density of plume emission sites is also high (Figures 8B and 9). The archive 1980 data indicates that the current methane emission sites have been active for decades, and their correlation with presently active emission sites overlying fault zones suggests an even longer-term geological process.

A major unresolved issue is the lack of any chemical or thermal indication of a deep source for the bubble plumes. In Puget Sound, the nearest terrestrial water-well with a mantle helium-3 signature is located on Bainbridge Island (Figure 1). This water-well is only 5 km distant from the nearest Puget Sound bubble plume vent site at Alki Point, and both the terrestrial and marine sites appear to be associated with the same branch of the Seattle Fault Zone (Figure 1). Terrestrial water-wells located near the Kingston Arch plume field have similar helium-3 signatures (Figure 1), suggesting that the helium-3 isotope results from the Bainbridge Island water well is not an isolated phenomenon. The proximity of terrestrial aquifers with clear helium isotope anomalies, identified on the same fault traces as the marine plume sites suggests but certainly does not prove, that the terrestrial and marine fluids could perhaps arise from a common sub-surface reservoir, but without a common helium-3 signature. This presents an unresolved paradox that requires additional study.

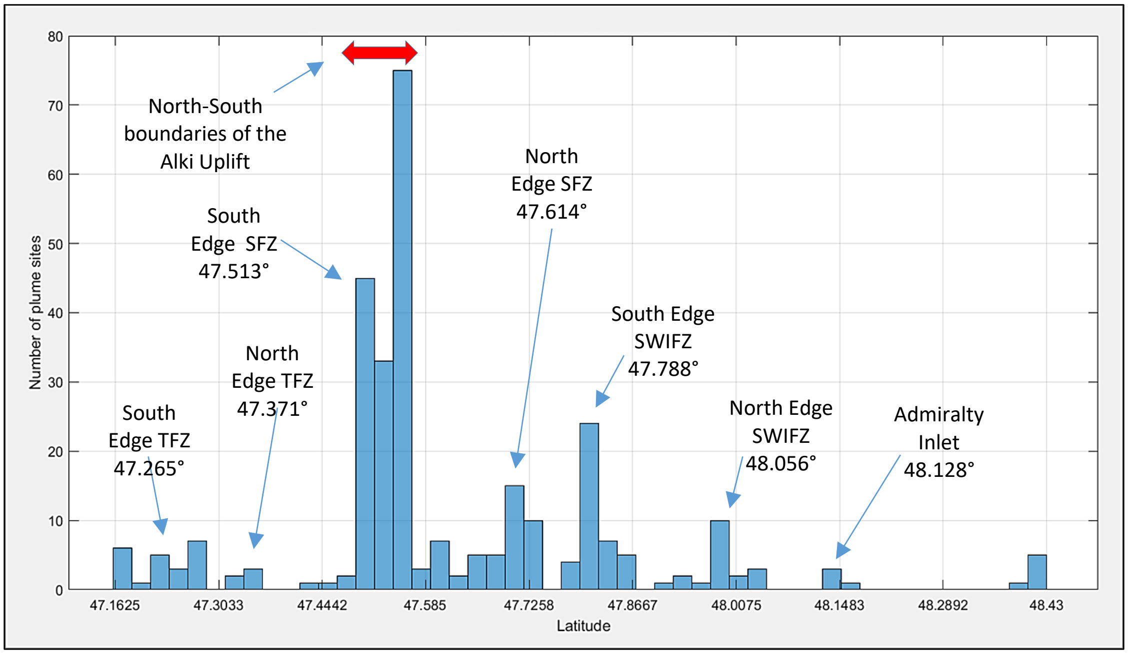


Figure 9: Histogram of plume site frequency with respect to the north-south latitude of the major fault zones, where TFZ is the Tacoma Fault, SFZ is the Seattle Fault, and SWIFZ are the South Whidbey Island Fault Zones. Admiralty Inlet is the northern boundary of the Puget Sound basins; sites farther north are within the Straits of Juan de Fuca. Note that the highest frequencies of plume sites are positioned at the bounding edges of the fault zones, and not within their central regions. The Alki Uplift is a prominent faulted block at the southern edge of the SFZ identified by ten Brink et al (2003). Kinston Arch is an uplifted anticline at the boundary of the Southern SWIFZ (Johnson et al., 1996). The large number of plume sites at the Alki Uplift is die in part a sampling bias due to frequent cruises to that general area.

4 Conclusions and the Paths Forward

Puget Sound contains an unexpectedly large number of methane bubble plumes that, along with a yet-unmeasured seasonal riverine input, maintain a high level of methane supersaturation with respect to the atmosphere. Extensive single-beam sonar surveys by the *RV CARSON* show that the bubble plume sites are distributed throughout the Sound, with clustered fields that are concentrated near three major regional fault zones. Although Puget Sound lies within the Cascadia Subduction Zone forearc and these regional fault zones appear to penetrate deeply into the forearc plate, our new data appears to strongly suggest a much shallower source region may be responsible for the methane plume gas and fluid.

Our limited Puget Sound plume chemistry does not support the hypothesis of a direct origin of the vent bubbles arising directly from the underlying Cascadia subduction zone plate interface. The low temperatures of the fluid also seem to exclude even a shallower sub-surface igneous rock reservoir such as the Siletz Terrain (Wells et al, 2017). We acknowledge that our data are incomplete, and our preliminary sampling and analysis of vent fluids are too limited to provide confirmation of any satisfying hypothesis. However, the presence of at least 330 individual methane bubble plumes within the Puget Sound forearc associated with the major deep-seated fault systems that penetrate the forearc crust merits presentation to the wider scientific community to provide context for more complete further studies.

Without more definitive data, a wide range of alternative explanations for the bubble plumes exists and a more compelling and satisfying hypothesis may be confirmed by subsequent field programs. Primarily these future field programs would include better methods for vent fluid sampling, more extensive chemical analyses, and high-resolution seismic imaging of the vent fields (Sassen et al., 2003). Comprehensive sampling programs of emission fluid chemistry at a wider range of marine sites, including the Kingston Arch field and the plumes located in the deeper, more thickly sedimented mid-basins. Additional sampling and chemical analysis of the three terrestrial water wells that show mantle helium-3 isotope signatures and are located adjacent to the marine vents could be helpful (i.e., on Bainbridge Island and west of Kington; Figure 1). These sampling programs could determine if the terrestrial wells and the marine vents originate from the same subsurface source area and use the similar migration pathways. Systematic long-term time-series measurements of the bubble emission flux could be correlated with external forcing including earth and seawater tidal forcing, storm surges, the 14-month cycle of Episodic Tremor and Slip events and/or the passage of distant seismic waves. These forcing processes have been shown elsewhere to modulate vent flow and therefore provide considerable insight regarding the depth of the source area and the properties of the transport pathways.

Some possible alternatives for the marine plume source include spring sapping flow from terrestrial aquifers that are adjacent to the aquifer (Brooks et al, 2021), the presence of an undetected/unrecognized sub-seafloor fluid reservoir beneath Puget Sound (Morikawa et al, 2016, Tauzin et al, 2017), or simple vertical compression of the seafloor sediments (Wang et al, 2018) where the extruded fluid migrates horizontally and then upward through disturbed sediment pathways defined by movement along the regional faults. Our data does not support or negate any of these hypotheses.

However, in spite of the limitations of this study, one observation remains. Puget Sound, an estuary within the deeply fractured forearc plate of the Cascadia Subduction Zone, hosts an abundance of methane gas plumes that correlate with these regional fault zones (Figure 9). Although the flux of methane emission from these plumes still remains unquantified, comparison with other continental margin vent sites suggests that the methane from the plumes may impact the general biological and chemical environment of the Puget Basin, as similar plumes do elsewhere (Schubert et al, 2006; Jordan et al, 2020). As the methane-rich seawater of the Sound is eventually discharged by estuarine circulation through the Straits of Juan de Fuca into the coastal NE Pacific, the influence of these bubble plumes, generated within an inland sea, may potentially extend farther west, into a far wider environment.

Acknowledgments, Samples, and Data

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1. The co-authors have no financial conflicts-of-interest with this study.
2. The 1983 NOAA/PMEL document that is the source for Figure 7 is available at (https://books.google.com.gi/books?id=C_UeAQAAIAAJ&source=gbs_navlinks_s)
3. All plume location data will be submitted to the appropriate federal database and

additionally available on the Corresponding Author's (H.P. Johnson) UW website (https://www.ocean.washington.edu/home/H_Paul_Johnson).

4. Rolling-Deck-2-Repository database for the 2011 *RV THOMPSON* cruise TN 265 is available at (https://www.rvdata.us/).

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Figure Captions

Figure 1: Map of Puget Sound and Hood Canal in north-central Washington State. Yellow circles are sites of individual bubble plume emissions based on CARSON single-beam 38 kHz sonar. Black lines are trace projections of the SWIF, SFZ and TFZ faults and the N-S Hood Canal and Puget Sound faults of Johnson et al., (1999). Black squares are urban sewer outfalls. Red triangles are locations of hot springs and water wells with helium isotope anomalies indicative of a deep subduction zone source (McCrory et al, 2016). Green triangles are CTD casts from the CARSON.

Figure 2: Panel A: Yellow circles show bubble plume locations from CARSON sonar acquired near Kingston Arch with red star indicating location of fault offset co-located in Panel B. Red line is MCS trackline for image in Panel B. **Panel B:** Offset sediment layers from the South Whidbey Island Fault Zone from archived MCS data (Johnson et al., 1996). Red stars in Panels A and B are co-located. **Panel C:** CARSON 38 kHz sonar image of bubble plumes at Kingston Arch, showing both gas bubbles (center) and fluid emissions (right) at position of red star in Panel A. Heavy red trace is the bottom reflection

Figure 3: Panel A: Yellow circles are bubble plume locations near Alki Point within the projections of the Seattle Fault zone (Figure 1). Red line is approximate trackline for MCS image in Panel B. Red stars in all 3 panels are co-located. **Panel B:** CARSON sonar image of gas plumes shown in Panel A. **Panel C:** Archive MCS profile of sub-surface structures beneath the bubble plume site, showing basement rock uplift.

Figure 4: Panels A and B: RV Carson 38 khz sonar profiles across Puget Sound over the projected locations of the Seattle Fault Zone, showing plume fields extending entirely across the Seattle basin (Figure 1). Sonar image locations are shown on lower map**. Panel C**: Map showing locations of sonar profiles in Panels A and B. Red stars are the center of the profiles in Panels A and B shown above. Yellow circles are individual methane plume locations.

Figure 5: CARSON survey RC0052 of the Alki Point plume field with the flow-through methane analyzer sampling of the surface waters. **Panel A:** Top curve shows methane concentrations (in nM) of the surface water with red stars indicating passes over the large cluster

of plume emissions. Thin black lines are unsmoothed data showing sharp turns where the seawater intake was lifted above sea level and grey curve is 7-point smoothing of the data. Bottom curve shows co-registered CH_4 carbon isotope ratio with both unsmoothed (black) and smoothed (grey) data. **Panel B:** Yellow line of ship track for survey in Panel A, with red dots showing methane emission sites. **Panel C:** Image of methane bubble plume from Alki Point field from CARSON 38 khz sonar during survey, red arrow points to plume location.

Figure 6: Top blue curve shows water depth above the bubble plume vent orifices at the Alki Point plume field, showing changes in elevation above the seafloor; shown to illustrate that the small temperature anomalies are not due to changes in altitude above seafloor. Bottom red curve shows the temperature data from the NOAA MAPR instrument. Data have abrupt changes due the sensor A2D converter resolving weak temperature variations. Arrows point to temperature anomalies associated with passage of the ROV directly over the vent orifices.

Figure 7; Figure extracted from a 1983 NOAA/PMEL Report showing contoured data from a series of CTD casts taken from Admiralty Inlet to South of the Tacoma Narrows (for locations, see Figure 1). Dissolved methane concentrations are in units of nM. Small black dots are the positions of the Niskin bottle samples. Red star is approximate position of the Alki Point plume field shown in Figure 1.

Figure 8A: Top Left – Red stars show locations of CTD casts and yellow arrow points to ROV dive site. Black lines are USGS fault trace compilations with the addition of the N-S Johnson et al (1999) fault lines from multi-channel seismic surveys. Gray regions are seawater and green areas terrestrial.

Figure 8B: Bottom: Expanded view of Alki Uplift site, red stars are CTD casts, yellow arrow points to ROV dive sites. Heavy red lines denote boundaries of the Alki Uplift of ten Brink et al (2006). Yellow circles are methane plume sites, showing locations that are both overlying the primary fault traces and also at the fault edges.

Figure 9: Histogram of plume site frequency with respect to the north-south latitude of the major fault zones, where TFZ is the Tacoma Fault, SFZ is the Seattle Fault, and SWIFZ are the South

Whidbey Island Fault Zones. Admiralty Inlet is the northern boundary of the Puget Sound basins; sites farther north are within the Straits of Juan de Fuca. Note that the highest frequencies of plume sites are positioned at the bounding edges of the fault zones, and not within their central regions. The Alki Uplift is a prominent faulted block at the southern edge of the SFZ identified by ten Brink et al (2003). Kinston Arch is an uplifted anticline at the boundary of the Southern SWIFZ (Johnson et al., 1996). The large number of plume sites at the Alki Uplift is due in part a sampling bias due to frequent cruises to that general area.