

## Supplemental Text

### Extensive decentralized hydrogen export from the Atlantis Massif

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## METHODS

### *Expeditions And Wireline Hydrocasts*

International Ocean Discovery (IODP) Expedition 357 utilized two seabed rock drills to core a series of boreholes on an east-west transect across the southern wall of the Atlantis Massif on the *RRS James Cook*, starting in October 2015. The water column was profiled directly over the drilling sites prior to coring to characterize the geochemistry prior to any drilling related disturbances. Samples for volatile analysis were collected near-bottom, typically <3 meters above the seafloor. Two additional casts were conducted away from the Atlantis Massif to collect background water and characterize the microbiological and geochemical signatures that are typical for seawater at these depths. A final cast was conducted specifically to capture fluids from the plume of the Lost City hydrothermal field. A total of 11 vertical casts (JC130\_001 through JC130\_11) were carried out (Table S2).

The sensors on the rosette included a Benthos 916T altimeter, SBE 3P primary and secondary temperature sensor, SBE 4C primary and secondary conductivity sensor, a Digiquartz pressure sensor, and a WETLabs BBRTD light scattering sensor.

The Lost City 2018 Expedition employed the *R/V Atlantis* and the ROV Jason to collect fluid and rock samples from the Lost City hydrothermal field and across the Atlantis Massif. During the expedition the water column was profiled with a total of 13 hydrocasts (AT4201-001 through AT4201-013), including 9 simple vertical casts, 3 modified vertical casts (where 2 to 4 separate positions were sampled during each cast and the CTD was brought to ~600 m water depth while the ship moved between lowerings), and one 13 km long two-yo.

The wireline rosette on the Lost City 2018 Expedition employed a *SeaBird 911plus* conductivity-temperature-depth (CTD) package, a VA-500 altimeter, an oxidation-reduction potential (ORP) sensor (PMEL-ORP4CTD, S/N 11) and a Optical Backscatter (OBS) sensor (Seapoint S/N 12325). Casts AT4201-001 through AT4201-009 also employed a second PMEL-OBS sensor (Seapoint S/N 12454). On casts AT4201-010 through AT4201-013 the second OBS sensor was replaced with a METS methane sensor (Franatech S/N 1507, calibrated 28-June-2018).

The first vertical cast (AT4201-001-V01) of the cruise was conducted during the transit to Lost City in deep waters. No samples were collected for methane and hydrogen concentrations. Seven samples were collected from the tow-yo (AT4201-002-T01) conducted along the east margin of the detachment fault east of Lost City. AT4201-003-V02 collected four different vertical profiles above the Lost City vent sites to capture the different standing plumes. Profile A was conducted over Marker H, profile B was conducted over Marker 6, profile C just south of Poseidon and IMAX, profile D over Marker 8. The next vertical CTD cast (AT4201-004-V03) was performed between Poseidon and Marker 6 with the aim to constrain the 750 m plume more thoroughly. AT42-005-V04 consisted of three vertical profiles conducted between Markers 8 and 2, along the ridge SW of Marker 8.

A high-resolution investigation was conducted on the Eastern wall of the Atlantis massif. Five vertical casts (AT4201-006-V05, AT4201-010-V09, AT4201-011-V10, AT4201-012-V11, and AT4201-013-V12) with bottles triggered every 10 meters were conducted to further investigate fluid flow from the wall and comparison with the fluids from the boreholes. All triggered bottles were samples for methane and hydrogen concentration analysis. AT4201-007-V06 was a deep cast at corner of Atlantis Fracture and the MAR with the goal to investigate for hydrothermal venting in the fracture. The two remaining vertical casts, AT4201-008-V07 and AT4201-009-V08 were conducted east of the MAR at the west end of the Atlantis fracture with the goal to investigate a massif similar to the Atlantis massif.

#### ***Expedition 357: Sensor Package And Niskin Bottles On Seabed Rock Drills***

On IODP Expedition 357 a sensor package with three 4L Niskin bottles was mounted on both the British Geological Survey (BGS) RD2 rock drill and the MARUM MeBo70 Rock drill. Since both drills use bottom-water as the drilling fluid, these samples should reflect a mixture of seawater with seafloor material. The bottles were mounted approximately 0.8 m above the seafloor on the RD2 rock drill and approximately 1.5 m above the seafloor on the MeBo. Two of the Niskin bottles were connected to teed tygon tubing that flushed water via a Seabird pump from a few centimeters away from the rock drill break out table through the bottles; the bottom end cap for these bottles was already in the closed position, while the tap end cap was left open to vent, with fluid flushing coming through the traditional sampling spigot which was left open. A one-way valve on the inlet prevented backflow out of the spigot. The third Niskin bottle was not flushed, with both end caps left open and the spigot closed. At the end of active drilling, or upon the request of the science party, the bottles were closed.

#### ***Lost City 2018: ROV Jason Sensors and Camera***

On the Lost City 2018 Expedition a series of sensors and cameras were mounted on the ROV Jason. A Miniature Autonomous Plume Recorder (MAPR) sensor was mounted on the brow of the vehicle on every dive. It is custom built at PMEL and incorporates a temperature sensor, an oxidation-reduction

potential sensor, and a high sensitivity optical backscatter sensor (Seapoint Turbidity Meters 0-5 NTU range).

A series of flow through sensors were mounted on the starboard side of the ROV, with a titanium intake on the front end mounted in the front basket of the ROV. The inline sensors consisted of a CTD sensor (Seabird 19*plus*V2 S/N 6952, courtesy of the JASON group), a METS methane sensor (Franatech S/N 1507), an ORP sensor (PMEL-ORP, S/N 3), and a pump. After passing through the sensors the fluids were shunted to an open 5 L Niskin bottle by connecting tubing from the output of the ORP sensor to the stopcock of the Niskin with the lower endcap closed and the upper endcap left open. At the end of the operational use of the flow through sensor system on each dive, the Niskin was allowed to flush for 10 minutes before closing the upper endcap. The water samples were then analyzed for methane and hydrogen for comparison to the METS sensor readings.

ORP is a sensitive indicator to the presence of nanomolar (or greater) concentrations of reduced hydrothermal chemicals (e.g.  $\text{Fe}^{2+}$ ,  $\text{HS}^-$ ,  $\text{H}_2$ ) that are out of equilibrium with the oxidizing ocean. Hysteresis and drift are characteristics of normal sensor behavior and prelude reporting a specific “background” value, but sudden and sustained decrease in voltage resulting in an overall drop  $>0.5\text{mV}$  is diagnostic of hydrothermal effluent (Walker et al., 2007; Baker et al., 2016).

A Multidisciplinary Instrumentation in Support of Oceanography (MISO) camera was pointed downward from the vehicle. It consists of an Ocean Imaging Systems model DSC 24000 digital still camera (S/N 003) with a domed optical port. Strobes were mounted on the two swing arms and operated simultaneously with image capturing.

### ***Volatile Concentration Analysis***

On both Expeditions volatile concentrations were determined by drawing 100 mL of bubble free water from the Niskins directly into 140 mL plastic syringes fitted with two-way stopcocks. A 40 mL headspace of ultrahigh purity helium was subsequently added. The sample was vigorously shaken and allowed to warm to room temperature of the analytical laboratory to reach equilibrium. Aliquots of the headspace gas were injected into an SRI 8610C gas chromatograph equipped with a 30m-length Molecular Sieve 5A column (Restek MXT-PLOT, 0.53mm inner diameter) run at 40°C and 18 ml per minute with ultra pure helium as carrier gas.  $\text{H}_2$  concentrations were determined with a pulsed discharge detector, and  $\text{CH}_4$  concentrations were measured with a flame ionization detector. Gas concentrations were calculated based on 100 ppm ( $\pm 5\%$ ) standards of  $\text{CH}_4$  and  $\text{H}_2$  in helium (Scotty Analyzed Gases) diluted to 1, 2, 5, and 10 ppm with helium.

## **References**

Walker, S. L., Baker, E. T., Resing, J. A., Nakamura, K., and McLain, P. D. (2007). "A new tool for detecting hydrothermal plumes: An ORP Sensor for the PMEL MAPR," in Proceedings of the American Geophysical Union, Fall Meeting 2007, (Washington, DC: AGU).

Baker, E.T., J.A. Resing, R.M. Haymon, V. Tunnicliffe, J.W. Lavelle, F. Martinez, V. Ferrini, S.L. Walker, and K. Nakamura (2016): How many vent fields? New estimates of vent field populations on ocean ridges from precise mapping of hydrothermal discharge locations. *Earth Planet. Sci. Lett.*, 449, 186-196, doi: 10.1016/j.epsl.2016.05.031.

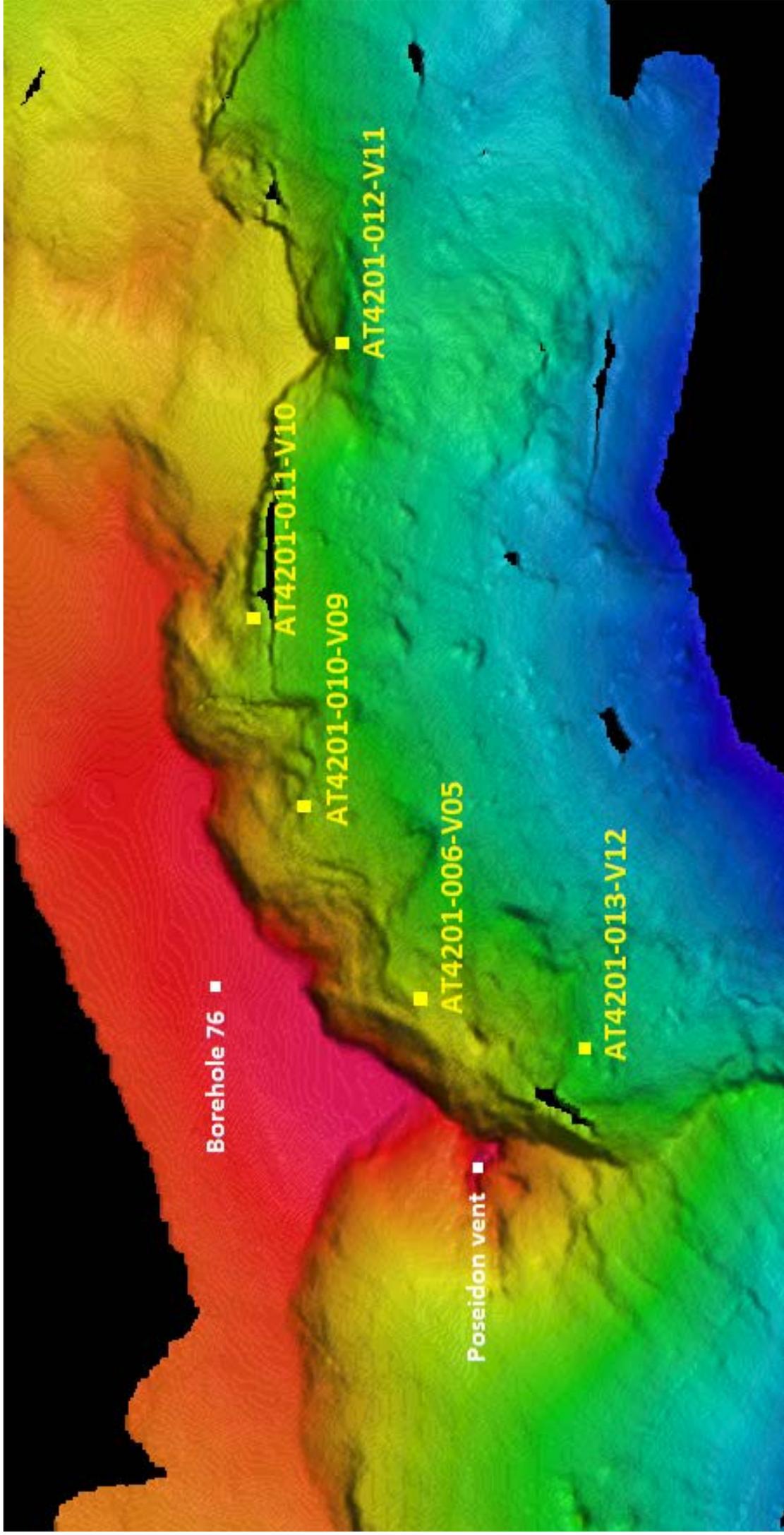


Figure S1. CTD casts along east wall (AT4201-006-V05, AT4201-010-V09, AT4201-011-V10, AT4201-012-V11, and AT4201-013-V12) are also shown over high-resolution ABE acquired bathymetry. Details on the casts can be found in Table S2.

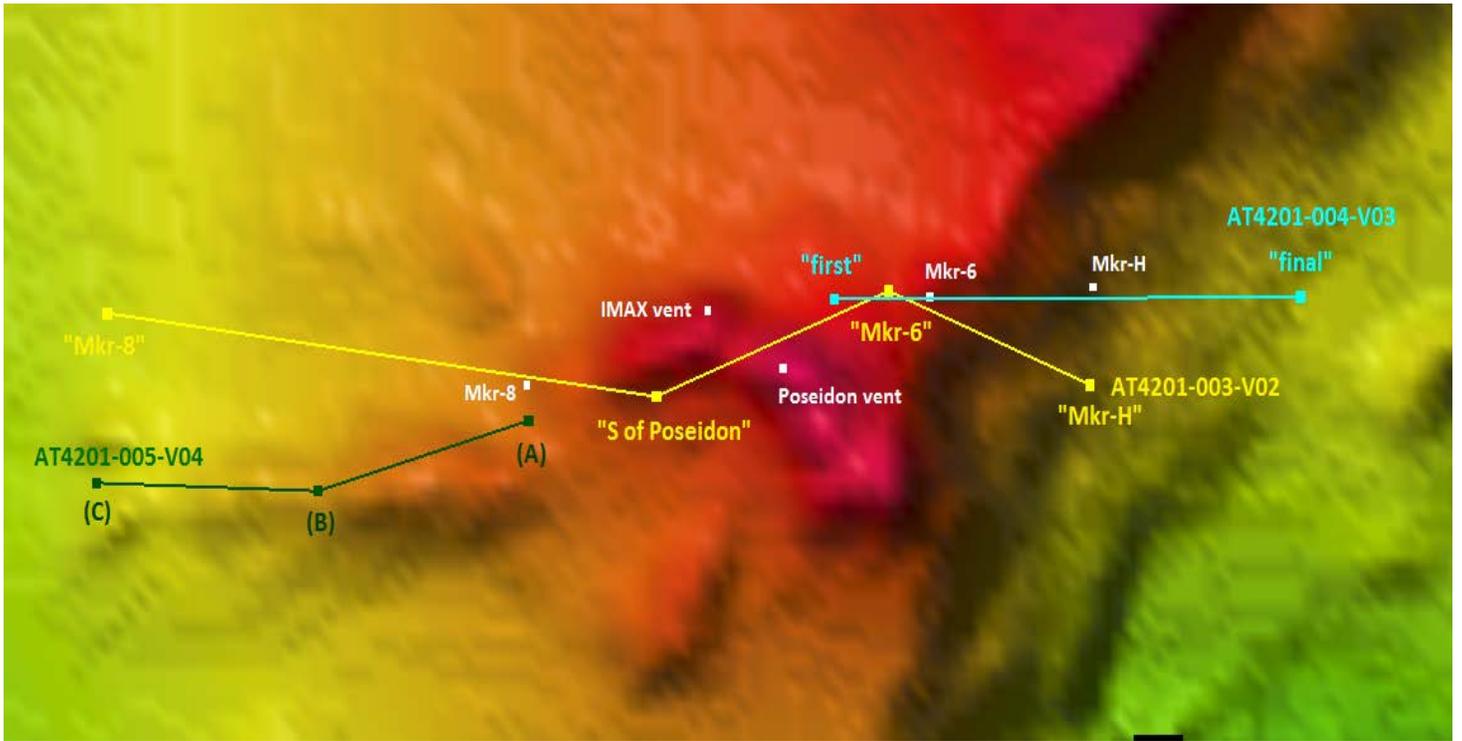


Figure S2. CTD casts over vents (AT4201-003-V02, AT4201-003-V02 and At4201-005-V04) to capture plume fluids shown over high-resolution ABE acquired bathymetry. Details on the casts can be found in Table S2.

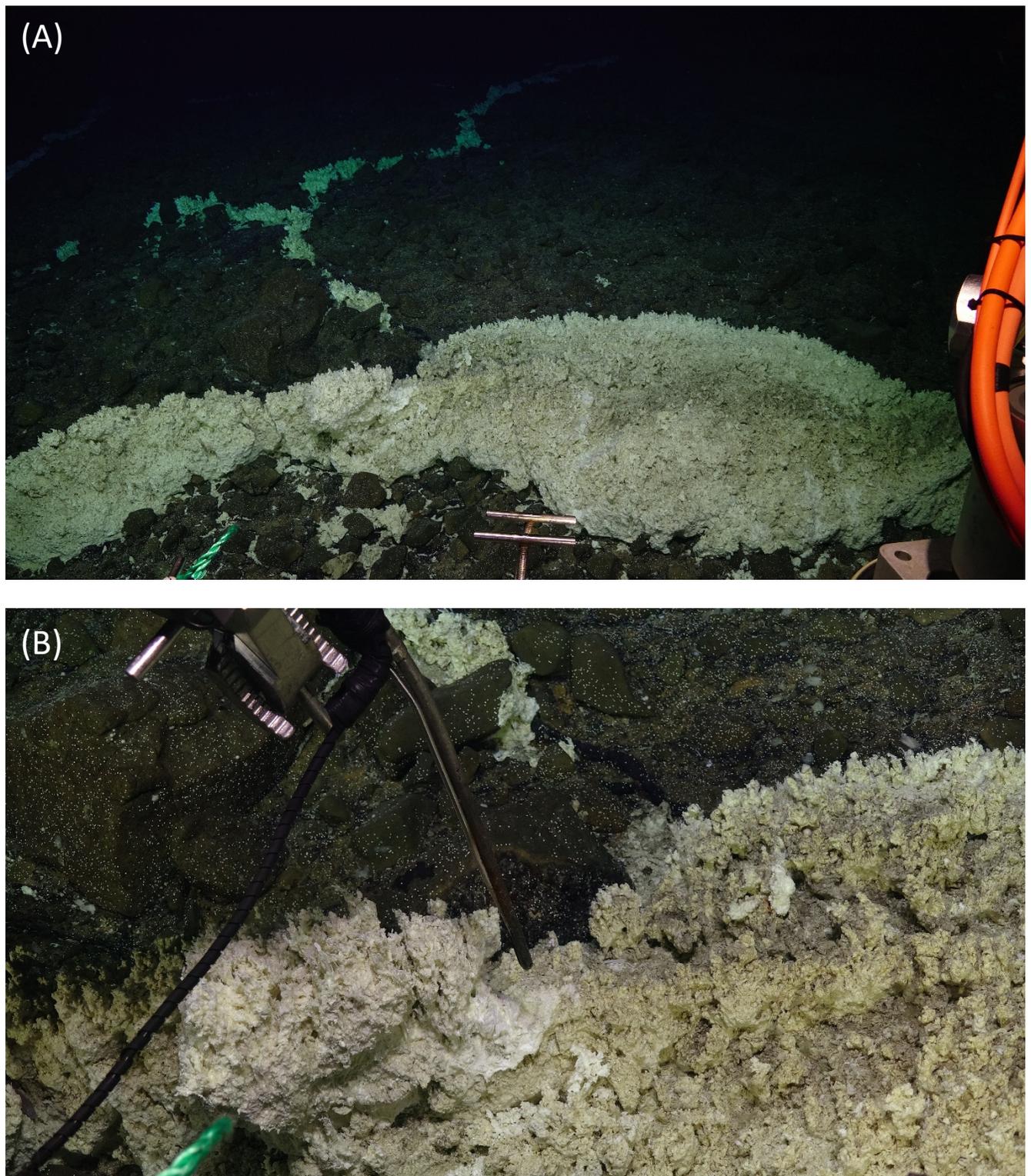


Figure S3. Cracks at the top of the Atlantis Massif, denoted as “carbonate veins” in Figure 1b. In (B) the high temperature probe of ROV Jason registered a reading of 1°C above background.

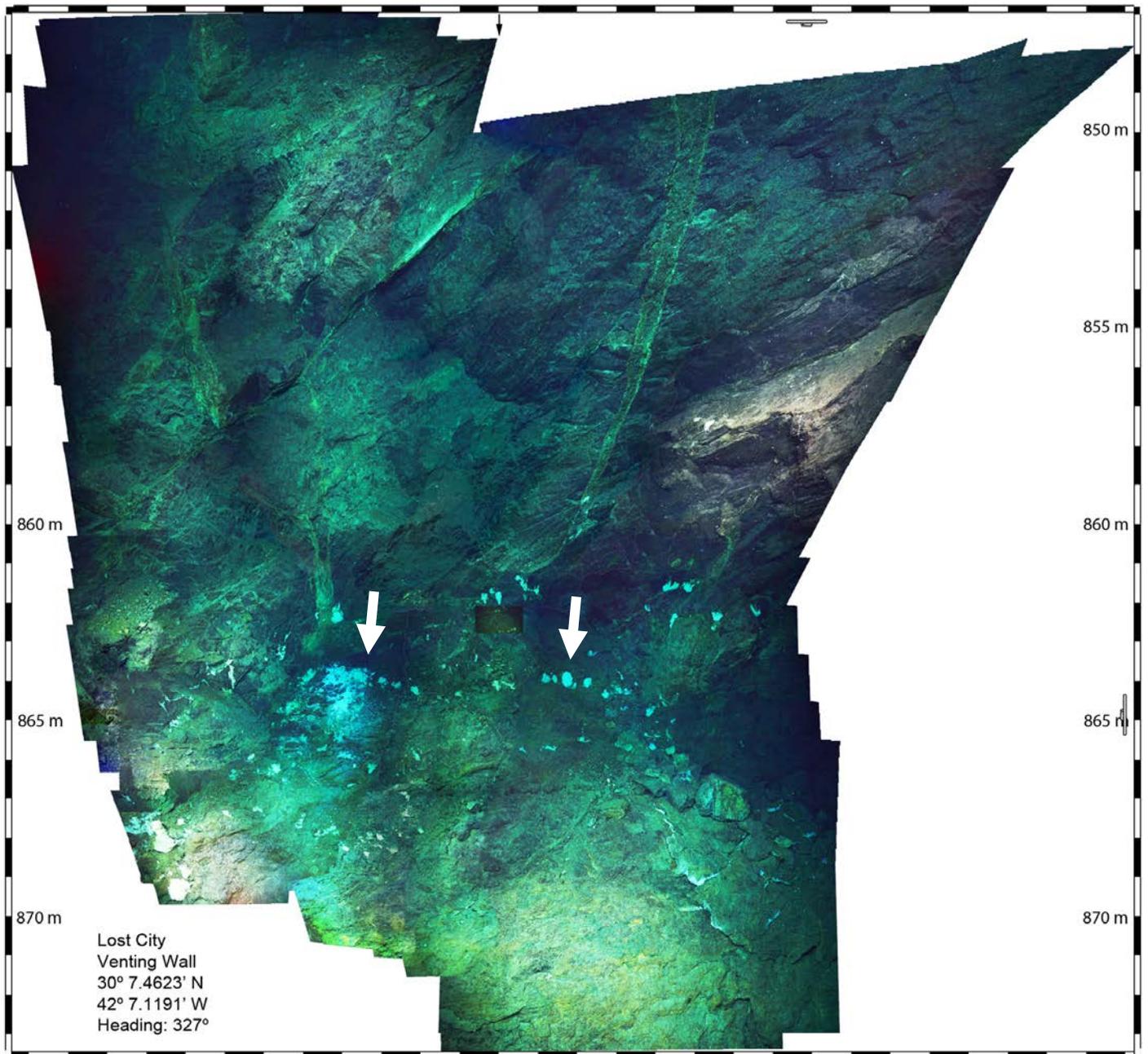


Figure S4. Photomosaic of carbonate deposits along the eastern wall of the Atlantis massif, denoted with a white star in Figure 1b. The white arrow on the left points to where a small chimney vented 22°C fluids; the chimney did not survive fluid sampling. The white arrow on the right points to the small chimneys depicted in Figure S3C.

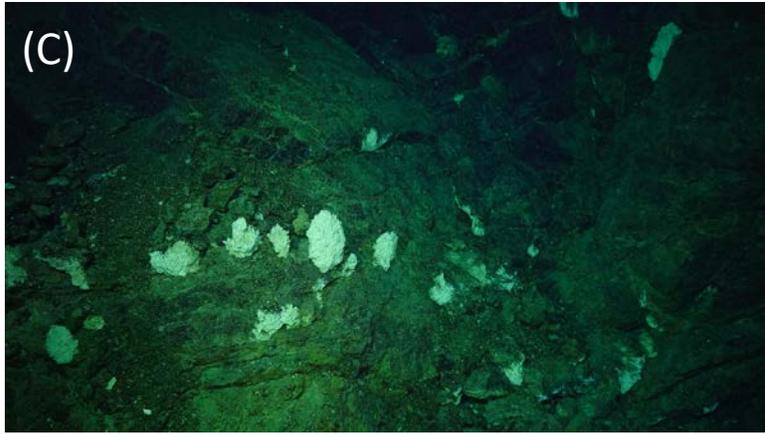
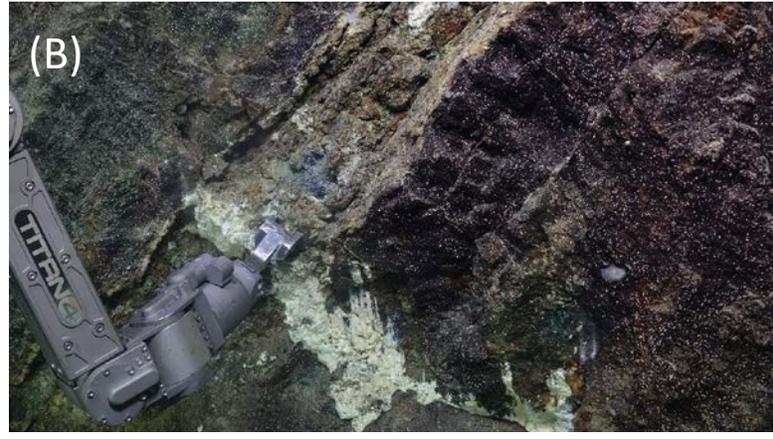
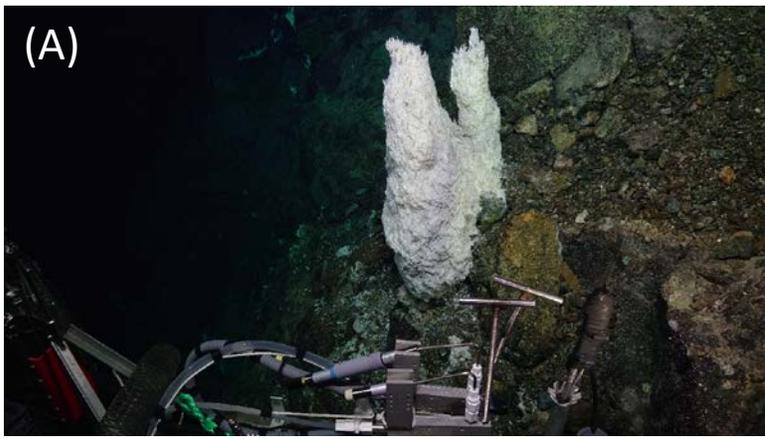


Figure S5. Diverse carbonate deposits indicating active or inactive fluid flow on the Eastern Wall of the Atlantis Massif. The structure in (A) actively vented 22°C fluids. The structures in (A) and (C) correspond to the white arrows in the mosaic in Figure S2. The remaining structures did not register elevated temperatures when tested.

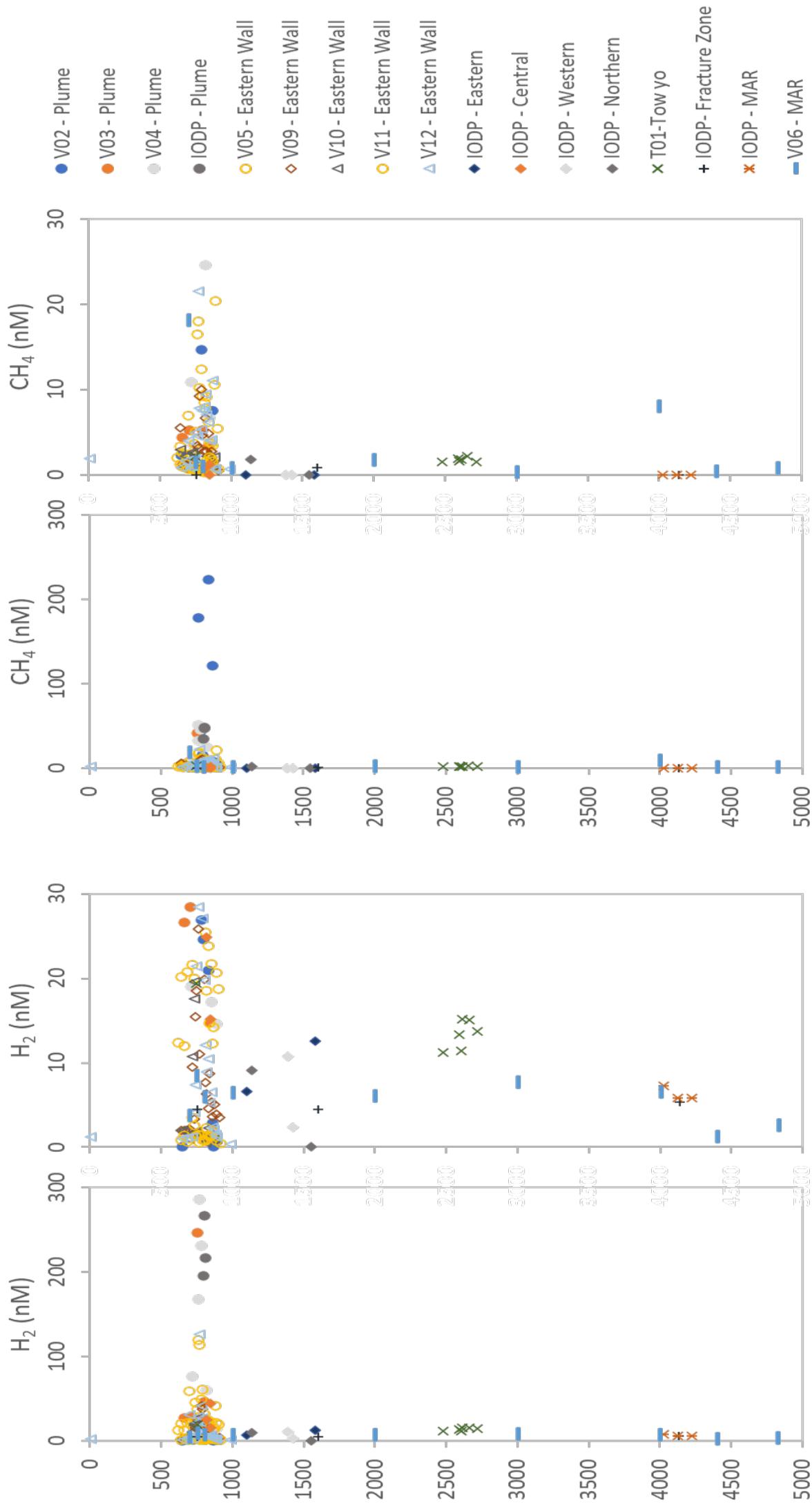


Figure S6. Concentrations of dissolved  $H_2$  and  $CH_4$  in nM from hydrocasts carried out during the two oceanographic expeditions. The specific hydrocasts are listed in Table S2. The second and fourth panels plot the same data as in the first and third panel, but focused on the lower concentration values.

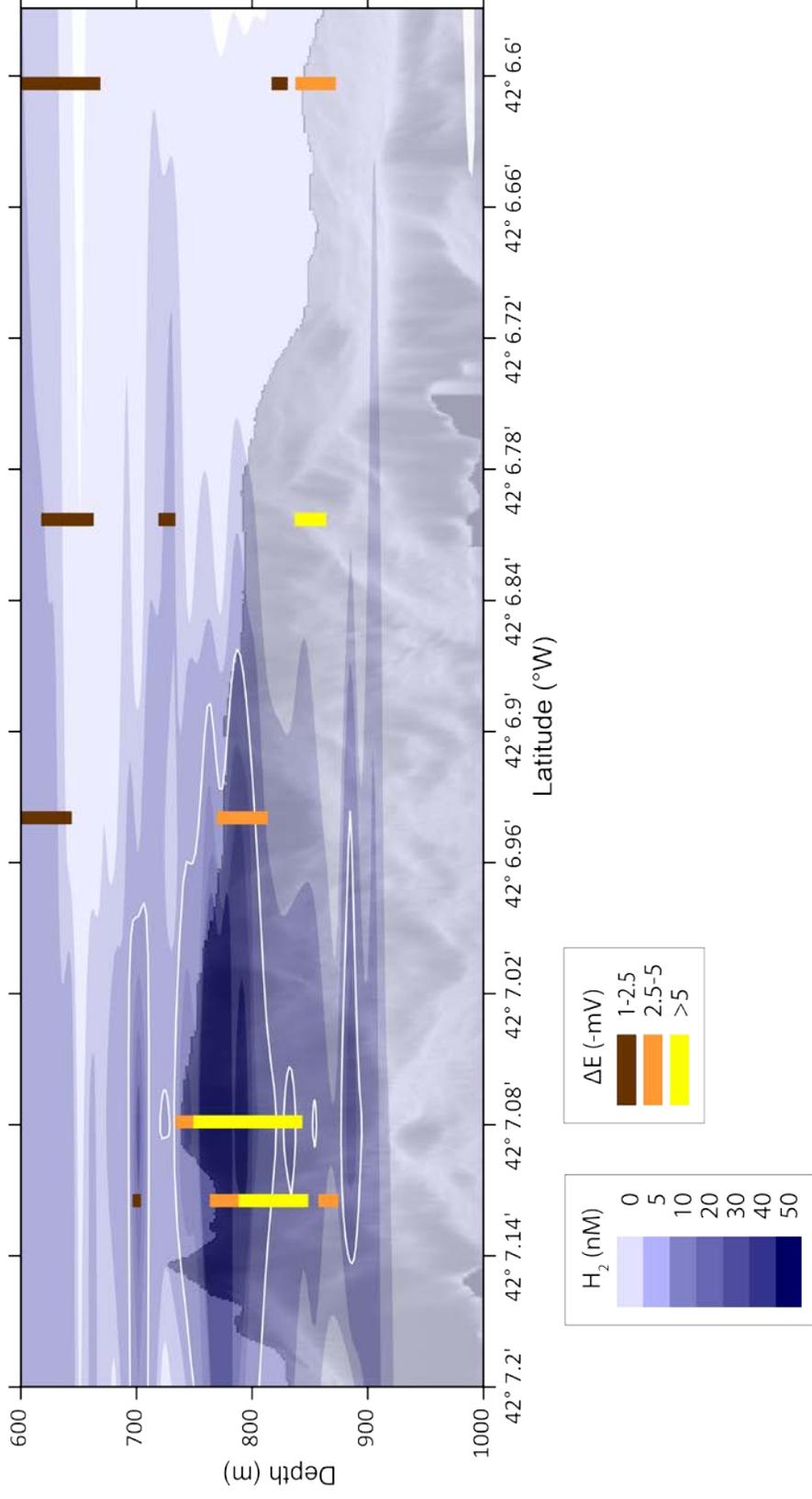


Figure S7. Hydrogen concentrations (contour lines) and  $\Delta E$  (vertical bars) from five vertical hydrocasts carried out across the eastern wall. Regions within the white contour lines have H<sub>2</sub> concentrations >20 nM.

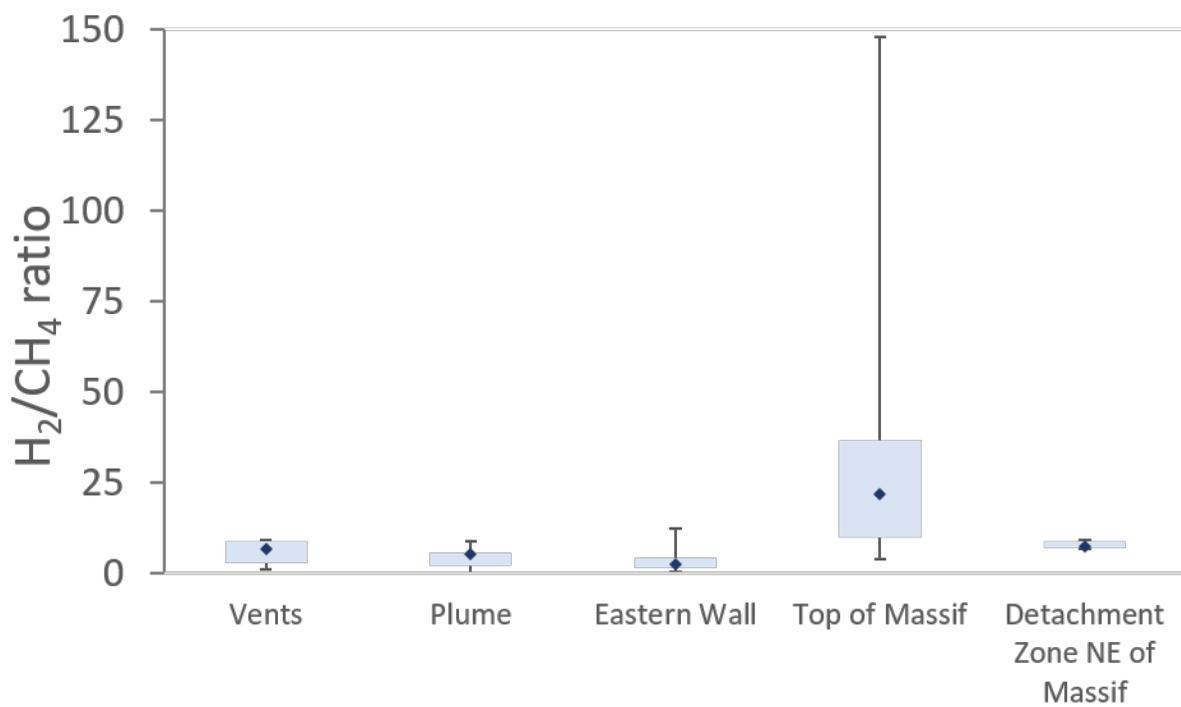


Figure S8. H<sub>2</sub>/CH<sub>4</sub> ratios of LCHF fluids and hydrocast samples.

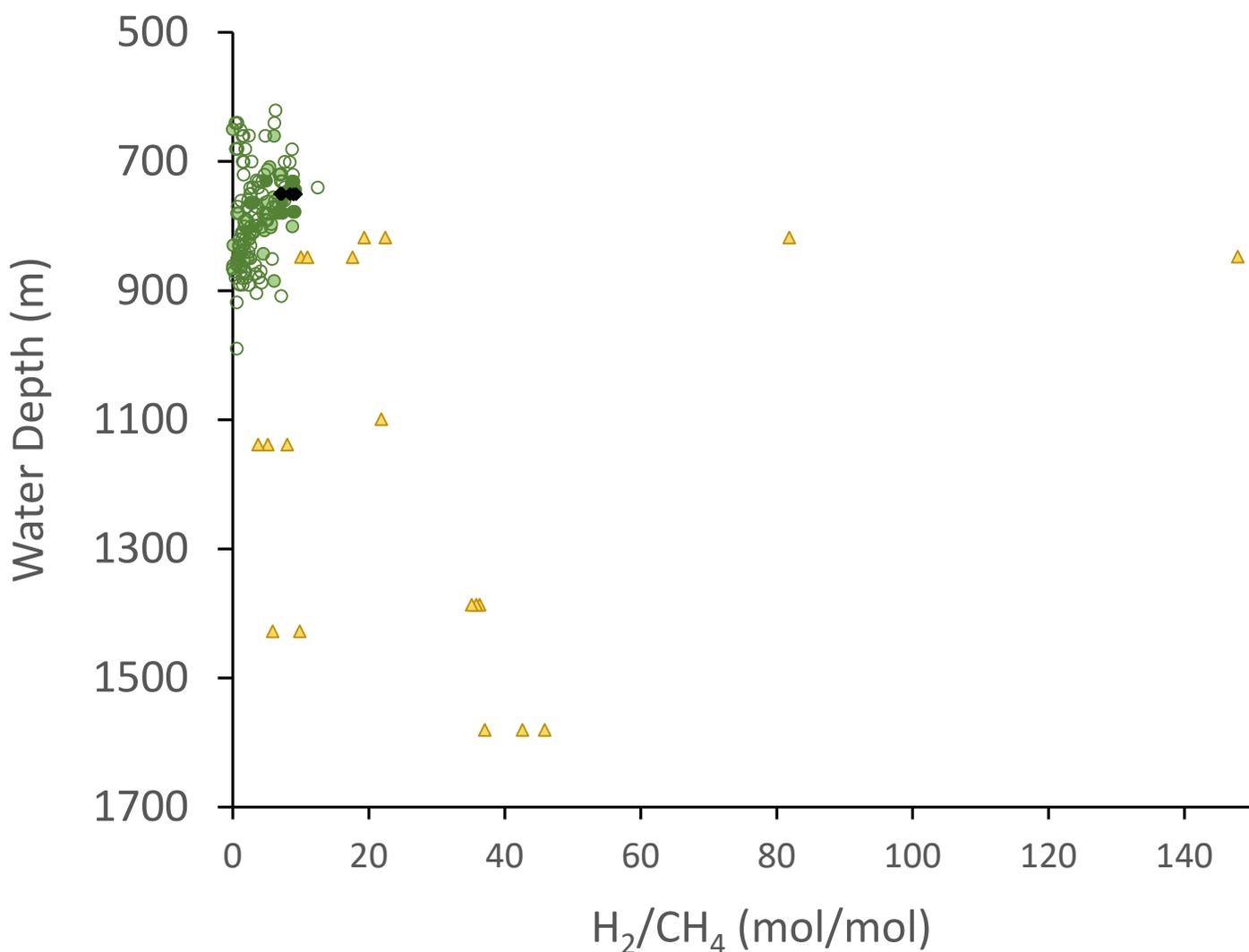


Figure S9. Water depth vs. H<sub>2</sub>/CH<sub>4</sub> ratios of fluid samples. Dark green circles are fluids collected from LCHF chimneys (Proskurowski et al., 2008). Light green circles are from the LCHF plume and empty green circles are from the eastern wall. Black circles are from the tow-yo along the termination of the detachment fault. These all group together, in contrast to the samples collected across the massif in yellow triangles.