

Variations in gas and water pulses at an Arctic seep: fluid sources and methane transport

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Introduction

We provide the basic information for the 12 study sites in this paper (Table S1). The porewater samples were collected during the three cruises in 2015 and 2016. We report the concentrations of dissolved sulfate, chloride, strontium, and lithium as well as the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios from the porewater for seven of the sites. For the other sites, only the sulfate profiles were shown to support our general observations. Sediment collection and porewater sampling procedures were detailed in Hong et al. (2017). Dissolved sulfate and chloride were measured by ion chromatography at the Geological Survey of Norway while dissolved strontium and lithium were measured by ICP-OES (Leeman Labs Prodigy) in the W.M. Keck Collaboratory for Plasma Spectrometry at Oregon State University using the axial viewing mode. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were measured with a Nu Plasma multicollector inductively coupled plasma-mass spectrometer (MC-ICPMS) in the W.M. Keck Collaboratory as described in Joseph et al. (2012; 2013). Porewater samples for cation analyses (<6 mL) were acidified onboard with 10 μL of 69% ultrapure grade nitric acid which lowered pH to <2. Prior to analyses, the porewater samples were diluted 20-fold with 1% quartz-distilled nitric acid. Repeated IAPSO and an in-house standard were measured every 11 samples to assess accuracy and precision of the measurements (accuracy: Li<2.8%, Sr<3%; precision: Li<1.3%, Sr<1.3%, n=12). For the $^{87}\text{Sr}/^{86}\text{Sr}$ analyses, a 30 μL aliquot of the acidified subsample was dried and brought up to 75 μL of 3M HNO_3 in preparation for porewater $^{87}\text{Sr}/^{86}\text{Sr}$ analyses. The samples were then passed through Eichrom “Sr-spec” chromatographic columns (Horwitz et al., 1992) to isolate strontium. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (at 30 ppb) were measured with a Nu Plasma multicollector inductively coupled plasma-mass spectrometer (MC-ICPMS) in the W.M. Keck Collaboratory as described in Joseph et al. (2012; 2013). Instrument mass bias was corrected using $^{88}\text{Sr}/^{86}\text{Sr}$ of 8.375209 and $^{87}\text{Sr}/^{86}\text{Sr}$ data were normalized to the NBS 987 standard, with a reported $^{87}\text{Sr}/^{86}\text{Sr}$ value of 0.710245, with an internal error of ± 0.000067 ($2\sigma_{\text{mean}}$, n=48). Replicate analysis of an in house standard yielded a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.708189 ± 0.000067 ($2\sigma_{\text{mean}}$, n=48), representing our external error. A data table is included in Dataset S1.

A gas sample was collected by a remotely operated vehicle above GHM3 in 2016 (Fig. 1c). The concentrations of hydrocarbons as well as the stable carbon and hydrogen

isotopes of methane were analyzed at Hydroisotop GmbH (Germany) with a GC-MS-IRMS system (see Sauer et al. (2015) for the details of analyses). The isotopic results were reported in ‰ (δ values) against the international standards Vienna Pee Dee Belemnite (VPDB) for carbon isotopes and Vienna Standard Mean Ocean Water (VSMOW) for hydrogen.

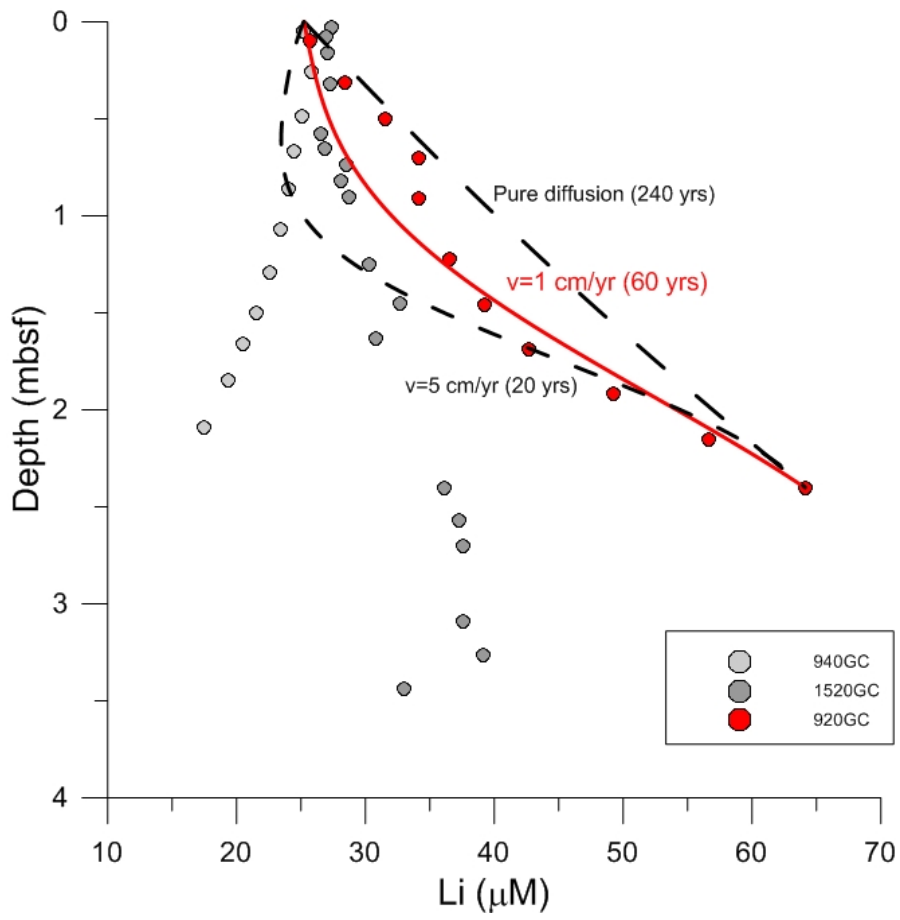
We also provide the details about our 1-dimensional model on the diffusion and advection of Li in the pore fluid. The current model was modified from the reduced model in Hong et al. (2017). We considered a 2.4-meter sediment column with the Li concentration set to be constants at the top and bottom boundary conditions (25.23 and 64.14 μM , respectively). Initial profile was assumed using the Li profile from 940GC. Only solute diffusion and advection are considered which were modeled in a sequential fashion: diffusion was simulated in the first half of the time step following the other half time step of advection. Such sequential fashion is advantageous for its easiness of implementation despite the smaller discretization needed to ensure accuracy (Peszynska et al., 2016). Diffusion step was solved by an implicit Crank-Nicholson scheme while advection was solved explicitly. Model results with several different advection rates used were shown in Figure S1.

Table S1. Water depth, recovery, and location of the six study sites

CORE ID	GHM ID	Water depth (m)	Recovery (m)	Lat	Lon
CAGE15-2-911GC	3	379	0.85	76.1069 N	15.9677 E
CAGE15-2-940GC	3	386	3.10	76.1069 N	15.9779 E
CAGE15-6-1520GC	3	386	2.90	76.1057 N	15.9661 E
CAGE15-6-1521GC	3	386	0.95	76.1060 N	15.9638 E
CAGE15-6-1522GC	3	388	3.20	76.1071 N	15.9579 E
CAGE15-2-918GC*	4	371	0.75	76.1168 N	16.0439 E
CAGE16-5-1081GC*	4	369	0.85	76.1170 N	16.0432 E
CAGE15-2-920GC	5	386	3.50	76.1117 N	16.0108 E
CAGE16-5-1048GC*	5	387	3.35	76.1123 N	15.9974 E
CAGE16-5-1068GC*	5	384	2.95	76.1123 N	16.0052 E
CAGE16-5-1069GC*	5	383	2.27	76.1120 N	16.0056 E
CAGE-16-5-1070GC*	5	385	3.26	76.1117 N	16.0027 E

* Sites with less completed dataset which will not be focused in the discussion.

Figure S1



Dataset S1. Concentrations of dissolved sulfate, chloride, strontium, and lithium as well as the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios in the porewater samples from seven of the sites. Only sulfate concentration data from the other additional sites were included.

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