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Supporting Information for

**Grand Challenge: Timescales and Processes of  
Methane Hydrate Formation and Breakdown, with  
Application to Geologic Systems**

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**Introduction**

The main contents in the supporting information are:

- Text S1 describes general information about the movies S1, S2, and S3
- Captions for Movies S1, S2, and S3

## Text S1

The Supporting Information contains descriptions of movies that demonstrate natural phenomena related to gas hydrate formation and breakdown as filmed at the seafloor of the northern Gulf of Mexico in 2014 (Lobecker et al., 2019) and 2017 (Kennedy et al., 2019) and the U.S. Atlantic margin offshore Virginia in 2019 (Cantwell et al., 2019). All videos were acquired by the remotely operated vehicle Deep Discoverer 2 (D2) during expeditions of the *Okeanos Explorer* research vessel. D2 is operated by the National Oceanographic and Atmospheric Administration's (NOAA's) Ocean Exploration and Research Program.

The movie clips provided here were extracted from public domain low-resolution movies using Adobe Photoshop software. The full-resolution movies can be found in data releases by Cantwell et al. (2019), Kennedy et al. (2019), and Lobecker et al. (2019).

Whenever two laser dots appear in these videos, their separation is 10 cm. These videos were mostly filmed in a close-up mode for which the lasers cannot be used.

**Movie S1.** Three-phase processes at the seafloor within the hydrate stability zone. This movie corresponds to Figure 7a in the main text. The video was filmed on the seafloor of the northern Gulf of Mexico at ~16:32 UTC on December 20, 2017, during *Okeanos Explorer* expedition EX1711 (Ruppel and Amon, 2017). The full-resolution video can be downloaded from Kennedy et al. (2019). The field of view for the portion shown in Figure 7a is fewer than 5 cm. The video shows vapor phase gas being emitted at the edge of a carbonate overhang where gas hydrate (orange) and sediment (gray) are intermixed. The seafloor at this location lies within the P-T conditions for stability of most common gas hydrates of methane or methane and higher order hydrocarbons. Contact with seawater could cause dissolution of the gas hydrate. High concentration of hydrate-forming gas dissolved in the bottom water would counteract dissolution, as would the presence of a thin layer of oil or other material on the surface of the hydrate (would not be visible in the video). The vapor phase gas being emitted at the edge of the overhang emerges as bubbles in some spots, and these immediately ascend through the water column and out of the frame of view. Vapor phase gas feeds into tubes of clear gas hydrate (looks like water ice) in other spots. Some hydrate-encrusted tubes have a bulbous protrusion at the end where the hydrate has formed around and "frozen in" a gas bubble. This process of vapor phase gas pushing forward within the hydrate stability zone and quickly acquiring a hydrate coating is similar to the process that may occur when vapor phase gas

invades sediments that are already within the P-T conditions for hydrate stability. While the camera is focused on the overhang, the center of the video shows one hydrate-encased bubble and tube that can be seen growing very slowly before it eventually detaches. Note that hydrate and gas are both buoyant relative to seawater, so the detachment may be driven by building gas pressure more than mere buoyancy of the hydrate and enclosed gas. The gas inside the tube does not form solid hydrate because insufficient water is available within the tube. In other spots along the overhang, gas hydrate has formed around an inverted teardrop-shaped bubble as it was emitted. During some of the video, migrating gas can be seen through the clear sides of the tallest hydrate chimney on the right. In this chimney, a bubble routinely ascends to the top, is held in place briefly, and then detaches and rises through the water column. Hydrate (slightly opaque frosty material) may be forming on the bubble when it is stuck at the top of the chimney, but this is difficult to verify visually. Near the end of the video, gas bubbles begin escaping from the side of this chimney, which then detaches.

**Movie S2.** Three phase processes at the seafloor within the hydrate stability zone. This movie corresponds to Figure 7b in the main text. The video was filmed on the seafloor of the northern Gulf of Mexico by D2 at ~19:41 UTC on April 12, 2014, during *Okeanos Explorer* expedition EX1402. The full-resolution video can be downloaded from Lobecker et al. (2019). The field of view is a few centimeters. It is relatively unusual for gas hydrate to form and persist on bare seafloor because ocean bottom waters are normally very undersaturated in hydrate-forming gas. This patch of seafloor has various rates of gas emission and different size bubbles. Some bubbles become coated in gas hydrate as they emerge, while others are emitted and ascend immediately out of view. Note particularly the repeated development of short columns of gas encased in hydrate and growing upward due to gas fed from below. The chimneys then detach and a new chimneys forms. Emission conduits with higher flux may produce bubbles, while lower flux may lead to the formation of hydrate cases around the gas. Gas bubbles may acquire a hydrate rind in the water column soon after emission although this process is not visualized here. This example also highlights the role of small scale variations in dissolved gas saturation in the bottom waters. Gas hydrate will form where flux is slow enough and the bottom water is saturated in the hydrate-former. Where dissolved gas concentrations are below the solubility limit, gas hydrate is expected to dissolve, and methane would more rapidly escape rising bubbles. Variations in the saturation of gas in pore waters within the hydrate stability zone could affect hydrate at small scales as well, with hydrate dissolving in some spots and forming in others. The pore water variations will tend to be homogenized in areas of high advection and persist longer in diffusion-dominated systems.

**Movie S3.** Inferred processes associated with gas hydrate dynamics in a deepwater seep field. The video is connected to the photograph in Figure 12a of the main text. The video was filmed on the seafloor on the Virginia margin at ~1500 m water depth by D2 at ~20:19 UTC on July 11, 2019, during *Okeanos Explorer* expedition EX1903 (Skarke et al., 2019). The full-resolution video can be downloaded from Cantwell et al. (2019). The video commences with a wide view of the low seafloor mound located within a previously unexplored area within the large Norfolk seep field described by (Skarke et al., 2014). Mounds like these are thought to be cored with gas hydrate. In some locations, seafloor hydrate mounds are described as hydrate pingos or pingo-like features to distinguish them from pingos cored by ice (Chapman et al., 2004; Waage et al., 2019). The sediment clouds in the bottom waters in the first half of the video are due to entrainment of sediment flowing out of the mound at small vents. The video focuses in on one such conduit. Dense fluid (presumably brine) mixed with coarse grained sediment flows downhill once emitted. The fine sediment portion ejected at the vent is entrained in the water column and transported by currents. Gas hydrate forming within the mound due to gas being fed from below excludes salts that then accumulate in pore waters, producing a briny fluid.

### **Disclaimer**

Any use of trade, firm or product name is for descriptive purposes only and does not imply endorsement by the U.S. Government.

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