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Key Points:

- We use machine learning to produce several inputs to deterministic models that estimate subsurface carbon degradation and methane generation.
- We estimate 0.8–2.2 and 1.1–3.0 × 10^6 Pg of carbon and methane, respectively, generated for subsurface sediments.
- Results yield global geospatial estimates of the maximum amount of methane possibly generated in situ and sequestered in marine sediments.

Supporting Information:
Supporting Information may be found in the online version of this article.

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Global Estimates of Biogenic Methane Production in Marine Sediments Using Machine Learning and Deterministic Modeling

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Abstract We have developed a model of geospatially estimating carbon accumulation and methanogenesis in seabed sediments that uses more accurate and sophisticated inputs to models than used in previous estimates. Using this hybrid stochastic and deterministic model, we estimate the maximum carbon available for methanogenesis in the global seabed, and subsequent microbial methane generated as a function of location and depth (including the gas hydrate stability zone). Global integration over present and previously microbially reactive sediments column yields total carbon and methane to be ∼0.8–2.2 × 10^6 and 1.1–3.0 × 10^6 Pg C and CH_4, respectively. Our improvements to accuracy include using geospatially machine learned estimates of seafloor inputs to which the methanogenesis modeling is most sensitive (e.g., total organic carbon, heat flux, porosity). Our improvements to model sophistication include geospatially dependent modeling (on a 5 × 5 arc-minute grid), a new model of sediment compaction (allowing for non-linear geothermal gradients), and variable age versus depth at each grid cell. A carbon reservoir of the magnitude we estimate here is consistent with the recent IPCC suggestion that long-term carbon sinks could explain imbalances in reduction of atmospheric CO2 over the last 50 million years. Our technique provides a foundation of using globally updateable machine learning parameters as the input to geologic and geochemical models, allowing for new observations to update global budgets of carbon available for methane, and subsequent total estimates of seabed methane.

1. Introduction

Organic carbon burial and degradation in marine sediments is an important factor in the regulation of Earth’s climate on geologic time scales (LaRowe et al., 2020) and has been suggested as a possible carbon sink, which can explain observed decreases in atmospheric CO2 over the past 50 million years (IPCC, 2021). Therefore, quantifying the amount and spatial distribution of organic carbon sequestered in marine sediments is fundamental to understanding global carbon cycling associated with climate variability via source and sink carbon interactions with the biosphere. Organic carbon is initially deposited on the seafloor through the outflow of terrestrial sources (i.e., rivers) and/or sinking of dead and decaying organisms (e.g., marine snow). Over time, this pool of shallow organic carbon deposited at the seafloor is either oxidized or buried where it can undergo a series of microbially driven redox reactions ultimately rendering methane (CH_4) (Middelburg, 2019). The majority of methane found in the subsurface is hypothesized to be of microbial origin (Kvenvolden, 1995), but abiotic thermogenic reactions also occur (Etiop & Sherwood Lollar, 2013).

Methane produced in the subsurface is commonly incorporated into one of the largest estimated free carbon pools on Earth, methane hydrate (Ruppel & Kessler, 2017). Methane at a concentration level that exceeds local solubility and occurs within a specific pressure-temperature regime, referred to herein as the hydrate stability zone (HSZ), may form methane hydrates. The carbon sequestered in this ice-like substance is estimated to comprise ~15%–50% of all global free carbon (Ruppel & Kessler, 2017). The HSZ generally occurs in water depths >300 m below sea level (mbsl), where pressures are high (~3–30 MPa) and seafloor temperatures are low (<25°C) (Max et al., 2006; Ruppel & Waite, 2020). The thickness of the HSZ is controlled by the temperature, pressure, and salinity gradients in the subseafoil. Direct, global subseafoil observations of the HSZ are sparse, therefore the identification of the HSZ is first-order dependent upon accurate estimates of input parameters such as pressure, temperature, and geothermal gradient.
Quantifying the amount of methane hydrate globally requires estimates of where and how much methane is generated in the sub-seafloor. Global estimates of organic carbon degradation and subsequent methane generated in the subsurface are based upon a variety of different models with varying degrees of complexity (Arndt et al., 2013; Freitas et al., 2021); however, all require some knowledge of environmental parameters as model input conditions. Direct measurements of geologic parameters are geospatially sparse, and coincident measurements of all required input model parameters (e.g., total organic carbon (TOC), porosity, temperature) are extremely rare. Accordingly, comprehensive assessment of seafloor properties and/or the resultant degradation potential, have always required some form of spatial interpolation. Input for previous efforts to model global organic carbon degradation and methane hydrate accumulation rely on using a globally assigned value (e.g., TOC = 1 percent-dry weight) or geospatial interpolation where direct observation does not exist (Piñero et al., 2013; Wallmann et al., 2012). Most of these efforts consider only geospatial proximity between observations. Recent advances in geospatial machine learning (GML) have improved upon spatial interpolation approaches, by allowing for statistically robust estimates of seafloor properties and quantification of associated uncertainty (Lee et al., 2019; Martin et al., 2015). In general, GML algorithms search for correlations in observed data (i.e., predictand) that may not be obvious and/or are simply too time-consuming to discern for an expert interpreter. These correlations between observed data and other environmental parameters (predictors) can then be used to estimate a value where no observation has been made. As long as sufficient predictors are available (comprehensive estimates of complementary geologic properties, e.g., bathymetry, see techniques in Lee et al., 2019), machine learning techniques can make statistically robust and reproducible predictions, with quantifiable uncertainties.

In contrast to previous approaches to estimate a global carbon and methane inventory, this work demonstrates the use of GML algorithms to constrain individual seafloor parameters as input into empirical and deterministic models. From deterministic models, at each 5-arc min location on the global seafloor, we identify the quantity of carbon and methane generated over sediments which are or have been biologically reactive (<100 degrees Celsius) including from within the HSZ. Although thermogenic CH4, may be incorporated into carbon and methane pools in the subsurface, this source of methane is not considered in this analysis since it is generally generated deep within the subsurface, and only exists in the shallow seabed due to fluid flow along faults and conduits (National Research Council, 2004). From our vertically distributed estimates, we integrate the amount of carbon and methane generated in situ for all marine sediments. We estimate only geospatial, not temporal, variability in our model results and present the 15.87 and 84.13 percentile from ensembles of model runs, where GML model inputs (Supplemental Table S1 in Supporting Information S1) are varied according to the input uncertainties. These machine learning estimates are easily updateable and when integrated in this model provide first-order constraints on how carbon and methane is sequestered in the marine environment. This modular approach can be used as a foundation that can be utilized with more sophisticated organic carbon degradation models, resulting in updated estimates of carbon and methane in the marine subsurface.

2. Materials and Methods

Our analysis is partitioned into three sections, each using previously published, fundamental, empirical models applicable on a global scale: (a) normal compaction of sediment (b) organic carbon degradation and methane accumulation, and (c) determining gas hydrate and free gas accumulation within the subseafloor. Relevant inputs and outputs for each of these sections, as discussed in the following sections, are shown schematically in Figure 1.

The input for each of these models use seafloor values predicted from a k-nearest neighbor regression (KNR) machine learning algorithm unless otherwise stated (Supplemental Table S1 in Supporting Information S1). We geospatially predicted each seafloor cell value globally at a 5 × 5 arc min spatial resolution and deterministically model at depths represented by a power law of 1.2 m below seafloor (mbsf) (e.g., 1.2² mbsf, 1.2³ mbsf, etc.). Each column of vertical cells is modeled independently for each geospatial cell. This depth discretization allows the upper sections of the subsurface (<1,000 mbsf) to be modeled at appropriately high resolution (small depth increments) while reducing computation time for deeper seated sediments.

In the analyses presented herein, two important general conditions are placed on our model. First, we do not consider the effects of fluid flux in the transport of dissolved methane, biological mediated processes such as anaerobic oxidation of methane, or of gas phase transport along conduits (e.g., seafloor seepage). Therefore, our
estimates for carbon in all seafloor sediments represent an upper limit (i.e., formed in place considering no flux). Lower estimates on all biologically generated methane represent a potential methane inventory, but not the lower limit of the true methane inventory, as further reduction in methane can occur via fluid flux and anaerobic oxidation. Estimates for methane in the HSZ represent in situ generation only since only methane generated within the HSZ will be sequestered there. If any fluid flux existed additional methane migrating near the seafloor would be trapped in the HSZ and would add to the amount we estimate here. Second, only geospatial, not temporal, variability of model inputs is considered. Although uncertainty estimates are available from our estimates of depth versus age (Lee et al., 2020) we do not use them in our ensemble process. There are many components of this model which can be varied in an ensemble analysis, however, this significantly increases the degrees of freedom adding detailed complexity resulting in difficulty interpreting our results.

Figure 2, discussed throughout the methods, shows several key sediment property profiles used in our model at the cell center nearest to National Gas Hydrate Program (NGHP) Site 118. This site location is indicated by approximate location of NGHP Site 119 on Figure S2b in Supporting Information S1.

2.1. Compaction Model

To model porosity versus depth, we used the Martin and Wood (2017) mechanical compaction model where the primary mechanism for porosity reduction is vertical effective stress resulting in grain rearrangement/rotation. Profiles from Figure 2c are relevant to this section.

2.1.1. Compaction Model Details

Surface porosity used as input to the compaction model \( (\phi_o; \text{Equation 1a}; \text{Martin \\& Wood, 2017}) \) was predicted using a KNR machine learning algorithm at a resolution of 5 x 5-arc min using observed data from Martin
In this KNR machine learning algorithm predictor (environmental) grids at observed data locations are correlated and ranked according to a Pearson's correlation coefficient. A uniform random noise grid is correlated and is also ranked and is used to determine a data-driven threshold of how many predictors to select. We use all predictors with a higher correlation coefficient than that of the uniform random noise grid. Further, the number of neighbors over which we calculate the weighted average value is selected as greater than one but less than 15 to avoid over and underfitting, respectively. The number of $k$ and predictors used for each grid predicted using GML is defined in Table S1 in Supporting Information S1. Our implementation of a machine learning KNR prediction and uncertainty estimation are discussed in further detail at Lee et al. (2019).

This compaction model, like many others in literature (e.g., Revil et al., 2002), requires estimates of the fraction of clay within the sediment. We do not have a predicted grid of global fraction clay, so we use our GML predicted seafloor porosity to estimate fraction clay particles. For each location on the global grid, we define end member porosity and fraction clay or sand from values outlined in Martin and Wood (2017). For example, given a sediment column containing 100% clay, the depositional (highest) and residual (lowest) porosities are 0.85 and 0.05, respectively. Conversely, for a sediment column containing 100% sand, we use depositional and residual porosities to be 0.39 and 0.22, respectively. We then solve for the fraction clay at each location by the rearrangement of Equation 1a found within Martin and Wood (2017) using our GML predicted porosity value. Using this fraction of clay, we solve for the residual porosity (Equation 1b).

Figure 2. Predicted profiles for the National Gas Hydrate Program Site 118 located in the Indian Ocean (Collett et al., 2014). The approximate location of this profile is indicated by Site 119 on Figure S2b in Supporting Information S1. The black dashed line across all profiles indicates the predicted base of hydrate stability. The green line indicates base run in the model. The dashed yellow lines indicate results from ensemble model runs. Profiles a-j show depth (in meters below seafloor) versus. (a) temperature in degrees Celsius; (b) pore pressure in megapascals; (c) porosity as a fraction; (d) age in million years; (e) labile portion of the total organic carbon in percent dry weight; (f) methane solubility in kilograms per cubic meter of seawater; (g) total methane generated per cubic meter of sediment; (h) methane in solution per cubic meter of water; (i) methane gas per cubic meter of sediment; (j) methane hydrate per cubic meter of sediment. Profiles g-h are per depth layer discretization and based on total in-situ methane generation.
\[ \phi_0 = f_{cly} \phi_{cly} + (1 - f_{cly}) \phi_{snd} \]  

(1a)

\[ \phi_r = f_{cly} \phi_{cly} + (1 - f_{cly}) \phi_{rsnd} \]  

(1b)

In Equations 1a and 1b, \( \phi_0 \) and \( \phi_r \) is depositional and residual porosity, respectively; \( f_{cly} \) is fraction clay. The terms \( \phi_{cly} \) and \( \phi_{cly} \) are depositional and residual porosities assuming 100% clay, respectively. The terms \( \phi_{snd} \) and \( \phi_{rsnd} \) represent depositional and residual porosities assuming 100% sand, respectively.

### 2.1.2. Simplifying Assumptions

This model is only accurate for the compression of unconsolidated sediment (i.e., normal compaction), and does not account for porosity reduction as a result of cementation or precipitation of minerals in the subseafloor, nor crushing or melting of grains. The biochemical production of methane and sequestration in the HSZ occurs at relatively low temperatures (<70°C) and shallow subseafloor depths (<2,000 mbsf) where mineral precipitation typically does not account for the significant porosity reduction (Bjørlykke, 1999). Additionally, we assume fraction clay is constant with depth when ideally, a variable fraction clay with depth would provide more accurate results within the model (particularly temperature, discussed in Section 2.2.2), however to date there exists no global fraction clay estimates with depth.

### 2.2. Organic Carbon and Methane Calculation

We follow an empirical function from Malinverno and Martinez (2015) which utilizes the Arrhenius equation (Equation 2) to determine the rate of organic carbon degradation, assumed to be primarily a function of temperature. Profiles from Figures 2a, 2d, 2e are relevant to this section.

#### 2.2.1. Organic Carbon Calculation Details

Seafloor TOC and uncertainty at a resolution of 5 × 5-arc min are GML results from Lee et al. (2019). This value serves as the initial seafloor organic carbon value used in a simple estimation of organic carbon degradation with depth in the subsurface. The Arrhenius equation (Equation 2) is used to determine this rate.

\[ k(t) = A e^{-\frac{E_a}{RT}} \]  

(2)

In Equation 2, \( k(t) \) is the organic carbon degradation reaction rate with respect to time; \( E_a \) represents the activation energy estimated to be 110 kJ/mole from Price and Sowers (2004). The term \( A \) is a constant derived from Malinverno and Martinez (2015) estimated to be \( 3 \times 10^{18} \) per million years. \( T \) is the temperature at a given depth and \( R \) is the universal gas constant. Two other input parameters besides TOC at the seafloor are needed for this degradation model, age of the sediment and temperature.

The age versus depth profile used for NGHP 118 is shown in Figure 2d. The simplest assumption to determine the age of sediment for a given depth in the subseafloor is a linear depth versus age model derived from the total sediment thickness and crust age estimate. A recent publication by Lee et al. (2020) allows for some variability in this linear depth versus age model by using isochore estimates for periods from present to Middle Miocene (15.97 million years; Ma). We integrate these isochore estimates to yield depth to age values for locations on a 5 × 5 arc-minute grid. For Figure 2d, NGHP Site 118, the incorporation of depth to age estimates can be seen as deviations from a linear depth versus age profile seen for depths 0 to ~100 mbsf. For profiles where the crust age estimate is greater than the isochore estimates (15.97 Ma; e.g., Figure 2d), we use a linear depth versus age profile using the total sediment thickness grid from Straume et al. (2019) and crust age estimate from Muller et al. (2008). The crust age estimate is originally at 6-min resolution and was bilinearly interpolated to 5 × 5 min. The linear depth versus age curve was used for depths greater than ~100 mbsf on NGHP Site 118 in Figure 2d.

The temperature in the subsurface is initialized by the predicted temperature at the seafloor. For each location on the globe, temperature at the seafloor was predicted using data from the World Ocean Atlas (Boyer et al., 2013). The initial data was derived from an average of in situ temperature within the water column over each a 1 × 1 arc-degree cell. To achieve higher spatial sampling of 5 × 5 arc-minute and extension to higher latitudes, a KNR machine learning algorithm was used to predict at native 1 × 1-arc degree resolution. Only predictors with complete global coverage were used in this process. Bilinear interpolation was then used to up sample to 5 × 5-arc minute resolution. Parameters, such as number of neighbors and predictors are shown in Supplemental Table S1.
in Supporting Information S1. Temperature within the subsurface was determined by calculating a geothermal gradient derived from predicted heat flow at the seafloor using observations primarily from the Global Heat Flow Compilation Group (2013). We assimilated several new observations from Hornbach et al. (2020) and Riedel and Collett (2017). Using these observations, we predicted heat flow and uncertainty at the seafloor using a KNR machine learning algorithm following the workflow outlined in Lee et al. (2019). We then derive the geothermal gradient for each depth (dT/dZ; Equation 3a) using the predicted seafloor heat flow (Q), and a calculated thermal conductivity (c) of the sediment (Equation 3b), which is based on porosity and fraction clay clay components obtained from Section 2.1.1.

The thermal conductivity of the pores considers only seawater as pore fill and is represented by 0.596 W/m/K, while the thermal conductivity of the grain component is calculated from the simple geometric mean using clay (smectite = 1.88 W/m/K) and sand (quartz = 7.69 W/m/K) (Goto & Matsubayashi, 2009). An example of temperature versus depth in sediment is shown in Figure 2a for the location nearest to NGHP Site 118.

\[ \frac{dT}{dZ} = \frac{Q}{c_{sed}} \quad (3a) \]

\[ c_{sed} = c_{pore} \times c_{grain}^{1-\phi} \quad (3b) \]

Once temperature and age within the subsurface are determined, we calculate the rate of organic carbon degradation using Equation 2. This rate is implemented within Equation 4 from Malinverno and Martinez (2015) to determine the remaining TOC concentration with time; \( G(t) \). In this equation, we assign the percentage of labile organic carbon available for microbially mediated methanogenesis as 28.6% of initial organic carbon deposited at the seafloor \( (G_0) \); Bradley et al., 2020). TOC versus depth is shown in Figure 2e for the location nearest to NGHP Site 118. The initial labile organic carbon value used for Site NGHP Site 118 in Figure 2e is indicated at 0 mbsf.

\[ G(t) = G_0 e^{\int_0^t k(t') dt'} \quad (4) \]

TOC concentrations calculated from Equation 4, where \( G(t) \), represents the amount of labile carbon remaining after microbial organic carbon degradation at geologic time, t, corresponding to a particular depth in the profile. To determine the amount of carbon that has been converted to methane at this geologic time and depth (i.e., carbon which has undergone the terminal redox reaction), we subtract the TOC remaining (at depth; \( G(t) \)) from the initial amount of TOC deposited (seafloor TOC; \( G_0) \). Equation 5 is used to determine the amount of organic carbon available for conversion to methane in percent dry weight.

Organic carbon concentrations with depth are expressed as a percentage of the dry-weight sediment, therefore we must convert the percent of organic carbon to mass. First, for a 5 x 5-min grid cell we multiply the length by width of the cell by discretization depth (i.e., voxel) to calculate the total volume of a voxel. For each voxel, the average porosity (e.g., Figure 2c) and total volume are used to determine the volume of pore space and solids. The solid volume (i.e., grain volume) is then converted to solid mass (Equation 6a) using the grain density, calculated from the weighted average of clay and sand components (Section 2.1.1) where clay and sand densities are 2.6 and 2.625 g/cm^3 respectively.

\[ OC_{\text{avail for CH}_4} = G_0 - G(t) \quad (5) \]

\[ \text{Mass}_{\text{solid}} = \text{Vol}_{\text{solid}} \times \left( (f_{cly} \times 2.6) + (f_{sand} \times 2.625) \right) \quad (6a) \]

The mass of organic carbon in methane (Equation 6b; \( \text{Mass}_{\text{OC}} \)) is the product of multiplying the mass of the solid component (Equation 6a; \( \text{Mass}_{\text{solid}} \)) by organic carbon available for methane in percent dry weight (Equation 5; \( OC_{\text{avail for CH}_4} \)), with respect to depth. This mass of organic carbon is converted to moles of carbon using the molar mass of carbon. As there is only one mol of carbon per methane molecule there is no conversion needed from moles of carbon to methane. The moles of carbon can be converted to grams of methane using methane's molar mass value. This final quantity of methane represents the amount of methane generated per voxel of sediment and the total amount of methane available for uptake in the aqueous, gas, and solid (i.e., hydrate) phases.

\[ \text{Mass}_{\text{OC}} = \text{Mass}_{\text{solid}} \times OC_{\text{avail for CH}_4} \quad (6b) \]
For the location nearest to NGHP Site 118, Figure 2g shows CH₄ generated in kg of CH₄ per cubic meter of sediment; here total mass methane generated for each sediment voxel is divided by the sediment voxel volume. Integration over the entire sediment column represents the amount of organic carbon degraded and subsequently converted to methane through geologic time.

### 2.2.2. Simplifying Assumptions

The focus of this paper is geospatial variability and not temporal and/or depth variability. Practical global modeling of marine organic carbon requires several simplifications;

1. The amount of organic carbon deposited at present-day is the initial amount deposited through all of geologic time. In reality, this initially deposited organic carbon is variable with age and geologic process (e.g., sea level changes particularly deglaciation; Cartapanis et al., 2016). However, at present, we have no constraint on initially deposited (i.e., pre-degradation) global organic carbon concentrations through geologic time. To date, only sparse estimates of organic carbon with age and/or depth exist. It is not clear how much a variable initial organic carbon concentration with depth would impact the results from this analysis.

2. We assume a single kinetic law where degradation of organic carbon occurs with as a function of a single rate constant. By aggregating these processes into one kinetic rate law and constant deposition of organic carbon in geologic time, we are broadly simplifying the organic carbon degradation process, and thus are ignoring preferential degradation of organic materials which has been highlighted to be an important aspect in the modeling of organic carbon degradation (Freitas et al., 2021). Specifically, some of the different parameters we are grouping into one rate which control organic matter reactivity would include source and composition, oxygen exposure time, terminal electron acceptor availability, microbial activity, sediment biological mixing, transport history, organic matter iron mineral adsorption, priming, and hydrogen sulfide exposure time (Freitas et al., 2021). Clearly, our assumption of a single global carbon pool is a simpler model than used by others. It is not clear if our methods result in an over or underestimated integration of organic carbon generated in geologic time.

Although, it is beyond the scope of this work to test other biodegradation models, it is possible that through our variability (i.e., ensemble) analysis, uncertainties in global input grid values (e.g., seafloor organic carbon concentrations, temperature) already encompass the effects that would be seen if we had used a model of different reactivities among carbon pools.

We assume a biological temperature limit of marine sediments, environment dependent, is ~100–120 degrees Celsius (Lagostina et al., 2021), at which point all organic carbon has been degraded or consumed and thus all methane generated from degradation for a given parcel of sediment is available for uptake in free gas, methane hydrate, or solution phases. Further, since we use a geometric mean to calculate sediment thermal conductivity and clay minerals have a lower thermal conductivity than other sediment components considered within this analysis (i.e., sand), the assumptions made in Section 2.1.1 on determining fraction clay minerals is likely to impact the temperature profile of the sediment. Specifically, a higher clay content would result overestimating the geothermal gradient and temperature with depth.

### 2.3. Estimating Base of Hydrate Stability and Solubility

To estimate the base of methane hydrate stability, we calculate the temperature of hydrate stability and solubility, using empirical equations from Dickens and Quinby-Hunt (1994), Davie et al. (2004), and Duan et al. (1992). Profiles from Figures 2a–2j are relevant to this section.

#### 2.3.1. Base of Hydrate Stability and Solubility Details

The base of HSZ is fundamentally a function of salinity, temperature, and pressure in the seafloor. Initial seafloor salinity and seawater density is from the 013 World Ocean Atlas (Boyer et al., 2013) and estimated by the same workflow as initial seafloor temperature discussed in Section 2.2.1. Hydrostatic pressure at the seafloor is calculated using atmospheric pressure, gravity respective of latitude, and seawater density. Pressure with depth in the seafloor is determined by the gravity respective of latitude, water depth, the thickness of a sediment voxel, and density of the sediment or seawater for lithostatic or hydrostatic, respectively. Elevation (i.e., water depth; bathymetry) grid (Tozer et al., 2019) was downsamples from SRTM15 + V2 at 15 arc-second to 5 × 5 arc-minute
resolution by the block median. An example pressure profile in megapascals (MPa) is shown in Figure 2b for NGHP Site 118.

For each sediment layer, given a pressure and salinity, the temperature at which pure methane hydrate is stable in seawater is determined using Equation 7 from Dickens and Quinby-Hunt (1994) where \( z \) is the depth in kilometers. If the temperature (°C) for a given voxel is less than or greater than this threshold temperature (\( T_{\text{hyd stable}} \)), hydrate is considered stable (i.e., in the HSZ) or not stable (i.e., outside of HSZ) respectively. For each 5 × 5 grid cell, the base of the HSZ is the final depth that hydrate is stable. The predicted base of the HSZ for NGHP Site 118 is shown in Figure 2 by the black dashed horizontal line.

\[
T_{\text{hyd stable}} = 11.726 + 20.5 \times \log_{10} z - 2.2(\log_{10} z)^2
\]  

To estimate how much gas hydrate and/or gas is present within or outside of the HSZ respectively, we calculate methane solubility. In this two-phase system, methane hydrate and/or free gas can only exist from methane concentrations in excess of methane solubility for a given voxel. Methane in excess of solubility is either free methane gas (outside of HSZ) or methane hydrate (within HSZ). The methane solubility is calculated within and outside of the HSZ using equations from Davie et al. (2004) and Duan et al. (1992) respectively. The methane solubility for NGHP Site 118 is shown in Figure 2f. We determined volume of pore fluid in liters (only water is considered) using the porosity with depth (\( \phi_z \)) estimate (Equation 8a). We multiply the calculated methane solubility in moles per liter to obtain the maximum capacity of methane in solution (Equation 8b).

\[
\text{Vol}_\text{pore} = \text{Vol}_\text{total} \times \phi_z 
\]

\[
\text{CH}_4\text{maxcap}_\text{solution} = \text{CH}_4\text{solubility} \times \text{Vol}_\text{pore}
\]  

Moles per liter is converted to kilograms per cubic meter for plotting purposes in Figure 2f. The amount of methane in solution is shown in Figure 2h for NGHP 118; Methane in solution never exceeds methane solubility (i.e., \( \text{CH}_4 \) Soluble) estimates. To determine the mass of methane hydrate or methane gas, the moles of methane in excess of solubility are converted to mass of free methane gas or methane hydrate assuming one mol of methane is approximately 16 g/mol free methane gas or 122.3 g of methane hydrate per moles \( \text{CH}_4 \) (Haeckel et al., 2004). For NGHP Site 118, shown in Figures 2i–2j, respectively, the total kilograms of methane gas or methane hydrate generated per sediment voxel was divided by the volume of the respective sediment voxel. For each grid cell at a 5 × 5-arc min resolution, the mass of methane hydrate and free methane gas was integrated over the column to achieve total amount sequestered within the sediment.

### 2.3.2. Simplifying Assumptions

Several simplifying assumptions made in this portion of the model include details regarding salinity, pressures, and maximum depth constraints on the base of hydrate stability.

In this model, similar to fraction clay and TOC, salinity is held constant with depth in the subsurface. Further, we assume, as some literature also suggests, that the pressures exhibited in sedimentary systems are not entirely hydrostatic but represent some small overpressures (Green & Edwards, 2018; Mann & Mackenzie, 1990). Therefore, we add 10% of the difference between lithostatic pressure and hydrostatic pressure to determine the pore pressure in the sediments. This is expressed below in Equation 9.

\[
P_{\text{pore}} = P_{\text{hydrostatic}} + (0.1 \times (P_{\text{lithostatic}} - P_{\text{hydrostatic}}))
\]  

Finally, the theoretical limit of the HSZ may extend beyond the sediment thickness estimate, but for our purposes, the base of the HSZ is limited to the sediment thickness.

### 2.4. Ensemble Range of Estimates

To estimate variability in our results, we perform 20 ensemble model runs where each member of an ensemble is shown in Supplemental Table S1 in Supporting Information S1. The variability in our global integrations is determined from the 15.87 and 84.13 percentiles, or ± one standard deviation of the 20 ensembles. We intentionally use percentiles rather than standard deviation as the latter term is specific to a symmetric (e.g., Gaussian) distribution and it is not clear our resulting distribution is symmetric. The range calculated within these percentiles.
represents the range of likely values of global integrations. 20 ensembles were used for an appropriate balance of accuracy and computation time. An example of 20 ensembles is displayed by dashed yellow lines for the location nearest to NGHP Site 118 in Figure 2.

For each ensemble run, at each location we determine a truncated normal distribution with the mean being the value at the grid cell and the standard deviation being the uncertainty estimate listed from Table S1 in Supporting Information S1. For each parameter and ensemble run, we sample within ± 1 standard deviation of a truncated normal distribution. We truncate our distribution, when necessary, to ensure that values occur within physically realistic ranges (e.g., porosity and salinity cannot be negative). Parameters which are used as input to the model, with their uncertainty estimation type and respective sources are shown in the Supporting Information within Table S1 in Supporting Information S1.

3. Results and Discussion

3.1. Global Integrations

For each grid cell on Earth, we integrate the mass of carbon and methane for all depths. As a whole this integrated carbon pool represents the quantity of carbon available for three phases of methane in the subseafloor - solid (i.e., sequestered within methane hydrate), gas, or in solution. Table 1 shows our predicted integrated global amounts of carbon sequestered in sediments and total amount of generated methane. Figure 3 displays our estimate and range of variability (Figures 3b and 3c; n = 20 ensembles) in the mass of carbon sequestered in methane per square kilometer of sediment at a single location. Figure 3d displays the total mass and variability (e-f; n = 20 ensembles) of methane generated per square kilometer of sediment. Integrated total carbon and methane in all global seafloor sediments is ∼1.6 and 2.2 × 10⁶ Pg C and CH₄ respectively. The integration of the 15.87 and 84.13 percentiles in the ensembles yields 0.8–2.2 and 1.1–3.0 × 10⁶ Pg C and CH₄ respectively.

The predicted base of HSZ is needed to partition our methane estimates into the solid, gas, and aqueous phases. Figure 4 displays our estimate in the base of the HSZ (a) and the 15.87 and 84.13 percentiles respectively (b-c) in 20 ensembles. For each 5 × 5-min grid cell, we integrated the solid (i.e., methane hydrate) and free gas phases over all depths shown in Figures 4a and 4d. The 15.87 and 84.13 percentiles in 20 ensembles for free methane gas and methane hydrate are shown in Figure 5 (b-c; e-f), respectively.

3.2. Qualitative and Quantitative Global Evaluation

There exist few, if any, estimates in the literature of carbon and methane sequestered in all seafloor sediments. Further, for all global quantities (e.g., carbon in methane, methane, hydrate, gas, base of HSZ), the majority of estimates in literature do not provide a geospatial uncertainty or variability in results. These values and ranges do not represent fluid flow and only represent the amount of methane generated in situ and are thus intended as first order constrains on methane generation through geologic time. Here, we compare and discuss similar analyses and results as compared this analysis.

A recent paper by Atwood et al. (2020) integrates carbon quantities over global marine sediments for the upper 1 m of the seafloor. Atwood et al. (2020) results do not account for any organic carbon degradation, and therefore yields a value of ~2,322 Pg C. Further, Lee et al. (2019) also does not consider organic carbon degradation and yields 87 Pg C integrated over the upper 5 cm. Our carbon integration estimates represent the amount of carbon in methane and are therefore orders of magnitude smaller (~0.43 Pg C) since microbial degradation of organic carbon in the upper meter of the sediment column can be significant (Arndt et al., 2013).

IPCC (2013) reports an estimate of 1.750 Pg C in shallow sub-seafloor sediments which includes both organic (150 Pg C in mixed layer) and inorganic carbon (1,600 Pg C) (Emerson & Hedges, 1988). The mixed layer is represented by Emerson and Hedges (1988) is ∼15 cm on average (10 cm open ocean, 20 cm continental margin). Our estimate is for the entirety of the sediment column. Additionally, our estimate has spatially variable porosity and sediment density whereas Emerson & Hedges, 1988 use an average organic carbon content, density and
Our estimates of organic carbon in the upper 1 m are already two orders of magnitude less than Emerson and Hedges (1988) which integrates over an average of 15 cm. The primary difference is likely attributable to differing degradation rates. In an effort to compare Emerson and Hedges (1988) estimates with those presented in this analysis, we do some simple calculations.

Figure 3. (a) Cumulative predicted organic carbon in methane in Teragrams (1 Tg = 1 × 10^{12} g) sequestered per square kilometer of sediment. (b)-(c) Respectively, the 15.87 and 84.13 percentiles in 20 ensembles of the cumulative predicted organic carbon mass available for methane conversion in Teragrams per square kilometer of sediment (d) Cumulative methane generated in Teragrams (1 Tg = 1 × 10^{12} g) sequestered per square kilometer of sediment (e)-(f) Respectively, the 15.87 and 84.13 percentiles in 20 ensembles of the cumulative mass of methane generated in Teragrams per square kilometer of sediment.
First, we assume the global mixed layer has a depth of 15 cm (10 cm in open ocean; 20 cm on continental margins). Assuming the organic carbon concentrations used for this paper (Lee et al., 2019, p. 87 Pg C over upper 5 cm) yields approximate 261 Pg C over the upper 15 cm (i.e., average mixed layer) versus 150 Pg C (Emerson & Hedges, 1988). Neither of these numbers consider degradation rates, and Lee et al. (2019) is approximate double that of the Emerson and Hedges (1988) estimate. To account for the amount of degraded organic carbon using Lee et al. (2019) values, we must determine an average fraction degradable from Emerson and Hedges (1988; ∼0.3–0.5–continental margin; 0.2–0.4–open ocean). Using 261 Pg C estimate of organic carbon within the average mixed layer (upper 15 cm) yields an estimated ∼91 Pg C of carbon degradable. From the Emerson and Hedges (1988) estimate (150 Pg C) we then calculate 53 Pg C degradable organic carbon for the mixed layer. Finally, to calculate the remaining organic carbon (which would be post degradation and available for incorporation into methane) we remove the degradable portions from the total portions of organic carbon for the mixed layer. This yields 97 Pg C and 170 Pg C for the Emerson and Hedges (1988) and Lee et al. (2019) estimate, respectively. Using this estimation of degradation, our estimates of 170 Pg carbon available in the methane phase are not vastly different from those of Emerson and Hedges (1988). Sources of discrepancies between this study and that of Emerson and Hedges (1988) may be numerous, because the modeling approach was significantly different. Some differences include this study's use of an exponential rate law which considers sediment age and temperature as opposed to a constant decay. Additionally, Emerson and Hedges (1988) assumed average sediment organic carbon concentrations, densities and porosities which is likely to cause large discrepancies among results.

Figure 4. (a) Predicted global base of methane hydrate stability zone (HSZ) in meters below seafloor. The maximum base of hydrate stability is limited by the sediment thickness. (b)-(c) The 15.87 and 84.13 percentiles in 20 ensembles of the predicted base of hydrate stability in meters below the seafloor. Tan values indicate masked values where there exists no predicted HSZ within the sediment column.
Qualitatively, the highest carbon and methane concentrations occur along the coastlines (Figures 2a and 2d). This is consistent with global estimates of seafloor TOC (Lee et al., 2019). Further, sediment thickness is generally thickest near coastlines (i.e., continental slope), as a result of terrestrial input resulting in more carbon and methane being integrated through geologic time (Emerson & Hedges, 1988). We find our results consistent with this by integrating total carbon over the continental slope (9.7 × 10^5 Pg C) and open oceans (6.6 × 10^5 Pg C) as defined by Paris et al. (2016) where the slope and open ocean are defined by <1211 and >1,211 mbsl.

Figure 5. (a) Cumulative mass of predicted total methane gas in gigagrams (1 Gg = 1 × 10^9 g) sequestered per square kilometer of sediment. (b)-(c) Respectively, the 15.87 and 84.13 percentiles in 20 ensembles of the cumulative mass of methane gas in gigatons per square kilometer of sediment. (d) Cumulative mass of total methane hydrate in gigatons (1 Gt = 1 × 10^9 g) sequestered per square kilometer of sediment. (e)-(f) Respectively, the 15.87 and 84.13 percentiles in 20 ensembles of the cumulative generated total mass of methane hydrate in gigagrams per square kilometer of sediment. Black indicates an explicit value of zero.
respectively. The highest variability in percentiles (in n = 20 ensembles) also exists in these regions with the highest carbon/methane accumulation.

We next consider gas hydrate. We use the predicted base of hydrate stability to approximate the global integration of free methane gas and methane hydrate. Given that free methane gas and methane hydrate accumulations are based on HSZ thickness, variability in the HSZ will cause variability in methane gas and hydrate estimates. Near the coastlines and on the continental shelves, the HSZ generally does not exist given lower pressures (i.e., water depths <300 m) which preclude the formation of hydrate (shown as tan in Figure 4). Areas that have the deepest base of the HSZ are along the continental slope, where temperatures and pressures (water depth >300 m) are appropriate for the stability of methane hydrate. Further, regions which have the broadest (i.e., distance perpendicular to the coastline) predicted base of HSZ occur on passive margins (e.g., U.S. East Coast, Brazil Margin, Western Africa, Eastern Australia). Intuitively this makes sense as these regions generally have lower geothermal gradients than active margins such as the U.S. West Coast (Cascadia) and Philippines. Our predicted results are consistent with previous estimates of the base of HSZ (Piñero et al., 2013; Wallmann et al., 2012). Several regions of the globe are similar in both our analysis and others within the literature, namely, offshore U.S. East coast, southeast Australia, and offshore Uruguay in South America. The base of HSZ is consistently deeper in these areas and exhibits values on the same (500–600 mbf) order of magnitude. Dissimilarly to similar to previous estimates, our results provide more spatial variability and are higher in resolution (5-min) than previous estimates (0.5 and 1-degree).

Similar to our carbon and methane estimates, most past estimates of the base of HSZ do not provide a geospatial estimate of uncertainty and/or variability. The percentile ranges in our 20 ensemble models (Figures 3b and 3c) shows the largest variability the HSZ thickness in regions with the deepest estimates of HSZ. However, if we compare the predicted results to the range represented via percentiles, the passive margins exhibit higher variability relative to the initially predicted depth to the base of the HSZ. For example, along the U.S. West Coast or Cascadia region, the predicted base of hydrate stability zone (BHSZ) is ~250 mbf. The variability (percentile) range in this region is approximately 200 mbf which is fractionally high. Conversely, offshore Uruguay the predicted BHSZ is in a small area is ~800 mbf where the variability in percentiles is fractionally lower, only ~400 mbf. The higher variability among the predicted base of HSZ ensembles is a result of larger uncertainties in the heat flow estimate, however more investigation is needed.

Finally, using our estimates of methane hydrate with depth and predicted base of the HSZ, we integrate only the mass of carbon occurring in methane hydrate for all sub-seafloor sediments. Figure 6 displays a modified bar graph from Piñero et al. (2013) showing our estimate in context of previous literature estimates. We calculate 1,469 Gt of C in methane hydrate globally. Although, all of these estimates consider different modeling parameters (e.g., carbon lability) and methods (e.g., fluid flow, organic carbon degradation) our value is generally consistent with previous estimates, particularly, the more recent estimates (Burwicz et al., 2011; Piñero et al., 2013; Wallmann et al., 2012). We calculate variability (or uncertainty) in our carbon in methane hydrate estimate using 15.87 and 84.13 percentiles of the 20 ensembles. Our estimated range, as indicated by error bars, of carbon sequestered inhydrate is 62–6,951 Gt. The tuning of model input parameters for each ensemble causes large variations in the estimated base of HSZ which results in large variability in the calculation of gas and hydrate values. Our estimated variability range is still within the broader range of carbon in hydrate estimates from Piñero et al. (2013).

3.3. Single Profile Analysis

To further quantitatively validate our estimated BHSZ, we compare our estimated HSZ thicknesses to observed drilling sites collated by Riedel and Collett (2017) shown in Table 2. Site locations are presented in their regional context within the Supplemental Information (Figures S1-S2 in Supporting Information S1). Site profiles (initial prediction in green, ensembles in yellow) are additionally available in the Figures S1 and S2 in Supporting Information S1. For many of these observed sites, a substantial amount of methane hydrate was found. However, qualitatively many of our predictions and subsequent ensembles yield little to no methane hydrate in the subsurface. While comparing our results/input to observed values (herein referred to as point observations), we note our inputs and results (herein referred to as cell averages) are representative of ~10 x 10-km areas, whereas point observations from Riedel and Collett (2017) are very discrete sampled points (scale of cm). Our cell averages represent areas and not necessarily discrete points; therefore, variability is inherent as our estimates actually
represent a 10 × 10 km area average. Geothermal gradient, water depth, and gas hydrate accumulation can be highly spatially variable (Malinverno & Goldberg, 2015) which can create discrepancies between point observations and our cell averages (Figures S1–S2 in Supporting Information S1; Table 2). Many of these drilling sites were specifically targeted to drill gas hydrate and are therefore biased toward finding the highest concentrations in the area. Further, for all sites, we find that the age of the sediment can significantly affect the rate of organic

Table 2
Comparison Between Data Collated From Observed (i.e., Point Observation) Drilling Sites From Riedel and Collett (2017) and Our Predicted (i.e., Cell Average) Input and Base of Hydrate Stability Zone at Associated Cell Centers

| Site id | Point observation Lat/ Lon (dd) | Offset of binned Lat/ Lon (km) | Water depth (point observation; meters) | Water depth (cell average; meters) | Thermal gradient (point observation; C/km) | Thermal gradient (cell average; C/km) | BGHSZ (point observation; mbsf) | BGHSZ (cell average; mbsf) | 15.87 and 84.13 percentiles in BGHSZ (ensemble n = 20) | Absolute error of BGHSZ (point observation–cell average; %) |
|---------|---------------------------------|-------------------------------|-----------------------------------------|-------------------------------------|------------------------------------------|---------------------------------------|-------------------------------|-------------------------------|--------------------------------------------------------------------------|--------------------------------------------------|
| DSDP 533 | 31.260, −74.869                 | 3.54                          | 3,191 ± 1                              | 3,187                               | 45.85                                    | 600 ± 5                               | 417.34                        | 357.1–521.9                  | 30.4                                                      |                                                  |
| IODP 311–1,325 | 48.811, −126.983                | 2.87                          | 2,201 ± 1                              | 1,584                               | 36                                        | 600 ± 5                               | 348.66                        | 315.3–480.2                  | 42.3                                                      |                                                  |
| NGHP 01–03 | 15.898, 81.899                  | 3.66                          | 1,076 ± 1                              | 961.5                               | 39 ± 2                                    | 200 ± 5                               | 248.59                        | 159–296.4                    | 24.3                                                      |                                                  |
| NGHP 01–17 | 10.753, 93.112                  | 4.49                          | 1,344 ± 1                              | 1,657.5                             | 19 ± 2                                    | 614 ± 6                               | 418.76                        | 346.1–504.7                  | 31.7                                                      |                                                  |
| NGHP 01–18 | 19.152, 85.772                  | 3.62                          | 1,374 ± 1                              | 1,510.5                             | 50 ± 2                                    | 215 ± 2.5                            | 263.73                        | 191.4–334.9                  | 22.6                                                      |                                                  |
| NGHP 01–19 | 18.977, 85.658                  | 4.14                          | 1,422 ± 1                              | 1,475                               | 53 ± 2                                    | 195 ± 7.5                             | 227.73                        | 181.3–275.7                  | 16.7                                                      |                                                  |
| ODP 164–994 | 31.785, −75.545                 | 0.77                          | 2,798 ± 1                              | 2,798                               | 36.4 ± 1.3                                | 428 ± 5                               | 316.72                        | 263.2–430.7                  | 26                                                      |                                                  |
| ODP 204–1,244 | 44.586, −125.119                | 4.32                          | 907 ± 1                                | 907                                 | 60 ± 1                                    | 125 ± 3                               | 119.78                        | 68.8–151.1                   | 4.2                                                      |                                                  |

Note. Point observation data shown are used only as validation to the prediction, and were not included in any of the machine learning or deterministic predictions. Cell averages represent areas (respective of latitude) as large as 1,000 km².

*Observed values come from Riedel & Collett, 2017. †Geothermal gradient value is extrapolated through BGHSZ. ‡Shown where applicable.
carbon degradation and subsequent generation of CH \(_4\) (e.g., distortion of labile TOC in Site DSDP 533). We proposed that future investigations should consider variable (i.e., not linear) depth versus age profiles to assess microbiologically produced methane accumulations in geologic time.

In all sites, except for three (DSDP 533, IODP 311, NGHP 117), the variability in the ensemble percentiles encompasses the point observation BHSZ. Our percentile range for the cell averages BHSZ is overestimated in IODP 311 and underestimated in sites DSDP 533 and NGHP 117. Both sites where the BHSZ is underestimated are on passive margins and the point observed BHSZ is relatively deep compared to other sites. This may indicate our estimates are biased to shallower BHSZ.

The absolute error between the cell averages and point observed base of HSZ varies for each site with the smallest and largest errors being site ODP 204 (5.22 mbsf) and NGHP 117 (195.04 mbsf). To investigate the difference between the point observations and cell averages of the BHSZ, we collate coincident point observations and cell average values including geothermal gradient and water depth. Additionally, we calculate the distance from point observations to binned latitude/longitude (i.e., offset of binned latitude/longitude) in Table 2. For sites with the largest discrepancies between cell averages and point observed base of HSZ (DSDP 533, IODP 311, and NGHP 117), the thermal gradient consistently differs by \(\sim 10^\circ\text{C/}\text{km}\). Conversely, in other sites where the thermal gradient differs by approximately the same amount (e.g., ODP 204) a relatively small residual between cell average versus point observation was calculated for the base of HSZ. Although this site, ODP 204, had the smallest error in the base of HSZ between cell average and point observation (4.2%), it also had one of the largest discrepancies between point observation and cell average of thermal gradient. Given both sites with the smallest (ODP 204) and largest (NGHP 117) errors have large discrepancies among the point observation and cell averaged thermal gradient, it is difficult to precisely say thermal gradient is the sole cause for mismatch between the observed and predicted BHSZ.

We investigate water depth has a relationship between larger water depth differences and increased absolute residual in the BHSZ. For example, sites NGHP 01–17 has a residual error of 31.7% and depth difference of 313.5 mbsf, followed by sites NGHP 01–18, NGHP 01–03, NGHP 01–19 with water depth differences and absolute residual of 136.5 mbsf (22.6%), 114.5 mbsf (24.3%), and 53 mbsf (16.7%), respectively. When the water depth point observation versus cell average difference is smaller than 20 mbsf there is no relationship in absolute residual (sites DSDP 533, ODP 164, ODP 204). Finally, we investigate residual versus offset of binned latitude/longitude and find no clear relationship between the two. In no sites, were all cell average values (offset distance, thermal gradient, water depth, and BHSZ) consistent with all point observation values. This makes it difficult to ascertain the source of discrepancy (i.e., residual) between predicted and observed base of HSZ values. Further, any number of the input grids or combination of input grids could have resulted in this discrepancy, not just the inputs discussed in Table 2.

### 3.4. Potential Areas of Model Improvement

Final estimates in the deterministic model are dependent on accurate input into the models. Machine learning provides a statistically robust parameters estimates where variables have previously not been observed. Each model input (e.g., predicted porosity) is dependent upon observed data and updating predictions will impact the final model results. As new observed data becomes available, assimilation of this data into a GML routine can result in improved input/predictions to the model. Any improvement in input parameters and uncertainty is likely to impact final estimates of carbon and methane. Because of the modular nature of our analysis, input parameter values are agnostic to the models that use them. Therefore, a different or more sophisticated sediment model can easily be coupled with machine learning predictions of seafloor values, as in Eymold et al. (2021) which consider vertical flux and distribution of methane.

Potential improvements to the methods presented here may include running ensembles where individual parameters are altered independently. This time-intensive and exhaustive analysis would help identify parameters that are most important to individual profile sites. Finally, estimates of depth versus age certainly impact results on methane generated and subsequently free gas and methane hydrate estimates. Our model incorporates some variability of age versus depth, however, only for present to middle Miocene. In future iterations, analysis may consider age versus depth data using sedimentation rates from recent sediments (e.g., Restrepo et al., 2020). Further, variable organic carbon deposition (as opposed to constant deposition through geologic time), and subsequently methane
generated in line with geologic time would provide insight and more accurate estimates of vertical methane distribution and budgets in the subsurface.

4. Conclusions

We have modeled, with quantitative uncertainties, global marine carbon and methanogenesis geospatially and with depth, using a hybrid of machine learning and deterministic techniques. This hybrid model quantitatively tracks organic carbon as it is deposited at the seafloor, is buried, heated, and is degraded through microbial processes to produce methane. The 5 × 5 arc min resolution grids (maps) are the highest resolution maps yet of cumulative carbon in marine sediments as carbon in methane, total methane, free gas and gas hydrate. These model results and uncertainties are easily updateable as new observed data and more accurate GML predictions become available. Many of our model results (e.g., base of the HSZ) are consistent with previous analyses, while other results (e.g., total methane generated all of geologic time) are the first of their kind. Integrating over the entire global ocean, using ensemble models, we estimate total carbon in methane and methane in all marine sediments to be between 0.8–2.2 and 1.1–3.0 × 10^6 Pg C and CH₄ respectively.

These new high-resolution estimates of total methane generated can provide key parameters that improve our ability to quantify carbon sequestration, capacity of long-term climate sinks, and may serve as a baseline for models which consider fluid flux to further our understanding of seafloor seepage. Understanding these estimates in geologic time can provide insights to how methane and methane hydrate is distributed with depth and age in the subsurface. Methane hydrate distributions are poorly understood, therefore, high resolution models which are variable spatially and with depth can help us further understand the extent of these massive reservoirs and how they evolve through time (You et al., 2020). Our final integrations represent a massive long term carbon sink. Carbon reservoirs of this magnitude may be consistent with IPCC (2021) suggestion that long-term carbon sinks such as these could explain imbalances in reduction of atmospheric CO₂ over the last 50 million years.

Data Availability Statement

Grids generated in this study are all publicly available and are included in the Supporting Information S1 and at https://doi.org/10.5281/zenodo.4776433.

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