

A photograph of a geological rock sample, likely a sedimentary rock, showing a distinct, elongated, and somewhat irregular shape. The rock is light-colored, possibly white or light gray, and has a rough, textured surface. A geological hammer is placed next to the rock for scale, indicating its size. The hammer has a dark handle and a metal head with a flat face and a pointed pick. The rock is set against a background of a rough, cracked, and layered rock face.

# A record of seafloor methane seepage across the last 150 My

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# Seafloor methane seepage

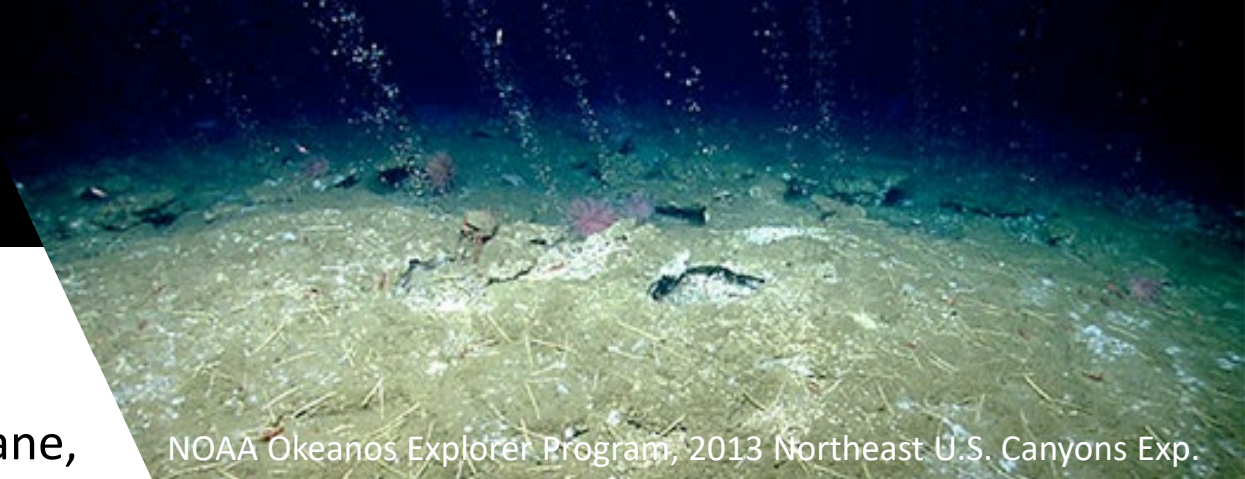
Sub-seafloor sediments host enormous volumes of methane, either as free gas or as hydrates

Part of this methane is continuously released in the water column by background seepage activity

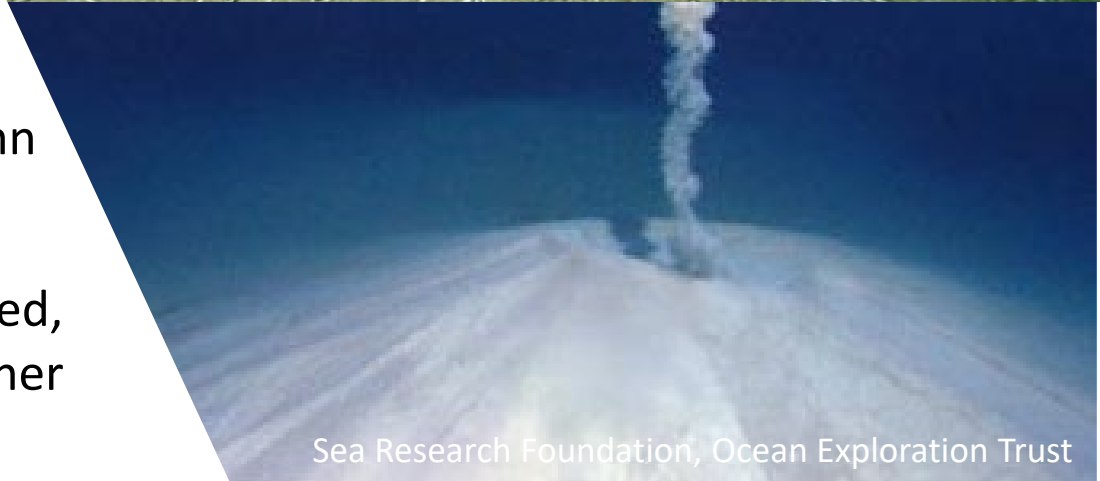
The total modern emission of seafloor methane is underestimated, and the volume of released methane is orders of magnitude higher than that reaching the sea surface

Perturbations in the environment, such as sea-level and temperature variations, may lead to the release of large methane volumes, which can have important local and global impacts

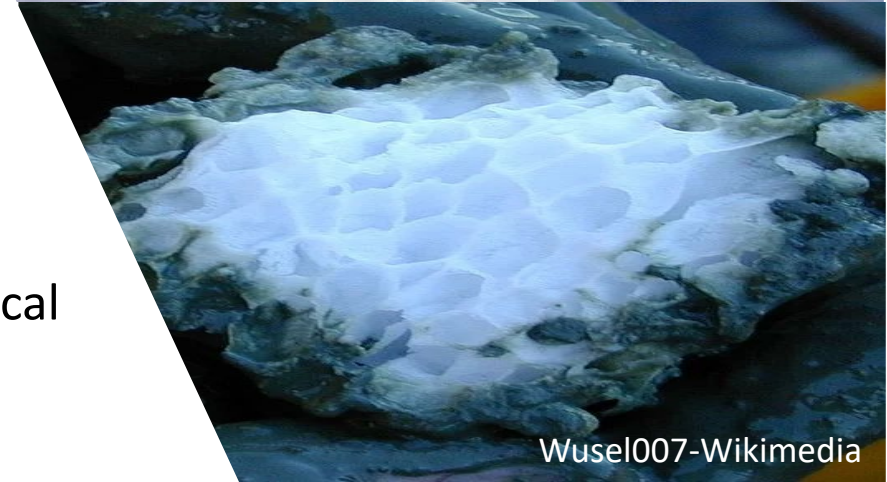
This scenario has been suggested to explain extreme events in the geological past



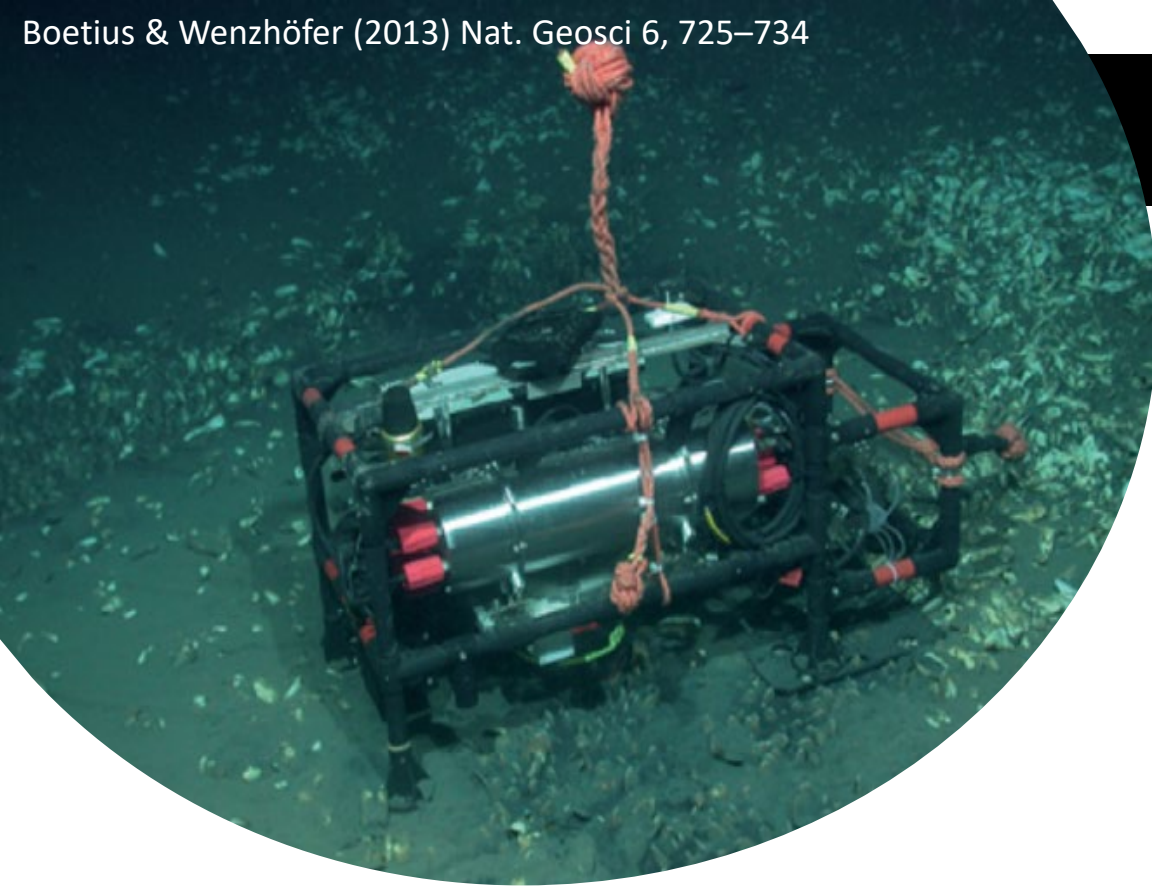
NOAA Okeanos Explorer Program, 2013 Northeast U.S. Canyons Exp.



Sea Research Foundation, Ocean Exploration Trust



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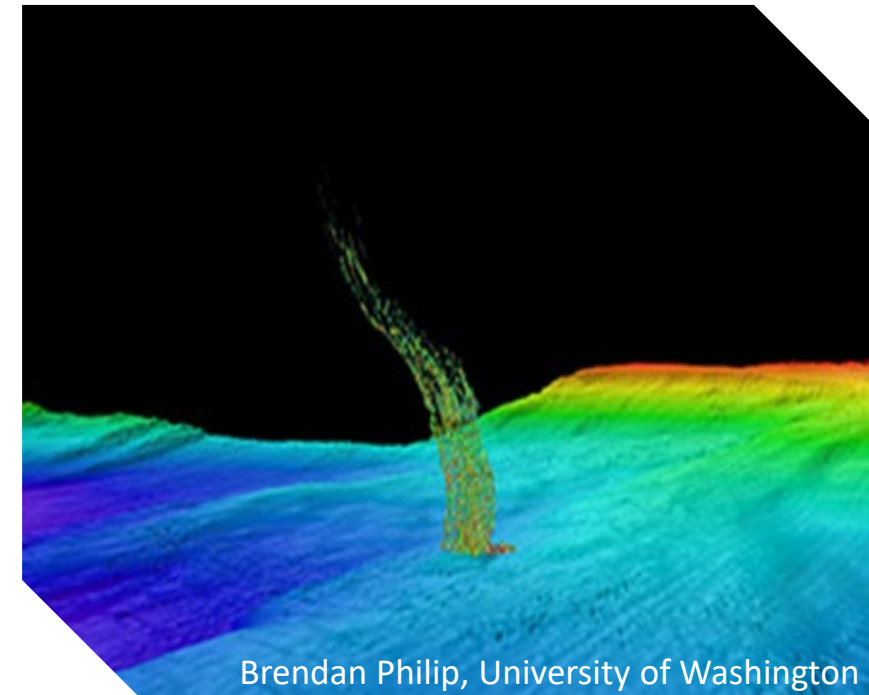
# Scientific gap

Rate of seafloor methane emission have varied through geological time and cold seepage systems can be regionally active on multi-million-year time scale

Past studies have primarily focused on either ancient or modern examples. Few have attempted to quantify long-term methane emission using compilations of globally distributed data

An exhaustive knowledge of the processes that may control methane release on long, geological time scales is still missing

Defining the processes involved in long-term cold seepage is necessary for establishing the precise relationship between geological and biochemical processes





# Materials & Methods

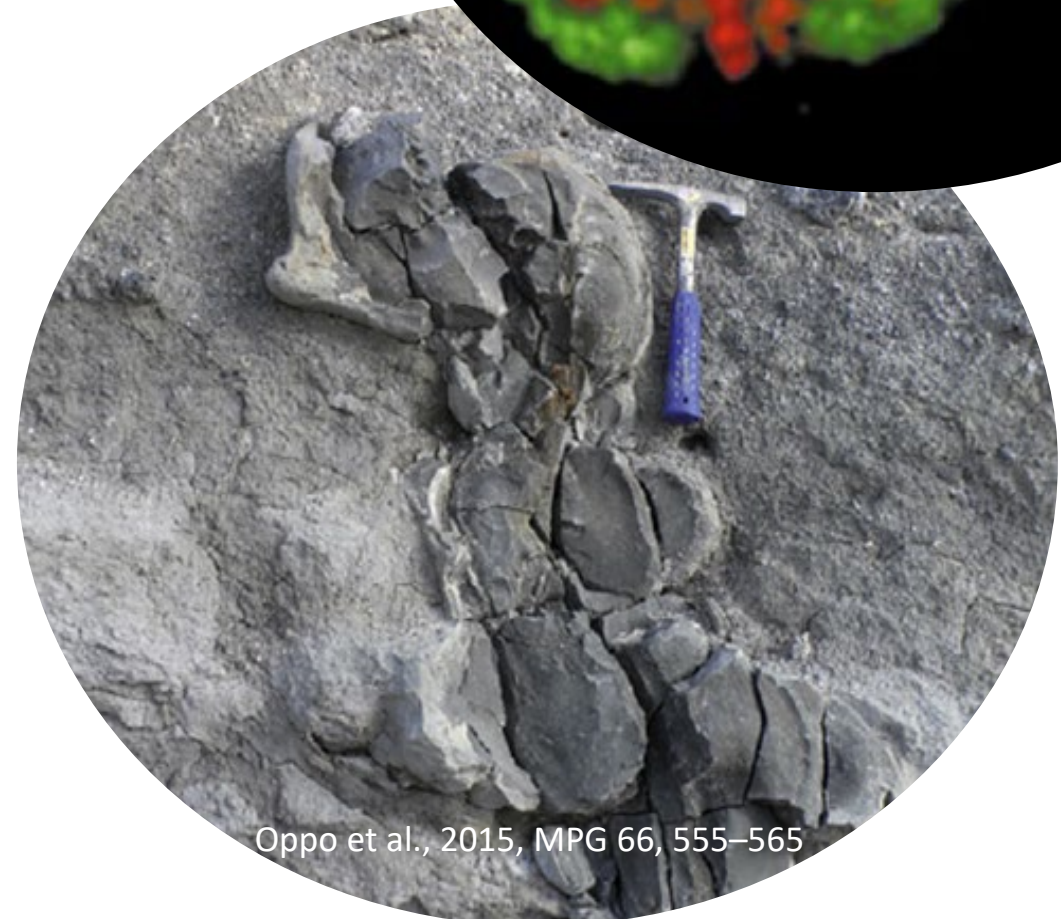
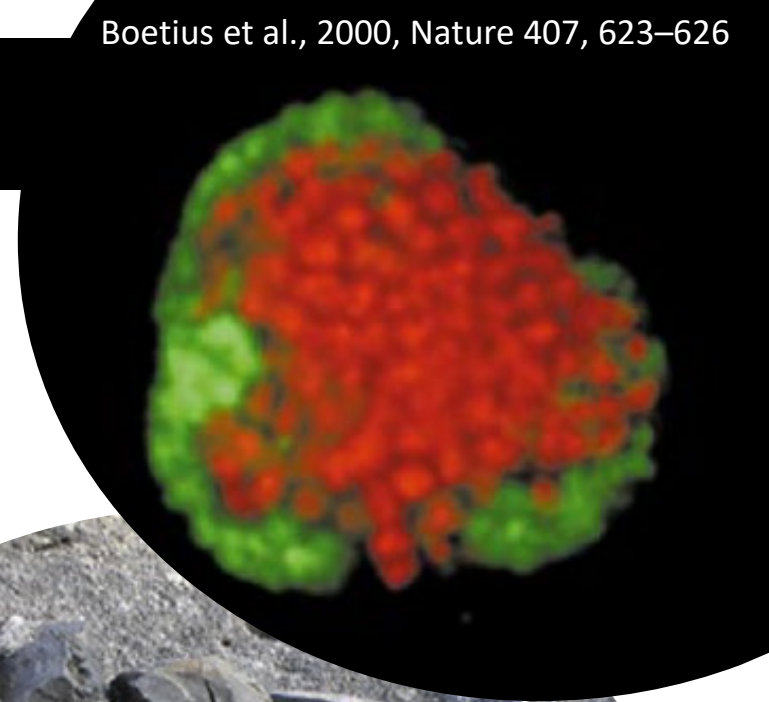
A robust proxy for estimating long-term seafloor methane emission is the record of Methane-Derived Carbonates (MDC)

Between 10% and 20% of methane oxidized within near-seafloor sediments precipitates as carbonate minerals, which are the main by-products of this process in marine environment

Because MDC are documented in sedimentary units ranging from the Neoproterozoic to Present, both modern and fossil MDC potentially record the trend of seafloor methane seepage across large intervals of geological history

We have compiled a database of worldwide occurrences of MDC and reconstructed the history of global MDC occurrence and natural methane emission from the seafloor across the last 150 My.

We selected data available in the literature and applied statistical and spectral techniques to (1) characterize the robustness and stability of the time series and (2) test the relative importance of global changes in sea level, deep ocean temperature, and organic carbon burial in mediating long-term methane release



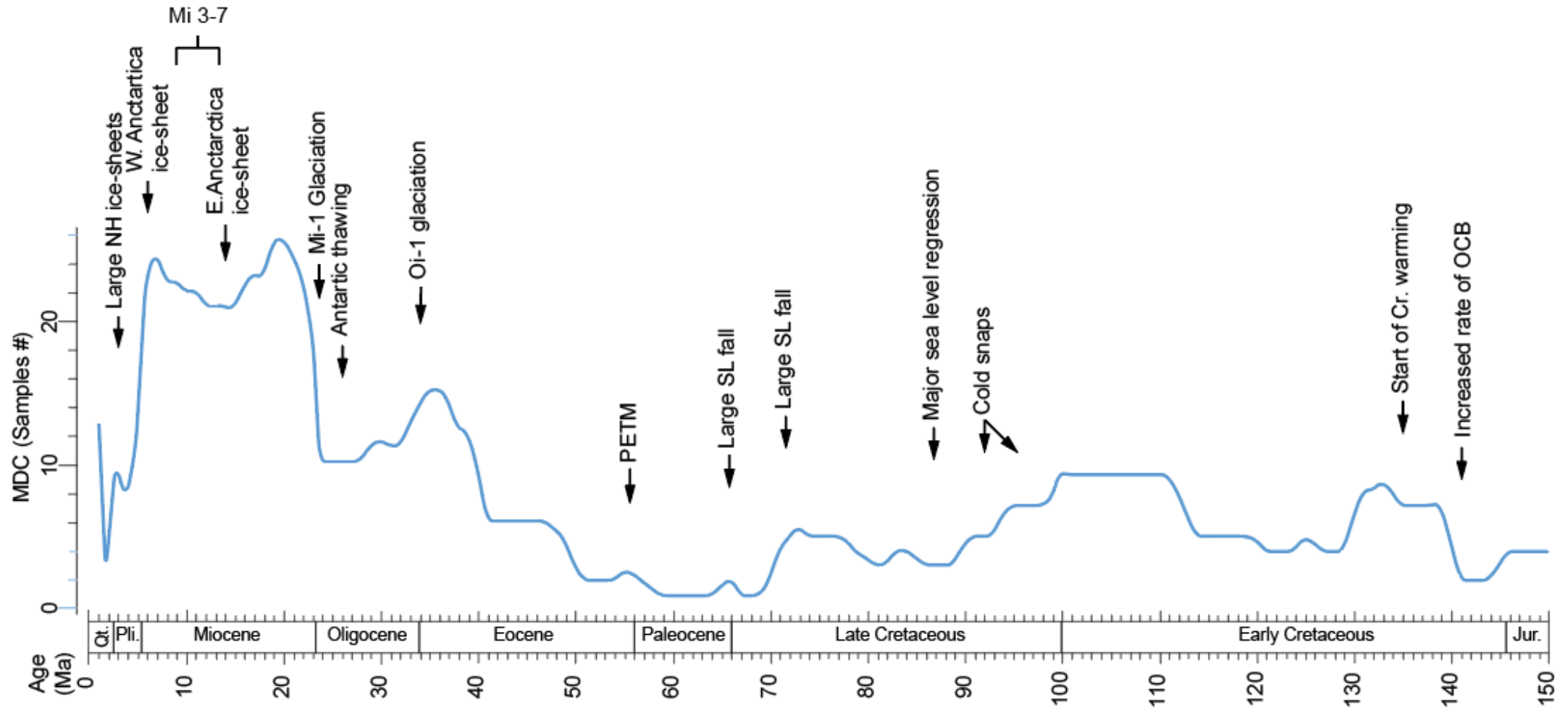
Oppo et al., 2015, MPG 66, 555–565

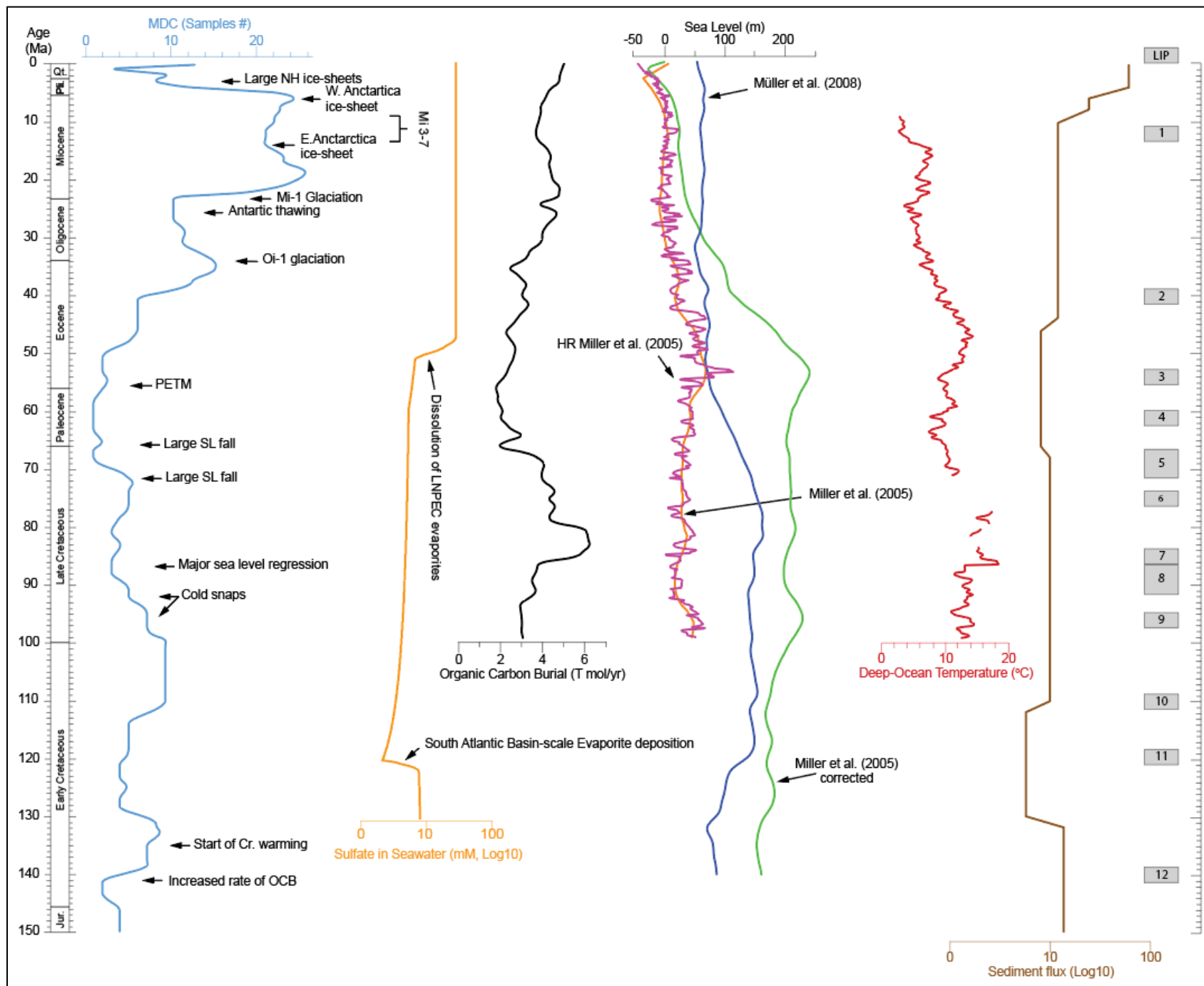
## Materials & Methods



Location of the methane-derived carbonates used in this study. Only one representative sample for each location/age combination has been considered

# MDC record across the last 150 My





Comparison of the MDC time series with the records of modelled Organic Carbon Burial (OCB), global sea level, deep-sea water temperature, seawater sulfate, global sediment flux, and large igneous provinces (LIP)

The MDC, OCB, sea level, and the temperature time series are interpolated to a 1 My time interval for the purposes of statistical analysis.

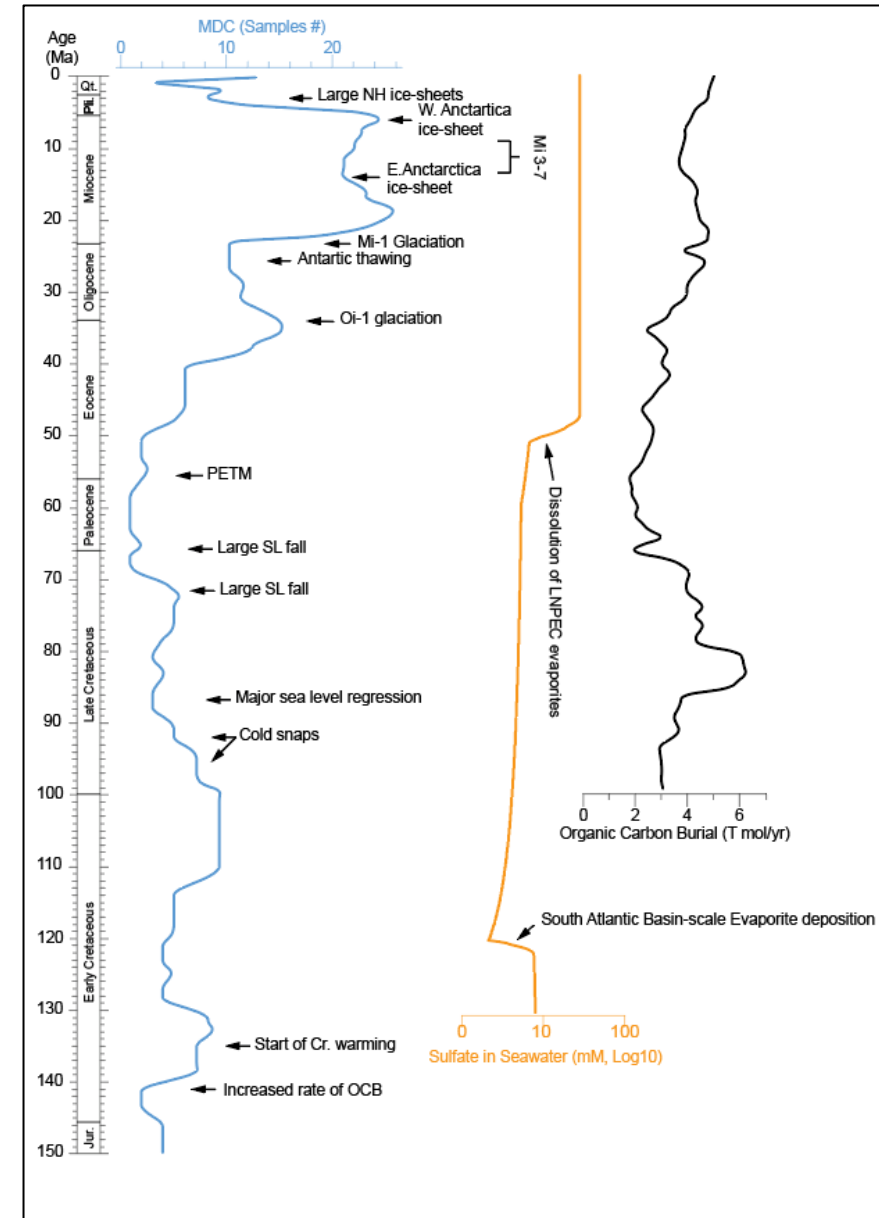
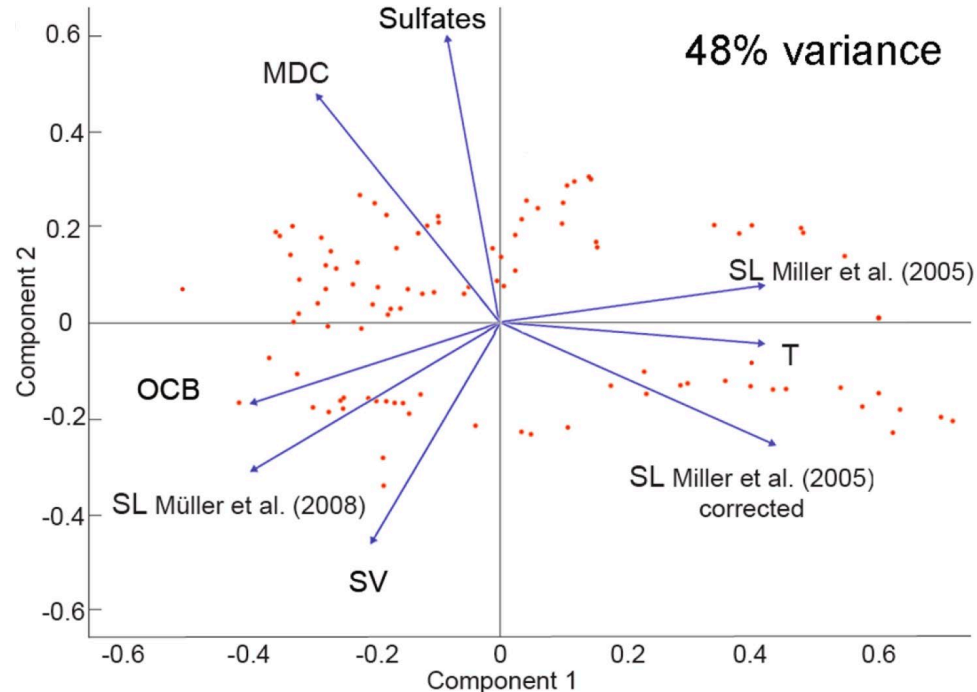
While prehistorical events are mapped on the MDC, inferences were made by statistical and spectral analyses.

See *Oppo et al., 2020. Scientific Reports 10:2562* for references to the time series

# Principal component analysis

Sulfate and MDC variations are best correlated for principal component analysis; however, sulfates have no cyclic behavior across the period considered. They act as an instantaneous sources of instability for MDC at ~50Ma and ~125 Ma.

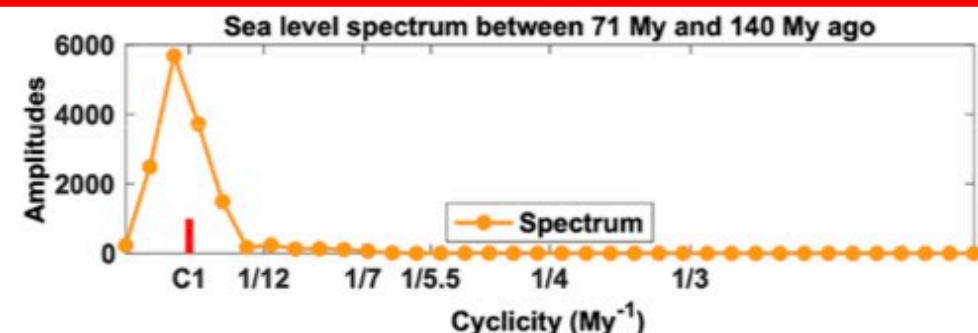
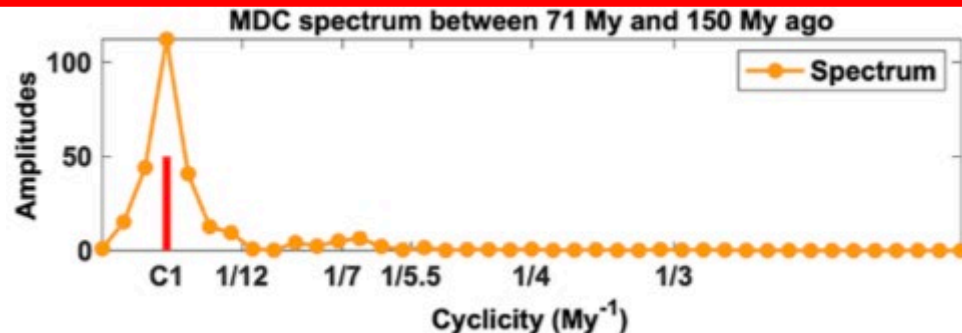
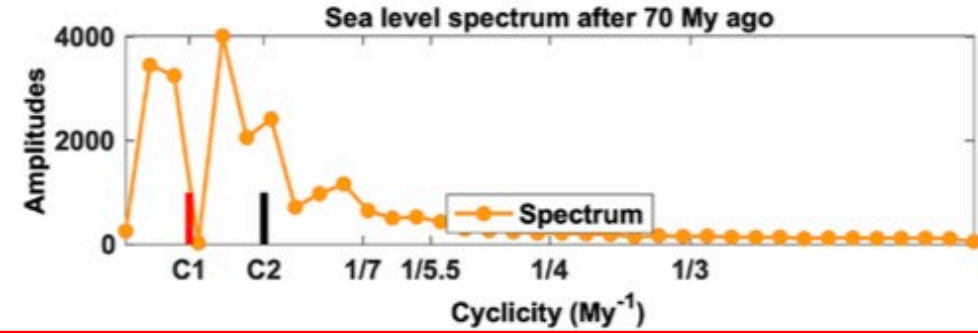
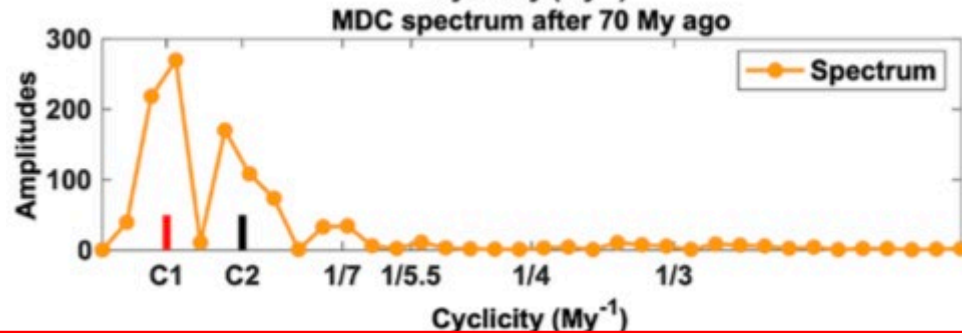
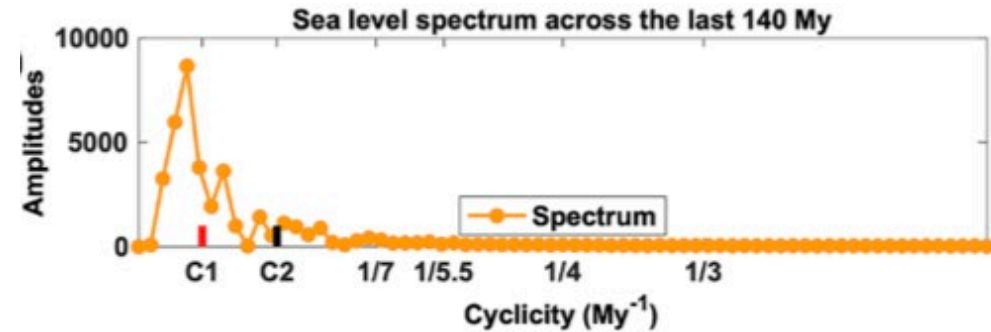
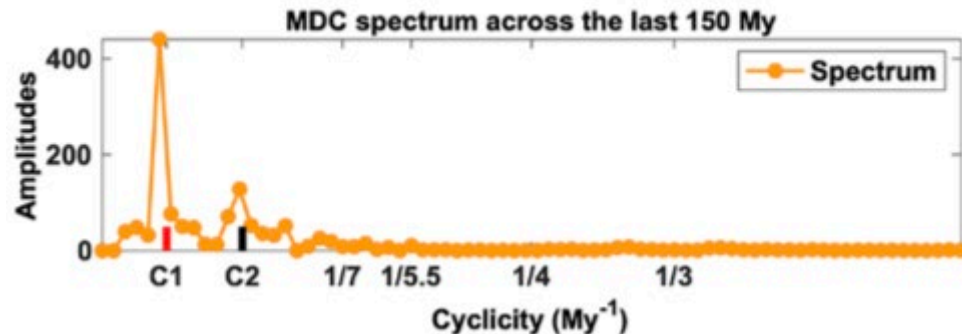
Once their effect is removed, the two most likely controllers of MDC remain variations in sea level (corrected sea level variations - SL - in the figure we show different models) and organic carbon burial (OCB).





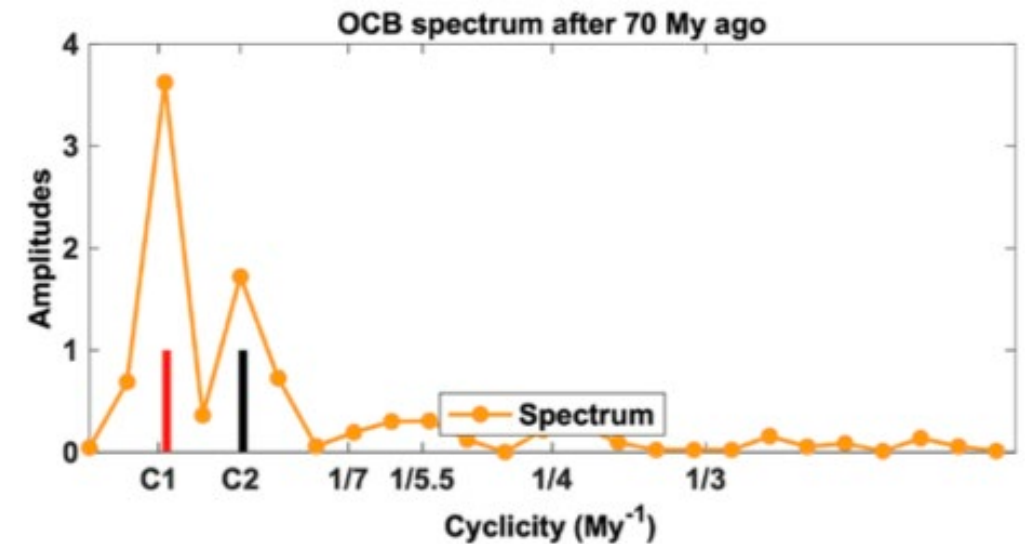
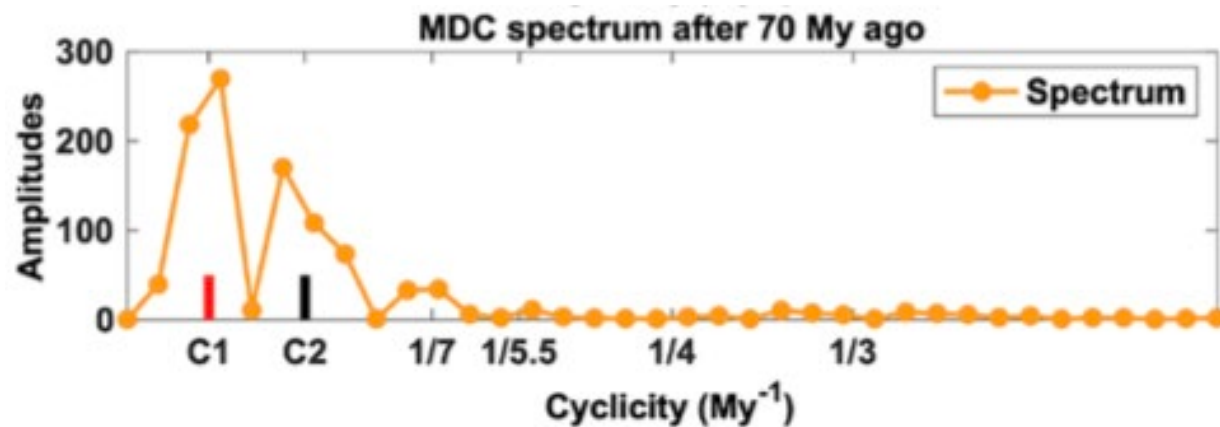
# Sea-level control on MDC

Sea level affects smooth trends in MDC on timescales of the order of tens of My (with the main cyclicity found at  $C1=26.66$  My), during which eustatic falls favored methane seepage. This is relevant especially between 71 My and 150 My but spectra differ in the last 70 My.



# Control of organic-carbon-burial on MDC

Although an increase of MDC abundance can ostensibly be associated with short-term sea level drops (< 5 My duration), most of the rapid sea level variations are not associated with significant cyclic variations in MDC abundance. The addition of cyclic variations over C2 = 12 My until 70 Ma highlights the similarity with OCB cyclic variations.



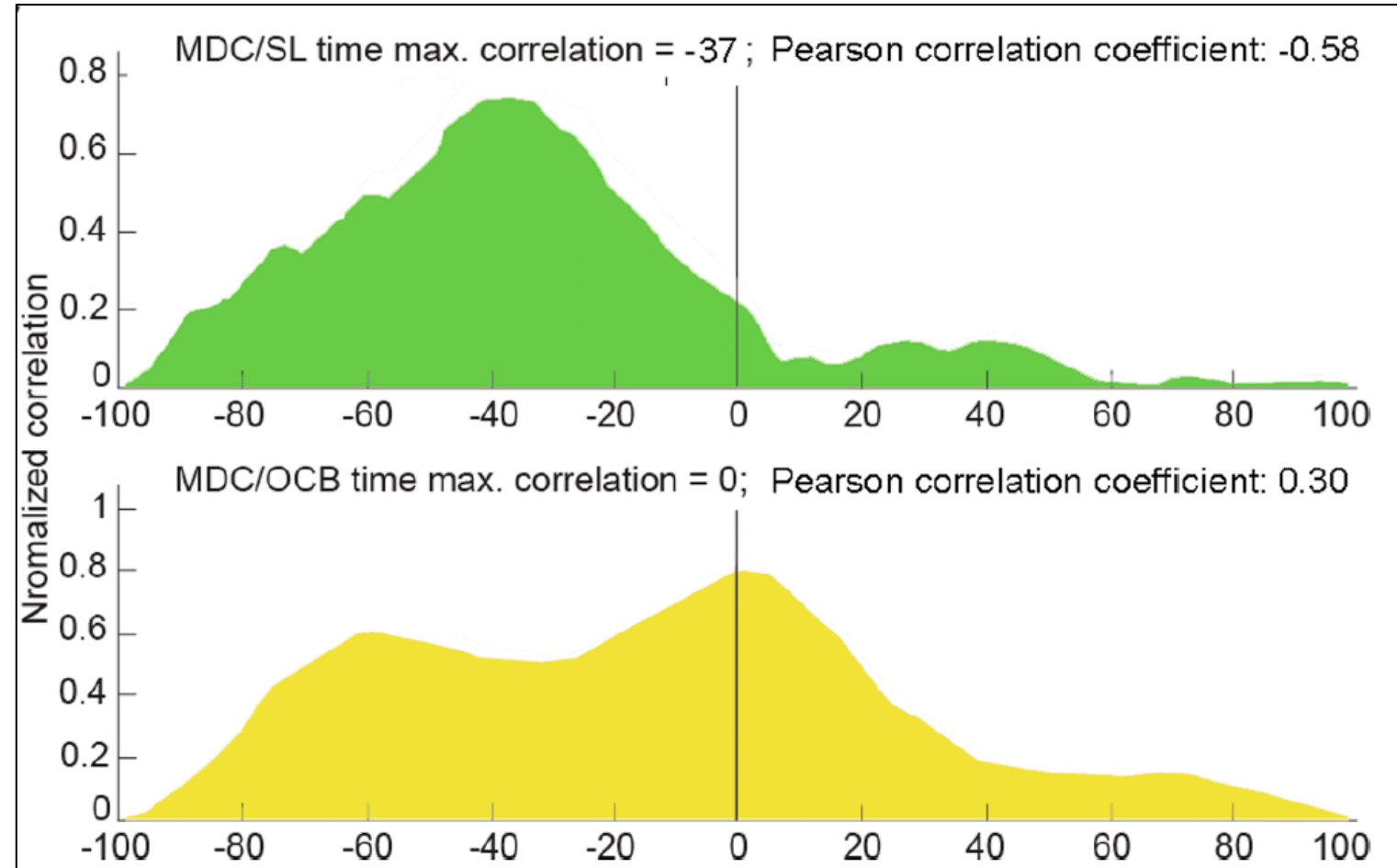


## OCB as instantaneous controller

The temporal correlation analysis confirms the lack of instantaneous correlation of SL variations, with best correlations obtained over the entire time-span of the process, from organic matter deposition to thermogenic methane seepage.

Instantaneous correlation is visible for OCB. Therefore, OCB likely represents a parameter that instantly influences seafloor methane emission over geologically very short (i.e. <1 My) periods.

We suggest that the 60 My time-lag reflects the signal of thermogenic methane, which acts as secondary contributor to the seafloor methane budget.



# Cyclicities

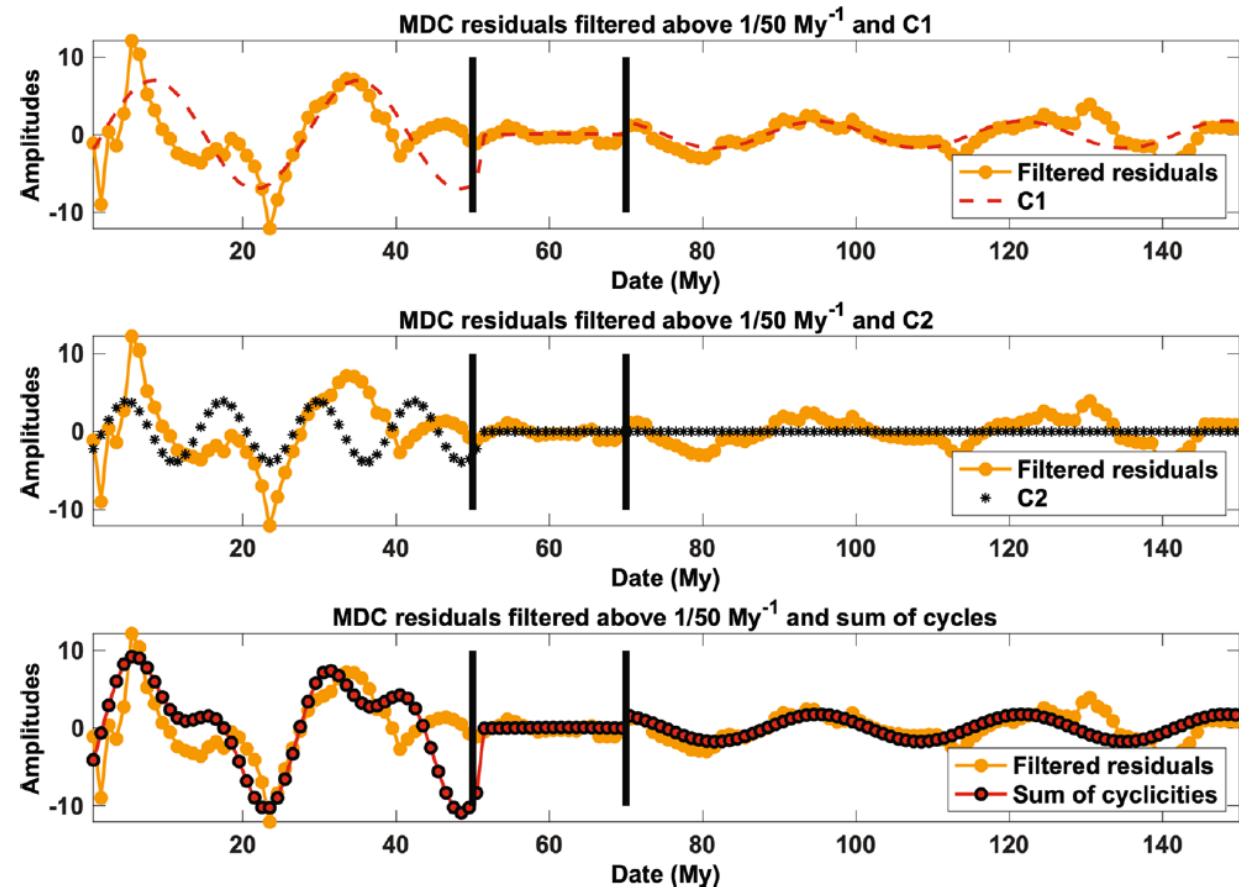
The two prominent cyclicities recognized by spectral analysis in MDC, sea level, and OCB data are :

- $C1 = 1/26.66 \text{ My}^{-1}$  - stable across the last 150 My
- $C2 = 1/12 \text{ My}^{-1}$  - out-of-phase cyclicity after 70 My

The fundamental cyclicity C1 is strikingly similar to the 26 My cyclicities observed in numerous global processes such as plate tectonic, atmospheric  $\text{CO}_2$ , oceanic anoxic events, and mass extinction events.

The C2 cycle is close to the  $\sim 9$  My cyclicity identified in Cenozoic/Mesozoic foraminifera  $\delta^{18}\text{O}$ , carbon-cycle, and sedimentological proxies

The addition of these two cyclicities provides an excellent first-order predictor of methane seepage, with major discrepancies induced by sulfate variations at  $\sim 50$  My and  $\sim 125$  My.





# Conclusions

The proposed reconstruction of seafloor methane seepage:

- Relates to a large spectrum of global phenomena
- Has key implications for a better understanding of methane cycling at the present day

The main controls influencing cyclic methane release are:

- **Sea level**, mainly affecting smooth trends on timescales of tens of My
- **Organic Carbon Burial**, instantly influencing methane emission over very short (<1 My) periods

The dynamics of methane escape to the global ocean undoubtedly involve interrelating additional factors, such as climate, sediment input and plate tectonics

**OPEN**

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