Goldschmidt 2022 Abstract
https://doi.org/10.46427/gold2022.10211

Reconstructing paleo-methane emission using the benthic foraminiferal stable sulfur isotopes

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Methane release from sedimentary environments represents an important component of the carbon cycle. Pore water geochemistry and modeling simulations allow the study of methane sources and sinks in modern environments. However, reconstructing methane dynamics through geological time is challenging as the interpretation of proxy-based investigations can be biased by the diagenetic alteration of the samples analyzed. In this study, we apply benthic foraminiferal stable sulfur isotopes ($\delta^{34}S$) to investigate changes in methane flux (diffusion vs. advection) in the fossil record. We used samples collected at Vestnesa Ridge (Arctic Ocean), a known site characterized by gas hydrates and seepage of microbial and thermogenic methane. Data were collected on *Cassidulina neoteretis*, a benthic foraminiferal species particularly abundant in the study area and widely utilized in paleoceanographic reconstructions.

Analysis of the benthic foraminiferal $\delta^{34}S$ revealed lower values (~20‰; modern seawater sulphate $\delta^{34}S$ = ~21‰) in the superficial sediment sample, which was not affected by methane release and oxidation. On the contrary, higher values (~25-27‰) were measured in samples deeper in the sediment column, which were affected by anaerobic methane oxidation and establishment of a sulphate-methane transition zone in the recent geological past. The use of the benthic foraminiferal $\delta^{34}S$ as a proxy to identify past migrations of the sulphate-methane transition zone was supported by the comparison of the benthic foraminiferal $\delta^{34}S$ data with the bulk benthic foraminiferal $\delta^{13}C$ data collected on the same samples. Very interesting were the results of the correlation between the benthic foraminiferal $\delta^{18}O$ and $\delta^{34}S$ values, as they provided information about methane flux. Specifically, the slope of the foraminiferal $\delta^{18}O$-$\delta^{34}S$ indicated that methane was advecting at the sampling site during the time frame of our study (early Holocene and the Younger-Dryas-post-Bølling). This finding provided the exciting opportunity to apply the benthic foraminiferal geochemistry to indirectly date methane emissions. In fact, the sulphate-methane transition zone is shallow when the methane flux is high (i.e., advection; slope of the benthic foraminiferal $\delta^{18}O$-$\delta^{34}S$ correlation = ~0.2) and this allowed us to interpret the signal recorded by the benthic foraminifera as almost coeval to the time of sediment deposition at the studied site.