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Subsurface heat and salts cause exceptionally limited methane hydrate stability in the Mediterranean Basin

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ABSTRACT

Knowledge of the global reservoir of submarine gas hydrates is of great relevance for understanding global climate dynamics, submarine geohazards, and unconventional hydrocarbon energy resources. Despite the expected presence of gas hydrates from modeling studies, the land-locked Mediterranean Basin displays a lack of evidence of extensive gas hydrate presence from samples and seismic data. We modeled the theoretical Mediterranean distribution of methane hydrate below the seafloor and in the water column using available geological information provided by 44 Deep Sea Drilling Project (DSDP) and Ocean Drilling Program (ODP) boreholes, measured geothermal gradients, and thermohaline characteristics of the water masses. We find that the pervasive presence of high-salinity waters in sediments, coupled with the unique warm and salty water column, limit the thickness of the theoretical methane hydrate stability zone in the subsurface and deepen its top surface to 1163–1391 m water depth. The theoretical distribution of methane hydrates coincides well with the distribution of shallow, low-permeability Messinian salt deposits, further limiting the formation of pervasive gas hydrate fronts and controlling their or distribution due to the prevention of upward hydrocarbon gas migration. We conclude that the Mediterranean Basin, hosting the youngest salt giant on Earth, is not prone to the widespread formation and preservation of gas hydrates in the subsurface and that the gas hydrate potential of salt-bearing rifted continental margins may be considerably decreased by the presence of subsurface brines.

INTRODUCTION

Shallow sediments of the ocean margins are known to host a globally widespread reservoir of natural gas hydrates, mainly methane, according to a thermodynamic stability field controlled by pressure, temperature, water salinity, and gas composition (Ruppel and Kessler, 2017; Mienert et al., 2022). In present-day continental margins, the methane hydrate stability zone (MHSZ) typically extends from ~300 m below sea level to nearly 900 m below the seafloor (Kretschmer et al., 2015). The maximum theoretical thickness of the MHSZ coincides with passive continental margins where the geothermal gradient is relatively low (Lucazeau, 2019).

The Mediterranean Basin is a geologically complex land-locked oceanic basin, in which the primary geophysical evidence of gas hydrates (bottom-simulating reflectors; BSRs) and other indicators in geological samples are scarce or absent. The only geological environment where the presence of gas hydrates is proven is that of mud volcanoes (e.g., Camerlenghi and Pini, 2009, and references therein), in which methane advection occurs in narrow mud conduits rather than being pervasive and widespread along continental margins. Despite several attempts to understand the gas hydrate system (Merey and Longinos, 2018; Tayber et al., 2019; Minshull et al., 2020; Obhodas et al., 2020; León et al., 2021), the Mediterranean lacks an assessment of its gas hydrate potential that considers measured thermohaline properties of interstitial waters.

We considered the full spectrum of physical and chemical parameters available from open-access databases and undertook a basin-wide approach to the modeling of the MHSZ in the Mediterranean Basin. We propose a brine-limited model for methane hydrate stability, which can be used to inform gas hydrate assessments in other rifted and salt-bearing continental margins globally. Our findings widen the results of previous studies recognizing the inhibiting role of excess salinity in pore waters of sediment cores on methane hydrate stability at the scale of salt diapir mounds in the Gulf of Mexico (Paull et al., 2005; Ruppel et al., 2005).

MODELING APPROACH

Our model uses the temperature and salinity of subsurface pore waters from 44 Deep Sea Drilling Project (DSDP) and Ocean Drilling Program (ODP) boreholes (Fig. 1A; see the Supplemental Material[1]). Water column thermohaline properties are from measurements (PERSEUS Oceanographic Mediterranean and Black Sea Data Management; https://isramar.ocean.org.il/perseus_data/) and model grids (Copernicus Marine Environment Monitoring Service, CMEMS; https://insitu.copernicus.eu/FactSheets/CMEMS/). The stability field of methane hydrate is simulated as a function of *in situ* pressure, temperature, and salinity using the equation of state of Moridis (2014) in each of the 44 boreholes and in the water column. Details and accuracy analysis are reported in the Supplemental Material.

MHSZ IN THE WATER COLUMN AND IN THE SUBSURFACE

The modeled intersection of the top of the MHSZ with the seafloor ranges from 1163 m to 1391 m below sea level (Fig. 1B), well beyond the continental shelf break and considerably deeper than in world continental margins. The top of the MHSZ is uniformly distributed in the Ionian and Herodotus-Levantine Basins,

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[1] Supplemental Material. Data and methods, Figure S1, Tables S1 and S2, and Plates S1 and S2 (the graphical results of modeling of DSDP and ODP boreholes, displayed with lithostratigraphy). Please visit https://doi.org/10.1130/GEOL.S.21606054 to access the supplemental material, and contact editing@geosociety.org with any questions.

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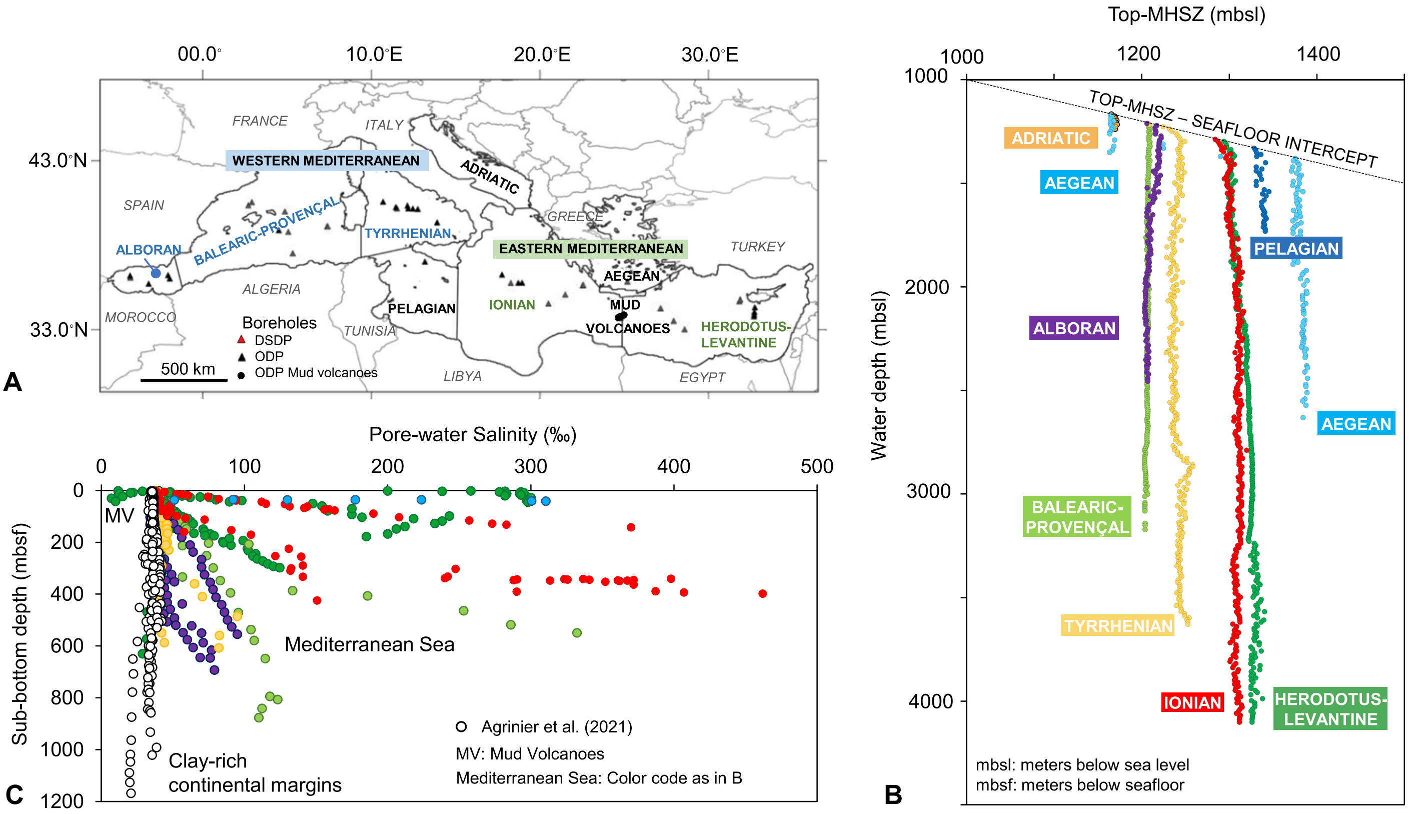

Figure 1. (A) Locations of 44 Deep Sea Drilling Project (DSDP) and Ocean Drilling Program (ODP) boreholes analyzed, and sedimentary basins considered, in this study. (B) Plot of the modeled top of the methane hydrate stability zone (MHSZ) in the water column versus water depth in the sedimentary basins outlined in A. The top-MHSZ–seafloor intercept line identifies the limit above which methane hydrates cannot be stable according to the Moridis (2014) model. Maximum error is 1.45% in the Herodotus-Levantine Basin, corresponding to a maximum depth variation of ±19 m (see data and methods in the Supplemental Material [see footnote 1]). (C) Comparison of borehole salinity from Mediterranean Sea boreholes and 13 typical clay-rich continental margin sedimentary successions, after Agrinier et al. (2021). Mediterranean pore-water salinity in excess of the typical range of seawater salinity (~33‰–37‰) explains the use of the term "subsurface brines" in this study. The original chlorinity in the data has been converted to salinity using the relation salinity (‰) = chlorinity (‰) × 1.80655.

reflecting a uniform water column in the Eastern Mediterranean. Conversely, the Tyrrhenian, Balearic-Provençal, and Alboran Basins in the Western Mediterranean display a shallowing trend of the top of the MHSZ, reflecting the modification of the water masses toward the Gibraltar marine gateway. The shallow Adriatic, Pelagian, and Aegean Basins display the highest variability.

In 33 of the 44 boreholes analyzed, we found important positive pore-water salinity anomalies (Figs. 1C and 2; Plates S1 and S2 in the Supplemental Material), indicating the pervasive presence of brines with concentrations of halite and gypsum as high as saturation (>300‰). In the Eastern Mediterranean, the thickness of the subsurface MHSZ is largest (as much as ~350 m) and the anomaly induced by subsurface brines is highest (~−300 m), with a thinning of the subsurface MHSZ by as much as 85% with respect to its thickness, calculated assuming constant pore-water salinity equal to bottom-water salinity. In the Western Mediterranean, Pelagian, Adriatic, and Aegean Basins, the MHSZ attains its minimum thickness of <100 m.

INHIBITING FACTORS OF METHANE HYDRATE STABILITY

Warm and Saline Mediterranean Water Column

The Mediterranean hosts an endogenic and dynamic overturning circulation driven by sinking of dense high-salinity waters produced primarily by wind-driven cooling (Pinardi et al., 2019). However, the bottom-water temperature is high, ranging from west to east from ~13 °C to ~14 °C, and salinity is always in excess of 38‰ (Zavatarielli and Mellor, 1995). The Aegean Basin is a remarkable exception in the Mediterranean because extreme spatial and temporal variability of water mass thermohaline properties (Mamoutos et al., 2021) produces the widest range of depth of the top of the MHSZ in the whole Mediterranean (Fig. 1B). In addition, events such as the Eastern Mediterranean Transient, generated in the Aegean Basin, can affect the thermal structure of the shallow sediments even in abyssal environments (Della Vedova et al., 2003), introducing a locally important variable in the distribution of the MHSZ in water columns and shallow sediments.

Age of the Underlying Lithosphere and Consequent Offshore Thermal Regime

The Eastern Mediterranean Basin floor results from nearly complete subduction of presumed Triassic oceanic lithosphere and incipient African and Eurasian continental collision, while the Western Mediterranean relatively young backarc basin has been undergoing stepwise opening since the Oligocene (e.g., Speranza et al., 2012; van Hinsbergen et al., 2020). Therefore, the regional thermal structure is bimodal, with low to very low heat flow (1–41 mW m^{-2}) in the Eastern Mediterranean and Adriatic Basins contrasting with high to very high heat flow (65 to >107 mW m^{-2}) in the Western Mediterranean, Pelagian, and Aegean basins (Davies, 2013). Due to warmer subsurface temperatures, the latter basins are less favorable to methane hydrate stability and generate a thin MHSZ in the subsurface (Fig. 2A).

Widespread Occurrence of Brines in the Shallow Subsurface

Western and Eastern Mediterranean, with scattered occurrences in the Tyrrhenian and

Aegean Basins, host the youngest salt giant on Earth (ca. 5.9–5.3 Ma, Messinian Age of the Miocene) at shallow sub-bottom depth, covered by Pliocene and Quaternary clays and silts (Haq et al., 2020). The areal extent of the MHSZ matches the distribution of the salt giant (Fig. 3).

The brines are generated by subsurface dissolution of most abundant and soluble salts (halite and gypsum; Elderfield and Summerhayes, 1978). Upward molecular diffusion of Cl^-, Na^+, Ca^{2+} and SO_4^- in the pore water of Pliocene and Quaternary hemipelagic sediments can attain linear concentration gradients from saturation at the top of evaporites to seawater concentration at the seafloor. Equilibrium is reached in $\sim$2 m.y. in a 200-m-thick hemipelagic cover (Camerlenghi, 1988). However, observed gradients in DSDP and ODP boreholes are not always linear and do not allow prediction of saturation concentrations at the top of the evaporitic layer (Plates S1 and S2). Deviations from diffusive gradients can be induced by several factors such as pore-water dilutions by dehydration of gypsum to anhydrite (Hoareau et al., 2011), compaction disequilibrium induced by rapid loading during evaporitic deposition (Dale et al., 2021), and/or osmotic and hyperfiltration effects (e.g., Agrinier et al., 2021). The complex mechanisms of subsurface evaporitic dissolution prevent predictable behavior of brine flow and ion diffusion at a basin scale. However, the spatial match between MHSZ and underlying Messinian evaporites implies that subsurface brines can be expected nearly everywhere within the MHZS of the Mediterranean Basin. Limited portions of the MHZS not underlain by evaporites are in the shallow margins of the Eastern Mediterranean and include parts of the Nile River deep-sea fan.

The influence of sediment pore-water salinity on methane hydrate stability on continental margins was described by Dickens and Quinby-Hunt (1997) and proven at shallow sub-bottom depth in focused fluid-flow sites in the Gulf of Mexico (Paull et al., 2005; Ruppel et al., 2005). However, pore-water salinity equal to that of bottom water is assumed in the vast majority of the estimations of the base of the MHSZ in continental margins. Our modeling results suggest that subsurface brines produced by buried salt rocks can produce dramatic reductions of the thickness of the MHSZ over an entire sedimentary basin, especially where the geothermal gradient is low, as in the Eastern Mediterranean. To qualitatively assess the relative importance of the limiting effects of subsurface brines and temperature on the thickness of the MHSZ, we have modeled synthetic cases to simulate Western and Eastern Mediterranean subsurface thermohaline conditions (Fig. 4). In the Western Mediterranean, the salinity effect is dramatically

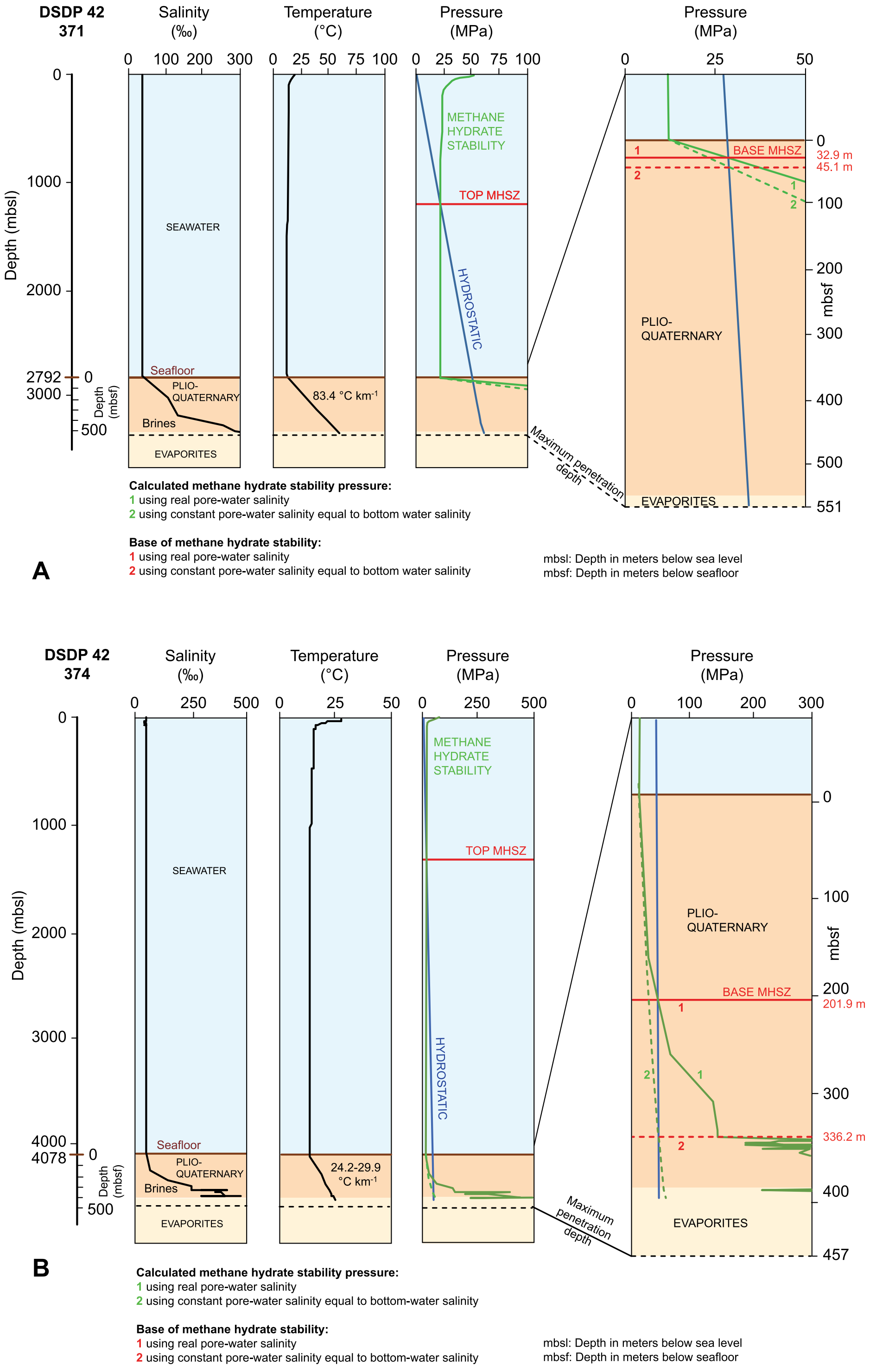

Figure 2. Calculation of the methane hydrate stability zone (MHSZ) in two end-member Deep Sea Drilling Project (DSDP) boreholes with presence of subsurface brines and contrasting geothermal gradient (the number 42 following DSDP identifies the drilling Leg number; the numbers 371 and 374 refer to the drilling Site). The MHSZ is calculated in each case for real and constant pore-water salinity. See the data and methods in the Supplemental Material (see footnote 1) for details and accuracy. (A) Western Mediterranean case of a high geothermal gradient. Thickness of the subsurface part of the MHSZ is 32.9 m (case 1). If calculated without brines, the thickness would be 45.1 m (case 2). The resulting brine-induced anomaly is –12.2 m. (B) Eastern Mediterranean case of a low geothermal gradient. Thickness of the subsurface part of the MHSZ is 201.9 m (case 1). If calculated without brines, the thickness would be 336.2 m (case 2). The resulting brine-induced anomaly is –134.3 m. Similar analyses for 44 boreholes are reported in Plates S1 and S2 in the Supplemental Material. Numerical results are provided in Table S1.

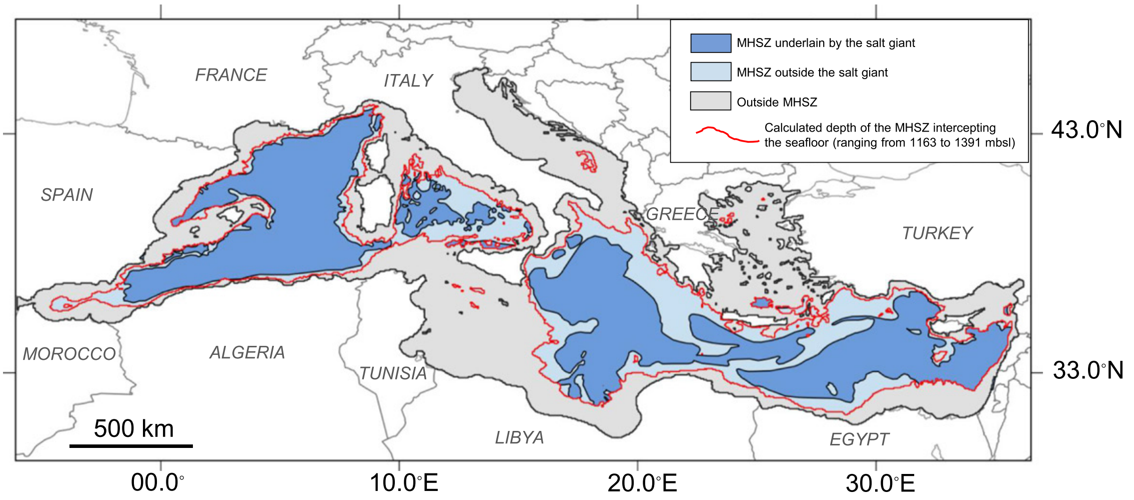

Figure 3. Superposition of the modeled subsurface areal extent of the methane hydrate stability zone (MHSZ) and the Mediterranean salt giant (after Lofi et al., 2018). Considering the 5% volumetric error in estimated distribution of evaporites by Haq et al. (2020), the majority of the seafloor hosting the MHSZ is underlain by the salt giant. Discrepancies are more common in the geothermally cold Eastern Mediterranean. mbsl—meters below sea level.

attenuated by the thermal effect that produces the most extensive thinning of the MHSZ.

Sealing Capacity of the Mediterranean Salt Giant

The evaporitic sequences in the deep Mediterranean basins, as in similar halite bodies worldwide, acts as effective barriers to upward migration of formation fluids (e.g., Warren, 2017). Important hydrocarbon gas plays are widespread along Mediterranean continental margins, and large methane reservoirs can be efficiently sealed by salts (e.g., offshore Egypt; Esestime et al., 2016). Bypass of the Mediterranean salt seal by subsalt gas migrating upwards into the MHSZ has occurred only through locally focused fluid migration paths (Bertoni and Cartwright, 2015), including mud volcanoes, with catastrophic seal failure and leakage from sub-salt biogenic petroleum systems induced by dramatic base-level oscillations associated with the Messinian salinity crisis (Al-Balushi et al., 2016). The only BSR reported outside mud volcanoes, on the Nile deep-sea fan (Praeg et al., 2022), is associated with the absence of underlying salts.

Therefore, with the exception of mud volcanoes and local breaches in the salt seal, the Mediterranean salt giant acts as an important regional-scale limiting factor to the formation of gas hydrates.

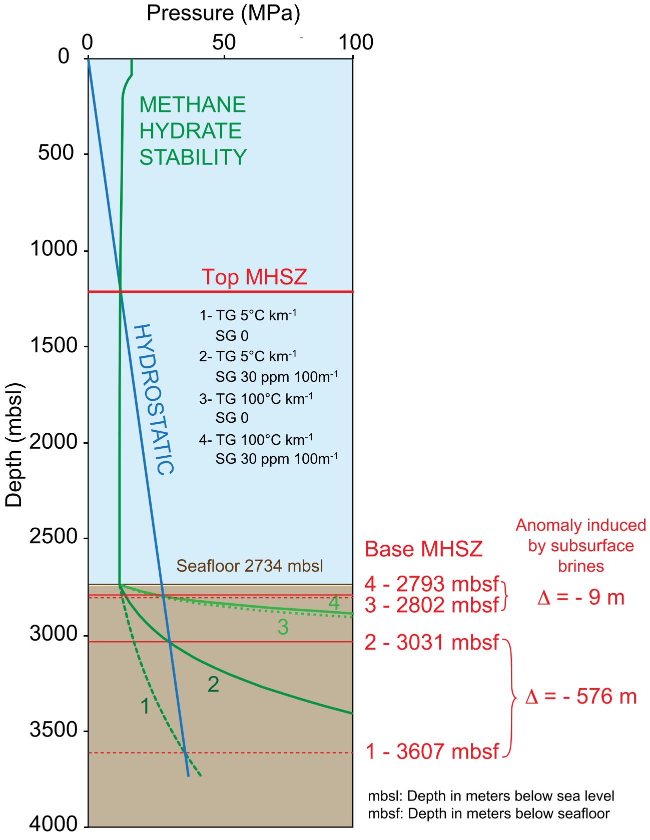

Figure 4. The presence of subsurface brines induces a striking thinning of the subsurface methane hydrate stability zone (MHSZ) only when the geothermal gradient is low. In the Eastern Mediterranean extreme case of low geothermal gradient (TG = 5°C km⁻¹), the realistic pore-water salinity gradient (SG = 30 ppm 100m⁻¹; case 2) induces thinning of the MHSZ exceeding 570 m with respect to the MHSZ calculated with constant pore-water salinity equal to bottom water salinity (SG = 0 ppm 100m⁻¹; case1). In contrast, in the Western Mediterranean extreme case of high geothermal gradient (TG = 100°C km⁻¹), the realistic pore-water salinity gradient (SG = 30 ppm 100m⁻¹; case 4) induces thinning of the MHSZ less than 10 m with respect to the MHSZ calculated with constant pore-water salinity equal to bottom water salinity (SG = 0 ppm 100m⁻¹; case 3). See the data and methods in the Supplemental Material (see footnote 1) for details on accuracy of the MHSZ determinations in the water column and in the subsurface.

Availability of *In Situ* Biogenic Methane in the MHSZ

The pore-water composition from ODP Leg 160 and Leg 161 boreholes demonstrates that variable organic-matter contents and sedimentation rate affect sulfate reduction in the pore water without generating sulfate depletion and methanogenesis, with the only exception in the evaporite-free Alboran Basin (Böttcher et al., 1998, 1999). In the deeper sedimentary sequence, close to the top of the evaporites, the sulfate ion concentration is increased by the brines, with authigenic gypsum precipitation as a result. Therefore, the pore-water chemistry expected above the Messinian evaporitic layer is unfavorable to *in situ* organic-matter fermentation and methanogenesis and acts as a further limiting factor to the generation of gas hydrates in the MHSZ.

CONCLUSIONS

Our study demonstrates that a combination of geological and oceanographic factors limits the theoretical stability field of methane in the water column and in the subsurface of the Mediterranean Sea. The study extends to an entire sedimentary basin the heat- and salt-induced limitation to gas hydrate formation recognized in shallow cores and on piercement structures in the Gulf of Mexico. The Western Mediterranean is where the combination of heat and salt produces the most dramatic reduction of the thickness of the subsurface MHSZ. Modeling and the environmental conditions support the lack of geophysical and geochemical evidence of gas hydrates in areas underlain by Messinian salts. If sufficient free methane can accumulate below the base of the MHSZ, a BSR on seismic reflection data may be expected in much deeper seafloor depth and shallower sub-bottom depth than in other continental margins globally. Given the occurrence of salt giants formed in different geological times on rifted margins of the Atlantic Ocean (including Gulf of Mexico outside piercement structures, and Barents Sea), Indian Ocean, and Red Sea, the pervasive presence of brines in the subsurface should be considered in future assessments of the gas hydrate global reservoir, the global carbon budget, and, where relevant, petroleum systems.

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