Sill-controlled salinity contrasts followed post-Messinian flooding of the Mediterranean

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14 5.33 million years ago, a mile-high marine cascade terminated the Messinian Salinity Crisis due to partial collapse of the Gibraltar sill that had isolated a largely desiccated 15 Mediterranean from the Atlantic Ocean. Atlantic waters may have refilled the basin within 16 2 years. Prevailing hypotheses suggest that normal marine conditions were established 17 across the Mediterranean immediately after the catastrophic flooding. Here we use proxy 18 data and fluid physics-based modelling to show that normal conditions were likely for the 19 western Mediterranean (wMed), but that flooding caused a massive transfer of salt from 20 21 the wMed to the eastern Mediterranean (eMed) across the Sicily sill, which became a hyper-22 salinity-stratified basin. Hyper-stratification inhibited deep-water ventilation, causing anomalously long-lasting organic-rich (sapropel) sediment deposition. Model-data 23 agreement indicates that hyper-stratification breakdown by diapycnal diffusion required 24 26,000 years. An alternative hypothesis that Atlantic reconnection occurred after the 25 Mediterranean had largely been refilled is inconsistent with our observations, as this would 26 27 have led to hyper-stratification and sapropel formation in both basins. Our findings offer insight into the role of stratification in delaying the re-establishment of normal marine 28 29 conditions following abrupt refilling of a previously desiccated ocean basin.

The 630,000-year Messinian Salinity Crisis (MSC) resulted from progressive closure of the connection(s) between the Mediterranean Sea and Atlantic Ocean ~5.96 Million years ago (Ma)^{1–3}. During the MSC, massive evaporite sequences were deposited. A phase of km-scale

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Mediterranean drawdown occurred⁴, resulting in a basin-wide Messinian erosional surface⁵ 33 and 2,500 m and 1,300 m deep Messinian canyons excavated beneath the Nile delta and the 34 Rhône River mouth, respectively^{6,7}. It has long been inferred that Mediterranean sea levels 35 were low, ~1,300 m to ~2,700 m below global sea level^{5,8–11}, prior to the earliest Pliocene 36 (Zanclean) megaflood event that terminated the MSC. Alternatively, geochemical and 37 paleobiological evidence from latest Messinian evaporites may imply elevated basin levels, 38 attributing the drawdown to an older Messinian phase¹². In that scenario, increased 39 Paratethyan and occasional Atlantic inflows are suggested to have largely refilled the 40 Mediterranean¹³ (a shallow Mediterranean base level below Atlantic, with interconnected 41 42 subbasins) with lower-salinity water overlying deep hypersaline fluids prior to complete Atlantic reconnection^{12–14}. We investigate the consequences of abrupt refilling of a partially 43 44 desiccated basin (a kilometer-scale drawdown in the megaflood hypothesis) and address the 45 alternative hypothesis in a sensitivity test (see Methods, Extended Data Figs. 6-8).

46 Evidence supporting the megaflood hypothesis

The megaflood hypothesis is based on seismic and borehole data from both the wMed and 47 eMed^{15–17}. Critical evidence at the Strait of Gibraltar comes from a massive erosive channel 48 in the Gibraltar arc/sill, which extends 390 km from the Gulf of Cadiz on the Atlantic side 49 toward the deep Alboran Sea on the Mediterranean side; the so-called Zanclean channel³. 50 51 Elongated megabar deposits detected alongside the main erosive channel in the eastern Alboran Sea have been related to the flooding event¹⁶. This sharply defined channel has 52 maximum depth and width of 650 m and 15 km, respectively^{3,16}. The dimensions of this 53 incision have led to model estimates of water fluxes of up to ~150 Sv (Sverdrup, $1 \times 10^6 \text{ m}^3\text{s}^{-1}$) 54 during the Zanclean flood³. 55

56 The most plausible flood-water passage from the wMed into the eMed is the Noto Canyon, which was carved into the Malta escarpment^{15,16}. Upslope, this canyon comprises a 400 m 57 deep and 4 km wide erosive channel, with Pliocene-Quaternary sediment infill¹⁵. At the Noto 58 Canyon outlet, seismic stratigraphy in the western Ionian basin reveals a buried chaotic 59 sediment body that extends over an 11,000 km² area, and reaches a 1,430–1,620 km³ 60 volume^{15,16}. This unit has been interpreted as a megaflood deposit of material that was 61 eroded, transported, and deposited during a catastrophic flooding event at the 62 Miocene/Pliocene transition^{15,16}. 63

The Miocene/Pliocene boundary was drilled at several Deep Sea Drilling Project (DSDP) and 64 Ocean Drilling Program (ODP) sites in the deep Mediterranean¹⁸. It is characterized by a sharp 65 66 lithological change, which has been attributed to an abrupt return to open-marine conditions across the Mediterranean, terminating the MSC^{16,18,19}. However, where continuous records 67 across the boundary are available, a major difference can be seen between Mediterranean 68 sub-basins (Supplementary Table 1, Extended Data Fig. 1). In the wMed, complete records 69 across the M/P transition from the Tyrrhenian Sea, Balearic margin, and Alboran Sea 70 (Supplementary Table 1) reveal earliest Pliocene foraminifer-nannofossil oozes over Late 71 72 Messinian evaporative sequences. This sequence suggests that normal marine conditions 73 were established rapidly in the wMed following flooding (Supplementary Table 1). In contrast, 74 eMed cores indicate deposition of a thick organic-rich layer (sapropel) in the earliest Pliocene, 75 which was first detected in DSDP Site 376 from the Florence Rise and named the "mystery sapropel"²⁰. ODP Site 969 on the Mediterranean Ridge contains a similarly thick, laminated 76 77 sapropel immediately above the flooding surface (Supplementary Table 1). Absence of 78 benthic foraminifera in this sapropel indicates a lack of eMed deep-water ventilation/ oxygenation in the immediate flood aftermath, in contrast to better wMed ventilation²⁰. 79

80 More evidence for the mystery sapropel

At eMed ODP Site 967 (Eratosthenes seamount), late Messinian brecciated carbonates (with 81 82 gypsum²¹) are overlain by dark grey to olive green earliest Pliocene sediments^{21,22}. 83 Throughout the sediments younger than ~3.2 Ma at Site 967, colour reflectance records and oxygen isotope stratigraphy indicate a regular pattern of sapropel occurrence, visibly 84 recognizable as dark layers^{21,23,24}. Between 3.2 Ma and 5.33 Ma, there are no visible sapropels, 85 but there are red intervals resulting from post-depositional sapropel oxidation^{21,23,24} 86 (Extended Data Fig.2). These red intervals contain similar Ba enrichments as sapropels, which 87 are associated with organic matter burial and remained present even after post-depositional 88 oxygenation^{25,26}; thus, Ba enrichment is a reliable proxy for original sapropel extents, and for 89 detecting sapropels that were initially present but that were later oxidized^{25–28}. Throughout 90 the last ~14 Myr, sapropel deposition (and associated Ba peaks) consistently occurred during 91 92 high-amplitude precession-driven insolation maxima, where the amplitude is modulated by orbital eccentricity maxima^{29,30}. 93

94 Here we present core-scanning X-ray fluorescence (XRF) and magnetic data across the M/P boundary at ODP Site 967 (Supplementary Information) that corroborate the deposition of an 95 96 organic-rich layer immediately following the M/P transition (Figure 1). From our results, this 97 layer comprises two Ba peaks with lower (but still substantially elevated) Ba levels in between 98 (Figure 1). This Ba pattern is mirrored by two Ti/Al minima with an intervening maximum (Figure 1). In the eMed sapropel stratigraphy, these mirrored fluctuations are typical of two 99 insolation maxima with an intervening minimum^{31–33}. Our chronology suggests that this 100 organic-rich sediment deposition persisted over a 26,000 year period (Extended Data Fig. 3, 101 102 Supplementary Table 2). Profiles of redox-sensitive elements and anhysteretic remanent magnetization (ARM) across the organic-rich layer (Figure 1) are also typical of 103 sapropels^{25,26,28}. This 26,000-year interval represents the only Neogene example of a sapropel 104 105 that extends through an insolation minimum; it breaks the well-understood relationship 106 between sapropel deposition and African monsoon maxima associated with northern hemisphere insolation maxima^{29,34–37}. In this conventional mode, monsoon maxima caused 107 108 extensive freshwater flooding into the Mediterranean^{36,38–40}, which drove both enhanced stratification (curtailing new deep-water ventilation/oxygenation) and enhanced organic 109 110 export production^{29,41,42}. Meanwhile, the monsoon maximum suppressed wind transport of Ti-rich dust to the eMed^{26,31,43}. Extension of the sapropel at the M/P boundary across an 111 African monsoon minimum (insolation minimum) is a clear indication of the operation of an 112 additional mechanism that was unique to this event with respect to the entire Neogene. 113

114 Establishment of normal marine conditions

The terminal Messinian was a period of surface dilution that resulted from increased 115 Paratethyan and riverine freshwater input (Lago Mare events), punctuated by local gypsum 116 deposition during times of enhanced evaporation^{32,44,45}. In our main scenario, deep 117 Mediterranean basins prior to the Zanclean flooding were filled with residual high-salinity 118 brines that in places exceeded 2 km in thickness^{9,32,44}. Here, we consider 'Late Messinian 119 brines' to have been derived from MSC Stage 2 fluid dilution (when halite precipitated in the 120 wMed and eMed⁴⁵), including potential contributions from halite re-dissolution. We set the 121 brine concentration to 140 PSU (see Methods for explanation on choice of brine salinity). A 122 qualitative conceptual refill scenario can then be formulated based on energy- and mass-123 124 balance arguments (the alternative hypothesis starting with a largely refilled late-Messinian

basin¹²⁻¹⁴ is addressed in a sensitivity test; see Methods). In the main scenario, the high-125 energy Atlantic floodwater cascading into a partially desiccated Mediterranean would have 126 encountered and vigorously mixed with residual wMed brines, while the wMed filled to the 127 128 height of the sill in the Strait of Sicily (Extended Data Fig. 4). Mixed wMed brine would then 129 have broken through and cut the Noto Canyon, cascading into the eMed. Meanwhile, wMed 130 sea level would have remained several hundred metres below Atlantic sea level, which continued the Atlantic cascade through the Zanclean channel, mixing with wMed brines. Thus, 131 we expect massive salt transfer from the steadily diluting wMed into the filling eMed. Once 132 133 eMed and wMed sea levels equalized, the level across both basins would have risen in unison, 134 until it equalized with Atlantic sea level across the Strait of Gibraltar. Approximately balanced 135 Atlantic inflow and Mediterranean outflow then drove gradual excess salt removal from the Mediterranean. At this stage, stratification was much less pronounced in the wMed than in 136 137 the eMed because of the prior wMed salt transfer into the eMed (as the latter filled). 138 Inhibition of deep-water ventilation during this hyper-stratified brine-filled eMed period— 139 accentuated by monsoon flooding during insolation maxima-may then explain the anomalously long duration of the "mystery sapropel" spanning two insolation maxima and an 140 141 intervening minimum.

142 Our qualitative concept requires quantitative assessment, especially to evaluate whether the 143 hypothesized mixing processes are realistic energetically and whether (and why) eMed stratification persisted for ~26,000 years, as derived from our XRF-based sapropel 144 stratigraphy and insolation tuning (see Methods, Extended Data Fig. 3). For this purpose, we 145 present a brine evolution model for the flooding event and its aftermath. Our model considers 146 post-Messinian Mediterranean basin evolution in two successive phases: (i) the Flooding 147 Phase and (ii) an Evolving Phase. The switch between phases occurred when Mediterranean 148 sea level matched Atlantic sea level across the Strait of Gibraltar. 149

The flooding phase is characterized by release of enormous gravitational potential energy. This potential energy is converted to kinetic energy as Atlantic floodwaters cascade into the wMed, which we estimate may have exceeded 1.6×10^{19} J per day at peak flood (Supplementary Figure 1), or more than 500 times the kinetic energy dissipation at Niagara Falls in one year⁴⁶. This energy is sufficient to mix (most of) the existing wMed brines with Atlantic inflow, forming a deep mixed layer atop potential residual brine (Figure 2, see 156 Methods). As the rising wMed sea level reached the crest of the Sicily Sill, mixed wMed waters started cascading into the eMed via Noto Canyon. Given a smaller channel cross-section in 157 Noto Canyon than in the Zanclean channel, we find that an even more energetic flow entered 158 159 the eMed than the wMed (Supplementary Figure 1). Note that our method calculates the minimum flow, restricted only by channel dimensions, and that more energy would have been 160 available in reality because of the steep drop in this passage^{15,16}. A massive amount of wMed 161 mixed brine was transferred into the eMed through Noto Canyon because the eMed volume 162 is ~2.5 times greater than the wMed, while—at the same time—Atlantic inflow through the 163 164 Zanclean channel continued to dilute the wMed mixed layer. We find that >95% of the wMed 165 excess salt ended up in the eMed, as this basin filled (Figure 2). Once eMed sea level reached 166 the Sicily Sill, energy transfer across the sill diminished as the cascade terminated gradually. 167 Later, while both basin levels rose together to the Atlantic level, enhanced mixing occurred 168 only in the vicinity of the Gibraltar Sill, further diluting the western basin (Figure 2, Extended 169 Data Figs. 4, 5).

At the flooding phase conclusion, dense (post-flood) brines had filled the eMed to the Sicily 170 Sill. For the Mediterranean to return to normal marine conditions, the salt in this brine must 171 have been transferred out of the basin, into the Atlantic Ocean. Eroding the deep brine layer 172 required mixing across the interface separating the brine from shallower, inflow-dominated 173 174 lower salinity waters. This mixing would have been governed by similar processes to those 175 operating in the modern global ocean: diapycnal mixing due to wind forcing, tidal interaction with bathymetry, and internal wave breaking⁴⁷. These processes lead to estimated diapycnal 176 diffusivity values within a 1-5 \times 10⁻⁵ m²s⁻¹ range⁴⁸; we use this range to estimate mixing 177 timescales and their uncertainties. The wide uncertainty range used here is derived from the 178 179 competing effects of mixing inhibition owing to stronger stratification and enhanced mixing within a smaller basin (with greater boundary interactions). 180

We model the evolving phase using diapycnal diffusivity in the stated range to remove salt by mixing. We obtain timescales of 10,000-40,000 years (Figure 3b). The ~26,000-year duration estimated from our proxy data agrees well with this range and corresponds to a diapycnal diffusivity of 2×10^{-5} m²s⁻¹. We calculate that this process would have released more than $7 \times$ 10^{16} kg of excess salt into the Atlantic Ocean within that time period, via Mediterranean outflow. Eventually, salt removal from the deeper eMed reduced stratification sufficiently to allow winter-cooled surface waters to attain densities conducive to new deep-water formation. This facilitated restart of deep-water ventilation and oxygenation, which ended the evolving phase marked by the "mystery sapropel." Downward oxidation of reduced sapropel sediments under an oxygenated water column caused a "burn down" oxidation front in the upper 35 cm of the sapropel^{26–28,49,50} (Figure 1).

192 We propose that absence of a wMed sapropel following the flooding resulted from wMed 193 brine transfer to the eMed during a high-energy mixing and refilling episode. Our sensitivity 194 test evaluates an alternative ending to the MSC, starting with a deep water column of residual brine overlain by lower-salinity brackish waters that had refilled the basin before Atlantic 195 reconnection^{12–14}. We find that there is insufficient energy in this scenario to remove brine 196 197 from the wMed (Extended Data Figs. 6-8, see Methods), and a long phase of anoxic (sapropel) 198 deposition would be expected in both the wMed and eMed, which conflicts with available observations (Supplementary Table 1). This offers strong support for a partially desiccated 199 200 Mediterranean state prior to Atlantic reconnection. To account for potential post-Messinian tectonic movements which may have affected the Sicily sill depth^{9,14}, and to test for the 201 minimum wMed and eMed base level drops below Atlantic level required to validate our 202 203 hypothesis, we perform additional sensitivity tests (Methods). Results of the analysis support 204 available observations, further strengthening our hypothesis (Extended Data Figs. 9, 10).

205 We find that only a kilometer-scale base level fall in both basins at the terminal Messinian 206 would have resulted in the observed proxy records and modelling outcomes. We conclude 207 that the transition from a partially desiccated Mediterranean basin to normal marine conditions was much less rapid than basin refilling; the full transition took ~26,000 years, 208 209 whereas flooding/refilling took only ~2 years (Figures 2, 3). The ~26,000-year timescale is corroborated quantitatively by our model, which suggests that a 10,000-40,000-year duration 210 211 is expected. Throughout this time, hyper-stratified eastern Mediterranean conditions caused persistence of an anomalously long interval of organic-rich sediment deposition that 212 213 extended through two precession-related insolation maxima and the intervening minimum.

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Author contributions

U.A. designed and led the study, and wrote the paper; U.A. developed the hypothesis and performed the modelling, with guidance from A.M.H. and E.J.R.; E.J.R., A.M.H., A.P.R., K.M.G. and D.H. contributed to data interpretation; K.M.G. calibrated scanning XRF data; S.G. performed WD-XRF analyses; X.Z. and P.H. assisted with magnetic measurements; D.L. performed XRF core-scanning; D.L. and T.W. developed the ODP967 composite depth splice; all authors contributed to manuscript development.

Competing interests

The authors declare no competing interests.

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Figure 1| XRF core scanning and magnetic data across the Miocene/Pliocene boundary from ODP 218 219 Site 967. The Miocene/Pliocene transition is tuned to the 65°N insolation maximum at 5.333 Ma (see 220 Extended Data Fig. 3). Return to normal marine conditions is marked by a sudden jump in Ca 221 concentration at around 5.307 Ma. Elevated Ba concentrations indicate organic matter preservation 222 between the first two Pliocene insolation maxima (Mystery sapropel). The intervening insolation 223 minimum records more than twice as high Ba concentrations compared to normal marine (NM) 224 conditions. (For Ba and Ti/Al, dotted lines in the background indicate original XRF data. Thick lines are 225 the 5-point moving average of the two records). The upper portion of the mystery sapropel has been 226 oxidized as indicated by Mn, S, and ARM profiles (Ox). Me, Messinian; FP, Flooding Phase (Zanclean 227 flooding surface); EP, Evolving Phase.

228 Figure 2 | Evolution of Mediterranean sub-basins during the Zanclean flood. a, Sketches of the basin 229 configuration during the latest Messinian (left) and Flooding Phase (right). A = Camarinal Sill; B = Sicily 230 Sill; Z_{wMed}/Z_{eMed} = Messinian drawdown in wMed/eMed; Z_{Sicily} = Depth to Sicily Sill; Z_{Atlantic} = Atlantic Sea 231 surface. b, Mixed layer evolution with time for western and eastern basins. Thick black line indicates 232 sea-level rise in both basins with time (wMed and eMed levels). Coloured, hatched, and dotted 233 backgrounds indicate the mixing extent at mixing efficiency (ME) = 0.2, 0.1 and 0.3, respectively. The 234 difference between the basin level curve and the mixed-layer curve gives the mixed layer thickness at 235 a given time. c, Reconstructed wMed and eMed hypsometry (left); salinity profile evolution (right) for 236 wMed and eMed at each 100-day interval. Salinity profiles (thick black lines) are given in PSU (from 0 237 to 140 in each diagram). ZFS 1 terminates as the wMed level reaches the Sicily Sill; ZFS 2 terminates 238 as eMed level reaches Sicily Sill; during ZFS 3 both basins rise to the Atlantic level simultaneously. ZFS, 239 Zanclean flood stage; AL, Atlantic Level; SL, Sicily Sill Level; WM, Western Mediterranean; EM, Eastern 240 Mediterranean.

241 Figure 3 | Mediterranean evolution during the Evolving Phase (EP). a, A sketch of the main processes involved in brine removal from eMed during the EP. Here, the eMed brine layer formed at the end of 242 the Flooding Phase, as a result of wMed salt transfer and mixing with eMed residual Messinian brines. 243 244 This layer should not be confused with 'residual Messinian brines' that existed before the flood. b, 245 Bottom salinity evolution with time. The thick line is the salinity evolution for diapycnal diffusivity K_s 246 = $2 \times 10^{-5} \text{m}^2 \text{s}^{-1}$. Upper and lower envelopes mark the evolution at diffusivities of $1 \times 10^{-5} \text{m}^2 \text{s}^{-1}$ and $5 \times 10^{-5} m^2 s^{-1}$, respectively. t_{k1} and t_{k5} are the durations taken to erode the brine at $1 \times 10^{-5} m^2 s^{-1}$ and 247 248 $5 \times 10^{-5} m^2 s^{-1}$, respectively. The time window between t_{k1} and t_{k5} demarcates all possible time frames 249 within mentioned diffusivity values. t_E is the duration recorded by proxy data (26,000 years). S_{LW} , 250 Levantine intermediate water salinity. c, Evolution of eMed salinity profile with time for K_s = 251 $2 \times 10^{-5} \text{m}^2 \text{s}^{-1}$. Numbers on each curve represent time evolved in thousand years (1 = 1,000 years, etc.)

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380 METHODS

381 Bulk sediment geochemistry

We present X-ray fluorescence (XRF) core-scanner data from ODP Site 967 for the 5.12-5.35 382 Ma interval. Scanning was performed on archive core-sections at MARUM-University of 383 384 Bremen on an Avaatech XRF core scanner. Core sections were covered with 4 mm-thick Ultralene film and measured at 50 and 30 kV with 0.55 mA current and with a Cu and Pd-thick 385 filter, respectively, and at 10 kV with 0.035 mA current (no filter); count time for all runs was 386 7 s. Element 'counts' for the entire interval were converted into element concentrations by 387 multivariate log-ratio calibration⁵¹, using new wavelength dispersive (WD)-XRF reference 388 element concentrations. For these, 38 bulk sediment samples were chosen to cover a range 389 390 of lithologies based on the XRF scan, then 1 cm³ dried ground sample was mixed with a lithium 391 tetraborate/lithium metaborate flux and fused into 39 mm diameter beads. Major element abundances were analysed by WD-XRF using a Bruker S8 Tiger™ spectrometer at Geoscience 392 Australia. Loss on ignition (LOI) was measured by gravimetry after combustion at 1000 °C. One 393 394 in every ten samples was duplicated along with multiple analyses of 3 international standards (NCS DC70306, MAG-1, ML-2) and an internal basalt standard (WG1). Quantification limits for 395 all major element oxides are <0.2% and reproducibility is within 1%. 396

397 Anhysteretic remanent magnetization (ARM)

398 ODP967 U-channel samples were sliced at 1-cm intervals into discrete non-magnetic 2×2×2 399 cm plastic cubes and measured for ARM on a 2-G Enterprises cryogenic magnetometer at the 400 Australian National University. The ARM was imparted using an alternating field of 100 mT 401 and a direct current bias field of 0.05 mT.

402 Mediterranean evolution

Evaporative Mediterranean drawdown during the Messinian left residual brines in the adjacent eMed and wMed basins, with thicknesses reaching more than 2 km in the deepest parts^{9,32,44}. We assume a water level drop of 2,000 m below Atlantic sea level in the eastern basin, and 1,750 m in the western basin prior to the megaflood^{5,8–10}. To relate Mediterranean refilling to the volume flux of the incoming flood, we use an intermediate reconstruction between Miocene and present-day Mediterranean hypsometry¹⁰. Before the flood, halite precipitation occurred at ~5.6 Ma in both basins at the Messinian Salinity Crisis (MSC) peak.
The final MSC stage contains gypsum deposition punctuated by surface-water freshening
events. A few drilling sites record gypsum below the flooding surface (Supplementary Table
1). Considering these observations, we set Messinian residual brine salinity to gypsum
saturation at the start of the flood (140 PSU⁵²). We test model outputs for a range of starting
salinities (60-240 PSU) in a sensitivity test (Extended Data Figs. 9, 10).

415 The Flooding Phase

To determine the flood velocity entering the western basin from the Strait of Gibraltar, we use a flood incision model, following Garcia-Castellanos et al. $(2009)^3$. This model computes the incision rate (dz_s/dt) at Camarinal Sill based on the following approach, where *dt* is the timestep and *dz*_s is the depth of erosion per time step:

420
$$\frac{dz_s}{dt} = k_b (\tau_b)^a, \tag{1}$$

421 where k_b and a = 1.5 are positive constants. To obtain a final sill incision depth (240 m^{ref. 3}), 422 we calibrate k_b to 1.8×10^{-4} m yr⁻¹ Pa^{-a}, where a is the constant mentioned above, while τ_b is 423 the basal shear stress at the sill, which is computed using:

424
$$\tau_b = \rho_s g(z_s - z_0)S, \tag{2}$$

425 where ρ_s is seawater density, (z_s-z_0) is the mean water depth at the sill, g is acceleration due 426 to gravity, and $S = z_h/L$ is the ratio between head loss (z_h) and length of the erosive channel 427 (L). Manning's formula³ is used to calculate the flow velocity:

428
$$v = \frac{1}{n} R_h^{\frac{2}{3}} S^{\frac{1}{2}},$$
 (3)

where *v* is the average flow velocity along the slope toward the Alboran basin, n = 0.05 is the roughness coefficient³ and R_h is the hydraulic radius of the passage between the Atlantic and Mediterranean³. Where channel width is substantially greater than the water depth, the hydraulic radius is approximated by (z_s - z_0). Flood discharge flux into the Mediterranean (Q) is then calculated using:

$$Q = W(z_s - z_0)v, \tag{4}$$

where $W = k_w Q^{a_w}$ is the incision channel width, which increases to a final 14 km value at the end of the flood. Here $a_w = 0.5$ is a constant, and we impose $k_w = 1.1$ to obtain the final channel width (see Methods in Garcia-Castellanos et al. (2009) for details³).

438 This model predicts a peak discharge flux that exceeds 100 Sv at a velocity > 40 ms⁻¹ at the Camarinal Sill³. In the initial flood stage, high-energy normal Atlantic seawater inflow 439 440 encounters much denser residual wMed brines. The extent of mixing between brine and flood waters depends on the kinetic energy of the flow that approaches the brine surface. Without 441 442 mixing, a seawater layer (ρ_s) would form on top of the denser brine (ρ_b). In contrast, if complete seawater mixing occurred with the brine, a single intermediate density layer (ρ_m) 443 would have formed. Mixing due to turbulent erosion of stratification can be accomplished 444 445 with a large enough energy to overcome the potential energy, raising denser fluid parcels while lowering lighter parcels⁵³. 446

If complete mixing occurred in the basin, the centre of gravity of the system must be raised,resulting in a potential energy gain given by:

449
$$PE_{Final} - PE_{Initial} = PE_{Gain} = \int_0^{H+h} \rho_m gz dz A(z) - \left(\int_H^{H+h} \rho_s gz dz A(z) + \int_0^H \rho_b gz dz A(z)\right),$$
(5)

450 where PE_{Gain} represents the energy (per day) required for complete mixing, A(z) is the basin 451 surface area with depth (z), h is the thickness of the seawater layer flooded into the basin per 452 timestep (per day), and H is the total brine column thickness prior to mixing.

453

454 Under the assumption of the Garcia-Castellanos et al. $(2009)^3$ model that the slope of Atlantic 455 water inflow into the wMed is constant over time, the flow velocity entering the brine surface 456 in the western basin, v_{wMed} is v. The kinetic energy per day of this inflow (KE_{wMed}) is then 457 calculated using:

458

$$KE_{wMed} = \frac{1}{2}\rho_s v_{wMed}{}^2 Q. \tag{6}$$

459

In our model, we calculate energy conversions at daily iterations, by integrating the energy expression in equation 6 over one-day intervals. Available potential energy of inflow into the wMed (at the sill), APE_{wMed} , is equal to $\rho_s gQL_{WMed}$, where L_{wMed} is the vertical distance between the brine surface and the Camarinal Sill crest. As the Atlantic inflow descended down-slope, gravitational potential energy was released to kinetic energy, and was lost in
processes such as Zanclean channel erosion and turbulent dissipation. Erosion along the slope
will have resulted in a headward-migrating erosion wave along the channel that may have
then triggered even higher flow and erosion rates, resulting in a more abrupt flood⁵⁴.
Therefore, our calculations offer a minimum estimate of flow kinetic energy into the wMed.

470 Most of the inflow kinetic energy into the basin is lost to viscous turbulent dissipation. 471 Laboratory experiments in stratified fluids suggest that ~20% of the turbulent energy loss goes 472 to irreversibly mixing the stratification (known as the mixing efficiency)^{55,56}. Thus, the energy 473 available for basin mixing (KE_{Av}) can be given as:

474 $KE_{Av} = \Gamma . KE_{wMed},$

where $\Gamma \approx 0.2$ is the mixing efficiency. Full-depth mixing can occur only if $KE_{Av} > PE_{Gain}$. If available kinetic energy is less than that required to mix the entire brine column thickness (*H*), partial brine mixing will occur. The mixing extent can then be computed using energy arguments. For this purpose, we introduce a mixing depth (H_{Mix}). Where mixing is incomplete, equation 1 can be rewritten as:

480
$$KE_{Av} = PE_{GPar} = \int_{H+h-H_{Mix}}^{H+h} \rho_m gz dz A(z) - \left(\int_{H}^{H+h} \rho_s gz dz A(z) + \int_{H+h-H_{Mix}}^{H} \rho_b gz dz A(z)\right),$$
 (8)

481 where PE_{GPar} is the potential energy gain during partial mixing. Hence, if available energy is 482 insufficient to mix completely, mixing will be restricted to a brine depth of H_{Mix} . If the flow 483 kinetic energy increases with time, H_{Mix} will increase accordingly, eventually completely 484 mixing the brine when KE_{Av} exceeds PE_{Gain} .

We use the following approach to calculate the salinity evolution (S_m) of the mixed fluid:

$$S_m = \frac{QS_{AW} + Q_bS_b}{Q_m} = S_{wMed}, \tag{9}$$

487 where Q_b is the brine volume (per day) mixed in the basin, S_b is the brine salinity, and Q_m is 488 the resultant volume of mixed fluid (per day). For salinity-density conversions, we include the 489 Gibbs Sea Water (GSW) Oceanographic Toolbox functions into our model.

490 Once wMed sea level reaches the Sicily Sill (430 m below sea level), wMed mixed fluid spills 491 into the eMed. We use equation (8) to compute the mixing extent in the eastern basin as it

(7)

fills. Noto Canyon is considered to be the path of flood waters into the Ionian basin (eMed) during the megaflood¹⁵. To approximate the flow velocity approaching the eMed brine surface, we consider that the volume flux spreads across a channel of width b_{eMed} , in a fluid layer of depth d_{eMed} travelling at velocity v_{eMed} . The erosive channel detected at the upper Noto Canyon is 4 km wide; thus, we impose b_{eMed} to be a 4 km maximum. The present average channel depth is 400 m^{ref 15,16} (d_{eMed}). Minimum flow velocity into the eMed is then given by:

498

$$v_{eMed} = \frac{Q}{A_{Noto}},\tag{10}$$

where A_{Noto} is the present-day maximum cross-sectional area of the Noto Canyon erosive channel. In reality, flow velocity entering the eMed would have been higher due to the steep slope toward the end of the passage^{15,16}. The kinetic energy of flow entering the brine is:

502

 $KE_{eMed} = \frac{1}{2}\rho_s(v_{eMed}^2)Q.$ (11)

503

Similar to the wMed approach, complete mixing is assumed if $1 > (\Gamma KE_{eMed})/PE_{Gain}$. As the eMed 504 505 is filled, wMed waters mix continuously due to inflow through the Strait of Gibraltar. Equation 506 (9) is used to calculate salinity evolution in both basins during this flood stage (S_{wMed} and S_{eMed} , 507 respectively). The water flow cascading into the Ionian basin (eMed) terminates once the 508 eMed level reaches the Sicily Sill level. Thereafter, both eMed and wMed levels rise 509 simultaneously to the Atlantic level. In this final flood stage, mixing occurs due to flow along 510 the Zanclean channel (Strait of Gibraltar), further diluting the western basin (thus, further 511 decreasing S_{wMed}).

512 The Evolving Phase

513 During the first part of our model (*Flooding Phase*), most wMed salt was transferred into the 514 eMed across the Strait of Sicily. As a result, the wMed had less saline waters, whereas the 515 eMed filled with a denser brine layer up to the Sicily Sill (the reconstructed early Pliocene 516 eMed volume with sea level equal to the Sicily Sill level is 2.04 x 10¹⁵ m^{3 ref. 10}). For the 517 Mediterranean to return to normal marine conditions, this enormous amount of brine should 518 then have been redirected to the Atlantic Ocean.

The processes required to erode this deep, dense brine layer would have involved mixing across the interface separating it from inflowing waters. This mixing would occur due to the same set of processes that operate in the modern global ocean; namely, diapycnal mixing due

to internal wave breaking^{47,57}. Generation of internal gravity waves results from a chain of 522 processes including barotropic tidal flow over topography, variations in wind force at the 523 524 ocean surface, lee waves produced by ocean currents, and eddies flowing over topography^{47,57}. The sum of all turbulent processes leads to an effective turbulent diffusivity 525 of $K_S = 1-5 \times 10^{-5} \text{ m}^2 \text{s}^{-1 \text{ refs. } 48,57}$. By assuming a turbulent diffusivity in this range, we estimate 526 the duration required to erode the deep layer. The salinity profile immediately after the 527 flooding event consists of an upper Mediterranean seawater layer, and a deep brine layer 528 below the sill depth. Mixing and advection is rapid in the x and y directions (horizontally); 529 530 thus, a good approximation for salinity, S, at any time is that it depends on height, z, only. The 531 diffusive timescale, *t*, can be estimated from the diffusion equation:

532
$$\frac{\partial S}{\partial t} = K_S \frac{\partial^2 S}{\partial z^2}.$$
 (12)

To solve Eq. (12) numerically, we divide the domain into N equally spaced layers, each having
thickness d_z. Basin hypsometry is characterized by the surface area (A_z) at each depth interval.
The salt flux between adjacent layers is proportional to the local salinity gradient given by:

536
$$F \sim K_s \frac{\partial s}{\partial z}$$
 (13)

537 Evolution of salinity in the *j*th layer of the domain is given by the flux divergence:

538
$$A^{j} \frac{\partial S^{j}}{\partial t} = \frac{K_{s}}{d_{z}} \left(A^{j+1} \left[\frac{S^{j+1} - S^{j}}{d_{z}} \right] - A^{j} \left[\frac{S^{j} - S^{j-1}}{d_{z}} \right] \right).$$
(14)

539 When this equation is further discretized in time, it can be shown that:

540
$$S_{\tau+1}^{j} = S_{\tau-1}^{j} + \frac{2K_{s}d_{t}}{d_{z}^{2}} \left(S_{\tau}^{j-1} - \left[\frac{A^{j+1}}{A^{j}} + 1\right]S_{\tau}^{j} + \left[\frac{A^{j+1}}{A^{j}}\right]S_{\tau}^{j+1}\right), \tag{15}$$

where d_t is the time step and time levels are indicated by the subscript (τ). With use of these equations, we compute the salinity profile evolution until the basin reached modern eMed salinity values. Diffusive transport of salt from deeper layers to upper eastern basin layers will be balanced by salt transport to the wMed, and ultimately into the Atlantic Ocean via the Mediterranean outflow.

546

547 Sensitivity test Part 1: Basin evolution for a largely refilled Mediterranean (alternative 548 hypothesis)

The nature of the MSC termination has long been debated. One hypothesis is a catastrophic termination (the main scenario of this paper) that resulted from Camarinal Sill collapse in the Strait of Gibraltar^{3,11,15,16}. This hypothesis considers a km-scale drawdown of water in Mediterranean sub-basins prior to the reconnection. An alternative hypothesis interprets an almost-filled Mediterranean during the final MSC stage (Lago Mare), which was connected to the Atlantic and/or Paratethys prior to the termination^{12,14,58}.

Here we present a sensitivity test to evaluate the validity of our hypothesis (a partiallydesiccated Mediterranean leading to catastrophic termination), compared to a scenario where the basin is filled to the Sicily Sill before the termination. We argue that the sapropel presence immediately after the Miocene/Pliocene boundary in eMed cores (Supplementary Table 1) resulted from basin stratification after the Zanclean megaflood. In contrast, no wMed sapropels were found following the M/P boundary. We suggest that absence of a wMed sapropel resulted from brine transfer to the eMed during an abrupt refilling event.

If the present-day Black and Caspian Seas were completely emptied into the Mediterranean, 562 this would result in a ~250 m thick layer at the Mediterranean surface¹³. This gives a high-end 563 564 estimate of Paratethyan inflow to the Mediterranean if the two basins were connected during the Lago Mare phase. It has been suggested that gypsum precipitation resulted from 565 566 evaporated Paratethyan surface waters (above denser residual brines) at a salinity of ~40 PSU⁵⁸. Therefore, we simulate emptying of a volume equivalent to the present-day Black and 567 Caspian Seas into the Mediterranean and allow it to evaporate to a salinity of 40 PSU. If the 568 top of such a layer sits at the crest of the Sicily Sill (430 m), it will be about 130 m in thickness 569 (calculated using a reconstructed Mediterranean hypsometry¹⁰), and will extend below the 570 sill crest to a depth of 560 m. 571

572 Numerical modelling has shown that halite precipitation during the MSC peak (Stage 2, 5.59-573 5.55 Ma) resulted from complete disconnection from the Atlantic⁵⁹. Complete isolation will 574 result in basin drawdown due to excess evaporation that will come to equilibrium within a 575 few thousand years (3 to 8 kyr)^{9,10}. The idea of a km-scale drawdown is also supported by the 576 existence of a basin-wide Messinian Erosional Surface (MES)⁵ and deep canyons excavated 577 beneath the Nile delta and Rhône River mouth^{6,7}. Similar to our main scenario, we assume a 578 wMed deep brine layer at gypsum saturation (140 PSU) below 1750 m, which formed as a 579 result of MSC stage 2 drawdown. From 560 m to 1750 m, we allow salinity to increase linearly 580 from 40 to 140 PSU, to obtain a conservative estimate (Extended Data Fig. 6a).

581 Starting with this basin configuration, we compute wMed basin evolution (Extended Data Fig. 6b) during a refilling event resulting from inflow through the Strait of Gibraltar using the 582 Flooding Phase model. Here we re-calibrate the channel width coefficient (k_w) to allow the 583 584 inflow channel to evolve up to the same dimensions as in our main scenario. We then compare the refilling and basin evolution in both scenarios (Extended Data Fig. 6c). Our 585 586 findings suggest that for a filled Mediterranean up to the crest of the Sicily Sill, reconnection with the Atlantic results in a much longer episode of refilling. The total kinetic energy released 587 588 during such a scenario will be >50 times smaller compared to a catastrophic termination, which is insufficient to erode the deeper brine in the wMed and would result in persistence 589 590 of a density-stratified wMed (Extended Data Fig.6b, c). In that case, we argue that a sapropel should be expected in the wMed after Atlantic reconnection. Absence of a wMed sapropel 591 provides strong support for catastrophic MSC termination and for a drawn-down 592 Mediterranean prior to Atlantic reconnection. We also test this scenario for even lower 593 594 salinity profiles between a thin surface layer and deep brines (from 560 m to 1750 m), to 595 assess if the flood energy was sufficient to erode wMed deep brines, if the mid-layer salinity 596 was lower (Extended Data Fig. 7a, b). This test run confirms that the energy is insufficient to remove wMed deep brines even for a 100% mixing efficiency. 597

598 Sensitivity test Part 2: Basin evolution at initial salinities < 140 PSU (Main scenario)

In order to test the validity of our hypothesis in case the residual Messinian fluid salinity was lower than 140 PSU, we computed the salinity evolution of the wMed and eMed using the same approach in the Flooding Phase used for the main scenario. We use a range of salinities from 60 to 120 PSU. Then we use the Evolving Phase to determine the duration of brine removal at different initial salinities (Extended Data Fig. 7c, d). At salinities lower than 140 PSU, all wMed salt is transferred to the eMed, resulting in a salinity approximated by:

$$S_{eMed} = \frac{\left(V_{eMed(Sicily)} - V_{b(eMed)} - V_{b(wMed)}\right) \times S_{Atl} + \left(V_{b(eMed)} + V_{b(wMed)}\right) \times S_{b}}{V_{eMed(Sicily)}}$$
(16)

21

where S_{eMed} is the final eMed salinity below the Sicily Sill, $V_{eMed(Sicily)}$ is the total eMed volume at the Sicily Sill, $V_{b(eMed)}$ and $V_{b(wMed)}$ are the residual Messinian volumes in eMed and wMed, and S_b is the initial Mediterranean fluid salinity. For a range of initial residual Messinian brine salinities from 60 to 120 PSU, we find that the salt removal period varies between ~14,000 to 22,000 years.

611 Sensitivity test Part 3: Basin evolution at initial salinities > 140 PSU (Main scenario)

612 In order to test the validity of the hypothesis in case the residual Messinian salinities were > 140 PSU, we compute the salinity evolution of the wMed and eMed using Flooding Phase 613 614 calculations for 160-240 PSU. We employ the Evolving Mode approach to estimate salt removal duration for both basins, when excess salt remains at the end of the flooding phase. 615 616 If wMed stratification was less than the resulting winter water density, convective overturn 617 would mix surface and deep waters, removing the additional salt. In the present-day wMed, surface salinities reach 38.0-38.4 PSU in winter, with reduced temperatures of 10-12 °C^{ref.29}. 618 We assume a constant brine temperature of 20 °C during the flooding phase. Therefore, we 619 calculate the sea water salinity at 20 °C for an equivalent wMed winter surface water density. 620 We use this as a limit in determining the brine removal period in the evolving phase (Extended 621 Data Fig. 8). Results of the sensitivity test indicate that all wMed salt would transfer to the 622 eMed up to initial brine salinities of 170 PSU. From 170 to 200 PSU, final mixed wMed waters 623 624 have a higher-than-winter water density. Above 220 PSU, residual Messinian brines remain in 625 the deep wMed, implying that flood energy was insufficient to erode the deep brines. Above 170 PSU initial salinity, we find that it would take about 4,000 to 12,000 years to remove 626 wMed salt by diffusion. Combining the information from Sensitivity Test Parts 2 and 3, we 627 conclude that our model results agree reasonably with the data for starting salinities across 628 the 60-170 PSU range. 629

630 Sensitivity test Part 4: Basin evolution with the change of Sicily sill depth

To test the effect of Sicily sill depth on basin evolution during the flooding mode, we modified our model for shallower and deeper than present Sicily sill depth. For this, we employed ~0.7 and ~1.3 times the present sill depth (300 m and 560 m, respectively). We find that the flooding mode length reduces/increases when the Sicily sill depth is shallower/deeper than present. We find that independent of the sill depth, a majority of the wMed Messinian salt is transferred to the eMed across the sill (Extended Data Fig. 9). Our model suggests that as the sill gets shallower, there is slightly more salt remaining in the wMed, compared to the deeper sill setting. Detailed palaeomagnetic studies have shown that the Sicily sill may have been uplifted to its present depth during central to eMed wide middle-late Pliocene tectonic events^{24,60}. Modelling experiments have also suggested a deeper Sicily sill during the Messinian^{9,10,14}. Our model results would agree with these observations, as more wMed salt would be transferred to the eMed for a deeper sill.

643 Sensitivity test Part 5: Effect of the initial base level on basin evolution

We have assumed initial base levels for the wMed (1,750 m below Atlantic level) and the 644 eMed (2,000 m) for our main scenario, based on available references. In sensitivity test part 645 1 (alternative scenario), we tested basin evolution when the Mediterranean is filled up to the 646 647 Sicily sill. To test the effect of intermediate base levels, and to define a minimum base level 648 which validates the hypothesis, we computed basin evolution by stepwise increasing the base level in 100 m increments in both basins. For each step of base level increase above the main 649 scenario base levels, we filled both wMed and eMed with lower salinity waters following the 650 steps of sensitivity test part 1. From 1,750 m up to 1,450 m base level in the wMed (2,000 to 651 1,700 m in the eMed), all the residual Messinian salt is transferred from wMed to the eMed. 652 When the base level is increased above this point, excess salt tends to remain in the wMed. 653 654 We have shown test results up to 1,250 m wMed base level; corresponding to a 1,500 m eMed 655 base level (Extended Data Fig. 10). A period of 2,500 to 4,500 years is required to remove the excess salt by diffusion when the wMed level is above 1,450 m. 656

Data availability

ODP Site 967 data from this study are available from Panagea (www.pangaea.de) under 'Scanning XRF and environmental magnetic data across the Miocene-Pliocene boundary from ODP Site 967 (Eastern Mediterranean)' and are also available as online Supplementary Data accompanying this article.

Code availability

All the figures in this manuscript are reproducible via Jupyter notebooks and instructions provided in the Github repository Med_evolution_megaflood⁶¹ (<u>https://doi.org/10.5281/zenodo.6528768</u>).

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Additional information

Supplementary information is available for this manuscript









