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Neodymium isotopes of central Mediterranean phosphatic hardgrounds reveal Miocene paleoceanography

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ABSTRACT

Understanding the causes of the formation of hardgrounds provides insights on the oceanographic evolution of a basin. Phosphate-rich hardground formation interrupted carbonate ramp deposition in the Mediterranean during the Miocene. We analyzed the ε_{Nd} record of three central Mediterranean hardgrounds to identify the origin of the phosphate-rich waters that formed them within the frame of Mediterranean Miocene paleoceanographic evolution. The Nd isotopes suggest that eastern Mediterranean deep waters were controlled by runoff, in contrast to Atlantic and Indian Ocean waters. This Nd isotope record attests to the weakening of Mediterranean circulation during the Miocene due to closure of the Indian Gateway. Limited exchange with Atlantic shallow seawater led to long residence times for deep waters in the basin. This record indicates the role of upwelling in formation of phosphate hardgrounds and shows the influence of global climate change and local paleoceanographic conditions.

INTRODUCTION

Phosphate-rich hardgrounds frequently occur in Miocene Mediterranean carbonate successions and are commonly associated with drowning platforms and depositional hiatuses (Föllmi et al., 2008, 2015; Brandano et al., 2020). The interpretations of the causes of their formation, related overall to increased phosphorus (P) burial rates, span from circulation changes linked to deepening of the substrate, global climate evolution and upwelling, to increased humidity and "washhouse" events (Föllmi, 1996; Mutti and Bernoulli, 2003; Böhme et al., 2008; Filippelli, 2008; Föllmi et al., 2008, 2015). Increased P availability in surface waters coincides with enhanced primary productivity and carbon cycle perturbations testified by positive $\delta^{13}C$ isotope shifts in carbonate successions. Enhanced P availability is recorded (1) at the Oligocene-Miocene transition, when a glacial maximum led to increased weathering rates (Zachos et al., 2001); (2) during the Monterey event, when global warming at the Middle Miocene Climatic Optimum (MMCO) favored biogeochemical weathering (John et al., 2003; Brandano et al., 2017); and (3) in the Tortonian, when the Carbon Maximum 7 (CM7) perturbation was linked to enhanced oceanic circulation rates (Brandano et al., 2016a, 2020). The geodynamic evolution of the Mediterranean area affected its circulations patterns, making this basin sensitive to climate changes and lapped by upwelling of phosphate-rich deep waters, which controlled the development of hardgrounds. The identification of the water masses that formed these hardgrounds is crucial for reconstructing the paleoceanographic evolution of the basin, i.e., the change through time of the main currents influencing the primary productivity in the Mediterranean. However, published literature has overlooked the problem of the origin of these water masses, focusing mostly on the link between hardground formation and the global P cycle and the role of nutrients on carbonate platform evolution (e.g., Föllmi et al., 2008; Brandano et al. 2016a). Neodymium (Nd) isotopes are a reliable proxy of paleoceanographic changes (Scher and Martin, 2008). However, published Nd isotope records are based mostly on planktonic foraminifera, which do not record the deep-waters fingerprint but rather provide a mixed signal derived from surface, bottom, and pore waters (Pomiès et al., 2002; Kocsis et al., 2008; Cornacchia et al., 2018). We focus on $^{143}Nd/^{144}Nd$ in shark teeth from hardgrounds, a proxy for deep-water masses. As a rare earth element, Nd is incorporated into apatite at or near the sediment-water interface, making $^{143}Nd/^{144}Nd$ in shark teeth a proxy for seawater chemistry (Scher and Martin, 2008). This principle makes Nd isotopes the best proxy for targeting the provenance of the deep waters. For this study, shark teeth were chosen because apatite is rich in Nd, is resistant to diagenetic alteration, and is a reliable proxy for seawater chemistry (Kocsis et al., 2009). Thus, this work aims to reconstruct the provenance of deep-water masses that formed hardgrounds within three Miocene carbonate successions of the central Mediterranean, identifying the control of geodynamics on paleoceanography and carbonate platform drownings.

GEOLOGICAL SETTING

The Apennines (Italy) consist mainly of Meso-Cenozoic limestones, relicts of an archipelago of carbonate platforms separated by deep basins (Bernoulli, 2001). The investigated phosphatic hardgrounds mark different Miocene drowning events of the Apulian and the Latium-Abruzzi platforms (Fig. 1).

The first sampled hardground crops out in the Fonte del Papa quarry on Maiella Mountain (Apulian platform, Central Apennines) and is late Burdigalian in age (ca. 16 Ma; Brandano et al., 2016b; Fig. 1). The second hardground crops out on the Salento Peninsula near the Porto Badisco locality (Apulian platform, southern Italy) and is late Serravallian in age (ca. 12 Ma; Föllmi et al., 2015; Fig. 1). Lastly, the third hardground crops out in the Tornimparte village (Latium-Abruzzi platform, Central Apennines) and is early Tortonian in age (ca. 11 Ma;

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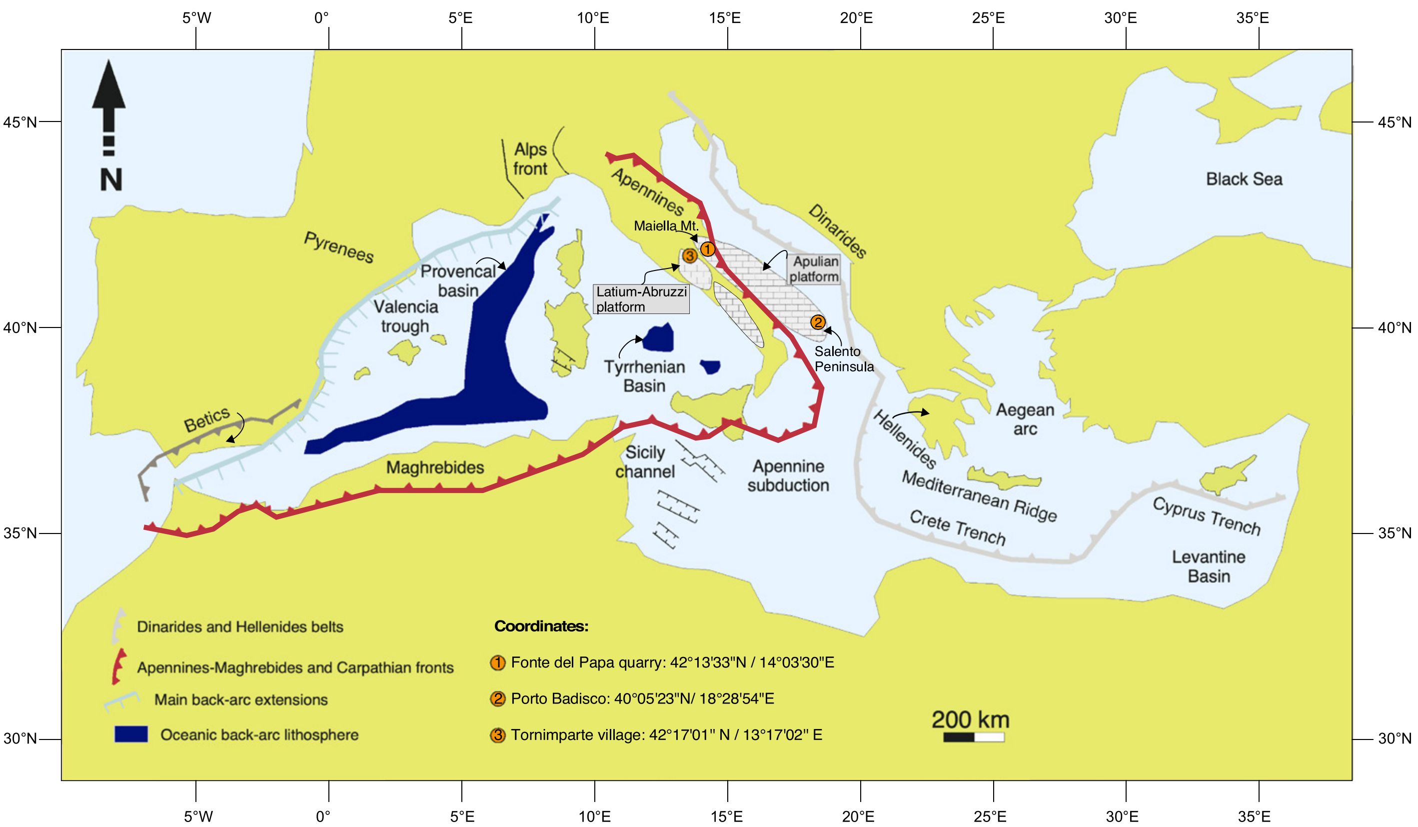


Figure 1. Mediterranean map with hardground locations. Modified from Agostini et al. (2010).

Brandano et al., 2020; Fig. 1). A detailed description of the stratigraphy of the carbonate successions to which these hardgrounds belong as well as log figures and outcrops photographs are provided in the Supplemental Material[1].

MATERIALS AND METHODS

We analyzed seven samples of shark teeth for $^{143}Nd/^{144}Nd$ ratios. $^{143}Nd/^{144}Nd$ ratios were measured with the Thermo Triton thermal ionization multicollector mass spectrometer of the Endogene Geodynamic Laboratory of the GeoZentrum Nordbayern (Friedrich-Alexander-Universität Erlangen-Nürnberg [FAU], Erlangen, Germany) after Nd purification.

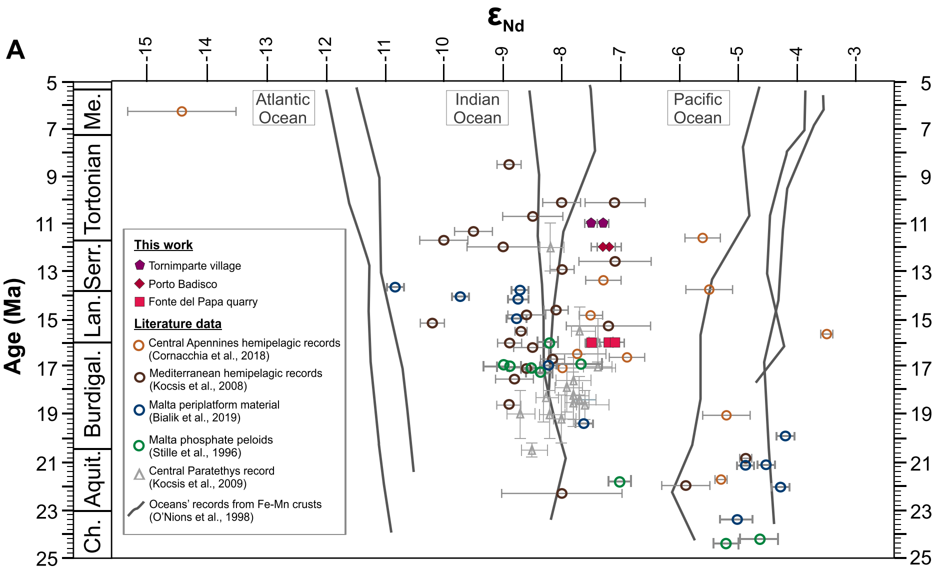
Neodymium isotope data were calibrated against the international La Jolla standard measured with the samples (Fig. 2). Values are corrected for the age of the samples assuming Sm/Nd ratio of fossils is the same as that of seawater (0.122; Piepgras and Wasserburg, 1980). Full details on the sample preparation, Nd purification, and applied corrections are available in the Supplemental Material.

[1]Supplemental Material. Detailed geological setting with logs of the stratigraphic sections to which the hardgrounds belong and outcrop photos (Figures S1–S4), methods, and a table of Nd isotope data. Please visit https://doi.org/10.1130/GEOL.S.19783126 to access the supplemental material, and contact editing@geosociety.org with any questions.

RESULTS AND DISCUSSION

The $^{143}Nd/^{144}Nd$ values are reported as ratios and in ε_{Nd} notation in Figure 2. They span from ε_{Nd} −7.5 to ε_{Nd} −7.1. Thus, not only are the $^{143}Nd/^{144}Nd$ ratios of the same hardground consistent, but the three hardgrounds show the same signature.

The ε_{Nd} values of the analyzed hardgrounds (ε_{Nd} −7.5 to −7.1) are similar to those of the deep Indian Ocean, central Paratethys, and North Alpine foreland basin records during the Miocene (Fig. 2A; O'Nions et al., 1998; Kocsis et al., 2009). This similarity is difficult to explain because the Indian Gateway closed in the Burdigalian (ca. 18 Ma) and reopened several times but remained very shallow until its definitive closure at the Langhian-Serravallian boundary (ca. 13.8 Ma; Fig. 3A; Popov et al., 2004). In this context, the Mediterranean ε_{Nd} remained similar to the Indian Ocean signature (Kocsis et al., 2008; Cornacchia et al., 2018; Bialik et al., 2019). In fact, Bialik et al. (2019), comparing the shallow water record of Malta and the Maldives, stated that the Indian Ocean connection was efficient during the Langhian. However, the modeling study of de la Vara and Meijer (2016) indicated that, after the Burdigalian (ca. 18 Ma), only the shallowest waters entered from the Indian Gateway, switching the Mediterranean circulation from estuarine to anti-estuarine. Cornacchia et al. (2018), analyzing the Umbria-Marche basinal succession, hypothesized that water exchanges between the Paratethys—characterized by an ε_{Nd} similar to that of the Indian Ocean—and the Mediterranean affected the Nd isotopes of the latter. Therefore, the ε_{Nd} of −7.5 and −7.1 of the uppermost Burdigalian (ca. 16 Ma) hardground on Maiella Mountain might be interpreted as a hint of efficient exchanges with the central Paratethys, which shows the same ε_{Nd} in that interval (Fig. 2A; Kocsis et al., 2009). However, the comparison of the Nd isotope records, besides proving water exchanges, does not identify the source of the P-rich waters, which might have developed in the North Alpine foreland basin and central Paratethys, as in the deep Mediterranean. Furthermore, according to the paleogeographic reconstructions of the Paratethys (Fig. 3A), only shallow-water exchanges with the Mediterranean occurred, thus potentially influencing the overall Nd isotope fingerprint of the eastern Mediterranean but not directly controlling the deep, P-rich waters forming the upwelling. Furthermore, the Maiella Mountain hardground is late Burdigalian in age (ca. 16 Ma), thus it developed during the MMCO when P availability was enhanced globally due to increased biogeochemical weathering (Föllmi et al., 2008). Lastly, the closure of the Indian Gateway weakened the overall Mediterranean circulation, potentially favoring the onset of deep-water oxygen-depleted zones. In turn, such oxygen-minimum zones favored the return of



B

Sample	Locality	Age (Ma)	$^{143}Nd/^{144}Nd$ (age corrected)	SE	$\varepsilon_{Nd(t)}$	±
TN-HG a	Tornimparte village	11	**0.512252**	0.000006	**-7.3**	0.1
TN-HG b	Tornimparte village	11	**0.512238**	0.000007	**-7.5**	0.1
S-HG b	Porto Badisco	12	**0.512248**	0.000011	**-7.3**	0.2
S-HG c	Porto Badisco	12	**0.512254**	0.000011	**-7.2**	0.2
M- HG a	Fonte del Papa quarry	16	**0.512233**	0.000007	**-7.5**	0.1
M- HG b	Fonte del Papa quarry	16	**0.512252**	0.000006	**-7.1**	0.1
M-HG c	Fonte del Papa quarry	16	**0.512248**	0.000007	**-7.2**	0.1
Measured La Jolla Standard			0.511841	0.000008		

Figure 2. (A) Neodymium isotope (ε_{Nd}) data of the investigated hardgrounds plotted against different Mediterranean and Paratethys records and oceanic curves. Our ε_{Nd} data are plotted as pink squares (Fonte del Papa quarry, Maiella Mountain, Apulian platform), purple diamonds (Porto Badisco, Salento Peninsula, Apulian platform), and violet pentagons (Tornimparte village, Latium-Abruzzi platform). See Figure 1 for locations. Oceans' curves identify deep-water record from ferromanganese crusts (O'Nions et al., 1998). Open light-brown circles refer to mixed planktonic foraminifera of the Umbria-Marche hemipelagic succession (Cornacchia et al., 2018); open green and blue circles refer to Malta successions (Stille et al., 1996; Bialik et al., 2019); open dark-brown circles refer to mixed fossil assemblages and fish debris from Mediterranean hemipelagic successions (Kocsis et al., 2008); open light-gray triangles refer to fish debris from the central Paratethys and Northern Alpine foreland basin (Kocsis et al., 2009). Horizontal bars represent $^{143}Nd/^{144}Nd$ standard errors (SE), and vertical bars represent age uncertainties. (B) $^{143}Nd/^{144}Nd$ reported with analytical error (SE) and as ε_{Nd} with relative error. Full details on age correction and ε_{Nd} calculation are given in the Supplemental Material (see footnote 1). Ch.—Chattian; Aquit.—Aquitanian; Burdigal.—Burdigalian; Lan.—Langhian; Serr.—Serravallian; Me.—Messinian.

phosphate from sediments to the water column, creating a positive feedback for hardground formation (Föllmi, 1996).

The same influence of the central Paratethys can not be easily inferred for the other two investigated hardgrounds. Simon et al. (2019) stated that at ca. 13.8 Ma, local tectonic changes and a sea-level drop of 50–70 m restricted the exchanges between the Mediterranean and Paratethys, the latter of which passed from open-marine to hypersaline conditions. Furthermore, brackish to freshwater conditions are attested in the shallow portion of the eastern Paratethys and in the gateways with the Mediterranean during the Middle to Late Miocene (Popov et al., 2004; Piller and Harzhauser, 2005; Fig. 3B). However, the Langhian (ca. 12 Ma) and lower Tortonian (ca. 11 Ma) hardgrounds show ε_{Nd} values between −7.5 and −7.2, thus characterized by a more radiogenic signature in comparison to central Mediterranean values, such as those of the Malta ε_{Nd} record, that (already across the Langhian-Serravallian boundary; ca. 14 Ma) spanned from −9 to −11 (Bialik et al., 2019; Fig. 2A). Furthermore, the studied hardgrounds are comparable with the Umbria-Marche hemipelagic record (Kocsis et al., 2008; Cornacchia et al., 2018). Kocsis et al. (2008) interpreted these ε_{Nd} values as a hint of locally evolved Mediterranean deep waters. Wu et al. (2019) reconstructed the Holocene record of the Mediterranean deep and surface waters by analyzing Nd isotopes on fish debris and foraminifera. The authors reported that the Levantine deep waters were significantly more radiogenic (ε_{Nd} between −5.7 and −7.4) than the western Mediterranean ones (ε_{Nd} −9.5) because the Strait of Sicily inhibited the eastern deep-water flow. This is also in agreement with Nd isotope data of the Mediterranean margins that show an ε_{Nd} east-west gradient from ∼−7 to ∼−12 (Jeandel et al., 2007). de la Vara and Meijer (2016), modeling the Miocene circulation of the Mediterranean, stated that with a shallow or closed Indian Gateway, the onset of an anti-estuarine circulation took place, promoting the exit of Mediterranean deep waters into the Atlantic and favoring a small Atlantic input of surface waters. Furthermore, in the eastern basins, deep waters at >500 m depth remained partially isolated (de la Vara and Meijer, 2016). In this framework, the newly provided Nd isotope data confirm the development of locally evolved Mediterranean waters—characterized by a weak circulation and few exchanges between the deep and the upper levels of the water column—whose chemistry was significantly more affected by regional runoff than by Atlantic input, which is characterized by ε_{Nd} between −11 and −12 in the Late Miocene (O'Nions et al., 1998; Figs. 2A and 4).

Therefore, after the late Burdigalian (ca. 18 Ma), we should assume a Mediterranean circulation similar to that of today, with the deep Mediterranean waters forming within the eastern Mediterranean remaining partially isolated and only the shallow ones flowing westward. This partial isolation of the eastern Mediterranean deep waters explains also the difference between the Nd isotope records of Malta and the investigated successions (Stille et al., 1996; Bialik et al., 2019; Fig. 2A). The Malta Plateau, in fact, has been relatively higher than the surrounding areas since the Mesozoic and experienced a further uplift during the Neogene (Jongsma et al., 1985; Micallef et al., 2016). Thus, the Malta Nd isotope record testifies to a shallower portion of the water column in comparison to the Central Apennines hardgrounds, and the Malta ε_{Nd} signature is relatively more influenced by the Atlantic inflow. Secondly, the Malta Plateau acted as a barrier for the deep-water currents, limiting the exchanges between the eastern and western Mediterranean. In this context, the Nd isotope record of the Langhian (ca. 12 Ma) to lower Tortonian (ca. 11 Ma) hardgrounds testifies to this major change in the Mediterranean oceanography, i.e., the onset of locally evolved deep waters in the eastern Mediterranean, affected by the regional runoff more than by the adjacent oceans. Furthermore, Karami et al. (2009), modeling the consequences of the closure of the Indian Gateway on Mediterranean circulation, stated that with the Indian Gateway closed, the evaporation in the Mediterranean exceeded the freshwater input, leading to a longer residence time of waters into the basin.

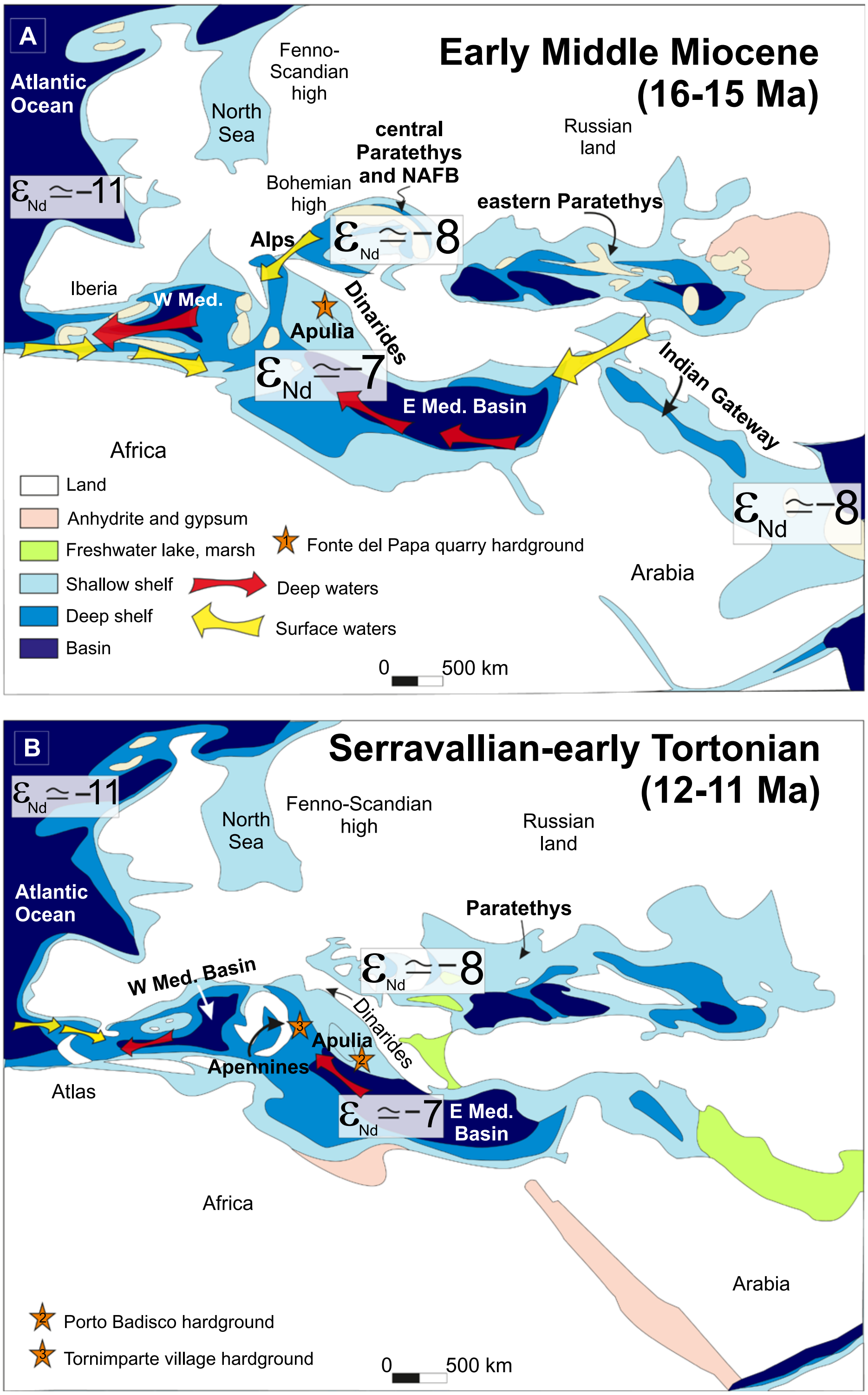


Figure 3. Mediterranean paleogeography during the early Middle Miocene (A) and Serravallian–early Tortonian (B) showing simplified circulation patterns, deep-water upwelling, and the average ε_{Nd} of the Mediterranean, central Paratethys and North Alpine foreland basin (NAFB), and adjacent oceans. ε_{Nd} of the Atlantic and Indian Oceans are from O'Nions et al. (1998); those of the central Paratethys and NAFB are from Kocsis et al. (2009). Modified from Cornacchia et al. (2021) after Popov et al. (2004).

Lastly, the Monterey event and the following CM7 perturbation coincided with a strengthening of ocean circulation and intensification of coastal upwelling (Holbourn et al., 2013). For the Mediterranean, after the closure of the Indo-Pacific connection, an oceanographic regime dominated by westward-oriented currents has been proposed by different authors (Moreno et al., 2004; Föllmi et al., 2015; Brandano et al., 2016a). The distinct Nd isotopic fingerprint of the analyzed central Mediterranean hardgrounds testifies to an eastern provenance of the deep waters that formed them. The deep water ascended to the eastern side of the Mediterranean ramps via upwelling before returning to the Atlantic, promoting a significant increase of primary productivity and favoring the development of phosphatic hardgrounds (Föllmi et al., 2008, 2015; Brandano et al., 2016a).

CONCLUSIONS

The Nd isotope record of three different phosphate-rich hardgrounds developed within Mediterranean shallow-water carbonate successions during the Miocene shows a persistent eastern signature. This record, framed in widely accepted paleogeographic reconstructions of the circum-Mediterranean region, attests to the onset of locally evolved deep waters in the eastern Mediterranean, proving that the closure of the Indo-Pacific connection changed Mediterranean circulation, leading to a longer residence time of waters and limited exchanges with the Atlantic Ocean. Lastly, the Nd isotope signature of the analyzed hardgrounds confirms their origin through upwelling of deep water. The onset of the upwelling is related to global climatic changes that affected the Mediterranean area.

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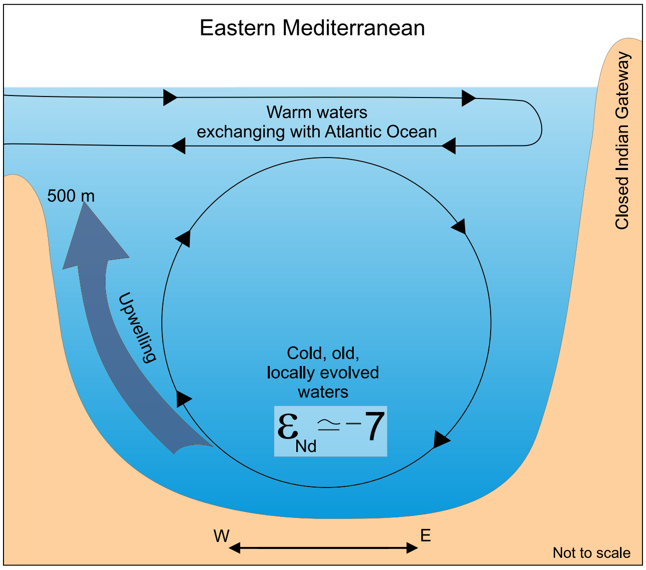

Figure 4. Simplified model of eastern Mediterranean anti-estuarine circulation after closure of the Indian Gateway, and upwelling formation. In the eastern basin, due to thermal stratification of the water column, deep cold waters below 500 m did not exchange with the upper level, and their Nd isotope signature was controlled by local and regional runoff. Modified after de la Vara and Meijer (2016).

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