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## To cite this version:

Hironao Matsumoto, Rodolfo Coccioni, Fabrizio Frontalini, Kotaro Shirai, Luigi Jovane, et al.. Long-term Aptian marine osmium isotopic record of Ontong Java Nui activity. Geology, 2021, 49, pp.1148 - 1152. 10.1130/g48863.1 . hal-03860568

GEOLOGY | THE GEOLOGICAL SOCIETY OF AMERICA®

https://doi.org/10.1130/G48863.1

Manuscript received 2 September 2020
Revised manuscript received 25 January 2021
Manuscript accepted 7 April 2021

# Long-term Aptian marine osmium isotopic record of Ontong Java Nui activity

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## ABSTRACT

**The early to mid-Aptian was punctuated by episodic phases of organic-carbon burial in various oceanographic settings, which are possibly related to massive volcanism associated with the emplacement of the Ontong Java, Manihiki, and Hikurangi oceanic plateaus in the southwestern Pacific Ocean, inferred to have formed a single plateau called Ontong Java Nui. Sedimentary osmium (Os) isotopic compositions are one of the best proxies for determining the timing of voluminous submarine volcanic episodes. However, available Os isotopic records during the interval are limited to a narrow interval in the earliest Aptian, which is insufficient for the reconstruction of long-term hydrothermal activity. We document the early to mid-Aptian Os isotopic record using pelagic Tethyan sediments deposited in the Poggio le Guaine (Umbria-Marche Basin, Italy) to precisely constrain the timing of massive volcanic episodes and to assess their impact on the marine environment. Our new Os isotopic data reveal three shifts to unradiogenic values, two of which correspond to black shale horizons in the lower to mid-Aptian, namely the Wezel (herein named) and Fallot Levels. These Os isotopic excursions are ascribed to massive inputs of unradiogenic Os to the ocean through hydrothermal activity. Combining the new Os isotopic record with published data from the lowermost Aptian organic-rich interval in the Gorgo a Cerbara section of the Umbria-Marche Basin, it can be inferred that Ontong Java Nui volcanic eruptions persisted for ~5 m.y. during the early to mid-Aptian.**

## INTRODUCTION

Oceanic anoxic events (OAEs) are major perturbations of the global carbon cycle, accompanied by the deposition of organic-rich sediments in various oceanographic settings. The early to mid-Aptian (Early Cretaceous) was marked by episodic burial of organic-rich sediments in the Tethyan region (Coccioni et al., 1987, 2012; Herrle et al., 2004), a gradual long-term decrease in the diversity of planktonic foraminifera (Coccioni, 2020), and a decline in the size of calcareous nannoplankton (Bottini and Faucher, 2020). The Selli Level, recognized not only in the Tethyan region but also in the Pacific Ocean, is one of the most studied Cretaceous organic-rich intervals and records the earliest Aptian OAE (OAE1a) (Coccioni et al., 1987; Price, 2003). In the Tethyan region, several minor organic-rich intervals occur above the Selli Level (Herrle et al., 2004; Coccioni et al., 2012). However, their significance in the geologic record is unclear because they are observed only in the Tethys at present and are poorly studied.

Radiometric ages of Ontong Java Plateau (OJP, southwestern Pacific Ocean) basalts (125–119 Ma; Mahoney et al., 1993; Tejada et al., 2002) roughly correspond to the depositional ages of the Selli Level (ca. 121–120 Ma; Malinverno et al., 2012), and massive volcanic episodes associated with the plateau emplacement have been ascribed as the trigger of the OAE1a and the associated marine biotic crises (e.g., Erba, 1994). Available geochemical data and radiometric ages of the Manihiki Plateau and the Hikurangi Plateau suggest that these plateaus, as well as the OJP, were part of a larger oceanic plateau, known as Ontong Java Nui (OJN) (Taylor, 2006; Chandler et al., 2012).

Marine Os isotopic ($^{187}$Os/$^{188}$Os) records have been applied to identify episodic submarine volcanic events (Turgeon and Creaser, 2008; Tejada et al., 2009; Bottini et al., 2012; Du Vivier et al., 2014). Open-ocean $^{187}$Os/$^{188}$Os is constant because of its long residence time (8–10 k.y.; Oxburgh, 2001) and reflects the balance between the Os input from the unradiogenic sources (e.g., hydrothermal activity, weathering of basaltic rocks, and cosmic dust) and radiogenic Os sourced from continental materials (Levasseur et al., 1999). Shifts to unradiogenic $^{187}$Os/$^{188}$Os in sedimentary records can, therefore, reflect massive input

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CITATION: Matsumoto, H., et al., 2021, Long-term Aptian marine osmium isotopic record of Ontong Java Nui activity: Geology, v. 49, p. XXX–XXX, https://doi.org/10.1130/G48863.1

of mantle-derived Os to the ocean via basaltic plateau emplacement. Early Cretaceous marine Os isotopic records are, however, limited to a short interval from the latest Barremian to the earliest Aptian (Tejada et al., 2009; Bottini et al., 2012), too short to trace the major OJN volcanic pulses ranging from early to mid-Aptian (Taylor, 2006). To assess the links between massive eruptions forming OJN and environmental perturbations in the early to mid-Aptian, this study extends the marine Os isotopic record to the entire Aptian using pelagic sediments collected from the Poggio le Guaine section (43°32'29.06"N, 12°34'51.09"E) and core (43°32'42.72"N, 12°32'40.92"E) in the Umbria-Marche Basin (central Italy) of the western Tethys (Coccioni et al., 2012; Fig. 1; Fig. S1 in the Supplemental Material[1]).

## RESULTS

The carbonate $\delta^{13}C$ ($\delta^{13}C_{carb}$) record of Poggio le Guaine core shows the highest values (4.7‰) just above the Selli Level, which is followed by a continuous decline to 3.1‰ toward the top of the studied interval in segment Ap10, at ∼13 m on the composite depth scale (Fig. 2; Fig. S2; Table S1). This trend is consistent with the $\delta^{13}C_{carb}$ records from the Gorgo a Cerbara section of the Umbria-Marche Basin and from the Vocontian Basin (southeastern France) (Herrle et al., 2004; Li et al., 2016). By integrating our new available $\delta^{13}C_{carb}$ data with those from the Poggio le Guaine and Gorgo a Cerbara sections (Coccioni et al., 2014; Li et al., 2016; Savian et al., 2016; Matsumoto et al., 2020), we constructed a composite $\delta^{13}C_{carb}$ record (Fig. 2) in which 21 carbon isotopic segments (Ap1 to Al6) can be identified, following Menegatti et al. (1998) and Herrle et al. (2004). Os and Re concentrations vary from 18 to 153 pg $g^{-1}$ and from 3 to 7100 pg $g^{-1}$, respectively (Fig. S3; Table S2). Age-corrected $^{187}Os/^{188}Os$ ($Os_i$) values above the Selli Level range from 0.27 to 0.55, with distinct excursions to unradiogenic values at the levels of two black shales at ∼3 m and ∼12 m (Fig. 2).

## DISCUSSION

The black shale horizon at 12 m on the composite depth scale near the boundary between the Ap9 and Ap10 carbon isotopic segments and within the *Globigerinelloides algerianus* planktonic foraminiferal zone can be correlated to the Fallot Level in the Serre Chaitieu section of the Vocontian Basin (Herrle et al., 2004). The black shale horizon located at ∼3 m on the composite depth scale falls in the Ap7 carbon isotopic segment and the *Leupoldina cabri* planktonic foraminiferal zone, which cannot be correlated to any named early Aptian black shales. Thus, we propose to name this undescribed black shale horizon as the "Wezel Level" (see also the lithological description in the Supplemental Material).

Upper Barremian $Os_i$ values from the Umbria-Marche Basin fluctuate around 0.7 and gradually decrease to 0.4 at ∼44 cm below the Selli Level (Fig. 2; Tejada et al., 2009). Upward, $Os_i$ values increase to 0.7 again before decreasing sharply to 0.2 in the lower part of the Selli Level (Fig. 2; Tejada et al., 2009). Based on the correspondence between the sedimentary age of the Selli Level (ca. 120–121 Ma; Malinverno et al., 2012) and the radiometric age of the OJP (125–119 Ma; Mahoney et al., 1993; Tejada et al., 2002), these two Os isotopic declines likely mark massive volcanic episodes during OJP emplacement (Tejada et al., 2009). $Os_i$ values increase to 0.5 above the Selli Level (∼2.4 m in Fig. 2), which may represent an interruption in submarine volcanic activity. However, $Os_i$ values drop to 0.27 around the Wezel Level (Fig. 2), recover back to 0.5, and show a small shift to unradiogenic values of 0.40 ∼1 m above the Wezel Level. A similar shift to unradiogenic $Os_i$ values of 0.36 is observed around the Fallot Level equivalent.

We estimated the unradiogenic Os input required to explain $Os_i$ variations using a box model, assuming constant continental weathering (cf. Tejada et al., 2009; Fig. S4; Table S3; and the box model calculation in the Supplemental Material). The three Os isotopic excursions, at the Wezel Level, 1 m above the Wezel Level, and at the Fallot Level equivalent, would have required at least ∼3100, ∼850, and ∼1150 t/k.y. increases in the unradiogenic Os input, respectively (Fig. 3). These values are 7.5, 2.1, and 2.6 times higher than the steady hydrothermal Os input associated with oceanic plate production of the uppermost Barremian (Fig. 3). To explain these negative Os isotopic shifts solely by a decrease in the flux of continental Os, a reduction of ∼60%–80% of continental-derived Os would be required. Given that continental weathering rates rose during the early Aptian (e.g., Blättler et al., 2011), this possibility is unlikely. The sedimentary ages of the early to mid-Aptian Os isotopic excursions (ca. 121–116.3 Ma; Malinverno et al., 2012) roughly correspond to the radiometric ages of the OJP (125–119 Ma; Mahoney et al., 1993; Tejada et al., 2002) and the Manihiki Plateau (126–117.9 Ma; Ingle et al., 2007; Timm et al., 2011), so the most probable source of unradiogenic Os is the hydrothermal activity associated with the

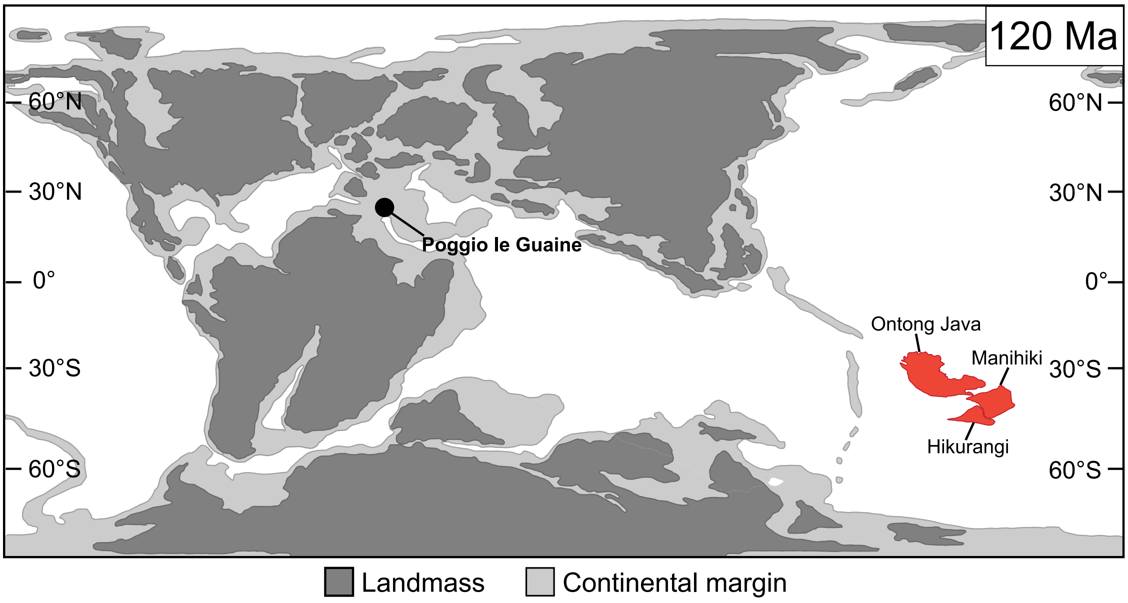

**Figure 1. Paleogeography at 120 Ma based on Chandler et al. (2012), showing location of the studied Poggio le Guaine section (Umbria-Marche Basin, Italy).**

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[1] Supplemental Material. Description of lithology and methods, Figure S1 (sampling site), Figure S2 (cross plots of $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$), Figure S3 (Re-Os data), Figure S4 (age-depth model), Table S1 (raw $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ data), Table S2 (raw Re-Os data), and Table S3 (age model). Please visit https://doi.org/10.1130/~~XXXXX~~ to access the supplemental material, and contact editing@geosociety.org with any questions.

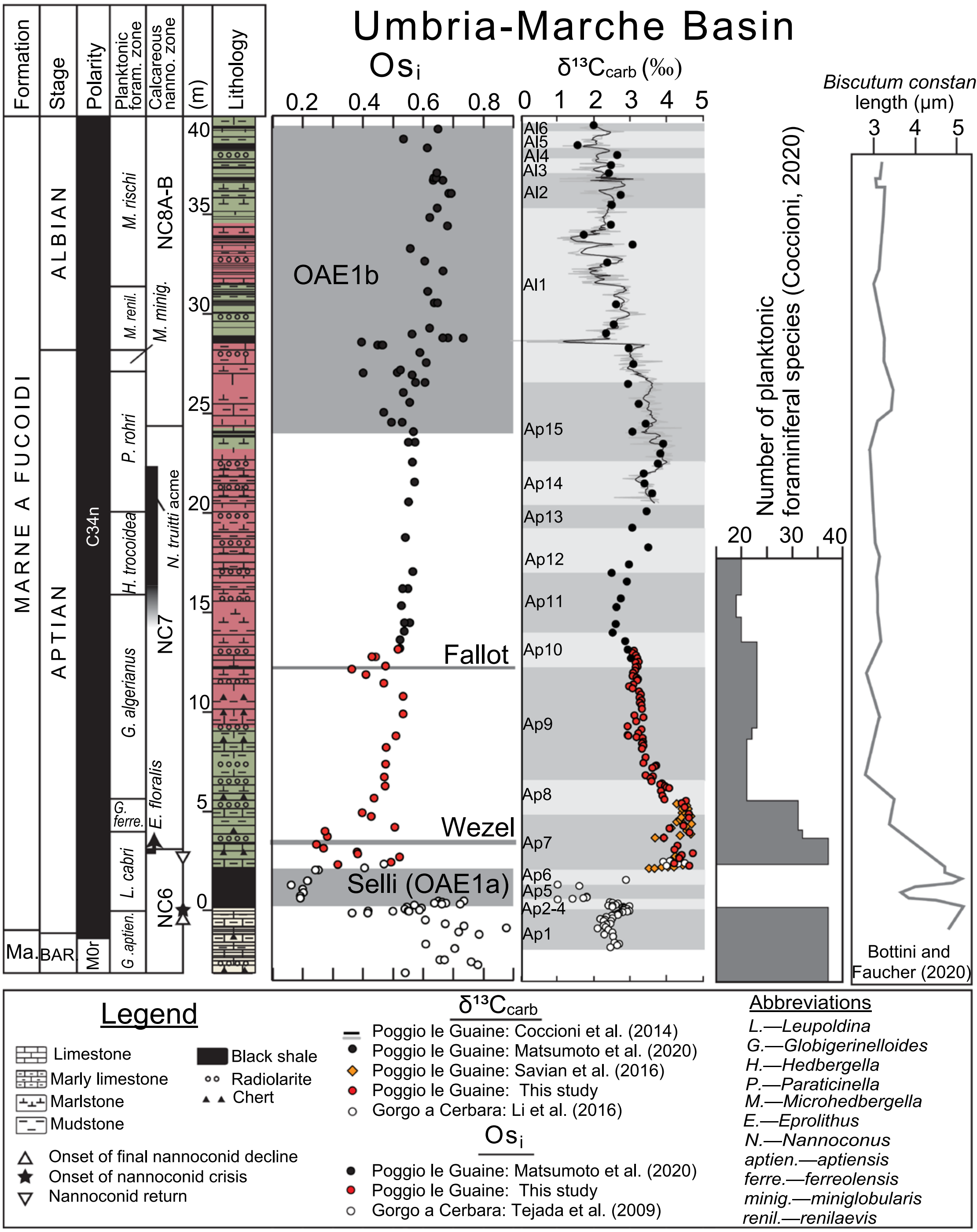

**Figure 2. Late Barremian to early Albian $^{187}$Os/$^{188}$Os (Os$_i$) and carbonate δ$^{13}$C (δ$^{13}$C$_{carb}$) records in the Poggio le Guaine (PLG) section and core and the Gorgo a Cerbara section of the Umbria–Marche Basin (Italy) Chronostratigraphy is after Coccioni et al. (2014), Coccioni (2020), and Matsumoto et al. (2020). Litho-, bio-, and magnetostratigraphy correspond to those of the PLG record, after Coccioni et al. (2012, 2014), Savian et al. (2016), Matsumoto et al. (2020), and this study. Biostratigraphy is slightly modified based on Coccioni (2020). Colors used in the lithological column are the schematic colors of the sedimentary rocks based on Coccioni et al. (2012). Os$_i$ is from Tejada et al. (2009), Matsumoto et al. (2020), and this study. Gray horizontal bars in the Os$_i$ plot represent the intervals of oceanic anoxia. δ$^{13}$C$_{carb}$ is after Coccioni et al. (2014), Li et al. (2016), Savian et al. (2016), Matsumoto et al. (2020), and this study. Gray and black lines in δ$^{13}$C$_{carb}$ plots represent the raw and smoothed curves of δ$^{13}$C$_{carb}$ at the PLG record, respectively (Coccioni et al., 2014). Number of planktonic foraminiferal species at the Gorgo a Cerbara section is after Coccioni (2020). Variation of *Biscutum constans* size in the Cismon (southern Alps, Italy) and Piobbico (central Italy) cores is from Bottini and Faucher (2020). Ma.—Maiolica; BAR—Barremian;foram.—foraminiferal; nanno.—nannofossil; OAE—Oceanic Anoxic Event.**

emplacement of these plateaus. The weathering of young basaltic plateau could be another candidate for the unradiogenic Os source. However, most of OJN was emplaced under submarine conditions, and subaerial weathering is considered to be unlikely to cause the large unradiogenic Os$_i$ shift (e.g., Shipboard Scientific Party, 2001), despite evidence of subaerial eruption on OJN (Thordarson, 2004).

The most probable trigger for the black shale deposition is global warming promoted by the massive release of volcanic gases associated with the emplacement of OJN. Global warming could have enhanced the continental weathering supplying nutrients into the ocean and, in turn, stimulated the primary productivity that ultimately led to the local marine anoxia and the subsequent deposition of the Wezel and Fallot Levels. These levels are currently reported only in Tethyan regions, possibly because the depositional environment may have been susceptible to nutrient supply from riverine input and upwelling. The sedimentary thickness of the organic-rich interval and δ$^{13}$C$_{carb}$ excursion at the Wezel (~20 cm and at most −1‰) and Fallot Levels (~6 cm and none) are smaller than at the Selli Level (~2 m and −2‰). The negative δ$^{13}$C excursion during OAE1a is ascribed to the thermogenic methane released from sill intrusion into marine organic-rich sediments (Adloff et al., 2020). Considering the smaller unradiogenic Os shifts at the Wezel and Fallot Levels (Fig. 3), the perturbations of the carbon cycle at these horizons may have been limited compared to that at the Selli Level. At each of the Wezel Level and the Fallot Level equivalent, the initial decline in Os$_i$ occurs ~60 cm below the onset of deposition of black shale horizons, which corresponds to ~200 k.y. (Fig. 3; Table S1). A shorter time lag (~59 k.y.) between unradiogenic Os$_i$ shift and the onset of positive δ$^{13}$C$_{carb}$ excursion (i.e., the potential organic-carbon burial)

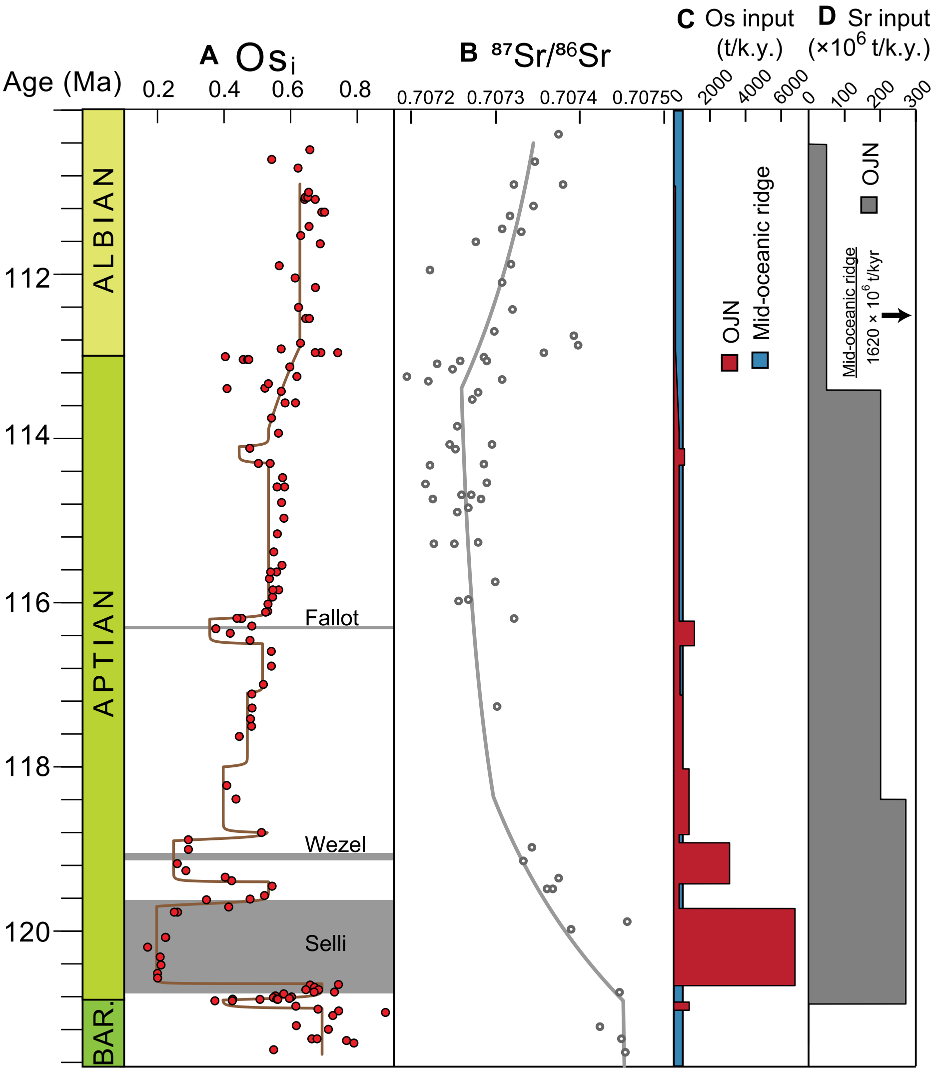

**Figure 3.** $^{187}$Os/$^{188}$Os (Os$_i$) (A) $^{87}$Sr/$^{86}$Sr (B) variations, unradiogenic Os (C) and unradiogenic Sr (D) inputs from Ontong Java Nui volcanism. Solid lines in A and B represent the estimated Os$_i$ and $^{87}$Sr/$^{86}$Sr variations caused by the hydrothermal Os and Sr input described in C and D. Os$_i$ is based on Tejada et al. (2009), Matsumoto et al. (2020), and this study. Gray horizontal bars in A represent the intervals of oceanic anoxia. $^{87}$Sr/$^{86}$Sr data are from Bralower et al. (1997). Time scale is after Coccioni (2020) and Matsumoto et al. (2020) (Fig. S4 and Table S3 [see footnote 1]). BAR.—Barremian; OJN—Ontong Java Nui.

is observed during OAE2 (Jones et al., 2020). The time lag could represent the required time from the onset of the volcanic outgassing to the outbreak of oceanic anoxia. Smaller Os isotopic shifts than at OAE2 around the Wezel and Fallot Levels imply decreased volcanic eruptions that may have led to tempered environmental perturbations and longer time lags. Above the Fallot Level equivalent, Os$_i$ shows stable values of ~0.55 to the end of Aptian (Matsumoto et al., 2020), lower than the latest Barremian pre-OAE1a values (~0.7) (Tejada et al., 2009), though estimated Barremian values have large variations (0.4–0.9) (Bottini et al., 2012). Because the oceanic crustal production rate during the Aptian is considered higher than that of the Barremian (Eldhom and Coffin, 2000), the intensified hydrothermal activity associated with plate production may have contributed to the low steady Os$_i$ values. Another possibility is that small-magnitude but continuous volcanism after the main volcanic pulse at OJN released unradiogenic Os into the ocean. Indeed, the basaltic rocks at the northeastern plateau margin (Rapuhia Scarp) on the Hikurangi Plateau show younger ages (118–96 Ma; Hoernle et al., 2010) than the peak ages of the OJP and Manihiki Plateau, which may support this possibility.

Marine strontium isotopic ratios ($^{87}$Sr/$^{86}$Sr) represent the balance between radiogenic Sr input from continents and unradiogenic Sr input through hydrothermal activity. Although $^{87}$Sr/$^{86}$Sr varies like Os$_i$, the residence time of Sr is much longer (~2.5 m.y.; Hodell et al., 1990) than that of Os (8–10 k.y.; Oxburgh, 2001), which makes the change in marine $^{87}$Sr/$^{86}$Sr more gradual than that of Os$_i$. The global marine $^{87}$Sr/$^{86}$Sr curve shows a marked decline toward unradiogenic values during the early Aptian (Fig. 3; Bralower et al., 1997), which supports enhanced hydrothermal activity induced by the submarine volcanic episodes. Although their timing is concordant, the increase in the input of unradiogenic Os is more drastic (2.6–16 times higher) compared to that of Sr (~15% increase) owing to their different geochemical behaviors.

The Os$_i$ fluctuations continued for ~5 m.y. during the early to mid-Aptian, which could reflect a long-term submarine volcanic eruption at OJN. The Os isotopic fluctuations during the early to mid-Aptian correspond to stepwise declines in the planktonic foraminiferal diversity, the demise of nannoconids, and a decrease in the shell size of nannofossils at least in the Umbria-Marche Basin (Fig. 2; Erba, 1994; Bottini and Faucher, 2020; Coccioni, 2020). Because calcareous organisms are strongly influenced by ocean acidification (Erba et al., 2010; Bottini and Faucher, 2020; Matsumoto et al., 2020), the biotic changes have been ascribed to at least local lowering of pH caused by the massive volcanic outgassing. After the three major Os isotopic declines, an increase in the heavily calcified planktonic foraminiferal species (Coccioni et al., 2014) and a blooming of nannoconids (*Nannoconus truitti* acme, Fig. 2; Coccioni et al., 2014) are reported, suggesting a weakening of the acidified conditions caused by volcanic activity at OJN. These pieces of evidence imply that the volcanic episodes at OJN could have had an impact on marine ecosystems at least locally from the latest Barremian to the mid-Aptian, potentially through marine anoxia and acidification.

## CONCLUSIONS

Our new Os isotopic record from the Poggio le Guaine in the Umbria-Marche Basin provides evidence of several Os isotopic shifts to unradiogenic values around two black shale horizons, namely the Wezel Level and the Fallot Level equivalent in the lower to mid-Aptian. Considering the lack of large-scale subaerial exposure on OJN, the Os isotopic variations are attributed to massive inputs of unradiogenic Os through hydrothermal activity. Integrating our Os isotopic data with published data from the late Barremian to early Aptian of the Gorgo a Cerbara section, we infer that intensive hydrothermal activity continued for ~5 m.y. during the early to mid-Aptian and was associated with the deposition of three major organic-rich intervals in the Tethyan region (Selli Level, Wezel Level, and Fallot Level equivalent). Considering the consistency of radiometric ages of OJN with the sedimentary ages of the Os isotopic shifts, the most probable cause for these volcanic signals is submarine volcanic eruptions at OJN.

## ACKNOWLEDGMENTS

Sincere gratitude is expressed to coauthor R. Coccioni for the suggestion of the naming of the undescribed early Aptian black shale horizon as the "Wezel Level" in honor of Forese Carlo Wezel for his undisputed international credits in the field of geology. We thank K. Suzuki, T. Nozaki, and Y. Otsuki for support in the Re-Os analysis, and K. Tanaka and N. Izumoto for support in the $\delta^{13}C_{carb}$ analysis. We thank Ian Jarvis and anonymous reviewers for valuable comments. This study was financially supported by a Grant-in-Aid for Japan Society for the Promotion of Science Research Fellow (grant 19J20708). We acknowledge the FUSP (Fundação de Apoio à Universidade de São Paulo)–Petrobras BARREMAG and 2405 projects for financial support of the drilling of the Poggio le Guaine core.

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