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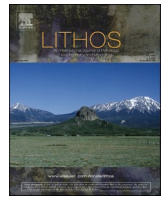
Publication Date

2023-04-01

DOI

10.1016/j.lithos.2023.107094

Peer reviewed



Letter

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ARTICLE INFO

Keywords
 Topography
 Rodinia
 Gondwana
 Continental arc

ABSTRACT

Widespread orogenesis associated with supercontinent assembly generates extensive collisional mountain chains with high elevations that are crucial for modulating the interaction between the atmosphere and lithosphere. However, not all supercontinents formed over Earth's history shared the same topography. This study investigates the temporal variation over the last 1.5 Ga in La/Zr ratio of continental-arc mafic volcanic rocks. We show that 0.6–0.5 Ga rocks related to Gondwana assembly are characterized by much higher La/Zr ratios compared with 1.2–0.9 Ga rocks formed during Rodinia assembly. Thermodynamic modeling reveals that the La/Zr ratios of continental-arc magmas are predominantly controlled by melting depth, which is directly tied to the thickness of the overlying arc crust. Thick continental arc decreases the extent of melting in the mantle wedge, leading to an increase in La/Zr ratios. Higher La/Zr ratios of continental-arc rocks during Gondwana vs. Rodinia assembly thus imply thicker continental arcs for the younger supercontinent. We suggest that the secular La/Zr ratio of continental-arc mafic magmas is an effective geochemical indicator of topography difference between the Rodinia and Gondwana supercontinents, with implications for coupling between the evolution of the lithosphere and atmosphere.

1. Introduction

Nutrient flux from the continents to the oceans is facilitated by elevated topography formed during orogenesis. The latter has been proposed to be a significant factor affecting the biosphere, surface oxidation, and climate over the Earth's history (Squire et al., 2006; Zhu et al., 2022). Global episodes of orogenesis that led to the formation of supercontinents are marked by extensive mountain chains, such as those associated with the Meso- to Neoproterozoic Rodinia supercontinent and its successor, the Gondwana supercontinent (Cawood et al., 2016; Spencer et al., 2013). Using detrital zircon provenance and isotopic proxies for crustal reworking, orogenesis during Gondwana assembly is thought to have yielded higher elevations, in contrast to Rodinia-related orogenesis, which is interpreted to produce a more subdued topography (Sundell and Macdonald, 2022; Zhu et al., 2022). This speculation is consistent with the crustal thickness estimate based on the detrital zircon Eu/Eu* proxy, indicating a thin (<45 km) continental crust at convergent margins during the Rodinia supercontinent tenure (Tang et al., 2021).

To test the above conclusions based on detrital zircon geochemistry, we turn to the composition of mafic volcanic rocks generated in continental arcs. Continental arcs, the site for generating felsic continental crust, tend to have a long life span (>100 Ma), and terminate by a collisional event (Cao et al., 2017; Ducea et al., 2015). The thick crust of continental arcs would yield high elevations (e.g., Andes) and impact the composition of arc magmas either derived via partial melting in arc mantle wedge or intracrustal differentiation (Luffi and Ducea, 2022; Turner and Langmuir, 2015a). We applied a bootstrap resampling statistical approach to the composition of mafic volcanic rocks associated with continental arcs throughout geologic time to establish the secular variations in La/Zr ratios. Supported by thermodynamic modeling, the

variation of La/Zr in continental-arc igneous rocks is found to be strongly influenced by melting depth in the arc mantle, which is directly tied to the thickness of the overlying arc crust. We propose that La/Zr of continental-arc mafic volcanic rocks is a sensitive proxy for continental-arc thickness and therefore relates to topography difference between the Rodinia and Gondwana supercontinents.

2. Methods and results

A global compilation of 5790 primitive mafic volcanic rocks erupted over the last 1.5 Ga were extracted from the geochemical database in Gard et al. (2019) (Fig. S1, Table S1), using the following data filtering criteria: 1) Samples were tagged as “igneous” and “volcanic” in the original database; 2) Only rock compositions with the SiO₂ and MgO contents falling into the ranges of 45–53 wt% and 6–13 wt%, respectively, were used to exclude highly evolved or cumulate rocks. 1545 continental arc-related data were further selected based on geochemical criteria of Th/Nb, (Nb/La)_{PM}, and Zr/Y (Th/Nb ≥ 0.2, (Nb/La)_{PM} < 0.75, and Zr/Y > 3) (Fig. S2) (Gao et al., 2022; Pearce, 1983, 2008).

The average values for elemental composition and interelement ratios (e.g., La, Zr, and La/Zr) for ≤1.5 Ga continental-arc volcanic rocks were calculated using Julia's weighted bootstrap resampling method based on the StatGeochem.jl package following Keller and Schoene (2012). The time bin for each compositional variable of interest was arbitrarily set at 60 Ma. This approach helps to alleviate sampling bias by using a built-in algorithm that assigns each sample a resampling probability, which is inversely correlated to spatiotemporal sample density (Keller and Schoene, 2012). Although there are potential limitations including preservation bias (Hawkesworth et al., 2009; Sundell and Macdonald, 2022), these effects can be limited by the use of independent geochemical proxies that yield a convergent conclusion.

<https://doi.org/10.1016/j.lithos.2023.107094>

Received 14 December 2022; Received in revised form 15 February 2023; Accepted 15 February 2023

Available online 18 February 2023

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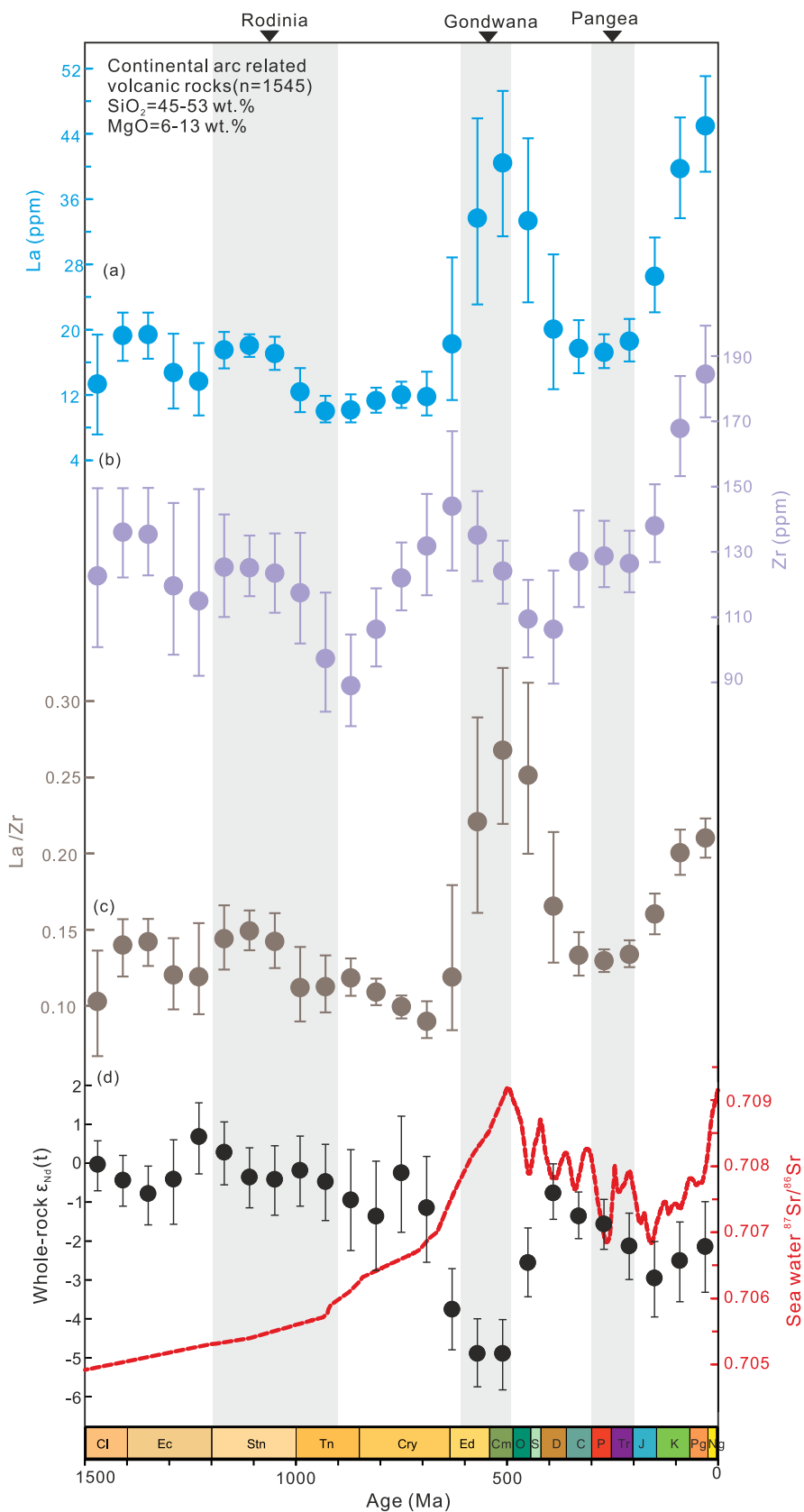


Fig. 1. (a-c) Secular variations of La, Zr, and La/Zr through time for screened continental-arc mafic volcanic rocks; (d) On the left panel is the scale for the whole-rock $\epsilon_{Nd}(t)$ vs. age plot. The whole-rock $\epsilon_{Nd}(t)$ data for the ≤ 1.5 Ga igneous rocks is from [Puetz and Condie \(2019\)](#). On the right panel is the scale for the seawater $^{87}Sr/^{86}Sr$ vs age graph constructed by LOWESS fit with the original data from [Chen et al. \(2022\)](#).

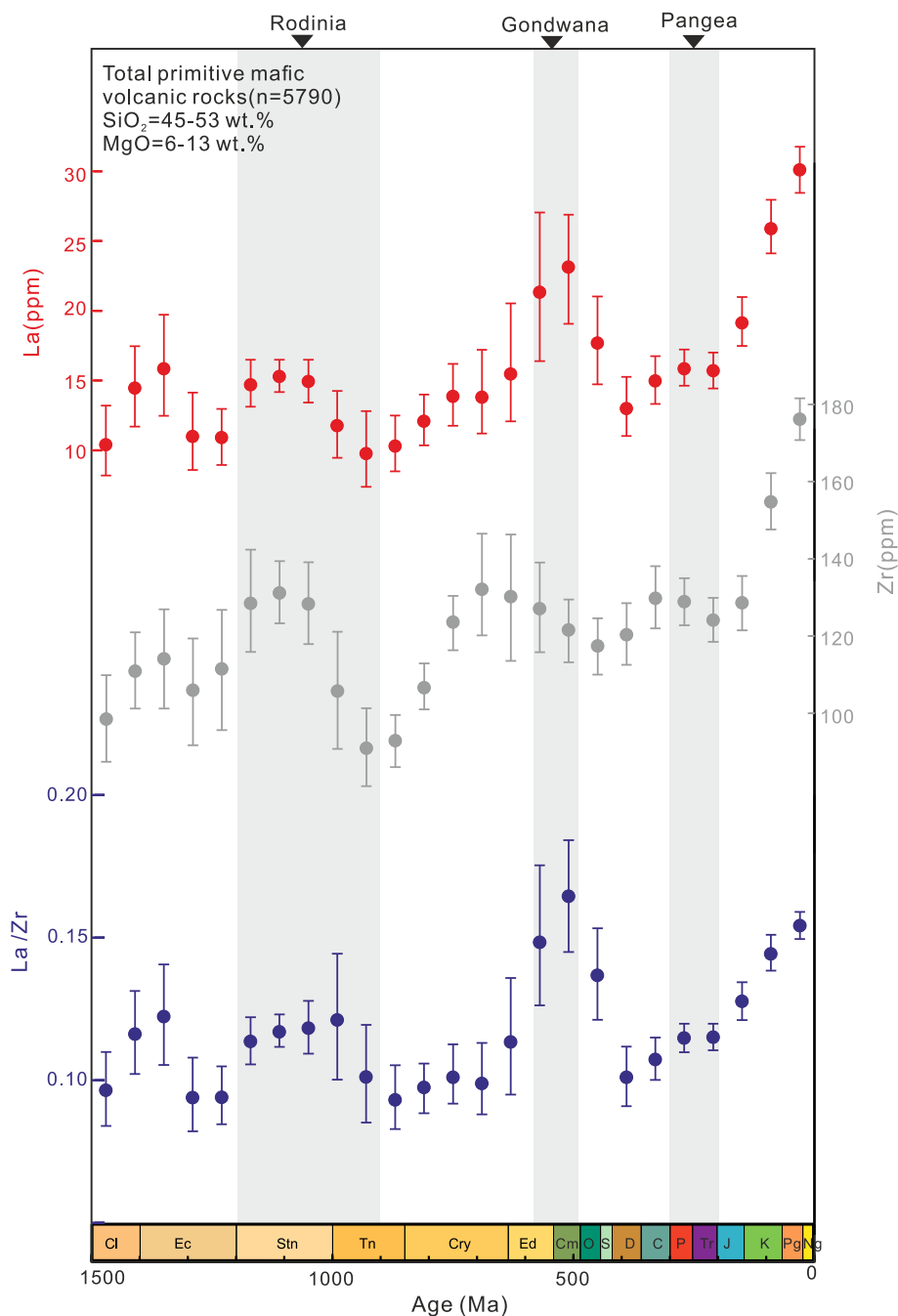


Fig. 2. Secular variations of La, Zr, and La/Zr through time for total mafic volcanic rocks compiled in this study.

To show how aqueous liquid, hydrous sediments, and altered oceanic crust (AOC) melts contribute to the geochemical character of the continental-arc mafic volcanic rocks (e.g., Turner and Langmuir, 2022; Xu et al., 2020), alphaMELTS_1.9 software was used to simulate melting in the mantle wedge of a subduction zone (Ghiorso et al., 2002; Smith and Asimow, 2005). The modeling procedure assumes that the partial melting occurred under an isobaric condition with constant temperature that approximates to or just below the original solidus of the mantle wedge, whose partial melting is induced by progressive addition of small amounts (mass = 0.01) of aqueous liquid/hydrous sediments/AOC melts into this source via many increments of melting models. For this open system, once the fraction of melt exceeds a fixed threshold (MINF = 0.01, here), the extra melt in each increment is extracted and the remaining system becomes the starting point for the melting model of next incremental addition of extraneous/subduction fluids/melts. The

depleted MORB mantle (DMM) is chosen to represent the primitive sub-arc mantle (Workman and Hart, 2005). The composition of slab-derived fluids and melts used in this study and associated references are presented in Table S2. The pressure is set at 2.0–4.0 GPa, roughly equivalent to the slab depth beneath arc volcanoes (Syracuse and Abers, 2006). The input data for various variables and starting settings for the flux-melting model are provided in supplementary material 2.

The 0.6–0.5 Ga continental-arc volcanic rocks erupted during Gondwana assembly have average La and Zr concentrations of 40 ± 9 to 34 ± 11 ppm, and 135 ± 14 to 124 ± 10 ppm, much higher than the 1.2–0.9 Ga rocks emplaced during Rodinia assembly (La = 18 ± 2 to 10 ± 1 ppm, Zr = 125 ± 15 to 97 ± 16 ppm), yielding average La/Zr ratios of 0.27 ± 0.05 to 0.22 ± 0.06 for Gondwana tenure and 0.15 ± 0.01 to 0.11 ± 0.02 for Rodinia tenure (Figs. 1a-c, Table S3). It is noteworthy that 5790 primitive mafic volcanic rocks without tectonic settings

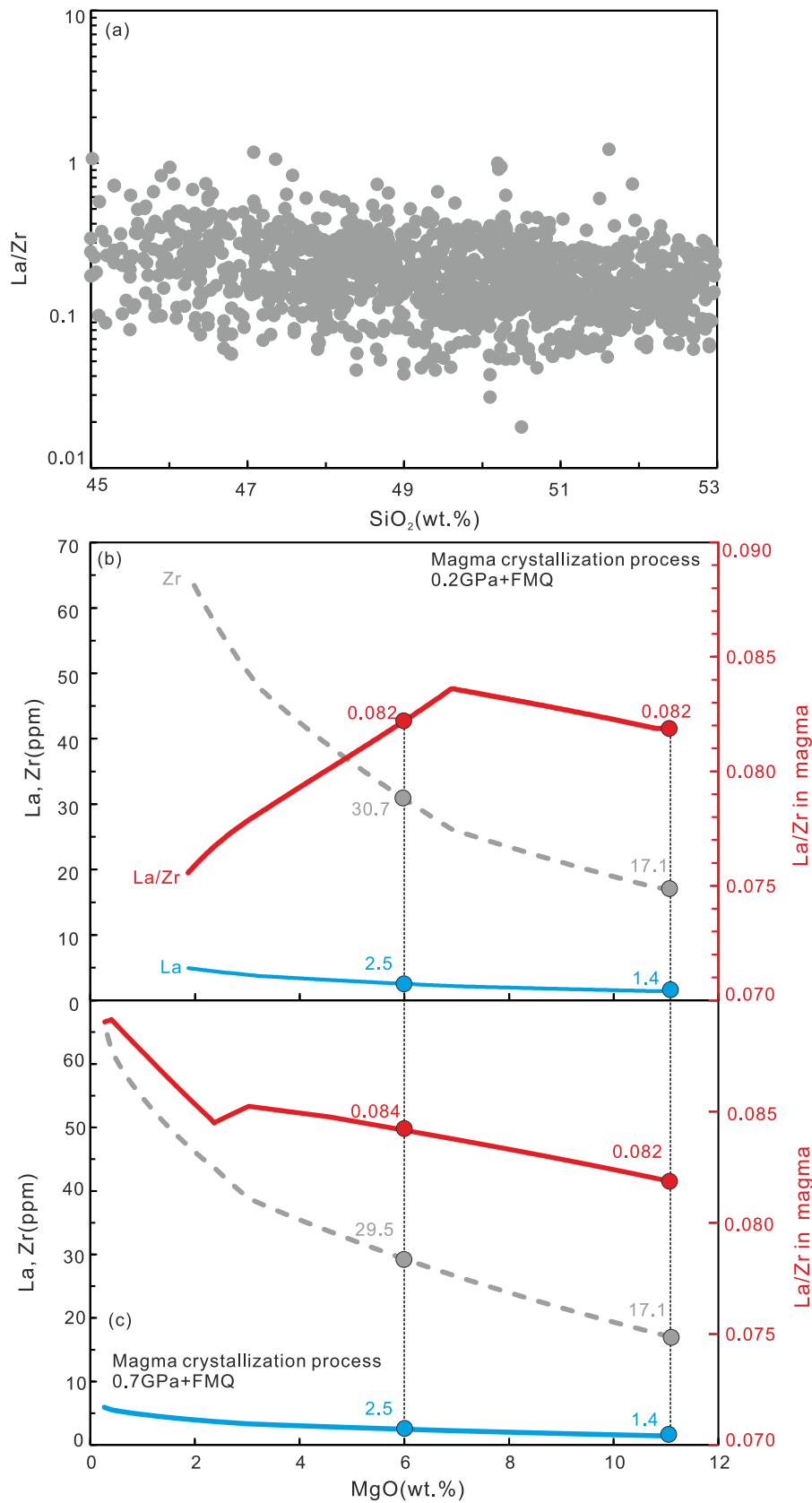


Fig. 3. (a) SiO₂ vs. La/Zr diagram for studied continental-arc volcanic rocks; (b-c) The results of fractional crystallization modeling under the conditions of high (0.7 GPa) and low (0.2 GPa) pressure using alphaMELTS 1.9 software (Ghiorso et al., 2002; Smith and Asimow, 2005). The starting material of primitive arc basalt (HPD1147R06) is from Tamura et al. (2013).

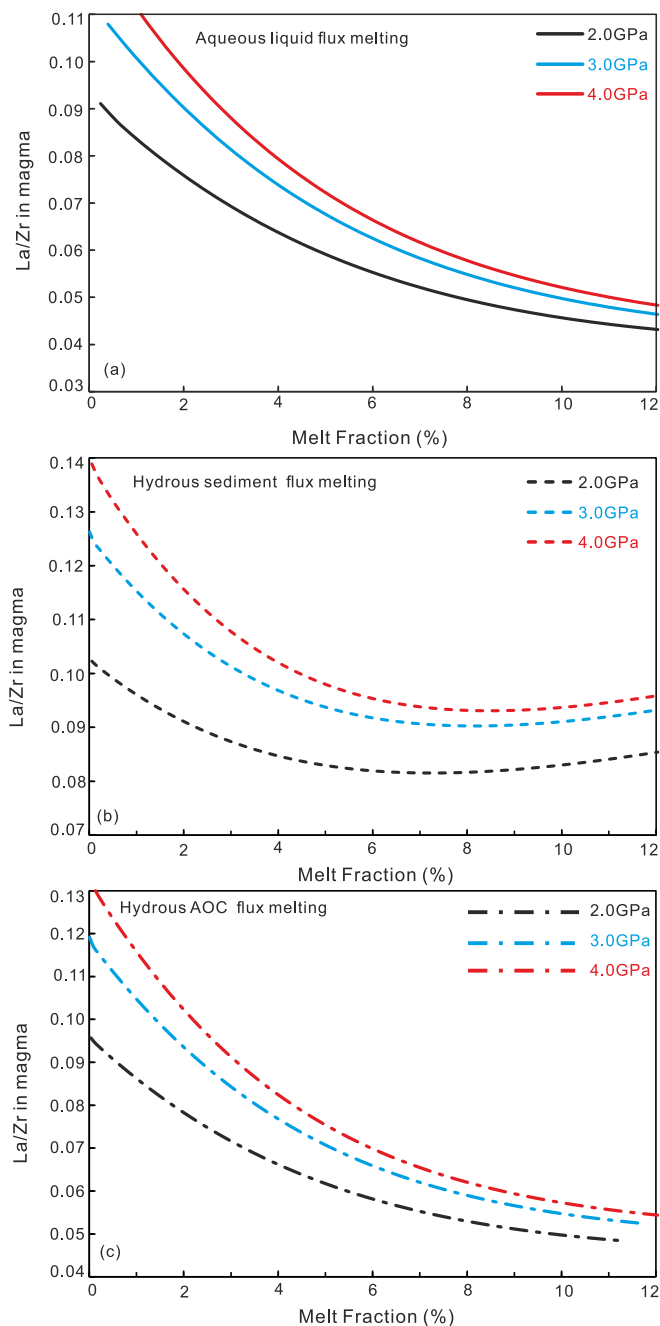


Fig. 4. (a–c) Variations in La/Zr in melts during flux-melting (aqueous liquid, hydrous sediment, and altered oceanic crust melts) in the continental-arc mantle wedge.

filtering yielded similar peaks in La, Zr, and La/Zr ratios during Gondwana assembly as those for continental-arc mafic volcanic rocks (Fig. 2).

3. Discussion

3.1. Factors controlling La/Zr in continental-arc mafic volcanic rocks

The highly incompatible elements La and Zr strongly partition into Earth's crust during mantle-crust differentiation (Rudnick and Gao, 2014), and they are generally deemed to have low mobility, thus resistant to post-magmatic weathering and alteration. Crustal contamination is readily eliminated as having a significant effect on the composition of targeted samples since all are relatively primitive volcanic rocks with high MgO contents (≥ 6.0 wt%). In addition, no

correlation is observed between SiO₂ contents and La/Zr ratios for targeted basalts (Fig. 3a), suggesting that the variations of La/Zr are not controlled by magma differentiation. This interpretation is further confirmed by magma differentiation modeling, which shows that fractional crystallization under the conditions of both high (0.7 GPa) and low (0.2 GPa) pressures is unlikely to substantially contribute to the observed La, Zr, and La/Zr variations when MgO ≥ 6.0 wt% (Fig. 3b–c).

The gradual increase in kimberlite magmatism after 1.2 Ga suggests that the mantle melting regime has changed since that time due to the interplay of secular mantle cooling and plate tectonics (Tappe et al., 2018). A colder mantle will yield a lower degree of partial melting than a hotter mantle (Campbell and Griffiths, 2014), which contributes to higher contents and ratios of incompatible elements (e.g., Wang and Becker, 2018). Further, the thermodynamic modeling shows that the lower degree of mantle melting results in higher La/Zr ratios (Fig. 4). In light of this, the higher La/Zr ratios during Rodinia assembly compared with those during Rodinia assembly could reflect a lower degree of melting of the mantle wedge due to a colder mantle. However, the following two reasons challenge this possibility: 1) Mantle temperature during Rodinia assembly was only slightly higher (<20 °C) than during Gondwana assembly (Ganne and Feng, 2017; Herzberg et al., 2010). 2) If the mantle cooling dominates the La/Zr variation trends, it would continuously increase through time, which is the opposite to the observed La/Zr peak during Gondwana time (Fig. 1c). Flux melting modeling using an aqueous liquid, hydrous sediment melts, and AOC melts at varying ranges of pressure conditions (2–4 GPa) demonstrates that higher La/Zr ratios can be obtained when melting occurs at a greater mantle depth regardless of what slab materials were involved (Figs. 4a–c). Further, the median La/Zr ratios of magmas from <100 Ma continental-arc volcanic rocks show a positive correlation ($R^2 = 0.52$) with their crustal thickness, confirming that the melting depth in the mantle wedge governs La/Zr ratios (Fig. S3). These findings are consistent with those of Turner and Langmuir (2015a), who showed that concentrations of LREEs and Zr correlate well with the crustal thickness of arcs. In addition to crustal thickness, the convergence rate and slab dip angle also regulate the melting degree of the mantle wedge, with faster plate convergence and steeper slab dip angle giving rise to a greater extent of melting at a given distance from the trench (Turner and Langmuir, 2015b). However, available paleomagnetic and passive margin data argue against a faster plate convergence during Rodinia assembly compared with Gondwana and Pangea assemblies (Bradley, 2011; Pesonen and Rolf, 2021). In summary, we deduce that the crustal thickness of the overlying continental arc is the first-order control over La/Zr ratio of continental arc mafic volcanic rocks through time.

3.2. Contrasting topography of the Rodinia and Gondwana supercontinents

Published detrital zircon trace element and Lu–Hf isotopic data indicate a higher production rate of S-type granites, extensive high-pressure metamorphism, and a more evolved, older crust-derived $\epsilon_{\text{Hf}}(t)$ during Gondwana assembly compared to prior and subsequent periods of supercontinent assembly (Spencer et al., 2013; Sundell and Macdonald, 2022; Zhu et al., 2020). This suggests a higher elevation for the Gondwana supercontinent (Zhu et al., 2022). The evolved, older crust-derived whole-rock $\epsilon_{\text{Nd}}(t)$ values for igneous rocks and radiogenic seawater Sr isotope ratios during Gondwana assembly are also consistent with this interpretation (Fig. 1d). Furthermore, the La/Zr ratios of continental-arc volcanic rocks during Rodinia (0.15 ± 0.01 to 0.11 ± 0.02) and Pangea (0.13 ± 0.01) assembly are comparable, but lower than those during Gondwana (0.27 ± 0.05 to 0.22 ± 0.06) assembly, directly reflecting thicker continental-arc crust during Gondwana assembly (Fig. 5). However, the inferred thickness of crust at convergent continental margins based on the detrital zircon Eu/Eu* proxy consistently increases over the Phanerozoic (Tang et al., 2021). This discrepancy may partly stem from the focus on different time slices in these

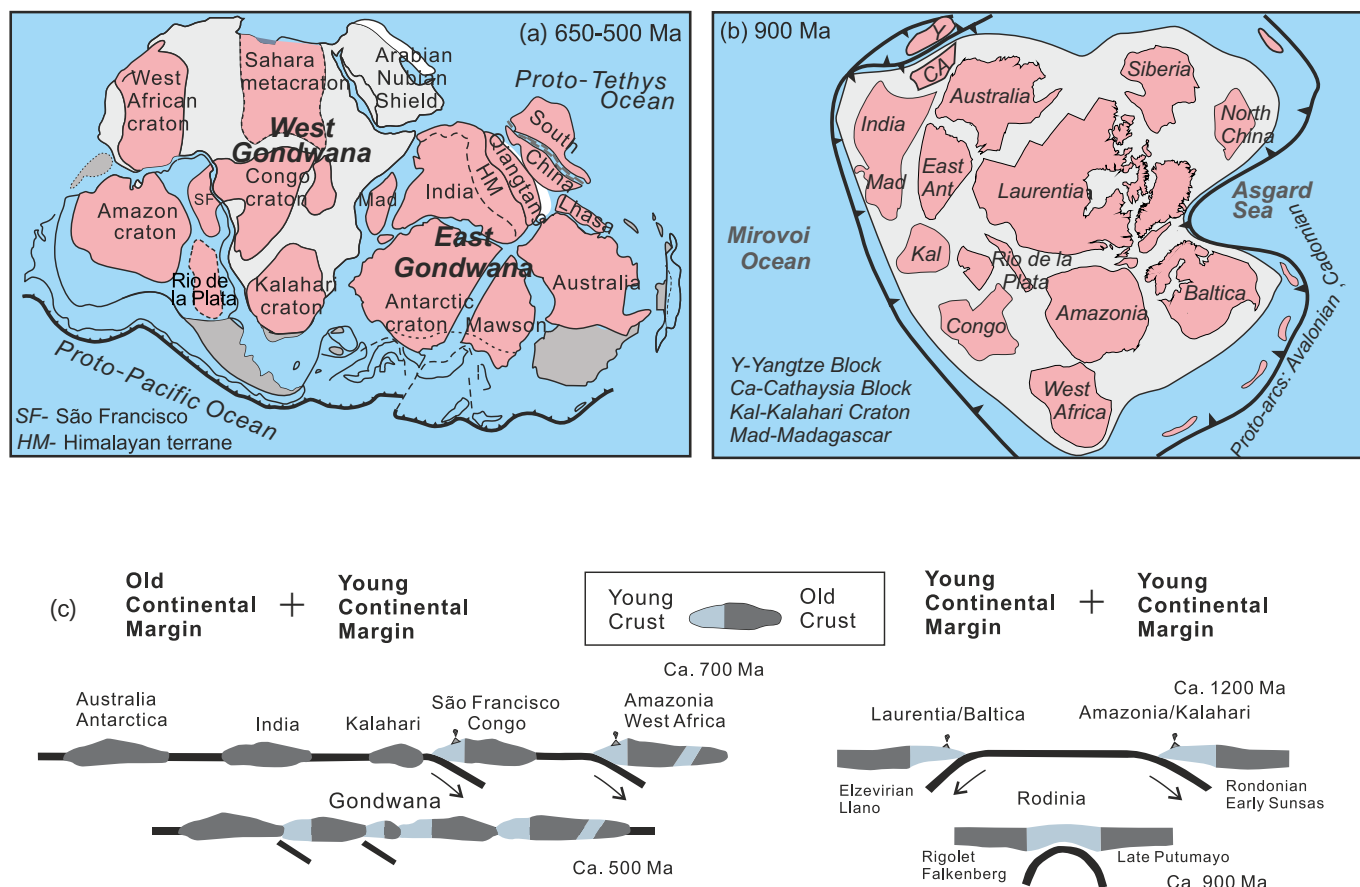


Fig. 5. (a-b) Schematic diagrams illustrating paleogeographic reconstructions for Rodinia and Gondwana supercontinents (modified after Wang et al., 2021); (c) contrasting supercontinent geodynamic models of Rodinia and Gondwana (modified after Spencer et al., 2013).

analyses, with our study concerned with continental arcs before collision, whereas Tang et al. (2021)'s study centered on continent-continent assembly that yields high elevations in their aftermath. Further, our findings are in good agreement with detrital zircon $\epsilon_{\text{Hf}}(t)$ patterns for Rodinia, Gondwana, and Pangea supercontinents (Sundell and Macdonald, 2022). It has been suggested that different subduction styles (dual-sided subduction of internal oceans during Rodinia assembly vs. single-sided subduction zones for external oceans during Gondwana assembly) with distinct continental margins (young continental margins for Rodinia vs. old continental margins for Gondwana) could reconcile the Nd-Hf isotopic signatures (Spencer et al., 2013; Martin et al., 2020, Fig. 5). We further propose that juxtaposed juvenile continental arcs in the dual-sided subduction system of Rodinia were characterized by thin arc crust due to subduction retreat and consequent outboard arc migration, whereas juvenile continental arcs in the single-sided subduction system of Gondwana were gradually thickened with subduction advancing and inboard arc migration (Fig. 5; e.g., Collins et al., 2011). In this scenario, the mafic volcanic rocks produced during Gondwana assembly should have higher La/Zr ratios reflecting lower degrees of melting of the mantle wedge due to the overlying thick arc crust compared to prior and subsequent supercontinents. Thus, La/Zr ratio of continental-arc mafic volcanic rocks indicates thicker continental arcs with higher elevations during Gondwana assembly compared with those associated with Rodinia assembly.

4. Conclusions

The higher La/Zr ratios of continental-arc mafic volcanic rocks are found during Gondwana assembly compared with those during Rodinia and Pangea assembly. Thermodynamic modeling shows that La/Zr

ratios of continental-arc magmas are predominantly controlled by melting depth in the mantle wedge, which is directly tied to crustal thickness of the overlying continental arc. We suggest that thicker continental arcs during Gondwana assembly compared to those associated with prior and subsequent supercontinents account for the observed La/Zr variation trend in continental-arc mafic volcanic rocks, implying contrasting topography between the Rodinia and Gondwana supercontinents. Greater elevation of Gondwana continental arcs had likely led to more intense weathering, erosion, and run-off, with consequences for atmosphere and ocean compositions, and climate.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgment

This study was supported by the National Natural Science Foundation of China (42272228 and 41972242), Fok Ying Tung Education Foundation (171013) and Guangdong Major Project of Basic and Applied Basic Research (2019B030302013). PAC acknowledges support from ARC grant FL160100168. AB participation was supported by NSERC Discovery and Accelerator grants and ACS PF grant 624840ND2. We gratefully acknowledge Di-Cheng Zhu for his expeditious editorial handling. The two anonymous reviewers are thanked for their constructive comments and suggestions, which helped us clarify the idea and improve the manuscript significantly. This is contribution No. IS-3317 from GIGCAS.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.lithos.2023.107094>.

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