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A red bole zircon record of cryptic silicic volcanism in the Deccan Traps, India

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ABSTRACT

Silicic magmas within large igneous provinces (LIPs) are understudied relative to volumetrically dominant mafic magmas despite their prevalence and possible contribution to LIP-induced environmental degradation. In the 66 Ma Deccan LIP (India), evolved magmatism is documented, but its geographic distribution, duration, and significance remain poorly understood. Zircons deposited in weathered Deccan lava flow tops ("red boles") offer a means of indirectly studying potentially widespread, silicic, explosive volcanism spanning the entire period of flood basalt eruptions. We explored this record through analysis of trace elements and Hf isotopes in zircon crystals previously dated by U–Pb geochronology. Our results show that zircon populations within individual red boles fingerprint distinct volcanic sources that likely developed in an intraplate setting on cratonic Indian lithosphere. However, our red bole zircon geochemical and isotopic characteristics do not match those from previously studied silicic magmatic centers, indicating that they must derive from yet undiscovered or understudied volcanic centers associated with the Deccan LIP.

INTRODUCTION

Silicic (>60% SiO₂) igneous rocks are commonly found within dominantly mafic large igneous provinces (LIPs), but their overall relationship to the development of LIPs is poorly understood (Bryan et al., 2002). In particular, the potential for silicic magmas to play an outsized role in driving environmental degradation during LIP emplacement is underappreciated. Because these magmas can release volatiles that reach the upper atmosphere during explosive eruptions (Robock, 2000), knowledge of the frequency and volumes of silicic volcanism is needed for a complete understanding of the relationship between LIP emplacement and mass extinctions (Courtillot and Renne, 2003; Kasbohm et al.,

The Deccan Traps (DT; Fig. 1) represent the flood basalt component of a LIP emplaced at ca. 66 Ma due to the intersection of the rising Réunion mantle plume with the Indian continental lithosphere (Glišović and Forte, 2017). A geochronologic and paleomagnetic timeline for DT eruptions established their temporal correlation with the end-Cretaceous mass extinction, raising the question of whether volcanism, in addition to the Chicxulub impact, was an important

contributor to the extinction event (Chenet et al., 2007; Schoene et al., 2019; Sprain et al., 2019; Hull et al., 2020). The Deccan LIP has long been known to have silicic members (e.g., Mathur et al., 1926), but they have been poorly studied compared to the basalts (e.g., Lightfoot et al., 1987; Sheth and Melluso, 2008; Sheth et al., 2011; Cucciniello et al., 2019). Recent geochronology suggests synchronicity of some of these bodies with the main phase of DT basalts (Parisio et al., 2016; Basu et al., 2020a; Sahoo et al., 2020), which indicates that silicic Deccan LIP activity was more extensive than previously recognized.

We explored the character and extent of explosive silicic volcanism coincident with the emplacement of DT flood basalts by leveraging volcanic zircons deposited throughout the basalt stratigraphy. Some deposits interbedded within the lava flows, particularly red-colored interflow horizons ("red boles"), contain zircons that record both the time of crystallization and compositional characteristics of the source magmas. U–Pb geochronology of hundreds of these zircons has been used to constrain the age of interbedded basalts (Schoene et al., 2015, 2019; Eddy et al., 2020) by inferring that the zircons

originated as airfall from evolved (andesitic to rhyolitic) volcanic sources rather than eolian or fluvial transport or direct crystallization in basaltic magmas. However, the volcanic sources of these zircons remain poorly understood. We present trace element and hafnium (Hf) isotopic compositions of previously dated zircon crystals to provide new constraints on silicic volcanism within the Deccan LIP and for comparison with the limited number of silicic magmatic centers thus far characterized by U-Pb zircon geochronology and Hf isotopes (Basu et al., 2020a). Our results are consistent with distinct volcanic origins for the zircons in red bole horizons, though their geochemical characteristics do not match well any of the considered silicic magmatic centers in western India.

SAMPLES AND METHODS

We analyzed residual dissolved material from Deccan zircons separated from samples collected in the Western Ghats and on the margin of the Malwa Plateau (Fig. 1) that had been previously dated by U-Pb geochronology (Schoene et al., 2015, 2019; Eddy et al., 2020). Each solution represented the residue after chromatographic separation of U and Pb from a single zircon crystal. We split this material into two aliquots that were then analyzed separately for trace elements (Schoene et al., 2010) and for radiogenic Hf isotope ratios (reported here as $\epsilon_{Hf},$ i.e., the deviation of age-corrected $^{176}Hf/^{177}Hf$ from the chondritic uniform reservoir [CHUR] model composition in parts per 10,000). The analysis included euhedral crystals isolated from (1) red boles, (2) an \sim 40-cm-thick tephra in the Mahabaleshwar Formation (samples DEC13-08/09/10), and (3) a single coarse-grained segregation within a basaltic flow in the Jawhar Formation (DEC13-30); we excluded xenocrystic/detrital zircon predating the end-Cretaceous (\sim 5% of zircons dated). For comparison, we

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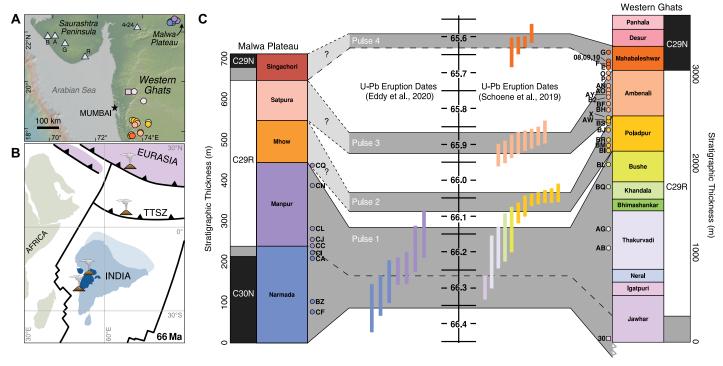


Figure 1. Red bole geochronology of the Deccan Traps (India). (A) Sample locations; circles and squares, color-coded by basalt formation—red boles, tephra, coarse basalt (Schoene et al., 2015, 2019; Eddy et al., 2020); triangles—silicic magmatic complexes in Gujarat (B–Barda, A–Alech, G–Girnar, R–Rajula, 4-24–Phenai Mata; Basu et al., 2020a). (B) Paleogeography of India at ca. 66 Ma. TTSZ—Trans-Tethyan subduction zone. (C) Basalt stratigraphy in the southern Malwa Plateau and the Western Ghats. Circles mark stratigraphic positions of zircon-bearing samples; vertical bars represent their U–Pb age estimates; gray backgrounds show a tentative correlation of eruptive pulses across the two areas (modified from Eddy et al., 2020).

included zircons from several magmatic centers in the Saurashtra Peninsula and the Phenai Mata complex in western India previously dated by Basu et al. (2020a; Fig. 1). Approximately 34% of the zircon trace element data (Schoene et al., 2015; Eddy et al., 2020) and ~14% of the Hf isotope data (Basu et al., 2020a) were previously published; the combined data set and details of the analytical protocols are presented in the Supplemental Material¹.

RESULTS

Zircon compositions (Figs. 2–4) have large variability overall but exhibit secular stratigraphic change (Fig. 2) and geochemically distinct populations within individual samples (Fig. 3). Lower Deccan red bole zircons display scattered $\varepsilon_{\rm Hf}$ values of -20 to +5, followed by a precipitous drop to values of -35 to -20 within the Ambenali Formation. While some trace element (TE) ratios (e.g., Lu/Gd; Fig. 2B) record weak trends stratigraphically, they do not mirror the distinct secular trend in Hf isotopes. Instead, zircon TE compositions

commonly cluster for individual samples when plotted against $\epsilon_{\rm Hf}$ (Fig. 3), indicating that zircon populations from individual red boles have distinct sources. Zircons from candidate source magmatic centers (Basu et al., 2020a) do not match those recovered from the Deccan basaltic stratigraphy, except for samples from the Girnar magmatic complex (Basu et al.'s samples G1–G9) that exhibit partial overlap in isotopic, age, and geochemical characteristics.

VOLCANIC IDENTITY OF DECCAN ZIRCON

Nearly all zircons analyzed in this study were sampled from horizons called red boles that occur between DT basalt flows. These prominent horizons are intervals of fine-grained material thought to develop through a range of processes that are dominated by in situ weathering of underlying basalt, but may include contributions of volcanic airfall or sedimentary material (Widdowson et al., 1997; Ghosh et al., 2006; Duraiswami et al., 2020). Schoene et al. (2019) found that \sim 20% of sampled red boles contained zircons, which were typically euhedral and often had morphologies that were unique to a particular horizon. U-Pb geochronology revealed age spectra typical of volcanic fallout, with \sim 5% pre-Deccan (>69 Ma) ages and eruption age estimates younging upward stratigraphically (Fig. 2C). Considering that detrital (eolian or fluvial) sources are generally associated with complex zircon age spectra, and that generation of zircon-bearing detritus on the slopes of a basaltic shield volcano is unlikely (Schoene et al., 2021), it was previously inferred that the red bole zircons were largely derived from volcanic fallout.

Our new geochemical and isotopic data are consistent with most zircon populations in individual red boles having originated as airfall from distinct eruptions (Fig. 3). While outliers are observed, some defining observations can be made in regard to each basalt formation (Figs. 2 and 3).

In the Mahabaleshwar Formation, three samples from different heights within the same 40-cm-thick tephra horizon yielded indistinguishable $\epsilon_{\rm Hf}$ and Lu/Gd ratios. While $\epsilon_{\rm Hf}$ is identical for all samples in this formation, suggesting an origin from a common magmatic center (see the Supplemental Material), Lu/Gd from sample E has much lower variance, and sample G trends toward higher values.

Zircons from the Poladpur Formation have similar age spectra and eruption age estimates, as do those from the Ambenali Formation. However, $\epsilon_{\rm Hf}$ and Lu/Gd ratios distinguish the zircon populations of each red bole sample, with the exception of samples BI, BM, and BR, which are indistinguishable. Several samples have bi- or polymodal distributions (e.g., BS, BH, and X), which is indicative of distinct sources, such as

¹Supplemental Material. Data tables, analytical protocols, and additional discussion. Please visit https://doi.org/10.1130/GEOL.S.17139284 to access the supplemental material, and contact editing@geosociety.org with any questions.

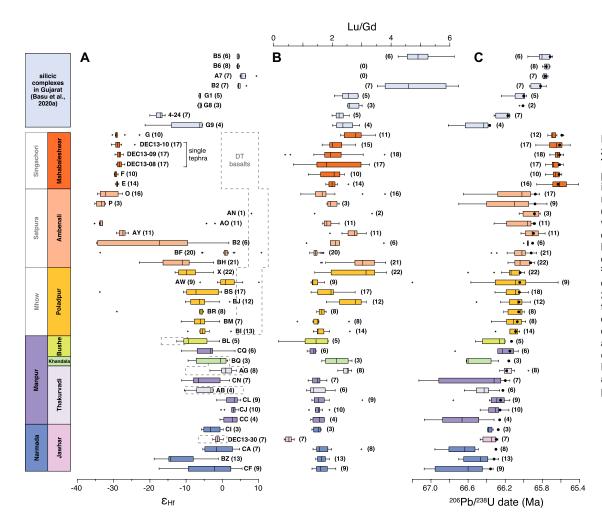


Figure 2. Compositions of zircon throughout Deccan Traps (DT) basalt stratigraphy compared to candidate intraplate sources in Gujarat (grouped by sample). (A) Hf isotope ratios (ε_{Hf}) with DT basalt ε_μ, (Basu et al., 2020b) shown as background boxes; (B) elemental Lu/Gd; and (C) ²⁰⁶Pb/²³⁸U dates (Schoene et al., 2019: Basu et al., 2020a; Eddy et al., 2020); the preferred eruption/ emplacement ages from each study are shown as dots. Boxes are colorcoded by DT formation. Number οf crystals analyzed is listed in parentheses.

multiple ashfall events within the time interval of red bole formation.

The lowermost formations in the Western Ghats are also easily distinguishable based on geochemistry, whereas in the Malwa Plateau, zircon populations from adjacent red boles tend to overlap compositionally. For example, red boles CC, CJ, and CL (Manpur Formation) show overlapping scatter, as do CN and CQ, which suggests there was no general change in the zircon source area over these intervals despite age populations that young upward stratigraphically.

The red bole zircon record is most consistent with volcanic airfall from distinct magmatic sources that change through time. This could result from variations in magma composition between consecutive eruptions from one magmatic center (particularly where ε_{Hf} remains unchanged), shifts to a separate volcano, or a general change of source area. However, considering that hiatuses in basaltic eruptions may have represented the bulk of the LIP's timeline, we cannot exclude some amount of reworking of originally volcanic zircons provided they are not transported far (given that the dated crystals were overwhelmingly euhedral).

CHARACTER AND LOCATION OF VOLCANIC SOURCES

The Hf and TE ratios also hold first-order clues about the geodynamic settings and locations of their source volcanoes. First, zircon Hf isotope ratios range from values that are generally in line with the ε_{Hf} of the coeval DT basalt formations (Jawhar through Bushe Formation) to values that are markedly lower (Poladpur-Mahabaleshwar Formation; Fig. 2A). While closedsystem DT basalt differentiation could plausibly explain the zircon $\epsilon_{\! Hf}$ values in the Bushe Formation and below, zircon ε_{Hf} of < -10 above the Bushe Formation requires large contributions of old (low- ε_{Hf}) crustal material in the source, perhaps bordering on pure crustal melting. Second, some zircon TE ratios are thought to be indicative of the tectono-magmatic setting of source melts (e.g., Grimes et al., 2015; Fig. 4B). Here, with the exception of the peculiar high-U "basaltic" zircons from sample DEC13-30, all red bole and tephra zircons span the range of compositions described by Grimes et al. (2015) as characteristic of intraplate ("ocean island-type") to continental arc-type volcanism.

An intraplate source would imply that the silicic magmas were genetically related to the activity of the Réunion hotspot and the Deccan

basalts, and therefore were likely emplaced into Indian continental crust. This scenario would be supported by the isotope ratios; the crust underlying the Deccan LIP is composed of old Precambrian cratonic domains (Ray et al., 2008; Bhaskar Rao et al., 2017) whose involvement would be sufficient to explain even the lowest ε_{Hf} observed. Some silicic rocks with an intraplate pedigree have been identified in discrete magmatic centers that are broadly associated with the major structural feature of the Narmada-Tapti-Son lineament in centralwestern India (Basu et al., 2020a). Outcrops of these complexes are dominated by plutonic and subvolcanic lithologies, with some explosive and effusive members (e.g., Sheikh et al., 2020; Sheth et al., 2021). While some evolved magma compositions have been attributed to near-closed-system differentiation of DT basalts (e.g., Cucciniello et al., 2019), radiogenic isotope ratios commonly indicate old crustal components (Chatterjee and Bhattacharji, 2001; Sheth and Melluso, 2008; Sheth et al., 2011; Basu et al., 2020a). Limited geochronologic data (Parisio et al., 2016; Basu et al., 2020a; Sahoo et al., 2020) reveal that many of these centers are in fact coeval with DT eruptions. Basu et al. (2020a) presented U-Pb zircon

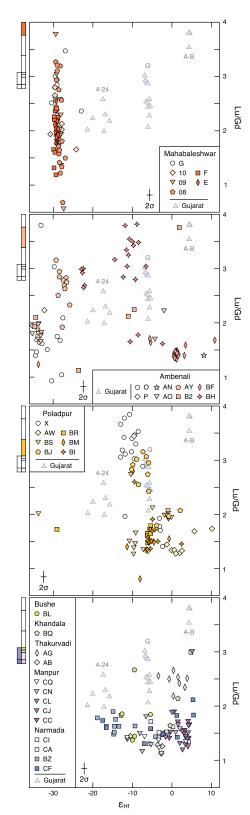


Figure 3. Hf-isotopic and Lu/Gd ratios of zircon from individual red boles compared to the analyzed silicic magmatic centers in Gujarat. Stratigraphic columns indicate the position of each sample group within the Deccan Traps.

geochronology and Hf isotopic compositions for zircon-bearing granophyres from five of these locations, though age and ϵ_{Hf} data rule

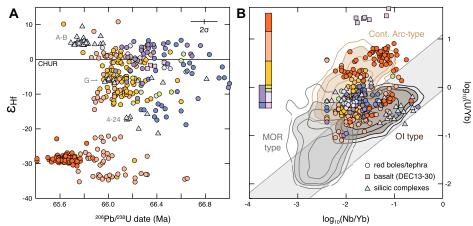


Figure 4. Compositions of Deccan zircons color-coded for basalt formation. (A) $\epsilon_{\rm Hf}$ variations with U–Pb date. CHUR—chondritic uniform reservoir. (B) Tectono-magmatic zircon provenance diagram outlining fields for mid-ocean ridge (MOR-type), ocean island and other plume-influenced (OI-type), and continental arc-type (Cont. Arc-type) zircon (Grimes et al., 2015).

them out as possible sources of most red bole zircons (Fig. 4A). One possible exception is zircon from the Girnar magmatic complex, which shows similarity to red bole zircons from the Poladpur Formation (e.g., sample BJ; Fig. 3).

An alternative to an intraplate source of the red bole zircons requires the presence of a volcanic arc active in the region at the time of DT emplacement. At 66 Ma, paleogeographic reconstructions place India at a latitude of 10-20°S, in the midst of its drift north toward the Eurasian continent (Fig. 1B). The nearest documented convergent margins at the time are the intra-oceanic Trans-Tethyan subduction zone located $\sim 10^{\circ}$ N, or ~ 2000 km north of India, and subduction beneath the Eurasian continental margin at $\sim 20^{\circ}$ N (Martin et al., 2020). Such distances are not entirely implausible for the deposition of zircon-bearing distal volcanic ash (e.g., \sim 1400 km in Smith et al. [2018]), but zircon delivery from these arcs to central India would have required repeated, large, explosive eruptions and a peculiar atmospheric circulation pattern. However, the observed low ε_{Hf} in red bole zircons effectively excludes intra-oceanic subduction sources (in the Trans-Tethyan subduction zone or elsewhere) due to the youthfulness and isotopically juvenile nature of all components involved in island arc petrogenesis (ϵ_{Hf} of +5 to +16 in the Kohistan-Ladakh Arc; Bouilhol et al., 2013). ε_{Hf} in continental arc magmas of the Eurasian margin recorded in the Gangdese arc and in Karakoram appear similarly juvenile at > +5 (Ravikant et al., 2009; Chu et al., 2011). Considering currently available data, we therefore find it unlikely that either of these two arcs was the source of the zircons found in Deccan red boles.

In summary, our analysis suggests that zircons contained in red bole horizons derive from intraplate magmatic centers located within cratonic India that were active synchronously with flood basalt volcanism. A mismatch between

geochemical and isotopic data from red boles and known magmatic centers, however, indicates that the exact source areas have yet to be identified.

CONCLUSIONS

Our database of ages, Hf-isotopic, and elemental compositions of red bole zircons identifies diverse explosive silicic volcanism coeval with the main phase of Deccan LIP flood basalt eruptions. Most red boles appear to contain zircons derived from single point sources or single eruptions. Given the distance to and isotopic incompatibility with the closest arcs at ca. 66 Ma, it is likely that the source magma bodies were associated with DT intraplate magmatism. However, comparison with our limited database from silicic magmatic centers in the region cannot yet fingerprint exact sources for the zircons interstratified in DT basalt flows. It is possible that continued work on zircons from intraplate silicic and alkaline magmatic centers in the Saurashtra Peninsula and Narmada rift areas will reveal candidate point sources. Alternatively, significant volcanic centers in southwest India have been lost to erosion or are now submerged due to subsequent rifting and could be located on the Indian continental margin or its counterpart, the Seychelles block. If so, incomplete surface exposures of silicic rocks may hint at a similarly incomplete basaltic record, hampering our ability to reconstruct basaltic fluxes, associated volatile release, and therefore models linking the Deccan LIP to climate change across the Cretaceous-Paleogene boundary.

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