



# Helium–oxygen–osmium isotopic and elemental constraints on the mantle sources of the Deccan Traps



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## ABSTRACT

The Deccan Traps, a 65 million-year-old continental flood basalt province located in western India, is the result of one of the largest short-lived magmatic events to have occurred on Earth. The nature and composition of its mantle source(s), however, have been difficult to resolve due to extensive assimilation of continental crust into the ascending Traps magmas. To circumvent this issue, using high-precision electron microprobe analysis, we have analyzed olivine grains from MgO-rich (up to 15.7 wt.%) lavas that likely erupted before substantial crustal assimilation occurred. We compare olivine, pyroxene and plagioclase mineral chemistry and He–O–Os isotope compositions with bulk rock major- and trace-element abundances and  $^{187}\text{Os}/^{188}\text{Os}$  for both bulk-rocks and mineral separates. Helium isotope compositions for the olivine grains generally show strong influence from crustal assimilation ( $<3 R_A$ ), but one ankaramite from the Pavagadh volcanic complex has a  $^3\text{He}/^4\text{He}$  ratio of 10.7  $R_A$ , which is slightly lower than the range of  $^3\text{He}/^4\text{He}$  measured for present-day Réunion Island volcanism ( $\sim 12\text{--}14 R_A$ ). Olivine-dominated mineral separates span a more restricted range in  $^{187}\text{Os}/^{188}\text{Os}$  (0.1267 to 0.1443) compared with their host lavas (0.1186 to 0.5010), with the separates reflecting a parental magma composition less affected by lithospheric or crustal interaction than for the bulk-rocks. Despite significant He–Os isotopic variations,  $\Delta^{17}\text{O}$  is relatively invariant ( $-0.008 \pm 0.014\text{‰}$ ) and indistinguishable from the bulk mantle, consistent with high- $^3\text{He}/^4\text{He}$  hotspots measured to-date.

Compositions of olivine grains indicate the presence of up to 25% of a pyroxenite source for Deccan parental magmas, in good agreement with  $\sim 20\%$  predicted from isotopic data for the same samples. Modeled pyroxenite signatures are similar to geochemical signatures expected to arise due to other types of mantle differentiation or due to assimilation of continental crust; however, we show that crustal assimilation cannot account for all of the compositional features of the olivine. Weak correlations exist between a global compilation of  $X_{\text{px}}$  (Deccan: 0.2–0.7) and  $^3\text{He}/^4\text{He}$ ,  $\delta^{18}\text{O}$  (Deccan olivine: 4.9–5.2‰) and  $^{187}\text{Os}/^{188}\text{Os}$ . Robust relationships between these parameters may be precluded due to a lack of two-reservoir source mixing, instead involving multiple mantle domains with distinct compositions, or because  $X_{\text{px}}$  may reflect both source features and crustal assimilation. Notwithstanding, geochemical similarities exist between Deccan Traps olivine ( $^3\text{He}/^4\text{He} = 10.7 R_A$ ;  $^{187}\text{Os}/^{188}\text{Os}_i = 0.1313 \pm 45, 2\sigma$ ) and Réunion igneous rocks ( $^3\text{He}/^4\text{He} = 12\text{--}14 R_A$ ;  $^{187}\text{Os}/^{188}\text{Os}_i = 0.1324 \pm 14$ ). These relationships imply that a characteristic geochemical ‘fingerprint’ may have persisted in the mantle plume that fed the Deccan Traps, since its inception at 65 Ma, to ongoing eruptions occurring on Réunion up to the present-day.

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## 1. Introduction

The elemental and isotopic compositions of basaltic lavas can reveal the nature of their mantle sources, including the identity

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of mantle lithologies involved in partial melting (e.g., Yaxley and Green, 1998), or evidence for the return of ancient subducted materials to Earth’s surface (Morgan, 1971; Hofmann and White, 1982; White and Hofmann, 1982). It has been shown that modification and metasomatism of peridotite mantle by subducted oceanic lithosphere may significantly alter the bulk composition of hotspot melts (Yaxley and Green, 1998), possibly resulting in the formation of mixed peridotite–pyroxenite/eclogite lithologies

(Hirschmann and Stolper, 1996; Sobolev et al., 2005, 2007; Day et al., 2009) with different bulk compositions than ambient peridotite mantle (e.g., Herzberg and O'Hara, 2002; Herzberg, 2011). Melting in the presence of these lithologies is thought to produce olivine with distinctive major and minor element compositions (Herzberg, 2011; Vidito et al., 2013; Herzberg et al., 2013, 2014). The degree to which these signatures correlate with olivine and bulk rock isotopic tracers, however, is poorly constrained.

A potential strategy to improve our understanding of these magmatic processes is to combine high-precision *in situ* elemental analyses with analysis of multiple radiogenic and stable isotope systems (e.g., He–O–Os). For example, noble gases provide evidence that primordial signatures exist in the mantle (e.g., Clarke et al., 1969; Lupton and Craig, 1975; Stuart et al., 2003) that have maintained high- $^3\text{He}/^4\text{He}$  (greater than the depleted upper mantle value of  $8 \pm 1 R_A$ ;  $R_A$  is the  $^3\text{He}/^4\text{He}$  ratio of air,  $1.38 \times 10^{-6}$ ; Graham, 2002) due to their high time-integrated  $^3\text{He}/(\text{U} + \text{Th})$  (e.g., Kurz et al., 1982; Allègre et al., 1983; Kaneoka, 1983; O'Nions and Oxburgh, 1983). In contrast, Os is highly compatible, resulting in low abundances in crustal materials and preferential incorporation into early-crystallizing mineral phases (e.g., Day, 2013). Consequently, the radiogenic  $^{187}\text{Re}$ – $^{187}\text{Os}$  system can provide robust, time-integrated information on the sources of mantle melts. Oxygen is a primary constituent of silicate and oxide minerals and a major element in mantle, crust and surface reservoirs. Oxygen isotopes ( $^{16}\text{O}$ ,  $^{17}\text{O}$  and  $^{18}\text{O}$ ) are mass dependently fractionated by both kinetic and equilibrium isotopic exchange processes, with fractionation decreasing as temperature increases (e.g., Bigeleisen and Mayer, 1947; Dauphas and Schauble, 2016). For this reason,  $^{18}\text{O}/^{16}\text{O}$  measurements can provide a sensitive record of relatively low temperature (<500 °C) hydrological and hydrothermal processes occurring in both the crust and lithosphere (Gregory and Taylor, 1981; Staudigel et al., 1995), which might then be recycled and returned at hotspot volcanoes (e.g., Eiler, 2001; Day et al., 2009). Combining these radiogenic and stable isotope data with mineral chemistry provides an opportunity to investigate mantle-derived lavas using a multivariate approach that tracks both petrological and geochemical processes.

Here we report the results of a multi-isotopic investigation into one of the largest short-lived magmatic events on Earth, the eruption of the Deccan Traps of western India. Deccan Traps lavas are well-characterized in terms of their major and trace element abundances and Sr–Nd–Pb isotope systematics, which reveal both crust- and MORB-like influences (Allègre et al., 1982; Mahoney et al., 1982; Melluso et al., 1995, 2006; Peng and Mahoney, 1995), as well as strong fractional crystallization and differentiation trends (e.g., Krishnamurthy, 1971), which ultimately serve to obscure information about the Deccan mantle source. To overcome this issue, we searched for olivine with relatively high forsterite contents ( $Fo > 80$ ) from olivine-phyric lavas for *in situ* elemental analyses. In other magmatic systems, high- $Fo$  (>88) olivines are thought to crystallize relatively early, prior to phases such as clinopyroxene, plagioclase and oxides. A limited number of basalts and picrites that display primitive geochemistry have been previously identified in the Deccan Traps (Krishnamurthy and Cox, 1977; Krishnamurthy et al., 2000). However, even  $Fo$ -rich olivines, when present in lavas that were erupted through thick crust and lithosphere, may have experienced late-stage modification that limits positive identification of their mantle source region(s) (Sobolev et al., 2007). Thus, using mutually complementary petrological and geochemical data represents an opportunity to identify the nature and composition of Deccan Traps parental magmas.

## 2. Samples and methods

Samples were collected from Gujarat and Maharashtra States in January 2014, and are from areas with lava flows and dikes associated with Deccan Traps magmatism (Table 1). Sample 47B (from the Pavagadh volcanic complex) is the sole ankaramite of the suite, and contains ~50% olivine and clinopyroxene phenocrysts that are relatively fresh despite matrix alteration. Although olivine, as the earliest crystallizing phase, is the best choice for studies determining the provenance of igneous rocks, not all samples contained a sufficient mass of olivine to complete all desired isotopic analyses. In these cases, clinopyroxene and plagioclase separates were substituted for He–Os isotope analyses, which require much more sample mass than O isotope analyses. Because He isotope analysis pulverizes separated minerals, we could measure the major and trace element contents of the crushed separates, referred to as “MAP” powders, to verify their mineral types and compare analyses with “fresh” powders. We found that Deccan olivine separates generally lack strong body color when observed under a binocular microscope, which in some instances made olivine difficult to distinguish from other phases and complicated efforts to obtain single-phase separates for destructive analyses (see *Supplementary Information* for more detail). Many mineral separates analyzed for O isotopes were not pure olivine separates, and we choose to focus our discussion on the two separates (from samples DC14-05 and –47B) that SEM analysis showed consisted of close to 100% olivine and yielded reliable olivine  $\delta^{18}\text{O}$  data and so enabled a faithful comparison with literature data. The remaining  $\delta^{18}\text{O}$  data for mixed mineral separates are reported in the *Supplementary Information*.

Bulk-rock major- and select trace-element abundance analyses were performed according to methods described in Peters et al. (2016) and Boyd and Mertzman (1987). Bulk-rock  $^{187}\text{Os}/^{188}\text{Os}$  and Re and Os abundance analyses were performed at the Scripps Isotope Geochemistry Laboratory using 1 g of homogenized powder according to methods described in Day et al. (2016), and were appropriately oxide, blank, and standard corrected. Rhenium, Ir, Ru, Pt and Pd abundances were determined by isotope-dilution using a iCAPq-c ICPMS, and Os abundance and  $^{187}\text{Os}/^{188}\text{Os}$  were determined by isotope-dilution using a ThermoScientific Triton thermal ionization mass spectrometer in negative ion mode. Olivine, pyroxene and plagioclase separate trace-element, HSE abundances and Os isotopes were determined using identical methods, but with between 0.1 and 0.4 g of powder, depending on the availability of clean mineral separates from each sample.

Helium isotope analyses were performed in the Fluid and Volatiles Laboratory at Scripps Institution of Oceanography. Gas inclusions containing He were released by crushing under vacuum for 150 s, which avoids release of structural He from the crystal lattice (Hilton et al., 1993; Graham et al., 1998; Scarsi, 2000). Procedural blanks were obtained by running the crusher without sample for the same period. The average blank was  $8.2 \times 10^{-11} \text{ cm}^3 \text{ } ^4\text{He}$  (STP), representing an average of 2.7%  $^4\text{He}$  released in the samples. Helium released by crushing was expanded into a stainless-steel gas preparation line and purified of Ne by exposing the sample gas to a Ti getter at 750 °C (30 min), cooling the getter to 400 °C, then sequentially expanding the gas into a liquid  $\text{N}_2$ -cooled charcoal trap, a SAES® Zr–Al getter at room temperature and a He-cooled charcoal trap. Following this purification, He was allowed into a dual collector noble gas mass spectrometer (MAP 215E) for static  $^3\text{He}/^4\text{He}$  analysis by peak jumping (Shaw et al., 2006). Neon concentrations were monitored to correct for contamination effects arising from imperfect instrument vacuum and/or for the presence of air in the mineral lattice (Hilton et al., 1992); total corrections to  $^3\text{He}$  were less than 1.5%. Raw sample ratios were normalized to SIO Pier air (1  $R_A$ ). Powders collected from

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