Lithos 252–253 (2016) 32–40

Contents lists available at ScienceDirect

Lithos

journal homepage: www.elsevier.com/locate/lithos

A proposed new approach and unified solution to old Pb paradoxes

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ARTICLE INFO

Article history:
Received 11 September 2015
Accepted 15 February 2016
Available online 27 February 2016

Keywords:
Mantle geochemistry
Marine carbonate recycling hypothesis
System of equations
Crustal recycling
OIB mantle sources

ABSTRACT

One of the most remarkable features of many and, perhaps, all oceanic basalts is that their Pb isotopic ratios ($^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$) are too radiogenic to be coming from the undifferentiated mantle or bulk silicate Earth. This has created three major concerns in the behavior of U, Th and Pb in the Earth's mantle that have been termed the Pb paradoxes. These are the unexpectedly long time-integrated high U/Pb (1st paradox), long time-integrated low Th/U (2nd paradox) and constant Ce/Pb and Nb/U (3rd paradox) in the mantle sources of oceanic basalts. The origins of such unexpected ratios have been the object of intense studies that produced several highly significant, but generally individualized results during the last four decades. Detailed analysis of available data shows that the paradoxes are closely interrelated as they all pertain to the mantle and have many common characteristic features. Thus, the Pb paradoxes constitute a system of equations that must be solved all together as each solution must satisfy every equation in the system. For example, compositional data for the voluminous mid-ocean ridge basalts (MORB) show that the 1st and 2nd paradoxes exhibit a long time-integrated enrichment of U and the Th/U and Nb/Th ratios are also constant. A single solution to simultaneously explain the paradoxes in MORB is possible if recycled materials with variable enrichments in incompatible trace elements, particularly U and its daughter Pb* plus Nb, Ce, and Th are added to or mixed with the depleted upper mantle. Significantly, a similar binary mixing solution has been proposed for the Pb paradoxes in ocean island basalts.

1. Introduction

The discovery of the highly radiogenic Pb (Fig. 1) and variable Sr isotopic ratios of young oceanic basalts indicates that the mantle has been compositionally heterogeneous for a long (b.y.) time (Gast et al., 1964). Continuing Sr–Nd–Pb isotopic studies show that to a first order, the heterogeneous mantle can be described in terms of four distinct end-member sources and that the compositional spectrum of oceanic basalts results mainly from variable degrees of partial melting and mixing of these end-members (Hart et al., 1992; Hofmann, 2003). The increase in $^{206}Pb/^{204}Pb$ is due to the decay of radioactive isotope ^{238}U over time and, thus, the highly radiogenic Pb isotopes of oceanic basalts, consisting of mid-ocean ridge basalts (MORB) and ocean island basalts (OIB), imply that one of the end-members has a long time-integrated U/Pb (more properly, high $^{238}U/^{204}Pb$ or high μ—HIMU) ratio that is much higher than the bulk silicate Earth (BSE) (Allegre, 1969; Allegre et al., 1986). Specifically, OIB with extremely radiogenic Pb isotopes from St. Helena, Mangaia and Tubuai islands are purportedly derived from a HIMU end-member source. The other proposed mantle end-member sources for OIB are the geochemically enriched mantle 1 (EM1) and enriched mantle 2 (EM2) whereas the geochemically depleted mantle is the proposed end-member source for MORB (DMM). As the bulk of oceanic basalts possess highly radiogenic Pb isotopic ratios that plot to the right of the geochron (Fig. 1), the proposed EM1, EM2 and DMM end-members also have the so-called HIMU effect (Vidal, 1992; Stracke et al., 2005; Castillo, 2015).

The high U/Pb ratio of the mantle sources of oceanic basalts is unexpected given that U is more incompatible than Pb and it is already enriched in the mantle-derived continental crust. This has given rise to a major concern regarding the concentration and behavior of U and Pb in the mantle that has been termed the main or 1st Pb paradox. Moreover, the decay of radioactive ^{232}Th increases $^{208}Pb/^{204}Pb$, which when plotted against $^{206}Pb/^{204}Pb$ ratios gives Th/U (or more properly $^{232}Th/^{238}U$ or κ) ratios for all oceanic basalts that are lower than the BSE κ value of ca. 4 (Tatsumoto, 1978; Galer and O'Nions, 1985; Elliott et al., 1999; Turcotte et al., 2001). Such low κ characteristics of oceanic basalts comprise the 2nd Pb paradox, which has also been termed 'kappa conundrum' (Elliott et al., 1999). Finally, Ce/Pb and Nb/U ratios have been observed to be constant (Hofmann et al., 1986). The constancy of the ratios is also unexpected given the known geochemical behaviors of Ce relative to Pb and Nb relative to U and, since both U and Pb are involved, has been termed the 3rd Pb paradox (Hofmann et al., 1986; Hofmann, 2003; Sims and DePaolo, 1997; Hart and Gaetani, 2006).

Significant efforts have been made to understand and solve the Pb paradoxes in the past three to four decades. However, the paradoxes were generally approached separately, resulting into a number of highly satisfactory and, thus, generally accepted albeit independent

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http://dx.doi.org/10.1016/j.lithos.2016.02.015
0024-4937/

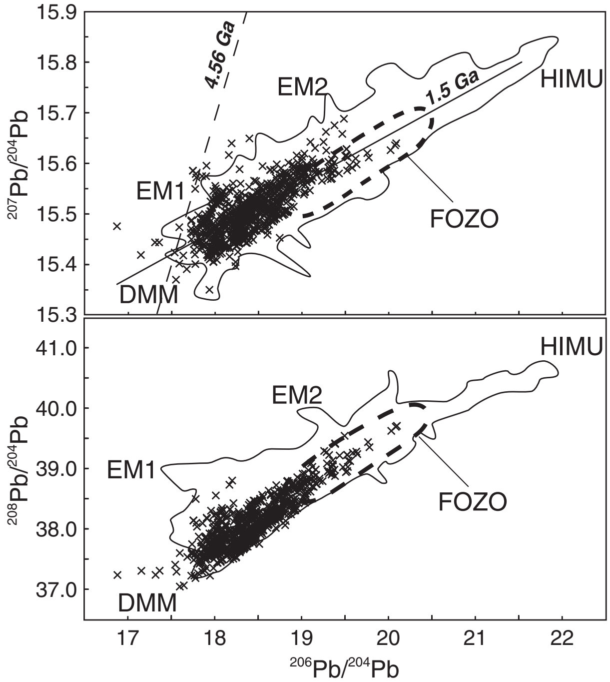

Fig. 1. (a) $^{207}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ and (b) $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ for oceanic basalts. Field for OIB is from Stracke (2012) and data for ALL MORB are from Gale et al. (2013). Also shown are 4.56 Ga geochron (White, 2010) and proposed DMM, EM1, EM2, HIMU, and FOZO mantle components (Hart et al., 1992). The ALL MORB $^{207}Pb/^{206}Pb$ array in (a) has a slope of 0.9146 and this gives an apparent age of 1.5 Ga.

explanations. Proposed solutions to the 1st paradox include the transfer of Pb into the core (Oversby and Ringwood, 1971; Vollmer, 1977; Vidal and Dosso, 1978; Allegre et al., 1986), preferential retention of Pb relative to U in the lower continental crust or subcontinental lithosphere (e.g., Doe and Zartman, 1979; Kramers and Tolstikhin, 1997) or in residual mantle sulfide (Hofmann, 2003; Hart and Gaetani, 2006), hydrothermal transfer of Pb from mantle to continental crust (Peucker-Ehrenbrink et al., 1994; Chauvel et al., 1995; Hofmann, 2003), preferential recycling of U into the mantle since Early Proterozoic (e.g., Staudigel et al., 1995; Hofmann, 2003), and crustal contamination of an early-formed (roughly 4.55–4.45 Ga) terrestrial mantle reservoir (Jackson et al., 2010). By and large, majority of the proposed solutions center on the decrease in Pb, not on the increase in U, in generating the 1st Pb paradox (Hofmann, 2003). Consequently, a number of later efforts to solve the 1st paradox have shifted focus to the search of the 'low μ' reservoir that complements the HIMU source (Murphy et al., 2003; Hofmann, 2003; Burton et al., 2012).

Proposed solutions for the 2nd paradox, on the other hand, center on the relative behavior of U to Th. These include a simple two-stage Th/U depletion model of the upper mantle (Galer and O'Nions, 1985) with a primitive, first stage κ value of BSE of ca. 4 changing abruptly to a second stage value of DMM at ca. 2.5, a continuous evolution model in which κ started from a reasonable BSE composition but decreased with time due to continental extraction (Tatsumoto, 1978; Allegre et al., 1986), and the aforementioned preferential recycling of U into the mantle due to oxidizing conditions at the Earth's surface since Early Proterozoic (Elliott et al., 1999; Andersen et al., 2015).

Finally, the constancy of Ce/Pb and Nb/U ratios or 3rd Pb paradox has been proposed as due to homogenization of the mantle through convective stirring later in Earth's history (Hofmann et al., 1986; Hofmann, 2003). Alternatively, it is due to similar silicate mantle-melt partition coefficient values (K_d) for Ce and Pb as well as for Nb and U during magmatic processes although most experimental studies show that Pb and U are more incompatible than Ce and Nb, respectively, in silicates (Sims and DePaolo, 1997), or for Ce/Pb (or Nd/Pb), retention of Pb in residual mantle sulfide during melting such that the bulk partitioning of Pb becomes equal to that of Ce (or Nd—Hart and Gaetani, 2006).

The main objective of this manuscript is to propose a new approach and a unified solution to the Pb paradoxes in oceanic basalts. This manuscript differs from the numerous significant solutions that have been published in three major ways. First, in contrast to the majority of prior studies, it introduces as well as emphasizes that the paradoxes are intricately connected to each other and to the radiogenic Pb isotopic composition of oceanic basalts. This simultaneous approach, in turn, necessitates related solutions. Second, focusing on the upper mantle, the approach shows that a unified solution to the Pb paradoxes in MORB is possible through binary mixing between small amounts of carbonatitic silicate melts (or their metasomatically-generated mantle phases) variably enriched in components derived mainly from the subducted crust through very low degree partial melting of the subducted oceanic lithosphere or slab and DMM (or its melt). Finally, a similar solution to the Pb paradoxes in OIB has already been proposed by the marine carbonate recycling hypothesis (Castillo, 2015). Thus, binary mixing between depleted and enriched mantle components presents a unified solution to the Pb paradoxes in both MORB and OIB.

In summary, the manuscript presents simple ideas and does not contain quantitative modeling of the paradoxes. The proposed solution is also not new as binary mixing of mantle sources in one form or another to explain the heterogeneity of oceanic lavas has long been proposed. For example, an early model to explain the wide spectrum of OIB compositions argued for binary mixing between the primitive or enriched lower mantle and depleted upper mantle (Wasserburg and Depaolo, 1979). However, later studies have clearly shown that this simplistic global process is not the case and/or such mixing is only between enriched OIB sources or their melts and DMM or its melt (e.g., White et al., 1993; Hanan and Schilling, 1997). Moreover, the unified solution is specific to the Pb paradoxes; it does not address other major features of oceanic basalts including the presence of still quite primitive signature of noble gases in OIB and other geochemical variations, such as those, e.g., by Hf and Os isotopes. Although all these are very important and related issues, quite simply they are beyond the scope of this manuscript.

2. An alternative new approach to old Pb paradoxes

The first concept being advocated here is that the paradoxes represent ratios of elements comprising a universal set (i.e., entire mantle) and its individual subsets (i.e., proposed end-member sources of oceanic basalts). These ratios therefore are all interrelated and exhibit features of the set's population. For example, an alternative but, perhaps, overlooked way to express the 2nd paradox is that the mantle sources of oceanic basalts have U/Th ratios higher than that of BSE. This non-conventional expression shows the obvious common feature of the 1st and 2nd paradoxes—the numerator U is higher than both the denominators in U/Pb and U/Th. This, in turn, indicates that both paradoxes can reasonably and simply be solved by a long time-integrated increase of U in the mantle sources of oceanic basalts (Castillo, 2015). Of course, such a solution has already been proposed, such as the preferential recycling of U into the mantle due to oxidizing conditions at the Earth's surface since Early Proterozoic (Staudigel et al., 1995; Elliott et al., 1999; Hofmann, 2003; Andersen et al., 2015). It is important to note, however, that only a few of the preferential recycling of U proposals emphasize the direct connection between long-time integrated enrichment of U in the mantle on the one hand and both 1st and 2nd Pb paradoxes on the other (e.g., Zartman and Haines, 1988; Kramers and Tolstikhin, 1997; Castillo, 2015). In general, it is not the favored solution to the 1st paradox (Murphy et al., 2003; Hofmann, 2003; Hart and Gaetani, 2006; Burton et al., 2012). Moreover, it has never been proposed as part of the solution to the 3rd paradox.

Besides providing a unified solution to the 1st and 2nd paradoxes, a long time-integrated enrichment of U has three additional significant