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Th/U and other geochemical evidence for the Réunion plume sampling a less differentiated mantle domain

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Abstract

Like Mid-Ocean Ridge Basalts (MORB), most Ocean Island Basalts (OIB) record a Th/U decrease during the history of their source. Notable exceptions are lavas sampling EM1 mantle end-member, which have super-chondritic time-integrated Th/U ($[Th/U]_{Pb}$). An intermediate situation is observed for the Réunion plume, where both present-day [Th/U] and $[Th/U]_{Pb}$ are similar to the bulk Earth value (weight ratio ~3.9). If [Th/U] measured in basalts reliably reflects the source ratio, then the Réunion plume source could have evolved in closed system with respect to Th/U. Th/U decrease in the mantle has been ascribed either to early continental crust extraction, or to preferential recycling of U over Th since oxidizing conditions appeared at the surface of the planet (~2.2 Ga). The primitive-like Th/U signature of Réunion is best explained by the absence of subduction influence since at least the Archean. This possibility is consistent with the $^{187}Os/^{186}Os$ signature, which is the less radiogenic of ocean island shield basalts. In addition, the difference in $^{3}He/^{4}He$ between Réunion ($R/R_A \sim 12.5$) and high-$^{3}He/^{4}He$ plumes (Hawaii, Iceland) most likely reflects a Réunion source less depleted in U and Th, rather than more degassed. In Sr–Nd–Pb–Os–He isotope space, Réunion signature plots in the region where OIB arrays converge, suggesting that the Réunion plume samples a component common to OIB, which is neither the source of MORB, nor the region where subducted plates are stored. It is suggested that the Réunion plume taps an early-depleted mantle domain subsequently influenced little, or not at all by recycling processes.

Keywords: Réunion; thorium/uranium ratio; primitive mantle; differentiation

1. Introduction

Geochemical models initially divided the mantle into a lower primitive layer and an upper layer whose composition has been depleted in incompatible elements during continental crust formation. Isotopic variations

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seen in oceanic basalts were thus interpreted as reflecting mixing between these two reservoirs. This model, and in particular the existence of a primitive deep reservoir sampled by hotspot volcanism, has been questioned for the last thirty years. Hofmann and White [1] pointed out that most Ocean Island Basalts (OIB) have positive εNd, which is indicative of long-term depletion of the source, whereas the concentration of trace elements in their mantle source must be higher than primitive. They proposed that recycling of oceanic crust accounts for this observation, and also solves the lead paradox (the unexpected U/Pb increase in the mantle) raised by Allègre [2]. Although the un-degassed isotopic signature of many OIB suggests their source has been less processed than that of Mid-Ocean Ridge Basalts (MORB), Hofmann and White's recycling model became widely accepted. Moreover, it appeared later that recycling of underlying lithosphere and overlying sediments is required to explain the composition and great isotopic diversity of OIB. The debate about the survival of a less differentiated reservoir was reactivated when Hart et al. [3] and Farley et al. [4] showed that the isotopic signatures of individual islands (or island groups) converge towards a common composition, a feature that possibly reflects lower mantle entrained into plumes [3,5].

The volcanic chain linking Deccan trapps to Réunion island displays geophysical and geochemical

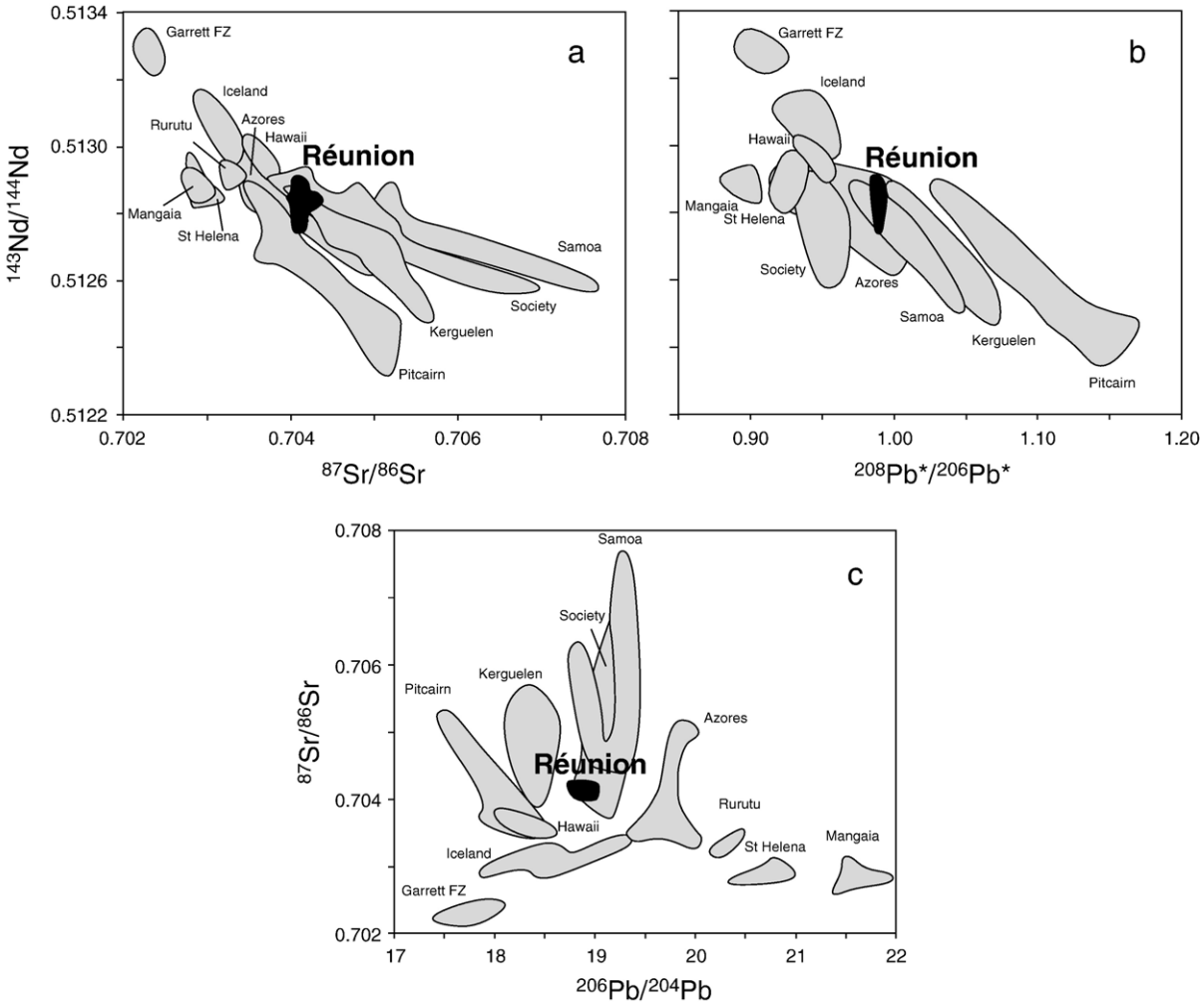

Fig. 1. Sr–Nd–Pb isotope signature of the Réunion plume. The Réunion isotopic field is inferred from the signature of the lavas erupted at Piton de la Fournaise since 0.53 Ma ([13,22] and unpublished data from D. Bosch). The signature of the depleted mantle, major plumes, and plumes sampling mantle end-components are shown for comparison. Samples whose compositions are used to represent the depleted mantle are from the Garrett fracture zone (East Pacific Rise, 13°28′S) [75]. Plumes isotopic fields have been drawn using most recently published data (references available on request). Data were compiled with the help of the GEOROC [76] and PETDB [77] databases.

characteristics which suggest that it results from a long-lived primary plume originating from the deep mantle [6]. Perhaps because of its homogeneity and intermediate isotopic signature, the Réunion plume source has been considerably less modeled than the sources of other primary hotspots (such as Hawaii and Iceland) or the sources exhibiting extreme isotopic signatures (mainly located in the south Pacific). The moderate Sr–Nd–Pb–He isotopic signature of Réunion lavas has been interpreted as resulting from simultaneous contributions of primitive material and recycled oceanic crust, or alternatively from ancient mantle differentiation [7–10]. In this study, we further explore the origin of the Réunion plume using the present-day and time-integrated Th/U ratios, which have been shown to vary systematically among mantle end-members [11]. Based on a new extensive data set (about one hundred samples measured for their Th–U content and Pb isotopic composition) covering the most recent activity (1998–2005) of Piton de la Fournaise ([12] and unpublished data), as well as

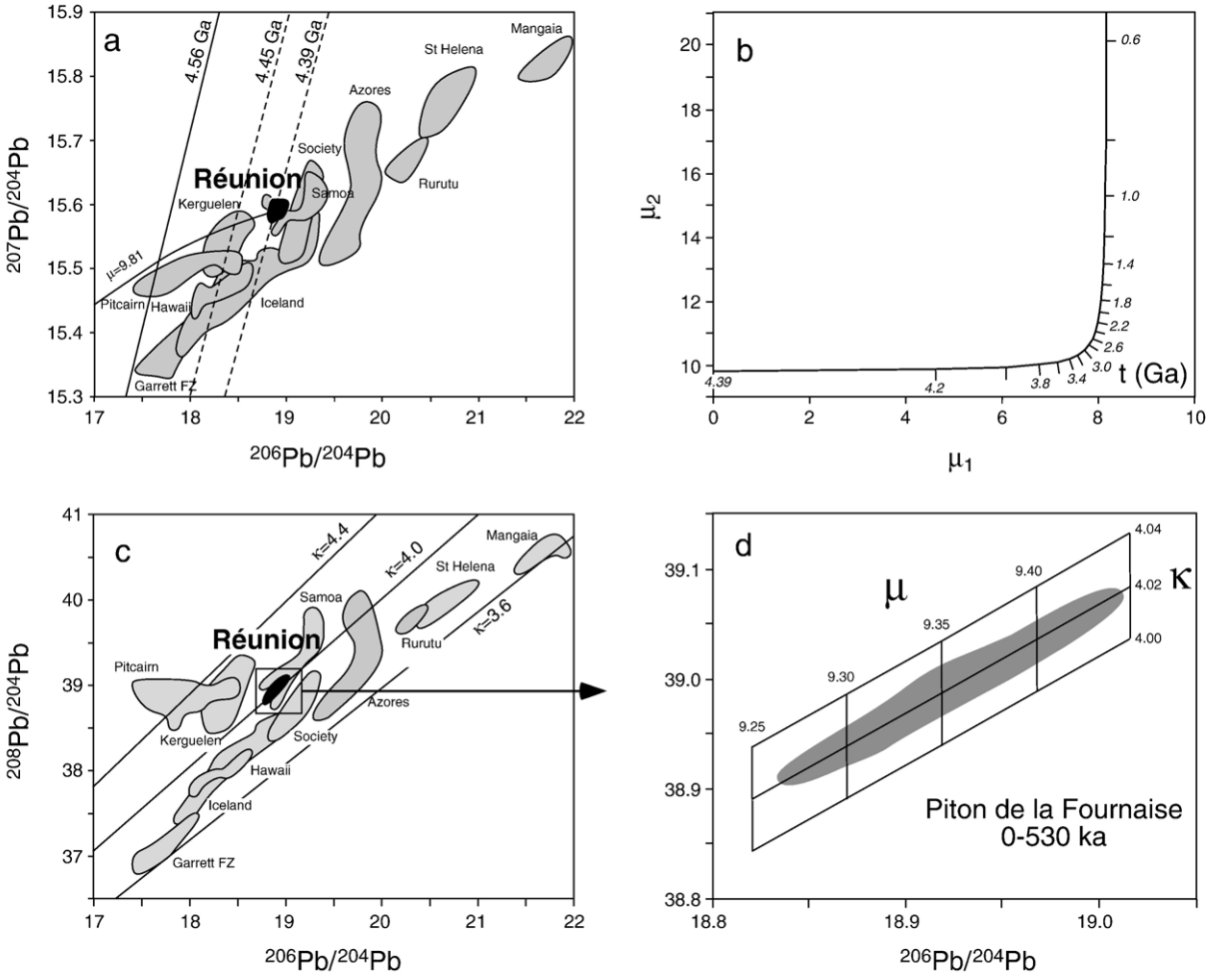

Fig. 2. Pb–Pb isotope signature of the Réunion plume. The Réunion isotopic field is inferred from the signature of the lavas erupted at Piton de la Fournaise since 0.53 Ma (unpublished data from D. Bosch). (a) The geochron (T=4.56 Ga, plain line) together with the 4.45 and 4.39 Ga isochrons (dashed lines) are shown. These lines represent the composition the primordial material would have today if it has evolved in closed system (with respect to U/Pb) since 4.56, 4.45 or 4.39 Ga. A single-stage isotopic growth curve starting 4.39 Ga ago from the composition of the Canyon Diablo meteorite and corresponding to a present-day $\mu=\{^{238}U/^{204}Pb\}$ of 9.81 is shown. (b) The $^{207}Pb/^{204}Pb$–$^{206}Pb/^{204}Pb$ signature of the Réunion plume is modeled by a two-stage evolution starting 4.56 Ga ago from the composition of the Canyon Diablo meteorite ($^{206}Pb/^{204}Pb$=9.307 and $^{207}Pb/^{204}Pb$=10.294) [78]. $\{^{238}U/^{204}Pb\}$ increased from μ_1 to μ_2 t years ago. The final composition used is that of recent lavas from Piton de la Fournaise ($^{206}Pb/^{204}Pb$=18.877 and $^{207}Pb/^{204}Pb$=15.590) [12]. The possible μ_1 and μ_2 values, together with corresponding t, are shown. (c) Each line indicates the composition the primordial material would have today if it evolved in closed system with respect to $\kappa=\{^{232}Th/^{238}U\}$ since 4.56 Ga. (d) In $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ space, Piton de la Fournaise data plot along a line of constant κ (~4.0) and varying μ.

recently available data covering the growth history of the volcano [13–16], it is shown that contrary to most other plumes, the Réunion plume could have preserved a primitive Th/U ratio. The possibilities that the Réunion plume originates from a less differentiated mantle domain are investigated.

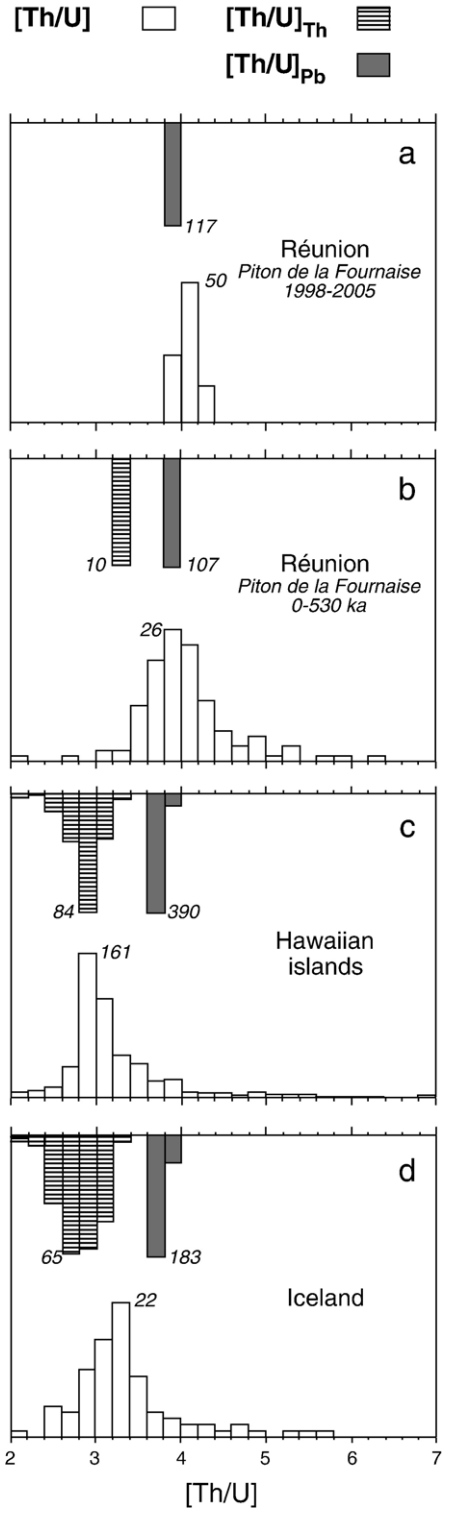

Fig. 3. Distributions of [Th/U], $[Th/U]_{Th}$, and $[Th/U]_{Pb}$ (weight ratios) in Piton de la Fournaise, Hawaii and Iceland lavas. $[Th/U]_{Th}$ is inferred from ($^{230}Th/^{232}Th$). $[Th/U]_{Pb}$ is the time-integrated Th/U ratio (single stage evolution with $T=4.56$ Ga) inferred from radiogenic $\{^{208}Pb/^{206}Pb\}$ (see text). Data source: (a) [12] and unpublished data, (b) [13,14] and unpublished data from D. Bosch, (c, d) [76]. Numbers on histograms indicate the highest frequencies.

2. Overview of the geochemical signature of the Réunion plume

The compositional evolution seen along the Deccan–Réunion volcanic chain, the youngest edifices having the less depleted signatures, has been ascribed to a reduction in the entrainment of asthenospheric material within the ascending plume, possibly reflecting the migration of the Central Indian Ridge away from the hotspot, or, alternatively, a decrease in plume productivity [17,18]. The lavas erupted during the shield building stage of the most recent edifices (Réunion and Mauritius Islands) are thus expected to carry the signature of the pure plume component. They display remarkably small compositional changes (for instance $^{87}Sr/^{86}Sr$ varies from 0.70397 to 0.70436) [10,13,19–21], suggesting a nearly homogeneous Réunion plume source over a period of 8 My. Although extreme homogeneity has been reported for He isotopes (R/R_A from 12 to 13.5) [7–9], high-precision studies have revealed significant isotopic shifts for Nd (εNd from +3.26 to +4.74) and Pb ($^{206}Pb/^{204}Pb$ from 18.79 to 19.01) during the growth history of Piton de la Fournaise [16,22], possibly reflecting a fine compositional zonation of the plume. Since Nd isotopes vary systematically through time, but Sr isotopes do not, $^{143}Nd/^{144}Nd$ and $^{87}Sr/^{86}Sr$ do not correlate at Piton de la Fournaise (Fig. 1a) [22]. $^{143}Nd/^{144}Nd$ does not correlate with radiogenic $^{208}Pb/^{206}Pb$, which is remarkably constant since 0.53 Ma (unpublished data from D. Bosch) (Fig. 1b). εHf is about +8 [20], but too few data exist (unpublished data from J. Blichert-Toft) to conclude about the behavior of hafnium isotopes at Piton de la Fournaise. ($^{230}Th/^{232}Th$) activity ratio is remarkably uniform (0.933 ± 0.009) since two millennia at Piton de la Fournaise [14], suggesting little change in the melting regime and source Th/U ratio.

For most isotopic systems (Sr, Nd, Pb, Hf, He) the Réunion plume signature is intermediate between mantle end-member compositions (Figs. 1 and 2). A possible exception, although based on only a single sample analysis [23], is Osmium, for which Réunion

shows the least radiogenic composition of ocean island shield basalts (see Fig. 4 of [24]). The isotopic signature of the Réunion plume may thus be similar to that of the component common to OIB (termed FOZO), as redefined by Hauri et al. [5,24].

3. Th/U estimates and notations

Mantle Th/U ratio may be estimated from three different approaches, including [Th/U], (^{230}Th/^{232}Th) or radiogenic {^{208}Pb/^{206}Pb}, where [ ] is for concentrations by weight, ( ) for activity ratios and { } for atomic ratios. Whereas [Th/U] and (^{230}Th/^{232}Th) give estimates of present-day mantle Th/U ratio, Pb isotopes provide a time-integrated value.

{^{232}Th/^{238}U}$_{Th}$, noted κ_{Th}, is inferred from ^{238}U–^{230}Th disequilibrium [25] according to:

$$\kappa_{Th} = \frac{\lambda^{238}/\lambda^{232}}{(^{230}Th/^{232}Th)}$$

where λ^{238} and λ^{232} are the decay constants for ^{238}U and ^{232}Th, respectively.

{^{232}Th/^{238}U}$_{Pb}$, noted κ_{Pb}, is the single-stage Th/U ratio recorded by Pb isotopes [26,27]:

$$\kappa_{Pb} = \frac{\{^{208}Pb\}^*}{\{^{206}Pb\}^*} \cdot \frac{e^{\lambda 238 \cdot T} - 1}{e^{\lambda 232 \cdot T} - 1}$$

$$\text{with } \frac{\{^{208}Pb\}^*}{\{^{206}Pb\}^*} = \frac{\left\{\frac{^{208}Pb}{^{204}Pb}\right\}_S - \left\{\frac{^{208}Pb}{^{204}Pb}\right\}_{CD}}{\left\{\frac{^{206}Pb}{^{204}Pb}\right\}_S - \left\{\frac{^{206}Pb}{^{204}Pb}\right\}_{CD}}$$

where T is the age of the system, {^{208}Pb}*/{^{206}Pb}* the radiogenic ratio, with subscripts S and CD referring to sample and Canyon Diablo meteorite, respectively.

To be compared to [Th/U], κ_{Th} and κ_{Pb} must be converted into weight ratios:

$$[Th/U]_{Th,Pb} = \frac{Ab^{238}}{Ab^{232}} \cdot \frac{W^{Th}}{W^{U}} \cdot \kappa_{Th,Pb}$$

where Ab^{238} and Ab^{232} are the natural abundances of ^{238}U and ^{232}Th; and W^U and W^{Th} the atomic weights of U and Th, respectively.

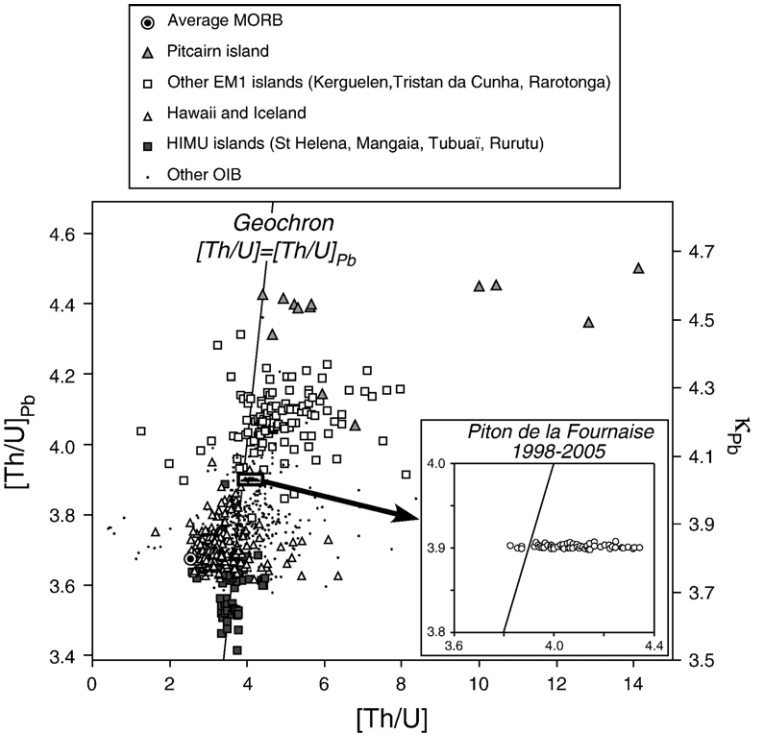

Fig. 4. Time-integrated Th/U versus present-day Th/U in Ocean Island Basalts (individual data). Correspondence between $[Th/U]_{Pb}$ weight ratio and {^{232}Th/^{238}U}$_{Pb}$ atomic ratio (noted κ_{Pb}) is indicated on y-axis. The average composition of MORB is also shown (from [35]). Iceland and Hawaii signatures plot dominantly to the left of the geochron, whereas EM1 islands plot dominantly to the right (data from [76]). The signature of the lavas recently (1998–2005) erupted at Piton de la Fournaise ([12] and unpublished data) plot on, or close to the geochron. Inset shows the absence of correlation between [Th/U] and $[Th/U]_{Pb}$ in these recent samples.

4. Th/U signature of the Réunion plume

Lavas erupted between 1998 and 2005 at Piton de la Fournaise have [Th/U]=4.07±0.11 (1σ) ([12] and unpublished data), which is in good agreement with the values previously reported for the last two millennia (3.95±0.08 (1σ)) [14] and for the last 530 ky (4.0±0.5 (1σ)) [13]. The time-integrated Th/U of these recent lavas is also close to 4 (κ_{Pb}=4.032±0.002 corresponding to $[Th/U]_{Pb}$=3.902±0.002) (Fig. 3a,b). The nearly perfect alignment of the data along the κ_{Pb}=4.0 line in $^{208}Pb/^{204}Pb$ vs. $^{206}Pb/^{204}Pb$ plot (Fig. 2c,d) indicates that $[Th/U]_{Pb}$ must have remained relatively constant during the last 0.53 Ma (unpublished data from D. Bosch). The absence of correlation between [Th/U] and $[Th/U]_{Pb}$ in 1998–2005 samples (Fig. 4), despite significant [Th/U] variations (from 3.83 to 4.32), indicates that a process fractionated Th/U shortly before eruption, and thus that the short-term [Th/U] heterogeneity is not a mantle feature.

Remarkably, both [Th/U] and $[Th/U]_{Pb}$ are similar to the chondritic value (weight ratio=3.9±0.2 [28]) in Piton de la Fournaise lavas. Despite greater [Th/U] dispersion, older lavas from Piton des Neiges ([29] and unpublished data from C. Deniel) and from the shield building stage of Mauritius Island [20] display a similar feature, supporting the idea that it is a robust fingerprint of the Réunion plume source. This feature is not shared by other major hotspots, such Hawaii or Iceland, where $[Th/U]<[Th/U]_{Pb}$ (Fig. 3c,d). The picture becomes more complex when considering $[Th/U]_{Th}$ (3.25±0.03 (1σ)) [14] which is much lower than both [Th/U] and $[Th/U]_{Pb}$ in recent lavas from Piton de la Fournaise.

5. Discussion

Albarède et al. [13] previously noted that Th/U in Piton de la Fournaise lavas is higher than in Hawaiian lavas, and suggested that U loss during hydrothermal alteration may account for the highest ratios measured in the oldest lavas from Rivière des Remparts. The analysis of very fresh samples (this study and [14]) and olivine-hosted melt inclusions [15] demonstrates that melts from the Réunion plume have [Th/U] close to 4.0. This value is intermediate between those measured in Hawaiian or Icelandic lavas (~3.2) and those of the EM1 lavas (>4.2), and is similar to that of bulk silicate Earth. Whether or not [Th/U] measured in lavas reliably reflects the source ratio is a long-standing debate (see [30] for a synthesis). If it does, then OIB spread on both sides of the geochron in the $[Th/U]_{Pb}$ vs. [Th/U] isochron plot, with Réunion plotting on or near to the geochron (Fig. 4). Conversely, if the source ratio is best recorded by $^{238}U–^{230}Th$ disequilibrium ($[Th/U]_{Th}$), as initially proposed by Allègre and Condomines [25], then the Réunion plume source as well as the mantle sources of nearly all oceanic basalts have $[Th/U]_{Th}<[Th/U]_{Pb}$ [27]. This point should be first discussed.

5.1. Mantle Th/U systematics

In the 1980s, when melting was considered to be an instantaneous event, the discovery of ^{230}Th excess in most oceanic basalts questioned the use of magmatic [Th/U] as a mantle proxy, and led to the idea that source Th/U is better inferred from ($^{230}Th/^{232}Th$) activity ratio [25–27]. However, it has remained difficult, if not impossible to reconcile the commonly measured ~20% ^{230}Th excess with the evidence that oceanic basalts, and in particular MORB, result from melting extents exceeding the critical value (about 1%) required to fractionate Th/U. Large Th/U fractionations inferred from $[Th/U]_{Th}$ have been tentatively explained by the presence of CO_2-rich melts [31], melting in the garnet stability field [32] or preferential melting of garnet lherzolites [33]. In another time-dependent approach, systematic $^{238}U–^{230}Th$ disequilibrium in oceanic basalts has been ascribed to ^{230}Th ingrowth during melting [34]. Considering a succession of finite melting steps, this model shows that pooled melt fractions must have ^{230}Th excess but not fractionated Th/U with respect to their source. A best estimate of present-day mantle Th/U ratios would thus be given by measured Th/U in the erupted lavas [35].

The two approaches have been examined by Elliott [30], who suggested that the ingrowth model accounts well for ^{230}Th excess when the rate of melting is slow and/or when the decompression melting column is tall. This idea is supported by the global inverse relationship between ^{230}Th excess and plume buoyancy flux [36]. If ^{230}Th has enough time for ingrowth during MORB formation, the situation is more problematic for OIB, which are thought to result from smaller degrees of melting within actively upwelling plumes. In a global compilation of OIB data, ^{230}Th excess does not correlate with partial melting extent (as inferred from the difference between measured Sm/Nd and Sm/Nd integrated by Nd isotopes), an observation that led Elliott [30] to suggest that ^{230}Th excess in OIB must include some ingrown ^{230}Th. In a detailed inspection of three OIB data sets (Hawaii, Iceland, Society), he concluded that the contribution of net Th/U fractionation (that is between source and extracted melts) to ^{230}Th excess is only evident in some differentiated lavas

from Hawaii, as previously proposed by Sims et al. [32]. A few years latter, Condomines and Sigmarsson [33] re-activated the debate. Based on the observation that (^{230}Th/^{232}Th) better correlates with ^{87}Sr/^{86}Sr than (^{238}U/^{232}Th) does, they proposed that measured (^{230}Th/^{232}Th) in melts reliably reflect (^{238}U/^{232}Th) of the mantle source. Thus, there is still no general agreement regarding which ratio, from (^{230}Th/^{232}Th) and [Th/U], better reflect the present-day Th/U ratio of the mantle.

Several lines of evidence indicate that alteration and extensive crystallization, rather than fractionation occurring during melting, dominantly limits the use of magmatic [Th/U] as a mantle proxy. For instance, [Th/U] often correlates with sample age or differentiation indices but rarely with trace element ratios sensitive to partial melting. In the case of fresh, tholeiitic or transitional OIB (erupted for example at Hawaii, Iceland or Réunion) magmatic [Th/U] is probably the most reliable estimate of mantle [Th/U] [30,35]. The existence of short-term [Th/U] heterogeneity, which is not a mantle feature (see Fig. 4), must however be taken into account (for instance through the use of averages). Conversely, in a global approach, it is safer to consider $[Th/U]_{Th}$ and [Th/U] as lower and upper bounds, respectively (Fig. 5). Unfortunately, the lavas having the highest [Th/U] and $[Th/U]_{Pb}$ ratios (Kerguelen and Pitcairn islands) are not documented for their ^{238}U–^{230}Th disequilibrium (mainly because of the rarity of young lavas) and only an upper bound of source Th/U ratio may be estimated. There is also no Th isotopic data from HIMU end-member, but trace elements and Pb isotopes suggest that both [Th/U] and $[Th/U]_{Pb}$ are low (Fig. 4).

As shown in Fig. 5, the main features of Th/U variations in the mantle are:

- As for MORB, the source of most OIB has lower Th/U compared to the value recorded by Pb isotopes. This feature may reflect the short residence time of Th and U in a low-Th/U upper mantle [26], or, alternatively, recycling of low-Th/U superficial material [37].
- The source of EM1 lavas has Th/U higher than the time-integrated value. The super-chondritic $[Th/U]_{Pb}$ (~4.4 in Pitcairn lavas) requires an increase of Th/U in the EM1 mantle reservoir, which is consistent with recycling of continental crustal material [38].
- The Th/U signature of the EM2 component plots near to the geochron, Society plotting slightly to the left ($[Th/U] < [Th/U]_{Pb}$) and Samoa plotting slightly to the right ($[Th/U]_{Th} > [Th/U]_{Pb}$). Interestingly, some lavas from Vailulu'u seamount, which samples the Samoan plume component, plot right on the geochron at [Th/U] ~4.0 ($\kappa \sim 4.1$) [39].
- The mantle array passing through the permissible ranges of [Th/U] yields a bulk Earth [Th/U] of ~3.9 ($\kappa \sim 4.0$), which is in agreement with meteorite constraints [28]. Modeling this array as an isochron yields a mean age of about 1.1 Ga for Th/U fractionation. This age is slightly older than that estimated for the MORB source (<1 Ga) [26,35], but this difference mainly reflects the lower bulk Earth κ (3.9 instead of 4.0) previously used. Most importantly, this age is much younger than the onset of Th/U decrease in the depleted mantle (>2 Ga) [37,40,41].
- The signature of Réunion lavas plots at the intersection of the mantle array with the geochron, suggesting their source has a near-primitive Th/U ratio.
- The signature of HIMU lavas probably plots below the main mantle array.

5.2. Evidence for the Réunion hotspot tapping a less differentiated mantle reservoir

5.2.1. A primitive Th/U signature

Given the relatively low buoyancy of the Réunion plume [42] and the characteristics of the recent lavas erupted at Piton de la Fournaise (transitional basalts derived from relatively large melting extents, about 7% [43]), the large ^{230}Th excess (20%) measured in historical samples [14] is best explained by the ingrowth model. In contrast, the slight difference between average [Th/U] (4.0) and average $[Th/U]_{Pb}$ (3.9) could reflect Th/U increase during melting. Thus, [Th/U] and $[Th/U]_{Pb}$ are consistent with the Réunion plume source having preserved a [Th/U] similar to that of bulk Earth (3.9±0.2 [28]). It is possible that the plume source had a complex history involving both periods of Th/U increase and periods of Th/U decrease, and that the primitive signature observed today is a pure coincidence. However, the most simple interpretation supported by the homogeneity of $[Th/U]_{Pb}$ (Fig. 2d), is that the Réunion plume source has evolved in closed-system with respect to Th/U. The possibility that the Réunion plume samples a remnant of primitive mantle can however be ruled out given the super-chondritic Sm/Nd and U/Pb time-integrated signatures and present-day Ce/Pb and Nb/U ratios. It is however possible that the Réunion plume samples a mantle domain that has undergone less differentiation than other plume sources.

Two dominant processes have been proposed to explain Th/U variations in the mantle: in the standard

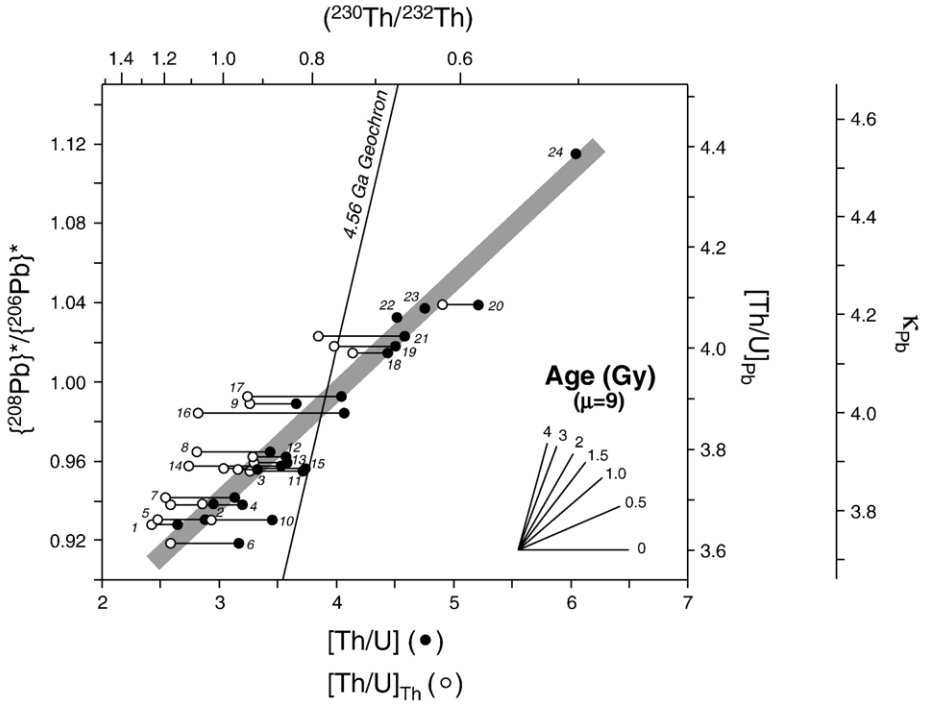

Fig. 5. Th/U systematic in Ocean Island Basalts (averaged data). Th/U integrated by Pb isotopes is plotted versus estimates of present-day Th/U. κ_{Pb} is the time-integrated $^{232}Th/^{238}U$ atomic ratio, which is inferred from $\{^{208}Pb\}^*/\{^{206}Pb\}^*$ assuming a single-stage evolution since 4.56 Ga (see text). $[Th/U]_{Pb}$ is the time-integrated weight ratio inferred from κ_{Pb}. Present-day [Th/U] of the mantle is constrained to range between magmatic [Th/U] ratio (closed circles) and the value derived from ($^{230}Th/^{232}Th$) activity ratio ($[Th/U]_{Th}$, open circles). For each locality, this permissible range is illustrated by a horizontal line connecting the two estimates. Th isotopes have not been measured in the samples having the highest magmatic Th/U ratios (EM1 lavas), but available [Th/U] data are nevertheless shown because they represent upper bounds. Our preferred mantle array (thick grey line), which takes into account the permissible range of [Th/U] for each locality, intercepts the geochron at [Th/U] ~ 3.9 ($\kappa \sim 4.0$), the chondritic value [28]. The slope–age relationship in this isochron diagram is inferred from:

$$\frac{\{^{208}Pb\}^*}{\{^{206}Pb\}^*} = \frac{\kappa_1\mu_1(\exp(\lambda^{232}\cdot T) - \exp(\lambda^{232}\cdot t)) + \kappa_2\mu_2(\exp(\lambda^{232}\cdot t) - 1)}{\mu_1(\exp(\lambda^{238}\cdot T) - \exp(\lambda^{238}\cdot t)) + \mu_2(\exp(\lambda^{238}\cdot t) - 1)}$$

where T=4.56 Ga, t is the age of Th/U fractionation, and λ^{232} and λ^{238} are the decay constants for ^{232}Th and ^{238}U. κ and μ are the $\{^{232}Th/^{238}U\}$ and $\{^{238}U/^{204}Pb\}$ atomic ratios, respectively. Subscripts 1 and 2 are relative to the intervals 4.56 Ga$-t$ and $t-0$ Ga, respectively. The slope–age relationship is shown for $\mu_1=\mu_2=9$ and $\kappa_1=4.0$. For localities 1 to 21 (with the exception of localities 3 and 16), data sets including Pb–Th isotopes and Th–U concentrations on the same samples have been considered (references available on request). For localities 22 to 24, data have been extracted from the GEOROC database [76], and outliers have been removed applying a 1σ filter. (1) Average MORB with [Th/U]<3.5 [77]; (2) Hawaii, Kilauea; (3) Hawaii, Loihi; (4) Iceland, Theistareykir; (5) Azores, Sao Jorge; (6) Azores, Terceira; (7) Azores, Pico; (8) Azores, Faial; (9) Azores, San Miguel; (10) Society, Moua Pihaa; (11) Society, Mehetia; (12) Society, Teahitia; (13) Society, Rocard; (14) Austral, McDonald; (15) Canary, Lanzarote; (16) Comoros, Karthala; (17) Réunion, Piton de la Fournaise; (18) Samoa, Vailulu'u; (19) Samoa, Savai'i; (20) Samoa, Malumalu, (21) Tristan da Cunha; (22) Inaccessible; (23) Kerguelen; (24) Pitcairn Island.

model, which is based on the difference of incompatibility of U and Th during melting, the upper mantle acquired a low Th/U ratio during continental crust extraction [26]. This model does however not explain the great Th/U heterogeneity of OIB. The second model is based on the difference of solubility of U and Th in aqueous environments. In order to explain the decrease of mantle Th/U through time (in particular since about 2.2 Ga), it has been proposed that U mobility increased when oxidizing conditions appeared at the surface of the Earth, resulting in a decrease of Th/U ratio in the alteration products that are ultimately recycled in the mantle [44,45]. This model, referred to as Post-Archean Uranium Recycling (PURE) [37], is very attractive since it can explain why most OIB showing geochemical imprints of recycling have today lower Th/U than their time-integrated value. In the standard model, the Th/U signature of the Réunion plume suggests that its source has not been influence by continental crust extraction, which is unlikely given trace element systematics (see

below). Within the frame of the PURE model, it can be suggested that the Réunion plume source has not been influenced by subducted components since at least the Archean. This possibility will be further tested using other geochemical data.

5.2.2. Other trace elements

Trace elements patterns from Réunion have been compared to those from localities sampling mantle end-members (Fig. 6), revealing the following first order distinctive features: (1) Enrichment in light rare earth elements is less than for lavas from Society, Pitcairn and Mangaia islands, and similar to alkali basalts from Hawaii. (2) Highly incompatible elements are slightly depleted relative to Nb and Ta. This depletion is however much less pronounced than for HIMU lavas. (3) The Th–U negative anomaly that characterizes Hawaiian and Icelandic lavas are absent. Trace element patterns from Réunion display some resemblance to the average OIB pattern, albeit with less pronounced enrichment in incompatible elements, and with a Ba depletion.

Hofmann et al. [46,47] explained trace element systematics in oceanic basalts in a two-stage model of mantle differentiation, which became widely accepted. In the first stage (referred to as "continental"),

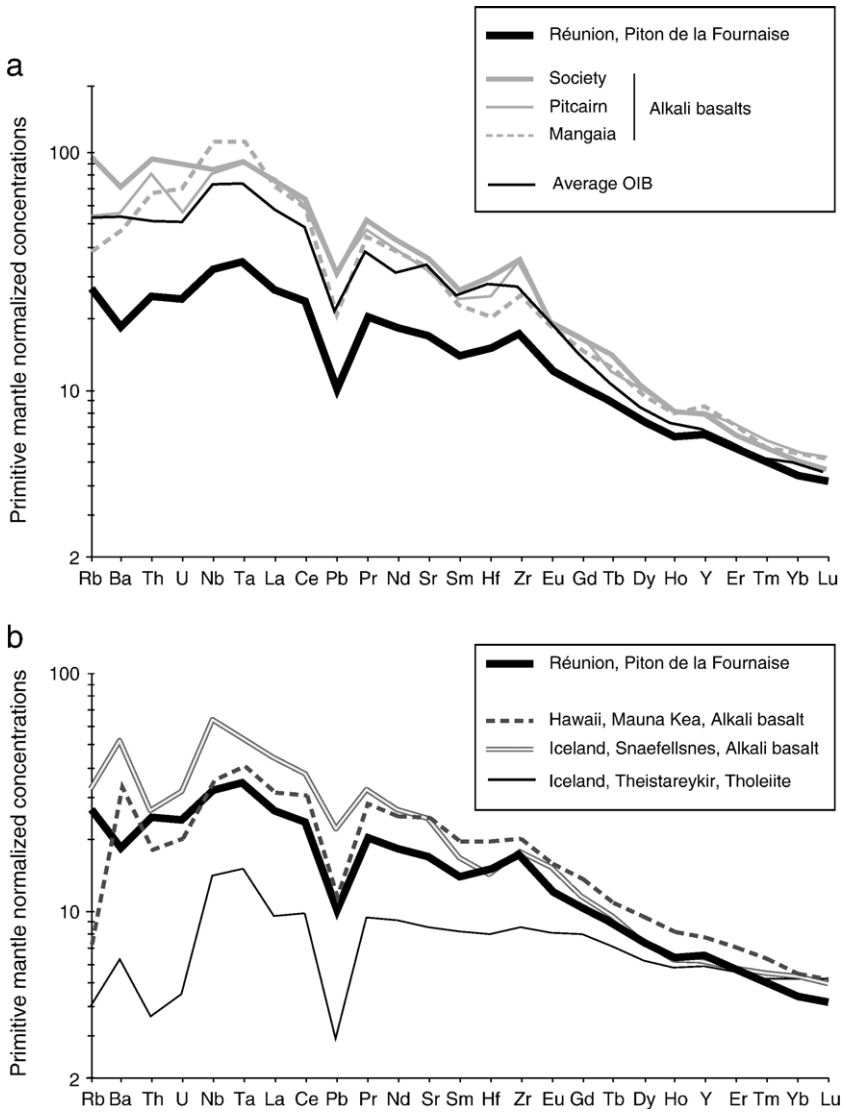

Fig. 6. Trace element pattern of Piton de la Fournaise lavas compared to patterns from other localities. Concentrations are normalized to the primitive mantle composition [79]. (a) The pattern of a recent sample from Piton de la Fournaise [12] is compared to patterns of mantle end-members (from [76]) and with the average pattern of OIB (from [80]). (b) Piton de la Fournaise pattern compared to those from Hawaii and Iceland (from [50–52]).

continental crust extraction depleted a part of the mantle, which was subsequently homogenized to form the source common to MORB and OIB. In the second stage (referred to as "oceanic"), this depleted reservoir internally differentiated into MORB and OIB sources through oceanic crust extraction and recycling at subduction zones. Hofmann's model is essentially based on two ratios, namely Ce/Pb and Nb/U, which were shown to strongly fractionate during the first stage and slightly during the second. Ce/Pb (26.9±2.1) and Nb/U (43.2±2.0) of recent lavas of Piton de la Fournaise are similar to the average values (25±5 and 47±10, respectively) common to MORB and OIB [46]. These far-from-primitive signatures indicate that the Réunion plume source, like other mantle reservoirs sampled by oceanic volcanism, did undergo the first, continental differentiation event. During the second stage, the recycling of oceanic lithosphere and overlying altered oceanic crust and sediments, introduced large compositional heterogeneities. Some trace element ratios were shown to record recycling of specific components. For example, recycling of oceanic crust may increase Ce/Pb and U/Pb (Pb loss), recycling of oceanic gabbros may decrease Th/La and U/La (Th and U loss), recycling of pelagic sediments may increase Ba/Th (Ba gain), and recycling of terrigenous sediments may decrease Ce/Pb (Pb gain) [48–50]. In particular the Ce/Pb ratio was found to be more sensitive to recycling of subducted components, and thus more variable than initially suggested by Hofmann et al. [46]. For instance, Chauvel et al. [11,51] noted values as low as 10 for Society basalts and Iceland picrites, and values as high as 50 for basalts from Tubuai. If the mean Ce/Pb value of oceanic basalts (~25) reflects that of the residual mantle after continental crust extraction and homogenization (i.e. Hofmann et al. [46] argument), then the Ce/Pb signature of the Reunion plume (~27) suggests that its source has been subsequently little affected by processes involving Pb loss or Pb gain. As previously observed [10], Réunion lavas have close to primitive Th/La ratios, and thus lack the Th–U negative anomaly identified in Hawaiian and Icelandic lavas, an anomaly ascribed to the presence of recycled oceanic gabbros in plume sources [50]. Although K/La is sub-chondritic in Réunion lavas (~300), it is nevertheless higher than at Hawaii (~240) [50] or Iceland (~260) [52] indicating that the Réunion source is less depleted in potassium. Again, the absence of recycled lithosphere stripped of its K in the Réunion source could account for this observation. Recycling of pelagic sediments in the Réunion source can also be ruled out based on the low Ba/La signature (see [10] for discussion about Ba depletion). It thus appears that the trace element signature of Réunion lavas lacks a clear imprint of recycled components in their source.

5.2.3. Isotopic constraints

5.2.3.1. Nd isotopes. The Sm–Nd isotopic system strongly constrains the timing and extent of silicate Earth differentiation because the initial compositions, both elemental and isotopic, of the two decay schemes (^{147}Sm–^{143}Nd, $T_{1/2}$=106 Gy; ^{146}Sm–^{142}Nd, $T_{1/2}$=103 My) have been preserved in some primitive meteorites. The positive ε^{143}Nd signature of most oceanic basalt indicates a long-term depletion of their mantle source, generally ascribed to early continental crust extraction. Recently, the discovery that the terrestrial ^{142}Nd/^{144}Nd signature, commonly used as a reference, is 20 ppm higher than that of chondritic meteorites questioned previous evolutions models based solely on ε^{143}Nd: the silicate earth must have differentiated very early, probably within the first 30 My of Earth's history, and the continental crust must have formed subsequently from an already depleted mantle [53]. According to the authors, early silicate earth differentiation implies (1) tiny ^{147}Sm/^{144}Nd fractionation. For example, increasing ^{147}Sm/^{144}Nd from 0.1966 to 0.21 4.53 Gy ago would raise present-day ε^{143}Nd up to ~+10, the MORB value. (2) In order to limit subsequent increase of ^{147}Sm/^{144}Nd in the mantle, continental crust formation must have affected a large portion of the mantle (~96%). Reconciling the recent developments of the Sm–Nd isotopic system with conventional model of mantle evolution is beyond the scope of this paper. For our concern, it is noted that the uncontaminated, primitive samples from the Deccan Province have the same ^{142}Nd/^{144}Nd signature as other terrestrial samples [54], indicating that the Réunion plume also samples the early depleted mantle reservoir. Thus, the short-lived ^{146}Sm–^{142}Nd chronometer suggests that (1) the Réunion mantle source acquired its slightly positive ε^{143}Nd very early, and (2) the degree of early ^{147}Sm/^{144}Nd fractionation was extremely low, so that the plume source composition may have remained very similar to that of the primitive mantle.

5.2.3.2. ^{206}Pb/^{204}Pb–^{207}Pb/^{204}Pb. As does that of most terrestrial samples, the Pb isotopic signature of Réunion lavas plots to the right of the 4.56 Ga geochron in the ^{207}Pb/^{204}Pb vs. ^{206}Pb/^{204}Pb diagram (Fig. 2a). Such a feature (often referred to as the first 'lead paradox') requires some U/Pb increase at some time

during the history of the accessible Earth. It is generally accepted that incorporation of Pb into the core during Earth accretion contributed to increase U/Pb in the mantle [55]. The bulk silicate earth Pb isotope composition is thus expected to lie to the right of the geochron, the compositional shift reflecting the accretion interval [56]. For the vast majority of OIB plotting to the right of the 4.45 Ga "core-corrected" geochron (Fig. 2a), additional U/Pb increase may have occurred subsequently through recycling of crustal material into the mantle, although the exact process involved is debated [49,57]. The Réunion signature plots on a 4.39 Ga isochron (Fig. 2a), raising the possibility that its source evolved with $\mu_1 \sim 0$ before 4.39 Ga and with $\mu_2 \sim 9.81$ after. This end-member scenario requires an accretion interval of 170 Ma, which is slightly greater than generally estimated [56]. Thus, an increase of U/Pb younger than core formation is likely. However, Fig. 2b shows that the later the U/Pb fractionation, the greater the increase. For example, U/Pb increase must be older than 3.8 Ga if $\mu_2 = 10$ and older than 2.4 Ga if $\mu_2 = 11$.

5.2.3.3. He, Ne, Ar. High $^3He/^4He$ in OIB has been commonly attributed to sampling a deep, less processed mantle reservoir, which kept a high concentration of primordial ^3He. The signature of Réunion lavas ($R/R_A \sim 13$), which is intermediate between MORB ($R/R_A \sim 8$) and the primitive helium mantle (PHEM, $R/R_A > 24$) [4], was thus interpreted as mixing between recycled and primitive components [7–9]. However, recent studies [58–61] have shown that the standard interpretation of He isotopic signatures is, to a first order, not correct. The main reason is that $^4He/^3He$ is nothing other than the time-integrated $(U+Th)/^3He$ ratio, which records not only ^3He loss but also U–Th heterogeneity. The importance of radiogenic ^4He in controlling $^3He/^4He$ variation is illustrated by a global relationship between $^3He/^4He$ and Th–U content of OIB [60,61]. The new model for the evolution of He isotopes [61] is consistent with the two-stage model of Hofmann et al. [46,47]: the source common to MORB and OIB, which is depleted in U–Th during continental crust extraction, evolved along a high $^3He/^4He$ curve. Subsequently, the superficial reservoir source of MORB, which loses more He than the deep reservoir source of OIB, progressively acquired a low $^3He/^4He$ signature. Formation of oceanic crust and recycling at subduction zones then introduced U–Th heterogeneities in the plume reservoir, thus generating various $^3He/^4He$ signatures. Since He is probably lost during subduction [62], the parameters that will control $^3He/^4He$ evolution of the plume source are the U–Th content of the material that is effectively recycled in the mantle, and the age of the heterogeneities. The relation between $^3He/^4He$ and Th content in OIB [61] suggests that the dominant parameter is the chemical heterogeneity. Recycling a U-rich crust–lithosphere assemblage may explain the low $^3He/^4He$ signature (~6–7 R/Ra) of HIMU lavas [63,64]. On the other hand, recycling of an oceanic lithosphere that has lost about 50% of its U budget during subduction [65] may lead to the high $^3He/^4He$ signature of many hotspot basalts [66]. For instance, the very high $^3He/^4He$ signature of some basalts from Hawaii or Iceland is consistent with recycling of U–Th depleted gabbros into their source [50,51]. Thus, considering the plume reservoir as uniformly degassed (i.e. constant ^3He concentration), the highest $^3He/^4He$ signatures found in OIB are best explained by a long-term U–Th depletion of their source, whereas intermediate $^3He/^4He$ values, as those found in Réunion, could result from closed-system evolution of $(U+Th)/^3He$.

The Ne–Ar isotope signature of Réunion is, as are helium isotopes, intermediate between MORB and Loihi-Iceland signature [9,67,68]. Because nucleogenic Ne ingrowth is coupled to the disintegration of Th and U isotopes, the intermediate signature of Réunion may be explained as is the He signature. Because $^{40}Ar/^{36}Ar$ is the time-integrated $^{40}K/^{36}Ar$ ratio, it is possible that the intermediate isotopic signature of the Réunion plume reflects an intermediate amount of K in its source. Indeed K/La indicates that the source of Iceland and Hawaii is depleted in K relative to the Réunion source, raising the possibility that the difference of Ar isotopic signature between these major hotspots reflect various amount of radiogenic ^{40}Ar in their source, rather than different degassing histories.

5.3. Origin of the Réunion plume

It has been a common practice to interpret the isotopic variations in OIB in terms of mixing mantle end-member compositions. However, this approach suffers from the lack of information about what these end-member compositions physically represent in the mantle. Nevertheless, it can be argued that because of its central signature in Sr–Nd–Pb isotope space, the Réunion plume results from mixing mantle end-member compositions. This possibility is highly unlikely because (1) the different components should have mixed perfectly and in constant proportions during the long life of the plume and, (2) any Sr–Nd–Pb mixing model is not compatible with the Os signature of the

Réunion plume, which is the least radiogenic of ocean island shield basalts [23,24].

Another way to look at OIB isotopic signature is to consider mantle processes, their extent, variety and age. For example, the moderately radiogenic Pb isotopic signature found in many OIB is unlikely to reflect the contribution of a very radiogenic, necessarily rare component in their source. Instead, such an intermediate signature may result from a similar process to that which produces the HIMU end-component (probably the recycling and storage of altered oceanic crust over time periods of 1.5–2 Ga) but occurring since more recently [69]. Although the Réunion plume has moderately radiogenic Pb, its elevated $^{207}Pb/^{206}Pb$ requiring an old U/Pb increase indicates that it is not sampling the young HIMU mantle.

Time-integrated Sm/Nd and U/Pb ratios, as well as Ce/Pb and Nb/U ratios are higher than primitive in Réunion lavas, ruling out the possibility that the Réunion plume samples a hypothetical undifferentiated mantle domain. Instead, the geochemical signature suggests sampling of an early-differentiated mantle domain that has been subsequently little influenced by recycling processes. The existence of such a reservoir has been previously invoked by Hofmann et al. [46,47] who proposed that early continental crust depleted a large fraction of the mantle, which was then homogenized, and that chemical heterogeneities were introduced subsequently through oceanic crust formation and recycling. The Ce/Pb and Nb/U ratios of Réunion lavas, close to the average values common to MORB and OIB, together with the homogeneity of the long-lived plume, are consistent with sampling the residual mantle resulting from continental crust extraction and homogenization. This scenario however requires that early crustal formation did not fractionate the Th/U ratio, which is supported by the relatively late (~2.5 Ga) decrease of Th/U in the mantle [37,40,41] and increase in continents [70].

The signature of Réunion lavas plots near to or within the isotopic field where OIB and MORB isotopic arrays converge [3–5,24,71], raising the possibility that the plume samples a source common to oceanic basalt. However, the signature of this common component is not precisely defined, and its origin is the object of a debate (see synthesis by Stracke et al. [72]). Possible origins include (a) less differentiated, lower mantle entrained into plumes [3,5] and (b) moderately old (0.3–2 Ga) recycled oceanic crust stored in the transition zone region [71] or dispersed in the entire mantle [72]. In addition, it has been proposed [72] that the isotopic convergence reflects a mean and widespread process rather than mixing with an independent reservoir. As part of the so-called "Dupal" islands, Réunion plots in Pb–Pb isotope space above the North Hemisphere Regression Line [73], which is equivalent to the MORB-FOZO array modeled by Stracke et al. [72]. Thus, the elevated $(^{207}Pb/^{204}Pb)/(^{206}Pb/^{204}Pb)$ signature of Réunion requires an older-than-average U/Pb increase, which is not compatible with a mean recycling process. It is nevertheless possible to explain the Réunion signature in the frame of the continuous differentiation model of Stracke et al. [72] by calling upon an additional enriched component. The unradiogenic Os signature of Réunion precludes involvement of upper crustal rocks and sediments. Instead, mixing the redefined FOZO component [72] with ancient subcontinental lithospheric mantle could be an alternative explanation for the origin of the Réunion plume signature.

Recycling of oceanic crust into hotspot sources was initially proposed [1] to account for the apparent contradiction between trace element abundances in OIB, which require elevated concentrations of incompatible elements in the source, and Sr–Nd isotopes, whose signature records a long-term depletion. However, alternative solutions to this long-standing problem exist: first, the discovery of very early mantle depletion [53] suggests that nearly the entire mantle must be depleted, but only slightly. Thus, mantle domains that have not been affected by subsequent differentiation events (oceanic crust extraction and recycling) may have kept incompatible element concentrations at a primitive level. Second, it has been shown [74] that mixing melt fractions originating from distinct mantle lithologies can produce trace element systematics of OIB without the need for a highly enriched source. Taken together, these recent results open up the possibility that the moderately enriched basalts (Fig. 6) of Réunion originate from an early, slightly depleted mantle domain that has been subsequently little, or not at all enriched by recycling processes.

6. Concluding remarks

Th/U measured in lavas from Piton de la Fournaise is similar to the value integrated by Pb isotopes since 4.56 Ga (weight ratio ~3.9). This suggests that U/Pb and Th/Pb fractionations have been tightly coupled during the history of the Réunion plume source, preserving a primitive-like Th/U signature. If Th/U fractionation in the mantle is the result of post-Archean subduction, then the Th/U signature of Réunion is best explained by sampling an early-differentiated mantle domain,

subsequently influenced little or not at all by recycled components. The other geochemical characteristics of the Réunion plume are shown to be consistent with such a scenario. In particular, (1) the geochemical signature of Réunion is systematically intermediate between mantle end-members with the exception of Osmium, whose isotopic signature is the least radiogenic of all plumes. (2) The difference in $^{4}He/^{3}He$ and $^{40}Ar/^{36}Ar$ between Réunion and the so-called primitive plumes (Iceland, Hawaii) may reflect a difference in the U, Th and K content of their source, rather than different degassing histories as previously proposed [9]. This increases the possibility that the intermediate rare gases ratios of Réunion reflect closed-system evolution of $(U+Th)/^{3}He$ better than extremely elevated ratios. The possibility that the Réunion plume source has been isolated from subduction influences since at least the Archean has the following implications: if subduction and convection are tightly coupled, which is likely, then the mantle domain sampled by the Réunion plume must be located off the main stream of convection. In addition, the lack of geochemical imprints of recycling at such a major hotspot indicates that plumes do not necessarily originate from a boundary layer where recycled components are stored.

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