Re-visiting barium isotope compositions of mid-ocean ridge basalts and the implications

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Graphical abstract

We estimate the average $\delta^{138/134}$Ba of the depleted MORB mantle (DMM) as $+0.05\%_{\pm 0.05\%} (2SD, n=16)$ based on D-MORBs.

Public summary

The $\delta^{138/134}$Ba of the global MORBs range from $-0.06\%$ to $+0.15\%$.

This study obtains the average $\delta^{138/134}$Ba of the DMM as $+0.05\%_{\pm 0.05\%} (2SD, n=16)$, which is much lower than the previous estimation.

Ba isotope compositions of the E-MORBs could be sourced by the incorporation of subducted AOC and/or sediments.

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Re-visiting barium isotope compositions of mid-ocean ridge basalts and the implications

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Abstract: Barium (Ba) isotopes can be used as potential tracers for crustal material recycling in the mantle. Determination of the Ba isotope composition of the depleted mantle is essential for such applications. However, Ba isotope data for mantle-derived basalts are still rare. In this study, we reported high-precision Ba isotope data of 30 oceanic basalts including 25 mid-ocean ridge basalts (MORBs) from geochemically and geologically diverse mid-ocean ridge segments and five back-arc basin basalts. The $\delta^{138/134}$Ba values of these samples varied from $-0.06\permil$ to $+0.11\permil$, with no systematic cross-region variation. Together with published data, we constrained the average $\delta^{138/134}$Ba of global MORBs to $+0.05\permil\pm0.09\permil$ (2 standard deviation, $n = 51$). Based on depleted MORBs that have (La/Sm)$_n < 0.8$, low $\delta^{87/86}$Sr ($< 0.70263$), and low Ba/Th < 71.3, we estimated the average $\delta^{138/134}$Ba of the depleted MORB mantle (DMM) as $+0.05\permil\pm0.05\permil$ (2SD, $n = 16$) that is significantly lower than the DMM ($= 0.14\permil$) reported previously. If a new estimation of the DMM is applied, it is unreasonable to infer that the Ba isotope signatures of the “enriched-type” MORBs (E-MORBs) could be attributed to pervasive sediment recycling in the upper mantle. We therefore, conclude that the Ba isotope compositions of the E-MORBs could be sourced from the incorporation of subducted altered oceanic crust and/or sediments depending on the Ba isotope composition and other geochemical information of the local mantle.

Keywords: barium isotopes; mid-ocean ridge basalt; upper depleted mantle; subducted crustal materials

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1 Introduction

Barium (Ba) is a highly incompatible element during mantle melting, thus, it is much more enriched in the upper continental crust\textsuperscript{1} and marine sediments\textsuperscript{2} than in the mantle\textsuperscript{3}. Ba is also fluid mobile during slab subduction\textsuperscript{4}. Ba has seven stable isotopes: $^{133}$Ba (0.11%), $^{135}$Ba (0.10%), $^{136}$Ba (6.59%), $^{137}$Ba (7.85%), $^{138}$Ba (11.23%), and $^{139}$Ba (7.10%)\textsuperscript{5}. Previous studies have found that Ba isotopic compositions ($\delta^{138/134}$Ba = [($^{138}Ba_{\text{sample}}$) / ($^{138}Ba_{\text{SRM3104a}}$) - 1] $\times$ 10000) of subducted crustal materials such as marine dimentes, altered oceanic crust (AOC), and continental rocks\textsuperscript{6-11} are distinct from the mantle (0.05\permil$\pm$0.06\permil)\textsuperscript{11}. For example, diamicrites have a substantial variation in $\delta^{138/134}$Ba from $-0.23\permil$ to $+0.47\permil$\textsuperscript{9}. Therefore, Ba isotopes have recently been suggested as sensitive tracers for crustal material recycling in mantle-derived igneous rocks\textsuperscript{7,8,11}.

Magma produced annually in mid-ocean ridge settings accounts for 75% of global magmatic output\textsuperscript{12}. Mid-ocean ridge basalts (MORBs) represent mafic melts derived from a simple melting history of the depleted upper mantle, providing a critical way to constrain the composition of the upper mantle\textsuperscript{13}. Substantial chemical heterogeneity has been observed in global MORBs\textsuperscript{14,15}. The ratios of highly incompatible trace elements (such as La/Sm) in MORB vary by more than an order of magnitude\textsuperscript{14}. A previous study has proven that Ba isotope variation in MORBs is not caused by mantle melting and/or fractional crystallization\textsuperscript{16}. Thus, the $\delta^{138/134}$Ba values of the MORBs should reflect their mantle source signatures. It is thus reasonable to use Ba isotope data to identify the contribution of recycled materials to the source of the MORB if the enriched composition in the mantle is related to the addition of materials from surface reservoirs.

Better knowledge of the Ba isotope composition of the upper mantle is essential for the application of Ba isotopes in tracing mantle heterogeneity. However, estimates of $\delta^{138/134}$Ba in the mantle are rare. Nielsen et al.\textsuperscript{7} reported $\delta^{138/134}$Ba values of MORB glasses from the Mid-Atlantic Ridge (MAR), East Pacific Rise (EPR), Juan de Fuca Ridge (JdF), Central Indian Ridge, and Southwest Indian Ridge ranging from $+0.02\permil$ to $+0.15\permil$. According to the composition of their most depleted MORBs, they defined the average $\delta^{138/134}$Ba of the depleted MORB mantle (DMM) as $= +0.14\permil$. Based on the measurement of mantle-derived carbonatites from Canada, East Africa, Germany and Greenland, Li et al.\textsuperscript{14} found that the $\delta^{138/134}$Ba of most carbonatites vary from $-0.04\permil$ to $+0.12\permil$, thus suggesting that the average $\delta^{138/134}$Ba of their mantle source is $+0.05\permil\pm0.06\permil$ (2SD). The average $\delta^{138/134}$Ba of the MORBs ($+0.07\permil\pm0.08\permil$, 2SD) reported by Nielsen et al.\textsuperscript{7} is generally indistinguishable from that of the carbonatites reported by Li et al.\textsuperscript{14}. However, the $\delta^{138/134}$Ba value of the DMM suggested by Nielsen et al.\textsuperscript{7} was much higher. This contradiction hinders the application of Ba iso-
samples were dried and diluted with 2% (m/m) HNO₃ for further purification. The yield of the purification process was > 99%. The total procedural blanks were < 0.5 ng. Purified samples were dried and diluted with 2% (m/m) HNO₃ for instrumental measurements.

Ba and Sr isotope measurements were conducted using a Neptune Plus multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS). Sr isotope ratios were measured in low-resolution mode under “wet” plasma conditions. The measured Sr isotope ratios were corrected for instrumental mass bias by normalizing them to “Sr/Sr = 0.1194. The results for the samples and reference materials are listed in Table 1. The ”Sr/Sr values of NBS-987 and BCR-2 are 0.710247±0.000011 (2SD, n = 25) and 0.705025±0.000009 (2SD, n = 4), respectively, which are consistent with the previous study[21]. Ba isotope ratios were measured using the double-spike (²⁹⁸Ba-²⁹⁶Ba) method in low-resolution mode under “dry” plasma conditions (Aridus II desolvating nebulizer). The background signal for (²⁹⁶Ba (< 5 mV) is negligible relative to the sample signal (~7 V). The results are reported in δ-notation relative to NIST SRM3104a: δ¹³⁸Ba (‰) = [(¹³⁸Ba-sample/¹³⁸Ba-SRM3104a) - 1] × 1000. For comparison, the δ¹³⁷Ba values of all the samples were calculated to be δ¹³⁸Ba following the mass-dependent fractionation (δ¹³⁸Ba = 1.33 × δ¹³⁷Ba). The external precision of δ¹³⁸Ba based on the measurement of two in-house standards (USTC-Ba and ICPUS-Ba) is ≲ 0.04‰ (2SD). We estimate the long-term external precision of δ¹³⁸Ba is ≲ 0.05‰ (2SD), as verified by Deng et al.[29]. The results for the samples and standard materials are listed in Table 1. The δ¹³⁸Ba values of BCR-2 and GSP-2 are in good agreement with previously published values[56-59].

4 Results
The Ba-Sr isotope data are presented in Table 1 and Fig. 1. The ”Sr/Sr values of the MORB and BABB samples range from 0.70243 to 0.70404. The δ¹³⁸Ba of the MORB samples range from −0.06 ‰ to +0.11 ‰. E-MORBs (−0.06 ‰ to +0.11 ‰, n = 7) show more scattered δ¹³⁸Ba than N-MORBs (−0.04 ‰ to +0.08 ‰, n = 18), although the difference is within the analytical uncertainty. There are no systematic cross-regional variations. Therefore, δ¹³⁷Ba of N-MORB varies from −0.03 ‰ to +0.08 ‰ in EPR MORBs (n = 13), from −0.02 ‰ to +0.11 ‰ in MAR MORBs (n = 6), and from −0.06 ‰ to +0.10 ‰ in GR MORBs (n = 6). One MORB sample in SEIR has a value of +0.04 ‰±0.03 ‰ (2SD). The δ¹³⁷Ba of the five BABB samples range from +0.01 ‰ to +0.08 ‰, which is within the range of MORBs. These values are slightly lower than those of previously reported MORB samples, despite substantial overlapping (+0.02 ‰ to +0.15 ‰)[7] (Fig. 2). To our knowledge, the five Lau Basin basalt samples are the first published Ba isotope data for BABBs. On average, δ¹³⁸Ba of our samples (+0.04 ‰±0.08 ‰, 2SD, n = 30) is indistinguishable from their average value (+0.07‰±0.08‰, 2SD, n = 21).

5 Discussion
5.1 Ba isotope systematics of MORBs
The basaltic oceanic ridge samples reported herein span a broad range of geographical distributions and chemical compositions. The inconsistent Ba isotope data between this study
Table 1. Elemental and isotope compositions for the samples and standard materials in this study.

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* $^{87}$Sr/$^{86}$Sr values of the samples with asterisks were measured in this study.
* La/Sm was normalized to CI-chondrite$^{[3]}$.
* 2SD = two times the standard deviation of the population of $n$ repeat measurements of a sample solution.
* $n$ represents the number of repeated measurements of the same solution using MC-ICP-MS.
* Data were calculated from the measured $^{137}$Ba/$^{134}$Ba values, as described in the text.
* Replicate = repeat sample dissolution, purified column chemistry, and MC-ICP-MS analysis.

PetDB= the Petrological Database website (http://www.earthchem.org/petdb).
and that in Nielsen et al. [7] could not reflect the different degrees of rock alteration because all these samples are fresh. Interlaboratory bias is also not considered as Ba isotope analyses of the igneous rock standards in our and their studies display consistent results within error [7, 24]. The most probable explanation is that this difference is due to practical Ba isotope variations among global MORBs.

Barium isotope variation in MORBs can potentially be induced by magmatic differentiation during the formation of these basalts or mantle heterogeneity. Fig. 3 shows that no observable correlations exist between $\delta^{138/134}$Ba and the indicators of magmatic differentiation (MgO and SiO$_2$) or degree of partial melting ($Na_8$) [27]. In addition, as has been verified previously [7], Ba is highly incompatible with the bulk distribution coefficient $D_{\text{solid/melt}}$ of 0.00012 during mantle melting [28], and ~99% of Ba tends to enter the melt after 1% partial melting. MORBs represent degrees of melting between 6% and 20% [29], as a result, Ba isotope fractionation between the starting mantle composition and these basaltic melts is negligible. Therefore, the Ba isotope compositions of the MORBs principally reflect variations in the $\delta^{138/134}$Ba values of their mantle sources.

Together, all the MORB data sets in this study and the literature [7] (51 samples, including BABB samples) provide a range of $\delta^{138/134}$Ba of $-0.06\%$ to $+0.15\%$, with an average of $+0.05\%\pm0.09\%$ (2SD, $n = 51$).

5.2 Estimation of the Ba isotope composition of DMM

According to the previous definition of N-MORB [19], 70% of
the MORB samples in PetDB are categorized as N-MORB, displaying a considerable range of major and trace element compositions\cite{19}. Only the most depleted samples were representative of the DMM. Therefore, we estimated the δ\textsuperscript{138/134}Ba of DMM based on the most depleted MORB (D-MORB) data. The selected 10 D-MORB samples showed typical depleted compositions with ([La/Sm]\textsubscript{N} < 0.8\textsuperscript{19}). We also used \textsuperscript{87}Sr/\textsuperscript{86}Sr (< 0.70263) and Ba/Th (< 71.3), which are lower than the average DMM\cite{22}, as the criteria for recognizing the D-MORBs (Figs. 1b and 1c). Along with the D-MORB samples measured in the literature\cite{4}, the average δ\textsuperscript{138/134}Ba of DMM was estimated to be +0.05‰±0.05‰ (2SD, n = 16). The JdF sample (+0.15‰) in Nielsen et al.\cite{7} is beyond the average δ\textsuperscript{138/134}Ba of all MORBs (+0.05‰±0.09‰, 2SD, n = 51) and was thus precluded when calculating the average DMM value. To avoid artificial bias in different laboratories, we also calculated a mean δ\textsuperscript{138/134}Ba solely based on our data of +0.05 %±0.04 % (2SD, n = 10). Our estimation of δ\textsuperscript{138/134}Ba of DMM is significantly lower than that reported by Nielsen et al.\cite{7} (+0.14‰).

It is necessary to evaluate different estimates of the average δ\textsuperscript{138/134}Ba values of the DMM. The estimation of the DMM by Nielsen et al.\cite{7} may be problematic for two reasons. First, the DMM and E-MORB mantle (EMM) were defined using only three MORB samples with the lowest and highest \textsuperscript{87}Sr/\textsuperscript{86}Sr respectively. The limited sample size leads to a lack of representativeness for the samples that estimate the DMM. Second, as pointed out by Wu et al.\cite{13}, using only \textsuperscript{87}Sr/\textsuperscript{86}Sr is not sufficient to choose appropriate samples to define the DMM. In fact, both the DMM and EMM reservoirs span a large range of radiogenic isotope ratios. In this case, the D-MORBs with \textsuperscript{87}Sr/\textsuperscript{86}Sr < 0.7024 exhibited a large δ\textsuperscript{138/134}Ba range (+0.03 ‰ to +0.14 ‰), overlapping with the δ\textsuperscript{138/134}Ba range (+0.02 ‰ to +0.11 ‰) of the MORBs with \textsuperscript{87}Sr/\textsuperscript{86}Sr > 0.7030. Therefore, one should rigorously choose MORB samples to estimate the average DMM. Based on all Ba isotope data for the D-MORBs, our study provides a more reliable estimate for the average δ\textsuperscript{138/134}Ba of the DMM (+0.05 ‰±0.05 ‰, 2SD, n = 16), which leads to new constraints on the nature of the E-MORB source.

5.3 The origin of the enriched source of E-MORB

There are long-term debates on how the mantle source of E-MORB was formed, i.e., melting of the enriched lower mantle\cite{8,9}, and recycling crustal materials into the mantle (e.g., crustal rocks, metamorphic fluids, and sediments)\cite{10,11,12}. Because Ba isotope compositions are unlikely to be modified during mantle melting and fractional crystallization from basaltic melt, Ba isotope variation in MORBs should reflect the introduction of recycled, Ba-rich crustal components into the mantle.

Based on the early estimates of average DMM data, Nielsen et al.\cite{7} proposed that Ba isotopic variation in MORBs is derived from pervasive mixing between isotopically heavy DMM and light EMM reservoirs. However, different insights into the formation of the EMM may be gained if the new DMM estimation is applied. The E-MORBs and BABBs show a slightly larger range of δ\textsuperscript{138/134}Ba (−0.06‰ to +0.11‰) relative to D-MORBs. The δ\textsuperscript{138/134}Ba does not show a simple trend with increasing ([La/Sm]\textsubscript{N} < 0.8\textsuperscript{19}), \textsuperscript{87}Sr/\textsuperscript{86}Sr, and Ba/Th (Fig. 1), which is inconsistent with previously observed hyperbolic mixing relationships\cite{15}. Nielsen et al.\cite{7} attributed the isotopically light Ba signature of EMM (≈ +0.03‰) to the pervasive addition of sedimentary materials to DMM (≈ +0.14‰), as they proposed that sediments have a narrow range of δ\textsuperscript{138/134}Ba (0.01‰±0.04 ‰, 2SD) and are similar to the E-MORB (+0.03‰±0.02‰, 2SD). However, in the new estimation is applied, the δ\textsuperscript{138/134}Ba values of such sediments will be similar to the DMM value (+0.01 ‰ vs. +0.05 ‰). Considering the observations of the MORBs and sediments together, it is unreasonable to infer that the Ba isotope signatures of E-MORBs are derived from pervasive recycled sediments in their mantle source. Additionally, the δ\textsuperscript{138/134}Ba observed in this study did not show a simple trend, but became more scattered with increasing \textsuperscript{87}Sr/\textsuperscript{86}Sr and Ba/Th (Figs. 1b and 1c).

Significant variations in δ\textsuperscript{138/134}Ba for crustal materials, such as marine sediments (−0.11‰ to +0.10‰)\cite{6}, AOC (−0.09‰ to +0.33‰)\cite{6}, and diamictites\cite{15} have been reported, which are much wider than the range used by Nielsen et al.\cite{7}. Although the AOC does not have much higher Ba content and \textsuperscript{87}Sr/\textsuperscript{86}Sr relative to the sediment, a study of the Tonga arc revealed that the addition of AOC-derived fluid to the mantle wedge is the most probable source of the high δ\textsuperscript{138/134}Ba in the arc lavas (up to +0.16‰)\cite{16}. In addition, Hao et al.\cite{11} identified the contribution of bulk AOC, aside from the sediment components, to the formation of the Fushui mafic rocks using Ba isotopes. Therefore, we suggest that subduction of sediments and AOC can account for the variation of δ\textsuperscript{138/134}Ba in the E-MORBs (−0.06‰ to +0.11‰). However, Ba isotopes can be considerably fractionated during subduction-zone fluid processes, resulting in a metamorphic fluid with much higher δ\textsuperscript{138/134}Ba than the subducted materials\cite{15}. Therefore, evaluating the contributions of different crustal components to the source of mantle-derived igneous rock requires careful consideration.

6 Conclusions

This study presents the Ba isotope compositions of a wide range of MORBs and BABBs from different localities, with variable δ\textsuperscript{138/134}Ba ranging from −0.06‰ to +0.11‰. Based on the D-MORB samples, we re-estimated the average δ\textsuperscript{138/134}Ba of the DMM as +0.05‰±0.05‰ (2SD, n = 16). Because of the similar δ\textsuperscript{138/134}Ba values of sediments and the DMM, it is unreasonable to conclude that Ba isotope signatures of E-MORBs are attributed to pervasive sediment recycling in the upper mantle. Instead, the Ba isotope data of the E-MORBs indicate that the EMM could reflect the incorporation of subducted AOC and/or sediments, depending on the Ba isotope composition and other geochemical information of the local mantle.

Supporting information

The supporting information for this article can be found online at https://doi.org/10.52396/JUSTC-2021-0276.

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Conflict of interest

The authors declare that they have no conflict of interest.

Biographies

Xiaoyun Nan is currently working as an associate research fellow at the University of Science and Technology of China (USTC). She received her PhD in Geology from the USTC in 2017. Her research focuses on metal–stable isotope geochemistry.

References


