Role of recycled oceanic basalt and sediment in generating the Hf–Nd mantle array

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Following its subduction, oceanic crust either contributes to the source of island-arc volcanic rocks or it is recycled into the mantle[1]. Most[2,3], but not all authors[4] believe that recycled crust is incorporated into the plume source of oceanic basalts. The hafnium (Hf) and neodymium (Nd) isotopic compositions of basalts from oceanic islands and mid-ocean ridges exhibit a linear relationship—the mantle array—which is thought to result from mixing between material from the depleted mantle and an enriched recycled component. Here, we model the Hf–Nd isotopic composition of oceanic basalts as a mixture of recycled oceanic crust and depleted mantle and find that recycling of basalt alone is not sufficient to reproduce the mantle array. We conclude that oceanic sediments, which have a relatively high ^{176}Hf/^{177}Hf ratio, must also be recycled. Combining oceanic sediments with recycled oceanic basalts and subsequent mixing with depleted mantle peridotite produces Hf and Nd isotopic compositions that coincide with the mantle array. The composition of bulk continental crust requires the existence of a complementary low ^{176}Hf/^{177}Hf reservoir, which we suggest is zircon-rich sediment.

Figure 1 shows the mantle array and the compositions of worldwide oceanic sediments in an $\varepsilon_{\mathrm{Hf}}$ versus $\varepsilon_{\mathrm{Nd}}$ plot. Ocean-island basalts (OIB) and mid-ocean ridge basalts (MORB) have an $\varepsilon_{\mathrm{Hf}}$ of $+1.28$ at $\varepsilon_{\mathrm{Nd}} = 0$, a value higher than the accepted bulk silicate Earth (BSE) value[5], but within error of the new values suggested by Bouvier *et al.*[6]. In contrast, sedimentary material exhibits a large range of Nd and Hf isotopic compositions, but almost all oceanic sediments have elevated $\varepsilon_{\mathrm{Hf}}$ at a given $\varepsilon_{\mathrm{Nd}}$, their position being related to the sedimentary process that produced them. Hydrogeneous sediments such as Fe–Mn crust and nodules have the highest $\varepsilon_{\mathrm{Hf}}$ at a given $\varepsilon_{\mathrm{Nd}}$, deep-sea clays and biogenic sediments have intermediate values and terrigeneous sediments have the lowest $\varepsilon_{\mathrm{Hf}}$ (ref. 7).

Using the average composition of MORB given in Table 1, we can calculate the evolution path on which such basalts would lie, had they been formed in a similar manner throughout Earth's history (see Supplementary Information, Fig. S1). This array, shown in Fig. 2a, is clearly different from the 'mantle array', suggesting that recycling of oceanic basaltic crust alone cannot account for the Nd and Hf isotopic compositions of MORB and OIB even if the recycled material is mixed with ambient depleted mantle. The 'mantle array' has a much shallower slope than the oceanic crust evolution path in Fig. 2a, requiring involvement of another component with low $\varepsilon_{\mathrm{Nd}}$ associated with high $\varepsilon_{\mathrm{Hf}}$. We suggest that this component is oceanic sediments.

The average Nd and Hf isotopic compositions of oceanic sediments are difficult to evaluate because of a paucity of $\varepsilon_{\mathrm{Nd}}$ and $\varepsilon_{\mathrm{Hf}}$ data and the large range in these data. Plank and Langmuir[8] calculated an average chemical composition for global subducted sediments (GLOSS; see Table 1) and provided estimates for most trace elements as well as for Sr, Pb and Nd isotopes. However, owing to the scarcity of Hf isotopic data, they could not provide an average Hf isotopic value. To estimate this value, an average of all measured values could be calculated. However, this could produce a questionable result because (1) the total number of analysed samples is not large (<200) and (2) two groups (the sediments present in front of the Lesser Antilles arc[9] and the Fe–Mn crusts and nodules[10,11]) dominate the database (see Fig. 1, inset). Mean values calculated from the entire sample set are $\varepsilon_{\mathrm{Nd}} = -7.5$ and $\varepsilon_{\mathrm{Hf}} = -1.3$. Alternatively, the average composition of the sedimentary pile sampled during Leg 185 in the western Pacific Ocean could be used. Details of the geochemistry of the sedimentary pile are found elsewhere[12]; here, we emphasize that the petrological and geochemical characteristics of these sediments are typical of deep-sea sediments and it is the first complete drill core on which both Hf and Nd isotopes were analysed. The $\varepsilon_{\mathrm{Nd}}$ of the sedimentary pile is -5.9 and its $\varepsilon_{\mathrm{Hf}} = +4.4$ (ref. 13; see Supplementary Information, Table S1). Finally, to associate an $\varepsilon_{\mathrm{Hf}}$ value with the Nd isotopic composition recommended for GLOSS, we used the trend defined by island-arc lavas in the Nd–Hf isotopic space (see Fig. 2b) arguing that sediments recycled into the mantle are similar to those that contaminate the mantle wedge source of island-arc volcanics. This technique provides an $\varepsilon_{\mathrm{Hf}}$ value of about 0 for the GLOSS $\varepsilon_{\mathrm{Nd}}$ value of -8.9, leading us to use an $\varepsilon_{\mathrm{Hf}}$ value of $+2$ (±3) in our modelling (see Table 1). These values, which are shown by a rectangle in Fig. 2b, lie significantly above the mantle array and overlap the fields of both ferromanganese crust and oceanic clay and mud.

Using the estimated compositions of oceanic sediments and oceanic basaltic crust (Table 1), we model the mantle array as a mixture of recycled oceanic crust and associated oceanic sediments, together with depleted mantle. We assume that the oceanic crust and sediments formed by similar processes and were recycled into the mantle for the past 3 Gyr. To calculate their initial isotope ratios

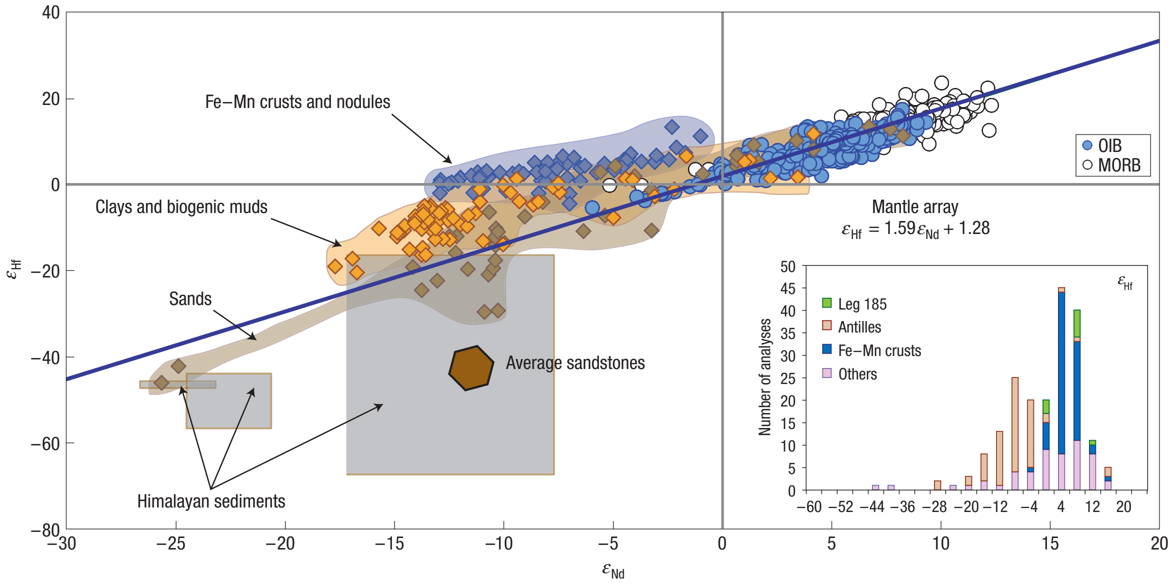

Figure 1 **ε_{Hf} and ε_{Nd} values of oceanic basalts and oceanic sediments.** OIB: blue circles, MORB: white circles, oceanic sediments: diamonds. Fields for Himalayan sediments were constructed using Nd whole-rock data and Hf data on zircons[20]. The large hexagon is the average sandstone value (Table 1). The OIB array follows the relationship $\varepsilon_{Hf} = 1.51\varepsilon_{Nd} + 1.39$. When OIB and MORB are combined, the relationship is essentially unchanged: $\varepsilon_{Hf} = 1.59\varepsilon_{Nd} + 1.28$. Data from the literature[9–11,13,18,20–27], GEOROC[28] and PETDB[29] databases, and unpublished data from C.C. and M.C. Inset: Histogram of ε_{Hf} values for sediments shown in the main panel. Sediments are grouped by colour according to their origin.

at different times in Earth's history, we assumed a linear relationship between the present-day isotopic ratios of average sediment and basalt and the Hf and Nd isotopic ratios of the Earth 4.55 Gyr ago. Both sediments and basalts are then modelled to evolve through time using the average Sm/Nd and Lu/Hf ratios of GLOSS and average MORB (see Table 1 and Supplementary Information, Fig. S1). The locii of the present-day isotopic compositions of sediments and oceanic crust that had formed at different times in the past are shown in Fig. 2a. The main observations are as follows. (1) The present-day average Hf isotopic composition of the sediments is indeed high ($\varepsilon_{Hf} = +2 \pm 3$) relative to its Nd isotopic composition ($\varepsilon_{Nd} = -8.9$), whereas the average basaltic crust has ε_{Hf} and ε_{Nd} identical to present-day MORB (+13.9 and +8.8). (2) The average $^{176}Lu/^{177}Hf$ and $^{147}Sm/^{144}Nd$ ratios of the sediments are low (0.0142 and 0.1296; see Table 1), which leads to a present-day position of old sediment that is displaced to the left of the mantle array (Fig. 2a). (3) The low $^{176}Lu/^{177}Hf$ ratio (0.0270) of basalt, associated with its high $^{147}Sm/^{144}Nd$ (0.1986), leads to low ε_{Hf} and high ε_{Nd} for oceanic crust formed during the Archaean era, which is plotted in the bottom right quadrant of Fig. 2a. Mixing arrays between sediment and basaltic crust with the same age, calculated using the concentrations and isotopic compositions in Table 1, are also shown in Fig. 2a.

Finally, we assume that the recycled oceanic crust and sediment remain together as they are stirred in the convection cells and are not totally mixed into the surrounding mantle.

Table 1 Nd, Sm, Lu and Hf concentrations and ε_{Hf} and ε_{Nd} values for oceanic sediments and basalts as well as for other reference materials or reservoirs.

Sample	Subducted oceanic sediments*	Bulk silicate Earth	Average MORB¶	Upper continental crust‖	Average sandstone**
Nd (p.p.m.)	27.00		10.43	26	
Sm (p.p.m.)	5.78		3.42	4.5	
$^{147}Sm/^{144}Nd$	0.1296	0.1967†	0.1986	0.1047	0.1047
$^{143}Nd/^{144}Nd$	0.51218	0.512638†	0.513088		
ε_{Nd}	−8.9	0	8.8		−12
Hf (p.p.m.)	4.06		2.480	5.8	10
Lu (p.p.m.)	0.413		0.480	0.32	0.1
$^{176}Lu/^{177}Hf$	0.0142	0.0332‡	0.0270	0.0077	0.0014
$^{176}Hf/^{177}Hf$	0.282829	0.282772‡	0.283164		
ε_{Hf}	+2 (±3)	0	13.9		−43
Nd/Hf	6.65	4.44§	4.21	4.48	

*Values published by Plank and Langmuir[8] for GLOSS except for the Hf isotopes from this study.

†Values from Jacobsen and Wasserburg[30].

‡Values from Blichert-Toft and Albarède[5].

§ Calculated using the primitive mantle Nd and Hf concentrations of Hofmann[16].

¶ Values from Su[17] except for Hf isotopes from this work (average of all published values for MORB).

‖Values from McLennan[31].

**Values calculated using the following parameters: composition given by Taylor and McLennan[19], ε_{Nd} is the average value suggested by Gallet *et al.*[32] for continental crust and ε_{Hf} is calculated using a 2 Gyr model age for the continental crust and the Hf and Lu contents listed in the table.

We carried out a Monte Carlo simulation to generate 50,000 mixtures of oceanic basalt and associated sediments created at different times. The proportion of sediment in the sediment–basalt mixture is constrained to be between 0 and 20%, in accord with the relative thicknesses of sediment and basaltic crust. These calculated mixtures, shown by density fields in Fig. 3a, define a band with a positive slope similar to that of the 'OIB array'. However, petrological constraints such as high Ni contents indicate that oceanic basalts are not created by melting of basalt and sediments alone: peridotite must also be present in the source. We therefore generated mixtures of subducted material and depleted mantle, as shown by density fields in Fig. 3b,c. For a proportion of recycled material between 20 and 30%, the field of calculated values (Fig. 3c) overlaps remarkably well with the compositions of OIB (Fig. 3d). In particular, the position of the OIB array above the BSE position is quite well reproduced. If the proportion of recycled material is maintained between 0 and 15%, a field of

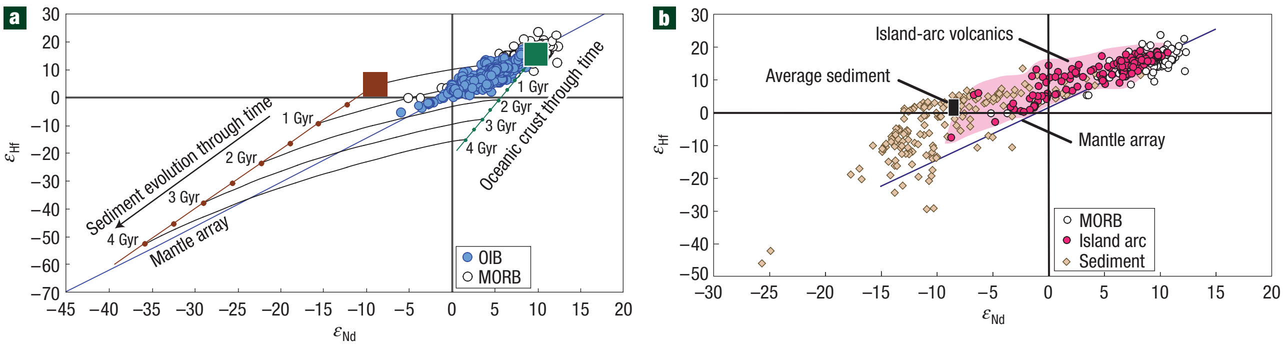

Figure 2 ε_{Hf} versus ε_{Nd} diagrams comparing modelled and measured data. a, MORB and OIB data. The present-day average compositions of oceanic basalt and sediments are shown with green and brown squares. The compositions of similar material formed at various times during Earth's history are shown as evolution paths (green and brown lines); mixing arrays between sediments and basalt are shown as curves. Data sources as in Fig. 1. **b**, Island-arc volcanics (from the GEOROC[28] database—pink circles) versus MORB and oceanic sediments. The black rectangle shows our preferred average recycled sediment value: $\varepsilon_{Nd} = -8.9$ from GLOSS, $\varepsilon_{Hf} = +2 \pm 3$ chosen to be consistent with (1) the extension of the island-arc field, (2) the Leg 185 average sediment composition and (3) the average composition of all analysed sediments.

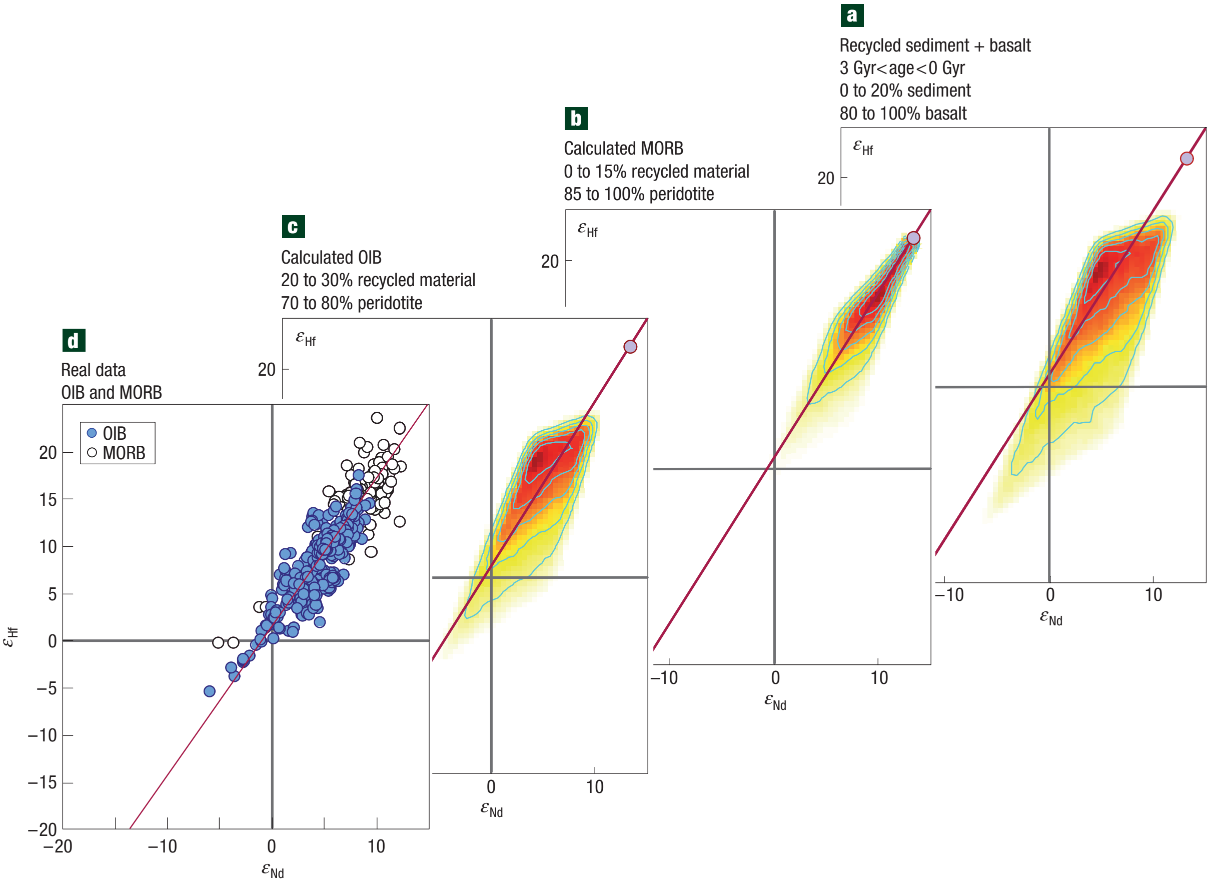

Figure 3 Comparison between our Monte Carlo simulations and measured values for OIB and MORB. a, A density plot of 50,000 runs simulating the composition of the recycled material (old oceanic crust and old sediment). Details of the calculation are given in the Supplementary Information. **b**,**c**, Density plots for simulated MORB and OIB using the procedure described in the Supplementary Information. The grey circles in **a**,**b** and **c** represent the composition of the depleted mantle. **d**, The same OIB and MORB data set as in Fig. 1.

calculated mixtures corresponding to MORB can also be generated (compare Fig. 3b and d). The amount of recycled sediment in the mixtures is always very low: 0–6% for the oceanic-island basalts and 0–2.2% for MORB. We recognize that 6% sediment in the source of OIB might be inconsistent with the Sr and Pb isotopic data and trace-element ratios such as Ce/Pb or Nb/U. However, our modelling ignores processes active in the subduction zones where elements such as Rb, Sr, U and Pb are preferentially removed from the subducted material[3], in contrast with Hf and the rare-earth elements which preferentially remain in the slab. The effect of a high sediment contribution to the OIB source is therefore minimized for all elements removed in subduction zones from the subducting material. Similar modelling using other isotopic systems (for example, Pb and Sr) would help constrain the impact of a high sediment contribution, but such modelling is beyond the scope of this manuscript.

The mantle array is generated by a model incorporating mixtures of recycled ocean crust and oceanic sediments: recycling of sediments alone does not work. Patchett *et al.*[14] and Salters and White[15] already noted that a mixture of deep-sea sediments and mantle peridotite cannot generate the OIB array because the elevated Nd/Hf ratio of the sediment produces an inappropriate mixing array in $\varepsilon_{\mathrm{Hf}}$ versus $\varepsilon_{\mathrm{Nd}}$ space. Incorporation of basaltic crust alone, which has trace element and isotopic characteristics similar to MORB[16,17] produces an array passing well below the BSE[15]. The low $\varepsilon_{\mathrm{Hf}}$ and high $\varepsilon_{\mathrm{Nd}}$ of old oceanic crust in Fig. 2a results from its low Lu/Hf and high Sm/Nd ratios, which generate such isotopic compositions as time passes. The Nd–Hf isotopic compositions of sediments in Fig. 1 reflect the dominant influence of authigenic processes and a minimal contribution from coarse-grained terrigeneous material. This might be explained by deposition of most sediments far from continents.

The average composition of oceanic sediments has an elevated $\varepsilon_{\mathrm{Hf}}$ compared with its $\varepsilon_{\mathrm{Nd}}$, a feature not seen in granitoid rocks from the continental crust[18]. It follows that processes acting during the formation of oceanic sediments have influenced their Hf isotopic composition[7]. This leads to the possibility that a reservoir with low $\varepsilon_{\mathrm{Hf}}$ at a given $\varepsilon_{\mathrm{Nd}}$ value is produced by sedimentary processes and isolated at the Earth's surface.

Given that the Nd/Hf ratios of deep-sea sediments and upper continental crust differ significantly (Table 1), a complementary reservoir with low Nd/Hf must have been generated by sedimentary processes. Patchett *et al.*[14] noted that zircon-rich sands, which are sequestrated on the edges of continents, have high Hf contents and low Lu/Hf ratios. Here, we propose that placers and beach sands rich in heavy minerals, parts of which are mined for Zr and related elements, could represent the sedimentary endmember with low Nd/Hf. We are currently analysing Nd and Hf isotopes in selected samples that might provide the key to unravelling terrestrial Nd–Hf isotopic systematics. Sandstones are common on continental platforms and represent 1% of the continental crust[19]. Table 1 shows a preliminary evaluation of their elemental and average present-day isotopic composition. The calculated $\varepsilon_{\mathrm{Nd}}$ and $\varepsilon_{\mathrm{Hf}}$ values lie significantly below the mantle array (Fig. 1) and clearly complement the high values found in the oceanic sediments. Sedimentary processes fractionate Lu/Hf and Sm/Nd ratios and Hf and Nd isotopic ratios more efficiently than magmatic processes, leading to large isotopic diversity with time. Vervoort *et al.*[18] already noticed that associated sandstones and shales share similar $\varepsilon_{\mathrm{Nd}}$ but $\varepsilon_{\mathrm{Hf}}$ is systematically lower in the sandstones. Given that the 'crustal array' of Vervoort *et al.*[18] is mainly based on fine-grained sediments with few coarse-grained sediments and few granitoids, it might not be representative of the continental crust as a whole. The continental crust might contain a higher proportion of low-$\varepsilon_{\mathrm{Hf}}$ coarse sediments than previously thought and the 'crustal array' could lie on or just below the 'mantle array' in $\varepsilon_{\mathrm{Hf}} - \varepsilon_{\mathrm{Nd}}$ space. If the currently accepted BSE value[5] is correct, zircon-rich detrital material could represent the low-$\varepsilon_{\mathrm{Hf}}$ 'hidden' reservoir; if the estimate of Bouvier *et al.*[6] is correct, this material merely represents the low-$\varepsilon_{\mathrm{Hf}}$ component of the continental crust.

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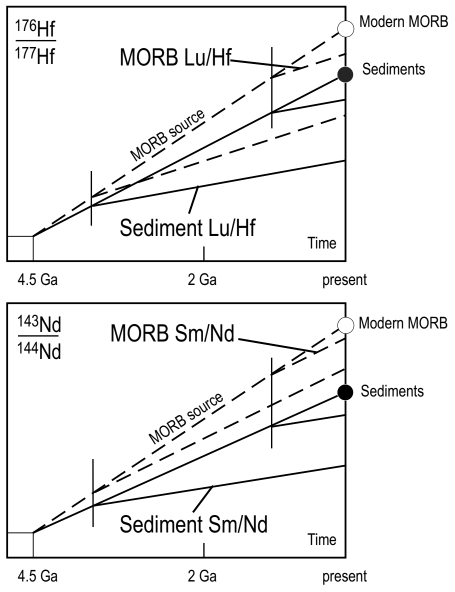
Author contributions

C.C. conceived the model and wrote the paper. E.L. made the numerical simulation. M.C. and J.-C.M. contributed to the data compilation and N.T.A. suggested several important ideas. All authors discussed the results and commented on the manuscript.

Supplementary Figure : Schematic ε_{Hf} and ε_{Nd} vs time diagrams describing the way the present day isotopic compositions of both recycled oceanic crust and sediments were calculated. The straight lines between the initial ratio of the Earth 4.5 Ga ago and the preferred present day average compositions of oceanic crust and sediments represent the locii of initial ratios for both reservoirs through time. The two vertical lines mark the times of separation of MORB and sediment from their source regions. Following this separation, the oceanic crust and the sediments follow shallower and different paths that are controlled by their own trace element ratios. Our modelling incorporates 50,000 examples of crust and sediment that separated at random times in the past.

Supplementary figure



Supplementary method presenting details about the modelling shown in figure 3.

The recycled material is a mixture of oceanic crust and sediments both with varying ages. The age distribution of oceanic crust is gaussian with a mean value of 0 Ga (present-day = maximum abundance) and a standard deviation of 1.5 Ga, truncated to remain in the interval [0 Ga, 3 Ga]. The distribution of sediment ages is conditioned by the age of the underlying oceanic crust: it is younger than the basaltic crust and follows a gaussian law, with a mean value of 0 Ga (contemporaneous with the oceanic crust) and a standard deviation of 0.5 Ga, truncated to remain in the interval [0 Ga, 3 Ga]. The Hf and Nd isotopic ratios of recycled oceanic crust and sediment with ages between 0 and 3 Ga were calculated and a MonteCarlo procedure was used to sample mixtures of basaltic crust and sediments within a range of sediment mass proportions between 0 and 20 %. Concentrations for sediment and basaltic crust are given in Table 1; isotopic ratios for the basaltic crust vary between $\varepsilon_{Nd} = +9.4$, $\varepsilon_{Hf} = +13.9$ for a 0 Ga crust and $\varepsilon_{Nd} = +3.7$, $\varepsilon_{Hf} = -7.3$ for a 3 Ga old crust; values for the sediments range between $\varepsilon_{Nd} = -8.9$, $\varepsilon_{Hf} = +2$ for 0 Ga old sediments, and $\varepsilon_{Nd} = -28.9$, $\varepsilon_{Hf} = -38.0$ for 3 Ga old sediments.

In Figures 3b and 3c, a second mixture is shown, between the recycled material of Figure 3a and an isotopically uniform depleted reservoir shown as the purple dot ($\varepsilon_{Nd} = 13.0$, $\varepsilon_{Hf} = 22.0$). This depleted mantle reservoir contains 1.04 ppm Nd and 0.25 ppm Hf (1/10 of that of the oceanic crust). The mass proportion of recycled material lies uniformly between 20 % and 30 % for the OIB simulation (3c), and between 0 % and 15 % for the MORB simulation (3b). In all cases, 50 000 points were calculated for each field.

If the present day ε_{Hf} value for sediments is changed to +5 or to -1 (to take the incertainty into account), the final result of the modelling is almost unchanged since the average value of the 50 000 simulated points for OIB is modified by only 0.5 epsilon unit.

This Monte-Carlo modelling constitutes a "direct problem": a series of parameters is discussed, preferred values such as the isotopic ratio and concentrations of the end-members and their mixing proportions are chosen, and finally the isotopic ratios of . the basalts are calculated and compared with measured values. A possible next step would be to invert the model by optimizing the input parameters to improve the agreement between the generated distributions and the observed ones. For this to succeed one needs to adjust the input parameters to approach a "best" *a posteriori* value, that is directly related to the sensitivity of the modelling. (for details see C.J. Allègre and É. Lewin. Chemical structure and history of the Earth : evidence from global non-linear inversion of isotopic data in a three-box model, Earth Planet. Sci. Lett. 96, 61-88, 1989). The sensitivity has to be evaluated and compared between the different input parameters, with respective variations scaled with a realistic uncertainty on each parameter.

In our case, changing a key parameter, such as the Hf isotopic ratio of the present-day sediment component, by +/- 3 epsilon units changes the final distribution by less than 0.5 epsilon units.

Conversely, the assumption that mixing proportions are uniform within a given range is governed by modelling simplicity (Ockham’s razor), because at present, it can neither be tested from the observational point of view, nor constrained by theory. The final result of our modelling may be sensitive to the mixing proportion law that we chose.

Supplementary Table : Hf isotopic composition of representative samples from Leg 185 drill core.

Supplementary Table

Hf isotopic compositions of representative sediments from Leg 185, Site 1149

Sample	Depth (mbsf)[a]	Unit[b]	^{176}Hf / ^{177}Hf ± $2\sigma_m$[c]	ε_{Hf}[d]
1149A 1H1 140-150	1.40	I	0.282964 ± 5	+6.8
1149A 10H3 140-150	84.60	I	0.282973 ± 4	+7.1
1149A 14H2 140-150	121.10	IIA	0.282960 ± 6	+6.6
1149A 20X1 140-150	171.20	IIB-III	0.282936 ± 5	+5.8
1149B 6R1 38-42	199.08	III	0.282873 ± 8	+3.6
1149B 17R1 14-17	292.14	IV	0.282678 ± 3	-3.3
1149B 25R1 19-23	368.89	IV	0.282715 ± 9	-2.0

[a] mbsf, meters below surface seafloor.

[b] Units as defined by Plank, T., Kelley, K. A., Murray, R. W. & Quintin Stern, L. Chemical composition of sediments subducting at the Izu-Bonin trench. Geochemistry, Geophysics, Geosystems 8, Q04I16 (2007).

[c] Normalized for mass fractionation to ^{179}Hf/^{177}Hf=0.7325.

[d] ε_{Hf} calculated using ^{176}Hf/^{177}Hf$_{CHUR}$=0.282772 after Blichert-Toft, J. & Albarède, F. The Lu-Hf isotope geochemistry of chondrites and the evolution of the mantle-crust system. Earth and Planetary Science Letters 148, 243-258 (1997).