Electronic Appendix B: Evolution of isotopic compositions as a function of time for metasomatized hydrous lithosphere

In Electronic Appendix B, we describe the methods we used to calculate the isotopic evolution of metasomatized hydrous lithosphere assuming that the liquids which generate this metasomatic enrichment are produced by partial melting of a MORB source (Fig. 1; main article). As indicated in the article, two possible scenarios have been proposed for the production of alkaline magmas by melting of metasomatized lithosphere: (1) Shortly after or coincident with metasomatism, the lithosphere experiences a thermal perturbation or decompression and thereby melts in situ; or (2) the metasomatized lithosphere is recycled into the convecting mantle by subduction or delamination and melts during later upwelling (e.g., in a plume or at a ridge). Therefore, the time interval between the metasomatism of this lithosphere and its subsequent melting (which produces the alkaline lavas) could range from 0 to 1–2 Ga (see the main article for further explanation).

Here we provide two examples of calculations for isolation times of 0.15 Ga and 1.5 Ga. The ranges in Sr, Nd, and Pb isotopic compositions for metasomatized lithosphere were calculated in two steps: (1) We calculated the isotopic composition of depleted MORB mantle (DMM to E-DMM) 0.15 Ga or 1.5 Ga ago assuming that the isotopic compositional range of these two MORB mantle end members was similar to the range of isotopic compositions observed in MORBs today, and that they were formed by an instantaneous depletion event at 2.5 Ga. (2) The isotopic evolution of the metasomatized lithosphere from 0.15 Ga, or from 1.5 Ga, to the present was calculated using the Rb/Sr, Sm/Nd, and U/Pb ratios calculated for each of the 25,000 Monte Carlo simulations.

1) Present day isotopic composition of depleted mantle

We estimated the present day isotopic composition for DMM and E-DMM in order to reproduce the compositional range observed in MORBs (data from the PetDB database). Since mixing between two fixed compositions will not reproduce the complete compositional range observed in MORBs, we introduced some isotope variability to our estimated DMM and E-DMM compositions; neodymium, strontium, and lead isotopic compositions for DMM and E-DMM are shown in Fig. B-1.

![Fig. B-1. a) $^{87}$Sr/$^{86}$Sr versus $^{143}$Nd/$^{144}$Nd and b) $^{206}$Pb/$^{204}$Pb versus $^{207}$Pb/$^{204}$Pb isotopic diagrams for the estimated compositions of DMM and E-DMM sources compared with MORB data (from the PetDB database). The light red points (forming a solid light red field) correspond to the present day calculated isotopic compositions of the model metasomatized hydrous lithosphere.](image)

The Rb/Sr, Sm/Nd, and U/Pb ratios of DMM and E-DMM were estimated assuming that these two reservoirs were produce simultaneously at 2.5 Ga from a primitive source. Figure B-2 shows the isotopic compositions of these two reservoirs as a function of time. For comparison, we have also plotted the Sr, Nd, and Pb isotopic evolution of the MORB source estimated by several other authors (see caption to Fig. B-2).
Electronic appendix B

Fig. B-2.  

2) Isotopic composition of metasomatic veins isolated for 0.15 Ga

To illustrate how we calculate the isotopic evolution of a specific composition of metasomatized hydrous lithosphere isolated for 0.15 Ga, we use the example described in Electronic Appendix A.

a) Composition of the source

In the example described in Electronic Appendix A, the source is composed of 51.9% of the E-DMM component (the remainder is DMM). We used these proportions and estimates of the isotopic compositions of DMM and E-DMM to calculate the isotopic composition and the Rb/Sr, Sm/Nd, and U/Pb ratios of the source.

Parameters of the mantle source (assuming that this source is composed of 51.9% E-DMM and 48.1% DMM):

<table>
<thead>
<tr>
<th></th>
<th>$^{87}$Sr/$^{86}$Sr</th>
<th>Rb/Sr</th>
<th>Sr (ppm)</th>
<th>$^{143}$Nd/$^{144}$Nd</th>
<th>Sm/Nd</th>
<th>Nd (ppm)</th>
<th>$^{206}$Pb/$^{204}$Pb</th>
<th>$^{207}$Pb/$^{204}$Pb</th>
<th>$^{208}$Pb/$^{204}$Pb</th>
<th>U/Pb</th>
<th>Pb (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>DMM</td>
<td>0.70245</td>
<td>0.0131</td>
<td>7.664</td>
<td>0.51316</td>
<td>0.3613</td>
<td>0.581</td>
<td>18.05</td>
<td>15.52</td>
<td>0.1290</td>
<td>0.018</td>
<td></td>
</tr>
<tr>
<td>E-DMM</td>
<td>0.70342</td>
<td>0.0194</td>
<td>9.716</td>
<td>0.51287</td>
<td>0.3354</td>
<td>0.703</td>
<td>18.87</td>
<td>15.53</td>
<td>0.1440</td>
<td>0.024</td>
<td></td>
</tr>
<tr>
<td>Source</td>
<td>0.70301</td>
<td>0.0167</td>
<td>8.731</td>
<td>0.51299</td>
<td>0.3466</td>
<td>0.644</td>
<td>18.53</td>
<td>15.53</td>
<td>0.1379</td>
<td>0.021</td>
<td></td>
</tr>
</tbody>
</table>

b) Isotopic composition of the mantle source 0.15 Ga ago

Using the trace-element ratios and estimates of the isotopic composition of the source, we can calculate its isotopic composition at 0.15 Ga.
Mantle source isotopic composition at 0.15 Ga:

<table>
<thead>
<tr>
<th>Source at time zero</th>
<th>87Sr/86Sr</th>
<th>Rb/Sr</th>
<th>143Nd/144Nd</th>
<th>Sm/Nd</th>
<th>206Pb/204Pb</th>
<th>207Pb/204Pb</th>
<th>208Pb/204Pb</th>
<th>U/Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Source 0.15 Ga ago</td>
<td>0.70301</td>
<td>0.0167</td>
<td>0.51299</td>
<td>0.3466</td>
<td>18.53</td>
<td>15.53</td>
<td>0.1379</td>
<td></td>
</tr>
</tbody>
</table>

These ratios correspond to the isotopic composition of the metasomatic vein formed 0.15 Ga ago.

c) Isotopic evolution of metasomatized hydrous lithosphere from 0.15 Ga to the present

The calculated Rb/Sr, Sm/Nd, and U/Pb ratios for the metasomatized hydrous lithosphere (hydrous cumulates + trapped liquid) shown in Fig. 5d (in the main article) were used to evaluate the isotopic evolution of the metasomatized lithosphere. By using only these ratios, we have neglected the role of the much larger volume of unmetasomatized lithosphere on the isotopic evolution of a parcel of heterogeneous lithosphere that consists of a small volume of a metasomatized hydrous component (hydrous cumulates + trapped liquid) and a much larger volume of unmetasomatized material. Two arguments support this assumption: (1) the metasomatized hydrous lithosphere is characterized by high trace-element contents relative to DMM and E-DMM, and thus the Rb/Sr, Sm/Nd, and U/Pb ratios of a heterogeneous lithosphere will essentially be those of the metasomatized hydrous lithosphere; (2) the relatively low temperatures in the lithosphere and the relatively short time scale of this calculation (0.15 Ga) limits the diffusive interaction between metasomatized and unmetasomatized lithosphere.

Isotopic compositions of metasomatized hydrous lithosphere isolated for 0.15 Ga:

<table>
<thead>
<tr>
<th>87Sr/86Sr</th>
<th>Rb/Sr</th>
<th>143Nd/144Nd</th>
<th>Sm/Nd</th>
<th>206Pb/204Pb</th>
<th>207Pb/204Pb</th>
<th>208Pb/204Pb</th>
<th>U/Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial composition of the metasomatized hydrous lithosphere 0.70291 0.0301* 0.51278 0.2311* 18.30 15.51 0.3160* 18.04 15.54</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Composition of the metasomatized hydrous lithosphere after 0.15 Ga of isolation 0.70309 0.51292 18.84 15.54</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* The Rb/Sr, Sm/Nd, and U/Pb ratios are calculated using the composition of metasomatized hydrous lithosphere from our Monte Carlo simulation (reported in Appendix A of the supplementary information and in Fig. 5d of the main article).

These four isotopic ratios represent one of the light gray points shown in Fig. 9a, panels I-IV (in the main article); light gray because the example in Appendix A has 18% trapped liquid.

A similar calculation was used to evaluate the isotopic evolution of metasomatized hydrous lithosphere over a period of 0.5 Ga; results are shown in Fig. 9b, panels I-IV.

3) Isotopic composition of metasomatic lithosphere isolated for 1.5 Ga

While melting of metasomatized lithosphere “in situ”, i.e., prior to being recycled into the convecting mantle, is capable of matching the isotopic compositions of many alkaline lavas observed in continental or oceanic settings, some alkaline magmas exhibit more extreme isotopic compositions and it has been suggested that such compositions can be produced by recycling the metasomatized lithosphere through the convecting mantle and thus greatly increasing the time for in-growth of extreme isotopic ratios (Halliday et al., 1995; Niu & O’Hara, 2003; Pilet et al., 2005). Here we calculate the isotope evolution of recycled metasomatized hydrous lithosphere that is isolated for 1.5 Ga. In contrast to the calculation above (involving a 0.15 Ga isolation time), here we cannot neglect the composition of the unmetasomatized lithosphere, since for isolation times of 1 to 2 Ga, some sizable fraction of the metasomatic veins can be expected to re-equilibrate with the surrounding peridotite (Kogiso et al., 2004). Thus we use the weighted average composition of a parcel of heterogeneous lithosphere that consists of metasomatized hydrous lithosphere and depleted peridotite (assumed to have a composition equal to DMM). This heterogeneous lithosphere we define as metasomatized peridotite. Based on field observations of metasomatic veins in the French Pyrenees (in Lherz in particular), we assume that the metasomatized hydrous lithosphere comprises 1% of the total parcel of heterogeneous lithosphere.

a) Isotopic composition of the mantle source 1.5 Ga ago
The mantle source composition is calculated using the same approach that we used in the example above (for an isolation time of 0.15 Ga). The composition of this depleted mantle source at present and 1.5 Ga ago is:

<table>
<thead>
<tr>
<th>Source at time zero</th>
<th>(^{87}\text{Sr}/^{90}\text{Sr})</th>
<th>Rb/Sr</th>
<th>(^{143}\text{Nd}/^{144}\text{Nd})</th>
<th>Sm/Nd</th>
<th>(^{206}\text{Pb}/^{204}\text{Pb})</th>
<th>(^{207}\text{Pb}/^{204}\text{Pb})</th>
<th>U/Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.70301</td>
<td>0.0167</td>
<td>0.51299</td>
<td>0.3466</td>
<td>18.53</td>
<td>15.53</td>
<td>0.1379</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Source 1.5 Ga ago</th>
<th>(^{87}\text{Sr}/^{90}\text{Sr})</th>
<th>Rb/Sr</th>
<th>(^{143}\text{Nd}/^{144}\text{Nd})</th>
<th>Sm/Nd</th>
<th>(^{206}\text{Pb}/^{204}\text{Pb})</th>
<th>(^{207}\text{Pb}/^{204}\text{Pb})</th>
<th>U/Pb</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.70202</td>
<td>0.51085</td>
<td>15.90</td>
<td>15.28</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

This composition corresponds to the isotopic composition of metasomatic veins formed 1.5 Ga ago.

**b) Rb/Sr, Sm/Nd, U/Pb trace-element ratios of metasomatized peridotite**

We estimate the trace-element ratios of metasomatized peridotite consisting of 1% metasomatized hydrous lithosphere embedded in 99% depleted peridotite similar to DMM. Based on the estimated DMM composition and the composition of metasomatized hydrous lithosphere (equal to metasomatic veins plus metasomatized peridotite), we calculated the Rb/Sr, Sm/Nd, and U/Pb trace-element ratios of re-equilibrate metasomatized peridotite.

Using the trace-element ratios that control the evolution of each of the isotopic systems, we recalculate the isotopic composition of metasomatized peridotite at 1.5 Ga.

**c) Isotopic evolution of metasomatized peridotite from 1.5 to the present**

After 1.5 Ga, the isotopic ratios of the metasomatized peridotite were calculated using the initial isotopic ratios and the corresponding trace-element ratios.

For long periods of isolation, the fraction of metasomatized hydrous lithosphere in the bulk system (depleted mantle + metasomatized hydrous lithosphere) controls the isotopic composition of the metasomatized peridotite (i.e., the bulk system). The following table lists the isotopic compositions of metasomatized peridotite after 1.5 Ga that contains between 1 and 100% metasomatized hydrous lithosphere.
The calculation shows, that as the fraction of metasomatized hydrous lithosphere increases, the isotopic compositions become more extreme given a 1.5 Ga isolation time.

References


