Methods

Sample selection and processing

All samples and previously published data used in this study must originate from a volcano that has been active within the Holocene [18] (Fig. 1), with the age of the sample estimated to be of Quaternary age (i.e., < 2.5 Ma). Essential criteria were that the samples are of known (precise) coordinates.

Obtaining previously published data

Previously published geochemical data was obtained from GeoROC [21]. Once downloaded the data-files were filtered to only include data within Ethiopia (including the Main Ethiopian Rift and Afar). These data were further filtered using the following criteria:

1. The values for the sample must relate to whole rock geochemistry, as opposed to mineral separates.
2. The individual sample must have major element, trace element, $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$, $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ isotope values available.
3. The coordinates must be specific to the individual sample's location rather than providing an average coordinate for a broader study area.

Analytical Geochemistry

Sampling and sample preparation

Ninety-three lavas, eleven welded tuffs and one pumice sample, from various volcanoes in Afar (Erta Ale Volcanic Segment, Ayelu, Abida, Yangudi, Dama Ali, Kerub, Ela, Didoli, Abbahu, Afdera, Tat Ali and Manda Hararo) were selected for geochemical analysis. The samples were collected during the CNR/CNRS projects in Afar during the 1960s [42] and stored in the Afar Repository at the University of Pisa, Italy,

24   (http://repositories.dst.unipi.it/index.php/home-afar). A further 52 samples from the Boset-

25   Bericha Volcanic Complex (BBVC) were collected during three field seasons, in November

26   2012, April-May 2015, and February 2017 [43].

27   Sample preparation for major, trace and isotope analyses was carried out at the University of

28   Southampton. Samples were cut with a saw to remove any sections, and any cut surfaces

29   ground down to reduce any potential contamination by metals from the saw blade. Rock

30   samples were then crushed using a fly press and placed in double-layered plastic bags prior to

31   crushing to minimise metal contamination.

32   The crushed material was separated into three size fractions (>1 mm, 0.5 mm to 1 mm, <0.5

33   mm) using Teflon sieves keeping the middle fraction (0.5 - 1 mm). The selected fraction was

34   cleaned by ultrasonicating in Milli-Q water then dried overnight in an oven at 85ºC. The

35   cleaned rock chips were then hand-picked under a microscope, to remove any non-rock

36   material. An aliquot of cleaned chips were used for Pb isotope analysis. For major element,

37   trace element, and $^{143}Nd/^{144}Nd$ and $^{87}Sr/^{86}Sr$ isotope analysis, the remaining rock chips were

38   ground to a fine powder using an agate mortar and pestle, to minimise contamination with

39   metals.

40   **Trace element analysis**

41   Samples were prepared for whole-rock trace element analysis using 0.05 g (for BBVC

42   samples) or 0.075 g (for all other samples) powdered sample. The powdered samples were

43   digested in sealed Savillex Teflon vials with 15 drops concentrated HNO_3 and 2 ml HF on a

44   hotplate at 130 °C for 24 hours (for all other samples), or with 50 drops HF and 2 ml HNO_3

45   on a hotplate at 130 °C for 24 hours (for BBVC samples). The HNO_3/HF was evaporated off,

46   and the samples were refluxed in 6M HCl for another 24 hours on a hotplate at 130 °C. The

47   6M HCl was evaporated off, and the samples were redissolved in 6M HCl. Mother solutions

48   were prepared by adding 6M HCl and Milli-Q water (total 30 ml) to the dissolved samples.

49   Daughter solutions were prepared using 0.5 ml of mother solution, diluted to 5 ml with 3%

50   HNO_3 (containing the internal standards In/Re/Be), resulting in an overall dilution factor of c.

51   4000.

52   Trace element analyses of the daughter samples were undertaken on the ThermoScientific

53   XSeries2 quadrupole inductively coupled plasma mass spectrometer (ICP-MS) at the

54   University of Southampton. Analytical standards and accuracy were monitored using

55   international standards JA-2, BCR-2, JB-2 (see Extended Data Table 3). Results in counts per

56   second were processed with the exact calculated dissolution factors and blank corrections to

57   produce concentrations in ppm for the trace elements analysed.

58   **Pb isotopic analysis**

59   For Pb isotope analysis, 0.3 g of cleaned, picked rock chips (0.5 mm to 1 mm) were weighed

60   into dedicated Pb Savillex Teflon vials and leached on a hotplate with 4 ml 6M HCl for an

61   hour (15 minutes for obsidian and pumice samples). Samples were rinsed several times in

62   Milli-Q water, then 0.5 ml concentrated HNO_3, before adding 3-4 ml of concentrated HF.

63   Samples were refluxed on a hotplate at 140 °C for 24 hours, and then evaporated to dryness.

64   0.5 ml concentrated HCl was added, and the sample evaporated to dryness, then 0.5 ml

65   concentrated HNO_3 was added and again evaporated to dryness. The final residue was

66   reconstituted in 0.5 ml HBr and refluxed for an hour. The samples were cooled and

67   centrifuged for 5 minutes. Pb was isolated using a single-stage HCl anion-exchange

68   chromatographic resin separation method [44], with AGX-1x8, 200 – 400 mesh resin.

69   Following this, the Pb isolate was dried down, redissolved in HNO_3, and analysed using the

70   double spike method of [45]. The samples were subsequently analysed on a ThermoScientific

71   Neptune MC-ICPMS at the University of Southampton (UK) with a NBS SRM 981

72   reproducibility of $^{206}Pb/^{204}Pb$ = 16.9404 ± 24 (142 ppm), $^{207}Pb/^{204}Pb$ = 15.4969 ± 26 (168

73   ppm), $^{208}Pb/^{204}Pb$ = 36.7149 ± 66 (180 ppm) (2sd; n=44). Pb isotope measurements of the

74   standard are within error of the accepted values ($^{206}Pb/^{204}Pb$ = 16.9412, $^{207}Pb/^{204}Pb$ =

75   15.4988, $^{208}Pb/^{204}Pb$ = 36.7233 [45]). Accuracy was 47 ppm for $^{206}Pb/^{204}Pb$, 123 ppm for

76    $^{207}Pb/^{204}Pb$, and 174 ppm for $^{208}Pb/^{204}Pb$.

77   **$^{143}Nd/^{144}Nd$ and $^{87}Sr/^{86}Sr$ isotopic analysis**

78   For Sr and Nd analysis, remaining mother solutions from the preparation of trace element

79   solutions (see method above) was used for all samples except those of the BBVC. An aliquot

80   of each mother solution was used, to give a volume of liquid containing at least 1 µg Sr and

81   200 ng Nd and evaporated to dryness in Savillex Teflon vials on a hotplate at 130ºC. Sample

82   residues were reconstituted in 200 µl 1.75M HCl. For the BBVC samples, rock chips were

83   leached in 4 ml 6M HCl or 30 minutes in Savillex Teflon vials (obsidian samples for only 15

84   minutes, to avoid dissolution of the sample). The samples were then rinsed with Milli-Q

85   water and HNO_3, and then the same digestion procedure as for trace element analysis (above)

86   was followed. The final mother solutions were made up using HCl and Milli-Q water to 30

87   ml for felsic samples and 20 ml for mafic samples.

88   All samples were then passed through ion exchange column chemistry, using a AG50-X8

89   200-400 mesh resin cation column to separate the Sr and Nd fractions. The sample fractions

90   were subsequently evaporated to dryness ready for further column chemistry.

91   Sr was further isolated through Sr-spec resin column, following the methodology of [46].

92   Samples were then evaporated to dryness, dissolved in 1.5 ml 1M HCl and loaded onto

93   outgassed tantalum filaments with 1 µl of Ta-activator. Sr isotopic analysis was performed on

94   a ThermoScientific Triton Plus (TIMS) at the University of Southampton. Reference material

95   SRM NIST987 ($^{87}Sr/^{86}Sr$ = 0.710258; GeoREM) was used to monitor accuracy and gave

96   average $^{87}Sr/^{86}Sr$ values of 0.710243 and samples are quoted relative to 0.710248, while

97   reproducibility was ± 0.000020 (28.2 ppm, 2sd; n=464). Accuracy was 21 ppm.

98   The Nd aliquot from the cation column was followed by an Ln-spec resin (50-100 μm) [47].

99   The samples were then evaporated to dryness and 3% HNO_3 was added to produce a solution

100   of 50 ppb. $^{143}Nd/^{144}Nd$ analyses were undertaken on the ThermoScientific Neptune multi-

101   collector inductively coupled plasma mass spectrometer (MC-ICP-MS) at the University of

102   Southampton. Reference material JNdi was measured as a standard ($^{143}Nd/^{144}Nd$ of 0.512124,

103   2sd; [81]) produced an average $^{143}Nd/^{144}Nd$ of 0.512115 with an external reproducibility of

104   ±0.000008 (2sd, 15.2 ppm) across 6 analysis sessions over 2 years. The total column blanks

105   (i.e., when blank acid is run through the column procedure) were negligible (<20 pg)

106   compared to the total amounts analysed (1 μg and 200 μg) for Sr and Nd, respectively.

Geophysical analyses

Shear velocity maps from joint inversion of teleseismic and ambient noise Rayleigh-wave phase velocities

110   We use the shear wave velocity model of [22] for analysis. The 3D velocity model is created

111   through a joint inversion of Rayleigh wave phase velocities from ambient noise and

112   teleseisms [22] [29]. The shear velocity model is parameterised every 5 km vertically with

113   0.1° x 0.1° pixel size for the upper 50 km. For deeper depths, an irregular spacing was used

114   increasing from 10 – 50 km spacings to match that of [31]. For further details on creation of

115   the velocity model, see [22] [29] and references therein.

116   For the analysis in this paper, the shear velocity model was interpolated to 1 km depth using a

117   linear interpolation; we then extracted 1D columns of velocity with depth at the same

118   resolution as our pixel size.

119 **Moho depths**

120 The gridded Moho depth map was produced from the Vs maps of [29] described above. The

121 Vs model was interpolated to a vertical grid spacing of 1 km and a velocity slice at the 3.75

122 km/s contour was extracted which mapped best to previous receiver function measurements

123 [61, 63, 64, 65, 66], active source experiments (e.g., [67]) and previous S-wave models (e.g.,

124 [68]).

125 ## Statistical analysis

126 **Models considered**

127 Five models were considered (see Extended Data Table 1), with each model being tested

128 using a linear fit and a spline fit (Fig. 2). We note that a spline fit can fit a linear pattern to the

129 data if that is the best-fitting line.

130 Empirical models are estimated for the variation of each of 14 geochemical quantities (each

131 of which is represented generically by random variable Y) as a function of distance $d \in [0,$

132 1800] km for five different models. Models are specified that explore the variation of Y with

133 d in increasing complexity. The simplest model (C1C) assumes the existence of a single

134 upwelling centre (at 11.192 ∘N 41.784 ∘E, Fig. 1), with respect to which d is defined for all

135 three rifts; the variation of Y with d is assumed common to all rifts. Model C3C assumes the

136 existence of three upwelling centres (at 11.192 ∘N 41.784 ∘E, 14.008 ∘N 40.458 ∘E & 6.626

137 ∘N 37.948 ∘E, Fig. 1) based on [30]; observations are allocated to the nearest upwelling

138 centre, facilitating calculation of a single d for each observation; like model C1C, the

139 variation of Y with d is assumed common to all rifts, regardless of upwelling allocation.

140 Model C1D assumes one upwelling centre (like C1C) for calculation of d, but now the

141 variation of Y with d is assumed to be different across rifts. Model C3D duplicates C3C for

142 estimation of d, but variation of Y with d is assumed to be different across rifts. Finally, in

143   model C3X we consider the presence of three upwelling centres, with different variation of Y

144   with d for each combination of upwelling and rift.

Data pre-processing

146   For models C1C and C1D, the distance between each sample and the upwelling locus centred

147   on Lake Abhe (11.192170 ∘N 41.783750 ∘E) is calculated. For models C3C, C3D and C3X,

148   the distance between each sample and each of the three upwelling locations (Fig. 1) is

149   measured, and then each sample is assigned to its nearest upwelling centre. The distance (d)

150   between two locations (i.e., upwelling and sample) is calculated using the spherical cosine

151   law:

152    $$d \;=\; R(\cos^{-1}(\cos(a)\cos(b) \;+\; \sin(a)\sin(b)\cos(C)) \qquad\qquad \text{(Eq. 5)}$$

153   where a is the angle (in radians) from the North Pole to the sample location, b is the angle (in

154   radians) from the North Pole to the upwelling location, C is the difference in radians between

155   the longitude values of the sample and upwelling, and R is the radius of the earth in meters

156   (6371×10^3).

Penalised B-splines

158   For each model, the variation of Y with d (possibly for a subset of the full sample) is

159   described using a penalised B-spline (e.g., [69, 70]), the characteristics of which are selected

160   to provide optimal predictive performance. First, for a large index set of locations equally

161   spaced on the domain of distance, we calculate a B-spline basis matrix, B (e.g., [71])

162   consisting of p equally spaced cubic spline basis functions. Then the value of Y on the index

163   set is given by the vector $B\beta$, for spline coefficient vector β to be estimated. The value of p is

164   specified to be sufficiently large to provide a good description of a highly variable Y. For a

165   given data set, we penalise the difference between consecutive values in β using a roughness

166   penalty, such that the penalised spline provides optimal predictive performance.

167 **Estimating optimal spline roughness and predictive performance**

168 For a sample of n_1 training data, consisting of vectors of geochemical quantities (y_1) and

169 distances (d_1), we first allocate each element of d_1 to its nearest neighbour in the index set,

170 and hence construct the appropriate spline basis matrix B_1 for the sample. We then assume

171 that $y_1 = B_1\beta + \varepsilon$, where the elements of ε are independently and identically distributed zero-

172 mean Gaussian random variables. We penalise the roughness of β using a first-different

173 penalty $\lambda\beta'P\beta$, where $P = D'D$ and D is a first difference matrix (with elements $D_{ij} = -1$ if $i =$

174 $j;\ = 1$ if $j = i + 1$; and $= 0$ otherwise (e.g., [72]). For a given choice of λ, we then find the

175 optimal value of β by minimising lack of fit:

176 $$\beta^*(\lambda) = \mathrm{argmin}\beta(y_1 - B_1\beta)'(y_1 - B_1\beta)' + \lambda\beta'P\beta \qquad \text{(Eq. 6)}$$

177 $$= (B_1'B_1 + \lambda P)^{-1}B_1'y_1 \qquad \text{(Eq. 7)}$$

178 We can evaluate the predictive performance of the resulting spline description using a tuning

179 set of n_2 observations (independent of the training set) represented by vectors y_2 and d_2. We

180 again start by finding the appropriate spline basis matrix B_2 for this sample. Then we can

181 calculate the predictive mean square error for the tuning sample

182 $$\mathrm{MSE}_2(\lambda) = \frac{1}{n_2}\big(y_2 - B_2\beta^*(\lambda)\big)'\big(y_2 - B_2\beta^*(\lambda)\big) \qquad \text{(Eq. 8)}$$

183 for each of a set of representative choices of values for λ. We can then select the optimal

184 value of λ using

185 $$\lambda^* = \mathrm{argmin}\,\lambda\mathrm{MSE}_2(\lambda) \qquad \text{(Eq. 9)}$$

186 The value $\mathrm{MSE}_2(\lambda^*)$ is a biased estimate of predictive performance since the value of λ^* was

187 tuned to minimise its value. We can obtain an unbiased estimate for the predictive

188 performance of the spline model using a test set of n_3 observations (independent of the

189   training and tuning sets) represented by vectors y_3 and d_3 (and corresponding spline basis

190   matrix B_3). Then the predictive performance is estimated using:

191   $\text{MSE} = \frac{1}{n_3}\left(y_3 - B_3\beta^*(\lambda^*)\right)'\left(y_3 - B_3\beta^*(\lambda^*)\right)$ (Eq. 10)

192

193   **Cross-validation and model comparison**

194   We exploit cross-validation to evaluate MSE, by partitioning the full sample of data into $k >$

195   2 groups at random, withholding one group for tuning, another group for testing, retaining the

196   remaining $k - 2$ groups for training. We then loop exhaustively over all possible

197   combinations of choice of train, tune, and test groups, evaluating overall predictive

198   performance on the test data over all iterations, noting that each observation occurs exactly

199   once in the test set. For models (that is, C1D, C3D, C3X) requiring separate model fits to

200   subsets of data, MSE is estimated using predictions from optimal predictive models for each

201   subset. Further, we can repeat the analysis for different initial random partitioning of

202   observations into k groups, to assess the sensitivity of overall predictive performance to this

203   choice. We are careful to use the same cross-validation partitions to evaluate each of the five

204   models, so that predictive performances can be compared fairly.

205   To quantify model performance over all 14 geochemical quantities, we define the overall

206   standardised MSE

207   $\text{SMSE} = \sum_{j=1}^{14} \frac{\text{MSE}_j}{s_j^2}$ (Eq. 11)

208   where MSE_j is the predictive performance for the jth quantity, and s_j^2 is the sample estimate

209   for the variance of that quantity. The estimation of the splines and the testing of their

210   predictive performance was repeated over 100 iterations. Results from each iteration and the

211   mean of the SMSE is shown in Fig. 3.

212 **Linear regression**

213 For comparison, we also evaluate linear regression models for the variation of Y with d. In the

214 current notation, these can be thought of as simple models with basis matrix $B = [1\ d]$, where

215 *1* is a vector of appropriate length with each element $= 1$. B in this case is a 2-vector with

216 elements corresponding to intercept and slope coefficients. Linear regression is approached

217 using penalised B-spline models as the roughness coefficient $\lambda \to \infty$. That is, linear

218 regression corresponds to a penalised B-spline model with very large λ. Therefore, a

219 penalised B-spline model is guaranteed to perform at least as well as linear regression.

220 **Principal component analysis**

221 Principal component analysis (PCA) requires each sample or object to have the same number

222 of values for each variable and so the dataset was reduced to 94 samples. PCA is only carried

223 out on radiogenic isotope compositions of the samples where data are available for the mantle

224 end members investigated (i.e., Afar plume, Pan-African Lithosphere, Depleted Mantle,

225 Enriched Mantle I, Enriched Mantle II, HiMU, Extended Data Fig. 4). Values used for the

226 end members is provided in Extended Data Table 4. Each object is standardised before being

227 included in the PCA:

228 $y_{\mathrm{std}j} = \dfrac{y_j - \overline{y_j}}{\sigma_j}$ (Eq. 12)

229 where $\overline{y_j}$ is the mean of variable j, and σ_j is the standard deviation of the variable j:

230 $\sigma_j = \sqrt{\dfrac{\Sigma\{(y_j - \overline{y_j})\}^2}{N_j}}$ (Eq. 13)

231 where N_j is the number of objects within variable j.

232   Approximately 90.5% of the variance is explained within the plane of the first two

233   eigenvectors, increasing to 95.5% when including the third eigenvector. The first principal

234   component (PC-1) is most influenced by ^{207}Pb/^{204}Pb, ^{208}Pb/^{204}Pb, whereas the second

235   principal component (PC-2) is dominantly influenced by ^{206}Pb/^{204}Pb and ^{87}Sr/^{86}Sr. The third

236   principal component (PC-3) is dominated by ^{207}Pb/^{204}Pb and ^{143}Nd/^{144}Nd (Extended Data

237   Table 5).

238   **K-means cluster analysis**

239   K-means cluster analysis [73] is carried out on the samples using the 13 standardised

240   variables, which are ^{206}Pb/^{204}Pb, ^{207}Pb/^{204}Pb, ^{208}Pb/^{204}Pb, ^{143}Nd/^{144}Nd, ^{87}Sr/^{86}Sr, Ce/Pb,

241   La/Sm, ΔNb, shear-wave speed at 40 km, 60 km, 80 km, 100 km, and 120 km depths. The K-

242   means algorithm assigns each object to a singular cluster that does not overlap with another

243   (i.e., partitional clustering), minimising the total sum of squared error (SSE) from the centre

244   point of each cluster, known as the centroid, to each object.

245   To find the optimum number of clusters (k), i.e., which reduces the within-cluster total sum

246   of squares error with the lowest number of clusters, we run the K-means algorithm specifying

247   k to be 1:20, over 1000 iterations for each k (Extended Data Fig. 3). We then select eight

248   clusters based on $k=8$ reducing the within-cluster total sum of squares by 75% from $k=1$, and

249   the range over the 1,000 iterations being minimised when $k \geq 8$. The cluster assignments for

250   each object, out of the 1,000 iterations, are selected by finding the iteration number that is

251   closest to the mean within-cluster total sum of squares of that k value (shown by the blue line

252   in Extended Data Fig. 3).

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