**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, 87Sr/86Sr, 143Nd/144Nd, 206Pb/204Pb, 207Pb/204Pb and 208Pb/204Pb 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, (http://repositories.dst.unipi.it/index.php/home-afar). A further 52 samples from the Boset- Bericha Volcanic Complex (BBVC) were collected during three field seasons, in November 2012, April-May 2015, and February 2017 [43].

Sample preparation for major, trace and isotope analyses was carried out at the University of Southampton. Samples were cut with a saw to remove any sections, and any cut surfaces ground down to reduce any potential contamination by metals from the saw blade. Rock samples were then crushed using a fly press and placed in double-layered plastic bags prior to crushing to minimise metal contamination.

The crushed material was separated into three size fractions (>1 mm, 0.5 mm to 1 mm, <0.5 mm) using Teflon sieves keeping the middle fraction (0.5 - 1 mm). The selected fraction was cleaned by ultrasonicating in Milli-Q water then dried overnight in an oven at 85ºC. The cleaned rock chips were then hand-picked under a microscope, to remove any non-rock material. An aliquot of cleaned chips were used for Pb isotope analysis. For major element, trace element, and 143Nd/144Nd and 87Sr/86Sr isotope analysis, the remaining rock chips were ground to a fine powder using an agate mortar and pestle, to minimise contamination with metals.

**Trace element analysis**

Samples were prepared for whole-rock trace element analysis using 0.05 g (for BBVC samples) or 0.075 g (for all other samples) powdered sample. The powdered samples were digested in sealed Savillex Teflon vials with 15 drops concentrated HNO3 and 2 ml HF on a hotplate at 130 °C for 24 hours (for all other samples), or with 50 drops HF and 2 ml HNO3 on a hotplate at 130 °C for 24 hours (for BBVC samples). The HNO3/HF was evaporated off, and the samples were refluxed in 6M HCl for another 24 hours on a hotplate at 130 °C. The 6M HCl was evaporated off, and the samples were redissolved in 6M HCl. Mother solutions were prepared by adding 6M HCl and Milli-Q water (total 30 ml) to the dissolved samples. Daughter solutions were prepared using 0.5 ml of mother solution, diluted to 5 ml with 3% HNO3 (containing the internal standards In/Re/Be), resulting in an overall dilution factor of c. 4000.

Trace element analyses of the daughter samples were undertaken on the ThermoScientific XSeries2 quadrupole inductively coupled plasma mass spectrometer (ICP-MS) at the University of Southampton. Analytical standards and accuracy were monitored using international standards JA-2, BCR-2, JB-2 (see Extended Data Table 3). Results in counts per second were processed with the exact calculated dissolution factors and blank corrections to produce concentrations in ppm for the trace elements analysed.

**Pb isotopic analysis**

For Pb isotope analysis, 0.3 g of cleaned, picked rock chips (0.5 mm to 1 mm) were weighed into dedicated Pb Savillex Teflon vials and leached on a hotplate with 4 ml 6M HCl for an hour (15 minutes for obsidian and pumice samples). Samples were rinsed several times in Milli-Q water, then 0.5 ml concentrated HNO3, before adding 3-4 ml of concentrated HF. Samples were refluxed on a hotplate at 140 °C for 24 hours, and then evaporated to dryness. 0.5 ml concentrated HCl was added, and the sample evaporated to dryness, then 0.5 ml concentrated HNO3 was added and again evaporated to dryness. The final residue was reconstituted in 0.5 ml HBr and refluxed for an hour. The samples were cooled and centrifuged for 5 minutes. Pb was isolated using a single-stage HCl anion-exchange chromatographic resin separation method [44], with AGX-1x8, 200 – 400 mesh resin. Following this, the Pb isolate was dried down, redissolved in HNO3, and analysed using the double spike method of [45]. The samples were subsequently analysed on a ThermoScientific Neptune MC-ICPMS at the University of Southampton (UK) with a NBS SRM 981 reproducibility of 206Pb/204Pb = 16.9404 ± 24 (142 ppm), 207Pb/204Pb = 15.4969 ± 26 (168 ppm), 208Pb/204Pb = 36.7149 ± 66 (180 ppm) (2sd; n=44). Pb isotope measurements of the standard are within error of the accepted values (206Pb/204Pb = 16.9412, 207Pb/204Pb = 15.4988, 208Pb/204Pb = 36.7233 [45]). Accuracy was 47 ppm for 206Pb/204Pb, 123 ppm for 207Pb/204Pb, and 174 ppm for 208Pb/204Pb.

**143Nd/144Nd and 87Sr/86Sr isotopic analysis**

For Sr and Nd analysis, remaining mother solutions from the preparation of trace element solutions (see method above) was used for all samples except those of the BBVC. An aliquot of each mother solution was used, to give a volume of liquid containing at least 1 μg Sr and 200 ng Nd and evaporated to dryness in Savillex Teflon vials on a hotplate at 130ºC. Sample residues were reconstituted in 200 μl 1.75M HCl. For the BBVC samples, rock chips were leached in 4 ml 6M HCl or 30 minutes in Savillex Teflon vials (obsidian samples for only 15 minutes, to avoid dissolution of the sample). The samples were then rinsed with Milli-Q water and HNO3, and then the same digestion procedure as for trace element analysis (above) was followed. The final mother solutions were made up using HCl and Milli-Q water to 30 ml for felsic samples and 20 ml for mafic samples.

All samples were then passed through ion exchange column chemistry, using a AG50-X8 200-400 mesh resin cation column to separate the Sr and Nd fractions. The sample fractions were subsequently evaporated to dryness ready for further column chemistry.

Sr was further isolated through Sr-spec resin column, following the methodology of [46]. Samples were then evaporated to dryness, dissolved in 1.5 ml 1M HCl and loaded onto outgassed tantalum filaments with 1 μl of Ta-activator. Sr isotopic analysis was performed on a ThermoScientific Triton Plus (TIMS) at the University of Southampton. Reference material SRM NIST987 (87Sr/86Sr = 0.710258; GeoREM) was used to monitor accuracy and gave average 87Sr/86Sr values of 0.710243 and samples are quoted relative to 0.710248, while reproducibility was ± 0.000020 (28.2 ppm, 2sd; n=464). Accuracy was 21 ppm.

The Nd aliquot from the cation column was followed by an Ln-spec resin (50-100 μm) [47]. The samples were then evaporated to dryness and 3% HNO3 was added to produce a solution of 50 ppb. 143Nd/144Nd analyses were undertaken on the ThermoScientific Neptune multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS) at the University of Southampton. Reference material JNdi was measured as a standard (143Nd/144Nd of 0.512124, 2sd; [81]) produced an average 143Nd/144Nd of 0.512115 with an external reproducibility of ±0.000008 (2sd, 15.2 ppm) across 6 analysis sessions over 2 years. The total column blanks (i.e., when blank acid is run through the column procedure) were negligible (<20 pg) 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**

We use the shear wave velocity model of [22] for analysis. The 3D velocity model is created through a joint inversion of Rayleigh wave phase velocities from ambient noise and teleseisms [22] [29]. The shear velocity model is parameterised every 5 km vertically with 0.1° x 0.1° pixel size for the upper 50 km. For deeper depths, an irregular spacing was used increasing from 10 – 50 km spacings to match that of [31]. For further details on creation of the velocity model, see [22] [29] and references therein.

For the analysis in this paper, the shear velocity model was interpolated to 1 km depth using a linear interpolation; we then extracted 1D columns of velocity with depth at the same resolution as our pixel size.

**Moho depths**

The gridded Moho depth map was produced from the Vs maps of [29] described above. The Vs model was interpolated to a vertical grid spacing of 1 km and a velocity slice at the 3.75 km/s contour was extracted which mapped best to previous receiver function measurements [61, 63, 64, 65, 66], active source experiments (e.g., [67]) and previous S-wave models (e.g., [68]).

**Statistical analysis**

**Models considered**

Five models were considered (see Extended Data Table 1), with each model being tested using a linear fit and a spline fit (Fig. 2). We note that a spline fit can fit a linear pattern to the data if that is the best-fitting line.

Empirical models are estimated for the variation of each of 14 geochemical quantities (each of which is represented generically by random variable Y) as a function of distance d ∈ [0, 1800] km for five different models. Models are specified that explore the variation of Y with d in increasing complexity. The simplest model (C1C) assumes the existence of a single upwelling centre (at 11.192 ◦N 41.784 ◦E, Fig. 1), with respect to which d is defined for all three rifts; the variation of Y with d is assumed common to all rifts. Model C3C assumes the existence of three upwelling centres (at 11.192 ◦N 41.784 ◦E, 14.008 ◦N 40.458 ◦E & 6.626 ◦N 37.948 ◦E, Fig. 1) based on [30]; observations are allocated to the nearest upwelling centre, facilitating calculation of a single d for each observation; like model C1C, the variation of Y with d is assumed common to all rifts, regardless of upwelling allocation. Model C1D assumes one upwelling centre (like C1C) for calculation of d, but now the variation of Y with d is assumed to be different across rifts. Model C3D duplicates C3C for estimation of d, but variation of Y with d is assumed to be different across rifts. Finally, in model C3X we consider the presence of three upwelling centres, with different variation of Y with d for each combination of upwelling and rift.

**Data pre-processing**

For models C1C and C1D, the distance between each sample and the upwelling locus centred on Lake Abhe (11.192170 ◦N 41.783750 ◦E) is calculated. For models C3C, C3D and C3X, the distance between each sample and each of the three upwelling locations (Fig. 1) is measured, and then each sample is assigned to its nearest upwelling centre. The distance (*d*) between two locations (i.e., upwelling and sample) is calculated using the spherical cosine law:

(Eq. 5)

where *a* is the angle (in radians) from the North Pole to the sample location, *b* is the angle (in radians) from the North Pole to the upwelling location, *C* is the difference in radians between the longitude values of the sample and upwelling, and *R* is the radius of the earth in meters (6371 x 103).

**Penalised B-splines**

For each model, the variation of *Y* with *d* (possibly for a subset of the full sample) is described using a penalised B-spline (e.g., [69, 70]), the characteristics of which are selected to provide optimal predictive performance. First, for a large index set of locations equally spaced on the domain of distance, we calculate a B-spline basis matrix, *B* (e.g., [71]) consisting of *p* equally spaced cubic spline basis functions. Then the value of *Y* on the index set is given by the vector *Bβ*, for spline coefficient vector *β* to be estimated. The value of p is specified to be sufficiently large to provide a good description of a highly variable *Y*. For a given data set, we penalise the difference between consecutive values in *β* using a roughness penalty, such that the penalised spline provides optimal predictive performance.

**Estimating optimal spline roughness and predictive performance**

For a sample of *n1* training data, consisting of vectors of geochemical quantities (*y1*) and distances (*d1*), we first allocate each element of *d1* to its nearest neighbour in the index set, and hence construct the appropriate spline basis matrix *B1* for the sample. We then assume that *y1 = B1β + ε*, where the elements of ε are independently and identically distributed zero-mean Gaussian random variables. We penalise the roughness of *β* using a first-different penalty *λβ′P β*, where *P =D′D* and *D* is a first difference matrix (with elements *Dij = −1* if *i = j; = 1* if *j = i + 1*; and *= 0* otherwise (e.g., [72]). For a given choice of *λ*, we then find the optimal value of *β* by minimising lack of fit:

(Eq. 6)

(Eq. 7)

We can evaluate the predictive performance of the resulting spline description using a tuning set of *n2* observations (independent of the training set) represented by vectors *y2* and *d2*. We again start by finding the appropriate spline basis matrix *B2* for this sample. Then we can calculate the predictive mean square error for the tuning sample

(Eq. 8)

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

(Eq. 9)

The value is a biased estimate of predictive performance since the value of was tuned to minimise its value. We can obtain an unbiased estimate for the predictive performance of the spline model using a test set of *n3* observations (independent of the training and tuning sets) represented by vectors *y3* and *d3* (and corresponding spline basis matrix *B3*). Then the predictive performance is estimated using:

(Eq. 10)

**Cross-validation and model comparison**

We exploit cross-validation to evaluate MSE, by partitioning the full sample of data into *k > 2* groups at random, withholding one group for tuning, another group for testing, retaining the remaining *k − 2* groups for training. We then loop exhaustively over all possible combinations of choice of train, tune, and test groups, evaluating overall predictive performance on the test data over all iterations, noting that each observation occurs exactly once in the test set. For models (that is, C1D, C3D, C3X) requiring separate model fits to subsets of data, MSE is estimated using predictions from optimal predictive models for each subset. Further, we can repeat the analysis for different initial random partitioning of observations into k groups, to assess the sensitivity of overall predictive performance to this choice. We are careful to use the same cross-validation partitions to evaluate each of the five models, so that predictive performances can be compared fairly.

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

(Eq. 11)

where *MSEj* is the predictive performance for the *j*th quantity, and is the sample estimate for the variance of that quantity. The estimation of the splines and the testing of their predictive performance was repeated over 100 iterations. Results from each iteration and the mean of the SMSE is shown in Fig. 3.

**Linear regression**

For comparison, we also evaluate linear regression models for the variation of *Y* with *d*. In the current notation, these can be thought of as simple models with basis matrix *B = [1 d]*, where *1* is a vector of appropriate length with each element *= 1. Β* in this case is a 2-vector with elements corresponding to intercept and slope coefficients. Linear regression is approached using penalised B-spline models as the roughness coefficient *λ → ∞*. That is, linear regression corresponds to a penalised B-spline model with very large *λ*. Therefore, a penalised B-spline model is guaranteed to perform at least as well as linear regression.

**Principal component analysis**

Principal component analysis (PCA) requires each sample or object to have the same number of values for each variable and so the dataset was reduced to 94 samples. PCA is only carried out on radiogenic isotope compositions of the samples where data are available for the mantle end members investigated (i.e., Afar plume, Pan-African Lithosphere, Depleted Mantle, Enriched Mantle I, Enriched Mantle II, HiMU, Extended Data Fig. 4). Values used for the end members is provided in Extended Data Table 4. Each object is standardised before being included in the PCA:

(Eq. 12)

where is the mean of variable *j*, and *σj* is the standard deviation of the variable *j*:

(Eq. 13)

where *Nj* is the number of objects within variable *j*.

Approximately 90.5% of the variance is explained within the plane of the first two eigenvectors, increasing to 95.5% when including the third eigenvector. The first principal component (PC-1) is most influenced by 207Pb/204Pb, 208Pb/204Pb, whereas the second principal component (PC-2) is dominantly influenced by 206Pb/204Pb and 87Sr/86Sr. The third principal component (PC-3) is dominated by 207Pb/204Pb and 143Nd/144Nd (Extended Data Table 5).

**K-means cluster analysis**

K-means cluster analysis [73] is carried out on the samples using the 13 standardised variables, which are 206Pb/204Pb, 207Pb/204Pb, 208Pb/204Pb, 143Nd/144Nd, 87Sr/86Sr, Ce/Pb, La/Sm, ∆Nb, shear-wave speed at 40 km, 60 km, 80 km, 100 km, and 120 km depths. The K-means algorithm assigns each object to a singular cluster that does not overlap with another (i.e., partitional clustering), minimising the total sum of squared error (SSE) from the centre point of each cluster, known as the centroid, to each object.

To find the optimum number of clusters (*k*), i.e., which reduces the within-cluster total sum of squares error with the lowest number of clusters, we run the K-means algorithm specifying *k* to be 1:20, over 1000 iterations for each *k* (Extended Data Fig. 3). We then select eight clusters based on *k=8* reducing the within-cluster total sum of squares by 75% from *k=1*, and the range over the 1,000 iterations being minimised when *k ≥8*. The cluster assignments for each object, out of the 1,000 iterations, are selected by finding the iteration number that is closest to the mean within-cluster total sum of squares of that *k* value (shown by the blue line in Extended Data Fig. 3).

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