

Do We Really Need Mantle Components to Define Mantle Composition?

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We discuss the concept of components in the Earth's mantle starting from a petrological and geochemical approach, but adopting a new method of projection of geochemical and isotopic data. This allows the compositional variability of magmatic associations to be evaluated in multi-dimensional space, thus simultaneously accounting for a large number of compositional variables. We demonstrate that ocean island basalts (OIB) and mid-ocean ridge basalts (MORB) are derived from a marble-cake mantle, in which different degrees of partial melting of recycled lithosphere, which are heterogeneous in age and composition, contribute to the magma genesis. This view is supported by the variability in the geochemical and isotopic signatures of OIB that are observed on the scale of a single ocean island as well as on that of an ocean, mostly varying between two extreme compositions, that are not strictly related to the commonly accepted mantle components (DMM, EMI, EMII, HIMU). Rather they are a distinctive feature of the mantle source sampled at each ocean island and are strongly dependent on the Pb isotope system. We recommend a change in perspective in studies of MORB–OIB geochemistry from one based on physically distinct mantle components to a model based on the existence of a marble-cake-like upper mantle. Although resembling the statistical upper mantle, this model implies that geochemical homogenization can be attained only within the limits of local mantle composition, so that a world-wide uniform depleted reservoir cannot be sampled by simply extending the volume of the region undergoing partial melting.

KEY WORDS: *geochemistry; isotope; mantle; OIB*

INTRODUCTION

The isotopic heterogeneity of oceanic basalts [mid-ocean ridge basalts (MORB) and ocean island basalts (OIB)] has been described by many researchers in terms of the interaction of a small number of mantle components

[Depleted Mantle (DMM); Enriched Mantle I (EMI); Enriched Mantle II (EMII); High- μ (HIMU); White, 1985; Zindler & Hart, 1986; Allègre *et al.*, 1987; Hart *et al.*, 1992; Hanan & Graham, 1996; Hofmann, 1997; Saal *et al.*, 2005; Stracke *et al.*, 2005]. Although these were not originally envisaged as distinct and physically accessible mantle portions, there has been some confusion as to what these components actually represent. The concept of a mantle component stems from the geochemical differences between MORB and OIB, respectively considered to originate from a degassed (low ^3He , ^{40}Ar), incompatible element-poor portion of the shallow mantle, and from mantle plumes sampling undegassed, lithophile element-enriched deep mantle (Jacobsen & Wasserburg, 1979; O'Nions *et al.*, 1979; Allègre *et al.*, 1980; Hofmann, 1997; Graham *et al.*, 2001; Saal *et al.*, 2002). The geochemical evidence for the existence of distinct, relatively homogeneous, upper and lower mantle reservoirs, and hence a two-layer mantle convection regime, was inferred from mass-balance calculations on heat production caused by radioactive decay and/or the isotopic budget of incompatible elements (Allègre *et al.*, 1996; Blichert-Toft & Albarède, 1997; Hauri & Hart, 1997; Albarède, 1998; Moreira & Allègre, 1998; Davies, 1999; Kellogg *et al.*, 1999; Allègre *et al.*, 2001; Helffrich & Wood, 2001; Turcotte *et al.*, 2001). Nevertheless, this model is hard to reconcile with geophysical evidence for whole-mantle convection (Hart *et al.*, 1992; Grand, 1994; Stein & Hofmann, 1994; van der Hilst *et al.*, 1997; Tackley, 2000). In addition, in the last decade geochemical characterization of the mantle components sampled by MORB and OIB has changed following improvements in analytical techniques for both radiogenic and stable isotope systems (e.g. Blichert-Toft *et al.*, 1997; Luais *et al.*, 1997; Galer, 1999; Maréchal *et al.*, 1999; White *et al.*, 2000; Nishio & Nakai, 2002). The new data have

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provided growing evidence that oceanic basalts are heterogeneous on different scales and do not represent a specific mixture of the well-known mantle components (DMM, EMI, EMII, HIMU). This has led to the conclusion that the correspondence between mantle components and specific mantle reservoirs is highly speculative. As a consequence, it is now more generally accepted that mantle processes, rather than a limited number of physically distinct components or reservoirs, are responsible for variability in the geochemical signatures of mantle-derived partial melts (e.g. Hauri, 2000; Kellogg *et al.*, 2002).

The conventional view on the origin of basaltic magmas (*sensu lato*) assigns a major role to variable degrees of partial melting of a peridotite source (McKenzie & O'Nions, 1991). At least at high pressures, and for relatively Ol-rich melts, experimental data on peridotite melting could explain the compositional variability of most near-primary OIB lavas (e.g. Keshav *et al.*, 2004). However, there is increasing recognition of the role played by olivine-poor sources in the genesis of mantle partial melts (e.g. Carlson & Nowell, 2001; Sobolev *et al.*, 2005; Fig. 1). Even if the dominant source of basalts is the convecting asthenospheric mantle, the process of subduction provides an efficient mechanism for the continuous recycling of pyroxenite-veined oceanic lithospheric mantle and of its cap of MORB and its differentiates. The Ca-Tschermak–Olivine–Quartz (CaTs–Ol–Q) diagram of Fig. 1 illustrates the compositions of a large dataset of OIBs along with relevant peridotite and pyroxenite melting relations. Following Hirschmann *et al.* (2003), it seems likely that less alkaline basic melts (tholeiites, picrites, picrobasalts) are mostly linked to high degrees of partial melting of a rather shallow peridotite source, whereas more alkaline melts (nephelinites, basanites, alkali basalts), although reflecting a deeper origin at lower partial melting degrees, may be also linked to an enriched source. Mantle sources sampled by these alkaline melts may be related to melting of metasomatic pyroxenite veins, as well as to the solid residuum of the extraction of silicic melts from an eclogitic source (Hirschmann *et al.*, 2003). Nevertheless, other researchers have denied any important role for pyroxenitic material in the chemical budget of OIB (Keshav *et al.*, 2004; for a recent review on this topic refer to Lustrino, 2006).

The wide variety of mantle mineral assemblages that can account for the genesis of the spectrum of MORB–OIB primary melts suggests that individual basic magmas should inherit an isotope signature reflecting the heterogeneity of the specific mantle portion in which partial melting occurred. Meibom & Anderson (2003) suggested that the upper mantle is characterized by a statistical distribution of small- to moderate-scale heterogeneities (the Statistical Upper Mantle Assemblage; SUMA) that could account for both MORB and OIB characteristics, without the need for any specific mantle reservoir or

thermal convection (see also Rudge *et al.*, 2005). The composition of oceanic basalts would thus reflect the complex history of their sources in which partial melting events and recycling by passive convection of oceanic and continental lithosphere cause heterogeneity, whereas homogenization is provided by the extent of sampling allowed by the volume of the source region and the degree of partial melting (Anderson, 2006). From this perspective, it is unlikely that specific compositions related to distinct mantle reservoirs or components will be identifiable.

In this paper we use the following definitions:

compositional parameter, a geochemical variable used to define the composition of samples (concentrations or elemental and isotope ratios);

mantle component, an extreme in geochemical composition existing independently of the chosen set of compositional parameters adopted for its identification, hence it should be identifiable in all the sub-spaces defined by the compositional parameters;

end-member, a fixed geochemical composition defined by a set of n compositional parameters; mixing of a given number of end-members is able to generate all the compositions found in a set of analyses of natural samples;

mantle reservoir, a mantle portion, physically distinct and potentially accessible, that may result from mixing of mantle components.

We discuss the concept of mantle component with respect to the evaluation of some key processes responsible for MORB–OIB heterogeneity. We assess the range of isotope and trace element variability of oceanic basalts and bind them simultaneously to a large number of compositional parameters using a novel multi-dimensional data plotting approach. This systematic and simultaneous evaluation of both trace element and isotopic data provides a more robust insight into the chemical structure of the mantle.

METHOD OF PROJECTION OF GEOCHEMICAL DATA

An n -dimensional space (R^n) relative to n ($n \geq 3$) independent variables can be projected into three-dimensional ($3D$) space (R^3) by choosing four points (hereafter referred to as 'end-members'; Table 1) that play the role of 'generating' the R^3 space into which the dataset in R^n is to be projected. End-members can assume an arbitrary position in R^3 and represent the image of four points in R^n : they must not be linearly dependent (i.e. non co-planar), both in R^3 and in R^n , and allow rescaling of the position of all data points in R^n into R^3 by projection (Albarède, 1995). For the sake of simplicity, end-members can be constrained to plot at the vertices of a regular tetrahedron (see Appendix A). Relationships in R^n are maintained in R^3 ; thus planar relationships in n -dimensional space are also

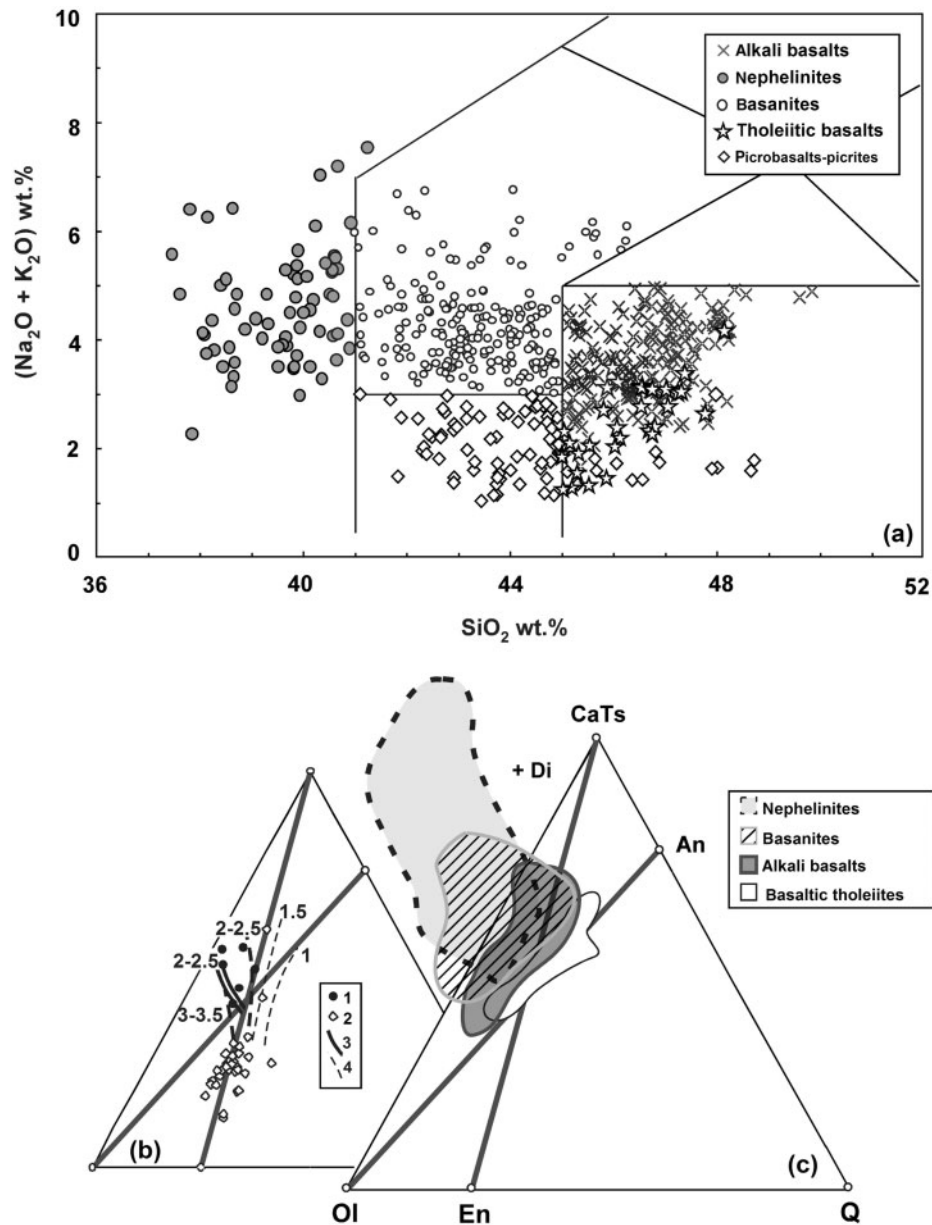


Fig. 1. (a) Classification of the OIB dataset according to the total alkalis–silica (TAS) diagram (Le Maitre, 1989). (b, c) Projection of major element data on the CaTs–Ol–Q face of the CaTs–Ol–Di tetrahedron, which is made by recasting the major element data according to O'Hara (1968) and the molecular proportions of Table 1. The CaTs–En join acts as a thermal divide at pressures >2 GPa and separates undersaturated compositions from Hy-normative ones; the Ol–An join separates Hy-normative lavas from Ol–Hy-normative ones. (b) Overview of experimental data on partial melts of pyroxenites from different sources: (1) Hirschmann *et al.* (2003); (2) Falloon *et al.* (2001); (3) Keshav *et al.* (2004); (4) Hirose & Kushiro (1993) and Falloon *et al.* (1997). (c) Fields of primitive (mg-number >63; details in the text) magmas from OIB settings; data from GEOROC database (<http://georoc.mpch-mainz.gwdg.de/Start.asp>).

planar in 3D space. Trace element and isotope ratios can be plotted simultaneously to represent the data. Such data share the characteristic not to be constrained to be closed to a constant sum. Mixing lines are not necessarily segments of straight lines. On the other hand, plots based on major element analyses are such that their sum is a constant. For major element data the processes of mixing or

subtraction of a phase(s) with a constant composition are represented by straight line segments (Pearce, 1987; Russel & Stanley, 1990). Performing some simulations, it is easy to check how a distribution of points, generated by adding random amounts of the end-members to an initial composition [e.g. the reference composition (RC); see Appendix B], generates a cluster of data points that

Table 1: Matrix to obtain a weight or molecular projection in the system *Q–Ol–CaTs–Di* in terms of CMAS components (O'Hara, 1968)

Mineral	Molecular proportions					Molecular proportions					Molecular weights			
	C	M	A	S		C	M	A	S		C	M	A	S
Q =	(0	0	0	SiO ₂)	=	0	0	0	1	=	0	0	0	60.09
Ol =	(0	2/3 MgO	0	1/3 SiO ₂)	=	0	2/3	0	1/3	=	0	26.69	0	20.03
CaTs =	(1/3 CaO	0	1/3 Al ₂ O ₃	1/3 SiO ₂)	=	1/3	0	1/3	1/3	=	18.69	0	33.99	20.00
Di =	(1/4 CaO	1/4 MgO	0	2/4 SiO ₂)	=	1/4	1/4	0	2/4	=	14.02	10.01	0	30.45

C = [mol. prop. (CaO – 3.33P₂O₅ + 2Na₂O + 2K₂O)] × 56.08; M = [mol. prop. (FeO + MnO + NiO + MgO – TiO₂)] × 40.31; A = [mol. prop. (Al₂O₃ + Cr₂O₃ + Fe₂O₃ + Na₂O + K₂O + TiO₂)] × 101.96; S = [mol. prop. (SiO₂ – 2Na₂O – 2K₂O)] × 60.09. Compositions are represented in molecular proportions in the system CaO–MgO–Al₂O₃–SiO₂ adopting for the four vertices of the tetrahedron: Q = S, Ol = M₂S, CaTs = CAS, Di = CMS₂. A dataset can be easily recast following the procedure of O'Hara (1968), according to a subset of components within CMAS, by simply changing the end-members and assigning their compositions with respect to the CMAS system.

projects within the tetrahedron, without any particular symmetry. The prevalent addition of a specific composition generates a trend lying on a mixing line. Distributions generated by random variations of each analytical compositional parameter show a symmetric elongation that is linked to the intrinsic variability of each component in the dataset (see Appendix B).

To allow evaluation of this new method of projection, two Excel files (anamorfosis.xls and data.xls) are provided for downloading from <http://www.petrology.oxfordjournals.org>. The use of these files is straightforward and self-explanatory. Details of the method of projection are given in Appendix A. The Excel files are set for the Italian language by default. Provide in cells A2–E2 the denomination for the graph window in the language your computer is set: it is required for the proper setting of the basic code. It is a requirement to assign the same set of *n* compositional parameters to describe the geochemical compositions of the four end-members, represented at each tetrahedron vertex (cells Q39–T48 in anamorfosis.xls), as well as to provide the data to process in data.xls. The data file of interest should be formatted exactly as in data.xls (e.g. using the copy/paste function of Windows and overwriting the analyses in the sample file), and its pathname has to be reported in cell N3 of the 'tetrahedron' spreadsheet of anamorfosis.xls. The number (*n*) of the compositional parameters to be plotted can vary between three and ten, and each parameter can be included or excluded from the evaluation of the tetrahedral coordinates (τ_1 , τ_2 , τ_3 , τ_4) of each point, by simply setting to one or zero the values given in cells U39–U48 in anamorfosis.xls. Macros in anamorfosis.xls allow the program to run all the calculations automatically to process the data for projection, by using the coloured buttons in the 'tetrahedron' spreadsheet. The comments in this sheet may be checked to view the defined

values and obtain more detailed information. Buttons are pressed to load ('Load data' button), then project ('ANAMORFOSIS') and finally plot ('Plot data') data, as well as to rotate ('vertical rotation' and 'horizontal rotation') the tetrahedron. The addition or suppression of one or more compositional parameters, as well as the choice of a different end-member, requires recasting of the projection by pressing again the 'ANAMORFOSIS' button in anamorfosis.xls. The spreadsheet can be saved with a new name but, in this case, the new name must be updated in cell N1 in sheet 'tetraedro'.

GEOCHEMICAL AND ISOTOPIC VARIABILITY OF MORB AND OIB

We compared trace element and isotopic (Sr, Nd, Pb) data for oceanic alkali basalts, basanites, nephelinites, picrites and tholeiites, representative of OIB magmatism, and MORB data, from the Pacific, Atlantic and Indian oceans from the GEOROC database (<http://georoc.mpch-mainz.gwdg.de/Start.asp>). Hf isotope data are too sparse to be processed together with the Sr–Nd–Pb isotope data. We selected about 1500 analyses for which loss on ignition is less than 2.2 wt % and mg-number is >63. In our selection, the composition of olivine in equilibrium with the bulk-rock ranges between Fo 63 and 85, assuming $K_d(\text{Fe/Mg}) = 0.30$ (Roeder & Emslie, 1970). Recent OIB magmatism is comprehensively represented on a world-wide scale (Fig. 2). We adopted the original classification of mantle components based on DMM, EMI, EMII and HIMU, as defined by Hart *et al.* (1992), by projecting their Sr, Nd and Pb ($^{206}\text{Pb}/^{204}\text{Pb}$) isotopic compositions ($n = 3$) onto the four vertices of a regular tetrahedron (Fig. 2). Once MORB and OIB data, the latter classified on the basis of their mantle component affinity

(e.g. Pitcairn-EMI, Society-EMII, etc.), are plotted, more than 90% of the data fall within the tetrahedron, suggesting that the range of compositions could result from the physical mixture of the selected end-members (See Appendix B). OIB mixing trends converge towards a focal zone (e.g. the FOZO, Focal ZONE; Hart *et al.*, 1992) on the DMM–HIMU edge (Fig. 2). Because the tetrahedron can rotate around two of its axes, trends relative to each ocean or archipelago are easily identifiable.

For a variety of basalts from MOR (Atlantic, Pacific, Indian) and EMI (Pitcairn), EMII (Society and Samoa) and HIMU (Tubuai, Rurutu and St. Helena) oceanic islands (<http://georoc.mpch-mainz.gwdg.de/Start.asp>), we calculated the average arithmetic values for the $^{208}\text{Pb}/^{204}\text{Pb}$ ratio and several incompatible trace element abundances, characterized by different ratios of charge/ionic radii, and geochemical behaviour in igneous processes. We selected suitable trace element ratios (e.g. Ce/Pb, Nb/U, La/Sm, Rb/Ba, etc.) to discriminate OIB on the basis of source- and/or process-related geochemical characteristics (Table 2). We constrained the geochemical compositions of the four mantle components of Hart *et al.* (1992) to n compositional parameters ($3 < n \leq 10$), thus using up to six extra trace element and/or isotope ratios. We note that even the addition of only one variable ($n=4$) to the three considered by Hart *et al.* (1992), either elemental or isotopic, significantly affects the distribution of data points. The combination of more than three geochemical parameters causes clustering of points towards the DMM–HIMU–EMI face of the tetrahedron, on a roughly even surface (here hence ‘pseudo-plane’), as highlighted by an opportune rotation of the tetrahedron (e.g. Fig. 3 and supporting file *anamorfosis.xls*). Because most of points fall outside the tetrahedron, such a distribution seems not to be generated by a physical mixture of the selected end-members (Appendix B, Fig. B1) and clearly recalls the OIB ‘plane’ described by Zindler *et al.* (1982).

For $4 \leq n \leq 10$, samples from each oceanic island (e.g. Canary Islands, St. Helena, Hawaii, etc.) describe sub-parallel alignments in the pseudo-plane, showing geochemical variability that may cover the entire compositional range observed for the OIB dataset as a whole (e.g. Zindler *et al.*, 1984). On average, such geochemical heterogeneity varies between two extreme compositions that: (1) are not constrained to any geographical location, and (2) never match or even converge to a unique end-member, as each extreme is distinctive of magmas feeding each ocean island or archipelago [e.g. the Canary end-members are not the same as the Iceland end-members or the Hart *et al.* (1992) end-members; Figs 3 and 4]; this holds even if they share similar geochemical characteristics, marked by clustering in neighbouring positions in R^3 .

Table 2: Average values for $^{208}\text{Pb}/^{204}\text{Pb}$ and several incompatible trace element abundances calculated from samples in the GEOROC database (<http://georoc.mpch-mainz.gwdg.de/Start.asp>) of a variety of basalts

Ratio	MORB	EMI	EM2	HIMU
$^{87}\text{Sr}/^{86}\text{Sr}^*$	0.70200	0.70530	0.70780	0.70285
$^{143}\text{Nd}/^{144}\text{Nd}^*$	0.51330	0.51236	0.51258	0.51285
$^{206}\text{Pb}/^{204}\text{Pb}^*$	18.00	17.40	19.00	21.80
$^{207}\text{Pb}/^{204}\text{Pb}^*$	15.39	15.47	15.61	15.86
$^{208}\text{Pb}/^{204}\text{Pb}$	38.42	38.85	38.86	40.12
La/Yb	1.47	15.11	19.68	28.13
La/Ce	0.33	0.42	0.45	0.51
La/Sm	1.36	3.72	4.45	6.23
La/Nb	1.10	0.91	0.95	0.74
Ce/Pb	25	21.36	21.15	31.47
Nd/Sm	2.77	4.49	4.77	5.55
Tb/Yb	0.28	0.48	0.50	0.57
Rb/Ba	0.073	0.074	0.095	0.077
Rb/Nb	0.67	0.71	1.06	0.36
Ba/La	12.13	9.63	11.79	4.78
Ba/Nb	6.78	8.72	11.19	4.87
Sr/Zr	1.44	1.70	2.05	2.90
Th/Yb	0.15	1.58	3.11	3.70
Th/Nb	0.27	0.15	0.09	0.09
Th/U	2.90	3.53	4.94	3.84
Nb/U	44.4	39.6	33.9	46.5
Zr/Hf	40.19	45.45	42.28	43.10

The basalts are representative MORB (Atlantic, Pacific, Indian) and EMI (Pitcairn), EMII (Society, Samoa) and HIMU (Tubuai, Rurutu, St. Helena) OIB, along with $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$, $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ data from Hart *et al.* (1992). Up to 10 of these values have been used to constrain the four end-members on the vertices of the regular tetrahedron.

*Data from Hart *et al.* (1992).

Each ocean island may show multiple extreme compositions, especially those characterized by complex petrogenetic history, e.g. note the various alignments described by Hawaii magmas (Gaffney *et al.*, 2005, and references therein; inset in Fig. 4a).

With the new choice of compositional parameters, samples with geochemical characteristics close to the Hart *et al.* (1992) mantle components (e.g. St. Helena = HIMU *sensu stricto*, Society = EMII *sensu stricto*, etc.) do not cluster close to the vertices of the tetrahedron but form sub-parallel elongated trends in the pseudo-plane, independent of the position of the Hart *et al.* (1992) end-members (Fig. 3). Significantly, MORB also spread out from the DMM vertex, revealing the intrinsic variability of their mantle

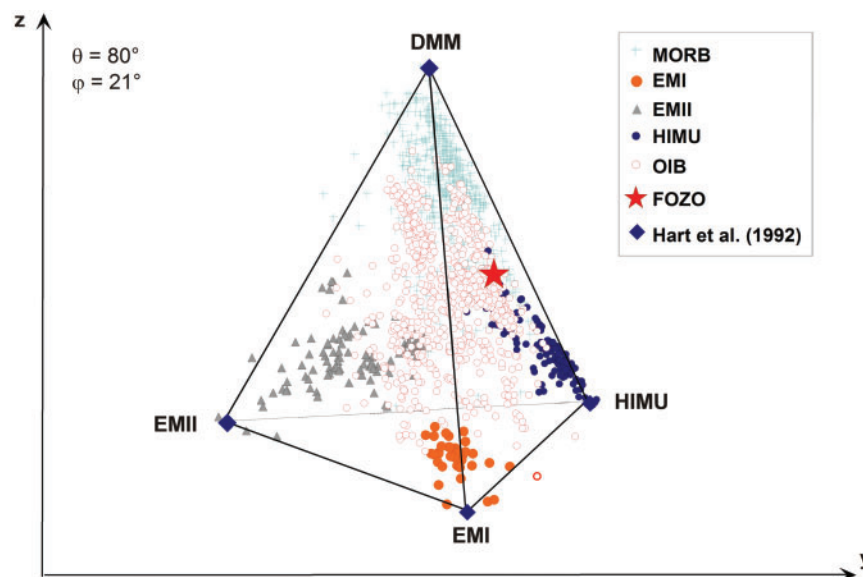


Fig. 2. A 3D plot of Sr, Nd and Pb ($^{206}\text{Pb}/^{204}\text{Pb}$) isotopic compositions ($n=3$) for worldwide OIB and MORB (<http://georoc.mpch-mainz.gwdg.de/Start.asp>), adopting the classification proposed by Hart *et al.* (1992) for the four end-members at the vertices of the regular tetrahedron (DMM, EMI, EMII, HIMU; data as in Table 2). From a mathematical point of view, OIB data within the tetrahedron, being a convex sum of the four end-members, could represent a physical mixture of the adopted end-members. θ and ϕ are the rotation angles with respect to the 'canonical orientation' of the tetrahedron ($\theta=0^\circ$; $\phi=0^\circ$), which provides the starting position.

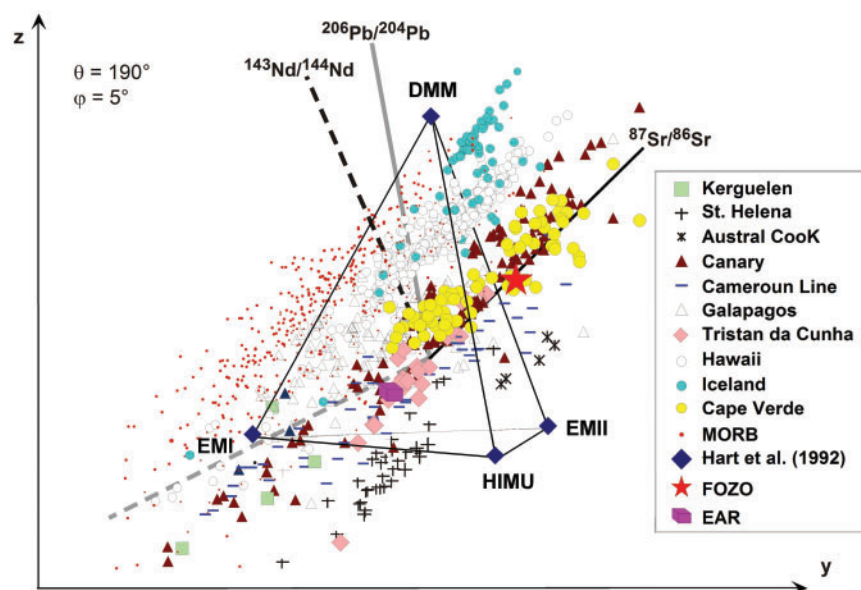


Fig. 3. Projection on the y - z plane of the 3D plot of Sr, Nd and Pb ($^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$) isotopic compositions ($n=4$) for representative OIB and MORB (see the text for details on the dataset). The addition of one compositional parameter (in this case the $^{207}\text{Pb}/^{204}\text{Pb}$ ratio) to the three considered by Hart *et al.* (1992) causes the loss of the data convexity. Points describe sub-parallel alignments between extreme compositions, independent of the four end-members adopted according to Hart *et al.* (1992). The OIB trends mostly lie on a planar surface, suggesting that OIB compositional variability is basically due to two degrees of freedom (see also Zindler *et al.*, 1982). Vectors (vectors of unitary module, used to indicate one particular direction) of $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$, $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ components are reported (see Appendix B and Fig. B1 caption for further details). FOZO and EAR are from Stracke *et al.* (2005) and Lustrino & Wilson (2006), respectively. An animated version of this figure is provided in the supplementary Excel file (anamorfoxis.xls). θ and ϕ are the rotation angles with respect to the 'canonical orientation' of the tetrahedron ($\theta=0^\circ$; $\phi=0^\circ$), which provides the starting position.

source (Fig. 3; e.g. Hofmann, 2003; Salters & Stracke, 2004; Workman & Hart, 2005). The so-called FOZO (Hart *et al.*, 1992) and EAR (Enriched Asthenospheric Reservoir; Hart *et al.*, 1992; Granet *et al.*, 1995; Stracke *et al.*, 2005; Lustrino & Wilson, 2006) do not hold any key position in this new projection (Fig. 3). Consequently, samples with the isotopic compositions of the Hart *et al.* (1992) components cannot properly describe the compositional variability of OIB and MORB, unless this is restricted to the isotopic ratios of Sr, Nd and Pb ($^{206}\text{Pb}/^{204}\text{Pb}$). This picture is not significantly affected by redefinition of the compositions of Hart *et al.* (1992) end-members; for example, adopting isotope ratios as suggested in the recent literature for DMM, EMI, EMII and HIMU (e.g. Workman *et al.*, 2004; Stracke *et al.*, 2005).

As the tetrahedron is simply a tool with which to 'visualize' the R^3 space for the projection of points from R^n , it may be dropped, with the focus being moved to the point distributions and clusterings. Thus it is possible to explore a more appropriate choice of end-members to obtain projections that better highlight the trends of interest; for example, allowing the dataset to reveal its planar distribution for $n \geq 4$ (Table 3; Figs 4 and 5), regardless of arrays' positions with respect to the tetrahedron. From this perspective, if samples are tagged on the basis of their major element composition (e.g. classified on the TAS diagram; Figs 1 and 4a and b), picrites, along with OIB tholeiites (most of the tholeiite samples are from the Hawaii database) and some alkali basalts, independent of their provenance, mainly cluster to form a well-defined alignment (see inset in Fig. 4a for Hawaii). The alkali basalts, which form part of this trend, have major element characteristics that are similar to those of picrites and fall on the picrite array ($P < 2$ GPa) in the CaTs–Ol–Q diagram shown in Fig. 1. Nephelinites, basanites and the other alkali basalts spread on OIB arrays that diverge from the picrite–tholeiite trend, pointing to different end-members (Figs 4 and 5). The two different alignments are not an artefact of the choice of adopting both process-related and source-related compositional parameters. Actually, the trends described are evident also if only tracers of source composition are selected as variables (e.g. isotopes, Th/U, Nb/U, etc.; Fig. 4b). This would suggest that the range of tholeiite–picrite vs basanite–nephelinite trends does not depend upon different degrees of partial melting.

Data for modern oceanic sediments (from Plank & Langmuir, 1998) plot mainly apart from the OIB trends, scattering far from the pseudo-plane together with some alkali basalt ($\pm$ picrite) samples [Fig. 4a; see also sediment distribution with respect to the 'plane' of Zindler *et al.* (1982)]. Although incorporation of modern sediments from the sea floor appears to modify the geochemistry of a minority of OIB mafic magmas, most of the OIB magmas thought to be characterized by recycling of sedimentary material in their mantle source (e.g. EMII) still plot on the pseudo-plane.

Also in the projection of Fig. 5, strong correlations between Sr and Nd isotopes and/or $^{207}\text{Pb}/^{204}\text{Pb}$ – $^{208}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$, such as the mantle–crust array or the Northern Hemisphere Reference Line (NHRL; Hart, 1984), do not affect the distribution of points in Figs 4 and 5 and do not define linear trends on the pseudo-plane.

Mixing equations involving trace element ratios in R^n (Langmuir *et al.*, 1978) can be used to calculate mixing curves in n -dimensional space, and the resulting curves can also be projected in R^3 . Physical mixing between the pairs of distinct end-members of each oceanic island or archipelago can account for its compositional variability (Fig. 5). Consequently, in this diagram OIB lie on a family of sub-parallel curves.

If incompatible trace element ratios alone ($n = 5, \dots, 10$) are adopted, or Pb isotope ratios are not included among the compositional parameters (e.g. selecting Rb/Ba, Th/Yb and La/Nb and/or $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios as in Fig. 6), the pseudo-plane defined by most of the OIB data collapses into a single alignment. The main trend lies on a mixing curve on which the various extreme compositions tend to merge into apparently only two end-members. Consequently, OIB compositional variability seems to be strongly controlled by two main end-members, in turn dependent on Pb isotope ratios. Even if data overlap is evident along this alignment, nephelinites–basanites–most alkali basalts tend to cluster near one of the two extreme compositions, whereas tholeiites–picrites–few alkali basalts are more representative of the composition of the other end-member. The basalts and sediments that in Fig. 4a plot away from the pseudo-plane spread perpendicularly to the trend of the main data array.

We emphasize that these conclusions can be verified for any sets of compositional parameters not including Pb isotopes (e.g. Ce/Pb, La/Sm, Nb/U and $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$). It should be noted that this 'Pb-effect' appears to be little influenced by the specific choice of the Pb isotope parameter plotted, as such spreading on the pseudo-plane is induced by any kind of Pb isotope ratio included among the compositional parameters. Otherwise, because of the scarcity of Hf isotope data in OIB databases, the effect of these isotopic ratios on the arrangement of data points in the tetrahedron cannot be strictly constrained. The limited Hf isotope data available confirm the similar behaviour of the Nd and Hf isotope systems, making it possible to switch from one compositional parameter to the other, without affecting the multi-dimensional distribution of data.

We made several attempts to verify this observation by replacing the arithmetic average values with extreme (minimum and maximum) values of $^{208}\text{Pb}/^{204}\text{Pb}$ and trace element abundances for the Hart *et al.* (1992) end-members, without noticing any significant deviation from the above conclusion. We further considered the possibility

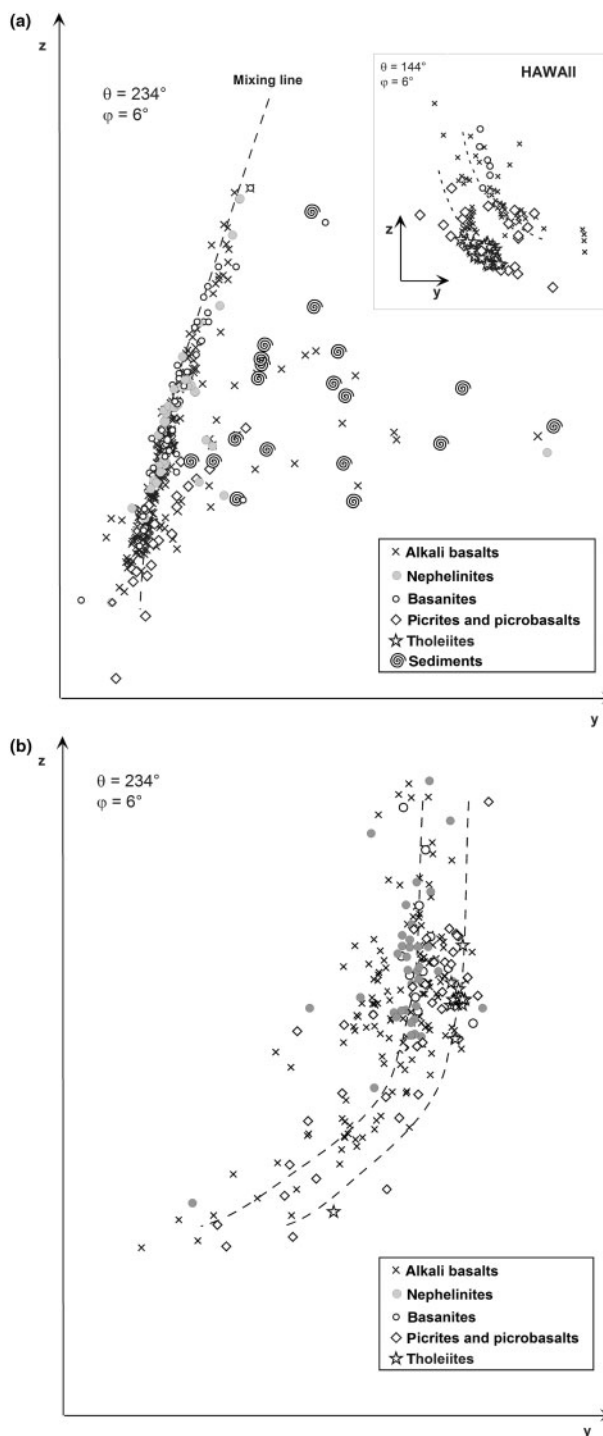


Fig. 4. (a) Projection on the y - z plane of the 3D plot of Sr, Nd and Pb ($^{206}\text{Pb}/^{204}\text{Pb}$) isotopic compositions and Rb/Ba, Th/Yb and La/Nb ratios ($n=6$) for representative OIB (GEOROC database, <http://georoc.mpch-mainz.gwdg.de/Start.asp>). The compositions of the four end-members (A–D; Table 3) are here chosen to highlight the planar surface on which the OIB arrays lie; the tetrahedron is not shown, to focus attention on the data arrangement. In this projection, it is possible to highlight spreading of some OIB, along with sediments, with respect to the pseudo-plane (here in section). In the inset,

that the use of compositional parameters involving U/Pb and/or Th/Pb ratios might account for spurious correlations, because of the presence of reliant variables, or distort intra-oceanic correlations because of the scarce correspondence between Pb and Sr–Nd isotope ratios. Nevertheless, even if we replace $^{206}\text{Pb}/^{204}\text{Pb}$ with $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ (the ratio of the radiogenic additions to the initial terrestrial lead, defined as $[(^{208}\text{Pb}/^{204}\text{Pb}) - (^{208}\text{Pb}/^{204}\text{Pb})_{\text{init}}]/[(^{206}\text{Pb}/^{204}\text{Pb}) - (^{206}\text{Pb}/^{204}\text{Pb})_{\text{init}}]$; Tatsumoto *et al.*, 1973; Hofmann, 1997), which linearly correlates with Sr and Nd isotope composition, or $\Delta 7/4$ Pb with $^{207}\text{Pb}/^{204}\text{Pb}$ (Hart, 1984), the distribution of data in multi-dimensional space ($n \geq 4$) still matches the above-mentioned correlations. We also verified that such relationships are still evident if only the most primitive OIB samples are selected (mg-number >70).

DISCUSSION

The proposed method of plotting multi-parameter datasets from n -dimensional space to $\mathbb{R}^3$ allows a comprehensive analysis of the effects of a large number of independent compositional parameters at the same time, once a proper set of end-members at the vertices of the tetrahedron has been defined. This represents a substantial improvement with respect to the adoption of a fixed reference system tied to a maximum of three independent variables (e.g. Hart *et al.*, 1992) that, as a matter of fact, is a projection in a 3D space that excludes the effect of (geochemical) data that were not taken in consideration. Such a drastic reduction of components to allow 3D or 2D projections may result in fictitious alignments or clusters that disappear when the complexity of the system is taken into account. In particular, for projections in $\mathbb{R}^3$ based on four end-members, data fall inside the tetrahedron defined by the end-members only when the compositional parameters adopted to describe MORB and OIB variability are the isotopes of Sr, Nd and Pb ($^{206}\text{Pb}/^{204}\text{Pb}$) (e.g. Hart *et al.*, 1992). However, when the compositional parameters are increased to four and include $^{207}\text{Pb}/^{204}\text{Pb}$ and/or $^{208}\text{Pb}/^{204}\text{Pb}$ ratios, the distribution within the volume

rotated 90° around the z axis with respect to the main figure, the distribution on the pseudo-plane of the Hawaii samples shows clustering of picrites, OIB tholeiites and a few alkali basalts. Conversely, nephelinites, basanites and most alkali basalts spread on OIB arrays that drift away from the picrite–tholeiite trend. (b) Projection on the y - z plane of the 3D plot of Sr, Nd and Pb ($^{206}\text{Pb}/^{204}\text{Pb}$) isotopic compositions and Th/Nb and Nb/U ratios ($n=5$) for representative OIB (GEOROC database, <http://georoc.mpch-mainz.gwdg.de/Start.asp>), classified according to the TAS. The compositions of the four end-members are from Table 2 [isotope data are from Hart *et al.* (1992)]. Tetrahedron is not shown, to focus attention on the data arrangement. By changing the compositional parameters (e.g. by selecting only source-tracers, such as isotopes and Nb/U and Th/Nb ratios), the split of the two trends is still evident. θ and φ are the rotation angles with respect to the ‘canonical orientation’ of the tetrahedron ($\theta=0^\circ$; $\varphi=0^\circ$), which provides the starting position.

Table 3: Sr, Nd and Pb ($^{206}\text{Pb}/^{204}\text{Pb}$) isotope data and Rb/Ba, Th/Yb, La/Nb, Ce/Pb, La/Sm, and Nb/U ratios of the four end-members

Ratio	A	B	C	D
$^{87}\text{Sr}/^{86}\text{Sr}$	0.70291	0.7053	0.70628	0.70285
$^{143}\text{Nd}/^{144}\text{Nd}$	0.51287	0.51236	0.51247	0.51285
$^{206}\text{Pb}/^{204}\text{Pb}$	20.891	17.4	19.171	21.8
Rb/Ba	0.55	0.07	0.02	0.08
Th/Yb	7.69	1.58	1.45	3.7
La/Nb	0.8	0.91	5.08	0.74
Ce/Pb	5.69	17.8	64.4	20.5
La/Sm	2.29	1.07	5.30	4.21
Nb/U	7.61	90.9	677.8	43.7

The end-members are chosen to enhance the even surface on which the OIB arrays lie. This choice is solely made on the geometry of the distribution, and thus is independent of the origin or petrology of the samples at the four vertices of the tetrahedron, which are: A, Rurutu (Austral-Cook Islands, Young Atiu-trend lavas: unique-ID 18583); B, EMI (Society, Samoa) of Table 2; C, Comoros (Grande Comore-Njazidja, Mt. Karthala: unique-ID 11913-D85-31); D: HIMU (Tubuai, Rurutu, St. Helena) of Table 2.

collapses into an approximately planar surface. This plane extends outside the tetrahedron (Figs 3–7). When the number of compositional parameters plotted is progressively increased to 10, this pseudo-plane becomes better defined, with the exception of the presence of a few OIB that seem to be significantly affected by incorporation of modern sediments from the sea floor (Fig. 4a). Conversely, exclusion of any compositional parameter related to Pb isotopes leads to the collapse of the pseudo-plane into a linear array, provided that $n > 4$ (Figs 6 and 7b).

Even if we have explored a large number of compositional parameters, in principle we cannot exclude that we may have missed some key geochemical tracers. Mathematically speaking, if any significant chemical species is responsible for the point spreading, its neglect is equivalent to perform a projection of the dataset onto an $R^{(n-1)}$ space that is perpendicular to the spread in R^n . This implies loss of the dataset variability in the chosen projection (i.e. it occurs when Pb isotopes are excluded).

The planar distribution of data in R^3 suggests that OIB compositional variability is characterized by two degrees of freedom. As three points are enough to define a planar distribution in R^3 , we might be tempted to relate the observed variability to three distinct end-members. Three mantle components have been previously invoked to explain the isotopic variability of OIB, distributed on a ‘plane’ in any 3–5-dimensional isotope space (Zindler *et al.*, 1982). However, the three end-members are not

geographically located, as extreme compositions on the pseudo-plane are not associated with any specific archipelago or, more generally, with the ocean of origin; they cannot be defined as ‘mantle components’, as the components necessary to define each magmatic suite are distinct and different, and depend on the choice of the compositional parameters (e.g. including or excluding Pb isotopes). The Hart *et al.* (1992) end-members do not represent extreme geochemical compositions within the entire MORB–OIB dataset and they are not appropriate to explain the observed variability of OIB sources by physical mixing, even if they can apparently satisfactorily describe their Sr–Nd–Pb isotopic characteristics.

Different pairs of compositional extremes could account for most of the compositional variability recorded in each archipelago (Fig. 7a). If Pb isotopes are not included in the set of compositional parameters, the distribution on the pseudo-plane collapses into a single array, thus the OIB geochemical variability can be related to only two distinct end-members. Three general considerations can help constrain the nature of the extreme components in MORB–OIB, by linking petrological and geochemical evidence:

- (1) picrites–tholeiites and some alkali basalts mostly cluster towards one of the two end-members and are probably characterized by shallow depths of origin and/or large degrees of partial melting of peridotitic, eventually pyroxenitic, sources (Figs 1, 4 and 5);
- (2) the sources of basanites–nephelinites and most alkali basalts show affinities with the other extreme composition and seem to be mostly related to deep mantle sources (Figs 1, 4 and 5);
- (3) The effect of including Pb isotope ratios seems to enhance the heterogeneity of OIB sources (Fig. 7a).

End-members in OIB and MORB mantle sources do not show a ‘fixed’ geochemical composition and are unlikely to reflect physically accessible mantle reservoirs. The complex variety of compositions along the array(s) may result from the various processes that cause mantle heterogeneity, such as depletion by melt extraction or enrichment by metasomatism, or recycling of subducted or delaminated lithospheric segments. If Pb isotopes are not among the compositional parameters, any set of end-members seems to generate a single linear distribution of OIB–MORB data (Fig. 7b). Magmas generated under largely different partial melting degrees mostly lie at the opposite extremes of this alignment, although both nephelinite–basanite–alkali basalt and tholeiite–picrite magmas overlap in composition. Therefore, diverse mantle processes (e.g. enrichment by recycling and depletion by melt extraction) appear to pervasively affect mantle sources independently of the depth at which they are located.

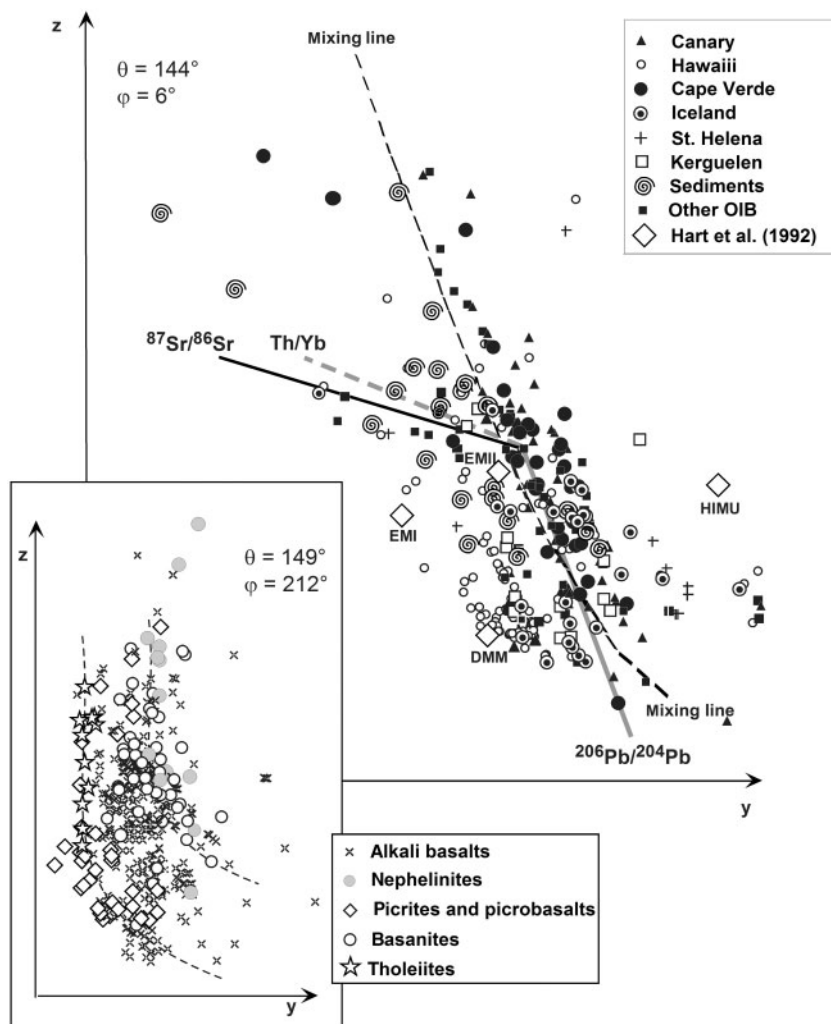


Fig. 5. Projection on the y - z plane of the 3D plot of Sr, Nd and Pb ($^{206}\text{Pb}/^{204}\text{Pb}$) isotopic compositions and Rb/Ba, Th/Yb and La/Nb (Ce/Pb, La/Sm, Nb/U in the inset) ratios ($n = 6$) for representative OIB (dataset as in Fig. 1). The compositions of the four end-members (A–D; Table 3) are chosen to highlight the planar surface on which the OIB arrays lie; the tetrahedron is not shown, to focus attention on the data arrangement. In this projection, the distribution of samples on the pseudo-plane with respect to their geographical location, or TAS-based classification (see inset), is highlighted. Magmatic suites of a given area describe sub-parallel alignments fitting mixing curves between diverse couples of extreme compositions, each distinctive for each volcanic district. The equation of the mixing line drawn is $z = 0.0535y^2 + 0.5121y - 128.55$. Versors of $^{87}\text{Sr}/^{86}\text{Sr}$, $^{206}\text{Pb}/^{204}\text{Pb}$ and Th/Yb components are reported (see Appendix B and Fig. B1 caption for further details). [For considerations on petrology trends, see comments in the inset of Fig. 4a (Hawaii arrays). See also Fig. 7 for further discussion.] θ and φ are the rotation angles with respect to the ‘canonical orientation’ of the tetrahedron ($\theta = 0^\circ$; $\varphi = 0^\circ$), which provides the starting position.

If OIB–MORB mantle sources were generated only from sources plotting at the two extremes, a marked bimodal arrangement should characterize the observed distributions. Similarly, if the source had an intermediate composition, a cluster of data points would be evident. In this case, each ocean island might be expected to record the fingerprint of a mantle component (or mix of mantle components), thus inheriting a well-defined and restricted geochemical composition. Moreover, this scenario would suggest the existence of ‘enriched’ *sensu stricto* (e.g. basanites–nephelinites–alkali basalts) or ‘depleted’ *sensu stricto*

(e.g. tholeiites–picrites) magmas, thus calling for the presence of distinct mantle reservoirs. Indeed, the continuous distribution of data along the OIB mixing trends testifies to the existence of a wide spectrum of melts in each ocean island. Both the collection of melts from a definite mantle source region and mixing of magmas from distinct mantle sources are able to explain the observed arrays of OIB data. Thus each ocean island reflects the scale of local mantle heterogeneity by showing its typical range of geochemical variability (Fig. 7b). Therefore, the concept of mantle component is strictly related to the mantle

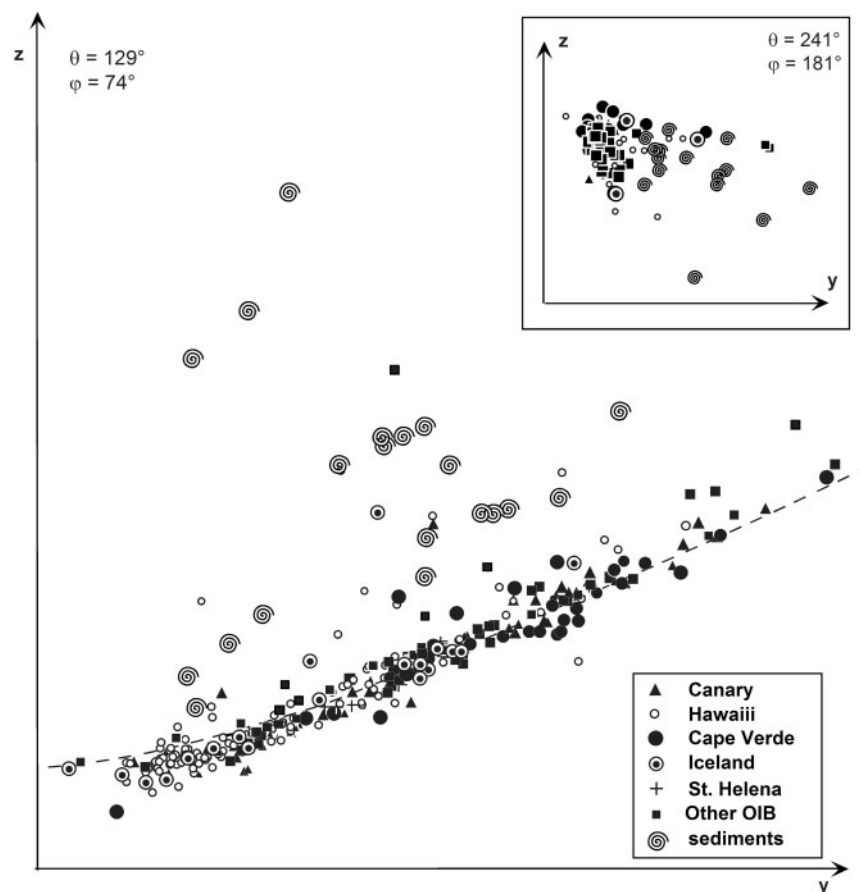


Fig. 6. Projection on the y - z plane of the 3D plot of Sr and Nd isotopic compositions and Rb/Ba, Th/Yb and La/Nb ratios ($n=5$) for representative OIB and MORB as in Fig. 4. The compositions of the four end-members (A–D; Table 3) are the same as in Fig. 4a and b. The pseudo-plane, defined by most of the OIB data, collapses into an alignment if Pb isotope ratios are not included in the selected variables, suggesting that OIB compositional variability is significantly dependent on Pb isotope systematics. The data are dispersed along the mixing line in R^3 , as can be clearly seen by opportune rotation around the z and y axis. The inset shows a particular rotation of the tetrahedron to set the alignment at right angles to the y - z plane. It should be noted that several hundred data points cluster in a relatively small area. θ and ϕ are the rotation angles with respect to the ‘canonical orientation’ of the tetrahedron ($\theta=0^\circ$; $\phi=0^\circ$), which provides the starting position.

portion(s) sampled at any specific ocean island (Figs 4 and 5). Extreme geochemical compositions and the whole range of intermediate compositions may coexist independently of the degree of partial melting (e.g. as commonly observed at Hawaii), supporting the hypothesis of a marble-cake mantle. However, we consider that the model of the statistical distribution of mantle heterogeneity (Meibom & Anderson, 2003; Anderson, 2006) should produce geochemical homogenization within the limits of local mantle variability, so that a uniform depleted reservoir cannot be sampled by simply extending the volume of the region undergoing partial melting (Fig. 7b).

The inclusion of Pb isotopic ratios among the compositional parameters plotted, and the consequent spreading of trends in the pseudo-plane, reveals that each oceanic island does not simply differ in terms of the compositional range between each extreme composition (Fig. 7b).

Because Nd and Sr isotope systematics, or elemental Pb in trace element ratios, do not induce differences in the data point distribution, we argue that such behaviour may be strictly related to the variation of U/Pb, Th/Pb and Th/U, and associated Pb isotope ratios, with respect to Sm/Nd or Rb/Sr in OIB mantle sources. This is confirmed by the use of $^{208}\text{Pb}^*/^{206}\text{Pb}^*$ and $\Delta 7/4$ as compositional parameters that do not cause the data to collapse into a single array, suggesting that the spreading of the OIB trends on the pseudo-plane is not a geometric effect of the scarce correlation between Sr–Nd and Pb isotopes.

The discriminant power of Pb isotopes clearly seems to indicate a key role for geological time in characterizing OIB–MORB geochemistry. Recycled oceanic crust, with its sediment veneer, is thought to be a common component in the mantle source of OIB, largely responsible for their chemical heterogeneity (Hofmann & White, 1982;

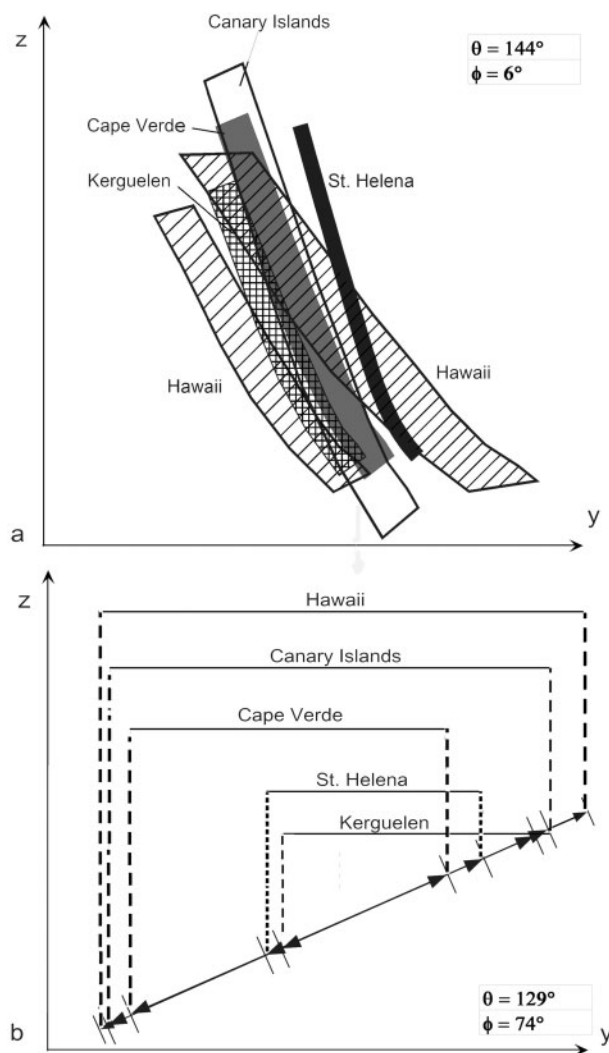


Fig. 7. Schematic view of Figs 5 and 6 in which the single array for each oceanic island is highlighted when Pb isotopes are included among the compositional parameters (a) or excluded (b). It should be noted that each volcanic suite shows characteristic geochemical compositions that vary between extremes distinctive to that suite.

Chauvel *et al.*, 1992; Lassiter & Hauri, 1998; Blichert-Toft *et al.*, 1999; Chauvel & Hemond, 2000; Eisele *et al.*, 2002; Workman *et al.*, 2004; Stracke *et al.*, 2005). U/Pb variability and variability in the age of different portions of subducted lithospheric slabs adds a distinctive Pb isotope fingerprint to the recycling process. Recycling of geochemically diverse subducted plates and delaminated lithospheric keels (e.g. Lustrino, 2005), each with unique U/Pb and Th/Pb values and residence time in the mantle, confers distinctive flavours to OIB mantle sources (e.g. Kelley *et al.*, 2005; Stracke *et al.*, 2005). These are reflected by characteristic Pb isotope ratios in OIB from different oceanic islands, and are thus probably responsible for the observed range of sub-parallel trends in the pseudo-plane (Fig. 7). Thus, each ocean island appears to be characterized by its own range

of compositional variability between its own geochemical end-members, with respect to recycling of peculiar lithospheric segments occurring in its local mantle source region. Sr and Nd isotopes in OIB do not seem to be such effective tracers of recycling processes, as differences in these isotope compositions in diverse subducted plates are not an effective discriminant (i.e. the fields of terrigenous and pelagic sediments largely overlap in the Sr–Nd isotope space). This might be related to the fact that Rb/Sr and Sm/Nd do not fractionate as strongly as U/Pb and Th/Pb during the recycling process, thus leading to the more homogeneous Sr–Nd isotope compositions of the enrichment agents. On the other hand, Pb isotope variability in OIB has traditionally been the basis on which the enriched mantle components or end-members (HIMU, EMI, EMII) required to describe mantle heterogeneity were defined (e.g. Weaver, 1991; Hofmann, 1997; Lustrino & Dallai, 2003; Stracke *et al.*, 2005), reflecting the age and composition of recycled lithospheric materials in their mantle source.

CONCLUSIONS

The chemical structure of the mantle can be constrained by the contemporaneous evaluation of the trace element and isotopic compositions of MORB and OIB. The choice of a particular set of geochemical parameters represents a filter through which mantle composition can be visualized, allowing a more independent perspective of the causes and nature of mantle heterogeneity. Multi-dimensional projections allow a broad view of large-scale mantle heterogeneities, accounting for several possible perspectives at once. The apparently complex projection scheme allows a change of coordinates to simplify the description of mantle compositions. This comprehensive evaluation of several compositional parameters reveals that each ocean island has its own local geochemical end-members, thus the so-called mantle components (DMM, EMI, EMII, HIMU; Hart *et al.*, 1992) are significant only in the Sr, Nd and Pb isotope perspective. The proposed method allows reconstruction of a relatively simple scenario in which mantle heterogeneity results from a range of processes that cause incompatible element enrichment and depletion, where time plays a major role. Different degrees of partial melting of peridotite and recycled eclogite could contribute to OIB petrogenesis. The geochemical signature of the enriching component, often deriving from recycled lithosphere with a distinctive age and Pb isotope signature, is crucial to characterize specific OIB mantle sources. The multi-dimensional evaluation of OIB–MORB mantle sources allows us to move away from a perspective based on physically distinct mantle reservoirs to a model based on a marble-cake structure for the mantle. Although this model is consistent with the SUMA model of Meibom & Anderson (2003), the highest degrees of partial melting of

the mantle source of each ocean island can reveal only the distinctive local depleted source composition, precluding the existence of a mantle-wide, common and uniform depleted DMM reservoir.

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SUPPLEMENTARY DATA

Supplementary data for this paper are available at *Journal of Petrology* online.

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APPENDIX A: THE PROJECTION SCHEME

The chemical composition of a system can be reported in terms of a set of n variables corresponding to a point in n -dimensional space. For a system in which four end-members, defined by a given set of components, represent a suitable basis (a basis for a vectorial space is a set of vectors that can generate the whole compositional space R^n to be projected into R^3) the projection scheme is based on well-known procedures reported in detail in basic textbooks of linear algebra. In the projection scheme an n -tuple of values that defines the composition of system $\mathbf{E} \approx (E_1, E_2, \dots, E_n)$ is recast choosing the four end-members:

$$\mathbf{A} \approx (A_1, A_2, \dots, A_n), \mathbf{B} \approx (B_1, B_2, \dots, B_n),$$

$$\mathbf{C} \approx (C_1, C_2, \dots, C_n), \mathbf{D} \approx (D_1, D_2, \dots, D_n).$$

The orthogonal projection of the point $\mathbf{E}^*$ in the 3D space through the end-members is obtained minimizing the n -dimensional Euclidean distance from the given point $\mathbf{E}$.

Given that $\mathbf{A}, \mathbf{B}, \mathbf{C}$ and $\mathbf{D} \in R^n$, we compute the vectors

$$\mathbf{v}_1 = \mathbf{A} - \mathbf{D}, \quad \mathbf{v}_2 = \mathbf{B} - \mathbf{D}, \quad \mathbf{v}_3 = \mathbf{C} - \mathbf{D}.$$

Set

$$\alpha = \frac{\mathbf{v}_2 \cdot \mathbf{v}_1}{\|\mathbf{v}_1\|^2} \text{ and } \beta = \frac{\mathbf{v}_3 \cdot \mathbf{v}_1}{\|\mathbf{v}_1\|^2}; \quad \mathbf{w}_1 = \mathbf{v}_1 \text{ and } \mathbf{w}_2 = \mathbf{v}_2 - \alpha \mathbf{w}_1$$

Lastly, set

$$\gamma = \frac{\mathbf{v}_3 \cdot \mathbf{v}_2}{\|\mathbf{v}_2\|^2} \text{ and } \mathbf{w}_3 = \mathbf{v}_3 - \beta \mathbf{w}_1 - \gamma \mathbf{w}_2.$$

For each point, we also compute the vector

$$\mathbf{E}' = \mathbf{E} - \mathbf{D}$$

and its orthogonal projection $\mathbf{E}''$ on $\langle \mathbf{w}_1, \mathbf{w}_2, \mathbf{w}_3 \rangle$, $\mathbf{E}'' = \lambda_1 \mathbf{w}_1 + \lambda_2 \mathbf{w}_2 + \lambda_3 \mathbf{w}_3$, where

$$\lambda_i = \frac{\mathbf{E}' \cdot \mathbf{w}_i}{\|\mathbf{w}_i\|^2}, \quad i = 1, 2, 3.$$

$\mathbf{E}'$ is the linear combination of $\mathbf{w}_1, \mathbf{w}_2, \mathbf{w}_3$ nearest $\mathbf{E}'$ in R^n . Please note the different apices appended to $\mathbf{E}$ and γ definition. As the distance is translation-invariant, it follows that $\mathbf{E}' = \mathbf{D} + \lambda_1 \mathbf{w}_1 + \lambda_2 \mathbf{w}_2 + \lambda_3 \mathbf{w}_3$ is the point in the 3D space passing through $\mathbf{A}, \mathbf{B}, \mathbf{C}, \mathbf{D}$ nearest $\mathbf{D} + \mathbf{E}' = \mathbf{E}$.

$\mathbf{E}'$ can be rearranged as

$$\begin{aligned} \mathbf{E}' &= \mathbf{D} + \lambda_1 \mathbf{w}_1 + \lambda_2 \mathbf{w}_2 + \lambda_3 \mathbf{w}_3 \\ &= \mathbf{D} + \lambda_1 (\mathbf{A} - \mathbf{D}) + \lambda_2 [\mathbf{B} - \mathbf{D} - \alpha (\mathbf{A} - \mathbf{D})] \\ &\quad + \lambda_3 [\mathbf{C} - \mathbf{D} - \beta (\mathbf{A} - \mathbf{D}) - \gamma [\mathbf{B} - \mathbf{D} - \alpha (\mathbf{A} - \mathbf{D})]] \\ &= (\lambda_1 - \lambda_2 \alpha - \lambda_3 \beta + \lambda_3 \alpha \gamma) \mathbf{A} + (\lambda_2 - \lambda_3 \gamma) \mathbf{B} + \lambda_3 \mathbf{C} \\ &\quad + (\mathbf{1} - \lambda_1 - \lambda_2 - \lambda_3 + \lambda_2 \alpha + \lambda_3 \beta + \lambda_3 \gamma - \lambda_3 \alpha \gamma) \mathbf{D} \end{aligned}$$

where the sum of the four coefficients of the end-members on the right-hand side of the above equation represents the tetrahedral coordinates τ_i of $\mathbf{E}^*$ with respect to the four vertices $\mathbf{A}, \mathbf{B}, \mathbf{C}, \mathbf{D}$:

$$\begin{aligned} \tau_1 &= \lambda_1 - \lambda_2 \alpha - \lambda_3 \beta + \lambda_3 \alpha \gamma \\ \tau_2 &= \lambda_2 - \lambda_3 \gamma \\ \tau_3 &= \lambda_3 \\ \tau_4 &= \mathbf{1} - \lambda_1 - \lambda_2 - \lambda_3 + \lambda_2 \alpha + \lambda_3 \beta \\ &\quad + \lambda_3 \gamma - \lambda_3 \alpha \gamma = \mathbf{1} - \tau_1 - \tau_2 - \tau_3. \end{aligned}$$

From $\sum \tau_i = 1$ it follows that each $\tau_i \leq 1$, thus the corresponding point lies within the array of the four end-members $\mathbf{A}, \mathbf{B}, \mathbf{C}, \mathbf{D}$ and represents a physical mixture of the end-members. If some coefficient $\tau_i > 1$ the constant sum imposes that one or more coefficient has to attain negative values; thus the point has to lie outside the envelope, a condition that suggests a different choice of end-members in order to express the data as physical mixtures of compositions plotted on the tetrahedron vertices.

The projection $\mathbf{E}'$ coincides with $\mathbf{E}$ if and only if $\mathbf{E}$ itself is a linear combination in R^n of the end-members with the sum of coefficients equal to one. Finally, we can map in R^3 the four end-members $\mathbf{A}, \mathbf{B}, \mathbf{C}, \mathbf{D}$, by choosing a set of four points $T_1, T_2, T_3, T_4 \in R^3$. The choice $T_1 = (0, 0, 1)$; $T_2 = (2 \times \sqrt{2}/3, 0, -1/3)$; $T_3 = (-\sqrt{2}/3, \sqrt{2}/3, -1/3)$; $T_4 = (-\sqrt{2}/3, -\sqrt{2}/3, -1/3)$ corresponds to a regular tetrahedron.

Finally, $\mathbf{E}$ can be mapped in R^3 onto $\mathbf{E}^{**}$ by the relation

$$\mathbf{E}^{**} = \tau_1 \mathbf{T}_1 + \tau_2 \mathbf{T}_2 + \tau_3 \mathbf{T}_3 + \tau_4 \mathbf{T}_4.$$

For a generic orientation of the tetrahedron, it is possible to express its vertices, $\mathbf{T}_i = \{x_i, y_i, z_i\}$, as a function of the angle φ of rotation around the z axis as

$$\begin{bmatrix} x'_i \\ y'_i \\ z'_i \end{bmatrix} = \begin{bmatrix} \cos \varphi & \sin \varphi & 0 \\ -\sin \varphi & \cos \varphi & 0 \\ 0 & 0 & 1 \end{bmatrix} \times \begin{bmatrix} x_i \\ y_i \\ z_i \end{bmatrix}.$$

The new coordinates are provided by multiplying the vectors representing the vertices by the rotation matrix. Analogously, the tetrahedron can rotate around the y axis by an angle θ . The rotation coordinates of its vertices are provided by

$$\begin{bmatrix} x''_i \\ y''_i \\ z''_i \end{bmatrix} = \begin{bmatrix} \cos \theta & 0 & -\sin \theta \\ 0 & 1 & 0 \\ \sin \theta & 0 & \cos \theta \end{bmatrix} \times \begin{bmatrix} x'_i \\ y'_i \\ z'_i \end{bmatrix}.$$

Given the tetrahedral coordinates $\mathbf{A}_T = \{\tau_1, \tau_2, \tau_3, \tau_4\}$ of a generic point $\mathbf{T}$, from the relation

$$\mathbf{E}^{**} = \tau_1 \mathbf{T}_1 + \tau_2 \mathbf{T}_2 + \tau_3 \mathbf{T}_3 + \tau_4 \mathbf{T}_4$$

it follows that the 3D coordinates of $\mathbf{T}$ with respect to the tetrahedron are

$$\begin{bmatrix} T_x \\ T_y \\ T_z \end{bmatrix} = \begin{bmatrix} \tau_1 & \tau_2 & \tau_3 & \tau_4 \\ \tau_1 & \tau_2 & \tau_3 & \tau_4 \\ \tau_1 & \tau_2 & \tau_3 & \tau_4 \end{bmatrix} \times \begin{bmatrix} x''_1 & x''_2 & x''_3 & x''_4 \\ y''_1 & y''_2 & y''_3 & y''_4 \\ z''_1 & z''_2 & z''_3 & z''_4 \end{bmatrix}$$

from which

$$T_y = \tau_1 y''_1 + \tau_2 y''_2 + \tau_3 y''_3 + \tau_4 y''_4$$

$$D_y = \tau_1 y_1 (-\sin \varphi + \cos \varphi) + \tau_2 y_2 (-\sin \varphi + \cos \varphi) + \tau_3 y_3 (-\sin \varphi + \cos \varphi) + \tau_4 y_4 (-\sin \varphi + \cos \varphi).$$

Then

$$T_y = (-\sin \varphi + \cos \varphi) \cdot (\tau_1 y_1 + \tau_2 y_2 + \tau_3 y_3 + \tau_4 y_4).$$

Analogously,

$$T_z = (\sin \theta + \cos \theta)(\tau_1 z_1 + \tau_2 z_2 + \tau_3 z_3 + \tau_4 z_4).$$

Coordinates T_y and T_z allow the orthogonal projection of point T onto a y - z plane from $\mathbb{R}^3$.

APPENDIX B: SOME NUMERICAL TESTS

Adopting four arbitrary end-members EM1, EM2, EM3, EM4 (their compositions with respect to the 10 compositional parameters are given in Table B1), we generated a reference composition RC to plot in the centre of the tetrahedron. RC is thus the average composition for each compositional parameter C1–C10 of the four end-members. RC is then manipulated to obtain four distinct datasets.

Dataset A is generated by adding random amounts of the end-members to RC, the resulting data point distribution is completely contained within the tetrahedron and does not show any symmetry (Fig. B1). Conversely, datasets B, C and D are generated by adding variable proportions of each compositional parameter C1–C10 to RC, through a random number generator. In set B, the same range of variability affects each compositional parameter and the resulting distribution is symmetric and perpendicular to C1 (the one with the maximum absolute values); moreover, points can plot outside the tetrahedron (Fig. B1). In sets C and D, different ranges of variability are allowed for each compositional parameter, the standard deviation of each one in set D is reduced by a factor of two with respect to set C. Specifically we took care to allow the largest standard deviation for C10 (the one with the minimum absolute value; Table B1).

Figure B1 shows the different symmetries and positions held by the four datasets. The resulting data arrays show a different spread; the wider distribution is attained by dataset C, which is the one characterized by the largest

Table B1: Compositions of the four end-members EM1, EM2, EM3 and EM4 with respect to the 10 compositional parameters C1–C10

Compositional parameter	End-members				Standard deviations			
	EM1	EM2	EM3	EM4	σ_{EM1}	σ_{EM2}	σ_{EM3}	σ_{EM4}
C1	712.47	442.91	14.85	149.58	25.48	18.56	46.92	22.27
C2	3.35	22.64	1.65	0.32	42.27	19.11	89.77	48.41
C3	17.46	0.17	8.91	20.94	21.52	17.12	13.46	61.28
C4	11.38	0.98	24.74	9.97	23.42	18.71	18.49	10.13
C5	0.20	0.23	0.10	0.11	10.74	19.47	20.58	12.17
C6	5.24	42.32	13.54	1.00	3.36	18.22	2.91	14.68
C7	8.31	0.20	20.38	11.97	22.61	18.48	30.83	17.83
C8	4.64	1.97	9.90	0.85	26.12	21.03	37.98	18.72
C9	1.34	0.98	11.88	44.87	37.54	19.00	40.64	1.91
C10	0.57	0.98	7.92	0.00	42.60	21.01	47.74	25.71

$\sigma(C_i, EM_i)$ is the matrix of the standard deviations of each C_i in the end-member EM*i* for the set C.

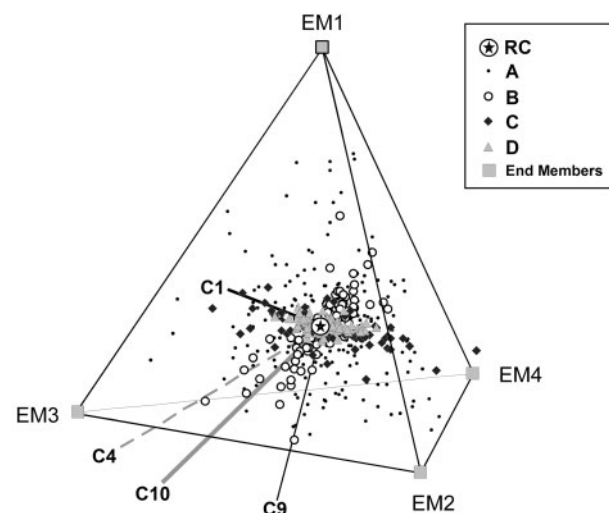


Fig. B1. Representation of the results of various numerical tests. The compositions of four arbitrary end-members, EM1, EM2, EM3 and EM4, are a combination of the 10 compositional parameters C1–C10 (Table B1). The reference composition (RC) is the average for each compositional parameter of the four end-members, and thus plots in the centre of the tetrahedron. The distributions of the four datasets are generated by the addition of: (set A) random amounts of the end-members to RC; (set B) the same random proportion of each of C1–C10 to RC; (sets C and D) variable random proportions of each C1–C10 to RC, allowing the largest standard deviation for C10 (the one with the minimum absolute value; Table B1). The standard deviation of each compositional parameter in set D is reduced by a factor of two with respect to set C. Some projections of the versors are reported. Each versor C_j is drawn by linking RC to a composition for which $C_{j \neq j} = 0$ and $C_j = 1$. Alignments of the data array in sets C and D is controlled by the versor of the compositional parameter with the largest absolute values.

standard deviation (Table B1). Figure B1 also reports the projections of the versors corresponding to some components. Each versor C_j is drawn by linking RC to a composition for which $C_{j \neq j} = 0$ and $C_j = 1$. Alignments of the data array in sets C and D are controlled by the

versor of the component with the largest absolute values. It is evident that the alignment of the data array in sets C and D are controlled by the direction (versor) assumed for the compositional parameter with the largest absolute values.