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The Pb isotopic compositions of lower crustal xenoliths and the evolution of lower crustal Pb

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Pb isotopic compositions for three suites of well-characterized granulite facies xenoliths from a diversity of crustal settings (the Chudleigh and McBride volcanic provinces, Queensland, Australia and the Eifel volcanics, West Germany) are presented here. All three suites plot to the right of the 4.57 Ga geochron, similar to the published Pb results of other mafic granulite xenoliths. Correlations between Sr, Nd and Pb isotopes in the three suites measured here point to an origin by mixing of mantle-derived basaltic magmas with lower crust at the time of basaltic underplating (i.e., < 100 Ma for Chudleigh, ~ 300 Ma for McBride, ~ 450 Ma for Eifel). Because the Pb concentration of the continental crust is much greater than that of mantle-derived basaltic magmas, the Pb isotopic compositions of the magmas are shifted dramatically by the mixing, allowing delineation of the isotopic characteristics of the lower crust. In all three cases, this lower crust had radiogenic Pb and Sr isotopic compositions and unradiogenic Nd isotopic compositions, yielding Proterozoic Nd model ages. Such radiogenic lower crust contrasts markedly with the Pb isotopic characteristics of most Precambrian granulite facies terrains. Whereas the Nd isotopes reflect the average age of crust formation, the Pb isotopic characteristics of the lower crust appear to be a function of the tectonothermal age of the crust: unradiogenic Pb can only develop in regions which have remained stable for long time periods (e.g., cratons), whereas in areas where orogenies have occurred subsequent to crustal formation, the Pb isotopic composition of the lower crust is “rejuvenated” through mixing with radiogenic Pb from upper crust and mantle-derived magmas. Thus, after orogeny, the Pb isotopic composition of the lower crust resembles that of the upper crust. On the basis of this proposed orogenic age-Pb isotope correlation, we estimate the Pb isotopic composition of the lower crust using the data for granulite xenoliths and granulite terrains. This, combined with an upper crust defined by modern sediments and ores, yields a model total crust composition which falls significantly to the right of the 4.57 Ga geochron. Thus, the lower crust does not appear to be sufficiently unradiogenic to balance the radiogenic upper crust and upper mantle Pb reservoirs.

1. Introduction

The continental crust is one of the earth's important Pb reservoirs and knowledge of its Pb isotopic composition is crucial in trying to constrain whole-earth Pb evolutionary models. Whereas the average Pb isotopic composition of the upper crust may be gleaned from the study of ore leads and sediments, that of the lower crust is harder to constrain. The lower crust is widely believed to be composed of rocks in the granulite

facies and a characteristic of granulites is their pervasive depletion in U [1,2], which gives rise to very low μ values ($\mu = {}^{238}\text{U}/{}^{204}\text{Pb}$). Thus, over time the Pb isotopic compositions of granulites evolve much more slowly than comparable upper crustal lithologies, causing the Pb isotopic compositions of granulites to be unradiogenic relative to that of the upper crust. For this reason, the lower crust has been proposed as the unradiogenic Pb reservoir needed to balance the radiogenic Pb of the depleted mantle and upper crust [3,4], and thereby resolve the Pb paradox [5]. (The Pb paradox is the apparent contradiction that both the upper crust and the depleted upper mantle have Pb more radiogenic than the bulk earth, as defined by the geochron).

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Generally, the Pb isotopic compositions of granulite terrains, which are often relatively unradiogenic (e.g., [6,7]), are used to model the lower crust [8]. However, granulite terrains presently lie at the earth's surface and, depending on the tectonics of their formation and exhumation, may or may not be representative of what lies in the present-day lower crust beneath these regions.

In contrast, granulite facies xenoliths, which are carried rapidly to the earth's surface by alkali basalts and kimberlites, provide direct information on lower crust composition for a variety of continental settings (see Rudnick [9] for a recent review). A lower crustal origin for most granulite xenoliths is documented by their high-pressure mineral assemblages and the presence of decompression melts, which, coupled with the occurrence of dense peridotite xenoliths in the same host rocks, collectively suggest that granulite xenoliths were carried rapidly to the earth's surface from great depths at the time of the host volcanism. In contrast with granulites from terrains, most granulite facies xenoliths have mafic compositions and, although some xenoliths may be related to earlier phases of the host volcanism [10,11], many are demonstrably older than this [12,13,14]. Thus, the mafic compositions of many xenoliths are not simply a function of their association with basaltic volcanism. Nevertheless, Nd and Sr isotopic compositions of many mafic granulite xenoliths point to their origin through mixing between a mantle-derived magma and compositionally evolved lower crust [10,15,16], the latter of which is infrequently sampled in xenolith suites. Granulite xenoliths therefore provide a window to the present-day lower crust, although compositionally speaking, they are not a totally representative sample.

Published Pb isotopic compositions for several suites of mafic granulite facies xenoliths show

them to be considerably more radiogenic than typical Precambrian granulite facies terrains (e.g., Camp Creek, Arizona [17]; Geronimo Volcanic Field, New Mexico [15]; Massif Central, France [18]), despite the fact that many of these xenoliths have Proterozoic Nd model ages and presently low μ 's. This observation may mean either that the xenoliths are old rocks, which have recently been metamorphosed to the granulite facies, or that the xenoliths are young mafic underplates which have assimilated crust having unradiogenic Nd and radiogenic Pb and Sr isotopic compositions. Distinguishing between these two interpretations is important for estimating crustal growth rates, delineating the importance of different crustal growth processes and understanding the evolution of Pb in the crust.

In this paper whole rock Pb isotopic measurements as well as U and Pb concentrations are reported for three suites of well-studied granulite facies xenoliths carried in Tertiary basalts: the Chudleigh and McBride volcanic provinces of northern Queensland, Australia and the Eifel basalts of West Germany. Examining the Pb isotopic compositions of xenoliths such as these, for which independent age information is available, provides constraints on interpretations of their Pb isotopic compositions. We use these data in conjunction with the literature data to evaluate the processes governing Pb isotopic evolution in the crust and to estimate the average Pb isotopic composition for the lower crust and the whole crust.

2. The samples

The three xenolith suites studied here come from continental areas of varied tectonic settings and ages, summarized in Table 1. As is true for

TABLE 1
Age and continental settings for granulite xenolith suites studied here

Suite	Setting	Age of protoliths	Age of metamorphism	Age of host
Chudleigh Prov., N. Queensland, Australia	Paleozoic fold belt	<100 Ma	<100 Ma	<1 Ma
McBride Prov., N. Queensland, Australia	Proterozoic block	1570 Ma and 300 Ma	~ 300 Ma	< 3 Ma
Eifel Volcanics, F.R.G.	Paleozoic massif, adjacent to Quaternary rift	1.5 Ga?	>170 Ma	<1 Ma

most lower crustal xenoliths, these three suites are dominated by mafic lithologies, many of which appear to have formed as crystal cumulates from mafic magmas.

The Chudleigh xenoliths were carried by Recent alkali basalts erupted on the boundary between the Proterozoic Georgetown Inlier and the Paleozoic Tasman fold belt of eastern Australia. These xenoliths have exclusively mafic compositions (see Rudnick et al. [10] and references therein) and equilibrated between 20 to 40 km depth. Their chemical and isotopic compositions reflect their origin as crystal cumulates derived from a mafic magma which underwent assimilation and crystal fractionation as it intruded and cooled in the lower crust [10]. Correlations between present-day Sr and Nd isotopic compositions and chemical composition (Mg# and various trace elements), coupled with their highly variable Sm/Nd ratios require these cumulates to be young (less than 100 Ma). They were therefore interpreted as being related to an earlier phase of the Tertiary basaltic volcanism [10].

In marked contrast to the Chudleigh xenolith suite, the McBride xenoliths were carried from depth (≥ 20 km) by Quaternary alkali basalts erupted within the Proterozoic Georgetown inlier. These xenoliths exhibit diverse compositions, ranging from mafic through felsic, although mafic compositions dominate (see Rudnick and Taylor [19] and references therein). Their chemical compositions reflect equally diverse protoliths for these rocks, including mafic cumulates, mafic melts, mafic restites, metapelitic sediments and felsic melts. The ages of the protoliths and the granulite facies metamorphism for these rocks have been determined by U–Pb zircon dating by ion microprobe [13]. Most protoliths formed at 300 Ma, a time of extensive calc-alkaline igneous activity in the region, but several protoliths formed during the Proterozoic at ~ 1570 Ma. All xenoliths underwent granulite facies metamorphism at 300 Ma, followed by slow cooling in the lower crust. A wide range of whole-rock $^{143}\text{Nd}/^{144}\text{Nd}$ (0.5130–0.5117) and $^{87}\text{Sr}/^{86}\text{Sr}$ (0.704–0.724) follows a mixing trend at 300 Ma [16], suggesting that most of these xenoliths were formed during large scale mixing between mantle-derived basalts and pre-existing crust at this time.

Recent alkali basalts and tuffs of the East Eifel

region of West Germany are associated with the uplift of the Caledonian Rhenish Massif, adjacent to the Rhein graben. These basalts carry abundant spinel peridotites as well as mafic granulites derived from ≥ 30 km (see Stosch and Lugmair [14], Looock et al. [20] and references therein). The granulites have the composition of mafic melts and cumulates derived therefrom. An unusual feature of the granulite xenoliths is the high abundance of amphibole, some of which is clearly secondary. The age of these xenoliths is uncertain. A Sm–Nd mineral isochron of 170 Ma provides a minimum age for the granulite facies metamorphism [14]. A three point whole rock Sm–Nd isochron of 1.5 Ga was suggested by Stosch and Lugmair [14] as the protolith age. Most samples lie to the left of the isochron, and were interpreted as reflecting the addition of a LREE-enriched metasomatic component, similar to that observed in the Eifel peridotite xenoliths. The amount of metasomatic component inferred to have been added on the basis of the Sm/Nd ratio correlates roughly with the amount of amphibole in the granulites. This, plus high Rb/Sr but low $^{87}\text{Sr}/^{86}\text{Sr}$ in the amphibole suggests that the metasomatic event was fairly recent.

3. Analytical techniques

Pb isotopic compositions were measured on whole rock powders in order to assure a representative sampling for these coarse-grained rocks. The powders were prepared by first removing weathered surfaces by sawing, then the samples were crushed in a steel jaw crusher. After splitting, approximately 50 g of sample was powdered in an agate (or WC, in the case of the Eifel samples) ring mill. Because of the potential of Pb contamination due to syn- or post-eruption natural processes (e.g., host-infiltration, weathering) or the above powdering procedure, all samples (~ 150 mg powder) were leached in hot 6N HCl for 1 h. The leachate and the residue were then analyzed for their Pb isotopic compositions, the results of which are discussed below. Separate dissolutions using unleached powders were performed for the isotopic dilution measurements of Pb, Th and U, using a ^{208}Pb spike and a mixed ^{230}Th – ^{235}U spike.

After dissolution in teflon bombs using HF and HNO_3 , Pb was separated on ~ 60 μl AG1-X8

TABLE 2
Pb isotopic results and Pb and U concentrations for lower crustal xenoliths

Sample	Pb (ppm)	Th (ppm)	U (ppm)	²⁰⁶ Pb/ ²⁰⁴ Pb (residue)	²⁰⁷ Pb/ ²⁰⁴ Pb (residue)	²⁰⁸ Pb/ ²⁰⁴ Pb (residue)	²⁰⁶ Pb/ ²⁰⁴ Pb (leachate)	²⁰⁷ Pb/ ²⁰⁴ Pb (leachate)	²⁰⁸ Pb/ ²⁰⁴ Pb (leachate)	μ	²⁰⁶ Pb/ ²⁰⁴ Pb (T)	²⁰⁷ Pb/ ²⁰⁴ Pb (T)
<i>Chudleigh granulite xenoliths (T = 0 Ma)</i>												
83-107	1.35		0.14	18.55	15.67	38.38	18.21	15.61	38.11	6.6		
83-110	0.44	0.03	0.11	18.18	15.60	38.31	17.56	15.54	37.43	15.8		
83-112	1.21	0.15	0.13	18.78	15.69	39.01	17.71	15.54	37.58	6.9		
83-114	1.15	0.03	0.02	18.57	15.63	38.39	17.93	15.58	37.81	1.1		
83-115	0.76	0.04	0.01	18.16	15.59	38.33	17.79	15.55	37.59	0.8		
83-117	0.87	0.02		18.29	15.56	38.13	17.74	15.56	37.63			
83-125	2.05	0.07	0.02	18.60	15.65	38.76	18.04	15.58	37.86	0.6		
<i>83-125 (unleached)</i>												
				18.58	15.64	38.69						
83-126	1.78	0.10	0.03	18.34	15.56	38.27	17.99	15.48	37.73	1.1		
83-127	0.47	0.04	0.02	18.28	15.60	38.41	17.80	15.58	37.80	2.7		
83-131	2.05	0.07	0.02	18.15	15.52	38.01	17.91	15.52	37.75	0.6		
83-140	1.15	0.11	0.05	18.67	15.68	38.35	18.00	15.56	37.76	2.8		
BC	0.28	0.03	0.01	17.79	15.45	37.57				2.2		
<i>McBride granulite xenoliths (T = 300 Ma)</i>												
83-157	8.3	1.14	0.44	18.40	15.62	39.83	18.36	15.57	39.22	3.4	18.24	15.61
<i>83-157 plagioclase</i>												
				18.28	15.63	39.90						
85-101	3.6	0.08	0.26	18.61	15.64	38.47	18.12	15.52	37.94	4.6	18.39	15.63
83-160	8.2	3.24	0.86	18.82	15.62	38.71	18.53	15.55	40.32	6.8	18.51	15.60
83-162	11.8	0.98	0.48	18.63	15.62	38.52	18.43	15.57	38.64	2.6	18.51	15.61
85-100	3.2	0.55	0.13	18.51	15.66	38.86	18.36	15.63	38.72	2.7	18.39	15.65
85-108	2.31	0.86	0.11	18.71	15.67	38.86	18.39	15.62	38.46	3.1	18.56	15.66
85-120	3.87	0.09	0.04	18.09	15.61	38.64	18.00	15.58	38.40	0.6	18.06	15.62
83-158	0.65	0.04	0.03	18.48	15.56	38.35				2.9	18.34	15.55
85-106	0.30	0.23	0.06	18.27	15.53	38.12	18.01	15.53	37.84	12.7	17.67	15.50
83-159	1.34	0.15	0.14	18.60	15.56	38.46	18.29	15.52	38.11	6.4	18.29	15.54
85-107	2.63	0.47	0.66	19.20	15.66	38.88	18.25	15.57	38.16	16.3	18.43	15.62
85-114	1.80	0.35	0.17	18.45	15.53	38.28	18.30	15.52	38.11	6.0	18.16	15.51
<i>Eifel granulite xenoliths (T = 450 Ma)</i>												
S-1	5.1	1.3	5.09	19.33	15.66	39.40	18.92	15.64	38.94	65.1	14.62	15.40
S-6	1.41		0.04	19.04	15.69	38.74	18.94	15.66	38.72	1.9	18.90	15.68
S-7	0.93	0.1	0.25	19.18	15.63	39.13	19.09	15.66	39.08	17.4	17.92	15.56
S-30	4.60		0.17	18.87	15.67	38.91	18.74	15.64	38.75	2.4	18.70	15.66
S-32	4.57	0.6	0.07	19.04	15.67	38.84	18.61	15.62	38.53	1.0	18.97	15.67
S-33	3.93		0.01	18.54	15.65	38.40	18.46	15.62	38.35	0.2	18.28	15.65
S-35	2.66		0.04	18.30	15.64	38.47	18.32	15.64	38.42	1.0	18.21	15.64
S-37	0.98		0.02	18.50	15.66	38.45	18.60	15.72	38.68	1.3	18.41	15.65
S-43	5.92	2.5	0.48	19.03	15.64	38.96	18.86	15.63	38.80	5.1	18.65	15.62

* Marked Pb and U concentrations are through spark source mass spectrometry, all others by isotope dilution. Th concentrations by spark source mass spectrometry for Chudleigh and McBride samples and by isotope dilution for Eifel samples. Average values for NBS 982 measured during the course of these analyses are: ²⁰⁶Pb/²⁰⁴Pb = 36.626, ²⁰⁷Pb/²⁰⁴Pb = 17.091, ²⁰⁸Pb/²⁰⁴Pb = 36.548 (n = 81). 2σ = 0.5%/amu which is taken to be the analytical error.

anion exchange columns using HBr and HCl. Total Pb blanks were ≤ 250 pg. Isotopic measurements were performed on a MAT 261 multicollector mass spectrometer in static mode. Correction factors of 1.53‰/amu for $^{206}\text{Pb}/^{204}\text{Pb}$ and 1.34‰/amu for both $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ were made based on multiple analyses of NBS 982 ($^{206}\text{Pb}/^{204}\text{Pb} = 36.738$, $^{207}\text{Pb}/^{204}\text{Pb} = 17.159$, $^{208}\text{Pb}/^{204}\text{Pb} = 36.744$), and account for both fractionation and cup bias. Estimated errors based on between-run precision of the standard are 0.5‰/amu (2σ , $n = 81$, see Table 2); in-run precision for individual analyses was much better than this. Each sample was run twice and the results averaged.

4. Results

The Pb isotopic compositions of both leachates and whole rock residues are presented in Table 2, along with U, Th and Pb concentrations and the Pb isotopic composition of one plagioclase separate from a McBride province xenolith. The similarity of Pb isotopic compositions for the plagioclase separate and the leached whole rock for sample 83-157 demonstrates that any possible effect on the Pb isotopes of the whole rock by host basalt infiltration has been removed by the leaching procedure. The whole rock data are plotted in Figs. 1-7.

Although strong leaching of a powder has the advantage of removing possible contaminants introduced through sample processing or host-xenolith interactions, it may also yield spurious results due to preferential leaching of U- or Th-enriched accessory phases (e.g., apatite). This is a particularly severe problem for samples in which significant radiogenic growth has occurred since the last mineral equilibration. For the granulite xenoliths the measured leachates are displaced, to variable degrees, towards lower $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ ratios compared with the residues (Fig. 1). This labile, less radiogenic Pb component is not easily explained as being derived from accessory phases, since they should contain more radiogenic Pb relative to the less easily dissolved, Th- and U-poor phases (e.g., orthopyroxene, clinopyroxene, plagioclase). The composition of the leachate could be explained as being due to a contaminant having a Broken Hill Pb composition

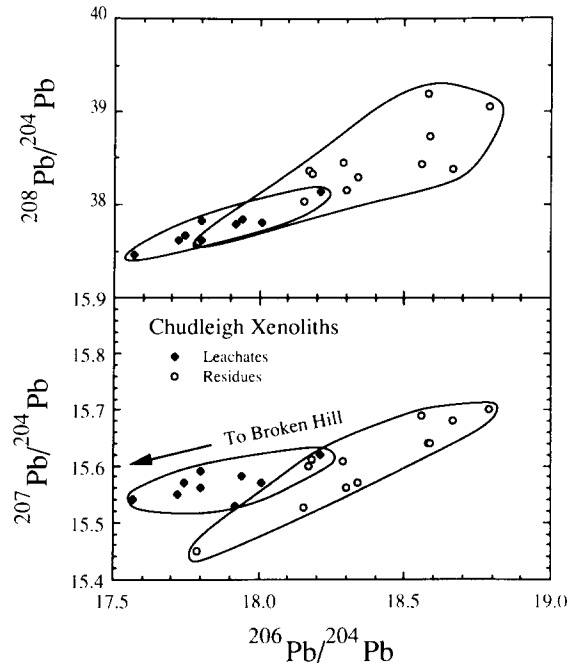


Fig. 1. $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ showing the compositions of Pb derived from leaching the Chudleigh sample powders in hot 6N HCl for 1 h (leachates) and the remaining whole-rocks (residues). Arrow points to Broken Hill Pb component at $^{206}\text{Pb}/^{204}\text{Pb} = 16.00$, $^{207}\text{Pb}/^{204}\text{Pb} = 15.39$ and $^{208}\text{Pb}/^{204}\text{Pb} = 35.65$.

([21], see Fig. 1), the ambient common Pb composition in both Australia and Europe [22]. This contaminant was presumably introduced during sample preparation through sawing and jaw crushing. The amount of contaminant present in each sample is unknown since the leachate represents a mixture of contaminant and rock Pb. However, the Pb contributed by the contaminant is not likely to be large since measurements of both an unleached and leached powder for one sample give comparable results (Table 2). The generally low U contents and relatively young mineral equilibration ages for these xenoliths ($< \sim 200$ Ma) also suggest that the leaching has not altered the whole rock Pb composition. Thus the results for the residues are likely to reflect the in situ lower crustal Pb isotopic compositions.

The Pb isotopic compositions of the three xenolith suites are all relatively radiogenic. They plot to the right of the geochron, well away from the fields of Archean granulite terrains, e.g., the unradiogenic Pb compositions characterized by

highly depleted (low μ) Archean granulite terrains (e.g., Scourian [6,23], Southern India [24], Labrador and Amitsoq [25,26,27] gneisses) and the terrains characterized by high $^{207}\text{Pb}/^{204}\text{Pb}$ ratios due to high μ conditions early on, followed by U-depletion, such as the Enderby Land [28] and Limpopo granulites [29] (see Fig. 9). The xenoliths are also distinctly more radiogenic than most Proterozoic granulite terrains. The Pb isotopic compositions of the xenoliths most closely resemble those of modern basalts.

5. Interpretations of individual suites

The Pb isotopic compositions of the Chudleigh province xenoliths are shown in Fig. 2 relative to the geochron and the fields for mid-ocean ridge basalts (MORB), Tertiary basalts of eastern Australia and modern sediment. The data follow a trend from the lower $^{206}\text{Pb}/^{204}\text{Pb}$ end of the MORB field to the field of modern sediments,

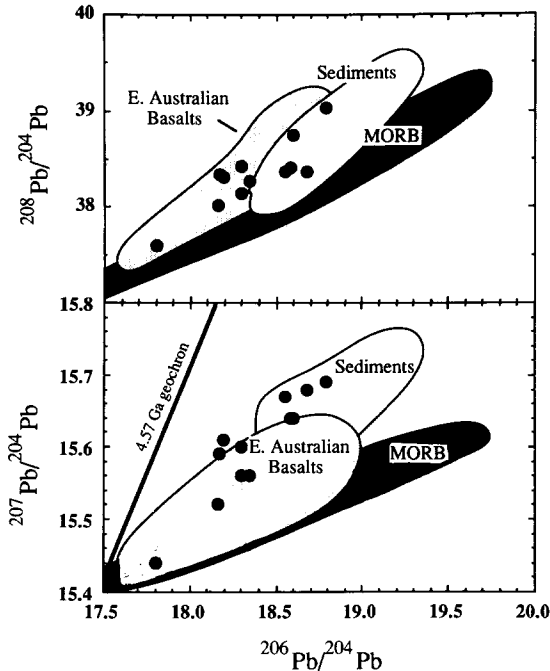


Fig. 2. Present-day Pb isotopic compositions of Chudleigh granulite facies xenoliths shown relative to the fields of mid-ocean ridge basalts, eastern Australian Tertiary basalts (from Ewart et al. [30]) and modern oceanic sediments (from White and Dupré [49]).

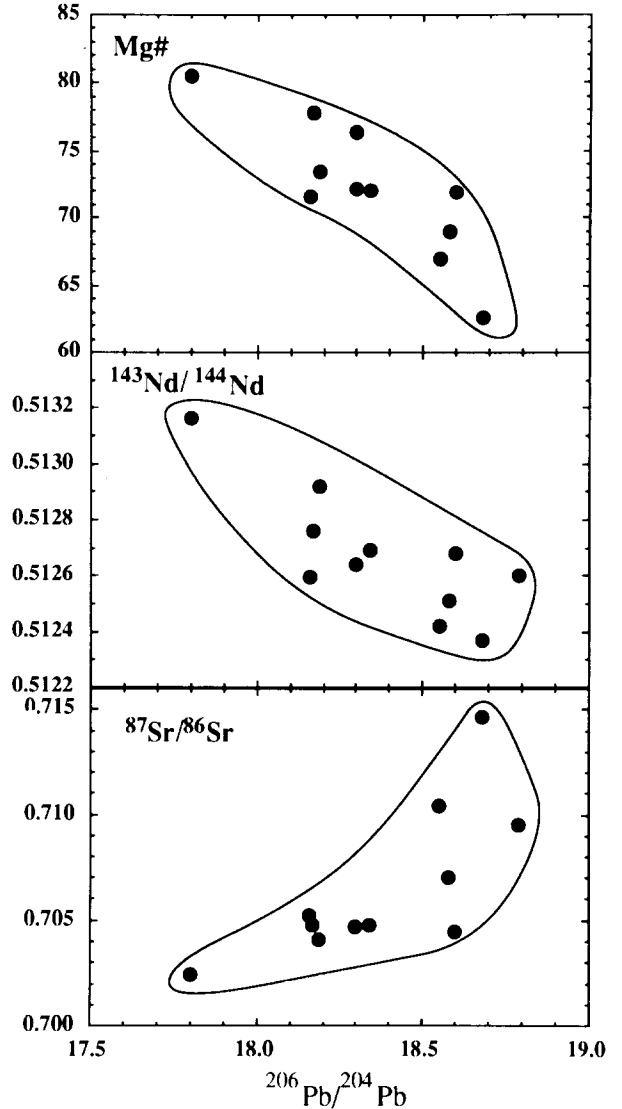


Fig. 3. Present-day $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$ and Mg# for Chudleigh granulite facies xenoliths. Nd, Sr and Mg# from Rudnick et al. [10].

completely overlapping the field of Tertiary basalts from eastern Australia. The Pb isotopic compositions of the eastern Australian basalts have been interpreted in terms of assimilation and fractional crystallization of mantle-derived basalts [30]. The Chudleigh data also reflect such processes, as indicated by the good correlation between the $^{206}\text{Pb}/^{204}\text{Pb}$ ratio and $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$ and Mg# for the xenoliths (Fig. 3), consistent with earlier interpretations based on geochemical and Sr and Nd isotopic results [10]. Thus the magma

giving rise to the Chudleigh cumulates had Nd, Sr and Pb isotopic compositions similar to those of MORB. This magma mixed with material in the lower crust having radiogenic Pb and Sr isotopes and unradiogenic Nd. The linear array on the $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $^{207}\text{Pb}/^{204}\text{Pb}$ plot thus has no age significance, but it does serve to characterize the isotopic nature of the lower crust into which these basaltic melts intruded.

The McBride xenoliths, shown in Fig. 4, have Pb isotopic compositions similar to those of the Chudleigh xenoliths, although they exhibit more scatter. The Pb isotopic composition of a plagioclase separate from one sample (83-157) is within error of the whole rock for the $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$, but has slightly lower $^{206}\text{Pb}/^{204}\text{Pb}$ (Table 2). This yields an age of ~ 240 Ma (assuming $\mu = 0$ in the plagioclase), similar to the age of granulite facies metamorphism in the lower crust (200–300 Ma, as determined by ion microprobe zircon dating [13]) and extensive calc-alkaline volcanism in this region (270–320 Ma). When the whole rock Pb data are viewed at 300 Ma, rough correlations between $^{206}\text{Pb}/^{204}\text{Pb}$ and

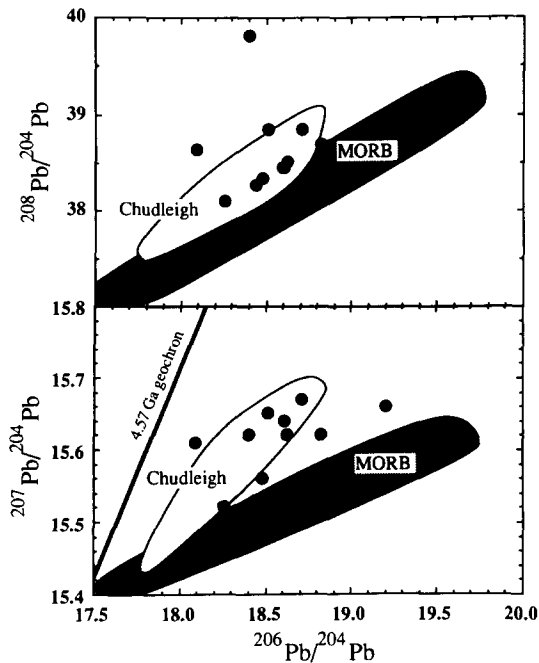


Fig. 4. Present-day Pb isotopic compositions of McBride granulite facies xenoliths shown relative to the fields of MORB and the Chudleigh xenoliths.

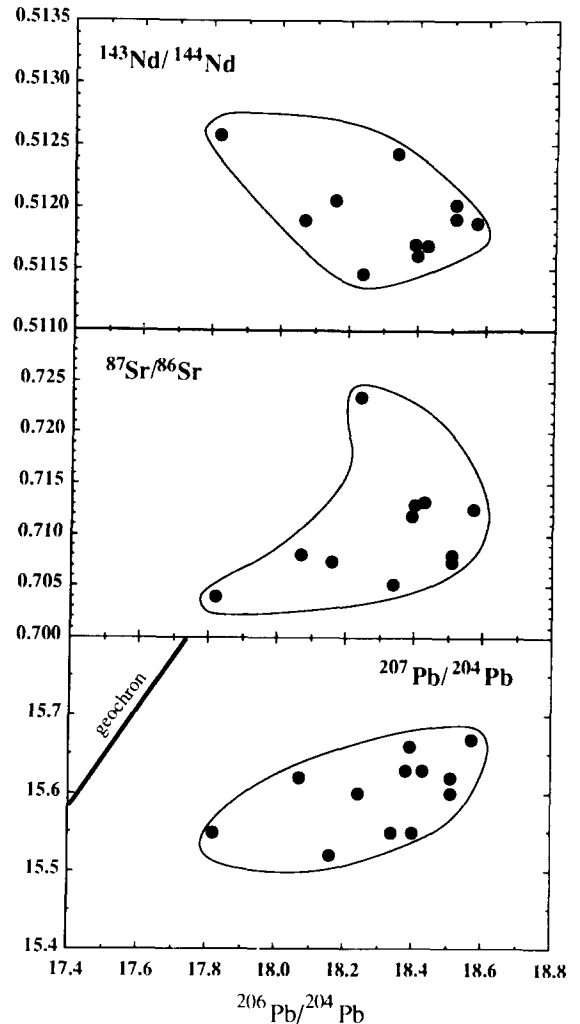


Fig. 5. $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $^{207}\text{Pb}/^{204}\text{Pb}$, $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ for McBride xenoliths at 300 Ma. Sr and Nd data are from Rudnick [16].

$^{207}\text{Pb}/^{204}\text{Pb}$, Nd and Sr isotopic compositions are apparent (Fig. 5). These trends, coupled with earlier interpretations of the Sr and Nd chemistry and zircon ion probe ages, are best interpreted as being due to mixing between basaltic melts, intruded at ~ 300 Ma, and older, isotopically evolved, continental crust. Both mixing end members are present in the xenolith suite. Thus, the lower crust in this region is composed of magmatically underplated 300 Ma old basalts, Proterozoic rocks and mixtures between these two rock types. The observation that the Proterozoic xenoliths

have radiogenic Pb isotopic compositions but presently low μ , suggests that they underwent granulite facies metamorphism at ~ 300 Ma, and were thus transported into the lower crust at this time.

The Eifel xenoliths plot in a horizontal field on the $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $^{207}\text{Pb}/^{204}\text{Pb}$ diagram, and show a positive correlation on the $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $^{208}\text{Pb}/^{204}\text{Pb}$ diagram (Fig. 6). These xenoliths overlap the lower $^{206}\text{Pb}/^{204}\text{Pb}$ end of the field of the Tertiary Eifel basalts, but most plot at significantly lower $^{206}\text{Pb}/^{204}\text{Pb}$ than the basalts. There is no correlation between the Pb isotopic compositions and the percentage of modal amphibole, nor with $^{143}\text{Nd}/^{144}\text{Nd}$ or $^{87}\text{Sr}/^{86}\text{Sr}$ [14]. All but two of the Eifel xenoliths have low μ values, and several of these low μ xenoliths are interpreted to be cumulates from basaltic magmas (e.g. [20,31]). Thus their low μ values are likely to be primary features, and their present-day Pb isotopic compositions are only slightly more evolved than their initial isotopic composition. Such radiogenic compositions are unlike the initial Pb isotopic compositions of Proterozoic or older basalts (assuming

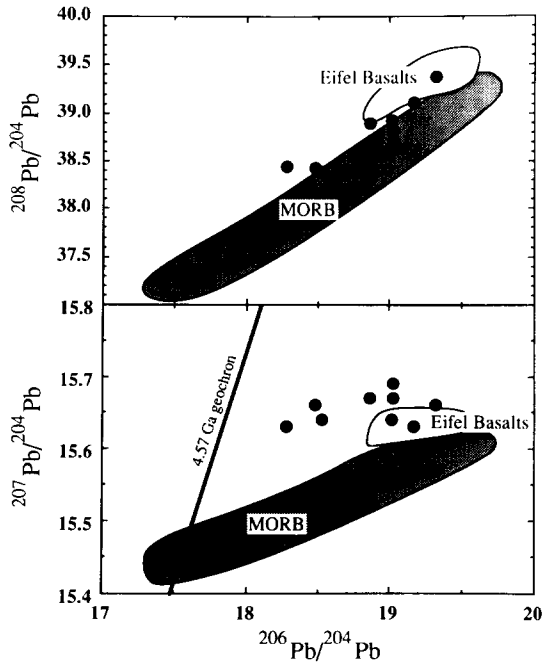


Fig. 6. Present-day Pb isotopic compositions of Eifel granulite facies xenoliths shown relative to the fields of MORB and Tertiary Eifel basalts (from Wörner et al. [50]).

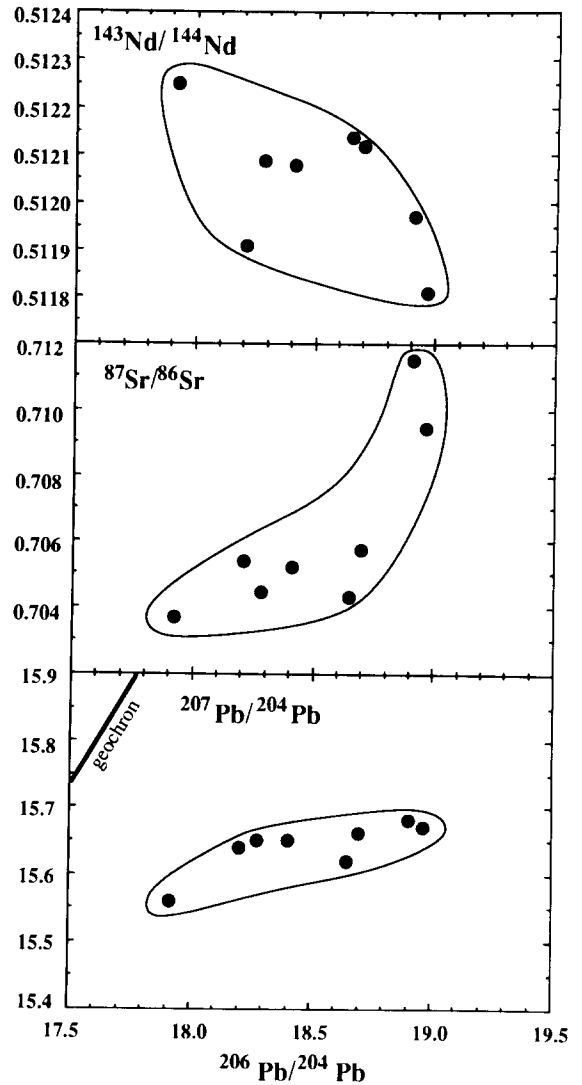


Fig. 7. $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $^{207}\text{Pb}/^{204}\text{Pb}$, $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ for Eifel xenoliths at 450 Ma. Sr and Nd data are from Stosch and Lugmair [14] and Lock et al. [20]. Note that sample S-1 is not shown here, as due to its very high μ , its $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ ratios are unrealistically low at 450 Ma. This sample comes from a different locality from the rest of the Eifel samples [20] and, considering peculiarities in its chemistry (particularly its high Th and U contents), may not be related to the other Eifel xenoliths.

that these magmas, like modern basalts, plotted slightly to the right of their respective geochron), suggesting that these mafic cumulates are younger than Precambrian. When the Eifel data are viewed at 450 Ma, a time of major orogeny (Caledonian) in this part of Europe, trends emerge between Pb,

Sr and Nd isotopic compositions (Fig. 7), excluding S-1, see figure caption). As with the Queensland xenoliths described above, these correlations may be explained as the product of mixing between underplated basalts and a Precambrian lower crustal component, having high $^{87}\text{Sr}/^{86}\text{Sr}$, $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ and low $^{143}\text{Nd}/^{144}\text{Nd}$ relative to the intruding magma. The Proterozoic Nd model ages therefore reflect assimilation of Proterozoic crust by ~ 450 Ma basaltic magmas.

6. Comparison with other suites

How do the Pb results presented above compare with those of other xenolith suites? The Pb isotopic compositions of 6 other granulite xenolith suites, reported in the literature, show a range in $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ (Fig. 8). Three of these suites have radiogenic Pb iso-

topic compositions, similar to those described above (i.e., Massif Central [18]; Camp Creek, Arizona [17] and the Geronimo Volcanic Field [15]). The remaining three suites have unradiogenic Pb isotopic compositions and plot well to the left of the geochron (these are: mafic granulites from Lesotho [32] and Lashaine [33] and metapelitic granulites from Kilbourne Hole, New Mexico [34]).

Following the conclusions reached above, the three suites of mafic xenoliths plotting to the right of the geochron (Massif Central, Camp Creek and Geronimo Volcanic Field) may be interpreted as relatively young (Phanerozoic) basaltic underplates. If crustal assimilation has occurred in these suites, either this crust was relatively radiogenic with respect to Pb, or only small amounts of crust with unradiogenic Pb were assimilated. The former possibility is supported by the presence of old Nd (giving Proterozoic Nd model ages) in many of these samples. The two suites of mafic xenoliths plotting to the left of the geochron (Lashaine and Lesotho) may also represent Phanerozoic basaltic underplates which assimilated very unradiogenic lower crust, or, perhaps more likely, older basaltic underplates that intruded with or without accompanying assimilation. The Kilbourne Hole metasedimentary xenoliths are Proterozoic granulites which have evolved with low μ since their metamorphism.

7. Discussion

Four important points about lower crustal xenoliths emerge from the above data and previous studies: (1) The dominance of mafic lithologies in many granulite xenolith suites point to the importance of basaltic underplating as a crustal growth process. (2) The cumulate xenoliths having radiogenic Pb isotopic compositions, in which low μ is a primary feature, are relatively young (Phanerozoic), but many have old Nd. (3) When independent age information is available, most mafic granulite xenoliths are shown to be older than the earliest phase of volcanism associated with the host. (4) Radiogenic Pb and Sr isotopic compositions and unradiogenic Nd (yielding Proterozoic Nd model ages) in mafic granulite xenoliths reflect mixing between basaltic underplates and pre-existing, chemically evolved, lower crust.

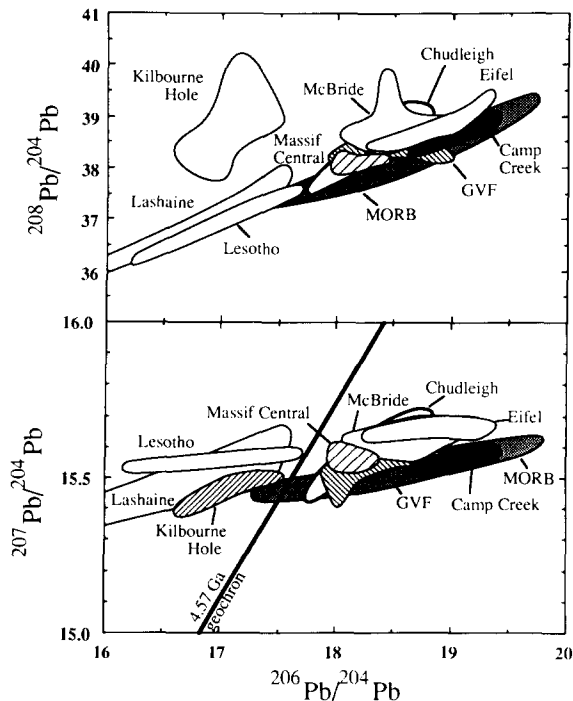


Fig. 8. Pb isotopic compositions of world-wide granulite facies xenoliths. Data are from this work and the following: Massif Central, France: Vidal and Postaire [18]; Camp Creek, Arizona: Esperança et al. [17]; Geronimo Volcanic Field, New Mexico: Kempton et al. [15]; Lesotho, southern Africa: Van Calsteren et al. [32]; Lashaine, Tanzania: Cohen et al. [33]; Kilbourne Hole, New Mexico: Reid [34].

These features may be used to develop a model for how and when the lower crust forms, and to predict its isotopic composition.

Although rarely sampled in xenolith suites, the presence of compositionally evolved lower crust is inferred in many instances by the mixing relationships exhibited in the Pb, Sr and Nd isotopic systems. These mixing relations may be used to characterize the lower crust prior to underplating, and in combination with the compositions of the underplates, be used to model the Pb isotopic composition of the lower crust. Because the ratio of Pb concentration between the upper continental crust and mantle-derived basalts is high (2–10) [35,36], higher than the ratios for Sr and Nd (0.5 to 2.0 and 1.0 to 5.0, respectively), the Pb isotopic compositions of contaminated magmas are shifted more readily toward the crustal value than either the Sr or Nd isotopic compositions (a good example of this is the Tertiary basalts of Skye, N.W. Scotland [37]). Thus, the Pb isotopic compositions of mafic xenoliths formed through intrusion and contamination of mantle derived basaltic magmas reflect the Pb isotopic composition of the total lower crust, even though not all lower crustal lithologies may be sampled as xenoliths.

Most granulite xenoliths have relatively radiogenic Pb isotopic compositions, and plot to the right of the geochron (Fig. 8). However, several suites fall to the left, illustrating that the Pb isotopic characteristics of the lower crust vary from region to region. Of the nine lower crustal xenolith suites shown in Fig. 8, six were carried by host magmas which traversed crust having Proterozoic crystallization ages (McBride, Camp Creek, Geronimo Volcanic Field, Kilbourne Hole, Lashaine and Lesotho). The remaining three suites (Eifel, Massif Central, Chudleigh) come from areas where crustal crystallization ages are Phanerozoic, but Nd model ages are Proterozoic, suggesting that some crust in these regions formed in the Proterozoic or earlier. Thus there is no apparent correlation between the Pb isotopic composition of the lower crust and the age of crustal formation.

What appears to be more crucial to Pb isotopic evolution in the lower crust is the timing since the last orogeny. If a granulite forms and remains in the lower crust undisturbed until transported to the surface by basaltic or kimberlitic volcanism,

then the lower crust in this region will be retarded in radiogenic Pb growth through time and the xenolith will have unradiogenic Pb. If, on the other hand, later orogeny tectonically transports upper crustal lithologies (having radiogenic Pb) into the lower crust, and this material is allowed to mix with pre-existing lower crust (characterized by unradiogenic Pb) as well as mantle-derived basaltic intrusions, then the Pb isotopic composition of the lower crust will be rejuvenated to more radiogenic Pb values. This latter scenario is quite similar to the model of the orogene put forth by Zartman and others in the plumbotectonics model, where lower crustal Pb, upper crustal Pb and mantle-derived Pb are re-mixed during an orogenic event [8,38,39]. The xenoliths having radiogenic Pb appear to have formed in a region thus rejuvenated.

8. Pb model composition

8.1 *The lower crust*

If this concept of lower crustal Pb holds, then it is possible to model the Pb isotopic composition of the lower crust on the basis of the age of the last orogeny. In doing this we assume that the bulk composition, hence Pb concentration, in the lower crust has not changed appreciably with time. However, the marked compositional differences between granulite terrains and xenoliths, coupled with the generally young age of xenoliths compared to terrains may suggest that the lower crust has become more mafic with time. We do not believe this to be the case on the basis of the following two observations: (1) the limited data available for granulite facies xenoliths erupted within Archean cratons show mafic compositions to be important (see [9], and references therein), and (2) the compositional features of granulite xenoliths outlined above suggest that they have commonly mixed with non-mafic lower crust, but this crust is seldom sampled in xenolith suites. These observations suggest that the compositional differences between terrains and xenoliths may be a function of their respective sampling mechanisms and do not require the lower crust composition to change with age.

To facilitate estimating the tectonothermal age of the crust, we have adopted the map of Sclater et al. [40], which divides the crust into age prov-

inces (i.e., 0–250; 250–800; 800–1700; and > 1700 Ma) on the basis of the timing of the last orogenic event. The Pb isotopic compositions of these crustal provinces can then be estimated by using the data for granulite xenoliths (which are principally derived from tectonically young crustal blocks) and granulite terrains (which should be representative of the Pb isotopic composition of the lower crust stabilized beneath Archean and Proterozoic shields—note that there are no published data for granulite xenoliths erupted within Archean cratonic regions). By using actual granulite data rather than model Pb compositions based upon single stage growth curves the following potential problems are alleviated: (1) the amount of radiogenic Pb growth occurring both before and after granulite facies metamorphism is accounted for, and (2) no assumptions need be made regarding the initial Pb isotopic composition of the lower crust.

Table 3 lists the crustal age provinces, along with their relative areal extent and the estimated lower crustal Pb compositions derived from granulites of various ages. It is apparent from Fig. 9 that, whereas the Pb isotopic compositions of Proterozoic and younger granulites fall into reasonably tight groupings, those of Archean granulites are widely variable. For this reason we have chosen two end-member compositions for > 1.7 Ga crust (Table 3): one plots near the lower

(unradiogenic) end of the Scourian and southern India fields, the other plots near the center of the 2.0 to 1.7 Ga granulites (Lapland, Lofoten, Molendo and Charcani) and near the lower end of the high $^{207}\text{Pb}/^{204}\text{Pb}$ Archean granulites (Limpopo and Enderby Land). This latter composition is probably better representative of > 1.7 Ga lower crust since the amount of crust stabilized before 3.0 Ga (hence having very unradiogenic Pb isotopic compositions like the Hebron and Amitsoq gneisses) is very small and not all Archean granulites are unradiogenic (e.g. Enderby Land and Limpopo). Nevertheless, we have considered Model 1 in order to maximize the amount of unradiogenic Pb potentially contained within the lower crust. The resulting Pb isotopic compositions are listed as Model 1 (unradiogenic) and 2 (radiogenic) in Table 3.

We have also modelled the evolution of lower crustal Pb on a purely theoretical basis assuming that the Pb isotopic composition of the lower crust is “rejuvenated” by the most recent orogenic event, but that after the segment is stabilized the Pb isotopes cease to evolve in the lower crust. The average Pb isotopic composition of the lower crust would then evolve independently of the upper crust, depending on the tectonothermal history of the continental crust. We have used a Stacey–Kramers model for upper crustal Pb isotopic evolution, and have calculated average Pb com-

TABLE 3

Crustal ages and model crustal Pb compositions, see text for explanation of models

Tectonothermal age (Ma)	0–250 Ma	250–800 Ma	800–1700 Ma	> 1700 Ma	
Proportion of total	19.1%	34.4%	14.2%	32.3%	
<i>Continental area</i> *				Model 1	Model 2
$^{206}\text{Pb}/^{204}\text{Pb}$	18.5	17.7	17.0	13.8	16.0
$^{207}\text{Pb}/^{204}\text{Pb}$	15.6	15.6	15.5	14.4	15.5
<i>Total lower crust models</i>					
	Model 1	Model 2	Model 3	Model 4	
	(see text)	(see text)	[40]	[42]	
$^{206}\text{Pb}/^{204}\text{Pb}$	16.5	17.2	16.9	16.63	
$^{207}\text{Pb}/^{204}\text{Pb}$	15.2	15.5	15.3	15.3	
<i>Range of total crust</i> ¹					
$^{206}\text{Pb}/^{204}\text{Pb}$	18.04–18.35				
$^{207}\text{Pb}/^{204}\text{Pb}$	15.51–15.62				

* From Sclater et al. [40].

¹ Assuming 7 ppm Pb in lower crust, 15–20 ppm Pb in upper crust, Pb isotopic compositions for upper crust: $^{206}\text{Pb}/^{204}\text{Pb} = 18.76$, $^{207}\text{Pb}/^{204}\text{Pb} = 15.66$, from Newsom et al. [4] and lower crust from models 1–4 in this Table.

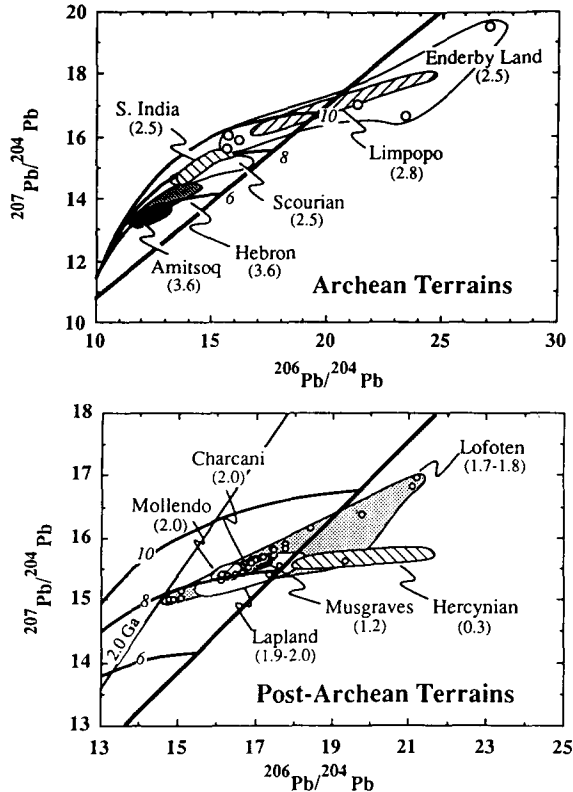


Fig. 9. Pb isotopic compositions of granulite facies terrains shown relative to a 4.57 Ga geochron and single-stage Pb growth curves of $\mu = 6$ to 10 (note scale change between upper and lower diagrams). Numbers in parentheses are metamorphic ages. Individual data points are shown for the Enderby Land (upper) and Lofoten (lower) granulites. Data are from: (upper diagram) Enderby Land, Antarctica: DePaolo et al. [28]; Limpopo, South Africa: Barton et al. [29]; Southern India: Peucat et al. [24]; Scourian, Scotland: Moorbath et al. [6], Whitehouse [23]; Hebron gneisses, Labrador: Schiøtte et al. [26]; Amitsoq gneisses, Greenland: Black et al. [25], Griffin et al. [27]; (lower diagram) Lofoten, N. Norway: Taylor [51] and Griffin et al., [52]; Charcani and Mollendo gneisses, Peru: Tilton and Barreiro [53] and Tilton, pers. commun., 1989; Hercynian granulites, Europe: Vidal and Postaire [18]; Musgrave ranges, Australia: Gray and Oversby [7]; Lapland, Finland: Bernard-Griffiths et al. [54].

positions for the lower crust for two crust stabilization curves (Fig. 10). One estimate (Model 3 in Table 3) is based on the tectonothermal age curve of Sclater et al. [40] (curve 2 in Fig. 10), and gives an average Pb isotopic composition for the lower crust similar to Model 2 described above. (Another, even younger, estimate of the tectonothermal age of the continental crust from Sprague and Pollack [41] is shown (curve 1 in Fig. 10),

which would yield an even more radiogenic lower crust.) The second calculation (Model 4 in Table 3) uses a hypothetical tectonothermal age curve after Veizer and Jansen [42], who showed that if a constant proportion of the crust is “recycled” during orogeny through geologic time, the mass-tectonothermal age curve can be described by a power law (see caption to Fig. 10). The exponent of the power law was chosen to be consistent with Sclater et al. [40] for young ages, but attributes a significantly larger proportion of continent to older tectonothermal provinces. The estimated lower crust Pb isotopic composition thus derived is similar to model 1 described above. It must be emphasized that these calculations give least-radiogenic estimates for the lower crust because post-cratonization radiogenic Pb growth is not taken into account.

The lower rows in Table 3 give the bulk lower crust Pb isotopic compositions calculated from the granulite data and the crust stabilization models. These are compared in Fig. 11 with previous estimates of lower crustal Pb isotopic compositions. Although all four of our compositions plot

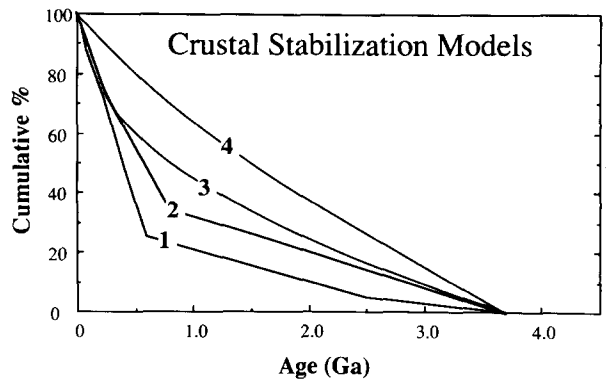


Fig. 10. Tectonothermal age curves depicting the amount of crust stabilized from further tectonic reworking as a function of age. Curve 1 is that of Sprague and Pollack [41], curve 2 is from Sclater et al. [40]. Curves 3 and 4 are hypothetical and assume the fraction of continent that has survived undisturbed since the most recent orogeny can be described by a power law (Veizer and Jansen [42]). In these cases, $F(t) = [1 - (t/T)^n]$, where t is age (presently $t = 0$), T the total time period considered (i.e., 3.8 Ga) and F the fraction of continent with tectonothermal ages greater than t . Values of n were chosen to be consistent with the Sclater et al. curve for young ages (curve 3, $n = 0.45$), and to create a lower crust unradiogenic enough so that the whole crust plots on the 4.57 Ga geochron (curve 4, $n = 0.7$).

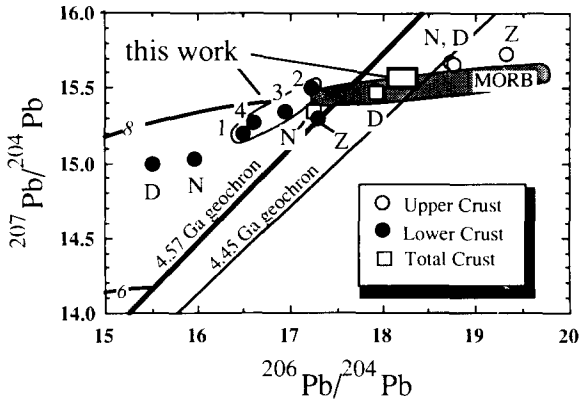


Fig. 11. Pb model compositions of the lower crust, upper crust and total crust shown relative to the 4.57 Ga geochron, the 4.45 Ga geochron and the field of MORB. *D* = Davies [3]; *Z* = Zartman and Doe [8]; *N* = Newsom et al. [4]. Model lower crusts 1 and 2 are calculated on the basis of the granulite data, models 3 and 4 are calculated based on the tectonothermal age curves shown in Fig. 10. The large box in the MORB field represents the range of our calculated total crust composition, assuming a upper crust composition of Newsom et al. [4].

to the left of the geochron, they are significantly more radiogenic than the lower crusts proposed by Davies [3] and Newsom et al. [4], and fall further to the left of the geochron than the estimate of Zartman and Doe [8]; see also Table 4.

8.2 The total crust

The above calculations can be carried one step further by calculating the average total crust Pb isotopic compositions, if the upper crustal Pb isotopic compositions and Pb concentration ratio between the upper and lower crusts are known. We have used average upper crustal Pb isotopic compositions similar to that of Newsom et al. [4], which is based on modern sediment compositions and is similar to modern ore Pb [43]. In contrast

TABLE 4

Pb contents (in ppm) of upper and lower crust

	T&M	Z&D	Davies	N et al.	This work
Upper crust	20	23.5	13	15	15–20
Lower crust	4	6.8	5.2	7.5	7

Where T&M = Taylor and McLennan [35]; Z&D = Zartman and Doe [8]; Davies = Davies [3]; N et al. = Newsom et al. [4].

to the above calculation of lower crustal Pb isotopic compositions, the calculation of average total crustal Pb isotopic compositions is critically dependent upon the Pb concentration of the lower crust, which we assume to be constant through time and probably lies between that of granulite facies terrains (10–14 ppm [2]) and granulite xenoliths (2 ppm [2]). We have therefore chosen a value of 7 ppm, which lies in the middle of the range chosen by previous investigators (Table 3).

Using this Pb concentration for the lower crust and an upper crustal Pb concentration between 15 and 20 ppm, and assuming that the lower crust represents half of the total crust, a range of total crustal Pb isotopic compositions is calculated from the isotopic estimates given in Table 3 and are shown as a box in Fig. 10. This range of compositions falls significantly to the right of the 4.57 Ga geochron and is more radiogenic than the estimate for the total crust of Newsom et al. [4], but similar to that of Davies [3].

Although a few MORB Pb isotope data lie to the left of the 4.57 Ga geochron, the majority of Pb from the suboceanic mantle lies to the right. In addition, because 75 to > 90% of the differentiated silicate earth's Pb resides in the continental crust (depending upon the fraction of mantle that has been depleted in order to form the continental crust [44,45,46]), the crust exerts a strong influence on where the bulk earth sits with respect to the geochron. If the total crust sits slightly to the left of the geochron, then the Pb paradox can be resolved. However, in the above models the total crust sits distinctly to the right of the 4.57 Ga geochron. There are only two ways to shift this composition to the left of the geochron: (1) by increasing the Pb content of the lower crust, or (2) by having a lower crust significantly less radiogenic than the models shown in Table 3. The estimated Pb content of the lower crust may indeed be too low if our model concentration is biased too heavily in favor of mafic compositions. However, in order for the Pb isotopic composition of the total crust to sit to the left of the 4.57 Ga geochron, the lower crust would need to have a *higher* Pb content than the upper crust. Since the median Pb contents of granulite terrains (10–14 ppm [2]) are lower than that of the upper crust, this is unlikely to be the case. With regard to the second point, we have calculated the tectono-

thermal age curve required to shift the lower crustal Pb isotopic composition to the unradiogenic values needed to place the whole crust to the left of the 4.57 Ga geochron. This is curve 4 in Fig. 10, which gives a lower crustal Pb isotopic composition similar to that of Newsom et al. [4]. (Note that it also assumes no post-cratonization Pb growth.) This curve requires that the stabilization of the continental crust occurred at a nearly uniform rate since 3.7 Ga, and appears to severely overestimate the amount of old craton compared to empirical tectonothermal age curves [40,41]. If our model linking the Pb isotopic composition of the lower crust to the tectonothermal age is correct, then the bulk crust must sit to the right of the geochron.

If the 4.57 Ga geochron represents the timing of the beginning of Pb evolution in the earth, then the lower crust is not sufficient as the unradiogenic Pb reservoir needed to resolve the Pb paradox. This suggests that either another unradiogenic Pb reservoir exists within the earth, or that terrestrial Pb evolution commenced after 4.57 Ga.

The continental lithospheric mantle may represent an unradiogenic Pb reservoir, especially if lower crustal foundering, as advocated by Arndt and Goldstein [47], is an important process. However, in order for the average Pb isotopic compositions of the continental lithospheric mantle (CLM) plus continental crust to lie on the 4.57 Ga geochron, the CLM (which is here assumed to be four times the mass of the continental crust) needs an average Pb concentration of 1.2–1.9 ppm (using the upper and lower continental crust as defined above in our total crust estimate and assuming that the Pb isotopic compositions of the CLM are equal to those of the lower crust). These assumptions maximize the role of the CLM in solving the Pb paradox—our assumed volume is larger than most estimates, with an average depth in the diamond stability field, and the assumption that the CLM is a low μ reservoir is probably unwarranted. Even so, the average Pb concentrations required in the CLM are higher than those observed in lithosphere-derived peridotite xenoliths [48]. We conclude that the continental lithospheric mantle can resolve the Pb paradox only if it contains hitherto unobserved domains with low μ and high Pb concentrations.

Finally, if terrestrial Pb evolution commenced

at ~ 4.45 Ga, as suggested by Stacey and Kramers [43] and Zartman and Haines [39], then the geochron would pass directly through the total crust box shown in Fig. 10 and there would be no Pb paradox.

9. Conclusions

The main conclusions to be drawn from this study are:

(1) The Pb isotopic compositions of many suites of lower crustal xenoliths are radiogenic (plotting to the right of the 4.57 Ga geochron).

(2) Correlations between Pb isotopes and Sr and Nd isotopes for the three suites investigated here suggest that the isotopic compositions of these xenoliths are the result of mixing in the lower crust between a mantle-derived melt and isotopically evolved crust. Because of the high Pb concentration ratios between crust and basaltic melt, the Pb isotopic composition of the crustal end member greatly influences the resulting isotopic composition of the final melt. The radiogenic Pb isotopic characteristics of the mafic xenoliths therefore suggests that the lower crust in these regions had *radiogenic* Pb isotopic compositions.

(3) Mafic xenoliths, which form as cumulates (in which low μ is a primary feature) and have radiogenic Pb isotopic compositions, are relatively young (Paleozoic or younger), even though they may have old continental Nd.

(4) The Pb isotopic characteristics of the lower crust vary according to the tectonothermal age of the crust (time since last orogeny) and are independent of its bulk composition.

(5) The estimated Pb isotopic composition for the total lower crust based on the tectonothermal age and Pb isotopic compositions for granulite terrains and xenoliths, and based on mass balance calculations, is unradiogenic, plotting slightly to the left of the 4.57 Ga geochron. This lower crustal Pb isotopic composition, in combination with that of the upper crust (based on modern sediments and ores), yields a bulk crust Pb isotopic composition which plots significantly to the right of the geochron, suggesting that the lower crust is not the unradiogenic Pb reservoir needed to balance the upper crust and upper mantle.

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