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Inheritance of early Archaean Pb-isotope variability from long-lived Hadean protocrust

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Abstract Comparison of initial Pb-isotope signatures of several early Archaean (3.65–3.82 Ga) lithologies (orthogneisses and metasediments) and minerals (feldspar and galena) documents the existence of substantial isotopic heterogeneity in the early Archaean, particularly in the $^{207}\text{Pb}/^{204}\text{Pb}$ ratio. The magnitude of isotopic variability at 3.82–3.65 Ga requires source separation between 4.3 and 4.1 Ga, depending on the extent of U/Pb fractionation possible in the early Earth. The isotopic heterogeneity could reflect the coexistence of enriched and depleted mantle domains or the separation of a terrestrial protocrust with a $^{238}\text{U}/^{204}\text{Pb}$ (μ) that was ca. 20–30% higher than coeval mantle. We prefer this latter explanation because the high- μ signature is most evident in metasediments (that formed at the Earth's surface). This interpretation is strengthened by the fact that no straightforward mantle model can be constructed for these high- μ lithologies without violating bulk silicate Earth constraints. The Pb-isotope evidence for a long-lived protocrust complements similar Hf-isotope data from the Earth's oldest zircons, which also require an origin from an enriched (low Lu/Hf) environment.

A model is developed in which ≥ 3.8 -Ga tonalite and monzodiorite gneiss precursors (for one of which we provide zircon U-Pb data) are not mantle-derived but formed by remelting or differentiation of ancient (ca. 4.3 Ga) basaltic crust which had evolved with a higher U/Pb ratio than coeval mantle in the absence of the

subduction process. With the initiation of terrestrial subduction at, we propose, ca. 3.75 Ga, most of the ≥ 3.8 -Ga basaltic shell (and its differentiation products) was recycled into the mantle, because of the lack of a stabilising mantle lithosphere. We argue that the key event for preservation of all ≥ 3.8 -Ga terrestrial crust was the intrusion of voluminous granitoids immediately after establishment of global subduction because of complementary creation of a lithospheric keel. Furthermore, we argue that preservation of ≥ 3.8 -Ga material (in situ rocks and zircons) globally is restricted to cratons with a high U/Pb source character (North Atlantic, Slave, Zimbabwe, Yilgarn, and Wyoming), and that the Pb-isotope systematics of these provinces are ultimately explained by reworking of material that was derived from ca. 4.3 Ga (i.e. Hadean) basaltic crust.

Introduction

In a common Pb diagram, initial isotope compositions of terrestrial continental rocks form a broad, concave array that is most clearly defined by samples ca. 2.5 to ca. 1.5 Ga in age (Fig. 1). The wide dispersal in $^{207}\text{Pb}/^{204}\text{Pb}$ at a given $^{206}\text{Pb}/^{204}\text{Pb}$ is *prima facie* evidence that long-lived (> 0.5 -Ga) sources with different $^{238}\text{U}/^{204}\text{Pb}$ (μ) existed in the crust-mantle system. Because of the rapid decay of ^{235}U (only ca. 1% of the original inventory remains today), continued differentiation into crust and mantle reservoirs is now no longer capable of producing a wide range of $^{207}\text{Pb}/^{206}\text{Pb}$ ratios. Rather, the wide $^{207}\text{Pb}/^{206}\text{Pb}$ dispersal presently observed reflects the fact that the mean age of exposed upper continental crust is 2–2.5 Ga, as indicated by Nd-isotopes (e.g. Goldstein and Jacobsen 1987). The large initial Pb-isotope variability is thus inherited from a time when more ^{235}U existed. The post-Archaean Pb-isotope evolution of the crust-mantle ensemble is best understood in terms of recycling of U and Pb from the crust into the mantle (e.g. Kramers and Tolstikhin 1997).

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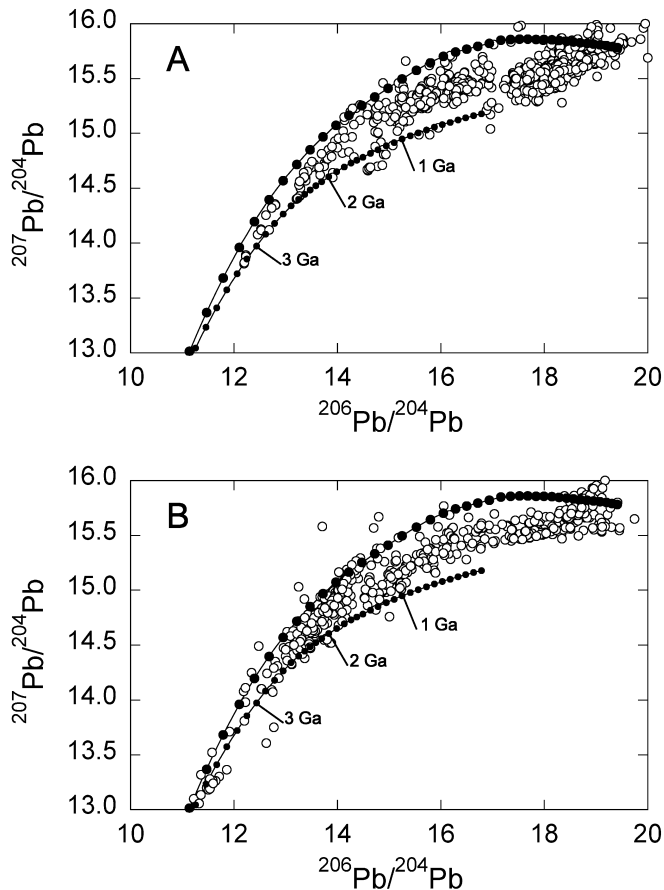


Fig. 1 Pb-isotope diagrams showing initial isotope compositions (*open circles*) recorded in ores (**A**) and leached feldspars (**B**). Data compilation by Kramers and Tolstikhin (1997) based on USGS Pb-isotope database, supplemented for this study with feldspar data from Sultan et al. (1992); Neymark et al. (1994); Hemming et al. (1996,2000); Van Wyck and Johnson (1997); Kamber and Moorbath (1998); Möller et al. (1998); Qiu and McNaughton (1999); De et al. (2000); Bodet and Schärer (2001); Klötzli et al. (2001); and Cheong et al. (2002). The majority of data plot between the 'old upper crust' (*curve connecting large solid circles*) and 'young lower crust' (*curve connecting small solid circles*) evolution curves modelled by Kramers and Tolstikhin (1997)

The chief effects of these processes are to reduce the difference in $^{207}\text{Pb}/^{206}\text{Pb}$ between the mantle and crustal reservoirs, to decrease the modern $^{232}\text{Th}/^{238}\text{U}$ (κ) of the Mid-Ocean-Ridge Basalt (MORB)-source mantle and to gradually increase the upper crustal κ .

At the other end of the time scale, the extent of U/Th/Pb fractionation on the early Archaean Earth is still poorly understood, but evidence has long been published that by 3.7 Ga considerable differences in $^{207}\text{Pb}/^{204}\text{Pb}$ at a given $^{206}\text{Pb}/^{204}\text{Pb}$ already existed between chemical sediments and gneisses of magmatic origin (orthogneisses). Moorbath et al. (1973) observed in the early Archaean rocks from West Greenland that the difference in calculated source μ indicates "different immediate precursors for these two very different rock formations". Since then, additional observations from other Isua Greenstone Belt (IGB) lithologies in West Greenland

have substantiated this early finding (Frei and Rosing 2001; Kamber et al. 2001), indicating that crustal and/or mantle sources remained isolated for extended periods of time during the early Archaean and possibly the Hadean (i.e. > 4.0 Ga ago). The geodynamic significance of source material isolation in the early Earth is difficult to evaluate in the absence of direct constraints on the extent of U/Th/Pb fractionation that could have been generated by early Archaean or Hadean processes. Kamber et al. (2001) showed that the difference in documented ca. 3.7-Ga initial Pb-isotope ratios between orthogneisses and chemical sediments requires sediment source isolation for ca. 0.5 Ga if U/Pb fractionation was similar to that presently observed within the crust. Alternatively, if U/Pb fractionation was more extreme, the relatively large isotopic difference could be generated within as little as 100 Ma. The latter possibility needs to be considered because many lunar samples appear to have been derived from sources with exceedingly variable but generally high U/Pb (Nyquist and Shih 1992).

Here we report an extensive set of new Pb-isotope data (whole rock and feldspar) for ca. 3.8-Ga tonalitic gneisses and amphibolites from the North Atlantic Craton, as well as a compilation of previously published and unpublished Pb-isotope data for a variety of lithologies, including Pb ores, of the 3.7- to 3.8-Ga IGB. These data define an unexpectedly large variation in interpreted initial isotope ratios, providing incontrovertible evidence that strong U/Pb fractionation occurred perhaps as far back as the Hadean. We explore whether the isotope array requires a multitude of source histories or whether an evolution model can be derived which operates with only one depleted and one enriched end-member. Finally, we discuss implications for terrestrial Pb-isotope models, which aim at explaining the present-day isotope composition of silicate Earth reservoirs (plumbotectonics) and argue that the early Archaean record implies existence of a long-lived Hadean protocrust.

Sample strategy in relation to field geology and previous studies

The Archaean Itsaq gneiss terrain in western Greenland is the largest, best-preserved, and most widely studied expanse of early Archaean rocks known on Earth. A smaller equivalent of these gneisses is exposed in the Hebron-Saglek segment of the Archaean gneisses in northern Labrador. Both areas form part of the North Atlantic Craton, which was contiguous prior to the opening of the Labrador Sea. These old gneiss complexes are predominantly composed of trondhjemitic, tonalitic and granodioritic (TTG) gneisses in which enclaves of supracrustal rocks (both metasediments and metavolcanic rocks) and older orthogneisses are found. In West Greenland, a special opportunity exists to study a large, well-preserved sequence of supracrustal rocks in the IGB, which contains a large variety of lithologies.

The best evidence for genuine, relatively undisturbed, 3.8-Ga continental crust has recently been published by Nutman et al. (1999) who described the geochronology of a gneiss complex south of the IGB. This complex (Fig. 2) has not been formally named, although it forms part of the regional Itsaq gneiss complex (Nutman et al. 1996, 1999). For the sake of clarity we here refer informally to this gneiss terrain as SOI (south of Isua) to distinguish it from other gneiss terrains in the region as a whole, and we refer to the studied rocks as SOI gneisses, SOI amphibolites, etc. In an attempt to substantiate claims for genuine 3.8-Ga magmatic precursors to these gneisses we present new ion-probe U-Pb zircon dates, although the main aim of our analytical effort was to reconstruct the source history of these and similar gneisses from northern Labrador using Pb-isotope data of separated feldspar. Below we describe in more detail the geological context of the studied samples.

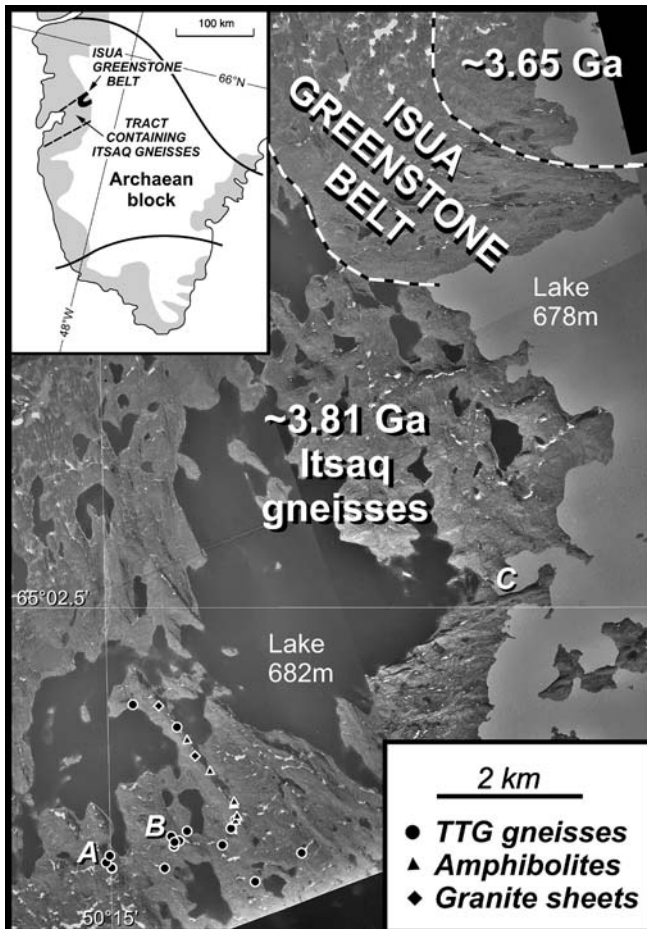


Fig. 2 Mosaic of aerial photographs of the low strain, ca. 3.81-Ga Itsaq gneiss terrain immediately SW of the Isua Greenstone Belt (IGB) which is delineated by dashed lines. Inset shows location of the IGB on a map of southern Greenland. Sample localities referred to in this paper are indicated, together with localities A, B and C of Nutman et al. (1999)

Greenland

In this paper, Pb-isotope data are presented and discussed for three principal early Archaean rock sequences from within the so-called Itsaq gneiss complex (Nutman et al. 1996) of southern West Greenland. These are (1) calc-alkaline (TTG) orthogneisses, still widely referred to as Amitsoq gneisses, from the Godthaabsfjord region along, and adjacent to, the coastal province south and east of Nuuk; (2) calc-alkaline (TTG) orthogneisses from the SOI area immediately south of the IGB, which has only recently begun to be investigated in earnest. Significantly, these gneisses show both similarities and differences to the Godthaabsfjord gneisses and also to the orthogneisses to the north of (within) the arcuate IGB; and (3) the IGB itself, some 150 km north-east of Nuuk, comprising metamorphosed volcanic and sedimentary lithologies some of which host minor sulphide mineralisation, including galena with the least radiogenic terrestrial Pb so far known.

Godthaabsfjord region gneisses

Since the early 1970s, much has been published about many aspects of these orthogneisses, of which their geochronology is the most relevant here. There is general agreement for a major period of emplacement of their magmatic precursors at ca. 3.65 Ga (e.g. Kamber and Moorbath 2000; McGregor 2000). However, debate continues on whether U-Pb zircon dates extending up to 3.90 Ga (Nutman et al. 1996, 2000) represent the magmatic protolith age (e.g. Nutman et al. 2001), or represent zircons inherited from older source regions (Whitehouse et al. 1999, 2001). In the strongly banded TTG gneisses, the additional possibility exists that genetically unrelated igneous rocks, ranging in age from 3.9 to 3.6 Ga, could have been intercalated as narrow bands.

The Godthaabsfjord region gneisses have long been known to contain the least radiogenic terrestrial silicate leads so far discovered (e.g. Black et al. 1971; Gancarz and Wasserburg 1977). Building on all previous work, combined with new analyses, Kamber and Moorbath (1998) tabulated and regressed the available Pb-isotope data from the Godthaabsfjord gneisses and concluded that all analysed whole rock and feldspar samples pointed to an age of ca. 3.65 Ga, even for some which contained zircons with significantly older portions. The Pb-isotope data for whole rocks and feldspars used in the figures and elsewhere are the ones listed in Table 1 of Kamber and Moorbath (1998) and represent some of the most typical gneiss units in the Godthaabsfjord region.

South-of-Isua (SOI) gneisses

This is an extensive terrain (Fig. 2) of varied, amphibolite-facies, TTG-type gneiss with numerous, large (up to several hundred meters) enclaves of older ultramafic rocks, metagabbros, layered amphibolites (\pm garnet) with interlayers of metamorphosed BIF and thin felsic

Table 1 Pb-isotope data compilation of Isua Greenstone Belt (IGB) lithologies and galenas. Numbers in columns entitled "Source" refer to analysts or data source: 1 previously unpublished Oxford data by Roy Goodwin, Stephen Moorbath and Paul Taylor; 2 data from Moorbath et al. (1975); 3 previously unpublished Oxford data by Roy Goodwin, Stephen Moorbath and Martin Whitehouse; 4 previously unpublished Brisbane TIMS data by Balz Kamber and

Peter Appel; 5 data from Richards and Appel (1987); 6 data from Appel et al. (1978); 7 data from Frei and Rosing (2001). All previously unpublished data are normalised relative to recommended NBS SRM 981 ratios of Todt et al. (1996). Accuracy (2 sigma) of the three Pb-isotope ratios is better than 0.25, 0.30 and 0.42% in the Oxford data set, and better than 0.17, 0.23 and 0.31% in the Brisbane data set

Sample	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	Source	Sample	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	Source
3.81-Ga felsic "volcanogenicmetasediment boulders"					3.71-Ga garnet-mica schists(eastern IGB) continued				
248412	16.236	14.803	37.700	1	SM/GR/93/58	16.742	15.156	37.076	3
248413	12.285	13.620	32.501	1	SM/GR/93/10	12.796	13.730	32.522	3
248414	11.842	13.420	32.001	1	SM/GR/93/11	12.262	13.619	31.838	3
248415	12.133	13.477	32.233	1	SM/GR/93/13	12.315	13.625	31.935	3
248416	13.169	13.838	33.356	1	SM/GR/93/15	13.601	13.913	33.169	3
248418	14.612	14.285	35.115	1	SM/GR/93/22	12.178	13.395	32.158	3
248419	14.121	14.186	34.412	1	SM/GR/93/28	12.187	13.609	31.853	3
248426	14.510	14.288	36.054	1	SM/GR/93/33	12.031	13.466	31.914	3
248431	13.666	13.993	33.898	1	SM/GR/93/75	12.680	13.761	32.528	3
248432	14.129	14.187	33.359	1	SM/GR/93/76	13.719	14.056	34.007	3
158493	13.109	13.671	33.550	2	"Tourmaline boulder" in garnet-micaschists				
158536	12.301	13.536	32.534	2	248483B	14.161	14.250	33.835	1
158537	12.238	13.541	31.941	2	248483E	13.963	14.213	33.485	1
158538	12.146	13.536	32.476	2	248483F	14.302	14.325	34.067	1
SM/GR/93/9	13.529	13.844	33.801	3	248483G	14.583	14.323	34.076	1
SM/GR/93/21	12.495	13.550	33.211	3	248483H	14.413	14.299	34.624	1
Matrix of 3.81-Ga"volcanogenic metased. boulders"					248483J	14.702	14.385	34.537	1
248421	14.696	14.353	35.980	1	Galenas (unpublished data)				
248422	14.344	14.168	35.102	1	SM/GR/93/18A	11.483	13.372	31.445	1
248423	14.278	14.130	35.160	1	SM/GR/93/18B	11.440	13.368	31.473	1
248424	13.045	13.694	33.270	1	SM/GR/93/18C	11.475	13.358	31.408	1
248425	12.864	13.627	33.308	1	215720	11.103	13.014	31.366	3
248428	13.201	13.840	33.636	1	215730	11.086	12.984	31.281	3
248429	12.560	13.617	32.842	1	HKS 450168A	11.034	12.898	31.035	3
248430	12.456	13.546	32.704	1	HKS 450168B	11.025	12.891	30.990	3
248433	14.046	14.133	34.461	1	HKS 450168C	11.022	12.882	30.978	3
3.71-Ga felsic "volcanogenicmetasediments"					HKS 450168D	11.024	12.917	31.093	4
248481A	24.526	17.582	46.121	1	HKS 462749	11.023	12.886	31.003	4
248481B	17.499	15.295	38.331	1	HKS 462750	11.039	12.913	31.095	4
248481C	23.033	17.071	44.546	1	HKS 462751	11.024	12.888	31.006	4
248481D	15.232	14.523	35.670	1	HKS 462752	11.029	12.899	31.042	4
248481E	15.965	14.766	36.435	1	HKS 462753	11.031	12.895	31.019	4
248481F	16.773	15.180	37.018	1	HKS 462759	11.121	12.984	31.072	4
248481G	16.958	15.224	37.431	1	HKS 462762	11.135	12.998	31.113	4
248481H	19.058	15.960	39.917	1	HKS 462768	11.141	13.019	31.184	4
248481J	15.708	14.847	35.867	1	HKS 462776	12.228	13.767	32.065	4
248481K	17.260	15.439	37.362	1	HKS 462754	12.478	13.707	32.070	4
248481L	20.062	16.170	40.978	1	HKS 467256	12.200	13.663	31.823	4
248481M	22.266	16.882	43.570	1	HKS 462757	12.136	13.637	31.756	4
SM/GR/93/59	14.916	14.416	35.477	3	Galenas (published data)				
3.71-Ga garnet-mica schists(single locality)					539	11.176	13.047	31.130	5
248484A	17.400	15.442	38.178	1	540	11.311	13.147	31.265	5
248484B	16.927	15.352	37.773	1	541	11.195	13.062	31.171	5
248484C	17.122	15.461	38.055	1	542	11.266	13.110	31.198	5
248484D	17.691	15.535	38.640	1	542	11.272	13.121	31.225	5
248484E	16.499	15.187	37.081	1	543	11.275	13.124	31.230	5
248484F	15.404	14.788	35.293	1	2946	11.434	13.345	31.424	6
248484G	17.886	15.615	39.088	1	2954	11.428	13.335	31.383	6
248484H	16.146	15.055	36.094	1	2926	11.314	13.158	31.301	6
248484J	15.864	14.964	36.202	1	2926	11.326	13.175	31.335	6
248484K	17.569	15.518	38.039	1	2966	11.146	13.025	31.148	6
248484L	14.536	14.439	34.782	1	2966	11.151	13.037	31.179	6
3.71-Ga garnet-mica schists(eastern IGB)					2966	11.164	13.052	31.216	6
SM/GR/93/56	16.072	15.130	36.992	3	460000-1	11.022	12.887	30.987	7
SM/GR/93/57	17.719	15.423	38.664	3	940001	11.131	13.004	31.126	7

bands of unknown origin. The entire assemblage is traversed by mostly discordant, amphibolite-facies, mafic dykes, regarded as equivalent to the mid-Archaean

Ameralik dykes elsewhere in the Itsaq gneiss complex. SOI TTG gneisses range from strongly banded (i.e. typical grey gneiss) to homogeneous varieties. The homo-

geneous gneisses are equigranular, but no *bona fide* igneous textural features are preserved, indicating that amphibolite-facies recrystallisation occurred under low strain. There is probably a greater proportion of homogeneous gneisses here than anywhere else in the Itsaq gneiss complex, as already noted by Nutman et al. (1999). Younger pre- and post-dyke granitoid sheets and pegmatites are locally present. There is evidence for pre- and post-dyke metamorphism and deformation.

Nutman et al. (1996, 1999) have convincingly shown that oscillatory-zoned prismatic grains with simple age structures dominate zircon populations from SOI tonalites and quartz-diorites, yielding single-population dates of mainly 3818 ± 8 Ma. This is in strong contrast to the geochronologically and structurally much more complex zircon populations of the Godthaabsfjord region, as well as the well-preserved, low-strain tonalite gneisses north of the IGB (Fig. 2), which have only yielded zircon U-Pb dates of 3.65–3.70 Ga, and not 3.8 Ga (Nutman et al. 1996). Zircon U-Pb ages from SOI gneisses thus yield the best evidence so far for the existence of in-situ ≥ 3.8 -Ga-old granitoid crust in the Itsaq gneiss complex.

Samples for further zircon U-Pb work (presented in this study) and for a comprehensive Pb-isotope survey (the main purpose of this paper) were collected by S. Moorbath and M.J. Whitehouse. We have closely followed the paper by Nutman et al. (1999) and concentrated sampling efforts on the outcrops either at, or surrounding (within ca. 2 km), their localities, where there are numerous fine exposures of homogeneous tonalitic gneisses, with or without biotite foliation, traversed by occasional discordant Ameralik dykes. The geology of the relevant localities (marked A and B) is discussed in detail by Nutman et al. (1999).

Seven samples of enclaves of layered amphibolites and garnet-amphibolites analysed for Pb-isotopes also come from outcrops within 1–2 km of localities A and B, whilst samples 409019 and 409030 consist of thin (ca. 20-cm), concordant, fine-grained granitoid sheets interlayered in the amphibolites.

Isua Greenstone Belt (IGB)

The oldest known, early Archaean sedimentary and volcanic rocks, as well as their mostly intense (amphibolite facies) metamorphism and deformation, have been closely studied for some 30 years. Opinions regarding the protoliths and the tectonic structure of the IGB are still evolving, as can be seen by comparing and contrasting, for example, the views of Nutman et al. (1984) with those of Fedo et al. (2001) and Myers (2001). There is also ongoing debate regarding the absolute ages of the various lithologies, although most investigators (ourselves included) agree that direct U-Pb zircon evidence by Nutman et al. (1997) points to a bimodal age distribution, with the majority of dated lithologies clustering tightly around 3.71 Ga in age, and a minority of directly dated lithologies extending to an age of 3.81 Ga. A model age of 3.81 Ga has also been claimed to explain the least radiogenic

Pb-isotope ratios of IGB galena (Frei and Rosing 2001). There is also evidence (from zircons in possible metasediments; Nutman et al. 1997) for the (former) existence of even older rocks of up to 3.9 Ga.

The Pb-isotope data for the IGB, which are tabulated (Table 1) and plotted in this paper, are based on representative (metamorphic) lithologies from the eastern sector of the belt, collected by S. Moorbath and P.N. Taylor. Within the framework of Nutman et al.'s (1997) chronology, our selection of rocks includes representatives of both the 3.81- and the 3.71-Ga age group: BIF; carbonates; metapelitic garnet-mica (\pm staurolite, \pm amphibole) schists probably derived from mafic, volcanogenic sediments; a tourmaline-bearing quartzo-feldspathic fragment enclosed within the metapelites; quartzo-feldspathic schists, probably derived from felsic, volcanogenic sediments; and interlayered quartzo-feldspathic rocks possibly derived from highly tectonised, metasomatised granitoid rocks (see Myers 2001). Kamber et al. (1998) obtained a whole-rock Sm-Nd regression age of 3742 ± 49 Ma for the garnet-mica schist suite, which agrees within error with the 3.71-Ga U-Pb zircon ages obtained for metasedimentary lithologies by Nutman et al. (1997).

Galena is found as small disseminations, patches and veins, together with other sulphides, within different lithologies of the IGB. The sulphide occurrences are not primary features of the lithologies but have formed (or were remobilised) during metamorphic events (Frei et al. 1999). All samples, except one, for which we present new data were collected by geologists of the Nunaoil Company and by Peter Appel of the Geological Survey of Denmark and Greenland. The exception is SM/GR/93/18, collected by S. Moorbath, which occurs as a scattered dissemination of galena in recrystallised chert at a locality in the eastern sector of the IGB, famous for the occurrence of ornamental fuchsite-quartzite. The uniquely (i.e. terrestrial) unradiogenic character of Pb in several IGB galenas was discovered by Appel et al. (1978), and further investigated by Richards and Appel (1987) and Frei and Rosing (2001).

Labrador

Sample selection for the northern Labrador segment of the North Atlantic craton was based on relative chronology established in exposures of low-strain gneiss domains. Two different groups of gneisses were chosen for Pb-isotope analysis (whole rocks and feldspar separates). First, the Uivak I (hereafter simply called Uivak) TTG gneisses dominate the early Archaean Gneiss Complex in the Saglek-Hebron area (Collerson 1983a; Collerson et al. 1989; Schiøtte et al. 1989). These gneisses are considered broadly similar in age and character to the TTG gneisses of the Godthaabsfjord area in Greenland (Collerson et al. 1989). The best U-Pb igneous zircon age estimate for the Uivak gneisses is 3732 ± 6 Ma (Collerson 1983a; Schiøtte et al. 1989). Like

many Godthaabsfjord TTG gneisses, the euhedral, zoned, igneous Uivak zircons sometimes contain rounded cores of up to $3,863 \pm 12$ Ma, which were interpreted as inherited from pre-existing evolved rocks or from the melt source (Schjøtte et al. 1989). Although complicated by later metamorphic events, whole-rock Rb-Sr, Sm-Nd and Pb/Pb regression dates are compatible with the 3.73-Ga age estimate and indicate derivation from a slightly depleted (relative to chondrite) mantle source (Collerson 1983a). There is only minimal difference in radiogenic isotope systematics between undisturbed Uivak and Godthaabsfjord gneisses.

The second set of gneisses, termed Nanok gneisses, is of different character and of distinctive iron-rich monzodioritic composition. Along the southern shore of Tigi-gakyut Inlet, between Hebron and Saglek, Nanok gneisses occur as easily recognisable enclaves in relatively undeformed Uivak gneiss. One specimen of Nanok gneiss (from Big Island, Saglek Bay) has yielded zircon U-Pb dates of 3.92 and 3.81 to ca. 3.7 Ga (Collerson 1983a). Based on the relative chronology (i.e. the observation that the Nanok gneisses predate Uivak gneisses), the ca. 3.7-Ga date was interpreted as the time of incorporation of enclaves into Uivak magma. It is presently unresolved which of the older two dates (3.81 or 3.92 Ga) corresponds to the igneous emplacement age. Regardless, for the purpose of the present paper it is sufficient to note that Nanok gneisses predate the Uivak gneisses by at least 80 if not 190 Ma.

Collerson (1983b) used secondary Rb-Sr isochrons and mineral ages to reconstruct the polymetamorphic history of the Uivak gneisses. Sr-isotope homogenisation events, on the mineral and compositional layer scale (for banded gneisses), were recorded at 2.8, 2.5 and 1.8 Ga. The oldest of these homogenisation events is also evident from U-Pb systematics of rims that developed on igneous zircons (Collerson 1983a; Schjøtte et al. 1989).

New U-Pb ion probe dates for SOI TTG gneiss

One sample of a typical, homogeneous SOI TTG gneiss (SM/GR/98/2) was collected during a reconnaissance visit in 1998 from a locality in the northern SOI gneiss terrain, ca. 4 km south of the IGB. We regard this sample as equivalent to Nutman et al.'s (1996) quartz-diorite sample 292128.

SIMS U-Th-Pb analyses were performed using a Cameca IMS1270 ion-microprobe located at the Nord-SIMS facility in Stockholm. Analytical details are given in the caption to Table 2. Results are reported in Table 2 and Fig. 3. Cathodoluminescence images of zircons from SM/GR/98/2 reveal relatively simple internal structures with concentric fine oscillatory zoning generally considered typical of magmatic growth (Fig. 3). This observation is in accord with that of Nutman et al. (1999) for zircons from their SOI terrain samples. In Fig. 3, $^{207}\text{Pb}/^{206}\text{Pb}$ ages exhibit a small spread along concordia

with the highest ages > 3.8 Ga. Given the simplicity of internal structure revealed by the CL images, and the differences in replicate analyses on the same growth phase (Table 2), we consider that $^{207}\text{Pb}/^{206}\text{Pb}$ ages < 3.8 Ga most likely represent ancient Pb-loss. The five grains with highest ages yield a weighted average $^{207}\text{Pb}/^{206}\text{Pb}$ age of 3813 ± 10 Ma (2σ , MSWD = 2.9). These same five grains also yield a concordia age (Ludwig 1999) of $3,812 \pm 14$ Ma (MSWD of concordance = 4.5; decay constant errors included). These ages, however calculated, are within error of ages reported from the SOI terrain by Nutman et al. (1996, 1999) and we agree with the conclusion of these authors that this represents the age of the magmatic protolith to these rocks. In conclusion, our new zircon data agree with the observations of Nutman et al. (1999) of relatively simple, igneous internal structure of SOI TTG gneiss zircons, which is reflected in comparatively (to the Godthaabsfjord TTG gneisses) uniform age distribution clustering between 3.81 and 3.82 Ga. This age is regarded as the most likely time of crystallisation of the gneiss precursors.

Pb-isotope results for TTG gneisses

Pb-isotope data for the SOI gneisses are presented in Table 3 and were obtained by multi-collector ICP-MS at the ACQUIRE Laboratory, University of Queensland. Analytical details are given in the table legend and followed closely those of Collerson et al. (2002). Pb-isotope data of the Labrador samples are listed in Table 4 and were obtained by TIMS in the Department of Earth Sciences of the University of California at Santa Cruz. Analytical details are given in the table legend and closely followed those of Palacz (1985).

Whole rocks

Whole-rock Pb-isotope data for SOI gneisses are plotted according to field-derived relative chronology (Fig. 4A). The two granitoid sheets that occur structurally concordant within amphibolites contain the most radiogenic Pb and, because of their unknown absolute age, will not be considered further. The whole-rock data of the tonalite gneisses range from very unradiogenic to typical crustal values, but on average, they are significantly more radiogenic than 3.65-Ga Amîtsoq-type gneisses from the Godthaabsfjord region (Kamber and Moorbath 1998). Furthermore, there is significant scatter such that the apparent regression age of 3,706 Ma has a large uncertainty of 190 Ma (2 sigma). Note that the high MSWD of 31,782 obtained from the MC-ICP-MS data (using external precision rather than the smaller internal error) cannot be compared with that for data obtained by conventional (unspiked) thermal ionisation mass spectrometry, which would yield an MSWD at least 100 times lower for the same degree of scatter. While the regression date agrees within error with the U-Pb zircon age range of

Table 2 Ion-microprobe U-Th-Pb data for zircons from sample SM/GR/98/2. Analytical parameters are identical to those reported by Whitehouse et al. (1999) and references therein. Errors on $^{207}\text{Pb}/^{206}\text{Pb}$ ratios are the raw counting statistic errors; Pb/U ratios

are calibrated relative to the 1,065-Ma Geostandards zircon 91500 (Wiedenbeck et al. 1995) and contain a propagated (and dominant) error component from the pooled standard analyses (ca. 2.5% 1 sigma for this analytical session)

Analysis ID	[U] (ppm)	[Pb] (ppm)	Th/U ^a	f_{206}^b (%)	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{206}\text{Pb}/^{238}\text{U}$	$^{207}\text{Pb}/^{206}\text{Pb}$ age (Ma)	$^{206}\text{Pb}/^{238}\text{U}$ age (Ma)
2b	190	231	0.63	0.07	26,116	0.3630 ± 0.0011	0.7951 ± 0.0186	$3,763 \pm 5$	$3,774 \pm 67$
2a	292	337	0.33	0.04	49,456	0.3684 ± 0.0008	0.7917 ± 0.0180	$3,785 \pm 4$	$3,761 \pm 65$
5b	128	150	0.48	0.70	2,668	0.3696 ± 0.0020	0.7937 ± 0.0181	$3,790 \pm 9$	$3,787 \pm 66$
1d	72	92	0.77	(0.05)	34,025	0.3704 ± 0.0013	0.8083 ± 0.0184	$3,793 \pm 5$	$3,819 \pm 66$
1c	142	175	0.60	0.08	23,079	0.3734 ± 0.0009	0.7986 ± 0.0182	$3,805 \pm 4$	$3,786 \pm 66$
5a	260	293	0.10	0.03	53,967	0.3745 ± 0.0010	0.8006 ± 0.0182	$3,810 \pm 4$	$3,793 \pm 66$
6a	85	111	1.01	(0.03)	58,651	0.3757 ± 0.0014	0.7896 ± 0.0179	$3,815 \pm 6$	$3,752 \pm 65$
3a	112	138	0.76	0.54	3,466	0.3758 ± 0.0016	0.7760 ± 0.0182	$3,815 \pm 6$	$3,718 \pm 67$
6c	147	193	1.06	0.15	12,793	0.3779 ± 0.0009	0.7801 ± 0.0179	$3,824 \pm 4$	$3,722 \pm 66$

^aCalculated from measured ThO intensity.

^bPercentage of common Pb detected, calculated from measured ^{204}Pb assuming 0-Ma Stacey and Kramers (1975) model. Common Pb correction has not been applied for samples for which this number is in parentheses due to low ^{204}Pb counts.

3.80–3.82 Ga, it is obvious that the scatter about the regression reflects either substantial initial isotope heterogeneity or disturbance of the U/Pb ratios. We will argue later that these rocks were affected by late Archaean and early Proterozoic regional open-system events that changed their U/Pb ratios and disturbed their isochronous Pb-isotope systematics. Nevertheless, the least radiogenic datapoints (as well as the intercept of the regression line with mantle evolution models) are still difficult to reconcile with a 3.80- to 3.82-Ga mantle origin, because the data plot well above, and to the right of, such an old model age. A forced regression to 3.81 Ga yields much too young an intersection with the Kramers and Tolstikhin (1997) mantle evolution curve at 3,685 Ma. Pb-isotope ratios of the amphibolite gneisses do not plot substantially off the tonalite regression line (Fig. 4Aa), but have themselves insufficient spread for meaningful regression.

Similarly, the whole-rock data for the Labrador gneisses are not well aligned (Fig. 4B), but plot roughly along a similar trend to the SOI data. It is clear that the whole-rock data on their own cannot be used to evaluate claims for 3.80- to 3.82-Ga (SOI), 3.81- to 3.92-Ga (Nanok) and 3.73-Ga (Uivak) emplacement ages, nor can the data be used to infer probable source histories with any reasonable confidence.

Feldspars

By comparison with the whole rocks, the feldspar Pb-isotope data are much better aligned in common Pb space. Pb-isotope ratios of (leached) feldspars are significantly less radiogenic than those of the whole rocks (Fig. 5). Data for the four main groups of rocks yield reasonably tight regression lines, which are readily identified as sec-

Fig. 3 Inverse (Tera-Wasserburg) concordia diagram for SIMS analyses from SM/GR/98/2 (Table 2). Cumulative probability diagram of $^{207}\text{Pb}/^{206}\text{Pb}$ ratios is shown at right-hand side with the secondary y-axis indicating age. Shaded ellipse shows the concordia age of $3,812 \pm 14$ Ma (95% confidence) obtained from the five samples with the highest $^{207}\text{Pb}/^{206}\text{Pb}$ ratios. Inset cathodoluminescence images are shown for two typical zircon grains from this sample

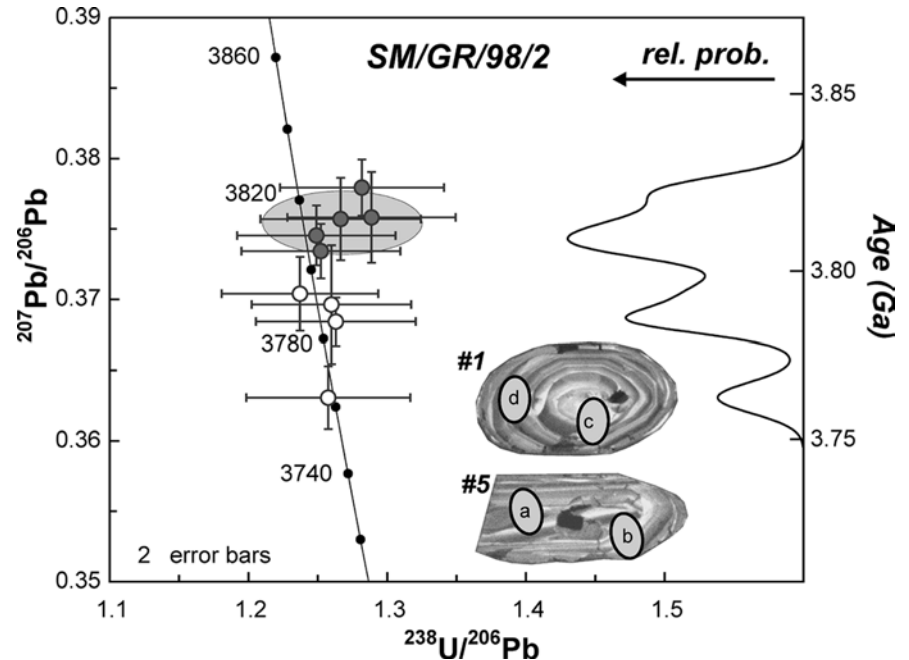


Table 3 Pb-isotope data for gneisses from SOI terrain. GPS localities use the WGS84 datum. Sample numbers (4090xx series) refer to the archives of the Geological Survey of Denmark and Greenland (GEUS). Letters in parentheses after the coordinates refer to localities studied by Nutman et al. (1999) and use the same designation. All samples collected by S. Moorbath and M.J. Whitehouse. Feldspar (mainly plagioclase) mineral separates were obtained by magnetic and density separation and were washed three times in ultrapure water in an ultrasonic agitator. They were first leached for 24 h in 2 N HCl on a 100 °C hotplate and then for 20 min in cold 14 N HF. Before final dissolution, the leached mineral separate was cleaned three times in ultrapure water.

Reproducibility of leaching procedure was tested with sample 409010 and was found to be excellent. Total procedural blank remained below 22 pg and was insignificant. Multi-collector ICPMS mass spectrometric procedures followed closely those of Collerson et al. (2002). Internal precision on all three ratios of all samples was 3–8 times better than two-sigma external reproducibility of NBS SRM 981 ($n=114$), which yielded $^{206}\text{Pb}/^{204}\text{Pb}$ of 16.941 (± 374 ppm), $^{207}\text{Pb}/^{204}\text{Pb}$ of 15.495 (± 490 ppm) and $^{208}\text{Pb}/^{204}\text{Pb}$ of 36.720 (± 634 ppm). For regression calculations, data points were assigned external errors which were always larger than internal precision

Sample	Location (lat. N, long. W)	Whole rocks			Feldspars		
		$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$
TTG gneisses							
SM/GR/98/2	65°02.70', 50°08.36' (C)	12.010	13.308	34.957	11.328	13.100	31.449
409001	65°00.761', 50°13.625'(B)	12.058	13.363	32.206	11.507	13.224	31.323
409002	65°00.761', 50°13.625'(B)	12.863	13.747	32.606	11.955	13.519	31.484
409003	65°00.761', 50°13.625'(B)	16.362	14.567	37.550	12.121	13.579	31.760
409004	65°00.64', 50°14.89'(A)	12.966	13.723	32.366	11.939	13.478	31.437
409005	65°00.64', 50°14.89'(A)	13.216	13.753	32.150	11.891	13.417	31.358
409006	65°00.64', 50°14.89'(A)	12.498	13.549	32.030	11.866	13.394	31.432
409007	65°00.64', 50°14.89'(A)	14.239	14.040	32.946	12.249	13.617	31.445
409008	65°00.64', 50°14.89'(A)	11.848	13.303	32.445	11.479	13.198	31.356
409010	65°00.64', 50°14.89'(A)	14.220	13.980	32.042	12.361	13.635	31.485
409010		Separately leached aliquot			12.387	13.638	31.478
409011	65°00.80', 50°13.75'(B)	13.752	13.996	31.825	12.188	13.639	31.332
409014	65°00.83', 50°13.48'	12.547	13.713	32.068	11.963	13.564	31.758
409015	65°00.83', 50°13.48'	12.759	13.614	33.600	11.739	13.342	31.582
409016	65°00.884', 50°12.644'	15.118	14.640	33.351	13.197	14.248	32.282
409021	65°00.754', 50°13.703'	15.944	14.758	36.212	13.282	14.219	32.718
409032	65°00.40', 50°12.40'	15.449	14.853	35.656	13.536	14.403	32.196
409033	65°00.70', 50°12.90'	16.259	14.939	34.600	13.344	14.337	31.969
409034	65°01.63', 50°13.50'	13.137	13.700	33.787	11.896	13.399	31.702
409035	65°01.83', 50°14.21'	12.195	13.448	33.015	11.617	13.275	31.500
409037	65°00.827', 50°12.718'	13.584	14.007	33.550	12.182	13.658	31.724
409038	65°00.820', 50°12.729'	14.948	14.412	35.540	12.797	13.926	32.215
409040	65°00.56', 50°13.91'	12.391	13.594	32.538	11.851	13.459	31.560
409042	65°00.691', 50°14.821'	12.445	13.737	32.244	12.037	13.624	31.715
Amphibolites							
409017	65°00.884', 50°12.644'	12.144	13.606	31.846	11.917	13.544	31.582
409018	65°00.92', 50°12.61'	12.137	13.507	31.976	11.794	13.449	31.618
409022	65°01.532', 50°13.343'	12.484	13.735	32.292	12.047	13.583	31.861
409023	65°01.532', 50°13.343'	13.682	13.864	34.088	12.224	13.575	32.395
409025	65°01.01', 50°12.65'	13.488	14.026	33.241	12.438	13.806	32.082
409026	65°01.03', 50°12.64'	13.337	13.864	34.094	18.608	15.597	32.813
409026					16.564	15.033	33.302
409031	65°01.28', 50°13.00'	12.030	13.305	31.565	11.477	13.214	31.382
Concordant granitoid sheets in amphibolites							
409019	65°01.80', 50°13.77'	16.367	14.908	37.323	13.088	14.113	32.448
409030	65°01.40', 50°13.23'	17.419	15.000	37.595	13.165	14.066	32.030

ondary isochrons by the fact that their slopes define apparent ages (of no geological significance whatsoever, but see Tera 2000) between 4.24 and 4.48 Ga well in excess of plausible intrusion ages. Rather, the slope of these secondary isochrons is determined by the relative timing of rock formation and feldspar recrystallisation (a process explained in detail by Rosholt et al. 1973).

Secondary feldspar-isochrons are a feature of the combined ^{235}U – ^{207}Pb and ^{238}U – ^{206}Pb systems, and occur because of the ability of feldspar to recrystallise with the Pb-isotopic composition of its host whole rock but without incorporating U, thus creating a snapshot of a particular point in Pb-isotopic evolution. If feldspars

record the isotopic composition of a set of consanguineous, isochronous host rocks at some time between rock formation and today, like in our examples, the slope defined in common Pb-isotope space yields an apparent age much in excess of the rock formation age. For a comprehensive discussion of secondary feldspar-isochrons the reader is referred to the excellent treatment of this subject by Rosholt et al. (1973; pp. 989–991).

In the case of our SOI tonalite gneiss feldspar regression line (which will henceforth be called a secondary 'isochron', where the inverted commas indicate that statistically these regressions are errorochrons), the slope is 0.585, which, if erroneously interpreted as an age,

Table 4 Pb-isotope data for Uivak and Nanok gneisses. These data were obtained by K.D.C. at the University of California (Santa Cruz). We retrospectively normalised the ratios to the recommended NBSSRM 981 values of Todt et al. (1996). Internal precision of all measurements was better than the standard

Sample	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	Sample	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$
3.73-Ga Uivak gneiss whole rocks				Nanok gneiss whole rocks continued			
KC-78-629A	14.075	14.420	34.624	KC-91-34A	17.312	14.889	40.251
KC-78-623C	13.793	14.009	35.374	KC-91-34B	14.933	14.424	38.831
KC-78-627B	13.408	14.079	38.547	KC-91-35A	15.516	14.690	36.589
3.73-Ga Uivak gneiss feldspars				KC-91-35B	14.650	14.413	35.634
KC-78-630A	13.017	14.190	34.397	KC-91-35H	13.672	14.224	34.664
Duplicate	13.241	14.290	34.672	KC-91-48B	15.037	14.411	37.094
KC-78-620D	12.554	13.832	32.653	KC-91-56B	13.800	14.278	34.766
KC-78-629A	13.323	14.313	33.832	KC-91-56G	13.500	14.147	34.200
KC-78-623C	12.335	13.722	32.471	Nanok gneiss feldspars			
Duplicate	12.478	13.791	32.799	KC-91-25A	14.152	14.727	32.819
KC-78-627B	12.519	13.840	32.885	Duplicate	14.213	14.711	32.821
Duplicate	12.718	13.992	33.222	KC-91-33A	12.812	13.999	32.753
KC-78-633D	12.878	13.998	33.287	Duplicate	12.922	14.056	33.181
Nanok gneiss whole rocks				KC-91-35B	12.867	14.077	32.678
KC-91-25A	15.215	14.440	40.287	Duplicate	12.902	14.109	32.891
KC-91-33A	15.154	14.393	40.220	KC-91-56G	12.673	13.971	32.258
KC-91-33C	15.076	14.452	37.044	Duplicate	12.716	14.000	32.513

reproducibility at Santa Cruz (that is, 0.16, 0.24 and 0.41%). In contrast to the new SOI gneiss feldspars, these samples were only leached in weak nitric acid, resulting in less-reproducible duplicates

would yield $4,480 \pm 77$ Ma (MSWD = 6,746). In the simplest case, a secondary feldspar-isochron is formed during a single (metamorphic) Pb-isotope homogenisation event at (t_h) from a suite of consanguineous samples. Provided that the magmatic age (t_m) of the suite is known, t_h can be calculated from the slope of the isochron. The combinations of rock formation ages and feldspar recrystallisation times that yield a slope of 0.585 are illustrated in Fig. 6A. Because all the SOI gneisses for which zircon has been studied in detail have a simple U-Pb age distribution and because the zircons have a simple structure, we assume that the magmatic protoliths formed at 3.82 Ga. The SOI TTG secondary feldspar-isochron then constrains the age of feldspar crystallisation to 1.95 Ga (Fig. 6A). The secondary feldspar-isochron minimum and maximum ages of 4,394 and 4,560 Ma translate into homogenisation at 1,808 and 2,216 Ma, respectively. The SOI gneisses and the coastal gneisses of northern Labrador clearly experienced a complex tectonic and metamorphic history which the simple interpretation of the secondary feldspar-isochron model does not reproduce. However, there are three features of the secondary 'isochron' that we believe justify the simple two-stage treatment for the purpose of Pb-isotope interpretation.

First, provided that the U/Pb ratio of the whole rock is not greatly disturbed in the course of Pb-isotope homogenisation of previous events, a valid secondary isochron can form during the last event of homogenisation. We recall that the whole-rock regression age, although scattered and imprecise, is still within error of the much more precise U-Pb zircon ages. This suggests that while the scatter does document a certain disturbance of the whole-rock U/Pb ratios, the general U-Pb isotopic integrity of the SOI TTG gneisses has remained preserved. Hence, although the secondary feldspar-isochron that

is now preserved may only be the latest in a whole succession, its slope still contains valid information about t_m , t_h and initial isotope composition.

Second, both in the IGB area and in northern Labrador there is evidence for Palaeoproterozoic isotope resetting. While there is no doubt that the strongest metamorphic overprints in both areas occurred at ca. 3.6 Ga and again between 2.6 and 2.7 Ga (e.g. Collerson 1983b; Baadsgaard et al. 1986), the important point for the secondary feldspar-isochron is that several studies have found reset mineral chronometers that require low-grade metamorphism or recrystallisation long after 2.6 Ga. Pankhurst et al. (1973) showed that K-Ar, Ar-Ar and Rb-Sr mica and amphibole ages from Itsaq gneiss samples were all reset and that a late Archaean and a Proterozoic (1.6–1.7 Ga) homogenisation event were recorded. Essentially identical early to mid-Proterozoic apparent ages were obtained by Baadsgaard et al. (1976) in their K-Ar, Rb-Sr and U-Pb mineral isotope study of the coastal Itsaq gneisses. In the IGB area, Baadsgaard (1983) found that discordant U-Pb sphene and apatite dates were unable to pin-point the exact date of secondary metamorphism in the range of 1,500–2,800 Ma. In a later study, Baadsgaard et al. (1986) documented two separate Sr-isotope resetting events in the IGB area at 2,550 and $1,623 \pm 65$ Ma, the latter within the limited resolution of t_h calculated from our secondary feldspar-isochron. In northern Labrador, there is strong evidence from homogenisation of Sr-isotope systematics in the Uivak gneisses that the last pervasive Sr-isotope homogenisation occurred at ca. 1,800 Ma. Collerson (1983b) discovered that individually cut compositional layers from two slabbed Uivak gneisses yielded secondary 'isochrons' with regression ages of 1,798 and 1,884 Ma, while the total rock sample still aligned along early Archaean reference lines. This

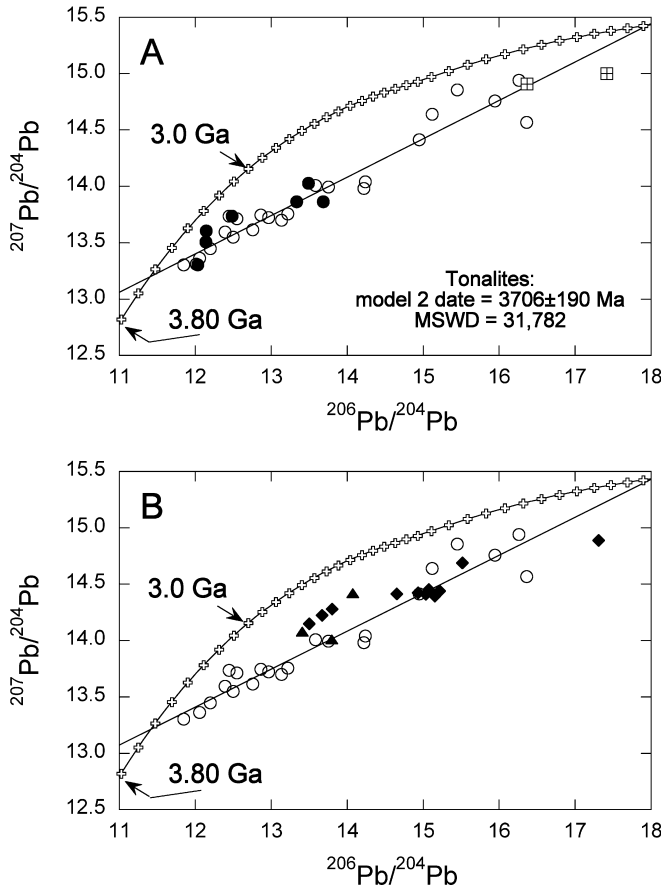


Fig. 4 Common Pb diagrams for whole-rock data. **A** Whole-rock data (Table 3) shown separately for the SOI tonalite gneisses (*open circles*), amphibolite gneisses (*solid circles*) and layer-parallel granite melts (*crossed squares*). Regression line is shown for tonalite gneisses only. The mantle evolution line (*solid fine curve connecting open crosses*) is that of Kramers and Tolstikhin (1997). Distance between *crosses* corresponds to a 100-Ma time step. **B** Labrador whole-rock data (Table 4) shown separately for Nanok gneisses (*solid diamonds*) and Uivak gneisses (*solid triangles*) compared to SOI tonalite gneisses (*open circles*). Regression lines and mantle model as in **A**

indicates that the Rb-Sr system was reopened at the centimeter-scale during Proterozoic events that affected the North Atlantic craton (Collerson 1983b).

Third, ‘homogenisation’ in the context of feldspar Pb-isotopes refers to partial or complete exchange of Pb-isotopes between the whole rock and the feldspar, which could have occurred due to dynamic recrystallisation (all our plagioclase samples are recrystallised) or diffusional exchange in response to a thermal event. Limited isotope exchange between the whole rock and only some portions of the feldspar still yields a valid slope (Fig. 6B) for simple systems that experienced only one isotope resetting. In other words, while the whole-rock-feldspar 2-point dates of partially homogenised feldspars will be older than t_h (like in our example, where they range between 2,700 and 3,500 Ma), the slope of the secondary feldspar-‘isochron’ remains valid. In more complex situations with multiple feldspar recrystallisations, partial homogenisation will

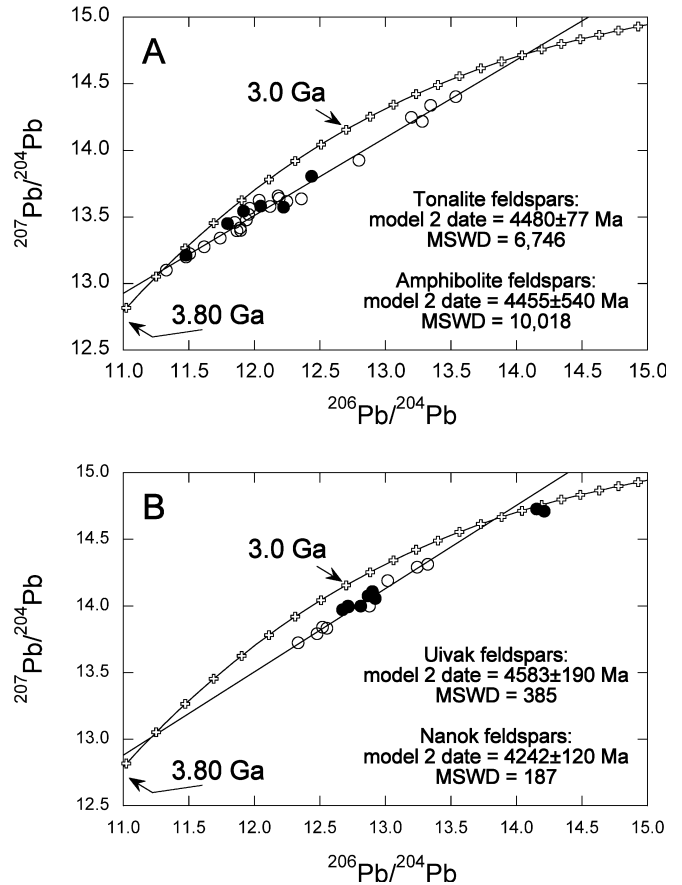


Fig. 5 Common Pb diagrams for feldspar data. **A** Feldspar data (Table 3) plotted separately for SOI tonalite (*open circles*) and amphibolite gneisses (*solid circles*). Regression line is shown for tonalite gneisses only. Mantle evolution line as in Fig. 4. **B** Labrador data (Table 4) shown separately for Uivak gneisses (*open circles*) and Nanok gneisses (*solid circles*). Regression line shown is for Uivak data only

introduce scatter into the isochron. The SOI gneiss secondary feldspar-‘isochron’ shows scatter outside the reproducibility that can be expected for the leaching protocol. This was estimated from the Pb-isotope ratios of one plagioclase (409010) that plots farthest from the secondary isochron. Its $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios were reproduced to within 0.2, 0.02 and 0.02% in two separate leaching experiments (see Table 3). The much larger scatter between samples is therefore not due to inefficient leaching of labile Pb but was caused by incomplete homogenisation or by superimposed effects of several homogenisation events.

In brief, the slope of the observed secondary feldspar-‘isochron’ reflects isotopic exchange between feldspar and whole rock during one or more metamorphic events, the latest of which occurred during the mid-Proterozoic.

Pb-isotope heterogeneity in the early Archaean

The main purpose of feldspar and ore Pb-isotope studies is not geochronological but to address whether analysed

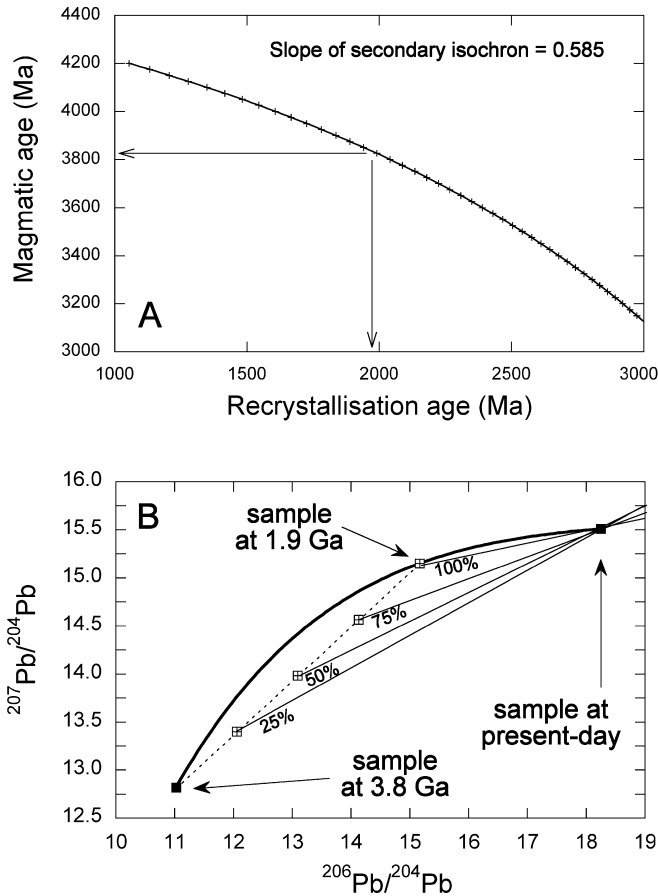


Fig. 6 **A** Combinations of magmatic ages and recrystallisation ages that yield a constant secondary isochron slope of 0.585, i.e. corresponding to an apparent age of 4,471 Ma. Because of the gentle curvature, relatively small changes in magmatic age require larger shifts in recrystallisation age. Arrows indicate likely magmatic age of SOI tonalite gneisses and corresponding recrystallisation age of ca. 1,950 Ma. **B** Effect of partial feldspar-whole-rock homogenisation on 2-point 'ages' and slope of secondary feldspar-isochron. Shown is a calculated evolution curve (*bold*) for a 3.80-Ga-old rock (with $\mu=9$) to the present day. In the case of complete homogenisation with the whole rock at 1.90 Ga, feldspar (*open crossed square*) plots along the evolution curve and the 2-point slope (*solid fine line*) with the present-day whole rock corresponds to the 1.90-Ga age of homogenisation. Partial homogenisation (three examples shown, corresponding to 25, 50 and 75% mixtures), however, produces less radiogenic feldspar Pb-isotope compositions that yield meaningless 2-point 'ages' (3,042, 3,447 and 3,664 Ga) with the whole rock. The important point is that all feldspar datapoints (fully and partially homogenised) still plot on the correct secondary isochron (*stippled line*)

samples show evidence for U/Th/Pb fractionation of their sources and, if so, to explore implications for terrestrial differentiation. In this regard, the Pb/Pb system has the advantage that in the early Earth's silicate reservoirs both $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ increased by several percent per 100 Ma, the latter especially benefiting from the much higher ^{235}U abundance at that time. Therefore, differentiation in the early Earth is expected to have left a record in initial isotope ratios recorded by samples with very low μ , such as feldspars, ores and chemical sediments. In order to compare the

initial Pb-isotope composition of different early Archaean rock types from SW Greenland and northern Labrador, it is necessary to refer to modelled evolution curves. The loci of these evolution lines reflect the rate of radiogenic ingrowth in particular terrestrial reservoirs. As such they depend on model assumptions which can be verified by comparing observed with modelled isotope composition.

There are two basic approaches to model Pb-isotope evolution curves. The first is to define a time series of input data to which a model curve is fitted by adjusting the μ of the reservoir in a series of successive 'stages'. This concept is widely applied to other radiogenic isotope systems, most notably Sm-Nd, where the isotope evolution of the 'depleted' mantle is defined by data from the most radiogenic basaltic rocks at any given time. Because of the mobility of U it is impossible to derive *bona fide* initial Pb-isotope ratios for ancient basaltic rocks, so that original attempts at modelling 'terrestrial' Pb-isotope evolution (naturally referring to the silicate Earth only) used data for U-free ores as input parameters. However, it was soon realised (e.g. Kanasevich 1968) that there exist strong isotopic variabilities between ores of the same age, indicating that Pb in those ores was derived from sources that had experienced evolution with different μ (and Th/U). Due to the degree of isotopic variability in ores and feldspars, illustrated in Fig. 1, it is necessary to make significant and equivocal assumptions regarding the origin of the Pb. Once the extent of variability in ore Pb-isotope ratios became appreciated [for a thorough discussion see Hofmann (2001)], distinction between 'conformable' and 'anomalous' was no longer warranted, so that most recent attempts at devising Pb-isotope evolution curves have used a less data-dependent approach.

The most widely known example of this second class of models was developed by Stacey and Kramers (1975). Rather than fitting an evolution line to a time-series of input data, this model was largely constrained by the following input values: the age of the Earth; the initial isotope composition of the Earth (as approximated by meteoritic troilite); and the Pb-isotope composition of present-day 'accessible' Earth. The latter parameter was estimated by measurements of MORB and pelagic sediments. As such, the Stacey and Kramers (1975) model provided only a single evolution curve which averages the various silicate Earth reservoirs and does not do justice to the fact that the ore and feldspar database (Fig. 1) demonstrates existence of strongly fractionated (in terms of U and Pb) reservoirs in the distant geological past.

More elaborate kinds of this type of model (e.g. Zartman and Haines 1988) have since addressed this problem and have proposed evolution curves for several crustal and mantle silicate Earth reservoirs. The evolution lines of 'old upper continental crust' and 'young lower continental crust' of Kramers and Tolstikhin's (1997) model encompass much of the variability seen in initial Pb-isotopes preserved in ores and feldspars

(Fig. 1). Recalling that the continental crust harbours more than a third of the terrestrial U and Th, it is clear that its extraction from and recycling back into the mantle have had the strongest effect on the present-day isotope composition of the MORB-source mantle. In contrast to the Sm-Nd system, which is relatively insensitive to extraction and isolation of large volumes of early Archaean crust (e.g. Nägler and Kramers 1998), the present-day differences in Pb-isotopes between MORB and average upper crust (as approximated by sediment) provide a strong control over the continental crust volume vs. time curve. In particular, this difference, combined with the average crustal Nd-isotope mantle derivation age of 2–2.5 Ga, implies that continental crust volume was small in the early Archaean and that the strongest net growth of the continents occurred between 3 and 2 Ga. Independent geochemical evidence supports the S-shape of the continental crust volume vs. time curve (e.g. Collerson and Kamber 1999).

However, these advances in plumbotectonics are of limited relevance to the interpretation of Pb-isotopes of the earliest Archaean samples (i.e. > 3.5 Ga). The reason for this is that most models (understandably) did not attempt to derive evolution lines for the primordial terrestrial crust that existed prior to formation and preservation of continental crust s.s. The model of Kramers and Tolstikhin (1997) does use a transient protocrust, whose Pb-isotope evolution, however, was not modelled. Rather, the protocrust in their model is fully recycled into the mantle at 4.3 Ga and its Pb-isotope evolution was not considered important for any preserved terrestrial samples. However, in the past few years, an increasing number of > 3.8-Ga zircons have been reported and the known terrestrial record now dates back to 4.4 Ga (Wilde et al. 2001), indicating that evolved rocks existed in the terrestrial protocrust. Kramers (2001) speculated that an enriched early crust that persisted for several 100 Ma might be the reason for the large variability in early Archaean $^{207}\text{Pb}/^{204}\text{Pb}$ ratios. Here we test how far our new Pb-isotope data for early Archaean samples are compatible with this proposal.

Our discussion is based on a comparison between data from the earliest accessible Earth and the MORB-source mantle evolution line of the Kramers and Tolstikhin (1997) model. Reference to a model facilitates comparison and construction of specific evolution curves. Because we are primarily interested in exploring the extent of early Archaean Pb-isotope heterogeneity it is not relevant whether the model MORB-source evolution line is correct. The locus of the model curve between 4.3 and 3.6 Ga is largely determined by increase in bulk silicate Earth μ as a function of metal and sulphide segregation into the core. If the prototerrestrial material had a substantially higher μ (e.g. 6–8) than carbonaceous chondrites, similar to the range inferred for ordinary chondrites, core formation would not have had a strong effect on silicate Earth Pb-isotope evolution and the MORB-source would have plotted at higher $^{207}\text{Pb}/^{206}\text{Pb}$ than that modelled by Kramers and Tol-

tikhin (1997). We will discuss this possibility in more detail later, but will initially focus on describing the extent of observed Pb-isotope heterogeneity.

There are five independent observations regarding the Pb-isotope systematics of the early Archaean Earth:

1. First, feldspars from the Godthaabsfjord 3.65-Ga Amitsoq gneisses, belonging to a relatively late and well-developed intrusive pulse (McGregor 2000), contain the least radiogenic silicate Pb known on Earth. This plots very close to the Kramers and Tolstikhin (1997) MORB-source curve at ca. 3.66 Ga (Fig. 7A). The 3.65-Ga age for the magmatic protoliths of these gneisses is well established from U-Pb zircon dating (e.g. Nutman et al. 1996; Whitehouse et al. 1999). They also yield 3.65-Ga whole-rock regression dates in the Pb-Pb, Rb-Sr and Sm-Nd systems (Kamber and Moorbath 1998). The combined whole-rock and feldspar Pb-Pb regression line intersects the Kramers and Tolstikhin (1997) MORB-source evolution at 3.66 Ga (Fig. 7A).
2. Second, the very high $^{207}\text{Pb}/^{206}\text{Pb}$ ratio of IGB BIF. Moorbath et al.'s (1973) BIF regression line yields the same 3.7-Ga age as Nutman et al. (1997), obtained by U-Pb zircon dating of a number of IGB metasediments. However, the regression line intersects the MORB-source evolution line at an impossibly young 'age' of 3.38 Ga (Fig. 7B). This observation prompted Moorbath et al. (1973) to postulate an immediate precursor to BIF that experienced a very different U/Pb history from the mantle. More recently, Kamber et al. (2001) have extended that discussion to IGB siliceous carbonates which indicate derivation of Pb from a similarly high- μ source (Fig. 7B).
3. Third, the position of IGB galena Pb-isotope ratios relative to the MORB-source curve (Fig. 7B). With the exception of galenas from one locality, they all plot to the left of the mantle evolution lines and presently define the least radiogenic terrestrial Pb known. This is not an artefact of inaccurate mass discrimination correction, as Frei and Rosing (2001) have demonstrated excellent reproducibility between data obtained on thermal and ICP mass spectrometers. Frei and Rosing (2001) regarded the offset between all IGB galena data and mantle evolution lines as significant and concluded that this required an elevated μ for the source of those galenas.
4. The SOI TTG secondary feldspar-'isochron' intersects the mantle evolution curve at unexpectedly radiogenic values, corresponding to a nominal date of 3.69 Ga, instead of the expected 3.81 Ga (Fig. 5a). The difference in $^{207}\text{Pb}/^{204}\text{Pb}$ between 3.69 and 3.81 Ga is very substantial and well outside the regression accuracy. However, if the linear feldspar array was formed in multiple events of incomplete homogenisation, it may have experienced some clockwise rotation, resulting in artificially 'young' intersection with the mantle evolution curve. However, there is no obvious bias in the regression line

Fig. 7 Pb-isotope variability in early Archaean lithologies expressed relative to the MORB-source mantle evolution curve of Kramers and Tolstikhin (1997), of which 100-Ma steps are plotted as *open circles* (the 3.8-Ga point is highlighted as a *larger circle with bold stroke*). **A** 3.65-Ga Godthaabsfjord Amîtsoq gneisses define a whole-rock (*solid squares*) and feldspar (*open crossed squares*) regression line that intersects the modelled evolution line at 3.66 Ga. Feldspar data points plot very close to the 3.7- to 3.6-Ga section of the curve. **B** IGB galena data (*solid squares*; see Table 1) plot significantly to the left of the model curve (with four exceptions, which were omitted for clarity), while BIF data points (*open crossed squares*) measured by Moorbath et al. (1973) plot along a 3.7-Ga regression line that intersects the evolution line at 3.38 Ga. Other possible chemical sediments [*solid diamond symbols*; data from Kamber et al. (2001)] also plot along the BIF regression line. **C** A 3.80-Ga isochron starting from coeval mantle composition plots significantly below the isotope composition of all IGB supracrustals [*open diamonds*; data from Table 1; Moorbath et al. (1973); Kamber et al. (2001)] and Akilia enclave metamorphic rocks [*open triangles*; data from Kamber and Moorbath (1998)]

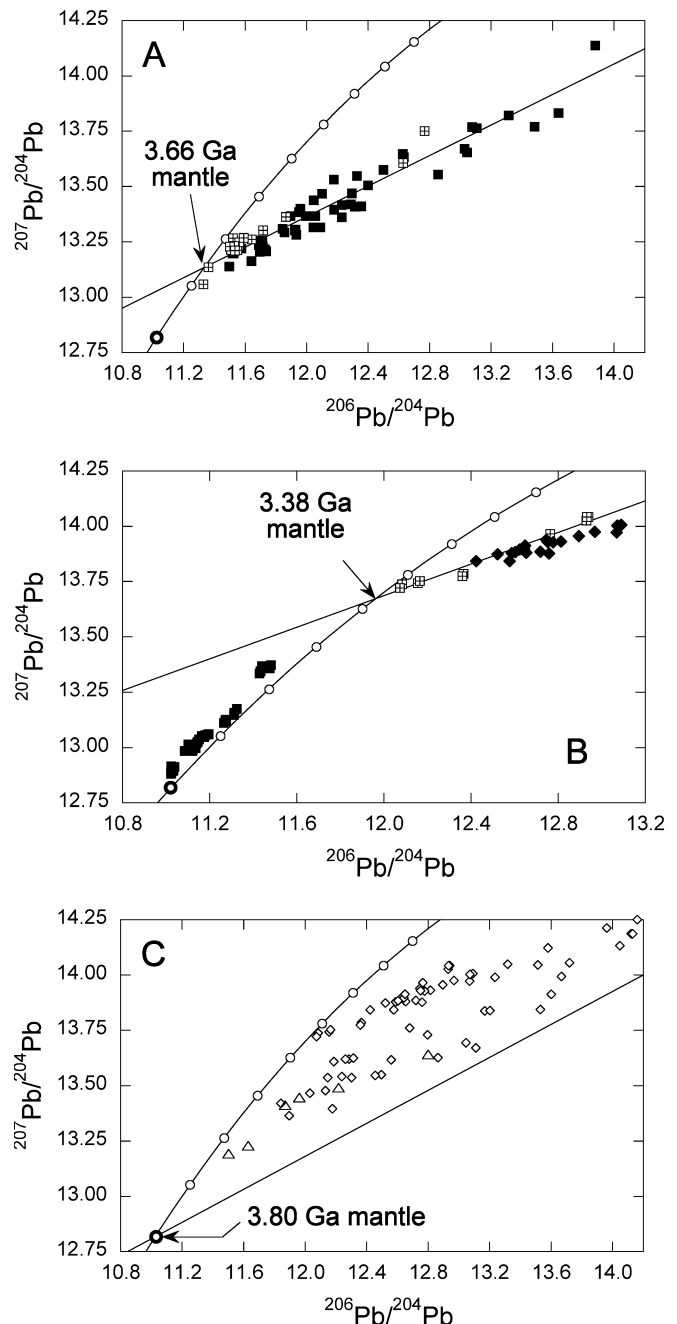
(Fig. 5a), which is defined equally well by the most and least radiogenic datapoints. This argues against systematic rotation. Furthermore, the whole-rock regression age, which is within error of the expected age of the rocks, also intersects the mantle evolution curve at an improbably young date of 3.63 Ga.

5. The Uivak feldspar regression line intersects the mantle evolution curve at 3.72 Ga (Fig. 5b), very similar to the U-Pb zircon age of 3.73 Ga.

In summary, two of the five lithologies (Amîtsoq and Uivak gneisses) have Pb-isotope systematics that are formally consistent with derivation from the mantle source modelled by Kramers and Tolstikhin (1997), while three types of lithologies (galena, chemical sediments and SOI gneisses) point to source areas that experienced a stage of significantly higher U/Pb evolution than the modelled mantle evolution line. In the following section we discuss the possible significance of this finding.

Model Pb-isotope evolution of high- μ source

The challenge of interpreting the observed early Archaean Pb-isotope heterogeneity is to identify the nature of the sources (i.e. crust vs. mantle) and to estimate the time of source separation necessary to generate the observed isotope variability. There are several possible scenarios which are encompassed by two end-member models. The first end-member assumes that the samples that require a source with a relatively low time-integrated U/Pb ratio were derived from a depleted portion of the mantle, while those that project to high initial $^{207}\text{Pb}/^{204}\text{Pb}$ ratios were derived from an undepleted or even enriched mantle domain. By contrast, the second end-member assumes that the mantle was not differentiated (i.e. neither depleted nor enriched) and that samples with radiogenic initial $^{207}\text{Pb}/^{204}\text{Pb}$ ratios were derived from a crustal source. There are three observations that favour the latter model. We discuss



these observations before we describe the parameters for our preferred second end-member scenario.

1. Because the bulk Earth μ is unknown and because terrestrial core formation could have been completed within 35 Ma (Schoenberg et al. 2002a), there is a possibility that the mantle evolution line to which we have compared the early Archaean data could plot at too low $^{207}\text{Pb}/^{204}\text{Pb}$ ratios at a given $^{206}\text{Pb}/^{204}\text{Pb}$ ratio. This is because in Kramers and Tolstikhin's (1997) model the μ of the silicate Earth gradually increased as Pb was removed with metal into the core. If, however, the core formed very rapidly, the period of retarded Pb-isotope evolution would have been

shorter, resulting in a mantle evolution line that plots at higher $^{207}\text{Pb}/^{204}\text{Pb}$ ratios. The most extreme case assumes a constant single-stage μ evolution, which corresponds to a bulk silicate Earth where core formation had no influence on Pb-isotope evolution. Indeed, a single-stage (4,568-Ma) mantle with a μ of 8.25 (Fig. 8A) plots significantly above the MORB-source mantle evolution line of Kramers and Tolstikhin (1997) and projects to the $^{207}\text{Pb}/^{204}\text{Pb}$ independently estimated for bulk silicate Earth by Galer and Goldstein (1996) and Kamber and Collerson (1999). However, as shown in the inset of Fig. 8A the IGB galena datapoints do not plot at sufficiently high $^{207}\text{Pb}/^{204}\text{Pb}$ to be derived from such an undepleted mantle source. If the μ of the single-stage model is adjusted to a value of 7.8, to yield a closer fit with the galena data (inset Fig. 8B), it becomes evident that such a mantle source would contain insufficient U to explain the present-day silicate Earth (i.e. the present-day composition of this source plots at lower $^{207}\text{Pb}/^{204}\text{Pb}$ than modern MORB; Fig. 8B). Further-

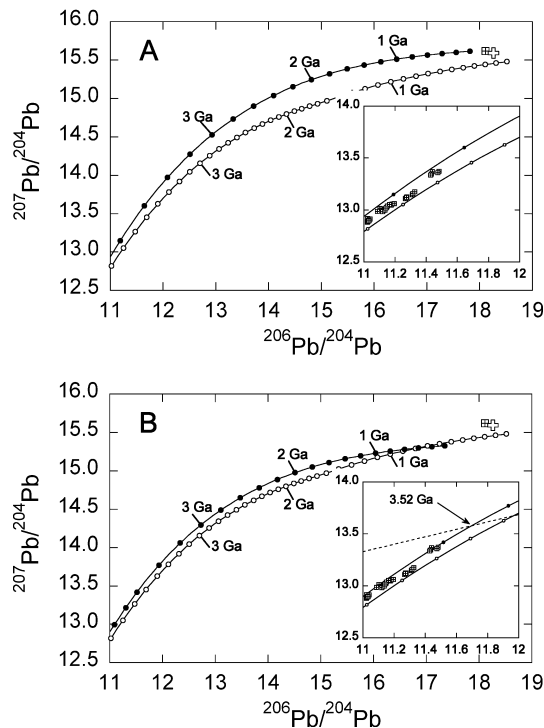


Fig. 8 Comparison of mantle evolution lines. **A** The MORB-source mantle evolution line (*open circles*) of Kramers and Tolstikhin (1997) plots well below a single-stage evolution with a constant μ of 8.25 (*solid circles*) that projects to the present-day Pb-isotope composition estimates of bulk silicate Earth: *crossed open square* by Galer and Goldstein (1996); *open large cross* by Kamber and Collerson (1999). *Inset* shows that the IGB galena (*small crossed open squares*) plot between both lines. **B** Similar comparison but showing a single-stage evolution (*solid circles*) with a lower μ of 7.8 to yield a closer fit to the IGB galena data (see *inset*). Note mismatch with present-day bulk silicate Earth estimates (*main graph*) and intercept with IGB BIF regression line (*stippled line in inset*) at 3.52 Ga

more, the single-stage mantle source fails to provide a solution to the IGB BIF data in that the BIF regression line intersects at too young an ‘age’ (inset Fig. 8B). In brief, the IGB galena data do not seem to outline a single-stage mantle evolution curve (such as could be expected for bulk silicate Earth).

- The second argument in favour of a crustal nature for the high- μ source is the observation that this source is most apparent in metasediments, and particularly chemical metasediments, while the lower- μ source is seen in the voluminous TTG gneisses (Godthaabsfjord Amitsoq and Uivak). Indeed, the highest $^{207}\text{Pb}/^{204}\text{Pb}$ ratios are found in IGB BIF and siliceous dolomites (Fig. 7B), which strongly implies a surface origin of the Pb that records derivation from a high- μ environment. Importantly, metapelites from the IGB also plot at considerably higher initial $^{207}\text{Pb}/^{204}\text{Pb}$ than contemporaneous mantle (Fig. 7C), lending support to the hypothesis that they represent erosion products of ancient terrestrial crust that may have persisted (and weathered) for several 100 Ma prior to deposition as clastic sediments. It is in these rocks that Schoenberg et al. (2002b) recently discovered ^{182}W -deficits, which they linked to accumulation of meteoritic W (from the heavy bombardment period) transported from weathered impact debris. Remelting of pre-existing high- μ crust could explain existence of TTG gneisses with high initial $^{207}\text{Pb}/^{204}\text{Pb}$.
- The third supporting argument is based on initial zircon Hf-isotope ratios from ancient zircons which, until recently, were interpreted to indicate derivation of the original melt from a strictly chondritic to mildly depleted reservoir. However, new experimental (Nir-El and Lavi 1998) and geological recalibrations (Scherer et al. 2001) of the ^{176}Lu decay constant fundamentally change this interpretation because of a significant shift in the chondrite reference Hf-isotope evolution. Amelin et al. (2000) and Scherer et al. (2001) noted that if the new ^{176}Lu decay constant is correct, initial Hf-isotopes recorded by early Archaean zircons would imply existence of an enriched reservoir. In order to evolve to the observed Hf-isotope compositions by ca. 4.0 Ga, this reservoir would have had to separate from convecting mantle at least 4.3 Ga ago (Scherer et al. 2001). The most likely candidate for the proposed enriched reservoir is a terrestrial protocrust.

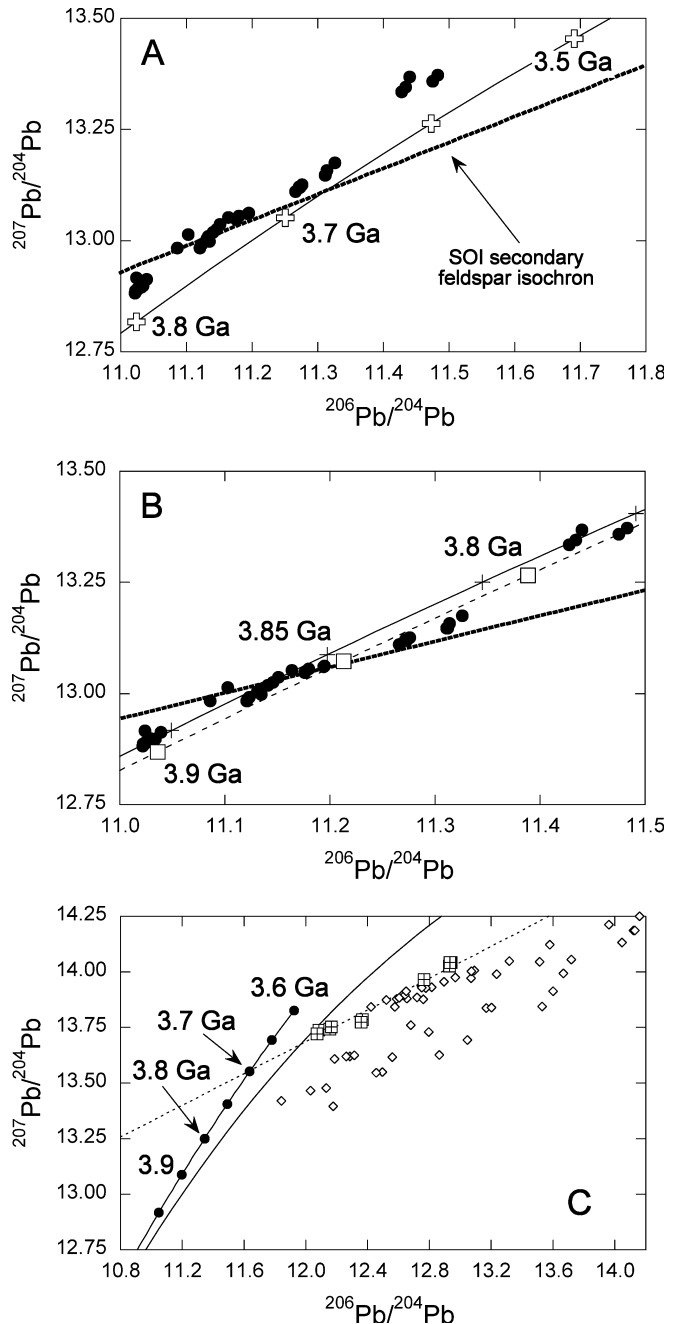
For these reasons we believe that the initial Pb-isotope variability seen in early Archaean rocks is best discussed in terms of existence of a terrestrial protocrust. This explanation is further supported by the fact that, unlike with mantle evolution lines, a single crustal model can be derived that provides an acceptable fit to all observations.

The difference in required μ between a sample source and contemporaneous mantle is a function of the source’s separation age. The longer the source spent in isolation from the convecting (model) mantle, the

smaller the difference in μ . For example, the 3.71-Ga IGB BIF source could have been isolated at 4.2 Ga and evolved with a μ of 13.8. Alternatively, if source separation occurred only at 3.80 Ga, a μ of 41 is required (Kamber et al. 2001). Similarly, the SOI tonalite gneiss source could have separated at 4.3 Ga and evolved with such a moderately high μ of 10.5, or it could have separated 200 Ma later with a μ of 12.5. In this manner, an infinite number of possible sources could be postulated for each available data set. However, the important observation is that one single model can be derived that provides acceptable fits to all observations. The evolution line of this high- μ Pb source is outlined by the galena datapoints, which define a curved array sub-parallel to the mantle evolution line (Fig. 9A). In order for a source to plot at significantly higher $^{207}\text{Pb}/^{204}\text{Pb}$ ratios than the mantle at a given $^{206}\text{Pb}/^{204}\text{Pb}$, without rapidly diverging from a parallel trajectory, a very early mantle separation (e.g. 4.3 Ga) with a moderately high μ is required. Later source separation (e.g. 4.1 Ga) with such a moderately high μ cannot explain the galena datapoints that plot farthest to the left of the mantle evolution line (Fig. 9B). The galena data therefore require an early source separation (Frei and Rosing 2001) for which we propose 4.3 Ga. An identical separation age for an enriched source was postulated by Scherer et al. (2001) in their reinterpretation of Hf-isotope ratios recorded by single early Archaean zircons. Slightly older (post-Moon formation) separation ages provide similar fits to the Pb data, but the quality of fit deteriorates rapidly for younger separation ages. For a 4.3-Ga source separation, the best fit to the combined galena, tonalite and BIF data was found with a source μ of 10.5. This describes a trajectory that encompasses all but the four most extreme galena compositions

Fig. 9 Pb-isotope evolution model. **A** Comparison between IGB galena data (*solid circles*; data sources as in Fig. 1) and mantle evolution model (*solid line connecting open crosses*) of Kramers and Tolstikhin (1997). The SOI tonalite secondary feldspar-‘isochron’ intersects the mantle model at ca. 3,680 Ma, which is at least 120 Ma younger than the magmatic age. The extent of mismatch in mantle model age is best appreciated by comparison with the model mantle composition at 3.80 Ga. The secondary ‘isochron’ projects directly into a prominent cluster of galena data. **B** Comparison of two source models for the SOI tonalite gneisses. The first (*broken line connecting open squares*) was calculated assuming a mantle separation age of 4.1 Ga and a μ of 12.5. It was modelled to intersect the secondary feldspar-‘isochron’ at 3.85 Ga but fails to explain the Pb-isotope compositions of all but the least anomalous galenas. A mantle separation age of 4.3 Ga with a μ of 10.5 (*solid line connecting + symbols*) results in a much better match with the galena data. It intersects the secondary feldspar-‘isochron’ at 3.87 Ga, which is within error of the resolution of the ‘isochron’s slope. **C** Comparison between mantle evolution line (*solid curve, no symbols*) with the 4.3-Ga, $\mu = 10.5$ model (*solid line connecting small solid circles, spaced at 50-Ma intervals*) derived in **B**. It can be seen that the Pb-isotope compositions of all IGB lithologies (*open diamonds*) project to between 3.70 and 3.82 Ga along appropriate reference isochrons (*stippled isochron corresponds to 3.70 Ga*). The 3.70-Ga BIF isochron (*stippled line connecting crossed squares*) of Moorbath et al. (1973) projects exactly to 3.70 Ga on the model source

(Fig. 9B). It yields a model age of 3.86 Ga for the SOI tonalite gneiss secondary feldspar-‘isochron’ (which is easily within error of the slope of that regression line), and a 3.70-Ga model age for the 3.71-Ga IGB BIF (Fig. 9C). Importantly, Pb-isotope compositions (Table 1) of all other typical IGB lithologies are also perfectly compatible with this model, as they extrapolate to intersections between 3.82 and 3.70 Ga, which is exactly the range found in zircon U-Pb ages (Nutman et al. 1997), Sm-Nd (Kamber et al. 1998) and Lu-Hf regressions [Villa et al. (2001); recalculated from data by Blichert-Toft et al. (1999)].



There is some scatter in the galena data (Fig. 9B), which probably exceeds analytical uncertainty. The spread of ratios *along* the model evolution line is easily explained as reflecting ore sources ranging in age from 3.75 to 3.91 Ga. This explanation differs from that of Appel et al. (1978) and Richards and Appel (1987) who attributed the variation in Pb-isotope ratios of galenas, which they regarded as late Archaean in age, to reflect slight differences in μ of a single ancient Pb source of a common age. Note, however, that Richards and Appel (1987) worked on a much smaller data array, which did not clearly define the sub-parallel character relative to the mantle curve. We prefer to interpret the variation in galena Pb-isotope ratios as largely reflecting differences in age of the Pb source rocks, allowing for the possibility that mineralisation occurred at 3.70 Ga (Frei et al. 1999). There is a large cluster of galenas whose model ages of ca. 3.86 Ga (Fig. 9B) exceed the age of the oldest known lithologies in the IGB. The SOI tonalite gneiss secondary feldspar-'isochron' projects into that data cluster, which indicates that the ore could have been derived by later remobilisation of Pb from feldspar in TTG similar to SOI. Figure 2 illustrates that the SOI gneisses are exposed in close proximity (0–30 km to the south) of the IGB. Furthermore, possibly detrital zircon grains from proposed IGB quartzites and from a paragneiss from the southern IGB cluster at exactly that age (Nutman et al. 1997; Nutman 2001). Zircon U-Pb dates of ca. 3.86 Ga have also been reported from Labrador, both from inherited xenocrysts of some Uivak gneisses (Collerson 1983a; Schiøtte et al. 1989) and from portions of detrital zircons in Nulliak quartzites (Nutman and Collerson 1991; Nutman 2001). The oldest U-Pb zircon dates from the North Atlantic craton date back to 3,897 and 3,910 Ma and are found in proposed IGB metasediments and Nanok gneisses (Collerson 1983a; Nutman et al. 1997), implying the existence of evolved crustal rocks of that age. In our model the magmatic hosts of these zircons were the source of the least radiogenic known terrestrial Pb, which is now stored in the IGB galenas with a 3.91-Ga model age. The deviation of some galena datapoints towards the right of the high- μ source (Fig. 9B) is explained by (minor) admixture of juvenile mantle source material. In summary, the Pb-isotope systematics of all IGB and SOI lithologies can be explained with a local source that separated from the mantle ca. 4.3 Ga ago and evolved with a μ of ca. 10.5.

Discussion

In the following discussion we follow the recent (informal) convention of defining Hadean as >4.0 Ga, but we emphasise that preservation of 3.8- to 4.0-Ga rocks is equally important and may hold the clues to preservation of even older Hadean isotopic memory.

Implications for early differentiation

The key observation regarding claims for the existence of radiogenic isotope heterogeneity within the early Archaean silicate Earth is the requirement for long-term (i.e. 300- to 500-Ma) source isolation. Differences in radiogenic isotope ratios recorded in earliest Archaean rocks (e.g. ca. 3.8 Ga) therefore require formation and isolation of enriched or depleted reservoirs in Hadean time. Here we postulate that Pb-isotope evolution of the early Archaean segment of the North Atlantic craton is explained by the early terrestrial upper mantle boundary layer model of Campbell and Griffiths (1993).

The mean age of oceanic lithosphere is presently ca. 60 Ma and models of oceanic crust production rate indicate that it may have been even shorter in the past. It is therefore impossible to create isotopic anomalies of the magnitude seen in studied lithologies over the lifetime of even the oldest known oceanic lithosphere (ca. 200 Ma) prior to subduction. However, as pointed out by Campbell and Griffiths (1993), in the absence of the subduction mechanism, a thick, relatively buoyant basaltic shell may have encased the mantle and survived for up to 500 Ma. Heat loss by conduction through stable basaltic lithosphere forms a cool, unstable, depleted boundary layer at the base of the crust. After 100–200 Ma, this mantle material cools to the point where it forms a cold plume (Campbell and Griffiths 1992), which sinks into the deep mantle with little entrainment of ambient mantle. Campbell and Griffiths (1993) commented on the possibility that this process could provide a means by which depleted mantle material was transported to the core-mantle boundary, from where it could be entrained into hot plumes (and brought back to the surface) at a much later stage. Here we stress the fact that the same process of cold plumes provides the early mantle with an inherent mechanism by which the most depleted material was removed from the uppermost mantle. In other words, in complete contrast to the present situation, it is more likely that isotope anomalies persisted in Hadean basaltic crust than in the upper mantle and that these crustal anomalies are 'enriched' in character.

Accepting the hypothesis of a stable Hadean basaltic crust for the terrestrial planets (e.g. Choblet and Sotin 2001), we now describe the geotectonic framework of the Pb-isotope evolution model for the early North Atlantic craton. The first step of this model involves creation of a thick basaltic crust at ca. 4.3 Ga with a μ of ca. 10.5. This represents an increase in μ by 26% from the coeval mantle value of 8.34 (Kramers and Tolstikhin 1997), which appears reasonable in view of the higher incompatibility of U. Importantly, because of the relatively low Pb concentration in MORB-like basalt [≤ 0.5 ppm on average; Jochum and Verma (1996)], creation of a thick global basaltic crust would not greatly affect the Pb-isotope evolution of the complementary depleted mantle. If the Hadean basaltic shell resembled enriched lunar basalts, which also have an elevated μ (Nyquist and Shih 1992), there is a possibility of significant complementary deple-

tion in the mantle. However, because of the aforementioned gravitational instability of the uppermost cooled mantle material, portions of the depleted residue would likely have sunk into the deepest mantle and been replaced by primitive mantle. We envisage that the basaltic proto-crust differentiated, by episodic remelting, to more silicic compositions. This could have occurred in response to exposure of the crustal base to convecting asthenosphere, immediately following departure of a cold plume. Melting of hydrated amphibolite, if present in the mafic proto-crust, could have produced silicic rocks of tonalitic or monzodioritic composition (e.g. Nutman et al. 1999). The oldest evidence in the IGB for differentiation is preserved as 3.90-Ga zircon (Nutman et al. 1997) and, if our model is correct, ca. 3.91-Ga-old source rocks for galena (Fig. 9B). Zircon evidence from the Acasta gneisses (Bowring and Williams 1999) and from Western Australia (e.g. Wilde et al. 2001) points to even earlier differentiation in these areas.

However, our model of a stable basaltic Hadean shell explicitly refutes operation of processes such as sediment recycling or melting in subduction zones. We therefore regard the interpretation of highly variable O-isotope ratios in Hadean zircons in terms of crustal assimilation or melting of greywacke protoliths at 4.3–4.4 Ga (Mojzsis et al. 2001; Wilde et al. 2001) as too speculative to question the fundamental constraints imposed by terrestrial cooling. The collection of recovered fragments from the Moon, which is patently depleted in water and volatiles, contains a considerable variety of evolved rocks, such as granites, granophyre and quartz-monzodiorites, some of which contain zircons (Nyquist and Shih 1992). Zircon U-Pb dates from these evolved rocks are in the range 4.37–3.90 Ga, but mostly fall in the older parts of this range (Meyer et al. 1996). The quoted authors conclude that the formation of lunar granite was not restricted to a single episode in lunar history, as, for example, during final solidification of the magma ocean. Rather, it appears to be the result of localised differentiation of individual plutons that intruded into the lunar crust or eventually erupted as felsic lavas.

In the IGB and SOI, differentiation, erosion and transformation of magmatic precursors into sediments proceeded at rapid pace between 3.90 and 3.80 Ga, but intrusion of the most voluminous TTGs, both in Greenland and Labrador, only started after 3.80 and peaked at 3.73 Ga (Labrador) and 3.65 Ga (Greenland). We view the creation of these granitoids as marking the initiation of subduction and rapid recycling of the Hadean basaltic shell (a process that may have been aided by the late heavy bombardment with meteorites).

Most importantly, both the 3.73-Ga Uivak gneisses and the most voluminous and dominant 3.65-Ga pulse of Amitsoq gneisses have Pb-isotope characteristics of the convecting mantle, and not of the high- μ Hadean basalt shell (Figs. 5B and 10A, respectively). This could imply a change in magma source from remelting the Hadean high- μ basalt shell to melting of the early Archaean convecting mantle. This view is supported by

concordant Rb-Sr and Sm-Nd regression lines for the Uivak and Godthaabsfjord TTG gneisses, which both yield intercepts corresponding to isotope ratios of contemporaneous, slightly depleted mantle (Collerson 1983a; Moorbath et al. 1997). The Pb-isotope characteristics of the oldest parts of the North Atlantic craton are thus testimony to the advent of subduction and provide a rare glimpse of Hadean differentiation.

Implications for Godthaabsfjord TTG gneiss Pb-isotope evolution

The co-existence of two reservoirs, from ca. 4.3 to ca. 3.65 Ga, with appreciably different μ (i.e. the high- μ Hadean basaltic crust and the mildly depleted mantle), complicates interpretation of Pb-isotope ratios of *individual* samples. This is best illustrated with the 3.65 Godthaabsfjord TTG gneiss whole-rock regression line (Fig. 10A). This intersects the depleted mantle evolution line at 3.66 Ga, which is the agreed age based on U-Pb zircon dating (e.g. McGregor 2000), and on the slope of the regression line itself. The intersection with the Hadean basaltic crust evolution line, however, is found at 3.85 Ga, because of faster evolution due to a higher source μ . Individual whole-rock datapoints that plot along the regression line cannot, therefore, be unambiguously assigned to the 3.65-Ga sample suite or to potentially older generations of TTGs (such as the SOI tonalite gneisses). The only incontrovertible direct Pb-isotope evidence for a ≥ 3.8 -Ga age of some of those samples would be a feldspar Pb-isotope composition that plots to the left of the mantle evolution line (akin to IGB galena). To date, such compositions have not been found (e.g. Kamber and Moorbath 1998). Indirect evidence against an age greater than 3.65 Ga for the samples studied by Kamber and Moorbath (1998) is provided by a comparison between feldspar and whole-rock data. In the case of the samples studied by Kamber and Moorbath (1998) and Gancarz and Wasserburg (1977), feldspar data, including the most radiogenic data points, plot closely along the 3.65-Ga whole-rock regression line (Fig. 10A). In the 3.80- to 3.82-Ga SOI tonalite gneisses, however, the feldspar Pb-isotope data define their own, much steeper trend (Fig. 10A), which we interpret as a secondary 'isochron'. Because such a trend is not seen in the Godthaabsfjord TTG gneiss data and because of the very low U-contents of igneous zircons, we argue that the very unradiogenic Pb of these samples is an original magmatic feature (Kamber and Moorbath 1998). However, we cannot exclude the possibility that a few of the whole-rock data points are from samples that are older than 3.65 Ga.

Implications for preservation of ≥ 3.8 -Ga rocks and minerals

Despite the fact that the early Earth, at least by 4.0 Ga, must have been covered by crust (probably basaltic),

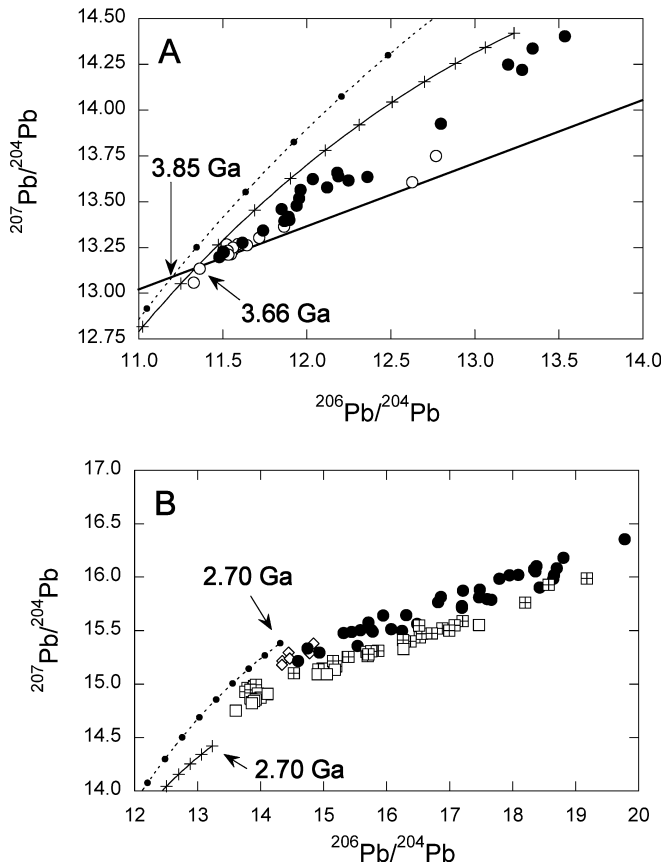


Fig. 10 Common Pb diagrams. **A** Intersection of 3.65-Ga Godthaabsfjord TTG gneiss regression line (*bold solid*; after Kamber and Moorbath 1998) with the depleted mantle curve (*solid fine line connecting + symbols*) of Kramers and Tolstikhin (1997) is at 3.66 Ga, while an intersection is found at 3.85 Ga with the Hadean basaltic crust evolution (*stippled line connecting small solid circles*). All leached plagioclase data points (*open circles*) compiled and presented by Kamber and Moorbath (1998) also plot along the 3.65-Ga regression line. This contrasts with the plagioclase data points for the SOI TTG gneisses (*solid circles*), which define their own, distinctively steeper trend. **B** Comparison of late Archaean model Pb-isotope compositions with data from late Archaean granitoid intrusions from high- μ cratons. *Solid line connecting (+) symbols* is depleted mantle curve by Kramers and Tolstikhin (1997) and *stippled line connecting small solid circles* is Hadean basaltic crust evolution derived in this study. Both evolution lines are shown to 2.70 Ga. Importantly, the least radiogenic datapoints of 2.6 to 2.73-Ga granitoids (typically from leached feldspars) from all cratons plot closely along a mixing line (not shown) connecting the depleted mantle evolution with that of the Hadean basaltic crust. The granitoids that plot closer to the 2.7-Ga Hadean basaltic crust evolution display a higher proportion of preserved Hadean crust in their source. *Symbols* and data sources for granitoids are: *solid circles* (Zimbabwe craton, northernmost Limpopo belt; Berger and Rollinson 1997); *open diamonds* (western Slave craton, Point Lake area; Davis et al. (1996); *crossed squares* (Wyoming craton, Beartooth Mountains; Wooden and Mueller 1988); and *open squares* (Yilgarn craton, Murchison province; Wang et al. 1993)

there is very limited direct evidence of its existence. The terrestrial situation contrasts with that of the Moon (almost complete preservation of ≥ 3.8 -Ga crust) and of Mars, where resurfacing did lead to appreciable denudation and reworking of ancient highlands (Hynek and

Phillips 2001). The scarcity of ≥ 3.8 -Ga material on Earth is a consequence of poor preservation. This implies tectonic instability and almost complete recycling of early crust. In the framework of an Earth encased with a relatively static basaltic shell (Campbell and Griffiths 1993), the disappearance of ≥ 3.8 -Ga crust is explained by the onset of subduction. Because the early basaltic shell was not created along subduction zones it lacked the buoyant lithospheric mantle keel, which is the key feature for survival of modern continental crust. Furthermore, in a plate tectonic framework, crust can only persist on a lithospheric mantle keel, it follows that only those fragments of early crust could survive that developed a keel *immediately* after onset of a tectonic framework driven by subduction. In other words, those portions of the early crust that experienced rapid addition of voluminous continental material (i.e. TTGs, granites and basalts) were also equipped with a complementary lithospheric mantle keel, which ultimately saved them from subduction. This conclusion appears to be supported by the geology of the other two major areas that preserve significant ≥ 3.8 -Ga material.

Bowring et al. (1989a), Bowring and Housh (1995), Stern and Bleeker (1998) and Bowring and Williams (1999) have reported U-Pb zircon dates of 3.8–4.03 Ga from the Acasta gneisses of the Slave craton (Northwest Territories, Canada). This provides strong evidence for existence of chemically evolved protoliths, which subsequently experienced a complex geological history. Importantly, the > 3.8 -Ga Acasta gneisses are surrounded by younger gneisses, with zircon dates between 3.6 and 3.8 Ga, identical to the situation in the North Atlantic craton. Geological events at 3.75 and 3.60 Ga can also be inferred from the complex U-Pb date patterns of those zircons that contain portions exceeding 3.8 Ga in age.

The oldest known terrestrial material by far (up to 4.4 Ga) are detrital zircons preserved in mid-Archaean metasedimentary rocks in the northwestern corner of the Yilgarn craton of Western Australia (Froude et al. 1983; Mojzsis et al. 2001; Wilde et al. 2001). The metasediments are tectonically juxtaposed against orthogneisses with interpreted magmatic ages between 3,730 and 3,380 Ma. The oldest in-situ rocks are represented by the 3,680-Ma Meeberrie gneiss and the 3,730-Ma layered intrusion of the Manfred complex. Detrital zircons of 3.5–3.75 Ga are also abundant in the Mt Narryer quartzite [see Fig. 2 of Kinny et al. (1990)], pointing to the former existence of significant components of early Archaean crust in the basement. Thus, the Mt Narryer gneiss complex contains the 3.75- to 3.60-Ga orthogneisses that we regard as crucial for preservation of ≥ 3.8 -Ga material.

There is only very limited further evidence for preserved ≥ 3.8 -Ga material. Dodson et al. (1988) reported 3.81-Ga U-Pb dates for detrital zircons from the Zimbabwe craton, which also contains 3.6-Ga granitoids in the Towke area (e.g. Taylor et al. 1991). Mueller et al.

(1992) discovered 3.96-Ga detrital zircons in quartzites of the Wyoming province, where the oldest known trondhjemitic gneisses date back to 3.5 Ga (Mueller et al. 1996). Although evidence for coupled occurrence of > 3.8-Ga material and 3.6- to 3.75-Ga TTG magmatism is incomplete, we stress, as our final conclusion, the observation that all the mentioned gneiss terrains occur on so-called high- μ cratons, as discussed below.

The high- μ signature as a remnant of Hadean basaltic crust

It has long been known that some (Archaean) cratons have a so-called high- μ Pb-isotope signature (e.g. Oversby 1975), while others (such as the Pilbara or Kaapvaal cratons) appear to have been derived from material with mantle-like U/Pb evolution. In the original definition, high- μ cratons are those that contain rocks whose initial Pb-isotope compositions require evolution in a source with a higher μ than MORB-source mantle. This does not necessarily imply that the rocks themselves inherit a high μ from the source. There is no agreed classification of cratons according to source μ , and we restrict our discussion here to the following examples: the Slave craton (e.g. Bowring et al. 1989b); the Yilgarn craton (e.g. Fletcher et al. 1988); the Zimbabwe craton (Taylor et al. 1991); the Wyoming province (Wooden and Mueller 1988); and the North Atlantic craton (despite its very unradiogenic present-day isotope composition). Explanations for the high- μ character of those cratons have favoured either sediment recycling or intra-crustal differentiation of U from Pb (e.g. Kramers and Tolstikhin 1997). Both explanations require the upper crust to assume a higher μ than the lower crust. In the sediment subduction model, sedimentary Pb of high- μ character (and ancient unradiogenic Nd) is cycled through arcs and ultimately reappears in the upper crust as arc magmas (Mueller and Wooden 1988). In the intracrustal differentiation model, U, Th and Pb are fractionated in the course of magmatic differentiation. High- μ Pb in such models is preserved by preferential erosion of juvenile material.

On the premise that the coexistence of > 3.8-Ga material, 3.6- to 3.75-Ga TTGs and a general high- μ character is not fortuitous, we propose a new model in which the high- μ character of the aforementioned cratons could be inherited from Hadean basaltic crust. We notice the good fit (Fig. 10B) between the modelled 4.3-Ga basaltic crust evolution line ($\mu = 10.5$) and initial isotope ratios from these cratons. The high- μ character of these cratons is well expressed in voluminous mid- to late-Archaean granitoids, which invariably contain Nd-isotope ratios indicative of a long crustal pre-history (e.g. Berger and Rollinson 1997). Furthermore, there is Os-isotope evidence from the Zimbabwe craton for initiation of growth of subcontinental lithospheric mantle at 3.8 Ga (Nägler et al. 1997).

We envisage that some Hadean high- μ basaltic crust and its differentiation products (such as the SOI gneisses and the source rocks of > 3.8-Ga zircons) were preserved in the emerging mantle-derived TTG-bearing cratonic nuclei which started forming along the earliest terrestrial subduction zones between 3.75 and 3.60 Ga. The rationale for this hypothesis is that widespread subduction-related magmatism between 3.75 and 3.60 Ga would have created a sufficiently deep subcontinental lithospheric mantle keel to stabilise the cratonic nuclei against destructive convectational forces. The basaltic high- μ rocks that were incorporated into the cratonic nuclei formed part of the source of later Archaean magmatism and, in that sense, the high- μ signature of these mid- to late-Archaean rocks may be hinting to the extreme antiquity of their source. Whether Hadean basaltic (and more differentiated) crust is properly preserved on Earth is still unknown, but the earliest pulses of non-mantle-derived orthogneisses (e.g. SOI, Nanok) are the most promising areas to harbour them.

Summary

Steep feldspar Pb-isotope arrays, which if interpreted as single-stage 'isochrons' yield apparent Hadean ages, are compatible with U-Pb zircon age evidence for an age of 3.80–3.82 Ga of SOI TTG gneisses in Greenland and an age of 3.81–3.92 Ga of the Nanok gneisses in northern Labrador. We interpret these arrays as secondary 'isochrons', which formed during feldspar-whole-rock isotope homogenisation, which last occurred in a 1.8- to 2.2-Ga tectonometamorphic event. The feldspar secondary isochrons are the first, albeit indirect, Pb-isotope evidence from terrestrial silicate samples for in-situ rocks older than 3.65 Ga.

Comparison of the secondary feldspar-isochrons with initial Pb-isotope information from metasediments and galena from the IGB and a combined feldspar-whole-rock regression line from the coastal 3.65-Ga Amitsoq gneisses demonstrates that Pb-isotope variability (mainly in $^{207}\text{Pb}/^{204}\text{Pb}$) existed in the early Archaean. Neither the magmatic precursors of the ≥ 3.80 -Ga TTG gneisses nor the metasediments or the galena in the IGB were derived from the mantle because they do not plot along trajectories that project to the Pb-isotope composition of present-day bulk silicate Earth. They require the existence of a terrestrial reservoir with a higher U/Pb ratio than mantle. Significantly, a single high- μ source that separated from the mantle at ca. 4.3 Ga with a μ of ca. 10.5 provides a good fit to all these observations. We propose that this source was the Hadean (basaltic?) crust which, in the absence of the subduction process, encased the early Earth. Differentiation of the early high- μ basaltic crust could have occurred in response to gravitational sinking of cold mantle material (Campbell and Griffiths 1992) or meteorite impact, and produced zircon-bearing magmatic rocks. The subchondritic

Hf-isotope ratios of ≥ 3.8 -Ga zircons support this model (Amelin et al. 2000) provided that the redetermined ^{176}Lu decay constant of Scherer et al. (2001) is correct. Our model of a stable basaltic Hadean shell explicitly refutes operation of processes such as sediment recycling or melting of hydrated material in subduction zones [as recently suggested by Mojzsis et al. (2001) and Wilde et al. (2001)]. Instead, we propose that initiation of terrestrial subduction occurred at ca. 3.75 Ga, at which stage most of the Hadean basaltic shell (and its differentiation products) was recycled into the mantle, because of the lack of a stabilising mantle lithosphere. Therefore, the preservation of ≥ 3.8 -Ga terrestrial rocks and minerals is not fortuitous, but reflects intrusion of voluminous granitoids immediately after establishment of global subduction because of complementary creation of a lithospheric keel.

Finally, we note that preservation of ≥ 3.8 -Ga terrestrial material (in-situ rocks and zircons) globally appears to be restricted to cratons with a high- μ source character (North Atlantic, Slave, Zimbabwe, Yilgarn and Wyoming). Although most of the direct early Archaean memory of these cratons was erased in later geological events, we suggest that their high- μ signature is an ancient isotopic memory pointing to the extreme antiquity of the source of these crustal domains and that the Pb-isotope systematics of these provinces are ultimately explained by reworking of material that was derived from Hadean basaltic crust.

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