Pb-isotope evidence for contrasting crustal contamination of primitive to evolved magmas from Ardnamurchan and Rum: implications for the structure of the underlying crust

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Synopsis

Using Pb isotope ratios we compare crustal contamination of primitive to evolved magmas from the Ardnamurchan and nearby Rum Igneous Centres, located on different crustal provinces in the British Tertiary Igneous Province (BTIP). The results confirm that compositional variations of parental cone-sheet magmas in Ardnamurchan can be explained by assimilation of granulite facies Lewisian gneiss at moderate crustal levels and subsequent contamination of evolved magmas with Moine schist metasediments within the uppermost crust during fractional crystallization. In contrast, samples from the Rum Centre have an uncontaminated mantle signature for the basaltic end-member, whereas the evolved rocks show Pb isotope evidence of contamination by Lewisian amphibolite facies rocks. Rum is separated from Ardnamurchan by a major thrust. The absence of a Moine-type isotopic influence in the Rum rocks supports an earlier interpretation, based on field evidence, that these overthrust rocks were eroded from Rum prior to the Palaeocene magmatic activity.

Introduction

Around 60 Ma ago intensive magmatism in NW Scotland and NE Ireland originated from lithospheric extension associated with the opening of the North Atlantic and formed the British Tertiary Igneous Province (BTIP). Besides widespread plateau lavas and dyke-swarms, several eroded remnants of ancient central volcanoes are present in western Scotland ('Igneous Centres', Fig. 1). There, primitive and evolved magmas encountered continental crust of various compositions during ascent and differentiation. Crustal contamination of these magmas is widely accepted and well documented in the BTIP from trace element and isotope studies (e.g. Carter *et al.* 1978; Dickin *et al.* 1981, 1984; Thompson *et al.* 1986; Morrison *et al.* 1985; Kerr *et al.* 1995, 1999; Geldmacher *et al.* 1998). Because of variable isotopic compositions and the limited number of published combined isotope and trace element data (e.g. parent/daughter ratios) of the crustal bedrock, accurate determination of the contaminating end-member can be difficult. Furthermore, large parts of the former uppermost crust have been lost through erosion since the emplacement of the magmatic rocks. Knowledge of the contamination history of intruding magmatic bodies could provide information about the structure of the surrounding country rock at the time of magma ascent and/or emplacement and hence, provide information on the structure of the underlying crust. In this study we compare the crustal lead isotope contamination of representative mafic and evolved rocks from the two Igneous Centres of Rum and Ardnamurchan, which are located on different crustal units separated by the major structural break of the Moine thrust (Fig. 1).

Geological setting and previous work

The crustal basement

The island of Rum and the peninsula of Ardnamurchan are located in NW Scotland at the transition of the Hebridean Terrane with the Caledonian orogenic belt (Fig. 1). The Hebridean Terrane represents one of the last phases of extensive crustal generation and is formed by the Archaean Lewisian gneiss complex (*c.* 2.9 Ga, Hamilton *et al.* 1979). A significant proportion of the Lewisian is made up of tonalitic to trondhjemitic gneisses with numerous mafic and ultramafic enclaves (e.g. Tarney & Weaver 1987). As a result of early high-grade metamophism, the Lewisan gneiss suffered metamorphism to granulite facies in the lower crust and to amphibolite-facies at upper crustal levels. During the metamorphism the granulites were strongly depleted in K, Rb, U and Th compared to the equivalent

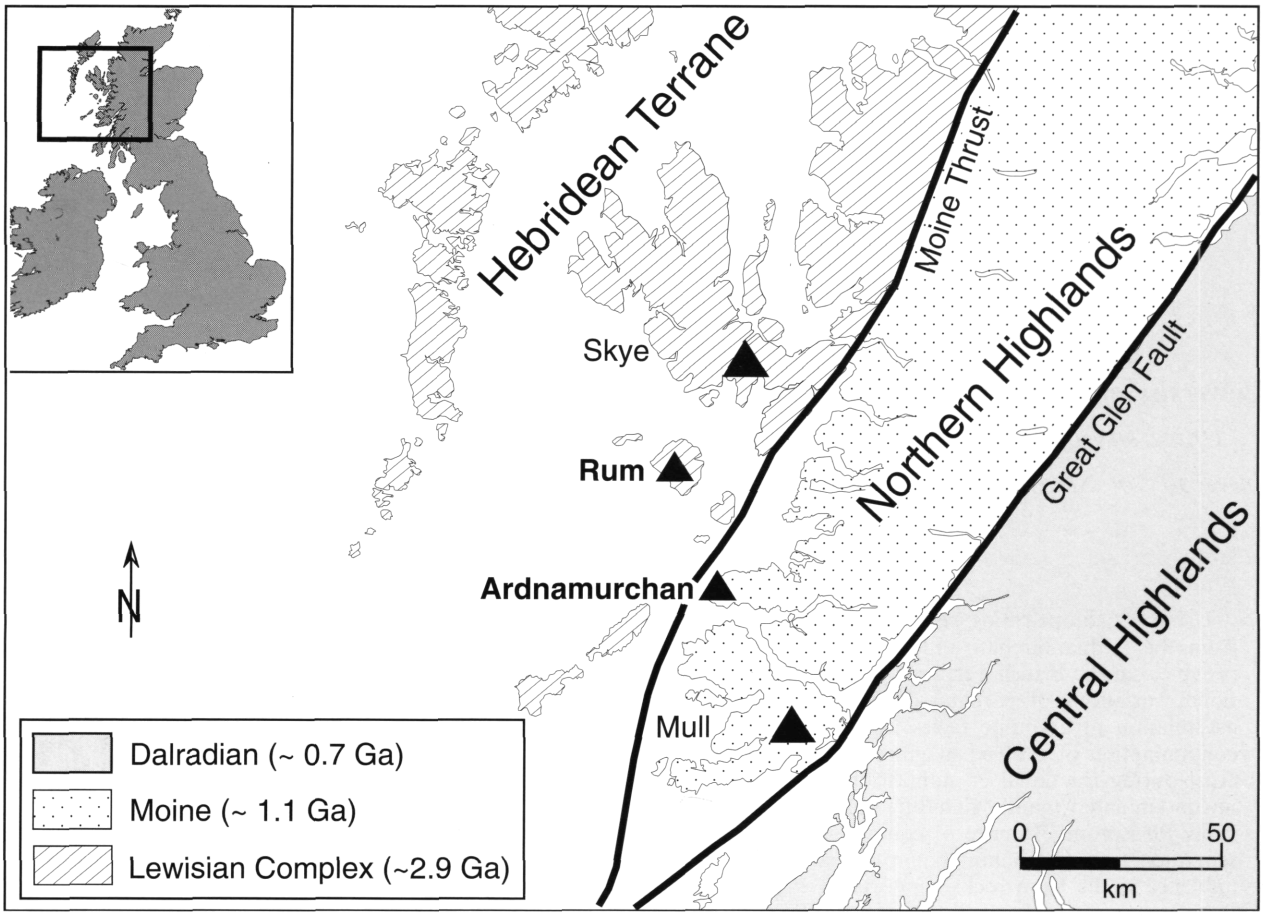

FIG. 1. Map of NW Scotland showing the major crustal provinces/terranes, the tectonic boundary features and the locations of several of the Tertiary Igneous Centres (black triangles), including the Ardnamurchan and the Rum Central Complexes. Mesozoic and younger rocks not shown. Simplified after Muir *et al.* 1992.

amphibolite-facies gneiss (e.g. Weaver & Tarney 1981), resulting in distinctly lower time-integrated $^{87}Sr/^{86}Sr$, $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, and $^{208}Pb/^{204}Pb$ ratios.

East of the Caledonian Moine thrust, the Lewisian gneiss is overlain by the Proterozoic rocks of the Moine Supergroup of the Northern Highlands. Deep seismic reflection profiles and geophysical evidence on physical rock properties (Hall 1987), however, suggest that the Lewisian or similar rocks form the basement of the Scottish mainland at least as far as the Great Glen fault to the south of the Northern Highland Terrane (Park 1991). The transition between amphibolite- and granulite facies Lewisian is generally assumed to be at depths of 6–14 km (Bamford *et al.* 1977). The overlying Moine schist succession is composed of mainly psammitic and pelitic metasediments that were metamorphosed in the late Proterozoic (*c.* 1.1 Ga; Harris & Johnson 1991). The thickness of the Moine rocks is unknown but several kilometres were suggested by Morrison *et al.* (1985).

The Moine thrust, that separates the Northern Highlands from the Hebridean Terrane in the north, dips at 20–25° to the east and carried the Moine metasediments over the Lewisian foreland (Smythe 1987). Therefore, the boundary between Moine schist and Lewisian gneiss must lie at a relatively shallow level beneath the Ardnamurchan Centre, which is located a short distance to the east of the thrust (Fig. 1). The thrust was active around 430 Ma ago (Halliday *et al.* 1987) during one of the main Caledonian deformation events.

The Tertiary Igneous Complexes of Rum and Ardnamurchan

The igneous complexes of Rum and Ardnamurchan belong to a belt of intrusive centres on the Scottish west coast (Fig. 1). The two complexes are located within 30 km of each other but are emplaced through different crustal provinces. The Rum Igneous Complex is located west of the Moine thrust on the Hebridean Terrane and the igneous rocks have intruded Lewisian gneiss and an unconformably overlying cover of mid-Proterozoic Torridonian sandstone with an approximate thickness of 2600 m. Outcrop of Lewisian gneiss is confined to the igneous complex where it occurs as large masses enveloped by the Palaeocene igneous rocks and smaller bodies tectonically emplaced along members of the Main Ring Fault system (Tilley 1944; Bailey 1945, Emeleus 1997). Its importance as a contaminant has been demonstrated for several units of the Layered Ultrabasic Suite (e.g. Palacz & Tait 1985; Tepley *et al.* 2000). From geological relationships Emeleus (1997) inferred that rocks of the Moine Supergroup were likely

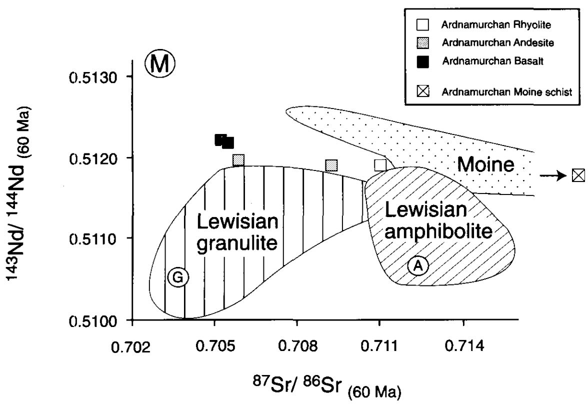

FIG. 2. Plot of $^{143}Nd/^{144}Nd$ versus $^{87}Sr/^{86}Sr$ isotope ratios of representative Ardnamurchan cone-sheets (after Geldmacher *et al.* 1998). Also shown are the fields for granulite and amphibolite facies Lewisian gneiss and Moine schist metasediments (see references therein). Fields and average composition (in circles) for Lewisian granulite (G), amphibolite (A) and sub-Hebridean Mantle (M) from Dickin (1981).

to have been eroded from Rum prior to the magmatic activity.

The igneous complex of Ardnamurchan is located slightly east of the Moine Thrust in the Northern Highlands (Fig. 1). Ring-dyke intrusions and numerous cone-sheets are emplaced in Moine Supergroup country rock partly overlain by a thin succession of Mesozoic sediments (for summary see: Emeleus 1982). The majority of the Palaeocene igneous rocks have a primitive basaltic composition, although some minor intrusions of more evolved rocks can be found.

Compositional variation of the dominantly tholeiitic to rhyolitic cone-sheets of the igneous centre of Ardnamurchan can be explained by crystal fractionation with simultaneous crustal assimilation (AFC). Based on combined trace elements and Sr- and Nd- isotope ratios, Geldmacher *et al.* (1998) postulated that basaltic cone-sheet magmas assimilated granulite facies Lewisian gneiss within the lower crust, whereas the andesite to rhyolite magmas formed by fractional crystallization from the contaminated basalt magma, and were further modified through the assimilation of Moine meta-sediments at uppermost crustal levels. Based solely on Sr- and Nd- isotope ratios, a clear discrimination between contamination of the andesite and rhyolite magmas by Moine or by amphibolite facies Lewisian remains, however, equivocal (Fig. 2).

Sampling and analytical techniques

Sampling, description, preparation techniques and major and trace element composition as well as Sr- and Nd- isotope ratios of samples from Ardnamurchan cone-sheets used in this study have been described in Geldmacher *et al.* (1998). Samples from Rum were collected from the Northern Marginal Zone of the complex where several composite sheets (samples CDCM-1, UDPN-3), plugs and ignimbrite outflow-sheets (sample STLH) of the earliest volcanic activity of the complex are exposed (Emeleus 1997; Troll *et al.* 1999, 2000; Donaldson *et al.* 2001). The sample of the Lewisian gneiss was collected from a Tertiary conglomerate underlying an icelandite lava flow of the Guirdil Member (Canna Lava Formation) on the south side of Fionchra in NW Rum (compare Emeleus 1997) (see Table 1). The cleaned samples were crushed and subsequently powdered in an agate mill. Lead isotope ratios and U, Th, and Pb concentrations of samples from Rum were determined from lithium tetraborate fusion digestions using an PE Elan 6000 inductively coupled plasma-mass spectrometer (ICP/MS) at the ACTLAB laboratory in Ontario/Canada (see website for details; http: //www.actlabs.com). Precision of trace element concentration is better than +/− 0.1 ppm for U and Th and +/−5 ppm for Pb (1σ). Accuracy of standard material concentration for those elements is better than 0.8 ppm SD. Precision of Pb-isotope ratios using the quadrupole ICP-MS at ACTLAB is between 0.2 and 0.5 % RSD (see 2σ errors in Table 1) which is significantly below the quality of a multi-collector mass spectrometer (e.g. TIMS). However, due to the large range of isotopic compositions of the measured magmatic samples and end-members, these errors are proportional. To evaluate the accuracy of isotopic data determined by ICP-MS, a duplicate of sample CDCM-1 was also measured at GEOMAR/Kiel on a Finnigan MAT 262 thermal ionization mass spectrometer (for details of chemical separation techniques see Hoernle & Tilton 1991). The isotope ratios (fractionation corrected to values given in Todt *et al.* 1996) lie within the analytical uncertainty of the values determined by ICP-MS for this sample (see Table 1). The samples STLH, SR321B and a plagioclase-separate from sample UDPN were also determined using TIMS. Analyses of Pb standard NBS 981 at GEOMAR gave $^{206}Pb/^{204}Pb$=16.893, $^{207}Pb/^{204}Pb$=15.433 and $^{208}Pb/^{204}Pb$=36.513 and analyses of standard SRM 981 at ACTLAB gave $^{206}Pb/^{204}Pb$=16.94, $^{207}Pb/^{204}Pb$=15.49 and $^{208}Pb/^{204}Pb$=36.72

Results

Based on major and trace element compositions, representative samples from Ardnamurchan (Geldmacher *et al.* 1998) and Rum (Troll 1998) were selected for Pb-isotope determination (Table 1). All isotope ratios were age-corrected to 60 Ma according to the time of igneous emplacement (see Emeleus 1991; Hamilton *et al.* 1998 for references).

The Moine schist sample (ARG 10) shows the most radiogenic initial Pb isotope composition ($^{206}Pb/^{204}Pb$=19.09, $^{207}Pb/^{204}Pb$=15.55; Fig. 3) that corresponds with isotopic data of crustal xenoliths from Streap Comlaidh (located around 50 km east of Ardnamurchan) reported by Halliday *et al.* (1993).

The basaltic (ARG 31) and the andesitic (ARG 50) samples from Ardnamurchan show less radiogenic initial $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios (16.93–17.01, 15.26–15.43, 36.81–36.88) but do neither match with early Tertiary sub-Hebridean (MORB)

TABLE 1
Pb-isotope data of rocks from Rum and Ardnamurchan

Sample	Rock type	Location (NGRS)	Excitat. Meth.	U ppm	Th ppm	Pb ppm	$^{206}Pb/^{204}Pb$ m.	$^{207}Pb/^{204}Pb$ m.	$^{208}Pb/^{204}Pb$ m.	$^{206}Pb/^{204}Pb$ (60 Ma)	$^{207}Pb/^{204}Pb$ (60 Ma)	$^{208}Pb/^{204}Pb$ (60 Ma)
Ardnamurchan												
ARG 31	Basalt	467/624	ICP-MS	0.10	0.36	1.24	16.98(6)	15.26(12)	36.86(22)	16.93	15.26	36.81
ARG 50	Andesite	456/641	ICP-MS	0.85	3.64	8.95	17.06(10)	15.43(18)	36.96(30)	17.01	15.43	36.88
ARG 9	Rhyolite	489/630	ICP-MS	2.44	10.0	17.7	17.60(14)	15.33(6)	37.67(22)	17.52	15.33	37.56
ARG 10	Moine schist	489/631	ICP-MS	5.01	15.4	17.2	19.27(12)	15.56(8)	38.82(30)	19.09	15.55	38.64
Rum												
UDPN-3	Rhyodacite	385/986	ICP-MS	0.72	5.29	13	16.13(20)	15.11(14)	38.27(38)	16.10	15.11	38.19
CDCM-1-1	Basalt	387/984	ICP-MS	0.54	2.03	10	18.40(10)	15.41(12)	38.14(38)	18.37	15.41	38.10
CDCM-1-2	Basalt	387/984	TIMS	0.54	2.03	10	18.349(1)	15.420(1)	37.999(3)	18.317	15.418	37.959
STLH	Rhyolite	387/981	TIMS	0.75	5.33	15	15.767(1)	14.968(1)	37.785(2)	15.740	14.967	37.720
SR321B	Lew. gneiss	336/004	TIMS	0.48	2.78	4.52	14.007(2)	14.558(6)	37.069(1)	13.950	14.556	36.960
UDPN-plg	Rhyodacite	358/986	TIMS	<0.10	0.07	1.49	15.535(4)	14.914(4)	37.702(1)	15.498	14.912	37.693

Pb-Isotope data. Location of sampling site coordinates refer to the British National Grid Reference System (NGRS). All lie within square NM (except for SR321B in NG). All errors are quoted at the 2 sigma level. Concentrations of Pb, Th, and U of Ardnamurchan samples for calculation of initial values are taken from Geldmacher *et al.* (1998)

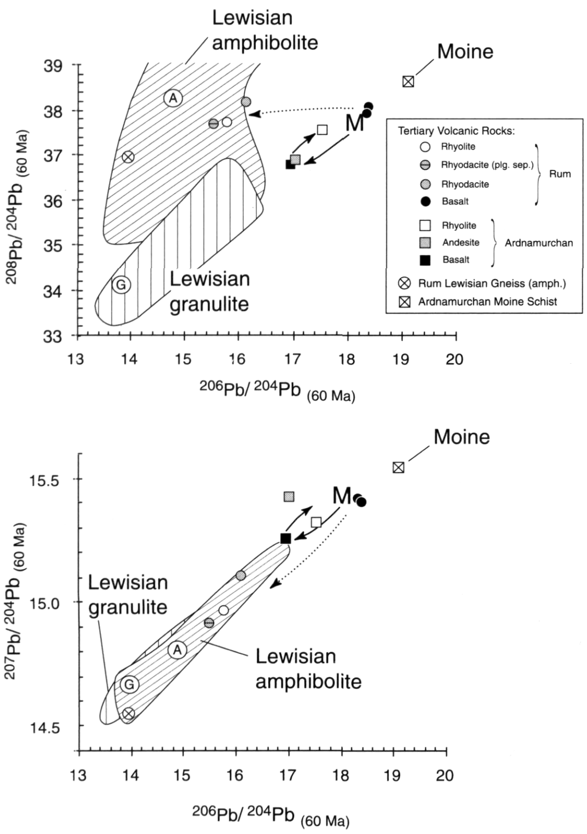

FIG. 3. **a, b**: Pb isotope ratios for representative Ardnamurchan and Rum samples. Ardnamurchan contamination trends (solid arrows): The basaltic sample is displaced from the sub-Hebridean Mantle composition (M) towards average Lewisian granulite, whereas the andesitic and rhyolitic samples form a contrary trend towards the Moine schist composition with increasing magmatic evolution. In contrast, basaltic samples from Rum (broken arrows) show an uncontaminated composition similar to sub-Hebridean mantle, whereas the evolved samples form a trend towards average amphibolite facies Lewisian. Fields and average composition (in circles) for Lewisian granulite (G), amphibolite (A) and sub-Hebridean Mantle (M) from Dickin (1981).

mantle composition ($^{206}Pb/^{204}Pb=18.16$, $^{207}Pb/^{204}Pb=15.46$, $^{208}Pb/^{204}Pb=37.72$) defined by Dickin (1981) nor with the recently proposed North Atlantic End-Member NAEM ($^{206}Pb/^{204}Pb\approx17.5$, $^{207}Pb/^{204}Pb\approx15.4$, $^{208}Pb/^{204}Pb\approx37.4$) of Ellam & Stuart (2000). Instead, the three Ardnamurchan samples are displaced towards average Lewisian granulite composition, with the basaltic sample having the least radiogenic and the rhyolitic sample (ARG 9) having the most radiogenic Pb isotope composition (Figs. 3a, b).

In contrast, samples from a composite sheet from the Rum Centre display lead isotope compositions resembling a sub-Hebridean mantle signature for the basaltic endmember (CDCM-1), whereas the rhyodacite through rhyolite rocks (UDPN-3, STLH) and plagioclase separates from the rhyodacite (UDPN-plg) show mantle-like $^{208}Pb/^{204}Pb$ but very unradiogenic $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{206}Pb$ ratios (Figs. 3a, b). This results in a strong shift of the data points towards the Lewisian amphibolite facies composition (including Lewisian sample SR321B from Rum).

Discussion and concluding remarks

The observed variations in Pb- isotope ratios point to subsequent contamination of mantle-derived magma with continental crust. Considering an AFC-like process for the magmatic evolution of the cone-sheets in Ardnamurchan, two contaminating end-members to the primitive, basaltic magmas of MORB-like composition are required: firstly, an average granulite facies Lewisian gneiss within the lower or middle crust, resulting in a contaminated basalt magma with relatively unradiogenic Pb isotope ratios (Figs. 3a, b) and only slightly enriched Sr isotope ratios (Fig. 2): secondly, at uppermost crustal levels, most likely in near-surface magma chambers, Moine-type metasediments. This latter contamination occurred while the magmas simultaneously fractionated through an andesitic to a rhyolitic composition. The radiogenic Pb isotope composition of Moine metasediments therefore led to higher $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ isotope ratios with increasing magmatic differentiation (Figs. 3a, b). The Pb isotope data from this study confirm the previously proposed model of two-stage crustal contamination of Ardnamurchan cone-sheets presented by Geldmacher *et al.* (1998), supporting a stratified crustal structure beneath Ardnamurchan with Lewisian granulites overlain by Lewisian amphibolites and Moine-type metasediments.

The representative samples from Rum, located west of the Moine thrust, show no evidence for assimilation of Moine type lithologies. The Pb isotope composition of the basaltic magma shows an uncontaminated mantle signature (Figs. 3a, b). If we were to consider a slightly less radiogenic sub-Hebridean upper mantle composition (e.g. NAEM) the Rum basalt magma would be slightly displaced towards the Moine composition. However, low Sr- isotope ratios of the Rum basalt (sample CDCM-1-1; $^{87}Sr/^{86}Sr_{60}=0.702923\pm9$; Troll *et al.* in prep.) preclude contamination with Moine type lithologies (compare Fig. 2). The decreasing $^{206}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ ratios of the rhyodacite through rhyolitic Rum rocks, on the other hand, can be interpreted as a result of increasing assimilation of average Lewisian amphibolite facies rocks forming the upper crust beneath Rum. Moreover, the somewhat more radiogenic Pb isotope ratios of the plagioclase separate relative to the whole rock imply either a high percentage of xenocrysts or, more probably, contamination by a high degree partial melt of a e.g. mica bearing country rock (cf. Duffield & Ruiz 1998).

Thus, the data corroborate earlier field observations that any originally overthrust Moine-type rocks in the Rum area must have been removed by erosion before the Mesozoic sediments were deposited (Emeleus 1997) and thus prior to the Palaeocene magmatic activity. Therefore rocks of the Moine Supergroup could not have been down-faulted within the ring-fault system of the Rum Igneous Complex and were not available as a contaminant when the igneous complex was emplaced.

These interpretations are consistent with the evidence from several other igneous centres of the area, notably those of Skye and Mull. The Skye Centre and the Isle of Eigg (NW of the thrust) are located on the same crustal segment as the Rum Centre and the Mull Centre (SE of the thrust) on the same crustal segment as the Ardnamurchan Centre. Whereas on Mull contamination of basaltic to acidic rocks by Lewisian and Moine type lithologies is well established (e.g. Thompson *et al.* 1986; Saunders *et al.* 1997; Kerr *et al.* 1995, 1999) on Skye and Eigg contamination of basaltic to acidic rocks is limited to Lewisian granulite and amphibolite facies compositions, i.e. no evidence for Moine type contaminants has been detected (cf. Dickin 1981; Dickin & Jones 1983; Saunders *et al.* 1997; Stuart *et al.* 2000). On Skye and Mull many of the early ascending magmas are thought to have ponded at lower crustal level where they experienced fractionation and contamination by lower crustal lithologies (Lewisian granulite) followed by a second contamination event at upper crustal levels (e.g. Thompson *et al.* 1986). Following their ascent to upper crustal levels, the magmas to the West and NW of the Moine thrust (e.g. Skye and Rum) encountered amphibolite facies Lewisian lithologies in the shallow parts of the Hebridean crustal segment. Those magmas that rose through the Northern Highland crustal Terrane to the East and SE of the Moine thrust (e.g. Mull and Ardnamurchan), encountered shallow-level Moine type sediments instead.

This brief comparison of the Rum and Ardnamurchan igneous rock samples therefore shows that chemical information locked in volcanic rocks can, in principle, be used as an indicator of the composition(s) of the underlying crust. Fractionated magma that assimilated crustal wall-rock(s) on ascent is thus considered as a valuable probe into the stratigraphy and structure of the continental crust.

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