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Clinopyroxenes and biotites from the Centre III igneous complex, Ardnamurchan, Argyllshire

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SUMMARY. Major- and trace-element analyses of the clinopyroxenes and biotites from the ring-intrusions of Centre III in the Tertiary igneous complex of Ardnamurchan are presented. It is shown that there are systematic differences in mineral chemistry between the eucrites, the gabbros, and the dolerites, and in particular the clinopyroxenes show an iron enrichment trend that is similar to that found in fractionated tholeiitic intrusions. The minerals from the intermediate rocks, the tonalite and quartz monzonite in the centre of the complex, do not show the further iron enrichment expected if these rocks had formed by continued fractionation of basic magma. On the basis of the chemistry of the clinopyroxenes and biotites, the intermediate rocks are interpreted as hybrids formed by the remelting and assimilation of country rocks into basic magma.

THE Tertiary igneous rocks of the Ardnamurchan peninsula, western Argyll, make up one of the six major Tertiary igneous centres of western Scotland. The geology of Ardnamurchan was extensively described by Richey, Thomas, and others in the Memoir of the Geological Survey in 1930, and their detailed mapping of the area established the three distinct intrusive centres for the igneous rocks (I, II, and III). The pyroxenes and biotites discussed in this paper are from the main rock types of the third and last of these centres.

Centre III is a plutonic complex of ring and arcuate intrusions situated between and cutting out parts of the two earlier centres, Centres I and II, and forms the largest and most complete set of ring intrusions in the British Isles. Richey *et al.* (1930) regarded the rocks of Centre III as outward-dipping ring-dykes becoming progressively younger towards the centre and formed by a process of cauldron subsidence. The structure proposed by Richey is not universally accepted and at the present time the shape of the intrusions is not known. Essentially, the rocks are coarse-grained basic gabbros with the development of a relatively small volume of intermediate rocks in the centre of the complex.

The divisions suggested by Richey are shown in fig. 1, and the list of these is given in Table I. The outermost ring intrusion of Centre III is the Quartz-gabbro of Faskadale (A), and this was regarded by Richey as the earliest formed ring-dyke. The partial ring of the Quartz-gabbro of Faskadale encloses the Fluxion gabbro of Faskadale (B), and these two intrusions, together with the small gabbro masses of Plocaig (C) and Meall nan Con (D) form the 'Outer' gabbros of Centre III.

The 'outer' gabbros surround the Great Eucrite (E in fig. 1) which occupies a ridge of high ground and makes up over one-half of the exposed rocks of the centre. Richey (1930) separated the Great Eucrite from the Biotite-eucrite (G) and the Inner

Eucrite (H) but subsequent work by Bradshaw (1961) suggested they were all part of a single funnel-shaped intrusion. The Great Eucrite encloses the 'Inner' gabbros: the Quartz-biotite Gabbros (J, J′, and J″ in fig. 1), which may originally have been part of one intrusion, and the Fluxion Biotite-gabbros of Sithean Mor (K), and the almost complete ring of the Fluxion Biotite-gabbro of Glendrian (L). In the very centre of the complex are the distinctive intermediate rocks: the Tonalite (M) and Quartz-monzonite (N).

In addition to these rocks there are, within the Eucrite ring, the Quartz-gabbros of Meall an Tarmachain summit, and the narrow ring of Quartz-dolerite veined with granophyre (F and I in fig. 1). The time of emplacement of these intrusions is not known for certain. The age relationships of some other members of Centre III is in doubt, but the over-all sequence with the first-formed rocks being the outermost intrusions and the central members being the last formed, is widely accepted. Until the structure of Centre III is known unequivocally the descriptive term 'ring-intrusion' is used here in preference to the genetic term 'ring-dyke'.

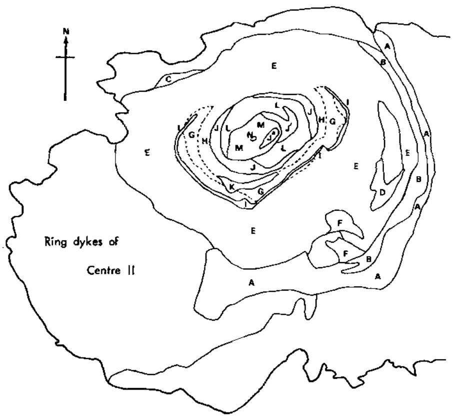

FIG. 1. Outline geological map of the rocks of the third igneous complex of the Ardnamurchan peninsula in Argyllshire, Scotland. (See Table I for list of intrusions.)

TABLE I.

Intrusions of Centre III, Ardnamurchan in proposed sequence of emplacement, following Richey *et al.* (1930).

(A)	Quartz-gabbro of Faskadale (earliest 'ring-dyke')
(B)	Fluxion Gabbro of Faskadale
(C)	Gabbro of Plocaig
(D)	Porphyritic Gabbro of Meall nan Con screen
(E)	Great Eucrite
(F)	Quartz-gabbro of Meall an Tarmachain
(G)	Biotite-eucrite
(H)	Inner Eucrite
(I)	Quartz-dolerite veined with granophyre
(J)	Quartz-biotite-gabbro
(J′)	Quartz-biotite-gabbro
(J″)	Quartz-biotite-gabbro
(K)	Fluxion Biotite-gabbro of Sithean Mor
(L)	Fluxion Biotite-gabbro of Glendrian
(M)	Tonalite
(N)	Quartz-monzonite

Clinopyroxene is an important mineral phase in all the rocks of the centre, with the exception of the granophyre within the dolerite, and was separated and analysed from each of the more important intrusions. Biotite is present only in small amounts in the eucrites and gabbros but can be more abundant locally. In the dolerite it is often present as about 4 % modally and in the Tonalite and Quartz-monzonite 7 % would be an approximate average modal abundance.

Clinopyroxene is an important constituent of the basic and intermediate rocks of Centre III. In the gabbro and eucrite intrusions it may locally form up to 50 % of the rock, although more representative modal analyses indicate 20 to 30 % clinopyroxene. It diminishes in amount in the intermediate tonalite and quartz monzonite, where in some samples it is replaced by primary amphibole. In most of the rocks orthopyroxene is present in only small amounts, but it can become more abundant locally, especially in the eucrites.

The petrographic work on the clinopyroxenes in the eucrites by Bradshaw (1961) indicated that the mineral varied in colour from pale brown to pale green, and was generally free from inclusions or exsolution, although it often had well-developed schiller structure. Clinopyroxene is recorded from all the more representative examples of the gabbros, and is generally homogeneous but frequently shows some alteration of the mineral in the gabbros of the Inner and Outer parts of the complex. Clinopyroxene was separated from 21 of the Centre III rocks, and major and trace-element analyses are given in Tables II and III. The analyses have been recalculated on the basis of 6 oxygen atoms and all have the number of cations in the (XY) and (Z) groups equal to $2{\cdot}00 \pm 0{\cdot}02$.

The Ardnamurchan Centre III pyroxenes are shown in terms of the components $CaSiO_3$–$MgSiO_3$–$FeSiO_3$ in the triangular diagram fig. 2. From this diagram it is clear that the pyroxenes separated are augites, following the nomenclature of Poldervart and Hess (1951), and that the variations in composition are relatively small.

The emphasis that has been placed on studies of the changes in composition shown by igneous pyroxenes lies in the changes that have been demonstrated in fractionating basic magmas. The most fully documented account of a fractionated basic igneous intrusion is the Skaergaard intrusion in east Greenland. The pyroxenes from the Skaergaard have been investigated by Muir (1951), Brown (1957), and Brown and Vincent (1963). In the Skaergaard intrusion the first formed rocks contain a clinopyroxene of composition $Ca_{45}Mg_{45}Fe_{10}$ and a coexisting orthorhombic Ca-poor pyroxene. With fractionation the composition of the pyroxene becomes richer in iron, and a point is reached at which the Ca-poor pyroxene ceases to crystallize. At this point (the two-pyroxene boundary) there is an increase in the Ca content of the clinopyroxene, in addition to the continued Fe-enrichment. This trend is illustrated in fig. 2. Studies on other areas, including those of Atkins (1969), Carmichael (1960), and Konda (1970), suggest that this trend of iron-enrichment in pyroxene composition is characteristic of fractionating tholeiitic magma.

For basic alkali magma there is much less evidence of a similar change in pyroxene chemistry. Pyroxenes from basic alkali rocks show only a small amount of iron enrichment, as demonstrated by Wilkinson (1957), Murray (1954), and Witkind (1969), although recently Gibb (1973) has suggested that for the clinopyroxenes from the Shiant Isles investigated by Murray a greater degree of iron-enrichment is apparent if analyses are made by electron-microprobe analysis.

For the Ardnamurchan clinopyroxenes the five eucrite samples analysed all have very similar compositions and, as fig. 2 shows, are the least iron-rich pyroxenes. The pyroxenes from the gabbros also have a small spread of points, but in comparison with the pyroxenes from the eucrites there is a small but consistent increase in iron content, with little or no decrease in calcium content. An exception to this grouping is the three clinopyroxenes separated and analysed from the small Meall an Tarmachain gabbro given in Table III; these show substantially greater iron-enrichment and are clearly separated in fig. 2 from the pyroxenes from all the other gabbro intrusions.

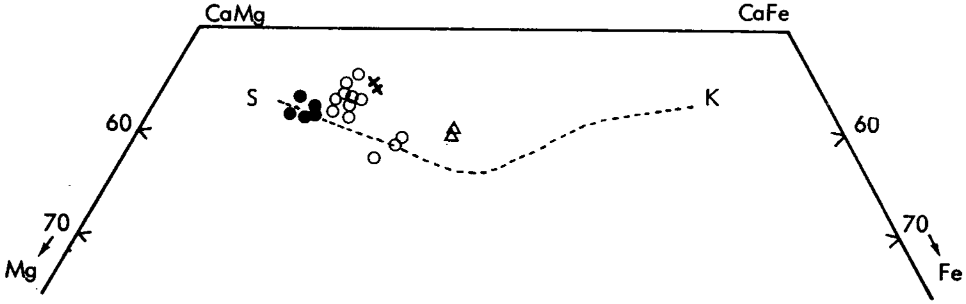

FIG. 2. Clinopyroxene compositions for the Ardnamurchan Centre III complex in the system $CaMgSi_2O_6$–$CaFeSi_2O_6$–$Mg_2Si_2O_6$–$Fe_2Si_2O_6$. ● eucrites; ○ gabbros; △ dolerite; × tonalite and quartz-monzonite. S—K Composition trend line for the clinopyroxenes of the Skaergaard intrusion. The plot illustrates the increase in Fe content for the Centre III basic rocks (eucrites, gabbros, and dolerite). The three gabbro pyroxenes that do not plot with the main group are all from the small Meall an Tarmachain gabbro (F in fig. 1).

The two pyroxenes from the dolerite of intrusion (I) (I 11 and I 21 in Table II) are almost identical in composition and show clear evidence of quite substantial iron-enrichment relative to the pyroxenes from the gabbros.

Thus the clinopyroxenes from the Ardnamurchan Centre III basic rocks (the eucrites, gabbros, and dolerites) show evidence of iron-enrichment. The pyroxenes from both eucrites and gabbros show a small range of compositions; pyroxenes from the gabbros, however, are invariably slightly richer in Fe. In the dolerites there is substantial iron-enrichment. This change in composition follows the differentiation trend for tholeiitic magmas, although there is only a small degree of Ca depletion.

The clinopyroxenes from the intermediate rocks, the Tonalite (M) and the Quartz monzonite (N), do not continue the compositional trend shown in the basic rocks. The two pyroxenes analysed (M3 and N4 in Table II) are virtually identical, and as fig. 2 shows, have only slight Fe-enrichment and no Ca depletion, when compared with the pyroxenes from the gabbros. They have substantially lower iron contents than the pyroxenes from the dolerites.

The average SiO_2 content of the Ardnamurchan eucrite rocks is 48·2 %; for the gabbros the average is 48·2 %, for the dolerites (from intrusion I in Table 1) 51·4 %, and for the intermediate tonalites and quartz monzonites 57·1 % (Walsh, 1971). From

TABLE II. *Analyses of clinopyroxenes from the intrusions of Centre III, Ardnamurchan*

	Eucrites					Gabbros									Dolerites		Tonalite	Quartz monzonite	
	E1	E5	E8	G3	H1	A4	B4	J1	J12	J′1	K5	K10	L1	L4	I11	I21	M3	N4	
SiO_2	51·61	50·97	51·44	51·95	52·38	51·74	51·50	50·50	51·28	50·67	51·24	50·07	50·47	50·69	50·64	50·45	51·62	52·26	SiO_2
Al_2O_3	2·63	2·42	2·19	2·36	2·17	1·39	1·57	2·52	1·30	2·46	2·41	4·20	2·54	2·39	1·63	1·65	1·20	0·85	Al_2O_3
TiO_2	0·45	0·76	0·52	0·63	0·51	0·65	0·44	0·87	0·33	0·71	0·64	0·51	0·69	0·86	0·51	0·53	0·24	0·28	TiO_2
Fe_2O_3	1·73	2·26	2·13	1·97	2·16	1·97	1·42	2·16	2·11	1·91	2·23	3·38	2·72	3·19	2·15	2·20	2·27	1·76	Fe_2O_3
FeO	5·92	6·65	6·60	6·36	5·20	7·84	9·56	7·70	9·09	8·78	7·43	6·63	7·37	7·15	13·96	13·82	8·68	9·21	FeO
MnO	0·21	0·24	0·23	0·27	0·21	0·32	0·29	0·29	0·34	0·37	0·31	0·30	0·28	0·27	0·43	0·46	0·77	0·76	MnO
MgO	15·54	15·48	15·36	15·68	16·32	14·70	14·46	13·95	13·80	14·16	14·45	12·80	13·88	14·00	11·04	11·30	13·13	12·90	MgO
CaO	20·84	20·43	20·55	20·18	20·30	20·27	20·04	21·09	21·01	20·44	20·47	21·09	21·30	20·70	18·58	18·52	21·71	20·94	CaO
Na_2O	0·40	0·36	0·37	0·34	0·39	0·52	0·44	0·45	0·38	0·48	0·46	0·37	0·43	0·47	0·40	0·46	0·39	0·37	Na_2O
K_2O	0·04	0·04	0·03	0·04	0·04	0·09	0·07	0·05	0·08	0·09	0·07	0·08	0·05	0·05	0·20	0·16	0·06	0·06	K_2O
TOTAL	99·37	99·61	99·42	99·78	99·68	99·49	99·79	99·58	99·72	100·07	99·71	99·43	99·73	99·77	99·54	99·55	100·07	99·39	
Si	1·917	1·899	1·918	1·922	1·930	1·938	1·934	1·897	1·934	1·899	1·914	1·877	1·894	1·898	1·944	1·937	1·943	1·974	Si
Al^{iv}	0·083	0·101	0·082	0·078	0·070	0·062	0·066	0·103	0·058	0·101	0·086	0·123	0·106	0·102	0·056	0·063	0·053	0·026	Al^{iv}
Al^{vi}	0·032	0·006	0·014	0·025	0·025	0·000	0·003	0·008	0·000	0·008	0·020	0·063	0·006	0·004	0·018	0·012	0·000	0·012	Al^{vi}
Ti	0·013	0·021	0·015	0·018	0·014	0·018	0·012	0·025	0·009	0·020	0·018	0·014	0·020	0·024	0·015	0·015	0·007	0·008	Ti
Fe^{3+}	0·048	0·063	0·060	0·055	0·060	0·056	0·040	0·061	0·060	0·054	0·063	0·095	0·077	0·090	0·062	0·064	0·064	0·050	Fe^{3+}
Fe^{2+}	0·184	0·207	0·206	0·197	0·160	0·246	0·300	0·242	0·287	0·275	0·232	0·208	0·231	0·224	0·448	0·444	0·273	0·291	Fe^{2+}
Mn	0·007	0·008	0·007	0·009	0·007	0·010	0·009	0·009	0·011	0·012	0·010	0·010	0·009	0·009	0·014	0·015	0·025	0·024	Mn
Mg	0·860	0·860	0·854	0·865	0·897	0·821	0·809	0·781	0·776	0·791	0·804	0·715	0·776	0·781	0·632	0·647	0·737	0·727	Mg
Ca	0·829	0·816	0·821	0·800	0·802	0·814	0·806	0·849	0·849	0·821	0·819	0·847	0·856	0·831	0·764	0·762	0·875	0·848	Ca
Na	0·029	0·026	0·027	0·024	0·028	0·038	0·032	0·033	0·028	0·035	0·033	0·027	0·031	0·034	0·030	0·034	0·029	0·027	Na
K	0·002	0·002	0·001	0·002	0·002	0·004	0·003	0·002	0·004	0·004	0·003	0·004	0·002	0·002	0·010	0·008	0·003	0·003	K
Z	2·000	2·000	2·000	2·000	2·000	2·000	2·000	2·000	1·991	2·000	2·000	2·000	2·000	2·000	2·000	2·000	1·996	2·000	Z
XY	2·004	2·009	2·004	1·994	1·993	2·006	2·014	2·010	2·023	2·019	2·002	1·983	2·009	1·999	1·993	2·001	2·013	1·990	XY
Ca	43·2	41·9	42·3	41·7	41·8	42·0	41·2	43·9	43·0	42·3	42·7	45·4	44·1	43·1	40·1	39·7	44·9	44·3	Ca
Mg	44·8	44·2	44·0	45·1	46·7	42·4	41·4	40·4	39·4	40·8	41·9	38·3	40·0	40·6	33·1	33·8	37·8	37·9	Mg
Fe	12·0	13·9	13·7	13·2	11·5	15·6	17·4	15·7	17·6	16·9	15·4	16·2	15·9	16·3	26·8	26·5	17·3	17·8	Fe
α	—	—	1·688	—	—	1·688	—	—	—	—	—	—	—	1·692	1·697	—	1·688	1·689	α
β	—	—	1·694	—	—	1·693	—	—	—	—	—	—	—	1·698	1·703	—	1·694	1·694	β
γ	—	—	1·708	—	—	1·711	—	—	—	—	—	—	—	1·713	1·720	—	1·716	1·716	γ
Sp.Gr.	—	3·342	3·364	—	—	3·353	3·363	3·343	—	—	—	3·370	—	3·354	3·407	—	3·388	3·409	Sp. Gr.
Co	30	30	30	30	45	40	45	30	50	30	30	20	35	30	40	40	20	20	Co
Cr	2000	430	1940	280	370	12	12	130	30	28	52	20	90	20	20	20	28	20	Cr
Cu	28	50	24	21	50	40	12	57	30	12	77	69	62	99	44	79	24	20	Cu
Ga	—	—	10	—	—	10	10	20	—	—	—	15	—	10	10	—	15	20	Ga
Li	5	3	5	4	2	8	4	5	12	—	—	8	6	4	9	—	29	31	Li
Ni	88	120	70	80	120	15	< 10	80	30	10	50	40	78	40	40	35	20	< 10	Ni
Pb	15	50	< 10	< 10	< 10	< 10	65	< 10	< 10	< 10	< 10	15	< 10	< 10	25	10	25	< 10	Pb
Sr	—	20	20	—	—	20	20	25	—	—	—	260	—	20	20	—	25	15	Sr
V	—	—	300	—	—	240	260	330	—	—	—	240	—	370	270	—	—	170	V
Zn	50	70	50	70	70	60	69	58	98	90	80	56	60	56	158	190	179	177	Zn
Zr	—	20	20	—	—	200	50	25	—	—	—	260	—	20	20	—	25	15	Zr

these values, substantial iron-enrichment would be expected in the pyroxenes of the intermediate rocks, if they had formed by a continuation of the fractionation process that is shown in the basic rock pyroxenes.

A significant feature of the pyroxene compositions from Centre III is the close chemical similarity of all the pyroxenes from the eucrite intrusions, and from all the gabbro intrusions. There are no obvious differences between the pyroxenes from the outer gabbros (A4 and B4 in Table II) and the inner gabbros, nor are there any noticeable differences between the fluxion gabbros (B4, K5, K10, L1, and L4 in Table II) and the other gabbros. Electron-microprobe analyses (K. Wills, personal communication) have confirmed this similarity of composition.

TABLE III. *Analyses of clinopyroxenes from the Meall an Tarmachain gabbro (intrusion F in fig. 1)*

	F1	F2	F5		F1	F2	F5
SiO_2	50·33	51·04	51·03	Si	1·907	1·919	1·925
Al_2O_3	2·97	1·78	2·53	Al^{iv}	0·093	0·079	0·075
TiO_2	0·55	0·64	0·61	Al^{vi}	0·040	0·000	0·037
Fe_2O_3	2·80	2·58	2·85	Ti	0·016	0·018	0·017
FeO	10·61	10·63	10·38	Fe^{3+}	0·080	0·073	0·081
MnO	0·37	0·39	0·38	Fe^{2+}	0·336	0·334	0·328
MgO	12·76	14·40	13·12	Mn	0·012	0·012	0·012
CaO	18·29	18·14	18·09	Mg	0·721	0·807	0·738
Na_2O	0·59	0·36	0·42	Ca	0·743	0·731	0·731
K_2O	0·10	0·04	0·08	Na	0·043	0·026	0·031
Total	99·37	100·00	99·49	K	0·005	0·002	0·004
Ca	39·3	37·4	38·7				
Mg	38·1	41·2	39·0	*Z*	2·000	1·998	2·000
Fe	22·6	21·4	22·3	*XY*	1·995	2·004	1·979

For most augites, Ca, Mg, Fe, and Si form over 96 % of the cations present, but, as Table II shows, significant amounts of other elements are found. Al_2O_3 varies in amount in the Centre III pyroxenes from 0·85 % to 4·2 %; it was determined both gravimetrically and by atomic absorption; the values obtained by the latter method, which are invariably lower, are given in Table II and are regarded as substantially more accurate. The Al_2O_3 values are generally low, with only one sample having more than 3·0 % and this pyroxene is from a rock with a high Al_2O_3 content. The lowest values were found in the pyroxenes from the tonalite and quartz monzonite, which are the most SiO_2-rich pyroxene-bearing rocks. A correlation between low Al_2O_3 content of the pyroxene and high SiO_2 of the host rock has been noted previously (Brown, 1967, p. 123). Recalculation of the analyses on the basis of 6 oxygen atoms (Table II) shows that with the exception of K10 pyroxene, most of the Al is present with Si in the tetrahedral site. The Fe_2O_3 content is quite high for clinopyroxenes, often exceeding 2·5 %, and possibly reflecting some alteration of the pyroxenes. Na_2O is consistent and close to 0·4 % for all the pyroxenes, but K_2O, although present in only small amounts, increases in the pyroxenes from eucrites to gabbros to dolerite, and then decreases in the pyroxenes from the intermediate rock, despite the much higher K_2O contents of the rocks. TiO_2 is lower in the pyroxenes from the tonalite and quartz monzonite than the basic rock pyroxenes. MnO increases with differentiation, and increasing Fe content, in the Skaergaard and Bushveld pyroxenes. The Ardnamurchan pyroxenes show a continuous increase in MnO from eucrite to gabbro to dolerite to tonalite and quartz-monzonite pyroxene, despite the lower Fe content of the pyroxenes of the intermediate rocks. It would therefore seem that the simple explanation of the Mn following Fe does not hold for these clinopyroxenes.

The trace-element contents of the Ardnamurchan pyroxenes are given in Table II. The Cr values show clearly the differences between the eucrites and gabbros. The chromium content of igneous pyroxenes is regarded as a good indicator of magmatic sequence and in the Skaergaard and Bushveld pyroxenes, for example, falls rapidly with fractionation. For the Ardnamurchan rocks, the pyroxenes from the eucrites have very much higher values than the gabbros. Similarly nickel falls from eucrite to gabbro pyroxenes, although not as rapidly as chromium.

The pyroxenes from the tonalite and quartz monzonite have higher lithium values than the other pyroxenes and the zinc contents are high (177 and 179 ppm), although the dolerites also have high zinc values.

Three pyroxenes were separated and analysed from the Meall an Tarmachain gabbro (F in fig. 1) and, as mentioned previously, these differ in composition to the pyroxenes from all the other gabbros; the reason for this is not clear: The Meall an Tarmachain gabbro is a small intrusion and its contact relationship to the surrounding eucrites is not well seen. Possibly it is not part of Centre III at all but a remnant of an earlier intrusion. Alternatively the difference in pyroxene chemistry might indicate that it formed by fractionation of the eucrite magma at a different level to the other gabbros.

Biotites. Richey *et al.* (1930) mapped an area in the south of the Great Eucrite as a separate intrusion—the Biotite Eucrite—but Bradshaw (1961) was unable to confirm such a separation. Bradshaw's detailed mapping of the eucrites indicated areas where biotite was found. He showed that biotite occurs sporadically in the eucrites and becomes more important in the Biotite and Inner Eucrites. Biotite occurs in only very small amounts in the eucrites collected for this study.

In the Centre III gabbros biotite is commonly present in small amounts and locally may become more abundant. The term 'Quartz-biotite-gabbro' was used by Richey for the three intrusions denoted J, J′, and J″ (fig. 1) and 'Fluxion Biotite-gabbro' for the Sithean Mor (K) and Glendrian (L) gabbros. Wills (1970) concluded from a petrographic examination of these intrusions that the term Biotite-gabbro was generally not justified. Biotite was found, but modal analysis usually gives figures of only 1 to 2 % biotite, although locally it may become more abundant. The petrographic work done on the samples used in this study, including the modal analysis, would support Wills's conclusion.

In the Quartz-dolerite component of intrusion (I) biotite is quite common (4 % from modal analysis). The Tonalite and Quartz Monzonite are characterized by more abundant biotite (7 % modally). The biotite often occurs as large flakes and in thin section its pleochroic scheme includes a characteristic foxy-red colour.

Biotites were separated from several intrusions and the analyses are given in Table IV. Some of the analyses are not complete because not enough material could be separated for H_2O and ferrous iron determination.

TABLE IV. *Analyses and partial analyses of micas from the intrusions of Centre III, Ardnamurchan*

	Gabbros						Dolerite	Tonalite	Quartz monzonite	
	J1	J23	J′60	J″2	K10	L22	I_D21	M3	N3	N4
SiO_2	—	37·53	37·60	37·40	37·40	37·15	—	37·29	37·84	37·84
Al_2O_3	11·92	—	—	11·97	12·99	13·63	13·39	12·69	12·73	12·59
TiO_2	4·58	4·93	5·87	5·04	5·60	5·03	4·65	5·03	5·12	5·02
Fe_2O_3	22·15*	20·23*	22·31*	20·16*	20·95*	22·32*	22·46*	3·20	2·63	3·14
FeO								14·74	15·26	14·64
MnO	0·14	0·14	0·18	0·18	0·15	0·15	0·17	0·23	0·20	0·23
MgO	11·94	13·07	11·88	12·82	11·95	11·89	11·49	13·16	12·75	13·17
CaO	0·57	0·41	0·27	0·69	0·34	0·26	0·43	0·54	0·60	0·70
Na_2O	0·26	0·23	0·33	0·44	0·30	0·20	0·30	0·37	0·45	0·51
K_2O	6·13	8·14	7·97	7·82	8·50	8·14	8·53	8·40	8·82	8·59
H_2O^+	—	—	—	—	—	2·36	—	3·19	2·32	2·39
H_2O^-	—	—	—	—	—	0·47	—	0·34	0·21	0·45
F	0·53	0·36	0·28	0·86	—	—	—	0·96	0·93	1·03
Total†	—	—	—	—	—	—	—	99·74	99·47	99·87
Fe/Mg	0·94	0·78	0·95	0·79	0·89	0·95	0·99	0·75	0·78	0·74
S.G.	—	—	—	—	—	3·056	—	3·063	3·066	3·061
β	—	—	1·657	—	—	1·659	—	1·653	—	1·654
Trace elements (ppm)										
Ba								2750	2600	2950
Co	80	120	54	20	92	120		80	60	57
Cr	<20	40	<20	<20	<20	55		15	15	15
Cu	—	52	52	65	76	290		49	49	58
Ga	—	—	—	—	—	—		15	20	15
Li	60	60	44	38	36	46		48	142	57
Ni	180	230	56	25	50	95		15	20	20
Pb	<20	15	<20	<20	<20	<10		10	10	30
Sr	—	15	—	10	15	15		15	40	20
V	—	—	—	—	—	—		330	330	360
Zn	455	278	300	615	310	250		320	250	290

* Total Fe as Fe_2O_3. † After subtraction of (O) equivalent of (F).

The analyses in Table IV show that the Ardnamurchan Centre III biotites are of generally similar compositions. The SiO_2 and Al_2O_3 contents do not differ greatly and K_2O remains around 8 % (with the exception of sample AJ1, which may be due to some chlorite impurity). The TiO_2 contents are quite high, and this may be due to the high opaque mineral content of these rocks. CaO and Na_2O are present in only small amounts. However, there are some small differences between the biotites, particularly in the Fe/Mg ratio. The biotites from the gabbros all have lower Fe/Mg ratios than the one biotite from the dolerite (I21 in Table IV). This was confirmed by partial microprobe analysis of two other biotites from the dolerite, which gave Fe/Mg ratios of 1·08 and 1·20.

In contrast to the biotites from the dolerite, the biotites from the tonalite (M3) and quartz monzonite (N3 and N4 in Table IV) have Fe/Mg ratios that are slightly lower than those of the biotites from the gabbros and are certainly lower than those of the biotites from the dolerite.

The relationship between Fe:Mg ratio and SiO_2 content of the host rock has been investigated for various biotites of igneous rocks. Deer (1937) showed that the Fe^{2+}:Mg ratio increases in the more acid rocks from the Carsphairn igneous complex. A similar trend of iron-enrichment in biotite composition has been demonstrated for the Garabal Hill and Glen Fyne complex (Nockolds, 1941), the Southern California batholith (Larsen and Draison, 1950), and the Idaho batholith (Larsen and Schmidt, 1958).

In the intermediate and acid rocks of Ben Nevis, Haslam (1968) has shown that there is no increase in the Fe:Mg ratio with increasing acidity of the rocks. For the biotites of that igneous complex the plot of (total iron)/(total iron+magnesium oxide) against CaO+MgO of the host rock shows a small fall, rather than the increase expected for the iron-enrichment trend. Haslam attributed this reversal of the usual trend to the partial pressure of oxygen remaining nearly constant under the prevailing calc-alkaline conditions.

Wones and Eugster (1965) have shown experimentally that in the crystallization of the assemblage biotite–sanidine–magnetite–melt, if the partial pressure of oxygen remains nearly constant, the Fe:Mg ratio of biotites may fall slightly. If the partial pressure of oxygen decreases, the biotites will show an increase in the Fe:Mg ratio.

The Ardnamurchan biotites show an increase in Fe:Mg ratio from gabbros to dolerites. This sequence may well reflect a decrease in the partial pressure of oxygen, which has been suggested during the differentiation of, for example, the Skaergaard magma (Wager and Brown, 1968).

The tonalites and quartz monzonites show a small decrease in the Fe:Mg ratio relative to the gabbros. It is suggested that this indicates an increase in the partial pressure of oxygen during the crystallization of these intermediate rocks.

In addition to the changes in the Fe:Mg ratio in the biotites, other elements show small differences. The MnO content of the biotites from the tonalite and quartz monzonite is above 0·2 %, in the gabbros it is less than 0·2. This is despite the fact that iron, which Mn is expected to follow, is higher in the biotites from the gabbro. The biotites from Ben Nevis (Haslam, 1969) show an increase in MnO, with 'constant' iron content for the biotites from more acid rocks.

The fluorine content of the biotites from the tonalite and quartz monzonite is significantly higher than that of the biotites in the gabbro, presumably reflecting the increase in volatiles in the magma.

There is not a large amount of published data on the trace elements of igneous biotites and comparisons are difficult to make. The Ba contents of all the biotites are high, and this has been noted in previous studies; Haslam (1969) gives Ba contents for the Ben Nevis biotites ranging from 850 to 2700 ppm. Co is quite high and rather variable. This is similar to Haslam's results; he quotes Co values of 46 to 100 ppm. In the other separated minerals, pyroxenes for example, Co is more consistent and it is not clear why it should have an erratic distribution in the biotites. The Li values are high and show that for these Ardnamurchan rocks, most of the Li is present in biotite. (The Li values are in fact generally lower than values given by Haslam.) Nickel concentrations are variable in the biotites from the gabbros, but are higher than the low values found for the tonalite and quartz monzonite. Lead and strontium are present at below, or close to, the detection limits for each element. Vanadium was determined in the biotites of the intermediate rocks, and was found to be present in quite large amounts. V^{3+} is thought to replace Fe^{3+} and some Fe^{3+} is present in these biotites as it is in the clinopyroxenes. However, the crystal-field stabilization energy for V^{3+} in octahedral coordination is greater than for Fe^{3+}. V would therefore be expected to be present in larger amounts in clinopyroxenes than the biotites, whereas the reverse appears to be the case for these rocks. Zinc is present in substantial concentrations in the biotites, ranging from 250 to 455 ppm.

Specific gravities and refractive indices were measured for some of the biotites, and are generally in agreement with published values (Deer, Howie, and Zussman, 1962, 3). The refractive indices of the two biotites from the gabbros are both slightly higher than for those from the intermediate rocks.

Conclusions. The major- and trace-element chemistry of the clinopyroxenes and biotites supports the conclusion drawn by Richey *et al.* (1930) that the more important intrusions of the area investigated may be attributed to one Centre with a common magmatic origin.

It is suggested that the Centre III intrusions may be grouped together into 'the eucrites', 'the gabbros', 'the dolerites', and 'the intermediate rocks' (the tonalite and quartz-monzonite). The minerals separated from the Great Eucrite (E), the Inner Eucrite (H), and the Biotite-eucrite (G) suggest that these can be considered together as a group. The pyroxenes from all the gabbro intrusions are similar, and different from those from the eucrites. There are no discernible differences between the fluxion gabbros and the quartz gabbros, nor between the Outer gabbros (A and B in fig. 1) and all the Inner gabbros (with the exception of the small Meall an Tarmachain gabbro mentioned earlier).

The pyroxenes from the basic rocks show iron enrichment from eucrite to gabbro, and especially dolerite. This trend is generally similar to that found in the differentiated basic intrusions such as Skaergaard and Bushveld. The intrusions of Centre III are closely associated geologically and there is no reason to believe they did not originate from a single magmatic source. It is therefore suggested that the basic rocks represent three stages in the fractionation of a basic magma. Differentiation must have occurred before the rocks were intruded, probably within the magma chamber from which they originated.

The clinopyroxenes from the intermediate tonalite and quartz-monzonite show that these two intrusions were not formed by a continuation of the differentiation process that formed the basic rocks. If they had, an increase, not a decrease, in iron content would be expected in the pyroxenes. The most obvious alternative to differentiation is assimilation, and it is therefore suggested that the magma that formed the tonalite and quartz-monzonite resulted from the assimilation of country rock into a basic magma. The ring-shaped structure of the intrusion, together with the slow rate of cooling envisaged, would provide favourable conditions for remelting the overlying rock, possibly the Moines. The similarity in composition of the minerals separated from the tonalite and the quartz-monzonite, including the pyroxenes, suggests a very simliar magmatic origin for the two intrusions.

The biotites from the basic rocks also show an increase in Fe content. The Fe/Mg ratio increases from gabbro to dolerite, and then decreases in the tonalite and quartz-monzonite. In fact the Fe/Mg ratio is lower in the biotites from the intermediate rocks than in those from either the dolerite or the gabbros. This is taken as further support for the theory that contamination, not differentiation, produced the tonalite and monzonite magmas.

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