Chemical characteristics of fluorine-bearing biotite of early Paleozoic plutonic rocks from the Sør Rondane Mountains, East Antarctica

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The chemical compositions of biotite grains have been determined for early Paleozoic plutonic rocks from the Sør Rondane Mountains, East Antarctica. The plutonic rocks are divided into three groups of the type-I granitoids, type-II granitoids and the Mefjell Plutonic Complex. Two types of biotite are found in the groups: pale yellow to dark green biotite in the type-I granitoids and brown to dark brown biotite in the type-II granitoids and the Mefjell Plutonic Complex. Mineral chemically, biotites from the type-I granitoids have higher SiO₂ contents, F contents, and \( X_{\text{Mg}} \) values with lower TiO₂ and Cl contents relative to biotites from the type-II granitoids. In the type-II granitoids and the Mefjell Plutonic Complex, biotite strongly enriched in FeO has lower F contents. Biotites in the type-II granitoids and the Mefjell Plutonic Complex have much higher \( \log(f_{\text{H}_2\text{O}})/(f_{\text{HF}}) \) than those from the type-I granitoids. The fluorine content in biotite from the type-I granitoids is similar to that from partial melting A-type granites in Lachlan Fold Belt of eastern Australia; and those from the type-II granitoids and the Mefjell Plutonic Complex are comparable to fractionated aluminous A-type granites from the Lachlan Fold Belt, and the Ambalavayal aluminous A-type granites of South India. Lower fluorine contents in biotite from the type-II granitoids and the Mefjell Plutonic Complex may be mainly controlled by late-magmatic fluid-rock interaction processes associated with melt and may not be indicative of original magma contents. Higher fluorine contents in biotite and whole-rock from the type-I granitoids may reflect fluorine-enriched magmatic source. Our results in biotites from the type-I granitoids as well as geochemistry support the models, which fluorine-rich A-type granites may be derived from partial molten crustal igneous rocks of tonalitic to granodiorite composition.

INTRODUCTION

It has been suggested by several authors (e.g., Loiselle and Wones, 1979; Collins et al., 1982) that the typical F content of biotite in A-type granites indicates a relatively high HF/H₂O ratio in the magma and that A-type granites form at low H₂O fugacities, low to moderate oxygen fugacities and high temperatures. However, low F contents in biotite from aluminous A-type granite have also been reported (King et al., 1997; Rajesh, 2000). The variation in measured fluorine in biotite from A-type granites is considered by Collins et al. (1982) to result from anion exchange with later hydrothermal activity, and may not be indicative of original magma contents.

Recent advances in crystal spectrometer design and operation allow for routine fluorine and chlorine analysis by microprobe, although long counting periods are required. Several researchers have studied F and Cl in biotite from plutonic and metamorphic rocks around the world (e.g., Markle and

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The Sør Rondane Mountains, East Antarctica have a well-documented history dominated by widespread felsic magmatism. Plutonic rocks are as the products of the late-Proterozoic to early-Paleozoic igneous activity and this activity is very important due to being regarded as one part of the Pan-African to post Pan-African events (Sakiyama et al., 1988; Takahashi et al., 1990). Presently, we classified early Paleozoic plutonic rocks into the type-I granitoids (Dufek and Lunckeryggen granitoids), type-II granitoids (Austkampane, Pingvinane, Rogerstoppane and Vikinghøgda granitoids) and the Mefjell Plutonic Complex on the basis of different field occurrence, chemical characteristics and isotopic data. We suggest that this classification is more suitable compared with previous classification of Tainosho et al. (1992) and Arakawa et al. (1994). In this paper, we present chemical characteristics of the biotite from the type-I granitoids, type-II granitoids and the Mefjell Plutonic Complex and use these data to determine their genesis and provide further information of A-type granite.

Fig. 1. Simplified geological map of the Sør Rondane Mountains (modified from Shiraishi et al., 1997). The type-I granitoids include: DG = Dufek granitoid and LG = Lunckeryggen granitoid. The type-II granitoids include: AG = Austkampane granitoid, PG = Pingvinane granitoid, VG = Vikinghøgda granitoid and RG = Rogerstoppane granitoid. MPC = Mefjell Plutonic Complex; LSC= Lunckeryggen Syenitic Complex; MSZ = Main Shear Zone; SRS = Sør Rondane Suture Zone.
**Regional geology and petrography**

The Sør Rondane Mountains (22°E to 28°E, 71.5°S to 72.5°S) in Dronning Maud Land, East Antarctica (Fig. 1), mainly consist of c. 1100–1000 Ma greenschist- to granulite-facies metamorphic rocks, and c. 500 Ma plutonic rocks with minor mafic dykes (Kojima and Shiraishi, 1986; Ishizuka and Kojima, 1987; Sakiyama et al., 1988; Takahashi et al., 1990; Shiraishi et al., 1991; Shiraishi and Kagami, 1992; Asami et al., 1992; Grew et al., 1992; Osanai et al., 1992; Tainosho et al., 1992, 1993; Arakawa et al., 1994; Osanai et al., 1996; Shiraishi et al., 1997; Ikeda and Shiraishi, 1998; Li et al., 2001a, b). Ages of c. 950 Ma and 500–530 Ma by the whole-rock Rb-Sr isochron method were obtained in the plutonic rocks (Takahashi et al., 1990; Tainosho et al., 1992). Geochronological studies indicate that younger plutonism in the Sør Rondane Mountains occurred at 500–530 Ma (Table 1). The region can be divided into two terranes on the basis of metamorphic grade: an amphibolite- to granulite-facies terrane to the northeast and an epidote-amphibolite- to greenschist-facies terrane to the southwest. The Sør Rondane Suture Zone separates the two and the Main Shear Zone cuts across the southwestern terrane (Osanai et al., 1996).

Granulite-facies rocks were produced at peak conditions of 750–850°C and 7–8 kbar for non-mylonitized gneisses and at 530–630°C and 5–5.5 kbar for mylonitized gneisses (Shiraishi and Kojima, 1987; Asami and Shiraishi, 1987; Osanai et al., 1988; Asami et al., 1992).

Several geologists (Sakiyama et al., 1988; Tainosho et al., 1992) have done geological and geochemical studies in older (late Proterozoic) and younger (early Paleozoic) plutonic rocks in the Sør Rondane Mountains. On the basis of field relationships, the Nils Larsen tonalite is the oldest, while the other plutonic units are younger and can be divided into concordant (Austkampane, Pingvinane, Rogerstoppane, Vikinghøgda and Mefjell) granitoids and discordant granitoids (Dufek and Lunckeryggen) by Takahashi et al. (1990). Isotopically and geochemically, early Paleozoic plutonic rocks have also been divided into the volcanic-arc type granitoids (Dufek, Lunckeryggen and Mefjell) and within-plate type granitoids (Austkampane, Pingvinane, Rogerstoppane and Vikinghøgda granitoids) by Tainosho et al. (1992) and Arakawa et al. (1994). The Mefjell Plutonic Complex is separated from the volcanic-arc type granitoids because it has different field occurrence and chemical characteristics (Li et al., 2001b) to the Dufek and Lunckeryggen granitoids.

### Table 1. Summary of geological, geochronological and initial Sr ratio characteristics of early Paleozoic plutonic rocks from the Sør Rondane Mountains

<table>
<thead>
<tr>
<th>Plutons</th>
<th>Related faults</th>
<th>Host rocks</th>
<th>Age (Ma)</th>
<th>Sr</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Type-I granitoids</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dufek</td>
<td></td>
<td>meta-tonalite and gneiss</td>
<td>528 ± 31&lt;sup&gt;b,c&lt;/sup&gt;</td>
<td>0.70372 ± 0.00029</td>
</tr>
<tr>
<td>Lunckeryggen</td>
<td></td>
<td>meta-tonalite</td>
<td>525 ± 31&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.70504 ± 0.00025</td>
</tr>
<tr>
<td><strong>Type-II granitoids</strong></td>
<td></td>
<td></td>
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<tr>
<td>Austkampane</td>
<td>SZ (inferred)</td>
<td>granulite-facies gneiss&lt;sup&gt;a&lt;/sup&gt;</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>Pingvinane</td>
<td>SRS, MSZ</td>
<td>gneiss</td>
<td>510&lt;sup&gt;b&lt;/sup&gt;, 500&lt;sup&gt;d&lt;/sup&gt;</td>
<td>0.70345–0.70680</td>
</tr>
<tr>
<td>Rogerstoppane</td>
<td></td>
<td>gneiss</td>
<td>—</td>
<td>0.7302&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
<tr>
<td>Vikinghøgda</td>
<td>MSZ</td>
<td>gneiss and meta-tonalite</td>
<td>525&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.7067–0.7184</td>
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<tr>
<td><strong>Mefjell Plutonic Complex</strong></td>
<td>SRS</td>
<td>granulite-facies gneiss</td>
<td>506 ± 43&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.70563 ± 0.00055</td>
</tr>
</tbody>
</table>

Note: SZ: Suture zone; MSZ: Main suture zone; SRS: Sør Rondane shear zone.

<sup>a</sup>Sapphirine-bearing gneiss.

<sup>b</sup>Rb-Sr whole-rock isotopic data by Tainosho et al., 1992; Takahashi et al., 1990; Arakawa et al., 1994.

<sup>c</sup>519 ± 98 Ma obtained from the Dufek granitoid (whole-rock Sm-Nd isotopic data, Arakawa et al., 1994).

<sup>d</sup>K-Ar biotite isotopic analysis (Takigami and Funaki, 1991).
The late Proterozoic Nils Larsen Tonalite and the early Paleozoic plutonic rocks are widely distributed. The Nils Larsen Tonalite is exposed in the southern part of the Sør Rondane Mountains, and was affected by regional mylonitization during the greenschist- to epidote-amphibolite-facies metamorphism (Kojima and Shiraishi, 1986). The Nils Larsen Tonalite is a medium- to coarse-grained biotite-hornblende tonalite, composed of plagioclase, quartz, hornblende and biotite with accessory apatite, zircon and magnetite. Hornblende and biotite grains have preferred orientation parallel to the gneissosity in the metamorphic sequence. Early Paleozoic plutonic rocks include the Dufek, Lunckeryggen, Austkampane, Pingvinane, Rogerstoppane and Vikinghøgda granitoids, the Mefjell Plutonic Complex and the Lunckeryggen Syenitic Complex. These were all emplaced after peak metamorphism and deformation (Osanai et al., 1996). The Lunckeryggen Syenitic Complex intruded into the Nils Larsen Tonalite and was itself intruded by the Lunckeryggen granitoid near Lunckeryggen (Sakiyama et al., 1988).

The Dufek and Lunckeryggen batholiths intrude across the gneissosity in the metamorphic rocks, and contain no foliation. The Austkampane, Pingvinane, Rogerstoppane and Vikinghøgda granitoids having gneissosity and schistosity, intrude the metamorphic rocks with contacts parallel to gneissosity and most of them are small in size, mainly occurring as sheets. The field occurrence and petrography of each pluton from the type-I granitoids, type-II granitoids and the Mefjell Plutonic Complex are described below, and their mineral assemblages and photomicrographs are given in Table 2 and Fig. 2 respectively.

**Type-I granitoids** The Dufek granitoid is exposed in an area of approximately 10 × 10 km² in and around Dufekfell (Fig. 1), south of the Sør Rondane Suture Zone (Fig. 1). The pluton intrudes across the gneissosity in the host gneisses and includes abundant xenoliths of tonalite in the north and large blocks of gneiss in the south. The pluton is composed of medium-grained biotite granite and fine-grained biotite hornblende granite. The medium-grained granite consists of plagioclase, K-feldspar, quartz and biotite with accessory amounts of titanite, apatite, zircon and magnetite with or without primary muscovite. The fine-grained biotite granite contains hornblende as long prisms and grains interstitial to K-feldspar. Hornblende typically contains inclusions of zircon, apatite and magnetite. K-feldspar (Or₉₅₋₈₃ Ab₅₋₁₇) is the most abundant mineral, forming tabular microperthitic crystals and granophyric intergrowths with quartz. It is accompanied by normally zoned plagioclase (An₃₃-An₃). Greenish-yellow biotite is mostly interstitial to K-feldspar but may form subhedral grains in the granophyric parts of the rock. Zircon, titanite, apatite and mag-

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**Table 2. Mineral assemblages of early Paleozoic plutonic rocks**

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<th>Qtz</th>
<th>Hbl</th>
<th>Bt</th>
<th>Cpx</th>
<th>Aln</th>
<th>Tn</th>
<th>Ap</th>
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<tr>
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</tbody>
</table>

**Notes:** MPC: Mefjell Plutonic Complex; Kfs: K-feldspar; Zrn: zircon; Grt: garnet; Mag: magnetite; Ilm: ilmenite; Ep: epidote; Ms: muscovite; Pft: fluorite; ++: major mineral; +: minor or accessory mineral; and ±: occasional occurring minerals.
Epidote occurs in some samples, coexisting with titanite. The Lunckeryggen granitoid is exposed in an area of $6 \times 6$ km$^2$ in the southern part of Lunckeryggen in the central part of the Sør Rondane Mountains (Fig. 1). It intrudes into gneissic lithologies, the Nils Larsen Tonalite and the Lunckeryggen Syenitic Complex, and contains angular xenoliths of these rocks. It is composed of massive coarse-grained biotite granite and biotite hornblende granite and is associated with dikes of fine-grained biotite granite. The coarse-grained granite is composed of quartz, K-feldspar, plagioclase and biotite with or without hornblende. Accessory minerals include titanite, apatite, zircon, magnetite and occasionally fluorite. Fluorite is common in the fine-grained biotite granite. Type-II granitoids The Austkampane granitoid is exposed in an area of $1 \times 3$ km$^2$ in the eastern part

![Photomicrographs showing mineral textures. (a) K-feldspar, plagioclase, quartz, biotite, hornblende and fluorite zircon. The type-I granitoids. Single polars. Length of width of the view is 1 mm. (b) K-feldspar, plagioclase, quartz, biotite and garnet. The type-II granitoids. Single polars. Length of width of the view is 2.5 mm. (c) Relationship among K-feldspar perthite, plagioclase, clinopyroxene, hornblende, biotite and quartz. Sympalctite of biotite and quartz and intergrowth of hornblende and quartz are also exhibited. Syenite from the Mefjell Plutonic Complex. Crossed Polars. Length of width of the view is 2.5 mm. (d) Relationship among K-feldspar perthite, plagioclase, quartz and biotite. Granite from the Mefjell Plutonic Complex. Crossed Polars. Length of width of the view is 2.5 mm. Mineral abbreviations are the same as in Table 2.](image-url)
of Austkampane (Fig. 1). This granitoid has a foliation defined by biotite and hornblende. The granitoid consists of quartz, plagioclase, K-feldspar, hornblende and biotite with accessory zircon, apatite, titanite and ilmenite. Plagioclase and hornblende are idiomorphic. Many small granitic sheets intrude into the metamorphic rocks and they are mostly fine- to medium-grained muscovite-bearing granite, containing garnet in places.

The Pingvinane granitoid (Fig. 1) intrudes into gneisses and includes a xenolith of host gneiss. It displays a weak schistosity parallel to the host gneissosity near contacts, but the central part of this granitoid is massive and coarse-grained. The granite is composed of K-feldspar, quartz, plagioclase, hornblende and biotite with or without clinopyroxene, and accessory titanite, apatite, zircon and magnetite.

The Rogerstoppane granitoid occurs at the southern end of Rogerstoppane (Fig. 1). It has a strong gneissosity and is interlayered with mafic gneiss. It contacts parallel to the host gneiss. The granitoid consists mainly of plagioclase, quartz, K-feldspar, biotite and hornblende with accessory allanite, epidote, zircon, apatite and magnetite with or without garnet.

The Vikinghøgda granitoid intrudes into gneiss parallel to the gneissosity (Fig. 1). This granitoid has gneissosity and includes many xenoliths of the host gneiss. It is mainly composed of quartz, plagioclase, K-feldspar, biotite and primary muscovite with accessory titanite, apatite, zircon and magnetite.

**Mefjell Plutonic Complex** The Mefjell Plutonic Complex is exposed in the Mefjell area of the central Sør Rondane Mountains. The Mefjell Plutonic Complex is composed of syenite (including charnockitic syenite) and granite. It was intruded by tonalite and diorite dykes and intruded into two metamorphic terranes.

A detailed petrographic description of the syenite and granite is provided by Li et al. (2001b). Both syenite and granite in the Mefjell Plutonic Complex are composed of K-feldspar, plagioclase, quartz and iron-rich biotite with accessory apatite, zircon, ilmenite, magnetite and titanite. The syenite contains iron-rich clinopyroxene, hornblende, and occasionally olivine, and the granite has occasional hornblende.

**Biotite and hornblende types in early Paleoozoic plutonic rocks**

Magmatic biotite grains occur in most of early Paleoozoic plutonic rocks as euhedral to subhedral flakes and are more or less altered to chlorite, sericite and iron oxide. Biotite is pale yellow to dark green in the type-I granitoids and dark brown to brown in the type-II granitoids and the Mefjell Plutonic Complex. Secondary biotite occurs in the Pingvinane granitoid and the Mefjell Plutonic Complex, and is petrographically distinct from magmatic biotite, occurring as aggregates of fine-grained flakes as product of recrystallization. It partially or completely replaces magmatic hornblende and/or pyroxene and also occurs as symplectites with quartz (Li et al., 2001b).

Hornblende is pale green to green in the type-I granitoids and pale brown to brown in the type-II granitoids and the Mefjell Plutonic Complex. Locally, pale green to green hornblende also occurs in the Rogerstoppane granitoid.

**GEOCHEMISTRY**

Major and trace elements of the samples were determined by a Rigaku 3270E XRF at Kobe University using the analytical methods of Yamamoto and Morishita (1997). Glass beads for major and trace element analyses prepared sample to flux ratios 0.5:5 and 1:2 respectively. The weight loss of the bead during the fusion is smaller than 1%. Rare earth elements were determined using ICP-MS at Shimane University, Japan following the methods described by Kimura et al. (1995) and Roser et al. (2000). Representative bulk chemical data of the type-I granitoids, type-II granitoids and the Mefjell Plutonic Complex are listed in Table 3. Both the type-I and type-II granitoids have a range of silica content (64.25 to 79.38 wt%) and high K$_2$O + Na$_2$O (6.80–11.40 wt%). They fall in the field of alkaline rocks (Fig. 3) on a total alkali-silica diagram (Cox et al., 1979). The type-I
Table 3. Representative bulk chemical compositions of early Paleozoic plutonic rocks

<table>
<thead>
<tr>
<th>Plutons</th>
<th>Type-I granitoids</th>
<th>Type-II granitoids</th>
<th>Mefjell Plutonic Complex</th>
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<td>108</td>
</tr>
<tr>
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<tr>
<td>Eu/Eu*₅ⁿ</td>
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<td>29.7</td>
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<tr>
<td>(La/Yb)₉ⁿ</td>
<td>108.6</td>
<td>137</td>
<td>83.6</td>
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</table>

Note: Fe₂O₃(T): as total of Fe₂O₃; chondrite normalized REE data after Taylor and McLennan (1985); LG: Lunckerygen granitoid; DG: Dufek granitoid; AG: Austkampane granitoid; PG: Pingvinane granitoid; RG: Rogerstoppane granitoid; VG: Vikinghøgda granitoid; MS and MG: syenite and granite from the Mefjell Plutonic Complex respectively.

granitoids have higher Sr and F contents, higher Sr/Ba ratios, and lower Ga/Al ratios than the type-II granitoids (Li et al., 2001a). In the Y vs. Nb and Rb vs. (Y + Nb) discriminative diagrams of Pearce et al. (1984), samples from the Austkampane, Pingvinane, Rogerstoppane and Vikinghøgda granitoids lie in the field of within-plate type granites, while those from the Dufek and Lunckerygen granitoids are volcanic-arc or collisional type granites. On ocean ridge-normalized spiderdiagrams, the type-I granitoids and type-II granitoids differ in Nb, Y and Zr depletion.
Fluorine contents from the type-I granitoids, type-II granitoids and the Mefjell Plutonic Complex are 100–2600, 50–1500 and 100–400 ppm respectively (Fig. 4).

MINERAL CHEMISTRY

Method

Chemical analyses of minerals were obtained using a JEOL-8900M electron probe microanalyzer at the Venture Business Laboratory of Kobe University. Element determinations were carried out using a beam size of 3 µm, an accelerating potential of 15 kV, a probe current of 12 nA, and a counting time of 20 s for each element analyzed except for F and Cl. For fluorine and chlorine analyses, natural pure fluorite (CaF₂) and a granular aggregate of halite crystal (NaCl) were adopted as standards (from Dr. H. Sato, Kobe University).

Although they have similar degrees of Nb-depletion. The type-II granitoids have high concentrations of total REE (264–696 ppm), Y (54.3–76.3 ppm), Nb (19.1–28.9 ppm) and HREEs with moderate to strong negative Eu anomalies. Among the type-II granitoids, the Austkampane and Pingvinane granitoids are weakly fractionated with respect to La/Yb ratios, while the Vikinghøgda and Rogerstoppane granitoids show almost flat MREE and HREE patterns and strongly negative Eu anomalies. The type-I granitoids have low total REE (173–184 ppm), Y (7.8–9.5 ppm) and HREE, and higher LREE/HREE ratios which lack Eu anomalies (Li et al., 2001a).

The syenite and granite of the Mefjell Plutonic Complex are alkaline (Fig. 2), high in FeO/ (FeO+MgO), Zr, Ba and Ga, and low in CaO, MgO, Rb, Y and Nb. The syenite has high LREE concentration with positive Eu anomalies whereas the granite has high LREE/HREE ratios with negative Eu anomalies in chondrite normalized REE diagram (Li, 2002, unpublished Dr. Thesis, Kobe University).

Fluorine contents from the type-I granitoids, type-II granitoids and the Mefjell Plutonic Complex are 100–2600, 50–1500 and 100–400 ppm respectively (Fig. 4).

Fig. 3. Chemical classification diagram of Cox et al. (1979) for early Paleozoic plutonic rocks.
The interference of Fe-Lα line with F-Kα line was removed by calculation. Counting times of the peak and background for F and Cl were set as 100 s and 50 s respectively. Matrix effects during analytical procedure were corrected using the ZAF software provided by JEOL. Accuracy of the fluorine and chlorine analyses examined by measurements of the fluorite and NaCl standards, is within the standard error of the analyses of c. 1 wt% (2 sigma). Detection limits of F and Cl are usually 0.01 wt% (3 sigma). Each microprobe analysis of biotite and hornblende is an average of three spot analyses. OH calculated assumes perfect stoichiometry on the basis of 22 oxygen formula units for biotite. Concentrations of F and Cl in biotite, hornblende, apatite and titanite are listed in Table 4.

**Mineral chemistry**

**Biotite** Representative chemical analyses of biotite are given in Table 5. Biotite analyses from the type-I granitoids, type-II granitoids, and the Mefjell Plutonic Complex have 0.480–0.687, 0.130–0.380 and 0.050–0.330 of Mg/(Mg+Fe) (in molar proportion, the same below) respectively (Fig. 5). The Pingvinane granitoid, a type-II granitoid, has the lowest Mg/(Mg+Fe) ratio (<0.050). The low Na and Mg contents of the biotite from the type-II granitoids and the Mefjell Plutonic Complex are comparable with those for biotites from alkaline rocks (Czamanske et al., 1977).
Biotite grains in type-I granitoids have mostly higher F contents than those in the other granitoids (Fig. 6). Biotites from type-I granitoids show that those from the Lunckeryggen granitoid and Dufek granitoid contain 1.67–2.50 wt% and 0.5–2.5 wt% fluorine respectively. Biotites from type-II granitoids have fluorine contents less than 0.8 wt%. In biotite from type-II granitoids, chlorine contents of less than 0.60 wt% are higher than those of <0.16 wt% in biotite of type-I granitoids (Fig. 6). On the other hand, biotite grains from samples of the Mefjell Plutonic Complex have fluorine contents less than 0.8 wt% (Fig. 6).

### Table 5. Representative chemical analyses of biotite from early Paleozoic plutonic rocks

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<tr>
<th>Sample No.</th>
<th>Type-I granitoid</th>
<th>Type-II granitoid</th>
<th>MPC</th>
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<td>LG</td>
<td>AG</td>
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<tr>
<td>12301B</td>
<td>12301D</td>
<td>11902A</td>
<td>11903B</td>
</tr>
<tr>
<td>68</td>
<td>20</td>
<td>3</td>
<td>59</td>
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<tr>
<td><strong>SiO₂ (wt%)</strong></td>
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<td><strong>Al₂O₃</strong></td>
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<td><strong>FeO(T)</strong></td>
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<td><strong>MgO</strong></td>
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<td>0.00</td>
<td>0.02</td>
</tr>
<tr>
<td><strong>Na₂O</strong></td>
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<td>7.64</td>
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<td><strong>F</strong></td>
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<td><strong>Cl</strong></td>
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<tr>
<td><strong>Total</strong></td>
<td>95.19</td>
<td>95.13</td>
<td>97.40</td>
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</table>

**Notes:** Calculating method for F and Cl is based on stoichiometry; calculated methods of IV(F) and IV(Cl) are from Munoz (1984); the fugacity ratios of log(f_H₂O)/(f_OH₂), log(f_H₂O)/(f_HCl) and log(f_H₂O)/(f_HCl) were calculated using the equations of Munoz (1992) under temperature of 380°C, which are based on the revised coefficients for F-Cl-OH partitioning between biotite and hydrothermal fluid (Zhu and Sverjensky, 1991; 1992); FeO(T) and Fe⁡²⁺ as total FeO and total Fe respectively; MPC: Mefjell Plutonic Complex; AG: Austkampane granitoid; PG: Pingvinane granitoid; RG: Rogerstoppane granitoid; VG: Vikinghøgda granitoid; DG: Dufek granitoid; and LG: Lunckeryggen granitoid.
very low F of less than 0.12 wt% and Cl contents.

Biotites from type-I granitoids have TiO$_2$ contents up to 1.80 wt%, whereas type-II granitoids have a wide range (0.01–3.60 wt%) of TiO$_2$, with the highest TiO$_2$ in the Austkampane and Pingvinane granitoids. Biotites from the Mefjell Plutonic Complex have higher TiO$_2$ contents (1.80–4.48 wt%). Biotite grains from type-I granitoids have TiO$_2$ contents comparable to those from the Nils Larsen Tonalite (Tainosho et al., 1993).

Hornblende Hornblendes from type-I granitoids have MgO contents of 7.80–12.68 wt% and fluorine contents of 0.45–1.22 wt%, which correspond to Mg/(Mg+Fe) ratios of 0.463–0.621 and F/(F+Cl+OH) ratios of 0.228–0.305. Hornblendes from the type-II granitoids have lower MgO contents of 0.08–0.41 wt% and F contents of 0.20–0.38 wt%. Hornblende from the Mefjell Plutonic Complex has up to 0.13 wt% in F and MgO/(MgO+FeO) ratios of 0.05–0.28 (11 samples). Chlorine contents are lower in hornblendes from the Lunckeryggen granitoids (0.20–0.30 wt%) than in those from the Pingvinane granitoids (0.31–0.35 wt%) in Table 4. K$_2$O contents in hornblendes from the type-II granitoids are much higher than in those from the type-I granitoids. Al$_2$O$_3$ contents in hornblende from the type-II granitoids and the Mefjell Plutonic Complex are higher (9–13 wt%) than those in the type-I granitoids (8–9 wt%). Hornblendes from the type-II granitoids with high Al$_2$O$_3$ contents are comparable to other anorogenic granites (Anderson, 1983).
Feldspar, apatite and titanite  Anorthite contents of plagioclase in the type-II granitoids (An_{5-15}) are lower than those in the type-I granitoids (An_{33} in the core to An_{3} at the rim in zoned crystals). Plagioclase in the Mefjell Plutonic Complex ranges from An_{25} in the core to An_{5} at the rim.

Chemical compositions of K-feldspar are similar between early Paleozoic plutonic rocks, and lie within the formula Or_{88–98}Ab_{2–12}.

Apatite occurs in the matrix of granitoids or as inclusions in plagioclase. Apatites from the type-II granitoids have higher fluorine contents, ranging from 3.21 to 7.20 wt%, compared to those from type-I granitoids (1.22–3.60 wt%, Table 4). Concentration of fluorine in apatite from the Mefjell Plutonic Complex ranges from 3.21 to 4.11 wt%.

Titanite has a low fluorine content in the Mefjell Plutonic Complex (0.23–0.50 wt%), less than 2.28 wt% in the type-I granitoids and relatively high (1.85–2.78 wt%) in the type-II granitoids (Table 4).

Biotite halogen chemistry and hydrothermal fluid fugacity ratios from early Paleozoic plutonic rocks

The halogen contents of biotite from the early Paleozoic plutonic rocks are shown in Table 5. Biotites from the type-I granitoids, type-II granitoids and the Mefjell Plutonic Complex have log(\(\frac{X_C}{X_{OH}}\)) values of –2.6 to –1.8, –1.3 to –3.2 and –1.6 to –3.2, and log(\(\frac{X_F}{X_{OH}}\)) values of –0.4 to –1.4, –0.7 to –1.9 and –1.3 to –2.9 respectively.

The fluorine and chlorine data in biotite were used to calculate log(\(f_{H_2O}/f_{HF}\)), log(\(f_{H_2O}/f_{HCl}\)), and log(\(f_{HCl}/f_{HF}\)) ratios for hydrothermal fluids (Munoz, 1992). The fugacity ratios were calculated using the equations of Munoz (1992), which are based on the revised coefficients for F-Cl-OH partitioning between biotite and hydrothermal fluid (Zhu and Sverjensky, 1991, 1992). These equations are:

\[
\log(\frac{f_{H_2O}}{f_{HF}}) = 1000 / T \left(2.37 + 1.1X_{Mg\text{ in biotite}}\right) + 0.43 - \log(\frac{X_F}{X_{OH}})_{\text{biotite}}
\]

\[
\log(\frac{f_{H_2O}}{f_{HCl}}) = 1000 / T \left(1.15 + 0.55X_{Mg\text{ in biotite}}\right) + 0.68 - \log(\frac{X_C}{X_{OH}})_{\text{biotite}}
\]

and

\[
\log(\frac{f_{HF}}{f_{HCl}}) = -1000 / T \left(1.22 + 1.65X_{Mg\text{ in biotite}}\right) + 0.25 + \log(\frac{X_F}{X_C})_{\text{biotite}}
\]

where

\[
X_{Sid} = [(3 – Si/Al)/1.75] (1 – X_{Mg})
\]

\[
X_{An} = 1 – (X_{Mg} + X_{Sid})
\]

and

\[
IV(Cl) = -5.01 – 1.93X_{Mg} – \log(\frac{X_{Cl}}{X_{OH}})
\]

The type-I granitoids, type-II granitoids and the Mefjell Plutonic Complex have 1.2 to 2.3, 1.2 to 2.4 and 1.5 to 3.5 in IV(F), and 4.2 to 6.2, 4.1 to 5.5 and 3.9 to 6.7 in IV(F/Cl) respectively.

Fluorine and chlorine data in biotite were used to calculate log(\(f_{H_2O}/f_{HF}\)), log(\(f_{H_2O}/f_{HCl}\)), and log(\(f_{HCl}/f_{HF}\)) ratios for hydrothermal fluids (Munoz, 1992). Fugacity ratios were calculated using the equations of Munoz (1992), which are based on the revised coefficients for F-Cl-OH partitioning between biotite and hydrothermal fluid (Zhu and Sverjensky, 1991, 1992). These equations are:

\[
IV(F)_{\text{biotite}} = 1.52X_{Mg} + 0.42X_{An} + 0.20X_{Sid} – \log(\frac{X_F}{X_{OH}})
\]

and

\[
IV(F/Cl)_{\text{biotite}} = IV(F) – IV(Cl)
\]
**DISCUSSION**

**Petrogenetic significance of halogens in biotite and hornblende**

The chemical data indicate that the early Paleozoic plutonic rocks of the Sør Rondane Mountains were derived from different fluorine-bearing source materials and/or crystallized under different conditions. Biotite and hornblende in the type-I granitoids generally have higher F and lower Cl contents at intermediate $X_{Mg}$ compared to the type-II granitoids. Biotite and hornblende in samples from the Mefjell Plutonic Complex have very low F and relatively higher Cl contents at low $X_{Mg}$. These indicate that a variation in fluorine content is associated with $X_{Mg}$ in biotite from early Paleozoic plutonic rocks. Furthermore, the high fluorine content in biotite from the type-I granitoids may indicate a relatively high HF/H$_2$O ratio and low-water fugacity in the source material. These features indicate that the Mefjell Plutonic Complex has a closer genetic affinity with the type-II granitoids than the type-I granitoids.

TiO$_2$ content in biotite is lower in the type-I granitoids than the type-II granitoids and rocks of the Mefjell Plutonic Complex. Anorthite contents of plagioclase are higher in the type-I granitoids than those from the type-II granitoids and the Mefjell Plutonic Complex. The Rogerstoppane granitoid has much higher fluorine contents in apatite than those from the Dufek granitoid. The reason for higher concentration of fluorine content in biotite with lower in apatite from the type-I granitoids compared to those from the type-II granitoids is not clear.

High-T stability of biotite is attained in water under-saturated conditions (Munoz, 1984). An increase of TiO$_2$ can further increase high-T stability. The type-I granitoids have high fluorine contents with constant amounts of TiO$_2$ in biotite consistent with relatively high temperature condition for biotite.

Micas provide an important residence for halogens. In synthetic trioctahedral micas, the OH group may be completely replaced by F, and even in natural biotites the molar ratio F/(OH+F+Cl)
may approach unity. In strong contrast, the amount of Cl substitution is very low [Cl/(OH+F+Cl) < 0.1] (Munoz, 1984). Mg and Fe contents in biotite are directly related to F-Cl-OH exchange between biotite and hydrothermal fluids (Munoz, 1984). Hydroxyl-bearing ferromagnesian silicate minerals with high $X_{Mg}$ tend to incorporate more F than comparable minerals with lower $X_{Mg}$. Biotites from the type-II granitoids and the Mefjell Plutonic Complex (Li, 2002) are Fe-rich and have relatively low F contents and those having higher $X_{Mg}$ values with less Cl in the type-I granitoids (Fig. 6), indicating that this is consistent with the substitution behaviour described by Munoz (1984). The scatter in log($X_{Cl}/X_{OH}$) and log($X_{F}/X_{OH}$) in biotites may be caused by a number of factors: fluid composition, the presence or absence of other halogen-bearing phases in the granitoid, local inhomogeneity of melt composition and the presence of volatile-rich fluids in the latest stages of granitoid crystallization.

The type-I granitoids generally have higher log($f_{HF}/f_{HCl}$) and lower log($f_{H,O}/f_{HF}$) values than those from the type-II granitoids and the Mefjell Plutonic Complex (Fig. 7). This is consistent with the formation of the type-I granitoids under lower water fugacity conditions than the type-II granitoids and the Mefjell Plutonic Complex. Evidence for A-type granites and a possibility for genetic explanation of the type-I granitoids

A-type granitic magmas typically contain higher amounts of fluorine and chlorine than I-type granitic magmas (Loiselle and Wones, 1979; Collins et al., 1982). Collins et al. (1982) suggested that if fluorine is relatively low in the melt, early amphibole crystallization may not occur and anorthite-rich plagioclase is the dominant crystallizing phase, resulting in the “plagioclase-effect” of Bowen (1945) and the peralkaline trend. Fluorine concentration in the melt is therefore considered to be critical in determining fractionation trends in A-type magmas. Most of early Paleozoic plutonic rocks from the Sør Rondane Mountains, East Antarctica are A-type granites (Tainosho et al., 1992), and they provide evidence that A-type granites contain low F contents in biotite. Biotites in syenite and granite from the Mefjell Plutonic Complex and the type-II granitoids have similar characteristics to those in fractionated aluminous A-type granites. The biotite and hornblende from the Mefjell Plutonic Complex are also suitable to use the explanation of co-precipitation of hornblende and biotite based on mineral texture, and low F contents in both biotite and hornblende might be consistent with the absence of fluorite as suggested by Rajesh (2000). Lower fluorine contents in biotite from the type-II granitoids and the Mefjell Plutonic Complex support that they may be mainly controlled by late-magmatic fluid-rock interaction process associated with melt (Li et al., 2001a).

Li et al. (2001a) suggested one possibility that the type-I granitoids from the Sør Rondane Mountains are derived from partial melting of the Nils Larsen Tonalite on the basis of geochemical and isotopic associations between the Nils Larsen Tonalite and the type-I granitoids. Furthermore, fluorine contents in biotite from the type-I granitoids of the Sør Rondane Mountains are similar to those from A-type granites derived from partial melting of a crustal source (Collins et al., 1982), and differ from fractionated aluminous A-type granites from the Lachlan Fold Belt, eastern Australia and elsewhere.

Several mechanisms have been postulated to explain the generation of A-type granites (see Clemens et al., 1986 for review), but the most cited model is that A-type granites are derived by the high-temperature melting of rocks containing F-enriched biotite and/or amphibole (Collins et al., 1982; Clemens et al., 1986; Whalen et al., 1987; Peterson et al., 1991). The most popular variant of this model holds that A-type granites are derived by partial melting of residual material that remains from production of I-type granites. Creaser and White (1991) argue against this variant of the model, and suggest that A-type granites may be derived from the partial melting of crustal rocks of tonalitic to granodioritic composition. Skjerlie and Johnston (1993) have done fluid-absent melting experiments on biotite (20 wt%) and...
hornblende (2 wt%) bearing tonalite gneiss. Their experiments show that the dehydration-melting of the tonalite gneiss that contains F-rich biotite produces F-rich granitic liquids with compositions within the A-type range. Their study supports the notion that A-type granites can be generated by H₂O-undersaturated melting of rocks of tonalitic composition (Creaser and White, 1991). Chemical affinity of major element and halogen chemistry in biotite and hornblende between the type-I granitoids and the Nils Larsen Tonalite (Table 4) further supports the arguments by Creaser and White (1991), Skjerlie and Johnston (1993) and Li et al. (2001a).

**SUMMARY**

(1) The chemical compositions of biotites have been systematically determined for early Paleozoic plutonic rocks from the Sør Rondane Mountains, East Antarctica. Two types of biotite from them were divided on the basis of color: pale yellow to dark green in the type-I granitoids and brown to dark brown in the type-II granitoids and the Mefjell Plutonic Complex.

(2) Chemical compositions of biotite from the type-I granitoids are distinctly different from that of biotite in the type-II granitoids and Mefjell Plutonic Complex. Biotites from the type-I granitoids have higher SiO₂ and F contents, and X_Mg (in molar proportions) with lower TiO₂ and Cl relative to biotite from the type-II granitoids and the Mefjell Plutonic Complex. X_F, X_Cl, X_OH and X_Mg in biotite have different in early Paleozoic plutonic rocks, reflecting change in such fluid composition and varied temperature condition during the crystallization history of the magma. Biotite in the type-II granitoids and Mefjell Plutonic Complex has much higher log (f_(H₂O))/(f_HF) than that from the type-I granitoids, indicating different magmatic or hydrothermal fluid components and condition.

(3) Compared with A-type granites worldwide, the fluorine content in biotite from the type-I granitoids is similar to that from partial molten A-type granites, whereas those from the type-II granitoids and the Mefjell Plutonic Complex are comparable to fractionated aluminous A-type granites. Our results in biotites from the type-I granitoids as well as geochemistry support the models, which fluorine-rich A-type granites may be derived from partial molten crustal igneous rocks of tonalitic to granodiorite composition.

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