

## AMMONIUM CONTENT OF BIOTITES FROM GRANITIC AND METAMORPHIC ROCKS IN THE SØR RONDANE MOUNTAINS, EAST ANTARCTICA

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**Abstract:** Ammonium content of biotites from granitic and metamorphic rocks in the Sør Rondane Mountains in East Antarctica has been determined in order to examine the difference in the  $\text{NH}_4^+$  content of biotites among six granite types. Ammonium content of biotites ranges from 12 to 23 ppm (average, 18 ppm) in the migmatitic granite, 23 to 80 ppm in the foliated granite (average, 46 ppm), 22 to 86 ppm (average, 66 ppm) in the massive granite with migmatitic margin, 11 to 66 ppm (average, 42 ppm) in the discordant granite, 28 to 106 ppm (average, 67 ppm) in the granite sheet and dyke, 76 to 109 ppm (average, 92 ppm) in the small granodiorite bodies, and 25 to 142 ppm (average, 52 ppm) in the metamorphic rocks. High  $\text{NH}_4^+$  content found in biotites from the small granodiorite bodies is probably due to a result of the interaction between the granitic magma and surrounding metamorphic rocks. Biotites from the migmatitic granite have lowest  $\text{NH}_4^+$  content in granitic rocks analyzed. The fact is in discord with the field evidence. The very low  $\text{NH}_4^+$  content in biotite from the Nils Larsen tonalite is in good accordance with the view postulated from petrochemical data that granitic magma of this tonalite was directly derived from the melting of subducted oceanic crust or the overlying mantle. In metamorphic rocks, the low  $\text{NH}_4^+$  content in biotites is probably due to liberation of  $\text{NH}_4^+$  from the rocks during high grade metamorphism (granulite facies).

### 1. Introduction

ITIHARA and HONMA (1979, 1983) examined the ammonium content of biotites in Cretaceous granitic and metamorphic rocks, with the view that the ammonium content of biotite may be more suitable for discrimination between meta-igneous and meta-sedimentary rocks. The results show that biotites from meta-sediments are high in their ammonium content. The high amount of ammonium of biotites from meta-sediments has been considered to be inheritance from organic matter in the original sediments. From this viewpoint, the present study has been carried out for granitic and

metamorphic rocks in the Sør Rondane Mountains, East Antarctica. The purpose of the study is to obtain geochemical data of nitrogen for plutonic and metamorphic rocks in East Antarctica. Samples were collected from rocks of the Sør Rondane Mountains and ammonium content of biotites of the rocks was determined.

## 2. Geological Setting of Biotite Samples

Geology of the Sør Rondane Mountains, East Antarctica, has been described by KOJIMA and SHIRAISHI (1986), ISHIZUKA and KOJIMA (1987), SHIRAISHI and KOJIMA (1987), ASAMI and SHIRAISHI (1987), ASAMI *et al.* (1989), GREW *et al.* (1989) and OSANAI *et al.* (1990). Figure 1 shows locations of the rocks used in this study. The Sør Rondane Mountains consist of metamorphic and granitic rocks. The metamorphic rocks of the Sør Rondane Mountains are divided into Talet-Vengen Group and Nils Larsen Group. The Talet-Vengen Group is exposed in central to eastern area and is composed mainly of pelitic and psammitic gneisses. They have been subjected to the granulite-to upper amphibolite-facies metamorphism (KOJIMA and SHIRAISHI, 1986, ISHIZUKA and KOJIMA, 1987; ASAMI *et al.*, 1989). Pressures and temperatures calculated from mineral compositions are  $\geq 7$  kbar and 700–750°C for early granulite facies event and 500–600°C for amphibolite facies event (GREW *et al.*, 1989).

The Nils Larsen Group is composed mainly of gneissose tonalite and basic schist which have been subjected to mylonitization under the conditions of the green schist to epidote amphibolite-facies (KOJIMA and SHIRAISHI, 1986). Plutonic rocks in the Sør Rondane Mountains are divided into six rock types based on the field occurrences, particularly along the boundary with the surrounding rocks (TAKAHASHI *et al.*, 1990b). These types are; 1. migmatitic granite, 2. foliated granite, 3. massive granite with migmatitic margin, 4. massive granite with discordant boundary (discordant granite), 5. granite sheet and dyke, 6. small granodiorite bodies. These granitic rocks intruded after the regional mylonitization represented by the Main Shear Zone except for migmatitic granite and foliated granite (SAKIYAMA *et al.*, 1988).

Migmatitic granite consists of well-banded or more homogeneous, biotite-streaked granite. The Vikinghogda granite, Bergersen granite and Vengen granite are included in this type (Fig. 1). Mafic minerals of the foliated granite show a parallel arrangement giving the rock gneissosity. The granite of this type intruded concordantly into the gneiss. The Austkampane granite and Mefjell granite are included in this type (TAKAHASHI *et al.*, 1990b). The Austkampane granite is coarse-grained hornblende-biotite granite. The Mefjell granite is medium-grained hornblende-biotite granite. Massive granite with migmatitic margin is equigranular and massive in main parts and has a migmatitic part along the boundary with the gneiss. The Pingvinane granite belongs to this type. Discordant granite is mostly massive and intrudes into the gneiss with a clear boundary. It often has leucocratic margin which is deduced to be chilled margin. The Lunckeryggen granite and Dufek granite belong to this granite type. Granite sheet and dyke are found all over the area, composed of granite and granodiorite. Small granodiorite bodies are medium-grained hornblende-biotite granodiorite. The relationship to the surrounding rocks is not clear. The Rogerstopane granite belongs to this rock type. The granites of these six types have emplaced during a short

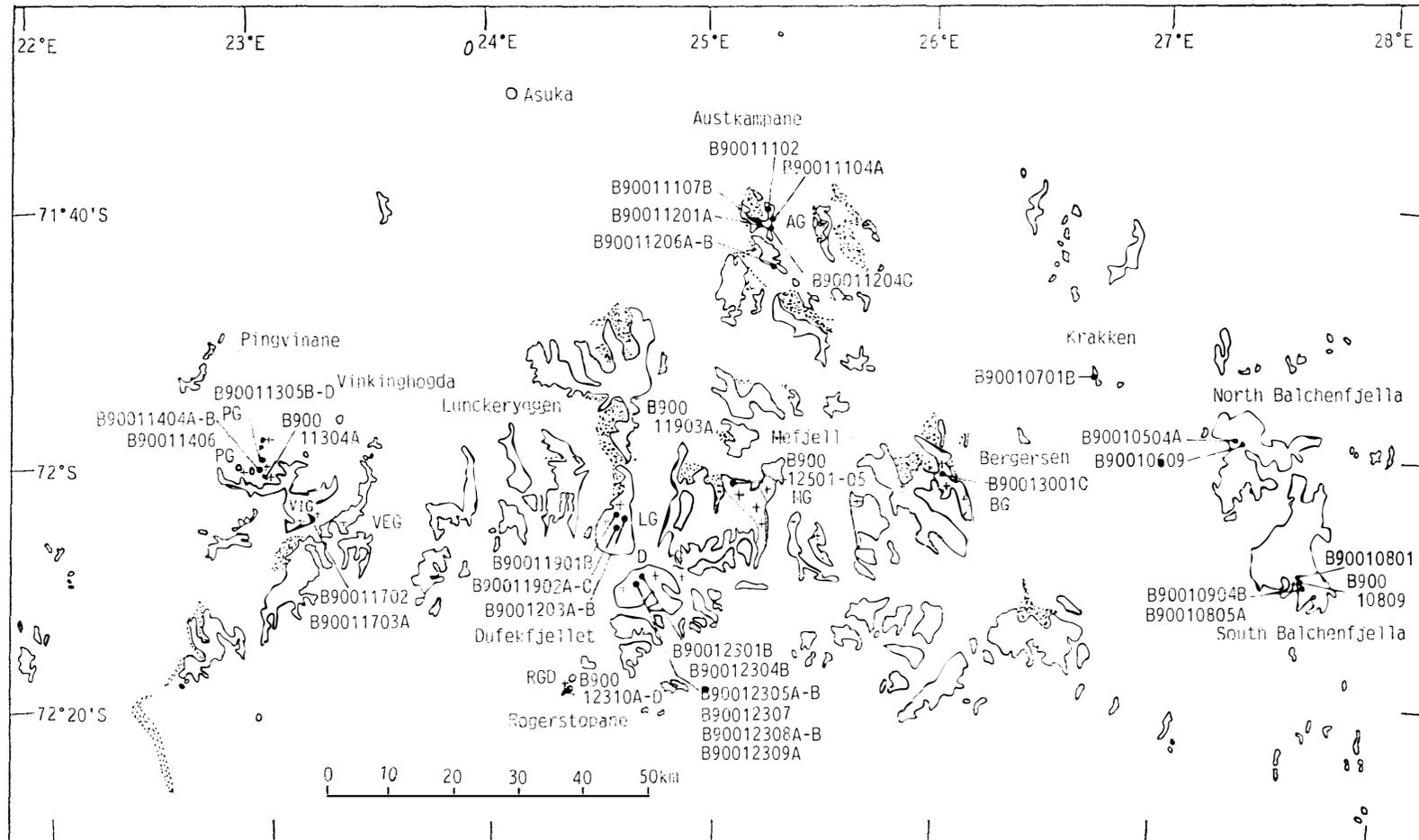


Fig. 1. Map showing distribution of the granitic rocks (TAKAHASHI *et al.*, 1990b) and location of the rocks used for biotite analyses. Numbers show sample localities and correspond to those listed in Table 1. Dotted symbols represent moraine, crossed symbols represent the granitic stocks, and the other parts bounded by solid line show metamorphic rocks.

VIG: Vinkinghogda granite, VEG: Vengen granite, PG: Pingvinane granite, LG: Lunckeryggen granite, DG: Dufek granite, MG: Mefjell granite, AG: Austkampane granite, BG: Bergersen granite, RGD: Rogerstopane granite.

period of 550 to 500 Ma (TAKAHASHI *et al.*, 1990a; TAKIGAMI and FUNAKI, 1990). The granites of these six types are characterized by high alkali, Zr and Nb content and low CaO content. Many of the granites belong to A-type granites (TAINOSHO *et al.*, 1991).

The Nils Larsen tonalite has foliation parallel to the structure of the surrounding rocks. It has a distinctively high MgO, CaO and low alkalis, and belongs to M-type granite based on chemical and petrographic features (TAINOSHO *et al.*, 1991). The age of the Nils Larsen tonalite is about 960 Ma (TAKAHASHI *et al.*, 1990a).

### 3. Analytical Techniques

Biotite was separated using an isodynamic separator and heavy liquids. The purity of samples was more than 90%. The ammonium content of biotite was determined by the sealed tube digestion method of STEVENSON (1960) who proposed this method for the determination of nitrogen in rocks and minerals. The procedure involves the digestion of specimens in concentrated H<sub>2</sub>SO<sub>4</sub> in sealed tubes at a temperature 420°C for two hours, after which the ammonium is distilled in a micro-kjedahl unit and estimated colorimetrically with Nessler's reagent. According to STEVENSON (1962), about 90% of the total nitrogen in biotite is fixed in crystal structure as ammonium nitrogen.

### 4. Results

#### 4.1. Biotite from granitic rocks

The analytical results are tabulated in Table 1. Ammonium content of biotites from migmatitic granite ranges from 12 to 23 ppm, an average being 18 ppm. These values are very low compared with those in other types of granite (Fig. 2). Sample B90011702 in Table 1 was obtained from gneissose fine-grained biotite granite which occurs along the Main Shear Zone and shows low NH<sub>4</sub><sup>+</sup> content. Contact relation between this granite and metamorphic rocks is clear. Ammonium content of biotites from foliated granite ranges from 23 to 80 ppm and the average is 46 ppm. The content is within the range of NH<sub>4</sub><sup>+</sup> content of biotites from S-type granite in Australia (Fig. 2) except for sample B90011107B, the Austkampane granite which shows a high NH<sub>4</sub><sup>+</sup> content, 80 ppm (Table 1). These values are slightly lower than those of the small granodiorite bodies. The Austkampane granite is a small intrusive body and contains biotite gneiss xenoliths. While the Mefjell granite intrudes into the gneiss with a clear boundary. Ammonium content of biotites from massive granite with migmatitic margin ranges 22 to 86 ppm, 66 ppm on the average. Among the samples of this group, sample B90011405B was obtained from migmatitic granite which occurs in the marginal part of the pluton. This sample shows low NH<sub>4</sub><sup>+</sup> content. Other samples have high NH<sub>4</sub><sup>+</sup> content (Table 1). Ammonium content of biotites from discordant granite ranges from 11 to 66 ppm and the average is 42 ppm. The content is within the range of NH<sub>4</sub><sup>+</sup> content of biotites from S-type granite in Australia (Fig. 2) except for B90012301B which shows a low NH<sub>4</sub><sup>+</sup> content, 11 ppm. Among the samples of this granite type, NH<sub>4</sub><sup>+</sup> content of the Dufek granite is slightly higher than those of the Lunckeryggen granite (Table 1). The Dufek granite has often dark inclusions. Ammonium content of

Table 1.  $NH_4^+$  content of biotites with description of the rocks. Average values are from ammonium content of the biotites.

Sample No.	Description of the rocks	Locality	$NH_4^+$ (ppm)	av.
Migmatitic granite				
B 90011702	gneissose fine-grained Bi granite	Vikingshogda	12	18
B 90011703 A	gneissose fine-grained Bi granite	(ditto)	5*	
B 90013001 C	fine-grained granite	Bergersen	23	
Foliated granite				
B 90011107 B	medium-grained Bi granite	Austkampane	80	46
B 90011206 A	gneissose Hb-Bi granite	(ditto)	8*	
B 90012502	gneissose Hb-Bi granodiorite	Mefjell	36	
B 90012504	Hb-Bi granodiorite (shear part)	(ditto)	45	
B 90013001 C	fine-grained granite	(ditto)	23	
B 90012501	gneissose Hb-Bi granodiorite	(ditto)	10*	
B 90012505	gneissose Hb-Bi granodiorite	(ditto)	18*	
Massive granite with migmatitic margin				
B 90011405 B	migmatitic granite	Pingvinane	22	66
B 90011305 B	migmatitic granite	(ditto)	18*	
B 90011305 D	coarse-grained Bi granite	(ditto)	81	
B 90011406	coarse-grained Bi-Hb granite	(ditto)	86	
B 90011304 A	Bi granite	(ditto)	9*	
Discordant granite				
B 90011902 A	medium-grained Hb-Bi granite	Lunckeryggen	41	42
B 90011902 B	fine-grained Bi granite	(ditto)	49	
B 90012003 B	Bi granite	(ditto)	28	
B 90012301 B	medium-grained Hb-Bi granite	Dufek	11	
B 90012304 B	medium-grained Hb-Bi granite	(ditto)	38	
B 90012305 A	fine-grained Bi granite	(ditto)	66	
B 90012305 B	medium-grained Bi granite	(ditto)	49	
B 90012307	mylonitixed granite	(ditto)	55	
B 90012308 A	coarse-grained Bi granite	(ditto)	54	
B 90012308 B	fine-grained Bi granite	(ditto)	12*	
B 90012309 A	coarse-grained Bi granite	(ditto)	1*	
Granite sheet and dyke				
B 90010609	fine-grained Bi granite	North Balchen	28	67
B 90011204 C	fine grained Bi granite	Austkampane	106	
Small granodiorite bodies				
B 90012310 A	Hb-Bi granodiorite	Rogerstopane	109	92
B 90012310 C	Hb-Bi granodiorite	(ditto)	76	
B 90012310 D	Hb-Bi granodiorite	(ditto)	90	
Nils Larsen tonalite				
B 90011901 B	gneissose Hb-Bi tonalite	Lunckeryggen	24	19
B 90011902 C	gneissose Hb-Bi tonalite	(ditto)	30	
B 90011903 A	gneissose Hb-Bi tonalite	(ditto)	10	
B 90012003 A	gneissose Hb-Bi tonalite	(ditto)	13	
Metamorphic rocks				
B 90010504 A	OPX-Ga-Hb-Bi gneiss	North Balchen	25	29
B 90010809	OPX-Hb-Bi gneiss	South Balchen	34	
B 90010701 B	Bi gneiss	Krakken	26	67
B 90010801	Bi gneiss	South Balchen	142	
B 90011104 A	Ga-Bi gneiss	Austkampane	74	
B 90011201 A	Hb-Bi gneiss	(ditto)	44	
B 90011404 A	coarse-grained Bi gneiss	Tanngarden	48	
B 90010805 A	Hb gneiss	South Balchen	4*	
B 90010904 B	amphibolite	South Balchen	36	
B 90011102	amphibolite	Austkampane	36	

Abbreviation for minerals; Bi: biotite, Hb: hornblende, Ga: garnet, OPX: orthopyroxene. Star symbol (\*) shows whole rock sample.

of biotites from granite sheet and dyke ranges widely from 28 to 106 ppm, with the average 67 ppm. In the Austkampane area, granitic sheet has high  $\text{NH}_4^+$  content, 106 ppm (Table 1). This rock concordantly intruded into gneiss and has gneiss xenoliths. Ammonium content of biotites from small granodiorite bodies ranges from 76 to 109 ppm, showing distinctively high  $\text{NH}_4^+$  content compared with those of other granite types. These rocks have weakly banded structure.

Biotite from the Nils Larsen tonalite has distinctively low  $\text{NH}_4^+$  content compared with those of other types of granitic rocks. The average is 19 ppm and the range is from 10 to 30 ppm. This range is within the range obtained from granites in non-metamorphic terrains of Japan (Fig. 2).

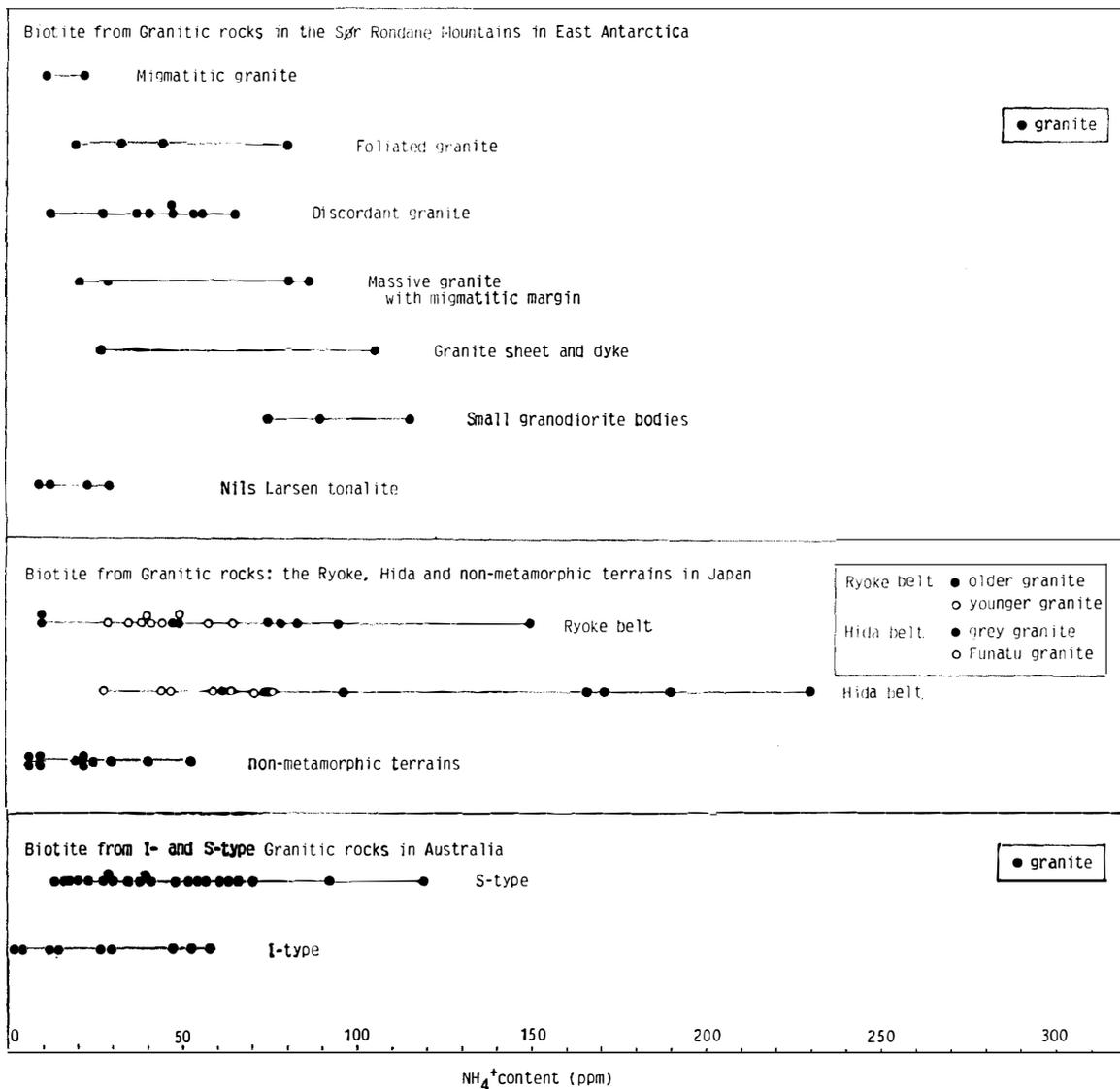


Fig. 2. Ammonium content of biotites from granitic rocks. Data for the Australian granitic rocks are from TAINOSHO and ITIHARA (1988). Data for granitic rocks in the Ryoke belt and non-metamorphic terrains are from ITIHARA and HONMA (1979). Data for granitic rocks in the Hida belt are from TAINOSHO and ITIHARA (1991).

#### 4.2. Biotite from metamorphic rocks

Ammonium content of biotites from metamorphic rocks in the Sør Rondane Mountains ranges from 25 to 142 ppm (Fig. 3), and the average is 52 ppm. Among the samples,  $\text{NH}_4^+$  content of biotites from hornblende-biotite gneiss (amphibolite facies) ranges 26 to 142 ppm, the average being 67 ppm. Ammonium content of biotites from orthopyroxene-hornblende-biotite gneiss (granulite facies) ranges 25 to 34 ppm, 29 ppm on the average. These values are low compared with those of hornblende-biotite gneiss. Ammonium content of biotites from two kinds of amphibolite is both 36 ppm. This value is low.

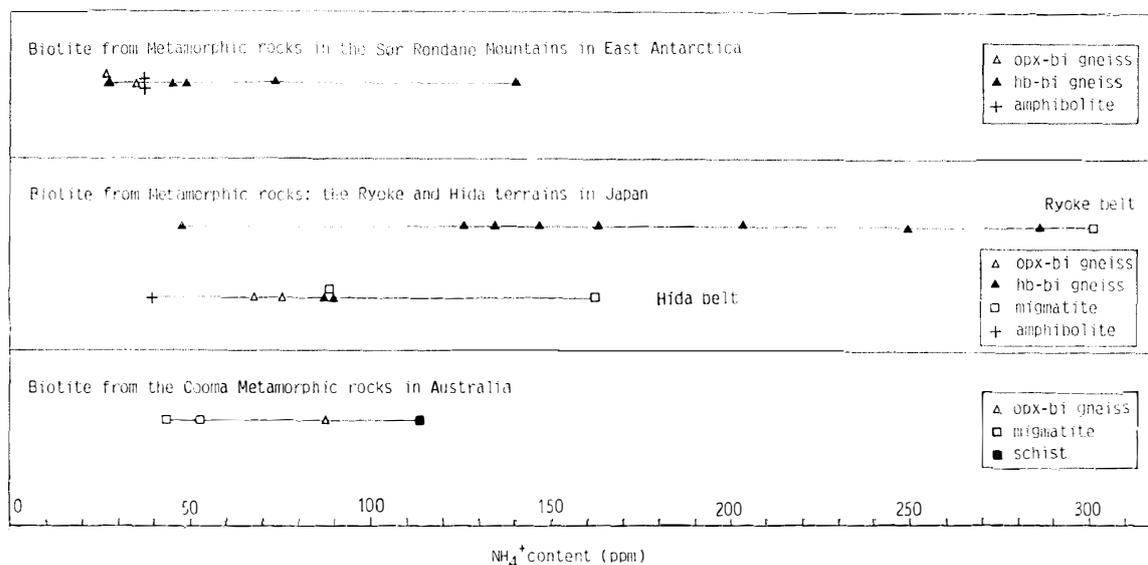


Fig. 3. Ammonium content of biotites from metamorphic rocks. Data for the Cooma metamorphic rocks in Australia are from TAINOSHO and ITIHARA (1988). Data for the Ryoke metamorphic rocks are from ITIHARA and HONMA (1979). Data for the Hida metamorphic rocks are from TAINOSHO and ITIHARA (1991).

## 5. Discussion

### 5.1. Variation in $\text{NH}_4^+$ content of biotites from granitic rocks

Ammonium content of biotites in the granitic rocks studied is closely related to the granite type. The  $\text{NH}_4^+$  content of biotites is very low in migmatitic granite, slightly low in foliated granite and discordant granite, somewhat higher in massive granite with migmatitic margin and in granite sheet and dyke. Biotites from small granodiorite bodies show distinctively high  $\text{NH}_4^+$  content. When the measured content is compared with those of biotites from migmatitic granite, the contrast is remarkable. The similarity of  $\text{NH}_4^+$  content between small granodiorite bodies and the granitic rocks in the Ryoke metamorphic belt probably indicates that the high  $\text{NH}_4^+$  content found in biotites from small granodiorite bodies is due to the inheritance of organic matter from the sediments, the origin of the granodiorite (Fig 2). However, owing to a lack of field evidence, a definite conclusion must be reserved for future study.

The Nils Larsen tonalite has low  $\text{NH}_4^+$  content ranging from 10 to 30 ppm.

These values, different from those of other types of granite except for migmatitic granite, are within the range of  $\text{NH}_4^+$  content of biotites from Cretaceous granitic rocks in non-metamorphic terrains of Japan (Fig. 2). These low  $\text{NH}_4^+$  content is also in good agreement with the view postulated from petrological and geochemical data that the granitic magma of the Nils Larsen tonalite was produced as a result of partial melting of upper mantle or an immature crust, and that sedimentary rocks played no significant role in the origin of these rocks judging from low  $^{87}\text{Sr}/^{86}\text{Sr}$  initial ratio (0.7024) (TAKAHASHI *et al.*, 1990a).

### 5.2. Variation in $\text{NH}_4^+$ content of biotites from metamorphic rocks

In many cases, biotites from meta-sedimentary rocks have very high  $\text{NH}_4^+$  content (ITIHARA, 1978; ITIHARA and HONMA, 1979). ITIHARA and HONMA (1979) concluded that the  $\text{NH}_4^+$  of biotites from these rocks originated from organic matter in the original sediments. However, the meta-sedimentary rocks in the Sør Rondane Mountains have low  $\text{NH}_4^+$  content compared with those of meta-sedimentary rocks in the Ryoke belt, except for one sample (Fig. 3). In the metamorphic rocks of the Sør Rondane Mountains,  $\text{NH}_4^+$  content of biotites from granulite is low compared with those of biotites from hornblende-biotite gneiss. Systematic decreasing tendency of  $\text{NH}_4^+$  content was recognized in biotite from amphibolite facies to granulite facies, namely,  $\text{NH}_4^+$  content of biotite decreases with increasing degree of metamorphic grade. In the Cooma metamorphic rocks (Fig. 3),  $\text{NH}_4^+$  content of biotites decreases with increasing metamorphic grade of the rock (TAINOSHO and ITIHARA, 1988). The Cooma gneiss has been highly metamorphosed (HOPWOOD, 1969) as well as the Sør Rondane Mountains metamorphic rocks. Ammonium in mica is not driven off below  $400^\circ\text{C}$  (HIGASHI, 1978, 1982), and there is also evidence that the maximum loss of  $\text{NH}_4^+$  occurs at  $800^\circ\text{C}$  in vacuum (KARYAKIN *et al.*, 1973). Thus,  $\text{NH}_4^+$  in mica is stable at low temperature, the  $\text{NH}_4^+$  content of biotites in metamorphic rocks will reflect the nitrogen content of the original rocks, if the metamorphic rocks did not undergo high temperature metamorphism ( $>600^\circ\text{C}$ ). However, if the rocks were under high temperature metamorphism such as granulite facies,  $\text{NH}_4^+$  might be liberated from biotite and the  $\text{NH}_4^+$  content of the biotites would not record the existence of nitrogen in the original rocks. This heat effect on  $\text{NH}_4^+$  in mica structure has been recognized in not only the metamorphic rocks in the Cooma district Australia (Fig. 3) but also in the metamorphic rocks of the Hida belt (granulites) (HOSHINO, 1979). The discussion leads to the following conclusion that the low  $\text{NH}_4^+$  content observed in the Sør Rondane Mountains gneisses seem to indicate liberation of  $\text{NH}_4^+$  from mica structure during high temperature metamorphism.

The biotites from amphibolites show very low  $\text{NH}_4^+$  content compared with those from meta-sedimentary rocks. This may reflect the fact that these amphibolites were formed from basic igneous rock which did not have organic matter.

### Acknowledgments

We are much indebted to the members of the 31st Japanese Antarctic Research Expedition, led by Prof. Y. NAITO (National Institute of Polar Research: NIPR), for

their kind supports during the field work. We would like to thank Messrs. K. TERAJ (NIPR), H. IKEGAMI (Nakanihon Air), T. UNO (Aero Asahi), K. URATANI (Aero Asahi) and T. TSUJI (Nakanihon Air) for their kind assistance in helicopter operation, and thank Mr. T. HAYASHI (Geographical Survey Institute) and Dr. S. HIRUTA (Hokkaido University of Education) for their collaboration in the field survey. One (TAINOSHO) of us thanks Dr. Y. ITIHARA of Osaka City University for her encouragement and advice during the analytical work. Thanks are also due to Dr. K. SHIRAIISHI (NIPR) for his generous support and kind assistance in the field survey.

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*(Received March 1, 1991; Revised manuscript received May 27, 1991)*