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Geochemical Study of the Sambagawa Metamorphic System in the Besshi District, Central Shikoku, Japan

Part II

General Tendencies in the Variation of the Bulk Chemical Composion of the Metamorphic Rocks

By

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Abstract

From the south to north in Central Shikoku, the rock facies of basic composition changes successively from the weakly metamorphosed basic rocks, through the greenschists (partly glaucophane schists) and the spotted hornblende schists, up to the amphibolites of various subfacies. The rock facies or the mineral assemblage changes more or less also within a single layer of the basic metamorphic rocks. The development of every facies and, in addition, the growth of minerals are largely dependent on the geological structures of both regional and local scale.

Chemical analyses of the basic metamorphic rocks of the various facies, which develop throughout the region, yielded the following general results :

(1) Tendencies in the fluctuation of the chemical composition of the metamorphic rocks are not uniform and are different from those recognized in the ordinary igneous processes and hydrothermalism. The degree of the fluctuation of the chemical composition within a certain area is more or less different from those for the basic igneous rocks.

(2) With the progressive change in the rock facies, as stated above, the contents of H_2O, CO_2, FeO, Fe_2O_3 and TiO_2 decrease and those of CaO and Al_2O_3 increase. The contents of SiO_2, MgO, Na_2O, K_2O and MnO are almost independent of the grade of metamorphism.

(3) Occurrence of the several special minerals in the basic metamorphic rocks is largely controlled by the bulk chemical composition of the rock and also by the degree of fluctuation in the concentration of the special chemical components within the area in which these minerals distribute.

Preface

Behaviour of a certain material, a chemical component in the present paper, in the metamorphic environment is dependent on a number of factors controlling the physical and the chemical property of minerals. Investigation of the relationships between these factors and the metamorphic phenomena may be one of the most fundamental works for petrologists. From the geochemical standpoint, however, on which the migration and the distribution of chemical components can be directly treated as the more important matters, it will be permitted to discuss about the metamorphism in direct connection with the mobility, the affinity or the activity of chemical components. From such a standpoint, several relationships between the local tectonic phenomena and the chemical composition of the Sambagawa metamorphic rocks in the Besshi district have been ascertained and summarized mainly in Part I.

In Part II, several significant geochemical informations, which have been obtained through the investigations from more general and regional viewpoint, will be described. For the statistical treatment of the chemical data, various possible methods may be provided in response to the purpose of study. However, the methods introduced into the present study shall be so devised as to suit the convenience of solving the problems on the metamorphic condition, especially on the migration of materials.

For the convenience of the statistical study, the chemical data on the basic metamorphic rocks which predominate in the amount of the data are grouped according to the general division of the regional geology, from north to south, as follows: (see Fig. 1, Part I)

(I) Amphibolite proper in the Tōnaru formations.

(II) Spotted hornblende schists in the Tōnaru formations.

(III) Common greenschists and glaucophane schists comprised in the Besshi and the Hiura formations.

(IV) Weakly matamorphosed basic rocks—relict minerals are common — comprised in the Mikabu system, the Chichibu palaeozoic formations and other younger formations.

Vein-forming greenschists of the cupriferous pyritic ore deposits and hydrothermally altered greenschists are grouped into other divisions respectively.

More critical and comprehensive exmination for the metamorphic condition, refering to the experimental results both on the physico-chemical stability of minerals and on the fugacity of materials, will be successively published in other series of papers.

Correlation of the geochemical characteristics for the Sambagawa system with those of other districts all over the world, from which several interesting facts confirming the results of the present study have been obtained, is expected to be written under the another title in the near future. But a few results of such a study, e. g., geochemical trends represented in the Al_2O_3 – $FeO \cdot CaO$ – $H_2O \cdot CO_2$ triangle are cited for reference.

The numerical values and most variation diagrams, which should be refered to, but are omitted from the present paper owing to the limited space, will be compiled in Part IV, the last paper on this serial study.

The words "the grade of metamorphism" or "metamorphic grade," which are frequently used for the convenience in the present papers concern only to such a successive change in the mineralogical facies as accepted customarily, and never to the static physical conditions[1], because the results of the present study suggest that the successive change in the mineral assemblage depends largely on the bulk chemical composition of the rocks and does not always imply the difference in the physical conditions of metamorphism.

Readers are particularly requested to make the correction for the Table 2 of Part I. The border-line between Ab.-spotted zone and Ab.-non-spotted zone ought to be drawn between the Besshi and the Hiura formations. *

Relationships of One Component to the Others in the Fluctuation of the Bulk Chemical Composition

In the several variation diagrams in Part I, it can be seen that the chemical components of the rocks fluctuate in their contents, being dependent on or quite independent of one another. Such relations will be recognized in many other variation diagrams which will be compiled in Part IV, though the mode of fluctuation varies case by case.

It seems hard to find any regularity in these relationships from the meagre analytical data, because the difference in both the amount and the combination

1 The words mean the conditions with such slow gradients of temperature and pressure as caused by deep burial only.

* The author is greatly indebted to Prof. H. Yoshizawa and Dr. T. Ueda for reading the manuscript.

of the constituent minerals in every metamorphic rock should intensify the complexity in the fluctuation of its bulk chemical composition. Such a study as substituting the bulk chemical composition for the bulk mineralogical composition, though it may be most desirable, is impossible under the present situation, because determination of the chemical composition of respective constituent mineral, generally intermingled and fine-grained, is a laborious work. Accordingly, with respect to the mutual relation between two components selected arbitrarily in the compositional fluctuation which is expected to depend on the metamorphic condition, some attempts have been made to solve the problem concerning the migration of materials.

The fluctuation tendencies which are recognized for each component in all variation diagrams inserted in the present serial are summarized as in Table 1.[2] Table 2 is the summary of those observed in the several variation diagrams of the typical basic igneous rocks selected for the comparison.

The correlations between the oxide-contents mainly of the basic metamorphic rocks from the region are illustrated in Figure 1 of Part II and Figure 8 of Part IV.[3] The general trends of the compositional variation for the basic rocks from the region, which can be obtained from these correlation diagrams, are added to Table 1, as No. 23.

From these tables and figures, the followings are possibly said as the results:

(i) Tendencies of the fluctuation in the content of the chemical components for the basic metamorphic rocks are not uniform, and vary with the locality and the extension from which the rock samples were obtained, and also with the mineral assemblage of the samples.

(ii) On the other hand, the tendencies for the basic igneous rocks, with a few exceptions, are comparatively constant and differ from any case of the basic metamorphic rocks.

(iii) Though it may be a matter of course, any of the tendencies for the basic metamorphic rocks differs from those for the pelitic schists and for the greenschists having been altered secondarily by the latest hydrothermal processes, (see Part III).

(iv) Correlation of the H_2O-content with the content of other components such as FeO, CaO and Al_2O_3, is relatively distinct, as to the Sambagawa rocks.

2 For investigation of the tendencies it is convenient to rearrange the columns sliced off from the variation diagrams.

3 Most of the correlation diagrams showing no regular tendency will appear collectively in Part IV, excepting the Na_2O-K_2O diagram.

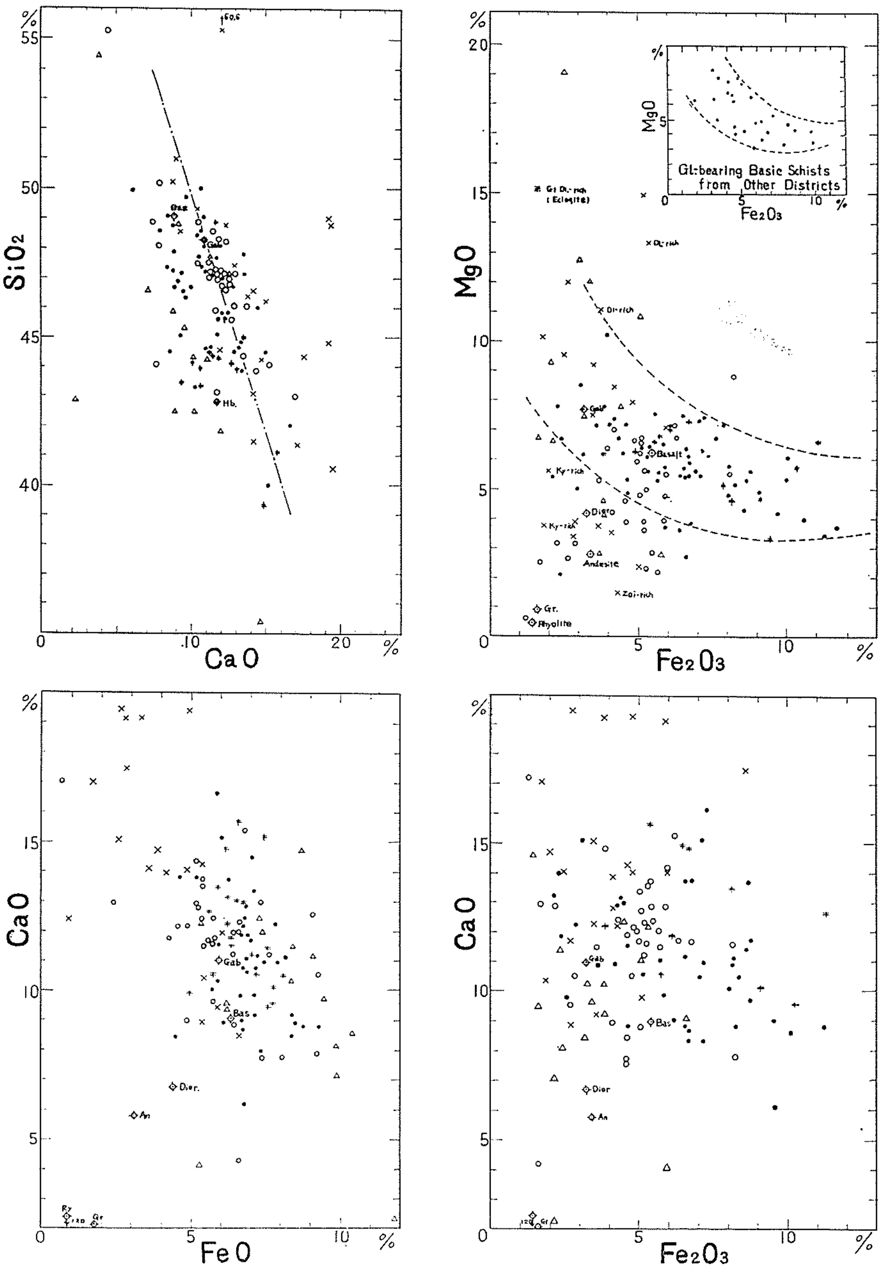
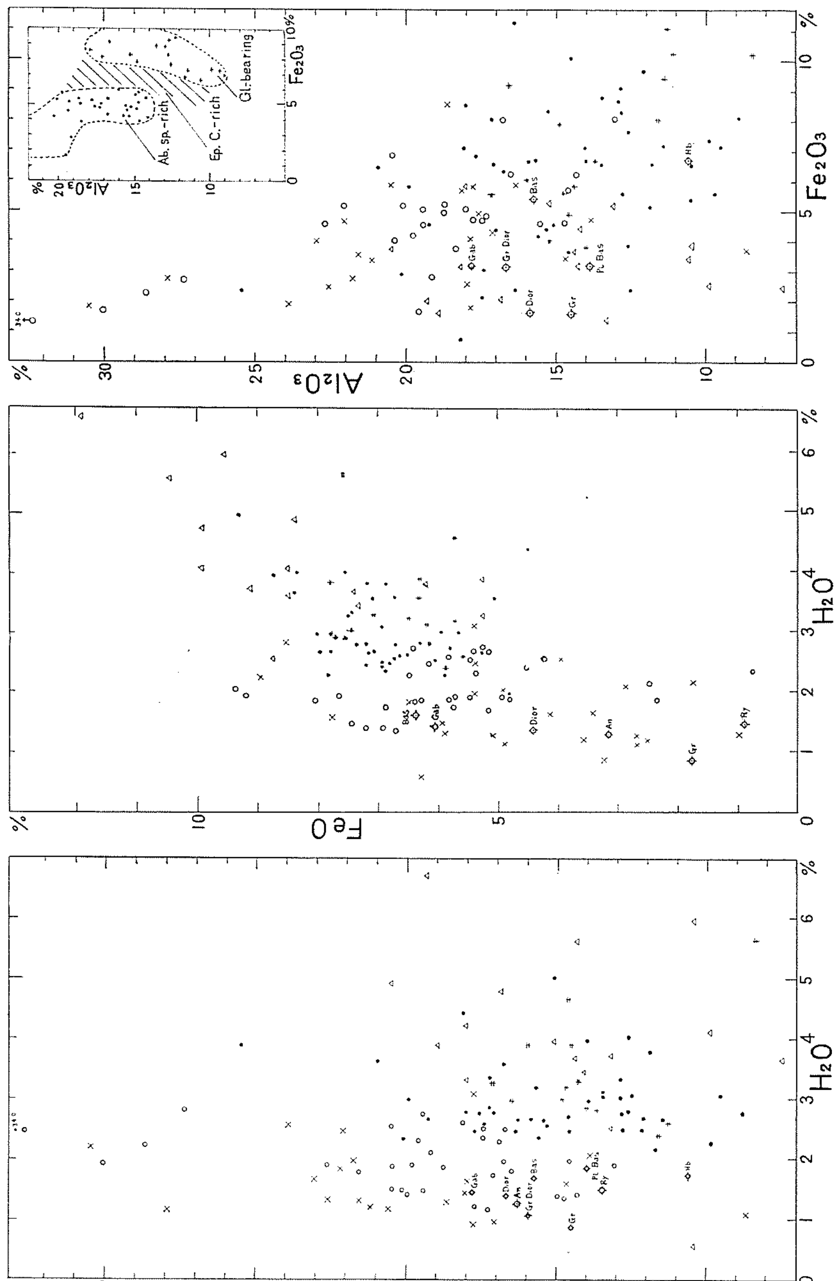
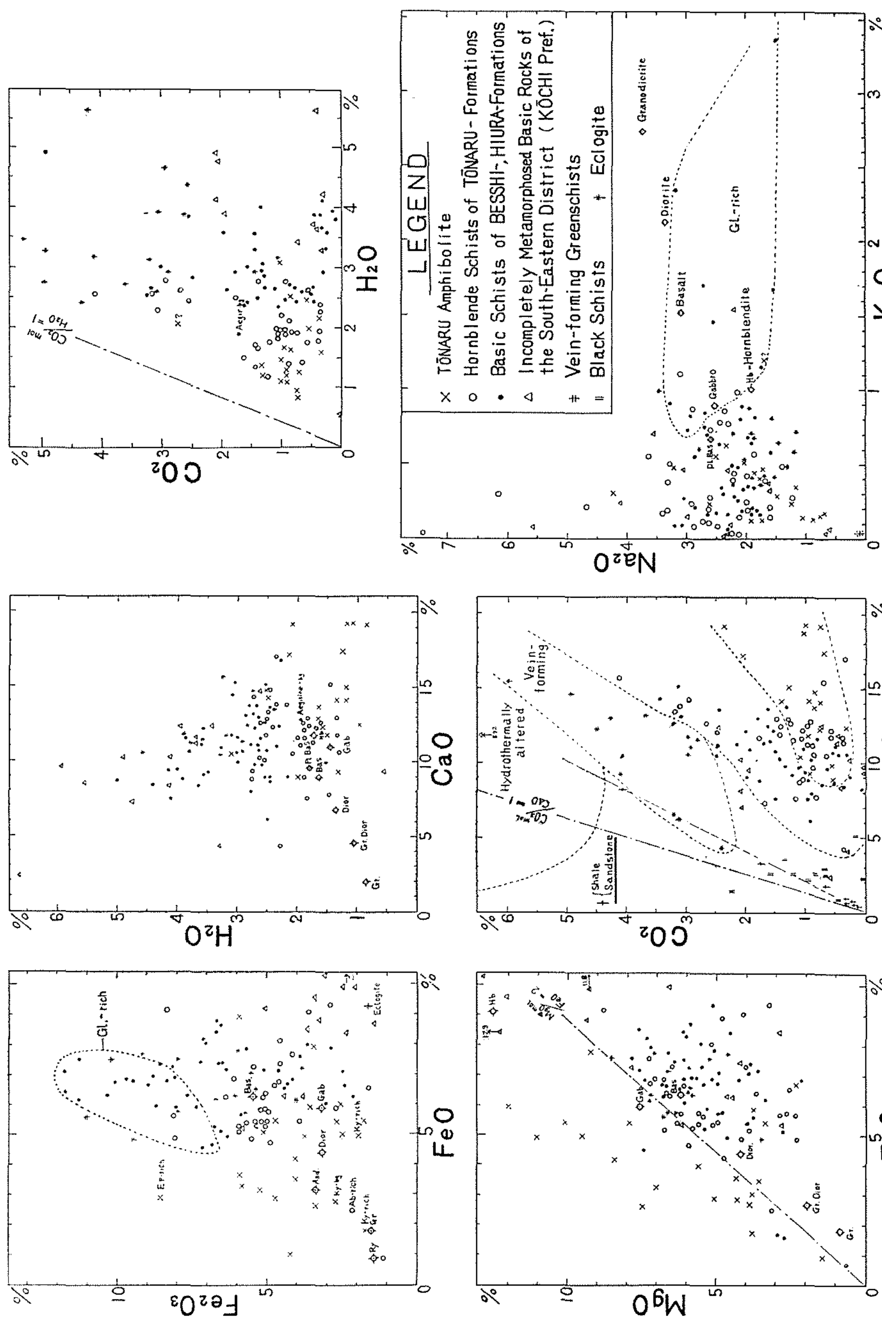


Fig. 1. Diagrams showing the correlations between the contents (in weight percentages) of the leading components in the basic metamorphic rocks from Central Shikoku. The values for the igneous rocks are from the Daly's averages. An. - Andesite, Bas. - Basalt, Dior. - Diorite, Gab. - Gabbro, Gr. - Granite, Hb. - Hornblendite, Ry. - Rhyolite, & Liparite.


Al2O3
Fe2O3
FeO
H2O
Ab. sp.-rich
Ep. C.-rich
Gl.-bearing


LEGEND
× TŌNARU Amphibolite
○ Hornblende Schists of TŌNARU-Formations
• Basic Schists of BESSHI-, HIURA-Formations
△ Incompletely Metamorphosed Basic Rocks of the South-Eastern District (KŌCHI Pref.)
‡ Vein-forming Greenschists
॥ Black Schists
✦ Eclogite
Fe_2O_3
FeO
H_2O
CaO
MgO
CO_2
Na_2O
K_2O
Gl.-rich
Granodiorite
Diorite
Basalt
Gabbro
Hb.-Hornblendite
Hydrothermally altered
Vein-forming
Shale
Sandstone

(v) Throughout the region, there is little correlation between FeO and Fe_2O_3, though oxidation and reduction may have some significances in the local development of the glaucophane schists as will be described in page 271.
(vi) There is negative correlation as a whole between CaO and CO_2, but CaO, when it is higher in the content, shows a positive correlation with CO_2 as to the rocks belonging to the same stratigraphic horizon.
(vii) The correlation between CaO and CO_2 is generally positive in the shales, sandstones and phyllites from the southern district, but it becomes rather negative in the pelitic schists from the Besshi district, (see also Fig. 7-J in Part IV).
(viii) There is a negative and clear correlation between Fe_2O_3- and MgO-content of the common greenschists, especially of the glaucophane-bearing one, (see also page 270).

These results seem to afford the evidences negative for an interpretation that development of the basic metamorphic rocks had proceeded in the closed system, reflecting faithfully the bulk chemical composition of the original rocks. It may be possibly said that the Sambagawa metamorphic rocks had experienced the allochemical changes within a certain extension in a rock-body, or, in other words, that the free migration of some chemical components within some extension in a single rock-body had taken place and destroyed their initial distribution to form the new geochemical tendencies. It will be readily assumed that hydration and dehydration depending on the physical conditions may be accompanied more or less by the migration of other components and also by the solution of calcite, which causes the escape of CO_2.

The above interpretation will be justified by other geochemical facts stated in the succeeding chapters.

Degree of Fluctuation of the Chemical Composition

Degree of the fluctuation in the chemical composition of the rock-samples from a single rock-body or from the several rock-bodies of the same kind in a certain area may give an important evidences to determine whether the migration or the redistribution of materials within a certain extension took place or not during the metamorphism. Moreover, it will suggest the range in which the chemical equilibria under a certain physical condition had been attained.

For investigating these matters, it should be first neccessary to determine critically (1) general geology of the vicinity, (2) extension and form of the rock-body in question, (3) distribution and concentration of the rock-forming

No.	SiO_2	Al_2O_3	Fe_2O_3	FeO	MgO	CaO	Na_2O	K_2O	H_2O	CO_2	Numbers of Sample	From Diagram
1	A	A	a	A	a	a	A	?	A	a	—	I-16-A, B, C
2	A	A	A	a	a	A	A	?	a	a	—	I-16-D, E
3	A	a	?	B	?	a	B	?	a	a	3	IV-7-A
4	A	?	a	B	a	a	A	?	b	a	6	IV-7-B
5	?	A	a	B	B	b	B	B	?	?	21	IV-7-C
6	?	A	a	B	a	?	B	?	?	?	10	IV-7-G
7	?	?	a	A	a	a	a	?	?	?	4*	I-9-B
8	A	B	?	?	B	a	?	?	?	b	5*	I-9-C
9	?	?	A	?	a	a	?	A	?	?	7	IV-7-D
10	A	?	B	?	?	b	?	?	A	b	8	I-11-B
11	?	A	A	a	a	B	?	?	a	b	13	III
12	?	?	?	?	?	A	B	?	b	A	11	IV-7-F
13	?	A	a	A	A	a	a	?	?	a	5*	I-9-A
14	A	B	A	a	b	A	B	?	a	?	10	IV-7-H
15	A	A	B	a	b	a	b	?	A	a	4	IV-7-I
16	?	A	a	A	A	B	?	a	A	B	7	I-11-A
17	?	?	?	A	A	a	a	?	A	a	6	III
18	A	A	?	?	?	?	A	a	a	a	18	III
19	?	A	a	?	a	a	a	A	A	A	18	III
20	A	?	B	a	C	C	B	?	a	?	9	IV-7-J
21	?	?	?	A	?	B	C	c	A	B	10	IV-7-J
22	A	B	a	a	?	a	?	B	b	A	3*	I-9-D
23	A	B	b	A	A	a	?	?	A	?	—	II-1
24	?	A	a	a	?	A	?	?	a	a	—	II-3

Table 1. Mutual relations of the chemical components in variation of their contents, as to the various rocks in the Central Shikoku. The components marked by the same letter vary with the tendency parallel to one another. Those marked by the different letters, e.g., A, B and C, vary independently of each other. The components marked by the small letters vary with the tendency reverse to that of the components marked by the large letters. A question-mark indicates "uncertain", or that number of the exception are more than a quarter of the total number of the samples. Notations for the diagrams, for example, IV-7-F indicates the diagram (F) in Figure 7 of Part IV. *: Closely spaced samples.

1. sp. Hb. - S, Tōnaru-F.; general trend in the folded structures.
2. sp. Hb. - S, Tōnaru-F. & greenschist of Besshi-F.; general trend in the sheared structures.
3. sp. Ep. Q. C. Hb. - S, Tōnaru-F.; in a rock-body; Loc., Uchiyoke, Niihama.
4. sp. Ep. C. Hb. - S, Tōnaru-F.; throughout three rock-bodies; Loc., Otoshi, Niihama.

5. Amphibolites of the various subfacies, Tōnaru-F.; in a rock-body; Loc., Tōnaru, Niihama.
6. sp. Hb. - S with the various mineral assemblages, Tōnaru-F.; in a rock-body; Loc., 23L, cx-cut, Shirataki Mine.
7. sp. Mus. C. Hb. - S, Tōnaru-F.; within a small area in a rock-body; Loc., 23L cx-cut, Shirataki Mine.
8. do.
9. Greenschists with the various mineral assemblages, Besshi-F. & Hiura-F.; throughout several rock-bodies; Loc., along the Otoshi-Kiyotaki route.
10. Greenschists of the various subfacies, excluding Gl.-rich one, Besshi-F.; along the greenschist on the Yokei-Hor.; Loc., near the Besshi Mine.
11. Act. Ab. Ep. Q. Cc. C. - S, adjacent to the Hiura ore-horizon, Hiura-F; within a small area in a rock-body; Loc., 14L, Fw. cx-cut, Besshi Mine.
12. Greenschists with the various mineral assemblages, Besshi-F & Hiura-F.; throughout several rock-bodies; Loc., along the Tai-Shirataki route.
13. Ab. Q. Cc. Ep. C. - S, Hiura-F.; within a small area in a rock-body; Loc., 22L, Fw. cx-cut, Besshi Mine.
14. So-called Mikabu green rock (metagabbro ?), Mikabu system; throughout a rock-body; Loc., along Osugi-Kochi route.
15. Weakly metamorphosed basic rocks, upper palaeozoic formations; throughout several rock-bodies; Loc., along Osugi-Kochi route.
16. Ab. Se Cc. Ep. Act. C. - S, vein-forming greenschists, Besshi-F.; throughout the ore-body; Loc., Besshi Mine.
17. Cp. Py. Q. C - S, poorly disseminated banded ore (SK), Besshi-F.; throughout the ore-body; Loc., Besshi Mine.
18. Se. Cc. - rich greenschists altered hydrothermally at the latest stage, Besshi-F. & Hiura-F.; throughout the several cases at shallower depth; Loc., Besshi Mine.
19. do., in the deeper levels; Loc., 22L, 23L & 26L, Besshi Mine.
20. Black schists of the various kinds, Sambagawa system; throughout many rock-bodies; Loc., several parts of the Besshi district.
21. Shales, sandstones and phyllites, Mikabu system and other non-metamorphic formations; throughout many rock-bodies; Loc., Kochi prefecture.
22. sp. Cc. Se. Q. G. - S, Besshi-F.; within a small area in a rock-body; Loc., 22L, Hw. cx-cut, Besshi Mine.
23. General trends for the basic metamorphic rocks of the Central Shikoku; obtained from the correlation diagrams (Fig. 1, Part II).
24. General trends for the basic metamorphic rocks, in relation to the successive facies change on a regional scale; obtained from the frequency distribution diagrams (Fig. 3, Part II).

minerals, (4) proper sampling interval and (5) the smallest amount of a sample representing a certain lot. Such preliminary investigations, however, are too laborious to be made.

With the view stated above, several tentative studies have been carried out, concerning the basic metamorphic rocks of the Central Shikoku.

The degree of fluctuation of a chemical content within a certain area

No.	SiO_2	Al_2O_3	Fe_2O_3	FeO	MgO	CaO	Na_2O	K_2O	H_2O	CO_2
1	?	A	a	B	b	b	B	?	B	—
2	?	?	A	a	b	b	B	B	b	—
3	A	A	?	a	a	a	B	B	?	—
4	A	A	?	?	a	a	A	A	?	—
5	A	A	A	a	a	a	A	A	?	—
6	A	A	a	a	a	a	A	A	?	—
7	A	A	A	a	a	a	A	A	A	—
8	A	A	a	A	b	a	B	B	?	—
9	A	B	?	b	b	b	A	?	?	—
10	?	A	?	a	a	A	a	?	a	?
11	A	A	a	a	?	A	A	A	?	?
12	A	A	A	a	a	A	A	a	a	?
13	A	A	a	a	a	a	?	?	a	—
14	A	a	?	a	a	a	A	A	a	—
15	A	A	a	a	a	a	A	A	a	—
16	A	a	A	?	?	a	A	?	A	A
17	A	?	?	a	a	A	A	?	a	a

Table 2. Tendencies of fluctuation for the chemical components of basic igneous rocks (SiO_2 42-57 in wt%). Notations are the same with those in Table 1.

1. Deccan basalts, after WASHINGTON (1922).
2. Oregon basalts, after WASHINGTON (1922).
3. Palisade diabase sills, after WALKER (1940).
4. Mount Wellington diabase sills, Tasmania, after EDWARDS (1942).
5. Olivine basalts and oceanites of Samoa, after MACDONALD (1944).
6. Basanitoid lava of Tahiti, after Williams, H. (1933), —— from TURNER and VERHOOGEN (1951).
7. Volcanic rocks of the Cascade province, do.
8. Lava of Mt. Fuji, after TSUYA (1937), —— from IWASAKI (1948).
9. Muroto gabbro, Shikoku, after YOSHIZAWA (1954).
10. do., coarse-grained part, closely spaced three samples, TAKUBO, analyst.
11. do., fine-grained part, closely spaced three samples TAKUBO, analyst.
12. do., from the center to the margin of the rock-body, TAKUBO, analyst.
13. Volcanic rocks of Japan, after IWASAKI (1948), p. 155, Fig. 12.
14. General trends from gabbro to diorite, after the DALY's averages (1933).
15. General trends from basalt to andesite, do.
16. Spilite association, after SANDIUS (1930).
17. Autometamorphosed basaltic diabases of the Central Kyusyu, Japan, after MUTA (1957).

No.	SiO_2	Al_2O_3	Fe_2O_3	FeO	MgO	CaO	Na_2O	K_2O	H_2O	CO_2
3	1.1	1.1	2.1	1.2	1.2	1.3	1.6	1.5	1.4	10.8
4	1.1	1.6	1.5	1.9	1.5	1.7	1.9	10.0	1.9	2.0
5	1.5	3.9	3.5	9.0	8.0	2.2	6.1	4.0	3.6	7.0
6	1.1	1.7	3.8	3.8	2.8	1.9	1.9	7.0	1.9	2.2
7*	1.0	1.1	1.1	1.1	1.8	1.1	1.3	1.2	1.1	1.4
8*	1.1	1.1	1.1	1.2	1.4	1.2	1.4	2.1	1.2	1.2
9	1.2	1.6	3.2	1.4	2.1	2.5	2.1	17.8	1.5	6.1
13*	1.0	1.2	1.4	1.2	1.2	1.2	1.3	1.5	1.1	1.3
10	1.1	1.2	1.4	1.6	1.3	1.2	1.8	5.8	1.1	1.4
11	1.1	1.1	1.2	1.4	1.4	1.3	1.2	2.7	1.2	1.3
16	1.1	1.9	2.9	1.3	2.4	1.6	1.7	2.7	2.2	1.9
A	1.3	2.2	6.9	3.8	3.8	4.1	5.7	5.0	2.1	10.0
B	1.3	2.8	5.1	2.1	2.4	2.5	2.1	8.4	2.0	∞
C	1.1	2.1	4.6	1.9	1.7	5.1	2.2	∞	1.9	∞

Table 3. The values for α/β, an indicator for the degree of fluctuation in chemical composition in a certain area, for the basic metamorphic rocks (SiO_2 42 to 57% in weight) from the Central Shikoku.

The remarks on Nos. 3, 4, 5, are the same as in Table 1 respectively. A: Spotted hornblende schists of the Tōnaru formations. B: Greenschists of the Besshi and the Hiura formations. C: Green rocks of the zones of lower metamorphic grade, excluding non-metamorphosed rocks.

*: Variation in a small area in a rock-body.

No.	SiO_2	Al_2O_3	Fe_2O_3	FeO	MgO	CaO	Na_2O	K_2O	H_2O	CO_2
1	1.1	1.2	2.0	1.5	2.1	1.5	2.0	4.3	4.4	?
2	1.2	1.1	3.4	1.5	3.2	1.5	1.5	3.9	9.0	?
3	1.1	1.8	2.7	1.5	5.2	1.5	2.7	3.0	1.7	?
4	1.1	1.3	3.0	1.6	3.1	1.4	2.0	3.0	4.0	?
5	1.1	1.1	2.8	1.5	1.6	1.0	1.2	1.5	2.5	?
6	1.1	1.3	3.2	1.9	3.5	1.6	2.1	3.2	1.9	?
7*	1.0	1.0	1.5	1.5	2.9	1.2	1.2	1.6	1.7	1.8
8*	1.1	1.0	1.6	1.1	1.3	1.1	1.3	1.5	1.2	3.1
9	1.1	1.6	2.3	1.9	3.5	1.6	2.3	3.2	2.5	?
10	1.1	1.3	2.8	1.6	3.2	1.4	2.0	3.2	3.7	?

Table 4. The values for α/β, for basic igneous rocks (SiO_2 42-57 in wt.%) from several part of the world.

1. Deccan basalts, after WASHINGTON (1922). 2. Oregon basalts, after WASHINGTON (1922). 3. Palisade diabase, after WALKER (1940). 4. Basaltic lava, Hawaii, from WASHINGTON (1917). 5. Basaltic lava, Mt. Fuji, after IWASAKI (1948). 6. Muroto gabbro, throughout the rock-body, after YOSHIZAWA (1954). 7. do., within a small area in the coarse-grained part, TAKUBO, analyst. 8. do., within a small area in the fine-grained part, TAKUBO, analyst. 9. do., throughout No. 7 and No. 8. 10. Average values from No. 1 to No. 9 excluding No. 7 and No. 8.

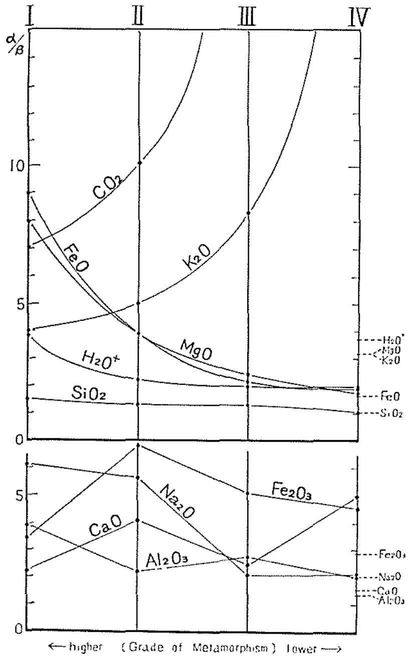


Fig. 2. Diagrams showing the values for α/β of chemical components for the basic metamorphic rocks from every zone in the Central Shikoku. Broken lines with chemical symbols indicate the average values for the basic igneous rocks (see No. 10, Table 4).

(I) Tōnaru amphibolite-body, (from No. 5 in Table 3).

(II) Spotted hornblende schists of the Tōnaru formations, (from A in Table 3).

(III) Greenschists of the Besshi and the Hiura formations, (from B in Table 3).

(IV) Green rocks of the Mikabu system and other non-metamorphic formations, (from C in Table 3).

in a rock-body may be approximately regarded as a ratio of the maximum content (α) to the minimum content (β) of a chemical component in a group of analysed samples which are collected from that area. Such an approximation is possibly applied also to a group of the rock-bodies which have the same mineral assemblage and stand on the same structural situation on a regional scale in a certain area.

Table 3 shows the values for the ratio α/β as to the basic metamorphic rocks from the region. The amount and the size of each sample conform to the rule described in page 58 of Part I. The data on the rocks which are estimated to have been affected by the post-metamorphic chemical processes, are omitted from codsideration, for example, a chlorite-rich and a sericite-rich part in the spotted hornblende schists. For the reference, the values for α/β of several typical basic igneous rocks are summarized in Table 4. Average value for every zone or formations is added to Table 3, as A, B and C, and is illustrated graphically as in Figure 2.

The data listed in these tables and figures do not completely satisfy the necessary conditions stated above. Above all paucity of the samples is so fatal that active examination for the problems proposed above should be avoided.

The following facts, however, may be recognized at least, with regard to the degree of fluctuation for the basic metamorphic rocks:

(i) Though it may be a matter of course, as to the basic metamorphic rocks, the value for α/β of every component fluctuates with the interval and the area of sampling and with the mineral assemblage. Whereas, the value for the basic igneous rocks is in general relatively constant for every component except H_2O, though most of the igneous rock-samples may have been collected at a distant interval and from the mutually separated rock-bodies. In this case also the fluctuation in a small area tends to become weaker, as recognized in the case of the Muroto Gabbro, (see No. 7 and No. 8 inTable 4).

(ii) As for the basic metamorphic rocks, the values for H_2O and MgO are generally smaller and the value for K_2O is larger than in the cases of the basic igneous rocks.

(iii) As shown in Figure 2, with successive increase of the metamorphic grade which can be recognized through the progressive change of the mineral assemblage in the basic metamorphic rocks, the value for α/β of every component except CaO, Al_2O_3 and F_2O_3, varies showing a regular and continuous tendency.

(iv) Since fluctuation of the chemical value is expected to reflect the difference in the mineralogical composition of the analyzed rock-sample, there must be some relations between the α/β-values and the mineral assemblage characterizing each zone or formations. In fact, local and deviated enrichment of the albite-porphyloblast, glaucophanitic amphiboles and some kinds of anhydrous silicate minerals, all of which are regarded as the indicator minerals for the grade of metamorphism in the region, can be attributed also to the degree of fluctuation as will be explained in the next chapter (see page 270).

Various interpretations are possible for the above results. Some results may possibly suggest that the chemical heterogeneity of a basic metamorphic rock had been intensified through the migration of several chemical components, which went on along the physical and the chemical concentration gradient during the Sambagawa metamorphism.

For reaching a rigid conclusion concerning the problem stated at the biginning of this chapter, further and systematic investigations should be neccessary.[4]

4 The ratio α/β for a component can be regarded as an indicator for the mobility, activity or affinity of that component, as well as for the chemical heterogeneity of a rock. It is interesting to compare the order of the degree of fluctuation, which can be readily obtained from Table 3, to the mobility series established by KORZHINSKY (1948) for the opened system under the higher heat effect.

Relationships between the Mineral Association and the Bulk Chemical Compostion of the Basic Metamorphic Rocks, mainly concerning the Incompatibility of Glaucophanitic Amphiboles with Porphyloblastic Albite

SUZUKI (1930) studied the relation between the mineral assemblage and the bulk chemical composition of the Sambagawa metamorphic rocks and concluded that the chemical composition of each rock is characteristic for its mineral assemblage. Similar result has been obtained from the present study.

Chemical peculiarities of the basic metamorphic rocks containing a characteristic mineral are noticed in many of the variation and the correlation diagrams. These peculiarities are summarized as shown in Table 5.[5] It should be noted that the chemical components suggesting the enrichment of a certain mineral do not necessarily coincide with those composing that mineral. Owing to the limited space, discussion on the above problem will concern only to the occurrence of glaucophane and albite in the basic metamorphic rocks. Both minerals have been frequently discussed by many authors. Their petrological and geochemical characteristics are summarized as follows:

(i) Glaucophanitic amphiboles in the basic schists occur as the mineral characteristic for the relatively low-grade zone, (see Table 2 in Part I). They form remarkable banded structures together with all or some of the minerals such as epidote, chlorite, quartz, white micas, albite, specularite, etc.

(ii) Whereas, an albite-porphyloblast increases in both its amount and grain-size, with general increase of the grade of metamorphism or with development of the prominent poli-axial folding in the upper formations. In general, it tends to be rich in the massive rocks at the crestal part of folds of the various forms, (see page 65, Part I).

(iii) Incompatibility of glaucophanitic amphiboles with albite-porphyloblasts, both on a regional and on such a small scale as being restricted within a single layer of the compositional banding, is also ascertained by the statistical study on the basic schists bearing albite and glaucophane, from every part of the world, enumerating the several tens of the samples from the Sambagawa system, (see Figure 9 in Part IV).

(iv) The contents of K_2O, F_2O_3 and Al_2O_3 in the basic schists are also important factors to control the relative amounts of glaucophane and albite, as shown in the Al_2O_3–Fe_2O_3– and the Na_2O–K_2O–diagram in Figure 1. The

5 The anomaly of the chemical composition, which suggests the higher concentration of amphiboles in a rock-sample, is hardly found as to the basic rocks of all formations. This probably suggests that the chemical composition of amphiboles varies in a wide range, depending upon the grade of metamorphism.

greenschists enriched in chlorite and epidote are plotted in the intermediate area between the rocks enriched in glaucophane and in the porphyloblastic albite, as shown in the Al_2O_3-Fe_2O_3 diagram in Figure 1.

(v) As for the basic schists containing the glaucophane, regardless of its amount in the sample, the content of MgO tends to vary inverse proportionally to that of Fe_2O_3 as shown in Figure 1. Similar relation of MgO to Fe_2O_3 is also obtained from the statistical study on the rocks from other districts in the world, as additionally illustrated in the Fe_2O_3-MgO diagram in Figure 1. Whereas, the above relation becomes irregular or rather sporadic for the basic schists characterized by the conspicuous enrichment of the albite porphyloblasts.

(vi) The degree of fluctuation of the content, i. e., α/β, of some chemical components well explains the mutually incompatible and localized concentrations of both minerals. That is, the value for α/β of the components such as K_2O, Fe_2O_3, Al_2O_3 and CO_2 increases in the Hiura and the Besshi formations which are characterized by the conspicuous development of the intercalated glaucophane schists. Most of these components are at the same time the characteristic components controlling the occurrence of glaucophane in the basic schists as stated above. In the same way, the successive increase in the amount of albite-porphyloblasts can be attributed to the locally deviated distribution of Na_2O in a rock body, which is suggested by its higher value for α/β, and not to the relative increase of Na_2O-content in a wide area, (see the next chapter).

Mineral	SiO_2	Al_2O_3	Fe_2O_3	FeO	MgO	CaO	Na_2O	K_2O	H_2O	CO_2	From Diagram
Aegirine				−				+	−	+	IV-7-F
Albite		+	−			−	+				II-I, etc.
Chlorites		+				−		−	+	−	II-I, etc.
Diopsidic Pyroxenes					+	+	−	−	−		II-I. IV-7-C
Garnets				+		−					IV-7-C, etc.
Glaucophanes		+	+			−		+		−	II-I, etc.
Kyanite		+	−	−	−						IV-7-C
Zoisite Minerals			−	−	−	+					IV-7-C

Table 5. Peculiarities in the bulk chemical composition of the basic metamorphic rocks (SiO_2 40—57 in wt.%), containing several characterstic minerals. + : relatively higher; − : relatively lower in content. Notation for the diagrams is the same as in Table 1.

Considering these facts all inclusively, the following interpretation will be given to the occurrence of glaucophanitic amphiboles incompatible with the albite-porphyloblasts.

(i) Presence of the glaucophanitic amphiboles does not necessarily imply the attendance of high load pressure having prevailed over the region. The more essential factor having controlled the genesis of glaucophane is not the static physical conditions, but the activity of the components such as Fe_2O_3, K_2O, Al_2O_3, etc. Redistribution of these components is assumed to have been affected by the strong shearing process accompanying more or less the local change of oxygen partial pressure, the local weak hydrothermalism and the formation of compositional bands, under the static physical conditions at moderate depth.

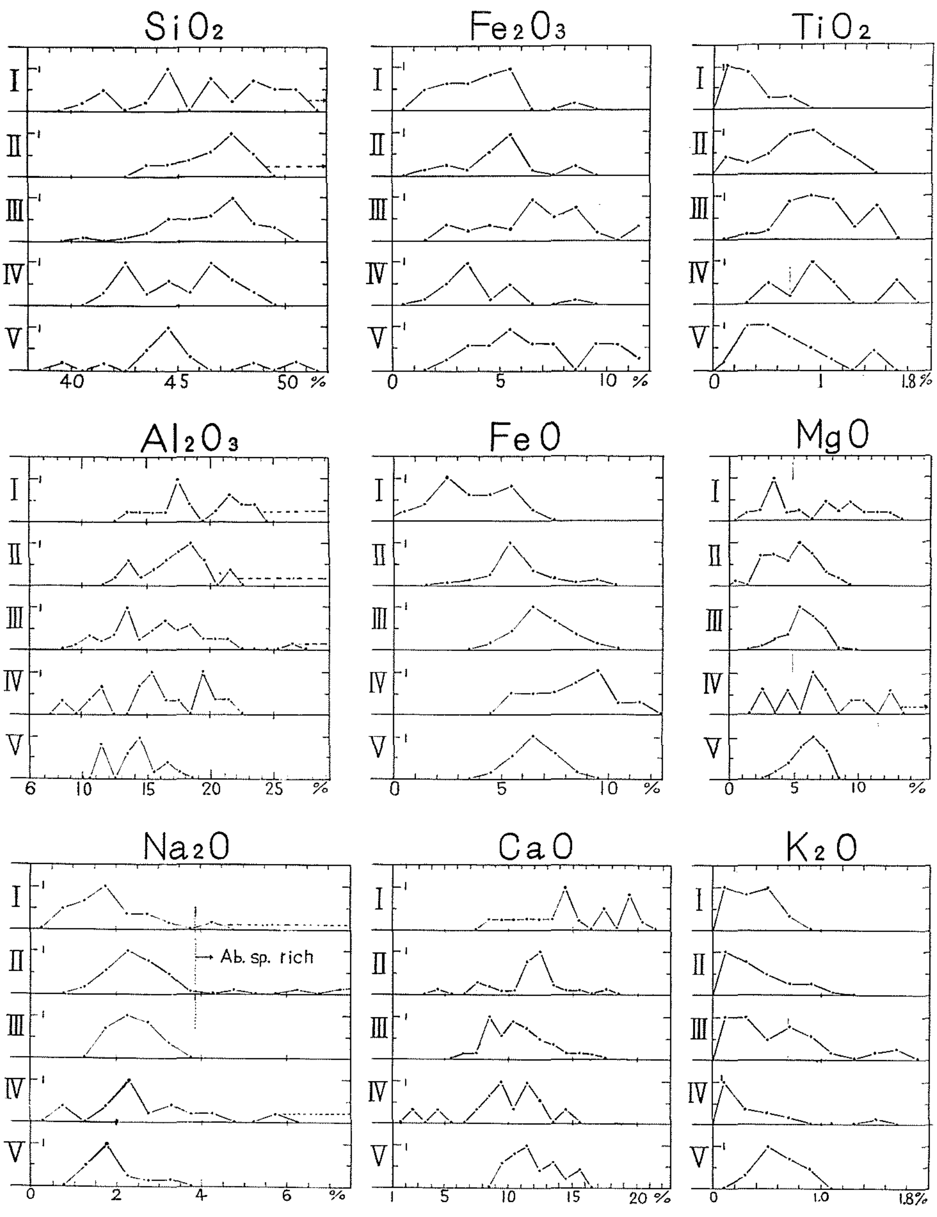
(ii) Free migration of Na_2O which had been restricted within the limit of a single rock-layer — not between the layers adjacent to each other — had caused the localized concentration of the macroscopic albite-porphyloblasts. The migration seems to have been controlled by the pressure gradients caused by the folding movement at moderate depth.

Above interpretation for the occurrences of albite and glaucophane on the basis of the tectonic differentiation is possibly applied also to the localized concentration of other anhydrous silicate-minerals such as diopsidic pyroxenes, pyralspite garnets and kyanite. Development of these minerals may be ascribed not only to the depth, but also to the local high confined pressure induced along a tight and complicated fold in the synclinorium. Thus mode and distribution of stress presure are considered to have played an important role in the redistribution of materials and accordingly in recrystallization of the minerals being characteristic of the Sambagawa metamorphic rocks.

Correlation of the Chemical Composition with the Grade of Metamorphism of the Basic Metamorphic Rocks

It is commonly accepted by many petrologists that the reactions in the progressive metamorphism from the original basic rocks through the greenschists up to the amphibolite proceed isochemically for most chemical components except the volatile components. Surely, when we plot the chemical data in the triangle having a $H_2O \cdot CO_2$- corner, the above assumption is understood to be nearly correct, as shown in Figures 4 and 5.

Figure 3 shows the frequency distribution of the oxide-contents for the basic metamorphic rocks of every zone or formations. From this figure and the



SiO2
Fe2O3
TiO2
Al2O3
FeO
MgO
Na2O
CaO
K2O
I
II
III
IV
V
40
45
50 %
0
5
10 %
0
1
1.8 %
6
10
15
20
25
%
0
5
10
%
0
5
10
%
0
2
4
6
%
→ Ab. sp. rich
1
5
10
15
20 %
0
1.0
1.8%

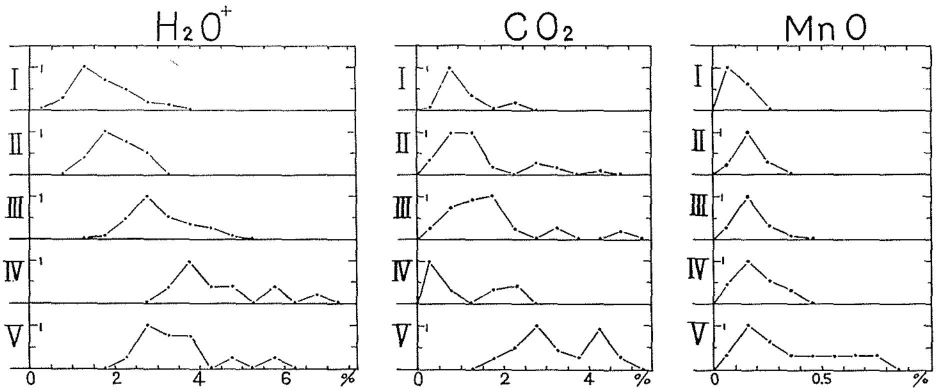


Fig. 3. Diagrams showing the frequency distribution for the oxide-contents of the basic metamorphic rocks of the Central Shikoku. The values for all intervals are converted as the maximum frequency equals one.
(I) Tônaru amphibolite-body, twenty samples. (II) Spotted hornblende schists of the Tônaru formations, thirty four samples. (III) The Besshi and the Hiura formations, forty four samples. (IV) Mikabu system and other non-metamorphosed formations, fourteen samples. (V) Vein-forming greenschists of the cupriferous pyritic ore-deposits in the Besshi formations, seventeen samples.

correlation diagrams in Figure 1, the following general trends as to the oxide-contents are obtained:

With the successive change of metamorphic facies, from the incompletely metamorphosed basic rock through the greenschist facies up to the amphibolite facies, the contents of H_2O, CO_2, FeO, Fe_2O_3 and TiO_2 of the rocks decrease, and those of CaO and Al_2O_3 increase. The contents of SiO_2, MnO, MgO, Na_2O and K_2O show no successive correlation with the grade of metamorphism.

As for Na_2O and K_2O, the simillar results to the BANNO'S (1961) are obtained from this study, (see page 50 of Part I). It is noteworthy that several components as well as H_2O and CO_2 vary more or less in their contents, with some regular trends, correspondingly to the progressive change in the mineral assemblage of the basic rocks in the region. This fact will be demonstrated also by the general trend illustrated in the Al_2O_3–FeO·CaO –H_2O·CO_2 triangle[6] as shown in Figure 4. As for the pelitic and the psamitic rocks, the progressive metamorphism from the sediments up to the crystalline schists does not also imply the decrease in the contents of the volatile components only, as shown in Figure 5, which is tentatively drawn for the statistical study.

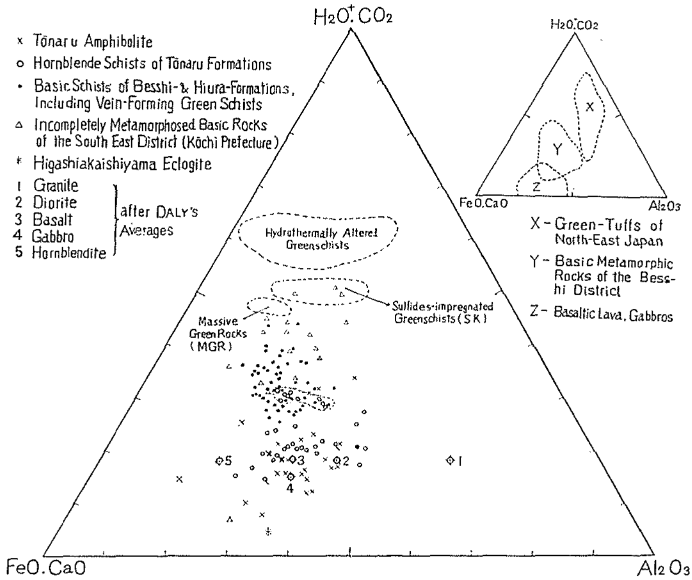


Fig. 4. General trends in the variation of chemical composition, for the basic metamorphic rocks of the Central Shikoku. The values are shown in mol %.

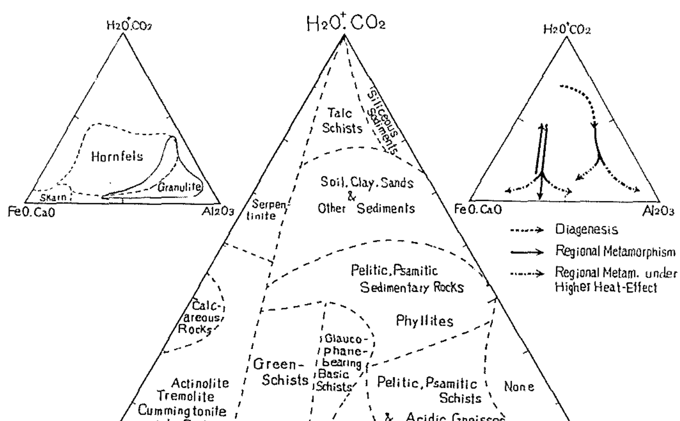


Fig. 5. Diagrams showing the distribution of various rocks and the general trends for the metamorphic processes, obtained from the statistical study on about a thousand and three hundred data from every part of the world. It should be noted that, for the most rocks except hornfelses, where $Al_2O_3/FeO \cdot CaO > 1$ in mol ratio, $SiO_2 > 53$ in wt. %.

Summary of the Geochemical Characteristics of the Basic Metamorphic Rocks of the Sambagawa System in the Besshi District

The prominent geochemical characteristics of the Sambagawa metamorphic rocks, which are recognized in the present study having been summarized in Part I and II, will be picked out as follows:

(i) The crystalline schists are chemically heterogeneous irrespectively of dimension of the rock, or in other words it is hard to find one block having the bulk chemical composition similar to that of the other block within a rock-body.

(ii) The mineralogical and the chemical compositions of the basic metamorphic rocks near the contact plane between two rocks being different from each other in such a way as pelitic and basic, do not show any regular variation.

(iii) On both a regional and a local scale, the mineralogical and the chemical compositions of the rocks vary with the close connection to the folded and the sheared structures in which the rocks are situated. In these variations there are some regular tendencies reflecting the tectonic career of the rocks.

(iv) Any geochemical tendency which is found in these variations of chemical composition of the Sambagawa metamorphic rocks is not similar to those which are commonly observed in the cases of igneous and hydrothermal processes.

(v) The degree of fluctuation of chemical composition or the ratio α/β, (α: maximum content, β: minimum content of a certain component), in a group of rock-blocks occurring in a certain area, is different, to an appreciable extent, from those for the basic igneous rocks. There are several chemical components indicating the close relationships between the degree of fluctuation and the successive change of the metamorphic grade indicated by the mineral assemblage and texture, as shown in Figure 2.

6 Representation of the chemical data by means of the ACF-diagram, which is commonly used for investigating the relationships of the chemical composition with both the mineral assemblage and the grade of metamorphism, seems not to be suitable for the Sambagawa basic metamorphic rocks. ACF-diagram clearly indicates that, as to the Sambagawa rocks, the change in the mineralogical facies accompanies more or less the change in the bulk chemical composition of the rocks and that the progressive facies change is hardly ascribed to the difference in the physical condition only.

If the metamorphic system is more or less open, the graphic representation of chemical data, in which the mobility of chemical components is respected, will be more suitable for the above purpose. On such a presumption, Al_2O_3-$FeO \cdot CaO$-$H_2O \cdot CO_2$ triangle has been devised.

(vi) Occurrence of several minerals typical for every zone or formations is strongly controlled by the bulk chemical composition of the metamorphic rocks.

(vii) With the successive change of the rock facies of the basic composition, from greenschists through spotted hornblende schists up to the amphibolites of various kinds, H_2O, CO_2, Fe_2O_3, FeO and TiO_2 decrease and CaO and Al_2O_3 increase in their contents. The content of other components such as MnO, MgO, Na_2O and K_2O, is independent to the grade of metamorphism. These geochemical tendencies seem to differ from those recognized in the course of basic igneous processes such as magmatic differentiation, spilitization and other hydrothermal processes.

From physical and chemical viewpoints, many explanations will be possible for these chemical characteristics of the Sambagawa metamorphic rocks. Some attempts from the geochemical standpoint have been made in order to solve the problem on "metamorphic conditions", as stated hitherto. The results, however, are so complicated that a simple and urgent conclusion for such a problem should be avoided, though the problem may be fundamental for the study of metamorphism. The results summarized above are expected to comprise some of peculiar geochemical characteristics which are hardly explained by the usual theories which respect only the static physical conditions and the chemical equilibrium during the regional metamorphism. Moreover, it should be noted that, though most of these facts have not been stressed in the present paper, there are the close relationships of the geological structures on both a regional and a local scale with the mineral assemblages and also with the bulk chemical compositions of the metamorphic rocks, and that the grade of metamorphism indicated by the successive change of mineral assemblage becomes higher in the "upper formations" or towards the apparently shallower depth, concomitantly with intensification of the deformation of rocks. On the present situation, it can be at least said that the effects of the stress pressure or of the pressure gradient caused by orogenesis are the indispensable factors for considering the migration of materials and the recrystallization of minerals during the Sambagawa metamorphism in the Besshi district.

"to be continued on Part III"

Literatures

BANNO, S., 1961, On the so-called "Soda-metasomatism" of the Sambagawa Schists in the Besshi District, (in Japanese): Jour. Geol. Soc. Japan, Vol. 67, pp. 615-525.

DALY, R. A., 1933, Igneous Rocks and Depths of the Earth : McGraw-Hill, New York.

EDWARDS, A. B., 1942, Differentiation of the Dolerites of Tasmania : Jour. Geol. Vol. 50, pp. 451-480.

IWASAKI, I., 1948, Chemistry of Volcanism, (in Japanese) : Kawade Shobo, Tokyo.

KORZHINSKI, D. S., 1948, Phase Rule and Geochemical Mobility of Elements : Internat. Geol. Congress, Rept. 18th Session, Part II, pp. 50-65.

MACDONALD, G. A. 1944, Petrography of the Samoan Islands : Geol. Soc. America, Bull., Vol. 55, pp. 1333-1362.

MUTA, K., 1957, On the Genesis of the Sangasho Type "Kieslager" as Related to Diabases, a Geochemical Study (1), (in Japanese) : Mining Geology, Vol. 7, pp. 254-264.

SUNDIUS, N., 1930, On the Spilitic Rocks : Geol. Mag., Vol. 67, pp. 1-17.

SUZUKI, J., 1930, Petrological Study of the Crystalline Schist System of Shikoku, Japan : Jour. Fac. Sci. Hokkaido Imp. Univ. Ser. IV, 1, pp. 27-111.

TURNER, F. J. and Verhoogen, J., 1951, Igneous and Metamorphic Petrology : McGraw-Hill, Co. Inc.

WALKER, F., 1940, Differentiation of the Palisade Diabase, New Jersey : Geol. Soc. America, Bull. Vol. 51, pp. 1059-1106.

WASHINGTON, H. S., 1917, Chemical Analyses of Igneous Rocks : U. S. G. S., Washington.

———, 1922, Deccan Traps and Other Plateau Basalts : Geol. Soc. America, Bull. Vol. 33, pp. 765-804.

YOSHIZAWA, H., 1954, On the Gabbro of the Cape of Muroto, Shikoku Island, Japan, Part II : Mem. Coll. Sci. Univ. Kyoto, Ser. B, 21, pp. 194-212.