Distribution of Major and Some Minor Elements in Pelitic Metamorphic Rocks in the Ryôke Zone of Central Japan, in Comparison with the Non-metamorphic 'Palaeozoic' Shales*

By

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(Received October 26, 1975)

Abstract

Compilation of chemical analyses of pelitic metamorphic rocks in the Ryôke zone and those of their non-metamorphic equivalents of central Japan is made, and the examination of distribution pattern of each major and minor element in metamorphic rocks is tried zone by zone. The mutual relations between elements are also investigated.

Except the volatile components, the pelitic metamorphic rocks seem to have preserved their original chemical compositions. Metasomatism had not played an important role during the Ryôke metamorphism, except as local phenomena.

Introduction

Isochemical or allochemical process during metamorphism has been controversial from the early days of researches (cf. MEHNERT, 1969). For the Ryôke metamorphic rocks too, metasomatism and its mechanism have been discussed by many authors (KOIDE, 1949, 1958; KANÔ, 1957; HAYAMA, 1962; MIYASHIRO and HARAMURA, 1962; ONO, 1969). In this connection, the comparison between the chemical compositions of the Ryôke metamorphic rocks and those of the 'Palaeozoic'** shales and slates as the non-metamorphic equivalents, has been tried (KOIDE, op. cit.; MIYASHIRO and HARAMURA, 1962, 1966). Recently it is generally accepted that the essential compositional changes during the Ryôke regional metamorphism had not taken place except for the volatile components, such as water and carbon dioxide. Especially MIYASHIRO and HARAMURA rejected the metasomatism as an

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* Read at the 82nd Annual Meeting of the Geological Society of Japan, held in Kyoto, on April 3, 1975 (KUTSUKAKE, 1975a).
** Formerly these formations were regarded as Carboniferous to Permian in age, but recently Triassic fossils and the upper Triassic conodonts are abundantly found from them. Therefore, it may be sure that these formations extend into Mesozoic (∼Jurassic) in age (YOSHIDA and KIMURA, 1974).

absurd thing.   Among the metamorphic zones as well as to those of the non-metamor-
phic equivalents, however, the detailed comparisons of the compositions of the met-
amorphic rocks have not been made so far.   Recently the geological map of the
Ryôke zone, central Japan (scale, 1:200,000) was presented by YAMADA *et al.*
(1974), in which metamorphic zoning is laid down.   Compiling the analyses of pelitic
metamorphic rocks and those of shales and slates, so far available, the comparisons
of chemical compositions between metamorphic rocks and non-metamorphic rocks
will be examined in this paper.

Anyhow, it is well known that pelitic rock is one of the most abundant sedi-
mentary rocks, and the estimation of the abundances of elements in it has been
eagerly made hitherto.   Also in Japan, the chemical data of pelitic rocks are ac-
cumulated more or less.   In this respect, the author wishes to discuss the chemical
characteristics of the pelitic metamorphic rocks in the Ryoke zone, central Japan.

General Geochemistry of Pelitic Rocks

Pelitic rock (shales and slates) is one of the most abundant sedimentary rocks in
space and time.   Chemical studies of pelitic rocks have been carried out from the
various points of view.   Some researchers (e.g., GOLDSCHMIDT, 1933) regarded that
pelitic rocks represent the average chemical composition of the upper earth's crust.
In these situations, the estimation of the abundances of elements in pelitic rocks
and the calculation of the average chemical compositions have been made by many
investigators.   The average chemical compositions for major elements and those
for minor elements in pelitic rocks are shown in Tables 1 and 2 respectively.

Since the pioneer work of MINAMI (1935), chemical data of pelitic rocks of
Japan have been accumulated.   Chemical analyses of Palaeozoic, Mesozoic and
Tertiary pelitic rocks for major elements have been carried out by several investi-
gators (HARAMURA, 1961a,b, 1962, 1963; KATADA *et al.*, 1963, 1964; KANBE *et al.*,
1969; and others).   Especially 'Palaeozoic' shales and slates have been analyzed in
large numbers partly in order to compare with their metamorphic derivatives. But,
for minor elements, data are very scanty, except for the analyses for V, Cr and Ni
done by HARAMURA (*op. cit.*).

According to KATADA *et al.* (1963), chemical characteristics of pelitic rocks in
the northern Kiso range and the Shiojiri-Tatsuno area, where the Ryôke meta-
morphic rocks grade into the non-metamorphic 'Palaeozoic' formations, are as
follows;

1) higher content of SiO_2,
2) higher content of Na_2O and higher ratio of Na_2O/K_2O,
3) lower content of CaO,

in comparison with the Palaeozoic shales of the world. These features can be read from Table 1.

Table 1. Representative average chemical compositions of pelitic rocks.

	A	B	C	D	E	F	G	H
SiO_2	58.9	61.54	58.38	60.15	58.35	61.42	65.10	62.67
TiO_2	0.78	0.82	0.65	0.76	0.83	0.62	0.647	0.73
Al_2O_3	16.7	16.95	15.47	16.45	18.74	17.68	16.98	17.21
Fe_2O_3	2.8	2.56	4.03	4.04	0.73	1.79	1.46	2.36
FeO	3.7	3.90	2.46	2.90	5.53	4.52	3.89	3.11
MnO	0.09	...	...	tr.	0.08	0.09	0.063	0.05
MgO	2.6	2.52	2.45	2.32	2.00	2.41	2.25	2.18
CaO	2.2	1.76	3.12	1.41	0.38	1.66	1.14	0.66
Na_2O	1.6	1.84	1.31	1.01	1.09	2.97	1.78	2.03
K_2O	3.6	3.45	3.25	3.60	2.84	2.70	3.35	4.04
$H_2O(+)$	5.0	3.47	5.02	3.82	4.25	3.05	2.17	2.87
$H_2O(-)$	...	...	...	0.89	1.09	...	0.44	0.59
P_2O_5	0.16	...	0.17	0.15	0.12	0.17	0.073	0.18
CO_2	1.3	1.67	2.64	1.46	0.53	...	...	...
SO_3	...	...	0.65	0.58	...	...	...	...
C	0.6	...	...	0.88	1.91	...	...	1.23
B	0.24	...	...	...	...	...	0.072	...
BaO	...	...	0.05	0.04	...	...	0.000	...
			FeS_2 0.81				ZrO_2 0.004	
							R_2O_3 0.012	
Total	100.27	100.48	100.46	100.46	99.47	99.08	99.43	99.91

A. Average composition of shales, mainly of geosynclines, after WEDEPOHL (1969).
B. Average composition of pelitic rocks, after SHAW (1956).
C. Average of composite analyses of 78 shales, after CLARKE (1916, p. 620).
D. Composite analysis of 51 Palaeozoic shales, by STOKES, after CLARKE (1924, p. 552).
E. Composite analysis of 36 Palaeozoic shales in Europe, after MINAMI (1935).
F. Composite analysis of 14 Palaeozoic shales in Japan, after MINAMI (1935).
G. Composite analysis of 17 non-metamorphosed Palaeozoic shales near Tajimi-chô along Chuô-sen, after KOIDE (1958).
H. Average of 14 pelitic rocks from Northern Kiso district, after KANBE et al. (1969).

Geological Outline of the Ryôke Zone of Central Japan

Geological sketch-map of the Ryôke zone of central Japan is shown in Fig. 1. The metamorphic rocks of this zone are derived mainly from sedimentary rocks and slightly from basic igneous rocks. The metamorphic zonation has been carried out mainly based on the mineralogical variations in pelitic rocks. This was first established by HAYAMA (1956, 1960, 1964) in the Komagane area, where the follow-

Table 2. Average abundances of minor elements in shales
(in parts per million).

	(a)	(b)		(a)	(b)
Li	66	60	Ge	1.6	2
B	100	100	As	13	6.6
F	740	500	Se	0.6	0.6
P	700	770	Br	4	6
Cl	180	160	Rb	140	
Ti	4600	4500	Sr	300	450
V	130	130	Zr	160	200
Cr	90	100	Mo	2.6	2
Mn	850		I	2.2	1
Co	19	20	Ba	580	
Ni	68	95	Ce	59	50
Cu	45	57	Pb	20	20
Zn	95	80	Th	12	11
Ga	19		U	3.7	3.2

(a) TUREKIAN and WEDEPOHL (1961)
(b) VINOGRADOV (1962)
Italicized elements are treated in the present paper.

ing four progressive zones can be distinguished, divided by the respective isograd indicating the metamorphic reaction;

        the biotite zone
    biotite+muscovite→cordierite+K-feldspar+water
        the cordierite zone
    andalusite→sillimanite
        the first sillimanite zone
    muscovite+quartz→sillimanite+K-feldspar+water
        the second sillimanite zone

In the geological map of the Ryôke zone of central Japan (scale, 1:200,000), recently compiled by YAMADA et al. (1974), the metamorphic zoning is based on the above mentioned criteria. But, the discrimination between the first and the second sillimanite zones is not shown in the map.

Chemical Compositions of Sedimentogeneous Metamorphic Rocks in the Toyone-mura Area

(1) Preliminary remarks

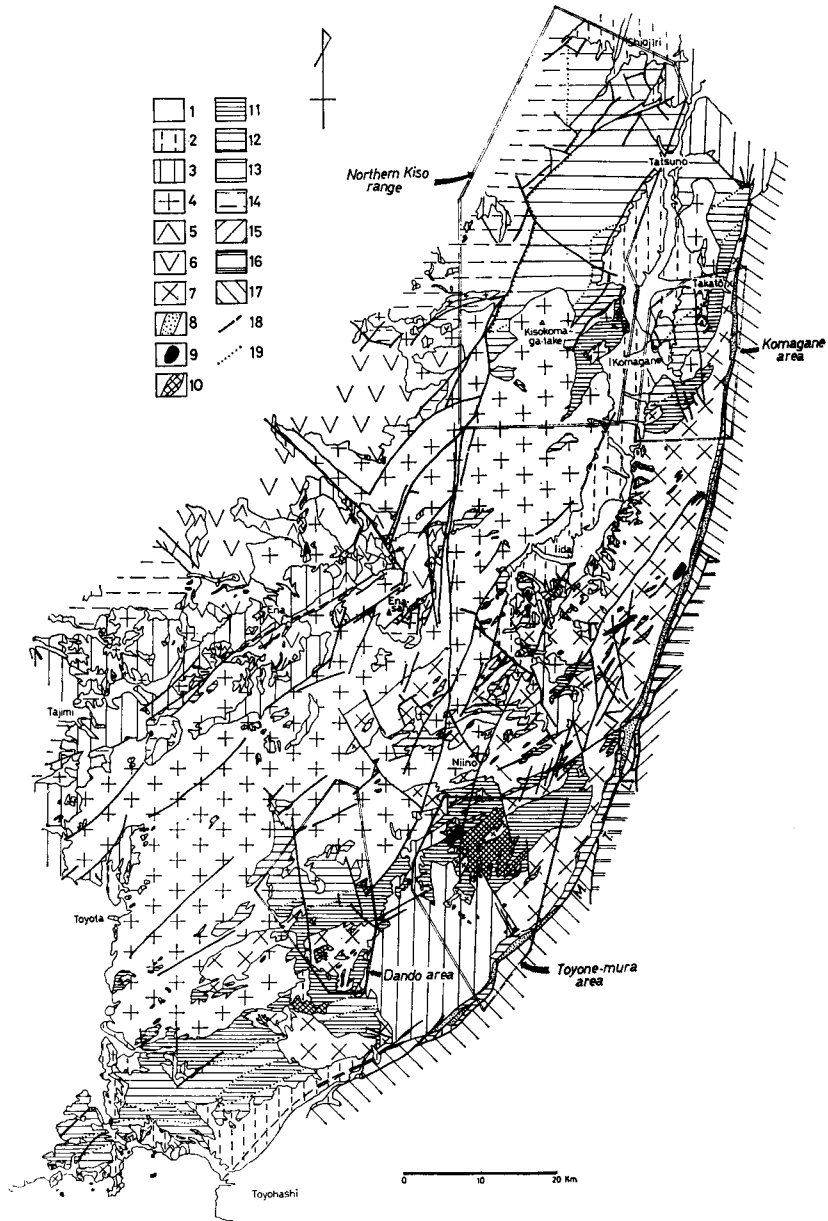During his work on the Ryôke metamorphic rocks in the Toyone-mura area, the

Fig. 1.   Geological sketch-map of the Ryôke zone of central Japan, simplified from YAMADA *et al.*
(1974).
1. Alluvium, 2. Terrace and fan deposits, 3. Neogene system, 4. Post-Nôhi granites, 5. Granite
porphyry, 6. Nôhi rhyolites, 7. Pre-Nôhi granites, 8. Mylonites, 9. Gabbros and cortlandtite,
10. Metadiabase, 11. The sillimanite zone, 12. The cordierite zone, 13. The biotite zone,
14. Non-metamorphic zone, 15. Shimanto formation, 16. Chichibu formation, 17. Sambagawa
crystalline schists, 18. Fault, 19. Boundary between metamorphic zones.

author carried out chemical study as well as their geological and petrological studies. To examine the relationships between the mineralogical compositions and the whole-rock chemical compositions and also to detect the compositional changes during the metamorphism, sixteen metamorphic rocks of sedimentary origin were analyzed for major and minor elements.

In this paper, only chemical compositions of the metamorphic rocks are presented, and the detailed descriptions of geology and petrography and petrological discussions will be given in another paper (KUTSUKAKE, 1975c).

(2) Geological outline

The Toyone-mura area occupies the southeastern part of the Ryôke zone of central Japan (Fig. 1). Brief description of geology of this area has already been presented (KUTSUKAKE, 1970, 1974, 1975b). This area is composed mainly of several kinds of granitic intrusives and their associated metamorphic complex derived from sedimentary and basic igneous rocks. Excepting the hornfels zone along the Median Tectonic Line, almost all part of the area belongs to the sillimanite zone. The hornfels zone belongs to the cordierite zone. In this area, the mineral assemblage of the highest-grade of pelitic rocks is represented by quartz-orthoclase-plagioclase-biotite-muscovite-cordierite-sillimanite, and shows the high-temperature part of the amphibolite facies. The geological map with sampling localities is shown in Fig. 2.

(3) Analytical methods

For major elements, conventional wet chemical analyses were employed except for determination of alkalies by flame photometry. For minor elements, colorimetric and atomic absorption spectrometry were carried out. Analytical method of each minor element is as follows;

V   Colorimetric by phosphotungstic acid after SANDELL (1959),

Cr   Colorimetric by diphenicalbadido after SANDELL (*op. cit.*),

Co ⎫
Ni ⎪
Cu ⎬ Atomic absorption spectrometric, by the method of TERASHIMA (1971),
Zn ⎪
Sr ⎭

Rb   Atomic absorption spectrometric, by the method of TERASHIMA (1973).

The apparatus used for atomic absorption spectrometric analyses is **HITACHI 308.**

Geological Map of the Toyone-mura Area,

Aichi Prefecture, Japan
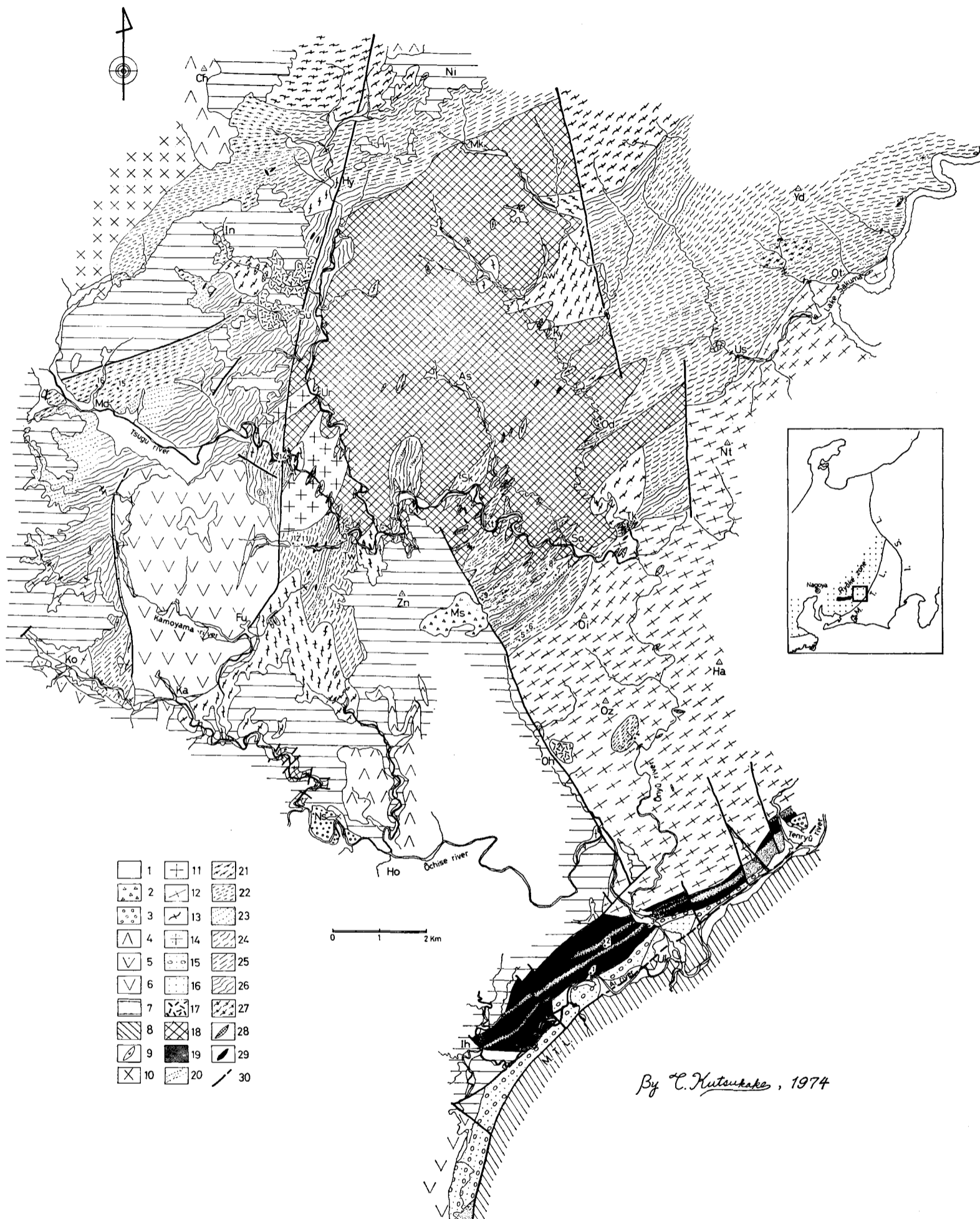
By T. Kutsukake, 1974

Fig. 2. Geological map and sampling localities of the Toyone-mura area.

1: Alluvium, 2: Debris, 3: Terrace deposit, 4: Basalt and andesite, 5: Dacite, 6: Rhyolite, 7: Tertiary sediments, 8: Sambagawa schists, 9: Busetsu granite, 10: Inagawa granite, 11: Mitsuhashi granite, 12: Tenryūkyō granite, 13: Kamihara quartz diorite, 14: Fine-grained biotite granodiorite, 15: Mylonite, 16: "Hälleflinta", 17: Gabbro, 18: Metabasite, 19: Hornfels (pelitic and psammitic), 20: Hornfels derived from chert, 21: Mica schist (pelitic), 22: Mica schist (psammitic), 23: Quartz schist, 24: Gneiss (pelitic), 25: Gneiss (psammitic), 26: Quartz gneiss, 27: Nebulitic gneiss, 28: Metamorphic rock derived from "Schalstein", 29: Marble, 30: Fault.

M.T.L.: Median Tectonic Line, I.S.L.: Itoigawa-Shizuoka Line.

Locality name As: Asakusa, Ch: Chausu-yama, Fu: Futto, Ha: Hanare-yama, Ho: Hongô, Hy: Hiyosawa, Ih: Ichihara, In: Inoshikori, Ka: Kami-Awashiro, Ko: Kobayashi, Kt: Kakinotaira, Ky: Kashiyage, Md: Midashi, Mk: Makinoshima, Ms: Misono, Ni: Niino-tôge Ns: Naka-Shitara, Nt: Nihon-ka-tsuka-yama, Od: Ôdachi, Oh: Ôhata, Oi: Ôiwa-dake, Os: Ôsawa, Ot: Ôtani, Oz: Ôzasa-yama. Sk: Shimo-Kurokawa, So: Sogawa, Su: Sakauba, Tk: Tashika, Ts: Tsugawa, Tw: Tawagane-tôge, Uk: Urakawa, Ur: Ure, Us: Urushijima, Yd: Yatsudake-yama, Zn: Zinno-yama.

Table 3. Chemical compositions of the sedimentogenous metamorphic rocks in the Toyone-mura area.

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
SiO_2	78.28	71.58	68.17	68.14	65.19	78.44	63.83	66.91	72.29	67.42	73.13	73.55	62.63	67.80	64.52	62.37
TiO_2	0.02	0.04	0.49	0.86	0.35	0.36	0.54	0.55	0.53	0.88	0.64	0.62	0.79	0.23	0.38	0.51
Al_2O_3	10.87	14.35	15.12	14.55	17.09	10.97	16.72	15.99	12.17	14.27	12.27	13.55	15.81	15.62	15.35	18.09
Fe_2O_3	0.33	0.55	1.04	0.64	1.02	0.41	0.39	0.55	0.77	1.63	0.28	0.22	0.38	1.19	1.49	1.48
FeO	3.16	2.56	4.22	5.16	3.52	2.17	4.75	4.10	3.23	4.95	4.68	2.67	5.88	5.16	4.31	4.72
MnO	0.36	0.11	0.03	0.01	0.03	0.03	0.07	0.06	0.02	0.22	0.05	0.20	0.09	0.34	0.06	0.06
MgO	1.62	1.33	2.01	1.38	2.54	0.99	2.37	2.25	1.31	2.33	1.73	1.59	2.74	2.49	2.01	2.39
CaO	0.50	1.96	0.58	1.68	1.05	1.35	1.35	2.68	0.79	1.88	0.20	0.89	2.28	0.37	0.70	0.28
Na_2O	1.92	2.49	2.00	2.07	2.33	1.86	1.10	1.90	2.26	1.67	1.42	0.72	4.08	1.67	2.27	2.21
K_2O	1.12	2.28	2.94	2.93	4.33	1.35	5.96	3.14	4.68	3.85	2.31	3.38	3.26	2.44	4.93	4.93
$H_2O(+)$	1.40*	2.64*	2.38*	2.28	1.60*	0.91*	2.87	1.71	1.25*	1.23	2.00*	1.74	2.01*	2.12*	3.23*	3.16*
$H_2O(-)$	0.08	0.15	0.24	0.09	0.27	0.31	0.12	0.06	0.27	0.04	0.35	0.22	0.12	0.12	0.27	0.16
P_2O_5	0.05	0.06	0.05	0.18	0.09	0.03	n.d.	0.17	0.13	0.02	0.01	0.05	0.09	0.09	0.14	0.32
Total	99.71	100.10	99.27	99.97	99.41	99.18	100.07	100.07	99.70	100.39	99.07	99.40	100.16	99.64	99.66	100.68

C.I.P.W. Norms

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
Q	56.60	40.38	38.58	36.35	26.74	56.97	24.70	32.21	35.63	32.39	50.12	50.36	13.16	40.38	25.42	23.98
C	5.71	4.38	7.75	5.30	6.83	4.18	6.02	4.89	2.14	3.98	7.04	7.14	1.53	9.67	5.30	9.07
or	6.67	13.34	17.24	17.24	25.58	7.78	35.03	18.35	27.80	22.80	13.90	20.02	19.46	14.46	28.91	28.91
ab	16.24	20.96	16.77	17.29	19.91	15.72	9.43	16.24	19.39	14.15	12.05	6.29	34.58	14.15	19.39	18.86
an	2.22	9.45	2.50	7.51	4.73	6.39	6.67	12.51	3.06	9.17	0.83	4.17	10.84	1.39	2.78	—
en	4.00	3.30	5.00	3.40	6.30	2.50	5.90	5.60	3.30	5.80	4.30	3.90	6.80	6.20	5.00	5.90
fs	6.20	4.49	6.20	7.52	5.15	2.90	7.66	6.34	4.36	6.73	7.39	3.80	9.37	8.71	6.20	6.86
mt	0.46	0.70	1.51	0.93	1.39	0.70	0.46	0.70	1.16	2.32	0.46	0.70	0.46	1.86	2.09	2.09
il	0.05	0.08	0.91	1.67	0.61	0.76	1.06	1.06	1.06	1.67	1.22	1.22	1.52	0.46	0.76	0.91
ap	0.13	0.13	0.13	0.34	0.20	0.07	...	0.34	0.34	0.03	0.03	0.13	0.20	0.20	0.34	0.67

* Ignition loss   Anal. T. Kutsukake

Table 4. Minor elements in the sedimentogenous metamorphic rocks in the Toyone-mura area (in ppm).

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
V	...	...	202	180	38	...	41	101	50	...	43	48	100	...	67	33
Cr	...	...	27	39	25	...	49	37	21	...	19	19	39	...	27	45
Co	19	10	17	16	24	25	26	34	24	...	43	16	26	16	17	17
Ni	34	37	47	33	39	32	58	55	43	...	53	47	71	83	52	53
Cu	24	16	30	37	13	4	16	46	18	...	32	42	27	2	6	17
Zn	62	68	109	91	117	66	102	338	123	...	101	95	123	126	112	112
Rb	65	80	138	218	189	52	154	124	114	...	138	125	146	88	180	204
Sr	151	229	160	176	169	37	159	245	245	...	125	147	195	230	135	153

... : not determined.   Anal. T. Kutsukake

(4)   Analytical results

Compositions for major elements and the calculated C.I.P.W. norms are given in Table 3.   Nos. 6 and 14 are of psammitic rock origin. Nos. 1, 2 and 9 are of highly siliceous shale origin.   The others are derived from normal pelitic rocks.

The analytical results of some minor elements in these rocks are given in Table 4.

Brief petrography of analyzed rocks is given in Appendix.

Chemical Compositions of Pelitic Metamorphic Rocks and Non-metamorphic 'Palaeozoic' Shales

(1)   Preliminary remarks

On pelitic metamorphic rocks of the Ryôke zone and non-metamorphic pelitic rocks in central Japan, chemical analyses are compiled here.   The latter rocks can be regarded as the equivalents of the original rocks of the formers.   The abundance of major and minor element in each metamorphic zone is shown as histogram.   The chemical variation between the zones and also the mutual relations between elements will be examined.

(2)   Source of data

The chemical analyses used in this report were taken from the literatures listed below.

The data of clearly non-pelitic and ambiguous rocks judged from their lithological nature are excluded from compilation.   The data of the rock without petrography are treated that the zone, it belongs to, can be estimated from its sampling locality.

i)   Non-metamorphic 'Palaeozoic' shales and slates
    Tatsuno (=Tatuno)-Shiojiri (=Siojiri) area
      HARAMURA (1961b)
    Northern Kiso range
      ÔKI (1958)
      KATADA et al. (1963)
    Tajimi area
      IKAWA (1942) and KOIDE (1958)
ii)   Pelitic metamorphic rocks
    Takatô-Shiojiri area
      ONO (1969)

Komagane area
  HAYAMA (1964)
Northern Kiso range
  ŌKI (1958, 1961)
  KATADA *et al.* (1963) and KATADA (1967)
Niino area
  SAKAKIBARA (1963)
Toyone-mura area
  KUTSUKAKE (1970, this paper)
Dando area
  IKAWA (1942) and KOIDE (1958)
Other areas
  SHIBATA (1968)

(3) Number of analyses used in the compilation

Zone	Number
Non-metamorphic	28
Biotite zone	20
Cordierite zone	19
Sillimanite zone	39
Total	106

(4) Presentation of data in the form of diagrams

Major elements

I) Range in composition

Histograms have been drawn for seven major oxides in terms of zonal classification (Fig. 3). For titanium, manganese and phosphorus oxides, the histograms are shown in Fig. 4 as well. With regard to the histograms, some characteristic features of each element will be mentioned.

i) *SiO_2*

Most of the non-metamorphic pelitic rocks fall in the range of 58 to 70 wt. per cent. But, the metamorphic ones frequently fall out of the above range. This feature is conspicous in both the biotite and the cordierite zones. It is probably due to the situation that the term "pelitic" in metamorphic petrology implies the wider meaning than that in sedimentary petrology. Accordingly, besides the normal pelitic rocks, highly siliceous ones and others have been included into "pelitic" metamorphic rocks.
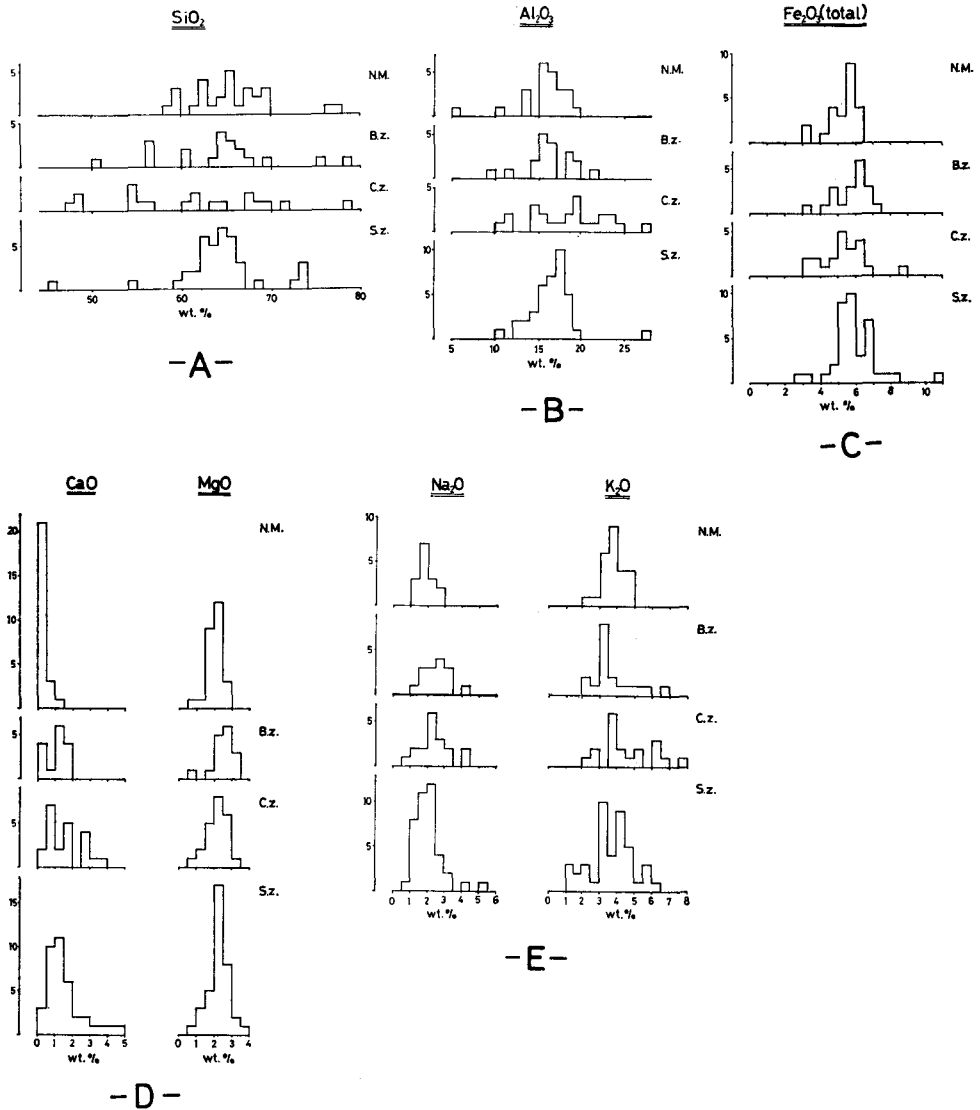
Fig. 3. Histograms for major element oxides of pelitic metamorphic and non-metamorphic rocks in zonal classification.
N.M.: Non-metamorphic, B.z.: The biotite zone, C.z.: The cordierite zone, S.z.: The sillimanite zone. Numbers of the ordinate represent the numbers of analyses.

ii) Al_2O_3

Most of the analyses fall in the range between 14 and 20 wt. per cent. In the
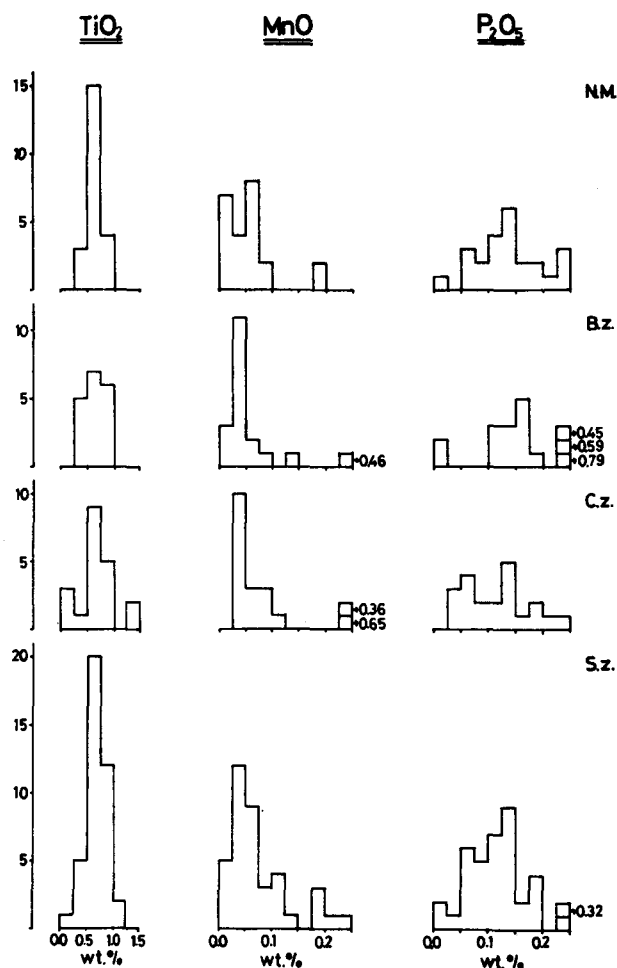
Fig. 4.   Histograms for titanium, manganese and phosphorus oxides.

cordierite zone, highly aluminous ones are persent.   It may be explained as follows; in that zone andalusite occurs as one of the typical minerals in highly aluminous rocks (usually $Al_2O_3 > 20$ wt. per cent, HAYAMA, 1964, pp. 333–334), and such rocks bearing andalusite are analyzed selectively to know their chemical characteristics.

iii) Fe_2O_3 (total)

In all of the zones, the concentration of analyses is recognized in the range between 4.5 and 7.0 wt. per cent.   Any particular tendency can not be detected in the histogram with increasing metamorphism.

iv) *CaO*

In the non-metamorphic pelitic rocks, the CaO content is less than 0.5 wt. per cent, but in the metamorphic ones, it is generally between 0 and 2 wt. per cent. In the higher metamorphic zones, the rocks with high CaO content (>2 wt. per cent) are present. And the apparent increase of CaO content with increasing metamorphism can be recognized. The similar feature has been reported by SHAW (1956) for the metamorphic rocks in the Appalachian region of New Hampshire. But, the present rocks, it is not necessarily attributed to the metamorphic effects, for the localities of analyzed specimens are maldistributed to the northern Kiso range where the rocks are characteristically poor in CaO (KATADA *et al.*, 1963). Incidentally the non-metamorphic shale in the Tajimi area (Table 1, G) is not so poor in CaO, and the value is almost the same as that of the metamorphic rocks. Thus, the original difference of CaO content has also to be taken into account.

v) *MgO*

It shows an unimodial distribution in every zone.   The differences of the range and the peak among the zones can not be detected.   The constancy is well guaranteed.

vi) *Na$_2$O*

Most of the analyses fall in the range between 1.0 and 3.5 wt. per cent.   With progressive metamorphism, neither increase nor decrease of soda can be detected.

vii) *K$_2$O*

The range in the non-metamorphic rocks is rather small, but the range in the metamorphic ones is wide and the distribution patterns are irregular.   In the metamorphic zones rocks over 5 wt. per cent K$_2$O are present, moreover, in the sillimanite zone low-potash rocks are also present.   However, the peak of abundance does not shift remarkably in any particular zones.

viii) *TiO$_2$*

The content of titania is almost constant in all of the zones, and it shows a normal distribution between 0.25 and 1.00 wt. per cent.

ix) *MnO*

MnO content is usually less than 0.1 wt. per cent in these pelitic rocks, but most of garnetiferous rocks of the metamorphic zone show more than 0.2 wt. per cent MnO.   There is a concentration around 0.05 wt. per cent in every zone.

x) P_2O_5

P_2O_5 shows no regular distribution patterns, scattering between 0.05 and 0.20 wt. per cent, and the weak concentration around 0.15 wt. per cent can be recognized. The abnormally high values, more than 0.50 wt. per cent, are worth to doubt the analyses.

Summing up the above mentioned features, the following statement could be presented as a rough conclusion.

Many oxides, except CaO and K_2O, fall within the ranges of non-metamorphic rocks, and no regular variations can be detected with increasing metamorphism. The presence of rocks with high CaO content in the high-grade zones is conspicuous and should be paid attention to. The range of K_2O content seems to become wider with increasing metamorphism.

II)   The relationships between the oxides

(A) Na_2O–K_2O

In Fig. 5 all the analyses are plotted. The most conspicuous feature in this diagram is that the non-metamorphic rocks are concentrated in small area, but the metamorphic ones are scatter in wide area. Most of the non-metamorphic ones fall in the limit of the Inner zone by MIYASHIRO and HARAMURA (1962). On the other hand, the metamorphic ones are abundantly present also out of the limit, although a slight concentration with some tendencies can be recognized.

N.M. : Non-metamorphic zone.
B.z. : The biotite zone.
C.z. : The cordierite zone.
S.z. : The sillimanite zone.
1   : The limit of the Inner zone,
O   : The limit of the Outer zone, after MIYASHIRO and HARAMURA (1962).
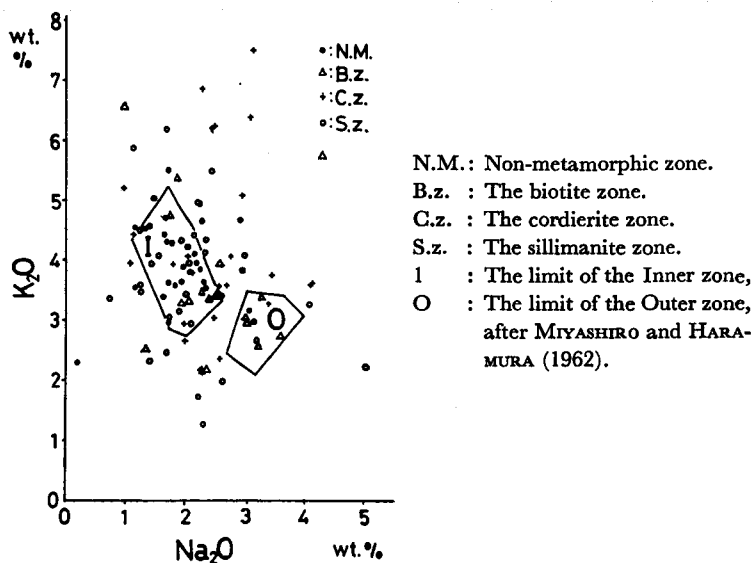
Fig. 5. $Na_2O - K_2O$ relations.

(B)   CaO–Na$_2$O–K$_2$O

In Fig. 6 the non-metamorphic rocks are in an area near the Na$_2$O–K$_2$O side with few exceptions. As already mentioned, the pelitic rocks in the northern Kiso range are poor in CaO, and its nature is revealed in the diagram. But, the original rocks of the metamorphic rocks are not necessarily so, as represented by the shale of the Tajimi area.
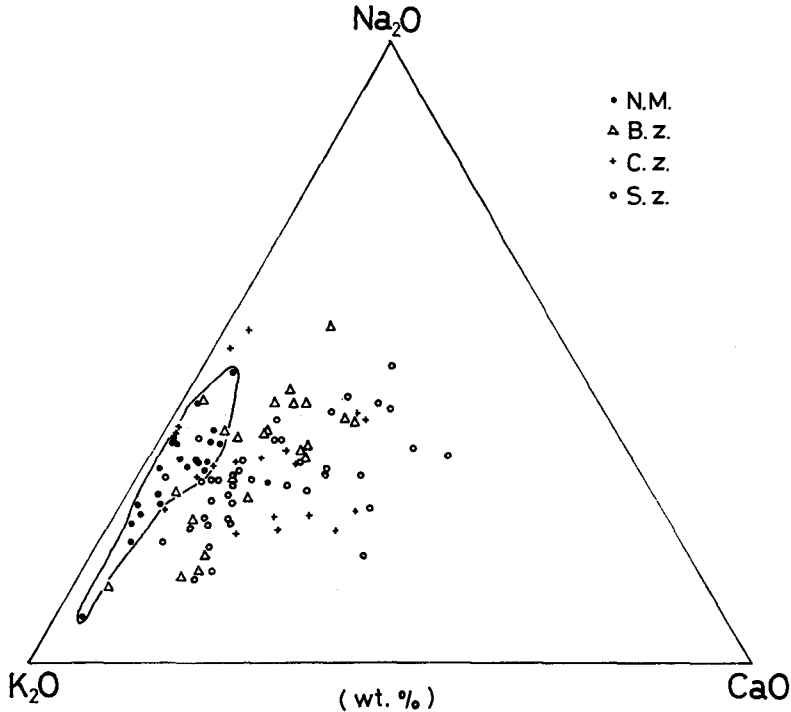

Fig. 6.   CaO–Na$_2$O–K$_2$O relations.
The symbols are the same as in Fig. 5.

(C)   H$_2$O (+)–FeO/ (FeO+Fe$_2$O$_3$)

Between the content of H$_2$O (+) and ratio of FeO/(FeO+Fe$_2$O$_3$), no causal relation seems to be existed. H$_2$O (+) is chosen as a representative of the volatile components, such as the commonest and the most abundant volatile, and measured in usual analyses.

Decrease of volatiles with increasing metamorphism has been reported from many metamorphic terranes, but in the diagram (Fig. 7), H$_2$O (+) decreases slightly with increasing metamorphism, especially in the rocks of the sillimanite zone.

On the other hand, it has been recognized in several metamorphic terrains that

reduction of iron from ferric to ferous advances with progressive metamorphism. In the Ryôke metamorphic belt, this fact was first pointed out by KOIDE (1958), and later by KATADA *et al.* (1963) and ONO (1969). This phenomenon can be theoretically explained as follows; graphite is oxidized to carbon dioxide during metamorphism, as the result reduction of iron takes place, as shown by MIYASHIRO (1964).

The above-mentioned both features are revealed in the diagram (Fig. 7), that the trend can be traced by a line declining toward right.
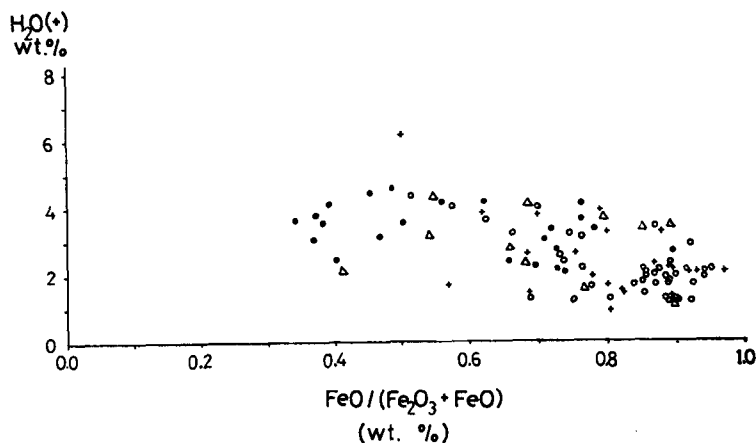
Fig. 7. $H_2O(+) - FeO/(FeO + Fe_2O_3)$ relations.
The symbols are the same as in Fig. 5.

Minor elements

Minor element data of pelitic metamorphic rocks in the Ryôke zone and of non-metamorphic ones are scarce, except for the data of V, Cr and Ni in 'Palaeozoic' slates (shales) by HARAMURA (1961a,b, 1962). For pelitic metamorphic rocks, only the author's data are available.

The histograms for minor elements are shown in Fig. 8. The content of Cr has almost the same range in both metamorphic and non-matamorphic ones. Ni content is slightly higher in the metamorphic rocks than in the non-metamorphic ones. V is similar in both. For other elements the comparison can not be made, as the data are absent for non-metamorphic rocks.

To compare the minor elements content in the metamorphic and the non-metamorphic rocks of the present subject with the average values (Table 2), the formers are rich in Co, poor in Cr, Ni, and Sr, and similar in V, Cu, Zn and Rb.

Cr–Ni relations

MIYASHIRO and HARAMURA (1962) demonstrated that the relation between Cr and Ni is slightly different in pelitic rocks of the Inner zone and the Outer zone of Southwest Japan.   As shown in Fig. 9, the data of the pelitic metamorphic rocks in the Toyone-mura area fall almost in the range of Inner zone, but some of them are slightly higher in Ni content.
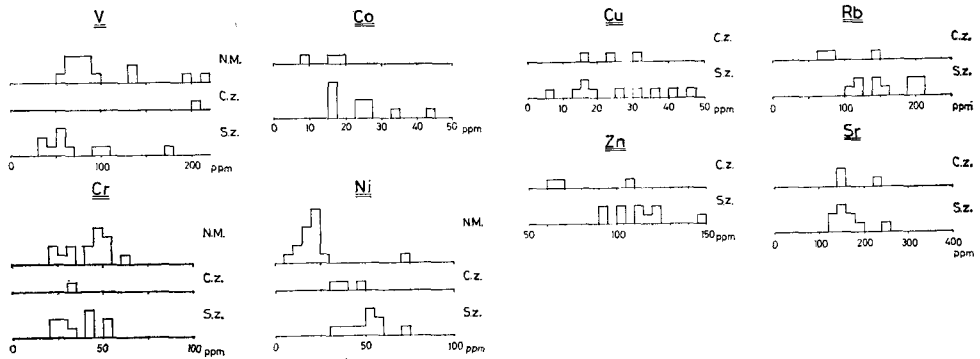
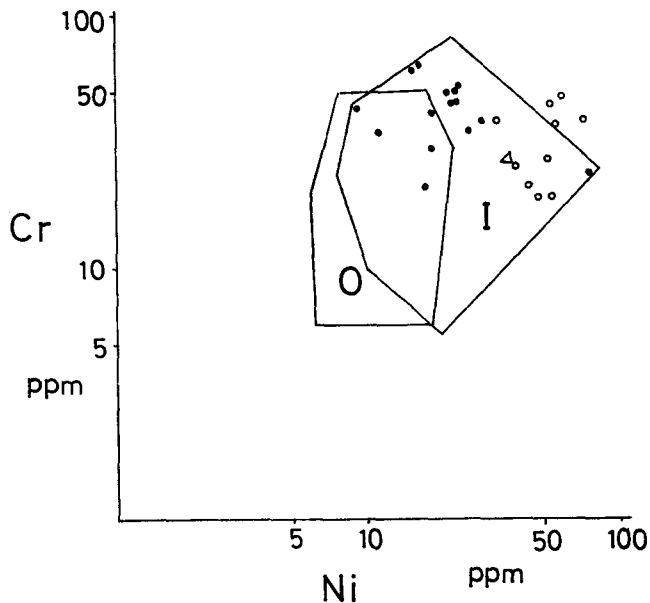Fig. 8.   Histograms for minor elements of pelitic metamorphic and non-metamorphic rocks in zonal classification.

Fig. 9.   Ni–Cr relations.
The symbols are the same as in Fig. 5.

Discussions

(1) General features of chemical compositions of pelitic metamorphic rocks in the Ryôke zone of central Japan

It has generally been accepted that the original heterogeneity of rocks is well preserved even after the metamorphism. In the study of a series of increasingly metamorphosed pelitic rocks from shales to sillimanite schists and gneisses of the Littleton formation of New Hampshire, Shaw (1954a,b) demonstrated that the concentration of elements remained constant during the metamorphism. Also, in the examination of 33 chemical analyses of phyllites, mica schists and sillimanite gneisses from the Central Pyrenees (Zwart, 1959), Mehnert (1969) showed that only a few significant changes can be observed during the progressive metamorphism. Thus, in normal progressive metamorphism remarkable compositional changes generally do not take place. Therefore, unless any distinct tendencies in compositional changes during metamorphism can be detected, to assume the metasomatism in the process is difficult to be accepted. As above mentioned, for major elements, any directional changes can not be detected for the Ryôke metamorphic rocks of central Japan, except for the fact that CaO content of the metamorphic rocks is higher than in the non-metamorphic rocks. But the reason for the variation in CaO content is not certain due to either metasomatic change or original difference. As a result, it can be stated that during the Ryôke regional metamorphism no essential changes in compositions of rocks had taken place. But, the migration of some elements in a limited extent must have operated through such processes as metamorphic differentiation (Koide, 1958) and metasomatic transfer due to the difference of water-vapour pressure place by place (Hayama, 1962).

In the Ryôke zone of the Yanai district, mixing of granitic materials with sedimentary metamorphic rocks is recognized and the migmatitic rocks are formed (Okamura, 1958). In that case, the exchange of materials between metamorphic rocks and granitic magma is well operative (Honma, 1974), so that the compositions of metamorphic rocks must have been considerably changed. But, in other areas of the Ryôke zone, as the preservation of original compositions of metamorphic rocks is safely assumed, excluding the volatile components, it may be possible that the metasomatism has not played an important role in the Ryôke regional metamorphism. However, in the case where the granitic intrusives are concerned, the features are not so simple; if the contact effects by granites are marked and the impregnation occurs, the reliable departure from the original compositions must have taken place.

(2) Na_2O-K_2O relations

Behaviour of Na and K is different from each other during metasomatism, as

discussed by many petrologists (e.g., MEHNERT, 1968). According to MEHNERT (1960), in the case of granitization in central Black Forest (Schwarzwald), Kinzigtal, Na_2O stays almost constant, but K_2O increases regularly with increasing granitization. For the Ryôke metamorphic rocks, KANÔ (1957) insisted on the different behaviour of Na and K in the Dando area from the data of KOIDE (1949). As already mentioned, Na_2O–K_2O relations of the pelitic metamorphic rocks in the Ryôke zone in central Japan show no such a regularity but remarkable randomness. K_2O does neither increase nor decrease in any particular zones. Large scale transfer or metasomatic movement of potassium would not have taken place during the Ryôke regional metamorphism.

In another paper (KUTSUKAKE, 1975b), the author discussed that the basic rocks have suffered intense alkali metasomatism during the Ryôke regional metamorphism. The reasons why the obvious metasomatism can not be detected in pelitic metamorphic rocks may be as follows;

1) Metasomatism plays different roles for different original rocks; the alkali-rich rocks such as pelitic ones may have been escaped from the effect.

2) Although they should accept the effect of alkali metasomatism, the result is not apparently revealed in chemical compositions and is almost negligible.

(3) Contact metamorphism and metasomatism

Near the contact of the granitic intrusive rocks, the metamorphic rocks have suffered contact effects, i.e., sillimanite is replaced by muscovite and large muscovite with the texture of porphyroblast-like cross-cutting is developed. They owe their reaction and formation to the effects by granite as shown by ONO (1969). Crystallization of muscovite requires potash first of all, which has been probably derived from the granite. This potash metasomatism would be very similar to the post-magmatic one in the course of the granitic magmatism (ESKOLA, 1956).

(4) Retrogressive metamorphism

Retrogressive metamorphism takes places during the upheaval of the metamorphic belt and in the retrograde stage of the regional metamorphism. The retrogressive metamor phism has been discussed for the Ryôke metamorphic rocks by several authors (KATADA, 1967; ISHIOKA, 1974). In this case, cordierite is replaced by pinite and/or sericitic mica and also sillimanite by epitaxial muscovite. In this case, similar to the case of the contact metamorphism, addition or subtraction of some elements must have taken place. But, precise detection has not been examined so far, it requirs for concrete study.

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Acknowledgements

The atomic absorption spectrometer, **HITACHI 308**, was purchased from Grant in Aid for the Scientific Research of the Ministry of Education, defrayed to Dr. D. Shimizu of Kyoto University.

Part of atomic absorption analyses was carried out by use of the spectrometer, supervised by Prof. K. Shigezawa of College of Arts and Sciences, Kyoto University. The author is very much indebted to these gentlemen.

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Appendix A Brief Petrography of Rocks Analyzed

No. 1   Specimen No. 71032608

   Hornfels, derived from siliceous shale

       Quartz-plagioclase-biotite-muscovite-garnet

       (Graphite (dusty)-apatite)

       Very fine-grained, quartz forms mosaic, biotite and muscovite are in parallel growth and show preferred orientation.

No. 2   Specimen No. 71032603

   Hornfels

       Quartz-plagioclase-K-feldspar-biotite-muscovite

       (Zircon-tourmaline-graphite-ore)

       Partly crushed. Plagioclase is polysynthetically twinned and weakly zoned. Myrmekite is developed inside the plagioclase which contacts with K-feldspar. Biotite is elongated flakes, pleochroic with X=yellow, Y=Z=reddish brown.

No. 3   Specimen No. 71032708

Hornfels

Quartz-plagioclase-muscovite-biotite-cordierite-andalusite

(Tourmaline (abundant)-graphite-ore-apatite)

Mica shows preferred orientation.   Muscovite is porphyroblastic and in parallel intergrowth with biotite.   Biotite is pleochroic with X=pale yellow, Y=Z=red brown.   Andalusite is replaced by muscovite from the periphery.   Cordierite shows sieve structure.   Tourmaline is zoned with greyish brown core and yellowish brown rim.

No. 4   Specimen No. 67122202

Mica schist

Quartz-plagioclase-K-feldspar-biotite-muscovite-cordierite-sillimanite

(Graphite-ore-tourmaline-apatite-zircon)

Fibrous sillimanite replaces part of biotite.   It is also developed in muscovite flakes.   Muscovite is usually in parallel intergrowth with biotite, but, larger crosscutting one is also present.   Cordierite is almost always replaced by sericitic muscovite and/or pinite.   K-feldspar shows fine streak-perthite structure.

No. 5   Specimen No. 68122004

Gneiss

Quartz-plagioclase-K-feldspar-biotite-muscovite-cordierite

(graphite-ore-zircon-apatite)

Banded with black biotitic seam and white quartzo-feldspathic seam.   Plagioclase is tabular and weakly zoned.   Myrmekite is developed.   K-feldspar is perthitic.   Cordierite is almost altered to pinite and/or sericitic mica.   Muscovite occurs as two forms; in parallel growth with biotite and large cross-cutting.   Biotite is pleochroic with X=yellow, Y=Z=brown.

No. 6   Specimen No. 68122005

Gneiss, psammitic

Quartz-plagioclase-K-feldspar-cordierite-biotite-muscovite-andalusite

(Ore-tourmaline-graphite-zircon-apatite)

Plagioclase is xenomorphic and polysynthetically twinned.   Cordierite is abundant and partly altered along crack, and it includes granular grains of quartz. Biotite is tiny scaly flakes and shows parallel arrangement.   It is pleochroic with X=nearly colourless, Y=Z=brown.   Muscovite is small in amount, and in parallel intergrowth with biotite.   Andalusite is irregular-shaped grains, sometimes associated with ore and/or biotite.

No. 7   Specimen No. 69121504

  Gneiss

    Quartz-plagioclase-K-feldspar-biotite-muscovite-cordierite-sillimanite

    (Graphite-ore-apatite-zircon)

    K-feldspar is very small in amount.   Biotite is pleochroic with X=yellow, Y=Z=red brown.   Muscovite is in parallel intergrowth with biotite, but larger cross-cutting ones are also present.   Cordierite is completely replaced by muscovite (lower birefringence, sericitic).   Sillimanite is fibrolite, partly derived from biotite and epitaxially replaced by muscovite.

No. 8   Specimen No. 68032908

  Gneiss

    Quartz-plagioclase-biotite-muscovite-cordierite

    (Ore-graphite-apatite-zircon)

    Cordierite is completely replaced by epitaxial mica.

No. 9   Specimen No. 70112201

  Gneiss, rather psammitic

    Quartz-plagioclase-K-feldspar-biotite-muscovite-cordierite-andalusite

    (Apatite-ore-graphite-zircon)

    Biotite is tiny scaly flakes and pleochroic with X=yellow, Y=Z=red brown. Muscovite occurs in two forms; small and in parallel intergrowth with biotite and larger cross-cutting.   Cordierite is frequently replaced by epitaxial muscovite. Andalusite is elongated crystals and replaced by muscovite from the margin.

No. 10   Specimen No. 68081902

  Gneiss

    Quartz-plagioclase-K-feldspar-biotite-muscovite-sillimanite-cordierite

    (Zircon-apatite-ore)

    Muscovite is large cross-cutting porphyroblastic flakes.   Sillimanite is needle-like crystals in muscovite flakes.   Cordierite always occurs as pseudomorph, replaced by muscovite.

No. 11   Specimen No. 68050203

  Gneiss

    Quartz-plagioclase-K-feldspar-biotite-muscovite-cordierite-sillimanite

    (Graphite-ore-apatite-zircon)

    Sillimanite occurs as felty aggregate of fibrolite.

No. 12   Specimen No. 68050202

Gneiss

Almost the same as No. 11.

No. 13   Specimen No. 70082502

Gneiss

Quartz-plagioclase-biotite-muscovite-cordierite

(Graphite-ore-apatite-zircon)

Plagioclase is weakly zoned.   Cordierite shows sieve structure, and partly replaced by muscovite.

No. 14   Specimen No. 70092003

Gneiss, psammitic

Quartz-plagioclase-garnet-biotite-muscovite-cordierite

No. 15   Specimen No. 70092102

Mica schist

Quartz-plagioclase-biotite-muscovite-sillimanite

(Tourmaline-opaques-zircon-apatite)

Spots of aggregate of biotite, muscovite and sillimanite are developed.

No. 16   Specimen No. 70092103

Mica schist

Quartz-plagioclase-andalusite-sillimanite-biotite-muscovite

(Tourmaline-opaques-zircon-apatite)

Biotite and muscovite partly change to sillimanite (felty aggregate of fibrolite). Andalusite is replaced by muscovite from the periphery.