

# Metamorphic white micas : definition of paragenetic fields

Autor(en): **Cipriani, C. / Sassi, F.P. / Scolari, A.**

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# Metamorphic White Micas: Definition of Paragenetic Fields

By *C. Cipriani* (Firenze)\*, *F. P. Sassi* and *A. Scolari* (Padova)\*\*)

With 12 figures and 4 tables in the text

## Summary

The present work is based on the statistical analysis, carried out globally and by classes, of about 400 chemical analyses of white micas (muscovite-phengite). In the first part, the frequency distributions of the element contents and their reciprocal correlations are studied. In the second part, by means of paragenetic fields, the influence of the intensive and extensive variables on the chemical composition of these minerals is shown. The results are discussed from the petrological point of view, and compared with the available experimental data.

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\*) Istituto di Mineralogia, Petrografia e Geochimica dell'Università di Firenze (Italia) e „Centro di Studio per la Geochimica e la Cristallografia dei minerali delle rocce del C.N.R.“

\*\*\*) Istituto di Mineralogia e Petrografia dell'Università di Padova (Italia) e „Centro di Studio per la Geologia e la Petrologia delle Formazioni Cristalline del C.N.R.“

Address: Dr. F. P. Sassi, University of Padova, C. Garibaldi 9, 35100 Padova (Italy).

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## 1. INTRODUCTION

The name "white micas" is generally given to those dioctahedral micas of petrological importance, above all muscovites, paragonites and phengites. While muscovite and paragonite are real mineralogical species, the phengites are to be considered as *ex compositione* varieties of muscovite or, better, intermediate terms between ideal muscovite and the hypothetical end members of "celadonite".

The white micas prevailing in metamorphic rocks are muscovites which frequently show clear deviations from the theoretical composition, owing to isomorphous substitutions both in the interlayer positions (towards paragonite end member) and in the octahedral and tetrahedral ones (towards phengite).

This research aims at establishing the range of such deviations by means of the statistical study of about 400 chemical analyses taken from the most recent literature, and to correlate such deviations with the environmental conditions which were active at the time of the metamorphic recrystallisation of the rocks.

Numerous investigations have shown that the composition of metamorphic white micas (muscovite-phengite group) are influenced by the environmental conditions. Among others, the contributions of the following authors have been significant in this respect: EUGSTER and YODER (1955a), LAMBERT (1959), CHINNER (1960), ERNST (1963, 1964), IYAMA (1964), EVANS (1965), VELDE (1965b, 1966, 1967a), CRAWFORD (1966), EVANS and GUIDOTTI (1966), BROWN (1967, 1968), BUTLER (1967), GRAESER and NIGGLI (1967), CIPRIANI, SASSI and VITERBO-BASSANI (1968), SCHWANDER, HUNZIKER and STERN (1968), GUIDOTTI (1969).

Excluding experimental investigations, the above-mentioned contributions were mainly concerned with the detection of compositional variations of white micas in relatively narrow thermal ranges, with the exception of the last three works. In particular, the work carried out by SCHWANDER, HUNZIKER and STERN (1968) over a large area of the Central Alps shows clearly a close agree-

ment between the zoneography deduced from the study of white micas and that established by other means, while CIPRIANI, SASSI and VITERBO-BASSANI (1968) pointed out the systematic variations of the Na, Mg, Fe and Si contents with the metamorphic grade in the whole stability field of white micas, for the barrovian-type facies series.

The recognition of systematic compositional variations of white micas depending on environmental conditions is not only important from the petrological point of view, but it can also be of enormous practical usefulness both in the study of metamorphic belts and of the clastic sedimentary formations. In fact, it would be possible to recognize, when more immediate criteria are lacking, a zoneography for the former, and for the latter the nature of the parent-rocks and therefore the paleogeographical reconstructions.

It is for this reason that we deal with the problem of the definition of paragenetic fields for the metamorphic white micas (excluding paragonite), as was done by DOBRETsov (1968) for metamorphic pyroxenes, and by KOSTYUK and SOBOLEV (1969) for metamorphic amphiboles.

To this purpose about 400 chemical analyses of white micas have been examined from the literature and, where possible, these have been classified into the following groups:

1. subfacies quartz – albite – muscovite – chlorite;
2. subfacies quartz – albite – epidote – biotite;
3. subfacies quartz – albite – epidote – almandine;
4. subfacies staurolite – almandine;
5. subfacies kyanite – almandine – muscovite;
6. subfacies sillimanite – almandine – muscovite;
7. orthoclase isograd;
8. granites;
9. pegmatites and veins;
10. glaucophanitic greenschist facies;
11. low-pressure facies series.

The first seven groups fall within the medium to high-pressure facies series: the pressure is higher than that of the triple point andalusite-kyanite-sillimanite, and lower than that of the appearance of glaucophane.

The global statistical analysis of chemical data allowed us to study, from the mineralogical point of view, the frequency distributions of the element contents and the correlations between them. On the other hand, the statistical analysis for classes allowed us to recognize, for each of the above-mentioned groups, well-defined paragenetic fields and, moreover, to detect the general trends of systematic variations in function both of temperature and of pressure; it was also possible to evaluate the effects of the bulk composition of the rocks.

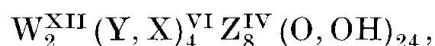
The chemical data are reported in the Appendix.

## 2. CALCULATION OF THE FORMULA

The examination of analytical data of white micas from most recent literature has shown that there is not yet unanimous agreement concerning the way to calculate crystallochemical formulas. Even if most authors carry out their calculations using  $O + OH + F = 24$ , calculations based on 44 valencies or 22 O are also used. ERNST (1963) justly states the problem of the impossibility of an exact calculation of the formula when precise data regarding the density and volume of the unit cell are lacking. However, of the three methods of calculation suggested by this author, the most logical seems to be the first one ( $O + OH + F = 24$  ions), owing to the fact that in any oxygenated mineral the structural framework is made up of oxygens and the calculation of any hydroxium ions is rather hazardous and also probably unfounded in the case of non-sedimentary micas.

For the treatment of the data, therefore, the formulas on the basis of  $O + OH + F = 24$  were recalculated for those micas on which the various authors had operated differently.

The calculations were carried out with reference to the crystallochemical formula usually accepted for muscovite:



were  $W = K, Na, Ca$ ;  $Y = Al, Fe^{+3}, Ti$ ;  $X = Mg, Fe^{+2}$ ;  $Z = Si, Al$ .

## 3. GLOBAL STATISTICAL ANALYSIS

## 3a. Frequency distributions

The frequency distributions of the contents of the various ions, the number of the interlayer and octahedral sites occupied (the tetrahedral ones being established at 8,00) and the number of charges (interlayer, octahedral, tetrahedral and total) were all examined.

For each of these parameters we have calculated the mean  $\bar{x}$ , the standard deviation  $s$ , the median  $me$ , the mode  $mo$  and the Pearson's skewness coefficient  $\alpha = \frac{\bar{x} - mo}{s}$  (Table 1).

The frequency distribution, obtained by grouping the data into classes of about  $s/2$  intervals, are almost all unimodal, generally more or less strongly asymmetrical with considerable departures from the normal distribution.

The distributions of the contents of the octahedral ions have reciprocal configurations, showing isomorphous substitution: the distribution of Al has a strongly negative skewness while the distributions of  $Fe^{+3}$ ,  $Fe^{+2}$  and Mg show positive skewness. The prevailing type is a muscovite not too far from

Table 1. Number of samples ( $n$ ), arithmetic means ( $\bar{x}$ ), standard deviations ( $s$ ), medians ( $me$ ), modes ( $mo$ ) and Pearson's skewness coefficients ( $\alpha$ ) of the element contents in white micas.

	n	$\bar{x}$	s	me	mo	$\alpha$
Si	361	6.163	0.385	6.153	6.116	0.12
Al	361	3.282	0.350	3.314	3.476	-0.57
Ti	342	0.067	0.039	0.059	0.063	0.10
Fe <sup>+3</sup>	315	0.247	0.177	0.198	0.125	0.69
Fe <sup>+2</sup>	315	0.133	0.089	0.111	0.102	0.35
Mg	376	0.308	0.200	0.254	0.194	0.57
K	376	1.735	0.224	1.729	1.724	0.05
Na	376	0.180	0.114	0.144	0.122	0.50
Ca	350	0.029	0.044	0.011	0.000	0.66
$\sum R^{VI}$	361	4.002	0.164	4.020	4.047	-0.27
$\sum R^{XII}$	376	1.947	0.196	1.933	1.934	0.07
[IV] <sup>+</sup>	315	30.180	0.416	30.180	30.141	0.09
[VI] <sup>+</sup>	315	11.601	0.511	11.634	11.647	-0.09
[XII] <sup>+</sup>	315	1.985	0.221	1.957	1.939	0.21
[O, OH] <sup>-</sup>	315	43.773	0.548	43.843	43.823	-0.09

the theoretical model, while the skewness is due to the gradual passage towards phengites. A particular characteristic is presented by the distribution of the content in Fe<sup>+3</sup>: this is the only one showing a bimodality, as well as positive skewness. It may be thought that for Fe<sup>+3</sup> there are two overlapping trends, one given by the common isomorphous substitution towards "ferrimuscovites" and the other one linked to the passage to "phengites".

The distribution of interlayer cations all show positive skewness, slightly for K, strongly for Na and Ca. In the light of the above mentioned criteria, these last two distributions seem justified, while a negative skewness would be expected for K.

Further information on distributions can be obtained using probability graphs (Fig. 1). As it is well known, these allow the graphic recognition of the normality of a distribution, through the aligning of the cumulative frequency points.

On arithmetical scale graphs, the distributions considered for the various chemical components are shown in four different ways:

1. a straight line with points more or less scattered (Si);
2. two intersecting straight lines (Al, Fe<sup>+3</sup>, Fe<sup>+2</sup>, Mg, Na);
3. two parallel straight lines jointed by an inflection (K);
4. concave curves (Ca, Ti).

The first case corresponds to a distribution which is not far from normality; the second is very often encountered in the frequency distributions of the content of a major rock component; this has been defined as a "bi-normal" distribution by GREEN (1963) and, in accordance with AHRENS (1964), interpreted as composed of two normal distributions truncated and juxtaposed.

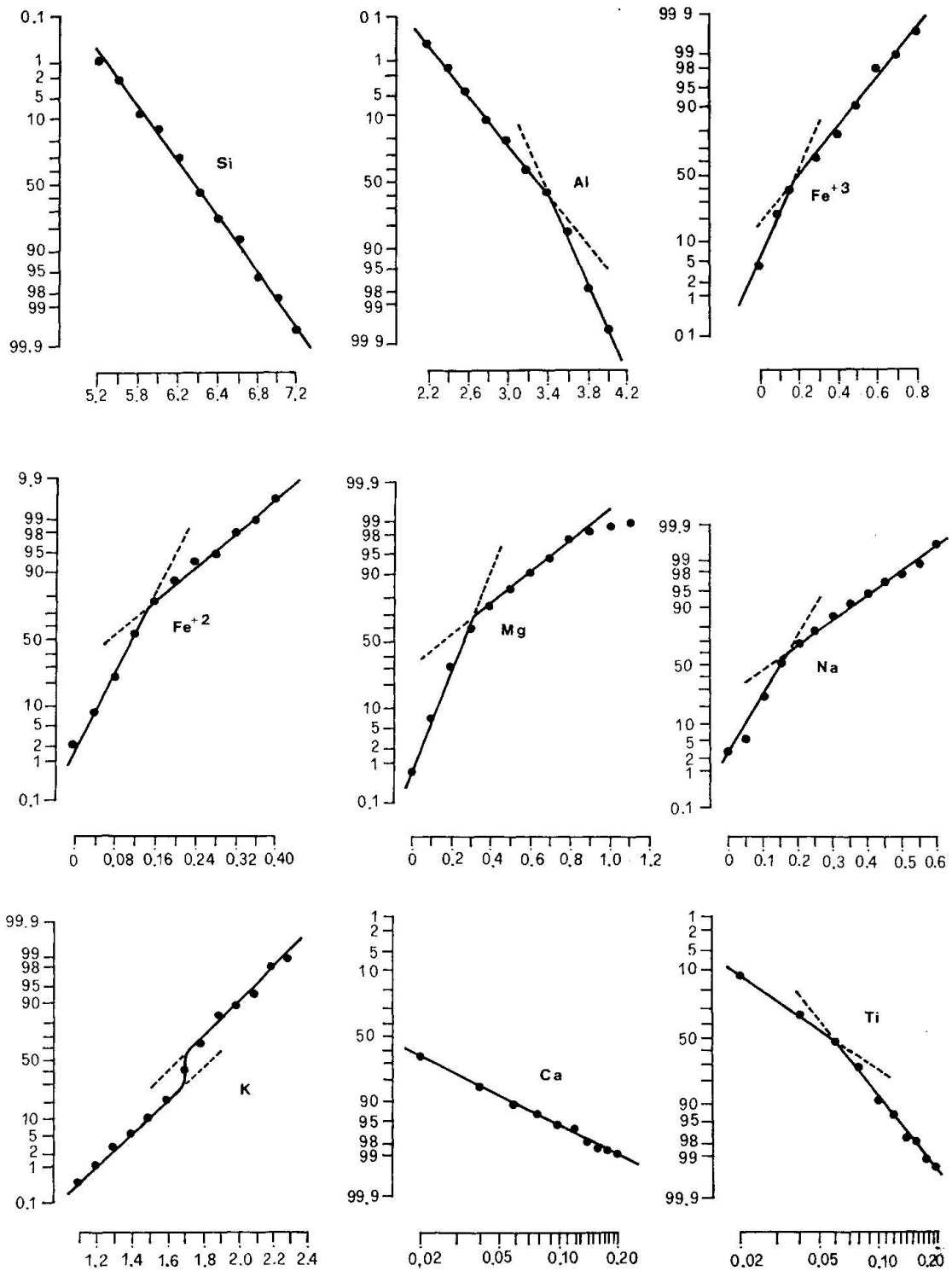


Fig. 1. Distributions of the element contents on probability graphs.

The third case is typical of the sum of two different normal distributions with identical standard deviations. Finally, the fourth corresponds to lognormal (Ca) and bi-lognormal (Ti) distributions, because the dots are on a straight line and on two intersecting straight lines using a logarithm scale.

The different behaviour of the distributions of the major components of white micas (Si, Al, K), must reflect a different behaviour of the phengitic and paragonitic substitutions. The components involved in isomorphous substitutions with regard to the major cation ( $\text{Fe}^{+3}$ ,  $\text{Fe}^{+2}$ , Mg, Na) all show bi-normal distributions, while lognormal distributions are shown by the minor components (Ca, Ti), although with a different tendency which reflects the differences of distribution between K and Al. The trends of the distributions of the major components also influence the distribution trends of the value of the interlayer charges as well as the octahedral and tetrahedral ones (this last is perfectly normal).

Seeing that all the octahedral components present bi-normal distributions, one could try to attribute the meaning of limits of composition between muscovites and phengites to the discontinuity points. These limits have been indicated by CIPRIANI, SASSI and VITERBO-BASSANI (1968) on the basis of an inversion of trend in the variations of the basal spacing in  $\text{RM} = 0,10$  (where RM stands for the sum of the molecular ratios of  $\text{Fe}_2\text{O}_3$ , FeO and MgO); and by GRAESER and NIGGLI (1967) in  $P = 12,5$  (where P is the ratio  $(\text{Mg} + \text{Fe}^{2+}) \times 100/\text{R}^{\text{VI}}$ ). Using the discontinuity points the following "limit" composition of the octahedral layer would be obtained:  $\text{Al}_{3,40} \text{Ti}_{0,06} \text{Fe}_{0,15}^{+3} \text{Fe}_{0,15}^{+2} \text{Mg}_{0,30}$ , not too far from that indicated in above mentioned works, because it implies that  $\text{RM} \cong 0,08$  and  $P \cong 11$ .

On the other hand, as the distribution of Na too is bi-normal with a discontinuity point on a percentage similar to that of the other components, the problem of discontinuity seems to move towards the existence of different populations for lithological rather than for mineralogical types. The average value of the discontinuity percentages, considering the skewness, can be placed at 57%, with a group of high values of the "phengitic" substituents (Mg = 68%;  $\text{Fe}^{+2} = 76\%$ ), one of low values of the normal "muscovitic" components (Al = 44%;  $\text{Fe}^{+3} = 43\%$ ; Ti = 48%) and one of intermediate values of the interlayer components (K = 55%; Na = 62%).

### 3b. Correlations

In Table 2 are shown the "r" values of the correlations for pairs of chemical components. As there are about 350 samples, we can consider the significance limit equal to  $\pm 0,12$  at 95% probability level, and  $\pm 0,16$  at 99% level.

We will now consider briefly the interpretations which can be given to significant correlations.



*Si.* The negative correlation with  $Al^{VI}$  and the positive ones with Mg and  $Fe^{+2}$  agree with the passage to phengite, which can be formulated in the following way:  $Si + Mg(Fe^{+2}) \rightleftharpoons Al^{IV} + Al^{VI}$ . Even with  $Fe^{+3}$  the positive correlation is not very strong and probably this is related to the very strong negative correlation  $Al-Fe^{+3}$ , due to the common isomorphous substitution and to the general increase of total Fe in the phengites. The negative correlations with alkaline components is also interesting: it is higher with K but not negligible even with Na. The first could be related with a parallelism between phengites and paragonites, but the second excludes this interpretation and renders more probable the hypothesis of formation of interlayer vacancies in the phengites, according to a mechanism of the type  $Si + \square^{XII} \rightleftharpoons Al^{IV} + K(Na)$ .

*Al<sup>VI</sup>.* Even the slight positive correlation with Na seems to support this last hypothesis. The very high negative correlation with  $Fe^{+3}$  is the obvious result of a typical isomorphous substitution. The analogous correlations with Mg and  $Fe^{+2}$  indicate also that the passage to phengites takes place through an identical mechanism of substitution, ion by ion, shown moreover by the frequency distributions of the octahedral sites. In other words, the phengites too remain dioctahedral, even if it is reasonable to suppose that the third octahedral site could be occupied by  $Fe^{+2}$ , the biggest ion, having the lowest correlation with Al.

*Ti.* This element presents only a positive correlation with  $Fe^{+3}$  and a negative one with Al, both at the limit of significance and perfectly justified by this element's role as isomorphous substitute of Al. These low  $r$  values and the absence of other significant correlations can be explained bearing in mind that this element is sometimes not determined and that it is however always present in small quantities: analytical and sample errors (i. e. inclusions) and rounding-off in calculations of formulas can be consequently rather important.

*Fe<sup>+3</sup>.* The positive correlations with  $Fe^{+2}$  and Mg indicate that the passage to the phengites involves also an increase in total iron: the substitution  $Al-Fe^{+3}$ , always present, increases in phengites.

*Fe<sup>+2</sup>.* The positive correlation with Mg is explained by the fact that both ions are typical phengitic components, and the low value of  $r$  can be determined by a certain degree of antagonism, because these elements have to enter the same lattice site, even if they are minor components. The negative correlations with the alkali metals, even if only K reaches the limit of statistical significance, seems to render plausible the hypothesis of interlayer vacancies in some phengites.

*Mg.* Same interpretations given for the correlations between  $Fe^{+2}$  and alkalis can be stated for this element too.

*K.* The strong negative correlation with Na is obvious and reflects the typical interlayer isomorphous substitution.

Table 2. Correlation coefficients between the element contents in white micas; underlining distinguishes the significant correlation coefficients.

	Na	K	Mg	Fe <sup>+2</sup>	Fe <sup>+3</sup>	Ti	Al <sup>VI</sup>	Si
Si	<u>-0.182</u>	<u>-0.467</u>	<u>0.512</u>	<u>0.359</u>	<u>0.197</u>	0.036	<u>-0.374</u>	/////
Al <sup>VI</sup>	<u>0.165</u>	-0.078	<u>-0.547</u>	<u>-0.277</u>	<u>-0.678</u>	<u>-0.142</u>	/////	
Ti	0.102	-0.002	<u>-0.076</u>	0.082	<u>0.174</u>	/////		
Fe <sup>+3</sup>	<u>-0.150</u>	0.058	<u>0.287</u>	<u>0.121</u>	/////			
Fe <sup>+2</sup>	<u>-0.065</u>	<u>-0.161</u>	<u>0.233</u>	/////				
Mg	<u>-0.118</u>	<u>-0.043</u>	/////					
K	<u>-0.483</u>	/////						
Na	/////							

Significance limits:  
 $|r| \geq 0.12$  at 95% prob. level  
 $|r| \geq 0.16$  at 99% prob. level

#### 4. COMPOSITIONAL FIELDS OF NON-METAMORPHIC WHITE MICAS

There is a clear compositional difference between metamorphic white micas belonging to medium and high pressure groups and granitic and pegmatitic micas, as can be seen from Figs. 2a and 2b. The slight overlapping of the granitic and pegmatitic micas with those of high-temperature metamorphites is to be emphasized; this is most easily explained by the convergence of the respective environment conditions of crystallization.

From the figures it is evident that both the compositional fields of the granitic micas and the pegmatitic ones are rather wide. The reason for this is to be found in the multiplicity of genetic processes for both types of rocks.

Regarding the granites, it is not surprising that the muscovites that crystallized from differentiated or anatectic melts differ in composition from the white micas of metasomatic granites. It should be remembered, in fact, that the metasomatic granites can be formed at the expense of metamorphites which have crystallized over a wide range of environmental conditions and that in these granites, generally, mica represents an inherited mineral formed through reactions in the solid state. On the other hand, even in those anatectic granites formed in T-P conditions lower than those of the muscovite breakdown, this mica can represent an inherited mineral, the composition of which is to be found only in the fields of the groups 6 or 7.

As regards pegmatites, analogous considerations can be made to justify the range of the compositional field of white micas; naturally the micas of magmatic pegmatites can show a wide compositional variability, owing to frequent occurrence of rare elements, which brings about a variation in the content of the other elements. Moreover, for the veins and pegmatoid bodies of meta-

morphic differentiation, which can be formed in a very extensive field of environmental conditions, it is not surprising that there is a compositional variability of white micas, also because it has now been shown (for example: MELGEREI, 1968; SASSI, 1968) that in such cases the composition of white micas is influenced by the composition of the country rocks as well.

##### 5. FACTORS CONTROLLING THE COMPOSITION OF METAMORPHIC WHITE MICAS

From Figs. 2a and 2b it can also be seen that the compositional field of metamorphic micas is even wider than that of granitic and pegmatitic micas. This depends on the fact that the composition of equilibrium of minerals in metamorphic processes is influenced by several intensive and extensive variables, with wide ranges of variation: a) pressure; b) temperature; c) chemical composition of the rock.

It is of course clear that chemical equilibrium is not always reached, above all because a favourable conjuncture between the kinetics of the metamorphic reactions and the time available does not commonly take place (that is, between the rates of mobilisation, migration and nucleation, and the lapse of time in which the intensive variables remain at a set value). Petrographic experience shows that situations of disequilibrium are far from rare, so that in a single rock micas of successive paragenetic assemblages can occur together, owing either to polymetamorphism or to the polyphase character of a single metamorphic event.

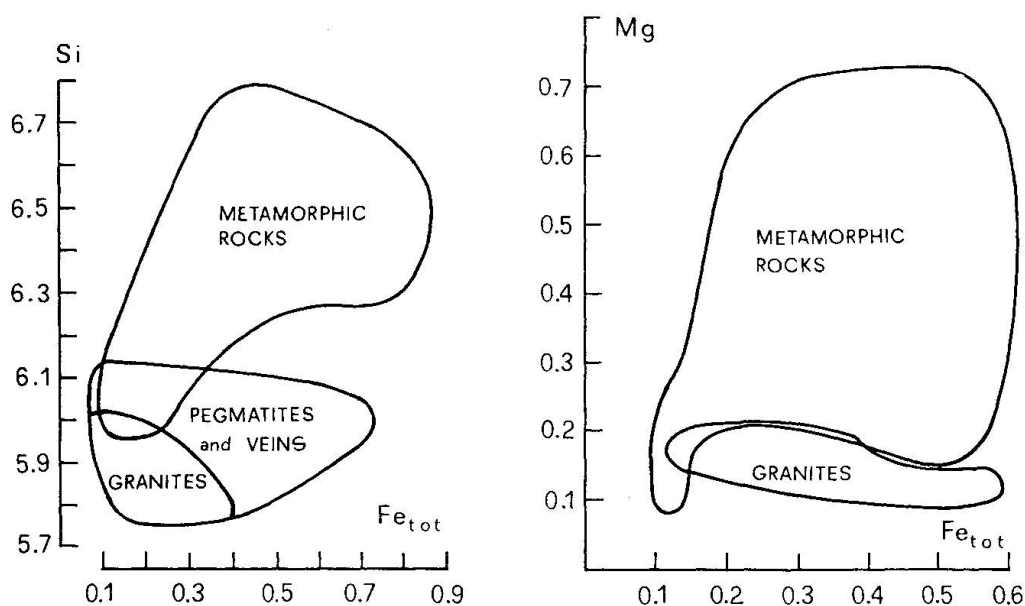


Fig. 2. Compositional fields of non-metamorphic white micas.

We now propose to identify the effects of each single intensive variable, and then the effects of the extensive variables, the latter being of secondary importance with respect to the former, as will be seen further on.

### 5a. Effects of pressures

The influence of pressure on the composition of white micas is illustrated by the fact that in the highest pressure metamorphites, i. e. in glaucophanitic greenschists, phengites are characteristically present (ERNST, 1963, 1964), and that phengitic micas are also common in the barrovian-type greenschist facies (CIPRIANI, SASSI and VITERBO-BASSANI, 1968).

The diagram in Fig. 3 is particularly suitable for the analysis of such an effect, as it is constructed so that the temperature effect occurs principally along the axis of the abscissa (as will be seen later on); the variations along the ordinates depend therefore mainly on pressure. Having only considered, for medium- to very high-pressure metamorphites, samples belonging to the greenschist facies, one can say that the thermal range, at least for fields HH and H, is the same, and that the only intensive variable responsible for their individuation is pressure: this statement is consistent with the fact that the variation range of the Na content is substantially the same for all three fields. The diagram thus obtained clearly discriminates the three compositional fields: field HH (= group 10) relative to very high-pressure micas, field H (= group 1-2-3) relative to medium- to high-pressure micas, and field L (= group 11) relative to low-pressure micas.

The evidence of these results unfortunately has its limits in the very low number of mica samples from the low-pressure facies series; it cannot be excluded, in fact, that, by increasing the population of this class, the field would become wider. However, seeing that valid explanations exist for the situation shown in Fig. 3, we do not believe that the widening of this field can indicate a substantially different trend<sup>1</sup>).

The effect of pressure described so far not only agrees with available experimental data, but also has a theoretical explanation.

From the experimental research carried out by VELDE (1965 b), it seems clear that the stability field of phengite is narrower than that of muscovite as regards temperature, and also that it narrows rapidly with decreasing pressure (Fig. 4, curve A) and that, the temperature being constant, an increase in pressure is

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<sup>1</sup>) When this work was in proof, we have had a good confirmation of this statement, plottig the chemical data of 5 low-pressures white micas belonging to the Ryoke metamorphic belt (chem. anal. in: A. ONO, J. Geol. Soc. Japan, 75, 521-536, 1969). The field L results only slightly widened towards lower Mg contents, with one exception.

The writers thank Professor A. MIYASHIRO for having kindly informed them on the availability of these chemical analysis.

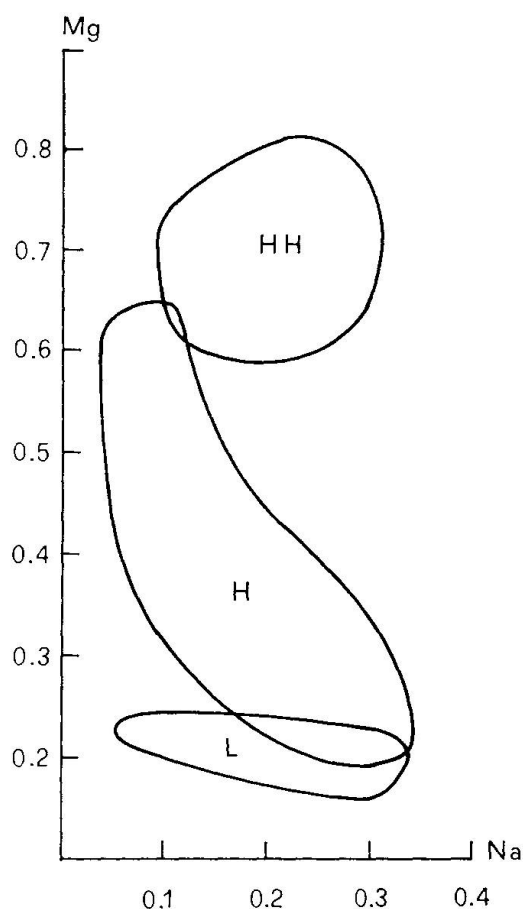


Fig. 3. Compositional fields of metamorphic white micas of different facies series (HH = very high pressures; H = medium-high pressures; L = low pressures).

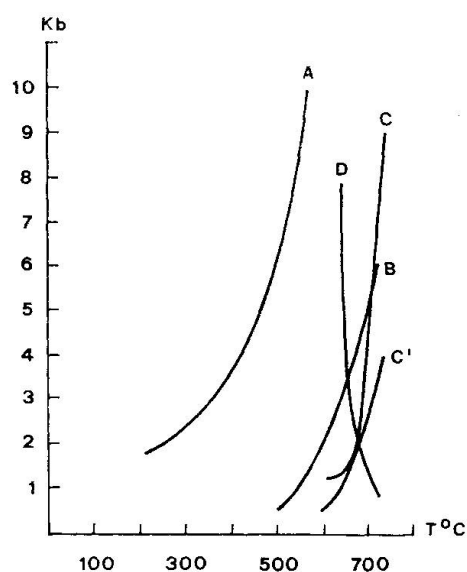


Fig. 4. Experimental data on white micas: A = upper stability curve of phengite (VELDE, 1965 b); B = reaction curve muscovite + quartz (EVANS, 1965); C = upper stability curve of muscovite (VELDE, 1965 b); C' = upper stability curve of muscovite (EVANS, 1965). D is the curve of beginning melting in the granite system (LUTH, JAHNS and TUTTLE, 1964).

connected with an increase of celadonite content in the mica and, in particular (VELDE, 1967 a, 1967 b), with a decrease of  $Al^{IV}$ .

The connection between these results with the behaviour observed by us is evident. In fact, the miscibility limit in the solid state of a given mineral with another of analogous crystal structure (in our case phengite in muscovite) is not independent of the stability limit of the solute mineral: in both cases the characteristics of the single ions (with the contribution that each brings to the lattice energy in the environmental conditions) determine the stability of a given crystal with a given composition. When the intensive variables are such that some ions are no longer stable in a given crystal structure, this will not only determine the disappearance of this mineralogical species, but will also have the effect of gradually making the relative isomorphous substitutions more and more difficult and finally impossible.

It can be foreseen that the decrease of ionic substitutions producing phengitic contents will become significant outside the phengite's field of stability;

in fact, at the limit of this field, there already exists an analogous crystal structure stable in itself, that of muscovite, which can still "admit" magnesium and iron ions even if their entrance does not cause an increase in lattice energy.

To justify this behaviour from another point of view, it should be remembered that phengite has a denser crystal structure than muscovite and that, therefore, its formation and its solid solution in muscovite are favoured by high-pressure conditions. Moreover, one must not forget that phengites have higher  $b_0$  values and lower basal spacings than those of muscovites (CIPRIANI, SASSI and VITERBO-BASSANI, 1968), which brings about a flatter structure in phengites compared to muscovites. One can thus consider that non-hydrostatic pressure plays some role in the appearance of phengitic micas (see also MCNAMARA, 1965).

The effects of  $P_{O_2}$  on the composition of white mica will be approached in the following paragraph, as  $P_{O_2}$  depends on temperature.

#### 5b. Effects of temperature

In various regional investigations, a systematic variation in the composition of white micas with temperature has been detected, even though only in relatively narrow ranges of metamorphic temperatures and often in not very large areas. Over the entire thermal range of the medium-high pressure facies series, CIPRIANI, SASSI and VITERBO-BASSANI (1968) have shown a progressive decrease of iron and magnesium contents related to an increase in temperature, and also a bell-shaped trend of the maximum Na-contents in the interlayer positions.

The influence of temperature on the composition of white micas is also indicated, at least for certain elements, by the experimental research of EUGSTER and YODER (1955a), IYAMA (1964) and VELDE (1965b, 1967a).

The following paragraphs describe the most significant effects of temperature on the composition of white micas and their various paragenetic fields. It is evident that the problem must be considered at constant pressure, or within a narrow range of pressure well defined from the petrological point of view. This, together with the scarcity of data on micas of low-pressure metamorphites, has led us to consider only the group from 1 to 7.

As a first approximation, these can be considered as functions of a single intensive variable (temperature), the values of which increase from group 1 to 7.

In each of these paragenetic groups, the frequency distribution for the contents (in cationic positions) of the various elements of the micas has been analyzed. Moreover, for each element and for each paragenetic group, the arithmetical means ( $\bar{x}$ ) and the relative standard deviations (s) have been calculated, as shown in Table 3.

The representation of the paragenetic fields has been carried out by ellipses,

Table 3. Number of samples ( $n$ ), arithmetic means ( $\bar{x}$ ) and standard deviations ( $s$ ) of the element contents for white micas of different metamorphic grade (for the distinctive numbers of the paragenetic groups, see Introduction).

para- gene- tic group	Si			Al <sup>IV</sup>			Fe <sub>tot</sub>			Mg		
	$\bar{x}$	s	n	$\bar{x}$	s	n	$\bar{x}$	s	n	$\bar{x}$	s	n
1	6.468	0.269	18	3.199	0.335	18	0.447	0.208	18	0.448	0.224	18
2	6.515	0.182	14	2.526	1.033	14	0.625	0.236	16	0.401	0.176	15
3	6.516	0.147	16	3.052	0.195	16	0.423	0.088	17	0.301	0.139	17
4	6.168	0.311	50	3.334	0.255	50	0.322	0.183	53	0.281	0.098	53
5	6.298	0.198	4	3.435	0.067	4	0.340	0.099	4	0.208	0.032	4
6	6.032	0.045	19	3.666	0.087	19	0.132	0.036	19	0.145	0.048	19
7	6.041	0.022	17	3.660	0.062	17	0.132	0.022	17	0.132	0.028	17
para- gene- tic group	Al <sup>VI</sup>			R <sup>+3</sup>			R <sup>+2</sup>			Fe <sup>+3</sup>		
	$\bar{x}$	s	n	$\bar{x}$	s	n	$\bar{x}$	s	n	$\bar{x}$	s	n
1	1.532	0.269	18	3.502	0.291	18	0.666	0.297	18	0.225	0.102	15
2	1.452	0.224	14	3.247	0.719	14	0.804	0.750	13	0.434	0.185	16
3	1.484	0.147	16	3.471	0.104	16	0.451	0.160	16	0.301	0.128	17
4	1.822	0.318	50	3.615	0.180	50	0.425	0.136	50	0.236	0.138	42
5	1.703	0.198	4	3.740	0.086	4	0.313	0.073	4	0.235	0.144	4
6	1.967	0.044	19	3.865	0.062	19	0.154	0.048	19	0.114	0.024	20
7	1.964	0.026	17	3.881	0.026	17	0.134	0.028	17	0.128	0.020	17
para- gene- tic group	$\sum R^{VI}$			Ti			Na			Fe <sup>+2</sup>		
	$\bar{x}$	s	n	$\bar{x}$	s	n	$\bar{x}$	s	n	$\bar{x}$	s	n
1	4.178	0.257	14	0.063	0.026	14	0.223	0.127	17	0.268	0.160	17
2	4.050	0.094	13	0.065	0.036	13	0.088	0.041	16	0.235	0.069	13
3	3.927	0.162	16	0.129	0.066	16	0.246	0.139	17	0.161	0.081	16
4	4.041	0.140	49	0.079	0.032	49	0.239	0.135	47	0.147	0.058	50
5	4.053	0.045	4	0.070	0.024	4	0.333	0.055	4	0.105	0.048	4
6	4.022	0.048	19	0.087	0.032	19	0.232	0.069	19	0.075	0.035	2
7	4.033	0.037	17	0.119	0.024	17	0.134	0.026	17			

the central points of which represent the arithmetical means of the contents shown in abscissa and in ordinate, the two semi-axes corresponding to the value  $1,96s/\sqrt{n}$ : the ellipses thus constructed therefore correspond to the fields within which the true value falls, at 95% probability level.

In Fig. 5 the more interesting paragenetic fields can be seen. They show clearly the influence of temperature on the composition of metamorphic white micas.

The position of field 1 turns out to be anomalous, owing to the low contents of Fe<sup>3+</sup> and total iron, and the high contents of Na and Al<sup>IV</sup>; the location of field 5 also turns out to be slightly anomalous. These anomalies do not dis-

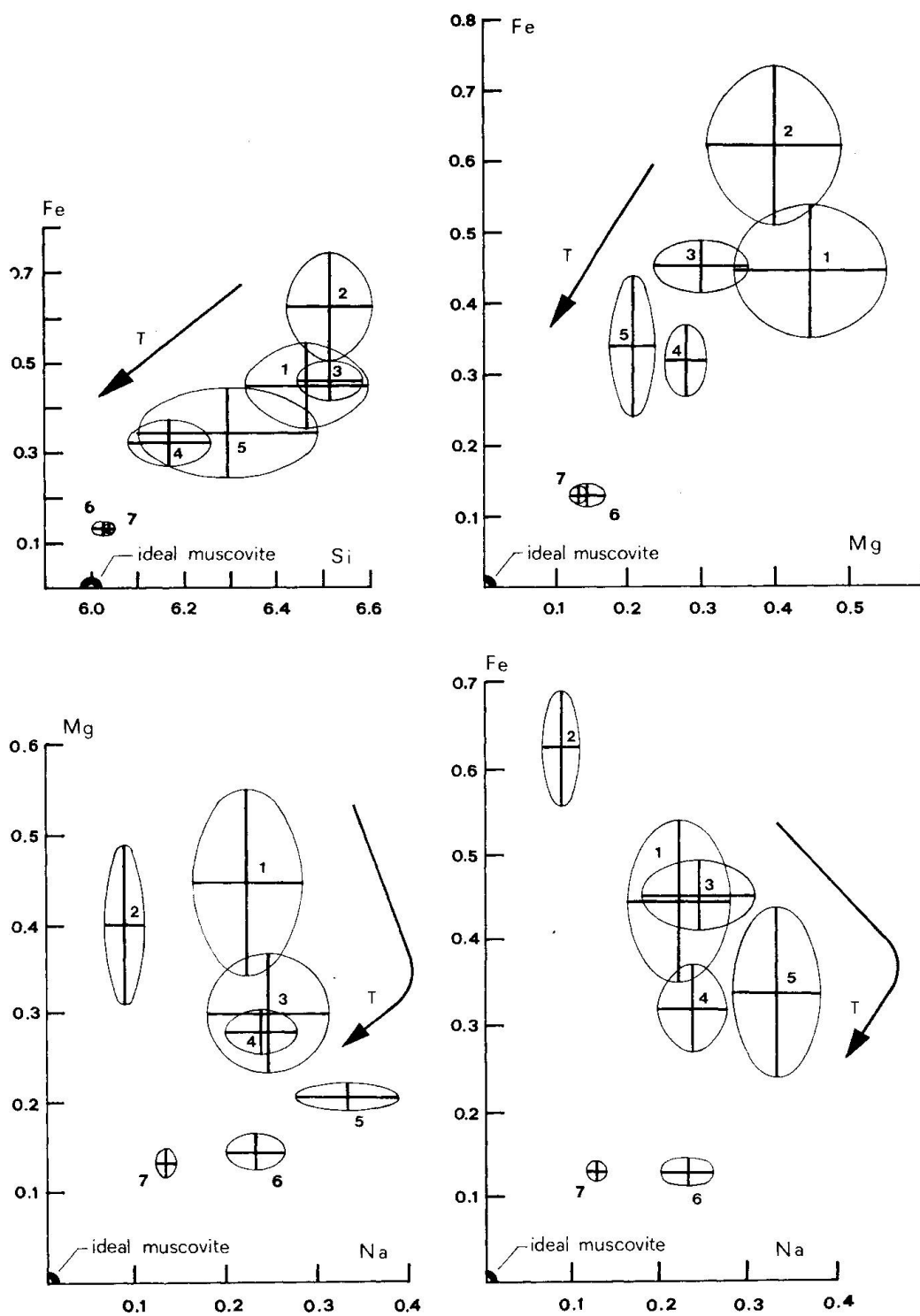


Fig. 5. Paragenetic fields of metamorphic white micas. Distinctive numbers are the same as those specified in the Introduction; arrows indicate increasing temperatures.

The upper diagrams are indicated in the text as Figs. 5a and 5b, the lower as Figs. 5c and 5d.



credit the trends which are evident; they can however be explained through the low population of these groups and/or the probable impurity of the analyzed samples, sometimes indicated by the authors themselves (see for ex. MCNAMARA, 1965).

In Fig. 5a not only can the positive correlation between Si and  $Fe_{tot}$  be noted (already pointed out using the global statistic analysis: see Table 2), but there is also an evident negative correlation with the temperature of the contents in both abscissa and ordinate: increasing the metamorphic grade the substitution of Si by  $Al^{IV}$  in the tetrahedral layer increases and, at the same time, Fe decreases in the octahedral layer. In this way, by increasing the metamorphic temperatures, the mica becomes purified, tending towards the composition of theoretical muscovite. Fig. 5b indicates that magnesium too decreases rapidly with temperature, and therefore by increasing the metamorphic grade the phengite content tends to disappear.

It should be emphasized that in Figs. 5a and 5b the fields 6 and 7 are not well discriminated, which means that the occurrence of Mg and Fe in octahedral sites is already scarce before arriving at the orthoclase isograd, that is, before the breakdown of muscovite takes place. These two fields on the other hand are well separated in Figs. 5c and 5d, where the interlayer cations are also taken into consideration. From these two diagrams it is evident that, when the metamorphic grade increases with a decrease of the phengite content, the paragonite content increases up to field 5, and then falls down rapidly within a very narrow thermal range (a few tens of degrees Centigrade). Particular emphasis should be laid on the rapid disappearance of the paragonite contents from group 6 to group 7.

A similar bell-shaped trend of the paragonite content in muscovite has already been shown by CIPRIANI, SASSI and VITERBO-BASSANI (1968, fig. 5); however, owing to a scarcity of samples relative to the kyanite-almandine-muscovite subfacies, the maximum was drawn erroneously at a lower temperature, in the staurolite-almandine subfacies.

Therefore, we can state that by increasing the metamorphic grade, the micas undergo total purification before reaching breakdown, and this happens at the tetrahedral, the octahedral and the interlayer levels. From a practical point of view, the diagram in Fig. 5b appears to be the most discriminating for the low and medium temperatures, while those of Figs. 5c and 5d, particularly indicative for extreme temperatures, are efficient over all the metamorphic thermal range.

In Fig. 6 the concomitant variations of phengite and paragonite contents with the metamorphic grade are shown by a triangular diagram: the upper apex shows the diadochy Na-K, and the basal apices the isomorphous substitutions in the octahedral sites. Speaking in terms of low, medium and high metamorphic grades, this diagram appears to be satisfactorily selective.

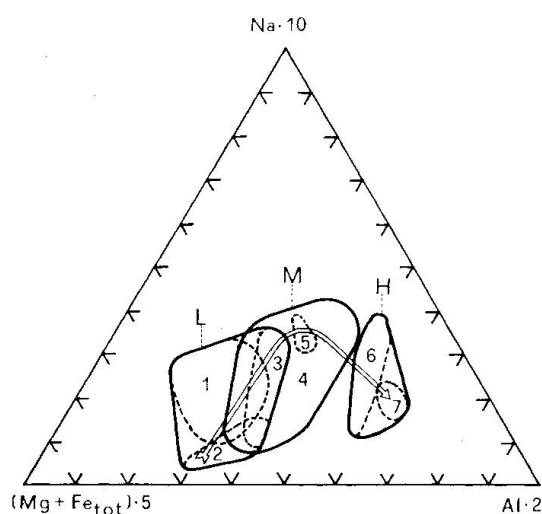


Fig. 6. Compositional fields of low-grade (L), medium-grade (M) and high-grade (H) metamorphic white micas. The sub-fields are relative to the paragenetic groups indicated in the Introduction. Arrow shows increasing temperatures.

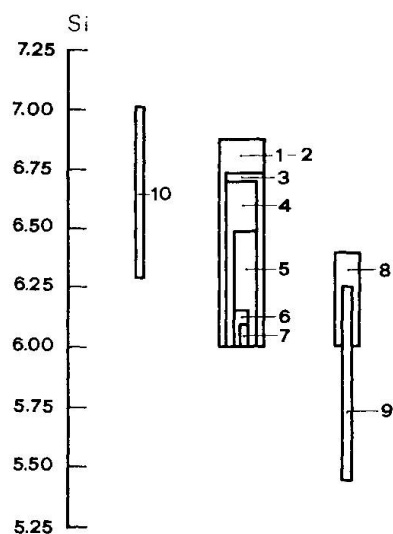


Fig. 7. Si content of the white micas of different paragenetic groups.

It seems to be opportune to add further considerations on the systematic compositional variations observed, giving, where possible, some petrological interpretations to link up with the available experimental data.

a) *Tetrahedral sites.* It is interesting to emphasize the progressive decrease of the ratio  $\text{Si} : \text{Al}^{\text{IV}}$  on increasing temperature, as shown in Fig. 7.

The increase of Si substitution by  $\text{Al}^{\text{IV}}$  under the influence of increasing temperature (which for phengites is clearly indicated by the experimental data of VELDE (1967a)<sup>2</sup>), has already been demonstrated by THOMPSON (1947) and HARRY (1950) for some minerals. The tendency to assume a lower coordination number with the increase of temperature is well known in polymorphous systems (see for example, the polymorphs of  $\text{Al}_2\text{SiO}_5$ ). The increase of  $\text{Al}^{\text{IV}}$  in function of T is by now a general rule for the silicates from metamorphic rocks, and has been verified for the plagioclases (WENK, 1962), the amphiboles (KOSTYUK and SOBOLEV, 1969), the biotites (OKI, 1961; BUTLER, 1967) and the pyroxenes (DOBRETSOV, 1968). Chlorites too, at least when associated with phengite, seem to exhibit similar behaviour (VELDE, 1967b).

b) *Octahedral sites.* One wonders by what mechanism the balances of charges are established with increasing metamorphic grade.

<sup>2</sup>) It should be noted that BROWN (1968) limited the validity of this author's interpretation to the  $\text{Si}^{4+}$  content of the phengites, showing – for the composition of the micas studied by VELDE – the existence of a control by the chemical composition of the rock. However, we believe that we should not deal with these considerations, as VELDE's results are not indispensable for the validity of our conclusions, and BROWN's objections do not affect it.

Using available data, and bearing in mind that the quantities of Ca in the interlayer positions are usually very small and anyway not related to the metamorphic grade, the balance must substantially lie in the octahedral layer, through a concomitant and progressive substitution of bivalent ions by trivalent ions.

The hypothesis formulated above finds wide confirmation in Fig. 8. In fact, in this diagram it is clear that not only  $Al^{IV}$  is positively and significantly correlated to  $R^{+3}$  ( $r=0,439$ ) and negatively with  $R^{+2}$  ( $r=-0,509$ ), but also that such correlations are strictly dependent on temperature, in the sense that the increase of the metamorphic grade favours the entrance of trivalent ions into octahedral sites and hinders that of the bivalent ions.

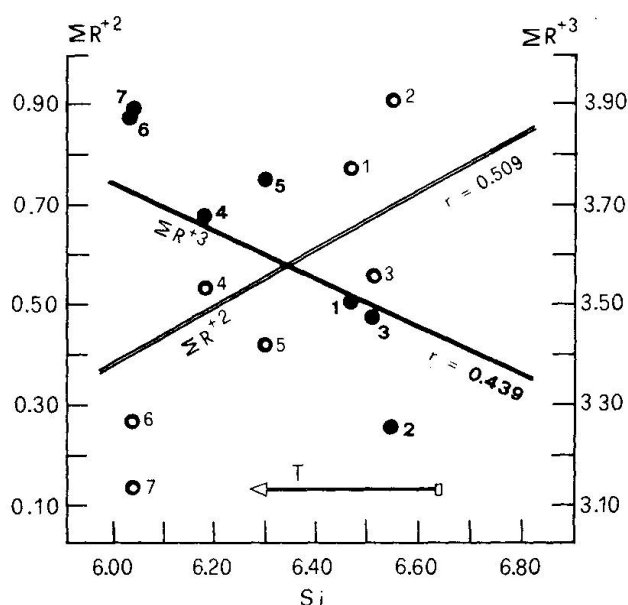


Fig. 8. Relationships between temperature and octahedral  $R^{+3}$  and  $R^{+2}$  contents; the correlation coefficients and the regression lines have been calculated in function of the Si contents, which are dependent from temperature.

On the other hand, in the white micas an increase of titanium with the metamorphic grade was also noted by us (see also KWAK, 1968). This behaviour too must be a general rule, because it is known for almost all the aluminosilicates of metamorphic rocks containing titanium: for the amphiboles (LEAKE, 1965; KOSTYUK and SOBOLEV, 1969), less evidently for the pyroxenes (DOBRETsov, 1968) and, above all, for the biotites (OKI, 1961; KWAK, 1968). For the latter, however, a progressive introduction of Ti into the tetrahedral sites with increasing temperature has also been postulated (SAXENA, 1966).

What we have stated so far also implies a positive correlation between  $Al^{IV}$  and  $Al^{VI}$  and this fact (that is, an increase of Al in the octahedral layers owing to increasing temperature) also explains the gradual disappearance of the phengites with the increase in metamorphic grade.

It is well known that  $P_{O_2}$  in the intergranular fluid increases with temperature, owing to the increasing thermal dissociation of water. It has also been recognized that the resulting oxygen reacts with the carbon present in the pelitic metamorphites to produce  $CO_2$ ; it has moreover been ascertained that the degree of oxidation of rocks generally decreases with the increase in metamorphic grade (SHAW, 1956; MIYASHIRO, 1964). Taking in account these statements one can check if the degree of oxidation of white micas too varies with temperature.

In Table 4 the average values of  $Fe^{+3}/(Fe^{+2} + Fe^{+3})$  and the relative standard deviations are shown, while in Fig. 9 the average values are plotted against Si, i. e. against temperature.

Table 4. Number of samples ( $n$ ), arithmetic means ( $\bar{x}$ ) and standard deviations ( $s$ ) of oxidation ratio  $Fe^{+3}/(Fe^{+2} + Fe^{+3})$  for the different paragenetic groups.

paragenetic group	$\bar{x}$	s	n
1	0.477	0.182	14
2	0.643	0.135	13
3	0.628	0.200	16
4	0.591	0.161	39
5	0.637	0.246	4
6	0.523	0.032	2

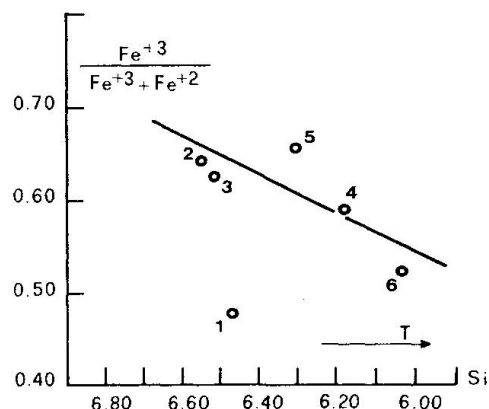


Fig. 9. Relationships between oxidation ratio of white micas and temperature, estimated using the correlation between  $Fe^{+3}/(Fe^{+3} + Fe^{+2})$  and Si contents.

In Fig. 9 clearly appears a negative correlation between T and the degree of oxidation; only the position of the first group is well outside the regression line and this could indicate that, during the first stages of metamorphism, the oxygen (present in relatively low concentrations because of the temperatures) is entirely consumed in reactions with the abundant organic material and is therefore not sufficient to oxidate the Fe of white mica (see also Fig. 11). On the other hand, one can also suppose that at low temperatures,  $Fe^{+3}$  prefers to enter the structure of feric minerals rather than micas.

The negative correlations represented in Fig. 9 also agrees with the results of CHINNER (1960), according to whom an increase in the degree of oxidation of the rock brings about an increase of  $\text{Fe}^{3+}$  and a decrease of  $\text{Fe}^{2+}$  in white mica. It is worthwhile remembering that also during the passage from shales to slates a partial reduction of iron has been noted (NANZ, 1953).

c) *Interlayer sites*. Regarding these structural sites, three phenomena should be considered: the  $\text{Na}^+ - \text{K}^+$  and  $\text{Ca}^{2+} - \text{K}^+$  substitutions, and the occurrence of vacancies.

The thermal control of the entrance of sodium has already been shown; and the bell-shaped trend has been related by CIPRIANI, SASSI and VITERBO-BASSANI (1968) to the lower decomposition temperature of paragonite with respect to that of muscovite. As a support for this interpretation, the same above-mentioned considerations (page 270) on the relationship between phengite's stability field and the limit of solid solution of phengite in muscovite can be used here.

A similar bell-shaped curve can also be seen in the phase diagram of EUGSTER and YODER (1955a), from which it can be deduced that the maximum content of Na in muscovites first increases with temperature (in coexistence with paragonite) and then, in a short thermal interval, rapidly decreases until it disappears (in coexistence with feldspars).

Moreover, such behaviour has also more recently been detected experimentally: at 1000 bars the solid solution limit of paragonite in muscovite varies in the following way:

500°	15%
600°	25%
650°	18%

These results, obtained by IYAMA (1964), match perfectly the behaviour we found: the decrease of solid solution takes place in a narrow thermal range (some tens of degrees) and the temperature of 600° found experimentally for the maximum solid solution coincides very well with the thermal range of the kyanite-almandine-muscovite subfacies (see further on, Fig. 12).

Concerning the substitution  $\text{Ca}^{2+} - \text{K}^+$ , the numerous data we considered allow us to state that no correlation exists between the very low contents of Ca and the metamorphic grade, and that, therefore, the entrance of this element into the interlayer sites is due to the chemical composition of the rocks and, mainly, to the presence of Ca in the intergranular fluid.

That calcium easily substitutes for potassium in such conditions, is understandable taking into account the characteristics of these two ions: it is possible then to consider the entrance of  $\text{Ca}^{2+}$  in the place of  $\text{K}^+$  as a capture process.

Finally, the possibility of interlayer vacancies is suggested, as we have

already seen, by the negative correlations between Si- and alkali-contents. The dependence on silicon would seem to support the hypothesis of a special mechanism of "phengitization"; however the statistical analysis by classes has not shown any systematic variation in the sum of interlayer ions.

### 5c. Effects of extensive variables

Up to now we have interpreted the compositional variations of white micas in function of temperature and pressure, although we did specify that other factors control the composition of this mineral in metamorphic rocks: chemical composition of the rocks and the presence of some other mineralogical species which utilize part of the chemical elements making up white mica.

It is worthwhile to consider the control exercised by the chemical composition of rocks. Several authors have stated this to be negligible (for example, SCHWANDER, HUNZIKER and STERN, 1968), while others have shown it to be significant (for example, BROWN, 1968).

Unfortunately, in the abundant bibliography which we consulted, the chemical analyses of the rocks from which micas have been separated are too few, so that it was not possible to resolve definitely this problem. However, we have ascertained that some control exercised by the bulk composition exists, even if ultimately it turns out to be of small importance with respect to that exercised by the intensive variables.

In order to study the influence of the chemical composition of the rocks, the effects of the intensive variables have been reduced to a minimum, taking into consideration only samples belonging to a single subfacies. The choice of group 4 was dictated by the fact that this group alone comprises a reasonable number of rock chemical analyses.

In these samples, a positive correlation between  $(\text{SiO}_2)_{\text{rock}}$  and  $(\text{SiO}_2)_{\text{mica}}$ , and between  $(\text{K}_2\text{O})_{\text{rock}}$  and  $(\text{K}_2\text{O})_{\text{mica}}$  has been noted, as was predictable. Moreover, it has been shown (Fig. 10a) that in Al- and Si-rich rocks, there occur white micas richer in ferric elements than the T-P conditions would permit. This is easily explained by the fact that in these rocks the only mineral which can accept these elements in its crystal structure is white mica (apart from possible accessories).

Another interesting characteristic is the correlation between  $(\text{Fe}_2\text{O}_3_{\text{tot}} + \text{MgO})_{\text{rock}}$  and  $(\text{Fe}_2\text{O}_3_{\text{tot}} + \text{MgO})_{\text{mica}}$  (Fig. 10b), the negative character of which is explained by the ever-increasing appearance of Mg- and Fe-rich minerals.

In all these correlations there are two regression lines, which only apparently are due to the regional character of the sampling; in reality this duplicity must depend on different values of the intensive variables in the two regions, as every subfacies has a range of P and T, and not simply single values for each variable.

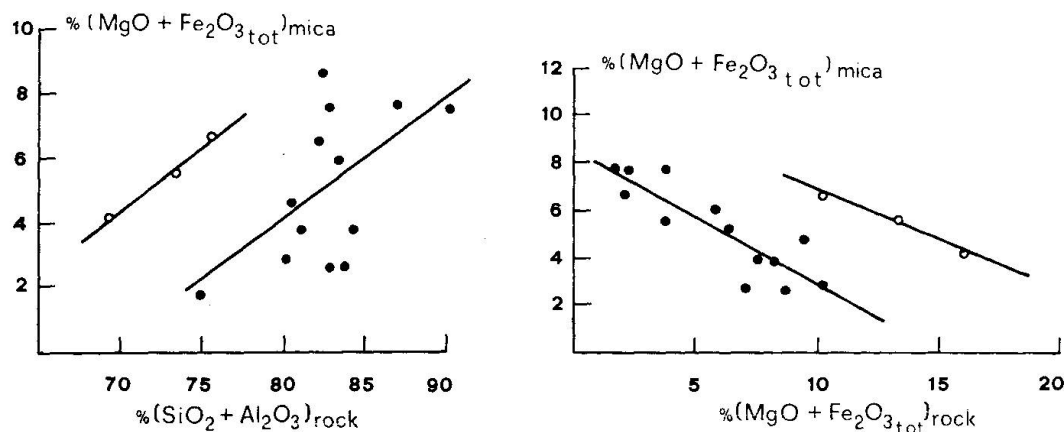


Fig. 10. Relationships between composition of white micas and that of the rocks: there are different regression line for different regions.

The validity of these considerations is unfortunately limited by the small number of rock analyses available.

For the coexistence of white mica with minerals which compete with it for the use of some chemical species, it must be remembered that this factor, genetically bound to T-P conditions, also depends on the chemical composition of the rock; we therefore wonder if the control of the latter on the composition of white mica is greater than that exercised by the intensive variables.

The results shown in the two last chapters, obtained by leaving out the composition of the rocks, indicate clearly that, for metamorphic white micas, the T-P controls (and above all that exercised by the temperature) are undoubtedly predominant, as has been shown by the existence of clearly distinct paragenetic fields.

This, however, does not allow us to neglect completely the extensive variables mentioned above, not only as regards the results of Fig. 10, but also for the following reason: if T and P were the only controls of the composition of white mica, we should have detected a systematic and continual variation of the distribution coefficients of single elements between mica and minerals which coexist with it over a large range of environmental conditions (for example: the distribution of Fe and Mg between white mica and chlorite in the greenschist facies and between white mica and biotite or garnet over almost all the metamorphic field; the distribution of Na between white mica and plagioclase, etc.). In fact we have

$$\left(\frac{\partial \ln K_D^{\text{el}}}{\partial T}\right)_P = \frac{\Delta H^\circ}{RT^2}; \quad \left(\frac{\partial \ln K_D^{\text{el}}}{\partial P}\right)_T = \frac{\Delta V^\circ}{RT}.$$

$\Delta H^\circ$  and  $\Delta V^\circ$  being the enthalpy and molar volume variations, R the gas constant, T the absolute temperature and  $K_D^{\text{el}} = \left(\frac{X_A}{1-X_A}\right)\left(\frac{1-X_B}{X_B}\right)$  the distribution

coefficient of the "el" element between two coexisting minerals A and B, in which "el" occurs in the molecular fractions  $X_A$  and  $X_B$  (KRETZ, 1961; ERNST, 1964).

These variations, however, did not find confirmation in the data we collected and analysed from this point of view; in fact, the  $K_D$  which we have calculated for about 70 mineral pairs did not show systematic variations with the temperature. The reason for this must lie in the fact that the  $K_D$  are controlled, at least to a certain degree, also by the concentration of the various elements in the chemical system examined, that is, by the bulk composition of the rock; in particular, they undergo sudden variations every time the crystallization environment determines the appearance of a new mineralogical phase which uses to a great extent the element under consideration (SAXENA, 1966, 1968; VELDE, 1967b). Moreover, all discussion on the  $K_D$  implies the attainment of chemical equilibrium (but this does not always occur), and presupposes that the two minerals which make up the pair are really coexistent minerals, that is, produced at the same time in the same situation of environmental conditions. Unfortunately, even recent literature is very scant from this point of view, not only because of the already criticised lack of chemical analyses of rocks, but also because the existence of the fundamental assumption is very rarely proved: that is, the effective syngeneses of the two minerals for which the  $K_D$  are studied (demonstrable only through a detailed microstructural analysis).

We believe, therefore, that the lack of systematic variations which we noted in the  $K_D$  is derived also from the frequent absence of the above-mentioned assumptions. In fact many rocks consist not of a single paragenesis, but of the overlapping of mineral phases (or assemblages) which crystallized successively. It can easily be admitted that each of these steps is characterized by different environmental conditions, not only in the case of polymetamorphism, but also in the case of a single polyphase metamorphic event.

This statement, generally valid, agrees with the results of some regional works in which the lack of equilibrium is shown by the occurrence of white micas of different composition (EMILIANI, 1957; CRAWFORD, 1966; BROWN, 1967; SASSI and ZIRPOLI, 1968; LIBORIO and MOTTANA, 1970). These occurrences are not at present very numerous, but their rarity is, we believe, apparent, and due to the insufficient attention given so far to this problem. It is also evident that in such conditions, the chemical analysis of a concentrate of white mica cannot be used for the calculations of the  $K_D$ ; nevertheless, it may be useful for identifying the paragenetic fields when, of two generations of mica, one is quantitatively prevalent over the other and substantially contributes to the analytical results, and when the scattering of the obtained values is minimized by statistical analysis.

The bulk of the observations made up to now brings us to consider the



area of the compositional fields of white micas as amplified by the facts considered in this chapter, starting from the very restricted fields determined by the T-P conditions. It can be admitted that the localization of the single paragenetic field also reflects the greater frequency of some lithotypes, that is, of some chemical compositions of rocks. From this, we can derive that a systematic variation of the  $K_D$  should be observable only if these are calculated, in a single metamorphic region, from chemical data which are directly comparable among themselves, as obtained for other pairs of minerals (see for example, HIETANEN, 1969). Otherwise, the different quality of the chemical analyses and the fact that each subfacies is related to an interval of intensive variables (so that the T-P conditions of a given subfacies found in any region can differ from those of the same subfacies in another region), render very hazardous generalizations obtainable from very sensitive parameters, like the  $K_D$ <sup>3</sup>).

In conclusion, having been able to identify well-defined paragenetic fields for white micas in function of the intensive variables, neglecting the extensive ones, we believe it clearly proved that the control exercised by temperature and pressure on the composition of these minerals is prevalent, while the influence exercised by other factors, although not insignificant, is of secondary importance.

## 6. PETROLOGICAL IMPLICATIONS

### 6a. Mechanism of compositional variations of white micas in progressive metamorphism

We have seen up to now that, depending on the intensive variables, the phengite and paragonite contents in solid solution in the white mica vary continuously. Let us now discuss the mechanism of these compositional variations.

Owing to the dependence of the chemical composition of white micas on the T-P conditions, it is evident that, as these intensive variables are continuous, the compositional variations too will have a continuous character. The fact that this character does not appear in our diagram depends only on the necessity of representing in a single group (subfacies) the points that should in reality be referred to a thermal interval, and of presenting the pressure

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<sup>3</sup>) At the conclusion of our work, we obtained confirmation of the above statement in a very recent and interesting paper by E. WENK (Contr. Mineral. Petrogr., 36, 1970, 50-61). This author, studying the distribution of Al between muscovites and biotites coexisting over a wide range of temperature in a very large region of the Central Alps, shows a systematic variation of  $K_D = Al_{Mu}^{VI}/Al_{Bi}^{VI}$ . The values range from 3 to 14 with increase of the metamorphic grade.

variations in a small number of facies series, thus artificially introducing into our diagrams a discontinuous character which is not real but only useful.

It follows that the compositional modifications of white micas, owing to their continuous character, cannot be explained as a sequence of reactions having a discontinuous character. The process should instead be related to ion exchange between the crystal structure of the white mica and the intergranular fluid; this mechanism allows a continuous adaptation of the mineral composition in respect to the ionic concentration of the intergranular phase, as response to the chemical potential of the various elements involved in the given environmental conditions and on the basis of the variations of lattice energy induced by the environmental conditions themselves.

#### 6b. Variations of the phengite content in the T-P diagram

We will now try to describe the trends of compositional variations of white micas in the T-P diagram of the metamorphic facies as proposed by WINKLER (1967, page 177).

In this respect, as regards phengite, the data provided by VELDE (1965b) assures us that: a) at  $T > 650^\circ$  approximately, phengite is no longer stable at any pressure; b) in the range of the amphibolite facies ( $T \geq 540^\circ$  approximately), phengite is only stable at pressures higher than about 8 Kb; c) in the field of the greenschist facies, that is, at temperatures between about  $400^\circ$  and  $540^\circ$ , the stability field of phengite is greatly narrowed with decreasing pressure, and that the appearance of this mineral requires a pressure not less than about 4 Kb in the subfacies of higher temperatures in the greenschist facies.

It is clear that in order to establish the real stability field of phengite, we cannot use equal T-P values obtained experimentally for its breakdown, as in common rocks the chemical system is more complex: phengite is associated with other minerals and therefore reactions between the respective chemical components are to be expected. Thus, the range of T-P conditions within which phengite can survive in natural rocks is really smaller than that limited by the experimental breakdown curve, as happens for muscovite (Fig. 4, compare curves B and C).

On the other hand, we have already shown that, *mutatis mutandis*, this behaviour must also be reflected in the values of solid solution of phengite in muscovite.

All these considerations allowed us to insert, for purely indicative purposes, the stability field of natural phengites from metamorphic quartz-rich pelites in the above-mentioned T-P diagram of WINKLER (Fig. 11). This field, although outlined by a indented line owing to the lack of specific experimental data, is consistent both with the results of experimental petrology and with the real

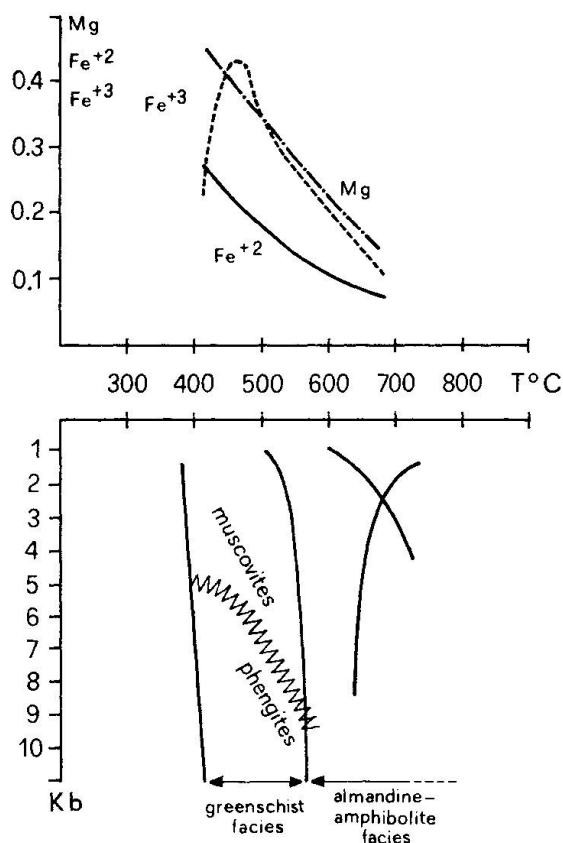


Fig. 11. Stability field of phengites in the T-P diagram, and variation trend of femic elements with temperature. The trends have been drawn on the basis of average values for each paragenetic group.

diffusion of natural phengites. In the same Fig. 11 we have inserted (for the medium-high pressure facies series) the variations of Fe and Mg with the temperatures resulting from the considered data. The antithetical trends at low temperatures of the Fe<sup>+3</sup>- and Mg- or Fe<sup>+2</sup>-variations should be noted: if it does not depend on the scantiness and impurities of samples of group 1, this behaviour may be explained by the entrance of Fe<sup>3+</sup> into the ore minerals and chlorite. It is in fact well known that this last mineral becomes more and more Mg-rich with increasing temperatures, the degree of oxidation remaining constant (ALBEE, 1962).

#### 6c. Variations of the paragonite content in the T-P diagram

Concerning the variation in solid solution of paragonite in muscovite, the petrological interpretation of the observed behaviour must take into consideration the following facts:

1. In the ambit of facies series of medium-high pressures, zones characterized by the K feldspar-sillimanite assemblage completely lacking in evident effects of incipient anatexis are not rare. Such occurrences cannot be explained by the reaction curve  $Mu + Q \rightarrow Kf + Al_2SiO_5$ , the T-P values of which established experimentally by EVANS (1965) are too high. Nor can they be explained by a lack of H<sub>2</sub>O, as the kinetics of the reactions and the mobility of the chemical

species imply the presence of this fluid. The appearance of anhydrous mineral assemblages (Kf + alm + sill) also requires the liberation, and therefore the availability, of conspicuous quantities of water.

2. In the ambit of medium-high pressures facies series, zones characterized by the stability of muscovite-sillimanite-orthoclase and muscovite-sillimanite assemblages are known (see bibliography quoted by: TURNER and VERHOOGEN, 1960, page 548; EVANS and GUIDOTTI, 1966, page 60; TURNER, 1968, pages 311–326; see also WINKLER, 1967, page 128). These findings led the first two above-mentioned authors to define the “sillimanite-almandine-muscovite” subfacies. This subfacies is not reported by WINKLER (1967) but it should deserve consideration, although adopting suitable precautions (FYFE and TURNER, 1966).

In WINKLER'S T-P diagram, for low-pressure ( $< 2,5$  Kb) and high-pressure ( $> 8$  Kb) values, it is difficult to understand the stability and the frequency of the above-mentioned mineral assemblages; in fact at  $P < 2,5$  Kb sillimanite occurs only together with Kf through muscovite breakdown, while at  $P > 8$  Kb the field of sillimanite is practically overlapped by that of incipient melting, and the reaction curve  $Mu + Q$  is not reached. On the other hand, at intermediate pressures, the appearance of sillimanite coexisting with muscovite is considered (“first sillimanite zone”), but is not connected with the appearance of neogenic Kf, whose first metamorphic crystallization would occur in the “second sillimanite zone” which, however, is overlapped by the field of incipient anatexis. The field of K feldspar + sillimanite, as outlined by the  $Mu + Q$  reaction curve, for  $P > 3$  Kb is practically non-existent, in the sense that these minerals should appear only when anatexis is not possible owing to the lack of water, or in the unmelted parts when there is evidence of incipient anatexis.

The considerations made in points 1. and 2. lead us to separate the first appearance of Kf + sillimanite from the reaction curve  $Mu + Q$ , and to put it at lower temperatures than those of incipient anatexis, even at relatively high pressures. Such a conclusion reaches greater importance if we consider the experimental data of EUGSTER and YODER (1955a, 1955b).

In fact, from the phase diagram of the muscovite-paragonite system prepared by these authors and from the results which they obtained in the albite-K feldspar-corundum-water system, it can be deduced that at 30.000 psi ( $\cong 2$  Kb) along the curve  $715^{\circ}$ – $660^{\circ}$  (dependent on composition) mica breakdown occurs, so that with the increase of T, the field of K feldspar + corundum is entered (a field which, in excess of  $SiO_2$ , would be of K feldspar +  $Al_2SiO_5$ ). Thus, it is this curve that, apart from its thermal values, corresponds to the one which petrologists have adopted as the “orthoclase isograd”, to mark the beginning of the sillimanite-cordierite-orthoclase-almandine subfacies (Abukuma-type metamorphism), and sillimanite-almandine-orthoclase subfacies (barrovian-type metamorphism).

Limiting the discussion to the muscovite side of EUGSTER and YODER's diagram, the quoted data indicate that, at thermal values lower than those of the curve  $715^{\circ}$ – $660^{\circ}$ , there exists a field in which mica-K feldspar-corundum coexist, and in which therefore, in excess of quartz, K feldspar- $\text{Al}_2\text{SiO}_5$  would exist, without the appearance of the latter two minerals implying the breakdown of muscovite. It is interesting to note that, according to EUGSTER and YODER, in this field the composition of muscovite, which at  $670^{\circ}$  has its maximum Na-content, gradually becomes more K-rich with increasing temperature, thus exhibiting a behaviour which is perfectly identical with the one we observed in the passage from groups 5 to 6 and 7. It is thus in this thermal range that the mineral assemblages (or, if preferred, the subspecies) of group 6 and partly of group 5, are to be placed.

These observations have been put into diagrammatic form in Fig. 12, obtained (for medium-high pressure facies series) from our data on the Na-content of micas, from the considerations made above and adopting for the subspecies reasonably acceptable temperature values on the basis of present

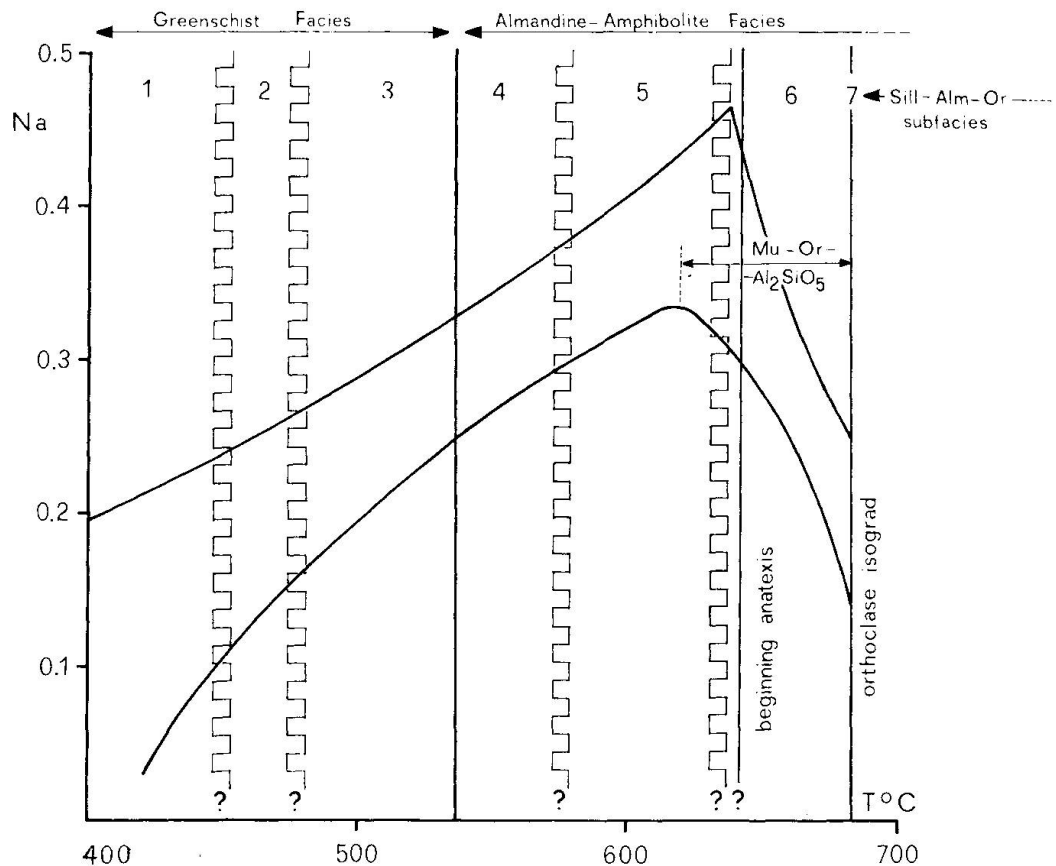


Fig. 12. Variation trend of Na content with temperature for groups 1-7, drawn on the basis of average values for each group (lower bell-shaped curve). The upper curve is the left part of the phase diagram Mu-Pg by EUGSTER and YODER (1955a).

knowledge. The obtained trend, well consistent with the phase diagram of EUGSTER and YODER (although with slight differences for T values) agrees with petrographic evidence and fits in perfectly with the results of modern experimental petrology.

3. Petrographic experience indicates that at extreme metamorphic grades and at temperatures lower than those of incipient anatexis, it is not rare to find an increase in the quantity of plagioclase (see for example, DESIO and ZANETTIN, 1970), which could partly be connected with the decreased solid solution of paragonite in muscovite.

Reactions of this type have been proposed (for example, by EVANS and GUIDOTTI, 1966), though it should be remembered that this process has to be considered in the wider context of a global adaptation of the rocks to the new conditions. This implies also the high Na-content in neogenic K-feldspar (HEALD, 1950; CHAPMAN, 1952; BARKER, 1961, 1962; GUIDOTTI, 1963; EVANS and GUIDOTTI, 1966), the appearance of Ti-richer biotites (OKI, 1961; KWAK, 1968) and the appearance of garnets which are Fe- and Mg-rich and Ca-poor (LAMBERT, 1959; STURT, 1962.)

#### 6d. Sensitivity of metamorphic white micas to other variables

Up to now we have considered the stability and the composition of equilibrium of white micas in function of T, P and the chemical composition of the rocks. However, other factors exist which limit the stability of white mica and presumably influence its composition.

For example, we quote postkinematic isochemical feldspathization, so frequent in pelitic metamorphites; in this process, inside the T-P stability field of white micas, these minerals are destroyed and partly replaced by feldspars. This indicates a sensitivity of white micas to other variables which we did not take into consideration.

SASSI (1971) has recently proposed a genetic model for this type of feldspathization: the model involves the increase in pH and in alkali and oxygen concentrations which occur in the intergranular fluid when the tectonic over-pressure disappears.

The same mechanism has also been proposed to explain some occurrences of paragonite; it is in fact possible that, during the postkinematic phase, partial paragonitization of muscovite takes place, even in rocks which did not previously contain sodic mica.

These are however particular phenomena, even if they are quite widespread in nature. It is therefore valid to state that the stability and composition of metamorphic white micas is mainly controlled by temperature and pressure, above all by the former.

## 7. CONCLUSIONS

The present work, based on the statistical elaboration of about 400 chemical analyses of white mica (excluding paragonite) and on the critical review of previous knowledge on this subject, demonstrates the sensitivity of this mineral to the intensive and extensive variables; it has been possible to select the effects of the different variables, and to individualize well separated paragenetic fields and systematic compositional variations for metamorphic micas crystallized under different T-P conditions.

The principal results obtained are:

1. *The cumulative distribution curves* of chemical components, drawn on probability graphs, show different trends which reflect the behaviour of the different components. Minor constituents (Ca and Ti) show log-normal distributions while the common isomorphous ions ( $\text{Fe}^{+3}$ ,  $\text{Fe}^{+2}$ , Mg, Na) show "bi-normal" distributions, which are caused by two truncated and juxtaposed normal distributions (Fig. 1).

2. *The correlations* between pairs of constituents (Table 2) reflect the isomorphous relationships and the passage to phengites, which takes place mainly through the mechanism  $\text{Si} + \text{Mg}$  (or  $\text{Fe}^{+2}$ )  $\rightleftharpoons \text{Al}^{\text{IV}} + \text{Al}^{\text{VI}}$ , but also through  $\text{Si} + \square^{\text{XII}} \rightleftharpoons \text{Al}^{\text{IV}} + \text{K}$  (or Na).

3. *The white micas of granites, pegmatites and veins* have a chemical composition different from that of medium-high-pressure metamorphic ones. The slight overlapping of the two fields is caused by the convergency of genetic conditions between those of granites and those of high-grade metamorphites. This width of the compositional fields of micas of granites, pegmatites and veins reflects the multiplicity of the genetic processes of these rocks (Fig. 2).

4. The composition of metamorphic micas is controlled to some extent by the *chemical composition of the rocks*, in the sense that, for example, in rocks very rich in silicon and aluminum, micas with relatively high phengite-contents can appear even under relatively high T and relatively low P conditions. However, the control of the chemical composition (Fig. 10) is unimportant with respect to that exercised by the intensive variables (mainly by temperature) as is shown by the fact that clearly distinct paragenetic fields have been recognized, disregarding the bulk composition of the rocks (Figs. 3 and 5).

5. White micas belonging to different facies series can be well distinguished on the basis of their phengite content in solid solution; in fact, an increase in *pressure* brings about an increase of Mg (Fig. 3) and Fe, and a decrease of  $\text{Al}^{\text{IV}}$ .

6. On increasing the metamorphic grade, a progressive decrease of oxidation of white micas can be observed (Fig. 9).

7. No correlation between the *Ca-content* and intensive variables has been noted; thus the entrance of  $\text{Ca}^{2+}$  ions must be related to compositional factors

of the intergranular fluid and/or of the rock and favoured by the fact that the substitution of K by Ca is a capture process.

8. The most marked effects are due to the increase in *temperature*, (i. e. metamorphic grade): increase in  $Al^{IV}$ , Ti,  $R^{+3}$  and decrease in  $Fe_{tot}$ , Mg and  $R^{+2}$  contents; gradual increase of Na to the kyanite-almandine-muscovite subfacies, and its successive rapid diminution in the short thermal range in which the white mica is still stable (Fig. 5).

9. The analytical data taken into consideration did not show systematic variations of the *distribution coefficients* of the elements between white mica and coexisting minerals. This result, however, cannot be interpreted by the absence of  $K_D$  variation rules, but is due to the non-homogeneity of the analytical data and the high sensitivity of the distribution coefficients<sup>4</sup>).

10. The compositional modifications of white mica, owing to their continuous character (due to the substantial dependence on the intensive variables) should not be explained by a series of discontinuous reactions which have a mica A in the left member and a mica B in the right member. Instead, the trend should be interpreted as a continuous mechanism of *ionic exchange* between white mica and intergranular fluid.

11. The field of natural *phengites* in quartz-bearing metamorphic pelites is inserted, purely indicatively, in the T-P diagram of Fig. 11, so that it agrees with the petrographic evidence and the available experimental data. In the same figure the variations in the Fe- and Mg-contents at varying temperatures are also shown. The antithetical trend at lower metamorphic temperatures should also be noted. This, if it does not depend on the scarcity and/or impurities of samples of group 1, would signify that  $Fe^{3+}$  at these temperatures prefers to enter other minerals rather than white mica, a behaviour that is attenuated and then inverted with increasing temperature.

12. Petrographic and experimental reasons exist for separating the first appearance of  $Kf + sillimanite$  (also at medium-high pressures) from the reaction curve  $Mu + Q$ . Fig. 12 tentatively illustrates the temperature values for this appearance, as well as the variations in the *Na-content* with increasing metamorphic grade, for medium-high pressure facies series. The diagram agrees both with petrographic evidence and with the results of experimental petrology, and in particular with the phase diagram of EUGSTER and YODER (1955a).

13. The isochemical postkinematic feldspathization at the expense of white mica demonstrates that the stability of this mineral is controlled by other variables too. This problem has been recently discussed by SASSI (1971), who has proposed a genetic model for this process.

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<sup>4</sup>) See footnote on page 282.



The same mechanism can produce compositional variations in potassic white mica and, at most, its later replacement by paragonite.

14. Polymetamorphism, the polyphase character of the metamorphic events and the lack of equilibrium in metamorphic rocks are frequent. These facts require a fine X-ray analysis on the concentrates of white mica before carrying out the chemical analysis: in fact, the occurrence in metamorphites of several generations of micas of different composition, crystallized at successive times and in different environmental conditions, is far from rare. On the other hand one should remember that diffractometric analysis of white micas may allow an evaluation of the composition of this mineral with a precision which is often superior to that necessary for studies of regional petrology (CIPRIANI, SASSI and VITERBO-BASSANI, 1968).

However, the chemical analysis of minerals should be accompanied by the chemical analysis of the corresponding rocks, as well as by microstructural data on the tectonic-metamorphic evolution.

15. The sensitivity of white micas to the intensive variables shown in this work is such that these minerals alone allow us to detect and map a metamorphic zoneography, when more immediate criteria are lacking, and to recognize the connection of any metamorphic complex or of any paragenesis with a facies series of high, medium or low pressure, in the absence of critical minerals. Finally, the results of the present research can be of great utility in the study of clastic sediments, in order to go back to the parent-rocks and to paleogeographic reconstructions.

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## APPENDIX

**Analytical Data and Respective References**

This appendix contains:

1. The analytical data used for this work recalculated, when necessary, on the basis of O + OH = 24.
2. The group to which each sample was attributed for the statistical analysis by classes; such groups are distinguished by the same number indicated in the Introduction.
3. The relative bibliographical sources, indicated by a progressive number.

For metamorphic micas the attribution to a given subfacies as been made, generally, on the basis of descriptions of rocks supplied by the various Authors. Such attribution was not always possible, and it cannot be excluded that, in some cases, mistakes have been made.

Ref.	Parag. group	Si	Al <sup>IV</sup>	Al <sup>VI</sup>	Ti	Fe <sup>+3</sup>	Fe <sup>+2</sup>	Mg	Mn	Na	K	Ca
1	4	6.18	1.82	1.90	0.18	0.21	0.01	0.11		0.34	1.47	
2	—	5.58	2.42	3.56		0.02	0.16	0.26		0.18	1.48	0.12
2	—	5.62	2.38	3.62		0.02	0.14	0.26		0.22	1.52	0.14
2	—	6.22	1.78	4.03		0.02	0.18	0.14		0.27	1.53	0.18
2	—	6.15	1.85	3.87		0.03	0.18	0.17		0.21	1.56	0.19
3	10	6.07	1.93	3.06	0.03	0.13	0.14	0.70		0.42	1.69	0.14
4	1	6.68	1.32	2.80	0.08	0.28	0.14	0.66		0.12	1.72	0.04
4	1	6.60	1.40	2.86	0.08	0.46	0.10	0.44		0.16	1.80	0.04
4	1	6.72	1.28	2.66	0.04	0.42	0.20	0.76		0.18	1.68	0.06
5	2	6.61	1.39	2.74	0.04	0.52	0.16	0.54	0.01	0.04	1.82	0.01
5	2	6.79	1.21	2.94	0.02	0.25	0.19	0.70	tr	0.04	1.78	0.01
5	2	6.58	1.42	3.07	0.02	0.25	0.24	0.58	tr	0.05	1.80	tr

Ref.	Parag. group	Si	Al <sup>IV</sup>	Al <sup>VI</sup>	Ti	Fe <sup>+3</sup>	Fe <sup>+2</sup>	Mg	Mn	Na	K	Ca
5	1	6.76	1.24	2.95	0.02	0.18	0.19	0.73	0.01	0.02	1.88	tr
5	1	6.16	1.84	3.56	0.02	tr	0.20	0.22	tr	0.18	1.66	tr
6	2	6.49	1.51	2.68	0.09	0.50	0.36	0.41	0.01	0.07	1.81	0.03
6	2	6.35	1.65	3.07	0.10	0.39	0.23	0.23	0.01	0.07	1.87	0.01
6	2	6.37	1.63	3.18	0.10	0.15	0.31	0.36	0.01	0.14	1.71	0.04
6	3	6.33	1.67	2.88	0.10	0.56	0.25	0.33	0.01	0.06	1.84	tr
6	3	6.27	1.73	3.15	0.09	0.21	0.31	0.36	tr	0.08	1.83	0.02
6	4	6.32	1.68	3.18	0.09	0.21	0.30	0.29	0.01	0.12	1.86	tr
6	4	6.37	1.63	3.05	0.08	0.30	0.22	0.40	0.01	0.12	1.85	0.04
6	4	6.37	1.63	3.00	0.10	0.46	0.19	0.46	0.01	0.12	1.81	tr
6	4	6.30	1.70	3.19	0.11	0.17	0.21	0.34	tr	0.11	1.81	0.02
6	4	6.21	1.79	3.40	0.10	0.15	0.11	0.29	tr	0.14	1.69	0.03
6	4	6.52	1.48	3.25	0.09	0.26	0.24	0.28	tr	0.13	1.78	tr
6	4	6.31	1.69	3.31	0.10	0.24	0.13	0.27	0.01	0.12	1.80	0.01
6	4	6.33	1.67	3.16	0.09	0.18	0.22	0.37	tr	0.10	1.82	tr
6	4	6.38	1.62	3.31	0.05	0.22	0.24	0.36	0.01	0.09	1.81	0.01
6	4	6.31	1.69	3.00	0.09	0.40	0.30	0.26	0.01	0.07	1.80	0.07
6	4	6.05	1.95	3.40	0.09	0.10	0.11	0.20	tr	0.30	1.52	tr
6	4	6.19	1.81	3.38	0.08	0.15	0.14	0.28	tr	0.19	1.75	0.02
6	4	6.12	1.88	3.44	0.08	0.16	0.12	0.30	tr	0.19	1.69	tr
6	4	6.31	1.69	3.23	0.10	0.21	0.19	0.33	tr	0.07	1.84	tr
6	4	6.25	1.75	3.07	0.09	0.42	0.18	0.31	tr	tr	1.85	0.02
6	4	6.21	1.79	3.43	0.11	0.20	0.10	0.22	tr	0.26	1.66	tr
6	4	6.13	1.87	3.61	0.11	0.09	0.10	0.19	tr	0.31	1.57	tr
7	8	6.61	1.39	2.24	0.06	0.14	0.10	0.27	tr	0.35	1.36	0.10
8	6	6.15	1.85	3.61	0.11	0.10	0.10	0.12	tr	0.22	1.65	tr
8	5	6.08	1.92	3.35	0.06	0.32	0.08	0.23	0.03	0.32	1.51	tr
8	-	6.08	1.92	3.04	tr	0.58	tr	0.28	tr	0.82	1.28	tr
8	-	5.85	2.15	3.45	0.06	0.41	0.03	tr	tr	0.48	1.71	0.06
9	10					0.26		0.57		0.28	1.72	
9	10					0.24		0.61		0.22	1.78	
9	-					0.38		0.22		0.07	1.93	
9	4					0.05		0.25		0.55	1.45	
9	4					0.16		0.25		0.44	1.56	
9	4					0.07		0.21		0.33	1.37	
9	9					0.12		0.08		0.42	1.48	
9	-					0.15		0.73		0.17	1.43	
9	2					0.50		0.59		0.11	1.79	
9	3					0.38		0.44		0.36	1.64	
9	2					0.21		0.59		0.17	1.83	
9	-					0.41		0.36		0.09	1.91	
9	-					0.48		0.28		0.11	1.89	
9	9					0.12		0.06		0.32	1.68	
9	9					0.15		0.05		0.20	1.80	
9	9					0.17		0.06		0.18	1.82	
10	1	6.40	1.60	1.70	0.05		0.24	0.38		tr	1.86	
10	1	6.25	1.75	1.74	0.04		0.25	0.35		0.08	1.86	
10	2	6.26	1.74	1.84	0.04		0.16	0.40		0.12	1.84	
10	2	6.08	1.92	1.44	0.05		0.17	0.28		tr	1.88	
10	2	6.22	1.78	1.87	0.04		0.19	0.33		0.14	1.86	
10	2	6.27	1.73	1.97	0.04		0.17	0.33		0.13	1.88	
10	2	6.25	1.75	1.78	0.04		0.18	0.32		0.14	1.84	
10	2	6.24	1.76	1.78	0.04		0.19	0.33		0.15	1.84	
10	4	6.14	1.86	1.59	0.03		0.17	0.28		tr	1.86	
10	4	6.11	1.89	1.52	0.03		0.18	0.29		tr	1.82	
10	4	6.00	2.00	1.28	0.03		0.18	0.26		tr	1.84	
10	4	6.12	1.88	1.57	0.04		0.15	0.26		tr	1.83	
10	4	6.25	1.75	1.94	0.05		0.11	0.25		tr	1.84	
10	4	6.04	1.96	1.47	0.05		0.12	0.22		tr	1.83	
11	-	7.06	0.94	2.44	0.08	0.72	tr	1.18	0.10	tr	1.94	0.04

Ref.	Parag. group	Si	Al <sup>IV</sup>	Al <sup>VI</sup>	Ti	Fe <sup>+3</sup>	Fe <sup>+2</sup>	Mg	Mn	Na	K	Ca
11	10	7.00	1.00	2.18	0.10	0.40	0.32	0.84	tr	0.30	1.26	0.38
11	2	6.54	1.46	2.54	tr	0.84	0.30	0.38	tr	0.04	1.88	tr
11	10	6.76	1.24	2.72	0.06	0.20	0.08	1.10	tr	0.24	1.60	0.02
11	10	6.62	1.38	2.72	0.02	0.46	0.02	0.80	0.04	0.14	1.62	0.02
11	10	6.66	1.34	2.80	0.06	0.38	0.26	0.58	tr	0.18	1.56	0.06
11	10	6.78	1.22	2.86	0.02	0.10	0.18	1.00	tr	0.14	1.74	0.04
11	10	6.18	1.72	3.02	0.02	0.16	0.20	0.80	tr	0.10	1.58	0.04
11	—	6.10	1.90	3.10	0.04	0.12	0.14	0.70	tr	0.42	1.70	0.14
11	9	6.14	1.86	3.44	0.04	0.26	0.18	0.16	tr	0.18	1.78	0.02
11	9	6.04	1.96	3.80	0.02	0.04	0.10	0.12	tr	0.20	1.72	tr
11	9	6.08	1.92	3.96	tr	tr	tr	0.02	tr	0.16	1.70	tr
12	10	6.80	1.20	2.60	0.12	0.38	0.30	0.58		0.26	1.48	0.16
12	10	6.84	1.16	2.98	0.04	0.12	0.40	0.54		0.16	1.56	0.08
12	—	7.00	1.00	2.28	0.10	0.40	0.32	0.84		0.30	1.26	0.38
12	—	6.66	1.34	2.80	0.06	0.38	0.26	0.58	tr	0.18	1.56	0.06
12	10	6.72	1.28	2.60	0.16	0.32	0.24	0.72	tr	0.14	1.70	0.02
12	10	6.96	1.06	2.64	0.06	0.48	0.14	0.68	0.04	0.16	1.60	0.02
13	6	6.10	1.90	3.55	0.07	0.18		0.22		0.08	1.84	
13	6	6.06	1.94	3.62	0.08	0.14		0.16		0.16	1.80	
13	6	6.09	1.91	3.57	0.13	0.10		0.21		0.16	1.76	
13	6	6.04	1.96	3.50	0.15	0.12		0.24		0.12	1.82	
13	6	6.02	1.98	3.72	0.07	0.12		0.12		0.20	1.72	
13	6	6.06	1.94	3.72	0.08	0.08		0.14		0.26	1.68	
13	6	6.00	2.00	3.76	0.06	0.12		0.11		0.28	1.64	
13	6	5.99	2.01	3.75	0.07	0.11		0.11		0.28	1.66	
13	6	6.05	1.95	3.58	0.12	0.11		0.22		0.16	1.76	
13	6	6.02	1.98	3.70	0.08	0.13		0.12		0.24	1.68	
13	6	6.00	2.00	3.75	0.05	0.13		0.10		0.28	1.62	
13	6	6.01	1.99	3.74	0.07	0.11		0.12		0.28	1.62	
13	6	6.00	2.00	3.71	0.08	0.11		0.12		0.30	1.62	
13	6	5.98	2.02	3.62	0.13	0.11		0.15		0.28	1.64	
13	6	6.01	1.99	3.74	0.09	0.11		0.11		0.30	1.58	
13	6	6.00	2.00	3.81	0.04	0.11		0.10		0.34	1.54	
13	7	6.00	2.00	3.64	0.10	0.13		0.14		0.18	1.76	
13	7	6.01	1.99	3.72	0.07	0.11		0.13		0.14	1.76	
13	7	6.03	1.97	3.66	0.12	0.12		0.11		0.12	1.82	
13	7	6.06	1.94	3.61	0.13	0.14		0.12		0.12	1.76	
13	7	6.04	1.96	3.72	0.05	0.14		0.13		0.12	1.78	
13	7	6.03	1.97	3.62	0.13	0.10		0.16		0.14	1.80	
13	7	6.03	1.97	3.59	0.13	0.12		0.16		0.14	1.78	0.01
13	7	6.06	1.94	3.53	0.12	0.20		0.16		0.10	1.84	
13	7	6.06	1.94	3.71	0.08	0.13		0.10		0.10	1.78	
13	7	6.03	1.97	3.68	0.08	0.13		0.13		0.12	1.80	
13	7	6.00	2.00	3.62	0.13	0.14		0.12		0.14	1.80	
13	7	6.02	1.98	3.64	0.12	0.13		0.11		0.16	1.74	0.01
13	7	6.01	1.99	3.58	0.14	0.11		0.19		0.14	1.78	
13	7	6.03	1.97	3.70	0.08	0.11		0.12		0.14	1.76	
13	7	6.04	1.96	3.69	0.09	0.13		0.12		0.12	1.78	
13	7	6.07	1.93	3.70	0.09	0.11		0.09		0.12	1.82	
14	9	6.05	1.95	3.28	0.02	0.40	0.11	0.19		0.10	1.72	0.05
14	9	6.06	1.94	3.22	0.02	0.34	0.15	0.19		0.17	1.66	0.03
14	9	5.99	2.01	3.36	0.03	0.21	0.13	0.23		0.14	1.78	0.01
14	9	6.09	1.91	3.22	0.03	0.30	0.10	0.40		0.18	1.67	0.02
14	9	6.05	1.95	3.48	0.03	0.22	0.14	0.18		0.14	1.64	0.03
14	9	6.02	1.98	3.48	0.04	0.22	0.10	0.18		0.14	1.79	0.01
14	9	6.10	1.90	2.91	0.04	0.55	0.21	0.27		0.17	1.70	0.02
14	9	5.95	2.05	3.16	0.04	0.51	0.10	0.24		0.12	1.66	0.05
14	9	6.01	1.99	3.11	0.02	0.57	0.10	0.21		0.15	1.65	0.02
14	9	6.02	1.98	3.31	0.05	0.43	0.09	0.19		0.10	1.54	0.03
14	9	6.03	1.97	3.30	0.02	0.39	0.09	0.15		0.15	1.77	0.02



Ref.	Parag. group	Si	Al <sup>IV</sup>	Al <sup>VI</sup>	Ti	Fe <sup>+3</sup>	Fe <sup>+2</sup>	Mg	Mn	Na	K	Ca
14	9	6.00	2.00	3.43	0.01	0.38	0.06	0.15		0.12	1.65	0.02
14	9	6.00	2.00	3.25	0.01	0.24	0.14	0.26		0.14	1.71	0.01
14	9	6.02	1.98	3.30	0.05	0.48	0.18	0.08		0.08	1.65	tr
14	9	6.06	1.94	3.26	0.04	0.59	0.07	0.09		0.08	1.65	0.03
14	9	5.79	2.21	3.47	0.05	0.16	0.09	0.10		0.26	1.23	0.06
15	3	6.60	1.40	3.17	0.02	0.10	0.14	0.57		0.18	1.70	0.02
15	3	6.56	1.44	3.24	0.04	0.10	0.24	0.39		0.07	1.78	0.05
15	11	6.06	1.94	3.58	0.02	0.12	0.09	0.18		0.06	1.84	0.02
16	—	6.00	2.00	3.94	tr	0.06	tr	tr		0.23	1.72	tr
16	—	6.22	1.78	3.67	0.02	0.07	0.09	0.18		0.12	1.81	0.02
16	—	6.17	1.83	3.75	tr	tr	0.02	0.11		0.15	1.70	0.04
16	—	6.54	1.46	3.36	tr	tr	0.16	0.44		0.12	1.83	0.02
16	—	6.70	1.30	3.44	0.02	0.14	0.05	0.50		0.51	1.68	tr
16	—	6.78	1.22	2.94	0.11	0.30	0.15	0.56		0.26	1.66	tr
17	—	6.22	1.78	3.67	0.02	0.07	0.09	0.18		0.12	1.80	0.02
17	—	6.94	1.06	2.61	tr	tr	0.64	0.48		tr	1.60	0.09
17	—	6.53	1.47	3.35	tr	tr	0.16	0.44		0.12	1.85	0.02
18	1	6.50	1.50	2.99	0.03	0.23	tr	0.84		0.08	1.64	tr
18	2	6.86	1.14	3.26	0.05	0.23	tr	0.36		0.16	1.63	0.03
18	4	6.30	1.70	3.48	0.04	0.13	0.14	0.20		0.50	1.46	0.02
19	8	6.35	1.65	1.24	tr	0.21	0.35	tr		0.51	1.66	
20	5	6.18	1.82	3.42	0.04	0.39	0.05	0.16		0.38	1.62	tr
20	8	6.39	1.61	3.12	0.08	0.46	0.20	0.10		0.15	1.60	0.10
20	5	6.49	1.51	3.51	0.08	0.15	0.15	0.22		0.37	1.51	0.04
20	5	6.44	1.56	3.46	0.10	0.08	0.14	0.22		0.26	1.57	0.06
20	4	6.38	1.62	3.38	0.10	0.15	0.19	0.24		0.40	1.59	0.06
20	4	6.61	1.39	3.14	0.09	0.35	0.15	0.30		0.31	1.60	0.05
20	4	6.48	1.52	3.52	0.09	tr	0.19	0.33		0.31	1.40	0.11
20	4	6.51	1.49	3.12	0.08	0.39	0.18	0.34		0.31	1.61	0.03
20	4	6.63	1.37	3.20	0.05	0.30	0.21	0.31		0.15	1.59	0.06
20	4	6.66	1.34	3.08	0.08	0.39	0.19	0.36		0.11	1.59	0.10
20	—	6.74	1.26	2.86	0.04	0.55	0.15	0.47		0.24	1.64	0.03
20	8	6.68	1.32	3.13	0.03	0.31	0.34	0.31		0.07	1.61	0.07
20	—	6.73	1.27	2.59	0.10	0.64	0.20	0.48		0.12	1.74	0.06
20	—	6.72	1.28	2.78	0.04	0.60	0.25	0.45		0.20	1.62	0.06
20	—	7.21	0.79	2.41	0.10	0.55	0.12	0.59		0.53	1.83	tr
20	—	6.11	1.89	3.08	0.04	0.13	0.14	0.70		0.42	1.70	0.15
20	—	6.92	1.08	2.93	0.05	0.23	0.15	0.71		0.09	1.60	0.09
20	—	6.83	1.17	2.85	0.07	0.30	0.11	0.84		0.37	1.69	tr
21	—	6.30	1.70	3.41		0.40	0.14	0.15	0.01	0.18	1.67	0.01
22	9	5.95	2.05	0.95	0.07	0.45	0.09	0.26		0.20	1.81	
23	4	5.92	2.08	3.54	0.07	0.07	0.08	0.24		0.36	1.50	0.02
23	4	5.45	2.55	2.98	0.17	0.49	0.16	0.29		0.13	2.10	tr
23	4	5.46	2.54	3.14	0.05	0.38	0.18	0.17		0.07	2.37	tr
23	4	5.91	2.09	3.48	0.01	0.17	0.17	0.12		0.13	2.01	0.01
23	4	6.63	1.37	3.37	0.07	0.25	0.12	0.17		0.31	1.38	0.04
23	4	6.00	2.00	3.55	0.09	0.08	0.06	0.24		0.43	1.41	0.01
23	4	5.62	2.38	3.71	0.08	0.22	0.06	0.16		0.42	1.65	tr
23	9	5.83	2.17	3.28	0.04	0.23	0.06	0.17		0.35	1.53	0.03
23	4	5.42	2.58	2.90	0.14	0.54	0.12	0.45		0.11	1.95	0.01
23	4	5.57	2.43	4.08	0.10	0.09	0.10	0.22		0.33	1.71	tr
23	4	6.00	2.00	2.73	0.09	0.44	0.16	0.38		0.11	2.22	0.01
23	4	6.34	1.66	3.12	0.05	0.17	0.10	0.66		0.08	1.83	0.01
23	4	5.48	2.52	3.18	0.16	0.38	0.11	0.34		0.11	1.94	tr
23	4	5.66	2.34	3.01	0.11	0.46	0.12	0.41		0.11	1.96	tr
24	10	6.61	1.38	2.72	0.02	0.46	0.02	0.80	0.04	0.14	1.62	0.02
25	10	6.40	1.60	2.80	tr	0.60	0.20	0.40	tr			0.20
26	10	6.73	1.27	2.16	0.06	0.95	0.23	0.59	0.04	0.10	1.74	0.01
27	3	6.55	1.45	3.39	0.08	0.24	0.23	0.15		0.24	1.76	tr
27	3	6.70	1.30	3.39	0.08	0.16	0.24	0.31		0.28	1.72	0.06

Ref.	Parag. group	Si	Al <sup>IV</sup>	Al <sup>VI</sup>	Ti	Fe <sup>+3</sup>	Fe <sup>+2</sup>	Mg	Mn	Na	K	Ca
27	4	6.20	1.80	3.46	0.07	0.14	0.13	0.30		0.26	1.65	tr
27	4	6.40	1.60	3.21	0.08	0.26	0.17	0.28		0.25	1.59	0.05
27	4	6.69	1.31	3.34	0.08	0.23	0.19	0.30		0.36	1.52	0.12
27	4	6.26	1.74	3.54	0.08	0.02	0.13	0.33		0.24	1.59	0.02
27	4	6.16	1.84	3.31	0.10	0.10	0.11	0.29		0.28	1.63	tr
27	2	6.63	1.37	3.00	0.09	0.67	0.09	tr		0.08	1.96	tr
27	2	6.28	1.72	2.96	0.09	0.47	0.24	0.34		0.08	1.94	tr
27	2	6.43	1.57	3.13	0.05	0.42	0.20	0.47		0.09	1.78	0.05
27	2	6.65	1.35	2.94	0.08	0.57	0.22	0.07		0.11	1.80	0.01
27	2	6.30	1.70	2.97	0.09	0.42	0.26	0.25		0.10	1.85	tr
27	—	5.96	2.04	3.75	0.09	tr	tr	0.04		0.43	1.49	0.07
27	—	5.95	2.05	3.98	tr	tr	0.03	0.01		0.51	1.48	tr
27	—	6.81	1.19	3.77	tr	0.01	0.24	0.44		tr	1.58	0.04
27	—	6.09	1.91	3.07	tr	0.58	tr	0.23		0.82	1.28	tr
28	4							0.13		0.20	1.37	0.07
28	4							0.12		0.20	1.32	0.06
28	4							0.26		0.10	1.67	0.05
28	6							0.15		0.18	1.50	0.07
29	6	6.00	2.00	3.52	0.07	0.18	0.06	0.10		0.26	1.77	0.01
30	1	6.10	1.90	3.40	0.08	0.13	0.14	0.17		0.21	1.51	0.12
30	1	6.81	1.19	2.88	0.06	0.15	0.52	0.29		0.25	1.23	0.02
30	1	6.29	1.71	2.94	0.07	0.25	0.41	0.72		0.50	0.94	0.10
30	1	6.49	1.51	3.16	0.09	0.13	0.43	0.48	0.01	0.42	1.25	0.07
30	1	6.55	1.45	3.24	0.10	0.20	0.28	0.44		0.29	1.42	0.02
30	1	6.31	1.69	3.51	0.07	0.15	0.16	0.29		0.24	1.67	0.03
30	1	6.39	1.61	3.26	0.10	0.17	0.33	0.26		0.19	1.76	0.05
30	2	6.29	1.71	3.28	0.04	0.25	0.25	0.36		0.17	1.67	0.03
30	2	7.06	0.94	3.27	0.07	0.27	0.67	0.60		0.38	0.86	0.09
30	2	6.06	1.94	4.03	0.03	0.11	0.05	0.07		0.32	1.42	0.03
31	—	6.37	1.63	3.26	0.05	0.09	0.01	0.63		tr	1.78	0.03
31	9	6.16	1.84	3.14	0.07	0.42	0.08	0.32		0.08	1.58	0.03
31	—	6.30	1.70	3.20	0.07	0.40	0.09	0.28		0.08	1.62	0.07
31	9	6.12	1.88	3.43	0.01	0.09	0.01	0.49		0.13	1.68	0.03
31	—	6.53	1.47	2.75	0.08	0.35	0.06	0.49		0.13	1.67	0.03
31	9	6.18	1.82	2.69	0.06	0.40	0.06	0.36		0.08	1.62	0.13
31	—	6.03	1.97	3.03	0.07	0.61	0.06	0.20		0.11	1.69	0.03
31	9	6.96	1.04	3.16	0.03	0.44	0.05	0.16		0.15	1.33	0.02
31	—	6.47	1.53	3.08	0.08	0.46	0.06	0.28		0.13	1.62	0.04
31	9	6.05	1.95	3.00	0.04	0.56	0.09	0.42		0.05	1.61	0.04
31	—	6.22	1.78	3.05	0.09	0.49	0.08	0.32		0.13	1.71	0.03
31	9	6.17	1.83	3.00	0.06	0.49	0.11	0.36		0.01	1.76	0.02
31	9	6.64	1.36	2.91	0.05	0.21	0.14	0.46		0.10	1.72	0.09
31	—	6.41	1.59	3.60	0.09	0.19	0.04	0.25		0.18	1.18	0.03
31	—	6.30	1.70	3.24	0.10	0.45	0.12	0.22		0.11	1.67	0.03
31	9	6.01	1.99	3.14	0.07	0.42	0.12	0.34		0.08	1.80	0.02
31	9	5.90	2.10	3.24	0.03	0.17	0.08	0.32		0.11	2.15	0.03
31	9	6.24	1.76	3.35	0.03	0.09	0.11	0.37		0.10	1.48	0.02
31	9	6.26	1.74	3.08	0.04	0.12	0.10	0.42		0.19	2.35	tr
31	9	6.50	1.50	3.25	0.01	0.08	0.05	0.58		0.10	1.33	0.03
31	9	5.84	2.16	3.50	0.05	0.08	0.12	0.24		0.10	1.65	0.04
31	9	5.45	2.55	3.12	0.05	0.38	0.18	0.17		0.07	2.37	tr
31	9	5.75	2.25	3.37	0.05	0.06	0.12	0.30		0.11	2.17	0.06
31	9	5.73	2.27	3.68	0.03	0.08	0.03	0.13		0.30	1.09	0.02
31	9	5.07	2.90	3.11	0.02	0.07	0.03	0.20		0.28	2.12	0.20
31	—	6.15	1.85	2.98	0.07	0.38	0.15	0.36		0.13	1.57	0.07
31	9	6.07	1.93	3.21	0.07	0.37	0.13	0.33		0.08	1.68	0.03
31	—	6.44	1.56	2.88	0.06	0.33	0.14	0.30		0.21	1.62	0.09
31	9	5.93	2.07	3.42	0.01	0.03	0.05	0.31		0.23	1.52	0.04
31	—	6.04	1.96	2.79	0.06	0.37	0.12	0.48		0.18	1.88	0.03
31	9	5.94	2.06	3.02	0.09	0.51	0.11	0.30		0.08	1.73	0.02

Ref.	Parag. group	Si	Al <sup>IV</sup>	Al <sup>VI</sup>	Ti	Fe <sup>+3</sup>	Fe <sup>+2</sup>	Mg	Mn	Na	K	Ca
32	10	6.72	1.28	2.34	0.08	0.76	0.10	0.72	0.02	0.10	1.82	0.06
33	8	5.88	2.12	3.67	0.11	0.11	0.14	0.19		0.15	1.79	tr
33	8	5.97	2.03	3.65	0.02	0.19	tr	0.17		0.19	1.71	0.13
33	8	5.84	2.16	3.63	0.01	0.16	0.20	0.17		0.24	1.71	tr
33	8	5.84	2.16	3.64	0.09	0.05	0.15	0.21		0.19	1.75	tr
33	8	5.93	2.07	3.83	0.06	0.13	tr	0.16		0.21	1.73	0.03
33	8	5.89	2.11	3.51	0.03	0.25	0.38	0.12		0.16	1.75	tr
33	8	5.87	2.13	3.77	0.05	0.04	0.12	0.18		0.17	1.80	tr
33	8	6.00	2.00	3.75	0.08	0.01	0.13	0.19		0.17	1.83	tr
34	6	6.05	1.95	3.73	0.05	0.06	0.05	0.15		0.21	1.88	
34	7	6.10	1.90	3.66	0.08	0.12	0.03	0.16		0.21	1.72	
34	11	6.01	1.99	3.43	0.09	0.30	0.05	0.13		0.19	1.71	
34	11	6.03	1.97	3.45	0.06	0.26	0.07	0.12		0.29	1.72	
35	8	6.36	1.64	3.44	0.06	0.22	0.12	0.14	0.06	0.19	1.72	0.07
35	8	6.34	1.66	3.33	0.08	0.28	0.10	0.17	tr	0.20	1.73	0.14
36	3	6.45	1.55	2.89	0.26	0.40	0.14	0.20		0.26	1.67	0.02
36	3	6.68	1.32	2.90	0.18	0.40	0.09	0.16		0.34	1.67	0.06
36	3	6.57	1.43	3.03	0.20	0.30	0.16	0.20		0.16	1.66	0.20
36	3	6.71	1.29	3.12	0.08	0.38	0.06	0.30		0.26	1.73	0.02
36	3	6.33	1.67	2.87	0.11	0.53	0.07	0.36		0.11	1.80	0.02
36	3	6.30	1.70	3.00	0.14	0.40	0.07	0.33		0.19	1.76	0.03
36	3	6.68	1.32	2.78	0.17	0.39	0.11	0.16		0.56	1.43	0.22
36	3	6.52	1.48	2.97	0.22	0.28	0.15	0.04		0.50	1.38	0.17
36	3	6.50	1.50	3.01	0.17	0.18	0.18	0.26		0.22	1.67	0.04
36	3	6.51	1.49	2.74	0.13	0.46	0.09	0.54		0.13	1.83	0.03
37	—	6.16	1.84	3.74		tr	0.03	0.12		0.16	1.71	
37	—	6.38	1.62	3.44		tr	0.13	0.26		0.02	1.54	
37	—	6.39	1.61	2.84		0.15	tr	0.53		0.03	1.60	
37	—	6.54	1.46	3.37		tr	0.16	0.44		0.12	1.85	
37	—	6.79	1.21	3.12		0.07	0.11	0.70		0.03	1.70	
37	—	7.06	0.94	2.37		0.12	tr	1.19		0.09	1.90	
38	—	5.83	2.17	3.65	0.06	0.17	0.08	0.33		0.11	2.07	0.02
38	9	5.95	2.05	3.29	0.03	0.12	0.09	0.16		0.17	1.83	0.02
38	9	6.76	1.24	3.50	0.01	0.08	0.23	0.16		0.25	1.43	0.04
38	—	6.86	1.14	3.57	0.10	0.08	0.11	0.26		0.10	1.73	0.03
38	—	5.99	2.01	3.55	0.11	0.09	0.10	0.26		0.11	1.93	0.03
38	—	5.92	2.08	3.09	0.08	0.28	0.06	0.30		0.10	1.86	0.03
38	9	6.37	1.03	3.27	0.07	0.11	0.08	0.24		0.08	1.86	0.03
38	—	6.22	1.78	3.30	0.09	0.10	0.09	0.22		0.26	1.82	0.06
38	9	5.91	2.09	3.04	0.02	0.40	0.12	0.18		0.13	1.80	0.03
38	9	5.86	2.14	3.22	0.03	0.37	0.11	0.18		0.13	1.85	0.03
38	—	6.23	1.77	3.33	0.08	0.17	0.06	0.28		0.18	1.98	0.04
38	—	6.05	1.95	3.35	0.11	0.13	0.08	0.26		0.15	1.80	0.04
38	—	6.31	1.69	3.39	0.08	0.11	0.12	0.26		0.23	1.66	0.04
38	—	6.49	1.51	3.47	0.09	tr	0.08	0.33		0.17	1.56	0.02
38	—	5.69	2.31	3.40	0.08	0.03	0.09	0.32		0.10	1.87	0.02
38	—	5.61	2.39	3.18	0.06	0.25	0.29	0.37		0.16	1.90	0.02
38	9	5.78	2.22	3.47	0.05	0.16	0.12	0.20		0.15	1.89	0.02
38	9	6.41	1.59	3.37	0.04	0.06	0.09	0.16		0.10	1.76	0.02
38	—	6.44	1.56	3.37	0.06	0.02	0.15	0.26		0.13	2.00	0.02
38	—	6.04	1.96	3.48	0.04	0.02	0.11	0.36		0.08	1.75	0.06
38	—	6.25	1.75	3.55	0.06	0.07	0.06	0.17		0.36	1.57	0.07
38	4	6.09	1.91	3.68	0.05	0.01	0.09	0.20		0.33	1.45	0.02
38	—	5.62	2.38	3.87	0.06	0.06	0.06	0.21		0.35	1.44	tr
38	9	7.03	0.97	3.37	0.01	0.10	0.06	0.06		0.59	1.30	0.04
38	—	6.11	1.89	3.30	0.06	0.12	0.05	0.16		0.18	1.91	0.02
38	—	5.96	2.04	3.00	0.09	0.36	0.10	0.30		0.21	2.14	0.02
38	9	6.01	1.99	3.36	0.04	0.13	0.14	0.21		0.18	1.68	0.02
38	9	5.93	2.07	3.17	0.03	0.41	0.05	0.24		0.18	2.01	0.02
38	9	5.79	2.21	3.45	0.03	0.37	0.01	0.24		0.18	1.86	0.02

Ref.	Parag. group	Si	Al <sup>IV</sup>	Al <sup>VI</sup>	Ti	Fe <sup>+3</sup>	Fe <sup>+2</sup>	Mg	Mn	Na	K	Ca
38	—	5.96	2.04	2.78	0.03	0.50	0.21	0.64		0.05	2.07	0.02
38	—	6.17	1.83	2.93	0.02	0.10	0.03	0.70		0.13	2.15	tr
38	—	6.22	1.78	2.54	0.08	0.16	0.13	0.37		0.17	1.72	0.04
38	9	6.06	1.94	3.42	0.04	0.19	0.06	0.21		0.17	1.64	0.02
38	4	5.90	2.10	3.55	0.02	0.07	0.06	0.04		0.31	1.59	0.02
38	4	6.02	1.98	3.79	0.04	tr	0.05	0.09		0.54	1.44	0.02
38	—	6.22	1.78	2.80	0.06	0.34	0.15	0.34		0.08	1.85	0.04
38	9	5.78	2.22	3.10	0.03	0.07	0.06	0.30		0.23	2.09	0.02
38	9	7.01	0.99	3.81	0.04	0.01	0.09	0.23		0.24	1.18	0.02
38	9	6.15	1.85	3.50	0.01	0.21	0.10	0.12		0.13	1.79	0.02
38	—	5.12	2.88	3.20	0.02	0.07	0.03	0.22		0.28	2.13	0.30
38	—	6.21	1.79	3.58	0.04	0.02	0.15	0.47		0.08	1.24	0.02
38	—	6.18	1.82	3.48	0.09	0.02	0.09	0.23		0.15	1.74	0.02
38	—	5.56	1.44	2.76	0.04	0.33	0.09	0.59		0.19	2.18	0.10
38	—	6.46	1.54	2.87	0.05	0.34	0.06	0.58		0.23	1.99	0.06
38	9	5.71	2.29	2.79	0.08	0.49	0.16	0.58		0.08	1.61	0.04
38	9	5.55	2.45	2.43	0.05	0.23	0.14	0.46		0.08	2.13	0.02
38	—	6.27	1.73	3.13	0.05	0.13	0.12	0.35		0.20	1.22	0.11
38	—	6.21	1.79	3.60	0.05	0.02	0.09	0.24		0.31	1.29	0.02
38	—	5.92	2.08	3.20	0.08	0.20	0.15	0.42		0.11	1.94	0.02
38	—	6.40	1.60	3.23	0.08	0.16	0.15	0.38		0.08	1.82	0.02
38	—	6.06	1.94	3.02	0.05	0.17	0.06	0.51		0.15	1.40	0.13
38	9	6.11	1.89	3.08	0.04	0.05	0.08	0.20		0.13	1.91	0.05
38	9	5.94	2.06	3.62	0.02	0.04	0.12	0.12		0.15	1.92	0.05
38	—	5.47	2.53	3.16	0.16	0.38	0.10	0.34		0.13	1.94	tr
38	—	5.66	2.34	3.01	0.11	0.46	0.12	0.41		0.11	1.96	tr
38	—	5.99	2.01	2.72	0.09	0.44	0.16	0.38		0.11	2.22	0.02
38	—	6.34	1.66	3.11	0.05	0.17	0.10	0.66		0.08	1.83	tr
38	—	5.83	2.17	3.28	0.04	0.23	0.05	0.17		0.35	1.53	0.03
38	—	5.29	2.71	2.63	0.14	0.53	0.12	0.44		0.10	1.90	0.01
38	—	5.39	2.61	3.68	0.10	0.08	0.09	0.22		0.31	1.65	tr
38	—	5.84	2.16	3.48	0.07	0.07	0.08	0.24		0.35	1.51	0.02
38	—	5.45	2.55	2.98	0.17	0.49	0.16	0.29		0.13	2.10	tr
38	—	6.01	1.99	3.57	0.09	0.08	0.06	0.24		0.43	1.41	0.02
38	—	5.62	2.38	3.71	0.08	0.22	0.06	0.16		0.42	1.66	tr
38	9	5.44	2.56	3.55	0.07	0.12	0.11	0.18		0.13	2.20	0.01
38	9	5.51	2.49	3.20	0.09	0.43	0.10	0.22		0.08	2.11	0.01
38	9	5.48	2.52	3.57	0.06	0.09	0.09	0.16		0.23	2.15	tr
38	9	5.50	2.50	3.58	0.06	0.12	0.14	0.20		0.13	1.93	0.01
38	—	5.62	2.38	3.56	0.06	0.14	0.14	0.18		0.13	2.05	0.01
38	—	6.16	1.84	3.28	0.06	0.18	0.18	0.24		0.08	1.99	0.03
38	—	5.44	2.56	2.76	0.09	0.49	0.10	0.30		0.08	2.07	0.01
38	9	5.60	2.40	3.35	0.06	0.20	0.14	0.18		0.13	2.09	tr
38	9	6.13	1.87	3.45	0.01	0.09	0.01	0.49		0.13	1.68	0.03
38	—	6.48	1.52	3.10	0.08	0.46	0.06	0.28		0.13	1.62	0.04
38	—	6.41	1.59	3.60	0.09	0.19	0.04	0.25		0.18	1.18	0.03
38	9	5.90	2.10	3.24	0.03	0.17	0.08	0.32		0.11	2.16	0.03
38	9	5.76	2.24	3.36	0.05	0.06	0.12	0.30		0.11	2.17	0.06
38	9	5.96	2.04	3.06	0.09	0.52	0.11	0.30		0.08	1.74	0.02
38	9	6.05	1.95	2.80	0.06	0.37	0.12	0.48		0.18	1.88	0.03
39	—	5.43	2.57	3.53	0.07	0.12	0.11	0.18		0.13	2.20	tr
39	9	5.60	2.40	3.38	0.06	0.20	0.14	0.18		0.13	2.09	tr
39	9	5.50	2.50	3.20	0.09	0.43	0.10	0.22		0.08	2.11	tr
39	—	5.49	2.51	2.87	0.09	0.50	0.10	0.30		0.08	2.09	tr
39	9	5.74	2.26	3.47	0.08	0.13	0.14	0.24		0.13	2.05	tr
39	9	5.47	2.53	3.50	0.06	0.15	0.14	0.18		0.13	2.06	tr
39	9	5.31	2.69	3.74	0.02	0.11	0.08	0.06		0.15	2.08	tr
39	9	5.49	2.51	3.58	0.06	0.09	0.09	0.16		0.24	2.16	tr
39	9	5.50	2.50	3.58	0.06	0.12	0.14	0.20		0.13	1.93	tr
39	—	5.61	2.39	3.54	0.06	0.14	0.14	0.18		0.13	2.05	tr

Ref.	Parag. group	Si	Al <sup>IV</sup>	Al <sup>VI</sup>	Ti	Fe <sup>+3</sup>	Fe <sup>+2</sup>	Mg	Mn	Na	K	Ca
39	9	5.08	2.92	3.12	0.08	0.10	0.12	0.33		0.27	2.60	tr
39	9	5.22	2.78	3.56	0.03	0.16	0.12	0.12		0.15	2.03	tr
39	9	5.48	2.52	3.57	0.03	0.26	0.14	0.12		0.13	1.63	tr
39	—	5.74	2.26	3.51	0.05	0.16	0.14	0.14		0.10	1.82	tr
39	9	5.62	2.38	3.48	0.05	0.09	0.09	0.14		0.13	2.01	tr
39	9	5.24	2.76	3.42	0.03	0.17	0.09	0.12		0.10	2.29	tr
39	9	6.38	1.62	3.28	0.07	0.11	0.08	0.24		0.08	1.87	0.03
39	9	5.97	2.03	3.32	0.03	0.12	0.09	0.16		0.23	1.83	0.03
40	11	6.54	1.46	3.26	0.04	0.18	0.32	0.28		0.16	1.76	0.02
40	11	6.48	1.52	3.28	0.02	0.24	0.36	0.18		0.08	1.52	0.02
41	10	6.32	1.68	2.20	0.08	0.40	0.11	1.30		0.32	1.78	0.04
41	10	6.43	1.57	3.02	0.08	0.38	0.09	0.56		0.26	1.63	0.01
41	10	6.49	1.51	2.86	tr	0.38	0.17	0.76		0.22	1.56	tr
41	10	6.35	1.65	2.77	0.07	0.45	0.15	0.57		0.19	1.50	tr
41	10	6.28	1.72	2.61	0.11	0.45	0.17	0.68		0.20	1.77	tr
41	10	6.73	1.27	3.37	tr	0.44	0.15	0.67		0.18	1.74	tr
41	10	6.52	1.48	2.99	0.02	0.38	0.10	0.58		0.25	1.64	tr
(+)	11					0.14	0.04	0.07		0.31		
(+)	11					0.14	0.08	0.22		0.33		

(+) New analysis of samples from Bosost Area.

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