

**Zeitschrift:** Schweizerische mineralogische und petrographische Mitteilungen = Bulletin suisse de minéralogie et pétrographie  
**Band:** 69 (1989)  
**Heft:** 1  
  
**Artikel:** b0 of muscovite in low and high variance assemblages from low grade Verrucano rocks, Northern Apennines, Italy  
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**DOI:** <https://doi.org/10.5169/seals-52780>

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# **$b_0$ of muscovite in low and high variance assemblages from low grade Verrucano rocks, Northern Apennines, Italy.**

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## **Abstract**

The regional distribution of the muscovite  $b_0$  parameter as function of metamorphic grade and assemblages in the Verrucano rocks of the Northern Apennines has been investigated. The equation relating  $b_0$  to R.M. ( $\text{MgO}/\text{M.W.} + \text{FeO}_{\text{tot}}/\text{M.W.}$ ) for the Verrucano muscovite is  $b_0 = 8.982 + 0.499 \cdot \text{R.M.}$  with  $r = 0.88$ . The mean  $b_0$  values increase from pyrophyllite or kyanite-bearing rocks (9.002 Å) to K-feldspar-bearing rocks (9.056 Å and 9.042 Å) for pyrophyllite and kyanite grade respectively. In the pyrophyllite or kyanite as well as in the K-feldspar-bearing rocks (low variance assemblages) the mean muscovite  $b_0$  values are very similar in all Verrucano areas and their standard deviations are small. On the contrary, in muscovites from pyrophyllite and K-feldspar-free samples (high variance assemblages) the mean  $b_0$  values vary within the different Verrucano areas and show a high standard deviation.

The muscovite  $b_0$  values in Al-rich low variance assemblages of Verrucano and other metamorphic areas with different P/T regime of metamorphism are discussed in relation to their use as a potential qualitative geobarometer.

**Keywords:**  $b_0$  of muscovite, mineral assemblages, geobarometer, Verrucano rocks, Northern Apennines.

## **Introduction**

Over the last twenty years many studies have shown that the  $b_0$  parameter of K-white mica is particularly useful for characterizing metamorphism in the low grade region. SASSI and SCOLARI (1974) first attempted to calibrate the muscovite  $b_0$  parameter as a qualitative geobarometer in  $\text{ms} + \text{ab} + \text{qtz} \pm \text{chl} \pm \text{carb} \pm \text{grp}$  (mineral abbreviations as in KRETZ, 1983; other abbreviations used in the text: carb = carbonates, grp = graphite, k-fs = K-feldspar, su = sudoite) high variance assemblages for low grade metamorphic rocks. Even though it was suggested a long time ago (GUIDOTTI and SASSI, 1976), very little effort has been spent on calibration of a geobarometer based on the  $b_0$  of muscovite from low variance assemblages.

This paper describes the variation of the muscovite  $b_0$  parameter in low grade metamorphic rocks of the Verrucano metasediments (Northern Apennines, Italy) as function of metamorphic grade, mineralogical assemblage and muscovite composition.

The aim of this study is to establish some constraints on the variation of  $b_0$  in a kyanite-type low temperature metamorphism and to lay the foundations for an empirical  $b_0$  reference scale for the muscovite from low variance assemblage in the model system KNaASH ( $\text{K}_2\text{O}-\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$ ), GUIDOTTI and SASSI (1976).

The Verrucano rocks are particularly suitable for this purpose because they show different degrees of Al-saturation and contain muscovite showing a wide range of chemical variation mainly in the phengite and ferrimuscovite components (FRANCESCHELLI et al., 1986).

## **Regional Setting**

The Verrucano metasediments crop out in discontinuous patches along the entire length of the Northern Apennines (Fig. 1). For details of the geology and stratigraphy of the Verrucano sequences refer to GELMINI (1969), RAU and TONGIORGI (1974), TONGIORGI et al. (1977), CASSINIS

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et al. (1979), COSTANTINI et al. (1985). Mineralogy, geochemistry and general aspects of the Verrucano metamorphism have recently been discussed by FRANCESCHELLI et al. (1986, 1987, 1989).

From those papers, some general features useful for the present discussion, are summarized below.

The Verrucano sequence is mainly composed of quartzites, metapsammites, metaconglomerates and minor metapelites metamorphosed and deformed during the Alpine orogeny. The rocks show weak to moderate schistosity. In thin section the texture is dominated by the superimposition of two main schistositities (S1 and S2, FRANCESCHELLI et al., 1986). Generally only S1 and subordinately S2 schistosity is associated with mineral crystallization.

The regional distribution of Al-silicates such as kaolinite, pyrophyllite and kyanite were taken as a basis for the metamorphic zonation of the Ver-

rucano rocks in the Northern Apennines. Four metamorphic zones have so far been distinguished: ka, (Perugia II boreholes), ka + prl + qtz transition (Monte Argentario, Monticiano-Roccastrada ridge and Monti Leoni pro-parte), prl + qtz (Monti Leoni, Monticiano-Roccastrada ridge, Monti Pisani and Punta Bianca areas) and ky + qtz zones (Massa area).

In these zones the pressures and the temperatures of metamorphism have been estimated by FRANCESCHELLI et al. (1986) as ranging from 3 to 5 kb and from 300° to 450°C respectively.

### Sample selection and analytical techniques

This study covers rocks characterized by a great variety of lithology and bulk chemistry in order to assess the variation of  $b_0$  of muscovite with mineral assemblages and metamorphic grade. The samples studied have been subdivided into three assemblage groups showing different degrees of Al-saturation. They are:

#### Al-rich:

pyrophyllite (or kyanite)-bearing assemblages  
 $ms + qtz + prl \pm su \pm cld$   
 $ms + qtz + ky \pm chl \pm pg \pm cld$

#### Al-intermediate:

pyrophyllite (or kyanite) and K-feldspar free-assemblages  
 $ms + qtz \pm chl \pm pg \pm ab \pm ctd$

#### Al-poor:

K-feldspar-bearing assemblages  
 $ms + qtz + k-fs \pm ab \pm chl$

Accessory minerals are magnetite, Ti-oxides, ilmenite, hematite, pyrite, zircon, epidote.

The d 060 spacing of muscovite was measured using a Philips PW 1793 diffractometer. The d 060 was determined on polished slabs of rock cut parallel to the cleavage using quartz as an internal standard, and  $b_0$  was calculated as  $6 \cdot d\ 060$  for Cu  $K_{\alpha}$  or  $K_{\alpha 1}$ . The standard deviation ( $1\sigma$ ) of a single d 060 measurement is estimated to be  $\pm 0.0005\ \text{\AA}$ .

Most of the muscovites examined show the d 060 reflection being strongly asymmetric or partially split into two peaks. The asymmetry or splitting degree was more pronounced in celadonite-rich muscovite. As shown in Fig. 2, when the Cu  $K_{\alpha}$  radiation is used the d 060 peak loses its partial splitting. This is the best evidence to demonstrate that the asymmetry or splitting of the 060 peak is due to the  $K_{\alpha 1}/K_{\alpha 2}$  doublet.

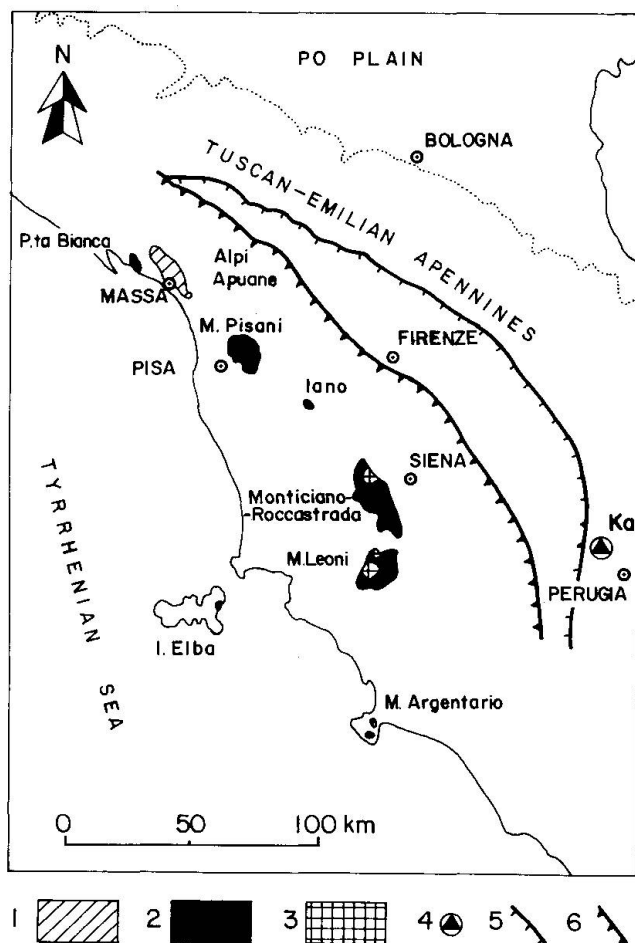


Fig. 1 Sketch map showing the metamorphic zonation of the Verrucano of the Northern Apennines and the geographical distribution of the localities sampled. (1) ky + qtz zone, (2) prl + qtz zone, (3) ka + prl + qtz zone, (4) ka + qtz zone boreholes, (5) Front of the Tuscan Nappe, (6) Front of the Cervarola flysch.

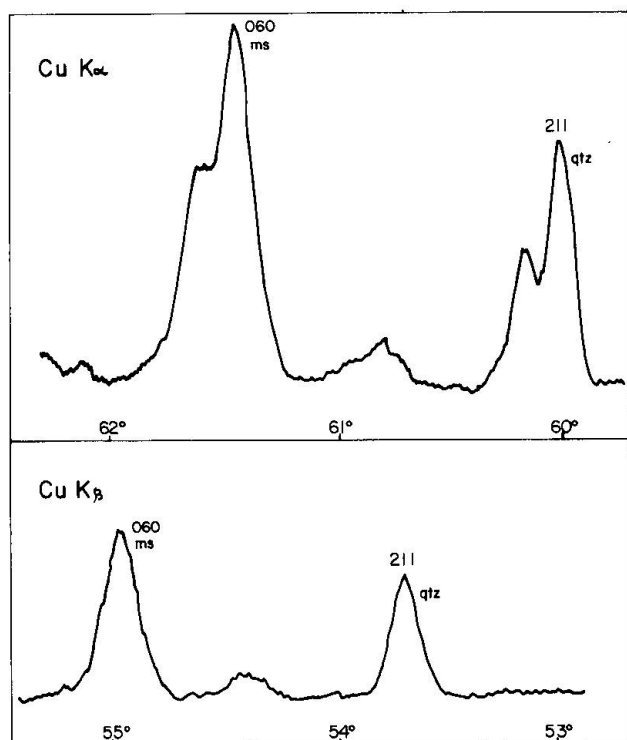


Fig. 2 XRD traces of Cu K $\alpha$  and Cu K $\beta$  of 060 peak of muscovite and 211 peak of quartz. See text for explanation.

### Analytical results

#### REGIONAL DISTRIBUTION OF $b_0$ VALUES

The regional distribution of the muscovite  $b_0$  values in the Verrucano rocks is shown in Fig. 3. The data are presented as a function of metamorphic grade.

For the pyrophyllite zone histograms representative for Monti Leoni, Monticiano-Roccastrada, Iano, Monti Pisani and Punta Bianca outcrops are also given. In all the histograms a further subdivision was made according to the three groups previously defined: pyrophyllite-bearing, pyrophyllite and K-feldspar free and K-feldspar bearing assemblages.

In the ka + prl + qtz transition zone (Monte Argentario area) the muscovite  $b_0$  values range from 8.990 to 9.045 Å with two maxima value densities at 9.005 and 9.040 Å (BERTAGNINI and FRANCESCHELLI, 1982).

The muscovite  $b_0$  spacing in samples of the prl + qtz zone range from 8.995 to 9.061 Å, with three maxima value densities around 9.008, 9.025 and 9.055 Å. Within this zone the range of  $b_0$  values of each outcrop is as follows: Monti Leoni 9.000 to 9.045 Å, Monticiano-Roccastrada area 8.990 to

9.050 Å, Punta Bianca 9.005 to 9.045 Å, Monti Pisani 8.995 to 9.065 Å, Iano 8.990 to 9.040 Å.

In the Verrucano rocks of the ky + qtz zone (Massa Unit) the muscovite  $b_0$  values range from 8.995 to 9.050 Å with a maximum density value close to 9.005 Å.

In each metamorphic zone or outcrop a close relationship between the  $b_0$  values and assemblage type was found. The  $b_0$  values in the pyrophyllite or kyanite-bearing samples range from 8.990 to 9.010 Å and no significant variations of the mean values in the three metamorphic zones seem to exist. In K-feldspar-bearing samples the values range from 9.045 to 9.065 Å and from 9.035 to 9.050 Å in the prl + qtz (Monti Pisani) and ky + qtz (Massa Unit) zones respectively.

K-feldspar bearing rocks, in the ky + qtz zone, occur in the Paleozoic sequence of the Massa Unit. However during the Alpine orogeny these rocks experienced the same metamorphic history as the Triassic Verrucano sequence.

Furthermore we observed that  $b_0$  values from pyrophyllite and K-feldspar-bearing assemblages never overlap and that muscovites from pyrophyllite and K-feldspar-free assemblages shows widely varying intermediate  $b_0$  values.

#### $b_0$ AND R.M. RELATIONSHIP IN MUSCOVITE

The  $b_0$  and R.M. (R.M. =  $\text{MgO}/\text{M.W.} + \text{FeO}_{\text{tot}}/\text{M.W.}$ ; M.W. = molecular weight) relationship for the Verrucano muscovites is shown in Fig. 4. The R.M. values were calculated assuming all the iron to be in the divalent state.

Table 1 gives the  $\text{MgO}/\text{M.W.}$ ,  $\text{FeO}/\text{M.W.}$ , 2  $\text{Fe}_2\text{O}_3/\text{M.W.}$ , R.M. and  $b_0$  values of some selected Verrucano muscovites from M. Pisani and Monticiano-Roccastrada ridge. Chemistry of these muscovite will be discussed in detail in an other paper (BALDELLI et al. in preparation).

On the assumption of a linear dependence of  $b_0$  on R.M., the following equation was computed by a least square method:  $b_0 = 8.982 + 0.499 \text{ R.M.}$  with a linear correlation coefficient  $r = 0.88$ . FREY et al. (1983) found a similar relationship directly relating the d (060+331) spacing to the R.M. values ( $d [060+331] = 1.4976 + 0.0819 \text{ R.M.}$ ). Expressing the R.M. as a function of  $b_0$ , FREY's equation transforms into  $b_0 = 8.986 + 0.491 \text{ R.M.}$  The equation relating the  $b_0$  to R.M. (defined as  $[\text{MgO}/\text{M.W.} + \text{FeO}/\text{M.W.} + 2 \text{FeO}/\text{M.W.}]$  by CIPRIANI et al. (1968) was:  $b_0 = 8.990 + 0.327 \text{ R.M.}$  The slight differences between CIPRIANI's, FREY's and our own equations may be due to various factors.

Apart from the different method of calculating the R.M. values, it is well known that the  $b_0$  musco-

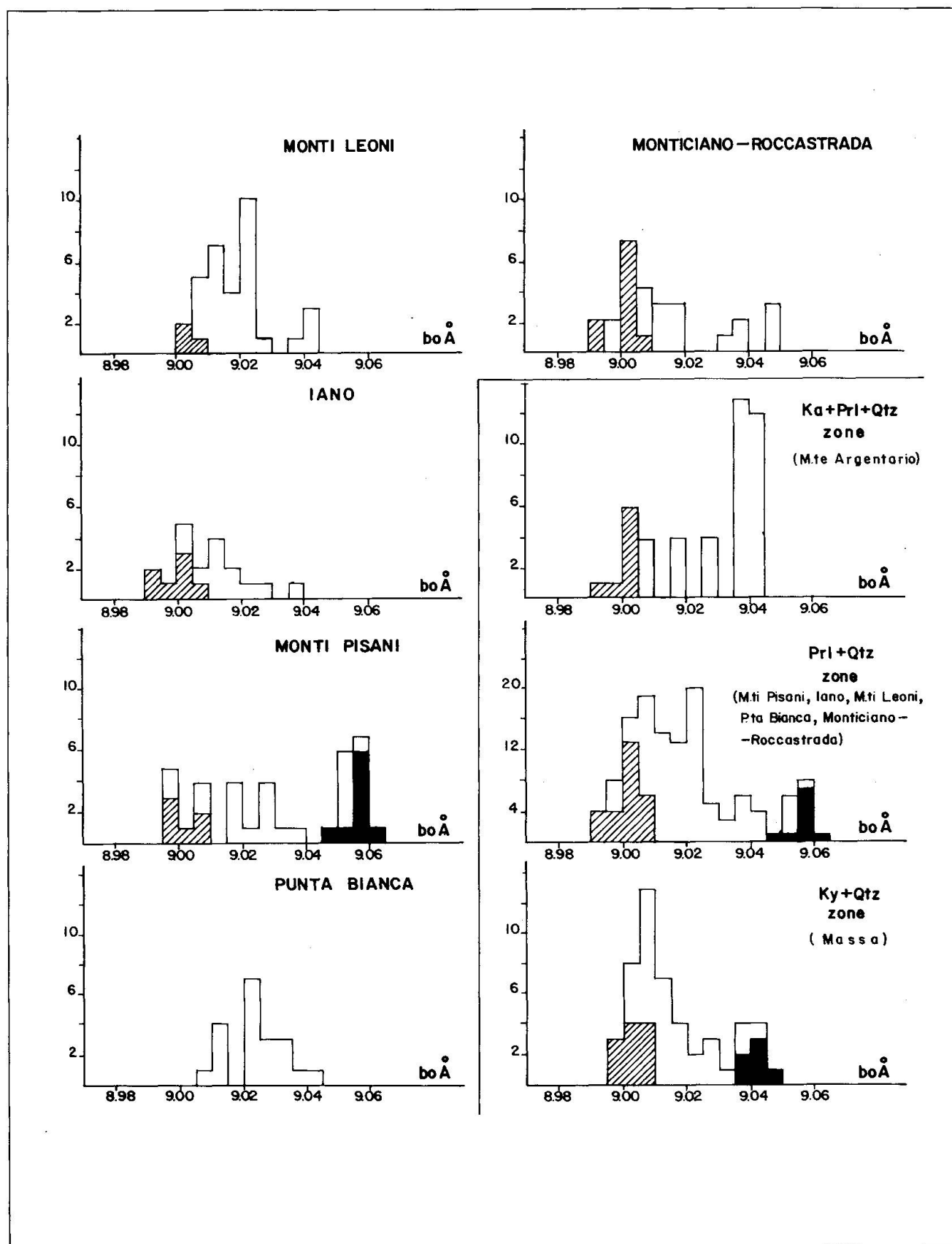


Fig. 3 Histograms showing the regional distribution of muscovite  $b_0$  values in the Verrucano of the Northern Apennines; hatched: pyrophyllite (or kyanite)-bearing samples; white: pyrophyllite (or kyanite) and K-feldspar-free samples; black: K-feldspar-bearing samples. On the right the  $b_0$  spacing of muscovite are arranged according to the metamorphic grade.

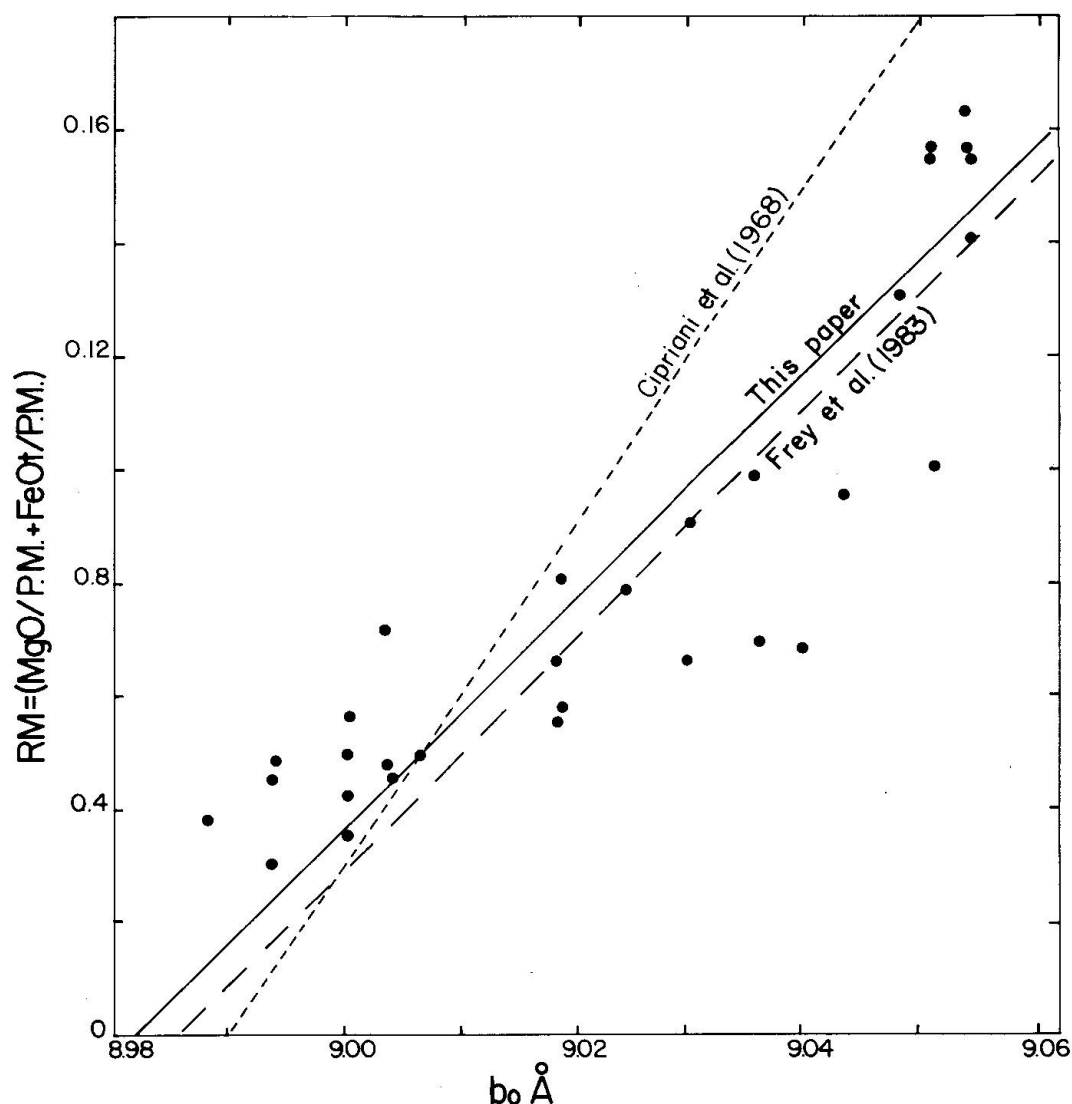


Fig. 4 Correlation of  $b_0$  and R.M. Dotted and broken lines represent the equation of CIPRIANI et al. (1968), FREY et al. (1983) respectively. The best fit line to 32 data of Verrucano muscovite is also shown.

vite parameter is not only  $(\text{Mg} + \text{Fe}_{\text{tot}})$  dependent, but is also somewhat influenced by the Na content (CIPRIANI et al., 1968) and by the ordering status (VELDE, 1980). Besides, despite their different ionic radius, usually no distinction among Mg,  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  cations is made.

For twelve muscovites whose analyses include  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  determinations (Tab. 1) (BALDELLI et al., in prep.) the contribution of the  $(\text{Mg} + \text{Fe}^{2+})$  (celadonite) and  $\text{Fe}^{3+}$  (ferrimuscovite) contents were separately evaluated. The linear equation computed by a least square fitting method was:  $b_0 = 8.982 + 0.548 (\text{Mg} + \text{Fe}^{2+}) + 0.522 (\text{Fe}^{3+})$ . This regression has been calculated assuming only  $(\text{Mg} + \text{Fe}^{2+})$  and  $\text{Fe}^{3+}$  as variables; 8.982 is the value resulting from the previous equation. This equation, compared with the one previously reported,

seems to suggest that, in the range of composition considered here (see Tab. 1), the celadonite and ferrimuscovite components similarly influence the  $b_0$  parameter.

The collected data indicate that the  $b_0$  parameter of the K-white mica may be considered a good measure of the octahedral sheet composition (the R.M. value of CIPRIANI et al., 1968) for the Verrucano muscovites.

### Discussion

#### $b_0$ IN LOW AND HIGH VARIANCE ASSEMBLAGES

In Tab. 2 the mean  $b_0$  values and standard deviation of muscovite from low and high variance as-

Tab. 1  $b_0$ , MgO/M.W., FeO/M.W.,  $2\text{Fe}_2\text{O}_3$ /M.W. and R.M. values of some selected muscovites from Monti Pisani and Monticiano-Roccastrada ridge. Chemical data and mineral assemblages from BALDELLI et al. in prep.) and FRANCESCHELLI et al. (1986).

Samp.	$2\text{Fe}_2\text{O}_3/\text{P.M.}$ $\times 10^{-3}$	$\text{MgO}/\text{P.M.}$ $\times 10^{-3}$	$\text{FeO}/\text{P.M.}$ $\times 10^{-3}$	R.M. $\times 10^{-3}$	$b_0$ ( $\text{\AA}$ )	Mineral assemblages
1	6.2	36.0	15.3	57.5	8.998	ms+qtz+prl+su+pg?
2	20.2	31.7	10.2	62.1	9.000	ms+qtz+prl+cld
3	10.8	27.0	13.2	51.0	8.998	ms+qtz+prl+su
4	30.0	46.6	10.7	87.3	9.000	ms+qtz+prl
5	26.0	40.7	6.9	73.4	9.000	ms+qtz+su+pg?
6	30.2	47.6	14.3	92.1	9.030	ms+qtz+chl
7	21.0	44.1	4.1	69.2	9.040	ms+qtz+chl+ab
8	48.4	56.6	3.4	108.4	9.052	ms+qtz
9	82.4	70.9	4.2	157.5	9.054	ms+qtz+ab
10	72.0	72.7	15.1	159.8	9.054	ms+qtz+chl?+ab+k-fs
11	82.0	76.4	5.6	164.0	9.054	ms+qtz+chl+ab+k-fs
12	77.6	69.0	8.4	155.0	9.055	ms+qtz+ab+k-fs

semblages (in the KNaASH system) are given for each metamorphic zone of the Verrucano complex.

The mean  $b_0$  values of muscovite from high variance assemblages are 9.032, 9.022 and 9.015  $\text{\AA}$  for the Ka + qtz + prl, prl + qtz and Ky + qtz zones respectively. According to SASSI and SCOLARI (1974) these values are typical of a medium-pressure type metamorphism despite the fact that many samples lack the peculiarity required for a correct application of the  $b_0$  geobarometer (absence of albite, presence of magnetite and/or hematite, etc., according to GUIDOTTI and SASSI, 1976). For muscovites from high variance assemblages the most relevant features of the  $b_0$  data reported in Tab. 2 are the wide range of the mean  $b_0$  value variation and the great dispersion of values within the same zone or outcrop. In the pyrophyllite zone the mean  $b_0$  values of Al-intermediate samples ranges from 9.013  $\text{\AA}$  (Iano) to 9.018  $\text{\AA}$  (Monti Leoni), 9.020  $\text{\AA}$  (Monticiano-Roccastrada ridge), 9.024  $\text{\AA}$  (Punta Bianca), 9.040  $\text{\AA}$  (M. Pisani).

Such a variation cannot be explained by invoking different metamorphic conditions but rather by the wide range of muscovite composition. This in turn is dependent upon the bulk chemistry variation of the Verrucano metasediments. In such a case, in fact, the mean  $b_0$  value and the associated standard deviation for a given outcrop are largely determined by the bulk chemistry variations of a certain lithotype, by the representativity of the lithotype in the field and, finally, by the frequency of sampling.

In the Verrucano metasediments the mean muscovite  $b_0$  value in the Al-intermediate group, as shown in Fig. 3, is actually obtained by averaging values covering the entire  $b_0$  range from low to high pressure-type metamorphism (SASSI and SCOLARI, 1974).

On the contrary, when the influence of bulk chemistry is reduced (i.e. in low variance assemblages), the mean muscovite  $b_0$  values are very similar in all the Verrucano areas and their standard deviations are considerably smaller.



Tab. 2 Average  $b_0$  values of muscovite from Al-rich, Al-intermediate and Al-poor groups for Verrucano rocks as a function of metamorphic grade. Figures in brackets represent the estimated standard deviation in terms of least units cited for the value to their immediate left.

Type of assemblage	Key mineral	$b_0$ av. value	Metamorphic zone	locality
Al -rich	prl	9.001(8)	ka+prl+qtz.	Mt. Argentario
	prl	9.001(8)	prl+qtz	(M. Pisani, Monticiana-Roccastrada, Iano, Punta Bianca, Monti Leoni)
	ky	9.001(4)	ky+qtz	Massa
Al-intermed	-	9.032(14)	ka+prl+qtz	Mt. Argentario
	-	9.022(16)	prl+qtz	(M. Pisani, Monticiana-Roccastrada, Iano, Punta Bianca, Monti Leoni)
	-	9.015(11)	ky+qtz	Massa
Al-poor	k-fs	9.056(5)	prl+qtz	Monti Pisani
	k-fs	9.042(6)	ky +qtz	Massa

In the prl + qtz zone, where it is possible to compare data from different outcrops, the mean  $b_0$  values are substantially the same, about 9.002 Å for all the outcrops.

For the Al-poor low-variance assemblages,  $b_0$  values from different outcrops of the same metamorphic zone are not available. In the prl + qtz (Monti Pisani) and ky + qtz zone (Massa Unit) the mean  $b_0$  values of muscovites from Al-poor assemblages are 9.056(5) and 9.042(6) Å respectively.

In this case the slight decrease in the  $b_0$  values may be related to the increase in metamorphic grade. It is well known that an increase in temperature reduces the (Mg + Fe<sup>2+</sup>) solid solution field in muscovite (GUIDOTTI and SASSI, 1976).

#### INTERREGIONAL COMPARISON OF $b_0$ FROM LOW VARIANCE ASSEMBLAGE

It is interesting to analyze the muscovite  $b_0$  values from low variance assemblages in low, medium and high pressure metamorphism.

Unfortunately, few studies mention the assemblages present in samples for which muscovite  $b_0$  data are given. We have compiled (Tab. 3) some of the available data from Iberian Massif and Sierra Baza (Spain), Grande Kabylie (Algeria), Calabria, Northern Apennines and Ligurian Alps (Italy), Glarus Alps (Central Swiss Alps), Crete (Greece). From the data of Tab. 3 it can be observed that:

i) In Al-rich low-variance assemblages (pyrophyllite or kyanite-bearing rocks) the mean  $b_0$  values vary from 8.954(5) (Hercynian metamorphites of Iberian Massif, Spain) to 8.996(6) Å (Hercynian metamorphites of Calabria, Italy and Sierra Baza, Spain) in the low-pressure type metamorphism.

ii) In the medium pressure type metamorphism (Verrucano metamorphites of the Northern Apennines and anchimetamorphic black shales of Glarus Alps) the mean  $b_0$  values are 9.002(4) and 8.982(7) respectively. As commented by FREY, "The reason



Tab. 3 Comparison of  $b_0$  muscovite mean values from Al-rich low variance assemblages from several metamorphic areas with different P/T metamorphic facies series. Figures in brackets represent the estimated standard deviation in terms of least units cited for the value to their immediate left. N = number of analyzed samples.

locality	facies series	age of metamorphism	Key mineral	$b_0$	N	source
Iberian M.	low	Hercynian	prl	8.954(5)	30	BRIME (1985)
Iberian M.	low	Hercynian	prl	8.961(4)	10	BRIME (1985)
G.Kabylie	low	Hercynian	prl	8.981(8)	6	BOSIERE et al.(1982)
Calabria	low	Hercynian	prl	8.996(6)	57	COLONNA et al.(1979)
Sierra Baza	low-medium	Hercynian	prl	8.996(6)	28	GOMEZ-PUGNAIRE et al.
Glarus Alps	medium	Alpine	prl	8.982(7)	38	FREY (1978) and written comm.
Northern Apennines	medium	Alpine	prl	9.002(3)	43	this paper
Northern Apennines	medium	Alpine	ky	9.002(4)	11	this paper
Ligurian Alps	high	Alpine	prl	9.010(7)	4	this paper
Crete	high	alpine	prl	9.013(7)	2	this paper

for the rather low  $b_0$  values in shales from Glarus Alps could be due to the paragonite content in muscovite of these rocks".

iii) In the pyrophyllite-bearing rocks from high pressure type metamorphism of the Ligurian Alps and Crete the  $b_0$  mean value is 9.010(7) and 0.013(7) Å respectively. SEIDEL (1977) gives an average value of about 9.024 Å for Al-rich rocks (chloritoid schists) from Crete.

The data presented in Tab. 3 clearly show that the mean  $b_0$  values in the Al-low variance assemblages tend to increase hand in hand with the P/T regime of metamorphism.

One could derive the following relation between the  $b_0$  average values in Al-low variance assemblages and the character of the metamorphism:

$b_0 < 9.000 \text{ Å}$	low pressure
$9.000 < b_0 < 9.010 \text{ Å}$	medium pressure
$b_0 > 9.010$	high pressure

This is a first step towards the calibration of the  $b_0$  geobarometer scale in ms + prl (ky) + qtz (plus a Mg,Fe silicate) assemblages. Nevertheless they cannot yet be used as a reference scale; other data are necessary to control its general validity especially in high pressure rocks.

The data given here seem to encourage the study of  $b_0$  in low variance assemblages (either in Al-rich or Al-poor) as a workable qualitative geobarometer even if the  $b_0$  variations due to pressure in Al-rich low variance assemblages appear to be too modest and comparable with the analytical error.

### Conclusion

In the Verrucano metamorphites of the Apennines the mean  $b_0$  values increase in the following order: Al-rich, Al-intermediate, Al-poor assemblages. In both Al-rich and Al-poor low variance assemblages the muscovite  $b_0$  spacing forms two

distinct groups. In these assemblages the mean  $b_0$  values change only slightly in different outcrops and/or metamorphic zones of the Verrucano and the values are characterized by a small standard deviation. Muscovites from Al-intermediate high variance assemblages, on the other hand, show highly variable  $b_0$  values. The mean values in each outcrop or metamorphic zone have a high standard deviation.

The pattern of  $b_0$  variation in muscovites, as it results from this study, seems to suggest that the use of this parameter to evaluate the P/T gradient of metamorphism, referring to high variance assemblages only, requires a great deal of attention. From the data given here, it clearly emerges that the mean  $b_0$  values of muscovite from high variance assemblages may be very different even in rocks metamorphosed under the same P/T gradient, therefore any comparison among different areas may prove to be thwarting.

On the other hand, when low variance assemblages are considered the  $b_0$  method of estimating the P/T gradients in metamorphic rocks from low grade regions is more reliable.

### Acknowledgments

We wish to express our sincere gratitude to Prof. B. Goffé and to Prof. E. Seidel for having kindly supplied some samples from Ligurian Alps (Italy) and Crete (Greece).

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Manuscript received November 20, 1988; revised manuscript accepted February 3, 1989.