PERIODICO di MINERALOGIA established in 1930 An International Journal of MINERALOGY, CRYSTALLOGRAPHY, GEOCHEMISTRY, ORE DEPOSITS, PETROLOGY, VOLCANOLOGY and applied topics on Environment, Archaeometry and Cultural Heritage

Chemistry and mineralogy of early Mesozoic pelite layers from continental redbeds «Verrucano», Peloritani Mountains, Sicily, Italy

PAOLA DI LEO^{1*}, GIOVANNI MONGELLI², SALVATORE CRITELLI³, ROCCO LAVIANO⁴, Emilia Le Pera³ and Vincenzo Perrone⁵

 ¹ C.N.R. Istituto di Ricerca sulle Argille, Via S. Loja-Area Industriale, I-85050 Tito Scalo (PZ), Italy
² Centro di Geodinamica, Università della Basilicata, Via N. Sauro, I-85100 Potenza, Italy
³ C.N.R. Istituto di Ricerca per la Protezione Idrogeologica nell'Italia Meridionale ed insulare, Via Cavour, I-87030 Roges di Rende (CS), Italy
⁴ Dipartimento Geomineralogico, Università di Bari, Via E. Orabona, I-70124 Bari, Italy

⁵ Istituto di Geologia, Università di Urbino, Località Crocicchia, I-61029 Urbino (PS), Italy

Submitted, July 1999 - Accepted, February 2000

ABSTRACT. - Mineralogical and chemical data on eight pelites and two metapelites from the Upper Triassic to Lower Jurassic continental sedimentary sequences of the Peloritani Mountains, Sicily, are reported. In both sediments and metasediments studied, the mineralogical assemblage is dominated by illite that prevails over kaolinite and chlorite. Chemical data suggest that a significant diagenetic control on the distribution of clay minerals can be excluded. The overall mineralogical features suggest derivation from a source area mostly composed by low- and medium- grade metamorphic terrains. The higher kaolinite and hematite abundance in the metasediments with respect to pelites, suggests derivation from a more weathered source. Metapelites underwent condition of P-T typical of low-grade metamorphism (anchizone) as indicated by the occurrence of pyrophyllite, likely deriving form the reaction involving kaolinite and quartz.

* Corresponding author, E-mail: pdileo@ira.pz.cnr.it

- 2 = E-mail: mongelli@unibas.it
- 3 = E-mail: critelli@irpi.cs.cnr.it
- 3 = E-mail: lepera@irpi.cs.cnr.it
- 4 = E-mail: Rocco.Laviano@geomin.uniba.it
- 5 = E-mail: perrone@uniurb.it

RIASSUNTO. - Nel presente studio sono riportati i primi dati mineralogici e geochimici delle peliti e metapeliti appartenenti alle successioni continentali del Triassico superiore-Giurassico inferiore che caratterizzano le coperture alpine, talune epimetamorfiche, di alcune falde a basamento cristallino dei Monti Peloritani (Sicilia nordorientale). L'associazione mineralogica riconosciuta sia nelle peliti che nelle metapeliti è dominata dall'illite che prevale sulla caolinite e sulla clorite, quest'ultima presente in tracce. La composizione chimica nel suo complesso suggerisce l'assenza, nei sedimenti studiati, di rilevanti modificazioni diagenetiche. L'associazione mineralogica indica che tali sedimenti provengono da un'area sorgente prevalentemene composta da terreni metamorfici di basso e medio grado. Nelle metapeliti, la presenza in quantità più elevate di caolinite ed ematite suggerisce un'origine da una area sorgente che ha subito un più alto grado di weathering. Le condizioni di P e T che hanno interessato le peliti metamorfosate sono quelle tipiche del metamorfismo di basso grado (anchizona) come testimoniato anche dalla presenza di pirofillite.

KEY WORDS: Continental Redbeds; Early Mesozoic; Sicily; pelite and metapelite; low-grade metamorphism.

INTRODUCTION

The Peloritani Mountains, the south-western end of the Calabrian Arc, are formed by a stack of Alpine nappes thrusted, during the Miocene, onto Meso-Cenozoic terrains, sedimented in the Maghrebian Flysch Basin Domain (Bonardi *et al.*, 1976 and references therein). The tectonic boundary between these two different domains is recognizable in the so-called «Taormina line» (fig. 1).

Some Peloritanian nappes only consist of a pre-Alpine crystalline basement (Fondachelli and Aspromonte Units), others include also a Meso-Cenozoic sedimentary cover (Longi-Taormina, Mandanici and Alì Units).

In these sedimentary covers the oldest strata are composed of upper Triassic-lowermost

Jurassic (Baudelot et al., 1988) continental redbeds (conglomerate, sandstone and pelite; Colacicchi, 1960; Colacicchi and Filippello, 1966; Lentini, 1975; Lentini and Vezzani, 1975; Bonardi et al., 1976; Gelmini, 1985) showing the typical facies of the «Verrucano», which represents the base of the Alpine cycle in some domains of both northern Apennines and Alps (Rau and Tongiorgi, 1974; Cassinis et al., 1979; Martini et al., 1986). The Tuscan «Verrucano», Triassic in age, outcrops in discontinuous patches throughout the entire lenght of the northern Apennines. It experienced a wide range of P-T conditions, linked to the Alpine metamorphism, and its compositive features have been extensively studied (Franceschelli et al., 1986a, 1986b, 1989; Giorgetti et al., 1998). In the northern



Fig. 1 – Schematic geologic map of the northeastern Sicicly: 1) Plio-Quaternary; 2) Etna volcanics; 3) Lower Pliocene-Upper Tortonian deposits; 4) Floresta Calcarenite Fm. (Lower Miocene) 5) High-grade metamorphics of the Upper Peloritanian nappes (Aspromonte and Mela Units; Pre-Triassic); 6) Alì Unit (Cretaceous-Triassic); 7) Sedimentary cover of the Lower Peloritanian nappes (Lower Miocene - Upper Triassic); 8) Low-grade metamorphics of the Lower Peloritanian nappes (Mandanici, Fondachelli, Longi-Taormina; Carboniferous-Cambrian); 9) Maghrebian Flysh Basin Units (Monte Soro, Troina-Tusa and Nicosia; Lower Miocene - Upper Triassic); 10) Maghrebian External Units (Panormide and Imerese; Middle Miocene - Middle Triassic); 11) Stratigraphic contacts; 12) Tectonic contacts; 13) Taormina Line; 14) Sampling localities.

Apennines the «Verrucano» mineralogical assemblage is not necessarily in equilibrium as some facies are pre-metamorphic and others developed during the Alpine stage of metamorphism.

The continental redbeds characterizing the Peloritanian nappes have been studied and sampled in all outcrops of the Peloritani Mountains, along two WNW-ESE transects from the Tyrrhenian Sea to the Ionian Sea. The southern transect, from Sant'Agata di Militello to Taormina (fig. 1), corresponds with the unmetamorphosed outcrops of the Longi-Taormina Unit, whereas the northern transect, from Capo Calavà to Alì, includes few outcrops of redbeds, affected by low-grade metamorphism, whose tectonic appartenance is still a matter of debate.

In these two transects we sampled both unmetamorphosed and metamorphosed pelite strata for the compositional study. The present paper focuses on the mineralogical and chemical (major and trace elements) features of the pelite and metapelite layers and its P/T evolution.

GEOLOGICAL AND STRATIGRAPHIC SETTINGS

The Mesozoic to Tertiary sedimentary sequences of the Peloritani Mountains nappes rest unconformably on the pre-Alpine crystalline basement of the Longi-Taormina, Alì and Mandanici units. The basements of these units consist of low-grade metamorphics (phyllite, metarenite, metalimestone and metavolcanic rocks) of Paleozoic age (Majesté-Menjaulas *et al.*, 1986; Spalletta and Vai, 1989; Messina *et al.*, 1996).

The Longi-Taormina Nappe constitutes the lowermost thrust sheets in the Peloritanian nappe stack (fig. 1) and shows the most extensive and complete Mesozoic to Cenozoic sedimentary successions, which include:

a) continental redbeds conglomerate, sandstone and pelite («Verrucano»), having variable thickness from few meters to over 200 meters. The oldest strata are Upper Triassic fluvial redbeds, followed by a few meters of Hettangian greyish continental and coastal deposits (Baudelot *et al.*, 1988). These deposits gradually pass vertically into

b) shallow-water limestone and dolostone, about 300 m in thickness, having a Hettangian(?) to Carixian age (Taormina Formation; Rigo and Barbieri, 1959); this carbonate platform facies pass into

c) Pelagic limestone and grey marl 200-300 m in thickness and Carixian-Domerian in age, passing into Toarcian-Bajocian ammonite-rich red marls («Ammonitico rosso inferiore»; Lentini and Vezzani, 1975). This slope facies pass into deep-water radiolarian chert, and cherty limestone, followed by new strata of ammonite-rich marls («Ammonitico rosso superiore»; Lentini and Vezzani, 1975), about 100 m in thickness and Bajocian to Tithonian in age. These terrains are followed by about 100 m thick Tithonian-Neocomian limestone and marly limestone, and then by Cretaceous to Oligocenic red, green and grey marls (Militello Formation). The sedimentary succession is capped by the

d) Turbidite quartzofeldspathic sandstone (Frazzanò Formation) having a lower Miocene age (Aquitanian; de Capoa *et al.*, 1997).

In the Longi-Taormina thrust sheets the continental redbeds are particularly thick (over 200 m), whereas they are few meters in the other areas. The Longi section shows Upper Triassic lenticular conglomerate and sandstone strata, representing fluvial channel-fill, interbedded with thin layers of red clays. They evolve into lower Hettangian yellow-white sandstone, clays and dolomite, showing the transition from continental to marine deposits (figs. 2 and 3). The fluvial sandstones are quartzarenite to sublitharenite, with abundant quartz, low-grade metamorphic, felsic volcanic, quartzite and chert lithic fragments, and dominantly quartz cement. The transitional sandstones are quartzarenite having carbonate cement and few bioclasts. The most of the pelite samples used in this study is from the fluvial portions of this section (samples VP1 to VP8).

In the Montagnareale-Gioiosa Vecchia area



Fig. 2 – Longi section (loc. Bosco Sottano): the section includes the Upper Triassic fluvial redbeds (Lower «Verrucano»; IV), that pass into Hettangian marine sandstone and clay (Upper «Verrucano»; uV), and Hettangian carbonate platform facies (L).

redbed deposits are affected by intense deformation and low-grade metamorphism, being involved in local thrust sheets with phyllite marble and calc-shist of the Mandanici Nappe (fig. 4). Two samples (VSP9, VSP10) have been collected close to the thrust contact which may be observed about 1 km north of Sorrentini, along the road to Russo (fig. 5).



Fig. 3 – Photographs of the main «Verrucano» outcrops in the Longi area. a) Lenticular conglomerate bodies and interbedded pelite layers; b) and c) quartzarenite and conglomerate strata and interbedded pelite (filled squares and arrows indicate the sample VSP5, in b) and the samples VSP6 (left) and VSP7 (right) in c); d) contact between Verrucano (V) and marine limestones (Li) near Galati.

23



Fig. 4 – Schematic cross-section showing the structure of the Verrucano strata outcropping in the Montagnareale-Gioiosa Vecchia area (modified after Caliri *et al.*, 1993). 1) Quaternary; 2) «Verrucano» redbeds (conglomerate, sandstone and pelite); 3) phyllite and marble of the Mandanici Unit; 4) fault; 5) thrust.



Fig. 5 – Thrust contact between Verrucano (V) and Paleozoic phyllite (Mandanici Unit). Samples VSP9 and VSP10 were collected along this shear zone.

METHODS AND TECHNIQUES

The mineralogy of the $<2 \mu m$ grain-size fraction and bulk Verrucano samples has been obtained by XRD (Siemens D5000, CuK_a radiation, graphite secondary monochromator, sample spinner) and the semi-quantitative distribution of the mineralogical components has been evaluated according to Laviano (1986). To estimate the illite crystallinity of the metapelites, the material was crushed in a hand mortar and then transferred to a plastic container for ultrasonic treatment for 2-3 min. After settling, the suspension containing the <2µm grain-size fraction was decanted, pipetted and dried at room temperature on glass slides to produce a thin layer, higly oriented aggregate with a particle density of at least 3 mg/cm². The illite crystallinity index (IC), defined by the width of the first-order illite basal reflection at 10 Å at half-height above the background (Kübler, 1967), was measured on both air-dryed and ethylen-glycol solvated slides following analytical technique and grainsize fractions as specified by Kish (1983, 1990, 1991). The results were calibrated using

interlaboratory standards (Warr and Rice, 1994).

The contents of major elements and Ba, Rb, Sr, La, Ce, Y, Zr, Nb, Cr, V and Ni on bulk samples were obtained by XRF methods described in Franzini *et al.* (1972) and Leoni and Saitta (1976). The precision for major elements are better than 5% and for trace elements are better than 10%.

The micro-texture has been investigated on thin section of selected samples by a SEM (Cambridge Stereoscan S360) equipped with an (LINK AN10000) energy dispersive spectrometer (EDS). Samples were sputtercoated with gold and mounted on carbon stubs. The working voltage was 21 kV.

MINERALOGY AND CHEMISTRY

The pelitic bulk samples contain progressively decreasing amounts of minerals as follows (Table 1): clay minerals >> quartz > hematite \approx feldspars. The illite, which appears ubiquitous in thin section, is the most abundant clay mineral and it largely prevails on kaolinite

TABLE 1

	bulk samples								<2 μm			
	Σ Phy	Qz	Р	Hm	F1	Ca	Do	Gy	Ill	Ka	Р	Qz
pelites												
VSP1	76	16	-	1	1	-	-	-	99	1	-	-
VSP2	79	11	-	1	1	1	tr.	-	99	1	-	-
VSP3	80	12	-	1	-	-	-	-	98	2	-	-
VSP4	72	18	-	1	1	-	-	-	97	3	-	-
VSP5	77	14	-	1	1	-	-	-	94	6	-	-
VSP6	76	17	-	1	1	-	-	· _	95	5	-	-
VSP7	81	14	-	1	-	tr.	tr.	-	95	5	-	-
VSP8	79	16	-	1	-	-	-	-	94	6	-	-
metapel	ites											
VSP9	78	8	2	3	1	-	tr.	tr.	71	10	19	tr.
VSP10	67	12	2	2	-	-	-	tr.	76	11	14	tr.

Mineralogical composition (wt%) of bulk samples and of $<2 \mu m$ size fractions.

 Σ Phy = phyllosilicates; Qz = quartz; Fl = feldspars; Ka = kaolinite; Hm = hematite; Ca = calcite; Do = dolomite; Gy = gypsum; Ill = illite; Ka = kaolinite; P = pyrophyllite; tr = trace.





Fig. 6 - SEM photo of detrital La-, Ce-rich phosfate and EDS specrum showing semiquantitative chemical analysis.



Fig. 7 – Pelite representative example of X-Ray diffraction patterns using powder preparation from the whole rock and Cu-K_{α} radiation. *a*) Packed into a plastic holder (unoriented mount). *b*) Oriented slides prepared by settling lcc of clay-water suspension onto a glass slide. *c*) Glicolated oriented slides (60°C overnight). *d*) Heated (250°C) oriented slides. (m.l. = mixed layers, i/ms = illite/muscovite, k=kaolinite, qz = quartz, hem = hematite).

and chlorite. The SEM-EDX observations reveal the occurrence of detrital grains of Tioxides, zircon, ilmenite and euhedral LREE- phosphate (fig. 6). The <2 μ m grain-size fraction is composed by illite prevailing on kaolinite and negligible amounts of illite-smectite mixed layers as suggested by the occurrence of small shoulders of 10 Å peak, towards the low angle side of the XRD patterns (fig. 7).

In the bulk metapelites the mineralogical composition is similar to the pelites excluding higher amounts of hematite and the occurrence of pyrophyllite. The 10 Å phase flakes are fine grained and mostly occur with a preferred orientation parallel to the cleavage and show a IC of 0.42° $\Delta 2\Theta$ (fig. 8). Pyrophyllite, largely present in the <2 µm grain-size fraction, is distinguished by its basal reflection at 9.2 Å and a strong diffraction maximum of 3.06 Å (fig. 9).

The distribution of the major and traceelements in the bulk samples is reported in Table 2. Normalized to the Upper Continental Crust (UCC, Taylor and McLennan, 1985), the samples are enriched in high field strenght elements (HFSE: Ti, Zr, Y, La and Ce) and transition elements (Fe, Ni, V, and Cr). Some large ion lithophile elements (Ca, Na, Sr), Mg and Nb are depleted (figs. 10 and 11). The metapelites, relative to the pelites, have higher contents of Na, Cr, V, La and Ce and lower Ca, Mg and Ba abundances.

DISCUSSION

The presence in the Verrucano pelites of the iron oxide coupled to the enrichment in the transition elements Ni, V, Cr indicates they formed in an aggressive climate. However, the low hematite abundance (see Table 1) may suggest that the erosion rate was high. The higher K_2O contents of the Verrucano pelites relatively to the Post Archean Australian Shales (PAAS, Taylor and McLennan, 1985) is also consistent with a high erosion rate not allowing a more evolved chemical and mineralogical stage. In addition, the values of the Al₂O₃/ K_2O ratio of these pelites are similar

TABLE 2

Elemental concentrations (wt% and ppm) in the «Verrucano» pelites and metapelites.

		Metapelites								
Samples	VSP1	VSP2	VSP3	VSP4	VSP5	VSP6	VSP7	VSP8	VSP9	VSP10
SiO ₂	60.14	56.44	62.62	62.90	60.60	62.24	61.85	62.26	52.17	58.23
TiO ₂	0.80	0.83	0.85	0.85	0.87	0.87	0.89	0.85	1.17	1.00
Al_2O_3	19.80	20.71	18.88	18.5	19.97	19.28	19.82	19.11	25.58	23.96
Fe ₂ O ₃	7.83	8.78	8.13	7.98	8.23	7.06	7.07	7.41	9.99	8.86
MnO	0.12	0.12	0.02	0.02	0.01	0.02	0.04	0.22	0.00	0.00
MgO	1.77	2.02	1.51	1.31	1.22	1.28	1.24	1.26	0.30	0.43
CaO	0.23	0.66	0.28	0.21	0.19	0.21	0.43	0.31	0.18	0.11
Na ₂ O	0.19	0.20	0.20	0.28	0.18	0.18	0.23	0.21	0.65	0.54
K ₂ O	4.94	5.26	4.49	4.13	4.50	4.57	4.08	4.20	4.30	3.71
P_2O_5	0.04	0.05	0.08	0.11	0.07	0.08	0.05	0.03	0.08	0.06
LOI	4.14	4.93	3.94	3.71	4.16	4.22	4.31	4.14	5.58	4.12
Ba	526	684	634	645	681	544	670	673	377	406
Rb	178	244	204	178	212	184	192	196	261	180
Sr	90	240	122	117	158	148	108	97	222	155
La	43	55	49	47	50	50	55	52	74	59
Ce	85	95	93	85	92	92	94	81	121	90
Y	42	49	44	46	46	48	47	40	49	40
Zr	258	166	279	255	251	289	263	233	259	241
Nb	17	14	18	17	16	18	19	22	22	18
Cr	80	97	86	75	92	81	89	93	143	110
V	118	132	122	110	132	120	125	132	168	134
Ni	30	38	40	33	37	55	41	40	27	31



Fig. 8 – SEM photo showing microtexture of metapelite (V10). Rounded grains are quartz; flakes oriented parallel to the cleavage direction are illite.

to the Al₂O₃/K₂O ratio observed in the PAAS (fig. 12), suggesting that illite, the dominant Kbearing phase, is mostly detrital and excluding a significant K uptake during diagenesis. The small difference in Al₂O₃/K₂O ratio with respect to PAAS may be due to uptake of some K⁺ during diagenesis as suggested by the presence of small amount of I/S mixed layers (80%; R=3 ordering). The IC values observed for the «Verrucano» sediments, in the range of $0.35-0.44^{\circ} \Delta 2\Theta$ (high diagenetic to low anchizone; see fig. 13) reflect both the P/T conditions of the source rocks and the effects of burial diagenesis in developing larger illite crystals. The limiting values for the anchizone condition are indeed 0.42 and $0.25^{\circ} \Delta 2\Theta$ (Kübler, 1967; Warr and Rice, 1994): higher values indicate diagenetic conditions, while lower values indicate epizone conditions. The high ordering of the mixed layers (R=3) and the high % of illitic layers in I/S mixed layers (80%) are also consistent with high diagenetic conditions. Thus, the overall mineralogical nature of the Verrucano pelites, dominated by illite, and the low abundances of quartz and feldspars suggest derivation from a source mostly composed by low- and medium-grade metamorphic terrains which are abundantly exposed in the Hercynian basement age (Majesté-Menjaulas *et al.*, 1986; Spalletta and Vai, 1989; Messina *et al.*, 1996).

In the metapelites the Al_2O_3/K_2O ratio is higher than in pelites (see fig. 12) as a consequence of higher kaolinite abundance. Micromorphological evidences indicate that this mineral is mostly detrital (fig. 14). Kaolinite

29



Fig. 9 – Metapelite representative example of X-Ray diffraction patterns using powder preparation from the whole rock and Cu-K_{α} radiation. *a*) Packed into a plastic holder (unoriented mount). *b*) Oriented slides prepared by settling lcc of clay-water suspension onto a glass slide. *c*) Glicolated oriented slides (60°C overnight). *d*) Heated (250°C) oriented slides. (m.l. = mixed layers, i/ms = illite/muscovite, p = pyrophyllite, k= kaolinite, qtz = quartz, kfs = k-feldspar, pl = plagioclase, hem = hematite).











Fig. 12 – K_2O and Al_2O_3 in pelites and metapelites from continental sequence of the Peloritani Mountains, Sicily. The K_2O/Al_2O_3 ratio of the pelites is similar to the Post Archean Average Shales (PAAS) whereas the metapelites exhibit lower ratios with respect to PAAS.



Fig. 13 – Illite crystallinity values. The limits of the lowgrade metamorphic zone are those of Kübler (1984).

distribution in sediments is also controlled by the intensity of the continental hydrolvsis (Chalmey, 1989) therefore, the abundance of kaolinite in Verrucano metapelites, coupled to higher hematite contents, is consistent with a derivation of such sediments from a more weathered source. The presence of pyrophyllite in the metapelites samples (see Table 1) suggests these sediments experienced condition of P-T typical of low-grade metamorphism (Winkler, 1979). This is consistent with the IC values observed for the metapelites ($\approx 0.30^{\circ}$ $\Delta 2\Theta$; see fig. 13). Pyrophyllite is often found in the clay-mineral assemblage in acid hydrothermal alteration system (Inoue, 1995) but it also a common low-grade metamorphic mineral (Winkler, 1979; Frey, 1987). Pyrophyllite is indeed an index mineral for the anchizone and low-grade epizone in very lowgrade metamorphic rocks with a pelitic composition: in aluminous, Fe-poor pelitic rocks, containing quartz pyrophyllite originates from the reaction (Frey, 1987)

$Al_2Si_2O_5(OH)_4 + 2SiO_2 \Leftrightarrow Al_2Si_4O_{10}(OH)_2 + H_2O$

This has been mapped, as a reaction isograd, in parts of the Swiss Alps (Frey, 1987) and in Verrucano metasediments from the northern Apennines (Italy), and in term of metapelitic zone has been located approximately at the late diagenetic-low anchizone transition (Wang *et al.*, 1996). Based on vitrinite reflectance and fluid inclusion data, the metamorphic condition

of the kaolinite-pyrophyllite isograd was estimated to about 200°C and 2.1 kbar, at $a_{\rm HO}$ of 0.1-0.2, and in the range of 240-260 °C, at $a_{\rm H,O}$ of ~ 0.8 and 1 kbar. The «cristallinity» values of illite from metapelites (on both normal and glycolated samples), the lowest observed among the whole analysed samples ($\approx 0.30^{\circ} \Delta 2\Theta$; see fig. 13), suggests metamorphic condition typical of the anchizone (temperatures are in the range 200-300°C; Jaboyedoff and Thélin, 1996 and references therein). At these temperatures the pyrophyllite is stabilised at the expence of kaolinite and quartz (Frey, 1987). Althought the temperature remains the principal physical factor determining illite crystallinity in some areas this parameter can be controlled by tectonic events (Frey, 1987; Fernandez-Caliani and Galan, 1992). The intense deformation occurred in the zone where metapelite were sampled - the Verrucano from Montagnareale-Gioiosa Vecchia area is involved in local thrust sheets – seems to be the main factor controlling the IC of metapelites, independently of the lithology and burial depth of the rocks. The microfabric, as represented by the stage of the cleavage development, is indeed representative of a shear zone.

CONCLUSIONS

Mineralogical and chemical data on pelites and metapelites from the «Verrucano» from Sicily are reported for the first time.

In both sediments and metasediments studied, the mineralogical assemblage is dominated by illite which prevails on kaolinite and chlorite. Chemical data suggest that a significant diagenetic control on the distribution of clay minerals can be excluded. The overall mineralogical features suggest derivation from a source mostly composed by low- and medium-grade metamorphic terrains characterized by a high erosion rate. Higher kaolinite and hematite abundance in the metasediments with respect to pelites, suggests derivation from a more weathered source.



Fig. 14 – SEM photo showing kaolinite crystals and EDS spectrum. The irregular shape of kaolinite crystals suggests a detrital origin. White arrow indicate spot position.

Metapelites experienced condition of P-T typical of low-grade metamorphism (anchizone) as indicated by the occurrence of pyrophyllite likely deriving form the reaction involving kaolinite and quartz. These sediments may be compared, from a mineralogical point of view, to the Verrucano metasedimets from the northern Apennines (areas of Monte Argentario, Monti Leoni and Monticiano Roccasarda; west of Siena) belonging to the kaolinite-pyrophyllite metamorphic zone of Franceschelli *et al.* (1986) characterized by the coexisting kaolinite and pyrophyllite.

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