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ORIGINAL PAPER

EVALUATION OF NUMIDIAN QUARTZ SANDSTONES FROM SOUTHERN ITALY FOR INDUSTRIAL APPLICATIONS

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ABSTRACT

Numidian quartz-rich sandstones collected from four different reconstructed sections along the Apennine chain of Southern Italy, have been studied for their textural, petrographic and geochemical characteristics to verify their potential industrial use. Sampling was carried out along a NW-SE transect: Monteverde and Aquilonia sections at NW (Campania Region) and Valsinni and Rotondella ones at SE (Lucania Region). The sandstones from NW successions are subarkoses with subordinate sublitharenites. High compositional maturity is shown by these sandstones containing high SiO_2 values (93wt% in average) and lowest amounts of CaO (~0.08 wt%), MgO (~0.1wt%), Fe_2O_3 (~0.9 wt%) due to the absence of carbonate components (cement and lithics) and scarce presence of clay-ferruginous cement. The sandstones from SE successions show lower contents of SiO_2 (87 wt% in average) and relatively higher contents of CaO (up to 7.4 wt%), MgO (up to 1.9 wt%) and Fe_2O_3 (up to 4.6 wt%) as compared to the previous ones, connected to the presence of calcite and clay-ferruginous cements; these can be classified as Fe-sands with subordinate sublitharenites. The sandstones from NW area show a better potential vocation to be used in the glass industry. However, the coloring elements contents are too high for clear or optical glass production. They are it is suitable as raw material for colored glass, insulating fibres, foundry molds or air Crete. With the aim of assessing the possible improvement of chemical characters, four granulometric fractions (A< 63 µm, B 63-125 µm, C 125-250 µm and D >250µm) were analyzed considering four samples from NW successions. Based on chemical composition, C and D fractions from Aquilonia area, are the most suitable for a coloured glass, while fractions A (<63 µm) and B (63-125 µm) mixed in proportions of 40 % (A) and 60 % (B), appropriately cleaned of clays, heavy minerals and iron hydroxides, could be used as raw material for the paint industry. The obtained results suggest a complete exploitation of this type of sandstones in the manufacturing industry, reducing waste materials and enhancing the economic value of these resources from southern regions of Italy.

1. INTRODUCTION

Silica sand represents the major constituent among natural raw materials, required for glass making or in different industrial fields as foundry and ferro-silica alloy, ceramic, fertilizer, abrasive and cement (Okereafor et al., 2020; Osseni et al., 2018; Phani, 2014). Natural deposits derived from both loosely consolidated sands and from crushing weakly cemented sandstones are used. However, among the quartz-rich sand and sandstones, only some types can be used in the glass industry, after purification carried out by wet or dry treatments; mainly by sieving and washing processes (Bide et al., 2020). The adequasy of these deposits must be evaluated based upon their chemical and physical properties. The SiO_2 contents and the grain sizes are the first essential parameters to be assessed (Bide et al., 2020). In this study, the evaluation of quartzose sandstones as potential raw material for glass industry has been performed, and, the required features for this aim have been discussed. The first property of the silica sands required for silicate glass production is related to high proportion (> 95 %) of silica (Bide et al., 2020). SiO_2 is one of the most utilized glass-former, with boron oxide for borosilicate glasses and phosphoric oxide for phosphate glasses (Lopes and Shelby, 2005). Another essential oxide for glass production is the soda (Na_2O), the most common flux agent used to reduce at 1600 °C the extremely high processing temperature required to vitreous silica production (2000 °C). The ferric and chromic oxides as well as heavy minerals (ilmenite, zircon, etc.) and ultra-stable minerals at high temperature (olivine, pyroxene, sillimanite, kyanite and garnet) are instead undesirable impurities which make the glass of poor quality (e.g. Amjad, 1998). In particular, the ferric oxides are critical at low concentrations (<0.3 %wt) in the raw material, causing a green coloration of the artifacts (Burkowicz et al., 2020; Osseni et al., 2018). Consequently, they must be kept within specific limits in the production of clear and colorless glass. Physical characters required to optimize the machining process are represented by the

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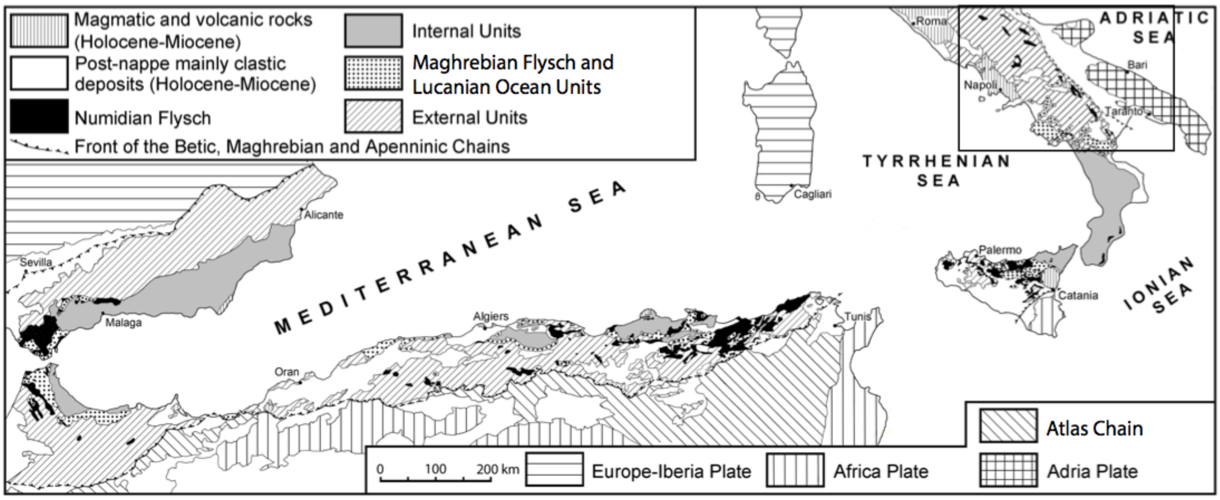

Fig. 1 Schematic geological map of Numidian Flysch outcropping in Western-Central Mediterranean area (modified from Hoyez (1989)). The box on the right indicates the studied area showed in Figure 2.

grain-size range of quartz sands that should be ranged between 0.1 and 0.5 or 0.71 mm in diameter together with a high degree of roundness of the quartz grains (Bide et al., 2020; Burkowicz et al., 2020; Osseni et al., 2018; Bloodworth, 2009). To obtain a high-quality glass silica sand, a set of processes must be employed, including most commonly screening to remove oversize, washing to remove the fine fraction (<0.1 mm) and sieving to produce appropriate sand grain ranges. The production of the raw materials with high quality standards requires further physical and/or chemical processing, such as gravity separation (spiral classifiers), froth flotation, high-intensity magnetic separation and acid leaching utilizing sulfuric acid (Bide et al., 2020). Finding suitable raw materials for glass production is therefore an important goal for country's industry because reduces production costs and influences employment levels.

In the Mediterranean area, quartz-rich sand and sandstones are widespread from North Africa, Spain, Southern France and Southern Italy (D'Errico et al., 2014). Impressive sequences of quartz-rich sandstones are found in Southern Italy and their chemical, petrographic and physical characteristics are comparable to those of rock sequences widespread in the Mediterranean area considered to belong to Miocene Numidian Flysch Formation (Fornelli et al., 2019, 2015).

The aim of present study is to assess the textural, petrographic and chemical features of Numidian quartz-rich sandstones from four different sites along Southern Apennines to evaluate their potential for the use in different productive processes, in particular for glass industry.

2. GEOLOGICAL BACKGROUND

The Numidian sandstones represent the most widespread sedimentary events in the Peri-Mediterranean Chains. They typified the Early-Middle Miocene depositional framework of the circum-Mediterranean foreland basin systems from Southern Spain (Betics), Northern Africa (Rif-Tell Chain), Sicily to Southern Apennines (Italy) (Fig. 1). The age of deposition changes from Early Miocene in the Betics, North Africa and Sicily to Early Langhian in the Southern Apennines (Wezel, 1967; Guerrera et al., 2005; Lentini et al., 2002; de Capoa et al., 2003; Perrone et al., 2008; Riahi et al., 2010; Belayouni et al., 2013; D'Errico et al., 2014; Pinter et al., 2016). The Numidian sandstones are interspersed with kaolinitic clays and were deposited mainly in deep-marine depositional systems (Fornelli and Piccarreta, 1997; Fornelli et al., 1998, 2015, 2019; Critelli et al., 2017; Critelli, 2018). These deposits consist of sediments derived by gravity-flows and are characterized by peculiar ultra-mature quartz-arenites containing mainly well-rounded monocrystalline quartz grains. These textural and mineralogical features make these sandstones ideal for use as raw materials in several industrial processes.

In the Southern Apennines, the Numidian Flysch (NF) sequences crop out along the external wedge of the Apennine Chain from Calabria-Lucania boundary to Molise (Fig. 2) and characterize the upper portion of the Cretaceous - Miocene sedimentary successions deposited in the Lagonegro-Molise Basin (Fig. 2).

The thickness of these sequences gradually decreases from south-east (600-1000 m) to north-west (few tens of meters) and, simultaneously, the sandstones become finer passing from quartz-arenites with scarce matrix and rounded grains to litharenites with abundant matrix and sub-angular – deformed quartz grains. Consequently, mineralogical and textural maturity decreases from south-east to north-west (D'Errico et al., 2014). The Numidian Flysch belongs to Sannio and Tufillo Serra Palazzo tectonic Units (Fig. 3) and crops out along the external wedge of the Apennine chain along a NW-SE transect from Lucania to Molise (Daunia) regions (Fig. 2). The quartz-rich sandstones of Numidian Flysch reach their

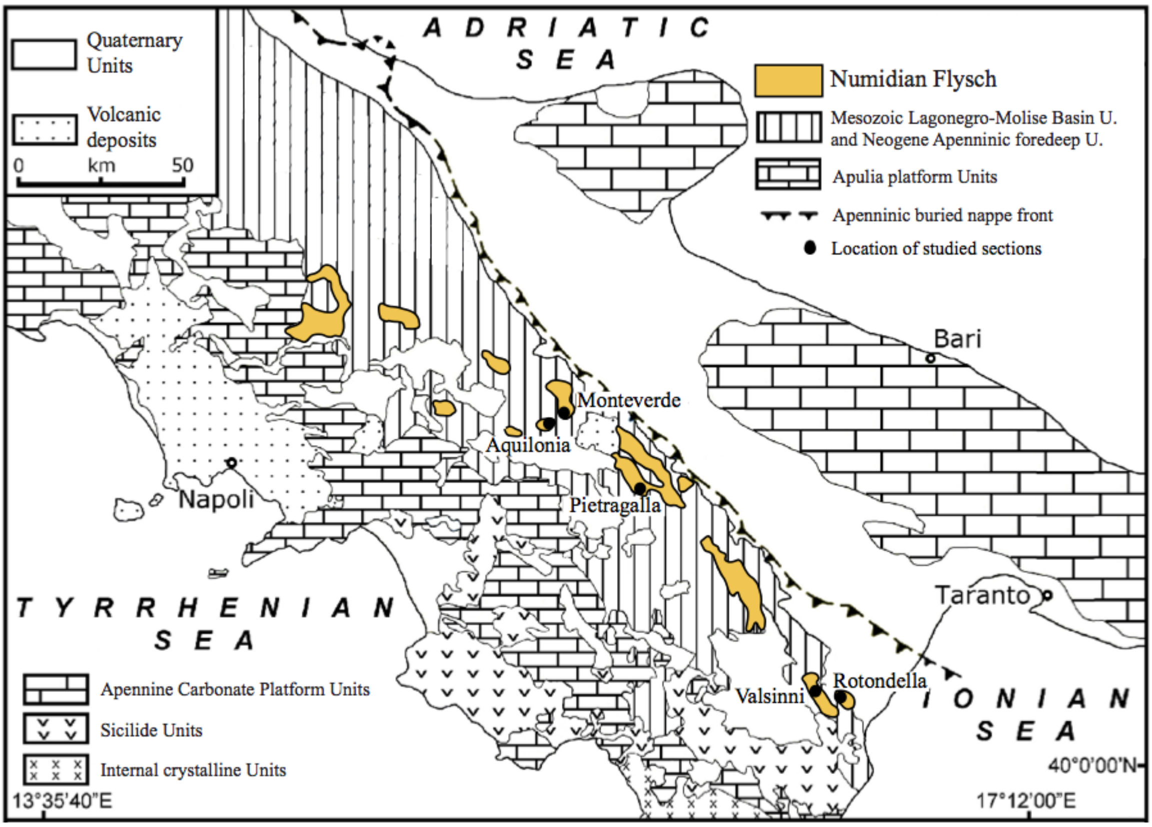

Fig. 2 Schematic geological map of Southern Apennines (Italy). Black circles represent the location of studied sections. Modified from D'Errico et al. (2014).

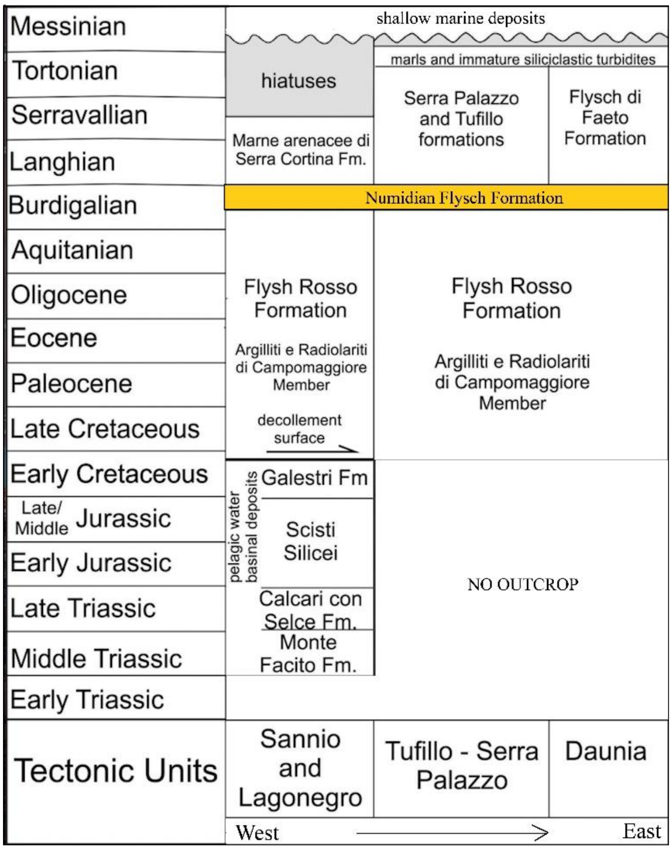

Fig. 3 Stratigraphic scheme up to Messinian of the tectonic units in which quartz-rich sandstones of Numidian Flysch (from west to east) occur in Southern Apennines (modified from Patacca and Scandone (2007) and Fornelli et al. (2019)).

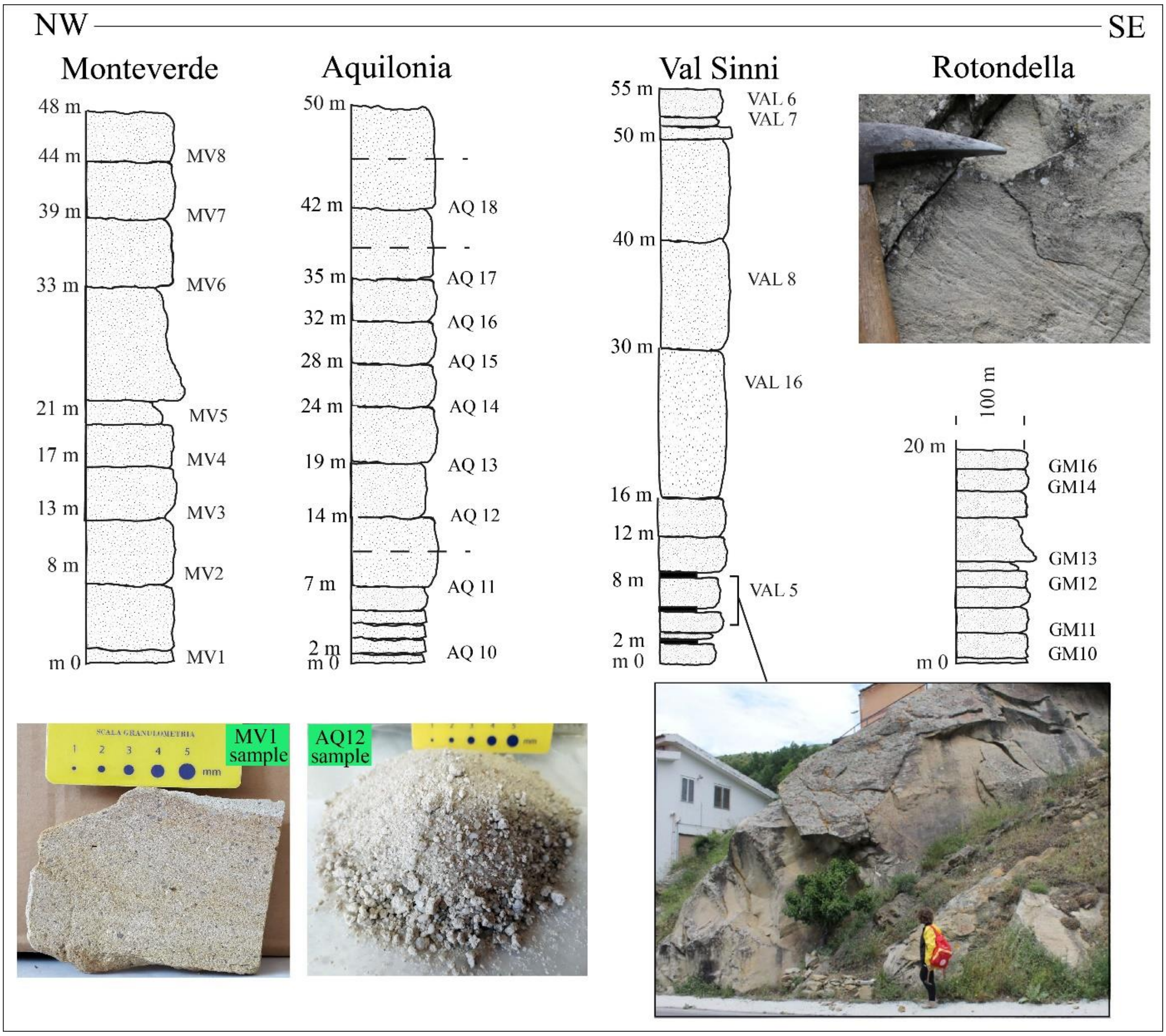

Fig. 4 Measured stratigraphic sections of Numidian Flysch sandstones in the studied sites with sample details.

maximum thickness (several hundreds of meters) and extension in the Sannio tectonic unit (Patacca and Scandone, 2007). These rest over the Cretaceous to Lower Miocene deep-sea successions of the Flysch Rosso Formation (Apat, 2007; Sabato et al., 2007) and passes upward to immature siliciclastic and/or calciclastic turbiditic successions of Langhian - Tortonian age (Fig. 3; Marne arenacee di Serra Cortina, Serra Palazzo Tufillo and Faeto formations) (e.g. Carbone et al., 1988; Pescatore et al., 1999; Gallicchio and Maiorano, 1999; Di Nocera et al., 2006; Pescatore et al., 2008; Di Nocera et al., 2011).

Four reconstructed sections of Numidian sandstones distributed along the NW-SE alignment (Fig. 2) were considered in this study: in the NW area, the Monteverde and Aquilonia successions (Campania Region); in SE area, the Valsinni and Rotondella ones (Lucania Region).

3. SAMPLING OF SANDSTONES

3.1. MONTEVERDE AND AQUILONIA SUCCESSIONS IN NW AREA

These successions crop out in the Campania Region (Fig. 2). The Monteverde section was sampled near the homonym village (Lat. N 40°59′37″, Long. E 15°31′50″).

In the field, the sandstones have yellow ochre colour and form amalgamated layers with a thickness ranging from 4-5 m up to 10 m (Fig. 4). These rocks show graded or massive structures with a grain size around 0.5-2.0 mm; however, coarse-grained sandstones are more common in the massive layers. Frequently, mud clasts are present in the fine-grained layers (0.5-1 mm). Eight samples (label MV) were collected along the section showing a thickness of about 48 m (Fig. 4).

The Aquilonia sequence was sampled near the Aquilonia village (Lat. N 40°99′40″, Long. E 15°49′15″) and nine samples (label AQ) were collected along a reconstructed section of about 50 m thick (Fig. 4). The sandstones show grey color and are

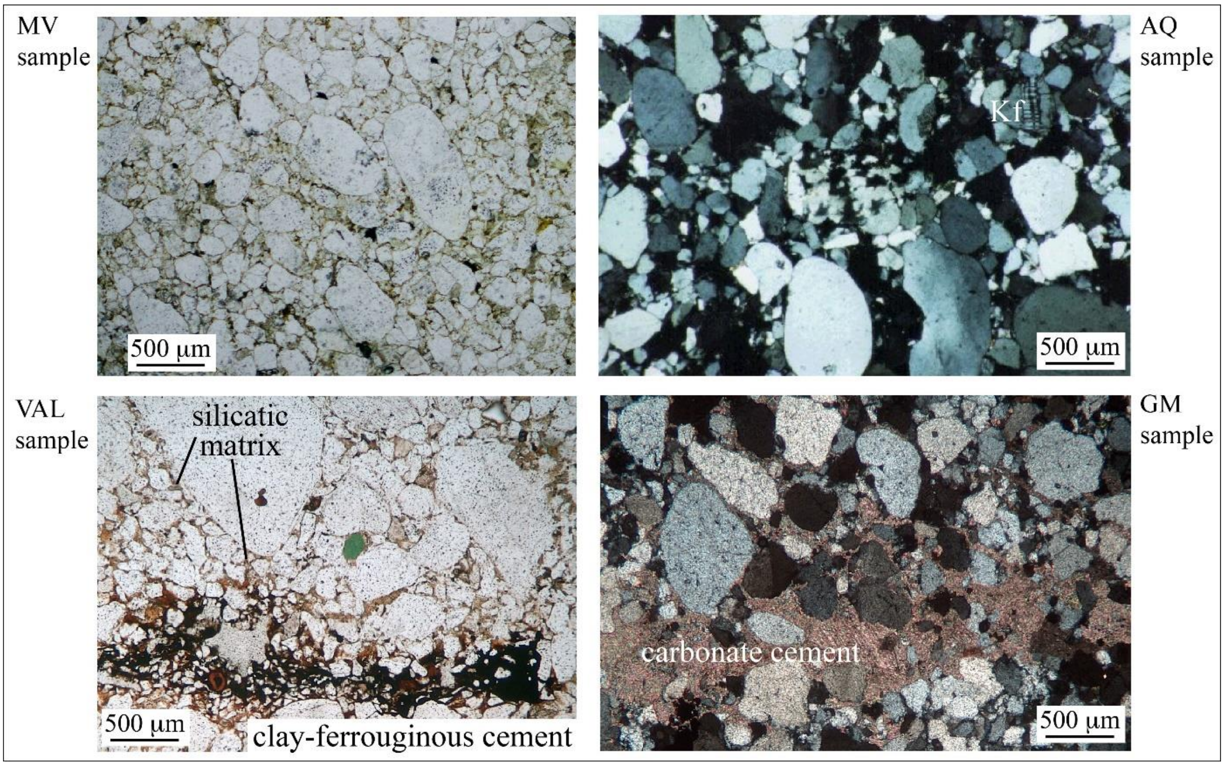

Fig. 5 Microphotographs of representative sandstones, under the polarizing microscope, from selected study areas: quartz grains coated by a very thin film of Fe-oxides in MV sample (plane-polarized light); the most abundant framework grain is the quartz as mono- or polycrystalline grains in AQ sample (Kf: potassic feldspar; cross-polarized light); clay-ferruginous cement in VAL sample (plane-polarized light); carbonate cement in GM sample (crossed-polarized light).

present both in graded and massive layers. The thickness of layers decreases from the bottom to the top of the sequence showing frequent amalgamation. The Aquilonia sandstones are less cemented and have a major degree of fracturing than those of Monteverde.

3.2. VALSINNI AND ROTONDELLA SUCCESSIONS IN SE AREA

These successions crop out in the Lucania Region. Near Colobraro and Valsinni villages (Fig. 2), the Numidian sandstones reaches a thickness of about 1000 m. The reconstructed Valsinni section (100 m thick) was sampled in the upper-medium portion of the entire succession (Lat. N 40°10’60”, Long. E 16°25’28”, label VAL). The grey-yellow sandstones outcrop as graded layers having a metric thickness and with intercalations of thin pelitic levels (Fig. 4).

The Rotondella deposits were sampled along the lower part of a continuous sequence of 100 m thick (Lat. N 40°10’05”, Long. E 16°31’11”, label GM) (Figs. 2, 4). These sandstones show the typical features of Numidian quartz-arenites: fine – to coarse grained, poor sorting with abundant matrix, rarely evident gradation and locally amalgamated beds.

4. ANALYTICAL METHODS

Modal analyses were performed under optical microscope on thirty samples of Numidian sandstones. Point counting was performed according to the Gazzi-Dickinson method (Ingersoll et al., 1984; Zuffa, 1985); 600 points were counted on each thin section distinguishing the different shape of quartz grains (angular or rounded). The proportion of quartz, feldspar and lithic grains for QFR diagram was obtained by recalculation to 100 % of collected data.

Chemical analyses were performed on thirty-one whole samples and sixteen granulometric fractions by X-ray fluorescence utilizing a Panalytical (Axios advanced) automatic spectrometer at Department of Earth Science and Geo-environmental, University of Bari Aldo Moro.

Grain size fractions were obtained by wet sieving using 63, 125 and 250 μm sieves, corresponding to 230, 120 and 60 mesh of ASTM series.

5. PETROGRAPHY

5.1. MONTEVERDE AND AQUILONIA SANDSTONES

The analysed arenites from Monteverde and Aquilonia sampling areas show similar texture and composition. Generally, these sandstones have a medium to a coarse - grained size and the most abundant framework grain is the quartz occurring both as single crystals or as polycrystalline grains (Fig. 5).

Table 1 Petrographic recalculated parameters from quartz-rich sandstones of the four selected Numidian successions.

SAMPLE	Q	F	R	Silicatic matrix	Clay-ferr Cement	Carbonate Cement	Q ang tot	Q rounded tot	Q ang/Q rounded to
MV1	96.47	3.13	0.4	25.02	8.98		25.59	36.93	0.69293
MV2	95.09	4.65	0.26	20.48	3.88		21.59	46.38	0.4655
MV3	92.34	7.66	0	23.51	4.14		28.33	33.49	0.84592
MV4	94	5.53	0.47	11.15	6.52		31.22	42.37	0.73684
MV5	90.6	9.4	0	18.69	6.54		22.14	32.12	0.68929
MV6	92.78	7.22	0	24.28	0.77		24.09	33.72	0.71441
MV7	94.55	4.97	0.48	20.88	5.37		26.45	34.47	0.76733
MV8	93	5.75	1.25	23.06	5.4		32.42	19.63	1.6516
Mean MV (n=8)	**93.6**	**6.04**	**0.36**	**20.88**	**5.2**		**26.479**	**34.889**	**0.82**
AQ10	91.21	8.79	0	12.77	1.16		21.05	31.51	0.66804
AQ11	91.81	7.94	0.25	11.76	1.47		22.27	36.34	0.61282
AQ12	91.91	8.09	0	12.12	0		23.71	35.83	0.66174
AQ13	92.09	7.91	0	13.48	0.19		22.45	32.22	0.69677
AQ14	94.83	5.17	0	13.69	0.57		21.67	31.55	0.68685
AQ15	87.31	12.26	0.43	10.93	2.19		25.85	23.13	1.1176
AQ16	91.24	8.76	0	13.02	1.18		23.87	27.71	0.86142
AQ17	88.92	11.08	0	12.88	0.94		26.22	13.58	1.9308
AQ18	91.31	8.69	0	16.55	0		27.65	15.73	1.7578
Mean AQ (n=9)	**91.18**	**8.74**	**0.08**	**13.02**	**0.86**		**23.86**	**27.511**	**0.999**
VAL1	97.59	0.7	1.7	1.1	1.4	4.6	33.5	66.5	0.503
VAL3	95.8	2.5	1.7	6.3	1.3	7.9	47	53	0.886
VAL4	91.21	8.2	0.5	7.2	6.4	4.8	49.1	50.9	0.964
VAL5	90	8.8	1.1	6.4	7.4	0	42.5	57.5	0.739
VAL6	87.6	8.6	3.7	3	6.2	0	46.8	53.2	0.879
VAL7	95.7	3.31	0.1	0.9	2.5	0	30.4	69.6	0.436
VAL8	90.9	4.8	0.8	2.4	3.5	2.5	42	58	0.724
VAL16	94	4.4	1.7				40.8	59.2	0.689
Mean VAL (n=8)	**92.85**	**5.164**	**1.413**				**41.512**	**58.488**	**0.728**
GM1	98.3	0.2	1.5	3.3	2.5	3.8	59.31	40.67	1.45
GM7	99.3	0.5	0.2	0	0	5.9	44.84	55.16	0.81
GM11	99.8	0.2	0	13.2	0	0	56.64	43.4	1.3
GM13	99.3	0.2	0.5	0 0	11.7	11	45.52	54.47	0.83
GM14	99.8	0.2	0	0	0	7	54.06	45.9	1.17
Mean GM (n=5)	**99.3**	**0.26**	**0.44**	**3.3**	**2.84**	**5.54**	**52.074**	**47.92**	**1.112**

Most of the quartz grains are coated by a very thin film of Fe-oxides. K-feldspar, plagioclase, micas, and heavy minerals (zircon, tourmaline, garnet and monazite) are present as accessory minerals of the framework grains. Lithic fragments are also present in small quantities: slate, chert and phyllite as fine-grained, or aphanitic lithics, and coarser phaneritic rock fragments ascribed to granitoids clastic supply. The sandstones are poorly cemented, sometimes by a siliceous precipitate (chalcedony, ~2–8 %), more often by clay-ferruginous materials (Fe-clay minerals and Fe-hydroxide) present in a variable quantity up to 9 % (9-0.8 % in MV samples, mean=5.2 % and 2.2-0.2 % in AQ samples). Carbonate cement, as well as fossil fragments or carbonate lithic grains, are completely absent. The matrix is siliciclastic and its abundance ranges from 11 % to 25 % in the samples of Monteverde section and from 11 % to 17 % in the samples from Aquilonia (Fig. 5, Table 1).

The principal composition is $Q_{90-96}F_{3-9}R_{0-1}$ for Monteverde sandstones (n=8) and $Q_{87-95}F_{5-12}R_{0-0.4}$ for those from Aquilonia sites (n=9) (Table 1), respectively. In both sections the sandstones can be classified as quartzarenites except two samples (AQ15-AQ17) from Aquilonia classified as subarkoses (Fig. 6).

From the analysis of Qang/Qround ratio (Table 1), it emerges that rounded quartz grains (single or polycrystalline) generally prevail with respect to angular clasts regardless of their internal textures (isotropic or anisotropic). In MV and AQ sandstones, the Qang/Qround ratio is in average 0.82 and 0.99 respectively; however, this ratio is higher than 1 in about one third of the AQ samples.

5.2. VALSINNI AND ROTONDELLA SANDSTONES

The sandstones of Colobraro-Valsinni sections (VAL) show coarse- grain size; the mono- and polycrystalline quartz grains are prevalent with respect to small quantities of K-feldspar, plagioclase, micas,

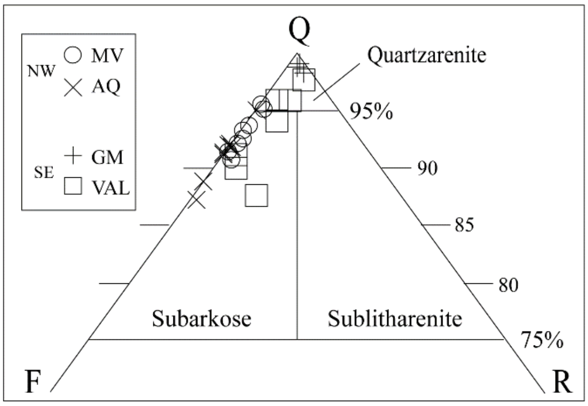

Fig. 6 QFR diagram (Folk, 1968) in which the principal composition of MV, AQ, VAL and GM samples is plotted.

opaques, glauconite and heavy minerals as zircon and tourmaline (Fig. 5). As shown in Table 1, matrix is siliciclastic in composition (1-7 %), whereas cement can be represented by carbonate (up to 8 %) and/or clayey-ferruginous precipitate (~1.5-7.5 %). The principal modal composition is $Q_{88-97}F_{1-9}R_{0-4}$ (n=8) evidencing a higher number of lithic grains (mainly phyllites) with respect to Monteverde and Aquilonia sandstones (Fig. 6, Table 1).

The sandstones can be classified as quartzarenites and subarkoses on the QFR diagram (Fig. 6). The shape of quartz grains is prevalently rounded as shown by Qang/Qround ratio comprised between 0.44 and 0.96 (0.73 in mean, Table 1). Among analysed samples a substantial difference is represented by the presence of carbonate cement, present only in some samples (around 2.5-8 %).

The Rotondella sandstones (GM) have a framework similar to that of other sandstones but they are characterized by a major amount of carbonate (4-11 %) and clayey-ferruginous (3-12 %) cement (Fig. 5). They are quartzarenites (Fig. 6) as the principal composition $Q_{98-99}F_{0.2-0.5}R_{0-1.5}$ indicates (n=5). The siliciclastic matrix reaches at most 13 %. The Qang/Qround ratio is in average 1.11 related to a higher quantity of angular grains (Table 1).

6. CHEMICAL COMPOSITION OF SANDSTONES

6.1. WHOLE ROCK GEOCHEMISTRY

Major and trace element composition was determined on thirty-one samples by X-Ray Fluorescence (Table 2).

Monteverde sandstones (MV, n=8) show high compositional maturity with high SiO_2 contents (92 wt% in average), related to the occurrence of abundant detrital quartz in the arenite framework. Y and Zr contents assume high values (in average 283 ppm) strongly depend on distribution of the accessory phases as zircon, more concentrate in the lower part of the section. In contrast, they show low contents of CaO (0.1 wt% in average), Sr (34 ppm), MgO (0.13 wt%), K_2O (around 1 wt%), Rb (26 ppm), and Fe_2O_3 (1.2 wt%) related to the scarce quantity of carbonate, clay-ferruginous components and micas.

Aquilonia sandstones (AQ, n=9) show high compositional maturity with high contents of SiO_2 (94 wt% in average), K_2O (around 1 wt%) and Rb (23 ppm). They show low contents of CaO (0.06 wt% in average), Sr (27 ppm), MgO (0.07 wt%) and Fe_2O_3 (0.7wt%) related to the total absence of carbonate component and very low clay-ferruginous cement. The distribution of Y and Zr is similar to that of Monteverde section with lower contents (Y+Zr around 230 ppm).

Valsinni sandstones (VAL, n=8) show high compositional maturity with high contents in SiO_2 (92 wt% in average). K_2O (around 0.4 wt%) and Rb (11 ppm) are very low related to small quantities of K-feldspar and micas. They show CaO contents around 0.7 wt% (in average), Sr about 35 ppm, MgO and Fe_2O_3 0.45 wt% and 1.38 wt% in average, respectively, connected to the occurrence of carbonate and clay-ferruginous cements, even the accessory phases are more abundant as indicated by higher contents of Y and Zr (in mean 326 ppm).

Rotondella sandstones (GM, n=6) show a minor compositional maturity with low SiO_2 amounts (83 wt% in average). K_2O (around 0.23 wt%) and Rb (8 ppm) show also low contents. They show high content of CaO (4.14 wt% in average, variable from 0.27 wt% up to 7.42 wt%) and Sr (133 ppm) due to the presence of abundant carbonate cement. They have high contents of MgO (0.87 wt%) and Fe_2O_3 (2.62 wt% in average, variable from 0.9 wt% to 4.6 wt%) related to widespread clay-ferruginous cement. The heavy minerals show a distribution similar to that of NW sandstones with Y and Zr varying around 263 ppm.

According to the geochemical classification of Herron (1988), the sandstones from NW area (Monteverde, Aquilonia) are subarkoses and sublitharenites (Fe_2O_3/K_2O ratio=0.89 in mean), whereas those from SE domains (Valsinni and Rotondella) plot mainly in the Fe-sand field (Fe_2O_3/K_2O ratio=7.51 in mean) (Fig. 7).

Chemical features of quartz-rich sandstones and in particular the high contents of SiO_2 and the scarce quantities of CaO seem to be promising for NW successions with respect to SE sandstones thinking about their use in the glass industry. To test in major detail their suitability, two samples from each section from NW area were crushed and sieved to obtain four different grain size fractions; each of these was weighed and analyzed for major and trace elements.

6.2. GEOCHEMISTRY OF SELECTED GRAIN SIZES

Four grain size fractions (A<63 μm, 63 μm< B < 125 μm, 125 μm < C < 250 μm and D >250 μm) were extracted from each selected sample: MV1 and MV7 from Monteverde section; AQ12 and AQ16 from the Aquilonia succession; the chemical composition and

Table 2 Chemical composition of Numidian sandstones from four selected successions (with calculated means). The mean of PQZ* samples (from Pietragalla) derives from Fornelli (1998).

Sample	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	P_2O_5	LOI	Rb	Sr	Y	Zr	Nb
MV1	90.58	0.29	4.54	1.43	0.18	0.13	0.36	1.02	0.02	1.43	25.7	37.1	7.1	300.2	4
MV2	92.49	0.32	3.88	0.92	0.1	0.1	0.34	0.99	0.01	0.85	24.2	35.3	8.4	439.5	2.9
MV3	91.78	0.29	4.05	1.11	0.09	0.08	0.38	1.13	0.01	1.08	23.9	34	5.2	280.1	2.3
MV4	87.57	0.4	6.05	2.35	0.34	0.18	0.45	1.39	0.02	1.24	32	46.80	15.8	321.1	3.6
MV5	93.13	0.23	3.61	0.82	0.12	0.09	0.36	0.99	0.01	0.64	23.9	35	3.4	246.5	2.3
MV6	94.24	0.21	3.13	0.71	0.03	0.04	0.28	0.88	0.01	0.47	21	24.4	2.8	203.9	2.5
MV7	92.77	0.22	3.86	0.8	0.05	0.06	0.35	1.08	0.01	0.82	26.6	32.8	3.4	215.2	2.4
MV8	91.51	0.23	4.27	1.19	0.16	0.08	0.36	1.06	0.01	1.13	27	29.2	3.5	208.3	2.3
Mean MV (n=8)	**91.76**	**0.27**	**4.17**	**1.17**	**0.13**	**0.095**	**0.36**	**1.07**	**0.013**	**0.96**	**25.54**	**34.33**	**6.2**	**277**	**2.79**
AQ10	93.21	0.21	3.55	0.64	0.13	0.1	0.27	0.82	0.01	1.05	21.8	21.9	1.6	191.	2.2
AQ11	94.27	0.26	3.04	0.7	0.04	0.04	0.25	0.71	0	0.68	21.9	20.6	3.2	318.2	2.3
AQ12	95.27	0.2	2.82	0.33	0.01	0.03	0.16	0.79	0	0.4	18.7	21.7	2.3	227.2	2.4
AQ13	94.64	0.23	3.05	0.44	0.03	0.03	0.19	0.95	0	0.44	21.7	23.9	3	264.5	1.4
AQ14	93.62	0.22	3.24	0.58	0.13	0.08	0.33	1	0	0.79	23.9	30.4	2.7	238.7	1.4
AQ15	92.34	0.26	3.97	0.86	0.08	0.06	0.39	1.26	0.01	0.76	28	33.3	3.6	243.9	1.7
AQ16	93.09	0.2	3.58	0.95	0.1	0.06	0.3	1.11	0.04	0.58	21.9	29.3	2.3	166.2	0.9
AQ17	93.47	0.2	3.26	0.73	0.06	0.04	0.32	1.07	0.01	0.85	23.3	29.9	2	175	1.2
AQ18	93.85	0.2	3.19	0.68	0.06	0.06	0.24	0.85	0.01	0.87	21.6	28.4	2.1	214	2.6
Mean AQ (n=9)	**93.75**	**0.22**	**3.3**	**0.66**	**0.07**	**0.06**	**0.27**	**0.95**	**0.009**	**0.71**	**22.53**	**26.6**	**2.53**	**227**	**1.79**
VAL1	94.78	0.39	2.51	0.65	0.24	0.13	0.5	0.27	0.02	0.5	7	21.5	12.1	724.2	7.5
VAL2	92.26	0.18	2.37	1.59	0.51	1.18	0.36	0.14	0.02	1.35	4.5	39.5	4.7	269.5	4.4
VAL3	87.97	0.27	5.01	1.98	0.68	1.15	0.75	0.48	0.04	1.63	13.8	49.6	7	287.4	6.6
VAL4	91.96	0.14	2.04	0.68	0.47	2.32	0.34	0.05	0.01	1.91	2.5	45.9	3.4	148.2	3.6
VAL5	90.78	0.27	4.65	1.15	0.35	0.19	0.59	1.03	0.04	0.94	26.4	36.1	7.9	260.6	6.8
VAL6	93.64	0.23	3.08	0.85	0.27	0.13	0.71	0.28	0.03	0.77	8.1	29.4	7.2	323.4	5.8
VAL7	87.87	0.26	4.88	3.44	0.84	0.16	0.45	0.44	0.03	1.6	13.4	34.6	8	252.1	6.7
VAL8	93.76	0.21	2.92	0.68	0.24	0.31	0.55	0.37	0.03	0.93	9.5	25.4	5.6	285.4	5.3
Mean (n=8)	**91.63**	**0.24**	**3.43**	**1.38**	**0.45**	**0.7**	**0.53**	**0.38**	**0.03**	**1.20**	**10.65**	**35.25**	**7**	**319**	**5.84**
GM10	88.43	0.22	3.09	0.87	0.37	3.40	0.57	0.27	0.01	2.74	8.3	103.9	5.8	236.4	4.9
GM11	89.72	0.27	3.9	1.38	0.53	1.48	0.67	0.26	0.04	1.74	7.6	59.3	5 0	258.6	5.7
GM12	80.11	0.24	4.03	3.25	0.91	5.85	0.63	0.14	0.03	4.71	5.7	148.1	8.5	237.5	5.3
GM13	79.33	0.31	3.5	2.74	0.74	7.42	0.56	0.15	0.02	5.19	5.3	217.9	8.3	367.8	6.2
GM14	75.1	0.32	5.92	4.59	1.89	6.41	0.77	0.22	0.03	4.7	8.6	228.9	8.6	248.6	6.2
GM16	87.25	0.26	5.82	2.89	0.77	0.27	0.86	0.34	0.04	1.47	9.5	38.6	8.1	198.4	6.9
Mean GM (n=6)	**83.32**	**0.27**	**4.38**	**2.62**	**0.87**	**4.14**	**0.68**	**0.23**	**0.03**	**3.43**	**7.5**	**133**	**7.38**	**256**	**5.87**
Mean PQZ* (n=16)	**92.9**	**0.22**	**3.65**	**0.84**	**0.1**	**0.11**	**0.35**	**1**	**0.02**	**0.81**	**27**	**46**	**6**	**264**	**6**

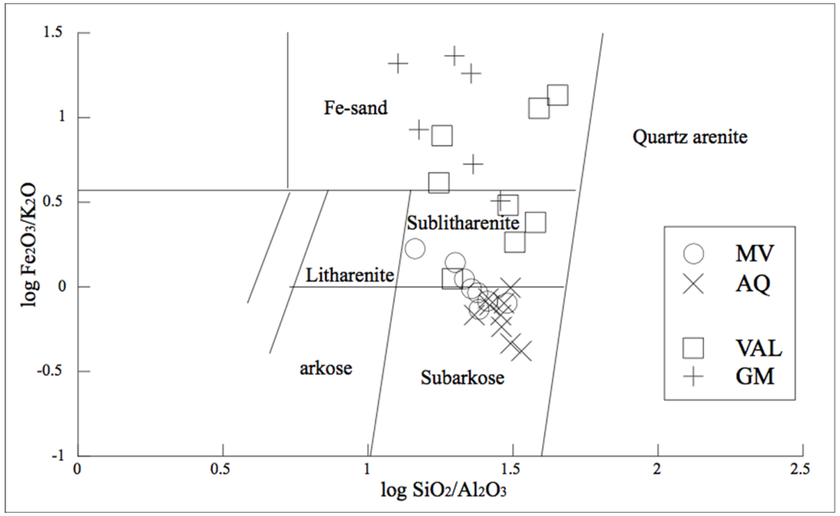

Fig. 7 Herron (1988) diagram (SiO_2/Al_2O_3 versus Fe_2O_3/K_2O) in which the chemical ratios of MV, AQ, VAL and GM samples are plotted.

Table 3 XRF analyses of different grain size fractions (four for each selected sample). Weight proportions are also indicated.

Sample	MV1	MV1	MV1	MV1	MV7	MV7	MV7	MV7	AQ12	AQ12	AQ12	AQ12	AQ16	AQ16	AQ16	AQ16
Weight %	68%	16%	10%	6%	57%	22%	15%	6%	60%	20%	11%	9%	51%	24%	14%	11%
Grain size (μm)	MV1<63	63<MV1<125	125<MV1>250	MV1>250	MV7<63	63<MV7<125	125<MV7<250	MV7>250	AQ12<63	63<AQ12<125	125<AQ12<250	AQ12>250	AQ16<63	63<AQ16<125	125<AQ16<250	AQ16>250
Fraction	**A**	**B**	**C**	**D**	**A**	**B**	**C**	**D**	**A**	**B**	**C**	**D**	**A**	**B**	**C**	**D**
SiO_2	77.21	90.33	93.78	92.41	80.35	91.93	94.74	94.28	84.64	93.74	96.2	96.43	82.64	92.77	95.66	95.27
TiO_2	0.83	0.34	0.18	0.23	0.84	0.26	0.13	0.15	0.66	0.29	0.15	0.13	0.56	0.21	0.12	0.13
Al_2O_3	10.7	4.45	2.76	3.63	9.98	4.11	2.71	2.90	8.31	3.5	2.18	1.97	8.4	3.72	2.29	2.46
Fe_2O_3	3.35	1.52	1.18	1.25	1.89	1.02	0.72	0.73	0.74	0.44	0.31	0.3	2.38	1.01	0.76	0.77
MgO	0.41	0.17	0.1	0.14	0.14	0.05	0.02	0.03	0.11	0.03	0	0	0.25	0.1	0.05	0.05
CaO	0.37	0.13	0.07	0.10	0.21	0.06	0.04	0.04	0.09	0.03	0.02	0.02	0.16	0.06	0.04	0.04
Na_2O	1.17	0.35	0.16	0.28	1.4	0.36	0.18	0.22	0.69	0.18	0.07	0.08	0.91	0.27	0.12	0.17
K_2O	2.91	1.11	0.56	0.77	3.11	1.26	0.75	0.76	2.88	1.1	0.54	0.49	3.00	1.24	0.61	0.69
P_2O_5	0.05	0.01	0.01	0.01	0.02	0	0	0	0	0	0	0	0.08	0.03	0.02	0.02
LOI	2.99	1.58	1.2	1.18	2.05	0.96	0.7	0.89	1.87	0.7	0.53	0.58	1.61	0.59	0.33	0.4
CaO/MgO	0.9	0.76	0.7	0.71	1.5	1.2	2	1.33	0.8	1	0	0	0.64	0.6	0.8	0.8
Ba	6502	257	158	190	729	323	196	198	636	255	139	119	628	277	142	158
Rb	73	33	19	20	80	37	22	21	74	29	13	11	67	28	14	15
Sr	129	49	28	28	123	45	25	25	95	33	15	11	103	39	18	19
Y	32	6	3	5	32	3	1	2	22	3	0	0	17	3	1	1
Zr	1215	412	126	220	1437	256	77	134	1129	453	91	109	684	251	61	105
Nb	11	5	2	2	11	4	1	2	9	4	1	1	7	1	0	0
V	42	20	12	14	28	12	8	8	21	11	6	5	31	12	9	9
Ni	23	19	17	10	18	14	11	12	8	8	7	6	13	9	7	7
La	39	14	11	14	32	10	9	9	27	10	7	8	17	11	8	9
Ce	69	24	19	17	64	13	11	10	43	14	10	9	33	13	13	12

the weight percentage of each fraction are reported in Table 3.

The fraction A (<63 μm) represents more than 50 % by weight from each sample indicating an important aliquote of this grain size in these sandstones (Table 3). This fraction shows lowest contents of SiO_2 (77-85 %) with high contents of Al_2O_3 (~8-11 wt%), K_2O (~3 wt%), Fe_2O_3 (0.7-3.4 wt%, 2 wt% in mean), Na_2O (0.7-1.4 wt%) and CaO (0.09-0.37 wt%). The LOI values range from 2.99 wt% to 1.6 wt% depending on the presence of a considerable quantity of clay minerals, micas and Fe-oxides/hydroxides, and the heavy minerals are concentrated in this fraction as suggested by high the Y and Zr contents (up to 1469 ppm).

The fraction B (63-125 μm) represents about 20 wt% of each sample (Table 3), the SiO_2 content increases ranging from 90 wt% to 94 wt%, whereas Al_2O_3 (3.5-4.5 wt%), MgO (0.05-0.17 wt%) and Fe_2O_3(0.44-1.52 wt%) decrease making cleaner this fraction with respect to fraction A being richer in quartz. The LOI values are again high in the MV fractions (1.6-0.96 wt%) whereas decrease up to 0.6-0.7 in the AQ ones. The Y and Zr contents decrease but remain high (456 ppm in the fraction of AQ12 sample and 254 ppm in the fraction of AQ16 sample).

The fractions C (125-250 μm) and D (>250μm) constitute ~ 20 wt% of the whole sample (Table 3), they show highest SiO_2 contents (94-95 wt% in MV and 96 wt% in AQ) and the lower values of Al_2O_3 (2.74 wt% in MV and 2.24 wt% in AQ), MgO (0.02-0.1 wt%), Fe_2O_3 (0.5-1 wt%), CaO (0.05 wt% in mean) and TiO_2 (0.15 wt% in mean). LOI values are high in the fractions of Monteverde samples (1.2-0.8 wt%) and decrease in the fractions of the Aquilonia samples (0.5-0.36 wt%), however, the LOI contents show a decrease towards the upper portions of both sections. The Y and Zr contents decrease in these fractions assuming the lowest values (100 and 84 ppm in mean) in AQ samples. The lower contents of volatile and heavy minerals are shown by C and D fractions from the AQ samples. The chemical composition of these fractions is related to higher concentration of quartz grains, however, the still high Fe_2O_3 contents in these fractions, almost in sample MV1, may be due to the ferruginous rims of the quartz grains and to the occurrence of Fe-oxides/hydroxides (Table 3 and Fig. 5).

7. DISCUSSION

The physical and chemical properties of Numidian quartz-rich sandstones from Aquilonia and Monteverde successions seem promising as raw materials for industrial uses including glass, foundry molds, ceramics, air Crete, adhesive, paints and many other. An assessment of the suitability for use of both bulk samples, and their different grain sizes, are presented as follows.

7.1. USE OF THE WHOLE-ROCK COMPOSITION OF THE NUMIDIAN SANDSTONES

Glass manufacture proposes many types of products with different chemical and physical properties (Bide et al., 2020). The principal glass products using silica sand include colorless and colored containers (bottles and jars), flat glass (windows, mirrors and vehicle glazing), and glass fibre (for reinforcement and glass wool insulation). In addition, the foundry industry has appreciated the advantage of the silica sands with specific physical and chemical standards mixed with a synthetic binder to produce a mold that retains shape and strength when metal is poured and cooled. The silica sands, as raw material, are usefully employed to produce air Crete characterized by low density cellular texture suitable for lightweight constructions (GWP Consultants LLP, 2010).

In the following, the peculiar features of the studied Numidian sandstones are compared with the specific technical requirements indicated by the industry for each specific use.

In glass industry, different types of silica sands are suitable depending on the type of glass to be produced. Generally, the sand includes a high silica content in the form of quartz and very low levels of deleterious impurities, particularly clay, iron oxides and refractory minerals, such as chromite, zircon and tourmaline (Burkowicz et al., 2020). In addition, they typically have a narrow grainsize distribution, generally in the range 0.5 to 0.125 mm (Bide et al., 2020). The very fine-grained sands (<63 micron) as well as coarse-grained sands (grain>0.71 mm) are discarded because they would cause defects in the final product (GWP Consultants LLP, 2010).

The general specifications about the chemical composition of the sands employed in the glass industry are shown in Table 4. The main

Table 4 Chemical standard for use of sands in different type of glass (modified by Khan et al. (2018); Edem et al. (2014); GWP Consultants LLP, (2010)).

Grade of glass		SiO_2	Fe_2O_3	Al_2O_3	CaO/MgO	TiO_2
Optical and opthalmic	**A**	99.7	0.013	0.2	0.1	0
Tableware and lead crystal	**B**	99.6	0.01	0.2	0.1	0
Borosilicate	**C**	99.6	0.01	0.2	0.2	0.012
Colourless container ("flint")	**D**	98.8	0.03	0.5	0.5	0.1
Clear flat	**E**	99	0.1	0.5	0.5	0.1
Coloured container	**F**	97	0.25	4	0.5	0.1
Insulating fibres	**G**	94.5	0.3	3	0.5	0.1

discriminating oxide contents in the use of sands to produce different types of glass are silica contents and iron and alumina oxides depending on the final uses of the products.

The whole-rock composition of Monteverde and Aquilonia sandstones seems to represent the most suitable typology for glass industry showing highest SiO_2 contents (92-94 %) and lowest amounts of contaminant elements (CaO~0.06-0.09 % and MgO~0.07-0.13 %); however, the Al_2O_3 (4.17-3.30 %) and Fe_2O_3 (1.17-0.66) contents do not match the required limits for the most of the uses in Table 4 except for the insulating fibers, if the iron contents are reduced. Sandstones from NW successions show a medium iron content ranging from 0.66 % (Aquilonia samples) to 1.17 % (Monteverde ones). The possibility of reducing the Fe_2O_3 content can be explored for the Aquilonia sandstones in which it takes lowest values. A procedure to clean quartz granules from iron patinas and to remove heavy minerals could be suggested to make these sandstones suitable for use as colored glass container (Tables 2-4). Cleaning of the rimmed granules and the gravity separation of the heavy minerals rich in Y, Zr, Ti and Cr would lead to a raw material with good chemical characteristics by reducing the impurities and the CaO/MgO ratio to approach the standard values around 0.5.

The characteristics of the silica sands for the foundry industry are less restrictive for the content of SiO_2 (95-96 %) and Fe_2O_3 (max 0.3), however, the contents of LOI (0.5), CaO (0.2) and Na_2O+K_2O (0.5) are also outside the ranges of the Monteverde and Aquilonia silica sands (Table 2). The grain size requirements include the grain size range 0.1-0.3 mm, and the exclusion of fraction < 0.1 mm as it would require too much binder in the preparation of the foundry molds. In addition, sub-angular quartz grains with good sphericity are preferred in silica sands for this use, implying less binder in the final foundry mold products. An evaluation of the overall characteristics of the Monteverde and Aquilonia sands suggests a possible use of these materials in the foundry industry, due to the chemical and physical features such as the SiO_2 content and the Qang/Qround ratio close to 1 (in average 0.82 and 0.99). Obviously, a specific particle size selection and removal of clays and heavy minerals must be carried out.

The Aquilonia and Monteverde sandstones seem promising as a component to produce aerated concrete blocks widely used as lightweight construction materials as Fe_2O_3 and Na_2O contents around 1 % and 0.3 % respectively, are tolerated for these products (Table 2) (Bide et al., 2020).

7.2. COMPARISON OF THE STUDIED SANDSTONES WITH QUARTZ-RICH SANDSTONES FROM PIETRAGALLA SUCCESSION

The chemical composition of the studied samples has been compared with that of similar quartz-rich sandstones constituting another Numidian succession, outcropping near Pietragalla village, in Lucania (Fornelli et al., 1998; D'Errico et al., 2014; Fornelli et al., 2015; 2019), with the aim to also test their suitability for industrial use. Pietragalla sandstones (n=16, PQZ label in Fornelli et al., 1998; Table 2) show a high compositional maturity with high contents in SiO_2 (93 wt% in average) and low contents of CaO (0.11 wt%), MgO (0.1 wt%) and Fe_2O_3 (0.8 wt%) owing to the total absence of crystalline and lithic carbonate components and clay-ferruginous cement. They show strong chemical and textural similarities with those from Aquilonia, so that in NW area of the Lucanian Apennines many Numidian sequences could be used as raw material in the glass and foundry industry and as building materials.

7.3. USES OF SPECIFIC GRAIN-SIZES OF THE NUMIDIAN SANDSTONES

Geochemical investigations on the selected granulometric fractions from the Monteverde and Aquilonia samples showed that the coarser grain sizes (C 125-250 μm and D>250 μm) in both sections have higher SiO_2 contents. However, the sum of the weight percentages of the two fractions is rather low ranging from 16 % to 25 %. The fine-grained fraction A (<63 μm) is prevalent representing the 51-68 % by weight (Table 3).

As expected, the coarse-grained fractions C and D are the most promising as specialist sands for industrial uses having SiO_2 in the range 92-96 %, CaO=0.02-0.1 %, MgO=0.02-0.14 %; unfortunately, deleterious elements such as Fe_2O_3 and TiO_2 still remain high, precluding the use of these sandstone for clear and colorless glass prodution. However, the chemical and granulometric characteristics of C and D fractions are more suitable after granule washing and further crushing of the >250 μm fraction, for use as colored glass container and insulating fibre production.

A different use might be suggested for the finer A and B fractions. Quartz sand in its finer form is used as a reinforcing filler and extender in paint preparations giving them greater hardness, scrubbing and wear resistance and improving their durability and flow capacity.

The finest grain size A represents a high percentage by weight after crushing of the Aquilonia sandstones with significant SiO_2 contents in the range 80.35 – 84.64 %. The possibility of mixing A and B fractions in different proportions, produced a raw material to be used in the preparation of paints, adhesives, grouts, fillers and extenders. In Table 5, the composition that would be obtained by mixing 40 % of fraction A with 60 % of fraction B is shown. The advantage of mixing the two fractions is to reduce the waste material, as the whole sample in all particle size fractions could be used: fractions C and D for insulating fibres and colored glass production, fractions A and B as fillers and extenders. Further cleaning processes as ultrasounds washing, chemical

Table 5 Calculated chemical composition mixing 40 % of fraction A and 60 % of fraction B from AQ12 sample.

Calculated composition AQ 12 sample	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K2O	LOI	CaO/MgO
40 % fraction A + 60 % fraction B	90.1	0.43	5.42	0.56	0.062	0.054	0.38	1.81	1.17	0.87

dissolution and gravity separation that remove heavy minerals in the finer fractions would result in a fully useable material.

8. CONCLUSIONS

Four Numidian sandstone successions along a NW-SE transect in the Campania-Lucania Apennine have been studied with the aim to suggest their suitability for industrial use. The petrography and geochemistry of the sandstones indicates that those from NW area are more promising for their high SiO_2 content (90.58-95.27) associated with lowest amounts of contaminant elements (CaO, MgO, Fe_2O_3, TiO_2) that could impart a dark pigmentation to the materials. However, the coloring elements, too high in content for clean or optical glass standard, suggest their use as raw material for colored glass, insulating fibers, foundry molds or air Crete. The comparison between Aquilonia and Monteverde sandstones and those from Pietragalla area suggests a strong compositional similarity extending the evaluable and exploitable rocks in a wider North-Western area (Fig. 2). Moreover, the study shows a relationship between grain size and sandstones chemical composition. The coarse-grained fractions C (125-250 μm) and D (>250 μm) show compositions more in line to the standard indications for colored glass, insulating fibers, foundry molds and cellular concrete, while finer grain sizes A (<63 μm) and B (63-125 μm), mixed in proportions of 40 % (A) and 60 % (B), properly cleaned of clays, heavy minerals and iron hydroxides can be used as raw material for the paint industry. In the circular economy, the almost complete use of these rock materials could lead to the development of the territory and to an accurate control of waste materials.

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