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

# Travertine, a building stone extensively employed in Umbria from Etruscan to Renaissance age: provenance determination using artificial intelligence technique

MAURIZIO PETRELLI\*, DIEGO PERUGINI, BEATRICE MORONI and GIAMPIERO POLI

Departimento di Scienze della Terra, Università di Perugia, Piazza Università, 1, 06100 Perugia, Italy

ABSTRACT. — This work is focused on determining the provenance of travertine stones employed in the construction of some monuments in Umbria (Italy) from the Etruscan to the Renaissance age. To this aim we propose a new methodological approach based on the combined use of petrographic observations and statistical analysis of geochemical data. Analyses are performed on samples from monuments and quarries whose activity is documented since ancient times. Statistical analysis is performed using the conventional Principal Component Analysis and two new methods based Artificial Intelligence (a Self-Organizing Map and a Fuzzy Logic System).

Results show that the Principal Component Analysis is a very poor technique to determinate travertine provenance because of its low discriminative power as stated using samples from the different quarries. On the contrary, the two Artificial Intelligence techniques show an excellent discriminative power and their application to monument samples produces very good and concordant results, although some uncertainties in the determination of travertine for some monuments are observed. These uncertainties can be solved, in most cases, by combining results of the statistical analysis with petrographic observations.

It is evidenced that a local provenance of traver-

tine employed in the construction of ancient buildings is a common feature at any age in the past. In addition, it is suggested that a non-local provenance may furnish information on the historical background in which a monument was conceived and built.

Results from this study indicate that the combined use of Artificial Intelligence techniques and petrographic observations is a powerful tool for provenance determination of travertine employed in the construction of ancient buildings.

RIASSUNTO. — Questo lavoro ha come scopo la determinazione della provenienza del travertino utilizzato per la costruzione di vari monumenti in Umbria (Italia) dal periodo Etrusco fino al Rinascimento. A tal fine viene proposto un nuovo approccio metodologico basato sull'uso combinato di analisi petrografiche e della statistica applicata ai dati geochimici. Sono stati analizzati i campioni provenienti dai monumenti e da cave la cui attività estrattiva è documentata sin dall'antichità. La statistica è stata eseguita applicando sia il metodo convenzionale dell'Analisi delle Componenti Principali sia utilizzando due nuovi metodi basati sull'Intelligenza Artificiale (la Self-Organizing Map ed un sistema basato sulla Logica Fuzzy).

I risultati dell'Analisi delle Componenti Principali sui campioni di cava mostrano ampie aree di sovrapposizione tra i campioni appartenenti alle diverse cave ed evidenziano quindi uno scarso

<sup>\*</sup> Corresponding author, E-mail: maurip@unipg.it

potere discriminativo del metodo. Al contrario, le due tecniche basate sull'Intelligenza Artificiale evidenziano un elevato potere discriminativo ed inoltre l'applicazione di queste due tecniche sui campioni provenenti dai monumenti produce buoni risultati sebbene si evidenzino alcune incertezze nella determinazione della provenienza di alcuni campioni. Le incertezze possono essere risolte nella maggior parte dei casi combinando i risultati delle analisi statistiche con quelli relativi alle osservazioni petrografiche.

Dalle analisi risulta che l'impiego di travertino locale in tempi passati fosse un'usanza estremamente diffusa. Inoltre, l'utilizzo di travertino di provenienza non locale potrebbe fornire informazioni sul contesto storico in cui il monumento è concepito e realizzato.

I risultati di questo studio indicano che l'uso combinato delle tecniche di Intelligenza Artificiale e degli studi petrografici è un mezzo estremamente utile nel determinare la provenienza del travertino impiegato nella costruzione degli edifici antichi.

KEY WORDS: Archaeometry, travertine, Umbria, artificial intelligence, self-organizing maps, fuzzy logic.

#### INTRODUCTION

Travertine is one of the most employed and appreciated building stones in Central Italy given its wide geographic diffusion and easy quarrying. Many travertine deposits with different origin, extension, economic validity, and historical importance are present in Tuscany, Umbria and Latium (e.g. Cipriani *et al.*, 1977), and a large number of them were sites of extraction of stones employed in the construction of monuments and works of art in different periods in the past (e.g. Rodolico, 1965).

Provenance determination of travertine is a complex archaeometric problem due to the wide textural and geochemical variability inherent to this lithology. Travertine, in fact, is the result of different overlapping processes, the most important being primary encrustation and secondary diagenetic processes that rapidly evolve in space and time generating a complex dynamical system in continuous evolution (e.g. D'Argenio and Ferreri, 1988). The result is a large lateral and vertical variability of the textural and geochemical properties of the stone with large dis-homogeneities within a single travertine formation (e.g. Ford and Pedley, 1996; Moroni and Poli, 1999).

Because of this heterogeneity, the combination of several methods, including petrographic and geochemical analysis, are needed for provenance determination studies. Petrographic analysis are useful for the identification of depositional environments and plays a fundamental role in paleo-geographic reconstruction. Geochemical analyses can be also related to the depositional environment of travertine. Elaboration of geochemical data can be performed using several statistical approaches. Among them, Principal Component Analysis (PCA) is widely used in archaeometry and many examples are present in literature (e.g. Buxeda et al., 2003a; Hall, 2001). In recent years, new analytical methods of data elaboration based on Artificial Intelligence (AI) have been developed and a wide variety of them have been made available in the literature (e.g. Fausett, 1994; Principe et al., 1999). The ability of AI methods to manage large amount of data by using «learning» algorithms makes them potential powerful methods to be used for archaeometric purposes (e.g. Remolà et al., 1996; Lopez-Molinero et al., 2000).

In this work we propose an approach based on the combined use of petrographic observations and statistical analysis of geochemical data using both classical (PCA) and AI statistical methods. We apply this approach crosscorrelating travertine samples belonging to quarries whose activity is documented from ancient times with the aim to determine provenance of travertine employed in the construction of some works of art in Umbria Region (Italy). In particular this study is focused on the Etruscan Arch and the medieval Fontana Maggiore in Perugia, on several monuments in Corciano, both etruscan and medieval and on the Orvieto Cathedral, built in the renaissance age.

#### MATERIALS AND METHODS

## Sampling and analysis

Travertine samples were collected from monuments and ancient quarries mentioned in historical documents as sites of extraction (Fig. 1). A total number of 195 travertine samples, 91 from monuments and 104 from quarries were sampled.

Regarding monuments 24 samples have been collected from the Etruscan Arch, 20 from the Fontana Maggiore, 7 from «Corciano

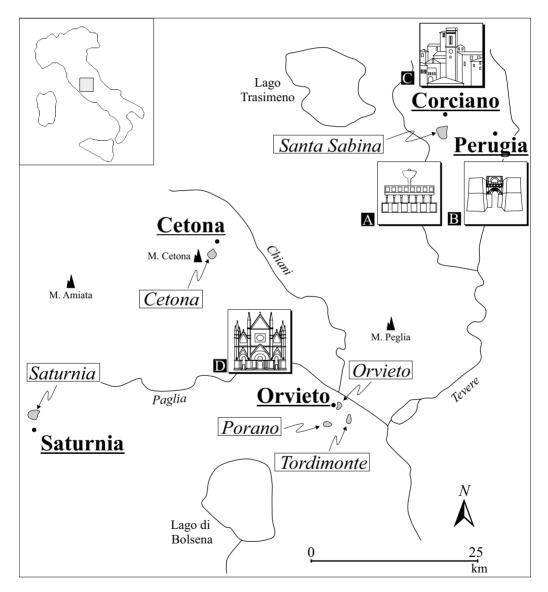


Fig. 1 – Schematic map showing the location of monuments (A: Fontana Maggiore, B: Etruscan Arch, C: «Corciano Monuments», D: Orvieto Cathedral), and travertine quarries (gray shaded regions indicated by black arrows).

Monuments» and 53 from the Orvieto Cathedral (Table 1A). Samples from «Corciano Monuments» were taken from some historical remnants in the town; in particular 2 samples are from two lion statues standing on the opposite sides of a flight of steps in the center of the village and of sure etruscan origin, 2 samples from the S. Cristoforo Cathedral, 1 sample from the tower of the town, 1 sample form the Church of S. Maria Assunta and 1 sample from the town walls, all of medieval origin.

Regarding quarries, 8 samples belong to the ancient quarry of Santa Sabina near Perugia, 53 samples are from a large quarry area in Saturnia, 5 samples were collected from small outcrops near Cetona, and 25 samples were collected from the «Orvieto district» (Fig. 1). For «Orvieto district» we intend three main outcrops located in the surroundings of the town of Orvieto: Tordimonte (15 samples), Porano (5 samples), and Orvieto (5 samples).

Sampling on monuments has been carried out in the hidden parts of the buildings using hammer and chisel with special care in order to preserve the aesthetic value of the monuments. With this method it is possible to collect only small samples (of the order of few cm<sup>3</sup>) from each travertine stone that may not fully represent the whole block. In order to minimize this problem several samples from the same travertine block have been collected with the aim to have a representative statistical number of samples. Special care was also taken to remove

# TABLE 1

# A) Number of travertine samples collected from the four studied monuments; B) number of travertine samples collected from quarries; C) major and trace elements utilized for the application of the SOM and the FLS.

Monuments	Age	Locality	N. of Samples
Etruscan Arch	Etruscan	Perugia	24
Fontana Maggiore	Medieval	Perugia	20
Corciano Monuments	Etruscan - Medieval	Corciano (Perugia)	7
Orvieto Cathedral	Renaissance	Orvieto	53
Total samples from Monuments			104
	(B)		
Qarries & Outcrops	Туре	Locality	N. of Samples
Santa Sabina	quarry	Perugia	8
Orvieto District	outcrop	Orvieto	25
Cetona	outcrop	Cetona	5
Saturnia	quarry	Saturnia	53
Total samples from quarries			91
	(C)		
Analyzed chemical elements			
Major Elements Trace Elements		Fe <sub>2</sub> O <sub>3</sub> , MnO, MgO, CaO, K n, Ga, Rb, Sr, Y, Zr, Nb, Ba	

(A)

the weathered layers in samples from both monuments and quarries.

All samples were characterized for their petrographic features and analyzed by X-ray fluorescence spectroscopy (XRF) to determine major and trace element abundances (Table 1C).

Analysis of some representative samples from monuments and quarries are reported in Table 2.

#### Principal Component Analysis (PCA)

Principal Component Analysis (PCA) is a statistical method for reducing the dimensionality of data sets while retaining as much information as it is possible about the sample population (Baxter, 1994). This technique consists in generating a new set of variables called Principal Components combining the variables of the original data set. The first principal component is the combination of variables that explains the greatest amount of variation within the data. The second principal component defines the next largest amount of variation and is independent from the first principal component and so forth. In practice, the first few Principal Components are able to explain the total amount of data given by all original variables with a minimum loss of information.

As drawbacks, PCA results are difficult to interpret when there are numerous samples or when the percentage of explained variance is not high enough (Remolà *et al.*, 1996). PCA method is widely described in literature (Baxter, 1994) and there is an extensive application of this technique for archaeometric purposes (e.g. Hall, 2001; Buxeda *et al.*, 2003b).

## Artificial Intelligence techniques

Artificial Intelligence is a branch of Science focused on the development of new methods to be used by computers to find solutions to complex problems in human-like fashion. This generally involves borrowing characteristics from human intelligence and applying them as algorithms. There are many different approaches to generate Artificial Intelligence algorithms; among them the Artificial Neural Networks (ANNs) are well studied and can be applied to a wide variety of problems, such as signal processing, control problems, pattern recognition, and classification problems (e.g. Fausett, 1994; Bishop, 1996; Principe *et al.*, 1999).

An Artificial Neural Network is an information-processing system that shares some performance characteristics with biological neural networks. A neural network consists of a large number of processing elements called neurons (Fig. 2). Each neuron elaborates elementary problems using a function called «activation function». Neurons are connected each other by communication links, each with an associated weight (w, Fig. 2). The weights represent the information being used by the network to solve a problem. In general the weight of each neuron is determined in a preliminary phase of training in which several examples of solutions are presented to the network; this phase is called «learning» of the network. Neurons are generally arranged in layers (Fig. 3) in which data are initially acquired and successively elaborated.

In this work two techniques based on Artificial Neural Networks are utilized in the attempt to determine provenance of travertine stones employed in the construction of the studied monuments.

The first technique is known as the Self-Organizing Map (SOM; Kohonen, 1998). It is a tool for the visualization of high-dimensional problems into regular low-dimensional grids and it is able to convert complex and non-linear statistical relationships between high-dimensional data into simple geometric relationships on a low-dimensional display.

The choice of using the SOM approach is related to the fact that travertine is a very complex lithology having wide geochemical heterogeneity. In order to keep into account the extreme variability inherent to this lithology many parameters (i.e., a large number of chemical elements; Table 1 and 2) must be considered at once. The capability of the SOM

# TABLE 2

# Geochemical analysis of some representative travertine samples from monument and quarries. Major elements (with the exception of MgO determined by wet chemical analyses) analyzed by XRF with full matrix correction after Franzini and Leoni (1972); trace elements by XRF after Kaye (1965).

Sample N	Vame FM1	FM6	FM7	AE2	AE3	AE4	OC11	OC13	OC33	CM1	CM2	CM3
Site	Fontana Maggiore	Fontana Maggiore	Fontana Maggiore	Etruscan Arch	Etruscan Arch	Etruscan Arch	Orvieto Cathedral	Orvieto Cathedral	Orvieto Cathedral	Corciano Monuments M	Corciano Aonuments	Corciano Monuments
Location	Perugia	Perugia	Perugia	Perugia	Perugia	Perugia	Orvieto	Orvieto	Orvieto	Corciano	Corciano	Corciano
Facies	МТ	ST	ST	РТ	МТ	РТ	DT	PT	ST	МТ	MT	МТ
Major I	Elements											
SiO <sub>2</sub>	0.13	5.84	3.06	0.43	10.44	3.80	10.76	11.31	7.50	0.51	0.88	2.35
TiO <sub>2</sub>	0.00	0.09	0.03	0.01	0.10	0.05	0.04	0.02	0.01	0.01	0.01	0.03
$Al_2O_3$	0.04	2.24	1.15	0.16	2.85	1.19	1.02	0.83	0.41	0.20	0.38	0.81
Fe <sub>2</sub> O <sub>3</sub>	0.06	0.82	0.42	0.08	0.70	0.47	0.34	0.24	0.18	0.11	0.14	0.22
MnO	0.05	0.06	0.07	0.04	0.06	0.07	0.08	0.04	0.07	0.03	0.02	0.02
MgO	1.25	1.28	1.48	0.48	0.86	0.54	0.69	0.89	1.01	0.52	0.30	0.27
CaO	54.10	46.85	50.06	54.35	43.07	50.18	46.93	46.01	48.74	54.77	54.68	53.60
K <sub>2</sub> O	0.01	0.41	0.23	0.02	0.72	0.28	0.23	0.26	0.17	0.02	0.05	0.14
$P_2O_5$	0.59	0.72	0.72	0.56	0.58	0.64	0.20	0.18	0.65	0.39	0.27	0.20
LOI	43.77	41.70	42.78	43.87	40.62	42.78	39.39	40.13	41.26	43.44	43.27	42.36
ТОТ	100.00	100.01	100.00	100.00	100.00	100.00	99.68	99.91	100.00	100.00	100.00	100.00
Trace E	Elements											
v	1	1	1	7	7	7	4	4	6	10	10	10
Cr	b.d.l.	6	b.d.l.	b.d.l.	8	6	2	5	b.d.l.	4	3	6
Co	b.d.l.	2	b.d.l.	5	3	4	5	1	6	1	1	1
Ni	b.d.l.	b.d.l.	b.d.l.	3	4	1	7	3	8	5	0	0
Cu	9	15	17	6	2	21	13	17	16	15	16	21
Zn	9	16	12	17	26	20	35	29	29	62	20	31
Ga	2	2	2	1	2	1	0	1	1	10	14	12
Rb	2	16	6	2	22	8	39	19	4	1	2	11
Sr	630	1000	1300	510	540	660	1864	1375	1989	579	704	651
Y	b.d.l.	b.d.l.	b.d.l.	3	3	3	1	1	0	0	0	1
Zr	22	43	43	15	24	24	65	38	49	21	21	26
Nb	2	3	3	4	2	3	5	1	1	1	2	2
Ba	2	19	5	3	17	16	125	245	21	9	0	22
La	36	40	45	21	16	17	15	21	0	6	8	5
Pb	2	274	696	8	8	14	13	35	17	9	8	30
			014	-	~	~	1.1	2	5	3	2	2
Ce	1	84	214	5	5	5	11	2	5	3	3	3

The precision is better than 15% for V, Cr, Ni, Co, Y, Ga and Nb, better than 10% for Cu, Zn, Rb, La, Pb, Ce, Th, Zr, Ba, and better than 5% for Sr. The accuracy has been tested on international standards and is better than 10%.

SAT53	SAT25	SAT9	CET5	CET3	CET1	OD11	OD9	OD1	SS7	SS5	SS1
Saturnia	Saturnia	Saturnia	Cetona	Cetona	Cetona	Orvieto	Orvieto	Orvieto	S.Sabina	S.Sabina	S Sabina
quarry	quarry	quarry	Outcrop	Outcrop	Outcrop	District	District	District	quarry	quarry	quarry
Saturnia	Saturnia	Saturnia	Cetona	Cetona	Cetona	Tordimonte	Porano	Orvieto	Perugia	Perugia	Perugia
DT	MT	DT	ST	ST	PT	PT	РТ	ST	DT	MT	PT
0.07	0.04	0.03	5.84	8.34	1.50	5.73	13.86	6.47	3.64	0.74	0.86
0.00	b.d.l.	b.d.l.	0.09	0.13	0.02	0.05	0.21	0.05	0.04	0.01	0.01
0.10	0.07	0.06	2.39	2.86	0.58	1.29	4.70	1.58	1.11	0.23	0.29
0.07	0.05	0.02	0.55	1.15	0.30	0.54	2.23	0.58	0.31	0.09	0.12
0.04	0.03	0.04	0.02	0.03	0.03	0.02	0.08	0.03	0.04	0.03	0.04
0.36	0.43	0.47	0.36	0.72	0.32	0.79	0.97	0.55	0.44	0.39	0.32
54.94	55.13	55.14	49.63	47.10	53.83	49.93	41.11	48.39	50.83	54.11	54.03
b.d.l.	b.d.l.	b.d.l.	0.30	0.46	0.06	0.25	1.08	0.44	0.22	0.05	0.05
0.53	0.49	0.47	0.09	0.16	0.09	0.22	0.30	0.27	0.54	0.61	0.55
43.88	43.76	43.78	40.66	38.89	43.23	41.00	34.23	41.43	42.83	43.75	43.73
99.99	100.00	100.01	99.93	99.84	99.96	99.82	98.77	99.79	100.00	100.01	100.00
7	7	10	12	15	10	10	52	10	7	7	7
2	b.d.l.	b.d.l.	28	29	1	5	6	4	10	b.d.l.	1
2	1	7	1	2	b.d.l.	0	6	2	b.d.l.	1	4
1	1	b.d.l.	4	15	b.d.l.	4	15	b.d.l.	3	b.d.l.	b.d.l.
12	15	16	14	20	17	23	24	20	8	7	4
7	9	2	21	48	33	19	46	29	12	9	13
0	b.d.l.	1	4	1	1	1	5	3	1	1	1
3	3	2	22	28	6	31	284	40	7	3	3
658	708	880	442	1477	559	1272	1716	1651	493	484	457
0	b.d.l.	3	13	3	3	1	9	1	4	3	3
12	13	15	31	58	26	52	132	66	17	16	17
1	2	1	4	4	3	4	8	4	1	2	1
5	5	2	38	75	10	92	417	188	3	3	3
24	21	17	21	10	19	21	42	19	16	7	11
5	7	2	9	9	10	12	228	13	2	6	2
5	5	3	6	7	4	9	126	10	5	5	5
0	b.d.l.	b.d.l.	0	2	3	2	14	5	4	4	2

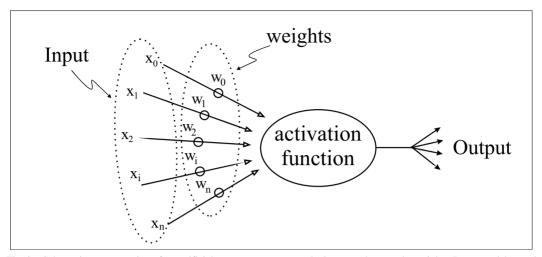


Fig. 2 – Schematic representation of an artificial neuron;  $x_i$  represents the inputs and  $w_i$  are the weights. Data are elaborated by the activation function which gives the output.

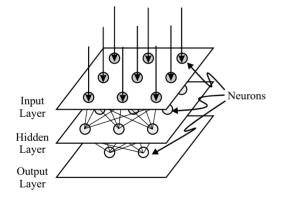


Fig. 3 – Typical structure of an Artificial Neural Network; neurons are arranged in layers. The Input layer receives the information to be processed, hidden layers elaborate data and, finally, the output layer gives the result.

technique to reduce and to present graphically the solution on two dimensional displays makes this technique a potentially useful tool to study and characterize travertine.

The used SOM (Petrelli *et al.*, 2003) consists of a two-dimensional regular grid of neuron  $W_{(j,k)}$  (Fig. 4) in which each neuron contains a number of weights  $w_{(j,k)i}$  equal to the number of elements in the input vectors

*Xs.* Input vectors are determined by geochemical analysis of travertine samples; in particular each travertine sample is described by 27-element vector *Xs* ( $x_{s1}$ ,  $x_{s2}$ ,  $x_{s3}$ ,...,  $x_{sn}$ , ...,  $x_{s27}$ ) in which each variable  $x_{sn}$  corresponds to the concentration of the chemical element *n* in the sample *s* (Fig. 4).

Learning is a self-organized process carried out in cycles, called «epochs» (t). One epoch is the period in which all input vectors Xs are presented to the network once. During the learning process, the SOM automatically adapts itself in such a way that similar input vectors Xs excite most of the neurons placed topologically close to each other. The result of this operation is a map in which the more similar input vectors Xs will be clustered, whereas dissimilar vectors will lie far away in the map.

The second AI technique is based on the principles of Fuzzy Logic (e.g. Klir and Yuan, 1996; Yang *et al.*, 2000). Fuzzy Logic theory deals with classes or groupings of data with boundaries that are not sharply defined and cannot be adequately represented by classical binary scheme classifications (e.g. 0-1, black-white). Therefore, Fuzzy Logic methods are useful for studying processes and phenomena

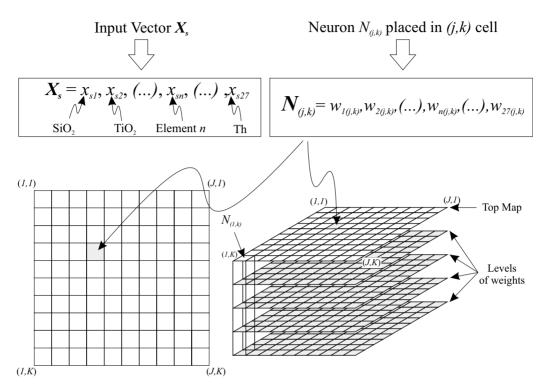


Fig. 4 – Schematic Kohonen SOM represented by a 10x10 block of neurons. Each input vector  $X_s$  is elaborated by several neurons  $N_{(j,k)}$ . In the case shown in the figure each neuron  $N_{(j,k)}$  is represented by a column of five weights [indicated in the figure as  $w_{(1,K)}$ ]. The top map displays the final output of the computation.

characterized by a continuous evolution in space and time.

The Fuzzy Logic System (FLS) that we developed, works by comparing the chemical features of samples belonging to monuments and those of samples belonging from quarries. This technique is detailed in Petrelli *et al.* (2003) and it is schematically illustrated in figure 5. It consists of a simple network which contains a number of input neurons equal to the number of elements in the input vector Xs (i.e., 27 input neurons, Fig. 5A, Step 1). Quarry sample compositions are used as internal reference database. Differently from the SOM approach, no learning process is utilized by the FLS. Each input monument sample (Xs) is elaborated by the artificial neurons

whose activation value  $a_n$  varies as a continuous function bounded between 0 to 1 (Fig. 5B). The more the concentration of chemical elements between the input monument sample and a quarry sample are similar, the more the activation value of the neuron approaches unity (Fig. 5B). Since analyzed chemical elements are 27, for each quarry sample there are 27 activation values, one for each neuron. The sum of these activation values indicates the degree of geochemical similarity between the unknown sample and each quarry sample and it is called Global Activation Value (I, Fig. 5A, Step 2). The procedure ends by associating the input monument sample to the quarry sample with the highest value of *I* (Fig. 5A, Step 3).

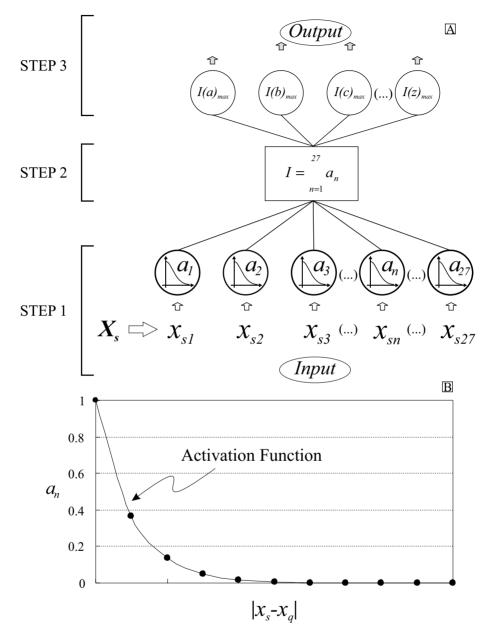


Fig. 5 – A) Schematic representation of the Fuzzy Logic System (FLS). STEP1: the unknown sample is introduced into the FLS as a 27-dimensional input vector  $X_s$  and activation values  $a_n$  between each chemical element of the input sample and the quarry samples are calculated; STEP 2: calculation of the global activation value (*I*) summing  $a_n$  values; STEP 3: the final output of the FLS associates the unknown sample to the quarry having the maximum degree of similarity (i.e., the maximum  $I_{max}$  value). B) Activation function:  $x_s$  is the chemical concentration of an element in the input sample,  $x_q$  is the concentration of the values of concentration are similar in the input and in the quarry sample) the higher is the value  $a_n$  of the activation function.

#### Petrography

## General features

All travertines, both from monuments and outcrops, are the result of calcium carbonate precipitation on microphyta, macrophyta and invertebrate supports. Four main travertine lithotypes were recognized, namely stromatolitic, phytohermal, microhermal and detrital (D'Argenio and Ferreri, 1988). Stromatolitic, phytohermal and microhermal travertines derive from chemical encrustation on vegetal supports (algal felts, macrophyta and microphyta, respectively) in growth position. Detrital travertines, instead, derive from biogenic processes of calcium carbonate encrustation involving macro- and microphyta fragments belonging to stromatolitic, phytohermal and microhermal travertines after mechanical transport. Detrital travertines may contain variable proportions of arenitic to siltitic calcareous sand.

Regarding monuments, stromatolitic, phytohermal and microhermal travertine is absent in the samples from Etruscan Arch, Fontana Maggiore and Orvieto Cathedral, respectively. Only microhermal and detrital travertine is present in the samples from «Corciano Monuments». In outcrops and quarries, stromatolitic and microhermal travertine is absent in the samples from S. Sabina and from «Orvieto district», respectively; only phytohermal and stromatolitic travertine is absent in the samples from Cetona, whereas the four lithotypes of travertine are all represented in the samples from Saturnia. Differences exist in the type and abundance of the fossil and siliciclastic component in the samples from different monuments and quarries. Table 3 reports a summary of the petrographic characteristics of travertine samples.

#### Monuments

The fossil component in the samples from monuments consists of Characeae oogonia, Gastropoda, Ostracoda and bivalvs. In the samples from Etruscan Arch only Characeae oogonia were observed, whereas in some samples from «Corciano Monuments», and in all samples from Orvieto Cathedral, Gastropoda and Ostracoda were recognized along with algal oogonia.

The siliciclastic component is scarse in the samples from Etruscan Arch, where it consists exclusively of quartz, and totally absent in the samples from Corciano and in the microhermal travertine from Fontana Maggiore, respectively. Stromatolitic and detrital travertines from Fontana Maggiore and all samples from Orvieto Cathedral contain a much varied siliciclastic fraction. In particular, samples from Fontana Maggiore contain plagioclase, muscovite and pyroxene whereas samples from Orvieto Cathedral contain K-feldspar, biotite, chlorite, titanite and epidote.

The depositional environment of all travertines, as inferred by the presence of Characeae oogonia in almost all samples, is lacustrine or paludal (e.g. Ford and Pedley, 1996). The presence of magmatic components within the travertines from Orvieto Cathedral, and within stromatolitic and detrital travertine samples from Fontana Maggiore is a clear evidence of lacustrine sedimentation occurring inside or in the proximity of volcanic areas. Composition of clinopyroxenes from these samples is similar to that of clinopyroxenes in volcanic products of the Roman Magmatic Province (Moroni and Poli, 2000). In particular, composition of clinopyroxenes from the samples from Orvieto Cathedral is consistent with that of samples from Orvieto district outcrops (Moroni and Poli, 2000).

# Quarries

The fossil component in the quarry samples consists of Characeae oogonia, Gastropoda, Ostracoda and bivalvs. Fossils are totally absent in the samples from Saturnia, and rare in the samples from Cetona, where they are represented by Ostracoda and Gastropoda, and S. Sabina where they consist of Caharaceae oogonia and bivalvs. In the samples from Orvieto district the fossil component is scarce and consists of Characeae oogonia, Ostracoda and Gastropoda.

TABLE 3	Summary of the petrographic characteristics of travertine samples from monuments and quarries. $ST = Stromatolitic Travertine;$ MT = Microhermal Travertine; PT = Phytohermal Travertine; DT = Detrital Travertine; f.c. = fossil component; s.c. = siliciclastic component; numbers refer to the number of samples; () rare; —— not present; Cha = Characeae; $Ostr = Ostracoda; Gastr = Gastropoda; Biv = bivalvs; Qz = quartz; K-fld = alkali-feldspar; Bi = biotite; Cpx = clinopyroxene; Ti = titanite; Chl = chlorite; Pl = plagioclase; Ep = epidote; Ms = muscovite; Pyr = pyroclastic fragments.$	
---------	--	--

	II = IIIanI	ue; Chl = chlo	rite; Fi = plagi	11 = 11 and $12 = 11$ and	idote; MS = m	uscovite; F	yr = pyrocta	suc fragments.
Μ	MONUMENTS	S QUARRIES						
Locality	Perugia	Perugia	Perugia	Orvieto	Cetona	Saturnia	Santa Sabina	Orvieto District
Description Etruscan Arch	Etruscan Arch	Fontana M.	Corciano Monuments	Cathedral	outcrop	quarry	quarry	outcrop
ST		8		21	2	ю		10
f.c.		Cha	)) –	(Ostr, Gastr, Cha)	I	I		(Cha, Ostr, Gastr)
s.c.		Qz, Ms, Cpx,	— (Qz,	(Qz, K-fld, Bi, Px, Ms)	- (5		)	(Qz, Px, K-fld, Pl, Pyr, Bi, Chl)
MT	12	6	9			6	1	
f.c.	Cha	Cha	(Cha, Gastr, Biv, Ostr.)		I		Cha, Biv	
s.c.								
PT	9			13	5	6	4	13
f.c.				(Ostr, Gastr) ((	(Ostr, Gastr)			(Ostr, Gastr)
s.c.	Qz			Px, Qz, k-fld (Ti, Chl, Pl, Ep)			Qz	Qz, Pl, Px (Bi, K-fld, Ep, Ti, Chl, Pyr)
DT	9	ю	1	19		32	ю	7
f.c.	Cha	Biv, Cha	Gastr, Biv, Ostr Ostr, Gastr	Ostr, Gastr			Cha	(Ostr, Gastr)
s.c.	Qz	Qz, Pl, Cpx		Px, Bi, K-fld			Qz	(Px, Pyr, Qz, Pl)

The siliciclastic component is absent in the samples from Cetona and Saturnia, rare (only a few quartz crystals) in the samples from S. Sabina, varied and abundant in the samples from «Orvieto district» where it is represented by the same mineral phases found in the samples from the Cathedral (clinopyroxene, K-feldspar, plagioclase, biotite, chlorite, titanite and epidote) along with a few pyroclastic fragments.

The depositional environment of travertines from Santa Sabina, «Orvieto district» and Cetona is lacustrine as evidenced by the faunal and algal fossil contents. Travertine from «Orvieto district» contains a magmatic siliciclastic component which is in accordance with the vicinity of the sampled area to the Vulsinian Volcanic District. The characteristics of the magmatic component and in particular the presence of pyroclastic fragments within the travertine, indicate lithoclast derivation from erosion of pyroclastic formations. The presence of macrophytes and the lack of any faunal remnants in the travertine from Saturnia can be assumed as an evidence of carbonate deposition in a gentle hydrothermal water regime.

#### STATISTICAL ANALYSIS

#### Principal Components Analysis (PCA)

The PCA technique has been initially applied to quarry samples in order to establish the discriminative power of the method before applying it for provenance determination purposes. Figure 6 shows the application of PCA method on quarry samples. All diagrams evidence wide overlapping regions among samples belonging to the different quarries. Samples from Saturnia and Santa Sabina always cluster as a single group with the only exception of figure 6C in which the two quarries are quite well discriminated. Orvieto district samples show very large scattering with respect to all Principal Components; this results in overlapping regions among these samples and all other quarry samples.

Similar to Orvieto samples, Cetona samples are scattered in all diagrams (Fig. 6) and they show large overlapping regions with all other quarry samples.

## Artificial Intelligence techniques

Like the PCA method, the two AI systems (SOM and FLS) have been initially tested on quarry samples in order to assess their discriminative power.

#### Application of the Self-Organizing Map (SOM)

Figure 7 shows a SOM for the 91 quarry samples. The SOM shows that samples belonging to each quarry are always well clustered. The only exception is related to a sample belonging to Cetona which is positioned on the right upper corner of the grid and lies far away form the other Cetona samples (Fig. 7, black arrows).

These tests evidence a good discriminative power of the used SOM regarding quarry samples. In detail, SOM is able to cluster samples belonging to the same quarry and well discriminate samples from different quarries.

The SOM has been applied using samples from both monuments and quarries. Figure 8 shows a SOM obtained utilizing the 91 quarry samples together with the 104 samples from monuments.

The structure of the SOM shows that Etruscan Arch samples are clustered with samples of the quarry of Santa Sabina. Travertine samples from the Fontana Maggiore are divided in two groups (Fig. 8) and they are not associated to any quarry. Besides, one sample is positioned far away from the areas occupied by other samples of the Fontana Maggiore (Fig. 8, black arrows). «Corciano Monuments» samples are clustered as single group and they are not associated with samples belonging to any quarry. Almost all samples from the Orvieto Cathedral and from quarries of the «Orvieto district» are clustered. Two samples from Orvieto Cathedral and three samples from «Orvieto district» quarries are

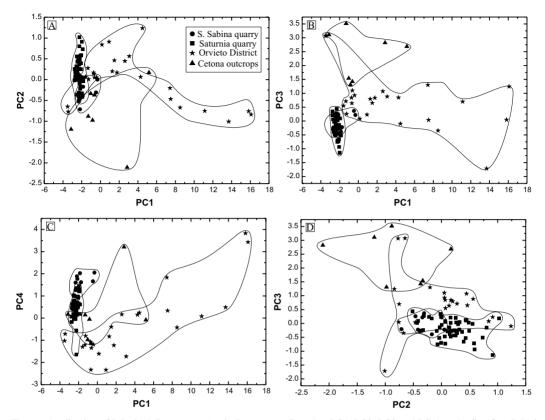


Fig. 6 – Application of Principal Component Analysis to quarry Samples. PC1, PC2, PC3 and PC4 are the first four Principal Components.

clustered in a different area (upper right corner of the map). No sample belonging to monuments are clustered With Saturnia samples. Three Cetona samples are clustered on the upper portion of the map and they are not grouped with any sample from monuments. The two other Cetona samples are positioned respectively in the lower portion and in the left side of the map.

## Application of the Fuzzy Logic System (FLS)

In order to test the discriminative power of the FLS, each quarry sample has been excluded from the quarry database and has been considered as unknown. The procedure has been repeated for all quarry samples considering each sample as an unknown sample. Table 4 shows the results of the test. The system is capable to recognize the exact provenance of 89 on 91 samples with a percentage of success of 97.8%.

## TABLE 4.

Results of the application of the FLS on quarry samples.

Qarry	N. of Samples	Correct	Uncorrect
Santa Sabina	8	8	0
Orvieto District	25	25	0
Cetona	5	3	2
Saturnia	53	53	0
Total	91	89	2

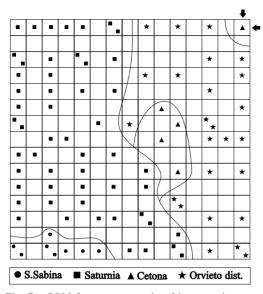


Fig. 7 - SOM for quarry samples. Lines are drawn to evidence the clustering of samples belonging to the same quarry. Black arrows indicate the coordinates of the Cetona sample positioned far away from other Cetona samples.

쇼 ☆ ☆

÷

☆

\*

\*\*

. .

☆  $\dot{\mathbf{x}}$ 

\*

☆

\* ☆ ☆

\* ☆

☆

☆

\*

O Etruscan Arch

Corciano Mon.

☆ ☆

☆

\* ☆

\*

 $\star$ 

\*

☆

\$ ☆

☆

۸ ÷

\*

\$ \$ ☆ \$ 0 ο

\*

☆ ☆ \$

÷

. •

0 0 0

 $\triangle$  Fontana Maggiore

☆ Orvieto Cath.

Δ 4

-

. -.

. 0

0 ်ဝ

Δ

. 

۸

.

•

0

С

c

0

• 0 ړه 0 0 0

0

+

\* á ۰. ^

۰.

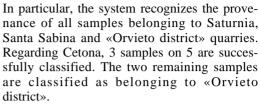
. 

> 0 0

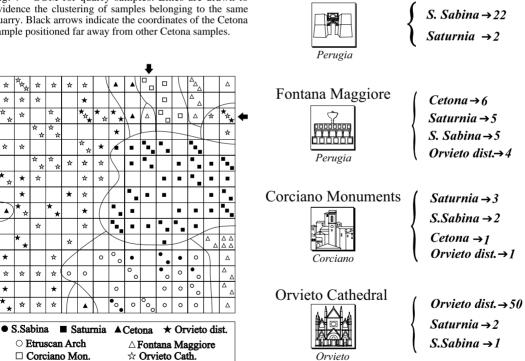
.

Δ

Δ



The FLS has been applied on travertine samples belonging to the four studied monuments. Results are schematically reported in figure 9. 22 samples from the Etruscan Arch are assigned to the quarry of Santa Sabina and 2 to Saturnia. The 53 samples of the Orvieto Cathedral are classified as it follows: 50 samples are assigned to the «Orvieto district», 2 samples to Saturnia and 1 sample to Santa



Etruscan Arch

Fig. 8 - SOM computed using both quarry and monument samples. Arrows indicate the sample from the Fontana Maggiore that deviate from the behavior of the other samples. Lines are drawn to evidence the clustering of samples.

Fig. 9 - Schematic report of the results obtained by the application of the FLS on monument samples. For each monument the number of samples assigned to the different quarries are reported.

Sabina. Regarding the 20 samples belonging to the Fontana Maggiore very dispersed results are obtained: 6 samples are assigned to the quarry of Cetona, 5 to Saturnia, 5 to Santa Sabina and 4 to Orvieto District quarries. Regarding «Corciano Monuments», the 2 samples from the lion statues are associated to Saturnia, the 2 samples from S. Cristoforo Cathedral to Santa Sabina and Saturnia, the sample from the tower of the town to Santa Sabina, the sample form the Curch of S. Maria Assunta to Saturnia and the sample from the town walls to Cetona.

#### DISCUSSION AND CONCLUSIONS

Figure 6 reports results of the PCA for samples belonging to the quarries and clearly indicates that there are large overlapping regions for samples belonging to different quarries. This means that the discriminative power of PCA is very low, and for this reason we discard it as a suitable statistical method for provenance determination of travertine stones.

The application of the Self-Organizing Map and the Fuzzy Logic System shows that they are powerful tools to determine provenance of travertine stones from ancient buildings. The two systems give, in fact, very concordant results evidencing a good discriminative power of both systems that are able to recognize the exact provenance of quarry samples with very little uncertainties. The only exception is related to samples belonging to the quarry of Cetona. These samples are quite well discriminated from the other quarry samples by SOMs, but FLS shows some uncertainties and it does not classify univocally all samples. This occurrence may be due to the paucity of samples from Cetona that does not allow to perform a statistically representative analysis.

Although the two systems indicate a good general agreement, interpretation of SOMs gives, in some cases, additional information which cannot be inferred using the FLS. SOMs structure, in fact, allows to cluster similar samples and to separate different ones (Kohonen, 1998), whereas FLS associates anyway the monument sample to a quarry for any value of *I*. For example, consider some samples from a monument for which there is no quarry sampled. When the monument samples are processed by the SOM system they will be cluster far away from any existing quarry. On the contrary, the same samples processed by the FLS will be associated to the quarries with the highest value of *I*. This evidence induces us to select SOMs as primary technique and use FLS as a test for the results obtained by SOMs.

Application of AI systems (SOM and FLS) on Etruscan Arch samples indicates a provenance from the quarry of Santa Sabina, close to the town of Perugia (Fig. 1). This is in agreement with the fact that facies of Etruscan Arch and Santa Sabina quarries, as deducible by the fossil and mineralogical contents and by the lithotypes association, are practically the same and markedly different from those of the other quarry samples. Therefore results of AI and petrographic analysis point to a local provenance of travertine. Our results are also in agreement with archaeological and historical data. In fact, provenance from a rich basin located near the site of construction is postulated by historians (e.g., Defosse, 1980) considering the great volume of material necessary to build the monument.

In the case of Orvieto Cathedral, sites of provenance of travertine stones are identified by AI techniques in quarries occurring in the surrounding of the town (Tordimonte, Porano, and Orvieto; Fig. 1). These data are in good accordance with textural analysis. In particular they agree for the lithological assemblage, the type and abundance of the fossil component and, remarkably, the composition of the magmatic siliciclastic component.

Our results point to a local provenance, from the travertine lenses inset in the local pyroclastic formation, for all the travertines from the Cathedral. It is to note that historical documents (Riccetti, 1988) report that part of the material employed in the building of the Cathedral was extracted in the area of Cetona. Our analysis, instead, shows that no sample employed in the Cathedral belong to such an area. In fact, SOM does not evidence any significant similarity between Cetona and Orvieto Cathedral samples and this result is confirmed by FLS. Furthermore, the total absence of any magmatic siliciclastic component in the samples from Cetona is in contrast with the presence of pyroxene, K-feldspar, biotite, chlorite, titanite and epidote in the samples from Orvieto Cathedral (see Table 3).

The hypothesis that Cetona did not provide the travertine for the Cathedral is not completely in contrast with historical sources, since the exact lithology of the materials from each quarry is not specified in the documents. Therefore it is possible that Cetona provided some kind of building material other than travertine or, more likely, that travertine was really extracted near Cetona, but due to its low value it was intended for the production of lime.

Regarding Fontana Maggiore results of SOM evidence two clusters of samples not attributable to any of the sampled quarries and outcrops. On the other hand, results of FLS point to affinities between samples from Fontana Maggiore and the four quarry districts taken under consideration. Although at first sight these results may appear contradictory, it is to note, as discussed above, that the great dispersion of results given by the Fuzzy Logic System can be also interpreted as related to the fact that, actually, outcrops of travertine employed in the Fontana Maggiore have not been sampled. Therefore travertine from Fontana Maggiore could come from two different quarry districts or zones of excavation.

Results of petrographic analysis point to the presence of stromatolithic, microhermal and detrital travertine lithotypes in Fontana Maggiore. Stromatolithic and detrital travertines are characterized by the presence of a magmatic component, whereas microhermal travertine lacks of any magmatic minerals (Table 3). Considering this textural evidence two groups of samples can be distinguished, the first constituted by stromatolithic and detrital travertine, the second represented by microhermal travertine, and these groups, on turn, exactly correspond to the clusters identified by the SOM. Therefore, results of petrographic analysis confirm those of AI techniques supporting the existence of two zones of provenance for the material employed in the Fontana Maggiore.

Composition of pyroxenes occurring within stromatolithic and detrital samples points to a provenance from travertine deposits inset in the volcanic units of the Roman Magmatic Province. However, the SOM system clearly separates these samples from those of «Orvieto district» quarries. In addition, values of Sr of these samples are in agreement with the values of travertines from northern Latium (Barbieri *et al.*, 1979). Therefore, a general provenance from northern Latium may be invoked for the pyroxene bearing travertine of Fontana Maggiore.

The employment of non-local travertine in the construction of Fontana Maggiore seems to be quite surprising considering the intense quarrying activity in Santa Sabina area up to recent times. However, it is important to note that travertine from Rome was employed in its construction (Ferretti, 2001) supporting the original employment of travertine from Latium as re-utilized material coming from ancient Roman buildings as in the case of marble (Moroni et al., 2002). The fountain suffered also a number of changes and substitutions during the centuries in order to remedy the decay of the constituent material. The employment of different types of travertine in the subsequent restorations suffered by the monument seems, hence, to be reasonable and they could be the microhermal samples for which no definitive inferences can be advanced.

In the case of the monuments from Corciano, results of SOM evidence a single group of samples not attributable to any of the sampled quarries and outcrops. On the other hand, results of FLS point to the presence of affinities between samples from «Corciano Monuments» and all the four quarry districts taken under consideration. As in the case of Fontana Maggiore, the great dispersion of results given by the Fuzzy Logic System can be interpreted as related to the fact that outcrops of travertine employed in the «Corciano Monuments» are not sampled.

Results of textural analysis on the travertines from «Corciano monuments» show great affinity of microhermal samples and facies relationships between microhermal and detrital samples. These results support the hypothesis of a single quarrying area for these travertines. Comparison between the samples from Corciano monuments and the quarry samples from Santa Sabina show general affinities among microhermal travertines, but differences regarding the presence of quartz and the relative abundance of the different lithotypes. These similarities and differences support, therefore, that the quarrying area was possibly located in surroundings of Santa Sabina.

In conclusion, results of the application of artificial intelligence techniques and petrographic analysis to the determination of provenance of travertines led to the identification of a strictly local provenance for the samples from Etruscan Arch, Corciano Monuments and Orvieto Cathedral, and a non-local provenance for at least one group of samples from Fontana Maggiore. Travertine is a material whose commercial value is generally not so high to support the costs of transport over long distances. For this reason, and in consideration of the great abundance of travertine formations of good quality outcropping all over Central Italy, local provenance can be considered a common feature of travertines employed at any age in the past. Non-local provenance furnishes information on the historical background in which a monument was conceived and built. In the case of Fontana Maggiore, for example, provenance from ancient Roman monuments clearly testifies the great cultural influence of roman customs over Perugia institutions in the period in which the Fountain was built.

The obtained results are promising enough to encourage future research based on integrated approaches involving petrography and Artificial Intelligence technique for archaeometric purposes.

#### ACKNOWLEDGMENTS

We are grateful to R. Mazzuoli and an anonymous referee for helpful suggestions and criticisms in reviewing the manuscript. The editorial handling of B. Messiga is gratefully acknowledged. This work was founded by CNR and «Centro di Eccellenza – Tecnologie Scientifiche Applicate alla Ricerca Archeologica agli Studi Storico Artistici» grants.

#### REFERENCES

- BARBIERI M., MASI U. and TOLOMEO L. (1979) Origin and distribution of strontium in the travertines of latium (Central Italy). Chem. Geol., 24, 181-188.
- BAXTER M. (1994) Exploratory multivariate analysis in archaeology. Edimburg University Press, Edimburg, 307 pp.
- BISHOP C.M. (1996) Neural Networks for Pattern Recognition. Oxford University Press, Oxford, 256 p.
- BUXEDA J., GARRIGOS I., CAU ONTIVEROS M.A. and KILIKOGLOU V. (2003a) — Chemical Variability in Clays and Pottery from a Traditional Cooking Pot Production Village: Testing Assumptions in Pereruela. Archaeometry, 45, 1-17.
- BUXEDA J., GARRIGOS I., JONES R.E., KILIKOGLOU V., LEVI S.T., MANIATIS Y., MITCHELL J., VAGNETTI L., WARDLE K.A. and ANDREOU S. (2003b) — Technology Transfer at the Periphery of the Mycenaean World: The Cases of Mycenaean Pottery Found in Central Macedonia (Greece) and the Plain of Sybaris (Italy). Archaeometry, 45, 263-284.
- CIPRIANI N., MALESANI P. and VANNUCCI S. (1977) — *I travertini di Rapolano Terme (Siena)*. Boll. Serv. Geol. It., **98**, 85-105.
- D'ARGENIO B. and FERREERI V. (1988) Caratteri sedimentologici e paleoambientali dei travertini pleistocenici dell'Italia centro meridionale. Atti 74° Congr. Soc. Geol. It.
- DEFOSSE P. (1980) Les remparts de Pérouse. Contribution à l'histoire de l'urbanisme préromain, in Mélanges d'Archéologie et d'Histoire de l'Ecole Francaise de Rome XCII, Paris, 725-780.
- FAUSETT L. (1994) Fundamentals of Neural Networks: Architectures, Algorithms, and Applications. Prentice Hall, Upper Saddle River, New Jersey, 461 p.
- FORD T.D. and PEDLEY H.M. (1996) A review of tufa and travertine deposits of the world. Earth Sci. Rev., **41**, 117-135.
- FERRETTI L. (2001) La Fontana Maggiore di Perugia: archeometria storica dei materiali lapi-

*dei*. Tesi di Laurea, Università degli Studi di Perugia.

- FRANZINI M. and LEONI L. (1972) A full matrix correction in X-ray fluorescence analysis of rock samples. Atti della Società Toscana di Scienze Naturali residente in Pisa. Memorie, A79, 7-22.
- HALL M.E. (2001) Pottery Styles during the Early Jomon Period: Geochemical Perspectives on the Moroiso and Ukishima Pottery Styles. Archaeometry, 43, 59-75.
- KAYE M.J. (1965) X-ray fluorescence determinations of several trace elements in some standard geochemical samples. Geochim. Cosmochim. Acta, 29, 139-142.
- KLIR G.J. and YUAN B. (1996) Fuzzy Sets, Fuzzy Logic, and Fuzzy Systems: Selected Papers by Lotfi A Zadeh. World Scientific, Singapore, 826 p.
- KOHONEN T. (1989) Self-Organization and Associative Memory. Springer-Verlag, Berlin, 312 p.
- KOHONEN T. (1998) *The self-organizing map*. Neurocomputing, **21**, 1-6.
- LOPEZ-MOLINERO A., CASTRO A., PINO J., PEREZ-ARANTEGUI J. and CASTELLO J. R. (2000) — Classification of ancient Roman glazed ceramics using the neural network of Self-Organizing Maps. Fresenius J. Anal. Chem, **367**, 586-589.
- MORONI B. and POLI G. (1999) Provenance of materials from the «Fontana Maggiore» of Perugia (Italy). Proceedings 2nd International Congress on «Science and Technology for the safeguard of cultural heritage in the Mediteranean Basin», Paris, France.
- MORONI B. and POLI G. (2000) Provenance of

materials employed in the construction of Orvieto Cathedral (Umbria, Italy). Per. Mineral., **69**, 143-163.

- MORONI B., PERUGINI D., POLI G. and TURI B. (2002) — Characterization of Marbles by Textural Analyses and Stable Isotope Geochemistry: Examples from the «Fontana Maggiore» of Perugia (Italy), Proceedings of The Second National Congress of Italian Association of Archaeometry, AIAr 2002 «Science and Cultural Heritage», Bologna, Italy, 437-446.
- OUENES A. (2000) Pratical application of fuzzy logic and neural networks to fractured reservoir characterization. Comp. Geosci., 26, 953-962.
- PETRELLI M., PERUGINI D., MORONI B. and POLI G. (2003) — Determination of travertine provenance from ancient buildings using Self-Organizing Maps and Fuzzy Logic. Appl. Artif. Intell., 17, 885-900.
- PRINCIPE J.C., EULIANO N.R. and LEFEBVRE W.C. (1999) — *Neural and Adaptive Systems*. John A. Wiley & Sons, New York, 672 pp.
- REMOLA J.A., LOZANO J., RUISANCHEZ I., LARRECHIL M.S., RIUS F.X. and ZUPAN J. (1996) — New chemometric tools to study the origin of amphorae produced in the Roman empire. Trends Anal. Chem., **15**, 137-151.
- RICCETTI R. (1988) Il Duomo di Orvieto. Edizioni Laterza, Bari.
- RODOLICO F. (1965) *Le pietre delle Città d'Italia*. Edizioni Le Monnier, Firenze.
- YANG Y., XU X. and ZHANG W. (2000) Design neural networks based fuzzy logic. Fuzzy Sets and Systems, 114, 325-328.