

NEW U-SERIES DATING OF A CARBONATE LEVEL UNDERLYING THE PEPERINO ALBANO PHREATOMAGMATIC IGNIMBRITE (COLLI ALBANI, ITALY)

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ABSTRACT

Increasing attention has been recently devoted to the last phases of the Colli Albani activity (Rome, Italy), in consideration of new stratigraphic data suggesting a possible Holocene activity of the volcanic complex located few kilometres to the south of the city of Rome. In particular the focus has been directed to the activity of the Albano *maar* and related pyroclastic deposits, the youngest eruptive centre of the Colli Albani volcano.

The most recent dated product from the Albano *maar* is known as Peperino Albano (PA) ignimbrite, which has been dated with several different techniques. The age of the PA spans between 51 and 19 ka. A recent study has shown that PA is overlain by at least other two pyroclastic deposits and several lahar deposits, all separated by paleosoils, making the recent eruptive history of the Albano *maar* rather younger than previously thought. The exact age determination of the recent succession is therefore a key issue to be tackled in order to evaluate the volcanic hazard in the Roman region.

The aim of this paper is to improve the chronology of the last eruptive episodes of the Albano *maar*, by dating a carbonate layer interbedded within the recent deposits using U-series disequilibria. The idea came out from the discordant results obtained by applying different dating methods to PA ignimbrite as a consequence of the difficulties connected with the application of classical isotope chronological methods to young volcanic products.

In order to date the carbonate layer, the total sample dissolution technique (TSD) has been preferred to using leachates alone because the leaching method gives reliable results only in the case of simple dissolution of the carbonate fraction, without any removal of U and Th isotopes from the detritic component. Three coeval sub-samples from the carbonate level underlying PA have been analysed, giving an age of 22.9 ± 6.7 ka, younger than other previous chronological data. The age limit of about 23 ka found for PA supports the hypothesis of a possible very recent activity of the Albano *maar*, justifying the increasing efforts devoted to improving the chronology of the last phreatomagmatic phases of Colli Albani volcano.

RIASSUNTO

Una crescente attenzione è stata recentemente dedicata alle ultime fasi dell'attività dei Colli Albani (Roma, Italia), in considerazione di nuovi dati stratigrafici che suggerirebbero una possibile attività olocenica del complesso vulcanico situato pochi chilometri a sud della città di Roma. In particolare, l'attenzione è stata focalizzata sul più recente centro eruttivo dei Colli Albani (*maar* di Albano) e sui depositi piroclastici ad esso associati.

Il più recente tra i prodotti datati del *maar* di Albano è un'ignimbrite conosciuta come Peperino di Albano (PA). Tale unità è stata datata con differenti tecniche e le età ottenute sono comprese tra 51000 e 19000 anni. Un recente studio mostra come PA sia ricoperto da almeno altri due depositi piroclastici e ulteriori due lahar, tutti separati da paleosuoli, dimostrando che la recente storia eruttiva del *maar* di Albano sia più giovane di quanto sinora ritenuto. L'esatta determinazione dell'età di questa successione risulta pertanto un obiettivo-chiave nella valutazione del rischio vulcanico nell'area romana.

Lo scopo di questo lavoro è fornire un approccio alternativo per migliorare la cronologia degli ultimi episodi eruttivi del *maar* di Albano, attraverso la datazione col metodo dei disequilibri nella serie dell'uranio di un livello carbonatico intercalato ai prodotti del *maar* sopra-mentionato. L'idea è scaturita dai risultati discordanti ottenuti applicando differenti metodi di datazione al peperino di Albano, riconducibili alle difficoltà spesso connesse alla datazione diretta di prodotti vulcanici recenti.

Al fine di datare il livello carbonatico, la tecnica della dissoluzione totale del campione (TSD) è stata preferita all'uso dei soli lisciviati poiché l'analisi di questi ultimi fornisce risultati attendibili solo in caso di una semplice dissoluzione della frazione carbonatica, senza la rimozione preferenziale di U e Th dalla componente detritica. Sono stati analizzati tre subcampioni coevi appartenenti al livello carbonatico sottostante il Peperino di Albano e l'età risultante è pari a 22900 ± 6700 anni. Questo dato è il più recente tra quelli sinora ottenuti e corrobora l'ipotesi di una possibile recentissima attività del *maar* di Albano, giustificando il crescente impegno speso nel migliorare la cronologia delle ultime fasi freatomagmatiche del vulcano dei Colli Albani.

Keywords: U-series dating, Peperino, ignimbrite, Colli Albani, Italy

Parole Chiave: datazioni, serie dell'uranio, Peperino, ignimbrite, Colli Albani, Italia

1. INTRODUCTION

The first attempt to date Colli Albani volcanic complex date back to about 35 years ago when Evernden and Curtis (1965) dated leucite crystals by K/Ar obtaining ages ranging from 706 to 268 ka. During the 1960's and 1970's the K/Ar method was further developed (Gasparini & Adams, 1969; Masi *et al.*, 1976; Biddittu *et al.*, 1979; Bernardi *et al.*, 1982) giving results compara-

ble with the previous data. In the same years other radiometric methods based on U-series disequilibria (Th/U isochrons and fission tracks) were used (Cerrai *et al.*, 1965; Taddeucci, 1969; Bigazzi & Ferrara, 1971) supporting the occurrence of recent volcanic activity in the interval 67-43 ka. In the 1980's and 1990's, $^{39}\text{Ar}/^{40}\text{Ar}$ method was associated to K/Ar and Th/U systematics (e.g. Karner and Renne, 1998). The application of the three methods to the main volcanic units permitted the

improvement of the chronological knowledge of the complex (Radicati Di Brozolo *et al.*, 1981; Andretta & Voltaggio, 1988).

Notwithstanding the numerous papers dedicated to define the chronology of Colli Albani activity, various problems related to the different methods arose. The first, connected with the application of K/Ar method, was suggested by Villa (1988) who envisaged the presence of excess argon in the analysed materials. Further difficulties arose from the direct comparison of $^{39}\text{Ar}/^{40}\text{Ar}$ and Th/U methods applied to the same products. Villa (1992) and Voltaggio *et al.* (1994) found in three samples out of four systematically $^{39}\text{Ar}/^{40}\text{Ar}$ ages older than those obtained by Th/U, presumably due to excess argon in microscopic inclusions in leucite crystals. According to Villa (1988) the presence of excess ^{40}Ar can be identified from an accurate examination of the spectra, but if the excess ^{40}Ar is uniformly distributed (Voltaggio *et al.*, 1994) there is no way of distinguishing between radiogenic and inherited argon. Also the Th/U isochron method can be affected by problems, the main of which being related to the initial isotopic heterogeneity of the thorium activity ratio.

This paper focuses on the age determination using the U-series disequilibria of a carbonate level found at the base of the Peperino Albano (PA) ignimbrite, one of the most recent products from the Colli Albani volcano, erupted from the Albano *maar*. The age determination should be independent from the above mentioned uncertainty factors, and its importance is therefore twofold:

- 1) it helps at determining the controversial age of the Peperino Albano ignimbrite, which has been dated using ^{14}C , U/Th disequilibrium and thermoluminescence techniques (Fornaseri and Cortesi, 1989 and references therein; Mercier, 1993), with reliable ages spanning between 51,000 years ago and 19,000 years ago and clustering around 29-30 ka (Fornaseri and Cortesi, 1989).
- 2) it constrains the recent activity of the Colli Albani volcano which is much more complex and recent than usually believed, as the carbonate layer has been found at the base of a newly exposed stratigraphic section, where the Peperino Albano is overlain by several other phreatomagmatic and lahar units (Funciello *et al.*, 2002)

2. GEOLOGICAL SETTING

The Colli Albani volcano is a large composite caldera complex located about 20 km south-east of the city of Rome (Fig.1). It is one of the several volcanoes belonging to the NW-trending volcanic belt parallel to the Tyrrhenian coast of central Italy active during the last 600 ka, belonging to the Roman Magmatic Province (e.g. Serri, 1990; Ferrara and Manetti, 1993). The Colli Albani volcano is at present considered a quiescent volcano (De Rita *et al.*, 1995a) characterised by seismic and hydrothermal activity (Amato and Chiarabba, 1995; Calcara *et al.*, 1995).

The style of activity of the Colli Albani volcano has been characterised mainly by explosive activity throughout its volcanic history. The stratigraphy has been subdivided in three main successions corresponding to

three epochs of activity. The lowest, named the Tuscolano-Artemisio succession, was emplaced between less than 600 ka and 350 ka and comprises seven main large-volume, caldera-forming, low aspect ratio ignimbrite units. The Tuscolano-Artemisio succession comprises the most voluminous and widespread deposits of the Colli Albani volcano (Fig. 1) (De Rita *et al.* 1995b). The last large-volume ignimbrite-forming eruption caused the final collapse of the central 10 km x 10 km wide caldera at 350 ka (De Rita *et al.* 1988; De Rita *et al.*, 1995b). After the collapse, the volcanic activity changed drastically into strombolian and effusive accompanied by a significant reduction of the average mass eruption rate. During this epoch of activity, named Le Faete Epoch which lasted approximately from 350 ka and 270 ka, a small stratovolcano named Le Faete edifice was formed within the collapsed area (De Rita *et al.*, 1995b). The most recent volcanic activity of the Colli Albani volcano, the final epoch of activity, has been characterised by several, small volume, monogenetic phreatomagmatic eruptions from eccentric maars, whose products form the Final Hydromagmatic succession (De Rita *et al.*, 1995b). Phreatomagmatic eruptions occurred from single and coalescent craters. The Albano *maar* is the most recent of these craters, wherefrom the Peperino Albano ignimbrite erupted (De Rita *et al.*, 1986; Giordano *et al.*, 2002).

Different attempts have been carried out to date the Peperino Albano. A first was conducted on unburned *Ulmus* and *Quercus ilex* wood samples giving a ^{14}C

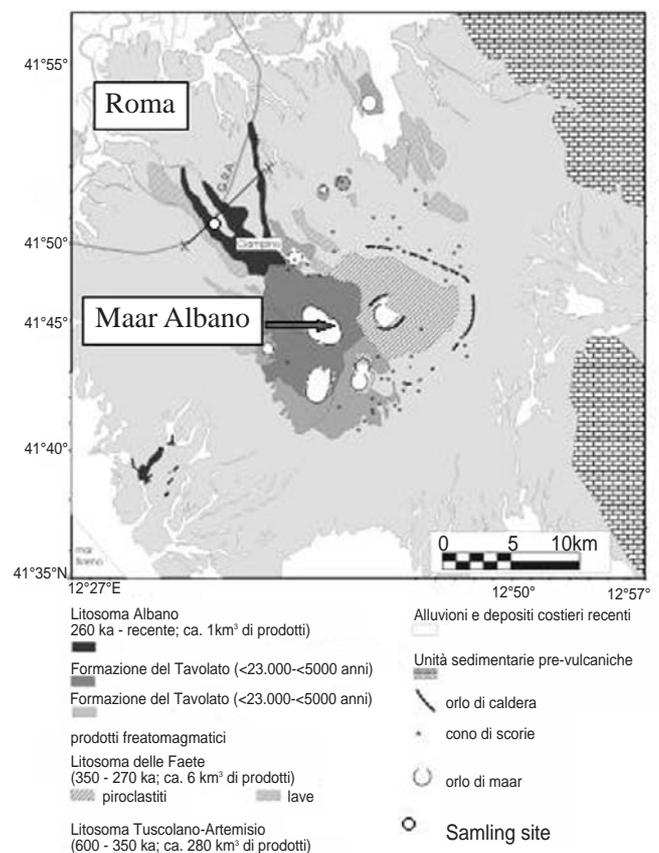


Fig. 1 - Geological map of Alban Hills area (Rome, Italy).

upper age of 29.7 ± 0.4 ka (De Vries, in Fornaseri *et al.*, 1963). Further radiocarbon determinations were performed on similar samples and the obtained ages were higher than 37 ka (Alessio *et al.*, 1966; Fornaseri, 1985). More recently Fornaseri and Cortesi (1989) reported a radiocarbon age of 29.9 ± 0.3 ka. This range is in agreement with Ar/Ar age determination from sediment-cores ascribed to PA eruption and drilled at the centre of the lake (Chondrogianni *et al.*, 1996). Also Villa *et al.* (1999) dated unaltered leucite crystals from other sediment-cores by Ar/Ar. They found that the evolution of the crater was a multistage process that took place in the last 45 ka. Two stages out of five (the first and the third), were dated at of 45 ± 3 and 26 ± 1 ka, respectively.

In addition, a Th/U isochron was realized using different mineral phases from the Peperino Albano (Andretta, 1988) with the resulting age of 51 ± 14 ka. Mercier (1993) used the thermoluminescence technique to date calcareous inclusions within PA obtaining an age of about 20 ka. Finally the recovering of uran-pyrochlore in sanidine ejecta of the IV Hydromagmatic units from the Albano *maar* has allowed to date the age of crystallization of these mineral phases by the U/Th method at 45 ± 1.3 ka (Votaggio and Barbieri, 1995).

3. STRATIGRAPHY

Figure 2 shows the stratigraphic section investigated in this study. This section was recently exposed thanks to roadworks related to the enlargement of the GRA freeway, in the Ciampino area, located along the NW slope of the Colli Albani volcano. The detailed stratigraphy of this section is reported in Funicello *et alii*, in press, to which the reader is referred to. The succession has been interpreted by the authors as the post-wurmian, possibly up to the Holocene filling of a paleovalley cut in the underlying Tuscolano-Artemisio epoch's deposits. The succession from base to top is composed by:

- a) fluvial lacustrine silt and sand
- b) phreatomagmatic surge deposit related to the IV eruption from the Albano maar
- c) fluvial sand and conglomerate
- d) Peperino Albano ignimbrite
- e) syn-eruptive reworked deposits from the Peperino Albano ignimbrite
- f) phreatomagmatic surge deposit
- g) debris flow lahar deposit
- h) hyperconcentrated flow lahar deposit

The carbonate level (fig. 3) was deposited at the top of unit a, which is composed of diffusely stratified silt and clay, likely from a CaCO_3 -rich swamp, similar to the

many swamps that occur in several places around the Colli Albani area, where CO_2 rich fluids upwell to the surface. It must be underlined that the Ciampino area is still the site of intense CO_2 release (e.g. Giordano *et al.*, 2000; Carapezza *et al.*, 2002)

4. SAMPLING AND METHOD

4.1 Theoretical considerations

Three coeval sub-samples from the carbonate level underlying the last erupted products from Albano *maar* were collected and analysed by U-series method, according to the total-sample dissolution (TSD) technique developed by Bischoff and Fitzpatrick (1991) for dating impure carbonates. The choice of such a method has been preferred to using leachates alone (Schwarcz and Latham, 1989) because the leaching method gives reliable results only in the case of selective dissolution of the carbonate fraction, without any removal of U and Th isotopes from the detritic component, or when U and Th are leached without any fractionation. Generally this condition is not verified because U and Th are often fractionated and Th can be reabsorbed onto the residual component. When analysing samples consisting of simple mixtures of carbonate and a detritic component, the

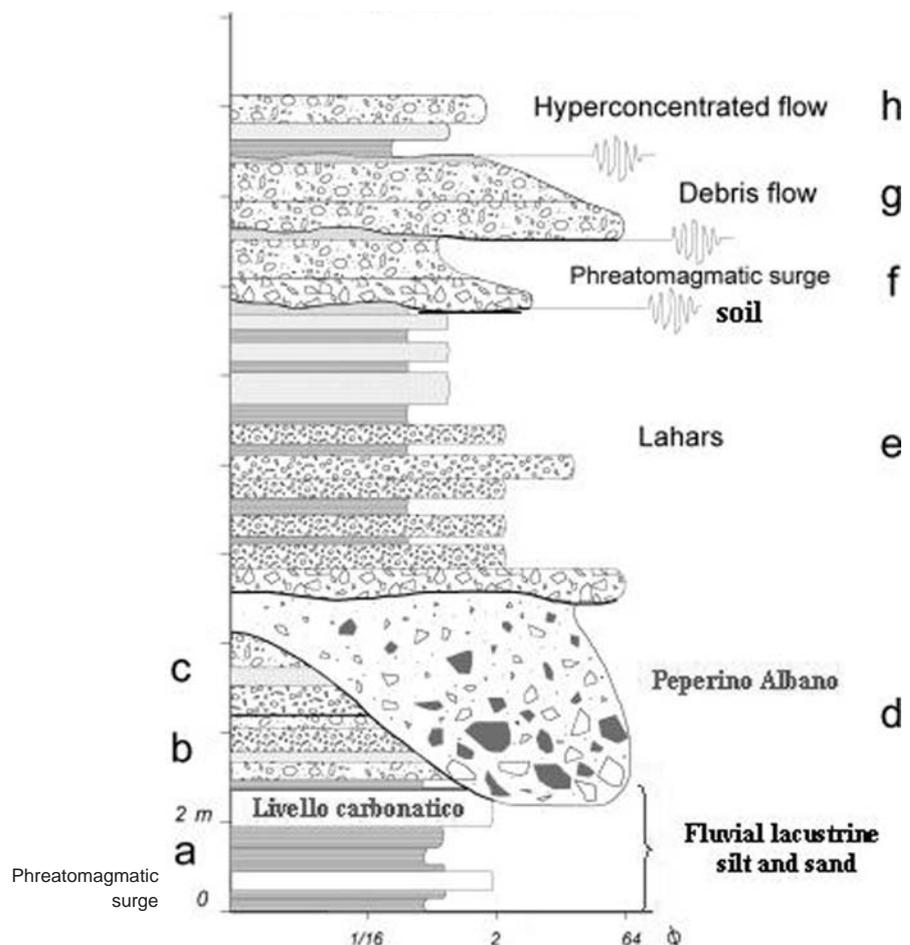


Fig. 2 - Stratigraphic section of Grande Raccordo Anulare (GRA) succession: a) fluvial lacustrine silt and sand, b) phreatomagmatic surge, c) fluvial sand and conglomerate, d) Peperino Albano, e) lahars, f) phreatomagmatic surge, g) debris flow, h) hyperconcentrated flow.

use of TSD method is to be preferred to determine the age because the sample is totally dissolved and consequently no preferential leaching or readsorption occur. In order to successfully apply this method it is required that the various coeval subsamples have different U/Th ratios, but the same ($^{230}\text{Th}/^{232}\text{Th}$) activity ratio when deposited.

TSD method assumes that: 1) the detritic component is homogenous and characterised by a constant U/Th ratio or 2) it is a heterogeneous mixture of two or more minerals with different U/Th ratio, but similar ($^{230}\text{Th}/^{232}\text{Th}$). In both cases the resulting age is that of formation of the carbonate.

The age of the carbonate level was calculated measuring the ($^{230}\text{Th}/^{232}\text{Th}$), ($^{234}\text{U}/^{232}\text{Th}$) and ($^{238}\text{U}/^{232}\text{Th}$) activity ratios of several coeval subsamples in order to obtain the value of ($^{230}\text{Th}/^{234}\text{U}$) and ($^{234}\text{U}/^{238}\text{U}$) activity ratios in the pure carbonate fraction. Such values are respectively calculated from the slopes of the regression lines in the ($^{230}\text{Th}/^{232}\text{Th}$) vs ($^{234}\text{U}/^{232}\text{Th}$) and ($^{234}\text{U}/^{232}\text{Th}$) vs ($^{238}\text{U}/^{232}\text{Th}$) isochron plots reported in Figure 4a and 4b. Calculations have been carried out using ISOPLOT, a plotting and regression program for radiogenic-isotope data (Ludwig, 1994).

4.2 Analytical technique

Samples were crushed and ultrasonically washed in deionised water. Fragments were also checked with a stereoscopic microscope to discard any recrystallised portions. About 10 g of sample were dissolved with 7 N HNO_3 . The insoluble residue was removed by centrifugation and successively digested in concentrated $\text{HF} + \text{HClO}_4 + \text{HNO}_3$, taken to dryness and redissolved in 7 N HNO_3 . The resulting solution was mixed with the carbonate solution and spiked with a tracer containing ^{228}Th in secular equilibrium with ^{232}U . U and Th were coprecipitated with $\text{Fe}(\text{OH})_3$ using NH_4OH and successively redissolved in 10 N HCl and loaded onto anionic Dowex AG1-X8 resin to separate Th from U. Thorium fraction was furthermore purified onto a second anionic Dowex AG1-X8 resin conditioned with 7 N HNO_3 and eluted with 10 N HCl . U and Th isotopic complexes were extracted from nitric solutions respectively at pH 3.5 and 1.5 using thenoyltrifluoroacetone (TTA) in toluene and loaded onto a glass planchet and flamed. U and Th were finally alpha-counted using high resolution ion implanted Ortec silicon surface barrier detectors.

5. RESULTS

Uranium and thorium concentration, activity ratios and ages are summarised in Table I. Quoted uncertainties are 1 standard deviation (± 1 sigma).

Resulting isochrons are shown in Fig. 4a and 4b. Data points are finely distributed on the ($^{230}\text{Th}/^{232}\text{Th}$) vs ($^{234}\text{U}/^{232}\text{Th}$) and ($^{234}\text{U}/^{232}\text{Th}$) vs ($^{238}\text{U}/^{232}\text{Th}$) isochron plots and arranged in two straight lines whose correlation coefficients R are higher than 0.96. The obtained age is equal to 22.9 ± 6.7 ka.

Uranium abundances of different subsamples range from 4 to 4.6 ppm, thorium contents vary from 4.7 to 9.6 ppm and ($^{234}\text{U}/^{238}\text{U}$) activity ratio in the pure carbonate fraction is around 1. Such data demonstrate that

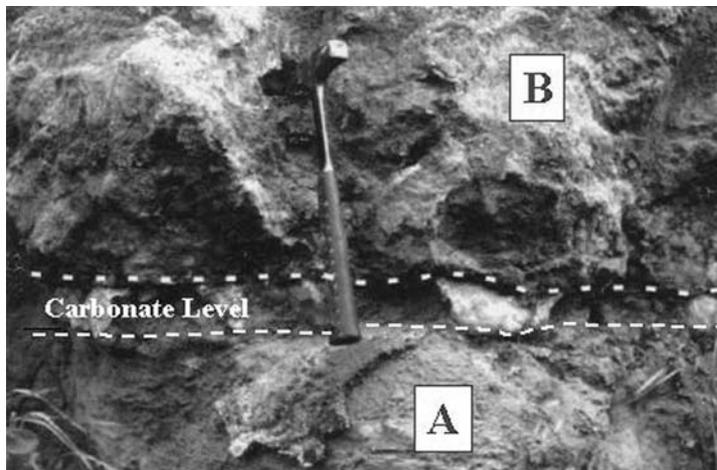


Fig.3 - View of the dated carbonatic level in layer A (fluvial lacustrine sand and silt), close to the limit with the overlying level B (phreatomagmatic surge), as in figure 2.

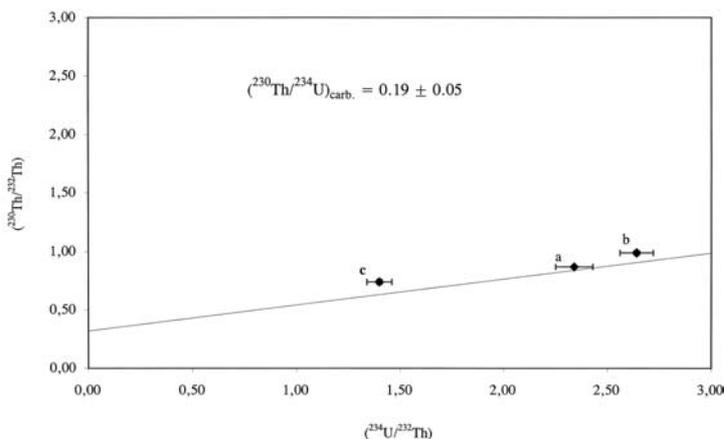


Fig. 4a - ($^{230}\text{Th}/^{232}\text{Th}$) versus ($^{234}\text{U}/^{232}\text{Th}$) isochron plot of the three carbonate sub-samples from GRA section.

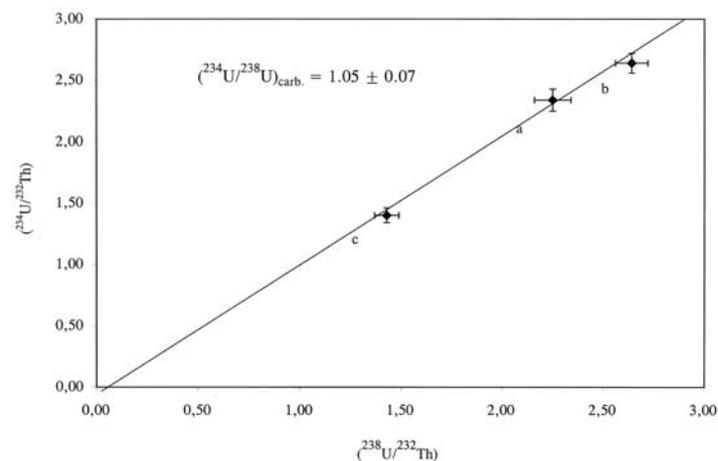


Fig 4b - ($^{234}\text{U}/^{232}\text{Th}$) versus ($^{238}\text{U}/^{232}\text{Th}$) isochron plot of the three carbonate sub-samples from GRA section.

TABLE I - U-series isotope data and resulting age of the carbonate level from GRA section.

Subsample	$(^{230}\text{Th}/^{232}\text{Th})$	$(^{238}\text{U}/^{232}\text{Th})$	$(^{234}\text{U}/^{232}\text{Th})$	U ppm	Th ppm	$(^{234}\text{U}/^{238}\text{U})_{\text{carb.}}$	$(^{230}\text{Th}/^{234}\text{U})_{\text{carb.}}$	Age (ka)
GRA a	0.87 ± 0.01	2.25 ± 0.09	2.34 ± 0.09	4.02 ± 0.01	5.37 ± 0.10			
GRA b	0.99 ± 0.02	2.64 ± 0.08	2.64 ± 0.08	4.21 ± 0.09	4.73 ± 0.12	1.05 ± 0.07	$0,19 \pm 0.05$	22.9 ± 6.7
GRA c	0.74 ± 0.02	1.43 ± 0.06	1.40 ± 0.06	4.64 ± 0.10	9.61 ± 0.30			

carbonate depositing fluids have circulated within a volcanic bedrock usually characterised by high uranium content and $(^{234}\text{U}/^{238}\text{U})$ activity ratios close to equilibrium. In addition, the high obtained thorium concentrations are consistent with a volcanic nature of the insoluble residue.

6. DISCUSSION AND CONCLUSION

The Peperino Albano ignimbrite erupted from the Albano maar has been dated several times by the application of different methods. Resulting ages differ significantly from each other and range between 51 and 19 ka. As mentioned before, data from the direct analyses of crystals of PA are affected by various problems of isotope disequilibria, but also ^{14}C determinations on carbon-rich samples in volcanic areas (leaves, branches etc.), where intense CO_2 emissions occur, can be unreliable (Calderoni and Turi, 1998). Actually, the carbon isotopic composition of organism living close to sites of intense CO_2 release in air can be presumably depleted in radiogenic carbon and consequently the obtained ages are not correct and result too old.

In light of this, it is necessary to produce an independent dating of PA in order to better constrain the chronology of last products from Albano maar. This aim has been reached with the determination of the age of the carbonate level outcropping in the GRA stratigraphic section and underlying PA. The U-series method using TSD (total sample dissolution) technique is to be preferred when analysing an impure carbonate with a relevant detritic component because it allows the overcoming of problems connected with the differential leaching and adsorption of U and Th isotopes from the insoluble residue. In view of this, the obtained results can be considered reliable.

The age of 22.9 ± 6.7 ka determined for the carbonate level indicate a very recent eruptive history of the Albano maar, rather younger than previously thought. This finding assumes a significant value especially in consideration of the recently described pyroclastic and lahar deposits overlying PA ignimbrite (see fig. 2). It is worth noting that such products are also separated by paleosoils indicating that a considerable span of time has to have been elapsed since the emplacement of the PA. This points to a possible Holocene age of the products filling the paleovalley cut into the Tuscolano-Artemisio volcanic succession to the NW of the Albano maar. Such considerations also allow the re-evaluation of datings from Andretta & Voltaggio (1988) who proposed a very recent activity of the Colli Albani complex, although that work did not clarify which stratigraphic layers were dated.

In conclusion, the recent activity of the Albano maar, needs more study in order to better define the volcanic hazard in the Colli Albani region because of the proximity of this area to a large city like Rome.

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