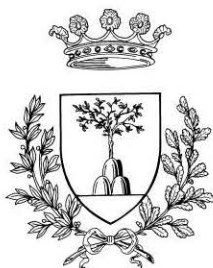


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Effect of deposits alteration on dating the animal teeth from Caune de l'Arago site by combined Electron Spin Resonance (ESR) and uranium series methods

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Abstract

Geochemical alteration process of the sediments in the archaeological site may affect the environmental radiation dose rate during the burial history, this will cause the inaccuracy of the age determination by radiation exposure dating methods, such as TL (thermoluminescence) or ESR (electron spin resonance) method. In Caune de l'Arago site, France, the original geochemical composition of the carbonate deposition in the layer Sol G, which found the Arago XXI skull, was modified, and formed a funneled phosphate section due to the acidification effect of guano during the historical period. Six teeth fossil samples from the different deposit environment of this layer were analyzed by combined ESR and U-series methods. The results show that the teeth samples from phosphate and carbonate area were consistent with the possible reference age – 450 ka in general, but the samples from non-altered area have given the age about 100 ka younger than others. The reason for this difference is still under discussion.

Keywords: geochemical alteration, Arago site, ESR dating, tooth fossil.

Introduction

The purpose of this study is to use the ESR/U-series combined dating on the middle Pleistocene site – Caune de l'Arago, France, and try to find the effect of different deposition environment on the dating results. The dating work of this site had been studied widely by many scientists in the recent thirty years, but due to the complex geochemical composition of the cave, there are still some chronological problems to this site. The analysis material in this study are mammalian teeth, which are from the Sol G of Arago site, combined ESR and U-series methods were used for comparing and discussing the uranium uptake model in the different deposit environment. The Caune de l'Arago is a large cavity on the limestone cliffs which border the Tautavel plain, in the Eastern Pyrénées of France. The thickness of the Quaternary deposits

in Caune de l'Arago is about 15 m and 25 archaeological layers have been observed. Each layer has reflected the human activities and their duration, and helps us to understand the type of human occupation (Fig. 1).

The archaeological layer Sol G comprises mainly the rock fragments brought by man and the bone fragments. This structure probably facilitates the circulation of water and therefore the water flowed from the cave wall can drain particularly passing through Sol G. Due to the acidification effect by the bat guano during the historical period, the original geochemical composition of the sediments in the cave had been modified and formed a funneled section. At present, the main three ensembles of sediments are aeolian sediments, stalagmitic floor, and phosphate breach (M. Icole *et al.*, 1981), the samples in this study were collected from these three sediments respectively (Tab. 1).

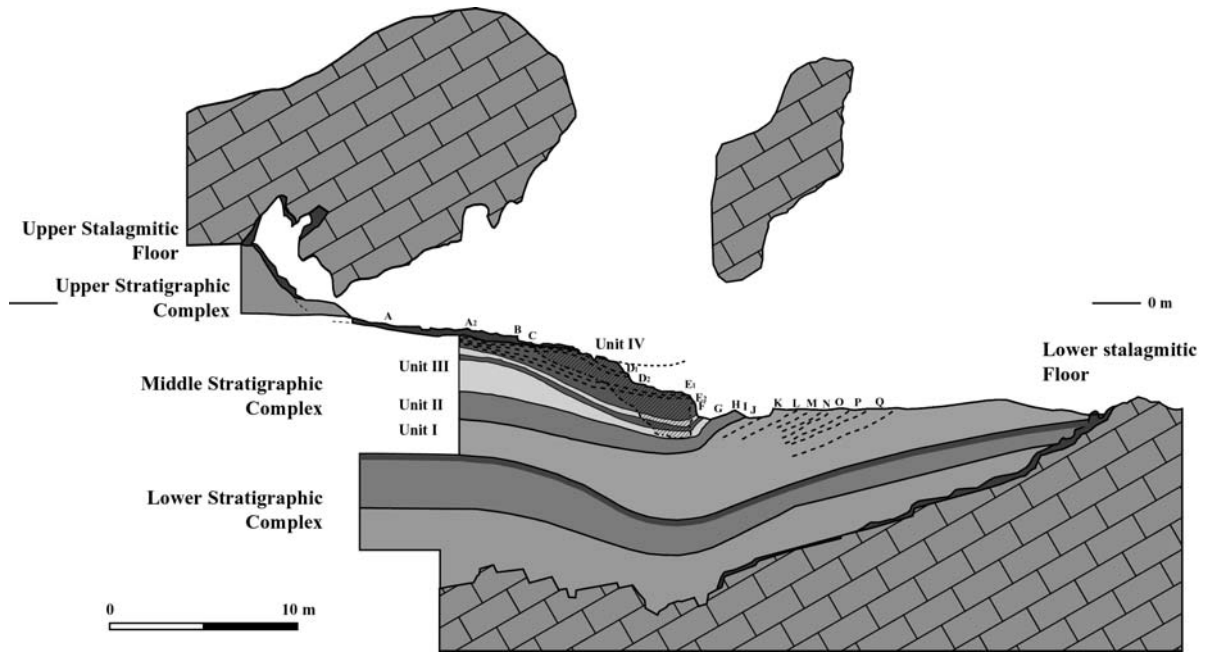


Fig. 1. Stratigraphic section of Caune de l' Arago (de Lumley *et al.*, 1984, modified by C. Falguères *et al.*, 2004).

lab No.	Genus(animal)	Type(tooth)	Spatial	Stratigraphic layer	No.inventory	X,Y,Z	Geochemical conditions
ARA0105	Ovis	M3 inf.	D18	Sol G	6179	61,100,624	G carbonate
ARA0106	Equus	M3 inf.	F20	Sol G	1718	54,49,584	G phosphate
ARA0108	Equus	P4 inf.	I18	Sol G	6864	63,54,553.5	G non altered
ARA0302	Ovis	M2.sup.(fragment)	D18	Sol G	5227	20,79,580	G carbonate
ARA0303	Equus	P4 inf.	F20	Sol G	3282	68,96,?	G phosphate
ARA0304	Rhinoceros	P3 inf.	J18	Sol G	6432	8,62,536	G non altered

Tab. 1. The location of the dating samples and their deposition environment.

Methods

The combined ESR (electron spin resonance) and uranium series methods were used in this study. ESR dating is an absolute dating method suitable for dating Quaternary sites. It is based on the detection of accumulated natural radiation damage resulting from environmental radioactivity. In this study, tooth enamel was used for analysis by ESR method, because it is the most fully mineralized and stable tissue of the tooth. There are two ESR spectra of tooth enamel, and we use the radiation sensitive signals at $g = 2.0018$ for ESR dating.

U-series dating methods are based on the measurement of the activity of uranium and its various daughter nuclides. In this study, the $^{230}\text{Th}/^{234}\text{U}$ dating method has been used to date each tooth tissues. The main problems encountered by U-series dating of dentine are

diagenesis and open-system behavior. The ideal situation for dating the teeth is that U taken up soon after burial and that the teeth remain closed system thereafter. In this paper, gamma and alpha particle spectrometry analysis has been used for U-series dating.

Although ESR dating can be used as a dating method of tooth enamel, it is mainly limited by the unknown uranium uptake history. The combination of ESR and U-series analysis has been considered as a powerful tool in the archaeological field, which permits to estimate the history of U-uptake by a p-value diffusion equation (Grun *et al.*, 1988). The whole procedure of combined ESR/ U-series dating of tooth enamel is illustrated by graph showing below (Fig. 2).

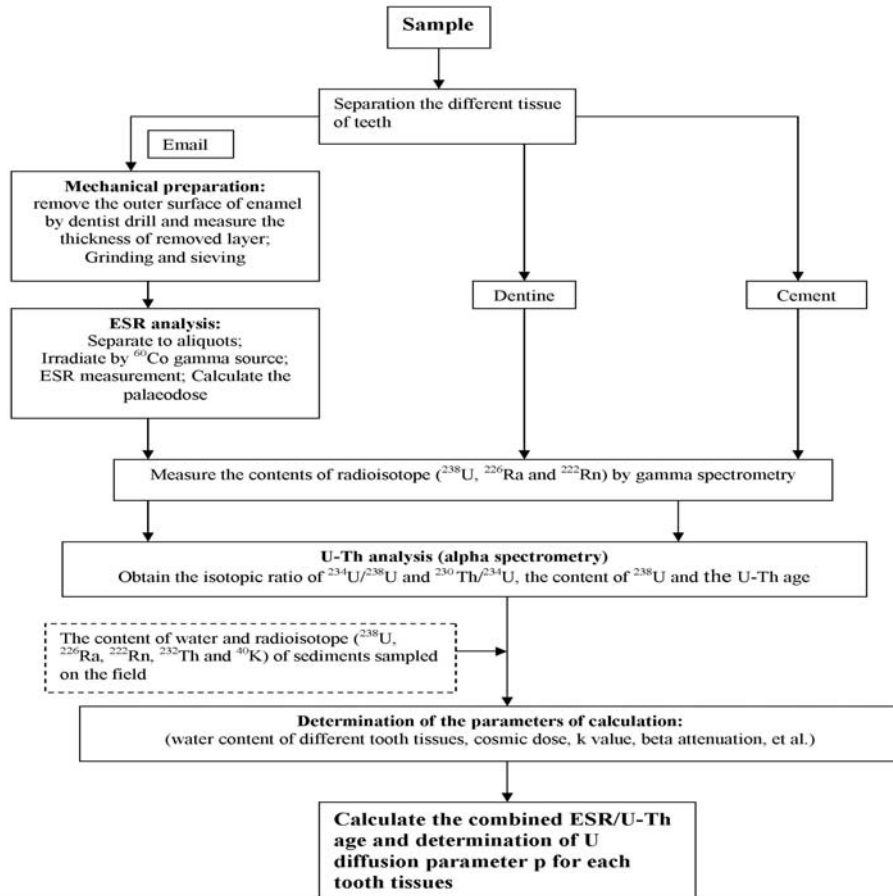


Fig. 2. The analysis procedure of combined ESR and U-series dating of tooth fossil.

Results

Table 2 presents the dose rate contribution and the combined US age with the p-values for each tissue. The calculated p-value range between -0.23 and 4.7 for enamel and from -0.64 to 0.04 for dentine, the US-ESR age are respectively 334 +33/-28 and 337 +19/ -20 ka, this results are younger than the other samples from phosphate and carbonate deposits, which show the p values varying from -0.87 to 5.07 (ARA0106 and ARA0303) and -0.73 to 6.64 for ARA0302 and ARA0105. It can be observed from the table that the samples for this study have the similar internal dose rate but much higher total dose for ARA0108 and ARA0304.

This is due to the higher external dose of these two samples than others (Figure 3).

Discussion and conclusion

According to the work of this study, the samples taken from different deposit

environment from Sol G in the Arago site have given us the consistent dating results in general

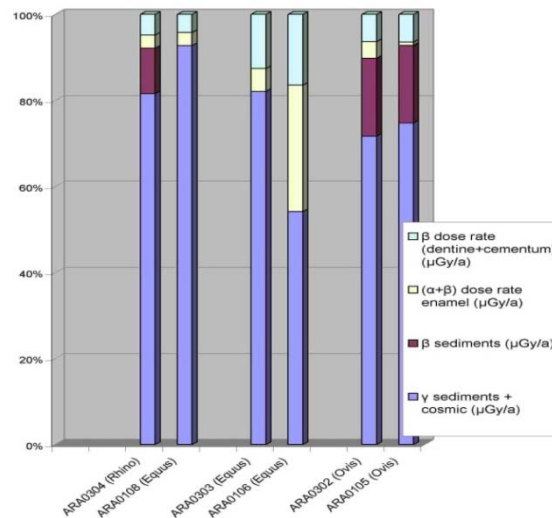


Fig. 3. The dose contribution from sediments, enamel and dentine to the total dose rate in percentage for samples from Sol G.

(Fig. 4), except the samples from non-altered area, which give unexpected younger age than other samples from other areas. This is quite possible caused by inhomogeneous deposit environment in non-altered area, which makes the accurate measurement of external dose of the samples very difficult. This phenomenon is not

only happened in Arago Cave, but also in other archaeological sites, when external dose rate makes a great contribution to total dose rate of the dating samples, so it is necessary to pay much attention on the external dose rate measurement in situ when taking samples for dating analysis.

Sample	γ sediments + cosmic ($\mu\text{Gy/a}$)	β sediments ($\mu\text{Gy/a}$)	US model				ESR and U-series combined data			
			Internal dose rate ($\alpha+\beta$) enamel ($\mu\text{Gy/a}$)	β dose rate dentine ($\mu\text{Gy/a}$)	β dose rate cement ($\mu\text{Gy/a}$)	Total dose rate ($\mu\text{Gy/a}$)	p enamel	p dentine	p cement	Age (ka)
non-altered area										
ARA0304 (Rhino)	662 ± 66	86 ± 10	25 ± 6	39 ± 5	N	812 ± 67	4.70	-0.64	N	334 +33 -28
ARA0108 (Equus)	662 ± 32	N	22 ± 14	14 ± 7	N	713 ± 37	-0.23	0.04	N	337 +19 -20
phosphated area										
ARA0303 (Equus)	352 ± 32	N	23 ± 17	39 ± 14	15 ± 5	428 ± 39	5.07	-0.28	0.03	551 +56 -50
ARA0106 (Equus)	352 ± 32	N	191 ± 49	102 ± 54	5 ± 20	649 ± 82	-0.83	-0.87	0	478 +38 -47
carbonated area										
ARA0302 (Ovis)	476 ± 47	120 ± 13	26 ± 13	42 ± 19	N	664 ± 54	-0.62	-0.55	N	474 +41 -39
ARA0105 (Ovis)	476 ± 47	115 ± 14	5 ± 3	41 ± 10	N	636 ± 50	6.64	-0.73	N	484 +48 -43

Tab. 2. Components of dose rates for US models of teeth and sediment from Sol G in Arago site.

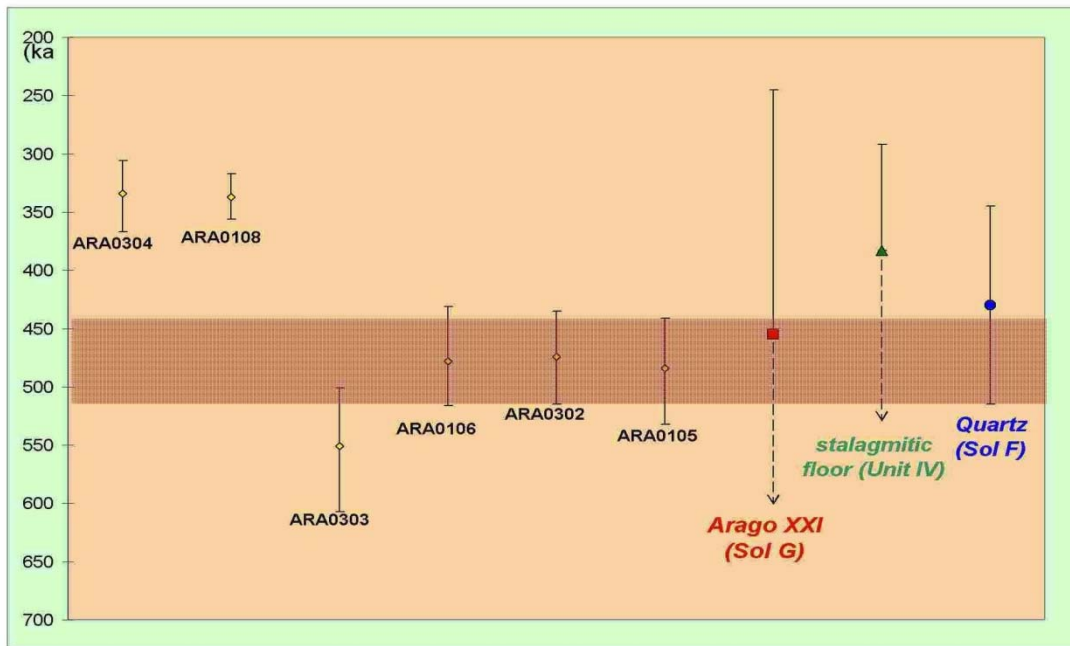


Fig. 4. The dating results in this study compared with previous date of different materials in Arago Cave.

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