Geologica Acta, Vol.5, Nº 1, 2007, 109-117
Available online at www.geologica-acta.com

U/Th dating of Quaternary travertines at the middle River Llobregat (NE Iberian Peninsula, Northwestern Mediterranean). Correlation with sea-level changes

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ABSTRACT

The dating of the fluvial terraces of the middle River Llobregat enabled one to establish a correlation between the sedimentation episodes in continental zones and in neighbouring deltaic and coastal areas in the NE Iberian continental margin. The fluvial Terraces 4 (+85-95 m above river level) and 3 (+55-65 m above river level) are made up by or include travertines, which were deposited around 350,000 and 111,000 years ago, respectively. These radiometric ages correspond to two high sea level periods: the Eemian interglacial (isotopic stage 5e) and the interglacial related to isotopic stage 9. However, other travertine units in the valley, such as the Can Tobella staggered cascade travertines, were formed during the Holocene, probably due to hydrothermal spring activity related to the tectonic contact between the Catalan Coastal Ranges and the Ebro Basin.

KEYWORDS **Travertine. U/Th dating. River dynamics. Sea-level changes. Pleistocene-Holocene.**

INTRODUCTION

The paleoenvironmental and paleoclimatic studies of Quaternary sediments have demonstrated the presence of high-frequency climatic fluctuations that affected the Earth at global scale (Bond et al., 1997; Broecker, 2000; Labeyrie, 2000; Ganopolski and Rahmstorf, 2001; Paillard, 2001; Schmittner, 2002). These global climatic changes, among other environmental factors, have given rise to high-frequency sea-level fluctuations (Dansgaard et al., 1989). These high-frequency eustatic changes have been detected by studying depositional architectures of fluvial deltas and coastal deposits (Mitchum and Van Wagoner, 1991; Stanley and Warne, 1993; Tornquist, 1994; Lowrie and Hamiter, 1995; Hernández-Molina et al., 1996; Lambeck, 1996; Somoza et al., 1997; Somoza et al., 1998). Sediment cores and seismic reflection profiles have improved our understanding of the resultant depositional geometry related to these high-frequency eustatic cycles (Somoza et al., 1998). The depositional geometry of the fluvial delta architecture (progradational, aggradational, retrogradational) results mainly from changes in sediment supply and relative sea level fluctuations (Postma, 1995). Moreover, the main factor that causes alternant erosion and sedimentation episodes in river valleys connected to sea is the development of these eustatic cycles.

The aim of this study is to start establishing a chronological model to date the erosion-entrenchment versus sedimentation episodes of the fluvial systems of the NE Iberian Peninsula, which resulted in the generation of fluvial terraces. The middle tract of the River Llobregat, which shows significant development of fluvial terraces, is taken as a particular case study to this end. The successive entrenchment and terrace generation of this river resulted in the deposition of typical fluvial clastic terraces and/or travertines. In this contribution, the term ''travertine'', in its broadest sense, refers to all non marine carbonate precipitates formed in/or near terrestrial springs, rivers, lakes, and caves (Sanders and Friedman, 1967).

The deposition of either clastic or travertine or mixed clastic-travertine fluvial terraces in the River Llobregat should be coeval to transgressive-high sea level situations (i.e. high base level). In contrast, the entrenchment episodes developed in the river valley zone should be coeval to regressive-low sea level situations (i.e. low base level).

Several authors have carried out geomorphological and stratigraphic studies in the Quaternary deposits of the River Llobregat (San Miguel and Marcet, 1928; Llopís Lladó, 1942; Solé Sabarís, 1953; 1963). The chronology of the Llobregat terraces had been determined previously on the basis of fossil mammal faunas collected from the terrace deposits (Solé Sabarís and Massachs, 1940; Solé Sabarís et al., 1957; Solé Sabarís, 1964). In this study, the radiometric dating of the travertines will allow us to establish a more precise chronology for the Late Pleistocene and the Holocene in the studied area. The determination of an absolute age for the fluvial terraces of the River Llobregat will improve our understanding of the factors that caused the successive entrenchment and sedimentation episodes. Moreover, this dating will constitute a reference chronological frame for the Quaternary sediments of the river basin as well as for future archaeological and palaeontological studies.

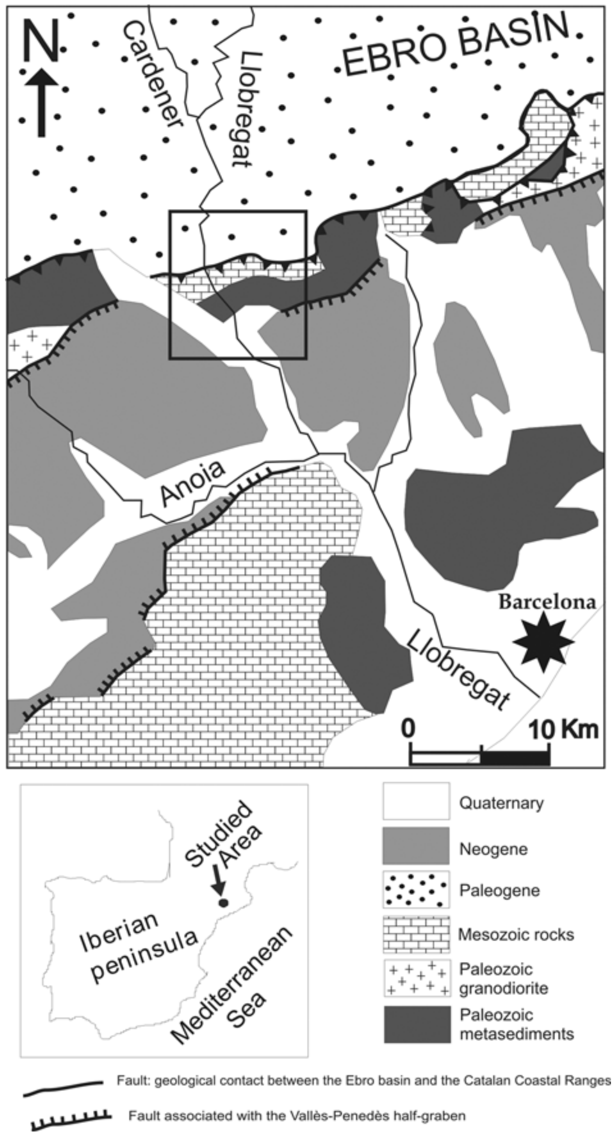

FIGURE 1 | **Geographical and geological setting of the studied area. The dated travertines are situated in the middle course of the River Llobregat (NE Iberian Peninsula). The middle course traverses sedimentary materials of Tertiary age crossing several geological units, which mainly constitute the contact between the Ebro Basin and the Catalan Coastal Ranges.**

GEOLOGICAL SETTING

The River Llobregat, which is located in the NE Iberian Peninsula, is 156.5 km long and has a drainage area of 4,948.4 km^2. The present study focuses on the travertine deposition that took place in the middle course of the River Llobregat (Figs. 1 and 2), in the province of Barcelona, between Esparreguera, Olesa de Montserrat and the Montserrat mountain (between 41º32'N-41º36'N, and between 1º51'E-1º53'E).

The middle course of the River Llobregat is underlain by Tertiary in age sedimentary rocks and crosses through several geological units that constitute the boundary between the so-called Ebro Basin and the Catalan Coastal Ranges. These geological units have been described in detail elsewhere (Roca et al., 1999; López-Blanco et al., 2000; López-Blanco, 2002). In the Ebro basin area, the River Llobregat flows through Paleocene-Eocene in age sedimentary sequences (e.g. the Montserrat Mountain). The River then traverses the Catalan Coastal Ranges, made up by a Variscan basement (Colonia Sedó area) unconformably overlain by a Mesozoic to Cenozoic cover (Can Paloma and Olesa de Montserrat areas, respectively).

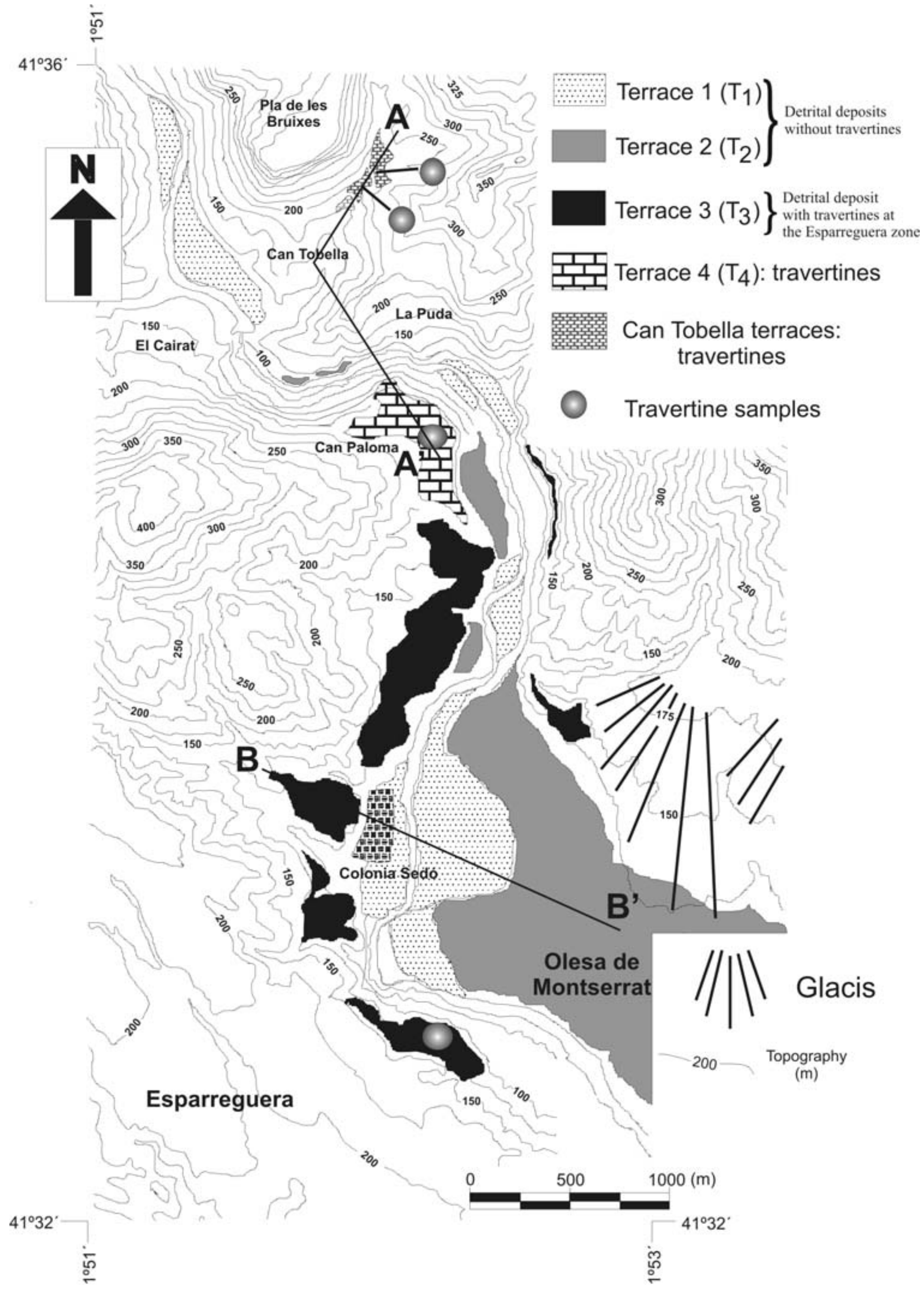

FIGURE 2 | **Fluvial terraces of the River Llobregat and location of the seven travertine samples. The travertine outcrops are situated at (from north to south): Can Tobella, Can Paloma (near La Puda), and near Esparreguera. Sections A-A’ and B-B’ correspond to geomorphological profiles shown in Figure 3A and 3B.**

Hydrothermal springs are frequent along this boundary as at La Puda spa, where a 20 m perched water table above the Llobregat riverbed occurs (Almela and Llopis Lladó, 1947). In addition to the hydrogen sulphide smelting rotten eggs (translation of the “La Puda” catalan name), chemical analysis report 129.28 $cm^{3}\cdot l^{-1}$ of carbonic acid and water temperatures between 32 and 28ºC (Arnús, 1855; Almela and Llopis Lladó, 1947; Bataller, 1933).

MATERIALS AND METHODS

Geomorphological mapping of the studied travertine units was carried out by means of aerial photography and fieldwork. The radiometric dating was performed on seven samples collected from travertine units located at diverse altitude in the fluvial valley (Fig. 2). Three samples were collected from the travertine deposits of Can Tobella (Can Tobella A, B-1 and B-2) that are characterized by their staggered arrangement and are not directly related to any fluvial terrace. Other three samples were gathered from the fluvial Terrace 4 (Can Paloma zone). Finally one sample was collected from the fluvial Terrace 3 at the vicinity of Esparreguera.

The travertine samples were dated at the Institute of Earth Sciences, CSIC (Barcelona), by the uranium-series disequilibrium dating method (Ivanovich and Harmon, 1992), using alpha spectrometry. The chemical separation and purification followed the procedure described by Bischoff et al. (1988). The isotope electrodeposition was carried out using the method described by Talvite (1972) and modified by Hallstadius (1984). Age calculations were based on the computer program by Rosenbauer (1991).

THE TERRACES OF THE MIDDLE RIVER LLOBREGAT

Geomorphological description

The Can Tobella travertines include three small travertine terraces, located near Can Tobella at the northwest of the study zone. This travertine system is staggered in a NE-SW oriented valley between 180-225 m a.s.l. (85-130 m above the present river level). This valley is very close to the tectonic contact between the Catalan Coastal Ranges and the Ebro Basin.

There are four fluvial terraces strictly related to the River Llobregat (Figs. 2, 3A and 3B). These terraces are distributed along an asymmetric valley, with steep slopes on the right margin (Esparreguera zone), and gentle slopes on the left margin (Olesa de Montserrat zone). Their relative heights above the present river level diminish progressively southwards. The Terrace 4 is located at 85-95 m above the river level. It only appears in La Puda zone, on the right river margin (Can Paloma). This terrace is characterized by the development of a laterally extensive travertine body that overlies unconformably the Mesozoic basement and shows changing thickness. The Terrace 3 is located at 55-65 m above the present river level. It mainly occurs on the right river margin and extends from La Puda to Esparreguera. It also occurs sporadically on the left river margin. This terrace includes some travertine deposits at Esparreguera. The Terrace 2 is at 20-30 m above the present river level. It constitutes the wide surface where the Olesa de Montserrat village is located. It appears sporadically on the right river margin near La Puda. The Terrace 1 is located at 8-10 m above the present river level. It crops out in the Olesa de Montserrat zone, in La Puda and in the Pla de les Bruixes zone.

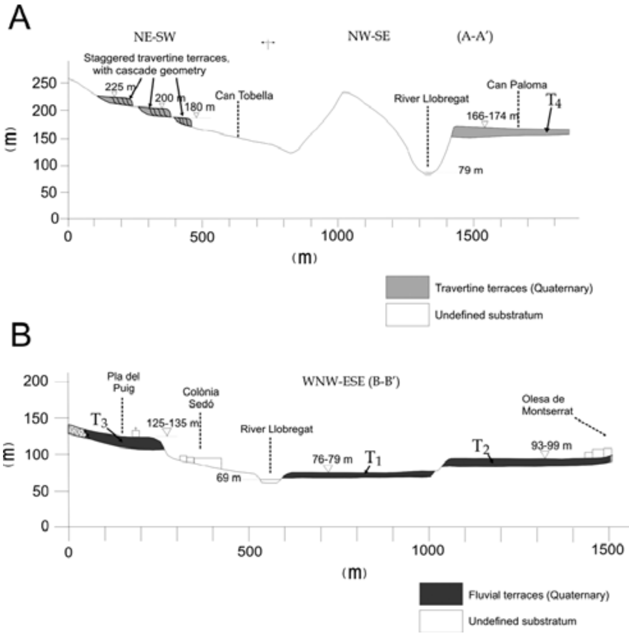

FIGURE 3 | **Geomorphological profiles of the studied area. The fluvial terraces of the River Llobregat (Terraces T_4, T_3, T_2 and T_1) and their absolute altitudes (m a.s.l.) are shown in the A and B profiles. Note that the staggered travertine terraces of Can Tobella show a cascade geometry. The situation of the profiles is indicated in Figure 2.**

Stratigraphy of the travertine deposits

Can Tobella travertine terraces

In the Can Tobella area (Figs. 2 to 4) the travertine bodies are staggered in three terrace levels and show clear cascade geometry. The lower terrace (terrace C) is situated at 180 m a.s.l. (85 m above the river level); the middle terrace (terrace B) is located at 200 m a.s.l. (105 m above the river level); and the upper one (terrace A) is situated at 225 m a.s.l. (130 m above the river level). These terraces show approximately similar thickness, ranging from 10 to

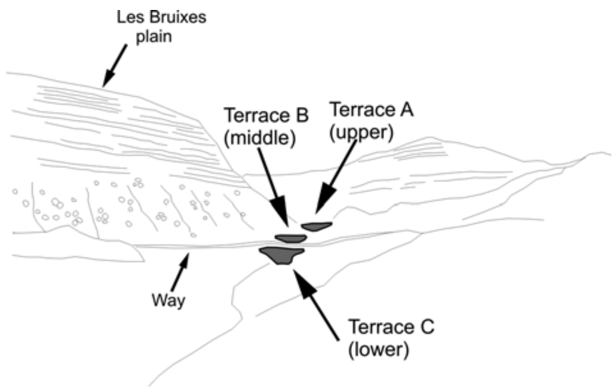

FIGURE 4 | **Schematic picture of the staggered travertine terraces of Can Tobella. The three small terraces are adapted to a SE-NW valley, which is located near the geological contact between the Ebro basin and the Catalan Coastal Ranges.**

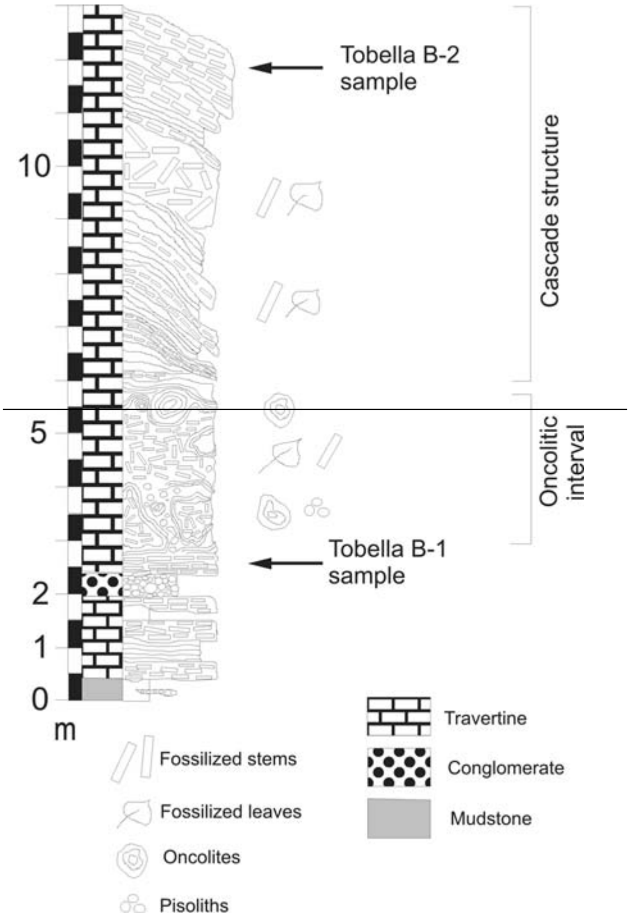

FIGURE 5 **Stratigraphy and sample position of the Can Tobella travertines (middle terrace of the staggered travertine system).**

12 m. Owing to erosion the inner structure for the middle terrace B is well observed. On he other hand, it is not easy to elaborate detailed stratigraphical sketches for terraces A and C, given the absence of erosion. The outcrop of the middle terrace of Can Tobella allows recognizing a complete sequence, from bottom to top (Fig. 5): 1) 0 to 2.5 m: Lower carbonate interval (fossil plant horizons) with detrital intercalations (shales and conglomerates). The encrusted stems parallelize the bedding surfaces; 2) 2.5 to 6 m: Intermediate oncolitic interval that bears decimeter in size oncolitic structures and conglomerate in size clasts. The encrusted stems are not parallel to the bedding; and 3) 6 to 13 m: Upper travertine body interval with a typical cascade structure that is well shown by the bedded horizons.

Can Paloma terrace (Fluvial terrace 4)

This terrace is a 25 m in thickness travertine body that overlies unconformably the Mesozoic substratum. The depocenter of this major travertine unit is mainly carbonate-dominated, whereas in the marginal zones (towards the S-SE) the detrital intervals become relatively frequent and the travertine beds are thinner. The stratigraphic log of Can Paloma was carried out in this marginal zone (Fig. 6). The dated samples were collected in an interval characterized by mudstone bed bearing travertine horizons.

Esparreguera terrace (Fluvial Terrace 3)

The Terrace 3 mainly occurs on the right river margin where it crops out from La Puda to Esparreguera. This terrace includes some travertine deposits at Esparreguera where the following lithological intervals can be distinguished (Fig. 7): 1) 0 to 4 m: Mainly reddish clay interval. It contains intercalated irregular in geometry polymictic breccias, as well as numerous carbonate nodules and small pisolitic levels. The clay beds are greyish in the upper meter of this sequence interval; and 2) 4 to 12 m: Travertines. At the bottom of this interval there is a 80 cm in thickness pisolitic horizon. The whole travertine interval displays pisolitic structures with encrusted stems.

Types of travertine bodies

The geomorphological, stratigraphical and sedimentological characteristics of the travertine units studied enabled us to establish two major types of travertine bodies:

1. Staggered travertines (cascade travertines) that correspond to the Can Tobella terraces. The cascade geometry indicates the perched character of these deposits. Plant fossils (mainly stem moulds and casts) are not broken and they remain parallel to the cascade bedding. Its geographic location, close to La Puda spa, suggests that these travertine terraces resulted from CO_2 outgassing from hydrothermal karstic springs (Arnús, 1855; Bataller, 1933; Almela and Llopis Lladó, 1947).

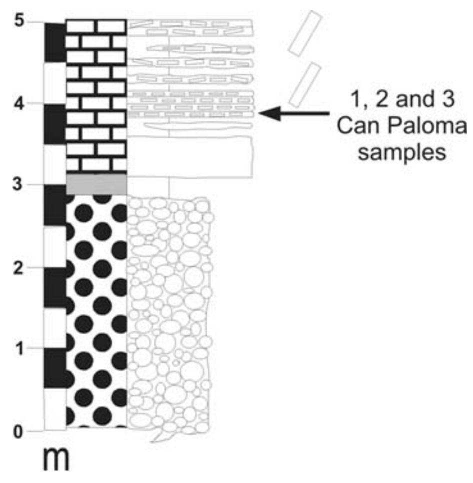

FIGURE 6 **Stratigraphy and samples position of the Can Paloma fluvial travertines (Terrace 4). See legend in Figure 5.**

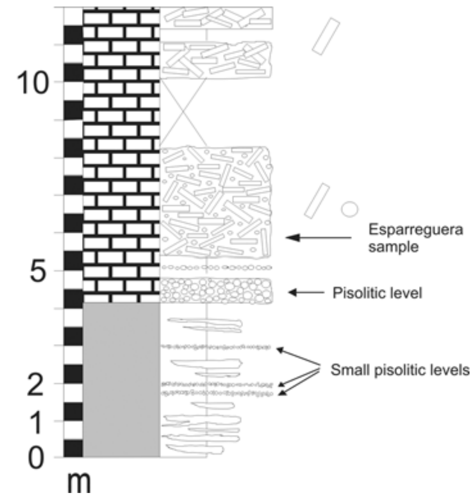

FIGURE 7 | **Stratigraphy and sample position of the fluvial Esparreguera travertines (Terrace 3, near Esparreguera). See legend in Figure 5.**

2. Fluvial travertines that correspond to the Can Paloma terrace (Terrace 4) and the Esparreguera terrace (Terrace 3). Textural and geometric features suggest a fluvial-related origin (i.e. riverine tufas after Andrews, 2006). Thus, the pisolitic structures and the horizontal geometry indicate that travertines had been deposited in marginal zones of the river. Karstic waters with a high carbonate content fed low flood plain areas affected by river flooding. The plant community that developed in this floodplain contributed to the precipitation of calcium carbonate. The intercalated mudstones would record successive river floods. These travertines are good indicators of ancient base levels of the River Llobregat.

U/Th DATING RESULTS

Results of the travertines dating are shown in Table 1. The different isotopic ratios and absolute ages obtained are given. The $^{230}Th/^{232}Th$ isotopic ratio is an indicator of the degree of sample contamination. The determination of this ratio indicates the reliability of the final results, because the absolute age shows older values when the sample is contaminated with terrigenous content. Julià and Bischoff (1991) suggest that when $^{230}Th/^{232}Th > 17$ the ages obtained are reliable, whereas when $^{230}Th/^{232}Th < 10$ the obtained dates for these samples are too old.

The Can Paloma fluvial travertine body (Terrace 4) yielded the oldest age of all the dated samples, showing an age in excess (or near the detection boundary) of the U/Th radiometric method (approx. 350,000 years). The Esparreguera travertine body (terrace 3) gave an intermediate age. In contrast, the Can Tobella staggered travertines yielded a Holocene age even though the $^{230}Th/^{232}Th$ ratios are <2.

DISCUSSION

Only two of the seven samples analyzed were slightly contaminated, one sample from Can Paloma 1 (Terrace 4), which yielded an age of 380,854 +79,521/-51,161 years; and the other sample from the Esparreguera zone (Terrace 3), which gave an age of 111,227 +3,518/-3,421 years.

We performed three radiometric analyses in one locality (Can Paloma 1, 2 and 3). The results (Table 1) show that this travertine sample does not provide evidence of high terrigenous contamination (since $^{230}Th/^{232}Th > 17$ is obtained by one analysis, and the other two yield values around 13). The obtained values suggest that travertine was deposited near the detection boundary of the U/Th radiometric method, around 350,000 years ago. The radioisotope ratios support this fact given that a) the relation $^{230}Th/^{234}U$ is very close to 1, indicating that both radioisotopes are close to equilibrium; and b) the calculated age is 380,854 years (+79,521/-51,161) for Can Paloma 1. By contrast, the age calculation did not converge (excess ^{230}Th) in the case of the samples from Can Paloma 2 and 3 (which are the most contaminated samples).

TABLE 1 | **U-series radiometric data and derived dates for travertines from the middle River Llobregat. Samples with significant detrital contamination are related to $^{230}Th/^{232}Th$ <10. This contamination, in general, renders too old nominal date calculated from $^{230}Th/^{234}U$. Samples of particularly high purity are related to $^{230}Th/^{232}Th$ >17 (in general, nominal dates in this category are of the highest confidence).**

U-series radiometric data

Sample	lab-ref	U ppm	Th ppm	$^{234}U/^{238}U$	$^{230}Th/^{234}U$	$^{230}Th/^{232}Th$	Age (years)
Esparreguera	38973	0.33	0.04	1.51 +/- 0.02	0.67 +/- 0.01	23.80	**111227** + 3518/-3421
Tobella A	39972	0.48	0.16	2.16 +/- 0.03	0.09 +/- 0.00	1.72	**9824** + 269/-268
Tobella B-1	40973	0.37	0.24	2.02 +/- 0.03	0.17 +/- 0.00	1.60	**19749** + 435/-433
Tobella B-2	50973	0.86	0.10	2.35 +/- 0.02	0.01 +/- 0.00	0.83	**1508** +/- 142
Can Paloma 1	21042	0.70	0.16	1.30 +/- 0.02	1.05 +/- 0.03	18.65	**380854** + 79521/-51161
Can Paloma 2	23043	0.68	0.23	1.35 +/- 0.01	1.07 +/- 0.02	13.31	**Age calculated do not converge**
Can Paloma 3	24041	0.72	0.25	1.35 +/- 0.02	1.09 +/- 0.03	12.96	**Age calculated do not converge**

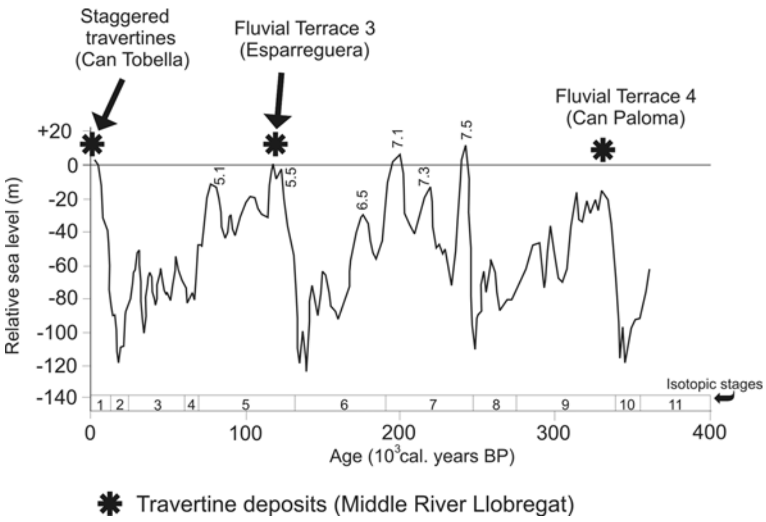

FIGURE 8 | **Relative sea-level fluctuations in the last 400,000 years (the last eleven isotope stages). The travertine deposits in the River Llobregat were accumulated during episodes associated with high sea levels. Thus, travertine from Can Paloma (Terrace 4) and from the Esparreguera zone (Terrace 3) were deposited during the isotopic stages 9 and 5e, respectively. Travertine from Can Tobella was deposited during the Holocene (isotopic stage 1), related to hydrothermal activity. Pleistocene sea-level curve is based on the detrended $\delta^{18}O_{water}$ record from Cocos Ridge core TR163-19 (adapted from Lea et al., 2002). Important sea-level high stands are indicated by their isotope stage numbering.**

This age, around 350,000 years ago, corresponds to the interglacial stage related to isotopic stage 9 (Fig. 8). During this time, River Llobregat was located at 85-95 m above the present river level (Terrace 4 level). Some time after the accumulation of the travertines, erosion gave rise to the Terrace 4 incision. This fluvial erosion led to a river level lowering (approx. 30 m) and must be associated with a sea-level drop.

Results given in Table 1 for Terrace 3 (Esparreguera fluvial travertines) show that the travertine sample does not provide evidence of terrigenous contamination (since $^{230}Th/^{232}Th > 17$), thereby demonstrating that the final result is reliable. The travertine body bottom was deposited approximately 111,000 years ago and was coeval to the Eemian interglacial stage, i.e. isotopic stage 5e (Fig. 8). During this time, the River Llobregat was located at 55-65 m above the present river level. Subsequently, the base level of the River Llobregat became stabilized, triggering the travertine deposition in Esparreguera (approx. 13 m in thickness). Some time after the accumulation of this travertine body, erosion produced the incision of Terrace 3. The river level lowering led to this erosion (approx. 35 m) and must be associated with a coeval sea level drop.

This study has not provided any chronological data related to the completion and end of the travertine growth at Esparreguera. Nevertheless, some previous studies carried out in other nearby travertine systems allow us to deduce the time when the Esparreguera travertine deposition stopped. Thus, the study performed in the travertine system of Abric Romaní (Barcelona), which developed in a geological setting similar to that of the Llobregat travertine system, have yielded additional information. The archaeological outcrop of Abric Romaní (Capellades area, River Llobregat basin, 15-20 km from Esparreguera) has enabled us to date the completion of the travertine sequence using also the U/Th method. The end of the travertine growth is dated at 43,300 (+/- 1,000) yr BP (Bischoff et al., 1988). Since the stratigraphical and topographical positions of the Capellades travertines are similar and correlatable with those of the Esparreguera travertines, a simultaneous deposition for both sequences can be assumed. Thus, the deposition of the Esparreguera travertines began at 111,000 (+/- 3,000) yr BP (Eemian interglacial or isotopic stage 5e) and ended at about 43,300 (+/- 1,000) yr BP (isotopic stage 3). Moreover, additional studies on the Alfaix travertines (Vera and Sorbas Depression, SE Iberian Peninsula) have demonstrated that the end of the travertine growth in this area occurred between 50,000 and 40,000 yr BP (Schulte, 2002).

The isotopic stages 7, 5 and 1 are commonly attributed to 'interglacial' climatic conditions, whereas the isotopic stages 6 and 4 are associated with 'glacial' conditions (e.g. Lea et al., 2002). Some palaeobiological records, such as the pollen sequences in the SE Iberian Peninsula (Florschütz et al., 1971; Pons and Reille, 1988; Burjachs et al., 1996; Schulte, 2002) and in the Abric Romaní (Burjachs and Julià, 1994), and the marine depositional record in the SW Iberian Peninsula (Sánchez-Goñi et al., 1999), provide evidence of wet conditions at the beginning of isotopic stage 3, thereby allowing the deposition of travertines. Furthermore, these studies also suggest wet climatic conditions during the isotopic stage 5.

The results from the Can Tobella travertines suggest terrigenous contamination, as indicated by the low values of $^{230}Th/^{232}Th$ (between 0.8-1.70). Therefore, the apparent dates of these samples are somewhat too old. Even so, the results (approx. 9,800 years, 19,700 years and 1,500 years) indicate that the Can Tobella terraces are Holocene. Thus, during the present interglacial, the karstic springs in the La Puda' area gave rise to the deposition of three perched travertine terraces which are not associated with the evolution of the ancient base level of the River Llobregat.

CONCLUSIONS

The travertine dating obtained from the middle River Llobregat fluvial terraces improve our understanding of the factors that gave rise to the successive entrenchment and depositional episodes in this river. Thus, fluvial travertines related to Terraces 4 (Can Paloma) and 3 (Esparreguera) were accumulated during high sea level periods in the Pleistocene. Terrace 4 was deposited near the detection boundary of the U/Th radiometric method, approx. 350,000 years ago (isotopic stage 9), whereas Terrace 3 was accumulated 111,000 years ago (Eemian interglacial or isotopic stage 5e), both at times of elevated base level. Nevertheless, the staggered Holocene travertines located at Can Tobella do not resulted from river base level evolution, but from the activity of likely hydrothermal karstic springs placed nearby the tectonic contact between the Catalan Coastal Ranges and the Ebro Basin.

ACKNOWLEDGEMENTS

We are indebted to Dr. Ferran Colombo who helped in the fieldwork and provided useful comments. We also acknowledge the help of Ana Maria Luque during the fieldwork. We should like to thank Graciela Monzón (Institute of Earth Sciences 'Jaume Almera', CSIC, Barcelona) for carrying out the radiochemical separation and the radioisotopic determinations.

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Manuscript received February 2005; revision accepted December 2006.