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Towards a New Chronostratigraphic Method Based on the Marine Sediment Radioactivity Variation

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ABSTRACT

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Sediment radioactivity (alpha and beta particle counting) from six cores from the Aegean and Ionian seas shows apparent variation identified with certain palaeoclimatic features during the Holocene and the last glaciation. The discussion of the radioactivity variation, the dating of the cores, the estimated sedimentation rates, and the palaeoclimatic recognition, is corroborated by the geotechnical properties of the sediments, some AMS C-14 dates, core intercomparison, and correlations with proxy climatic data.

ADDITIONAL INDEX WORDS: *Sediment, radioactivity, Aegean, uranium, spectrum, alpha counting, beta counting, palaeoclimate, Holocene.*

INTRODUCTION

The variation of the radioactivity with depth in lake and marine sediment was attributed to palaeoclimatic variation (DEMENOCAL *et al.*, 1992; LIRITZIS *et al.*, 1994; LIRITZIS and GALLOWAY, 1995) while possibly related solar periodicity has been noted (XANTHAKIS *et al.*, 1992). Reported variation in thermoluminescence down a core (*e.g.* CASTAGNOLI *et al.*, 1990) may well be related to variations in radioactivity.

The leaching/transportation of radioisotope-bearing minerals/clays and their deposition to lake/ocean basin occurs in a manner proportional to the climate variation (precipitation, temperature, wind). During these weathering-transportation processes, the natural radionuclides and partitioned in a way dependent upon their trapped sites (minerals) and the geochemistry of the reactions. (WEDEPOHL, 1978; IVANOVICH and HARMON, 1982).

Indeed, in geology it has been repeatedly documented that there is an almost universal tendency for an increase in the concentration of uranium and thorium in proportion to the concentration of later members—potassium for example—in any "igneous or metamorphic differentiation series". The linear and/or hyperbolic partitioning tendency of the three radioelements U, Th and K in the detrital rocks have also been extensively investigated. The K_2O% versus beta dose-rate relationships were derived for geological materials and archaeological ceramics (LIRITZIS, 1985, 1989; QUINIF *et al.*, 1982).

Moreover, uranium may be fractionated, U-234 being enriched in ground waters with respect to its parent, U-238. Uranium may be carried into the lake/ocean bottom in minerals such as zircon. Thorium is carried by minerals too, and potassium follows the geochemistry of thorium in most rock types and is carried also by minerals such as the feldspars. In warm periods, potassium (K) concentrations are low which implies a limited degree of weathering, *i.e.*, stability in soil production which is related to dense vegetation. The K:U:Th ratios in various rock types have been studied by various authors (QUINIF *et al.*, 1982; LIRITZIS, 1985, 1989; ADAMS *et al.*, 1959).

The rationale of the climate–sediment radioactivity relationship unfolds as follows: as the ultimate agent for weathering/erosion/transportation/deposition processes is climate, from purely geochemical reasons the deposition of sedimentary layers involves a partitioning of their radioisotopic content. Hence, their emitted energy in terms of absorbed dose (dose-rate) varies with depth in a proportional manner (U and Th are alpha- and beta-particle emitters, and K is only β-particle emitter). This radioactivity change with depth is measured here, and refers to alpha and beta particle counting per successive layers (LIRITZIS and GALLOWAY, 1995; XANTHAKIS *et al.*, 1992).

This report augments previous sediment radioactivity measurements on two cores from Lac du Bouchet in France (GALLOWAY, 1995a; LIRITZIS *et al.*, 1994), one core from Meerfelder Maar in Germany (GALLOWAY, 1995b, 1996), and two cores from the Black Sea (LIRITZIS and GALLOWAY, 1995).

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Since the commencement of this project (XANTHAKIS *et al.*, 1992), the reason for a continuing interest in the variation of the radioactivity with depth in lake and marine sediment core is towfold: a) such variation indicates paleoclimatic features, and b) variability offers time markers to date cores (*i.e.* it potentially offers a new, efficient, swift and unexpensive tool towards the establishment of a chronostratigraphy).

The present paper is based on the measurements from six representative cores along the Aegean sea, and off Corfu in the Ionian sea. These measurements show drastic change in sediment radioactivity at the end of the last glaciation.

The Aegean region is a marginal sea in the NE Mediterranean sea. Due to their complex geotechnic evolution it consists of numerous small scale basins (up to 1000 m depth) with extensive marginal plateau (MASCLE and MARTIN, 1990).

The Cretan Sea in the southern Aegean is the largest basin with mean depths of about 1600–1800 m. The sedimentation rates in the Aegean sea are generally low and vary between 5–15 cm·Kyr^{-1} in the basin margins and between 10–30 cm·Kyr^{-1} within the basins (PIPER and PERISSORATIS, 1991; LYKOUSIS *et al.*, 1995). Relatively lower sedimentation rates have been estimated in the southern Aegean relatively to the central and northern Aegean.

Therefore, the Aegean Sea should be regarded as a marginal sea with numerous small marginal sub-basins and consequently the sedimentary evolution is controlled by shallow rather than deep water processes. Besides, due to the very limited river input, the sedimentation rates are fairly uniform of basin wide origin, and reasonable correlations between sediment cores from different marginal sub-basins could be made.

METHODOLOGY

The direct measurement of radioactivity in this context has been by a-particle counting and β-particle counting (GALLOWAY and LIRITZIS, 1991). While β-particle counting of samples of natural radioactivity require substantial shielding against background radiation from the surroundings, the short range of α-particles ensures that no significant shielding problem arises for a particle counting with a detector insensitive to other radiations. The long established ZnS screen (LIRITZIS and GALLOWAY, 1982 a,b) has this property of sensitivity to α-particles and insensitivity to β-particles or γ-rays. With natural radioactivity the counting rate is inevitably low, so that with a single detector only one or two samples can be measured each day. To measure the hundreds of samples required to provide a detailed depth dependence from a sediment core of significant length it is attractive to use many detectors simultaneously. That ZnS α-particle counters do not require massive shielding and are basically simple to use and maintain, makes them attractive for this purpose. For this reason a system comprising 24 ZnS scintillation detectors was assembled and used to make measurements on five cores along a north-south transect of the Aegean sea (Nos T4, T21, T10, T18, AEG-3), and one core from the Ionian sea (PK-11) offshore the island of Corfu (Figure 1).

Sediment core sampling was carried out by the R/V Aegaio using a 3 m BENTHOS INSTR. (USA) gravity corer. Position fixing was obtained by an integrated positioning system (GPS) the TRIMBLE-4000 surveyor with an accuracy of ±50 m. AMS radiocarbon dating analysis on selected samples was performed in the BETA ANALYTIC (USA) laboratories.

RESULTS-DISCUSSION

The following describe the radioactivity measurements from each core and associated information regarding sedimentation rates, geotechnical characteristics, comparison between themselves, and spectrum analysis in two cores.

Core T18 (Length: 210 cm, Water Depth: 1050 m)

This core was recovered from a relatively deep marginal sub-basin in the eastern Aegean sea, but nevertheless it is directly affected by shallow water processes due to the land proximity (LYKOUSIS *et al.*, 1995).

Figure 2 shows the alpha and beta particles counting against depth, spanning two meters and covering the last 11–14 kyr. Two AMS C-14 calibrated (by us) dates, made on sapropelic clay, provide two tied points at 118 cm (8.1 ± 0.06 kyr B.P.) and at 145 cm (9.8 ± 0.06 kyr B.P.). The segments 0–118 cm and 0–145 cm give provisional sedimentation rates of 14.5 cm/ka and 15.3 cm/ka respectively, or an average value of 15 ± 0.5 cm/ka. Assuming this rate, the last ~60 cm accumulated during an ~4 kyr interval. Thus, the 212 cm core length corresponds to at most 14 kyr. Geotechnical properties (grain size, CaCO$_3$%, density, water content, shear strength) reinforce the above estimations.

At about 55 cm or about 3.6 kyr, a drastic change (drop) occurs in alpha counting, evident, but to a lesser degree in beta counting. This variation corresponds to the sub-boreal (see below). Alpha counting drops from 180 cm to 145 cm (end of slope). A more pronounced slope occurs in core T21 (the dating being deduced from tight dated points, see below), because of the slower sedimentation rate there; almost half of the T18 sedimentation values. This gives support of the rationale that high rates reflect reduced slope in counting.

Fourier spectrum analysis and significance tests (Kolmogorov-Smirnov, Randomness test) produced significand perriodicities of around 0.40, 0.60, 0.90, 1.70, 3.0 and 5.1 kyrs. The approximated time-scale (~±5%) together with the accuracy of the fourier method of spectrum analysis, are both reflected as a 10% error for the obtained periodicities.

Four features are identified with Greek letters (α, β, γ, δ) and attributed to certain climatic episodes.

Features α/β

The 55–60 cm (~3–3.2 kyr) variation rather corresponds to sub-Atlantic/sub-Boreal period. Perhaps, the minimum at (β) defines (milder) glacier readvancing, since the c.1 kyr B.C. marks end of post-glacial warmest time (*e.g.* glaciers in Alps, wetness setting in abrubtly in northwest Europe registered in the Irish, Welsh and North European peat bogs, the North and South America cold oscillation in the Rockies (LAMB, 1972). At circa 1 kyr B.C. there appears, also, the drastic

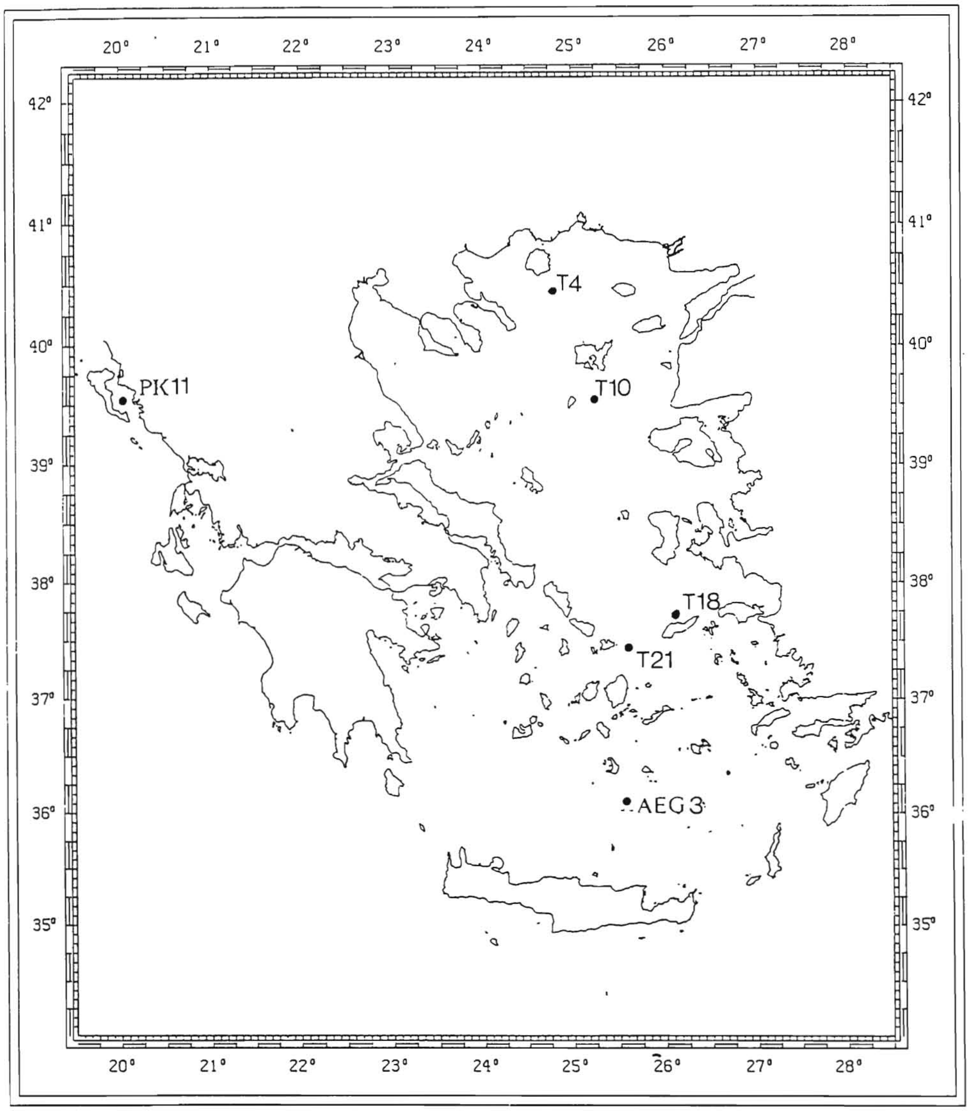

Figure 1. Map of the Aegean and Ionian seas and location of cores.

onset of sea-level rise (JELGERSMA and TOOLEY, 1995). Moreover, during 50–60 cm depth (~1–1.2 kyr B.C.) frequent climatic changes occurred in the sub-boreal stage, evidenced from archaeological and Holocene soils analysis in Greek mainland, especially regarding dramatic sedimentation changes (PAEPE *et al.,* 1995).

As a result of this climatic episode, the drastic change between α/β may be due to sudden intrusion of sediments from higher sedimentation rates for a short period of time, *e.g.* within 500 years, reinforced, also by somehow greater silt content (coarser *i.e.* high shear strength in kPa, that is more terrigenous input).

Features γ and δ are identified with the boreal and Preboreal/Young dryas (cool/wet to cold/drier). It is worth noting possible sub-phases within the Pre-boreal phase.

Core 21 (Length: 240 cm, Water Depth: 410 m)

This core was obtained from a relatively shallow small scale marginal basin in the eastern part of central Aegean sea. Sedimentologically, it represents conditions and processes over the greater shallow Cyclades plateau, *i.e.* the major morphological feature in the central Aegean sea.

Figure 3 shows the alpha and beta-counting versus depth. Generally it consists of muddy sediments, although deeper than 85–100 cm turns to sandy muds. The discernible change in natural agents is due to sea level drop. Such an effect indicates the pre-Holocene period for depths higher than about one meter.

The depth of 67 cm corresponds to a C-14 date of 9500 ± 80 years B.P. Compared with PK11, the depth of ~55 cm corresponds to ~7.5 Kyrs.

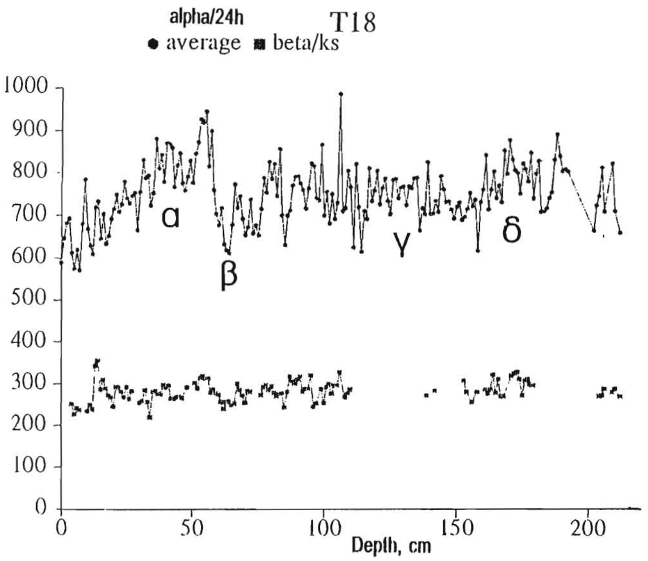

Figure 2. Core T18. Variation of a) alpha-counting and b) beta-counting, against depth. The greek letters refer to paleooclimatic phases.

Drastic changes are seen for the depths at 50 cm, 130–150 cm and 170–190 cm. The depth 75–80 cm corresponds to the begining of the Holocene, while the peak at 140 cm is at 18 kyrs, since the depositional period 80–140 cm, where a drop in radioactivity occurs, corresponds to the fast drop in eustatic sea level (Holocene transgression).

The ~140 cm peak resembles the γ-peak at the depth of 380 cm -ice advance- in the alpha-counting diagram for lake Bouchet (XANTHAKIS *et al.,* 1992; LIRITZIS and GALLOWAY, 1995; GALLOWAY, 1995a), and the cold maximum in core B-3 (LIRITZIS and GALLOWAY, 1995).

The depth at 160 cm, in which a little but drastic uprise in alpha counting (not in beta counting) occurs, refers to ash layer, possibly Y-5, dated to 22 ± 2 Kyrs (LYKOUSIS *et al.,* 1995). From ~180 cm the sedimentation rate increases (Pleni-Wurm period). Thus, the high peak at ~185–195 cm is related to the two large short-term successive glacial peaks inferred from the oxygen isotope ratio from Greenland ice dated to 27–29 Kyrs B.P. (DANSGAARD *et al.,* 1971).

Based upon the above and from comparisons made with other radioactivity and temperature reconstruction diagrams ($\delta^{18}O$), the following sedimentation rates are cestimated: 0–9 kyrs (~67 cm), 7.5 cm/kyr; 0–18 kyr (140 cm), 7.7 cm/kyr; 0–20 kyrs (~150 cm), 8 cm/kyr, with an average of 7.7 ± 0.3 cm/kyr. This dates the 237 cm core to 31 ± 1 kyrs.

There is a similarity in the variation curves of alpha and beta counting, in particular it is striking the trends and some peaks at depths of about 20, 25, 40–80, 130–150, 180–200, and 210–230 cm.

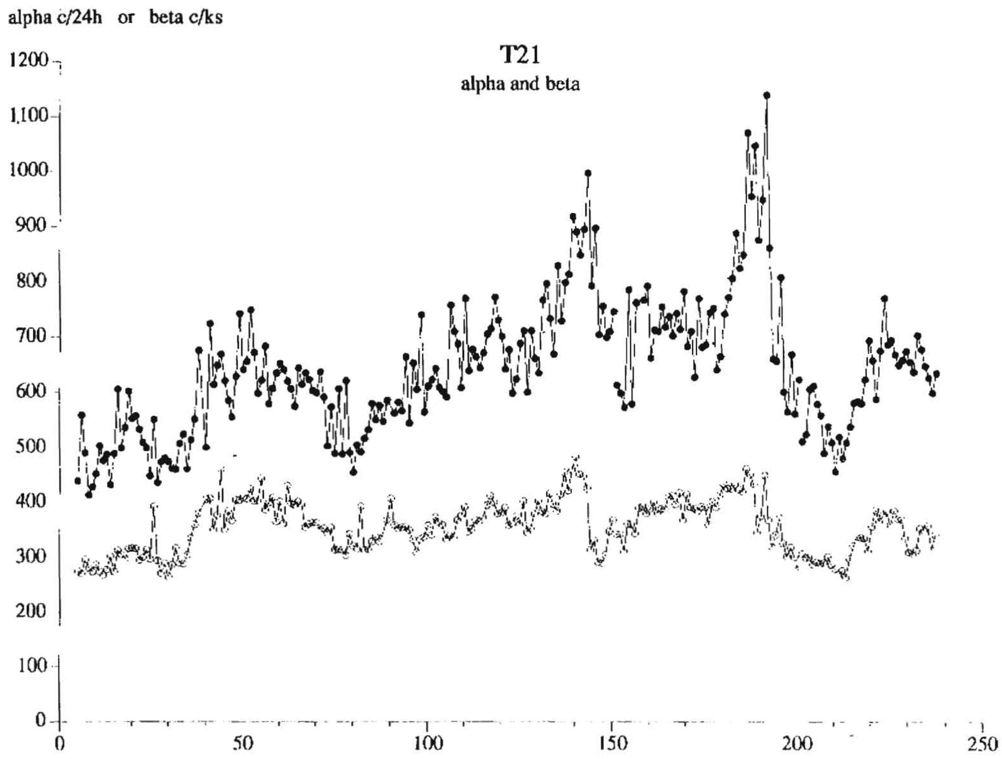

Figure 3. Core T21. Alpha-counting and beta-counting against depth.

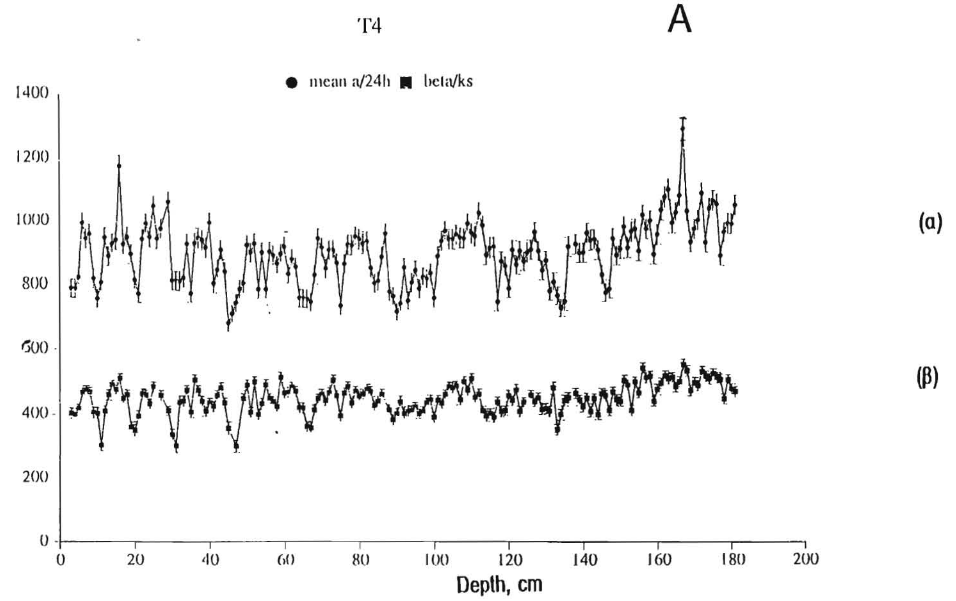

Figure 4. Core T4. Variation of a) alpha-counting, and b) beta-counting, against depth.

Core T4 (Length: 190 cm, Water Depth: 665 m)

This core was recovered from the northern Aegean marginal sub-basin and consists mainly from fine silt. Preliminary comparison with core T10 attribute to this core length the age span of ~12 kyrs: the ~60 cm corresponds to 4 kyrs (subboreal) and the 150 cm to ~10 kyrs.

Commensurable variations are noted between the alpha and beta counting diagrams at 20–30 cm, ~50 cm, 75 cm, 110 cm, 130–145 cm, as well as the general trend and increase beyond 150 cm (Figure 4).

In our effort to investigate possible periodic nature of these variations the alpha radioactivity was smoothed with a 3-term moving average and a 2nd order polynomial was subtracted. Figure 5a shows this statistical smoothing. The randomness test on the residuals verified the non-random nature of the variation. Subsequently, Fourier (FFT) spectrum analysis of the alpha counting series revealed high variance to the periodic terms of ~3, ~2 and 0.6–0.75 kyrs (Figure 5b). Similar analysis for the beta counting revealed same significant periodicities at ~0.32, ~0.6–0.7 and ~1.5–2.0 kyrs.

Core T10 (Length: 160 cm, Water Depth: 550 m)

This silty-clay core was obtained from the extensive relatively shallow Lemnos-Lesbos plateau.

Figure 6 show the alpha and beta counting. Characteristic changes are observed for the a-counting curve, four of which are identified with Greek letters α, β, γ, δ, likewise those in core T18, for which two layers are dated by radiocarbon. This core rather resembles core T18. Thus, the depths of 212 cm and 55–60 cm for T18 correspond to depths of 120 cm and 30 cm respectively, in T10. That is, the 120 cm of T10 cover the period 11–14 kyrs. These age/depth correlations were based upon the presence of sapropels (7–9 Kyrs BP) and the presence of older stiffer horizons due to the late glacial-holocene transgression.

Figure 6 exhibit a long term trend superimposed by short-term variation. The estimated rate of sedimentation is about 12.5 cm/1kyr.

The two smoothing ways of alpha-counting time-series with the subtraction of a 2nd order polynomial, as well as a 3-terms moving average followed by subtraction of a 2nd order polynomial, followed by Fourier spectrum analysis, revealed the periodic terms, 2.4, 1.2, 0.76, 0.57, 0.35–0.42, and 0.17 kyrs of high significance. The trend appears to inhere a quasi-periodicity of ~12 kyrs, perhaps identified with the known period of the precession of equinoxes (~24 kyrs).

Core AEG-3 (Length: 190 cm, Water Depth: 1600 m)

This relatively deep core from the south Aegean (Cretan sea) consists of uniform mud and displays low sedimentation rates.

Figure 7 shows the alpha counting variation versus depth, where drastic fluctuation of cycles per 25 cm sediment thickness are observed.

Core PK11 (Length: 190 cm, Water Depth: 35 m)

This is the shallowest core from a uniform depositional environment.

Figure 8 shows the alpha radioactivity versus depth. It rather derives from the Holocene and consists from homogeneous silty-clay (distal pro-delta sedimentation). It has an increased rate of sedimentation, 30–40 cm/kyr, perhaps due to the increase rate during recent times. Preliminary estimations of depth-to-time scale in three depths are as follows; ~3 kyrs for the 60 cm (sub-Atlantic, high radioactivity); 4–5 kyrs for 100–120 cm (sub-boreal, low radioactivity), and 5–7

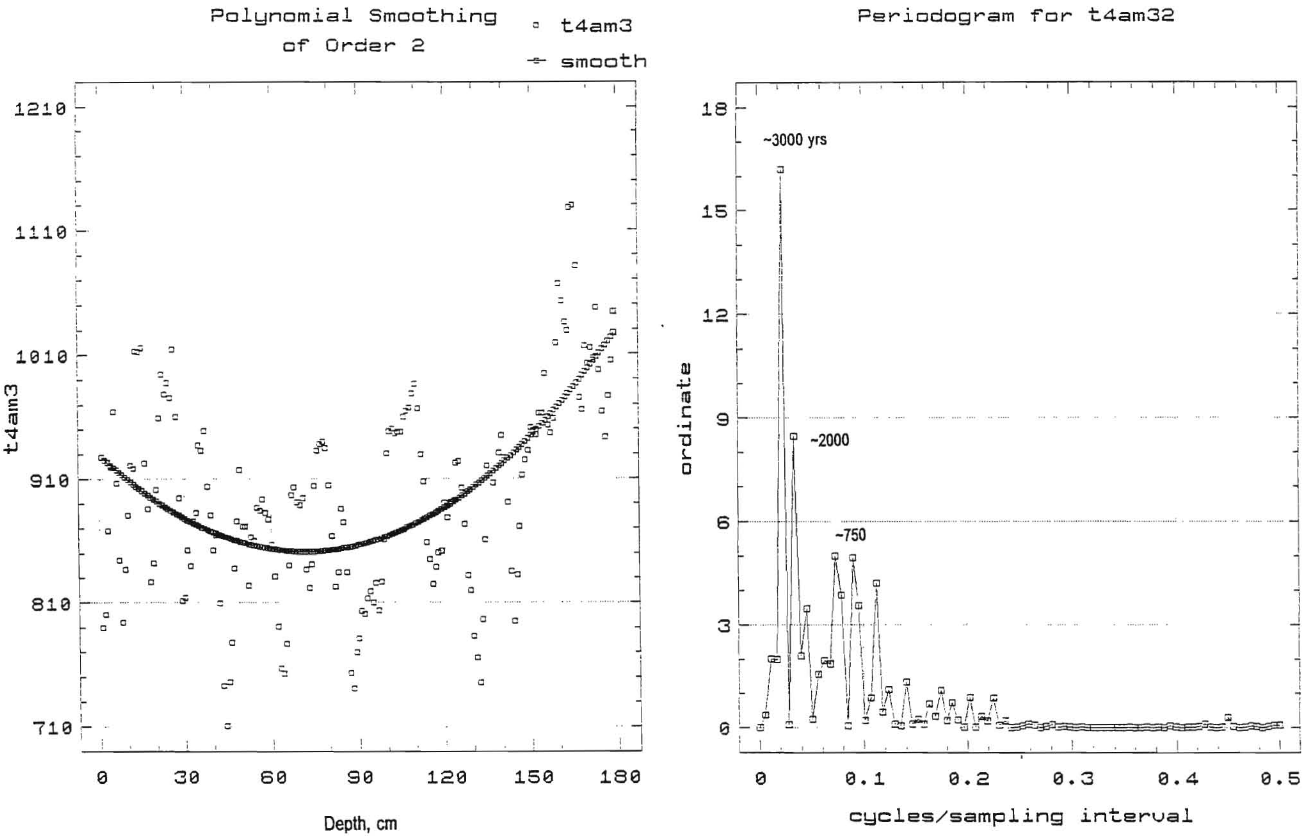

Figure 5. Smoothing of alpha-counting of T4 with 3-terms moving average (squares) and a polynomial of 2nd order (continuous line), against depth, b) Fourier spectrum analysis.

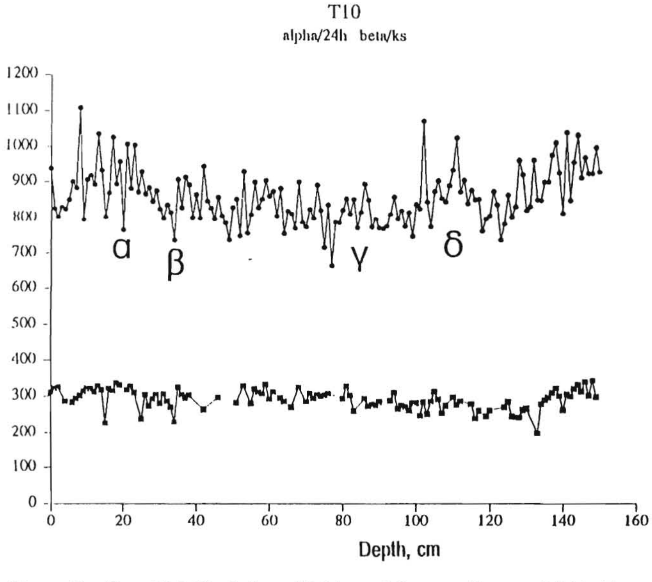

Figure 6. Core T10. Variation of (a) top, alpha-counting, and (b) bottom, beta-counting, against depth.

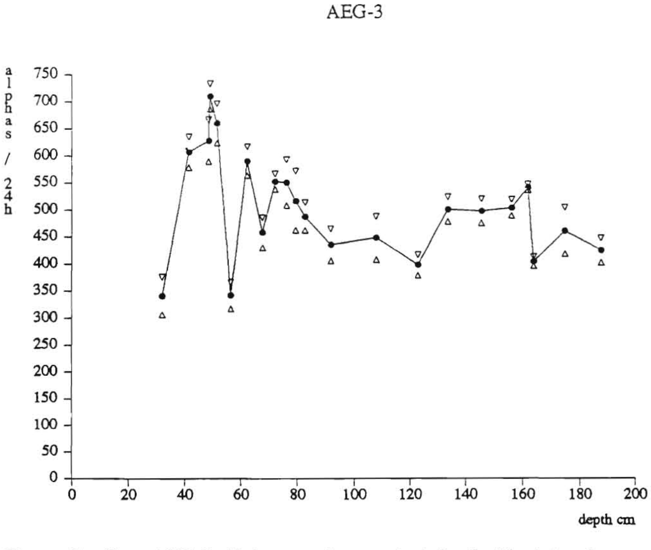

Figure 7. Core AEG-3. Alpha-counting against depth. The triangles are error bars.

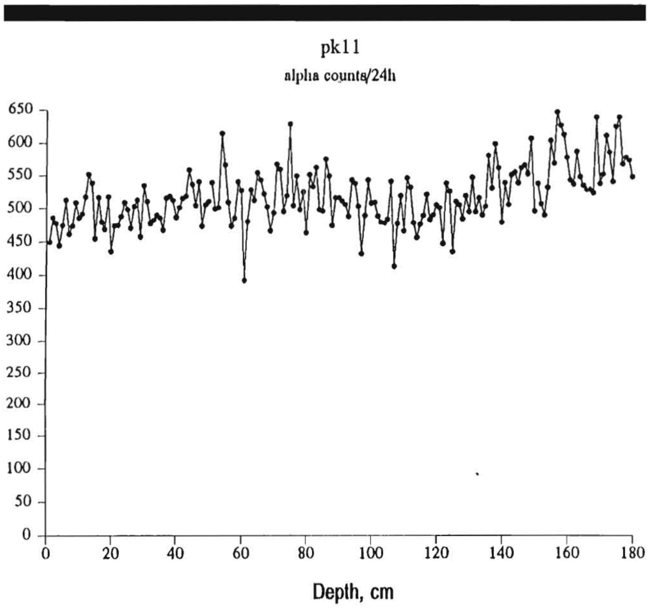

Figure 8. Core PK-11. Alpha-counting against depth.

kyrs for 160–180 cm (Atlantic, high radioactivity). Similar correlations between alpha radioactivity and palaeoclimatic events have been reported for the German lake Meerfelder Maar (Galloway, 1995 a,b).

The preliminary periodic analysis for T4 and T10 cores has shown similar periods of 0.6–0.7, 1–2, 3–3.5 kyrs. Similar cyclic pulses of drought and humidity during the holocene have been found in land records from geoarchaeological sites in Attica (Greece) of 0.22, 0.30, ~0.50, ~1.00 and 2.50 kyrs. (Paepe *et al.,* 1995). The ~2.5 kyrs has been reported, also, as a climatic cycle from O-18 analysis of foraminiferas from deep-sea cores in the Indian Ocean (Pestiaux *et al.,* 1987), and in landforms as high geomorphic activity in the European mountains (Starkel, 1995), together with short violent interuptions of 0.1–0.3 kyrs duration. The periodic and quasi-periodic variation of climatic changes, are usually embedded within a chaotic behavior of the system: atmosphere, climate, solar activity, planetary impulses (alignements, conjuctions). Such a climatic behavior is recorded in proxy climatic recorders such as the sediment radioactivity (Schove, 1987; Xanthakis *et al.,* 1992; Fairbridge and Sanders, 1987; Fairbridge, *pers. comm* to IL).

CONCLUSION

The alpha and beta radioactivity measurements of sediments from six cores from the Aegean and Ionian sea, Greece, exhibit changes which broadly reflect palaeoclimatic phases within the Holocene and during the last glaciation.

The comparison of the depth dependence of radioactivity measured by alpha particle counting and by beta particle counting shows the latter to provide a more smooth variation with depth, possibly because the short range of alpha particles in the sample means a smaller volume of the sample, by about two to three orders of magnitude, contributes to the counting and so the result may be more subject to variation due to sample inhomogeniety than in the beta particle case. The interpretation of the beta particle counting rate data in relation to climate is, however, in some cases (T18, T10) less clear either because the mineral content of the sediment is different or because the low potassium content as the major contributor to beta counting is less sensitive to striking climatic change. It may be that the variation in beta particle counting rate is at least in part due to variation in mineral composition, but that variation may in turn be due to climatic variation when discussing sediments of basin and sub-basin marginal depositional environment.

The drastic radioactivity changes can be used as chronological markers, useful for the establishment of chronostratigraphic scale. In contrast to other dating tools, such as ^{14}C or $\delta^{18}O$, this new dating approach has the following advantages: i) almost inexpensive, ii) very fast; counting time per sample of 1 hour for beta particles and 24 hours for alpha particles, thus with the automatic multisample detectors 24 samples can be measured in one day, iii) much less tedious, iv) requires a few grams of mass, and the satisfactory accuracy.

The present results, thus, provide the basis for the establishment of type curves for the broad Aegean geographical region; thus, introducing a new method for dating, and identification of palaeoclimatic features in the southern Mediterranean region.

Preliminary spectrum analysis of sediment radioactivity, from two Aegean cores, present a periodic nature, which reflects climatic cycles.

Therefore, such measurements offer additional means investigating climatic cycles from proxy climatic data.

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