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SEDIMENTOLOGICAL AND GEOCHEMICAL CHARACTERIZATION OF HOLOCENE SEDIMENTS, FROM ALIKES LAGOON, ZAKYNTHOS ISLAND, WESTERN GREECE

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Abstract

The study area is the Alikes wetland (an old salt pan) which is located in the northern part of Zakynthos island. In the Alikes lagoon an exploratory borehole (GA-1) were drilled in a depth up to 21.20m. A total number of forty six (46) samples were analysed for their particle size, calcium carbonate and total organic carbon content. Moreover, bulk sample chemical analyses for major and trace elements were carried out on 12 samples, as well as mineralogical analysis for bulk, oriented and the clay fraction (<2 μm) were determined by powder X-Ray diffraction. The grain size characteristics, statistical parameters and TOC, for the Holocene analysed samples, suggest a coastal environment (restricted-shallow) with reduced salinity such as a lagoon margin and in a tidal flat and/or marsh particularly. Depositional environments and the source material affecting the geochemical signature indicating that that Al, Mg, K are mainly associated with the aluminosilicate fraction of the sediments analyzed, Fe and Ti consistent with detrital phases, while Ca, Si, P, and Mn showing different geochemical patterns.

***Key words**: sedimentology, geochemistry, Holocene sediments, Alikes, Zakynthos.*

1. Introduction

There are different approaches in reconstruction the Holocene coastal – lagoon environmental changes, which are based on geological, geochronological, sedimentological and geochemical analyses methods. Studies in western Greece, investigating and reviewing the Holocene environmental changes in coastal lagoonal areas, have been carried out by Kontopoulos and Avramidis (2003), Kraft et al. (2005), Vött (2007), Avramidis et al. (2008), Kontopoulos and Koutsios (2009), Avramidis & Kontopoulos (2009) and Engel et al. (2009).

Geological processes that affect the chemical composition of sediments include hydraulic sorting, weathering and diagenesis (Fralick and Kronberg, 1997). Significant improvement has been made in geochemical approaches to monitoring sediment provenance, especially through the introduction of discrimination diagrams based on the relationship of major and trace elements (Condie, 1993; Cullers, 1994; Bauluz et al., 2000). Whilst trace element geochemical studies have tended to focus on aspects of sediment provenance, their application to paleoclimate reconstructions has been rela-

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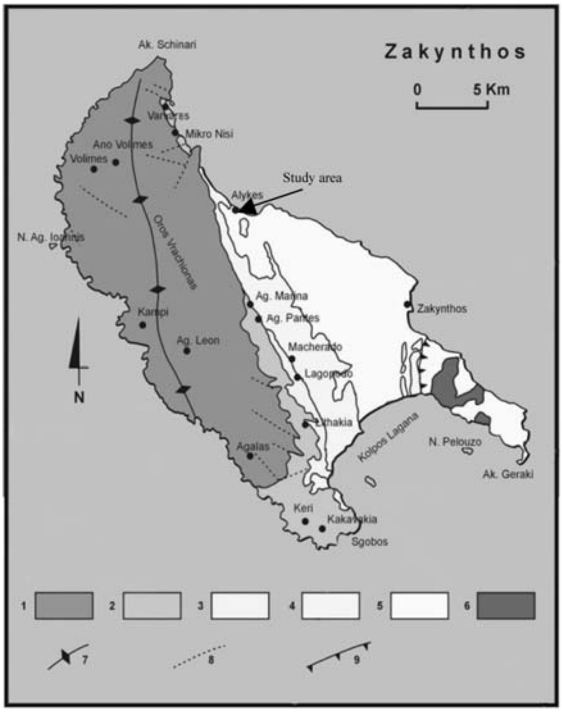

Fig. 1: Simplified geological map of Zakynthos Island. Legend: 1. Cretaceous carbonates, 2. Eocene carbonates, 3. Oligocene marly carbonates, 4. Miocene clastics and carbonates, 5. Pliocene-Quaternary alluvial deposits, 6. Ionian evaporites and breccias, 7. Anticline, 8. Fault, 9. Main thrust.

tively neglected, with most paleoclimate studies focusing on biogenic components, such as carbonate, opal, organic carbon and authigenic elements (Elderfield, 1990). On the other hand, the geochemical behaviour of trace elements in natural systems may often reflect changes in the chemical, biological, and physical conditions of the environment. For example, Mn and Fe change their chemical forms and concentrations according to the redox condition (Stumm and Morgan 1996), while distribution profiles of some bio-elements such as Ca, P and Si, are sensitively affected by biological activity (Riley and Chester 1983).

The purpose of the present paper is to study the Holocene sedimentological environments of the Alikes lagoon, Zakynthos island (Fig. 1) as well as the geochemical and mineralogical changes, based on a 21.2m sediment core. An effort is made to record the Holocene palaeonvironmental evolution – changes of the area and to correlate them with the sediment geochemical and mineralogical distribution.

2. Geological setting

The study area is the Alikes wetland (an old salt pan) which is located on the northern part of Zakynthos island (Fig. 1), and expand in an area of 400.000 m^2. The Zakynthos island is characterised by two geotectonic zones the Pre-Apulian and the Ionian zone. It is located near the north-western terminus of the Hellenic Trench, very close to the convergent boundary between the African and European plates and the diapirism area of the evaporates, that belong to the Alpine basement. The sedimentological evolution of the island was influenced by both compresional and extensional tectonics (Zelilidis et al,. 1998) while sedimentation can be distinguished in carbonates Cretaceous to Miocene and clastic Plio – Quaternary (Fig. 1).

Fig. 2: The narrow inlet in the northern part of the lagoon.

Fig. 3: General view of the lagoon with maximum depth up to 45cm.

In the study area, Alikes lagoon, during the last decades human activities such as agriculture, irrigation works and building development influenced, the water surface of the lagoon. On the northwest side, the Alikes lagoon is separated from the Ionian Sea by a low relief sand barier and has limited communication with the open sea with a sort and narrow inlet (Fig. 2). The maximum water depth of the lagoon is up to 45cm and an average depth only up to 15 cm (Fig. 3). Holocene sedimentation rates for Alikes lagoon area have been estimated by Avramidis & Kontopoulos (2009) and reveal two different stages of sedimentation rate: the first one between 8280 BP and 5590 BP (5.3 mm/yr) and the second one between 5590 BP and modern times (1.03 mm/yr).

3. Methods

The exploratory core GA-1 (Fig. 4) was bored in February 2008, at the geographical position N $37^0 50' 32''$, E $20^0 45' 51''$. The drilling equipment was a rotation Longyear 38, with single tube core barrel with tungsten carbide bit and 101mm diameter. All the segments of the core were sealed with cling film. Sediment types, structure, colour, as well as contact depths and bed characteristics, were recorded. Colour were identified using a Minolta CM-2002 hand held spectrophotometer. Standarised sedimentological analysis were carried out, on 46 samples including particle size analysis, calcium carbonate and organic carbon content. Particle size distribution was made using a Malvern Mastersizer 2000, while moment measures were calculated using GRADISTAT V.4 and based on Folk (1974) nomenclature. The organic carbon content was estimated by the titration method according to Gaudette et al. (1974).

Bulk sample chemical analyses for major and trace elements were carried out on 12 samples at the Activation Laboratories and were performed using a Thermo Jarrell-Ash ENVIRO II ICP for INAA and a Perkin Elmer Optima 3000 ICP. Silica, SiO_2 was determined using the X-ray Fluorescence method after $LiBO_2$ fusion and Zr was measured by the xylenol orange colorimetric method (Roser et al., 2000).

The mineralogical composition of bulk samples and of the clay fraction (<2 μm) separated by sedimentation was made on 12 samples and were determined by powder X-Ray diffraction (XRD), using a Bruker D8 advance diffractometer, with Ni-filtered CuKα radiation. Powders from oriented samples were prepared by the dropper method and were scanned at $1^o 2\theta$/min from 3 to $70^o 2\theta$. Random powder mounts were prepared by gently pressing the powder into the cavity holder. For each < 2 μm specimen, the clay minerals were identified from three XRD patterns (after air-drying at 25^o C).

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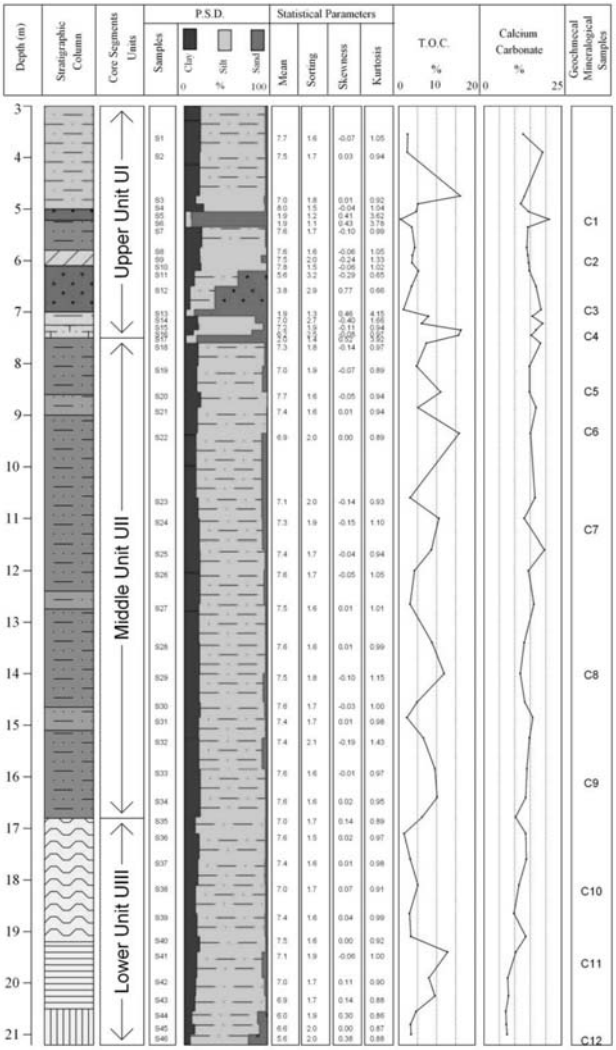

Fig. 4: Borehole profile GA-1 modified after Avramidis & Kontopoulos (2009), showing the sedimentary units, the P.S.D. - statistical parameters, TOC, carbonate content and geochemical – mineralogical analyzed samples.

4. Results

4.1 Sedimentology

The sedimentological core description is based on Avramidis & Kontopoulos (2009) discrimination of three major sedimentary units. The upper unit UI (3.00–7.50m), middle UII (7.50–16.80 m) and lower UIII (16.80–21.20 m) (Fig. 4).

Upper Unit (3.00-7.50 m): refer to the upper part of the core segment between depth 3.00m to 7.50m (Fig. 6), we didn't take into account material between depths 0.00–3.00m as it is classified as filling material (pave) for road construction. The unit consist of poorly to very poorly sorted, grayish olive, laminated fine to medium silt, rich in organic matter with bioturbation structures and shell fragments. Silt

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is intercalated by three main horizons of poorly sorted, olive brown to dark olive, fine to medium sand (5.00–5.22 m, 6.00–7.00m and 7.25–7.50 m). Contacts between silt and sand are characterised as slightly erosional. The sand fraction consists of foraminifera, ostracodes and molluscs 15 in proportion less than 2% to the clastic grains. Also, there are carbonate – cementing aggregates in abundance in the sand fraction. The calcium carbonate content in the upper unit ranges from 11.90 to 21.00% with an average value 13,07%, while TOC between 0.54 to 16.30% with an average value 5.36% (Fig. 4).

Middle Unit (7.50-16.80 m): consist of poorly sorted, very dark greenish gray fine silt, with shell fragments, thin layers rich in organic matter with plant remains and abundant shell fragments and 20 *Cardium* shells. The sand fraction contains mostly micro- and macrofossils and abundance plant remains. Caliche-like evaporate, largely of a calcium carbonate type, is found cementing some of the grain together into aggregates. Internally to this unit we distinguish three layers (8.60–9.00 m, 12.40–12.60m and 14.65–15.10 m) consisting of poorly sorted grayish olive fine silt and is characterised by ripple lamination. The calcium carbonate content in the middle unit ranges from 11.71 to 19.48% with an average value 14,97%, while TOC between 2.16 to 15.79% with an average value 7.90% (Fig. 4).

Lower Unit (16.80–21.20 m): consist of poorly sorted, greenish gray fine to coarse silt. This unit is characterized by the presence of *Cardium, Cerithium* and *Hydrobia*, *Ammonia* shells and abundant organic matter. Also, there are carbonate-cementing aggregates in abundance in thesand fraction. In the lower part of the unit we observe an increase in sand participation and we distinguish a layer of poorly sorted, light olive gray to grayish olive, coarse silt. The calcium carbonate content in the lower unit ranges from 7.09 to 13.65% with an average value 9.94%, while TOC between 1.4 to 12.81% with an average value 5.23% (Fig. 4).

4.2 Geochemical analysis

Results of chemical analysis for the sediments collected from the Alikes Lagoon, are summarized in Table 1. The SiO_2 contents of the samples range from 37.92 to 64.02 wt.% and are enriched relative to Clay Shales (CSH). The Al_2O_3 contents of the samples are higher than of CSH (8 wt.%), except of the samples with codes C2 and C5. By contrast, the CaO contents are remarkably higher than that of CSH (2,20 wt.%). Most samples show that the Na_2O contents are enriched relative to CSH, while K_2O contents shown slightly lower values relative to CSH, with their average values being of 1.70 wt.% and 2.13 wt.% respectively. MgO contents of the samples range from 0,92 to 4,49 wt.%, with their average value being higher (3,42 wt.%) than the CSH (1,50 wt.%). The Fe_2O_3 concentrations are slightly higher than the CSH values, with the samples C2, C4, C5 and C12 showing lower concentrations than CSH. The MnO content of the samples ranges from 0.043 to 0.112 wt.%, while TiO_2 concentrations range from 0.08 to 0.686 wt.% being in the same magnitude with the CSH values. The P_2O_5 contents of the samples are enriched compare to the CSH concentration, with their average value being 0,089 wt.%. The LOI (Lost Of Ignition) values ranges from 9,89 to 21,21 wt.% with an average value of 17,98 wt.%.

4.3 Mineralogical analysis

The mineralogical composition of samples C1-C5 is characterized by the dominant presence of quartz, calcite, albite, chlorite and illite, while lepidocrocite presence was detected in samples C4, C5 (Fig. 5). The sample C6 shows a different mineralogical composition and halite and gypsum coexist with calcite, albite, chlorite, quartz and illite (Fig. 5). Samples C7-C12 are characterized by the dominance of illite and the presence of quartz, albite, chlorite and calcite and traces of lepidocrocite.

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Table 1. Results of chemical analysis for the sediments collected from the Alikes Lagoon borehole GA-1.

Upper Unit: core segment between depths 3.00m to 7.50m															
Sample Code	*Core Depth*	*SiO_2*	*Al_2O_3*	*Fe_2O_3 (T)**	*MnO*	*MgO*	*CaO*	*Na_2O*	*K_2O*	*TiO_2*	*P_2O_5*	*LOI***	*Total*	*Rb*	*Cs*
	(m)	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	ppm	ppm
C1	5,12	39,13	10,94	5,89	0,103	4,49	13,41	1,82	2,21	0,574	0,13	20,63	99,32	93,0	4,5
C2	5,9	61,74	2,78	1,16	0,043	0,92	15,74	1,14	1,09	0,088	0,04	15,49	100,2	36,0	1,0
C3	6,85	39,23	11,55	5,81	0,097	3,95	13,5	1,63	2,49	0,577	0,11	20,5	99,44	111,0	6,2
C4	7,35	44,72	9,19	4,5	0,091	3,31	14,02	1,71	2	0,452	0,1	19,42	99,5	81,0	4,0
Middle Unit : core segment between depth 7.50–16.80m															
Sample Code	*Core Depth*	*SiO_2*	*Al_2O_3*	*Fe_2O_3 (T)**	*MnO*	*MgO*	*CaO*	*Na_2O*	*K_2O*	*TiO_2*	*P_2O_5*	*LOI***	*Total*	*Rb*	*Cs*
	(m)	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	ppm	ppm
C5	8,42	55,94	5,57	2,78	0,067	1,86	14,97	1,17	1,41	0,26	0,06	16,45	100,5	53,0	2,3
C6	9,2	37,92	11,24	6,11	0,112	4,3	13,47	1,96	2,37	0,563	0,09	21,21	99,36	111,0	6,6
C7	11,1	40,11	11,52	6,38	0,087	4,02	12,81	1,78	2,36	0,59	0,09	19,59	99,33	108,0	5,9
C8	13,9	40,59	11,47	5,58	0,091	3,97	13,15	1,86	2,34	0,581	0,09	19,92	99,64	101,0	5,2
C9	16	40,77	12,31	6,45	0,094	4,28	12,42	1,56	2,51	0,629	0,1	19,02	100,1	106,0	6,0
Lower Unit : core segment between depth 16.80–21.20m															
Sample Code	*Core Depth*	*SiO_2*	*Al_2O_3*	*Fe_2O_3 (T)**	*MnO*	*MgO*	*CaO*	*Na_2O*	*K_2O*	*TiO_2*	*P_2O_5*	*LOI***	*Total*	*Rb*	*Cs*
	(m)	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	wt.%	ppm	ppm
C10	18,1	48,62	11,8	5,97	0,084	3,63	10,13	1,82	2,33	0,658	0,1	15,62	100,8	110,0	5,6
C11	19,5	42,97	13,49	6,71	0,058	3,82	8,84	1,95	2,61	0,686	0,09	18,01	99,23	138,0	7,9
C12	21,1	64,02	10,07	4,26	0,067	2,48	5,51	1,97	1,8	0,645	0,07	9,89	100,8	79,0	3,5

* (T): Total, **LOI: Lost of Ignition

Increasing depth the amount of chlorite as well as calcite is decreasing while illite is increasing. Vermiculite is present in samples C9-C12 (Fig. 5).

5. Discussion

The particle size distribution throughout the core indicate a relative uniform distribution of grain size. Sediment types are fine to coarse silt with three intercalations of medium sand in the upper unit. The presence of sand is low, ranging between 0 to 5%, clay percent ranges between 2 to 20% and silt between 69 to 85% (Fig. 4). Almost all the analysed sediments are characterised as poor to very rich in organic matter (TOC>0.5), while carbonate content participate in a portion >8%.

Based on statistical parameters, in the upper core segment, the silt material is characterised as poorly to very poorly sorted, while skewness indicate basically a symmetrical distribution and values of kurtosis propose a mesokurtic distribution. The sand layers of the upper unit appear to be poorly to very poorly sorted while skewness in all sand samples indicate very fine skewed distribution while kurtosis propose mainly an extremely leptokurtic distribution. The very fine skewed value is caused by the presence of a fine tail in the sand-dominate size frequency curve. This fine tail in combination with the poorly to very poorly sorting and extremely leptokurtic kurtosis may be suggesting an event analogous with a fluvial flood. This event may occur in a coastal environment such as a sandy tidal flood. If this happens then the silt beds of this unit which have a very low sand content, poorly to very poorly sorting and normal distribution, have deposited in a sheltered muddy tidal flat or marshy area. The middle unit indicate a uniform distribution of poorly sorted fine silt while skewness and kurtosis in-

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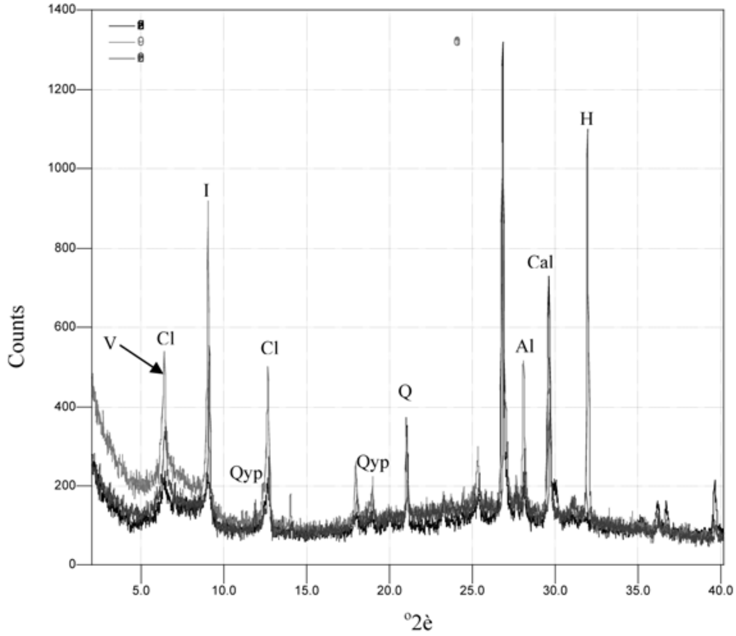

Fig. 5: X-ray diffraction patterns of representative oriented samples C3, C6, C10. Cl:Chlorite, I:illite, Qyp: Gypsum, Q: quartz, Al:Albite, H: Halite.

dicate mainly a symmetrical and mesokurtic distribution. The participation of sand is very low <5% (Fig. 4). These grain size statistical parameters are the same as those of the silt beds of the upper unit. This event and the very low sand content suggest also a sheltered muddy tidal flat or marshy area. The lower unit of the core is characterized by poorly sorted fine to medium silt and the increase of the sand participation in the lower part, depth 20.55– 21.20 m, where we have the presence of coarse silt. Participation of sand is low <7% with exception the lower part of the unit where increases up to 23%. Skewness present a symmetrical to fine skewed distribution, while kurtosis a mesokurtic one. According to the grain size statistical parameters the mainly part of the unit III has deposited also in a sheltered muddy tidal flat or marshy area. The lower part of the unit III where the sand content increases, the skewness is near symmetrical to fine skewed and the kurtosis is slight platykurtic, may have deposited in a mixed flat environment.

However geochemical analyses may contain additional new paleoclimate information that provides important new constrain on sediment depositional environment and climate. In this case, we examine the geochemical characteristics of the sediments from Alikes Lagoon, Zakynthos island, Western Greece, by calculating enrichment factors for the elements relative to CHS (Clay Shale; Salomons & Forstner, 1984). The enrichment factor (EF) for any element is given by the following equation:

$$EF= (C_{sample} / C_{Al\ sample}) / (C_{standard} / C_{Al\ standard}),$$

where C_{sample} is the concentration of the element and $C_{Al\ sample}$ is the concentration of Al in the sample (Yan et al., 2007). The Al normalization is utilized because concentrations of most elements show correlation with Al_2O_3. CSH is used as the standard. Enrichment factor diagrams are shown in Figure 6a-d.

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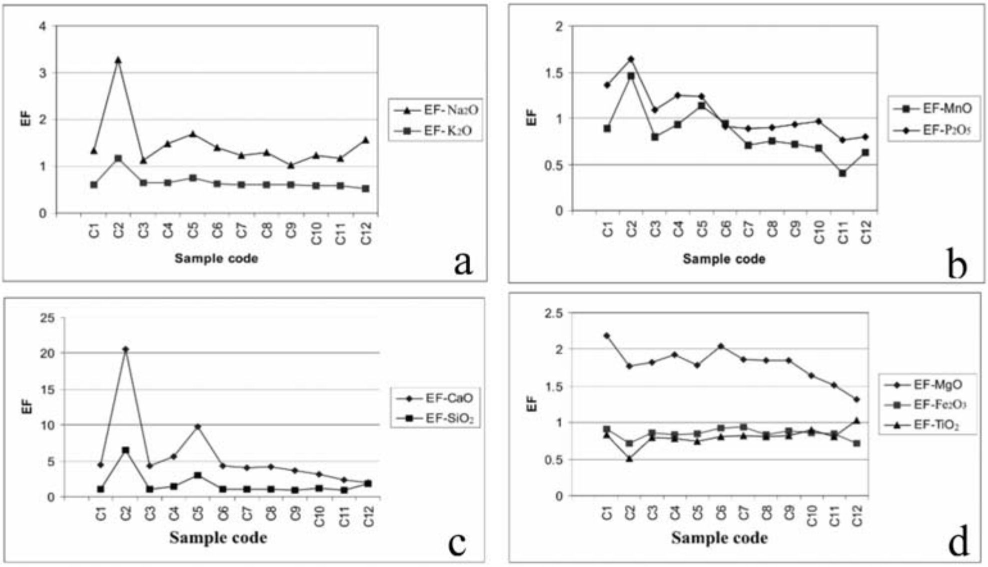
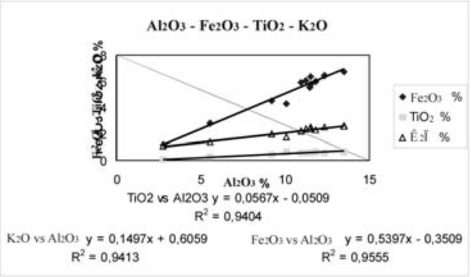


Fig. 6: Element enrichment factor (EF) plot (relative to CSH) for the samples from the Alikes Lagoon, Zakynthos island, Western Greece: a) EF for K_2O, Na_2O, b) EF for MnO, P_2O_5, c) EF for CaO. SiO_2 d) EF for MgO, Fe_2O_3, TiO_2. (CSH composition from Salomons & Forstner, 1984).



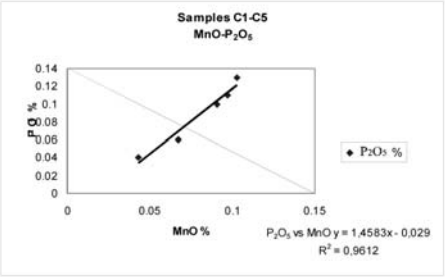


Fig. 7: The *x–y* plots among Fe_2O_3, TiO_2, K_2O and Al_2O_3 data from the Alikes Lagoon, Zakynthos island, Western Greece sediments.

Fig. 8: The *x–y* plot among MnO and P_2O_5 data from the Alikes Lagoon, Zakynthos island, Western Greece, sediment samples C1-C5.

The samples show that Na, Ca and Mg content are enriched relative to CSH (Fig. 6a, c and d), while Mn and P enrichment factors show variations relative to CSH (Fig. 6b). K, Fe and Ti enrichment factors show depletion relative to CSH (Fig. 6a and d). Samples C2 and C5 display an abnormally high enrichment factor for Ca, and Si thus it is possible these data related to the carbonate cementing aggregates, the shell fragments and the presence of quartz and calcite as it is shown from the XRD analysis. By contrast, Mg enrichment factor for C2 and C5 samples having a tendency to degrease due to the lower amounts of chlorite. The same pattern is obvious for Fe and Ti, although their enrichment factors are lower than CSH. It is known that the chemical composition can be influenced by weathering, dissolution and adsorption-desorption processes and these may produce irregular chemical variations in some major and trace elements (Mass and Mc-

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Culloch, 1991). This is consistent with the significant positive linear correlation between Al, Fe and Ti classified as a detrital element group, (Al-Fe: R^2=0.9555, Al-Ti: R^2=0.9404, Fig. 7), while strong positive linear correlation between Al and K (R^2=0.9413), indicating their association with the phyllosilicate fractions such as illite and chlorite (Bauluz et al., 2000), which are present in our samples (Fig. 5).

In the case of Mg, its significant negative linear correlation with Ca in the horizon extended between 5m and 8m of the core (R^2=0.988) implies that this element is consistent with the aluminosilicate fraction.

In addition Mn is a redox-sensitive element that easily reduced or oxidized by changes of the biogeochemical conditions of the environment (Stumm and Morgan 1996). Mn geochemical behaviour shows differences between 5m to 8m, and 9m to 21m downcore. In the upper part of the sediment column there is a variation in the Mn enrichment factor relative to CSH, while its significant negative correlation with Ca (R^2=0.9855) and the very strong positive correlation with Al (R^2=0.9678), indicating Mn association with the clay fraction and/or oxide-hydroxide phases. In the down part of the sediment core, its depletion compare to CSH (Fig.6b), is compatible with the dissolution of Mn-solid phases due to the presence of abundant organic matter.

The Si enrichment factors (Fig. 6c), together with its significant positive linear correlation with Ca in the upper part of the core (R^2 = 0.9937) suggesting the biogenic origin of the element. From 9m to 21m sediment depth, Si is mainly associated with the fraction of resistance aluminosilicate minerals, by having a strong negative linear correlation with Ca and TOC (Ca-Si: R^2 = 0.8324, TOC-Si: R^2 = 0,7362). Additionally in the core-top samples, P is mainly related with manganese oxides/hydroxides phases due to their strong positive linear correlation (R^2 = 0.9612, Fig. 8, Yuan-Hui Li et al., 2007).

However, Ti usually behaves as conservative element in a weathering profile, and thus Ti normalized ratios help to track the behaviour of elements during chemical weathering. Additionally chemical weathering affected from environmental changes such as climate and redox conditions (Yan, et. al 2007). Furthermore, the Ti content show no covariance with SiO_2 content (R^2 = 0,2828) and significant inter-sample variations, hence Rb and Cs are normalized to Ti. The Rb and Cs are very sensitive to climatic influences and may become leached during chemical weathering. As chemical weathering is largely controlled by moisture and temperature, a wet and warm climate may enhance the chemical weathering (Nesbitt et al., 1980). Variation of Rb/Ti, Cs/Ti ratios, $CaCO_3$ contents and TOC values imply a warm and wet climate which result in greater vegetation cover (Yan et al., 2007).

6. Conclusions

Although the sediment chemical properties variation, can be related to different sediment sources, the rich Ca and Si content of the samples indicate high primary production rates during deposition in the past. The higher rate of sedimentation in the Alikes lagoon until mid-Holocene from 5.3 mm/yr to 1.03 mm/yr in modern times, the grain size statistical parameters, the silt beds and the low sand content of the sediment samples analyzed, suggesting a warm, sheltered muddy tidal flat and/or marshy coastal deposition environments. The grain size statistical parameters, suggests a coastal environment (restricted-shallow) with reduced salinity such as a lagoon margin and in a tidal flat and/or marsh particularly. Depositional environment and the source material affecting the geochemical signature indicating that Al, Mg, and K are mainly associated with the aluminosilicate fraction of the sediments analyzed, while Ca, Si mainly related with the biogenic fraction of the sediments in the core top samples. Iron and Ti classified as a detrital element group, while the P and Mn correlation in the top core samples showing its relation with manganese oxides/hydroxides phases.

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