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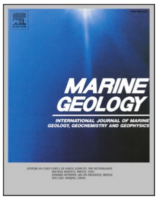

Research Article

Refining Late-Holocene environmental changes of the Akko coastal plain and its impacts on the settlement and anchorage patterns of Tel Akko (Israel)

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ABSTRACT

Akko/Acre, a UNESCO World Heritage site since 2001, is one of the oldest continuously inhabited sites in the eastern Mediterranean. Tel Akko was a major maritime centre of the southern Levant from the Middle Bronze to the Late Persian period. The city was then moved 1500 m to the west on the Akko promontory where the 'Old City' of Saint-Jean d'Acre is located. The natural and anthropogenic evolution of Tel Akko area is reflected by persistent geographical and habitation pattern changes. We combine sedimentological and faunal analyses of radiocarbon dated cores as well as identification of ceramic sherds found in the cores with ground penetrating radar investigations to propose an up-to-date palaeogeographical reconstruction of landscape/environmental changes of the Akko coastal plain in order to understand the extent to which environmental pressures have played a role on the position of anchorage and habitation patterns. We highlight how the local population make use of the natural advantages of the area and adapted to environmental pressures. Following a constant sedimentary input and simultaneous coastal progradation of the Akko coastal plain the main anchorage areas where forced to move. While the 2nd Millennium BCE anchorage was on the southern area of the tell, the late-1st Millennium BCE (Phoenician-Persian) anchorage was relocated on the western area. Vicissitudes in settlement pattern noted in archaeological excavations and surveys on Tel Akko have, most likely, been the consequence of the changes in the position of the coastline.

1. Introduction

Over the past decades, the question of human reaction, adaptation, and resilience to environmental changes has gained increased interest, particularly for those related to coastal societies. These locations are specifically considered as vulnerable to environmental changes (Wong et al., 2014). During the Holocene, a transition from a nature-dominated to a human-dominated environment is noted (Messerli et al., 2000). In the Early-Holocene, coastal societies were strongly affected by the rise in sea level that led to the submersion of prehistoric coastlines (Benjamin et al., 2011, 2017; Galili and Rosen, 2011; Galili et al., 2017). Humans living along the coasts tended to adapt to coastal changes by the relocation of their communities further inland. In the eastern Mediterranean, the oldest coastal defence against sea level rise was found in the form of a seawall dated to ca. 7500–7000 BP discovered at the submerged Neolithic settlement of Tel Hreiz along the Carmel coast of Israel (Galili et al., 2019). However, the seawall was only a temporary solution and finally the village was abandoned and submerged. After the diminished rate of sea level rise at ca. 7000/6000 years BP and its stabilisation since ca. 4000 years, coastal communities became increasingly

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sedentary with the enhancement of maritime trade and the development of anchorages and harbours (Artzy, 1997, 2006; Faust and Ashkenazy, 2009; Sherratt, 2016). In the Bronze Age, populations favoured natural features as anchorage areas whereas most artificial harbour basins and related port facilities started to be built mainly during the Iron Age (Blue, 1995; Marriner et al., 2014, 2017). Along the Levantine coast, the first artificial harbours are dated to the Iron Age, the "Phoenician" period – the first part of the 1st Millennium BC – at Tabbat el-Hammam (Braidwood, 1940; Frost, 1972) and Atlit (Haggi and Artzy, 2007; Haggi, 2010).

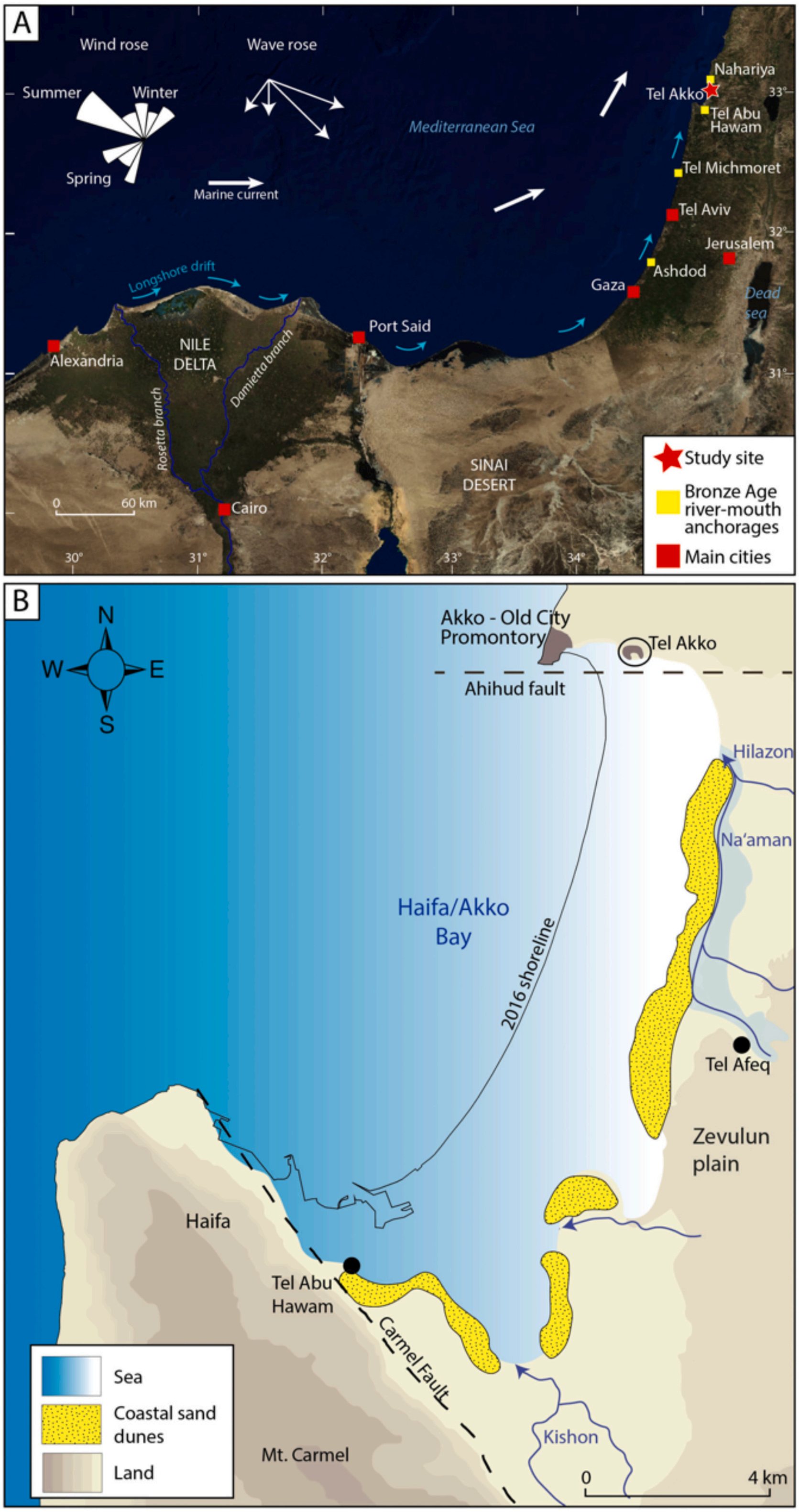

Fig. 1. A. Regional location of the Haifa/Akko bay in the Eastern Mediterranean (after Inman, 2003; image CNES). Wind and wave patterns at Alexandria are from Stanley and Bernhardt, 2010). B. Geomorphological reconstruction of Haifa/Akko bay 4000 years BP (after Elyashiv et al., 2016).

It is widely recognized that during the Late-Holocene, Mediterranean coastal/deltaic plains have been impacted by human related environmental impacts – mainly linked to watershed deforestation that led to soil erosion and an increase in fluvial sediment supply – that accentuated the expansion of coastal plains (Stanley and Warne, 1997; Bellotti et al., 2004; Simeoni and Corbau, 2009; Maselli and Trincardi, 2013; Anthony et al., 2014). This phenomenon was favoured in the context of the glacio-eustatic stabilisation of sea level since ca. 7000/6000 years. The development of the coastal plains happens particularly early in the eastern Mediterranean. In ria type deltas, the limited accommodation space for the deposition of fluvial sediment led to particularly important progradation of the coast (Anthony et al., 2014). In Cyprus, the infilling of the Gialias ria has been elucidated with the discovery of a succession of coastal palaeoenvironments (alluvial plains, lagoons, coastal spits) consistent with the eastward migration of the coastline during the past 6000 years. This progradation of the waterfront forced the local inhabitant to keep pace with the shifting coastline by relocating their anchorages since the Bronze Age (Devillers, 2008). The progradation of coastal plains and the infilling of lagoons had important negative impacts on the navigability of coastal water bodies. In Cyprus, infilling of the Larnaca Salt Lake area – leading to the gradual isolation of the Salt Lake's lagoon from the surrounding marine environment through sedimentation – eventually lead to the abandonment of the settlement at Dromolaxia-Vyzakia (Hala Sultan Tekke) in the early 12th century BCE (Devillers et al., 2015). Along the coast of northern Levant, the increasing rate of coastline progradation is illustrated by the formation of the tombolos of Tyre and Ras Ibn Hani. After 3000 years BCE, a particularly rapid phase of sand bank accretion is observed and characterised by a large increase in sedimentation rates (Marriner et al., 2007, 2012). Along the southern Levant, increasing rate in coastal plain progradation appears to be consistent with a pulse in sediment supply driven by Bronze and Iron Age soil erosion in local catchments indicating more extensive farming to sustain a growing population (Palmisano et al., 2019). These data suggest that at a regional scale, early human impacts were significant in modulating geomorphological change in downdrift coastal areas of the Levant.

Using a multidisciplinary approach, the present study aims to reconstruct geomorphological evolution of the Akko coastal plain. We aims to understand: What was the natural environment at the foundation of Tel Akko? How did the coastal landscape evolved and how fast? How did local inhabitants deal with coastal changes? Answering these question will allow us to precisely reconstruct the impact of coastal changes on the evolution of the habitation and anchorage patterns from the Bronze Age onward.

2. Regional setting

Tel Akko is located on the northern edge of the Haifa/Akko Bay, the only bay along the coast of the southern Levant (Fig. 1). Zviely et al. (2006) and Elyashiv et al. (2016) have provided details of the Holocene geomorphological evolution of the bay in relation to sea level changes and sedimentary inputs. During the Early-Holocene rates of sea level rise were primarily linked to modifications in ice volumes and the Earth's response to the changing ice-water load. This sea level rise was the main driver of coastal changes in this tectonically stable region (Sivan et al., 2001; Avnaim-Katav et al., 2012). During the last 6000 years, the ice-equivalent melt-water input is considered minimal leading to an important decrease in the rate of sea level rise (Sivan et al., 2001; Lambeck et al., 2004, 2014; Vacchi et al., 2016, 2018). The associated large sedimentary inputs lead to the westward progradation of the coastline. The main source of sediments leading to the infilling of the bay was Nile-derived sand quartz. Zviely et al. (2007) have estimated the quantity of Nile-derived quartz sands trapped in the bay over the last 8000 years to be 700 million m^3. Marine sediment infill is supplemented by fluvial sediment inputs from the Qishon and the Na'aman Rivers at the southern and northern extremities of the bay (Vachtman et al., 2013; Zviely et al., 2006; Fig. 1.B).

Locally, the Na'aman River's geomorphological evolution played an important role in the environmental changes at Tel Akko. The river drains the western Lower Galilee Mountains and its watershed is formed by two subsidiary river-basins: (i) the Hilazon River basin drains 240 km^2, and (ii) the Na'aman River basin (drainage area of ca. 70 km^2), and is mainly fed from the Apheq spring (Lichter et al., 2009). Due to anthropogenic use of the water from the Apheq spring, the river discharge has undergone significant decrease (Lichter et al., 2009). The Na'aman is presently artificially channelized 800 m to the south of the tell (Figs. 2 and 3), but in the Late-Holocene it flowed closer to the tell (Morhange et al., 2016; Giaime et al., 2018). In a map dating to the beginning of the 20th century, a relic arm/meander of the Na'aman flowed closer to Tel Akko can be observed (Fig. 2). Lichter et al. (2009), who studied historical maps and aerial photographs, have demonstrated that the Na'aman mouth migrated by 1.5 km (both north and south) along the coast during the last 200 years.

3. In search of Tel Akko's anchorage(s): (geo)archaeological context

The site of Tel Akko has a strategic geographical location, near the coast and in close proximity to the Na'aman river mouth on the northern edge of the Haifa/Akko Bay (Fig. 1). Besides its favourable position near the coast, the panoramic view from Tel Akko includes the entire bay, the north-south route, as well as routes leading from the coast to Syria via the fertile Jezreel valley and across to Transjordan (Artzy, 2018). The first archaeological remains at Tel Akko are dated to the beginning of the Early Bronze Age, ca. 5000 years ago (Table 1). Monumental structures – a rampart and a gate – dating to the Middle Bronze *Age* IIa (MBA; 2000–1750 BCE) were found on the northern, high section of the tell (Dothan, 1976; Raban, 1983, 1991; Be'eri, 2008; Fig. 3; Area A and AB). Tel Akko acted as a major anchorage site from at least the MBA to the Late Persian period. Written sources from 2nd Millennium BCE, found in the el-Amarna Letters dated to the 14th century BCE indicate that it was an important harbour utilized by the Egyptians (Artzy, 2018). In the Persian Period, it was a centre for the Persian army and its mercenaries in the quest to conquer and control Egypt (Abu Hamid and Artzy, 2021; Abu Hamid, 2021).

From a palaeo-environmental perspective, pioneer research has suggested that fluvio-coastal dynamics linked to sea level changes and high sedimentation rate played a major role in the evolution of the Akko coastal plain (Inbar and Sivan, 1983). At the scale of the tell, past geo-archaeological studies have shed light on the evolution of the ancient coastal interface (Morhange et al., 2016; Giaime et al., 2018). These results pointed toward an important coastal progradation on the southern area of the tell during the last two millennia BCE and induced a shift in the location of the ancient coast through time. Recent work confirmed the presence of the coastline, and reinforced the evidence of a possible anchorage or even a proto-harbour, in the southwestern area of Tel Akko during the first part of the 1st Millennium BCE (Giaime et al., 2021).

Considering the dynamic coastal interface of the city (Morhange et al., 2016; Giaime et al., 2018), the different anchorages were not in a fixed location over the centuries. The question of the Bronze Age anchorage was first raised by Raban (1985, 1991). He proposed that like many other Bronze Age Levantine coastal cities, Tel Akko's inhabitants took advantage of the Na'aman River mouth to install their anchorage in a naturally protected environment (Raban, 1985). When viewed from above, the present shape of Tel Akko is reminiscent of a "moon crescent" (Artzy and Quartermaine, 2014; Artzy et al., 2021). Part of the indentation in the crescent was assumed by Raban to have been an inner harbour. The assumption of a Bronze Age anchorage in the vicinity, if not in the indentation, of Tel Akko was reinforced by the discovery of numerous imported ceramics dated from the 2nd Millennium BCE in areas P and PH (Fig. 3; Raban, 1991).

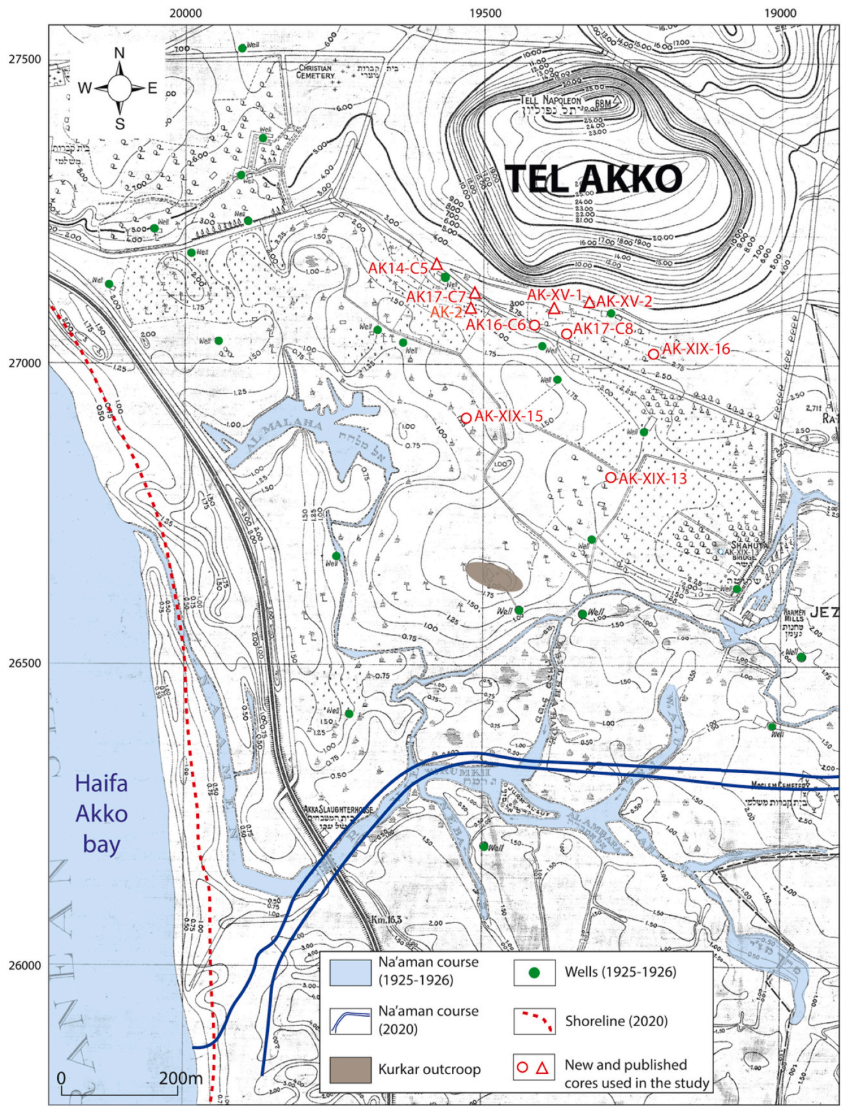

Fig. 2. Map of the Akko coastal plain in 1925–1926 (Unpublished map of Joseph Treidel). The map shows that the Na'aman River was flowing closer to Tel Akko in the beginning of the 20th century.

4. Material and methods

To reconstruct Late-Holocene geomorphological changes in the area, we combined a sedimentological and faunal study of radiocarbon dated sedimentary cores with geophysical investigations of the sub-surface sediments.

4.1. Collection and bio-sedimentological study of the cores

Five cores were collected in plastic liners from a Geoprobe DT22 percussion corer. The core locations (Easting and Northing) were obtained using a GPS-RTK system providing a spatial accuracy of 0.02 m (Fig. 3). The elevation of each core was measured with a total-station using a National Datum point MAPI 3835 as reference benchmark. This datum point has a mean sea level elevation of 33.40 m. This elevation was established by the Survey of Israel Center in 1999, based on 18.6 years of continuous orthometric measurements of sea level of the Mediterranean Sea.

The chronology of the cores is based on nine Accelerator Mass Spectrometry (AMS) radiocarbon dates. The preparation of the samples, which includes chemical pre-treatment, fractionation of their carbon components followed by oxidation and reduction to graphite was completed at the Radiochronology Laboratory of the Centre d'Études Nordiques (University of Laval, Canada). AMS measurements were performed at the Keck Carbon Cycle AMS Facility (University of California-Irvine, USA; Table 2). Samples submitted were grape seeds, pieces of charcoal, and marine shells. Dates were calibrated using Calib 8.2 (Stuiver and Reimer, 1993; Stuiver et al., 2020) with IntCal20 (Reimer et al., 2020) and Marine20 (on shells) curves (Heaton et al., 2020). In addition, ceramic fragments collected during coring give a high-precision relative chronology for the stratigraphic units.

The core liners were opened and stratigraphically described. Sampling was based on the stratigraphic breaks in the cores. The general sediment texture, including gravel (> 2 mm), sand (63 μm-2 mm) and silty-clay fractions (<63 μm), was determined by the wet sieving of 50 g of dry sediments. Ostracods were picked from the sand fraction >125 μm and identified using atlases and specific literature (e.g. Athersuch et al., 1989; Lachenal, 1989; Frenzel and Boomer, 2005; Avnaim-Katav et al., 2016) while molluscs were identified in the >2 mm fraction (D'Angelo and Garguillo, 1978; Avnaim-Katav et al., 2016). Statistical analyses were performed using the paleontological statistical software PAST (Version 2.14, Hammer et al., 2001). The standardized dataset is based on the relative abundance of each ecological group (freshwater, lagoonal, marine lagoonal and coastal/marine). Principal Component

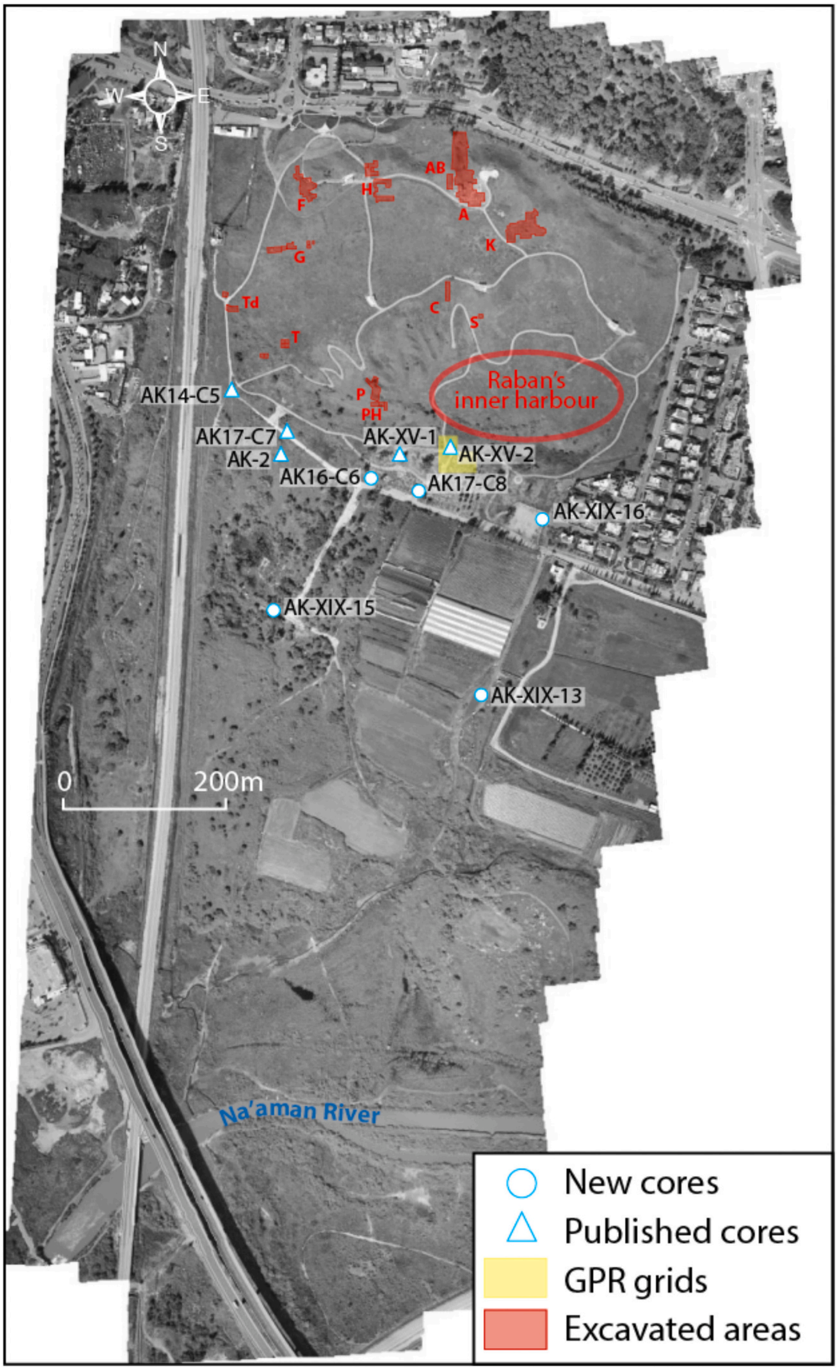

Fig. 3. Location map of the data used in this article collected in the Tel Akko coastal plain. Background image taken by drone (© P. Bauman, 2018).

Table 1
Main archaeological periods of Tel Akko.

Period	Years BCE
Early Bronze	3500–2200
Middle Bronze	2200–1550
Late Bronze	1550–1200
Iron Age I	1200–1000
Iron Age II (Phoenician)	1000–586
Iron Age III	586–539
Persian	539–332
Hellenistic	332–63

Analysis (PCA) was undertaken on the variance-covariance matrix to test the ordination of samples by assessing major changes in palaeoenvironmental proxies. We did not include macrofauna data in the PCA because they were too scarce and not found in all the cores.

4.2. *Ground penetrating radar (GPR)*

GPR has been widely used to investigate geoarchaeological as well as coastal geomorphic environments (Jol and Bristow, 2003; Conyers and Goodman, 1997; Buynevich et al., 2009; Goodman et al., 2009). This non-invasive method is a near-surface geophysical surveying technique which utilizes pulses of electromagnetic waves to image the subsurface stratigraphy and associated human-made disturbances. The resulting

Table 2
AMS-14C data expressed in calibrated years BP and BC at the 95% interval confidence level (2σ) using Calib 8.2 (Stuiver and Reimer, 1993; Stuiver et al., 2020) and the IntCal20 (Reimer et al., 2020) and Marine20 curves (Heaton et al., 2020).

Sample	Lab number	Material	Depth (cm below surface)	Depth (cm below mean sea level)	Age 14C	2 sigma cal BP min; max	2 sigma cal BC/AD min; max	Historical period
AK16C6 670–680	ULA-9186	Marine shell (*Cythara*)	675	356	4105 ± 15	3540; 4010	2060; 1600 BCE	Middle Bronze
AK17C8 306–310	ULA-9132	Grape seed	308	47	2425 ± 15	2350; 2670	720; 410 BCE	Late Iron Age - Persian
AK-XIX-13624–631	ULA-9185	Marine shell (*Conus*)	627	410	4535 ± 15	4100; 4580	2630; 2150 BCE	Early Bronze
AK-XIX-15300	ULA-9133	Charcoal	300	137	1955 ± 15	1830; 1930	20; 120 CE	Roman
AK-XIX-15586	ULA-9130	Grape seed	586	449	2975 ± 15	3070; 3210	1260; 1130 BCE	Late Bronze - Early Iron Age
AK-XIX-15638	ULA-8699	Charcoal	638	475	2990 ± 20	3070; 3320	1370; 1120 BCE	Late Bronze - Early Iron Age
AK-XIX-16388	ULA-8696	Charcoal	388	3	2220 ± 15	2150; 2320	370; 200 BCE	Hellenistic
AK-XIX-16550–560	ULA-9131	Olive pit	555	170	2250 ± 15	2160; 2340	390; 210 BCE	Hellenistic
AK-XIX-16740–750	ULA-9181	Marine shell (*Bittium*)	745	360	5730 ± 15	5580; 5970	4020; 3630 BCE	Chalcolithic

profiles provide reflections of subsurface horizons and are dependent on local sediment properties including dielectric permittivity, electrical conductivity, and attenuation (Jol and Bristow, 2003). The use of GPR in identifying and imaging archaeological sites is a growing field within geoarchaeology (Conyers, 2014).

On the southern area of Tel Akko, multiple transects and grid surveys were collected using a Sensors & Software pulseEKKO (PRO) GPR system with antennae frequencies ranging between 225 MHz and 500 MHz. The highlighted profile was collected with an antennae frequency of 225 MHz. The antennae were shielded and separated by 0.25 m. Traces were collected every 0.1 m along the transect and were vertically stacked 8 times with a sampling rate of 800 ps. The digital profiles were processed and plotted using pulseEKKO software. Basic processing included AGC, signal saturation correction, trace stacking (horizontal averaging) and point stacking (running average) as well as other routines when necessary. To measure depth, near surface velocity measurements were calculated based on hyperbolic measurements and resulted in a near surface velocity of 0.06 m/ns with depths corroborated by local boreholes. The application of radar stratigraphic analysis (distinct signature patterns) on the collected data provides the framework to investigate both lateral and vertical geometry and stratification of the interpreted coastal deposits.

5. Results and interpretations

5.1. *Chronology of the cores*

Nine radiocarbon-dated samples were taken from various depths, providing a chronological framework covering all periods of human occupation and anchorage development at Tel Akko. The most recent sample is dated from the Early Roman period (ULA-9133 in core AK-XIX-15) whereas the oldest sample is dated from the Chalcolithic period (ULA-9181 in core AK-XIX-16). In core AK-XIX-16, the dating of samples ULA-9131 (550–560 cm depth) and ULA-8696 (388 cm depth) to the Hellenistic period is problematic. If we consider both dates, the core might have recorded an important sedimentation rate that can explain the low density of ostracods in the samples. However, the uppermost sample ULA-8696 may also be problematic due to the possible old wood effect of the charcoal compared to the dating of the short lived olive pit.

Dating of marine shells reveal a discrepancy with the chronology obtained from the reading of the pottery reflecting a possible deviation regarding the age reservoir of the sea water. This is the case for the oldest samples in cores AK-XIX-13 and AK-XIX-16 (Table 2). Siani et al. (2000) have estimated the mean marine reservoir age to be 390 ± 80 years in the Mediterranean Sea, which hides important spatial disparities between the different sub-basins (western and eastern) of the Mediterranean. Nevertheless, temporal deviations in this reservoir age have also been demonstrated in more recent studies, in particular in lagoonal environments (Sabatier et al., 2010). Rather than using this mean value proposed in Siani et al. (2000), we used a local ΔR of 142 ± 66, based on the average of 8 published dates of marine shells from around the site (http://calib.org/marine/).

In additions, the presence of ceramic sherds in the coastal/marine units of the cores allow us to put in perspective the geomorphological evolution of the area with changes in the anchorage and settlement pattern. The oldest sherds, dated to the 2nd Millennium BCE (Middle and Late Bronze periods) were found on the south-eastern area of the tell. More recent ceramics, dated to the mid-1st Millennium BCE (Late Iron Age period), were identified in the cores collected on the south-western side of the tell .

5.2. *Numerical analyses on the ostracod dataset*

PCA ordination was chosen because it is a non-parametric statistical method that extracts the most important information from complex datasets. Previous analysis of an ostracod dataset has been shown to be useful in linking ecological changes in ostracod assemblages with environmental evolution (e.g. land locking of waterbodies) in coastal settings (e.g. Giaime et al., 2017). PCA undertaken on the standardized ostracod dataset shows that the main variance is loaded by the PCA-Axis 1 (94% of the variance). PCA-Axes 2 and 3 respectively account for 4.3% and 0.6% of the total inertia. The PCA-Axis 1 has been plotted on a linear depth-scale along each studied core (Figs. 4-8). PCA-Axis 1 has high positive scores for samples dominated by lagoonal fauna with the presence of subsidiary freshwater species and negative ones for those comprising coastal/marine and marine lagoonal species. This axis is interpreted as a gradient of waterbody confinement. Samples with a positive score are identified as a restricted (confined) lagoonal environment whereas samples with negative scores are typical of an open marine environment.

5.3. *Bio-stratigraphy of the cores*

- Core AK-XIX-16 is 900 cm long and was drilled close to the southern limit of Tel Akko (Figs. 2 and 3), 385 cm above msl (Fig. 4).

Unit A, below 815 cm depth (-430 cm msl) is composed of dense brown silty-clay containing no fauna.

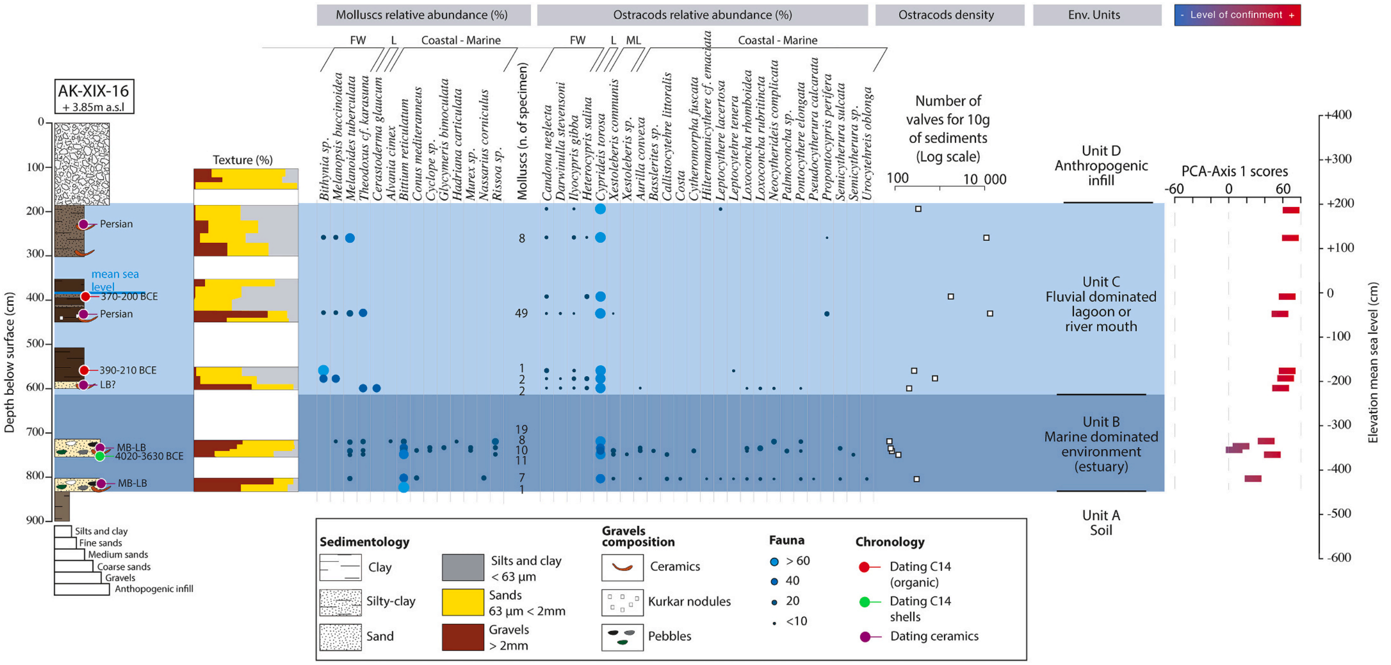

Fig. 4. Sediment texture and faunal analysis of core AK-XIX-16. Four different environmental units were identified. The succession of the units shows the progressive land locking of the area as demonstrated by the PCA scores. See Fig. 3 for location of the core.

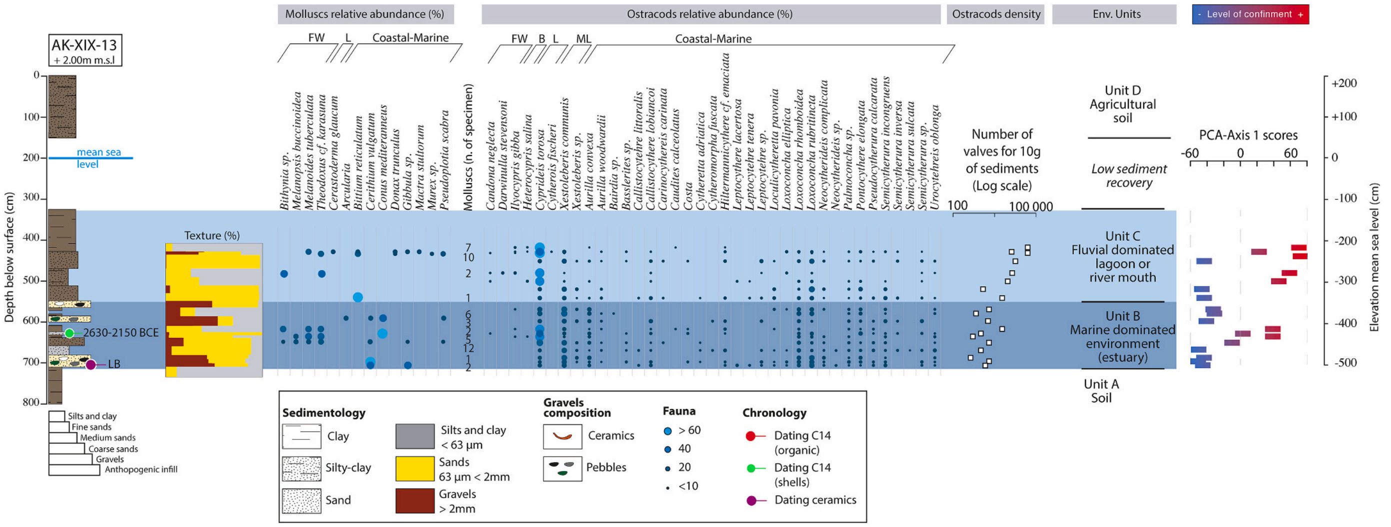

Fig. 5. Sediment texture and faunal analysis of core AK-XIX-13. Four different environmental units were identified. As with AK-XIX-16 (Fig. 4), the succession of the units shows the development of a lagoon – over a coastal environment – due to the progradation of the coast and the river-mouth. See Fig. 3 for location of the core.

Unit B is found between 815 and 590 cm depth (-430/-205 cm msl). This unit is mainly composed of coarse sands (48%) and gravels (41%). The gravel fraction is formed of small pebbles, shells, and fragments of *Cladocora caespitosa* (marine cushion coral). Marine shells dominate the assemblage (e.g. *Bittium reticulatum*, *Conus mediteranneus*, *Murex* sp.), and specimens of freshwater and brackish molluscs were observed in the upper sections (e.g. *Bithynia* sp., *Melanopsis buccinoidea*, *Theodoxus* cf. *karasuna*). The ostracod assemblage is dominated by *Cyprideis torosa* (lagoonal) which represents 62% of the total assemblage (Fig. 4). The recovered density is low with ca. 100 valves per 10 g of sediments. This low density may be explained by the impact of higher wave energy on the preservation of shells.

Unit C, between 590 and 200 cm depth (-205/+185 cm msl) has a silty-clay to silty sand texture. The gravel fraction is composed of shells, small pebbles, or ceramic fragments. The mollusc assemblage is dominated by freshwater and brackish species. The ostracod assemblage is dominated by lagoonal species (*C. torosa* represents 82% of the total) with subsidiary freshwater and coastal-marine species.

Unit D represents the upper unit of the core, and is formed by recent anthropogenic infill between 0 and 200 cm depth (+185/+385 cm msl). The unit is mainly composed of coarse sediment deposited on the south of Tel Akko for land reclamation.

- Core AK-XIX-13 (+200 cm msl) is 800 cm long and was drilled 200 m south of AK-XIX-16 (Figs. 2, 3 and 5).

Unit A is similar as AK-XIX-16's unit A, it is composed of dense brown silty-clay with no fauna below 710 cm depth (-510 cm msl).

Unit B, between 710 and 510 cm depth (-510/-310 cm depth), is composed of intercalated layers of yellowish and brownish sediments. In

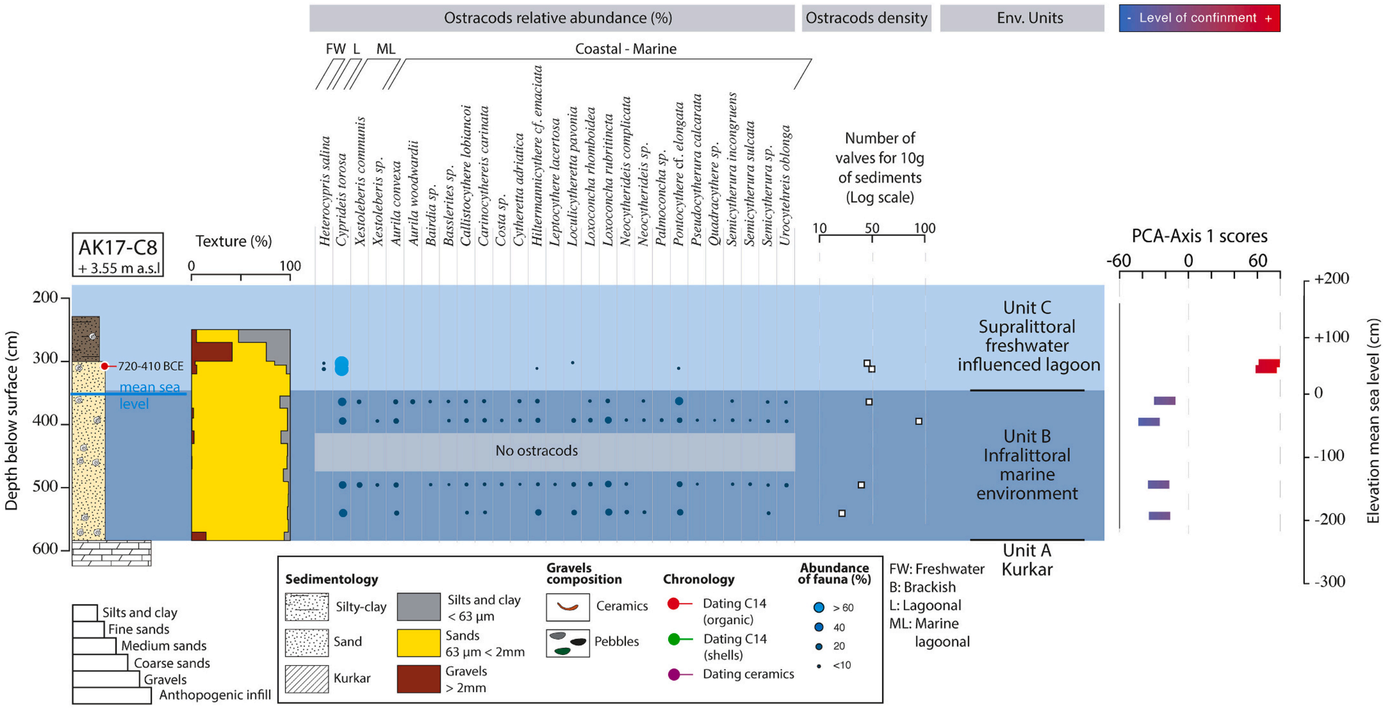

Fig. 6. Sediment texture and faunal analysis of core AK17-C8. The different environmental units are characteristic of the progradation of the coast. The development of a coastal lagoon is dated from 720 to 410 cal years BCE. See Fig. 3 for location of the core.

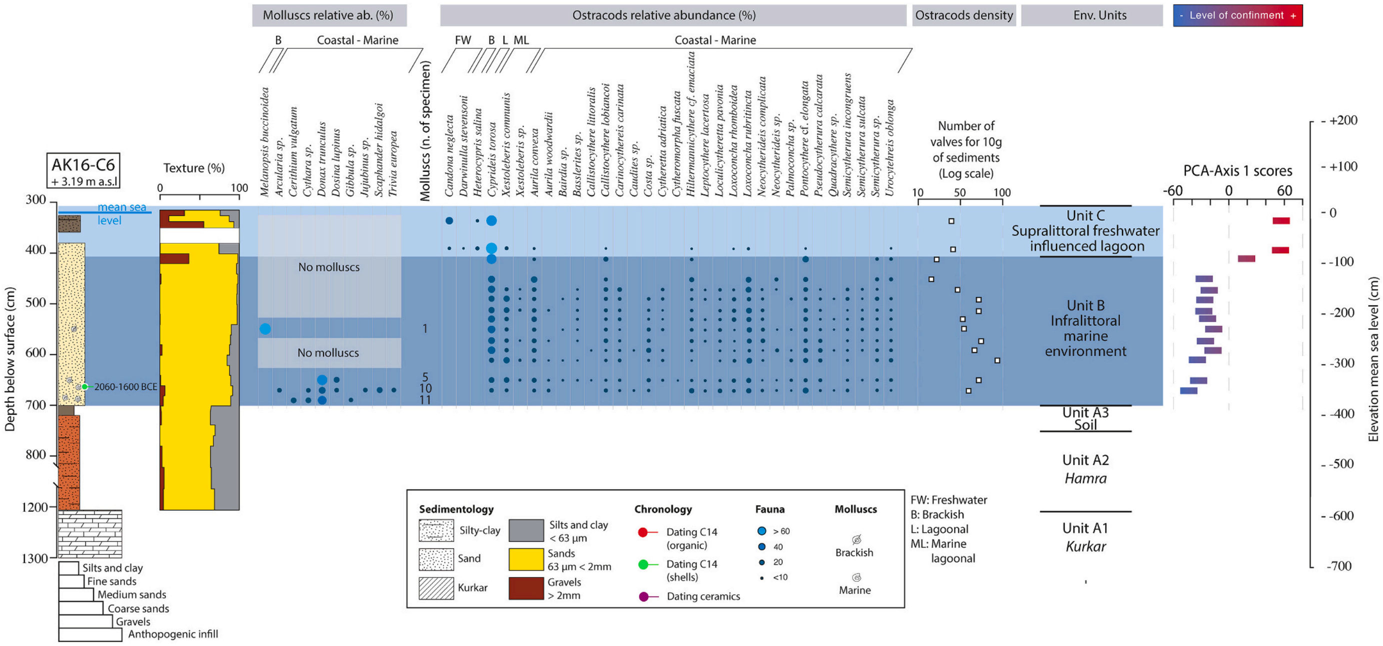

Fig. 7. Sediment texture and faunal analysis of core AK16-C6. The different environmental units are characteristic of the progradation of the coast. The marine transgression is dated before 2450–1980 cal year BCE at the base of unit B. See Fig. 3 for location of the core.

the yellowish layers, gravels are composed of small pebbles, shells and fragments of *C. caespitosa* (marine cushion coral) indicating a marine origin. The mollusc assemblage is composed of marine and freshwater species. Coastal/marine species dominates the ostracod assemblage (61%). Freshwater (*Ilyocypris gibba*, *Candona neglecta*), lagoonal (*C. torosa*) and marine lagoonal (e.g. *Xestoleberis*) species represent 1%, 32% and 6% respectively of the assemblage. The density varies between 200 and 1000 valves per 10 g of sediments.

Unit C, between 510 and 320 cm depth (-310/-120 cm msl), presents a silty-clay to silty-sand texture with some layers of pure sand. The ecology of molluscs and ostracods demonstrate a twofold influence from the sea and the river with the presence of freshwater, lagoonal, marine lagoonal and coastal-marine species. This is confirmed by the ostracod density which is higher than in unit B with maximum densities of ca. 50,000 valves for 10 g of sediment.

Unit D, between 150 and 0 cm depth (+50/+200 cm msl), represents the upper unit of the core, formed by a recent agricultural soil.

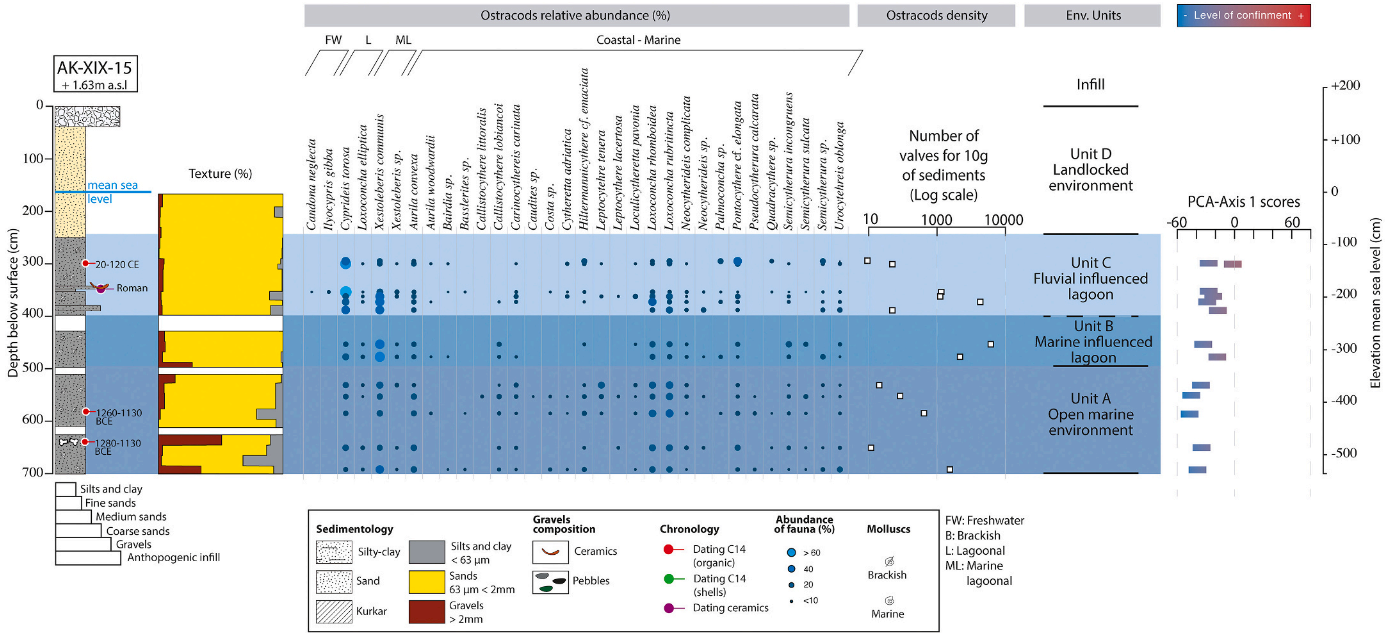

Fig. 8. Sediment texture and ostracod analysis of core AK-XIX-15. Four different environmental units were identified and show the progressive isolation of the area from the sea and the progradation of the river-mouth from the Early Iron Age to the Roman period. See Fig. 3 for location of the core.

- AK17-C8 (600 cm long) was drilled 355 cm above msl, 50–60 m south of Tel Akko and 170 m west of AK-XIX-16 (Figs. 2, 3 and 6).

Unit A is formed by the *Kurkar* sandstone substratum, below 600 cm depth (below -245 cm msl).

Unit B is situated between 600 and 310 cm depth (-245/+45 cm msl). Very few fragments of marine shells are present. The ostracod assemblage is dominated by coastal and marine species (72%). Lagoonal species (*C. torosa*) and marine lagoonal species count for 24% and 4% of the assemblage respectively. In this unit, the density of ostracods is very low (<50 valves for 10 g of sediment) attesting to a high energy coastal environment.

The transition to unit C is marked by the changes in sedimentary texture. Between 310 and 250 cm depth (+45/+95 cm msl), the sediment texture is characterised by an increase in silts and clay to 24% of the total. Sands still comprise more than the half of the sediments (62%), gravels represent 14% of the sediments. In this unit, the lagoonal ostracod *C. torosa* represents 97% of the total assemblage. Few freshwater specimens of the ostracod *I. gibba* are present. *I. gibba* likes permanent water bodies with a clayed or sandy bottom (Athersuch et al., 1989). This unit is typical of a coastal lagoon separated from the sea by a sand spit.

- Core AK16-C6 (1300 cm long) was drilled 320 cm above msl (Figs. 2, 3 and 7).

Unit A is situated between 1300 and 710 cm depth (-780/-390 cm msl). The bottom of the unit – unit A1 (1300–1210 cm depth) – is composed of *Kurkar* sandstone. This unit is overlain by unit A2, a 500 cm thick (1210–710 cm depth) reddish silty to sandy unit (locally known as *Hamra*). Following, unit A3 is a thin layer (between 710 and 700 cm depth) of very compact dry brown clay (as in cores AK-XIX-13 and AK-XIX-16).

Unit B, between 700 and 390 cm depth (-380/-70 cm msl) is composed mainly of sands (88%). Silts and clay represent 8% of the total and gravel 4%. At the bottom of the unit, the gravel fraction reaches 6% between 670 and 700 cm depth; shell fragments and few fragments of *C. caespitosa* were found.

Unit C, between 390 and 320 cm depth (-60/0 cm msl), is still mainly composed of sands (56%). However, the proportion of silts and clay increases to 28%. The gravel fraction (16% of the total texture) is composed of organic remains, shell fragments, and small pebbles. Molluscs are absent from the unit. The ostracod assemblage is clearly dominated by the lagoonal species, *C. torosa* with few individuals living in fresh to brackish water environment (*C. neglecta* and *I. gibba*).

- Southernmost core AK-XIX-15 (700 cm long) was drilled 165 cm above msl (Fig. 8). It is located 300 m south of the limit of tell (Figs. 2 and 3).

Unit A, between 700 and 500 cm depth (-535/-335 cm msl) is characterised by fine grey sands composed of quartz, and marine organisms (shells, foraminifera, ostracods; Fig. 8). This unit contains few marine (eg. *Cerithium vulgatum*, *Conus mediteraneus*, *Donax trunculus*) and brackish (e.g. *Cerastoderma glaucum*, *Melanoides tuberculata*, *Melanopsis buccinoidea*) shells. The ostracod assemblage is composed of coastal-marine species (eg. *Aurila convexa*, *Loxoconcha rhomboidea*, *Loxoconcha rubrincinata*, *Pontocythere elongata*) representing 50–80% of the species, lagoonal (*C. torosa*) species representing 7–16% of the species, and marine lagoonal (*Xestoleberis*) representing 10–30% of the total of the valves identified. The density varies from 10 to 6000 valves for 10 g of sediment.

Unit B, between 500 and 400 cm depth (-335/-235 cm msl) is dominated by fine sands. The ostracods assemblage is dominated by marine and coastal species counting for 54% of the species while marine lagoonal and lagoonal species count for 33% and 13% of the assemblage respectively.

Unit C, between 400 and 250 cm depth (-235/-85 cm msl) is, as the previous units, dominated by fine marine sands. The ostracod assemblage is still dominated by marine and coastal species (>50%) but the proportion of lagoonal (*C. torosa*) and marine lagoonal (*Xestoleberis*) species increases. The ostracod density varies between 10 and 5000 valves for 10 g of sediments.

Unit D, between 250 and 30 cm depth (-85/+135 cm msl), is composed of fine beige quartz sands barren of fauna.

Unit E, represents the upper unit of the core and is formed by recent anthropogenic infills between 30 cm depth and the top of the core (+135/+165 cm msl).

5.4. *Ground penetrating radar*

The representative profile shows subsurface reflections which are interpreted as stratigraphic horizons. The lower radar facies package of continuous to semi-continuous horizontal to sub-horizontal reflections from ~1.5 m to 80 cm is interpreted as *Kurkar* sandstone (Fig. 9). This reflection package is truncated abruptly at 37 m along the transect which we interpret as the location where the *Kurkar* sandstone plateau drops off (erosional truncation). The upper 80 cm are characterised by semi-continuous to discontinuous sub-horizontal to slightly dipping reflections. This upper radar facies is interpreted as fill that has accumulated upon the *Kurkar* sandstone (slope wash from the tell and archaeological materials). A coring program has corroborated the significant change in *Kurkar* sandstone depth below the surface at this location of the tel.

6. Discussion: Tel Akko in its regional context

6.1. *Palaeoenvironmental phases recorded in the cores*

The study of the sedimentary sequences of the Tel Akko coastal plain have recorded the succession of four main environments in the Holocene (Figs. 10 and 11). We put in perspective these changes in a broader context of environmental changes and human habitation patterns (Fig. 12).

6.1.1. *Phase 1: continental pre-transgressive environment*

Cores drilled close to the tell have reached the pre-transgressive continental substratum (Fig. 10). This succession of environments has been well recorded in core AK16-C6 (Fig. 8). We observe a succession of *Kurkar* sandstone, reddish silty to sandy soil and coastal brown silty-clay. The *Kurkar* sandstone is commonly found along the coastal plain of Israel. In some areas, as Tel Akko, it emerges from the plain to form small to medium elevated hills. The reddish palaeo-soil, locally named *Hamra*, is widespread along the coast of Israel (see Porat et al. (2004) for a discussion about mode and timing of *Kurkar* and *Hamra* formation). The brown silty-clay pre-transgressive soil is present in most of the bay. Zviely et al. (2006) have dated the top of this soil unit to 8300 ± 50 BP (9130–9450 cal. Years BP) at a depth of -500/-545 cm msl in the centre of the Haifa/Akko Bay. To the south, a radiocarbon date at the base of a marine unit (at -1350 cm msl) covering the paleo-soil shows an age of 8240 ± 60 BP (8160–8640 cal. Years BP; Avnaim-Katav et al., 2012). At Tel Akko, it has been identified in cores AK16-C6, AK-XIX-13 and AK-XIX-16 between -380 and -510 cm msl (Fig. 10). 8000 years ago, sea level was not higher than -13.5 to -16.5 m (Sivan et al., 2001) and thus the coastline was located ca. 4.5 km west of its present position (Zviely et al., 2006). The Early-Holocene soil identified in our cores may be attributed to fluvial sedimentary inputs by the Na'aman River in the context of increasing precipitations between 9000 and 7500 years BP (Elyashiv et al., 2016). Different studies based on distinct proxies such has pollen data from Sapropel S1 (Rossignol-Strick, 1999) and δ18O from cave speleothem (Bar-Matthews et al., 2000; Cheng et al., 2015), point toward warm-humid conditions during the Early Holocene in the southern Levant (Fig. 12).

6.1.2. *Phase 2: marine transgression and development of a coastal environment during the Chalcolithic/Bronze Age period*

The maximum sea incursion in Haifa/Akko Bay occurred before 4000 years ago flooding the Zevelun plain under several meters of sea water and leading to the displacement of the shore some 4 km landward (Zviely et al., 2006; Elyashiv et al., 2016). At that time, the sea level stabilised and the progradation of the coast started because of marine (Nile quartz sands transported by the longshore drift currents) fluvial (Qishon and the Na'aman Rivers) sediment inputs to the bay. In the Mediterranean, sea level variations over the last 4000 years are still poorly understood (Dean et al., 2019), although they have had a fundamental role on sedimentary dynamics and coastal urbanisation (Giaime et al., 2019). Based on the available data, sea level was above -1 m msl around 2000 years BC (Sivan et al., 2001; Porat et al., 2008; Dean et al., 2019). A shallow marine environment dating back to the Chalcolithic period onward, has been identified in all cores by the presence of marine organisms such as molluscs, ostracods and coral fragments found

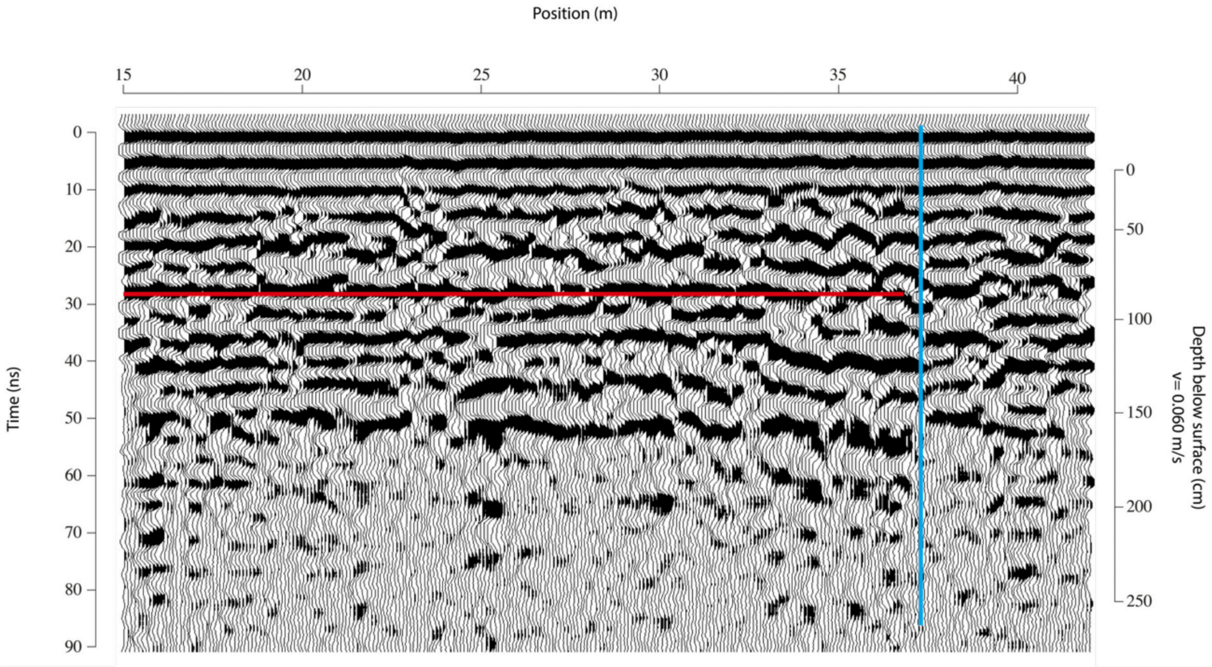

Fig. 9. A representative GPR transect collected from north to show on the southern portion of Tel Akko using a Sensors and Software pulseEKKO system with 225 MHz antennae. The horizontal line (red) represent the upper limit of the *Kurkar*sandstone layer. Above, the sedimentation corresponds to the deposition of sediments eroded from the tell and archaeological materials. The vertical line (blue) show the erosional truncation linked to the drop of the *Kurkar*sandstone plateau. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

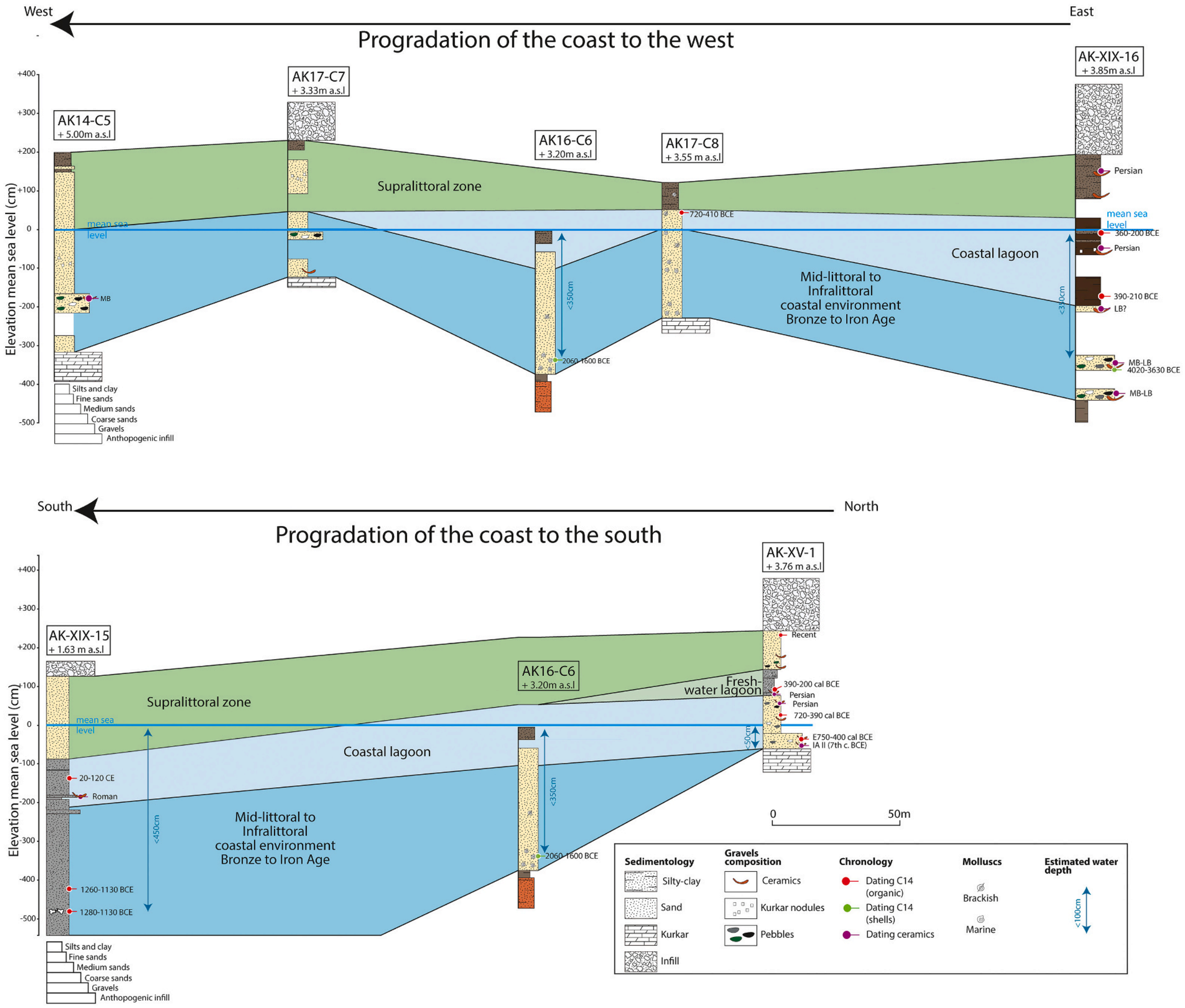

Fig. 10. Correlation of sediment cores retrieved from the south of Tel Akko. Location of cores and transects are given in Figs. 3 and 11. These transects show the progradation of the coast and the land locking of the environment. They illustrate the southward and westward displacement of the coast from the Bronze Age to the Early Roman period. Costal zonation in the Mediterranean after Pérès and Picard (1964) and Pérès (1982). Supralittoral zone: not submerged but impacted by sea spray; Mid-littoral zone: area affected by waves (and tides); Infralittoral zone: coastal zone: totally immersed (its lower limit corresponds to the limit of light penetration). Core AK-XV-1 is from Giaime et al. (2018). Radiocarbon dates have been calibrated using the new calibration curve IntCal20 (Reimer et al., 2020).

in marine sands (Figs. 10 and 11). The marine coral species *C. caespitosa* typically develops on shallow (<10 m) or deeper (>30 m) rocky substrates and is able to thrive in turbid waters at relatively low irradiance (El Kateb et al., 2016). This is consistent with the nearby presence of the Na'aman River mouth that transports fine sediments (mainly silts and clay) and freshwater fauna discovered in this unit (Avnaim-Katav et al., 2016). The *Kurkar* sandstone, which is not always covered by the clay soil (AK17-C8) may have constituted a perfect substrate for the coral to fix and grow. PCA analysis of the data shows a gradient of confinement toward the south-eastern corner of the tell. AK-XIX-16 shows moderate level of confinement (Fig. 5), which is associated with the preponderance of *C. torosa* in most of the samples. The absence of freshwater ostracods and the few number of freshwater to brackish molluscs bring us to interpret this unit as a sheltered coastal environment with variable salinity due to the influence of freshwater inputs. AK-XIX-13 PCA's scores demonstrates a lower degree of confinement than in AK-XIX-16 (Fig. 4). The level of confinement further decreases toward the west as demonstrated by the PCA scores obtained in cores AK16-C6 and AK17-C8 (Figs. 6 and 7). In these cores, the coastal environment is shown by the presence of fine to medium quartz sands with the presence of coastal ostracod species.

6.1.3. Phase 3: infilling of the southern area of Tel Akko from the Bronze to the Iron Age (2nd and 1st millennium BCE)

At the scale of the Haifa/Akko Bay, available data (Zviely et al., 2006, 2007; Porat et al., 2008) point toward a rapid progradation of the bay, from the Bronze Age onwards, up to an average of 40–50 cm/year (Fig. 12). However, this assumption is based on a few sedimentological and geochronological data points and not making it possible to highlight spatial variability in the infilling of the bay. At a much higher resolution, the apparent sedimentation rate appears reduced at Tel Akko, illustrating that local spatio-temporal variabilities are evident. Comparison of cores AK16-C6 and AK17-C8 demonstrate that from 2450 to 1980 cal. Years BCE (Early to Middle Bronze) to 720–410 cal. Years BCE (Late Iron Age), the area at the foot of the tell experienced a vertical aggradation rate of 2.06 ± 0.27 cm/years due to the deposition of marine sands (Fig. 10). We assume that this low comparative sedimentation rate is due to the retention of sediments by geomorphological features in the bay. Close to the tell, the presence of a *Kurkar* sandstone outcrop (Figs. 3 and 11) must have acted as a natural coastal groyne or dyke preventing marine sediments, moved northward by the longshore drift, to reach Tel Akko (Giaime et al., 2021). When the area located down drift of the *Kurkar* sandstone outcrop was filled, large quantity of quartz sands was able to by-pass the outcrop and reach the foot of the tell as shown in the

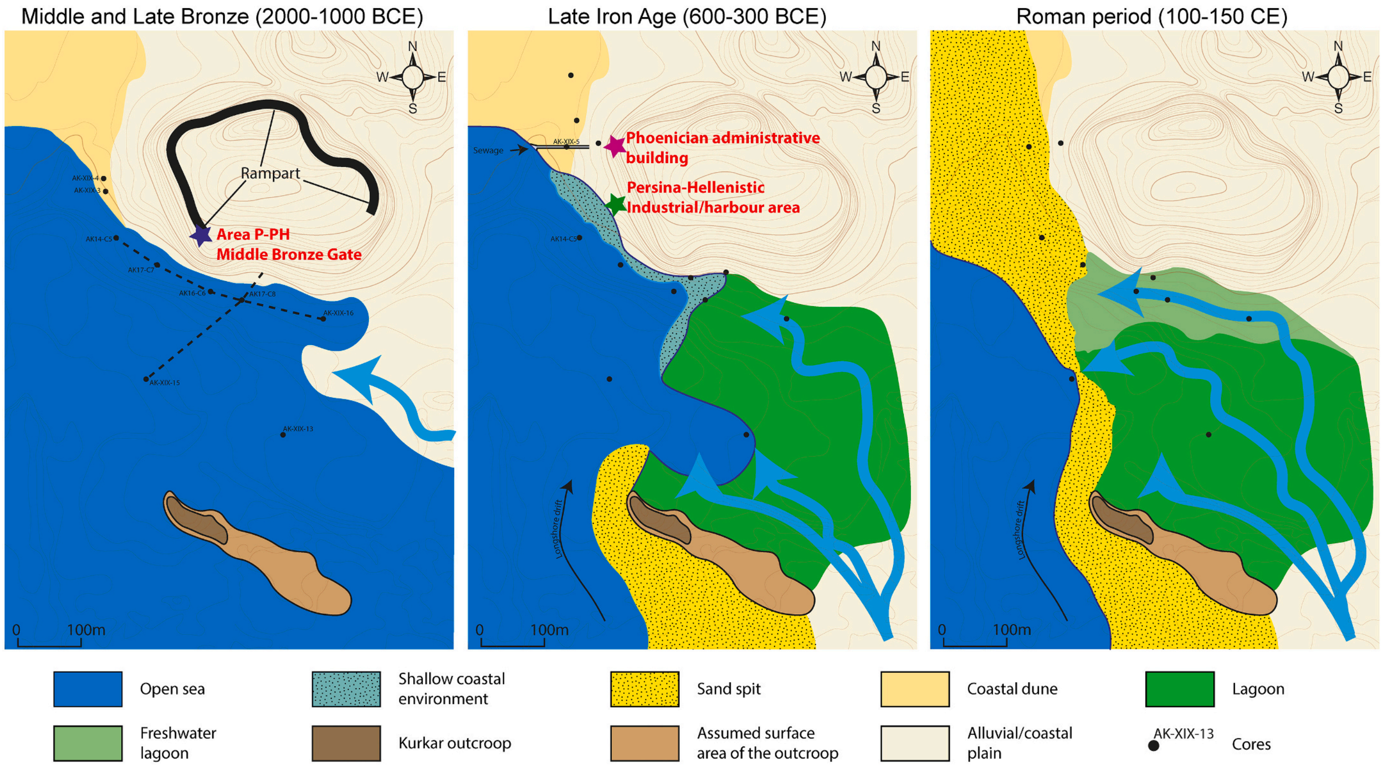

Fig. 11. Palaeoenvironmental evolution of Tel Akko maritime interface from the Bronze Age to the Early Roman period. The different environments have been identified based on this study and from previous results published in Morhange et al. (2016); Giaime et al. (2018) and Giaime et al. (2021). Topography was adapted from the Treidel map shown in Fig. 2.

collected cores.

The land locking of the area, with the evolution to a coastal lagoon is dated to the middle-1st millennium BC, namely the Phoenician-Persian period (Figs. 10 and 11). In its early stage of confinement, our data show that the lagoon seems to have received moderate fluvial influence with the exception of core AK-XIX-16 which is dominated by freshwater and lagoonal ostracods and molluscs and silty-clay sediments (Unit C). In contrast, unit C of core AK-XIX-13 shows that the marine influence, in the form of coastal-marine fauna was more important further south (Fig. 11.B). It is only during the Late-Persian period that the area close to the tell lost its coastal setting and was occupied by a fluvial-dominated lagoon as demonstrated by unit C in cores AK16-C6 and AK17-C8 (Figs. 10 and 11.B). This freshwater influence was probably linked with a meandering arm of the Na'aman River. The coastal lagoon, separated from the open sea by the sand barrier spit was progressively infilled by fluvial sediments (Fig. 11.B).

6.1.4. *Phase 4: complete land locking of Tel Akko in the Hellenistic period*

The area 300 m south of Tel Akko, is characterised by a permanence of the marine environment as demonstrated by core AK-XIX-15. The marine dominated infralittoral environment identified during the Early Iron Age (late 2nd Millennium BCE) progressively shows signs of confinement as demonstrated by the evolution of the PCA scores for core AK-XIX-15 (Fig. 8). Nevertheless, the scores remain negative for all the samples which demonstrate that the land locking of the area was in an early stage during the Early Roman period (Fig. 11.C). At the same period (1965 ± 35 BP; 40 cal. Years BCE – 200 cal. Years CE), Morhange et al. (2016) identified a marshy environment close to the tell, ca. 200 m north of core AK-XIX-15. This marshy environment is followed by the development of a freshwater influenced lagoon during the 1st millennium CE and shows the complete land locking of the area at that time. This is confirmed by the development of a supralittoral environment in our cores (Fig. 12).

6.2. *How did Tel Akko's inhabitants deal with coastal changes?*

6.2.1. *A perfect coastal setting at the beginning of the Bronze Age settlement*

Like many other Levantine Bronze Age maritime sites, Tel Akko's inhabitants have exploited the unique coastal setting provided by the *Kurkar*/sand hills along the shore. Thus, over the centuries, Tel Akko's settlement pattern has been strongly linked to the coastal geomorphology of the area. In the Middle Bronze Age, the habitations were situated on top of the hill and linked to the anchorage area with a gate, located in area P on the southern side of the tell, where numerous imported ceramics have been found in Raban's excavations (Fig. 2). It is also assumed to have been the location where a main entrance to the tell was situated in the Bronze Age periods (Raban, 1991; Artzy, 2018). Considering a sea-level elevation of -1 m msl at that time, the water depth was >2 m at the immediate proximity of the tell, allowing boats to approach (Fig. 11). This environment was perfect for the anchoring and unloading of boats at the immediate proximity of the habitation centre. Moreover, the southern limit of the tell was naturally protected from possible external invasions which explains the absence of fortification in contrast to the northern side (Raban, 1991). At that time, in the absence of proper harbours along the coast, beaching of seafaring ships has been proposed as an important practice (Votruba, 2017 and references therein). Votruba (2017) proposed that the beaching practice was commonly used for small size ships while larger ones would have been anchored along the shore. This practice is presented in lithographs, such as Robert's from the 19th century CE (Roberts, 1843). Lighters may have been used to unload larger merchantmen anchored in naturally protected area but this would necessitate organized anchorages, as Akko's might have well been in the 2nd and 1st Millennia BCE. We can imagine the development of proto-harbours, characterised by the installation of light structures such as quays or moles. The high *Kurkar* sandstone ridge on the southern side of the tell could have also been used to facilitate unloading of goods. However, such structures have left few, if any,

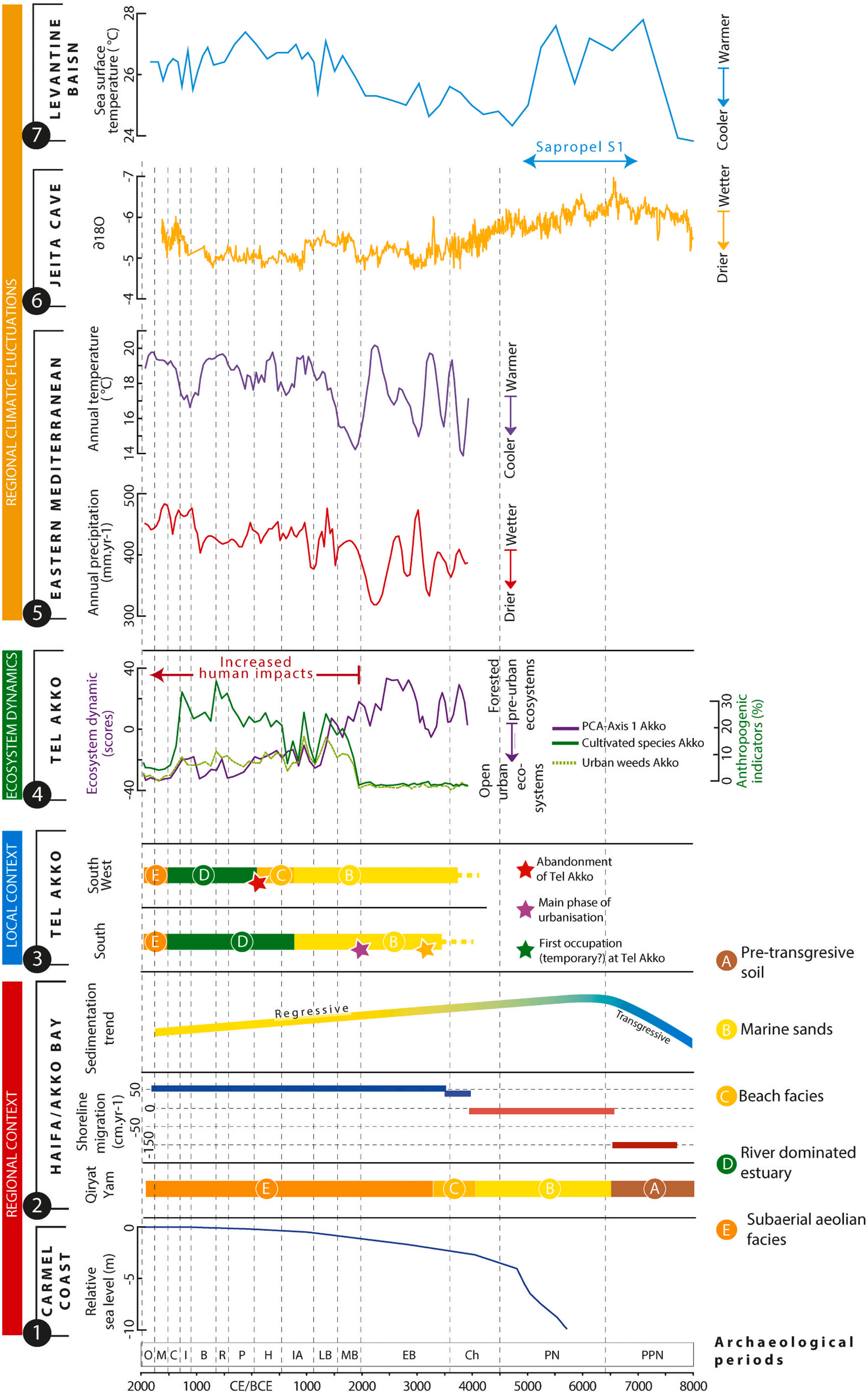

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Fig. 12. Summary diagram of Tel Akko's environmental changes associated with regional climatic data. The base of the diagram shows geomorphological changes at regional and local scale. The upper part present climatic changes. Sea level trend is from Lambeck and Purcell (2005). Geomorphological evolution of the Haifa/Akko Bay has been reconstructed using Zviely et al. (2006), Porat et al. (2008) and Elyashiv et al. (2016). Tell Akko ecosystem dynamics and regional precipitation and temperature reconstructions from Kaniewski et al. (2013). Delta O18 data from Jeita Cave obtained from Cheng et al. (2015). Reconstructed sea surface temperature from Castañeda et al. (2010). Archaeological periods: PPN (Pre-Pottery Neolithic); PN (Pottery Neolithic); Ch (Chalcolithic); EB (Early Bronze Age); MB (Middle Bronze Age); LB (Late Bronze Age); IA (Iron Age); P (Persian); H (Hellenistic); R (Roman); B (Byzantine); I (Islamic); C (Crusaders); M (Mamluk); O (Ottoman).

traces in the archaeological and sedimentary record.

Raban (1991) proposed that the Bronze Age harbour was located on the lower area of the tell, in the inner part of the "moon crescent" and directly associated with the Na'aman River mouth and the sea (Fig. 3). However, his assumption that the inner part of the tell was used as an "inner harbour" is problematic. The GPR profile and coring negates Raban's 'inner harbour' theory. We found that the *Kurkar* sandstone is too high and hence the water column too shallow for any boat, even a barge, to enter the area. Fig. 10 shows that the water column was never >50 cm. A rapid drop in the *Kurkar* sandstone is noted in the GPR transect and in the cores precisely in the waterfront area where boats could have sailed from the coast toward Tel Akko, but not in the depression itself (Fig. 11.A).

At the scale of the bay, a number of tells are dated from the Bronze Age period (Fig. 1). However, they likely did not benefit from a direct and permanent access to the sea as Tel Akko (Fig. 1B; Zviely et al., 2006). Tel Abu Hawam was the second main coastal/anchorage site of the bay, located in its southern margin, in the Qishon river mouth (Fig. 1B). Both Tel Akko and Tel Abu Hawam (TAH), were advantaged by the rivers for the terrestrial routes to the economic hinterlands and associated successful commercial activities (Hamilton, 1934; Balensi, 1985; Artzy, 1997, 2018). TAH, however never reached the same expansion as Tel Akko, probably because of a much more dynamic environment. Located directly in the mouth of a river, the tell was indeed directly impacted by important sedimentary inputs from the sea and the river. Tel Akko was part of the regional maritime network in the Middle Bronze IIA period (Stager, 2001), TAH's important role as a maritime interface only started in the Late Bronze IIB period. TAH's environmental limitations may be seen by its non-continuous occupation (TAH was settled only in limited periods, mainly in the Late Bronze II, the Iron Age and the Persian/Hellenistic periods) and the changes in settlement patterns identified during excavations.

The favourable environment found by the earliest inhabitant of Tel Akko allowed the creation of an important urban maritime centre. This urban development has important consequences on the landscape, with a gradual disappearance of the forest cover toward a much more urbanised landscape (Kaniewski et al., 2013. Fig. 12.4). This appears in a context of reduced climate variability and dried climate compared to the Early Holocene period (Fig. 12.5–7). Along the Israel coastal plain, an increasing urbanisation phase is attested in the Middle Bonze Age (Faust and Ashkenazy, 2009).

6.2.2. *Impact of coastline progradation and associated changes in anchorage and habitation patterns*

Hyper sedimentation has always been a major threat for maritime cities located in deltaic contexts leading to the displacement of the anchorages or harbour basins through time (Goiran et al., 2015; Morhange et al., 2015; Giaime et al., 2019). At Tel Akko, the migration of the anchorage areas is clearly illustrated by the ceramics found in the cores. For instance, the cores collected in the south-eastern part of the tell, show coastal/marine sediments at the foot of the tell that contain ceramics dated to the 2nd Millennium BC (namely MB and LB periods). Cores collected on the south-western part of Tel Akko, contain a large number of mid-1st Millennium (Late Iron Age) ceramics along with few examples of earlier Iron Age ones again, from coastal/marine units. Thus, the Bronze Age maritime activities associated with the tell are located mainly below its central southern foothill. Iron Age – Phoenician-Persian – maritime interface of Tel Akko was, however, centred on the south-western side of the tell (Fig. 11.B; Bergevin et al., 2020; Giaime et al., 2021). When Tel Akko lost its access to the sea by the Hellenistic period, it was abandoned in favour of the settlement on the Akko promontory (Old City) where a harbour was constructed (Artzy, 2012; Sharvit et al., 2021).

7. Conclusion

With the advent of urbanisation in the early 2nd Millennium BCE, numerous maritime cities flourished along the Levantine coast. Tel Akko inhabitants exploited the unique coastal setting provided by the *Kurkar*/sand hills along the shore. The nearby presence of freshwater springs and of the Na'aman river allowed the development of agriculture leading to the decrease of the pristine forested ecosystem at local scale.

From a palaeoenvironmental perspective, Tel Akko is one of the most investigated sites of the Mediterranean coast of Israel. High-resolution studies of new sedimentary cores, radiocarbon dating along with dating obtained from ceramic sherds, and GPR data enabled us to elucidate the geomorphological setting of the Akko coastal plain, south of Tel Akko and its evolution during the Late-Holocene (last 5000 years). Our research demonstrate that the Tel Akko history, as a major urban and maritime centre, is deeply influenced by its access to the sea. Vicissitudes in habitation patterns within Tel Akko and its environs dating back to the Early Bronze and Middle Bronze periods, were noted during the archaeological excavations and can be explained in terms of the changes that have taken place in the coastal plain. Our results demonstrate that the area shows a classic geomorphological evolution defined by a maximum marine transgression before 4000 BP, rapidly followed by coastal progradation due to the accumulation of marine sands.

During the Middle Bronze Age an impressive defensive rampart was constructed especially on the northern side of the tell. An entrance was left in the defences on the south, bordered by the sea which protected Tel Akko from invasions. An anchorage was likely in function along the tell's coastline. This area was progressively infilled because of marine sedimentary deposition by longshore drift currents as well as the sedimentary infilling by the river. In the middle of the 1st Millennium BCE (Phoenician and Persian periods), Tel Akko's direct access to the sea was reduced and the maritime activities were relocated to its western side where public administrative buildings have been found along a higher energy coastline. In the last part of the 1st Millennium BCE, the increasing sedimentary inputs lead to further progradation of the coastline to the west and the demise of anchorage possibilities close to the tell.

While the 2nd and most of the 1st Millennia BCE anchorages were in close proximity to the tell, the decision of the local inhabitants and authorities to build an artificial harbour in the bay, where the Old City of Akko is presently located, in the Hellenistic period (2nd century BCE) seems to have been mainly driven by environmental pressures. This, newly occupied, sector of the bay was far from any fluvial influence and the marine sedimentary inputs were greatly reduced. The relocation of the harbour activities led to the progressive end of the city of Tel Akko and its eventual abandonment in the 2nd or the early 1st century BCE.

The finds demonstrate that Tel Akko's ancient anchorages – and as a consequence habitation patterns – moved toward the west (the sea) is driven by coastal changes over the millennia and is an archetypal of the race to the sea of maritime cities located in a deltaic context.

Data availability statement

Data used in this study are accessible through the Universitat Autónoma de Barcelona Digital Repository of Documents: 10.5565/ddd.uab.cat/257530.

Author contributions

Conceptualization: M.G., M.A., H.J.; Data curation: M.G., M.A., H.J., Y.S., G.L., A.A.H.; Formal analysis: M.G., M.A., H.J.; Funding acquisition: M.G., M.A., H.J.; Laboratory analysis: M.G., M.A., H.J.; Methodology: M.G., M.A., H.J.; Project administration: M.A., H.J.; Writing: M. G., M.A., H.J.; All the authors revised the original manuscript draft and agreed on the submitted version.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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