Research paper



# Evaporite evidence of a mid-Holocene (c. 4550–4400 cal. yr BP) aridity crisis in southwestern Europe and palaeoenvironmental consequences

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## Abstract

Sedimentological evidence for an abrupt dry spell in south-eastern Spain during the middle Holocene, from *c*. 4906 to 4384 cal. yr BP, is presented. This phase was determined primarily from halite beds deposited between muddy slimes in a lagoon system of Puerto de Mazarrón (Murcia province) with a peak phase from *c*. 4550 to 4400 cal. yr BP. A multi-core, multi-proxy study of 20 geotechnical drills was made in the lagoon basin to identify the main sedimentary episodes and depositional environments. The results suggest that this halite bed, more than 80 cm thick, was conditioned by climate change and was accompanied by a generalized drying-out of the basin. Halite precipitation was linked with palaeoecological changes, including forest and mesophyte depletions and increasing cover and diversity of xerophytic plant species. Archaeological evidence indicates a demise of the population at this period probably due to resource exhaustion. An overall picture of the biostratigraphy and palaeoclimates of the region is given in a broader geographical context.

#### **Keywords**

aridity crisis, climate change, halite, Holocene, lagoon, pollen tests, Western Mediterranean

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## Introduction

This paper is the result of a broader multidisciplinary research project, whose objectives deal with the reconstruction of human cultures and environments over the coastal lines of south-western Europe throughout the late Pleistocene and Holocene. Two beds of pure halite were found in 1 of the 20 geotechnical logs drilled in the location of former lagoons; these lay several metres deep, in between muddy deposits. The thicker of the two beds, about 1 m thick, was radiocarbon dated between 4540 and 4384 cal. yr BP.<sup>1</sup> We will discuss that this finding suggests a cold, arid period, within a warm and wet trend in the mid-Holocene climate (Cacho et al., 2010). The Mazarrón saline event is supported by the non-presence of pollen and the identification of microfossils cold water.

Two halite beds are located near the coast of the Puerto de Mazarrón (Murcia province, south-eastern Spain), an area that was formerly occupied by coastal salt flats (Figure 1). Geologically, the study area lies in the Betic Zone *s. str.*, belonging to the Betic Cordillera, and corresponds to a Neogene-Quaternary basin infilled with evaporates of detritic and clay deposits. The area is a tectonically unstable sector, at least since the late Pleistocene, where neotectonic faults had greater importance than fluvio-littoral processes in shaping the landscape (Rodríguez-Estrella et al., 2011).

## Methods

The multidisciplinary project involved the following studies: sedimentological description of 20 geotechnical boreholes (Figure 2), radiometric dating, palynological analysis, anthracology, micropaleontological data and archaeology of the study area.

Five consecutive halite core samples from borehole MAZ-17 were selected for mineral and geochemical characterization. As the halite samples were poorly cemented, they were first washed with pure ethanol in order to avoid mineral precipitation from pore water. Mineral identification used an Environmental Scanning Electron Microscopy FEI Quanta 200 coupled with an energy-dispersive spectroscopy (EDS) spectrometer EDAX-Genesis. Chemical composition of primary brines (fluid inclusions) during halite precipitation was determined by means of the

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Figure 1. Geographical location of the study area.



**Figure 2.** Situation of the investigation boreholes and of the geological transects. Source: Rodríguez-Estrella et al. (2011).

cryo-scanning electron microscopy (SEM)–EDS method, described in García-Veigas et al. (2009, 2011) using a SEM JEOL-840 with an Oxford-CT1500C cryo-unit stage and an x-ray spectrometer (Oxford-INCA). Sulphate minerals in saline beds

(mainly gypsum) were dissolved and re-precipitated as BaSO<sub>4</sub> and then the  $\delta^{34}S_{V-CDT}$  was determined with a Carlo Erba 1108 elemental analyser and the  $\delta^{18}O_{V-SMOW}$  with a thermo-conversion elemental analyser (TC–EA) unit, both coupled to an IRMS

Boring	Sample depth	Laboratory code	BP date	Cal. yr BP date <sup>a</sup>			
				Confidence interval (2σ, p = 0.954)	Median	$2\sigma$ most probable range	
MAZ-12	-3.90 -7.10 -8.27	Poz-26491 Poz-26492 Poz-28022	5820 ± 40 13,600 ± 100 25,690 ± 430	6503–6730 16,431–16,984 29,603–31,093	6628 16,748 30 473	6528–6730 16,431–16,984 29,603–31,093	
MAZ-20	-4.20 -6.00 -7.80	Poz-31907 Poz-31098 Poz-31909	$7440 \pm 60$ 5790 ± 50 5170 + 40	8074–8387 6470–6726 5758–5998	8266 6589 5930	8162–8387 6470–6679 5884–5998	b
	-1.00 -14.70	Poz-31910 Poz-31911 Poz-26497	$10,410 \pm 70$ $8090 \pm 60$ $7670 \pm 60$	12,060–12,542 8773–9251	12,287 9022	12,060–12,542 8773–9144	b
MAZ-15	-7.28 -1.40	Poz-28123 Poz-21033 Poz-28023	$7480 \pm 50$ 1665 ± 35 1850 + 50	8192–8381 8192–8382 1420–1693 1628–1897	8299 1569 1785	8192–8381 8192–8382 1513–1634 1691–1897	
	-2.22 -2.48	Poz-28025 Poz-32654	3220 ± 30 3700 ± 35	3371–3553 3927–4149	3432 4038	3371–3481 3962–4104	
	-3.27 -3.39	Poz-32655 Poz-28026	3900 ± 50 4100 ± 40	4155–4506 4447–4819	4331 4621	4220 <u>444</u> 0 4516 <u>4</u> 728	
	-3.54 -4.08 -4.80 -5.86 -6.15	Poz-28083 Poz-21034 Poz-28084 Poz-28028 Poz-28029	4055 ± 35 3335 ± 35 3810 ± 35 3785 ± 35 6550 ± 50	4424–4687 3472–3680 4087–4402 3999–4288 7333–7567	4525 3569 4201 4165 7463	4424-4627 3472-3641 4087-4298 4079-4259 7415-7567	b
MAZ-13	-6.50 -4.95 -5.60 -8.00 -14.00	Poz-21101 Poz-31012 Poz-31011 Poz-26493 Poz-26494	$6760 \pm 50$ $3925 \pm 35$ $3210 \pm 35$ $4180 \pm 40$ $4440 \pm 40$	7517–7684 4244–4506 3362–3553 4581–4839 4876–5282	7617 4360 3426 4718 5050	7563–7684 4244–4440 3362–3483 4606–4769 4876–5083	b
MAZ-14	-7.80 -8.80	Poz-31010 Poz-26495	4125 ± 35 4340 ± 40	4528-4820 4839-5035	4666 4914	4528–4729 4839–4978	
MAZ-17	-2.00 -2.40 -5.40	Poz-29937 Poz-31015 Poz-31014	$1590 \pm 35$ $2550 \pm 35$ $3720 \pm 35$	1400–1548 2493–2752 3933–4219	1470 2632 4060	1400–1548 2681–2752 3972–4155	
	-6.00	Poz-29974 Poz-26544	3070 ± 30 3950 ± 50	3215–3362 4245–4524	3296 4403	3215–3362 4245–4524	b
	-8.30 -9.20 -10.60	Poz-31013 Poz-26498 Poz-29972	4065 ± 35 4330 ± 35 4300 ± 35	4436–4801 4839–5027 4829–4961	4553 4899 4861	4436–4645 4839–4973 4897–4961	

Table 1. C14 datings from different sediment samples in the boreholes drilled.

Shown in grey shading the dates related to the Mazarrón saline event (MAZ-17) and the pollen diagram (MAZ-15).

<sup>a</sup>Calib 14C Calibration Program, v. 6.0. 2010 (Stuiver et al., 2010).

<sup>b</sup>Fluvio-torrential inputs to the lacustrine environment (modified from Rodríguez-Estrella et al., 2011).

Thermo Finnigan Delta Plus XP. Bromine content was determined by x-ray fluorescence on pressed powder tablets after being crushed in ethanol.

For calcareous and siliceous microfossils, sediment samples were prepared according to the standard randomly distributed method (Flores and Sierro, 1997). In the case of siliceous microfossils, HCl and  $H_2O_2$  were previously added to a known weight of dry sediment to remove carbonates and organic matter. Sediment was rinsed several times with bi-distilled water in order to get acid-cleaned material. For calcareous nanofossils, counts were done using a Leica DMRP polarizing microscope at 1000× magnification, while for diatoms, a Leica DMLB with phase-contrast illumination at 1000× magnification was used. In both cases, percentages of the most significant palaeotaxa were calculated.

Sediment samples were collected at different depths in 10 of the 20 boreholes for radiocarbon data. A total of 34 datings were made about total organic carbon (TOC) in the Poznan Radiocarbon Laboratory. Their calibration had been made by 14C Calibration Program, v. 6.0. 2010 (Stuiver and Reimer, 1993); as an argument for discussion has been taking into consideration, the median probability under calibration ranges to  $2\sigma$  (95.4 confidence interval; Table 1). Dating and interpretation of sedimentary events and their absolute dating already made by Rodríguez-Estrella et al. (2011) can be summarized as follows:

- 1. The peak of the Flandrian Transgression took place on 7463 cal. yr BP in the studied basin.
- The Mazarrón saline event occurred between 4899 and 4403 cal. yr BP.
- 3. At the time of the post-Flandrian marine regression, fluvial episodes continued under an increasingly arid climate. These episodes remobilized earlier deposits giving rise to a chronological inversion with anomalous evidences (Rodríguez-Estrella et al., 2011). Fluvial evidence deposits occurred in a warm and wet period, after the arid phase, characterized by torrential rainfall conditions.



**Figure 3.** Chronostratigraphic correlations in the Mazarrón coastal salt flats. P: Pliocene lumachels; Me: Marls, sandstones and gypsums from the upper Tortonian and lower Messinian; P-T: Permo-Triassic carbonate and metamorphic rocks; Ca: Caliche (Calcrete); S: Marine; L: lagoonal; F: fluvial; A: anthropogenic.

(Modified from Rodríguez-Estrella et al., 2011).

In borehole MAZ-15, a complete palynological study was carried out, correlating its lithostratigraphy with the other analysed sections; sedimentary records in sections were contextualized by means of anthracological and archaeological data from population episodes before, during and after the halite deposition, both in the immediate area and beyond (south-eastern Iberia). Extraction and concentration of palynomorphs from the sediments in borehole MAZ-15 were made following the standard procedure described in Moore et al. (1991). Exotic Lycopodium clavatum tablets of a known concentration were added to calculate pollen concentrations. Palynomorphs were concentrated by means of ZnCl<sub>2</sub> solution flotation. Palynological identification and counting were performed using an optical microscope and the reference collection of Department of Plant Biology at the University of Murcia. Identification of non-pollen palynomorphs followed Van Geel et al. (1989), Boros et al. (1993), Strother (1996) and Carrión and Van Geel (1999). In the case of fungal palynomorphs, we used the nomenclature by Kalgutkar and Jansonius (2000). Microscopic charcoal particles larger than 50 µm were also counted on pollen slides. Psimpoll software (Bennett, 2007) was used to plot the pollen diagram. Percentages of each taxon were based on a pollen sum (c. 119-532 grains), excluding the indeterminable pollen, pteridophyte spores, hydro- and hygrophytes, Amaranthaceae and non-pollen microfossils.

## **Results and interpretation**

## Halite genesis

According to Rodríguez-Estrella et al. (2011), the land occupied by the former Puerto de Mazarrón coastal salt flats comprises a double tectonic trough: one to the east and the other to the west separated by a sedimentary threshold. Salt flats grade westwards to the Moreras delta cone (Figure 3).

The Quaternary base of the western lagoon has a maximum age of 30,473 cal. yr BP, and the Las Moreras delta cone dates to 9022 cal. yr BP. Halite beds found in the borehole MAZ-17 occur in the eastern lagoon (Figure 4).

The base of the MAZ-17 section consists of sands with shell remains which are interpreted to be the result of a 'marine transgression', when the sea flooded the newly formed trough. Above these sands, a marine-lacustrine series with fluvial beds of fine detritic and mud deposits occur. Two beds of halite are intercalated within this stratigraphic level: the lower halite layer, some 20 cm thick, precipitated in 4906 cal. yr BP, while the upper one, approximately 1 m thick, formed between 4540 and 4584 cal. yr BP. Lateral changes in halite thickness suggest an isolated brine body gathered in a shallow depression followed by the sporadic seawater influx over a rudimentary sandbar. Halite precipitation indicates the onset of a phase of very arid climate. Their origin is linked to reduction and supersaturation of residual brackish waters that overlay the impermeable base of dark silts and sandy muds (Navarro-Hervás et al., 2009). An acute phase of arid climate would have caused the gradual crystallization of the salt over a minimum of 115 and a maximum of 380 consecutive years, with an intermediate interruption of less acute aridity.

The fact that halite is present in borehole MAZ-17 but not in the nearby borehole MAZ-13 is attributed to the Mazarrón Fault reactivation. Subsidence of the northern block gave rise to a larger volume of water and sediments, thus impeding supersaturation of the salt (Figure 4).

Taken as a whole, the former eastern lagoon can be typed as a moderately deep coastal linked to fluvial action, located in a flood plain (Daley, 1973), in which lutites and seashells predominate. Finally, the lake can be classified as eutropic (Kukal, 1971) and almost dystrophic. Of the two models of sedimentation with abundant organic material considered by Picard and High (1981), this lagoon belongs to the type with an increasing concentration of organic muds or sapropels towards the centre of the basin. In the study area, the organic deposits have been conserved in a reducing environment with abundant pyrite and strong smell of hydrocarbons in the cores extracted from the boreholes.

It belongs to the type of small estuarine or coastal saline ponds and lakes, as defined by Collinson (1978). These types of lakes are typically short-lived features because of rapid silting up; nonetheless, in our case study, the lake ought not to have been ephemeral enough because of its subsidence caused by the neotectonic movements.

It can also be classified as a meromictic lake, with a strong, stable density stratification – the bottom water is stable because of



Figure 4. Lithology and chronology of borehole MAZ-17.

the steep gradient of chemical solutes. There is a strong change in density over the pictocline, which divides the deep, stable monimolimnion from the rest of the water body (Hutchinson, 1975). This ancient phenomenon can be detected even today in the groundwater beneath the former lake. Hydrochemical analyses of samples from borehole 16 indicate a transition from conductivity of 68,800  $\mu$ S/cm at a depth of 0.67 m to 145,278  $\mu$ S/cm at a depth of 3 m.

# Mineralogy, geochemistry and micropaleontology of the saline beds

Saline beds in the borehole MAZ-17 consist of centimetre-sized halite crystals showing cubic textures indicative of subaqueous precipitation. Meanwhile, vertically oriented 'chevron textures' (which would indicate formation of the halite crystals on the floor of very shallow saline ponds) were absent. Abundant millimetre-sized gypsum (CaSO<sub>4</sub>·2H<sub>2</sub>O) crystals exhibiting idiomorphic prismatic habits are scattered between the halite crystals. Minor amounts of micrometric celestine (SrSO<sub>4</sub>) arranged in radiated aggregates are present, as well as framboid pyrite. Detrital fraction is very scarce and consists of quartz, K-feldspar, calcite and clay minerals together with calcareous nanoplankton (coccolithophorids).

The composition of the fluid inclusions, expressed in moles per kilogram H<sub>2</sub>O, averages  $5.1 \pm 0.1$  (Na<sup>+</sup>),  $0.7 \pm 0.7$  (Mg<sup>2+</sup>),  $0.2 \pm 0.1$  (K<sup>+</sup>),  $0.3 \pm 0.1$  (SO<sub>4</sub><sup>2-</sup>) and  $6.1 \pm 0.1$  (Cl<sup>-</sup>). All these

concentrations fit well with those expected for evaporated seawater at the beginning of halite precipitation (McCaffrey et al., 1987). Bromine content in the Mazarrón halite crystals ranges between 55 and 78 ppm, close to the concentration obtained in the first halite precipitated from modern marine brines (Holser, 1966; Valyashko, 1956).

The sulphate isotope composition of the Mazarrón samples averages +17.8‰  $\pm$  0.6‰ and +14.8‰  $\pm$  0.5‰ for  $\delta^{34}$ S and  $\delta^{18}$ O, respectively. Dissolved sulphate in modern seawater has a  $\delta^{34}$ S value close to +21‰ (Böttcher et al., 2007), whereas  $\delta^{18}$ O shows a broader range from +8‰ to +11‰ with an average value of +9.45‰ (Longinelli, 1989). Sulphate minerals (gypsum) precipitated in the Mediterranean coastal saline ponds of Isla Cristina (Huelva, Spain) give isotope compositions of +21.1‰ ( $\delta^{34}$ S) and +12.4‰ ( $\delta^{18}$ O). Consequently, there is no coincidence between the sulphate isotope compositions of the Mazarrón saline deposit and modern marine salts.

The absence of vertically oriented chevron halite points to a moderately deep brine body (several metres deep). The bromine content and the fluid inclusions in the salt deposit indicate that it was marine in origin; this finding suggests that the Mazarrón saline deposit was formed during the early stages of halite precipitation from evaporated seawater. Coccolithophores are almost exclusively marine, proliferating in the surface of the euphotic zone in coastal environments. However, sulphate isotope compositions ( $\delta^{34>}$ S and  $\delta^{18}$ O) did not coincide with modern seawater.



**Figure 5.** (1–4) Coccolithus pelagicus subsp. Pelagicus, (5–6) Gephyrocapsa oceanica, (7) small Gephyrocapsa (<3 µm) and (8) Umbilicosphaera sibogae. Petrographic microscope Nikon eclipse 80i 1000× coupled to camera Nikon DS-Fi1. Scale bar 50 µm.

Isotopic fractionation related to bacterial sulphate reduction (supported by the presence of framboidal pyrite) would increase both the sulphur and oxygen isotope signals in brines. In contrast, the isotope composition of the Mazarrón salt beds are depleted in  $\delta^{34}$ S but enriched in  $\delta^{18}$ O compared with the concentration expected for evaporated seawater. Accordingly, there must be an additional source of dissolved sulphate. The most plausible explanation is an input of dissolved sulphate from the recycling of Triassic (Keuper) and Neogene (Miocene) gypsum deposits, which are abundant in the hinterland.

The lagoonal sediments are characterized by the presence of calcareous coccolithophorid phytoplankton and diatoms. Coccoliths are scarce in the log from MAZ-17, being concentrated in the 6-10 m depth interval, which indicates conditions of a marine influence. The well-preserved coccolithophorids are characterized by Gephyrocapsa (<3 µm), which make up more than 50% of the association, typically indicating an environment of cold water, rich in nutrients (Flores et al., 2000). Moreover, there is a significant increase in Coccolithus pelagicus subsp.Pelagicus (Figure 5; Narciso et al., 2006) at 8.3 m. The abundance of this organism triples (from 7.4% to 29.5%) between the base of the first halite layer and its peak abundance at the base of the second halite layer at about 8 m in the core. The preservation of diatoms is moderate to low, while other siliceous microfossils (spicules of sponges and phytoliths; Figure 6) are well represented throughout the marine sequence analysed. Nevertheless, the presence of Thalassionema nitzschioides indicates nutrient-rich cold waters (Bárcena et al.,

2001) at the base of each halite layer. Above the second halite layer, in the sample corresponding to 6.10 m, the association of diatoms is more diverse and dominated by *Thalassiothrix*, indicating changing conditions in a stratified water column. In turn, the presence of benthic species like *Hanzschia* and *Cocconeis* indicates a shallow water column.

## Climatic and vegetational dynamics

Palynological data from core MAZ-15 (Figure 7) in the sedimentary interval correlated with halite beds show an increase in xerophytic vegetation and a decrease in forest cover altogether with a decline in mesophytic and thermophytic species. It is worth mentioning a drop in the abundance of fungal spores (Figure 8), which could be connected with a decline in plant biomass, since most fungi might be saprophytic. In addition, the fungal reduction in these strata can be related to a decrease in rainfall implying less fluvial sediment input to the basin. Microcharcoal particles, indicative of regional fire events, exhibit a pattern of lower fire occurrence than previous and subsequent periods, suggesting less biomass available for burning.

Between 3.32 and 3.28 m, the pollen diagram (Figure 8) shows a short gap (no pollen) that could be related with the abovementioned aridity event. Salt crystallization is a well-known cause of mechanical damage on palynomorphs (Carrión et al., 2009). Interestingly, this palynological hiatus is preceded by an oxidized bed. Increased aridity could have also provoked lake-level



Figure 6. Sponge bristles: monoaxon (left) and triaxon (right). Petrographic microscope Nikon eclipse 80i. Magnification 400×.



Figure 7. Correlations between the boreholes MAZ-15 and MAZ-17.

oscillations with episodes of temporal desiccation causing exile deterioration (Campbell and Campbell, 1994; Holloway, 1989) and biased or null preservation (Carrión et al., 2009; Davis, 1990; Navarro et al., 2002). The continuous curve of *Pseudoschizaea* associated with decreased total pollen concentration seems to support the existence of erosive contexts (Pantaleón-Cano et al., 1996, 2003) in concert with lithostratigraphical changes from peaty to terrigenous sediments (Carrión et al., 2009; Sancho et al., 2011).

Overall, the palynological record in the borehole MAZ-15 exhibits a change towards arid conditions that occurred about 4500 cal. yr BP, as observed in a number of pollen sequences from south-eastern Spain such as Antas, Roquetas de Mar and San Rafael (Pantaleón-Cano et al., 2003), Cabo de Gata (Jalut et al., 2000), El Cautivo (Nogueras et al., 2000), Cañada de la Cruz (Carrión et al., 2001b), Siles (Carrión, 2002), Villaverde (Carrión et al., 2001a), Gádor (Carrión et al., 2003), El Sabinar (Carrión e



Figure 8. Synthetic palynological diagram, microcharcoal concentration and lithostratigraphy of borehole MAZ-15. 'Thermophytes' include Chamaerops, Phillyrea, Myrtus, Maytenus, Pistacia, Cistaceae, Ephedra fragilis, Ziziphus, Withania, Periploca, Osyris, Calicotome, Lycium and Nerium. 'Mesophytes' curve includes Alnus, Fraxinus, Acer, Corylus, Betula, Ulmus, Salix, Juglans and Sambucus. 'Hydro- and hygrophytes' include Ranunculaceae, Epilobium, Cyperaceae, Typha, Apium, Nuphar and Myriophyllum.'Xerophytes' curve includes Ephedra fragilis, Artemisia, Lamiaceae, Asphodelus, Lygeum, Thymelaeaceae, Tamarix, Ziziphus, Periploca and Lycium. 'Fungi' include Inapertisporites, Monoporisporites, Hypoxylonites, Spirotremesporites, Diporisporites, Foveodiporites, Biporipsilonites, Dicellaesporites, Diporisporites, Brachysporisporites, Polyadosporites, Dicellaesporites, Diporisporites, Polyadosporites, Polyporisporisporites, Chaetomium, Gelasinospora, Sordariaceae, Staphlosporonites, Lacrimasporonites and Chaetosphaerites. 'Thorny shrubs' include Genisteae, Rhamnus, Berberis, Periploca, Ziziphus, Withania and Calicotome.

2004), Carril de Caldereros (Fuentes et al., 2005) or Baza (Carrión et al., 2007). In all of these sequences, progressive xerophytization is noticed from c. 5000 to 4500 cal. yr BP onwards. This new pollen record supports the notion that opening of the landscape and local extinction of broad-leaf trees accounted before in the thermomediterranean belt, and coastal territories (Jalut et al., 2000; Pantaleón-Cano et al., 2003), moving thereafter to higher elevation and continental locations (Carrión et al., 2001a, 2001b, 2003, 2007; 2010b). Regional anthracological data are strongly supportive of this picture with available information for full (4630 and 4350 cal. yr BP) and late Chalcolithic in Los Millares (Andarax Basin) and Vera Basin (4640 and 4350 cal. yr BP). Charcoal analyses show predominance of sclerophyllous matorrals with lentisks and wild olives, together with Ephedra and Lycium intricatum. A clear trend to riparian forest reduction is also observed (Rodríguez-Ariza, 2000; Rodríguez-Ariza and Vernet, 1991).

# Archaeological indicators in the time window of the arid event

The known archaeological indicators in the study area are in line with the neotectonic, climatic and palaeobotanic changes outlined above. Dating of the prehistoric settlements in the vicinity of the marine lagoons wherein the halite deposits formed indicates that a group of people lived on the Cabezo del Plomo site, towards the end of the Neolithic/Chalcolithic Period. The settlement's location strategy would have been connected to the habitat and ecological diversity of the vicinity, a key factor for the development of the production pattern. Further stability would have been derived from its proximity to the alluvial terraces of the old mouth of the Rambla de las Moreras and to the shellfish and other resources of the sandy coast (which was much closer at that time). In addition, the presence of brackish lagoons would have formed a backdrop for hunting and livestock activities. The post-Flandrian regression exposed the coastal plain in many places ('emergent coastline') and on a low coastal promontory (now known as La Punta de los Gavilanes). This new settlement appeared more than a millennium later, part of the Bronze Age cultural sphere, which was engaged in the exploitation of marine resources in the immediate vicinity.

The oldest known settlement in Cabezo del Plomo dates back to  $5170 \pm 90$  BP (SUA-1474), while the most recent (SUA-1476) occurred at  $4930 \pm 120$  BP (Muñoz Amilibia, 1993). Calibration of both of these dates using shell debris (Calib 14C REV6.0.0., Marine09.14c; Reimer et al., 2009; Stuiver and Reimer, 1993) taking a correction factor for the marine reservoir effect of  $\Delta R =$  $64 \pm 16$ ) places the older settlements in the interval 5270–5645 cal. yr BP and the most recent one between 4845 and 5451 cal. yr BP. The first occupation on Punta de los Gavilanes was dated to  $3730 \pm 30$  BP (KIA-32355), and calibration places this in the interval 3981–4154 cal. yr BP (Reimer et al., 2009; Stuiver and Reimer, 1993; Stuiver et al., 2010).

The two settlements therefore indicate occupancy before and after the deposition of the halite beds; this fact, together with the presence of productive natural resources and the climate and vegetation indicators, that we have derived in studying this event allows us to provide the following observations:

 The chronology of the settlements, with respect to the episodes described in the lagoon environment of Puerto de Mazarrón, sets a new archaeological indicator of the climate changes that occurred during the Holocene. These environmental changes are detected from about 5500 cal. yr BP on as indicators of greater aridity (Cacho et al., 2010; Fletcher and Sánchez Goñi, 2008; Frigola et al., 2007, 2008; Jalut et al., 2009; Martínez-Cortizas et al., 2009). Climate change intensified *c*. 4500 BP (Martín-Puertas et al., 2008), as reflected in the increase of xeric markers at the -8.30 m level in borehole MAZ-17.

- 2. The middle date of the time interval (at  $2\sigma$  confidence interval) calculated for the more recent settlement at Cabezo del Plomo is close to 5147 cal. BP. The mid-date for the formation of the halite beds (calculated from borehole MAZ-17, at -9.20 m depth) are *c*. 4906 cal. yr BP, giving a difference of 200 years between the two events. We can also state that the probability interval of the first datation (5451–4845 cal. yr BP) correlates to the probability range of the second (4973–4839 cal. yr BP).
- 3. Before this last event, the diverse ecological setting had created a unique niche consisting of stable alluvial plots that were easily cultivated alongside recurrent brackish lagoons that would have been easily cultivated. The settlement would be in proximity of a coastal shelf and dunes and with Mediterranean forest on the surrounding mountain slopes. It seems reasonable to consider, at least for the littoral setting of the Bay of Mazarrón at the beginning of the 5th millennium cal. BP, that the effects of these climate change would have significantly modified the formed biodiversity, replacing by one less predictable and productive because of the dramatic salinity increase in soils. Such changes would have led to relocation of the settlements to other areas, nearby or further away, less exposed to human pressure and best tolerant to the ever-increasing hydric and saline stress caused by the climate change in environmental conditions.
- Tectonic reactivation before and during this event, not only in the study area but also in the Guadalentín Valley between Lorca and Librilla (Calmel-Ávila et al., 2005), would have brought further risks such as watercourse deviations, and the modification and segmentation of the alluvial fans previously deposited by tributaries on the left bank of the Guadalentín River, coastline changes and marine intrusion into newly subsident zones as the western lagoon at Puerto de Mazarrón became even more compartmentalized. In fact, during this period, settlements were abandoned and relocated next to the old watercourses of the Guadalentín Valley. In Puerto de Mazarrón, mediumgrade earthquakes would have accompanied reiterative movements of the Mazarrón Fault, which was responsible for an abnormally high sedimentation rate of 23 mm/yr in the southern sector of the eastern lagoon.
- 5. The subsequent trend towards a wetter climate, indicated from borehole logs in the former Mazarrón salt flats, in particular MAZ-17 (-7.25 m depth; Poz-26544: 4245–4524 cal. yr BP), together with other archaeological indicators in the interior of the Guadalentín valley and elsewhere in south-eastern Iberia (Castro Martínez et al., 1996), supports a human occupation of the sandbank. At that point, the sandbank had almost formed the new coastline, except at the eastern end. Therefore, at around 3730  $\pm$  30 BP/3981–4154 cal. yr BP, a small group established itself on the coastal promontory of Punta de los Gavilanes. Over a long period of the second millennium BC, this group exploited the best opportunities for diversification (Rodríguez-Estrella et al., 2011).

## Discussion

Sedimentary, anthracological and pollen records and geochemical information (accompanied by an increasing amount of micropalaeontological information in aquatic environments – both continental and marine) allow us to contextualize the arid event in MAZ-17 (from 9.20 to 7.25 m depth) at various spatial scales. The aridity event evidenced in MAZ-17 was probably a regional response to climate changes that, in recent years, are being identified on a global scale, therefore changing the perception of the Holocene as a climatically stable period during the Quaternary.

The subsections of this discussion, below, consider this correlation with other contemporary episodes and with the same or similar climate and vegetation characteristics, for micro-spatial and regional settings, as well as in the context of the Iberian Peninsula, the Mediterranean region and worldwide.

### Regional context

There are several sedimentological and palynological records that confirm and contextualize the arid event identified from the sedimentary profile of MAZ-17. Silva et al. (2008) studied the alluvial fan at Lorca, situated in the Guadalentín depression and demonstrated how sediment aggradation was interrupted c. 5400 cal. yr BP. This date is associated with the first Neolithic occupation at the apex of the alluvial fan. Occupation persisted well into the Calcolithic Period, c. 4400-4200 cal. yr BP. At that time, there was a transition from the stable phase to one when the incision of the alluvial fan apex began. The stable phase implies a significant lack of rainfall, and it coincides climatically with a period of greater aridity. Not only does the arid event detected on the Mazarrón coast support this finding in Lorca. Radiocarbon analyses point that the time span of this event can be precisely sequenced and, taking into account its proximity, might be extrapolated to the Guadalentín basin.

The phase of down-cutting in the Lorca alluvial fan coincided with climate instability elsewhere: in the mid-valley of the Guadalentín River, Calmel-Ávila (2000) highlighted episodes of climatic irregularity in the Holocene glacis sequence of this tectonic depression. One instance of this was found at the confluence of the Librilla and Algeciras watercourses and the Romeral reservoir. Here, between  $4305 \pm 55$  BP and  $3888 \pm 60$  BP (4862-4329 cal. yr BP), fluvio-torrential facies, some 7–8 m thick, overlie a lower energy fluvio-lacustrine sequence. On the raised plains of Gebas and Fuente-Librilla, similar facies were described at the same time interval (Calmel-Ávila, 2000). The position of this fluvio-torrential detritic facies enveloped by fine sediments can be associated with an arid phase of irregular but torrential precipitation, which is compatible with the arid process detected at Mazarrón.

In the palynological context, as well, the data from the Bay of Mazarrón and the prelittoral Guadalentín depression are remarkably similar. Likewise, in the coastal lagoon of San Rafael (Almería province, south-eastern Spain), an arid period has also been determined between 5000 and 4500 cal. yr BP (Pantaleón-Cano et al., 2003). Geochemical analyses on carbonized seeds from archaeological sites obtained similar results, concluding that, regionally, the period 7000–5000 BP was wetter than later within the Holocene (Araus et al., 1997; Burjachs et al., 2000).

#### Iberian peninsular context

According to the humid–arid transition manifested between 7 and 5 kyr cal. BP (Cacho et al., 2010), the arid event identified in Mazarrón means that the aridity should have intensified towards the end of this period, lasting until at least 4400 cal. yr BP.

In the Sierra Nevada (Betic Cordillera, south-eastern Spain), eight periods of slope instability have been identified from the middle Holocene (Oliva et al., 2010), in the lakes of Aguas Verdes, Río Seco and Lagunilla de Río Seco. These processes were evidenced by an increase in the clastic material towards the lakes, corresponding to dry and cold climatic rhexistasic phases. The Sierra Nevada event (dated to between 5.2 and 4.6 kyr cal. BP) is concordant with the arid event in our study area.

In the north-east of the Iberian Peninsula, an accentuation of the aridity was also identified by means of a study of trace element analyses in peaty sequences (Fábregas Valcarce et al., 2002). In the Huerva River (in the Ebro Basin), seven sections were differentiated in the slope of Peña Enrique. The basal section corresponds to a badlands relief from the Calcolithic period, dated to 2540 cal. yr BP, which has been associated with semiarid or arid environmental conditions (Peña Monné et al., 2011). Similarly, Fabián et al. (2006) noted a population hiatus in the Amblés valley (Ávila province) in the Sub-Boreal period, which was interpreted to be related to the existence of abrupt, short-lived climatic crises. One of these population hiatus is dated to 4000-3700 BP, associated with an event around 4.0 kyr BP (4500-4100 cal. yr BP), corresponding to an acute climatic crisis characterized by extreme aridity. This coincides neatly with the one detected in Mazarrón.

## Western Mediterranean context

Analysis of pollen and sediment records for various places in Western Europe and the Mediterranean Basin (Burjachs et al., 1997; Carrión et al., 2007; Juliá et al., 2001) manifested conditions compatible with the Mazarrón episode. Likewise, studies of marine-lacustrine fluctuations (Magny, 1993, 2004; Menotti, 1999) also imply that a significant regression in sea level occurred. Geological, geomorphological and stable isotope analyses from various continental logs from central southern Italy (Giraudi et al., 2011) denote a general reorganization of climate and of the environment between 5000 and 4000 cal. yr BP.

On the Franco-Spanish Mediterranean coast, Jalut et al. (2009) dated the arid phase between 4500 and 4000 BP (5300-4200 cal. yr BP). This is consonant with data from other points in the Western Mediterranean, like the Iberian Peninsula (Caroli and Caldara, 2007), the Balearic Islands (Yll et al., 1997) and Corsica (Reille and Pons, 1992). This phase is contemporary with the event recorded in MAZ-17 and correlates with the changes detected in the vegetation cover through the Western Mediterranean. The findings of Jalut et al. (2000) are clearly confirmed by the increase in thermophilous and xerophytic taxa and the decline in temperate deciduous forest, associated with changes in the seasonal pattern of precipitation, which are related to one of the cold intervals recorded on a worldwide scale. Similarly, Fletcher et al. (2013) remarked on the significant reduction in Mediterranean forest over the interval 5400-4500 cal. yr BP based on the MD95-2043 marine sediment core from the Alboran Sea.

Pollen records from marine cores in the Alboran Sea (MD95-2043) and from north of Menorca (MD99-2343) are consistent with the arid episode identified in our study area. They also tally with those highlighted in the terrestrial sequences of the Betic mountain ranges and in the lowlands of Almeria province (Cacho et al., 1999; Carrión et al., 2007, 2010a; Frigola et al., 2007; Jiménez-Moreno and Anderson, 2012; Pantaleón-Cano et al., 2003; Pons and Reille, 1988). Sea-level periodical variations, related to climatic changes, have been reported during the Holocene based on littoral progradational beach studies in the Granada and Almería provinces (Fernández Salas et al., 2009).

Together with other records from the Moroccan Rif and Atlas mountains (Lamb and Van der Kaars, 1995), these data point to a cooling trend in the climate from *c*. 5400 cal. yr BP, accompanied by greater aridity in low-lying areas (Fletcher and Sánchez Goñi, 2008). On the Moroccan coast, an abrupt, short-lived arid phase is inferred from the lowering of the water level in Lake Tigalmamine between 4500 and 4050 BP (Hassan, 1997).

Mercury et al. (2011) link the trend of increasing aridity in various environments in the Mediterranean and North Africa with the increase in anthropogenic influences on the corresponding environments. Nevertheless, the cold, dry episode registered in MAZ-17 seems to indicate the fundamental role played by climate changes in the mid-Holocene, rather than simply answer to anthropogenic factors.

Other authors have related the consequences of this period of climate change with significant cultural changes in various areas of the Mediterranean Basin and as far as Asia (De Menocal et al., 2000; Gibbons, 1993; Peiser, 1998; Weiss et al., 1993). However, these considerations have to be viewed within singular parameters of latitude and longitude, in consonance with the socio-economic, technological and political differentials of each cultural region over that period.

## Global context

This event is possibly one of the most pronounced of the Holocene, and its expressions have been recorded in various regions across the world. Its origin is still uncertain, although one suggestion is that it relates either to changes in solar activity, as measured by teleconnections in a complex cryosphere–ocean–atmosphere system (Magny, 2004). Other suggestions are that it arose due to variations in the Earth's magnetic axis, or the collision of a meteorite in the Middle East, which would have caused forest fires and a great arid episode that would have affected even the Iberian Peninsula (Courty, 1998; Weiss et al., 1993). Certainly, there is climatic concordance between the event studied in this paper and the 4.2 kyr BP event identified by Bond et al. (1997, 2001) when the climate was very dry and cold and the water in the Atlantic cooled at  $1-2^{\circ}$ C.

The onset of the dry, cold event in Mazarrón (4839–4973 cal. yr BP) could be associated with 1 of the 15 phases of water-level rise in Alpine lakes during the mid-Holocene (Magny, 2004). Specifically, it coincides with episode 8 (4850–4800 cal. yr BP), which was testified by an acute enlargement of Lake Leman, evidenced by a terrace that lies more than 3 m above the current water level and dated between 5029 and 4832 cal. yr BP (Gallay and Kaenel, 1981).

In the North African case, the abrupt increase in eolic terrigenous deposits until 5.5 cal. kyr BP was related to the end of the African Humid Period and the rapid change to greater aridity, which persisted to date (De Menocal et al., 2000). Related to this evidence, the marine sediment core MD03 2705 drilled in the Atlantic coast of North Africa confirms the greater input of eolic terrigenous material into the ocean, as well as an increase in the species of warm coccolithophorid associations and a rapid fall in the cold species, such as *Gephyrocapsa muellerae*. This circumstance, supported by the record of freshwater phytolytes and diatoms, is related to end of humid conditions and the sudden warming of water masses as a result of greater aridity in the region (Mejía-Molina et al., 2006).

On the Asian continent, a phase of climate degradation, associated with short cold events followed by dramatic aridification, has been deduced between 4500 and 4200 BP, based on the GT40 sediment core taken in the Hexi Corridor in the dry lands of NW China (Yu et al., 2006).

## Conclusion

Here, we describe a lithological signal for an arid crisis in southeastern Spain between c. 4550 and 4400 cal. yr BP, evidenced by the precipitation of a halite bed in the eastern lagoon at Puerto de Mazarrón. The salt deposit is a mixture of halite and gypsum, with a relatively high amount of celestine. The presence of nanoplankton, together with the composition of the fluid inclusions, indicates that the salt is basically marine in origin. The presence of framboid pyrite and bituminous material suggests reducing conditions during the burial diagenesis (not in the sedimentary environment). Bromine content and the isotope composition of the sulphates (mainly gypsum) indicate an additional source of continental water, rich in  $CaSO_4$ , derived probably from the dissolution of nearby deposits of Neogene evaporites. This input did not modify the elemental composition of the halite brines, since they were already saturated in gypsum, but simply caused a greater amount to be precipitated and affected its isotope signature. The fact that the evaporites are intercalated between sapropels indicates that, prior to and after the arid phase, there were two separate episodes of more humid conditions and/or groundwater inflows.

In the study area, the middle Holocene is characterized by an unstable climate, alternating between big floods and very cold recurrent droughts. A period of drought between 7 and 5 cal. kyr BP affected the southern part of the Iberian Peninsula. Towards the end of this period, the drought episode intensified in the Mazarrón. This phase lasted until at least 4400 cal. BP. The log from borehole MAZ-17 in Mazarrón corresponds to this more acute arid phase (4436–4645 cal. yr BP) and peaks at *c*. 4550 cal. yr BP. The peak extends until *c*. 4400 cal. yr BP (with distribution range: 4245–4524 cal. yr BP) in the vicinity of the Bay of Mazarrón.

The formation of this halite deposit in Mazarrón occurred during a period of significant neotectonic reactivation that seems to coincide with the one identified in the Guadalentín Valley, which lies quite nearby. This fact needs to be borne in mind in the future, for two reasons. First, it suggests a potential relationship with global climate forces through both the Pleistocene and the Holocene. Second, there is also a potential relationship with the restructuring of the human population detected over the second half of the 3rd millennium BC in the Mediterranean and the Iberian Peninsula, especially in south-eastern Spain. The confirmation that the population restructuring in this area of south-eastern Iberia and in the region as a whole, during that time interval coincident with drought conditions and forest regression, reinforces the idea that changes in the Holocene ecosystem, caused by climatic and edaphic changes, had an impact on a human level. Settlements were faced with the need to reorganize social and productive aspects of previously inhabited land. In some cases, previously stable resource niches became unpredictable for some time and had to be abandoned. In short, environmental change provided new opportunities for cultural change.

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## Note

 All the radiometric calibrated data contained in this paper reflect the median of 2σ (95.4%) probability intervals. Dating BP and their corresponding standard deviations are reflected in Table 1.

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