Dear Editor and Referees,

Firstly, we would like to thank you the time devoted in reviewing this manuscript and your comments, which we found appropriate and relevant.

As a general comment, syntax, spelling and grammar were checked by an English native professional translator, although these changes are not marked in the marked-up manuscript version to avoid hampering the identification of the changes related to the Referees' comments.

The main changes undergone by the text in relation to each of the Referees' comments are explained below and marked in text. Please note that brief specific comments not mentioned here have been accepted and changes have been made accordingly to the Referees' suggestions.

Referee #1

Material and Methods chapter, section 2.3 (lines 98-120): I miss a justification for the splitting of E. huxleyi and G. oceanica into two size classes. Did the author implemented this splitting because of differences in size classes between fossil and modern taxa (ie. no large E. huxleyi and/or G. oceanica in the modern dataset)? Other reason(s)?

-The size criteria to split *E. huxleyi* responds to the well-known existence of two sized morphotypes (< 4 µm and > 4 µm) with different ecological and biostratigraphic significance already demonstrated in the study area (Colmenero-Hidalgo et al., 2002; Colmenero-Hidalgo et al., 2004). *G. oceanica* size distinction follows a classical size criteria (medium: > 3 < 5 µm; large: > 5 µm). Despite there is no evidence to our knowledge that this separation has a demonstrated ecological significance, we usually carry out it due to a potential use in Ecology. A short explanation for this has been added to the text. Nevertheless, it has to be noted that abundance of *G. oceanica* > 5 µm is very low, 0.24 % on average and 2 % as maximum.

Throughout the manuscript: the term "range of SSS values" is often used, but might refer to either the amplitude of SSS changes within a particular event, or to the complete range of measured (modern data set) or reconstructed (downcore CEUTA10PC) SSSs. The authors should better define what they mean by "range off SSS values". The same question applies to the frequent and sometimes incorrect use by the authors of "SSS gradients".

-We concur with the Referee that the terms "range of SSS values" and "SSS gradients" might be confusing. We have clarified those terms where necessary.

Section 4.1, lines 358-360: The authors recall a finding by Knappertsbuch (1993) of a positive relationship between G. oceanica abundance (in modern samples) and Atlantic-derived waters (and thus SSSs) in the western Mediterranean Sea. The authors should comment this finding later-on in section 4.3 and 4.4 based on the downcore distribution of G. oceanica within core CEUTAP10PC (Ausin et al; Paleo3, 2015). A visual comparison of the downcore SSS patterns and G. oceanica abundance changes within this core indeed shows a high degree of similarity between these two datasets. Such a comparison would therefore work in favor of both reliable SSS reconstructions, as well as the main influence of Atlantic water inflow upon SSS in the Alboran Sea.

-We appreciate the Referee's comment on the finding by Knappertsbusch (1993). However, we find this good visual comparison is a logical consequence of the "use" of this (but not only) relationship between *G. oceanica* and low SSS by the transfer function to reconstruct SSS, causing a circular argument.

Section 4.3, lines 383-391. The reference to Bollmann and Herrle (2007) is not accurate. Bollmann and Herrle' modern micropaleontological dataset does not include any coccolith of E. huxleyi larger than 4 microns. Only their LGM dataset contain coccoliths close to but < 4 microns for higher latitude sites (north of 35_N). Rather than discussing Bollmann and Herrle (2007), the authors should relate the non-analog situation (linked with large coccoliths of E.

huxleyi) to input of cold polar-subpolar derived Atlantic waters, based on the modern distribution of extant population of E. huxleyi type B (coccoliths larger than 4um) in subpolar environment such as in the southern Ocean (e.g. Poulton et al, Mar. Ecol. Progr. Ser., 2011).

-We agree with the Referee reference to Bollmann and Herrle (2007) is not entirely correct. Certainly their modern dataset does not include any specimen of *E. huxleyi* > 4 µm, so this has been corrected in the text. However, we still believe their findings are worth mentioning and discussing briefly. Their LGM dataset does include *E. huxleyi* > 4 µm (see Fig. 6 and section 4.2 in that study), where specimens of *E. huxleyi* of 4-6 µm length in samples located in the Canary Island region led to an overestimation of the salinity values compared to values published by other authors (Table 2 in that study). Their results (and also interpretation) suggest that the large morphotype in ancient sediments represents an extinct morphotype. To be the case, *E. huxleyi* > 4 µm would lack an analog in modern sediments, as shown by the results of the present paper and therefore supporting our interpretation.

Section 4.4.1 and 4.4.2: the authors often compare paleo SSS changes with pollen based changes in aridity/precipitation, hereby suggesting that precipitation changes might explained to a high extent the surface salinity changes (ie. line 414: "This change is not supported by the findings of Fletcher....."). This connection between paleo SSSs and paleo-precipitation is definitively ruled out by the authors in section 4.4.3 (Younger Dryas and Holocene) based on modern observations (incoming AW drive SSS in the Alboran Sea) and a comparison off SSSs reconstructions with Uk37- derived paleo SSTs. The comparison of reconstructed SSSs with paleo-precipitation records over the nearby continent should therefore only recall previous works such as Fletcher et al (Clim. Past 6, 2010) who illustrated and thoroughly discussed the phasing of high latitude cooling (cool inflowing Atlantic water through Gibraltar strait) with dryness of the western Mediterranean climate. A thorough investigation of the ocean-atmosphere coupling in the western Mediterranean region throughout the last deglaciation and Holocene is out of scope of B. Ausin's manuscript.Section 4.4: Keeping-on the discussion on comparing paleo-precipitation records with paleo SSS reconstructions (which, according to my previous comment should be drastically reduced) : Although very similar to the pollen-derived record of Combourieu-Nebout et al. (2009), the dataset by Fletcher et al. (2010) was produced from the same sediment core as the one from which the illustrated SST record is derived, core MD952043 (Cacho et al., 2001). Also, beside TMF (%) variability, Fletcher et al. (2010) provided additional indexes of interest such as quantitative paleo-precipitation reconstructions and/or pollen-based precipitation index (Ip). The authors might therefore consider illustrating Fletcher et al. (2010) record (rather than the ODP site 976 record), although mentioning that both MD952043 and ODP 976 pollen-records are highly coherent.

-The discussion about atmospheric conditions in the study area has been ruled out as suggested by Referee#2. Accordingly, any pollen record has been removed from Fig. 6.

Section 4.4.3: a lot of emphasis is put on brief periods of low SSSs. No discussion is made on the exceptionally high values of SSS (higher than during late glacial and YD) during the short time interval of ca. 10-10.5 cal. ka BP. This event should definitively be discussed by the authors.

-A brief discussion for the SSS increase that occurred from 10.5 to 10 ka cal. BP has been added to the text.

Referee #2

As a general comment, we tried to emphasize our results with those by Oviedo et al. (2015) and to interpret them in more relation to the oceanographic context.

Page 3763, Line 21: are samples core tops, like stated here and throughout the manuscript, or surface sediments (caption Fig. 1)?

-*Stricto sensu,* these are "core-top samples", later referred to as "surface sediment samples" to avoid repetition of a technical term. In any case, we have changed caption in Fig. 1.

Section 2.3 Micropaleontological analyses: Gephyrocapsa caribbeanica is not included in living coccolithophore species (Young et al., 2003; Jordan et al., 2004). It is an important component of Atlantic Ocean surface sediment assemblages and further analysis/explanation should be nice. For instance electron microscope analysis may provide further taxonomic details and may rule out that they are not specimens of G. oceanica or G. muellerae. Calciosolenia murrayi is in my opinion not discernible by Calciosolenia brasiliensis and Calciosolenia corsellii. Much better Calciosolenia spp. More information is needed for the eliminated 29 samples (page 3765, lines 19-26). Why did you choose the 10% reworked treshold. As later discussed (page 3770, lines 18-21) also coccoliths with a compatible age (still living and long-range taxa) may be displaced or reworked. Why did you not simply rule out reworked specimens from counts and include all samples?

-The species we found in our samples is a small specimen (< 3 μm) that shares many morphological features under the optic microscope with *G. caribbeanica* Boudreaux & Hay, 1967. We have already observed this small *G. caribbeanica* in Holocene sediments from many locations and previous investigations conducted by our group at Salamanca University using SEM pointed it could be an overcalcified small *Gephyrocapsa*. Nevertheless, we prefer not to lump together these latter species due to a possible ecological significance. Certainly the name "*G. caribbeanica*" for this small specimen is not appropriate and it has been changed by *Gephyrocapsa* cf. *caribbeanica* in the text.

-Most of micropaleontological studies, even those aimed at stratigraphic purposes, have to deal with the possible presence of reworked specimens within the "long-range" taxa and are based on the assumption that such presence, which cannot be quantified, is not significant. As reworked, we could only identify and quantify the *obviously* reworked specimens (in our case, those taxa pertaining to older stratigraphic levels with no representative in modern oceans and then lacking a relationship with modern surface water conditions). We counted 500 coccoliths per sample. Bellow 450 coccoliths, the statistical representativeness of the sample for studying the main species starts to decline (Fatela and Taborda, 2002), and this further affects minority species. This means that if we removed reworked specimens in those samples where their relative abundance is > 10 % (sometimes up to 80 %), the rest of the sample would not be statistically representative of the "in situ assemblage". Therefore, samples with obviously reworked specimens above 10 %, even considering that these specimens do not necessarily imply that the rest of the association is also allochthonous, were ruled out as a precautionary measure to not compromise the representativeness of the sample. We later explored the spatial distribution of reworked specimens in the remaining samples and observed: i) that samples far from river influence contained very low percentages of reworked specimens (usually < 2 %), and ii) that those samples with the highest % of reworked (but still below 10%) were located close to river mouths. From this we interpreted that higher % of reworked (but below 10%) were linked to terrigenous inputs associated to river discharges and therefore it could be assumed that the rest of the assemblage is autochthonous. For this reason, samples with a 10% of reworked specimens were retained in the modern training set, although of course, the reworked are excluded from the analyses.

Page 3767, Lines 18-26: I did not understand. Please carefully explain this passage. Photic zone down to 300 metres depth?

-This analysis was developed by Telford (2013) for planktonic foraminifera. Some species of these organisms present a clear zonation along the upper part of the water column and bloom in a particular season. Therefore, different fossil species preserved in the same sediment layer, might have lived under the influence of different conditions (different SST, SSS, nutrients, etc). However, calibrations in transfer functions are usually performed in a mechanical way using annual-averaged data of the environmental parameters at 10 m depth, therefore assuming that properties at that depth (10 m) and "season" (annual) were the most influential for the assemblage. This should be tested before, since it is possible that, let's say, summer-SST at 70 m influenced the assemblage more than annual-SST at 10 m. For this, several reconstructions are performed, each of them using for calibration data of the water column from a different depth and season. The reconstruction that more variance explains in the fossil assemblage will tell us the depth and season at which the transfer function has to be calibrated. The implementation of this analysis makes more sense when using foraminifera since the majority of coccolithophores inhabit in the upper photic zone, although still some species have a clear preference for deeper parts (e.g. *F. profunda*) or bloom in a particular season. Nevertheless, one of the aims we pursued with this work was to apply a wide array of new statistical approaches on our data and to show them in a logical series of steps that one should consider when developing a transfer function. This was intended to target the wide public that works on transfer functions, not only based on coccolithophores but on any other nanno/microfossil. We have tried to explain better this analysis on the text. As pointed out by the Referee, the photic zone does not reach 300 m (we took the same depths considered in Telford et al., 2013), and coccolithophores do not inhabit there anyway, so we excluded this depth too.

Page 3768-3769, Lines 23-25 and 1-13: the description is quite poor and in any case the spanish-african coast comparison is possible just in a very narrow area. In my opinion, Figure 2 is quite self-explanatory and should be complemented by meso-scale oceanographic features (partially plotted in Fig. 1). Then the spatial distribution description of coccoliths on the sea floor will likely follow local hydrology.

-Meso-scale features have been plotted for the reader in Fig. 2 following the Referee's suggestion and *G.* cf. *caribbeanica* has been eliminated from this figure in line with previous comments. Despite this section has been rewritten placing more emphasis on the biogeographic pattern, description has been kept brief, since we agree this figure is self-explanatory and the likely relationship with the oceanographic context is discussed further in section 4.1). Regarding this section, it has also been rewritten following Referee's suggestions.

Page 3770, Lines 18-21: which long-range taxa? Percentage and abundance? Can you rule out them and identify the in situ assemblage (see comment Section 2.3).

-The yellowish color of this sample had already been noticed and written down when the counts were performed, but it was included in the initial modern training set because it only contains extant species. The fact that both methods (MAT and WA-PLS) pointed this sample as outlier along with its anomalous color made us suspect it had been subjected to diagenetic processes. Consequently, we preferred not to include it within the modern training set. It must be added that any anomaly/observation was written down when the counts were performed, and a revision of these notes let us discard that none other sample presented this color or similar characteristics.

Page 3771, Lines 5-6: it is not clear from Fig. 5, how much is the SSS error range? How was it calculated? It is important to assess the error given that current geochemical methodologies (temperature corrected d18O) are affected by a huge full propagation error that makes SSS reconstructions unreliable (Rohling, 2007).

-Sample-specific reconstruction errors are automatically derived for the fossil samples by the C2 software as follows: At each bootstrap cycle we obtain a slightly different value for our estimate. The standard error of these bootstrap estimates for each modern and fossil sample is calculated by C2, being this the prediction error due to errors in estimating species coefficients (i.e. the optima in WA). It is worth to note that we have added to this error a second error component that we had not considered before, and have plotted errors accordingly in Fig. 5a. This second component is a constant that represents errors in the calibration function, calculated by C2 as the root mean square across all training set samples of the difference between the observed environmental value and the mean bootstrap estimate. Due to an extensive explanation is required to clarify how these errors are calculated, we have indicated the reader where to find detailed information about it in section 2.4.3. The amplitude of the error has been added to the discussion whenever possible.

Page 3772, Lines 3-13: please re-write. Ecological preferences here speculated need to be formulated taking into account 1) previous reports and 2) a detailed hydrological setting (oceanography once again). So, in my opinion, it is wrong to suppose fresher water preference *(low sea surface salinity) for Florisphaera profunda, a deep photic zone taxon (> 50 m depth). Low salinity in surface water is likely linked to anything else that influences and controls species distribution and abundance. Also very striking is the presumed preference for rather saline waters of Helicosphaera. High abundance of Helicosphaera carteri, arguably the main species you found, was often associated to fresher waters in many studies (i.e. Colmenero-Hidalgo et al., 2004; Narciso et al., 2010; Grelaud et al., 2012). Even in Ausin et al. (2015, PALAEO3) Helicosphaera spp. peaks are interpreted 'as being linked to the low-salinity inflowing Atlantic water' (Section 5.1).*

-This section has been re-written following Referee's suggestions.

Page 3772, Lines 14-29 and Page 3773: both discussion and references deal with the influence of salinity on coccolith weight mass, species type (Emiliania huxleyi), coccolith calcification, alkenone production and so on. All them are profoundly different from the influence on coccolithophore assemblages.

-We agree with the Referee and are aware of the differences between the "species" and "assemblage" approaches. We refer to those works for two main reasons: i) we believe that the evidences that exist on the relationship between coccolithophores and salinity (even at a species level) support the salinity influence over the total assemblage, and ii) there is a lack of works dealing with the influence of environmental variables over the total coccolithophore assemblage using comparable methods. Nevertheless, it has to be mentioned that the work by Oviedo et al. (2015) explores this influence at a species level, but also at a hetero- and holo-coccolithophore assemblage levels, and their results are discussed in this regard. In any case, we have tried to clarify the difference between approaches in the text.

Page 3776, Lines 2-5: these estimates are however affected by a huge propagation error (see comment above).

-The propagation error that possibly affects other records and its implications for the comparison with our results have been included in the discussion.

Page 3777, Lines 1-20: Alpine meltwater may have also had a role in ORL1 lower salinity (Rohling et al., 2015).

-We thank Referee's valuable comment on Rohling et al. (2015), which has been included in the discussion. We have ruled out mentioning the work by Rogerson et al. (2008), which is already discussed and (the equations used therein) corrected by Rohling et al. (2015).

Page 3777, Lines 17-20: how the Intra-Allerød Cold Period (IACP) was identified? Apparently there is no basis for the identification of the IACP by oxygen isotopes in Ausin et al. (2015, PALAEO3) and in fact it is not mentioned there. If so, you cannot identify it by the SSS increase. There is also an inextricable confusion about (among others) the IACP in Figure 6 (see comment below).

-Indeed the IACP was not identified from our records, and consequently, has been ruled out.

Page 3777, Lines 21-26: the only plot with Younger Dryas (YD) is Figure 6 and I do not understand it. YD and GS-1 are synonyms, simply from different records (the latter from Greenland ice cores) but in Fig. 6 there is a clear mismatch. The Greenland nomenclature from the column is out of phase with the grey shadow of YD. The mismatch also involves the upper part of the Bølling-Allerød (B-A), the IACP should be in coincidence of GI-1b. There is also an evident problem with the timing of the base of B-A, YD and Holocene that should be respectively at 14.75 (or 14.65) ka, 13 ka and 11.7 (or 11.5) ka. Please check the plot. Without this basic information it is difficult to understand the salinity trend during the YD.

-We thank the Referee for their comment on Fig. 6. Certainly there was a huge mismatch with Greenland chronology, which has been corrected following Rasmussen et al., (2014). According to these authors, the "Oldest Dryas" term has been also excluded since it is poorly defined in literature and it has been changed by Termination 1b, already identified and interpreted in CEUTA10PC08 core by Ausín et al. (2015).

Page 3778, Lines 13-18: In my opinion the visual inspection of Fig. 6 does not establish a firm correlation between SSS decreases and Alboran cooling (AC) events. In any case why are AC associated with SSS drops? There was not any significant amount of icebergs close to the Iberian Margin like for Heinrich 1 and Heinrich 2 layers (i.e. Bard et al., 2000; de Abreu et al., 2003). Why AC2 does not match with a SST deecrease? Authors should better explain their reasoning providing a mechanism that led to SSS decreases. In my opinion the comparison with terrestrial records (pollen and stalagmites) is extremely difficult and should be significantly shortened or ruled out.

-In order to better explain the SSS drops and its possible relation with AC events in the Alboran Sea, we argue a mechanism based on the Bond events (Bond et al., 1997). We have also included an explanation for the absence of SSS drop at times of AC2. Comparison with terrestrial records has been ruled out.

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DEVELOPMENT OF COCCOLITHOPHORE-BASED TRANSFER FUNCTIONS IN THE WESTERN MEDITERRANEAN SEA: A SEA SURFACE SALINITY RECONSTRUCTION FOR THE LAST 15.5 KYR

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Abstract

A new dataset of 88 marine surface sediment samples and related oceanic environmental variables (temperature, salinity, chlorophyll-*a*, oxygen, etc.) was studied to quantify the relationship between assemblages of coccolithophore species and modern environmental conditions in the Western Mediterranean Sea and the Atlantic Ocean, west of the Strait of Gibraltar. Multivariate statistical analyses revealed that coccolithophore species were primarily related to ~~the~~ sea surface salinity (SSS~~) gradient,~~), explaining an independent and significant proportion of variance in the coccolithophore data. A quantitative coccolithophore-based transfer function to estimate SSS was developed using the Modern Analog Technique (MAT) and weighted-averaging partial-least square regression (WA-PLS). The bootstrapped regression coefficient (R^2_{boot}) was 0.85_{MAT} and $0.80_{WA\text{-}PLS}$, with root-mean square error of prediction (RMSEP) of 0.29_{MAT} and $0.30_{WA\text{-}PLS}$ (psu). The resulting transfer function was applied to fossil coccolithophore assemblages in the highly resolved (~65 yr) sediment core CEUTA10PC08 from the Alboran Sea (Western Mediterranean) in order to reconstruct SSS for the last 25 kyr. The reliability of the reconstruction was evaluated by assessing the degree of similarity between fossil and modern coccolithophore assemblage, and comparison of reconstruction with fossil ordination scores. Analogs were poor for the stadials associated with Heinrich Event 2 and 1 and part of the Last Glacial Maximum. Good analogs indicate more reliable reconstruction of the SSS for the last 15.5 kyr. During this period, several millennial and centennial SSS changes were observed and associated with sea-level oscillations and variations in the Atlantic Water entering the Alboran~~ Sea, sea-level oscillations, and arid or humid atmospheric conditions in the Western Mediterranean~~.

Key words: transfer function; coccolithophores; salinity; Western Mediterranean Sea; Atlantic Ocean; Last Glacial Maximum.

1. INTRODUCTION

Coccolithophores are one of the major components of marine phytoplankton. They are sensitive to changes in many environmental variables, such as nutrients, temperature and salinity and are widely used in qualitative paleoenvironmental studies (Baumann et al., 2005; Guerreiro et al., 2013; Guerreiro et al., 2014). These studies provide general insight into the response of coccolithophores to environmental variables, but quantitative studies (e.g. transfer functions) allow assessing these relationships in a more rigorous and clear manner. Transfer functions are based on the calibration of the modern relationship between organisms and environmental conditions, and this information is in turn used to reconstruct past environmental variables. Different statistical approaches based on coccolithophores have been proposed in order to generate quantitative paleoreconstructions of different ecological variables. Giraudeau and Rogers (1994) used factor analyses and multiple regressions to estimate chlorophyll-*a* from coccolithophore census counts in surface sediment samples in the Benguela upwelling area. Several authors (Beaufort et al., 2001, 1997; Incarbona et al., 2008) calibrated the relative abundance of the coccolithophore *Floriphaera profunda* in surface sediment samples with respect to primary productivity and reconstructed past variations of this parameter in the Indian and Pacific oceans and in the Central Mediterranean Sea. Saavedra-Pellitero et al. (2011, 2013) used linear regression methods to derive past SST estimates in the southeast Pacific Ocean from coccolithophore census counts and accumulation rates. Bollmann et al. (2009) and Bollmann and Herrle (2007) applied multiple linear regressions to morphometric measurements of the coccolithophore *Emiliania huxleyi* from globally distributed core-top and plankton samples to obtain modern and past sea surface salinity (SSS) estimates.

To date, no coccolithophore-based transfer function has been applied in the Western Mediterranean Sea, a semi-enclosed basin situated at mid-latitudes (Fig. 1a). In this region evaporation exceeds precipitation plus runoff, such that water budgets tend to be balanced by the advection of relatively less saline Atlantic Water (AW) through the Strait of Gibraltar (Bèthoux, 1979). The AW flows eastward while mixing with Mediterranean water to form the Modified Atlantic Water (MAW) at the surface (100-200 m) (Millot, 1999). This distinctive feature affects the spatial distribution of some environmental parameters such as SST and SSS, leading to the development of well-defined longitudinal gradients between the Atlantic Ocean and the Western Mediterranean in annual terms. In this confined basin, the estimation of changes in those environmental parameters is essential for determining Atlantic-Mediterranean water mass exchange through the Strait of Gibraltar in the past (Rohling and Bigg, 1998; Schmidt, 1998). This exchange depends on variations in the hydrological cycle, ice-volume effects, and Mediterranean circulation patterns, which have a thermohaline origin (MEDOCGROUP, 1970).

The aim of this study is to explore the potential of coccolithophores for the development of quantitative reconstructions in the Western Mediterranean Sea. We study the response of coccolithophore assemblages from surface sediment samples from Atlantic Ocean and Mediterranean Sea to environmental variables. The resulting calibration model (transfer function) for salinity was used to reconstruct SSS changes at high-resolution in the Alboran Sea (Fig. 1a) for the last 25 kyr. The reliability of the reconstruction was assessed by analysis of the similarity between fossil and modern coccolithophore assemblages, and fossil ordination scores. Finally, centennial and millennial SSS changes are described and discussed, and compared with regional records of SST and organic matter preservation~~ and continental aridity~~.

2. MATERIALS AND METHODS

2.1. Modern training set

2.1.1. Surface sediment samples

Initially, 117 core tops located around a horizontal transect along the Western Mediterranean Sea and near the Gulf of Cadiz in the Atlantic Ocean were selected. They had been retrieved at varying water depths ranging from 70 to 2620 m during several oceanographic surveys and were stored at the University of Vigo and at the Core Repository of the Institute of Marine Sciences- CSIC in Barcelona. The first cm (or the second, if the first was unavailable) of the 117 core tops was sampled, assuming that it essentially represents present-day conditions.

2.1.2. Environmental variables

Data on temperature (Locarnini et al., 2013), salinity (Zweng et al., 2013), chlorophyll-*a* (Boyer et al., 2013), oxygen content and saturation (García et al., 2014a), nitrate, phosphate, and silicate (García et al., 2014b) were obtained from the 2013 World Ocean Atlas (WOA13), and mixed layer depth (Monterey and Levitus, 1997), total alkalinity (T_{ALK}) and total dissolved inorganic carbon (DIC) (Goyet et al., 2000) for the training set sites were taken for a grid of 1° longitude by 1° latitude, using weighted averaged gridding by Ocean Data View (ODV) software (Schlitzer, 2014). Similarly, data on pH and carbonate (CO_3^{2-}) were calculated using the 'derived variable' tool of ODV software. These data have been averaged annually and seasonally (for summer and winter) from 1955 to 2012 and were selected at 10, 20, 30, 50, 75, 100, 125, 150, and 200 ~~300~~ meters water depth.

2.2. Fossil data set

The fossil coccolithophore data set used for the reconstruction comprises coccolithophore census counts from core CEUTA10PC08 (36°1′22″N, 4°52′3″W; 914 mbsl), located in the Alboran Sea, previously published by Ausín et al. (2015). Fossil assemblages show a good-to-moderate degree of preservation. Location of this core lies under the modern path of the AW at the surface, near the Strait of Gibraltar (Fig. 1a). Sediment core chronostratigraphy was based on 15 radiocarbon ages and covered the time span from 25 to 4.5 ka calibrated BP at a ~65 yr temporal resolution (Ausín et al., 2015). All dates reported in this study are given in calibrated ages BP.

2.3. Micropaleontological analyses

Both modern (surface sediment) and fossil (downcore) samples were prepared for coccolithophore analyses according to the techniques proposed by Flores and Sierro (1997). A polarized-light microscope at 1000x magnification was employed to identify and count at least 500 coccoliths in each sample, belonging to 21 different taxa. Species whose relative abundance was < 1 % in the first count were considered later in 20 visual fields in order to estimate their abundance accurately. The final relative abundance of each species in each sample was then recalculated. *Gephyrocapsa* specimens smaller than 3 µm were lumped together and designated "small *Gephyrocapsa"* (Flores et al., 1997). The "medium *Gephyrocapsa*" group was made up of *Gephyrocapsa* whose size was between 3 and 5 µm. Two sizes of morphotypes of *E. huxleyi* (< 4 µm and > 4 µm) were considered owing to their different ecological and biostratigraphic significance in the study area. Similarly, *G. oceanica* was split according to a size criterion of < 5 and > 5 µm owing to their comparable potential ecological significance. Other taxa identified in this study were *Calcidiscus leptoporus*, *F. profunda, Gephyrocapsa* cf. *caribbeanica, Gephyrocapsa muellerae*, *Helicosphaera* spp., and *Syracosphaera* spp. (as dominant taxa). The rare taxa identified were *Braarudosphaera bigelowii, Calciosolenia* ~~*murrayi,*~~spp., *Coccolithus pelagicus* subsp. *braarudii, Coccolithus pelagicus* subsp. *pelagicus, Oolithotus fragilis, Pontosphaera* spp., *Rhabdosphaera clavigera, Umbilicosphaera* spp. and *Umbellosphaera* spp.

Twenty-nine samples were finally eliminated from the initial modern data set owing to their high content (> 10 %) in obviously reworked nannofossils. These taxa belong to older stratigraphic levels (consistently older than the Pliocene in this study), meaning that they were resuspended and transported from their original location to the sample site, and they lack any relationship with modern environmental conditions. 10 % of reworked specimens was chosen as an acceptable threshold below which the sample could be retained in the modern training set, after ruling out these reworked specimens, without compromising the statistical representativeness of the major species (Fatela and Taborda, 2002). Later examination of the spatial distribution of reworked specimens in the retained samples revealed that those with the highest percentages were close to river mouths, relating reworked specimens to river discharges and suggesting that the rest of the assemblage could be considered autochthonous. Thus, the final training set (supplementary material) comprised 88 surface samples (Fig. 1b): 78 from the Western Mediterranean (58 from the Balearic Sea and 20 from the Alboran Sea) and 10 from the Atlantic Ocean.

2.4. Statistical analyses

2.4.1. Relationship between coccolithophore assemblages and environmental variables

Prior to statistical analyses, environmental variables were checked for unimodal distribution. Only mixed layer depth, T_{ALK} and DIC were $\log_{10}$ transformed, since the transformation of other variables did not cause noticeable changes. Principal Component Analyses (PCA) was performed on this initial data set to assess the major environmental gradients and collinearity among the variables.

Coccolithophore relative abundances were square-root transformed to stabilize their variances. The species *Braarudosphaera* sp., *Calciosolenia* ~~sp~~spp., *Coccolithus pelagicus* subsp. *braarudii*, *Coccolithus pelagicus* subsp. *pelagicus*, *and Pontosphaera* spp., were excluded from the modern (and consequently from the fossil) coccolithophore assemblages since their maximum relative abundance was not > 1 % in at least two samples. Detrended Correspondence Analysis (DCA) was then performed on the modern coccolithophore assemblage to estimate the length of the environmental gradient. A length of the first DCA axis > 2 Standard Deviation (SD) units indicates the unimodal responses of the organisms (Birks, 1995; Ter Braak and Prentice, 1988), while shorter lengths indicate linear responses.

Akaike's information criterion (AIC) was used in an ordination analysis to identify the minimum number of variables (subset) that, being statistically significant, explained the maximum variation in the modern coccolithophore assemblage. Canonical Correspondence Analysis (CCA) was used to evaluate the influence of this environmental subset to explain coccolithophore distribution in the modern training set.

The ratio between the first constrained axis and the first unconstrained axis (λ_1/λ_2) was used as a diagnosis to test the strength of a single environmental variable when the effects of those remaining were excluded from the analyses (ter Braak and Juggins, 1993). If $\lambda_1/\lambda_2 \geq 1$, the variable under study is considered to be important for explaining the distribution of the species. The proportion of the variance in the coccolithophore training set explained uniquely by each significant environmental variable was calculated using variance partitioning.

Ordination analyses and variance partitioning were performed using the 'vegan' package v.2.3. (Oksanen et al., 2015) for R (R Core Team, 2015).

2.4.2. Transfer function

Calibration models were calculated for the variable of interest (and each variable by means of exploratory analysis) using the weighted-averaging-partial least squares (WA-PLS) method (ter Braak and Juggins, 1993; ter Braak et al., 1993) and the Modern Analog Technique (MAT) (Prell, 1985), both implemented in C2 version 1.4.3 software (Juggins, 2007). All models were calculated for the cross-validation predictions by bootstrapping (999 permutation cycles) (Birks, 1995). In MAT, the number of analogs resulting in the maximum coefficient of determination (R^2_{boot}) between the observed and predicted values and the lowest root-mean square error of prediction (RMSEP) (Telford et al., 2004) was calculated using an optimization set together with the usual training and test sets implemented in the 'analoge' package for R (R Core Team, 2015). In WA-PLS, a decrease of 5 % or more in RMSEP was required to retain the next component (Birks, 1995; ter Braak et al., 1993).

Many coccolithophore species inhabit at depths within a specific range of the photic zone and are subject to environmental seasonality (Winter et al., 1994). Therefore, the depth and season considered for calibration and reconstruction should be those that most influenced the coccolithophore fossil assemblage. Following the procedure described by Telford et al. (2013), we reconstructed the variable of interest based on summer-, winter- and annual-averaged data at 9 different depths of the photic zone from 10 to 200 m using the ‘paleoSig’ package v.1.1-1 (Telford, 2012) for R (R Core Team, 2015). The reconstruction that explains the highest proportion of variance in the fossil data being statistically significant reflects the depth and season that most influenced the coccolithophore fossil assemblage and hence provides the most suitable calibration.

Outliers may reduce the power of prediction of the calibration model as well as introducing undesirable effects in model coefficients (Birks, 1995). Potential outliers were determined as those whose absolute residual was higher than the mean SD of the observed values (Edwards et al., 2004).

A combination of the highest R^2_{boot} and the lowest RMSEP was used as a criterion for the quality prediction of the model. The graphical representations of the observed values against the values predicted by the model and the residuals against the predicted values were used as a diagnosis of the model.

2.4.3. Derived reconstruction and evaluation

MAT and WA-PLS were applied to the fossil coccolithophore assemblages of core CEUTA10PC08, which were previously square-root transformed, using C2 version 1.4.3 software (Juggins, 2007). Sample-specific reconstruction errors under bootstrapping were derived automatically by C2 software, considering the prediction error due to: i) errors in estimating species coefficients, and ii) errors in the calibration function (further details may be found in Birks et al. (1990)). In order to assess the quality of the modern analogs for the fossil (downcore) samples, the squared chord distance between each fossil sample and each sample in the modern training set (Overpeck et al., 1985) was calculated with MAT by C2 software. A squared chord distance below the 10th percentile would be considered good, while values above this cutoff would represent assemblages with poor analogs (Simpson, 2007).

The first axis of the PCA analyses of the fossil dataset ($PC1_{fossil}$) shows the most important changes in the composition of the fossil coccolithophore assemblage. Comparison between $PC1_{fossil}$ and the reconstructed variable of interest was used to assess whether the reconstruction could be considered representative of the major ecological changes of the fossil assemblage (Juggins, 2013).

3. RESULTS

3.1. Geographical distribution of coccolithophores

The small placoliths (small *Gephyrocapsa* and *E. huxleyi* < 4 µm) are the dominant taxa (Fig. 2b, c) constituting on average 83 % of coccolithophore assemblages. Small *Gephyrocapsa* shows higher abundances near the Spanish coast and southeast of the Balearic Islands. *E. huxleyi* < 4 µm is more abundant in the Balearic Sea and around the Ebro River Delta (Fig. 2c). *G. muellerae* (Fig. 2d) concentrates southeast of the Balearic Islands and shows a patch of 2 % in the northern Alboran Sea. *C. leptoporus* and *Helicosphaera* spp. (Fig. 2e, f) are almost absent in the Alboran Sea and show similar patchy distributions between the Catalan and the Balearic fronts and east of the Balearic Islands. *F. profunda* (Fig. 2g) is more abundant in the Atlantic Ocean (up to 16 %) and gradually decreases eastward. It shows two patches (up to 4 %) south of the Ebro River mouth. *G. oceanica* (< 5 µm) (Fig. 2h) is mostly distributed near the Strait of Gibraltar. It also shows a patch (up to 3 %) around the Andarax River mouth.

3.2. Relationship between coccolithophores and environmental variables

The PC1 explains 56.1 % of the variance within the environmental data set (Fig. 3a) and is highly correlated with CO_3^{2-}, salinity, pH and T_{ALK}. PC2 explains 22.3 % of the total variance and primarily summarizes the information on temperature and phosphate.

The ordination based on the AIC revealed that only salinity, nitrate, phosphate, silicate and oxygen are needed to explain the maximum variation in the modern coccolithophore assemblage and are significant at the 95 % level when added individually to the model via a forward selection procedure. The first axis of the DCA performed on the modern coccolithophore assemblage was 2.6 SD units. Accordingly, unimodal methods were followed. The CCA (Fig. 3b) revealed sites and species distribution along this environmental subset. The others were also plotted as passive variables to avoid overfitting. The vectors show that salinity exhibits the longest gradient and is strongly correlated with the CCA1, indicating a strong relationship with coccolithophore distribution. Some sites from the Alboran and Balearic Seas and the taxa medium *Gephyrocapsa* and *E. huxleyi* (> 4 µm) were found to be distributed along the CCA2. Individual CCAs (Table 1) to calculate λ_1/λ_2 showed that salinity was the most important variable among those found to be significant. Variance partitioning revealed that these significant variables accounted for 38.9 % of the cumulative variance in the coccolithophore training set and salinity explained a large proportion of this variance (15.5 %).

3.3. Transfer functions

Salinity explained the largest amount of variation in the coccolithophore assemblages and was therefore chosen to develop the coccolithophore-based transfer function. Additionally, comparison among the R^2_{boot} from preliminary calibration models for each variable confirmed the best predictive power for salinity (Table 1).

Among the WA-PLS models for salinity, the two-component model (WA-PLS2) was chosen as the most suitable since it afforded a reduction of 6.4 % in the RMSEP. The ideal number of analogs for MAT was six.

The analyses of the amount of down-core variance explained by the summer, winter, and annual salinity reconstructions at 9 different depths and their statistical significance revealed that the mean-annual reconstruction at 10 m explained the highest variance. Hence, the reconstruction for core CEUTA10PC08 was based on the mean-annual salinity data at 10 m depth and referred to as SSS reconstruction.

Five samples showed higher residuals than the SD of salinity and were preliminary identified as potential outliers (supplementary material). However, only one of these samples (CO-81-2/TK-2) was identified as an outlier in both MAT and WA-PLS regression methods. This had a bright yellowish color under the microscope, likely due to the effect of diagenetic processes. In order to retain the maximum number of observations representing modern environmental conditions, only this sample was removed from subsequent model implementations, leading to an improvement of the MAT and WA-PLS2 R^2_{boot} coefficient of 3.4 % and 6.6 %, respectively, and reducing both Max_Bias_{boot} and RMSEP (Table 2).

The final MAT and WA-PLS2 models showed similar quality predictions (Table 2). The salinity values in the modern training set vary from 36.2 to 38.2 psu. Intermediate values (37.1-37.6 psu) are less well represented by the observations (Fig 4a). MAT- and WA-PLS2-predicted values are shown in Figs. 4b, c. The predicted *versus* observed values from both models approach the diagonal of slope one (which indicates perfect predictions) reasonably well (Fig. 4d, e). The residuals for the MAT and WA-PLS2 models (Fig. 4f, g) are equally distributed around zero and show no apparent trends.

3.4. SSS reconstruction

SSS trends and values reconstructed for the CEUTA10PC08 core derived from both MAT and WA-PLS2 are very similar (Fig. 5a, b). These only differ during the stadials associated with Heinrich Events 2 and 1 (H2 and H1), when the WA-PLS2-estimated SSS shows more pronounced salinity decreases.

The SSS reconstructions obtained from core CEUTA10PC08 (Fig. 5a) can be divided into three intervals: i) the period from 25.5 to 15.5 ka is characterized by higher values~~, ranging~~ that oscillate between 37.8 and 37 psu. Lower values are found from 20 to 18 ka, followed by a drop of 0.8 psu at 17.3 ka; ii) the period from 15.5 to 9 ka shows fast, large-amplitude changes. An abrupt decrease from 37.9 to 36.9 psu can be recognized at 15 ka, followed by large peaks of high values at 12.8, 11.1, and 10.2 ka; and iii) the period from 9 to 4.5 ka records the lowest values, which vary between 37 and 36.5 psu, and shows a general decreasing trend.

On average, the errors associated with both SSS reconstruction are of a similar magnitude: ±0.15 psu for MAT and ±0.17 psu for WA-PLS (Fig. 5a). Squared chord distances between fossil and modern assemblages (Fig. 5b) revealed that many samples from 25.5 to 16 ka were above the 10th percentile. A comparison between $PC1_{fossil}$ and the SSS reconstruction is depicted in Fig. 5c, showing general good agreement, especially for the last 16 kyr.

4. DISCUSSION

4.1. Geographic coccolithophore distribution and SSS

E. huxleyi (< 4 µm) and small *Gephyrocapsa* are widespread in the Western Mediterranean, as previously reported for surface sediment and water column samples (Álvarez et al., 2010; Knappertsbusch, 1993; Oviedo et al., 2015). These taxa, especially *E. huxleyi* (< 4 µm), are cosmopolitan and tolerate wide ranges of temperature and salinity (Winter et al., 1994). *G. muellerae* abundance is higher southeast of the Balearic Islands, where the MAW encounters more saline and warmer Mediterranean waters, and close to the Alboran Front, possibly reflecting its preference for rich-nutrient waters, as reported for sediment trap samples in the Alboran Sea (Bárcena et al., 2004; Hernández-Almeida et al., 2011). *C. leptoporus* and *Helicosphaera* spp. (Fig. 2e, f) show similar spatial distributions and abundances. Interestingly, the CCA suggests that *Helicosphaera* spp. have a preference for more saline waters (Fig. 3b). By contrast, in paleoceanographic works this species has been linked to fresher and turbid waters in the Mediterranean Sea (Ausín et al., 2015; Colmenero-Hidalgo et al., 2004; Grelaud et al., 2012). *Helicosphaera* spp. abundance in surface sediments from the northeastern Balearic Island has also been related to upwelling events (Álvarez et al., 2010). Similarly, the abundance of *C. leptoporus* in the Alboran Sea has been linked to nutrient-rich waters (Bárcena et al., 2004). The similar patchy pattern shown by both species may be related to the temporary upwelling of nutrient-rich waters associated with frontal structures in the area limited by the Balearic and Catalan fronts (Font et al., 1988). In agreement with this interpretation, the co-occurrence of both species in other Mediterranean locations has already been linked to high coccolithophore productive periods and pre-upwelling events (Hernández-Almeida et al., 2011; Ziveri et al., 2000). The CCA (Fig. 3b) suggests that *F. profunda* and *G. oceanica* (< 5 µm) woul be associated to less saline waters. This notion may partly be a consequence of their higher abundance in Atlantic waters (Fig. 2d). *G. oceanica* has already been proposed as a tracer for AW influx in the Western Mediterranean Sea (Álvarez et al., 2010; Bárcena et al., 2004; Knappertsbusch, 1993; Oviedo et al., 2015). Similarly, the *F. profunda* spatial distribution reflects the path of the Algerian current (Fig. 2a, d), formed by recent and fresher MAW (Fig. 2a, d). Low percentages of *F. profunda* spottily distributed south of the Ebro River and in the Catalan-Balearic Sea suggest this species may be also affected by the influence of river discharges (Álvarez et al., 2010). These results suggest that *F. profunda* and *G. oceanica* proliferate mainly in waters of Atlantic origin, but not exclusively, as indicated by their presence in the Eastern Mediterranean (Knappertsbusch, 1993; Malinverno et al., 2008) where Atlantic influence becomes diluted.

Salinity was highly correlated with CO_3^{2-} and pH (Fig. 3a, b). Oviedo et al. (2015) have found exactly the same variables as being the most important factors to account for changes in heterococcolithophore assemblages from the study of coccolithophore distribution from water column samples and *in situ* environmental measurement in the Mediterranean Sea. In our study, multivariate analyses revealed that salinity was significant and was the most important variable of those studied in explaining the variance in coccolithophore data in this modern training set. However, the individual importance and proportion of variance explained by each of the significant variables was not assessed in the study of Oviedo et al. (2015). Despite this, the authors discarded salinity as a final explanatory variable, arguing that *E. huxleyi*, the most abundant and ubiquitous extant coccolithophore (Cros and Fortuño, 2002), inhabits at a wide salinity range, suggesting a negligible ecological effect of salinity on coccolithophores. Contrary to this reasoning, the direct relationship between varying salinities and the morphology of *E. huxleyi* has been demonstrated by several authors (Bollmann and Herrle, 2007; Bollmann et al., 2009; Fielding et al., 2009; Green et al., 1998; Paasche et al., 1996; Schouten et al., 2006) in both culture experiments and marine surface sediment samples. Oviedo et al. (2015) later explained the high and negative relationship that they found between salinity and *G. oceanica*, *G. muellerae* and *E. huxleyi* morphotype B/C distributions as being a consequence of their carry-over by the AW through the Mediterranean. Instead of this, however, we interpret the AW influx as promoting the optimal conditions for these species to thrive in the Mediterranean Sea. Therefore, the coccolithophore relationship with salinity would reflect the different water masses where coccolithophore species prefer to inhabit.

It is worth mentioning that salinity influences the solubility of CO_3^{2-} via several pathways: the solubility of free carbon dioxide in water, the solubility product constants, the concentration of hydrogen ions, and the quantity of calcium in the water (Trask, 1936). Accordingly, salinity could influence coccolithophores through coccolith calcification processes. In contrast, Bollmann and Herrle, (2009) have proposed that salinity influences coccolithophores through cell turgor regulation linked to osmotic processes. Although there is no clear consensus about the mechanism through which salinity influences coccolithophores, many other studies point to a strong influence of this variable on molecular compounds only produced by coccolithophores and on specific species. In the Japan Sea, salinity has been proposed to have an ecological or physiological influence on the production of alkenone and alkenoates, which are organic compounds mainly produced by the genera *Emiliania* and *Gephyrocapsa* (Fujine et al., 2006).

In the Baltic Sea, alkenone unsaturation ratios have been found to be significantly correlated with salinity (Blanz et al., 2005). In the Mediterranean Sea, Knappertsbusch (1993) found that *G. oceanica* distribution was linearly correlated with salinity. Based on such evidences, we propose that the assemblage composition may be conditioned by the optimum salinity range preferred by each species. Moreover, salinity has proved to be important to other marine unicellular planktonic groups such as diatoms (Jiang et al., 2014; Li et al., 2012) and dinoflagellate cysts (Jansson et al., 2014, and references therein), reinforcing the hypothesis of salinity as an important variable for planktonic communities in semi-enclosed basins.

4.2. Transfer function quality

A general good fit can be deduced for both models, although MAT was seen to perform slightly better from a higher R^2_{boot} and a lower RMSEP (Table 2) and plotted predicted values compared with observed values (Fig. 4). Intermediate salinity values (37.1- 37.6 psu) are less well represented than the more extreme values (Fig. 4d, e). Unevenness can bias the RMSEP leading to overestimation of the predictive power of the model (Telford and Birks, 2011).While an even distribution would be always desirable, unevenness is a feature inherent to most training sets from oceanic environments. In this case, it is not severe and the observations, although unevenly distributed along the salinity gradient, do not leave gaps. The distribution of the residuals (Fig. 4f, g) indicates the adequacy of the model.

4.3. Downcore SSS reconstruction

The derived MAT and WA-PLS2 SSS reconstructions (Fig. 5a) are very similar. Nevertheless, WA-PLS2 shows more marked salinity decreases than MAT during the H2 (25.2-23.7 ka) and H1 (17.4-15.9 ka). Unlike WA-PLS, MAT does not consider the entire dataset when calculating the species optima, only the most taxonomically similar analogs, and is more sensitive to local conditions (Telford and Birks, 2009). Fossil samples lack good analogs for the H2 and H1, coinciding with large peaks of *E. huxleyi* (> 4 μm) (Fig. 5b). H2 and H1 have been linked to the entry of cold and fresher water originating from the North Atlantic ice melting in the Western Mediterranean Sea (Cacho et al., 1999; Melki, 2011; Sierro et al., 2005), suggesting the preference of *E. huxleyi* (> 4 μm) not only for cold waters (Colmenero-Hidalgo et al., 2002; Colmenero-Hidalgo et al., 2004) but also fresher waters in the past. By contrast, Bollmann and Herrle, (2007) reported a current positive correlation between the size of *E. huxleyi* up to 4 μm and increasing salinities from the study of globally distributed core-top samples. These authors used this relationship to estimate salinity values during the LGM. Interestingly, they observed several overestimations with regard to other published values in samples characterized by high relative abundances of larger specimens of *E. huxleyi* (> 4 μm). These discrepancies suggest that *E. huxleyi* (> 4 μm) in ancient sediments lacks an analog in modern assemblages, as indicated by the high dissimilarity between fossil samples with high percentages of this species and modern samples (Fig. 5b).

Because MAT is strongly dependent upon on the analogs selected (Telford and Birks, 2009) and since the WA-PLS2 reconstruction for H2 and H1 is more coherent with a freshwater inflow scenario, it seems that WA-PLS2 affords more reliable values than MAT. Consequently, WA-PLS2-estimated SSS was chosen for our final interpretations.

Transfer functions assume that the ecological response of organisms to either the environmental variable of interest or to the linear combination of this important variable with others has not changed significantly over the time span represented by the fossil assemblage (Birks, 1995). The good agreement observed between $PC1_{fossil}$ and the reconstructed SSS patterns from 16 ka onwards (Fig. 5c) suggests that the SSS transfer function fulfills this assumption back to 16 ka. Larger differences are observed from 25 to 16 ka, possibly promoted by the lack of analogs during this time span, discussed above. Consequently, the SSS reconstruction from 25 to 16 ka will not be discussed further.

4.4. SSS changes in the Alboran Sea over the last 15.5 kyr

4.4.1. Termination 1b (T1b)

A decrease in salinity of about 0.6 ± 0.15 psu occurred from 15.4 to 14.6 ka (Fig. 6a). The global sea-level rise of ~20 m during meltwater pulse 1a (mwp-1a) has been dated between 14.6 and 14 ka (Stanford et al., 2006, and references therein). Since this section covers 3,000 yr with no control point (Fig. 6a), it could be an artifact of poorly constrained chronology for this time interval. Nevertheless, this seems unlikely because other authors (Duplessy et al., 1992; Emeis et al., 2000; Kallel et al., 1997) have reported SSS decreases in different regions of the Mediterranean Sea and Atlantic Ocean at this time from a combination of oxygen isotope ($\delta^{18}O$) and SST data. These salinity decreases are larger than that observed for the CEUTA10PC08 core. For instance, Duplessy et al. (1992) identified a salinity drop of about 2.5 psu in an Atlantic core west of the Strait of Gibraltar. It is worth mentioning that the salinity changes estimated by this method depend strongly on the accuracy of the SST record (Schmidt, 1998) and the unknown salinity-seawater $\delta^{18}O$ relationship in the past (Rohling, 1999), being sensitive to several deviations and uncertainties that are difficult to assess (Rohling, 2000; Rohling and Bigg, 1998; Schmidt, 1999). Although the uncertainty in the chronology prevents a robust correlation, the smaller SSS decrease identified in the SSS reconstruction could be related to the Laurentide ice sheet melting and retreat at ~15.5 ka (Clark et al., 2001). This event has already been proposed to be the cause of the freshwater input identified at 15.3 ka south of Iceland via advection within the North Atlantic Current

(NAC) and subsequently its northern branch (Thornalley et al., 2010). Similarly, the southeastern branch of NAC could have advected freshwater to the study area.

4.4.2. Bølling−Allerød (B−A)

The SSS values are generally low for the B−A, the Bølling being fresher than the Allerød (Fig. 6a). Owing to the global sea-level rise during the B−A, a greater volume of AW would have entered through the Strait, decreasing the average SSS. This period of reduced salinity also coincides with the highest values of total concentration of C_{37} alkenones, a proxy of organic matter preservation, from a nearby core located off the coast of Malaga (Ausín et al., 2015) (Fig. 6b). This accumulation of high amounts of organic matter resulted in the formation of the so-called Organic-Rich Layer (ORL-1) (Cacho et al., 2002) in the Western Mediterranean, although its origin is still under debate (Rogerson et al., 2008; Rohling et al., 2015). The joint effect of a salinity reduction of 0.8 psu and a temperature increase of 3 ºC (Cacho et al., 2001) (Fig. 6c) would have led to a significant reduction in sea surface density, possibly prompting stagnation of the upper water column. This, along with increased organic matter export to the seabed (Ausín et al., 2015) and reduced deep-basin ventilation (Martínez-Ruiz et al., 2015), would have hampered organic matter mineralization, reinforcing the formation of the ORL-1 in the Alboran Sea. According to Rohling et al. (2015), the origin of ORL-1 lies in hydraulic changes in the Strait of Gibraltar (Bernoulli aspiration depth) and/or the inhibition of deep water formation in the Gulf of Lion, both resulting from a drastic reduction in seawater density. Those authors have shown that the mwp-1a and the monsoon flooding into the Eastern Mediterranean were insufficient to trigger these mechanisms, and demonstrated that the Alpine melt-water input into the NW Mediterranean at this time (Ivy-Ochs et al., 2007) may have played an essential role as freshwater forcing.

4.4.3. Younger Dryas (YD) and the Holocene

The YD exhibits a shift from higher to lower SSS values, decreasing by a total of 0.6 psu along its two phases: YDa and YDb (Fig. 6a). Several large short-term SSS fluctuations occurred as from the onset of the YD throughout the early Holocene (up to 8 ka). This time span coincides with a sea level rise of ~ 30 m (Peltier and Fairbanks, 2006) due to short-lived freshwater inputs associated with residual melting of the northern hemisphere ice sheets (Andrews and Dunhill, 2004; Elmore et al., 2015; Seidenkrantz et al., 2013; Tornqvist and Hijma, 2012). Six brief periods of a SSS decreasing trend were identified at 12.77-12.06, 11.95-11.71, 11.24-11.00, 10.09-9.83, 9.30-9.12, and 8.95-7.90 ka (Fig. 6a). REDFIT spectral analysis reveals a periodicity of 770±40 years (Fig. 6d), very similar to the 730±40 years cycle found by Cacho et al. (2001) in a SST record in the Alboran Sea, which was punctuated by the so-called Alboran cooling (AC) events (Fig. 6c). Although this similarity does not necessarily imply a causal relationship, the timing of SSS decreases is comparable to that of the AC events (Table 3), suggesting a common origin. Cacho et al. (2001) have associated the AC events to influxes of cold Atlantic waters in the Alboran Sea during ice-rafted debris discharges (so-called Bond events) (Bond et al., 1997) (Table 3). These latter authors noted that the oxygen isotopic record showed no evidence of any of the coolings found for each Bond event during the Holocene and argued that the cooler surface waters may have also been fresher, offseting the expected temperature-driven $\delta^{18}O$ enrichment in their records. Similarly, the highly-resolved $\delta^{18}O$ profile reported by Cacho et al. (2001) does not show any of the expected oxygen isotopic enrichments associated with the AC events, supporting the presence of fresher waters at those times. We suggest that freshwater advection (FA) events (as well as AC events) would have resulted from the influx of fresher and colder Atlantic waters in the Alboran Sea related to the southeastward drifting of meltwater from the Labrador, Greenland and Iceland seas (Bond et al., 1997).

FA events only occurred during the early Holocene, while AC and Bond events have also been identified through the middle and late Holocene. Wenner et al. (2014) concluded that, unlike those occurring later, early Holocene Bond events originated from changes in the meridional overturning circulation due to meltwater pulses from the Northern Hemisphere ice-sheets. It is likely that FA events would only have been noticeable when this mechanism was operating (i.e. the early Holocene), since very little meltwater was present after that period (Elmore et al., 2015).

An SSS increase of 0.87±0.15 psu is observed from 10.7 to 10 ka. Because the Western Mediterranean is a semi-enclosed basin, local conditions may have played a role as additional feedbacks for this rapid high-amplitude variability. For this brief period, Frigola et al. (2008) have demonstrated the most pronounced weakening of the Mediterranean thermohaline circulation for the last 50 ka. The consequent reduction in Atlantic-Mediterranean water exchange, along with the maximum summer insolation and inland aridity (Fletcher et al., 2010), would have led to more saline surface waters.

FA1 includes the 8.2 ka event (Alley et al., 1997), which has been linked to a sub-thermocline freshening of 0.5 psu in the North Atlantic (Thornalley et al., 2009). However, no distinctive SSS changes are observed in relation to this event, suggesting that it would have had a negligible effect on surface salinity in the Alboran Sea. Minimum SSS values are recorded at 7.8 ka, possibly related to maximum high-stand conditions reached at 7.4 ka (Zazo et al., 2008), along with the influence of the African Humid Period (AHP; 11-5.5 ka) over the study area, especially up to its decline at 7.4 ka (deMenocal et al., 2000). From 7.8 to 4.5 ka, salinity values level off around 36.6 psu, close to present SSS values.

5. CONCLUSIONS

Multivariate statistical analyses show that coccolithophore distribution of modern coccolithophore assemblages in the Atlantic Ocean, west of the Strait of Gibraltar, and the Western Mediterranean was mainly influenced by annual mean salinity at 10 m depth. MAT and WA-PLS2 calibration models show similar outcomes. These models were applied to coccolithophore assemblages from a fossil core to reconstruct SSS at high resolution for the last 25 kyr in the Alboran Sea. Statistical analyses reveal assemblages lacking good modern analogs in relation to the species *E. huxleyi* > 4 µm during H2 and H1 and part of the LGM, preventing further interpretations for these periods. A low SSS was found for the B−A, possibly due to the post-glacial sea-level rise~~ and regionally wetter conditions in the adjacent continental areas.~~. The consequent reduction in sea surface density is suggested to have reinforced the formation of the ORL-1. During the YD and Holocene, six brief, abrupt SSS decreases at 12.77-12.06, 11.95-11.71, 11.24-11.00, 10.09-9.83, 9.30-9.12, and 8.95-7.90 ka were linked to the advection of fresher and colder AW related to the southeastward drifting of meltwater in the North Atlantic ~~and continental aridity~~. No evidence of the 8.2 ka event is found in the reconstructed SSS, which reached its lowest values at 7.8 ka, coinciding with high-stand conditions in the Alboran Sea and the onset of the decline of the African Humid Period. SSS remained low from 7.8 to 4.5 ka, close to its present values.

A broader understanding of the ecological link between coccolithophore species and environmental parameters would be desirable in order to be able to place coccolithophore-based transfer functions within the ecological context in future works. Nevertheless, the diverse statistical tests performed in this study and the strong emphasis placed on assessing the validity and reliability of both the model and the reconstruction do reveal the potential of coccolithophores for developing transfer functions. The derived transfer function provides a potential independent proxy for quantitative reconstructions of SSS changes in other locations of the Western Mediterranean Sea over the last 15.5 kyr.

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Table 1.

Multivariate analyses results. λ_1/λ_2: individual CCA. Preliminary model coefficients from MAT and WA-PLS2. R^2_{boot}: bootstrapped coefficient of determination between the observed and predicted values. RMSEP: root mean square error of prediction.

			MAT		**WA-PLS**	
Variable	**λ_1/λ_2**	**% Explained variance**	**Boot_R^2**	**RMSEP**	**Boot_R^2**	**RMSEP**
*Salinity	1.38	15.47	0.83	0.30	0.75	0.33
*Nitrate	0.65	8.14	0.45	0.32	0.39	0.33
*Phosphate	0.25	4.89	0.36	0.02	0.19	0.02
*Silicate	0.22	8.93	0.56	0.24	0.40	0.26
*Oxygen	0.1	1.46	0.15	0.05	0.05	0.05
Chlorophyll-*a*			0.61	0.05	0.58	0.05
Temperature			0.12	0.52	0.07	0.53
Oxygen Saturation			0.20	1.04	0.18	1.02
Mixed Layer Depth			0.31	0.19	0.25	0.19
CO_3^{2-}			0.74	0.02	0.70	0.02
pH			0.70	0.02	0.67	0.02
T_{ALK}			0.70	0.00	0.67	0.004
DIC			0.51	13.31	0.48	13.16

*Variables determined by ordination based on AIC.

Table 2.

Model coefficients from final MAT and WA-PLS2 cross-validated by boot-strapping for SSS, after removal of one outlier. R^2_{boot}: bootstrapped coefficient of determination between the observed and predicted values. Max_Bias_{boot}: bootstrapped maximum bias. RMSEP: root mean square error of prediction (psu).

	MAT	**WA-PLS2**
R^2_{boot}	0.85	0.80
Max_Bias_{boot}	0.23	0.22
RMSEP	0.29	0.30

Table 3.

Timing (given in ka cal. BP) of: freshwater advection events (FA) deduced from SSS decreases in the CEUTA10PC08 core (this study) and their magnitude; Alboran cooling (AC) events from core MD 95-2043 (ACYD-AC3, Cacho et al., (2001)); and Bond events in the North Atlantic (Bond et al., 1997).

SSS decreases	SSS change (psu)	AC events	Bond events
FAYD	0.79±0.15	ACYD	
12.77-12.06		13.1-12.0	12.5
FA5	0.22± 0.16	AC6	---
11.95-11.71		11.9-11.65	
FA4	0.41± 0.16	AC5	
11.24-11.00		11.21-10.95	11.1
FA3	1.0± 0.15	AC4	
10.09-9.83		10.34-9.95	10.3
FA2	0.42± 0.15	---	
9.3-9.12			9.4
FA1	0.57± 0.15	AC3	
8.95-7.9		9.08-7.56	8.1

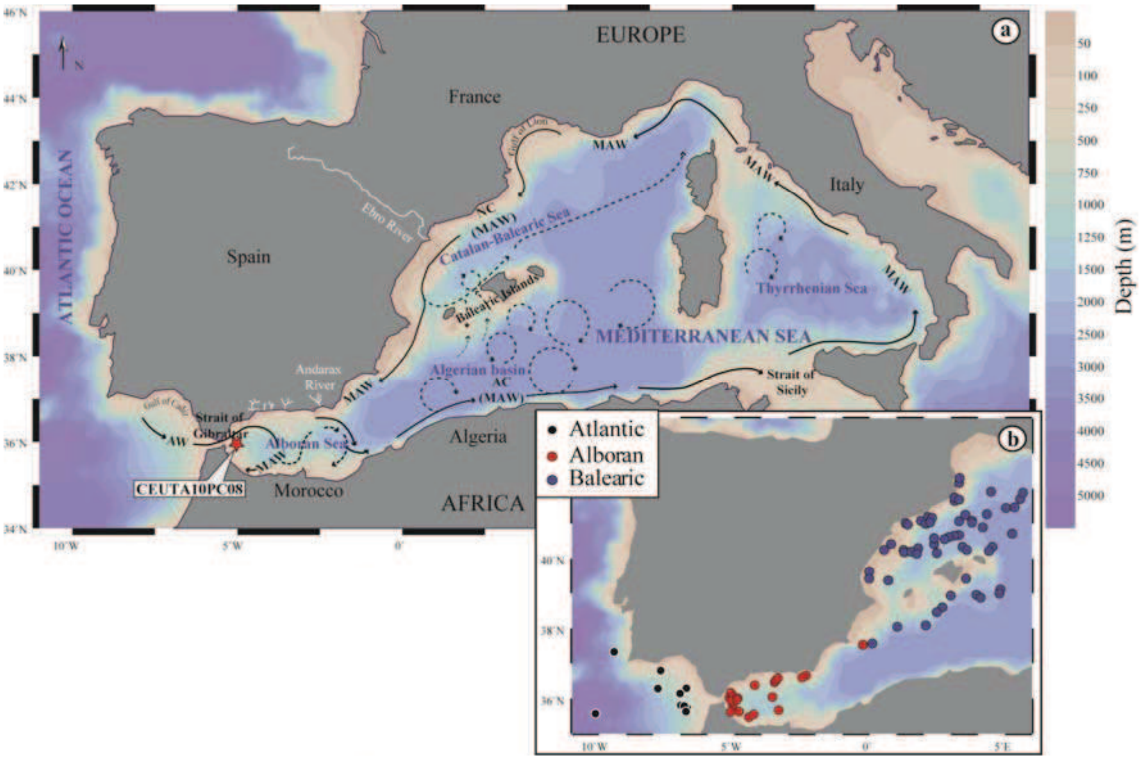

Figure 1. Maps of the study area. a) Location of the CEUTA10PC08 core (red star). Black arrows trace general surface circulation. Legend: AW: Atlantic Water. MAW: Modified Atlantic Water. AC: Algerian Current. NC: Northern Current. b) Location of the 88 core-top samples used for final calibrations. Maps generated with Ocean Data View software (Schlitzer, 2014).

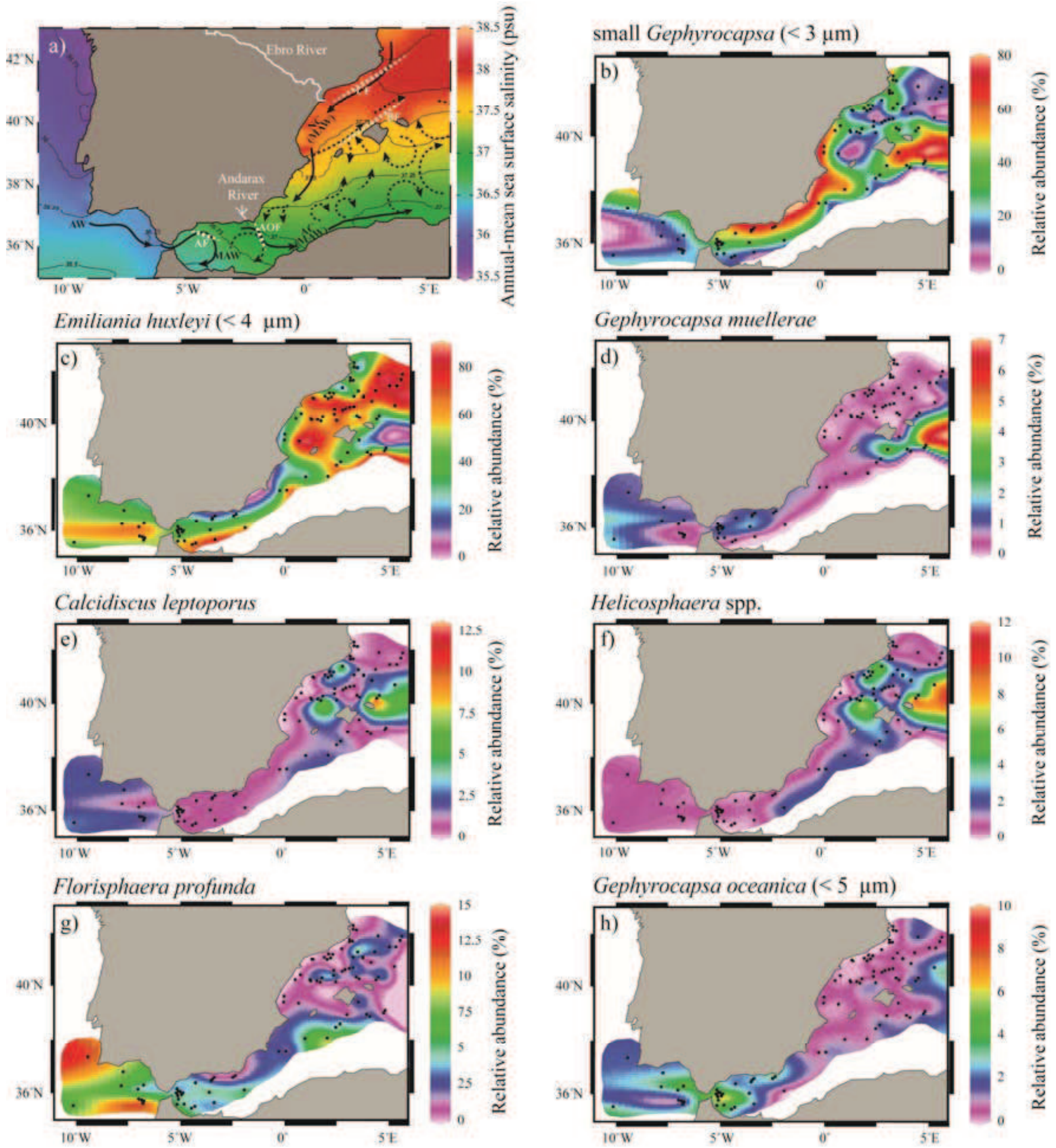

Figure 2. Geographical distribution of the main coccolithophore taxa. a) Annual-mean salinity at 10 m depth and surface circulation patterns in the study area: permanent trajectories (black arrows) and semi-permanent meso-scale features (dashed arrows). Legend: AF: Alboran Front; AOF: Almería-Orán Front; CF: Catalan Front; BF: Balearic Front; AW: Atlantic Water; MAW: Modified Atlantic Water; AC: Argelian Current; NC: Northern Current. Distribution, according to their relative abundance (%), of: b) small *Gephyrocapsa* (< 3 µm); c) *E. huxleyi* (< 4 µm); d) *G. muellerae*; e) *C. leptoporus*; f) *Helicosphaera* spp.; g) *F. profunda*; h) *G. oceanica* (< 5 µm).

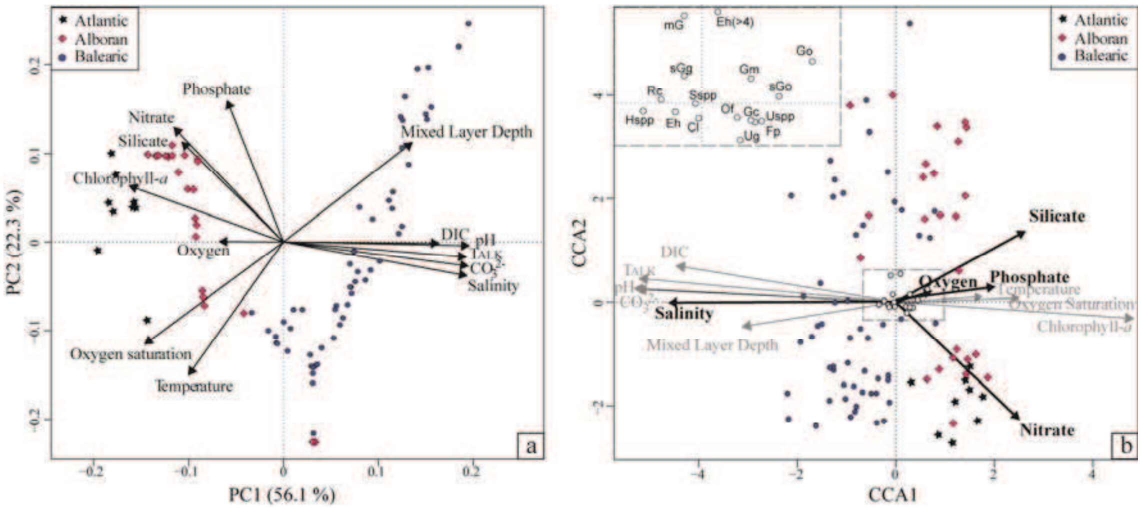

Figure 3. Multivariate analyses. a) PCA based on the thirteen initial environmental variables. b) CCA ordination plot with the site scores scaled by eigenvalues. The 88 sites are represented regarding their location in the Atlantic Ocean, Alboran Sea or Balearic Sea. Active and passive environmental vectors are represented by black and gray arrows, respectively. Scaling for the 16 taxa scores (open circles) is shown at the top left corner. mG: medium *Gephyrocapsa*; Eh(>4): *E. huxleyi* (> 4 µm); Eh: *E. huxleyi*; sGg: small *Gephyrocapsa*; Gm: *G. muellerae*; Go: *G. oceanica*; sGo: small *G. oceanica*; Rc: *R. clavigera*; Sspp: *Syracosphaer*a spp.; Of: *O. fraglis*; Gc: *G. cf. caribbeanica*; Hspp: *Helicosphaera* spp.; Cl: *C. leptoporus*; Uspp: *Umbellosphaera* spp.; Ug: *Umbilicosphaera* spp.; Fp: *F. profunda.*

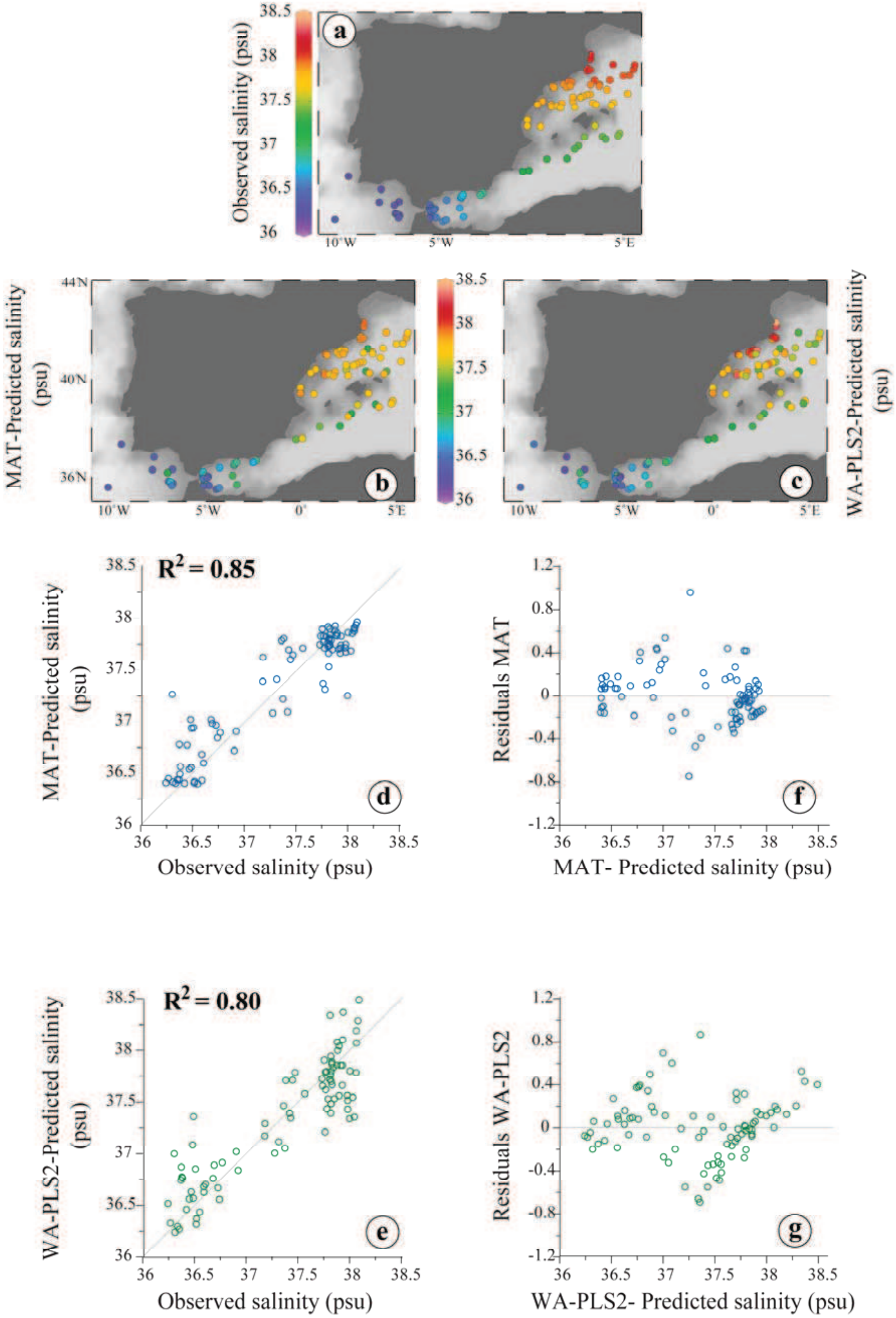

Figure 4. Diagnostic graphs of the models: a) Observed salinity values. b) MAT-predicted salinity values. c) WA-PLS2-predicted salinity values. d) Observed *vs* MAT-predicted salinity values. e) Observed *vs* WA-PLS2-predicted salinity values. f) MAT-predicted salinity values *vs* residuals. g) WA-PLS2-predicted salinity values *vs* residuals.

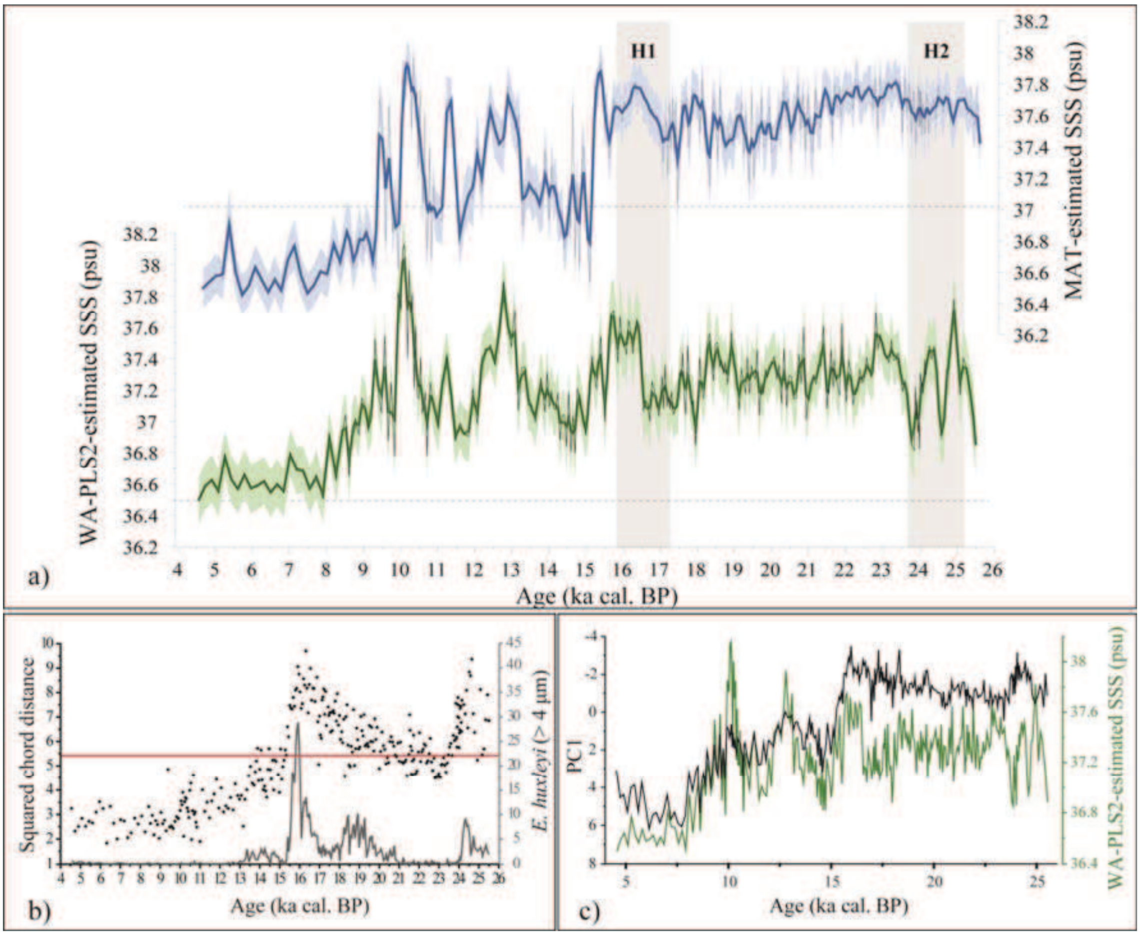

Figure 5. a) SSS reconstructions for the CEUTA10PC08 core derived from MAT (blue) and WA-PLS2 (green). The thin black lines represent the estimated values. The thick blue/green lines represent these original data fitted to a 3-point moving average smoothing spline. Pale blue/green shadows represent the error range, and dashed lines indicate current annual mean SSS in the Alboran Sea from the WOA13 (Zweng et al., 2013). b) Dissimilarity between modern and fossil assemblages (black dots) measured by squared chord distance (left axis) plotted *vs* age. The red line indicates the 10th percentile. Relative abundance of the species *E. huxleyi* (> 4 µm) (%; right axis). c) Profiles comparing the $PC1_{\mathbf{fossil}}$ (black line) and WA-PLS2-estimated SSS (green line).

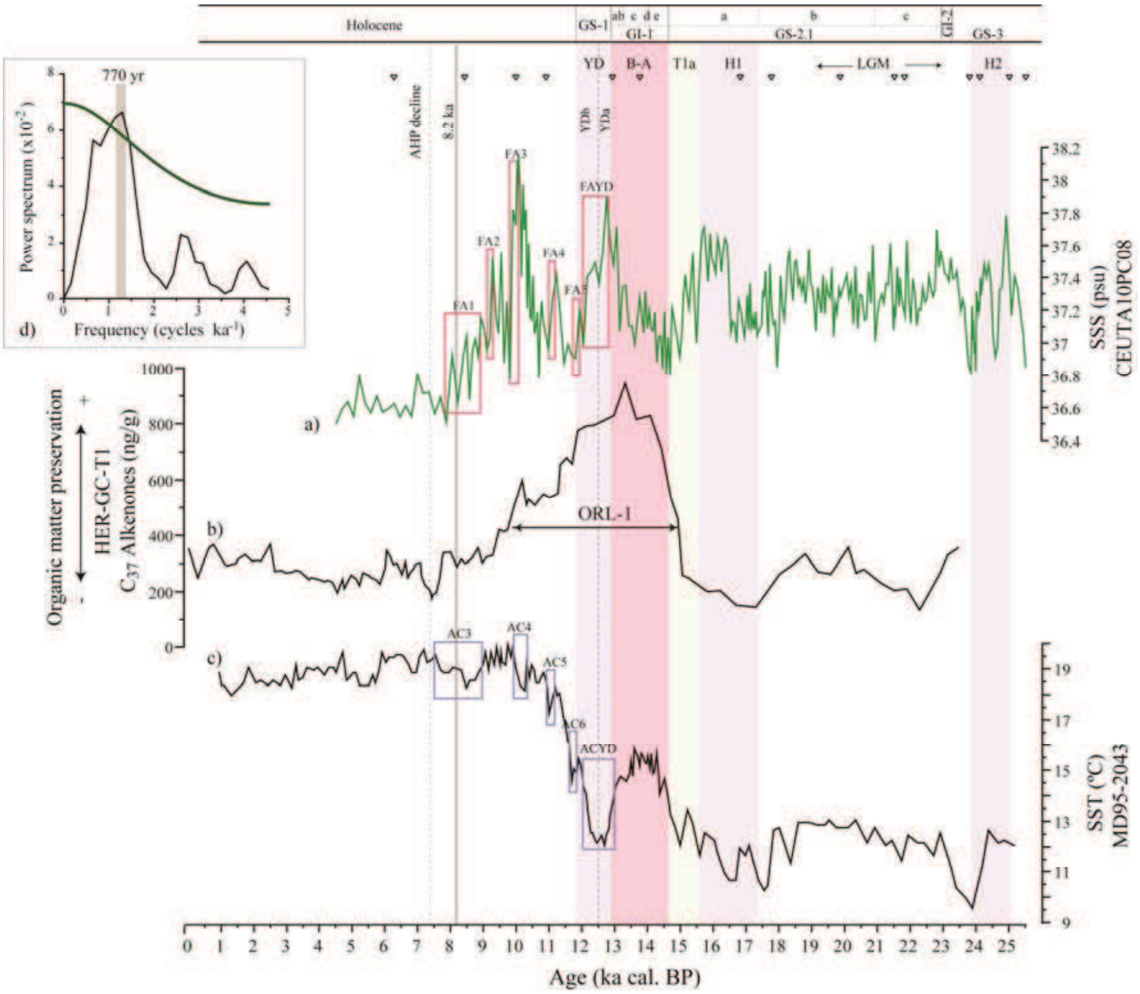

Figure 6. Paleoenvironmental records in the Alboran Sea: a) WA-PLS2-SSS reconstruction for CEUTA10PC08 core; age control points marked by triangles. b) C_{37} Alkenones from core HER-GC-T1 (Ausín et al., 2015). c) Alkenone-SST from core MD95-2043 (Cacho et al., 2001). Red boxes represent the Alboran cooling events (AC). d) REDFIT periodogram of the SSS reconstruction for the Holocene. The gray bar marks the only significant peak at the 95 % significance level (green line).