

Carbon Accumulation and Storage in a Temperate Coastal Lagoon under the Influence of Recent Climate Change (Northwestern Adriatic Sea)

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Abstract

Pialassa Baiona is a shallow temperate coastal lagoon influenced by a variety of factors, including regional climate change and local anthropogenic disturbances. To better understand how these factors influenced modern organic carbon (OC) sources and accumulation, we measured OC as well as stable carbon isotopes ($\delta^{13}\text{C}$) in ^{210}Pb -dated sediments within a vegetated saltmarsh habitat and a human impacted habitat. Relative Sea Level (RSL) at the nearby tide gauge station data and four different Sea Surface Temperature (SST) data sets were analyzed starting from 1900 to assess the potential effect of sea ingression and warming on the coastal lagoon sedimentary process. The source contribution calculated from the MixSIAR Bayesian model revealed a mixed sedimentary organic matter (OM) composition dominated by increasing marine-derived OM after the 1950s, parallel with decreasing autochthonous saltmarsh vegetation (*Juncus* spp.) in the saltmarsh habitat and riverine-estuarine-derived OM in the impacted habitat. RSL rise in the area ($8.7 \pm 0.5 \text{ mm yr}^{-1}$ in the period 1900-2014) has been mainly driven by the land subsidence, especially during the central decades of the last century, enhancing the sea ingression in the lagoon. Annual SST anomalies present, starting from the eighties, a continuous warming tendency from 0.034 ± 0.01 to $0.044 \pm 0.009 \text{ degC yr}^{-1}$. No direct effect on sedimentary properties was detected; however, RSL influenced OM sediment properties, although this effect was different between the two habitats.

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Carbon Accumulation and Storage in a Temperate Coastal Lagoon under the Influence of Recent Climate Change (Northwestern Adriatic Sea)

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Key Points:

- The relative contribution of allochthonous marine-derived organic matter increased over the years reflecting local relative sea level
- Decreased contribution of autochthonous-derived organic matter from C3 vegetation is evident in the saltmarsh habitat since the 1950s
- Relative sea level rise influenced sedimentary organic matter although this effect was different between and within habitats

Abstract

Pialassa Baiona is a shallow temperate coastal lagoon influenced by a variety of factors, including regional climate change and local anthropogenic disturbances. To better understand how these factors influenced modern organic carbon (OC) sources and accumulation, we measured OC as well as stable carbon isotopes ($\delta^{13}\text{C}$) in ^{210}Pb -dated sediments within a vegetated saltmarsh habitat and a human impacted habitat. Relative Sea Level (RSL) at the nearby tide gauge station data and four different Sea Surface Temperature (SST) data sets were analyzed starting from 1900 to assess the potential effect of sea ingression and warming on the coastal lagoon sedimentary process. The source contribution calculated from the MixSIAR Bayesian model revealed a mixed sedimentary organic matter (OM) composition dominated by increasing marine-derived OM after the 1950s, parallel with decreasing autochthonous saltmarsh vegetation (*Juncus* spp.) in the saltmarsh habitat and riverine-estuarine-derived OM in the impacted habitat. RSL rise in the area ($8.7 \pm 0.5 \text{ mm yr}^{-1}$ in the period 1900-2014) has been mainly driven by the land subsidence, especially during the central decades of the last century, enhancing the sea ingression in the lagoon. Annual SST anomalies present, starting from the eighties, a continuous warming tendency from 0.034 ± 0.01 to $0.044 \pm 0.009^\circ\text{C yr}^{-1}$. No direct effect on sedimentary properties was detected; however, RSL influenced OM sediment properties, although this effect was different between the two habitats.

Plain Language Summary

Coastal vegetated ecosystems (mangroves, saltmarshes, seagrasses) are highly efficient in removing carbon dioxide from the atmosphere through plant growth and thus play an important role in climate regulation, limiting the greenhouse effect. The term “blue carbon” has been devised to refer to carbon captured from the atmosphere and stored as organic matter in the sediments of these habitats. On the other hand, climate changes and human activities can affect coastal ecosystems and the quality and quantity of blue carbon. The goal of the present study was to analyze how the characteristic of the organic matter stored in the sediment of a Mediterranean coastal lagoon changed over the time from year 1850 to year 2010. These changes were compared with changes in sea water temperature and Relative Sea Level over the same period. Results showed that the importance of marine organic matter entering the lagoon from outside increased after the 1950s, while the contribution of organic matter produced inside the lagoon decreased. It is likely that Relative Sea Level, dominated by subsidence in the area, enhanced sea ingression and thus inputs of marine organic carbon. Such changes will continue to have an impact on the accumulation and storage of blue carbon.

1 Introduction

Carbon captured and stored in sediments from coastal vegetated habitats (wetlands, saltmarshes, tidal flats), where the rates of organic carbon (OC) accumulation from multiple sources is high, constitutes a relevant and active fraction in the global carbon sink, and plays an important role in climate regulation and mitigation (Kirwan and Mudd, 2012). However, a global inventory of this coastal ‘blue carbon’ storage remains a challenge, as observations of OC accumulation and stock in coastal ecosystems are labor intensive, expensive, scarce, and unevenly distributed, with few records even for relatively well-studied temperate areas in the Northern Hemisphere (Beaumont et al., 2014). Recent reviews (Duarte et al., 2005; Wilkinson et al., 2018) report a mean organic carbon accumulation rate of $151 \text{ g C m}^{-2} \text{ yr}^{-1}$ for saltmarshes (maximum $1720 \text{ g C m}^{-2} \text{ yr}^{-1}$), $41.4 \text{ g C m}^{-2} \text{ yr}^{-1}$ for lagoons (maximum $340 \text{ g C m}^{-2} \text{ yr}^{-1}$), and $62.9 \text{ g C m}^{-2} \text{ yr}^{-1}$ for coastal wetlands (maximum $335.8 \text{ g C m}^{-2} \text{ yr}^{-1}$) exceeding the mean burial rate of estuaries and continental shelves ($17\text{--}45 \text{ g C m}^{-2} \text{ yr}^{-1}$).

The rate of accumulation of ‘blue carbon’, mostly stored in soil and sediments within coastal vegetated ecosystems, is sensitive to rapidly changing climate factors (e.g. warming, sea level rise, inundation frequency) and non climatic anthropogenic drivers (e.g. eutrophication, landscape development) (Arriola, 2017; Cuellar-martinez et al., 2019; Ewers Lewis et al., 2018; Kelleway et al., 2017; Kirwan and Mudd, 2012; Macreadie and

Saintilan, 2019; Macreadie et al., 2013; Negandhi et al., 2019; Pendleton et al., 2012; Rogers et al., 2019; Ruiz-fernández et al., 2018; Simpson et al., 2017). Between 20 and 90% of the existing area occupied by coastal vegetated ecosystems is projected to be lost at global level by 2100, depending on sea level rise projections and warming under future emission scenarios (IPPC, 2019).

Carbon stable isotopic composition ($\delta^{13}\text{C}$) in combination with supporting geochemistry, specifically the ratio of organic carbon to total nitrogen (C/N) and total organic carbon content (OC), have been used as tracers to differentiate sources of organic matter (OM) that characteristically accumulate in sediments of coastal vegetated habitats near the Westerschelde Estuary (SW Netherlands), in Massachusetts and South Carolina (USA) (Chen et al., 2016; Goñi et al., 2003; Middelburg et al., 1997), and in Australian coastal wetlands (Saintilan et al., 2013), as well as past sea level indicators in the Thames Estuary (Khan et al., 2015b), in tidal-dominated wetlands in North West Europe (Wilson, 2017), in a back-barrier lagoon system in New Jersey, USA (Kemp et al., 2012), and in Hudson Bay, Canada (Godin et al., 2017).

The signature of bulk organic sediment $\delta^{13}\text{C}$ and C/N analysis in sediment records in European coastal vegetated habitats is primarily to distinguish between OM derived from autochthonous C3 and C4 saltmarsh vascular vegetation (i.e. coastal blue carbon; $\delta^{13}\text{C}$ -12‰ to -30‰ , C/N 5.80 to 41.10; Khan et al., 2015b), and allochthonous sources including fluvial and marine particulate organic matter (OM), the latter mainly derived from freshwater or marine phytoplankton ($\delta^{13}\text{C}$ -12‰ to -30‰ , C/N 5 to 9; Lamb et al., 2006).

There is, however, little knowledge about the spatial and historical distribution, and the sources of sedimentary OM in coastal vegetated habitats in the Mediterranean Region. In this study, carbon stable isotopic composition ($\delta^{13}\text{C}$) and C/N ratios were measured, and Pb-210 chronology reconstructed in sediment cores from two contrasting habitats (a semi-natural saltmarsh habitat and an impacted habitat) within a coastal lagoon (Pialassa Baiona) connected to the Northwestern Adriatic Sea. This temperate shallow coastal ecosystem is subject to ongoing sea level rise, sea warming (Carbognin and Tosi, 2002; Cerenzia et al., 2016; Tsimplis et al., 2012; Mariano et al., 2021), land subsidence, hydrological alterations, coastal erosion, maintenance dredging, embankments (Airolidi et al., 2016; Guerra et al., 2009).

The objectives of this work were to investigate the changes in OM accumulation and sources, and to assess the climatic factors and local anthropogenic disturbances influencing the spatial and temporal changes in OM 'blue carbon' storage in this temperate coastal lagoon (Pialassa Baiona) located in the Mediterranean Region.

2 Materials and Methods

2.1 Study area

Pialassa Baiona is a temperate coastal lagoon ($44^{\circ} 28' \text{N}$ and $44^{\circ} 31' \text{E}$) adjacent to the Northwestern Adriatic Sea (Figure 1). The area forms part of the Natura 2000 European network and is a wetland of international importance under the Ramsar Convention. The climate is hot-summer Mediterranean (Köppen-Geiger) with a continental influence, with a mean annual temperature of 14.1°C and a maximum of 29.2°C (Antolini et al., 2016). Annual rainfall ranges from 650 mm to 696 mm, mainly from October to March (Mollema and Antonellini, 2012).

The lagoon consists of shallow water bodies of saline, brackish or fresh water wetlands (~ 0.5 m average depth) completely or partially isolated by levees and crossed by a dendritic network of artificial channels dug in 1850 (~ 5 m maximum depth). The inner channels converge into a main channel connected to sea through the shipway channel. Salinity in the lagoon (25–35 psu) is mainly controlled by tidal flushing through this channel into the channels network resulting in delayed tidal oscillation and low water exchange with the open sea. On average, the water covers an area of 10 km^2 with a tidal range of 0.3–1 m, and usually vast shallow areas emerge during low tides.

The northern area is composed of unvegetated zones alternated with zones dominated by halophile species characteristic of temporarily inundated saltmarshes (*Juncus maritimus* and *Juncus acutus* and to a lesser

114 extent by *Salicornia* spp.; Merloni and Piccoli, 1999), with marginal areas with reedbeds of *Phragmites*
115 *australis* (Ferronato et al., 2016).

116 The southern area is a subtidal habitat consisting of tidal wetlands separated by discontinuous artificial
117 embankments receiving inputs from agriculture runoff, and urban and industrial wastewater discharges through
118 the main inflow channel (Airoldi et al., 2016). Vegetation is sparse and mainly present on the embankments.
119 This area has been subject to local anthropogenic disturbances (chemical and thermal pollution) resulting in
120 higher than expected concentrations of mercury, chlorinated compounds and polycyclic aromatic hydrocarbons
121 (PAHs) in sediments (Covelli et al., 2011; Guerra et al., 2014) and mussels (Capolupo et al., 2017), and
122 alteration of the benthic community (Guerra et al., 2009; Ponti et al., 2011). The high THg concentrations found
123 in the sediment of this area are in good agreement with the Hg inputs following the booming of petrochemical
124 industry located near Pialassa Baiona lagoon in the mid-1950s. Chemical plants producing acetaldehyde and
125 vinyl chloride from acetylene and using mercury salts as catalysts, released an estimated 100–200 tons of Hg
126 directly into the southern area of the lagoon during the 1958–1978 period (Miserocchi et al., 1993). In 1973,
127 wastewater treatment began, the acetaldehyde plant was shut down and the use of Hg catalysts in vinyl chloride
128 and acetaldehyde production was drastically reduced and finally discontinued by 1991 (Fabbri et al., 1998).

129

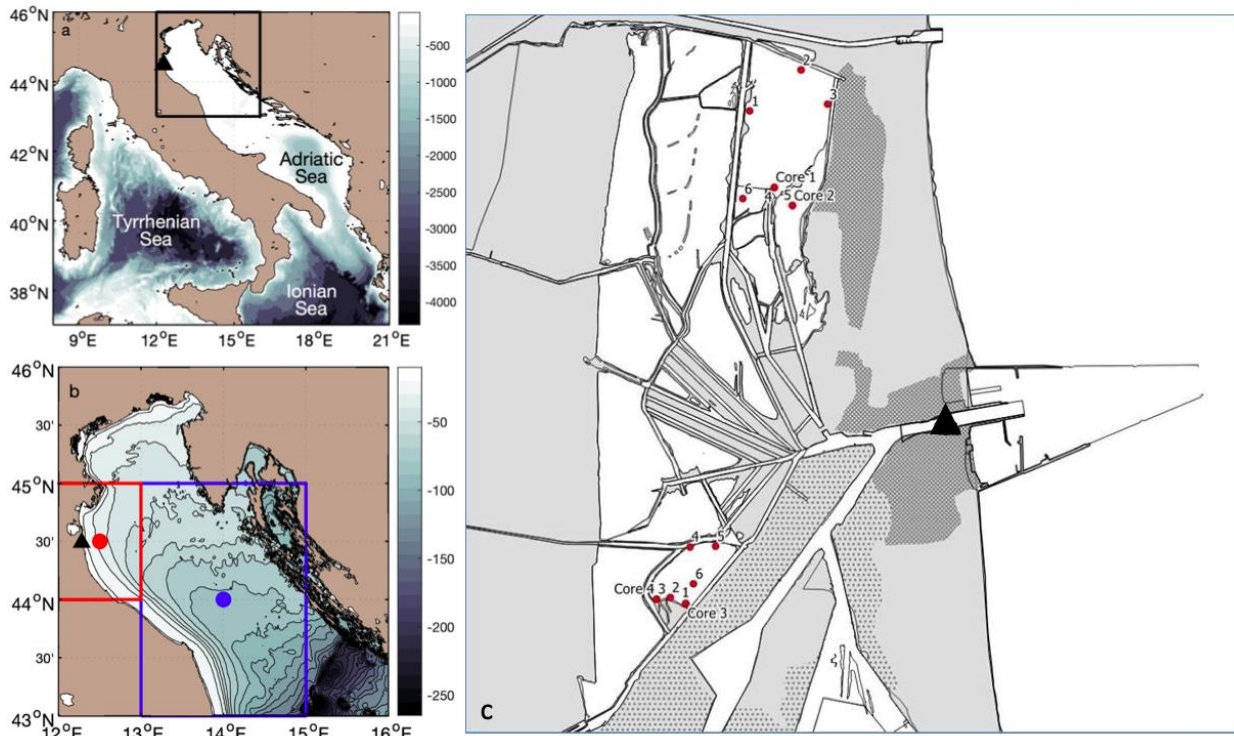


Figure 1. (a) Overview map and location of the study (black triangle) area overlain on the bathymetry from GEBCO Compilation Group (2019) GEBCO 2019 Grid. (b) Zoom in the Northern Adriatic Sea, corresponding to the black square in the left panel, and relative bathymetry. The red square and dot correspond to the HadISST and COBE-SST mesh grid, while the blue square and dot correspond to the ERSST mesh grid. (c) Map of the Pialassa Baiona coastal lagoon and station locations of surface sediment samples and core samples in the saltmarsh habitat and impacted habitat. The black triangle indicates the Porto Corsini/Marina di Ravenna tide gauge location.

2.2 Sampling

Sampling took place in April and July 2008 at two contrasting habitats within Pialassa Baiona coastal lagoon: a) a saltmarsh habitat located northward in a relatively flat area with the presence of saltmarsh vegetation, and b) a human impacted habitat located southward close to anthropogenic source inputs (Figure 1). At each habitat, sediment cores were sampled at two random locations by inserting one cylindrical Plexiglas hand corer (5 cm diameter, 50 cm long) into the sediment to a depth of 20–25 cm. The cores were extruded in the field, sectioned into 1–2 cm intervals, and analyzed for total organic carbon (OC), total nitrogen (TN), carbon isotope ratio ($\delta^{13}\text{C}$) and dry bulk density (upper 20–25 cm). In addition, a composite of two sediment cores with no evidence of physical disturbance was taken at each habitat to reconstruct the chronology of sediment accumulation. These two cores were sampled within a 1-m² area within each habitat to attenuate variation due to spatial heterogeneity (Bernal and Mitsch, 2012). Composite core intervals were bulked together to provide satisfactory amounts, which were needed for effective ²¹⁰Pb dating and dry bulk density.

Additionally, a total of 12 surface sediment samples (0–5 cm) were collected with a stainless-steel grab sampler at six stations in the saltmarsh habitat and six stations at the impacted habitat for organic matter description (OC, TN and $\delta^{13}\text{C}$) (Figure 1).

2.3 Laboratory analysis

Freeze-dried sediment samples were treated with HCl (1.5 M) to remove inorganic carbon (carbonate). Aliquots of approximately 20 mg were added into silver capsules for the measurement of total organic carbon (OC), total nitrogen (TN), and carbon isotopes ($\delta^{13}\text{C} = [({}^{13}\text{C}/{}^{12}\text{C})_{\text{sample}}/({}^{13}\text{C}/{}^{12}\text{C})_{\text{standard}} - 1] \times 1,000$) using a FINNIGAN Delta Plus XP mass spectrometer directly coupled to Thermo Fisher FLASH 2000 CHNS Elemental Analyzer. OC and TN content were expressed as the weight percentage of dried sediment, and carbon isotope results were reported in the standard delta notation (‰) with respect to the international standard Vienna Pee Dee Belemnite (VPDB). Standard deviation based on replicates of reference standard (IAEA-CH7) was $< \pm 0.2\text{‰}$.

For radiometric analysis, packed sediments (~40–50 g) were set aside for at least 21 days to allow ${}^{222}\text{Rn}$ ingrowth and to establish secular equilibrium between ${}^{226}\text{Ra}$ and its granddaughter ${}^{214}\text{Pb}$. The ${}^{210}\text{Pb}$ and ${}^{226}\text{Ra}$ activities were measured by γ -ray spectrometry, and the excess ${}^{210}\text{Pb}$ activity was estimated by subtracting the ${}^{214}\text{Pb}$ from the total ${}^{210}\text{Pb}$ activity with self-absorption corrections (Cutshall et al., 1983). Detailed information on the radiometric technique is provided in Guerra et al. (2019). The excess ${}^{210}\text{Pb}$ was used to determine ages of sediment intervals using the ‘simple’ Constant Flux Constant Sedimentation model (CF-CS; Sanchez-Cabeza and Ruiz-Fernandez, 2012). Sediment accumulation rates (SAR, cm yr^{-1}) and mass accumulation rates (MAR, $\text{g cm}^{-2} \text{yr}^{-1}$) were estimated from the profile of excess ${}^{210}\text{Pb}$ activity concentration versus sediment depth (cm) and mass sediment depth (calculated using bulk density). An aliquot of sediment was dried at 60°C to measure the water content and to calculate bulk density assuming a sediment density of 2.65 g cm^{-3} and a water density of 1.034 g cm^{-3} .

The OC accumulation rate ($\text{g OC m}^{-2} \text{yr}^{-1}$) was estimated by multiplying the OC fraction ($\text{\%OC}/100$) in each core interval by the MAR ($\text{g m}^{-2} \text{yr}^{-1}$). Total C stock (g C m^{-2}) was calculated by integrating OC accumulation rates over the c.a. last century (1900–2008).

Total mercury (THg) was determined with a Perkin Elmer Optima 3200 XL inductively coupled plasma-optical emission spectrometer coupled to a FIAS 400 hydride generation system following the US EPA method 6010C (USEPA, 2000). Sediments were digested with concentrated nitric acid (HNO_3 , Suprapur, Merck; 65%) and hydrochloric acid (HCl, Suprapur, Merck; 37%). QA/QC for THg analysis was carried out using sample replicates, method blanks, and certified reference materials (CRMs, Marine Sediment PACS-2 and MESS-3 from NIST, USA). The accuracy of the instrument was consistently within their certified range (3.04 ± 0.20 , and $0.91 \pm 0.009 \text{ } \mu\text{g g}^{-1}$, respectively), and all measured samples showed less than 10% deviation from CRMs.

2.4 Climatic data

Existing Sea Surface Temperature (SST) and Sea Level (SL) climatic data sets have been analyzed in order to assess the main climatic characteristics and changes happened after 1900 in the study region and relate them to the accumulation and variations in the pool of blue carbon through multivariate analysis.

One of the longest SL time series (1987–2014) available within the Northern Adriatic region has been recorded at the Porto Corsini/Marina di Ravenna tide gauge station (44.49°N , 12.28°E), located nearby the entrance to the Piallassa Baiona (Figure 1c), along the main shipway channel connected to the sea (Cerenzia et al., 2016; Zerbini et al., 2017). The Relative SL (RSL) data, given by the combined effect of vertical land movements and SL, used in the climatic analysis are the annual means (Figure 3) over the time period 1900–2014 analyzed and provided by Cerenzia et al., (2016). They derive from different data sources and have been properly homogenized relying upon the information on benchmarks and data overlaps. The three main RSL data sources are: (1) the Permanent Service for Mean Sea Level (PSMSL) archive for the time period 1897–1922;

(2) the Hydrology Annals of Bologna for the 1934–1979 time period and (3) the Institute for Environmental Protection and Research (ISPRA) for the period 1980–2014.

The centennial RSL change presents anomalous rate of increase ($8.5 \pm 0.2 \text{ mm yr}^{-1}$) within the Northern Adriatic and the Northern Mediterranean region. Cerenzia et al., (2016) found out that land subsidence represents the main cause of local RSL rise measured by the tide gauge due to a total land lowering from 1.8 to 0.2 m above sea level in the time period 1897–2014.

The rate of absolute SL change during the last decades, obtained by subtracting the rate of land subsidence from the rate of RSL change, is equal to $2.2 \pm 1.3 \text{ mm yr}^{-1}$, consistently with other tide gauge observations nearby (i.e. Venice, Trieste) and the estimate of 3 mm yr^{-1} computed from altimetry in the region from Bonaduce et al., (2016). Another long-term sea level trend estimate from Bruni et al., (2019) at Porto Corsini/Marina di Ravenna tide gauge gives a value of $1.25 \pm 0.16 \text{ mm yr}^{-1}$ over the time period 1873–2016.

Sea Surface Temperature gridded data sets covering the time period 1900–2018 have been retrieved from three global climatic data products with the aim to obtain a robust estimate of the long term temperature variation of the sea water entering and exiting the coastal lagoon under investigation to be used for the multivariate analysis. Three global monthly SST data sets have been considered: (1) the HadISST from Met Office Hadley Centre for Climate Prediction and Research; (2) the ERSST from NOAA National Centers for Environmental Information and (3) the COBE-SST from the Japan Meteorological Agency (JMA). These products might not fully capture regional details, as highlighted by (Li et al., 2019) thus a fourth, most recent and accurate regional satellite-based data set (Pisano et al., 2016) distributed by the Copernicus Marine Environment Monitoring Service (CMEMS) has been considered as the reference to validate the three global products and select the most representative of the area for the successive analysis. The CMEMS SST is the longest satellite SST time series (1982–2018) at 4 km resolution for the Mediterranean Basin and it is spatially complete, accurate, homogeneous and stable, i.e. free of spurious trends, all essential characteristics for products usability for climate applications. The main characteristics of the considered SST data sets have been summarized in Table 1 and detailed in the supporting information.

Table 1. Main characteristics of the SST data sets used and displayed in Figure 3.

SST Product	Reference	Time Coverage	Time resolution	Space resolution [degrees]
HadISST	(Rayner et al., 2003)	1882 onwards	monthly	1x1
ERSST	(Huang et al., 2017)	1854 onwards	monthly	2x2
COBE-SST	(Ishii et al., 2005)	1891 onwards	monthly	1x1
CMEMS REP L4	(Pisano et al., 2016)	1981–2018	daily mean	0.0417x0.0417

The closest sea grid point to the study area (Figure 1) has been selected from each global data set to first inspect the time series. HadISST and COBE-SST sea grid points are coincident and the grid cell includes the coastal strip [12–13°E; 44–45°N] up to approximately the 40 m bathymetric (red square in Figure 1b). The ERSST grid cell (blue square in Figure 1b) is wider than the others, due to its coarser resolution and it encloses a central area of the Northern Adriatic sea [13–15°E; 43–45°N], which does not include the considered coastal strip. The CMEMS SST sea grid points falling inside the HadISST and COBE-SST grid cell have been selected from daily fields to compute monthly averages and represent the best SST estimate of the coastal area under investigation.

A preliminary consistency analysis of the monthly SST time series has been conducted and is provided in the supporting information (Figure S1). The outcome suggests a consistency among the centennial SST data sets and CMEMS SST and their potential usability to characterize the long term SST variations of the domain under investigation.

SST annual averages and relative anomalies have been computed subtracting from each time series its relative time average (i.e. 1900–2014 for the centennial datasets, 1982–2018 for CMEMS SST). The resulting annual SST anomalies are displayed in Figure 3 together with the annual RSL values from Cerenzia et al., (2016).

The correlation between the annual anomalies from the centennial SST datasets and the reference CMEMS SST have been computed over the time period 1982–2018 in order to select the most correlated one to be used in the successive multivariate analysis. The correlation is highly significant and it is equal to 0.85 for COBE-SST, 0.86 for HadISST and 0.88 for ERSST. The correlation among all four SST annual anomalies and the annual RSL has been computed too and it results minimum for COBE-SST (0.42) and maximum for CMEMS SST (0.63), while it is equal to 0.54 and 0.55 for HadISST and ERSST respectively.

The analysis of the available centennial datasets suggested to select the ERSST annual averages for the successive analyses on sediment OM data due to its highest correlation with both CMEMS SST and RSL, even if its grid mesh does not include the area under investigation (Figure 1). The reason could depend from the largest availability of in situ data within the ERSST widest grid box on which the gridded data product relies, making its estimate more robust.

2.5 Multivariate statistical analysis

The constrained ordination method of redundancy Analysis (RDA, Legendre and Legendre, 2012) was employed to evaluate the relation between sediment OM data (response variables) and the climatic ERSST and RSL data (explanatory variables). The analysis was carried out using the R package VEGAN. The overall significance of the RDA model and of the single explanatory variables was tested using the `permutest()` and `anova.rda()` functions of VEGAN, which are based on permutation methods. Due to the availability of RSL and SST data, the period prior to 1897 and the periods 1922–1935, 1941–1945 and 1990–1994 could not be included in the RDA. Organic matter data and climatic variables were further averaged during the period comprised by each core section. Four distinct RDA were performed, one for each core. Correlation between single explanatory and response variables and between variables and RDA axes was quantified and tested using the Pearson's *r* coefficient.

3 Results

3.1 ^{210}Pb geochronology

The excess ^{210}Pb profile showed a decreasing trend with depth, and activities ranged from 0.8 to 33.5 Bq kg^{-1} in the saltmarsh habitat, and from 14.4 to 58.7 Bq kg^{-1} in the impacted habitat, with an overall mean value of 23.1 ± 17.0 Bq kg^{-1} . The period covered by each sedimentary core is longer than 100 years, and the time span from 1900 to 2008 (Figure S2). The average sediment accumulation rates (SAR, 0.15 ± 0.06 cm yr^{-1} and 0.21 ± 0.06 cm yr^{-1}) and mass accumulation rate (MAR, 0.16 ± 0.07 and 0.17 ± 0.06 g cm^{-2} yr^{-1}) were comparable in the saltmarsh habitat and the impacted habitat, respectively.

The modern (last decades to century) sediment chronologies estimated from the excess ^{210}Pb CF-CS model were compared to the profiles of THg concentration (Table S1). Generally, the THg concentration increased from low near background regional values in the bottom layers to the maximum values in the sub-surface layers followed by a rapid decrease towards the upper layers (Figure S3). THg concentrations were <0.12 mg kg^{-1} up to 9 cm and 12 cm depth in the saltmarsh and impacted habitat cores, respectively (1950 ± 4

and 1953±4). The onset of THg was recorded in the middle layers of the impacted habitat core and the saltmarsh habitat core (11 and 8 cm, respectively; year 1957±3). The highest values of THg were found in the middle layers of the impacted habitat core (6–9 cm, from year 1967±3 to 1977±2), where the concentrations of THg were always higher than 20 mg kg⁻¹, with a maximum value of 25.3 mg kg⁻¹ (7 cm, year 1977±2).

The mean differences of geochronology (1958 and 1978) between the results from excess ²¹⁰Pb and THg are 1.0±0.7 years and 2.5±5.1 years, respectively. The relatively consistent estimation results of ²¹⁰Pb and THg indicated the geochronology was valid, although the biggest uncertainty (6.1 years) was found in the saltmarsh core (Table S1).

3.2 Elemental and isotopic composition of sedimentary organic carbon

The OC content, C/N ratio and δ¹³C in the surface sediment ranged from 1.04 to 2.62%, from 6.92 to 10.99, and from -21.87 to -18.21‰ in the saltmarsh habitat, and from 0.68 to 2.62%, from 6.98 to 11.1, and from -26.81 to -20.91‰ in the impacted habitat (Table S2). δ¹³C values were significantly enriched in the saltmarsh habitat when compared to the impacted habitat (-20.06±1.31 ‰ and -23.34±2.00 ‰; p < 0.007), whereas C/N ratios did not show any significant difference between the surface sediments from the saltmarsh and impacted habitats.

On the basis of ²¹⁰Pb chronology and historical Hg concentrations, three time periods were defined: before 1957, 1957–1977, and after 1977.

In the saltmarsh habitat cores, OC content ranged from 0.38 to 1.38% and from 0.22 to 1.31% (core 1 and core 2), and from 2.06 to 3.74% and from 2.53 to 3.94% in the impacted habitat cores (core 3 and core 4); C/N ratio varied from 6.62 to 12.73 and from 6.32 to 15.80 in the saltmarsh habitat (core 1 and core 2), and from 6.64 to 10.56 and 6.74 to 11.33 in the impacted habitat (core 3 and core 4). The range of variation of δ¹³C was -22.19 to -19.19 ‰ and -20.25 to -17.74 ‰ in the saltmarsh habitat (core 1 and core 2, respectively), and -26.26 to -22.09 ‰ and -27.21 to -22.76 ‰ in the impacted habitat (core 3 and core 4, respectively) (Figure 2 and Table S3).

In the saltmarsh habitat, δ¹³C values were slightly but significantly depleted in the sediments deposited before the 1950s than in recent years (1980s–2008) (-21.15±0.60 ‰ and -19.41±0.21 ‰, p < 0.001) in core 1; conversely, core 2 did not display any significant difference over time (-19.12±0.55 ‰ and -19.18±1.03 ‰, respectively). In the impacted habitat, δ¹³C values in recent sediments (-22.54±0.39 ‰ in core 3 and -23.61±0.62 ‰ in core 4) displayed a marked and significant enrichment in comparison to the 1960s–1970s intermediate layers (-24.82±0.69 ‰, p < 0.001 in core 3, and -25.84±1.43 ‰, p = 0.003 in core 4) and older layers deposited prior to the 1950s (-25.49±0.73 ‰, p < 0.001 in core 3 and -25.62±0.77 ‰, p = 0.002 in core 4).

In the saltmarsh habitat, C/N ratios did not display any significant increase from recent sediments (1980s–2008) to the older layers deposited prior to the 1950s in core 1 (7.98±1.20 and 9.04±1.56) and in core 2 (8.38±1.05 and 9.71±3.47). In the impacted habitat, C/N ratios slightly but significantly increased from recent sediments (1980s–2008) to the older layers deposited prior to the 1950s both in core 3 (8.11±0.61 and 9.44±1.20, respectively; p = 0.038) and core 4 (7.85±0.70 and 9.41±0.79, respectively; p = 0.004).

Accumulation rates of OC ranged from 8.0 to 14.6 g m⁻² yr⁻¹ and 4.5 to 15.5 g m⁻² yr⁻¹ in the saltmarsh habitat (core 1 and core 2), and from 32.4 to 57.6 g m⁻² yr⁻¹, and 22.2 to 54.7 g m⁻² yr⁻¹ in the impacted habitat (core 3 and core 4). In the saltmarsh habitat, OC accumulation rates were slightly but significantly higher in intermediate and recent sediments (1960s–2008) than in the older layers deposited prior to the 1950s both in core 1 (12.97±1.35 g m⁻² yr⁻¹ and 9.35±1.23 g m⁻² yr⁻¹, respectively; p = 0.003) and in core 2 (13.79±1.25 and 8.40±3.69 g m⁻² yr⁻¹, respectively; p = 0.047). In the impacted habitat, there were no significant differences in OC accumulation rates between the older, intermediate and recent layers in core 3 (44.82±8.30, 39.04±4.48, and 41.63±4.68 g m⁻² yr⁻¹, respectively). In core 4, OC accumulation rates were significantly higher in the older

layers deposited prior to the 1950s than in intermediate (1960s–1970s) and recent sediments (1980s–2008) (43.57 ± 7.20 , 24.51 ± 2.11 , and 33.78 ± 8.20 g m⁻² yr⁻¹; $p < 0.001$ and $p = 0.034$, respectively) (Figure 2).

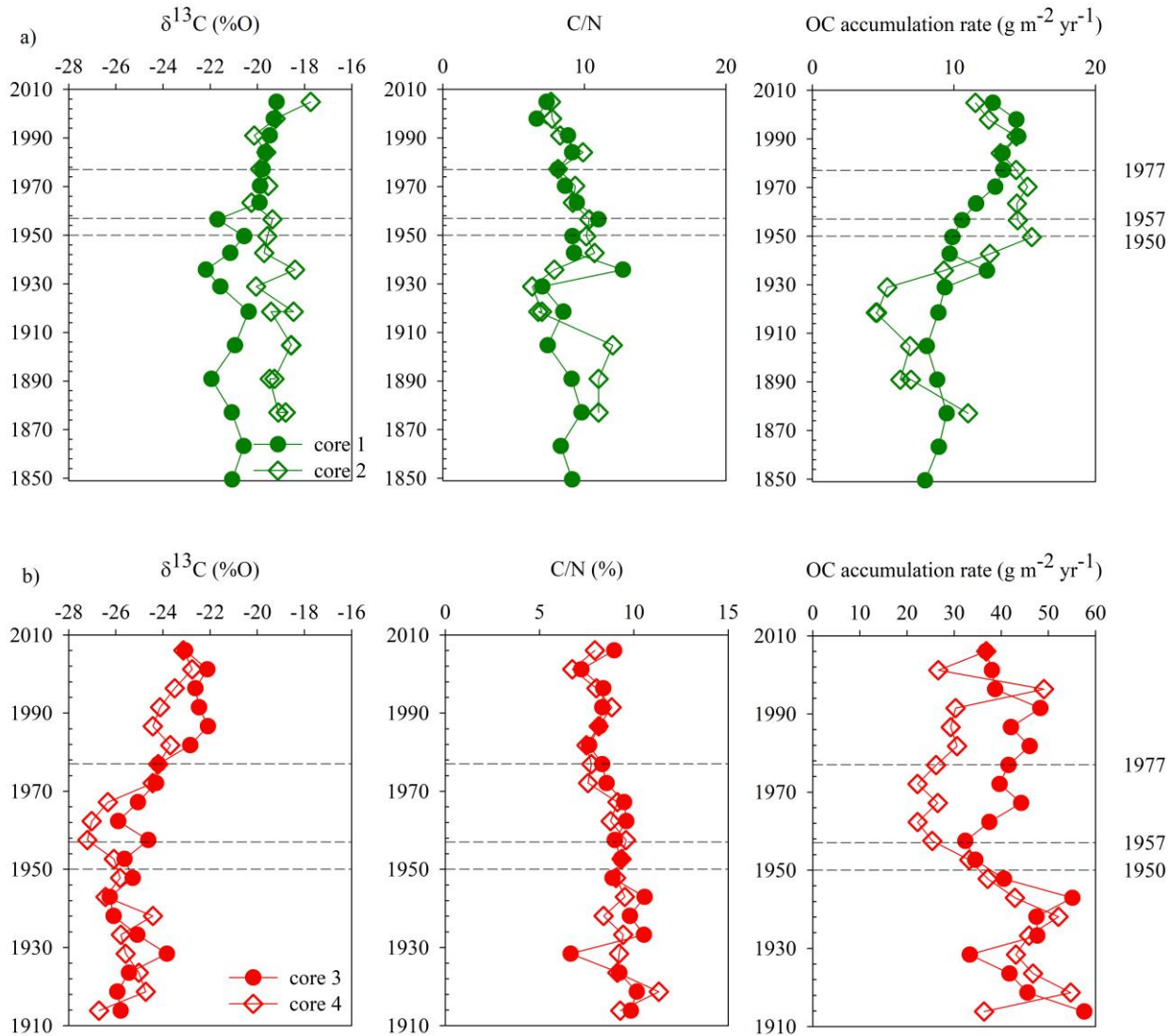


Figure 2. Vertical distributions of stable carbon isotope ($\delta^{13}\text{C}$), C/N ratio, and OC accumulation rates in recent deposited sediments (1978–2008), intermediate layers (1957–1977) and older layers (deposited prior to the 1950s) within Pialassa Baiona coastal lagoon from a) the saltmarsh habitat and b) the impacted habitat.

3.3 Climatic site characterization

The RSL time series over the considered 1900–2014 time period (Figure 3) is characterized by a total variation of 98.4 cm with a continuous rise starting from the '40s until present, characterized by a steep increase from the '40s to the '70s. The RSL trend is equal to 8.7 ± 0.5 mm yr⁻¹, which reduces to 7.0 ± 1.4 mm yr⁻¹ in the most recent 1982–2014 time period.

Cerenzia et al. (2016) highlight the dominant role of land movement on the RSL rise in this area and report a natural subsidence trend of about 6 mm yr⁻¹ in the time period 1897–1950 followed by a noticeable increase over the period 1950–1970, corresponding to a rate of about 24 mm yr⁻¹. Bruni et al., (2019) also

describe a moderate RSL rising tendency before 1940, a steep increase from the '40 to the 70s and its significant reduction after 1980.

The effect of human activities in the study area is thus predominant in the area with respect to SL rise, especially from the '40s until the 1970s, when subsidence due to groundwater withdrawal from surface aquifers and gas extraction have superimposed onto the regional tectonic component. As a consequence the sea water ingress within the coastal lagoon increased in time, modulated by tidal flow.

SST annual anomalies (Figure 3) appear mainly negative during the first two decades (1900–1920), they fluctuate approximately between $\pm 0.5^\circ\text{C}$ from 1920 to 1970 with the most pronounced ERSST negative anomalies during the 1940s. Between 1970 and the early 1980s the anomalies become mainly negative, but since that period the SST start a continuous warming tendency characterized by always positive anomalies approximately after the year 2000. Consistently, CMEMS SST anomalies appear mostly negative before the year 2000 and positive afterwards.

The total ERSST variation over the 1900–2018 time period is equal to 1.0°C with a constant increase of $0.009 \pm 0.002^\circ\text{C yr}^{-1}$. SST increases mainly from the early 1980s, thus the linear trends in the time period 1982–2018 is provided too: ERSST trend is equal to $0.034 \pm 0.010^\circ\text{C yr}^{-1}$ and CMEMS SST is $0.044 \pm 0.009^\circ\text{C yr}^{-1}$. The latest value is comprised within the estimates by Pisano et al. (2020) for the whole Mediterranean Sea ($0.041 \pm 0.006^\circ\text{C yr}^{-1}$) and the Adriatic Sea ($0.045 \pm 0.007^\circ\text{C yr}^{-1}$).

Starting from the '80s the effect of subsidence slowed down due to mitigation effects but SL rise continues at a rate of about $2.2 \text{ mm} \pm 1.3 \text{ mm yr}^{-1}$ (1990–2011; Cerenzia et al., 2016) and contemporary SST increases with unprecedented rate.

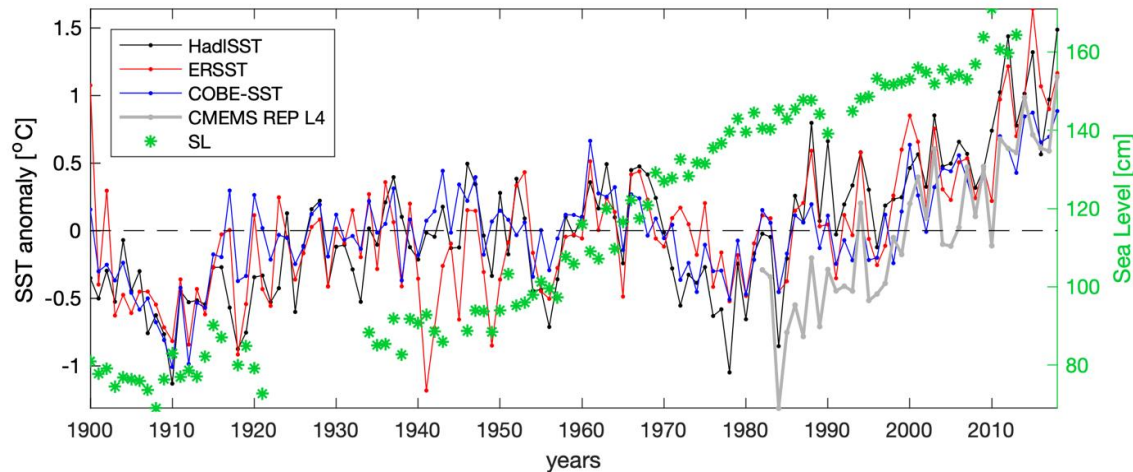


Figure 3. Annual mean time series of RSL (1900–2014) at the Porto Corsini tide gauge (Cerenzia et al., 2016) and SST anomalies (1900–2018) from three global data products (HadISST, ERSST, COBE-SST) and a reference regional data product from CMEMS (1982–2018). SST data products characteristics are summarized in Table 1.

4 Discussion

4.1 Organic matter sources

Coastal wetlands ecosystems represent the convergence of terrestrial and marine systems, hence stable carbon isotopes ($\delta^{13}\text{C}$) and the ratio of organic carbon to total nitrogen (C/N) values of these environments will

depend on the relative contributions of carbon derived from saltmarsh vascular vegetation, phytoplankton, seagrasses, algae, and particulate organic carbon (POC) transported by rivers and tides (Lamb et al., 2006). $\delta^{13}\text{C}$ and C/N together are able to differentiate sources of OM that accumulate in coastal depositional records, specifically between C3 and C4 vegetation and freshwater and marine organic matter (Kemp et al., 2012, 2010; Khan et al., 2015b; Oreska et al., 2018).

In order to estimate the relative contribution of different OM sources supplied to and stored in sediments of Pialassa Baiona coastal lagoon, we combined $\delta^{13}\text{C}$ and C/N values of saltmarsh habitat cores ($n = 38$), impacted habitat cores ($n = 40$), and saltmarsh and impacted habitat surface sediments ($n = 6$ and $n = 6$, respectively) (Table S2 and S3 in the supporting information). The $\delta^{13}\text{C}$, C/N and OC composition of modern sediments (c.a. last century) in the two habitats of the lagoon generally indicated their spatial and temporal distribution (Figure 3 and Figure 4). Despite this distinct pattern, a significant positive correlation ($p < 0.0001$, $n = 88$) between contents of OC and TN was observed with a strong linear relationship and a close to zero intercept ($\text{TN} = 0.01312 \cdot \text{OC} + 0.0048$; $r^2 = 0.9459$). This clearly indicates common OM sources in the two habitats, and suggests that nitrogen in sediments was predominantly bound to sedimentary OC, thus is probably in an organic form (Hedges et al., 1986).

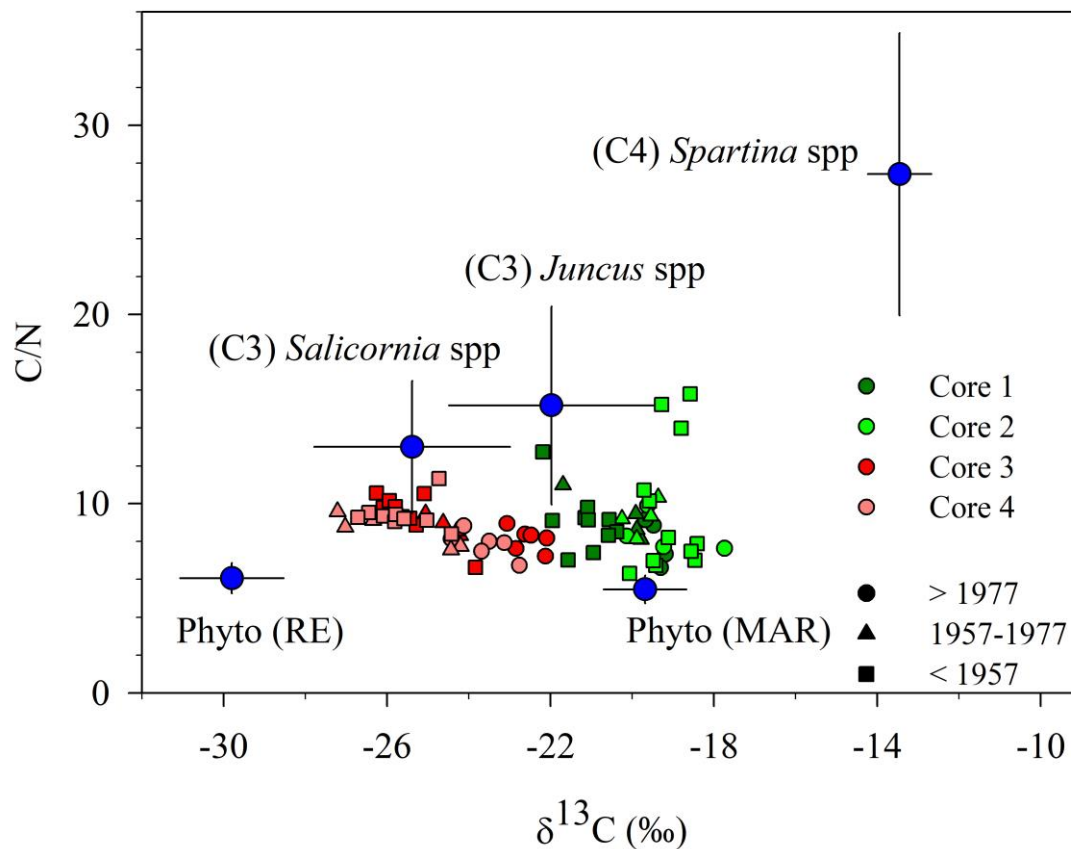


Figure 4. Plot of the stable OC isotopic compositions ($\delta^{13}\text{C}$) and atomic carbon: nitrogen ratios (C/N) in recent deposited sediments (1980s–2008), intermediate layers (1960s–1980s) and older layers (prior to the 1950s) from the saltmarsh habitat (core 1 and core 2) and the impacted habitat (core 3 and core 4) within Pialassa Baiona coastal lagoon (Gebrehiwet et al., 2008; Guerra et al., 2013; Hughes and Sherr, 1983; Kelleway et al., 2017; Kemp et al., 2012, 2010; Lamb et al., 2006). The compositions of five possible OM sources: marsh (C3) vascular vegetation (*Juncus* spp. and *Salicornia* spp.), marsh (C4) *Spartina* spp., riverine-estuarine phytoplankton (Phyto (RE)), marine phytoplankton (Phyto (MAR)), are plotted to show the relative contribution of allochthonous and autochthonous OM.

The C/N ratio has been used as effective index to characterize predominant sources of OM in coastal marshes and lagoons (Cloern et al., 2002; Khan et al., 2015b; Meyers, 1994; Wilson, 2017). Previous studies reported C/N ratios for C3 and C4 saltmarsh vegetation OM that are typically >12, whereas values for marine OM are <8; intermediate values are thought to be mixed. C/N values varies little between the saltmarsh and impacted habitats sediments and is consistently <10 with very few exceptions (Figure 3 and Figure 4). The C/N ratio indicates that the sedimentary OM in the saltmarsh and impacted habitats had a mixed source of freshwater- and marine-derived OM, and the freshwater source of OM was mainly distributed in the impacted habitat, while the saltmarsh habitat was dominated by marine source OM. Despite the overall low C/N values, still a slight but consistent increasing trend from recent layers (1980s–2008) to the pre 1950s sediments can be identified in the impacted habitat (Figure 3b). Significant changes in C/N typically occur in early stages of OM microbial degradation (Dai et al., 2005), and this may have resulted in in situ lower surface sediment C/N compared to the older deposited layers.

Carbon isotope signatures clearly allocated sediments in two distinct groups, with enriched $\delta^{13}\text{C}$ values found in the saltmarsh habitat, and relatively depleted $\delta^{13}\text{C}$ values in the impacted habitat lying between values for corresponding marine and riverine-estuarine end-members (Figure 4). These $\delta^{13}\text{C}$ values infer a change in OM sources from the saltmarsh to the impacted habitat, and reflect contributions from mixed sources of sedimentary OM suggesting that allochthonous freshwater/riverine sources may have a local influence in the impacted habitat, while allochthonous marine sources seem to predominate in the saltmarsh habitat. The $\delta^{13}\text{C}$ values of the saltmarsh and impacted habitats partially overlay the isotopic signatures of saltmarsh vegetation of a C3 photosynthetic pathway origin, which have a $\delta^{13}\text{C}$ extent of -29.88 to -20.2 ‰ and a C/N of 9.70 to >20 (Khan et al., 2015a; Lamb et al., 2006). So far C4 saltmarsh plants are either absent or have been recently introduced in North European saltmarshes (Preston et al., 2002); to our knowledge there is no record of C4 *Spartina* spp. presence in Pialassa Baiona coastal lagoon in the scientific and grey literature.

Locally produced (autochthonous) biomass is generally considered to contribute significantly to the total OC pool in saltmarsh sediments (Chen et al., 2016). Among saltmarsh vascular vegetation, species of the genus *Juncus* is the prevailing C3 high marsh plant of Pialassa Baiona coastal lagoon (Merloni and Piccoli, 1999). Allochthonous sources are the marine-derived OM, imported through tidal flow from the main channel connecting to the sea, and the riverine-estuarine derived OM, brought in by the main inflow channel located on the southern edge of the lagoon, while *Juncus*-derived OM is likely produced in situ, and thus represents an autochthonous source in the sedimentary OM of the saltmarsh and impacted habitats.

To estimate the proportional contribution of the different sources to the sediment OM in each of the cores we applied MixSIAR, a Bayesian mixing model framework implemented as an open-source R package (Stock et al, 2018). MixSIAR, and mixing models in general, require tracer data that characterize the chemical or physical traits of both the sources and the mixtures; these traits are assumed to predictably transfer from sources to mixtures through a mixing process. In the present application $\delta^{13}\text{C}$ and C/N are used as tracers. Marine phytoplankton, riverine/estuarine phytoplankton and *Juncus* spp. are considered the potential sources while sediments are the mixtures.

Tracer values for marine phytoplankton ($\delta^{13}\text{C}$: -19.7 ± 1.0 ‰; C/N: 5.5 ± 0.7) and for riverine/estuarine phytoplankton ($\delta^{13}\text{C}$: -29.8 ± 1.3 ‰; C/N: 6.1 ± 0.8) were obtained from Tesi et al. (2007) and Guerra et al. (2013). Values for *Juncus* spp. ($\delta^{13}\text{C}$: -22.0 ± 2.5 ‰; C/N: 15.2 ± 5.2) were obtained from Gebrehiwet et al. (2008) and Kelleway et al. (2017).

Regardless of the tracers or mixing system considered, all mixing model applications are rooted in the same fundamental mixing equation:

$$Y_j = \sum_k p_k \mu_{jk}^s$$

where the mixture tracer value, Y_j , for each of j tracers is equal to the sum of the k source tracer means, μ_{jk}^s , multiplied by their proportional contribution to the mixture, p_k .

MixSIAR, through the application of Markov chains Monte Carlo sampling, estimates the probability density function of each pk value, i.e. the proportional contribution of each source to the final mixture. Statistics like mean and standard deviation are then calculated from this estimated distribution. Bayesian mixing models improve upon simpler linear mixing models by explicitly taking into account uncertainty in source values. MixSIAR, in particular, has the ability to incorporate categorical and continuous covariates to explain variability in the mixture proportions and offers several options for error parametrization.

In our analysis, two categorical covariates, core and time period, were taken into account. Error was assumed as “residual only”, in accordance with the indications of Stock et al. (2016) for the application of mixing models to sediment sourcing. The probability density functions of the proportional contributions of the three sources to the sediment OM are shown in Figure S4, separately for each core and time period. Means, standard deviations and percentiles of the same distributions are reported in Table S4.

The marine-derived OM increased from ~50% in the sediments deposited prior to the 1950s to ~70% in recent decades (1980s–2008) in the saltmarsh habitat. Similarly, marine-derived OM increased from ~15% in the sediments deposited prior to the 1950s to ~40% in recent decades (1980s–2008) in the impacted habitat. Riverine-estuarine OM sources accounted for <10% and ~50 % of sedimentary OM in the saltmarsh and impacted habitats over the last century; these sources decreased from ~60% in the pre 1950s period to ~40% in recent decades (1980s–2008), while marine-derived OM contribution increased simultaneously after the 1950s within the impacted habitat. These findings indicate that the relative contribution of marine-derived OM to recent sediments are larger than in sediments deposited prior to the 1950s, and this contribution is by ~30% higher in saltmarsh habitat sediments than in the impacted habitat. The explanation of the increasing contribution of tidally-derived OM is related to an increasing inflow of sea water into the lagoon over the last decades supported by the positive RSL trend recorded by the tide gauge.

$\delta^{13}\text{C}$ signatures and C/N showed that an important fraction of sedimentary OM stored in sediment within Pialassa Baiona coastal lagoon is derived from the autochthonous C3 saltmarsh plant *Juncus* spp. Overall, this fraction is slightly higher within the saltmarsh habitat, where C3 saltmarsh vegetation-derived OM comprised on average ~33% of the OM sedimentary pool when compared to the ~26% fraction in the impacted habitat sediments. The mixing model evidenced a change in C3 saltmarsh vegetation OM contribution from ~40% to ~20% in the sediment records from the saltmarsh habitat in the pre 1950s period to recent decades (1980s–2008) concomitant with the increased marine-derived OM contribution from the 1950s forward, and from ~30 to ~25% in the impacted habitat, respectively. The most plausible explanation is that $\delta^{13}\text{C}$, OC and C/N geochemistry of sediments varied in relation to mixing of the autochthonous OM from in situ C3 saltmarsh vegetation with an increased contribution from allochthonous particulate OM from marine sources, which in turn, is controlled primarily by tidal inundation.

4.2 Relationship between organic matter and climatic variables

Redundancy analysis (RDA) was employed to determine if the observed spatial and temporal variability in sedimentary OM data was related to regional climatic changes (Figure 5). RDA model, tested by using the permutest procedure, is highly significant for all the cores except core 2 (Figure 5b). RDA axes account from 38% (core 4) to 66% (core 1) of the total variation, thus a large fraction of the differences in sediment OM properties among core sections is not explained by the RSL and SST (Figure 5, Table S5). The first axis (RDA1) accounts for 86% (core 3) to 97% (core 1) of the explained variance and was positively correlated with both RSL and SST, although RSL had higher scores on this axis. The second axis (RDA2), which accounts only for 3% to 14% of explained variance, was mostly related to SST. As a consequence, RDA1 can be interpreted as the conjunct variation of RSL and SST, while RDA2 as the fraction of SST variation that is not related to the RSL. However, SST presents little independent explanatory power since its effect, tested using the anova.rda procedure, is in all cases not significant. On the other hand the effect of RSL is highly significant in the three cores (1, 3 and 4) where the RDA model is also highly significant.

Recent years (after 1977) show high values on RDA1 axis (except for core 2 not significant) well separated from older years (before 1957), which are instead characterized by low values. Years 1957–1977 score intermediate values and overlap with both recent and older years. This result reflects the steady rise in RSL and SST changes over the last century (Figure 3): low RDA1 scores before 1957 when both RSL and SST rise are low, intermediate RDA1 scores between 1957 and 1977 when SST is not increasing while RSL rapidly increases mainly under the effect of subsidence, while high RDA1 scores appear after 1977 when both RSL and SST increase.

TN was positively correlated with RDA1 and C/N ratio was negatively correlated with RDA1 in all four cores. On the other hand, the relation between other OM variables and RDA1 was not consistent among the four cores. OC content was positively correlated with RDA1 and TN content in the saltmarsh habitat (cores 1 and 2), while independent from RDA1 in the impacted habitat (cores 3 and 4). Correlation between OC accumulation rate and RDA1 was positive in the saltmarsh habitat, negative in the impacted habitat, although correlation was significant only for cores 1 and 4. $\delta^{13}\text{C}$ was positively correlated with RDA1 in all cores with the exception on core 2 (saltmarsh habitat).

In summary RDA suggested that climatic variables, in particular RSL due to the cumulated effect of land movement and SL rise, influenced OM properties of the sediments. However, the effect of this influence was not consistent between the two habitats, and differences were observed even between cores from the same habitat.

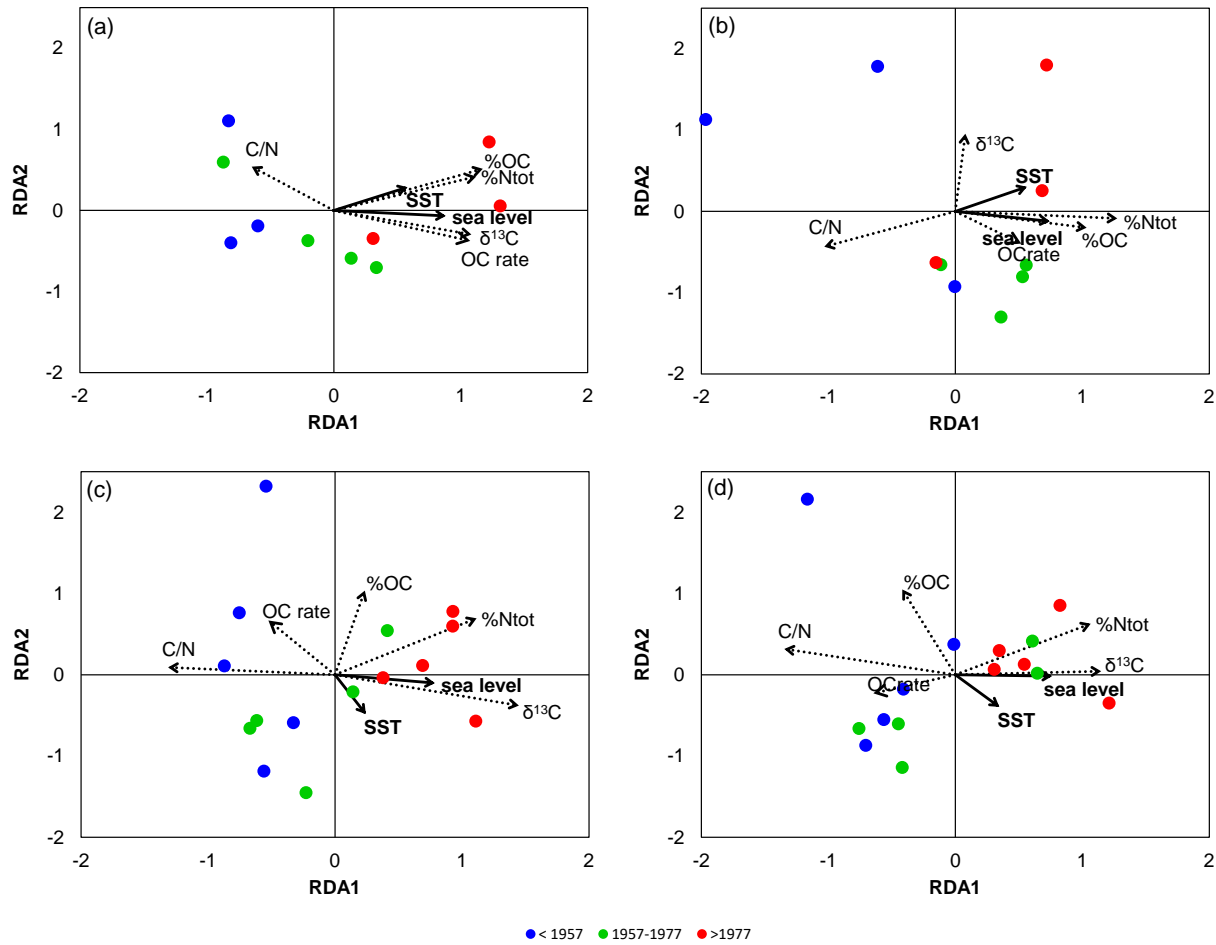


Figure 5. Plot of RDA analysis with response sedimentary OM variables (OC, $\delta^{13}\text{C}$, OC accumulation rates) and explanatory regional climatic variables (RSL and SST): (a) saltmarsh habitat core 1, (b) saltmarsh habitat core 2, (c) impacted habitat core 3 and (d) impacted habitat core 4.

4.3 Organic matter accumulation

Core dating using excess ^{210}Pb confirmed that the sediment bed has accreted due to sediment accumulation in the saltmarsh and impacted habitats over the last century. The differences in geochronology resulting from excess ^{210}Pb and THg can be attributed to complex depositional conditions and biogeochemical cycling with northward migration of Hg causing a delayed THg peak (Covelli et al., 2011; Fabbri et al., 2001; Trombini et al., 2003).

Pb-210 derived SARs are comparable with published accretion rates measured within Oregon tidal saline wetlands ($0.13\text{--}0.29\text{ cm yr}^{-1}$) and Long Island Sound ($0.1\text{--}0.36\text{ cm yr}^{-1}$) (Peck et al., 2020; Hill and Anisfeld, 2015), in microtidal coastal wetlands in the Gulf of California and the Caribbean Sea ($0.04\text{--}0.65\text{ cm yr}^{-1}$) (Ruiz-fernández et al., 2018), and within Venice lagoon under rising sea level, subsidence and increasing frequency of high tides flooding called ‘acqua alta’ ($0.13\text{--}0.44\text{ cm yr}^{-1}$) (Bellucci et al., 2007).

Salt marshes are vulnerable and dynamic systems that can extend landward in response to lateral erosion and accrete vertically by accumulating sufficient materials to compensate for rising sea level (Fagherazzi et al., 2020; Kirwan and Megonigal, 2013; Mariotti and Carr, 2014). Sediment accretion is further promoted by the presence of halophytic plants producing OM, which in turn, can be partially buried favoring net sediment accumulation (Cahoon et al., 2021).

To explore whether Pialassa Baiona lagoon is a net depositional or erosional area, excess ^{210}Pb (Bq cm^{-2}) inventories were estimated at sediment core habitats according to a standard technique (Appleby and Oldfield, 1992), and then compared to the annual atmospheric flux of ^{210}Pb within the latitudinal range 40–50°N ($0.0155 \pm 0.0075 \text{ Bq cm}^{-2} \text{ yr}^{-1}$) (Baskaran, 2011). The mean excess ^{210}Pb inventory for sediment cores at the saltmarsh and impacted habitats is $0.37 \pm 0.27 \text{ Bq cm}^{-2}$, which is lower than the steady state inventory of atmospheric ^{210}Pb ($0.50 \pm 0.24 \text{ Bq cm}^{-2}$) at 40–50°N, and thus the study area is not net depositional.

Averaged SAR over the period ~1900–2010 ($\sim 0.2 \text{ mm yr}^{-1}$) is lower than the local RSL (Cerenzia et al., 2016) increase owing to rapid land subsidence in the 1950–1970, thus indicating the accretion balance is negative and the area is less likely to keep pace with the ongoing SL rise. SL rise scenarios for the year 2100 and relative inundations map for the subsiding Northern Adriatic coastal plain, predict that the coastal area under investigation will be most probably submerged by that time (Antonioli et al., 2017; Mariano et al., 2021).

Although OC accumulation rates averaged over 1900s–2008 are lower than the averages for tidal wetlands soils and salt marsh sediments (Ouyang and Lee, 2014; Wang et al., 2019), they compare well to mean OC rate in modern sediments (ca < 200 yrs) in coastal and inland aquatic ecosystems (Wilkinson et al., 2018). In terms of carbon sequestration, our data suggests that modern sediments in the study area are similar to recently deposited sediments soils in vegetated U.K. salt marsh habitats, as well as salt marshes in the Blackwater, Sheldth, and Guadiana estuaries (Table 2).

The variation of modern OC accumulation rates between the two habitats is remarkable, with the highest values found in the impacted habitat located in the southern lagoon, an area that historically received allochthonous inputs of OC and nutrients from agriculture runoff and wastewater discharges through the main inflow channel draining an intensely cultivated watershed (Giordani et al., 2005). Lower OC values were found in the saltmarsh habitat located in the northern area of the lagoon, distal from the main freshwater and anthropic inputs from inland activities.

Before the 1950s, OC accumulation rates in the impacted habitat were on average higher and negatively correlated with RSL, a trend confirmed as statistically significant for core 3. This could be explained by a decrease in the amount of suspended solids entering the inflow channels from the watershed. While there are no direct measurements available for the time period under consideration, this appears to be in line with the recorded hydrological changes in the area (Buscaroli et al., 2011).

In the nineteenth century, the inland of Pialassa Baiona, which is now entirely reclaimed agricultural land, was predominantly formed up of wetlands, which were connected to the lagoon through channels. The Lamone River, which previously flowed directly into the Adriatic Sea north of Bialassa Baiona, was redirected to flow into the wetlands southwest of the lagoon in 1846, to reclaim agricultural land by filling the wetlands with sediments brought by the river. When the southern wetlands were filled, the river was shifted northward, finally returning to run directly into the Adriatic Sea north of Pialassa Baiona at the turn of the 1950s. As a result of the reclamation process, the lagoon was gradually cut off from the Lamone river's suspended solids flow.

Our hypothesis is that the balance between increasing sedimentation of marine OM (due to RSL rise) and decreasing riverine OM was somewhat negative in the impacted habitat, resulting in a decrease in OC accumulation rate over time. Conversely, the saltmarsh habitat was already excluded from riverine OM contributions at the turn of the nineteenth century, and thus increased sedimentation of marine OM resulted in an increase of OC accumulation rate.

A plausible explanation of the lower OC accumulation rates found in this habitat could be attributed to the predominance of marine phytoplankton sources increasing with RSL rise over the last century with a minor contribution from prevailing C3 saltmarsh vegetation (*Juncus* spp.) as evidenced by the isotopic ^{13}C signals and the MixSIAR model outputs. Marine phytoplankton predominantly consists of macromolecules (pigments, lipids, carbohydrates, and aminoacids) that show a steady decline through the water column into the sediments (Hama et al., 2004; Wakeham et al., 1997) in contrast to intrinsically refractory components (for example lignin and lipid biomarkers) typical of terrestrial angiosperms (Hedges and Mann, 1979), and of marine and wetland

vascular plants including *Juncus* spp (Clifford et al., 1995; Goñi and Thomas, 2000; Klap et al., 2000) that are selectively preserved and accumulated into the sedimentary OC pool of coastal systems (Bianchi et al., 2018; Goñi et al., 1998; Kuzyk et al., 2008; Tanner et al., 2010, 2007).

OC storage and sink capacity of blue carbon ecosystems, including saltmarshes, are impacted at spatial and temporal scales by rapidly changing climate and anthropogenic factors, comprising SL rise, land subsidence, warming, hydrological alterations, and landscape development (Ouyang and Lee, 2014; Tan et al., 2020), and these disturbances resulted in estimated areal losses of $0.2\text{--}7\%$ yr^{-1} at global level (Spivak et al., 2019). Human-caused and climatic changes have also affected the physical environment of the Pialassa Baiona coastal lagoon and its adjacent areas throughout the previous century, particularly in the middle of the 1900s due to the combination of SL rise and fast subsidence rate, resulting in changes in OM accumulation and sources in their sediments.

4.4 Carbon stock

For the 1900–2008 period, the carbon stock estimated from the OC accumulation rates is 1253 ± 12 g C m^{-2} for the saltmarsh habitat, and 4223 ± 40 g C m^{-2} for the impacted habitat. Overall, the estimated amount of OC stored in the two habitats are 1223 ± 24 g C m^{-2} and 3800 ± 20 g C m^{-2} , respectively, of which ~30% are ‘blue carbon’ from in situ C3 saltmarsh vascular vegetation.

Between habitat variability has important implications for attempts to characterize blue carbon based on modern environmental conditions (c.a. last century). Our results suggest that contemporary changes in the RSL and allochthonous sources are likely to affect both C stocks and OC accumulation rates. That is, we found approximately threefold differences in OC accumulation rates and C stock between the saltmarsh habitat and the impacted habitat.

Over a century scale (1900–2008), the total C pool stored in the sediments is 7379 Mg C and 13682 Mg C in the saltmarsh and impacted habitats (589 ha and 324 ha, respectively), totaling 21061 Mg C corresponding to an area of ~ 10 km^2 . According to the global dataset of carbon dioxide emissions (World Bank, 2019), Italy per capita CO_2 emissions went from 2.20 in 1960 to 7.11 tons in 2010. If we consider the averaged country’s national CO_2 emissions per capita over this period (6.40 tons), the estimated C pool stored in the two habitats of the Pialassa Baiona amounted to ~ 77000 Mg CO_2 , or the equivalent of ~ 12000 individuals Italian peoples CO_2 emissions per year. These estimates, while small in terms of national CO_2 emissions (338 Mt CO_2 ; Friedlingstein et al., 2019), are equivalent to 3% of the annual estimated CO_2 emissions from fossil fuel use and combustion of the closest town (Ravenna) (INEMAR-ER, 2019).

4 Conclusions

OC accumulation rates, OC sources and carbon stock were estimated in ^{210}Pb -dated sediment cores from a salt marsh habitat and an impacted habitat within Pialassa Baiona coastal lagoon (Northwestern Adriatic Sea) and paralleled to the temporal trends and changes of SST and RSL in the region. The main findings of this work can be summarized as follows:

- OC accumulation rates showed a contrasting pattern from the 1950s forward, with a significant increasing trend in the saltmarsh habitat and a decreasing trend in the impacted habitat, though significant only in one core;
- Allochthonous marine-derived OM sources showed a consistent increase in 1980s–2008 when compared to the pre 1950s period, parallel with a decreased contribution from autochthonous saltmarsh vegetation (*Juncus* spp.) reflecting an increased sea water inflow into the lagoon as confirmed by RSL positive trend in the area;
- RSL change in the coastal region has been continuously increasing due to the sum of prevailing land subsidence of both natural and anthropogenic origin, and the SL rise. It has been faster during the central decades of the last century, due to the land lowering caused by ground fluids extraction, while it slowed down during the last decades, when subsidence returned to natural rates after the adoption of mitigation measures.
- SST analysis indicate quite stable SST anomalies until the 1960s, negative anomalies in the 1970s, a rapid and continuous SST increase from the eighties with a trend between $0.034 \pm 0.010^{\circ}\text{C yr}^{-1}$ and $0.044 \pm 0.009^{\circ}\text{C yr}^{-1}$.
- A direct effect of SST on sedimentary C has not been detected, however RL rise and SST warming are not independent, due to a contribution of thermal expansion to SL rise ranging from 30% to 40% approximately (Storto et al., 2019).
- Substantial differences have been observed between the saltmarsh and the impacted habitats as to sediment OM composition and sources, rate of accumulation, as well as the temporal trends of these properties and their relationship with climatic variables. Even cores collected few meters apart within the same habitat were appreciably different.
- The estimated total C pool stored in sediments equals ~3% of the annual estimated CO₂ emissions from the nearby town.

The findings of this study confirm that Pialassa Baiona coastal lagoon is an important coastal blue carbon ecosystem in sequestering carbon from atmospheric CO₂ emissions. However, with an annual loss rate of 1–2% between 1980 and 2000 (Spivak et al., 2019) under increasing SL rise and warming, this trend is likely to compromise the capacity of coastal vegetated habitats like Pialassa Baiona coastal lagoon for atmospheric CO₂ uptake and storage, unless proper management and coastal protection is implemented. According to SL rise scenarios and inundations map this coastal vegetated habitat is likely to be submerged by 2100, and thus specific measures should also be implemented to mitigate sea ingression.

There are significant data gaps on carbon stocks in coastal vegetated habitats in the Mediterranean Region, and the characteristic variability of these habitats, is a significant challenge to the goal of a global coastal blue carbon inventory. Further research and mapping of C stock and greenhouse gas fluxes including methane (CH₄) in coastal vegetated habitats at regional and national level is strongly recommended.

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The data sets reported in Table S2 and Table S3 (Supplementary Information) are freely available and accessible at SEANOE, <https://doi.org/10.17882/73534>.

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933 Table 2. Carbon accumulation rates ($\text{g OC m}^{-2} \text{yr}^{-1}$) in coastal wetlands in European Seas and worldwide averages.

934

Area	type	time span	OC accumulation rate ($\text{g m}^{-2} \text{yr}^{-1}$)	Dominant halophyte species/genera	Reference
Global (coastal and inland ecosystems)	sediment	<200 yr	15.6 - 73.2	<i>various</i>	(Wilkinson et al., 2018)
NW Atlantic	tidal wetlands soil	ca 100-150 yrs	172.2±18.1	<i>various</i>	(Ouyang and Lee, 2014)
US tidal wetlands	salt marsh sediments	nr ^a	154.3±8	<i>various</i>	(Wang et al., 2019)
<i>NE Atlantic</i>					
Norwegian Arctic coastal wetlands	low marsh	ca 100 yrs	19-390	<i>Juncus gerardii</i>	(Ward, 2020)
North Sea and Skagerrak Sea	continental shelf	ca 100 yrs	0.02-66.18	nr	(Diesing et al., 2021)
Baltic Sea	continental shelf, estuaries, fjords	nr	13–54	nr	(Winogradow and Pempkowiak, 2014)
Wadden Sea, Germany	semi-natural salt marsh	ca 50 yrs	112-149	<i>Spartina anglica, Salicornia europaea, Puccinellia maritima</i>	(Mueller et al., 2019)
Coastal margin habitats, UK	dry dune habitat	140 yrs	58±26	<i>Carex arenaria, Hypochaeris radicata, Ammophila arenaria</i>	(Jones et al., 2008)
	wet dune habitat	140 yrs	73.0±22	<i>Carex arenaria, Hypochaeris radicata, Ammophila arenaria</i>	(Jones et al., 2008)
	salt marsh	60 yrs	64-219	nr	(Beaumont et al., 2014)
	machair	60 yrs	34.9±15.7	nr	(Beaumont et al., 2014)
Blackwater Estuary, England	salt marsh	ca 10 yrs	6.1-66	<i>Salicornia</i> spp., <i>Spartina</i> spp., <i>Aster tripolium</i> , <i>Puccinellia maritima</i>	(Adams et al., 2012)
Scheldt estuary, Netherlands	salt marsh	<60 yrs	88-151	<i>E. athericus</i>	(Van De Broek et al., 2016)

	salt marsh	<60 yrs	2177 ± 503	<i>Spartina anglica</i>	(Van De Broek et al., 2016)
	salt marsh	<60 yrs	22 ± 10	<i>Mixed vegetation</i>	(Van De Broek et al., 2016)
	embanked salt marsh	<64 yr	86.6± 7.5	nr	(Van de Broek et al., 2019)
	embanked brackish marsh	ca 100 yrs	111.3± 11.4	nr	(Van De (Van de Broek et al., 2019)
Guadiana Estuary, Portugal	salt marsh	13,000 - 6900 cal yrs BP	164-314	nr	(Boski et al., 2021)
Guadiana Estuary, Portugal	salt marsh	after 6900 cal yrs BP	46-52	nr	(Boski et al., 2021)
Mondego Estuary, Portugal	salt marsh	2 yrs	<100	<i>Scirpus maritimus</i>	(Couto et al., 2013)
	salt marsh	2 yrs	100-200	<i>Spartina maritima</i>	(Couto et al., 2013)
	salt marsh	2 yrs	300-400	<i>Zoostera nolti, Spartina maritima</i>	(Couto et al., 2013)
	salt marsh	2 yrs	213-218	<i>Spartina maritima</i>	(Sousa et al., 2010)
Tagus Estuary, North East Atlantic	salt marsh	<100 yrs	150-340	<i>Spartina maritima, Halimione portulacoides</i>	(Caçador et al., 2004)
Ria de Aveiro, Portugal	low marsh	nr	119.5 ± 5.3	<i>Spartina maritima</i>	(Sousa et al., 2017)
Ria de Aveiro, Portugal	mid & high marsh	nr	132.7 ± 5.8	<i>Juncus maritimus</i>	(Sousa et al., 2017)
Ria de Aveiro, Portugal	mid & high marsh	nr	157.3	<i>Halimione portulacoides</i>	(Sousa et al., 2017)
Ria de Aveiro, Portugal	mid & high marsh	nr	152	<i>Sarcoconia perennis</i>	(Sousa et al., 2017)

Odiel marshes	salt marsh	2-3 yrs	104	<i>Spartina maritima</i>	(Curado et al., 2013)
Bay of Cadiz, Spain	tidal coastal wetland	ca 100 yrs	40-107	<i>Spartina maritima</i> , <i>Zoostera nolstei</i>	(Jiménez-Arias et al., 2020)
Palomones Estuary	salt marsh	nr	560	<i>Sarcoconia perennis</i>	(Palomo and Niell, 2009)
<i>Mediterranean Sea</i>					
Venice lagoon, North Adriatic, Italy	salt marsh	nr	132	<i>Salicornia veneta</i> , <i>Spartina maritima</i> , <i>Limonium narbonense</i> , <i>Sarcocornia fruticosa</i> , <i>Juncus maritimus</i>	(Roner et al., 2016)
Rhône River Delta, France	riverine sites	3 yrs	357	<i>Juncus maritimus</i> , <i>Phragmites australis</i> , <i>Scirpus lacustris</i> , <i>Scirpus litoralis</i>	(Hensel et al., 1999)
	marine sites	3 yrs	87.9	<i>Arthrocnemum fruticosum</i> , <i>Halmione portulacoides</i> , <i>Suaeda fruticosa</i> , <i>Limonium vulgare</i>	(Hensel et al., 1999)
	impounded sites	3 yrs	72.1	<i>Juncus maritimus</i> , <i>Arthrocnemum fruticosum</i>	(Hensel et al., 1999)
Ebro River Delta, Spain	tidal marsh	ca 3 yrs	94-126	<i>Phragmites australis</i> , <i>Tipha</i> spp.	(Calvo-Cubero et al., 2014)
	salt marsh	<100 yr	39-293	<i>Sarcoconia fruticosa</i>	(Fennessy et al., 2019)
	brackish mash	<100 yr	32-495	<i>Phragmites australis</i> , <i>Cladium mariscus</i> , <i>Juncus maritimus</i> , <i>Sarcocornia fruticosa</i> , <i>Scirpus maritimus</i> , <i>Spartina versicolor</i> , <i>Paspalum</i> spp.	(Fennessy et al., 2019)