Journal of Marine Systems 158 (2016) 26–33

Contents lists available at ScienceDirect

Journal of Marine Systems

journal homepage: www.elsevier.com/locate/jmarsys

The imprint of anthropogenic CO_2 emissions on Atlantic bluefin tuna otoliths

Igaratza Fraile [a,*], Haritz Arrizabalaga [a], Jeroen Groeneveld [b], Martin Kölling [b], Miguel Neves Santos [c], David Macías [d], Piero Addis [e], David L. Dettman [f], Saadet Karakulak [g], Simeon Deguara [h], Jay R. Rooker [i]

[a] AZTI–Tecnalia, Marine Research Division, Herrera Kaia Portualdea z/g, 20110 Pasaia, Spain
[b] MARUM-Center for Marine Environmental Sciences, University of Bremen, 28359 Bremen, Germany
[c] Instituto Português do Mar e da Atmosfera (IPMA I.P.), Avenida 5 de Outubro s/n, 8700-305 Olhao, Portugal
[d] Spanish Institute of Oceanography, Corazón de María 8, 28002 Madrid, Spain
[e] Department of Life Science and Environment, University of Cagliari, Via Fiorelli 1, 09126 Cagliari, Italy
[f] Environmental Isotope Laboratory, Department of Geosciences, University of Arizona, 1040 E. 4th Street, Tucson, AZ 85721, USA
[g] Faculty of Fisheries, Istanbul University, Ordu Cad. No 200, 34470, Laleli, Istanbul, Turkey
[h] Federation of Maltese Aquaculture Producers, 54 St. Christopher Str., Valletta, VLT 1462, Malta
[i] Department of Marine Biology, Texas A&M University, 1001 Texas Clipper Road, Galveston, TX 77553, USA

ARTICLE INFO

Article history:
Received 9 March 2015
Received in revised form 23 December 2015
Accepted 29 December 2015
Available online 2 February 2016

Keywords:
$\delta^{13}C$ and $\delta^{18}O$
Mediterranean Sea
CO_2 emission
Suess effect

ABSTRACT

Otoliths of Atlantic bluefin tuna (*Thunnus thynnus*) collected from the Mediterranean Sea and North Atlantic Ocean were analyzed to evaluate changes in the seawater isotopic composition over time. We report an annual otolith $\delta^{13}C$ record that documents the magnitude of the $\delta^{13}C$ depletion in the Mediterranean Sea between 1989 and 2010. Atlantic bluefin tuna in our sample ($n = 632$) ranged from 1 to 22 years, and otolith material corresponding to the first year of life (back-calculated birth year) was used to reconstruct seawater isotopic composition. Otolith $\delta^{18}O$ remained relatively stable between 1989 and 2010, whereas a statistically significant decrease in $\delta^{13}C$ was detected across the time interval investigated, with a rate of decline of 0.05‰ yr^{-1} (−0.94‰ depletion throughout the recorded period). The depletion in otolith $\delta^{13}C$ over time was associated with the oceanic uptake of anthropogenically derived CO_2.

1. Introduction

The increase of anthropogenic emission of carbon dioxide (CO_2) and other greenhouse gases to the atmosphere since the industrial revolution is receiving growing attention in recent years (Pachauri and Meyer, 2014). The concentration of CO_2 in the atmosphere has increased almost 40% since preindustrial times in 1850 and continues to increase (Francey et al., 1999; Le Quéré et al., 2014). Carbon and oxygen stable isotope ratios $^{13}C/^{12}C$ and $^{18}O/^{16}O$ (expressed in parts per thousand in relation to Vienna Pee Dee Belemnite, hereafter represented by $\delta^{13}C$ and $\delta^{18}O$ respectively) in the atmosphere and ocean fluctuate temporally due to both natural and anthropogenic processes. Atmospheric $\delta^{13}C$ has decreased by ~2‰, from −6.4‰ in 1850 (Suess, 1955; Friedli et al., 1986) to −8.4‰ in 2012 (Keeling et al., 2005 and http://scrippsco2.ucsd.edu/data/mlo.html). Global $\delta^{13}C$ and CO_2 trends in the atmosphere are anticorrelated at both seasonal and annual scales (Keeling et al., 2011). Plants have less ^{13}C relative to the atmosphere due to fractionation during photosynthesis (Farquhar et al., 1989), and thus, the burning of most fossil fuels releases relatively light carbon isotopes. Hence, CO_2 released to the atmosphere by human activities is enriched in the lighter carbon isotope (^{12}C), causing a noticeable depletion in atmospheric $\delta^{13}C$. Additionally, the lighter carbon isotope (^{12}C) is preferably absorbed by plants to perform photosynthesis (O'Leary, 1981), and deforestation also contributes to the atmospheric depletion of $\delta^{13}C$. The combination of both elements leads to a depletion in atmospheric $\delta^{13}C$, which is commonly referred to as the "Suess effect" (Suess, 1955; Keeling, 1979; Verburg, 2007).

Atmospheric CO_2 partly dissolves into the ocean and the relative proportion of dissolved inorganic ^{13}C in the oceans is steadily decreasing over time (Quay et al., 1992). Determining the rate of oceanic CO_2 uptake is of considerable importance in this context, and recent attention has focused on air–sea CO_2 exchange (e.g., Sarmiento et al., 2010; Raupach et al., 2014; Sitch et al., 2015). The air–sea gas exchange leads to the dissolution of atmospheric CO_2 into the ocean, and consequently the ^{13}C-Suess effect is reflected in the marine environment (Suess, 1955; Broecker and Maier-Reimer, 1992; Lynch-Stieglitz et al., 1995; Bacastow et al., 1996). Additionally, biological processes, including carbon fixation by primary producers, soft part organic tissue degradation and hard-part $CaCO_3$ remineralization, may play an important role in controlling oceanic $\delta^{13}C$, which varies regionally and with

* Corresponding author at: AZTI-Tecnalia, Marine Research Unit, Herrera Kaia Portualdea z/g, 20110 Pasaia, Basque Country, Spain.
E-mail address: ifraile@azti.es (I. Fraile).

http://dx.doi.org/10.1016/j.jmarsys.2015.12.012
0924-7963/

time and depth (Gruber et al., 1999; White, 2015). The marine ^{13}C-Suess effect has attracted interest because it provides a means for estimating the ocean's uptake rate of anthropogenic CO_2 (Quay et al., 1992; Bacastow et al., 1996).

While surface ocean $\delta^{13}C$ changes are linked to complex interactions between biological, thermodynamic and kinetic processes, (Gruber et al., 1999; Tagliabue and Bopp, 2008), oxygen stable isotope ratios in seawater are related to water temperature and salinity, and seawater $\delta^{18}O$ varies with ice volume, precipitation, evaporation or continental runoff. Higher $\delta^{18}O$ values indicate lower temperatures and higher salinities, and thus, $\delta^{18}O$ values of seawater display seasonal, interannual and long-term variations related to hydrological cycle and climate changes (Craig and Gordon, 1965; Killingley and Berger, 1979).

Calcified structures such as shells or fish skeletons precipitate in equilibrium with seawater, recording seawater chemistry and other environmental conditions (Kerr and Campana, 2014). Carbon and oxygen stable isotopic differences associated with marine microfossils and corals are widely used in paleoceanography for climate reconstructions (e.g. Spero et al., 1997; Taricco et al., 2009; Felis and Rimbu, 2010). However, studies using structures from marine vertebrates like fish are scarce; principally because due to their high solubility in seawater, their occurrence in marine sediments is rare (Woosley et al., 2012). In contrast to plankton records, fish skeletal structures offer a continuous temporal record spanning multiple years or decades, with the possibility of obtaining an integrated annual signature. Otoliths are accretionary aragonite structures found within the inner ear of teleost fishes and involved in fish's balance, orientation, and sound detection (Campana and Neilson, 1985). Given that the chemical composition of otoliths is linked to ambient seawater conditions these calcified structures are a powerful tool to investigate environmental histories of fishes. Following this line, otoliths of the long lived species Atlantic bluefin tuna (*Thunnus thynnus*) have been previously used to reconstruct interdecadal variations in oceanic $\delta^{13}C$ and $\delta^{18}O$ (Schloesser et al., 2009).

Here we investigate temporal variation in the otolith $\delta^{13}C$ and $\delta^{18}O$ of Atlantic bluefin tuna during the age-0 period within the Mediterranean Sea as a proxy for examining changes in oceanic $\delta^{13}C$ (Suess effect) and $\delta^{18}O$. During the first year, juvenile bluefin tuna can swim over vast geographic distances, but remain mainly in surface waters (Tudela et al., 2011; Galuardi and Lutcavage, 2012). Individuals produced in the Mediterranean Sea appear to stay within this basin (including the Strait of Gibraltar) during the first year of life (Rey, 1978), but as highly mobile predators, they may carry out local migrations to foraging areas throughout the Mediterranean Sea (Rooker et al., 2007). Therefore, otolith $\delta^{13}C$ and $\delta^{18}O$ accreted during the first year (hereafter referred to as otolith core) is likely reflective of seawater conditions within this basin. By analyzing stable isotopic composition of tuna otoliths we produced annual-resolution otolith $\delta^{13}C$ and $\delta^{18}O$ records for the Mediterranean Sea between 1989 and 2010, which was used to reconstruct seawater stable isotope composition shifts in this region. The time series of otolith $\delta^{13}C$ and $\delta^{18}O$ presented here contribute to our understanding of the oceanic carbon cycle, and enables to estimate the uptake rate of anthropogenic CO_2 by the Mediterranean Sea over the past two decades.

2. Material and methods

Bluefin tuna ($n = 632$) were captured between 2009 and 2011 from different locations in the Mediterranean Sea ($n = 150$), Strait of Gibraltar ($n = 174$) and the Bay of Biscay ($n = 308$) in the North Atlantic under an international biological sampling program by the International Commission for the Conservation of Atlantic Tunas (ICCAT, Fig. 1). Bluefin tuna from the foraging area in the Bay of Biscay were collected in the summer (June–September). Specimens from the Strait of Gibraltar were captured by Spanish traps in May, and by Portuguese traps mainly between late August and October. Sampling in the Mediterranean Sea included the eastern (Levantine Sea), central (region neighboring Malta in the Ionian Sea and waters around Sardinia in the Tyrrhenian Sea), and western (Balearic Sea) regions of the basin. Previous studies have demonstrated that the majority of bluefin tuna captured in these areas have originated in the Mediterranean Sea (> 95% in the Bay of Biscay and 100% in the Mediterranean Sea and Strait of Gibraltar based on Rooker et al., 2008, 2014; Fraile et al., 2015). Therefore, bluefin tuna used to characterize temporal shifts in otolith $\delta^{13}C$ and $\delta^{18}O$ were assumed to have resided within the Mediterranean Sea during their age-0 life period.

Fork length of each individual (FL, cm) was also recorded during the sample collection. Age–length relationship was used to estimate the age of individuals from length data by an inverse von Bertalanffy function (ICCAT, 2012, Cort, 1991). The sizes of individuals ranged from 52 to 282 cm FL with corresponding ages of 1 to 22 years, and birth year was back-calculated based on the estimated age and collection date (Table 1). The contribution of older individuals to the temporal patterns of otolith $\delta^{13}C$ and $\delta^{18}O$ was tested by selecting a subgroup of bluefin tuna <15 years and comparing these results with the complete

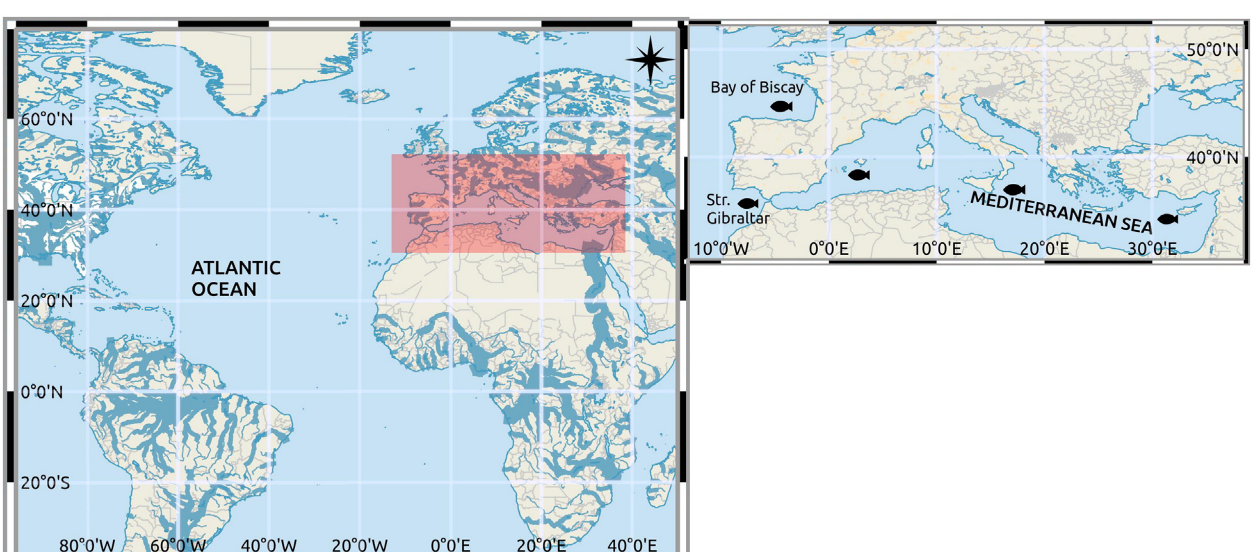

Fig. 1. Location of the areas used as sample sites in the North Atlantic Ocean (Bay of Biscay, Strait of Gibraltar) and Mediterranean Sea (western, central and eastern Mediterranean).

Table 1
Sample size, mean length, weight, and estimated age of Atlantic bluefin tuna (*Thunnus thynnus*) collected from the Bay of Biscay, Strait of Gibraltar and Mediterranean Sea between 2009 and 2011. FL = fork length (cm); N = sample size; age estimation based on growth curve from Cort (1991).

	FL(cm)		Age (year)		N
	Range	Mean	Range	Mean	
Bay of Biscay	52.5–182	103.5	1–8	3.3	308
Strait of Gibraltar	161–278	209.6	6–21	10.7	174
Mediterranean Sea	112.7–282	206.1	3–22	10.7	150

dataset. We used individuals of different age classes to investigate interannual variations over the last 22 years and represent the overall trend of isotopic values by capturing the different potential bluefin tuna subpopulations of the Mediterranean Sea.

After extraction, sagittal otoliths of bluefin the tuna were cleaned of tissues, soaked briefly in dilute nitric acid (1%), and then rinsed with deionized water. One otolith per fish was randomly selected and embedded in an Epofix resin (Struers) to cut a transverse section of approximately 1.5 mm thick across the core using an IsoMet® low-speed saw. Sections were then glued in a sample plate with thermoplastic glue. The otolith zone corresponding to the first year of growth was isolated and powdered using a high-resolution New Wave Micromill System consisting of a microscope and imaging system, controlled by computer software. The drill path corresponding to the yearling period was the same as the one used in previous studies (Schloesser et al., 2009; Rooker et al., 2014). The aragonite powder samples were analyzed for carbon and oxygen stable isotopes on an automated carbonate preparation device (KIEL-III) coupled to a gas-ratio mass spectrometer (Finnigan MAT 252) from the Environmental Isotope Laboratory of the University of Arizona. All the measurements are reported in the standard δ notation. The isotope ratio measurements were calibrated based on repeated measurements of National Bureau of Standards (NBS) NBS-19 and NBS-18, and the precision of isotope ratio measurements were ± 0.10‰ for $\delta^{18}O$ and ± 0.08‰ for $\delta^{13}C$ (1σ). Multiple isotopic measurements from a single otolith (0.03‰ for $\delta^{13}C$ and 0.04‰ for $\delta^{18}O$) indicated relatively high analytical precision for otolith $\delta^{13}C$ and $\delta^{18}O$.

For otoliths collected in the Bay of Biscay, quadratic discriminant function analysis (QDFA) was performed to identify potential western Atlantic migrants; only individuals from Mediterranean nursery origin were selected for this study (Fraile et al., 2015). No prior classification was performed in samples from the Strait of Gibraltar and Mediterranean Sea because previous studies have revealed that 100% of bluefin tuna from these areas are originated within the Mediterranean Sea (Rooker et al., 2014). Kendall's tau test was used to identify temporal trends in otolith $\delta^{13}C$ and $\delta^{18}O$ values, and the statistical meaningfulness of the trend was further evaluated using the methodology developed by Bryhn and Dimberg (2011). Values of $R^2 \geq 0.65$ and $p < 0.05$ were considered as statistically significant. Additionally, linear models (LM) were used to determine whether otolith $\delta^{13}C$ and $\delta^{18}O$ values varied significantly with birth year. When a significant relationship was identified, a linear regression was fitted to model the relationship between birth year and isotope values. Normal distribution assumption was tested with Shapiro–Wilk's normality test applicable for isotope data to ensure that both $\delta^{13}C$ and $\delta^{18}O$ values were normally distributed (i.e., no prior transformation was needed). Further, the Breusch–Pagan test was used to check the assumption of homoscedasticity of the residuals from the lineal regression, and no significant heteroscedasticity was detected (p-value > 0.05). To examine the effects of influential points on the linear regression, isotopic values corresponding to the oldest bluefin tuna from 1989 and 1990 were analyzed by computing Cook's distance. Additionally, model fit was evaluated using standard diagnostic plots (residuals versus fitted values, standardized residuals against quantiles of the standard normal distribution, standardized residuals against fitted values and Cook's distance plot), and the distribution of the residuals as well as the variance structure was found to be adequate to meet model assumptions.

Otolith $\delta^{13}C$ values from 1989 to 2010 were then compared to environmental data (sea surface temperature (SST), sea surface salinity (SSS), surface chlorophyll-α concentration (chl-α) and CO_2 emissions to the atmosphere) for the same time period in the Mediterranean Sea. Given the strong internal gradient in physicochemical properties within the Mediterranean Sea, reference values from the Strait of Gibraltar (located at the western entrance to the Mediterranean Sea), Ionian Sea (central Mediterranean Sea) and Levantine Sea (eastern Mediterranean Sea) were used to represent the longitudinal range of environmental conditions within the basin. Night-time sea surface temperature (SST) data were derived from AVHRR Pathfinder Version 5.2 data, obtained from the US National Oceanographic Data Center and GHRSST (http://pathfinder.nodc.noaa.gov; updated from Casey et al., 2010). Statistical values averaged for 22.5° × 22.5° (latitude × longitude) grid were used, and mean August temperature was computed to represent summer SST variation. Eastern Mediterranean Sea SST corresponded to the average value East of 22.5° E, central Mediterranean SST represented the average temperature from 0 to 22.5° E, and the Strait of Gibraltar was considered the region west of 0°. Summer SSS of the Mediterranean Sea was derived from the Mediterranean Sea Physics Reanalysis (1987–2013) generated using Copernicus Marine Environment Monitoring Service (CMEMS) products. Salinity estimates of three reference points have been selected to represent different Mediterranean regions: Levantine Sea representing eastern Mediterranean Sea (34°N 30°E), Ionian Sea representing the central Mediterranean Sea (34°N 15°E) and the Strait of Gibraltar representing the western limit of the age-0 tuna habitat (36°N 2°W). SeaWiFS monthly estimates of surface chl-α concentration in the Mediterranean Sea were used to describe summer chl-α variation between 1997 and 2010 (satellite launched in August 1997 by NASA's Earth Observing System; Hooker and Esaias, 1993). Data from the Levantine Sea (34°N 30°E), Ionian Sea (34°N 15°E) and the Strait of Gibraltar (36°N 2°W) were used to represent eastern, central and western Mediterranean Sea regions (L3 monthly dataset at a resolution of 9 Km available from https://podaac.jpl.nasa.gov). The mean August chl-α value in the Strait of Gibraltar, central Mediterranean Sea and eastern Mediterranean Sea were used to represent summer chl-α concentration (mg m^{-3}). Emissions of CO_2 to the atmosphere from fossil-fuel combustion were estimated using the Carbon Dioxide Information Analysis Center (CDIAC), and emissions from countries surrounding the Mediterranean Sea were computed from 1989 to 2010 (Boden et al., 2013). Correlation analysis was used to examine the relationship between otolith $\delta^{13}C$ and CO_2 emissions.

3. Results and discussion

Otolith $\delta^{13}C$ and $\delta^{18}O$ values of bluefin tuna ranged from −10.24 to −6.96‰ and from −1.38 to 0.02‰, respectively. Kendall's Tau test indicated a negative relation between otolith $\delta^{13}C$ and birth year ($p < 0.05$) and the statistical meaningful of the trend was found to be significant ($p < 0.05$), with otolith $\delta^{13}C$ becoming more depleted as the birth year approached 2010. Likewise, the LM analysis and showed a statistically significant ($p < 0.05$) depletion in $\delta^{13}C$ values from 1989 to 2010. In contrast, temporal pattern in otolith $\delta^{18}O$ was not significant based on Kandall's Tau test, the statistical meaningfulness tests of Bryhn and Dimberg (2011) and the LM analysis (Fig. 2). The isotopic values corresponding to otoliths of bluefin tuna born before 1993 were not influential on the overall trend, with Cook's distance less than 1.

Otolith $\delta^{13}C$ values decreased 0.94‰ from 1989 to 2010, which corresponds to annual depletion rate of 0.05‰. The marine Suess effect has been previously reported using otoliths from Atlantic bluefin tuna collected in the western Atlantic Ocean from 1947 to 2006, and the

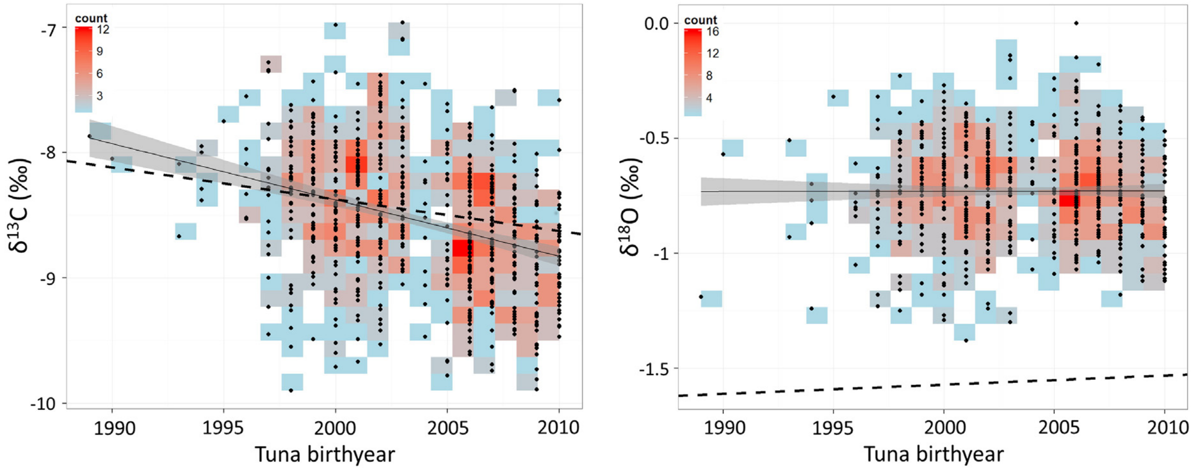

Fig. 2. Temporal variation in the $\delta^{13}C$ and $\delta^{18}O$ values measured in milled otoliths of Atlantic bluefin tuna (*Thunnus thynnus*) originated in the Mediterranean Sea. Solid lines represent a linear regression of $\delta^{13}C$ and $\delta^{18}O$ values in relation to birthyear. The depletion rate of $\delta^{13}C$ estimated using bluefin tuna otoliths from the western North Atlantic Ocean (dashed line) is shown for comparison (from Schloesser et al., 2009). The average standard deviation (SD) of the measurements is approximately 0.03‰ for $\delta^{13}C$ data and 0.04‰ for $\delta^{18}O$ data. Colours represent counts of isotopic data in rectangular bins of one birth year and 0.13‰ variation for $\delta^{13}C$ and 0.06‰ for $\delta^{18}O$.

depletion rate for otolith $\delta^{13}C$ reported in that study was 0.026‰ per year (Schloesser et al., 2009). The $\delta^{13}C$ depletion rate reported for the western Atlantic Ocean using bluefin tuna otolith was twice as low as that estimated for the Mediterranean Sea (Schloesser et al., 2009), which implies that the high depletion rate found in our tuna otoliths is not a species specific characteristic, but may be due changes in seawater $\delta^{13}C$ in the Mediterranean Sea over the last two decades. Similarly, the Suess effect has been documented in other biogenic carbonates of the world's oceans (e.g., corals [Pelejero et al., 2005; Swart et al., 2010; Dassié et al., 2013], mussel shells [Butler et al., 2009; Pfister et al., 2011] or planktonic foraminifera [Black et al., 2011; Beveridge and Shackleton, 1994; Bauch et al., 2000]), as well as in $\delta^{13}C$ reconstruction based on chlorofluorocarbons (CFCs) (Körtzinger and Quay, 2003). Although species specific carbonate fractionation or habitat use may influence the isotopic composition of their carbonates, $\delta^{13}C$ depletion rates estimated for the tropical North Atlantic Ocean using different marine organisms are of similar magnitude for a similar time frame ($-0.02‰\ yr^{-1}$, $-0.027‰\ yr^{-1}$ and $-0.03‰\ yr^{-1}$ based on sclerosponges, corals and planktonic foraminifera, respectively [Böhm et al., 1996; Swart et al., 2010; Black et al., 2011]). On the other hand, CFC-derived water ages have shown that the $\delta^{13}C$ depletion rates vary from region to region, with overall depletion rates being higher in tropical and subtropical oceans compared to polar regions (Sonnerup et al., 1999). Furthermore, an inter-ocean comparison based on $\delta^{13}C$ measurements in corals have revealed higher depletion rates in the Atlantic Ocean compared to Pacific Ocean and Indian Ocean (Swart et al., 2010). However, the magnitudes of depletion rates reported on these studies for the period 1960 – early 2000s were lower than the $0.05‰\ yr^{-1}$ recorded by fish otoliths in the Mediterranean Sea. The high depletion rates found in the Mediterranean Sea may partly be due to the special characteristics of this semi-enclosed basin with narrow connection with the Atlantic Ocean making particularly sensitive to anthropogenic perturbations and climate change (Diffenbaugh and Giorgi, 2012). Due to the higher total alkalinity of the Mediterranean Sea compared to open ocean, it has a greater capacity to absorb anthropogenic CO_2 (Palmiéri et al., 2015), leading to an amplification of the Suess effect. However, a recent study using mussels shells from the southern California estimated a $\delta^{13}C$ depletion rate of $-0.07‰\ yr^{-1}$ from 1999 to 2009 (Pfister et al., 2011). The unusually high depletion rates found by Pfister et al. (2011) coupled with our observed rate of depletion may indicate an acceleration of the Suess effect over the last few decades.

An increase in sea surface temperature of the Mediterranean Sea, may also have contributed to the observed decline in otolith $\delta^{13}C$ values (Santoreli et al., 1995; Skliris et al., 2012). In order to quantify the effect of seawater temperature on otolith $\delta^{13}C$, summer sea surface temperatures (SST) from eastern Mediterranean Sea, central Mediterranean Sea and Strait of Gibraltar were derived from AVHRR Pathfinder Version 5.2 for the period 1989–2010 (Fig. 3). An increasing trend was visible for all localities, although this warming trend during the studied period was significant only in the eastern Mediterranean Sea (p-value < 0.05). The estimated increase of SST from the linear regression of satellite data was of 0.4 °C in the central Mediterranean Sea and Strait of Gibraltar and of 1.9 °C in the eastern Mediterranean Sea during the period studied. Using the thermodynamic relationship between $\delta^{13}C$ in otoliths and temperature from Thorrold et al. (1997) and assuming constant seawater $\delta^{13}C$ concentration, we estimated a depletion of 0.07‰ (in the Strait of Gibraltar and central Mediterranean) and 0.34‰ (in the eastern Mediterranean) caused by the increase in temperature. The magnitude of the $\delta^{13}C$ depletion estimated in our study (0.94‰ over a 21 yr period) exceeds that expected from the temperature–$\delta^{13}C$ relationship presented by Thorrold et al. (1997), since an increase in surface temperature of approximately 5 °C would be needed to explain a depletion in otolith $\delta^{13}C$ of 0.94‰. Still, thermodynamic effect in carbonate fractionation may be important, especially in the eastern Mediterranean Sea where SST raise in the last decades has been more pronounced (Fig. 3). The depletion in otolith $\delta^{13}C$ after substracting thermodynamic effect on $\delta^{13}C$ fractionation was estimated to be between 0.6‰ and 0.87‰ over the studied period, which turns in a depletion rate of about 0.03‰–$0.04‰\ yr^{-1}$. Alternatively, decreased primary production (reducing isotopically light carbon in seawater) could also contribute in decreasing otolith $\delta^{13}C$. However, chl-α concentration estimated from remote sensing data over the last 12 years for the three Mediterranean Sea regions indicated that the variation in summer phytoplankton production in these regions was not significant, suggesting that any effect on otolith $\delta^{13}C$ depletion is likely inconsequential (Fig. 3). An increase in summer surface salinity was detected from 1989 to 2010; although this temporal variation was significant only in the eastern Mediterranean Sea. Given that otolith $\delta^{13}C$ shows a positive relationship with salinity (Elsdon and Gillanders, 2002), the influence of salinity otolith $\delta^{13}C$ values of bluefin tuna was also assumed to be unimportant. Our findings suggest that seawater $\delta^{13}C$ variation is likely the main factor influencing the observed temporal pattern in otolith $\delta^{13}C$.

The accelerated depletion of $\delta^{13}C$ observed in the otoliths of bluefin tuna may be linked to increased CO_2 emission rates observed in the last decade, and attributed primarily to increases in fossil fuel combustion and cement production (Le Quéré et al., 2014). Global atmospheric

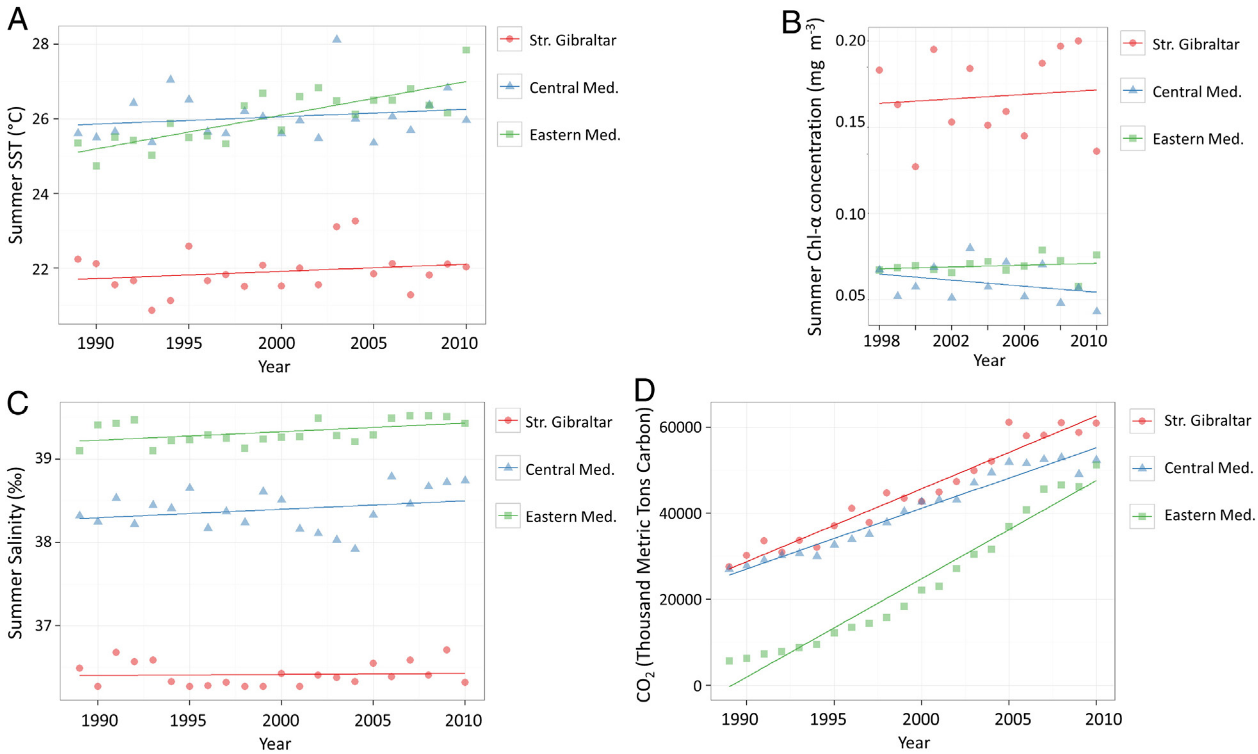

Fig. 3. A) Linear regression applied on AVHRR-derived SST data from 1989 to 2010 in the eastern Mediterranean Sea, central Mediterranean Sea and Strait of Gibraltar as indicated by night-time satellite observations and corresponding $\delta^{13}C$ depletion rate for the same period. Average August SST was used to represent summer temperature (Strait of Gibraltar and Ionian Sea: slope (b) = 0.02, standard error (SE) = 0.02, p-value > 0.05; Levantine Sea: b = 0.09, SE = 0.01, p-value 0.05; Ionian Sea: b = −0.002, SE = 0.001, p-value 0.05). C) Summer sea surface salinity from 1989 to 2010 in the eastern Mediterranean Sea, central Mediterranean Sea and Strait of Gibraltar (from Mediterranean Sea Physics Reanalysis [1987–2013] generated using CMEMS products) (Strait of Gibraltar: b = 0.001, SE = 0.005, p-value > 0.05; Ionian Sea: b = 0.01, SE = 0.008, p-value > 0.05; Levantine Sea: b = 0.01, SE > = > 0.004, p-value 2 emissions to the atmosphere between 1989 and 2010 around the Mediterranean countries (from Boden et al., 2013) (Strait of Gibraltar: b = 1.7·103, SE = 85, p-value < 0.05; Ionian Sea: b = 1.4·101, SE = 69, p-value < 0.05; Levantine Sea: b = 2.3·103, SE = 110, p-value < 0.05).

CO_2 concentration and the uptake of CO_2 by ocean and terrestrial "CO_2 sinks" are still increasing with time, leading to declines in both atmospheric and oceanic $\delta^{13}C$ (Le Quéré et al., 2014). Anthropogenic CO_2 emissions to the atmosphere from Mediterranean countries have increased significantly during the past few decades (Fig. 3). Between 1989 and 2010, CO_2 emissions and annual mean otolith $\delta^{13}C$ values showed a strong negative correlation (correlation coefficient of −0.86), confirming the influence of anthropogenic CO_2 on otolith $\delta^{13}C$ (Fig. 4).

Despite of the temporal decline in otolith $\delta^{13}C$ values, the data exhibit much variability over the entire time period. This high variability can be explained by the differences in the migratory movements among individuals, meaning that individuals in our sample likely experienced different environmental conditions or water masses during the young

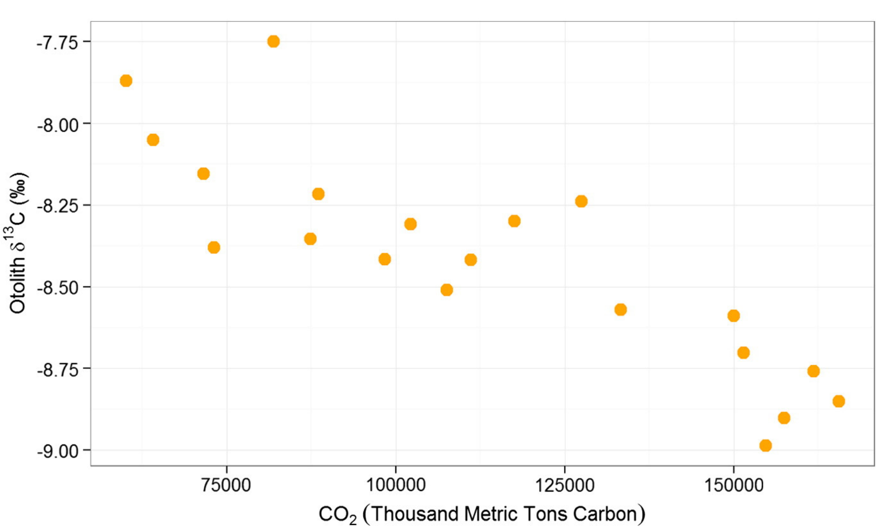

Fig. 4. Relationship between yearly mean otolith $\delta^{13}C$ values between 1989 and 2010 measured on bluefin tuna (*Thunnus thynnus*) from the Mediterranean Sea and annual CO_2 emissions to the atmosphere around the Mediterranean countries (data from Boden et al., 2013).

of the year period. Movements of young individual bluefin tuna within the Mediterranean Sea are still not well resolved, but by the end of the age-0 period these fish are capable to swim long distances. When different schools of bluefin tuna occur at the same region, they tend to mix and regroup themselves into new school units (Hilborn, 1991). During these migrations, bluefin tuna feed on different type of prey and reside in water masses of distinct physico-chemical and biological properties, being exposed to conditions influencing $\delta^{13}C$ incorporation into their otoliths. Thus, the high variability of carbon isotopes may be linked to differences among individuals in the diet, behavior and habitat types (with differing temperature and productivity regimes) during the first year of live. Additionally, seasonal and interannual variations in the comparatively fresher North Atlantic water mass entering through the Strait of Gibraltar may also introduce additional variability by influencing the isotopic signature of individuals that reside in the western part of the Mediterranean Sea (Macías et al., 2008). While individual $\delta^{13}C$ values vary considerably, annual mean $\delta^{13}C$ values reflect an integrated signal of the Mediterranean basin.

Although individual movements result in the variability within a certain year, the observed decadal-scale trend in $\delta^{13}C$ record that we observed in Atlantic bluefin tuna otoliths appears related to the carbon cycle in the Mediterranean Sea. Swart et al. (2010) suggested that $\delta^{13}C$ in corals is an important tracer for oceanic uptake of anthropogenic CO_2, as they reflect $\delta^{13}C$ of the dissolved inorganic carbon of the surface ocean. Schloesser et al. (2009) found a strong correlation between $\delta^{13}C$ in otoliths and athmospheric $\delta^{13}C$, and concluded that bluefin tuna otoliths effectively track changes in atmospheric $\delta^{13}C$. The progressive decline in otolith $\delta^{13}C$ in the current study also could be ascribed to the Suess effect in the Mediterranean Sea. A recent study by Palmiéri et al. (2015) simulated Mediterranean Sea's uptake of anthropogenic CO_2 during the last two centuries, and found an steep increase in anthropogenic carbon since the industrial period. Likewise, an increase in dissolved inorganic carbon over the past few decades has been reported for the western and central Mediterranean Sea (Touratier and Goyet, 2009; Luchetta et al., 2010; Geri et al., 2014).

Results derived from the LM analysis indicated a lack of temporal pattern in otolith $\delta^{18}O$ (Fig. 2). Seawater $\delta^{18}O$ varies principally due to evaporation, precipitation and river input. The difference in otolith $\delta^{18}O$ values of bluefin tuna measured by Schloesser et al. (2009) and the current work is likely due to salinity differences between the Mediterranean Sea and western North Atlantic Ocean (Fig. 2). The small but significant variation in otolith $\delta^{18}O$ found by Schloesser et al. (2009) was associated to changes in salinity regimes around the Gulf Stream. In contrast, an increase in salinity between 1989 and 2010 was only significant in the eastern Mediterranean Sea (Fig. 3), with minor implications for the overall otolith $\delta^{18}O$ trend. Moreover, previous studies have shown that $\delta^{18}O$ fractionation in otoliths is also temperature dependent, with inverse relationship between otolith $\delta^{18}O$ and water temperature (Thorrold et al., 1997). Assuming a SST increase between 0.4 and 1.9 °C (AVHRR Pathfinder dataset) and using the temperature–otolith $\delta^{18}O$ relationship presented by Thorrold et al. (1997), depletion in otolith $\delta^{18}O$ between 0.08‰ and 0.39‰ could be expected due to thermodynamic effects in $\delta^{18}O$ fractionation. However, an increase in salinity is related with an enrichment of otolith $\delta^{18}O$ (Elsdon and Gillanders, 2002), which could partly balance the thermodynamic effect (between 0.01‰ and 0.06‰ enrichment [Legrande and Schmidt, 2006], assuming otolith $\delta^{18}O$ is deposited in equilibrium with seawater). Thus, the lack of temporal variation observed in bluefin tuna otoliths could be explained by the combined influence of elevated temperature and salinity on $\delta^{18}O$ fractionation.

Similar to $\delta^{13}C$, pronounced variability was also observed for otolith $\delta^{18}O$ over the time period investigated and may be explained by changes in seawater $\delta^{18}O$ composition within Mediterranean basin. Seawater $\delta^{18}O$ mirrors the distribution of surface salinity, and hence, enriched $\delta^{18}O$ values are found in the eastern Mediterranean Sea, compared to the more depleted values of the western Mediterranean Sea (Pierre, 1999; Gat et al., 1996). Variability in otolith $\delta^{18}O$ within a certain year could be expected due to differences in habitat use among individuals of the eastern and western Mediterranean Sea. Additionally, Pierre (1999) showed that interannual variations in oceanic $\delta^{18}O$ are larger in the western Mediterranean Sea due to the variability in the inflow of North Atlantic water throughout the Strait of Gibraltar, compared to more stable values of the eastern Mediterranean Sea. These changes could introduce additional variability to the otolith $\delta^{18}O$ values, especially for those individuals that occur near the Strait of Gibraltar during the age-0 period.

4. Conclusions

We present an annual resolution $\delta^{13}C$ and $\delta^{18}O$ record from Atlantic bluefin tuna otoliths that reflects changes in oceanic $\delta^{13}C$ of the Mediterranean Sea from 1989 to 2010. A year to year depletion in otolith $\delta^{13}C$ was observed for the study period, which correlated with the increase of atmospheric CO_2 emissions around the Mediterranean countries. As a result, otolith $\delta^{13}C$ depletion could be attributed to the increase in anthropogenic CO_2 emissions (Suess effect). A depletion of 0.94‰ was estimated over a 22 yr period, which translates in an annual depletion rate of 0.05‰. This depletion rate is higher than previously reported, and was interpreted as a consequence of combined thermodynamic effect on isotopic fractionation and the Suess effect. The increase of SST between 1989 and 2010 could explain part of the depletion observed in bluefin tuna otoliths. After subtracting thermodynamic effect, the net $\delta^{13}C$ depletion between 0.6‰ and 0.87‰ was estimated to be due to the oceanic Suess effect, which turns in a depletion rate of about 0.03‰–0.04‰ yr^{-1}. This is still higher than most of the previously reported values in the tropical and temperate North Atlantic Ocean, suggesting that the Suess effect between 1989 and 2010 in the Mediterranean Sea may be magnified compared to open ocean regions. We also found little variation in otolith $\delta^{18}O$ over the time period investigated, likely due to the opposite effect of increasing SST and salinity in $\delta^{18}O$ fractionation.

Interdecadal changes recorded in the otoliths of bluefin tuna demonstrate the promise of the approach for tracking temporal changes in the seawater chemistry and past environmental conditions experienced by bluefin tuna.

Acknowledgements

We would like to express our deepest gratitude to L. Naval, the staff from "La Bacaladera" and the crew from "Attalaya Berria" for sampling help in the Bay of Biscay. We also thank all the people that actively participated in sample collection across the Mediterranean Sea and the Strait of Gibraltar. The authors are indebted to the two anonymous reviewers for their many helpful insights and suggestions which improved the manuscript considerably. This work was carried out under the provision of the ICCAT Atlantic Wide Research Programme for Bluefin Tuna (GBYP), co-funded by the European Union (grant SI2/585616), by several other ICCAT CPCs, the ICCAT Secretariat and by other entities (see http://www.iccat.int/GBYP/en/Budget.htm). The contents of this paper do not necessarily reflect the point of view of ICCAT or of the other funders. This work was also supported by grant CTM2011-27505 by the Spanish Ministry of Economy and Competitiveness (MINECO), National Geographic Society (Grant W132-10) and the Basque Government (Grant 351BI20080022). This paper is contribution number 733 from AZTI-Tecnalia (Marine Research).

References

Bacastow, R.B., Keeling, C.D., Lueker, T.J., Wahlen, M., Mook, W.G., 1996. The 13C Suess effect in the world surface oceans and its implications for oceanic uptake of CO_2: analysis of observations at Bermuda. Glob. Biogeochem. Cycles 10, 335–346. http://dx.doi.org/10.1029/96GB00192.

Bauch, D., Carstens, J., Wefer, G., Thiede, J., 2000. The imprint of anthropogenic CO_2 in the Arctic Ocean: evidence from planktic d13C data from water column and sediment surfaces. Deep Sea Res., Part II 47, 1791–1808. http://dx.doi.org/10.1016/S0967-0645(00)00007-2.

Beveridge, N.A.S., Shackleton, N.J., 1994. Carbon isotopes in recent planktonic foraminifera: a record of anthropogenic CO_2 invasion of the surface ocean. Earth Planet. Sci. Lett. 126, 259–273. http://dx.doi.org/10.1016/0012821X(94)90111-2.

Black, D., Thunell, R., Wejnert, K., Astor, Y., 2011. Carbon isotope composition of Caribbean Sea surface waters: response to the uptake of anthropogenic CO_2. Geophys. Res. Lett. 38, L16609. http://dx.doi.org/10.1029/2011GL048538.

Boden, T.A., Marland, G., Andres, R.J., 2013. Global, Regional, and National Fossil-Fuel CO2 Emissions. Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory, U.S. Department of Energy, Oak Ridge, Tenn., U.S.A. http://dx.doi.org/10.3334/CDIAC/00001_V2013.

Böhm, F., Joachimski, M.M., Lehnert, H., Morgenroth, G., Kretschmer, W., Vaceot, W.C., 1996. Carbon isotope records from extant Caribbean and South Pacific sponges: evolution of δ13C insurance water DIC. Earth Planet. Sci. Lett. 139, 291–303.

Broecker, W.S., Maier-Reimer, E., 1992. The influence of air and sea exchange on the carbon isotope distribution in the sea. Glob. Biogeochem. Cycles 6, 315–320. http://dx.doi.org/10.1029/92GB01672.

Bryhn, A.C., Dimberg, P.H., 2011. An operational definition of a statistically meaningful trend. PLoS One 6, e19241. http://dx.doi.org/10.1371/journal.pone.0019241.

Butler, P.G., Scourse, J.D., Richardson, C.A., Wanamaker, A.D., Bryant, C.L., Bennell, J.D., 2009. Continuous marine radiocarbon reservoir calibration and the ^{13}C Suess effect in the Irish Sea: results from the first multi-centennial shell-based marine master chronology. Earth Planet. Sci. Lett. 279, 230–241.

Campana, S.E., Neilson, J.D., 1985. Microstructure of fish otoliths. Can. J. Fish. Aquat. Sci. 42, 1014–1032.

Casey, K.S., Brandon, T.B., Cornillon, P., Evans, R., 2010. The past, present and future of the AVHRR Pathfinder SST Program. In: Barale, V., Gower, J.F.R., Alberotanza, L. (Eds.), Oceanography from Space. Springer. http://dx.doi.org/10.1007/978–90-481-8681-5_16.

Cort, J.L., 1991. Age and growth of bluefin tuna (*Thunnus thynnus* L.) of the Northwest Atlantic, collective volume of scientific papers. ICCAT 35, 213–230.

Craig, H., Gordon, L.I., 1965. Deuterium and oxygen 18 variations in the ocean and marine atmosphere. In: Tongiogi, E. (Ed.), Stable Isotopes in Oceanographic Studies and Paleotemperatures, Spoleto, Italy, pp. 9–130.

Dassié, E.P., Lemley, G.M., Linsley, B.K., 2013. The Suess effect in Fiji coral δ13C and its potential as a tracer of anthropogenic CO_2 uptake. Palaeogeogr. Palaeoclimatol. Palaeoecol. 370, 30–40.

Diffenbaugh, N.S., Giorgi, F., 2012. Climate change hotspots in the CMIP5 global climate model ensemble. Climate Change 114, 813–822.

Elsdon, T.S., Gillanders, M., 2002. Interactive effects of temperature and salinity on otolith chemistry: challenges for determining environmental histories of fish. Can. J. Fish. Aquat. Sci. 59, 1796–1808.

Farquhar, G.D., Ehleringer, J.R., Hubick, K.T., 1989. Carbon isotope discrimination and photosynthesis. Annu. Rev. Plant Physiol. Plant Mol. Biol. 40, 507–537.

Felis, T., Rimbu, N., 2010. Mediterranean climate variability documented in oxygen isotope records from northern red sea corals—a review. Glob. Planet. Chang. 71, 232–241.

Fraile, I., Arrizabalaga, H., Rooker, J.R., 2015. Origin of Atlantic bluefin tuna (*Thunnus thynnus*) in the Bay of Biscay. ICES J. Mar. Sci. 72, 625–634. http://dx.doi.org/10.1093/icesjms/fsu156.

Francey, R.J., Allison, C.E., Etheridge, D.M., Trudinger, C.M., Enting, I.G., Leuenberger, M., Langenfelds, R.L., Michel, E., Steele, L.P., 1999. A 1000 year high precision record of δ13C in atmospheric CO_2. Tellus 51B, 170–193.

Friedli, H., Lötscher, H., Oeschger, H., Siegenthaler, U., Stauffer, B., 1986. Ice core record of 13C/12C ratio of atmospheric CO_2 in the past two centuries. Nature 324, 237–238. http://dx.doi.org/10.1038/324237a0.

Galuardi, B., Lutcavage, M., 2012. Dispersal routes and habitat utilization of juvenile Atlantic bluefin tuna, *Thunnus thynnus*, tracked with mini PSAT and archival tags. PLoS One 7 (5), e37829. http://dx.doi.org/10.1371/journal.pone.0037829.

Gat, J.R., Shemesh, A., Tziperman, E., Hecht, A., Georgopoulos, D., Basturk, O., 1996. The stable isotope composition of waters of the eastern Mediterranean sea. J. Geophys. Res. 101, 6441–6451.

Geri, P., Yacoubi, S.E., Goyet, C., 2014. Forecast of sea surface acidification in the Northwestern Mediterranean Sea. J. Comput. Environ. Sci. 1-7.

Gruber, N., Keeling, C.D., Bacastow, R.B., Guenther, P.R., Lueker, T.J., Wahlen, M., Meijer, H.A., Mook, W.G., Stocker, T.F., 1999. Spatiotemporal patterns of carbon-13 in the global surface oceans and the oceanic Suess effect. Glob. Biogeochem. Cycles 13, 307–335. http://dx.doi.org/10.1029/1999GB900019.

Hilborn, R., 1991. Modeling the stability of fish schools: exchange of individual fish between schools of skipjack tuna (*Katsuwonus pelamis*). Can. J. Fish. Aquat. Sci. 48, 1081–1091.

Hooker, S.B., Esaias, W.E., 1993. An overview of the SeaWiFS Project. *EOS Trans. Am. Geophys. Union* 74, 245–246.

ICCAT (International Commission for the Conservation of Atlantic Tuna), 2012a. Report of the 2012 Atlantic Bluefin Tuna Stock Assessment Session. Madrid, Spain, September 4–11 (124 pp.).

Keeling, C.D., 1979. Recent trends in the 13C/12C ratio of atmospheric carbon dioxide. Nature 277, 121–123.

Keeling, C.D., Piper, S.C., Bacastow, R.B., Wahlen, M., Whorf, T.P., Heimann, M., Meijer, H.A., 2005. Atmospheric CO_2 and $13CO_2$ exchange with the terrestrial biosphere and oceans from 1978 to 2000: observations and carbon cycle implications. In: Ehleringer, J.R., Cerling, T.E., Dearing, M.D. (Eds.), A History of Atmospheric CO_2 and its Effects on Plants, Animals, and Ecosystems. Springer Verlag, New York, pp. 83–113.

Keeling, C.D., Piper, S.C., Whorf, T.P., Keeling, R.F., 2011. Evolution of natural and anthropogenic fluxes of atmospheric CO_2 from 1957 to 2003. Tellus 63B, 1–22.

Kerr, L.A., Campana, S.E., 2014. Chemical composition of fish hard parts as a natural marker of fish stocks. In: Cadrin, S.X., Kerr, L.A., Mariani, S. (Eds.), Stock Identification Methods: Applications in Fisheries Science. Academic Press, San Diego, pp. 205–234.

Killingley, J.S., Berger, W.H., 1979. Stable isotopes in a mollusk shell: detection of upwelling events. Science 205, 186–188.

Körtzinger, A., Quay, P.D., 2003. Relationship between anthropogenic CO_2 and the ^{13}C suess effect in the North Atlantic Ocean. Glob. Biogeochem. Cycles 17, 1005. http://dx.doi.org/10.1029/2001GB001427.

Le Quéré, C., Moriarty, R., Andrew, R.M., Peters, G.P., Ciais, P., Friedlingstein, P., Jones, S.D., et al., 2014. Global carbon budget 2014. Earth Syst. Sci. Data Discuss. 7, 521–610.

LeGrande, A.N., Schmidt, G.A., 2006. Global gridded data set of the oxygen isotopic composition in seawater. Geophys. Res. Lett. 33, L12604. http://dx.doi.org/10.1029/2006GL026011.

Luchetta, A., Cantoni, C., Catalano, G., 2010. New observations of CO_2-induced acidification in the northern Adriatic sea over the last quarter century. Chem. Ecol. 26, 1–17.

Lynch-Stieglitz, J., Stocker, T.F., Broecker, W.S., Fairbanks, R.G., 1995. The influence of air–sea exchange on the isotopic composition of oceanic carbon: observations and modeling. Glob. Biogeochem. Cycles 9, 653–655. http://dx.doi.org/10.1029/95GB02574.

Macías, D., Bruno, M., Echevarria, F., Vazquez, A., Garcia, C.M., 2008. Meteorologically-induced mesoscale variability of the north-western alboran sea (Southern Spain) and related biological patterns. Estuar. Coast. Shelf Sci. 78, 250–266.

O'Leary, M.H., 1981. Carbon isotope fractionation in plants. Phytochemistry 20, 553–567.

Pachauri, R.K., Meyer, L., 2014. Climate Change 2014, Synthesys Report. 2014. IPCC.

Palmiéri, J., Orr, J.C., Dutay, J.C., Béranger, K., Schneider, A., Beuvier, J., Somot, S., 2015. Simulated anthropogenic CO_2 storage and acidification of the Mediterranean Sea. Biogeosciences 12, 781–802. http://dx.doi.org/10.5194/bg-12-781-2015.

Pelejero, C., Calvo, E., McCulloch, M.T., Marshall, J.F., Gagan, M.K., Lough, J.M., Opdyke, B.N., 2005. Preindustrial to modern interdecadal variability in coral reef pH. Science 309, 2204–2207.

Pfister, C.A., McCoy, S.J., Wootton, J.T., Martin, P.A., Colman, A.S., Archer, D., 2011. Rapid environmental change over the past decade revealed by isotopic analysis of the California mussel in the Northeast Pacific. PLoS One 6, e25766. http://dx.doi.org/10.1371/journal.pone.0025766.

Pierre, C., 1999. The oxygen and carbon isotope distribution in the Mediterranean water masses. Mar. Geol. 153, 41–55.

Quay, P.D., Tilbrook, B., Wong, C.S., 1992. Oceanic uptake of fossil fuel CO_2: carbon-13 evidence. Science 256, 74–79.

Raupach, M.R., Gloor, M., Sarmiento, J.L., Canadell, J.G., Frölicher, T.L., Gasser, T., Houghton, R.A., Le Quéré, C., Trudinger, C.M., 2014. The declining uptake rate of atmospheric CO_2 by land and ocean sinks. Biogeosciences 11, 3453–3475. http://dx.doi.org/10.5194/bg-11-3453-2014.

Rey, J.C., 1978. Resultados de la campaña de marcado de atún rojo, *Thunnus thynnus* (L.), juvenil en la costa mediterránea española. Collect. Vol. Sci. Pap. ICCAT, 7, 318–321.

Rooker, J.R., Alvarado, J., Block, B., Dewar, H., De Metrio, G., Corriero, A., Kraus, R.T., et al., 2007. Life history and stock structure of Atlantic bluefin tuna (*Thnnus thynnus*). Rev. Fish. Sci. 15, 265–310.

Rooker, J.R., Secor, D.H., De Metrio, G., Schloesser, R., Block, B.A., Neilson, J.D., 2008. Natal homing and connectivity in Atlantic bluefin tuna populations. Science 322, 742–744.

Rooker, J.R., Arrizabalaga, H., Fraile, I., Secor, D.H., Dettman, D.L., Abid, N., Addis, P., et al., 2014. Crossing the line: migratory and homing behaviours of Atlantic bluefin tuna. Mar. Ecol. Prog. Ser. 504, 265–276.

Santoreli, R., Böhm, E., Schiano, M.E., 1995. The sea surface temperature of the Western Mediterranean Sea: historical satellite thermal data. In: La Violette, P.E. (Ed.), Seasonal and Interannual Variability of the Western Mediterranean Sea. American Geophysical Union, Washington D.C., pp. 155–176.

Sarmiento, J.L., Gloor, M., Gruber, N., Beaulieu, C., Jacobson, A.R., Mikaloff Fletcher, S.E., Pacala, S., Rodgers, K., 2010. Trends and regional distributions of land and ocean carbon sinks. Biogeosciences 7, 2351–2367. http://dx.doi.org/10.5194/bg-7-2351-2010.

Schloesser, R.W., Rooker, J.R., Louchuoarn, P., Neilson, J.D., Secor, D.H., 2009. Interdecadal variation in seawater d13C and d18O recorded in fish otoliths. Limnol. Oceanogr. 54, 1665–1668.

Sitch, S., Friedlingstein, P., Gruber, N., Jones, S.D., Murray-Tortarolo, G., Ahlström, A., Doney, S.C., et al., 2015. Recent trends and drivers of regional sources and sinks of carbon dioxide. Biogeosciences 12, 653–679.

Skliris, N., Sofianos, S., Gkanasos, A., Mantziafou, A., Vervatis, V., Axaopoulos, P., Lascaratos, A., 2012. Decadal scale variability of sea surface temperature in the Mediterranean Sea in relation to atmospheric variability. Ocean Dyn. 62, 13–30.

Sonnerup, R.E., Quay, P.D., McNichol, A.P., Bullister, J.L., Westby, T.A., Anderson, H.L., 1999. Reconstructing the oceanic 13C Suess effect. Glob. Biogeochem. Cycles 13, 857–872. http://dx.doi.org/10.1029/1999GB900027.

Spero, J., Bijma, J., Lea, D.W., Bemis, B.E., 1997. Effect of seawater carbonate concentration on foraminiferal carbon and oxygen isotopes. Nature 390, 497–500.

Suess, H.E., 1955. Radiocarbon concentrations in modern wood. Science 122, 415–417.

Swart, P.K., Greer, L., Rosenheim, B.E., Moses, C.S., Waite, A.J., Winter, A., Dodge, R.E., Helmle, K., 2010. The 13C Suess effect in scleractinian corals mirror changes in the anthropogenic CO_2 inventory of the surface oceans. Geophys. Res. Lett. 37, L05604. http://dx.doi.org/10.1029/2009GL041397.

Tagliabue, A., Bopp, L., 2008. Towards understanding global variability in ocean carbon-13. Glob. Biogeochem. Cycles 22, GB1025. http://dx.doi.org/10.1029/2007GB003037.

Taricco, C., Ghil, M., Alessio, S., Vivaldo, G., 2009. Two millennia of climate variability in the Central Mediterranean. Clim. Past 5, 171–181.

Thorrold, S.R., Campana, S.E., Jones, C.M., Swart, P.K., 1997. Factors determining δ13C and δ18O fractionation in aragonitic otoliths of marine fish. Geochim. Cosmochim. Acta 61, 2909–2919.

Touratier, F., Goyet, C., 2009. Decadal evolution of anthropogenic CO_2 in the Northwestern Mediterranean Sea from the mid-1990s to the mid-2000s. Deep-Sea Res. I 56, 1708–1716.

Tudela, S., Sainz-Trápaga, S., Cermeño, P., Hidas, E., Graupera, E., Quílez-Badia, G., 2011. Bluefin tuna migratory behavior in the western and central Mediterranean Sea revealed by electronic tags. Collect. Vol.Sci. Pap. ICCAT 66, 1157–1169.

Verburg, P., 2007. The need to correct for the Suess effect in the application of d13C in sediment of autotrophic Lake Tanganyka, as a productivity proxy in the Anthropocene. J. Paleolimnol. 37, 591–602.

White, W.M., 2015. Isotope Geochemistry. Wiley-Blackwell, UK, p. 496.

Woosley, R.J., Millero, F.J., Grosell, M., 2012. The solubility of fish-produced high magnesium calcite in seawater. J. Geophys. Res. 117, C04018. http://dx.doi.org/10.1029/2011JC007599.