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Tracing freshwater provenance in palaeo-lagoons by boron isotopes and relationship with benthic foraminiferal assemblages. A comparison from late Quaternary subsurface successions in Northern and Central Italy

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KEY WORDS - Quaternary lagoon, palaeoenvironment, boron isotopes, benthic foraminifera, geothermal fluids, freshwater provenance.

ABSTRACT - In order to evaluate for the first time the application of boron isotope ($\delta^{11}B$) composition as a proxy for freshwater provenance in transitional environments, measurements taken from mollusk shells from palaeo-lagoonal deposits are presented herein. The mollusk specimens were sampled from late Quaternary lagoonal successions observed in the cores 187-S1 and Vignarca, respectively from drill-sites south of the Po River delta (Northern Italy) and the Cornia River plain (Central Italy). The latter locality occurs nearby the Larderello boron-rich geothermal area.

A freshwater influence in the palaeo-lagoons has been highlighted by benthic foraminiferal analyzes, revealing two main assemblages: the first, dominated by Ammonia tepida *(Cushman, 1926) and* Ammonia parkinsoniana *(d'Orbigny, 1839a), is consistent with high freshwater discharge into the palaeo-lagoon, whereas the second assemblage includes large numbers of Miliolacea, considered as reflecting low freshwater discharge. The inferred salinity variations are possibly constrained between 22‰ and 41‰, based on comparison with modern assemblages observed in various lagoons of the north Adriatic Sea and Tyrrhenian Sea coastal areas (e.g., Zampi & D'Onofrio, 1986; Coccioni, 2000). Additional Sr isotope data, comparable with coeval marine values, are consistent with the salinity range inferred by the fossil assemblages.*

Boron isotopes clearly record a continental-dominated provenance source of boron in the lagoonal mollusks collected from the Vignarca core. The low $\delta^{11}B$ and boron-rich component currently observed in the continental water deriving from the geothermal area, was possibly also present in the freshwater feeding the palaeo-lagoon and gives a unique isotopic imprint to the analyzed samples. Conversely, in the lagoonal mollusks collected from core 187-S1, the $\delta^{11}B$ component reveals a dominant provenance source of boron from seawater. In the analyzed successions, boron isotopes have proved useful for recognizing the provenance of the freshwater feeding the palaeo-lagoon, especially in areas characterized by continental water enriched in boron and showing remarkably different $\delta^{11}B$ values with respect to marine values.

RIASSUNTO - [Studio della provenienza dell'acqua dolce nei paleoambienti lagunari attraverso gli isotopi del boro e relazioni con le associazioni a foraminiferi. Un confronto da successioni tardo quaternarie di sottosuolo dell'Italia Settentrionale e Centrale] - *Misurazioni del rapporto isotopico del boro ($\delta^{11}B$) sono state eseguite su conchiglie di* Cerastoderma glaucum *(Poiret, 1789),* Bittium reticulatum *(Da Costa, 1778) e* Cerithium vulgatum *Bruguière, 1792 raccolte in depositi tardo quaternari di ambiente lagunare, per valutare la loro utilità nel distinguere la provenienza delle acque continentali nei paleoambienti transizionali. Gli esemplari analizzati sono stati prelevati nelle successioni dei sondaggi 187-S1 e Vignarca, perforati rispettivamente a sud del delta del Fiume Po (Italia Settentrionale) e nella pianura del Fiume Cornia (Italia Centrale), una zona quest'ultima ubicata in prossimità dell'area geotermica di Larderello e caratterizzata da fluidi ricchi in boro.*

L'influenza dell'acqua dolce nelle paleolagune è stata evidenziata dall'analisi delle associazioni a foraminiferi bentonici che ha permesso di riconoscere due associazioni: la prima, composta principalmente da A. tepida, A. parkinsoniana, Cribroelphidium *spp.,* Elphidium *spp. e* Haynesina *spp., indica condizioni lagunari fortemente influenzate dall'acqua dolce; la seconda, dominata da Miliolacea, è ritenuta indicativa di scarsa influenza di acque continentali. Queste associazioni sono confrontabili con quelle localmente presenti nelle lagune attuali del Mediterraneo con salinità compresa fra il 22‰ e il 41‰.*

Le misure del rapporto isotopico dello stronzio hanno fornito valori simili a quelli registrati in altri carbonati biogenici marini e nell'acqua di mare. Dati di letteratura indicano che ciò è compatibile con la salinità delle paleolagune desunta dalle associazioni a foraminiferi.

I rapporti isotopici del boro sono invece molto diversi nelle paleolagune dei due sondaggi, in particolare i valori di $\delta^{11}B$ delle conchiglie raccolte nel sondaggio Vignarca sono negativi e notevolmente più bassi di quelli registrati nei carbonati biogenici marini; mentre i valori di $\delta^{11}B$ misurati negli esemplari del sondaggio 187-S1 sono positivi e sono tendenzialmente confrontabili con quelli riportati in letteratura per i carbonati biogenici marini. Questo indica che il boro presente nelle conchiglie del sondaggio Vignarca proviene principalmente da acque dolci arricchite in boro con basso $\delta^{11}B$. Acque con queste caratteristiche e provenienti dalla vicina zona geotermica di Larderello sono attualmente presenti nell'area del sondaggio Vignarca e probabilmente lo erano anche durante la deposizione della successione lagunare esaminata. Invece i valori del $\delta^{11}B$ provenienti dal sondaggio 187-S1 indicano una provenienza tendenzialmente marina del boro e non forniscono informazioni sulla provenienza dell'acqua dolce nella paleolaguna. Nelle successioni analizzate, il rapporto isotopico del boro appare utile per riconoscere la provenienza dell'acqua dolce nelle paleolagune per le aree caratterizzate da acque continentali arricchite in boro e con $\delta^{11}B$ notevolmente differente rispetto a quello marino.

INTRODUCTION

Recent studies carried out in the Adriatic and Tyrrhenian plains (Central Mediterranean) provide evidence for an alternation of continental, lagoon and shallow marine sediments deposited during the late Quaternary, as response to relative sea-level variations (Mazzini et al., 1999; Amorosi et al., 2003, 2004a, 2009;

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Carboni et al., 2010). Lagoonal sediments, overlying glacial alluvial deposits, are key-stratigraphic intervals of these subsurface successions, marking relative sea-level rise with on-land marine ingression, and periods of rapid environmental changes (retrograding barrier-lagoon-estuary systems), that may involve drainage reorganization in continental areas (e.g., Mazzini et al., 1999; Curzi et al., 2006; Amorosi et al., 2007) and remarkable variation of river discharge in palaeo-lagoon environments (Dinelli et al., 2012).

Lagoonal sediments include foraminiferal assemblages commonly dominated by species of *Ammonia*, *Cribroelphidium, Elphidium*, *Haynesina* and Miliolacea, which record even subtle palaeoenvironmental changes and variations of freshwater influx (e.g., Carboni et al., 2002, 2010; Amorosi et al., 2003; Bergamin et al., 2006). Comparable assemblages have been also observed in modern lagoons from the same areas (e.g., D'Onofrio et al., 1976; Zampi & D'Onofrio, 1986; Coccioni, 2000; Carboni et al., 2009; Frontalini et al., 2011) allowing palaeoenvironmental interpretation based upon recent analogues. In particular, Mediterranean lagoon environments with relatively high freshwater influx are typically dominated by few foraminiferal species, such as *A. tepida, A. parkinsoniana, Haynesina germanica* (Ehrenberg, 1840)*, Elphidium* spp. and *Cribroelphidium* spp.; in contrast, lagoon environments with low freshwater influx commonly show more diversified assemblages (e.g., Albani & Serandrei Barbero, 1990; Coccioni, 2000; Carboni et al., 2009).

The source of freshwater feeding the palaeo-lagoons is often inferred from the geochemical composition of the deposits, that provides sediment provenance related to rivers drainage-basin (e.g., Curzi et al., 2006; Dinelli et al., 2012). This method alone does not add any constraints on the physico-chemical characteristics of palaeo-lagoon water, whereas isotope measurements of biogenic carbonates occurring in these palaeoenvironments may provide useful information, especially when coupled with analyses of fossil assemblages (Reinhardt et al., 1998; Anadón et al., 2002). For instance, δ^{18}O, δ^{13}C and ^{87}Sr/^{86}Sr values are routinely measured on fossil assemblages to evaluate salinity and evaporation (e.g., Ingram & Sloan, 1992; Reinhardt et al., 1998, 2003; Widerlund & Andersson, 2011). Application of boron isotopes is mainly focused on open marine biogenic carbonates for palaeo-pH reconstruction of seawater and for assessing the evolution of atmospheric CO_2 (e.g., Sanyal et al., 2000; Joachimski et al., 2005; Rae et al., 2011) and until now it has not been tested in lagoon palaeoenvironments.

Boron is concentrated in the non-organic parts of shell matrices of mollusks, and its content is related to the boron concentration, pH, and salinity of ambient waters (Furst et al., 1976). Specifically, Roopnarine et al. (1998) documented a positive relationship between B concentration in mollusk shells and salinity of nearshore environment (<6 m water depth). Boron isotopes are fractionated during shell formation, ^{10}B being preferentially incorporated with respect to ^{11}B in biogenic carbonates (Vengosh et al., 1991; Hemming & Hanson, 1992). As B isotopic composition of marine carbonates is controlled by pH and δ^{11}B value of the water in which carbonates precipitated (Lemarchand et al., 2002), lagoon biogenic-carbonates possibly reflect pristine B isotopes of the mixture fresh- and seawater of the lagoon.

This study aims to investigate the relationships between foraminiferal assemblages and boron isotopic composition of mollusk shells from Quaternary lagoonal deposits, in order to trace the provenance of freshwater feeding the palaeo-lagoons. Additional Sr isotope measurements have been performed to integrate the foraminiferal-based palaeoenvironmental reconstruction.

GEOLOGICAL SETTING AND SUBSURFACE STRATIGRAPHY

Data analyzed in this work were obtained from two late Quaternary lagoonal successions located in Fig. 1:

a) the Cornia River alluvial plain, an area characterized by freshwater enriched in B (Minissale, 1991; Bianchini et al., 2005; Pennisi et al., 2006, 2009) located in the proximity of the Larderello geothermal fields;

b) the Po River coastal plain, an area with relatively low B concentration in freshwater (Tartari & Camusso,

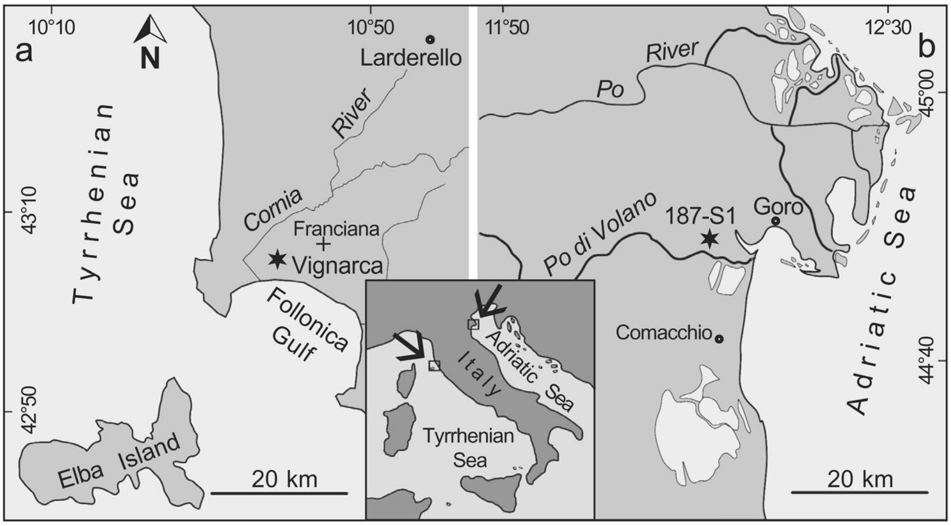

Fig. 1 - Location map of cores Vignarca (a) and 187-S1 (b). The position of the core Franciana (+) is included.

1988), where the late Quaternary succession was mainly deposited under a glacio-eustatic control (Amorosi et al., 2004a).

The depositional history and chronological framework of the studied successions and relationships with late Quaternary climate and sea-level variations are reported in Amorosi et al. (2004b, 2007) and Bondesan et al. (2006).

Cornia River alluvial plain

The Cornia River alluvial plain is located in western Tuscany, facing the Elba Island (Fig. 1a). It represents the infilling of a tectonic depression originated by the extensional regime developed since the Tortonian and still active, as a consequence of the Tyrrhenian sea rifting (Carmignani et al., 1994). The Cornia River catchment area includes part of the geothermal field of Larderello, in which hydrothermal circulation is associated to the worldwide known Larderello field, that developed since the Early Pliocene, in response to magmatic intrusion in the area (D'Amore et al., 1987; Dini et al., 2005). Before the industrial exploitation of the field, B-rich hydrothermal fluids emerged at the surface and gave rise to the natural manifestation in the so called "boraciferous region", well known since the Roman times. Isotope data and concentration of B and Sr in the groundwater of the Cornia River alluvial plain (Pennisi et al., 2006) show, in most samples, δ^{11}B and ^{87}Sr/^{86}Sr values lower than marine waters. In particular, at less than 3 km from the analyzed succession (samples 161, 273, 352, 749, and 775 in Pennisi et al., 2006), δ^{11}B is lower than marine values of about +40‰ (Lécuyer et al., 2002; Gonfiantini et al., 2003) and ranges between -6.1‰ and -2.3‰. In contrast ^{87}Sr/^{86}Sr ratios are well constrained between 0.70870 and 0.70898 and approach the marine water ratio of 0.70917 (e.g., McArthur, 1994). In this area, high concentration of B (up to 6.6 mg L-1) and relatively low concentrations of Sr (up to 1.7 mg L-1) were recorded in bicarbonate waters (Pennisi et al., 2006).

Po River coastal plain

The Po River plain represents the infilling of a perisutural basin bounded by two mountain chains, the Apennine to the south and the Alps to the north.

The Po River coastal plain is located in the eastern part of the Po River plain (Fig. 1b), which extends till the Adriatic Sea. It is infilled by 700-800 m-thick sediments accumulated since the Pliocene in a NE-verging thrusting of the Apennine front towards Adriatic foreland (Pieri & Groppi, 1981; Castellarin & Vai, 1986). Although evidences of compressional tectonic are shown by seismic data (e.g., Pieri & Groppi, 1981), the late Quaternary succession of the Po Plain coastal area has been deposited under relatively undisturbed conditions (Amorosi et al., 2003). Detailed analyses of tens of continuously cored boreholes, promoted by the Geological Survey of the Regione Emilia-Romagna, reveal that this succession consists of a cyclic stacking pattern of shallow-marine to continental sediments formed under a predominantly glacio-eustatic control (Amorosi et al., 2003, 2004a; Curzi et al., 2006).

Boron isotope data of the Po River water are not available, however B concentrations were measured by Tartari & Camusso (1988) and show low values (up to 0.07 mg L^{-1}) that are consistent with data from world's main rivers (Lemarchand et al., 2002). The ^{87}Sr/^{86}Sr ratio of the Po River water (0.7097, Brass, 1976; Müller et al., 1990) is slightly higher than Sr isotope ratio of seawater (0.70917, McArthur, 1994) and possibly reflects the presence of silicate and metamorphic rocks in the western part of the catchment area (Dinelli & Lucchini, 1999).

MATERIAL AND METHODS

The analyzed succession of the Cornia River alluvial plain came from the 105 m long core Vignarca, drilled 3 km landward from the Follonica Gulf (Fig. 1a). This core consists of rhythmical alternation of floodplain and fluvial channel deposits, with the local occurrence of crevasse and levee sediments (Amorosi et al., 2004b). A single interval recording lagoonal clays, with subordinate fine sands and common mollusk shells, was recognized between 21 and 18 m core depth (Fig. 2a). Associated pollen data reveal an interglacial climate conditions attributed to the Marine Isotope Substage 5.5 (Amorosi et al., 2004b).

The analyzed succession of the Po River plain came from core 187-S1, drilled in proximity of the Po River delta, 6 km landward from the shoreline (Fig. 1b). This core is 188 m long and includes two thick continental bodies, intercalated by three paralic and marine intervals, corresponding to the last two interglacial phases (Marine Isotope Substage 5.5 and Marine Isotope Stage 7) and to the Holocene post-glacial interval (e.g., Amorosi et al., 2007; Rossi & Vaiani, 2008). The lagoonal deposits are present in the lower part of the core, between 176 and 170 m core depth (Fig. 2b) and have been attributed to Marine Isotope Stage 7 (Bondesan et al., 2006). These deposits include clays, silts and fine sands; mollusk shells are locally abundant.

Nineteen samples from core Vignarca and 72 samples from core 187-S1 were treated for palaeontological analyses as follows: 1) dried at 60°C for 8 hours, 2) soaked in water, 3) wet sieved at 63 µm, 4) dried again and 5) dry sieved at 125 µm. Samples are deposited in the Dipartimento di Scienze Biologiche, Geologiche e Ambientali, University of Bologna.

The grain size fraction between 63 and 125 µm was qualitatively analysed to describe the foraminiferal assemblages, substantially showing the same taxa observed in the coarser fractions, although with a remarkable amount of juvenile specimens. Thirty-six samples (14 from core Vignarca and 24 from core 187-S1), which contain perfectly preserved benthic foraminifera in the size fraction >125 µm, were split into small aliquots of at least 300 specimens and counted (details in Supplementary online material: Vaiani_Pennisi_fossil counts.xls). This size fraction was selected to avoid juvenile forms of uncertain classification in quantitative analysis (e.g., Cann et al., 1988; Donnici & Serandrei Barbero, 2002).

The foraminiferal distribution in core Vignarca within the size fraction > 175 µm was reported in Amorosi et al. (2004b); in the present work these foraminiferal assemblages are recounted in the size fraction > 125 µm, in order to compare the foraminiferal distribution of both cores using the same size fraction. Quantitative data from three new samples are added.

The identification of the foraminifera is supported by the original descriptions and follows selected papers, such as Jorissen (1988), Albani & Serandrei Barbero (1990), Hayward et al. (1997) and Fiorini & Vaiani (2001). As commonly reported in other palaeoenvironmental studies on these areas (e.g., Aguzzi et al., 2007; Rossi & Vaiani, 2008; Amorosi et al., 2009), *A. tepida* and *A. parkinsoniana* were grouped into a single taxonomic unit for the common recognition of intermediate morphotypes (e.g., Jorissen, 1988; Rasmussen, 2005). Results of quantitative analyses were reported in a matrix for cluster analyses including the eight most abundant taxa and taxa groups (those > 4% in at least one sample, according to other studies on benthic foraminiferal assemblages of transitional environments and palaeoenvironments; e.g., Coccioni et al., 2009; Carboni et al., 2010; Elshanawany et al., 2011), and was processed with a palaeontological statistical program (PAST-PAlaeontological STatistic-ver. 2.17 by Hammer et al., 2001).

A hierarchical R-mode cluster analysis was performed to define the main foraminiferal assemblages in the cored successions. The robustness of the foraminiferal groups was tested by running selected unweighted pair group (UPGMA) cluster analyses with different similarity indexes, consistent with the nature of our data. Horn's (1966) modified version of Morisita's (1959) index was selected, because it provides the most realistic output data, down weighting the more abundant species. A Q-mode cluster analysis using Bray-Curtis distance (1957) was performed in order to produce groups of samples with similar foraminiferal assemblage.

Boron and Sr isotopic composition were not determined on foraminifera because of insufficient amount of perfectly preserved specimens in the samples necessary to perform the Thermal Ionization Mass Spectrometry (TIMS) analyses. Consequently, these analyses have been performed on selected mollusk species: *C. glaucum, B. reticulatum* and *C. vulgatum*, commonly found in Mediterranean lagoons (e.g., Koutsoubas et al., 2000; Sfriso et al., 2001). In particular, *C. glaucum* is extensively used to investigate palaeoenvironmental conditions (Nikula & Vainola, 2003), and as a proxy for bioaccumulation of heavy metals in the soft tissue (Machreki-Ajmi & Hamza-Chaffai, 2006).

Isotopic analyses in the core Vignarca were performed at -19.60 m and -20.60 m core depth. Perfectly preserved bivalve shells of *C. glaucum* were collected in both samples, a gastropod shell of *C. vulgatum* was collected uniquely at -20.60 m core depth, whereas the other sample includes only rare and poorly preserved gastropods, suggesting a possible transport from nearby environments or reworking of these specimens.

Isotopic analyses in core 187-S1 were performed in two closely spaced samples at -174.80 and -174.90 m core depth, as both samples yield perfectly preserved specimens of *C. glaucum*. The gastropod *C. vulgatum* was not observed in this core, therefore a specimen of *B. reticulatum*, a different species from the same family (Cerithiidae), was analyzed at -174.80 m core depth.

The surface of the analysed specimens was first mechanically cleaned using an electric wheel; samples were then crushed to small fragments and ultrasonically cleaned in ultrapure water to remove the organic and pelitic residues from the carbonate. Washing procedure was repeated till the sample was completely clean when observed under binocular microscope. Sample was then dried and finally ground to a fine powder in an agate mortar. For B isotopic composition, 500 mg of sample were fused using ultra-purified K_2CO_3, and B separated using the ion exchange procedure described in Tonarini et al. (1997, 2003). Following this procedure, no residual organics remained. Boron isotopic composition was determined by positive TIMS, on a VG Isomass 54E. Isotopic fractionation associated with the mass-spectrometer analysis was corrected using repeated analyses of the NIST SRM 951 standard taken through the full chemical procedure, which gave a $^{11}B/^{10}B = 4.0541 \pm 0.002$. Boron isotopic composition is reported in the conventional delta notation ($\delta^{11}B$), as permil deviation from the certified composition of the NIST SRM 951 standard. Duplicate analyses of $\delta^{11}B$ were performed, with the exception for the specimen collected at -174.80 m in core 187-S1 because of scanty material. Boron concentration was determined by isotope dilution using the NIST SRM 952 standard (^{10}B enriched).

For Sr isotopic analyses, 50 mg of material was dissolved in 2.5N ultrapure HCl, centrifuged, and Sr separated from the matrix through conventional cation-exchange procedures. Strontium isotopic compositions were measured on a Finnigan MAT 262V multi-collector mass spectrometer. Measured $^{87}Sr/^{86}Sr$ ratios were normalized to $^{86}Sr/^{88}Sr = 0.1194$. Replicate analyses of the SRM-NIST 987 standard measured in the same period gave an average ratio of 0.710244, with the error expressed as 2 standard deviation (2s.d.) of $\pm 15 \times 10^{-6}$ (n = 8).

As no significant variations in seawater Sr-isotope composition have been recorded from the time of deposition of the lagoon successions, measured values in this work are compared with $^{87}Sr/^{86}Sr$ ratio of modern seawater (0.70917, e.g., McArthur, 1994).

FORAMINIFERAL ASSEMBLAGES

Foraminiferal distribution in the analyzed successions are reported in Fig. 2; the R-mode CA defines two clusters that represent two main assemblages (Fig. 3a). Specifically, Cluster Ra includes *A. tepida*, *A. parkinsoniana*, *Cribroelphidium* spp., *Elphidium* spp. and *Haynesina* spp.; whereas Cluster Rb, is dominated

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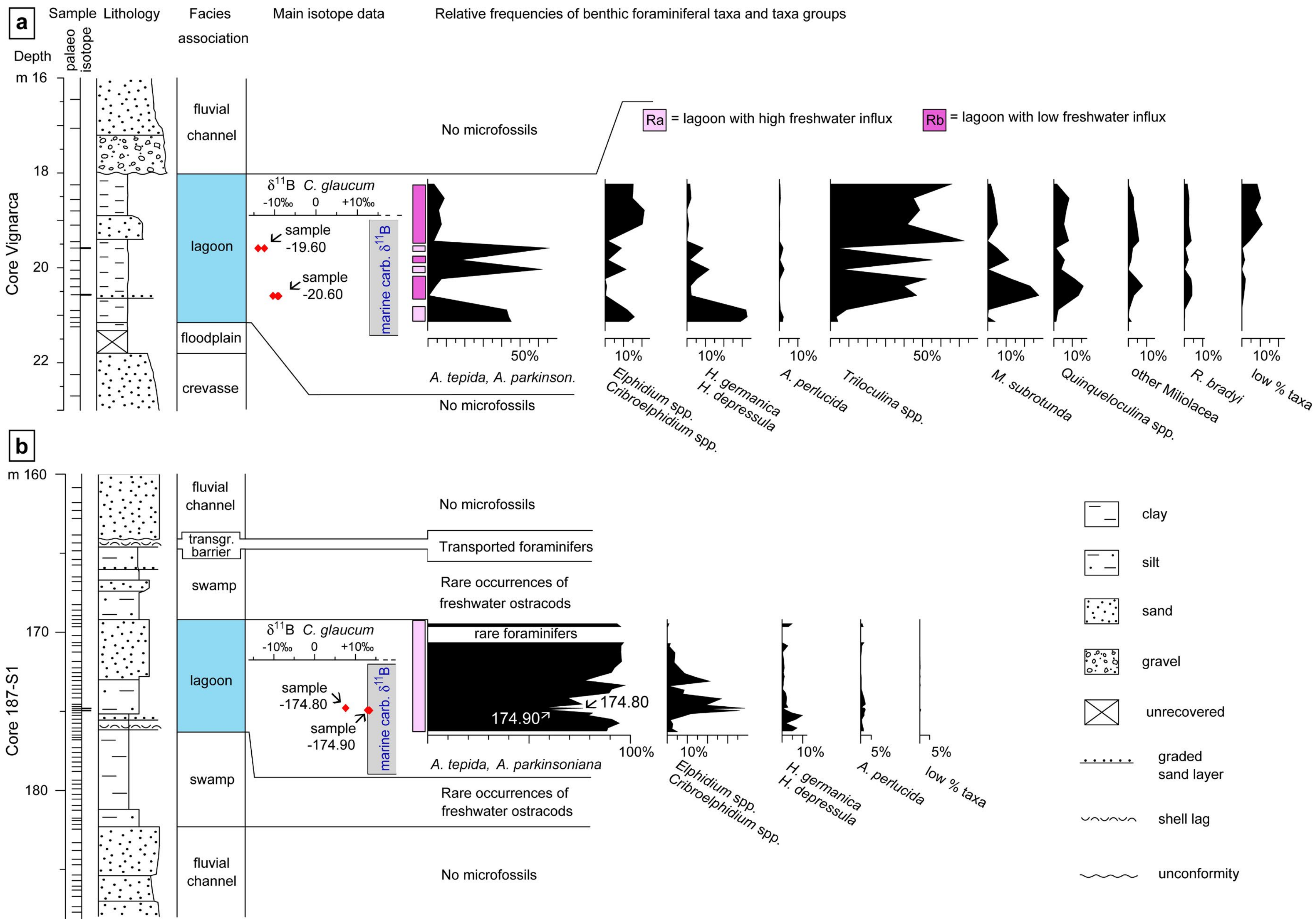
Fig. 2 - Stratigraphy, foraminiferal distribution and $\delta^{11}B$ values of the analyzed parts of the cores (a: core Vignarca; b: core 187-S1). Two foraminiferal assemblages are observed: the first (Ra), dominated by *A. tepida* and *A. parkinsoniana*, is indicative of high freshwater discharge in the palaeo-lagoon; the second (Rb), dominated by Miliolacea, mainly *Triloculina* spp., is considered to reflect low freshwater discharge. A rhythmical alternation of high and low freshwater influx is recorded in the palaeo-lagoon of core Vignarca, whereas only high freshwater influx is observed in core 187-S1. The low values of $\delta^{11}B$ measured in *C. glaucum* of core Vignarca are indicative of a continental-dominated source of boron in the palaeo-lagoon, in contrast, the higher $\delta^{11}B$ values recorded in core 187-S1 reveal a dominant source of boron from seawater. Facies associations and ostracod data after Amorosi et al. (2004b, 2007) and Bondesan et al. (2006).


a
Sample
palaeo
isotope
Lithology
Facies association
Main isotope data
Relative frequencies of benthic foraminiferal taxa and taxa groups
Depth
m 16
18
20
22
Core Vignarca
fluvial channel
lagoon
floodplain
crevasse
No microfossils
Ra = lagoon with high freshwater influx
Rb = lagoon with low freshwater influx
δ11B C. glaucum
-10‰ 0 +10‰
sample -19.60
sample -20.60
marine carb. δ11B
50%
A. tepida, A. parkinson.
10%
Elphidium spp. Cribroelphidium spp.
H. germanica H. depressula
A. perlucida
Triloculina spp.
M. subrotunda
Quinqueloculina spp.
other Miliolacea
R. bradyi
low % taxa
b
m 160
170
180
Core 187-S1
transgr. barrier
swamp
Transported foraminifers
Rare occurrences of freshwater ostracods
rare foraminifers
174.80
174.90
sample -174.80
sample -174.90
100%
A. tepida, A. parkinsoniana
5%
clay
silt
sand
gravel
unrecovered
graded sand layer
shell lag
unconformity

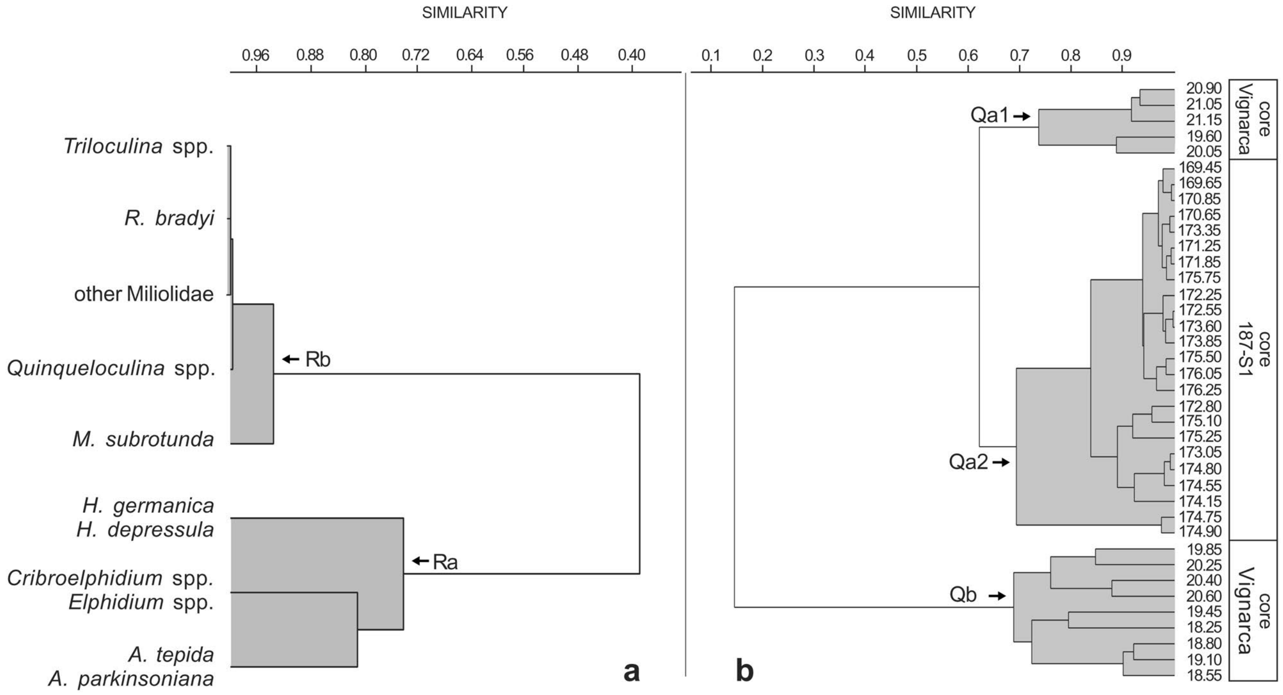


Fig. 3 - Dendrogram classifications from cluster analysis based on the relative abundance of the eight most common foraminiferal taxa and taxa groups. a) Foraminiferal assemblages obtained by R-mode analysis. The two clusters are considered to reflect assemblages indicative of high (Ra) and low (Rb) freshwater influence in the palaeo-lagoon environment. b) Sample associations obtained by Q-mode analysis show clusters Qa and Qb grouping samples with assemblage Ra and Rb, respectively. Qa has been also divided in two subclusters that include samples with assemblage Ra from core Vignarca (Qa1) and 187-S1 (Qa2). Core 187-S1 is uniquely characterized by high freshwater influence in the palaeo-lagoon, in contrast core Vignarca shows both high and low freshwater influence.

by Miliolacea in association with *Rosalina bradyi* (Cushman, 1915). These assemblages are substantially consistent with the dendrogram of sample groups obtained by Q-mode analyses (Fig. 3b) that shows clusters Qa and Qb grouping samples with assemblage Ra and Rb, respectively. Minor differences in the foraminiferal distribution recorded in samples of Cluster Qa provide evidence for two Subcluster Qa1 and Qa2 (Fig. 3b), that include samples with assemblage Ra from cores Vignarca and 187-S1, respectively. In details, core Vignarca includes both assemblages (Figs 2a, 3a): the first one (Ra) is dominated by *A. tepida* and *A. parkinsoniana,* in association with *H. germanica*, *Cribroelphidium* spp. (mainly *Cribroelphidium oceanensis* [d'Orbigny, 1826]), *Elphidium* spp., *Aubignyna perlucida* (Heron-Allen & Earland, 1913), *Haynesina depressula* (Walker & Jacob, 1798) and Miliolacea. The second assemblage (Rb) is dominated by Miliolacea, mainly *Triloculina rotunda* d'Orbigny, 1826, with subordinate *Miliolinella subrotunda* (Montagu, 1803)*, Quinqueloculina laevigata* d'Orbigny, 1839b*, Quinqueloculina* cf. *seminulum* (Linnaeus, 1758)*, Triloculina schreiberiana* d'Orbigny, 1839a and *Triloculina* spp. (Figs 2a, 3a). Other species of Miliolacea (reported as "other Miliolacea" in Fig. 2a) are rare and consist mainly of *Adelosina elegans* (Williamson, 1858)*, Adelosina* sp., *Cycloforina costata* (d'Orbigny, 1826)*, Siphonaperta aspera* (d'Orbigny, 1826) and *Spiroloculina communis* Cushman & Todd, 1944. Low amounts of other foraminifera, such as *A. tepida, A. parkinsoniana, R. bradyi, Cribroelphidium* spp. (mainly *Cribroelphidium granosum* [d'Orbigny, 1846] and *Cribroelphidium lidoense* [Cushman, 1936]) and *Elphidium* spp. (mainly *Elphidium fichtellianum* [d'Orbigny, 1846]), are observed. Samples with assemblage Rb from the upper part of the succession show few specimens of several other species (low % taxa in Fig. 2a), mainly *Lobatula lobatula* (Walker & Jacob, 1798), *Nonion* sp., *Peneroplis pertusus* (Forskål, 1775), *Peneroplis* sp. and *Planorbulina mediterranensis* d'Orbigny, 1826.

Core Vignarca is characterized by a cyclic alternation of both assemblages up to -19.60 m core depth; the upper part of the palaeo-lagoon succession solely shows assemblage Rb (Figs 2a, 3a).

Foraminiferal distribution in the analysed part of the core 187-S1 (Fig. 2b) includes uniquely assemblage Ra and it is dominated by *A. tepida* and *A. parkinsoniana. Elphidium* and *Cribroelphidium* species (mainly *C. granosum* and *C. oceanensis*) are common (>20%) in five samples between -174.90 m and -173.05 m core depth*,* in contrast other species (*A. perlucida, H. germanica* and *H. depressula*) are substantially rare. This assemblage is similar to assemblage Ra found in core Vignarca, although *A. tepida* and *A. parkinsoniana* are on average more abundant and Miliolacea were not observed.

PALAEOENVIRONMENTAL INTERPRETATION

Ammonia tepida and *H. germanica* dominate the assemblage of the Goro Lagoon in the Po River delta (Coccioni, 2000), an area with high freshwater discharge and average bottom-water salinity of 26.9‰ (Coccioni, 2000). Both species are typically euryhaline, although their relative abundance may vary in response to food source; specifically *A. tepida* is known to feed on many sources, including refractory material (Modley et al., 2000), in contrast *H. germanica* prefers labile organic material such as diatoms (de Nooijer et al., 2007). *Ammonia tepida, A. parkinsoniana, H. germanica* and selected *Cribroelphidium* species (including *C. granosum* and *C. oceanensis*) are commonly present with high frequencies in shallow lagoons and coastal lakes (less than 7 m depth) of the Tyrrhenian coast with salinity ranging between 22‰ and 37‰ (e.g., Carboni et al., 2009; Frontalini et al., 2009). *Haynesina depressula, Q. laevigata, Q. seminulum* and *R. brady* are locally recognized in transitional environments within assemblages dominated by *A. parkinsoniana* and *H. germanica*, with relatively lower amount of *A. tepida* (Carboni et al., 2009). *Aubignyna perlucida* is commonly observed in north Adriatic lagoons and deltaic areas (e.g., Jorissen, 1988; Coccioni et al., 2009). *Elphidium fichtellianum* and *C. lidoense* are shallow marine species (Jorissen, 1988; Hayward et al., 1997).

The observed species of Miliolacea are commonly found in Mediterranean lagoons and shallow-marine environments (e.g., Zampi & D'Onofrio, 1984; Sgarrella & Moncharmont Zei, 1993; Murray, 2006). *Triloculina rotunda* is abundant (up to 24%) in a lagoon of the Tyrrhenian coast located 75 km south of the Vignarca core, at which seasonal salinity varies between 30‰ and 41‰ (Zampi & D'Onofrio, 1986). Foraminiferal assemblages with high amount of *Triloculina* and *Pseudotriloculina* species are present in confined lagoons possibly with hypersaline water or shell bottom indicating the availability of calcium carbonate (Zaninetti, 1982; Debenay et al., 2001; Debenay & Guillou, 2002).

Adelosina elegans, L. lobatula, P. pertusus, P. mediterranensis, R. bradyi, S. aspera, S. communis and *T. schreiberiana* are taxa commonly found as epiphytic or typical of shallow marine and transitional environments with vegetation cover at the bottom (Langer, 1988; Sgarrella & Moncharmont Zei, 1993; Fiorini & Vaiani, 2001).

Foraminiferal assemblages dominated by *A. tepida* and *A. parkinsoniana* are found in both cores and reflect deposition in lagoon subjected to remarkable freshwater influx and salinity constrained between 22‰ and 37‰ (according to similar assemblages found in lagoons and coastal lakes of the Tyrrhenian coast by Carboni et al., 2009 and Frontalini et al., 2009). The relatively higher amount of *H. germanica* found in this assemblage of core Vignarca is considered to reflect major availability of labile organic material in the palaeoenvironment. The higher frequencies of *A. tepida* and *A. parkinsoniana* paralleled by lower amount of *H. germanica* recorded in core 187-S1 are possibly consistent with an increasing freshwater discharge and refractory material, although the local occurrence of remarkable amount of *Cribroelphidium* spp. and *Elphidium* spp. would suggest temporary conditions of slightly lower freshwater discharge.

Foraminiferal assemblages dominated by Miliolacea are interpreted to have formed within a confined lagoon with low freshwater influx and possibly high salinity and availability of calcium carbonate. The occurrence of epiphytic taxa, mainly in the upper part of core Vignarca, is considered to reflect vegetation cover at the bottom.

The benthic foraminiferal distribution recorded in the core Vignarca reveals, in the lower part, the rhythmical alternations of high and low freshwater influx in the palaeo-lagoon, replaced by conditions of substantially low freshwater influx (Fig. 2a); in contrast the relatively homogeneous foraminiferal assemblages observed in core 187-S1 are indicative of high freshwater influx (Fig. 2b).

ISOTOPE ANALYSES

Sample selection for isotope analyses

In order to test the importance of B isotopes as proxy of freshwater provenance and the relationship between B and Sr isotopes and palaeontological data, the analysis has been focused on samples with foraminiferal assemblages particularly representative of the degree of freshwater influence.

Two samples were collected in both cores, specifically in core Vignarca (Fig. 2a), sample at -19.60 m includes the highest abundance of the euryhaline foraminifera *A. tepida* and *A. parkinsoniana* (assemblage Ra, Fig. 3a), which is considered to reflect the highest freshwater influence. In contrast, the second sample (-20.60 m) corresponding to the Rb assemblage (Fig. 3a), includes the lowest concentration of *Ammonia* species, which is consistent with relatively low freshwater influence.

In core 187-S1 (Fig. 2b), sample at -174.80 m is strongly dominated by specimens of *A. tepida* and *A. parkinsoniana* (76.6%), indicating high freshwater discharge in the palaeo-lagoon, whereas sample -174.90 m includes relatively lower amount of *Ammonia* species (59.4%) and abundant *Cribroelphidium* spp. and *Elphidium* spp. (36.1%), suggesting a possible increase of marine influence.

Results

In both cores, the $^{87}Sr/^{86}Sr$ ratios (Tab. 1) show minor variations in the analyzed specimens ($^{87}Sr/^{86}Sr$ ratio: 0.70914-0.70917) and these values are comparable considering data uncertainty (about $\pm 2x10^{-5}$).

In core Vignarca, $\delta^{11}B$ ranges from -9.1‰ to -20‰ and the measured B contents are 4.4 and 12.2 μg/g. In core 187-S1, $\delta^{11}B$ ranges from 1.3‰ to 13.4‰ and the measured B contents are 3.2 and 5.9 μg/g (Tab. 1).

Boron characteristics show significant differences in the two sites, and within each given core, specifically:

1. $\delta^{11}B$ values and boron content measured in gastropods (*B. reticulatum* and *C. vulgatum*) are lower than those recorded in the bivalves (*C. glaucum*) from the same sample. Specifically in sample -20.60 m of core Vignarca, the $\delta^{11}B$ of *C. vulgatum* are -20.0‰ and -19.6‰ and the boron content is 4.4 μg/g. In this sample, the $\delta^{11}B$ of *C. glaucum* ranges from -10.4‰ to -9.1‰ and the boron content is 12.2 μg/g. In sample -174.80 m of core 187-S1, the $\delta^{11}B$ of *B. reticulatum* is 1.3‰ and the boron content is 0.44 μg/g. In this sample, the $\delta^{11}B$ of *C. glaucum* is 5.0‰ and the boron content is 5.9 μg/g.
2. $\delta^{11}B$ values in specimens of core Vignarca are remarkably lower (from -20.0‰ to -9.1‰) than those measured in specimens of core 187-S1 (from 1.3‰ to 13.4‰).

Core / sample / palaeoenvironment / analyzed taxon		δ^{11}B	permil rsd	B µg/g	^{87}Sr/^{86}Sr	error x10^{-6}	isotopic results
Core Vignarca	sample at -19.60 m core depth: lagoon with high freshwater influx						
	analyzed taxon: *Cerastoderma glaucum*	-14.1	0.50				B from FW
	C. glaucum (analyses repeated)	-12.6	0.62				B from FW
	sample at -20.60 m core depth: lagoon with low freshwater influx						
	analized taxon: *Cerastoderma glaucum*	-9.1	0.60	12.2	0.709160	±16	B from FW; Sr from SW
	C. glaucum (analyses repeated)	-9.2	0.60				B from FW
	C. glaucum (analyses repeated)	-10.4	0.37				B from FW
	analyzed taxon: *Cerithium vulgatum*	-19.6	0.43	4.4	0.709140	±20	B from FW; Sr from SW
	C. vulgatum (analyses repeated)	-20.0	0.65				B from FW
Core 187-S1	sample at -174.80 m core depth: lagoon with high freshwater influx						
	analyzed taxon: *Cerastoderma glaucum*	5.0	0.52	5.9	0.709141	±15	B from FW and SW; Sr from SW
	C. glaucum (analyses repeated)				0.709142	±13	Sr from SW
	analyzed taxon: *Bittium reticulatum*	1.3	0.44	3.2	0.709175	±18	B from FW and SW; Sr from SW
	sample at -174.90 m core depth: lagoon with high freshwater influx						
	analyzed taxon: *Cerastoderma glaucum*	13.1	0.44				B from SW
	C. glaucum (analyses repeated)	13.4	1.02				B from SW

Tab. 1 - Boron and strontium isotope data and boron concentrations in analyzed mollusk shells. Boron isotopes and concentration record a continental-dominated source of boron from geothermal areas in lagoonal mollusks of the core Vignarca; in contrast data from core 187-S1 reveal a dominant source of boron from seawater, with a possible partial contribution of B from freshwater in mollusk shells collected in sample at -174.80 m. The measured ^{87}Sr/^{86}Sr ratios from both cores are substantially comparable with marine water values. The dissolved Sr appears dominated by the marine components. Repeated isotope analyses have been performed in most specimens. FW: freshwater; SW: seawater.

3. The B content of *C. glaucum* from core Vignarca is higher (12.2 µg/g) than that observed in this species from core 187-S1 (5.9 µg/g).

4. δ^{11}B values in *C. glaucum* from core Vignarca, collected in deposits characterized by high freshwater influx (-14.1‰ and -12.6‰), are lower than those collected in deposits with low freshwater influx (-10.4‰, -9.2‰ and -9.1‰).

RELATIONSHIP BETWEEN ISOTOPE DATA AND FORAMINIFERAL ASSEMBLAGES

The measured ^{87}Sr/^{86}Sr ratios from both cores are substantially consistent with modern marine water (0.70917, e.g., McArthur, 1994). The dissolved Sr appears dominated by the marine components, consequently these values are not useful for palaeosalinity evaluation. The low Sr concentration in freshwater and the relatively subtle differences in ^{87}Sr/^{86}Sr ratios between fresh- and seawater, currently characterizing the continental water of Po and Cornia plains, were probably present at the time of deposition of lagoon sediments and hamper any measurable influence on Sr isotope ratios in palaeo-lagoon mollusks. The observed results are in agreement with those reported in other biogenic carbonates from transitional environments, whose Sr isotopic signature deviates from the marine one only when salinity drops down to values (12‰ for weighted world average river according to Bryant et al., 1995), remarkably lower than that estimated by foraminiferal assemblages.

The differences observed in the B isotope data of gastropods and bivalves collected in the same sample (core Vignarca sample at -20.60 m and core 187-S1 sample at -174.80 m) are considered to reflect species-dependent B isotope effects and B fixation, as already evidenced in other biogenic carbonates (e.g., Sanyal et al., 1996; Honisch et al., 2004). This offset hampers reliable interpretations based on comparison of data from different species, and, in addition, the low number of analyses carried out on gastropods may not provide certain inferences. Consequently, the interpretations of

B isotope data are mainly based upon measurements collected in the species *C. glaucum*, analyzed in all samples (Tab. 1).

δ^{11}B values in core Vignarca are negative and remarkably lower than those recorded in marine biogenic carbonates, while in core 187-S1, δ^{11}B values are positive, and consistent to the range +13‰ to +27‰ reported for marine biogenic carbonates (e.g., Vengosh et al., 1991; Lécuyer et al., 2002; Pagani, et al., 2005). These results indicate that the dominant source of B incorporated in mollusk shells of core Vignarca is continental and probably associated to freshwater enriched in B, with δ^{11}B much lower than the seawater value of +40‰ (Lécuyer et al., 2002; Gonfiantini et al., 2003). The low B concentration of the Po River water (Tartari & Camusso, 1988) and the unavailability of δ^{11}B measurements on this continental water hamper a clear detection of the continental source of B in the palaeo-lagoon of core 187-S1. In particular, the B isotope composition of *C. glaucum* collected at -174.90 m (Tab. 1) is included in the lower part of the range reported for marine biogenic carbonates (Vengosh et al., 1991; Pagani, et al., 2005), revealing a dominant source of B from seawater. The lower δ^{11}B value of *C. glaucum* in sample -174.80 m (Tab. 1) suggests a partial contribution of B from freshwater with isotopic composition possibly constrained in the lower part of the range of the world's main rivers (δ^{11}B from -6‰ to 42.8‰, Lemarchand et al., 2000). The possible higher influence of continental B observed at -174.80 m is paralleled with the higher freshwater influence revealed by foraminiferal assemblages; in details, sample at -174.80 m includes greater amount of *A. tepida* and *A. parkinsoniana* (76.6%) than those recorded in sample -174.90 m (59.4%; Fig. 2b).

The δ^{11}B range of mollusk specimens from core Vignarca (-9‰ to -20‰) overlaps the δ^{11}B values reported for fine sediments (-9.5‰ to -18.8‰) from the same area (core Franciana in Fig. 1a, Pennisi et al., 2009). According to the model reported by Pennisi et al. (2009), it could be suggested that similar conditions have governed both clay minerals equilibration with B bearing fluids and mollusk shells precipitation from a boron-rich palaeo-lagoon water. At this stage, data on mollusk shells are too scarce, and parameters driving the B isotope fractionation in lagoon environments are poorly constrained for a quantitative discussion in terms of δ^{11}B, pH and salinity of the palaeo-lagoon water in the analyzed cores. However, the available measurements from core Vignarca clearly record the influence of freshwater from nearby geothermal area with a specific boron isotopic signature, at time of carbonate precipitation.

The continental origin of B observed in core Vignarca is also evidenced by the comparison of δ^{11}B measured in specimens of *C. glaucum* deposited under different lagoon conditions: in lagoon with high freshwater influence, as highlighted by foraminiferal assemblages (Fig. 2a), the δ^{11}B is remarkably lower than that measured in lagoon with low freshwater discharge (Fig. 2a; Tab. 1). This difference is considered to reflect the different amount of continental water from geothermal areas in the palaeo-lagoon. In contrast, the palaeo-lagoon of core 187-S1, deposited under high freshwater discharge (Fig. 2b), but in an area not influenced by geothermal activity, shows δ^{11}B values remarkably higher (Tab. 1).

As discussed for boron isotopes, boron concentration of *C. glaucum* from core Vignarca also supports the hypothesis of higher B entrapment in biogenic carbonates under the influence of B-enriched fluids, as shown by its significant higher B content with respect to that measured in specimens of *C. glaucum* living in B-impoverished waters, as indicated by the results obtained in core 187-S1 (Tab. 1).

CONCLUSION

1. In the Cornia and Po rivers plains the palaeoenvironmental evolution of the lagoon successions, defined by foraminiferal assemblages, reveals conditions of high and low freshwater discharge in the palaeo-lagoons.
2. The B isotopic imprint observed in mollusk shells (*C. glaucum* and *C. vulgatum*) from the palaeo-lagoon of core Vignarca is consistent with discharge of B-enriched and low δ^{11}B continental water from the nearby geothermal area at time of carbonate precipitation and allows a clear recognition of freshwater provenance. In contrast, B isotope data from *C. glaucum* and *B. reticulatum* in the palaeo-lagoon of Po River plain show a dominant source of B from seawater preventing to detect the provenance of freshwater.
3. This research encourages further applications of B isotopes in transitional (palaeo) environments to confirm the applicability of this proxy for detecting freshwater provenance in geological settings that drive B mobilization, as geothermal and magmatic ones.

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