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Influence of the upwelling events on the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of the benthic bivalve shells of the South Western Continental Margin of India

Josia Jacob¹ · Prosenjit Ghosh^{1,4} · K. U. Abdul Jaleel² · B. R. Smitha² · K. R. Abhilash³ · V. N. Sanjeevan²

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Abstract In this study, the influence of the spatial and temporal variability of upwelling intensity and the associated biological productivity observed during different phases of summer monsoon along the southwestern continental margin of India (SWCMI) on the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of the inorganic biogenic carbonate shells was investigated. Multispecies benthic bivalve shells (1–5 mm) separated from ten surface sediment samples of SWCMI (off 12°N, 10°N and 9°N) collected during the onset (OSM) and peak (PSM) phase of the summer monsoon of 2009 were analysed for $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$. Sea surface temperature along the study region indicates prominent upwelling in PSM than in OSM. A comparison of analytical and predicted values for $\delta^{18}\text{O}$ in the bivalve shells confirmed their in situ origin during both the sampling periods. During PSM, the $\delta^{13}\text{C}$ values in the benthic bivalve shells were more depleted in ^{13}C than during OSM which recorded lower values of $\delta^{13}\text{C}$ in dissolved inorganic carbon of bottom waters expected in the study region in PSM due to the upwelled waters, high surface productivity and the associated high degradation of the organic matter in the subsurface and bottom waters. However, this depletion of $\delta^{13}\text{C}$ was not observed in benthic bivalve shells obtained

from 10°N, since it is influenced by high export fluxes of carbon from the Cochin estuary since early monsoon months.

Keywords $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ · Benthic bivalves · Upwelling · Summer monsoon · South western continental margin of India

Introduction

The biogenic carbonate shells (calcite and aragonite) of the marine organisms (foraminifera, molluscs, corals, fish otoliths, etc.) are precipitated throughout the life span of the organism and, therefore, record the environmental characteristics of the region they inhabit. The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ isotopes in these biogenic carbonates can be correlated with the characteristic composition of the ambient seawater in which they were grown (Epstein et al. 1953; Grossman and Ku 1986). Hence many of them have been used as a proxy in paleoclimatic and paleoceanographic studies (Wefer and Berger 1991; Lynch-Stieglitz and Fairbanks 1994; Smith et al. 1997; Khim et al. 2001; Sadler et al. 2012). Fine-scale incremental subsampling at intervals of 1 mm or less of the mollusc shells and analysis of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ are used to infer annual or seasonal variations of salinity and temperature in modern as well as ancient marine and estuarine environments (Khim et al. 2001; Dettman et al. 2004; Latal et al. 2006). The stable isotope geochemistry of the larval or juvenile mollusc shells has also been found to reflect the environmental conditions and, hence, they are proposed to be useful proxies in short-term ecological studies as indicators for upwelling, eddies, larval migration, etc. (Owen et al. 2008).

The western continental margin of India is characterized by southwesterly and/or weak northerly winds, equator ward moving surface currents (West India Coastal Current) and a

✉ Josia Jacob
josiajacob@gmail.com

¹ Centre for Earth Sciences, Indian Institute of Science, Bangalore 560 012, India

² Centre for Marine Living Resources and Ecology, Ministry of Earth Sciences, Kochi 682 037, India

³ Department of Marine Biology, Microbiology and Biochemistry, School of Marine Sciences, Cochin University of Science and Technology, Kochi 682 016, India

⁴ Divecha Centre for Climate Change, Indian Institute of Science, Bangalore 560 012, India

pole ward propagating undercurrent (West India Under Current) during the summer monsoon season (Shetye and Gouveia 1998; Lekshmi et al. 2007). The region experiences upwelling of moderate intensity during this season, mainly driven by alongshore winds. Intensity of upwelling varies from south to north as summer monsoon progresses in response to the strength of the alongshore component of the wind (Shetye 1984; Shetye et al. 1985, 1990). However, remote forcing other than the wind driven mechanism is also proposed as the probable mechanism for upwelling observed along the region and it is also argued that upwelling along the coast is non-uniform (McCreary et al. 1993; Smitha et al. 2008). The biological production along the west coast of India exhibits temporal and spatial variation in response to the intensity of upwelling during the summer monsoon season (Habeebrehman et al. 2008; Lathika et al. 2013). One of the major biogeochemical consequences of the coastal upwelling observed along the region during summer monsoon is the bottom and subsurface oxygen deficiency associated with upwelling attaining peak intensity in September–October. The development of this seasonal hypoxia is natural in origin mainly due to the high organic production triggered by upwelling and also by the vertical stratification due to the monsoon rainfall and runoff which reduces the ventilation. However, intense reducing conditions are observed north of 12°N (Banse 1959; Naqvi et al. 2000, 2006).

Several studies have reported that a combination of factors such as water depth, sediment texture, temperature, organic matter content and dissolved oxygen together is responsible for determining the distribution of the benthic population and biomass along the western continental margin of India (Jayaraj et al. 2007, 2008; Joydas and Damodaran 2009; Abdul Jaleel et al. 2014). The main objective of the present study is to examine the effect of the spatial and temporal variability observed in the upwelling intensity and the associated biological production during the onset (OSM) and peak (PSM) phase of summer monsoon on the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of the inorganic biogenic carbonates (benthic) along the South Western Continental Margin of India (SWCMI). $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of the benthic bivalve shells (multispecies of 1–5 mm size) obtained from ten surface sediment samples belonging to three transects along the SWCMI collected during June 2009 (OSM) were compared with those collected during August 2009 (PSM). Analysis based on samples collected during OSM and PSM of the same year is relevant as the process of coastal upwelling shows considerable interannual variations (Kurian et al. 2011).

Materials and methods

Surface sediments were collected from ten locations along the SWCMI during the OSM and PSM of 2009. Sampling was done onboard *FORV Sagar Sampada* during the 267

and 270 cruises conducted from 29th May–14th June and 4–21st August, 2009, respectively. The samples were taken along three transects of the SWCMI at 9°N, 10°N and 12°N (Fig. 1). The sediment samples were collected using Smith McIntyre grab (0.2 m²) and were immediately transferred and stored at −20 °C till analysis. The physico-chemical characteristics of the water column above the sediment for each sampling location were obtained from the vertical profiles of temperature and salinity processed at one meter interval measured using the Conductivity-Temperature-Depth (CTD Seabird 911 plus) Profiler. However, the temperature and salinity data were not available for the 100 m station off 9°N. Bulk SST (<28 °C) or temperature at 5 m water depth and the mixed layer depth are used as indicators of upwelling.

The thawed sediment samples were then washed in a sieve of size 63 μm using distilled water. The benthic organisms (of all size ranges) retained on the sieve were oven dried at 40 °C (Ishimura et al. 2012). From this mixture of benthic organisms, juvenile bivalve shells of the size range 1–5 mm from each station (~5–7 numbers) were separated, identified and powdered for analysis of the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$. Multispecies analyses of the separated bivalve shells were done as the bivalve shells were very few in many of the stations. Also, the distribution and abundance of benthic fauna in the region are found to be varied with depth, latitude and sediment nature (Jayaraj

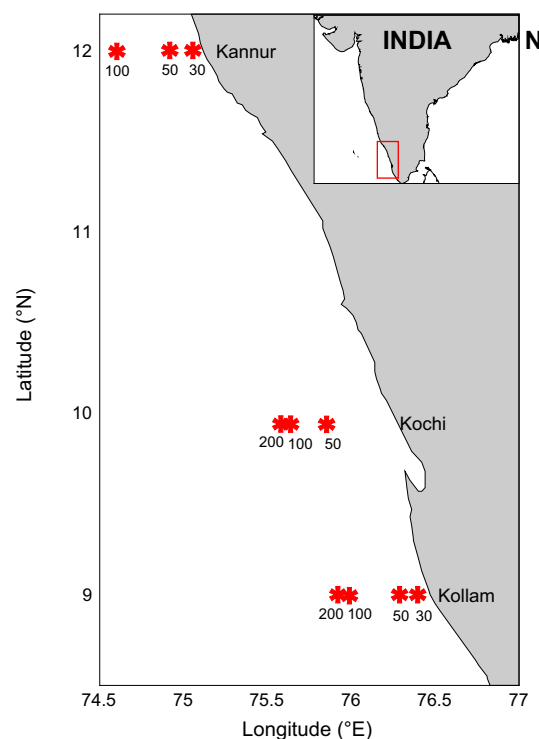


Fig. 1 Map showing the station positions. Depth of the stations is also given

et al. 2008). The major bivalve species identified were *Venus chemnitzii*, *Donax faba*, *Paphia malabarica*, *Dosinia* sp. *Donax scortum*, *Cardium flavum*, *Venus reticulata*, *Pecten tranquebaricus*, etc. The growth rate reported for one of the species studied, i.e. *P. malabarica* varies from $58.6 \pm 0.09 \mu\text{m day}^{-1}$ in fertilized egg to $206.4 \pm 0.3 \mu\text{m day}^{-1}$ in post-metamorphic larvae (Gireesh and Gopinathan 2008). Hence, the size fraction considered is suitable for the intraseasonal comparison between OSM and PSM as done in the present study.

Analysis of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ in the bivalve shells was carried out by the acid digestion method using the Gas-Bench II peripheral of the MAT 253 Isotope Ratio Mass Spectrometer (IRMS). The powdered and homogenized shell samples were transferred to exetainer vials which after flushing with Helium gas were reacted with 105 % phosphoric acid at a constant bath temperature of 70 °C maintained in the GasBench II peripheral. Isotopic analysis of the CO_2 evolved was done in the IRMS. The isotopic composition of carbon and oxygen in the bivalve shells was reported as delta notations ($\delta^{13}\text{C}_{\text{sample}}$ and $\delta^{18}\text{O}_{\text{sample}}$) with respect to the V-PDB scale as permil (‰):

$$\delta \text{ in } \text{‰} = \left[\left(\frac{R_{\text{sample}}}{R_{\text{standard}}} \right) - 1 \right] \times 1000$$

where $R = {}^{13}\text{C}/{}^{12}\text{C}$ or ${}^{18}\text{O}/{}^{16}\text{O}$.

An inter-laboratory standard (MARJ1) and an international lab standard (NBS-18) were used for the quality control of analytical procedures. The reproducibility of $\delta^{13}\text{C}$ based on replicate analysis of the standards was within $\pm 0.05 \text{‰}$. For $\delta^{18}\text{O}$, the reproducibility was within ± 0.02 for MARJ1 and $\pm 0.1 \text{‰}$ for NBS-18. Repeated analysis of the multispecies benthic bivalve shells gave a reproducibility of $\pm 0.13 \text{‰}$ for $\delta^{13}\text{C}$ and $\pm 0.23 \text{‰}$ for $\delta^{18}\text{O}$.

The analytical values obtained for $\delta^{18}\text{O}$ in the benthic bivalve shells were compared with $\delta^{18}\text{O}$ values calculated for inorganic carbonate shells precipitated under equilibrium conditions using known relationship between carbonate $\delta^{18}\text{O}$, temperature and $\delta^{18}\text{O}$ of seawater. Powdered benthic bivalve shells from two of the stations were analysed using XRD (Bruker-D8 Advance) technique for mineralogical identification. Compositionally, the major fraction was aragonite with the presence of minor calcite minerals. Because several species of bivalve shells of variable mineralogical composition were used, estimates for both calcite and aragonite were calculated. The bottom water temperature and salinity recorded at each station were used in the calculation. Salinity and $\delta^{18}\text{O}$ of the seawater covary linearly (Craig and Gordon 1965), hence the linear regression parameters for the relationship in the open and coastal regions of the South Eastern Arabian Sea (SEAS) are established (Singh et al. 2010; Deshpande et al. 2013; Jacob et al. 2014). Bottom water salinity was converted to $\delta^{18}\text{O}$ values using the linear regression

equation for the $\delta^{18}\text{O}$ versus salinity relationship for the subsurface waters (50–500 m) of the SEAS (Slope 0.55 ± 0.16 ; Intercept -18.5 ± 5.6) proposed by Singh et al. 2010. The predicted calcite shell $\delta^{18}\text{O}$ was obtained from Epstein et al. (1953) and Epstein and Lowenstam (1953) (Eq. 1), while the predicted aragonite shell $\delta^{18}\text{O}$ was obtained from Grossman and Ku (1986) (Eq. 2), respectively.

$$T(^{\circ}\text{C}) = 16.5 - 4.3 (\delta^{18}\text{O}_{\text{calcite}} - \delta^{18}\text{O}_{\text{water}} + 0.27) + 0.14 (\delta^{18}\text{O}_{\text{calcite}} - \delta^{18}\text{O}_{\text{water}} + 0.27)^2, \quad (1)$$

where T is the seawater temperature in °C, $\delta^{18}\text{O}_{\text{calcite}}$ is expressed relative to VPDB and $\delta^{18}\text{O}_{\text{water}}$ is expressed relative to VSMOW.

$$T(^{\circ}\text{C}) = 20.6 - 4.34 (\delta^{18}\text{O}_{\text{aragonite}} - \delta^{18}\text{O}_{\text{water}}), \quad (2)$$

where T is the seawater temperature in °C, $\delta^{18}\text{O}_{\text{aragonite}}$ is expressed relative to VPDB and $\delta^{18}\text{O}_{\text{water}}$ is expressed relative to mean ocean water which is equivalent to VSMOW minus 0.2 ‰ (Grossman 2012).

Results

The temperature and salinity profiles of the water column overlying the sediment at the sampling locations during OSM and PSM are shown in Fig. 2 and the hydrographical characteristics are given in Table 1. PSM observations off 9°N recorded cold upwelled waters recording bulk Sea Surface Temperature (SST) between 23.8 and 26.3 °C and a shallow mixed layer (ML) (6–12 m) over the entire shelf while at 10°N upwelling was moderate (bulk SST 25.1–26.6 °C; ML 7–8 m). Off 12°N cold waters were confined to the coast up to 30 m depth region (bulk SST 23.2 °C), whereas the offshore waters were warmer between 28.1 and 28.6 °C. The observations on Sea Surface Salinity (SSS) indicated the influence of local precipitation and runoff in PSM which prevents the upwelled waters reaching the surface layer.

During OSM, upwelling was in the initial stage with relatively cool surface waters (27.3–27.8 °C) and thin ML (9–12 m) along the near coastal sites (30–50 m) off 9°N compared to the northern region. Offshore waters along 9°N and north of 10°N, the surface waters were warmer (bulk SST >28.8°C). Influence of freshwater runoff was observed up to 50 m depth zone of 10°N in OSM.

The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ in the multispecies bivalve shells obtained from the sediments of the SWCMI during the OSM and PSM are given in Fig. 3 and Table 1. The $\delta^{13}\text{C}$ in the benthic bivalves ranged from -0.25 to 1.35‰ (av

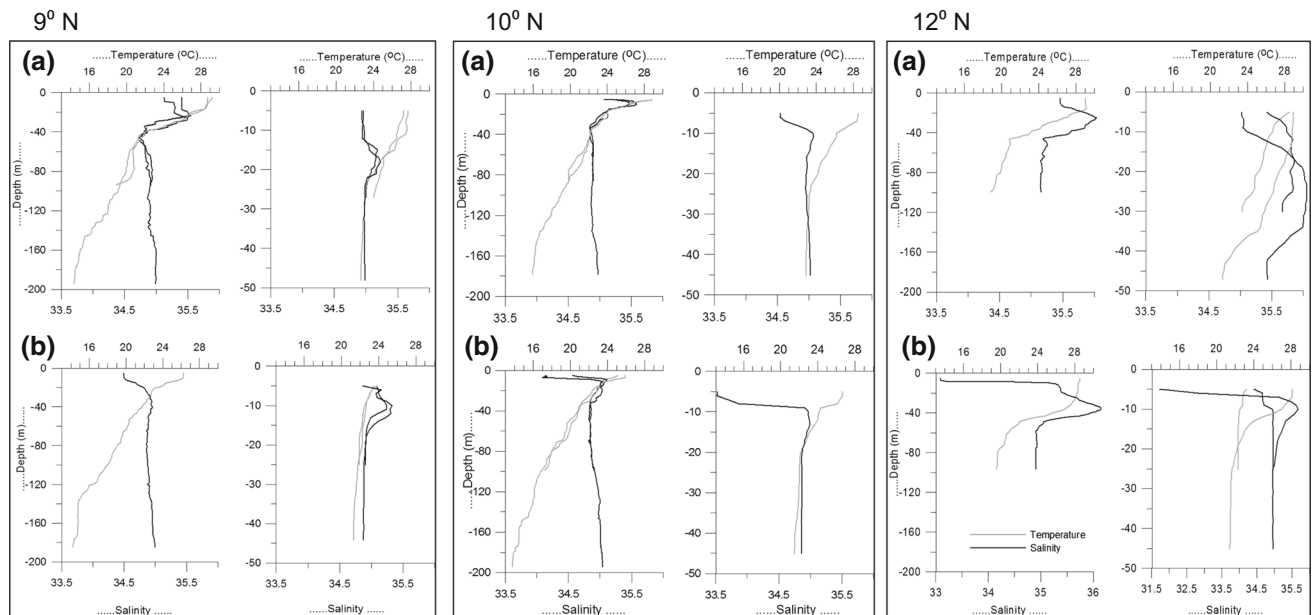


Fig. 2 Temperature and salinity profiles of the water column overlying the sediment sampling locations during **a** OSM and **b** PSM

Table 1 Characteristics of the water column overlying the sediment sampling locations, $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of the multispecies benthic bivalve shells

Transect	Depth (m)	SSS (5 m)	SST(5 m) (°C)	ML (m)	$\delta^{13}\text{C}$ (‰)	$\delta^{18}\text{O}$ (‰)
12°N						
OSM	30	35.4	28.6	6	1.22	−0.43
	50	35.0	29.0	11	1.35	−0.66
	100	35.5	29.1	17	0.96	0.44
PSM	30	34.4	23.2	^a	−0.12	−0.75
	50	31.7	28.1	9	0.87	−0.71
	100	33.1	28.6	31	0.74	0.57
10°N						
OSM	50	34.6	28.5	9	−0.25	−1.00
	100	35.3	28.9	7	1.12	0.04
	200	35.1	28.9	9	0.15	0.58
PSM	50	33.5	26.6	7	0.82	−1.21
	100	34.6	25.1	7	1.14	0.12
	200	34.1	25.9	8	0.42	0.59
9°N						
OSM	30	35.0	27.3	9	0.88	−0.35
	50	34.9	27.8	12	0.66	−0.27
	100	35.1	29.2	10	0.82	−0.50
	200	35.4	28.7	17	0.65	1.64
PSM	30	34.9	23.8	6	−0.46	−0.83
	50	35.1	23.4	8	−0.37	−1.42
	100	—	—	—	−0.15	0.62
	200	34.5	26.3	12	0.40	1.70

^a Well mixed up to the bottom

$0.76 \pm \text{sd } 0.49$) and from $−0.46$ to 1.14 ‰ (0.33 ± 0.57) during the OSM and PSM, respectively (Fig. 3a–c). In the benthic bivalves, the $\delta^{18}\text{O}$ varied from $−1.00$ to 1.64 ‰

($−0.05 \pm 0.77$) during OSM and $−1.42$ to 1.70 ‰ ($−0.13 \pm 1.00$) during PSM (Fig. 3d–f). The $\delta^{13}\text{C}$ values in the benthic bivalve shells collected at 12°N and 9°N

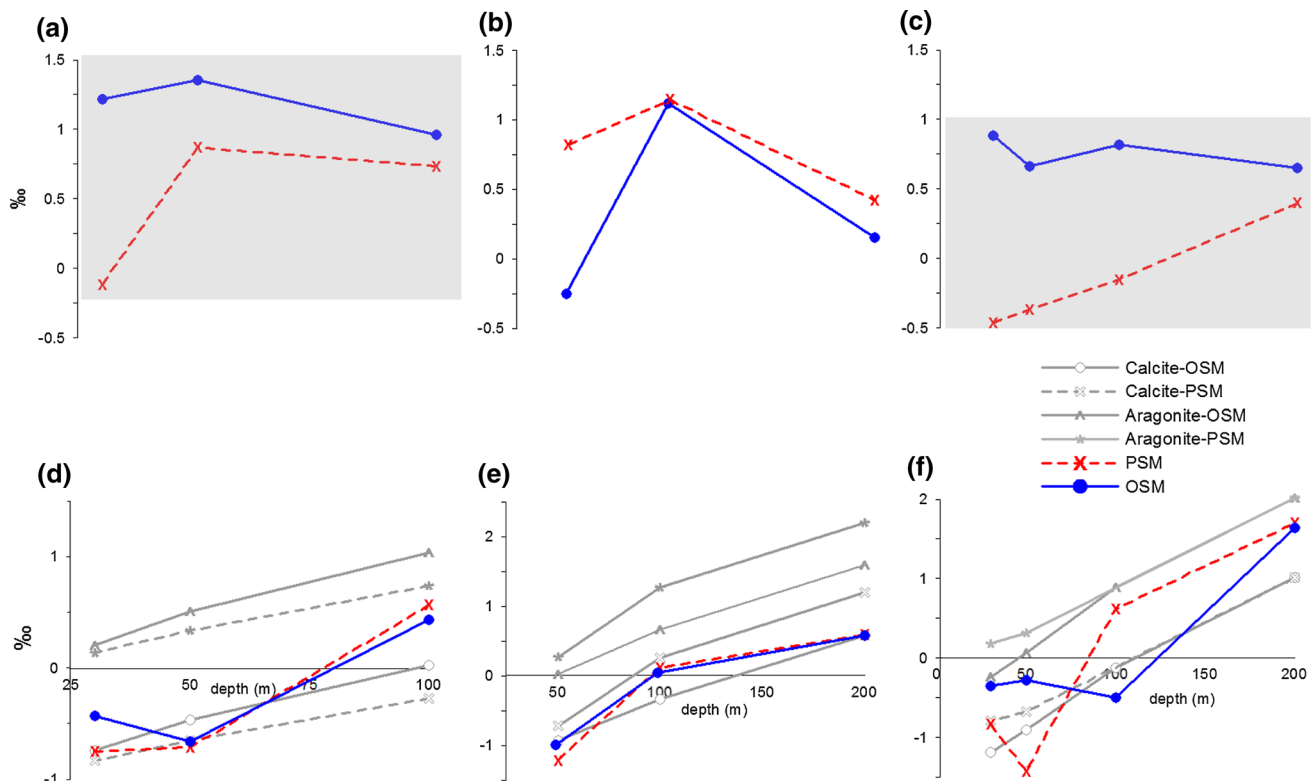


Fig. 3 Observed $\delta^{13}\text{C}$ in the benthic bivalve shells from **a** 12°N, **b** 10°N, and **c** 9°N. The observed $\delta^{18}\text{O}$ in the benthic bivalve shells along with the predicted calcite and aragonite $\delta^{18}\text{O}$ at **d** 12°N, **e** 10°N, and **f** 9°N

were lower in PSM than during OSM (Fig. 3a, c). However, the depletion in ^{13}C was not observed at 10°N (Fig. 3b). The $\delta^{18}\text{O}$ of the benthic bivalves from most of the stations did not vary much between the sampling periods (Fig. 3d–f). There was no correlation between $\delta^{13}\text{C}$ and the $\delta^{18}\text{O}$ in the benthic bivalve shells for both the sampling periods ($r = 0.02$ in OSM and $r = 0.23$ in PSM).

Discussions

The $\delta^{18}\text{O}$ values of the biogenic carbonate shells of marine organisms precipitated under equilibrium conditions with the surrounding seawater reflect the calcifying temperature and salinity, while the latter is directly related to $\delta^{18}\text{O}$ of the seawater (Epstein et al. 1953; Craig and Gordon 1965; Grossman and Ku 1986). The $\delta^{13}\text{C}$ of these biogenic carbonate shells mostly reflects the composition of the dissolved inorganic carbon (DIC) (McConnaughey and Gillikin 2008). However, isotopic disequilibrium during the calcification is caused by several factors known as the vital effects which represent kinetic and metabolic processes. Kinetic effect results in fractionation against the heavier isotopes of carbon and oxygen during CO_2 hydration and hydroxylation as well as the disparate incorporation of lighter isotopes due to the higher diffusivity of

HCO_3^- molecules enriched in lighter carbon and oxygen isotopes. Metabolic effect results in changes in the internal DIC pool, which is a combination of both seawater DIC and internally produced metabolic or respiratory carbon (McConnaughey 1989a, b). Kinetic effects generally affect both ^{18}O and ^{13}C and hence exhibit a good correlation between them. The effect is minimal for juvenile shells compared to the adults. In the present study, kinetic effects were negligible as evident from the poor correlation between $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values of the benthic bivalve shells. The $\delta^{18}\text{O}$ values observed in the benthic bivalve shells in the present study were compared with $\delta^{18}\text{O}$ obtained under equilibrium condition (at known temperature) between water and calcite or aragonite. The observed $\delta^{18}\text{O}$ values in general agree with the trend recorded in the predicted values (Fig. 3d–f) and suggest in situ occurrence of bivalve shells. During PSM, the $\delta^{18}\text{O}$ values in the benthic bivalves from 12°N at 30 and 50 m stations were similar to the estimated values for calcite while at 100 m station it closely resembles the estimated values derived from aragonite precipitation. The same observation was extended for stations 50 and 200 during OSM and at 100 m station in PSM along 10°N. At 30 and 200 m stations off 9°N in PSM, the observed $\delta^{18}\text{O}$ values in the bivalve shells were similar to the predicted values for the aragonite. Based on these set of observations, we established that the bivalve shells at these

stations were produced under equilibrium conditions with sea water. However, at some stations offsets were observed in the estimated and observed values of $\delta^{18}\text{O}$. This could be explained after taking into account the uncertainty in $\delta^{18}\text{O}$ values of the bottom waters which were input in our calculation and were derived from the established $\delta^{18}\text{O}$ versus salinity equation for the study region. An additional reason for such a discrepancy can also occur due to minor variability in the mineralogical composition (proportion of calcite and aragonite) of $\sim 5\text{--}7$ multispecies bivalve shell specimens used here for isotopic analysis. $\delta^{18}\text{O}$ values for the bottom waters from these sampling stations measured on a seasonal scale might help to understand the discrepancy.

The $\delta^{13}\text{C}$ values of the seawater DIC ($\delta^{13}\text{C}_{\text{DIC}}$) in the surface layers of the ocean are influenced by the biological productivity and the atmospheric exchange. High biological productivity in the surface layers leads to a preferential uptake of the ^{12}C during photosynthetic process, whereby the seawater gets enriched in ^{13}C . The oxidation of the organic matter in the subsurface and deeper water column releases the fixed ^{12}C making the $\delta^{13}\text{C}_{\text{DIC}}$ lighter or more negative. Thus, the biological pump creates an isotopic gradient with ^{13}C enriched waters at the surface and ^{13}C depleted waters at the bottom. During upwelling, water enriched in ^{12}C , nutrient rich and cold waters from the intermediate/deeper layers reach the surface (Freeman 2001). Hence, the $\delta^{13}\text{C}$ values in mollusc shells are used to study the seasonal and long-term variability of the upwelling and primary production (Killingley and Berger 1979; Sadler et al. 2012). Isotopically light (more negative) $\delta^{13}\text{C}$ values have been observed in several benthic bivalve shells which record changes in $\delta^{13}\text{C}_{\text{DIC}}$ during periods of high biological productivity occurring either by upwelling or by nutrient-rich riverine inputs (Killingley and Berger 1979; Arthur et al., 1983; Krantz et al. 1987, 1988; Jones and Allmon 1995; Sadler et al. 2012). However, the surface water of the coastal and continental margins also receives waters from the runoff and the ground water discharge enriched in ^{12}C (Freeman 2001).

Upwelling waters observed along the southernmost transects during OSM were stronger and progressed towards the northern transects up to 12°N in PSM. Hence, $\delta^{13}\text{C}_{\text{DIC}}$ values noted for the subsurface and bottom waters could be expected to be lower in PSM than in OSM due to upwelling and high biological production and the associated sinking and degradation of the organic matter in the subsurface and bottom waters. $\delta^{13}\text{C}$ values in the benthic bivalve shells were observed to be lower during PSM compared to OSM along the stations from two of transects out of the three studied (9 and 12°N). The lower $\delta^{13}\text{C}$ values of the bivalve shells studied during PSM would be corresponding to the expected lower $\delta^{13}\text{C}_{\text{DIC}}$ values of the

bottom waters. The high productivity can also cause high metabolic effects during the season. But the shell carbon in marine molluscs derives mainly from ambient DIC (McConnaughey and Gillikin 2008). Jacob and Ghosh (2010) have earlier reported a similar observation of the lower values of $\delta^{13}\text{C}$ of the benthic bivalve shells during late summer monsoon compared to the pre-monsoon season along the western continental shelf of India. However, it was interpreted as due to the greater organic matter accumulation in the surface sediments of the region during late summer monsoon season (Jacob et al. 2009) compared to the pre-monsoon season.

The observation from the 10°N was an exception since the region is reported to be influenced by the discharges from the adjacent estuarine ecosystem, i.e. the Cochin estuary during the summer monsoon months. Gupta et al. (2009) observed significant amount of inorganic and organic carbon export from the Cochin estuary to the adjacent sea from the early monsoon months. They proposed that these export fluxes together with upwelling cause the prevailing hypoxic conditions along the coastal regions of 10°N during the summer monsoon. Jyothibabu et al. (2006) from planktonic studies done in the Cochin backwater system have also proposed that a major portion of the primary carbon either settles down or gets transported to the coastal regions during the monsoons.

Summary

In the present study, the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ of benthic bivalve shells collected from surface sediments of the SWCMI during OSM and PSM of 2009 were analysed and related to the seasonal oceanic process. Comparison of the $\delta^{18}\text{O}$ values in benthic bivalve shells in PSM and OSM with the predicted equilibrium calcite and aragonite $\delta^{18}\text{O}$ proves the in situ origin of the shells. The $\delta^{13}\text{C}$ values in the benthic bivalve shells obtained from the surface sediments off 12°N and 9°N were lower in PSM than during OSM. The $\delta^{13}\text{C}$ in the bivalves studied recorded the expected depletion in ^{13}C of the $\delta^{13}\text{C}_{\text{DIC}}$ values of the bottom waters during PSM due to the seasonal upwelling, the high surface productivity and the associated greater degradation of the organic matter in the surface and bottom waters. However, this depletion of $\delta^{13}\text{C}$ was not observed in the benthic bivalve shells obtained from the 10°N transect, probably because of the influence of the drainage of inorganic and organic carbon-rich waters into the coastal region from the adjacent estuarine ecosystem, i.e. the Cochin estuary during the monsoon months. More observations on stable isotopic composition in benthic bivalve shells of uniform size and species together with determination of $\delta^{13}\text{C}_{\text{DIC}}$ and $\delta^{18}\text{O}$ in the bottom waters during the different periods of

summer monsoon will expand the scope of understanding the seasonal/intraseasonal variability and usefulness of stable isotope in bivalve shell in past environmental reconstruction studies. However, the present observations report the effect of the variable upwelling intensity and the associated biological response observed along the SWCM I during the onset and peak phase of summer monsoon on the biogenic carbonates. The study indicates that the $\delta^{13}\text{C}$ of juvenile benthic bivalve shells along with other suitable geochemical proxies can be used to infer the temporal or seasonal scale characteristics of the coastal and/or other dynamic paleo-environments.

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