

Contrasting patterns in pH variability in the Arabian Sea and Bay of Bengal

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Abstract:

Continuous understanding of the ongoing ocean acidification (OA) is essential for predicting the future impact of OA on marine ecosystems. Here we report the results of open ocean time-series measurements (19 cruises) of seawater pH in total hydrogen ion scale (pH_T) and associated parameters in the Arabian Sea (AS) and the Bay of Bengal (BoB). During Southwest monsoon (SWM) the pH_T within the 30 to 100 m water column shows the maximum difference between the two basins with BoB pH_T being lower (up to ~ 0.39 units) than AS which could be due to freshwater influx from rivers, mixed layer dynamics, and cold-core eddies. However, during Spring inter-monsoon (SIM), the pH_T of BoB follows the trend of AS. A contrasting finding is that the lowest pH_T occurs at 350 to 500 m in the BoB while it is ~ 1000 m in the AS. The pH_T within the 150 to 1500 m layer of these two basins shows lower values by 0.037 (± 0.03) in the BoB as compared to the AS. The possible reasons for the low pH_T within the BoB oxygen minimum zone (OMZ) could be due to intrusion of western Pacific water in the BoB, freshwater influx from rivers, variations in OMZ of the two basins, higher temperature ($\sim 2^\circ\text{C}$) within the OMZ of the AS, denitrification in the AS and also due to the differences in lithogenic material in the two basins. The pH_T in both the basins (500 to 1000 m) is lower than in the North Atlantic and higher than in the North Pacific waters, however, the pH_T in the 200 to 500 m is lower in the BoB than in all these basins. This study highlights the under-saturation of calcium carbonate at very shallow depths (~ 100 m) in the BoB, indicating that the plankton in the BoB are facing a major risk from OA compared to the AS and need further investigation.

Keywords: ocean acidification; pH; Arabian Sea; Bay of Bengal; oxygen minimum zone

1. Introduction

The ocean acts as a sink for ~30% of human-induced CO₂ emissions (Sabine et al., 2004; Khatiwala et al., 2013; Le Quéré et al., 2018; Gruber et al., 2019). This carbon uptake by the ocean results in a decrease in the oceanic pH, a process termed ocean acidification (OA) (Feely et al., 2004), and with time the anthropogenic CO₂ gets transported to deeper layers, lowering the pH of the deeper waters as well (Caldeira and Wickett, 2005). Since the Industrial Revolution, the global surface ocean pH has declined by approximately 0.1 (from 8.2 to 8.1) (Caldeira and Wickett, 2003; Orr et al., 2005; Garcia et al., 2021). Jiang et al. (2019) report a similar global decrease of ~0.12 units from 8.19 to 8.07 from 1750 to 2010. OA is a major threat to marine ecosystems (Fabry et al., 2008; Gattuso et al., 2015) and can affect the shells of calcified marine organisms (Steinacher et al., 2009) and even affect fishery resources (Tai et al., 2021). OA can also affect the carbon and silicon cycle by influencing community composition and vertical fluxes of diatom (Petrou et al., 2019; Taucher et al., 2022). However, little is known about how OA might affect the complex interaction of processes that govern marine biogeochemical cycles (Blackford and Gilbert, 2007). Moreover, little is known about the net effect of OA on the whole ecosystem (Riebesell, 2004), for which continuous long-term time-series pH data is required that will help in better understanding the trends and time scales of variability in OA.

There are 7 major open ocean time-series stations for monitoring carbon and acidification. These sites include 2 in the subpolar North Atlantic and 5 sites in the tropical and temperate Atlantic and Pacific Ocean (BATS, ESTOC, HOT, CARIACO, and Muninda). Bates et al. (2014) have summarized the results highlighting that all these stations show a drop in pH with values ranging from -0.0013 to -0.0026 pH units yr^{-1} . In the Atlantic Ocean, a drop in pH is reported in several studies at the regional scale, eg. Leseurre et al. (2020) have reported a drop in pH of -0.0017 yr^{-1} during 1993–2017 in the North Atlantic subpolar gyre. García-Ibáñez et al. (2016) reported a drop of -0.0016 pH units yr^{-1} in the Irminger and Iceland basins, while Omar et al. (2019) reported -0.0024 pH units yr^{-1} in the North Sea. For the period 1995–2010, the average surface pH in the Northeast Atlantic Ocean declined by 0.051 pH units (Davila and Casiano 2023). The acidification rate calculated for the North Atlantic Central Water from 2005 to 2015 is 0.0036 pH units yr^{-1} (Flecha et al., 2019). Fransner et al. (2022) have also reported a decreasing annual pH trend in the Nordic Seas from 1981 to 2019 ranging from -0.002 to -0.003 pH units yr^{-1} . In the Pacific Ocean also, a drop in pH has been reported (Dore et al., 2009). Midorikawa et al. (2010) reported a long-term decreasing trend of -0.0019 pH units yr^{-1} in the Northwest Pacific. They also highlighted the seasonal differences whereby the pH decreased by -0.0013 pH units yr^{-1} in summer and -0.0018 pH units yr^{-1} in winter from 1983 to 2007. Wakita et al. (2013) investigated pH changes in the subArctic Northwest Pacific

ic from 1997 to 2011 and found that pH in the winter mixed layer decreased at a rate of -0.0011 pH units yr^{-1} . Their study also revealed a decline in pH at a rate of -0.0051 between 200 and 250 m. Ishizu et al. (2021) reported patterns of acidification (south of 25°N) and alkalization (north of 25°N) in the North Pacific with rates of -0.008 to 0.004 pH units yr^{-1} . Their study also reported that strong acidification or alkalization occurs below 200 m depth. Although, earlier studies have suggested that OA is more pronounced in the <500 m deep layer (Krimsky et al., 2020), the pH decrease can also be detected down to 2000 m depth (Fransner et al., 2022).

The Indian Ocean significantly influences the global carbon cycle as it accounts for $\sim 20\%$ of the global CO_2 uptake (Takahashi et al., 1997). However, it is one of the under-sampled regions concerning surface ocean pCO_2 compared to other oceans and pH remains elusive because of insufficient observations (Chakraborty et al., 2021). The seasonal cycle of pCO_2 and carbon fluxes was studied in the AS (George et al., 1994; Goyet et al., 1998; Sarma et al., 1998; 2003; 2013; Chakraborty et al., 2021; De Verneil et al., 2022;), and in the BoB (Sardessai et al., 2010; Sarma et al., 2012; 2015; 2021; Chakraborty et al., 2021). The factors controlling CO_2 variabilities are physical transport, such as upwelling and convective mixing in the AS; whereas freshwater input by rivers and deposition of atmospheric pollutants in the BoB (Sarma et al., 2000; 2012). The BoB acts as a net sink, whereas AS acts as a net emitter of CO_2 to the atmosphere (Valsala and Maksyutov, 2010; Sarma et al., 2012; Valsala and Maksyutov, 2013). The AS has pCO_2 reaching up to $700 \mu\text{atm}$ (Sarma 2003), with emissions of $\sim 160 \text{ Tg C yr}^{-1}$ to the atmosphere with high emissions during summer (Verneil et al., 2022). The BoB is a perennial sink for atmospheric CO_2 (Takahashi et al., 2009). A few other studies also reported BoB to be a sink for atmospheric CO_2 (Kumar et al., 1996; Sarma, 2003; Sharada et al., 2008; Takahashi et al., 2009; Sarma et al., 2012; Valsala and Maksyutov, 2013), while Ye et al. (2018) reported it as a source of CO_2 during April to August. Sarma et al. (2012) highlighted the role of river discharge into BoB in determining whether BoB will be a source or sink of CO_2 during southwest monsoon (SWM). Recently Roy et al. (2021) reported the southern BoB as a source of CO_2 . Based on data from 2010 to 2019, Shanthi et al. (2022) recently reported the BoB as a weak sink during post-monsoon and pre-monsoon seasons, while a strong source of CO_2 to the atmosphere during monsoon season. Chakraborty et al. (2018) highlighted that the region east of Srilanka experiences dominant phytoplankton blooms and it is of prime importance to the carbon cycle of the BoB. Recently, Sarma et al. (2023) reported that the Indian Ocean north of 18°S is found to be the mean annual source ($0.04 \pm 0.05 \text{ Pg C yr}^{-1}$) whereas a net sink ($-0.23 \pm 0.11 \text{ Pg C yr}^{-1}$) in the south of 18°S .

The AS hosts one of the world's largest and most intense OMZs (Naqvi et al., 2000; 2006), and recent reports indicate the expansion of AS OMZ (Lachkar et al., 2018, 2019). Large-scale denitrification

(25.3 Tg N Yr⁻¹) takes place in the AS (Anju et al., 2021), whereas the denitrification rates are inhibited by 50% in the BoB (Naqvi, 2006; Johnson et al., 2019). Both the AS and BoB are undergoing rapid changes in biogeochemistry with the AS witnessing a temperature rise (Roxy et al., 2014; Dinesh Kumar et al., 2016; Sun et al., 2019) and CO₂ (Kanuri et al., 2017) and a drop in pH (Piontkovski and Queste, 2016; Shetye et al., 2020). However, very recently Sarma et al. (2023) reported that the emission of CO₂ to the atmosphere from the AS is decreasing due to the warming of the basin that led to stratification and a decrease in upwelling intensity in the western basin. Also, Sridevi and Sarma (2021) indicated a freshening of the surface waters of the BoB over the past two decades due to an increase in the warming and melting of Himalayan glaciers (Goes et al., 2020). As the pH of the Himalayan rivers such as the Ganges and Brahmaputra are relatively alkaline this has led to an increase in pH for the past two decades in the central and eastern BoB (Sridevi and Sarma, 2021).

Despite all these above-mentioned studies, an understanding of pH changes in the sub-surface and deeper layers of the AS and BoB is still missing. As compared to the surface CO₂ only a few studies have reported open oceanic pH in the northern Indian Ocean (NIO). The studies available on pH from BoB and AS are either from the coastal region (Sarma et al., 2015; Shetye et al., 2020), or from the open ocean surface waters (Sridevi and Sarma 2021), while Sreeush et al. (2019) and Chakraborty et al. (2021) used models to show seasonal variations in pH in the surface waters of the NIO. This clearly shows the lack of pH data in the surface and deep waters of the NIO. In this context, a valuable approach was to have time-series stations in the NIO. In 2014, a long-term study on precise pH_T measurements was initiated by the biogeochemistry group at CSIR-National Institute of Oceanography (CSIR-NIO) through which we documented the first-ever detailed spatial, seasonal, and temporal changes in pH_T and its controlling factors in the NIO.

2. Methodology

2.1 Sampling

Here we used the data from 19 open ocean cruises (13 cruises in AS and 6 cruises in BoB) conducted from 2014 to 2023. These cruises were conducted on CSIR-NIO *Research Vessels Sindhu Sadhana* (SSD) and *Sindhu Sankalp* (SSK) covering different seasons (Northeast monsoon (NEM), Spring inter-monsoon (SIM), Southwest monsoon (SWM), and Fall inter-monsoon (FIM)) as detailed in Supplementary Table 1. Water column sampling was carried out along a meridional transect at 68°E extending from 8°N to 21°N cutting across the AS OMZ under the CSIR funded INDIAS-IDEA project. In addition, more frequent measurements were carried out at the Arabian Sea Time Series station (ASTS; 17°N 68°E) and the Bay of Bengal Time Series station (BoBTS; 18°N; 89°E)

established under the SIBER-INDIA program (funded by the MoES). The data collected at ASTS was compared with previous US-JGOFS data collected in 1995 at the nearest station (N7; 19.16°N; 67.15°E) in the AS. The US- JGOFS pH data was converted from the seawater scale to the Total scale using the alkalinity, dissolved inorganic carbon, temperature, salinity, and nutrients using the CO₂SYS program (Lewis and Wallace, 1998). Also, the pH_T data from the NIO is compared with data from the HOT (Hawaii Ocean Time-series) program in the North Pacific and the CARIACO (Carbon Retention in a Colored Ocean) program in the North Atlantic.

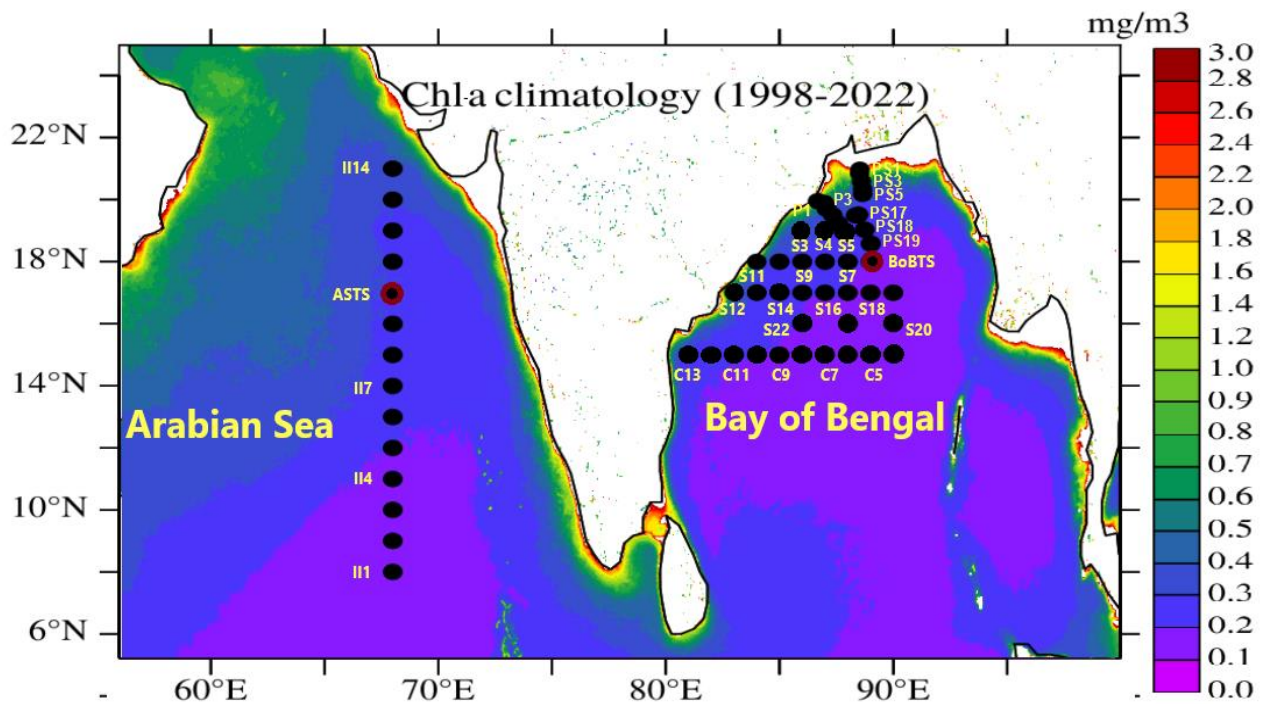


Fig 1 (a) Study area map showing the station locations in the AS and BoB. The background shows the annual mean Chlorophyll - *a* in the northern Indian Ocean from 1998 to 2022.

2.2 Biogeochemical parameters

A CTD (SBE 19 plus, Sea-Bird Electronics, USA) was used to get high-resolution temperature and salinity data. The accuracy of temperature and conductivity for the CTD system was $\pm 0.001^\circ\text{C}$ and $\pm 0.0001 \text{ S m}^{-1}$, respectively. The Wetlabs fluorescence sensor connected to the CTD measured the fluorescence. The Chlorophyll - *a* concentration is estimated from measured fluorescence data using scale factor (SF) and the dark count (DC) data provided along with the sensor using the following equation:

$$\text{Chlorophyll - } a = \text{SF} (\text{counts} - \text{DC})$$

The establishment of spectrophotometric pH measurement (Byrne and Breland 1989; Clayton and Byrne 1993; Dickson 1993) has made it possible to detect even minute scale changes in oceanic pH.

The samples are collected directly from Niskin sampler into a 10 cm pathlength cuvette. After bringing the samples to 25°C in a water bath (Polyscience), measurements of pH_T were made onboard spectrophotometrically using purified *m*-cresol purple as an indicator (Patsavas, 2013) and pH_T was calculated following equations of Liu et al. (2011). Analytical precision was ~0.002 for pH_T . Alkalinity was analyzed by potentiometric titration using VINDTA 3S and the precision was 2.5 $\mu\text{mol/Kg}$. The saturation state of aragonite (Ω_{Ar}) was calculated from alkalinity and pH_T using the CO₂SYS program by Lewis and Wallace (1998). The dissolved oxygen (DO) samples were fixed and analyzed following the Carpenter modification of the Winkler method (Carpenter, 1965). This methodology gave a precision of 0.1 μM with an accuracy of better than 1%. Nitrite was measured spectrophotometrically following Grasshoff et al. (2009). Total Organic Carbon (TOC) was measured using a Shimadzu TOC-LCPH analyzer following the combustion catalytic oxidation method (Dickson et al., 2007) as detailed in Shetye et al. (2021). Total bacterial counts (TBC) were done using an Olympus BH2 epifluorescence microscope (Olympus Corp., Tokyo, Japan) following Parsons et al. (1984).

2.3 Water masses, Mixed Layer Depth, stratification and eddy identification

The water masses were identified using a T (temperature)-S (salinity) diagram. The monthly mean ocean parameters such as temperature and salinity, with a spatial resolution of 0.25 x 0.25 degree were taken from the European Centre for Medium-Range Weather Forecast (ECMWF) Ocean Reanalysis System 5. We defined Mixed Layer Depth (MLD) as the depth at which density exceeds 0.2 kg m^{-3} from its surface value. The ocean stratification (Bruñt Vaisala frequency) in the upper 100 m water column was calculated using the temperature and salinity data.

Gridded daily sea level anomaly data with spatial resolution 0.25 x 0.25 degree resolution for 13 September 2019 was obtained from <https://cds.climate.copernicus.eu/cdsapp#!/dataset/satellite-sea-level-global?tab=form>. Closed contours of positive sea level anomaly indicate the presence of warm core eddies and negative sea level anomalies indicate cold core eddies.

2.4 Sediment trap sampling

As a part of the SIBER-INDIA project, a mooring consisting of sediment traps was deployed at the Northeastern AS trap station (NEAST) in the AS (near ASTS) at 600 m and 1200 m and BoB trap station (near BoBTS) at 430 m and 1020 m depths. The biogenic fluxes were calculated based on carbonate, opal, and organic matter (POC x 1.8) contents and the lithogenic flux was computed as the difference between the total flux and the sum of biogenic fluxes (Kurian et al., 2023). The lithogenic fluxes calculated for the years 2017-2018 in the AS and 2018-2019 in the BoB are used in the present study.

3. Results

3.1 Physico-chemical differences in the AS and BoB

The temperature and salinity can influence the pH_T in the water column and therefore we calculated the average temperature and salinity in the different layers in the AS and BoB. The average sea surface temperature (SST) in the AS (at ASTS) was 28.17°C while in the BoB (at BoBTS) it was 29.13°C indicating high temperature in BoB. Figures 2 and 3 clearly show warmer SST in the BoB. Figures 2a and 3a also show cooler waters at intermediate depths coming from the south in the AS. The water column up to 150 m has an average temperature of 24.6°C in both the basins. However, below this depth, the temperatures are high in the AS, the average temperature for the water column from 150 to 1000 m is 12°C and 9.75°C in AS and BoB, respectively indicating a difference of >2°C in the two basins. Similarly, for the water column between 1000 m and 2000 m, the average temperatures are 5.4°C and 4.4°C in AS and BoB, respectively. Overall the 2000 m water column in the AS is warmer with average temperatures of 9.7°C and 8.1°C in the AS and BoB, respectively. The water column up to 150 m has an average salinity of 36.3 and 33.9 in AS and BoB, respectively. The Arabian Sea High Saline watermass comes from the north in the AS (Fig. 2c and 3c), while the low saline riverine water is seen in the BoB (Fig. 2d and 3d). The average salinity for the water- column from 150 m to 1000 m is high in AS with 35.6 and 35 in AS and BoB, respectively. Similarly, for the water column between 1000 m and 2000 m, the average salinities are 35.04 and 34.85 in AS and BoB, respectively. Overall the 2000 m water column in the AS is more saline with an average salinity of 35.4 and 34.84 in AS and BoB, respectively.

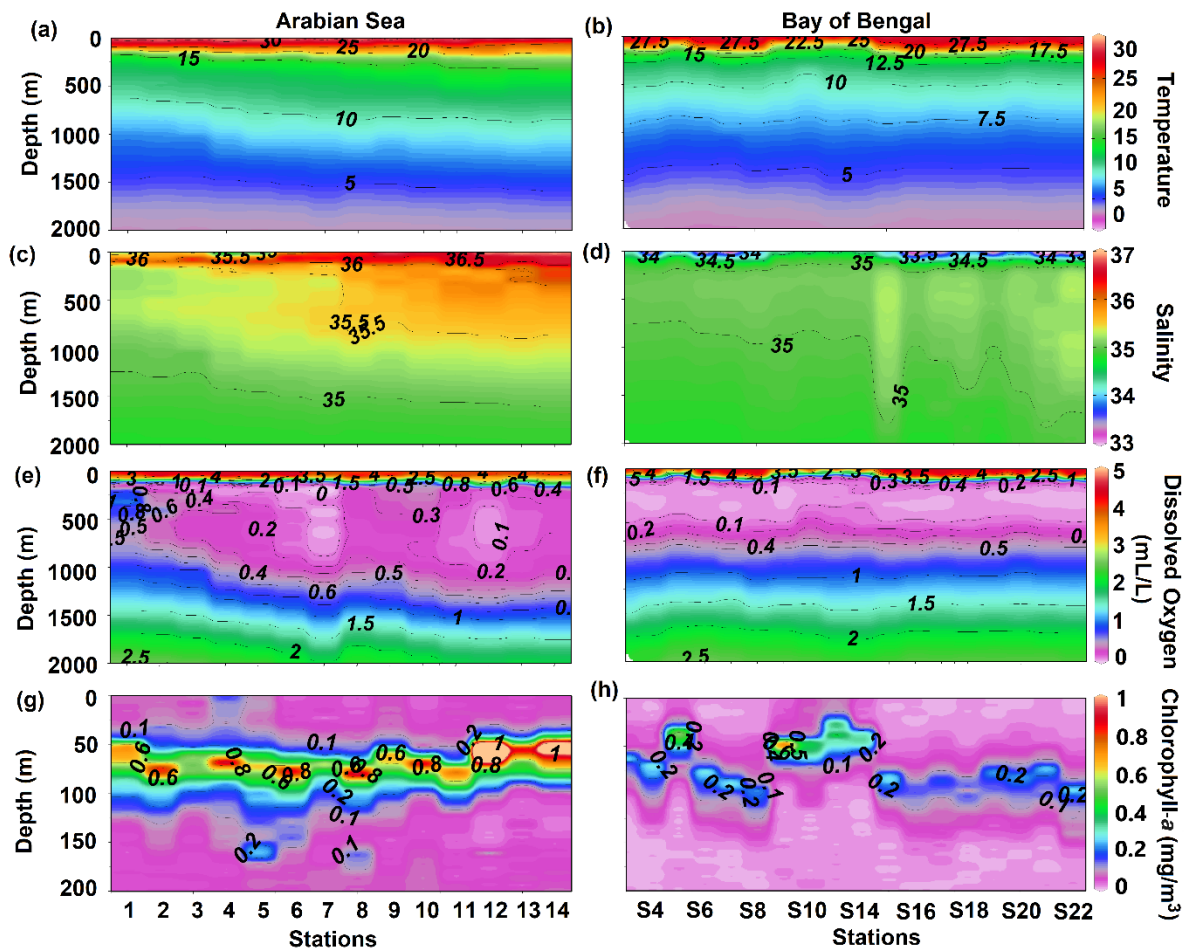


Fig 2 Variations of temperature ($^{\circ}\text{C}$), salinity, dissolved oxygen, and Chlorophyll - *a* (200 m) in the AS (left panel) and BoB (right panel) during SIM

The water column up to 150 m has an average dissolved oxygen of 2.3 mL/L and 2.1 mL/L in AS and BoB, respectively. The average dissolved oxygen for the water column from 150 m to 1000 m is low in AS with 0.09 mL/L and 0.24 mL/L in AS and BoB, respectively. The OMZ is broader in the AS as compared to the BoB (Fig 2 e, f and 3 e, f), starting from ~ 110 m to around ~ 1230 m in the AS and from 110 m to ~ 820 m in the BoB. This means that the OMZ in the AS is thicker (~ 1100 m) than the BoB (~ 700 m). The AS OMZ thickness tapers towards the south as the water mass having high oxygen enters into the AS from the south (Fig. 2 e and 3 e). The water column between 1000 m and 2000 m reported an average dissolved oxygen of 0.97 mL/L and 1.59 mL/L in the AS and BoB, respectively. The 2000 m water column has low dissolved oxygen in the AS with an average of 0.7 mL/L and 1.04 mL/L in AS and BoB, respectively. The Chlorophyll - *a* clearly showed higher biological productivity in the AS during SIM, especially the northern station (II-14) had high Chlorophyll - *a* reaching 1.36 mg/m^3 (Fig 2 g and h), while during SWM the BoB had high Chlorophyll - *a* (Fig 3 g and h).

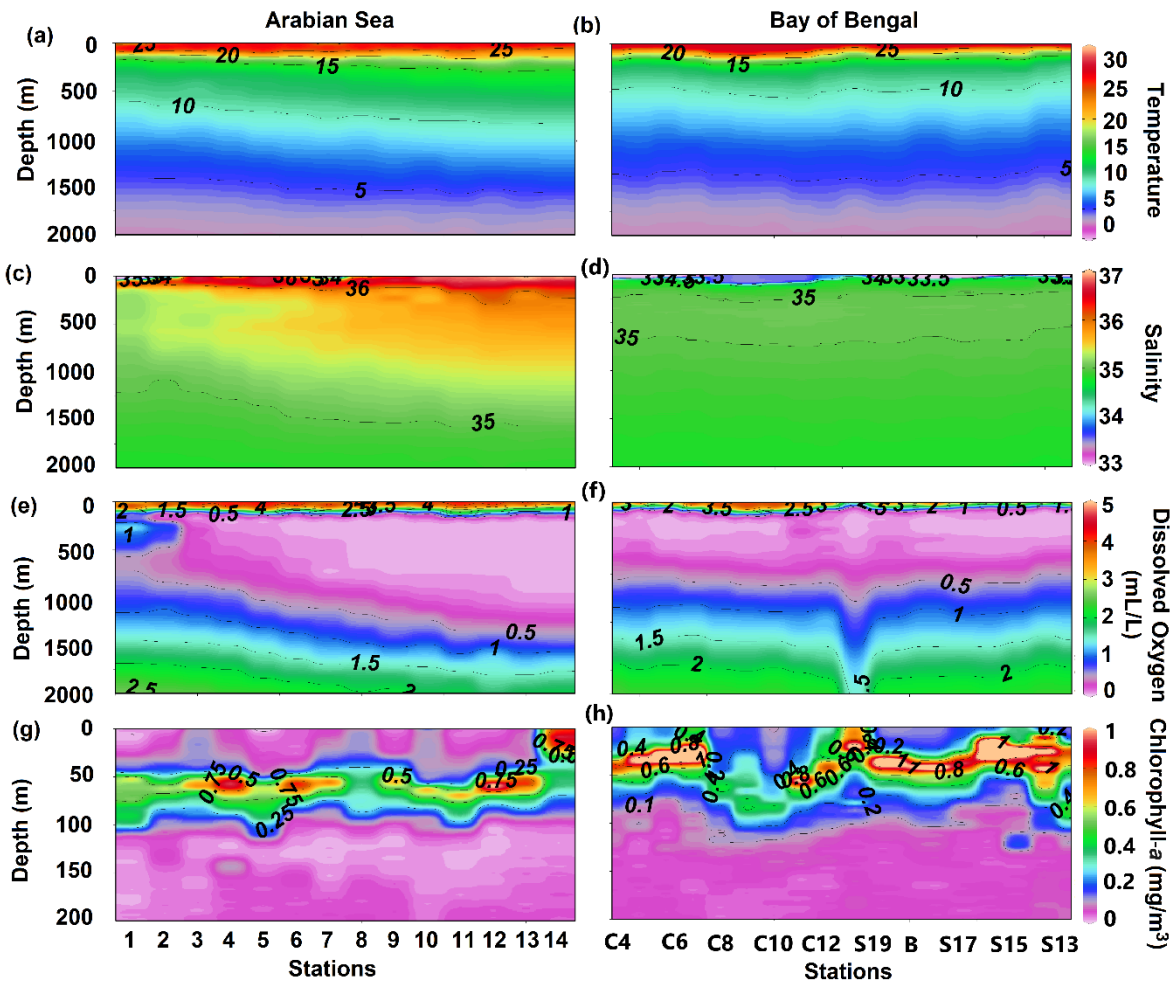


Fig 3 Variations of temperature ($^{\circ}\text{C}$), salinity, dissolved oxygen, and Chlorophyll - *a* (200 m) in the AS (left panel) and BoB (right panel) during SWM.

The above-mentioned results of high temperature and low salinity in surface layers of BoB as compared to AS are better presented in Fig. 4 a and b. Also, the thicker OMZ in AS as compared to the BoB is evident in Fig. 4c. Although the difference in temperature and salinity between the AS and BoB is minimal in the deeper waters (2000 m), dissolved oxygen in BoB remains high as compared to the AS. The low dissolved oxygen in the AS led to denitrification and NO_2 concentrations of $\sim 2 \mu\text{M}$, while no such build-up of NO_2 was observed in the BoB (Fig. 4 d). The TBC was high in AS (maximum counts: 0.8×10^9 cells/L) as compared to BoB (maximum counts: 0.27×10^9 cells/L) throughout the water column (Fig. 4e). The high TBC was associated with high TOC in the AS as compared to BoB with a difference of $\sim 13 \mu\text{M}$. However, at deeper depths, the BoB TOC was lower by only $5 \mu\text{M}$ (Fig 4f).

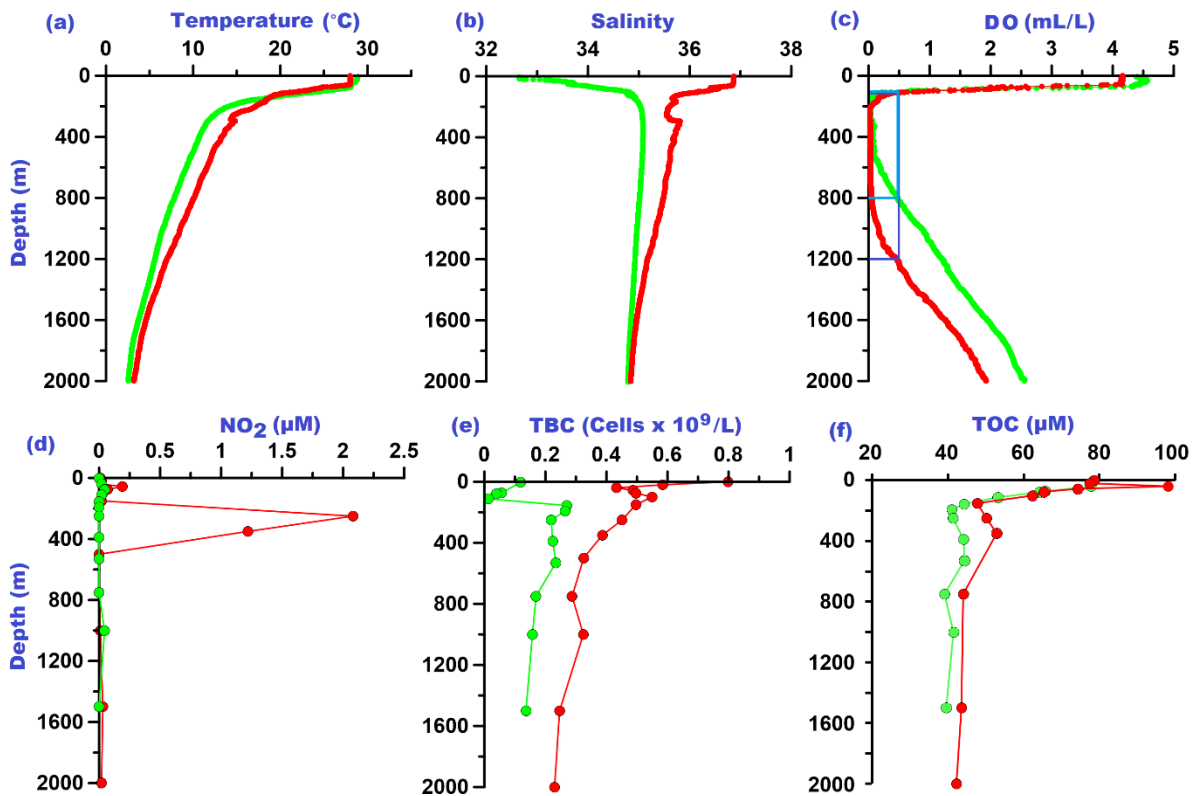


Fig 4. Vertical profiles of physico-chemical and biological parameters at the ASTS (red) and BoBTS (green) during 2016. (a) Temperature (b) Salinity (c) DO (d) NO_2^- (e) TBC and (f) TOC

3.2 Seasonality in the AS and BoB

The AS and BoB show strong seasonality in the physico-chemical parameters as seen in Fig. 5. The temperatures are high during SIM and FIM in the AS, while in BoB the temperatures are high during FIM (Fig 5 a and b). The AS temperature shows a minimal change in the top 70 m with a deeper MLD during the NEM (Fig 5a, supplementary table 2). The temperatures in the BoB during SIM follow the AS trend with minimal changes in the top 70 to 80 m (Fig 5 a and b). The salinity is high during SWM in the AS, while it's low in the BoB during SWM dropping below 28 in the upper 20 m. Salinity remains relatively high in BoB during SIM (>32) (Fig 5 c and d). During NEM, salinity also follows a similar trend of temperature in the AS wherein there is minimal change in the upper 70 m (Fig. 5c). Chlorophyll - *a* in the BoB was high as compared to the AS during SWM reaching up to 2 mg/m^3 (Fig 5 e and f). Also, the Chlorophyll - *a* peak occurs at a much shallow depth ($\sim 30 \text{ m}$) in the BoB during SWM while the peak was deep during SIM for both basins.

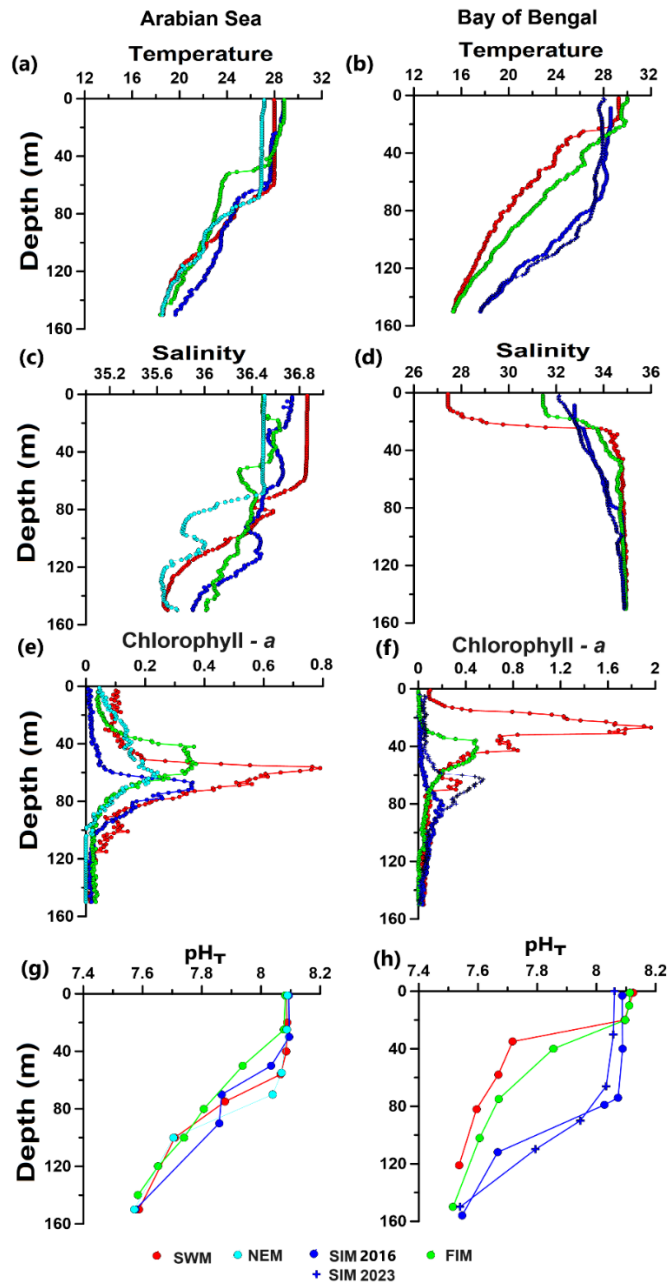


Fig 5 Seasonal variations of temperature ($^{\circ}\text{C}$) (a, b), salinity (c, d), Chlorophyll - *a* (mg/m^3) (e, f), and pH_T (g, h) at the ASTS and BoBTS respectively. Please note that the scales are different for salinity and Chlorophyll - *a*.

3.3 pH_T variations in the AS and BoB

pH_T in the AS and BoB followed the trends of temperature. While the pH_T in the AS didn't show major seasonal variations in the surface waters, the BoB had marginally lower values in the SIM as compared to SWM and FIM. During SIM pH_T in the BoB followed the trend of AS with the higher surface values upto 80 m depth, whereas pH_T drops rapidly from 20 m to 40 m and reaches as low as 7.72 during SWM and FIM in the BoB (Fig 5 g and h).

In the AS the high pH_T (~ 7.95) isolines deepened to ~ 100 m except for the northern region (Fig 6 a-f). During NEM some upliftment in the isolines (pH_T 8) was observed in the central stations (15 to 16 °N) of the AS (Fig 6 e). Also, a comparison between the SWM of different years (2016, 2017, 2020, and 2021) shows shallowing of the isolines (pH_T 8.05), especially in the southern stations (Fig 6 d). The pH_T in the BoB is low in the top 100 m as compared to the AS, especially between 30 and 100 m (Fig 6). Figure 6 (g - i) clearly shows shoaling of the 7.7 pH_T isoline in the BoB as compared to the AS, especially during SWM.

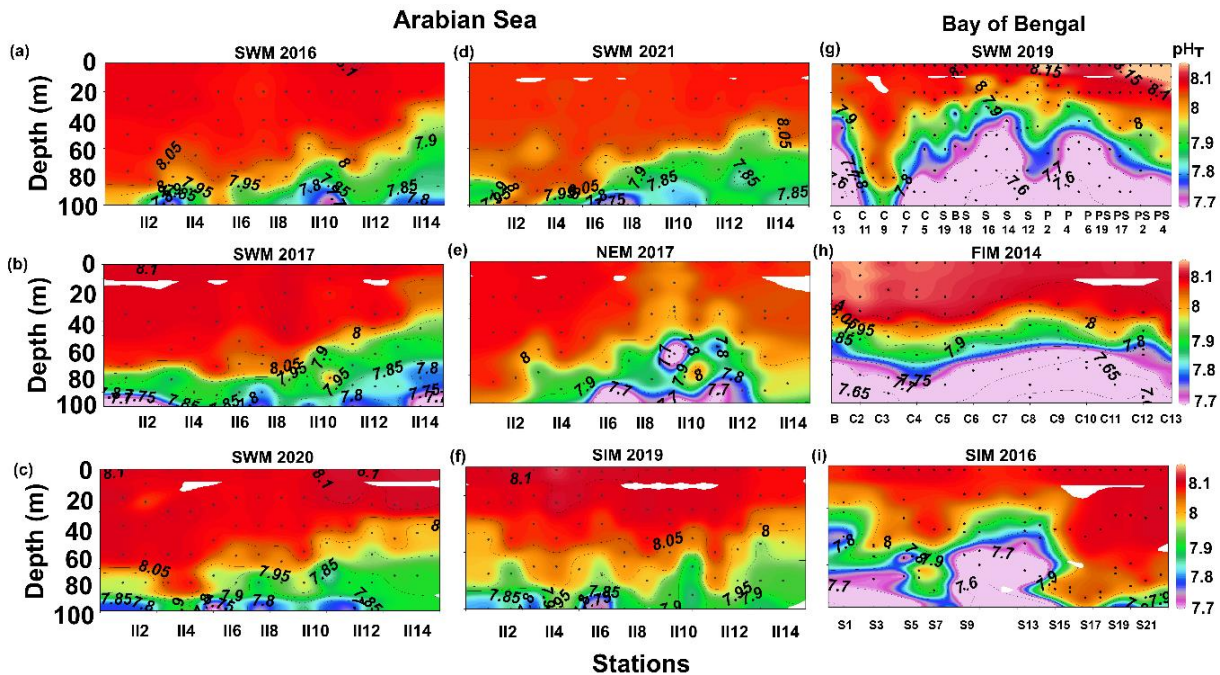


Fig 6. Seasonal variations of pH_T in the AS (left and middle panels (a - f) and BoB (right panel (g - i)) highlighting the variations in the two basins within the 100 m depth range.

Figure 7 highlights the seasonal variation of pH_T in the 2000 m in the AS and BoB transects, clearly showing higher pH_T in the AS OMZ as compared to the BoB. But in the AS low pH_T waters are centered around $12^\circ N$ to $17^\circ N$ (stations II 5 – II 10). Also, low pH_T waters are seen coming from the north at 1000 to 1500 m depth (Fig 7). A comparison between the SWM of different years (2016, 2017, 2020, and 2021) shows low pH_T levels within the AS OMZ, wherein the low pH_T (7.44) isolines reach south up to $13^\circ N$. Also, the NEM had comparatively higher pH_T within the OMZ as compared to SWM or SIM.

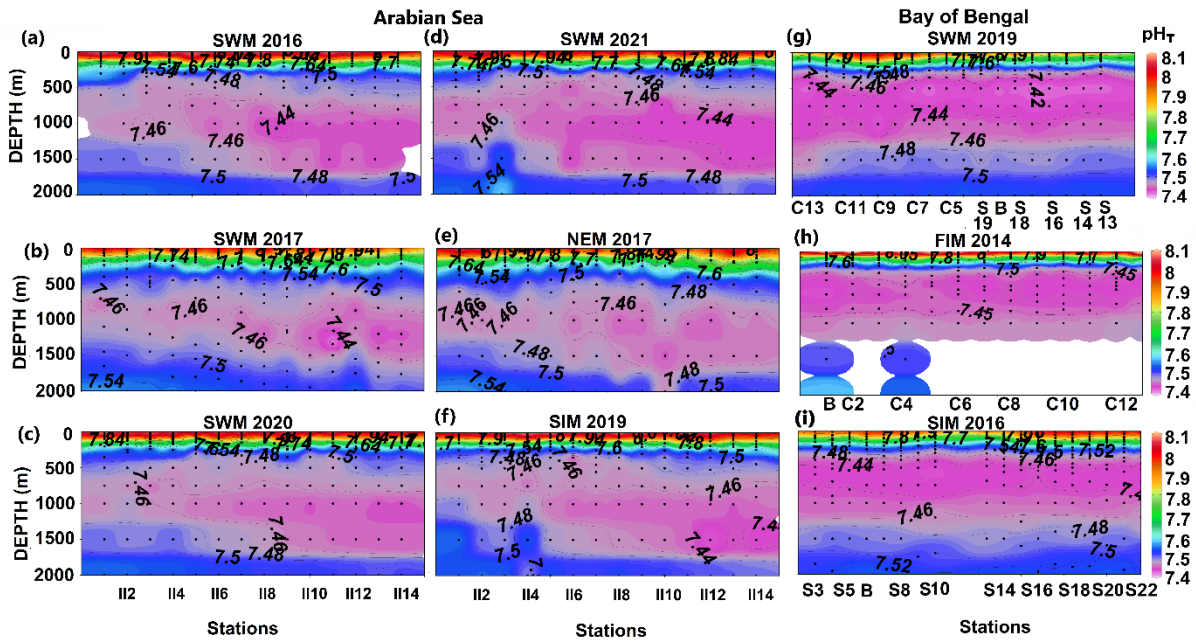


Fig 7 Seasonal variations of pH_T in the AS (left and middle panels (a - f)) and BoB (right panel (g - i)) highlighting the variations in the two basins within the 2000 m depth range.

Figure 8a shows the pH_T variations at ASTS and BoBTS, indicating a decreasing trend from surface to intermediate depth before slightly increasing below ~ 1500 m. The average pH_T (all stations) in the surface layer in AS ($n = 628$) is $8.078 (\pm 0.024)$, while in BoB ($n = 300$) it is $8.045 (\pm 0.111)$. While, the average pH_T within the top 50 m at ASTS and BoBTS is 8.069 and 8.064, respectively. The pH_T within the surface layer (< 50 m) of AS and BoB shows lower values by $0.033 (\pm 0.03)$ in the BoB. The pH_T within the 150 to 1500 m layer in these two basins shows lower values by 0.03 ± 0.02 in the BoB. The average pH_T (all stations) within the 150 to 1500 m layer in AS is 7.49 ± 0.05 , while in BoB it is 7.46 ± 0.05 . While, the average pH_T within the 150 to 1500 m layer at stations ASTS and BoBTS is 7.486 ± 0.01 and 7.457 ± 0.015 , respectively. As seen from Figure 7a, the lowest pH_T in BoB occurs at shallow depth (~ 350 to 500 m) as compared to AS (~ 1000 m). The lowest and highest pH_T values in the AS are 7.43 and 8.12, while in the BoB it is 7.41 and 8.24, respectively.

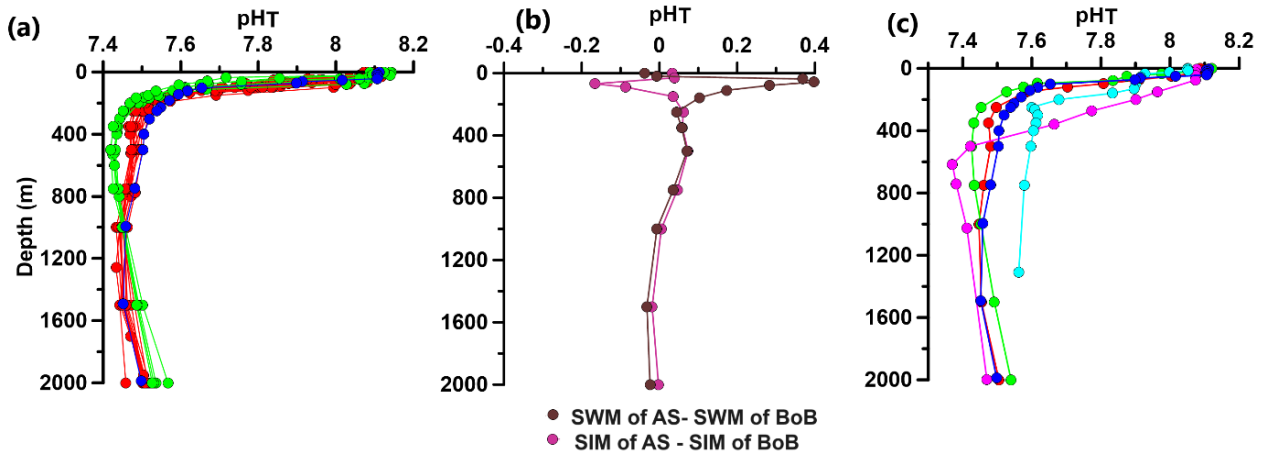


Fig 8 (a) Vertical profiles of pH_T up to 2000 m at the ASTS (present data in red and US-JGOFS data in blue) and at BoBTS (green). (b) The difference in pH_T between AS and BoB during SWM and SIM. (c) Mean of pH_T up to 2000 m at the ASTS (present data in red and US-JGOFS data in blue), BoBTS (green), North Pacific data from HOT (pink), and North Atlantic data from CARIACO (Cyan). The details of sampling (year and season) are given in supplementary table 1.

Figure 8b highlights the seasonal difference in pH_T between the AS and BoB (AS-BoB). During SWM the depths between 35 to 110 m showed a difference ranging from 0.17 to 0.39 indicating low pH_T in BoB within these depths. The difference between the AS and BoB during SIM is -0.165 at 70 m indicating high pH_T waters in the upper 100 m water column in the BoB.

3.4 pH_T comparison with Atlantic Ocean, Pacific Ocean and US-JGOFS

In the North Atlantic, the pH_T decreases rapidly in the 250 m with the lowest value of ~ 7.6 and then slightly increases with the depth (Fig. 8c). While in the North Pacific, the pH_T decreases even more rapidly with the lowest value of 7.37 at ~ 600 m, but then it increases gradually with the depth (Fig. 8c). The pH_T in both AS and BoB (500 to 800 m) is lower than the North Atlantic and higher than the North Pacific waters, however, the pH_T in the 200 to 500 m is lower in the BoB than both these basins (Fig. 8c). A comparison with the US-JGOFS pH_T data gives a long-term decreasing trend of $\sim 0.0009 \pm 0.0005 \text{ yr}^{-1}$ within the 150-1500 m layer of the AS and this rate is lower than the pH_T drop in the surface waters ($0.0011 \pm 0.0005 \text{ yr}^{-1}$).

4. Discussion

The global oceanic surface water pH is 8.07 ± 0.02 , and this is also the same for the Indian Ocean (Jiang et al., 2019). The future predictions indicate a drop in average global surface pH by $\sim 0.33 \pm 0.04$ units from 2000 to 2100 (Jiang et al., 2019). Considering the significant role of Indian Ocean in the global carbon cycle, it is important to understand the variability in the pH_T and the factors regulating its

variability. The NIO being unique due to its landlocked nature has several possible and varying factors that can regulate the variability of pH_T within the AS and BoB. Earlier studies have related pH or pCO_2 variabilities in the AS and BoB with upwelling, convective mixing, temperature, salinity, alkalinity, DIC, biological productivity, aerosol deposition, riverine runoff, etc. (Sarma et al., 2012; 2015; 2023; Sridevi and Sarma 2021; Chakraborty et al., 2018, 2021). Although the variabilities for the surface waters are better understood at regional levels using both *insitu* and modeled studies, the variability in the deeper layers remains elusive. In this study, we present the pH_T variability in the NIO along with its possible controlling factors.

4.1 pH_T variability in the top 150 m of the AS and BoB

The pH_T in the present study is high in the AS as compared to the BoB. The low pH_T in surface waters in the BoB could be attributed to the following factors.

4.1.1 Role of freshwater influx

The AS receives lower volumes of river runoff ($0.3 \times 10^{12} \text{ m}^{-3} \text{ yr}^{-1}$) compared to BoB ($1.6 \times 10^{12} \text{ m}^{-3} \text{ yr}^{-1}$) (Gauns et al., 2005). The BoB is strongly stratified as a result of the warmer and less saline surface water (Figures 2-4). The stratification index confirms higher stratification in the BoB than in the AS (supplementary Fig 1). Strong stratification prevents nutrient supply to the surface waters (Prasanna Kumar et al., 2002) thus making the BoB less productive than the AS (Madhupratap et al., 2003). Also, the primary productivity in the AS is reported to be 8 times higher than the BoB (Gauns et al., 2005). However, recent studies show comparable results in both the basins. Singh and Ramesh (2015) suggest that the BoB could be as important as the AS in its efficiency in exporting carbon to the deep ocean. The TOC in Fig. 4 indicates similar concentrations at deeper depths in the AS and BoB. Also, the productivity in the BoB during SWM is high as compared to the AS.

The river water can influence the pH_T of BoB by bringing in alkaline water, affecting the mixed layer dynamics by inducing strong stratification leading to shallow MLD (supplementary table 2), and supplying nutrients that are required for phytoplankton. The pH_T in the BoB follows more seasonality than the AS. Among all the seasons the SIM had low pH_T in the surface waters, however, the levels remained uniform up to 100 m and the pH_T followed the trend of temperature (Fig. 5). Despite having higher pH_T at the surface during the SWM and FIM, the levels dropped drastically from 20 to 40 m. The drop in pH_T coincided with the fall in salinity indicating that due to shallow MLD (supplementary table 2), the surface values are high. Chakraborty et al. (2021) suggested that the governing mechanisms of the seasonal cycle of pH in the AS and BoB are different, the former being due to the upwelling/downwelling mechanism and the latter due to the upper ocean mixed layer dynamics. Figure

9b clearly shows the input of freshwater having higher pH_T during the SWM. High pH_T values in the northern bay can be attributed to alkaline waters from the Himalayan rivers, as also suggested by Sarma et al. (2012) and Sridevi and Sarma (2021). Chakraborty et al. (2021) through a modeling study highlighted the role of freshwater spreading and suggested that salinity plays a unique role in inducing seasonal variability of pH_T in the northern head BoB. The reduced freshwater input during SIM results in relatively low pH_T levels (Fig. 9a), apart from other possible contributions such as aerosol deposition (Sarma et al., 2015; 2021).

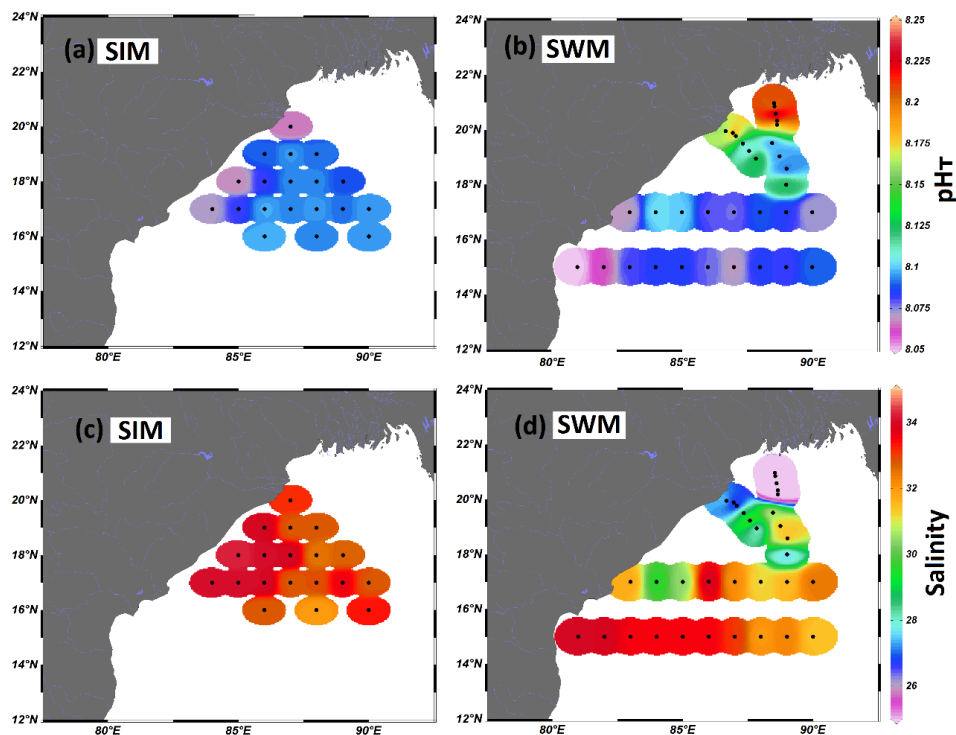


Fig 9 Spatial variation of pH_T and salinity in the BoB surface waters during SIM and SWM

4.1.2 Role of eddies

Eddies play an important role in the biogeochemistry of the BoB by transferring nutrients into the euphotic zone and increasing primary production (Gomes et al., 2000, Prasanna Kumar et al., 2004; Singh et al., 2015). The dominant warm-core eddies in the BoB oxygenate the subsurface thereby reducing the vertical extent of the OMZ (Sarma et al., 2018). The Sea Surface Height Anomaly (SSHA) suggests that the entire BoB is covered by several warm-core and a few cold-core eddies during the SWM of 2019 (Fig. 10a). The SSHA was > 0.3 m in the case of the warm-core eddy. Dandapat and Chakraborty (2016) reported that although the number of cold-core eddies is less than warm-core during most parts of the year, the cold-core eddies have a longer lifetime and are more

energetic. Sarma et al. (2019) reported eddies of variable sizes are present in BoB, with warm-core eddies ranging from 400-600 km and cold-core eddies from 50-150 km. The shoaling of 7.7 pH_T isoline up to ~50 m was seen at the cold-core eddy region but the same was deepened to ~100 m in the warm-core eddy region (Fig. 10b). Similar findings were reported by Sarma et al. (2019), showing an upsloping of 7.8 pH isoline to 50 m in cold-core eddies as compared to 125 m in the rest of the region.

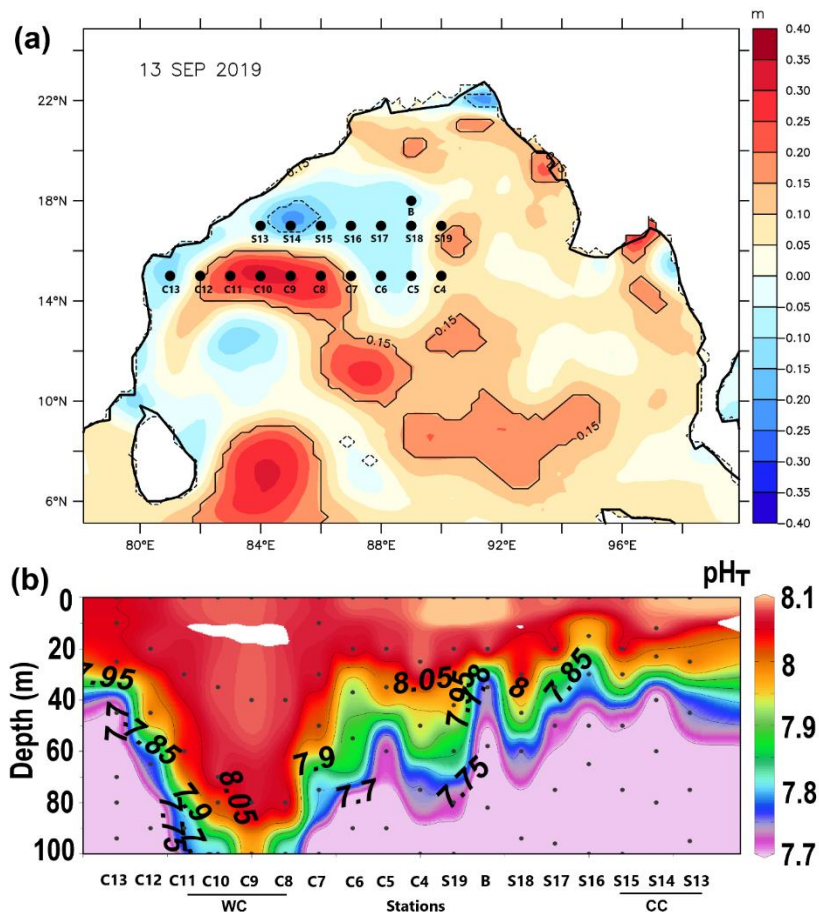


Fig 10 (a) Variations of SSHA and (b) pH_T in the BoB during SWM of 2019. WC represents stations within the warm-core eddy region while CC represents stations within the cold-core eddy region.

4.2 Plausible factors regulating the contrasting pH_T patterns in OMZ of the AS and BoB

The pH_T within the 150-1500 m of these two basins differs by 0.03 (± 0.02) with lower values in the BoB. A significant finding is that the lowest pH_T in BoB occurs at 350-500 m, while in AS it occurs at ~1000 m. Although the anthropogenic uptake of CO_2 is expected to be transported to deeper waters, the observed changes within the two basins can be governed by circulation patterns and

biogeochemical changes. In the following sections, we discuss the possible factors regulating the contrasting pH_T patterns in the AS and BoB.

4.2.1 Role of water masses

Both AS and BoB experience surface warming in April-May and October and surface cooling during SWM and winter months (DineshKumar et al., 2016). They also reported that from 1960 to 2009, the rate of SST increase was almost uniform at $0.12\text{ }^\circ\text{C/decade}$ in both AS and BoB. The BoB intermediate waters are cooler as compared to the AS and therefore can hold more CO_2 (Fig. 4 a). The water masses in the AS and BoB have been plotted in Figure 11a with a comparison between 1995 and 2018 which showed that the water masses in the two basins have not changed from 1995 to 2018. The AS has Arabian Sea high saline water (ASHSW) and the Persian Gulf water (PGW), while the BoB has the BoB Water mass (BoBW), the Mixed Zone (MZ), and the Indonesian Through Flow water mass (ITF). The MZ water mass is identified at the boundary of BoBW and ITF and can be seen in the depth range between 60 m and 100 m (Sengupta et al., 2013). Earlier Sengupta et al. (2013) reported the presence of ITF water between 100 and 500 m in the BoB. After entering the Indian Ocean, the ITF spreads through three key pathways, one each at the surface (0-50 m), intermediate depth (50-200 m), and deep ocean (200-500 m) (Valsala and Ikeda, 2007).

The ITF is derived from Pacific Ocean Central Water and is considerably older than BoBW (Dutta and Bhusan, 2012). Figure 8c clearly shows that the BoB is influenced by the Pacific water. Earlier, Culberson & Pytkowicz (1970) had given a similar trend of pH for the BoB as that of the eastern Pacific with the lowest pH between 350 and 500 m. Recent data from HOT (Pacific) and CARIACO (Atlantic) stations clearly shows that the pH_T in BoB is more influenced by the Pacific waters with low pH_T up to 7.31 (Fig 8c). The interesting highlight is that the potential density (27 kg/m^3) of the lowest pH_T depth (~650 m) in the North Pacific matches the depth of the lowest pH_T in the BoB (~500 m). Based on particle tracking and tracers, Valsala and Ikeda (2007) reported the different pathways of ITF water reaching the BoB. The detailed pathways of the ITF transport to the northern BoB through South Java current (SJC) are given in Figure 6 and Figure S2 of Makarim et al. (2019). Their study also showed that the presence of ITF water was observed in the depth range between surface and 500 m and it takes about 9 months (~10 cm/s) to reach off Sumatra (Schott et al., 2009). It further moves northward and reaches the BoB.

A few earlier studies also suggest longer residence time of water masses in the BoB than AS (Howell et al., 1997; Sarma, 2002a; b), and such older waters can lead to low pH_T in the BoB. Figure 11 shows the different water masses in the South Indian Ocean (SIO) and ITF for the period 1995 and 2018. The

water mass properties look similar in the ITF, SIO, and BoB below 500 m. Earlier studies confirmed the advection of the western Pacific water in the SIO through ITF (Lee et al., 2015; Makarim et al., 2019; Vidya et al., 2020). The ITF water from the western Pacific advects to the BoB through the SIO as suggested by the TS plots of BoB that match with that of SIO and ITF (Fig. 11 a & b). Recently Rahman et al. (2021) also showed the presence of Pacific water coming through ITF water in the BoB. Moreover, the Andaman Sea also has a role in transporting equatorial waters to the BoB (Chatterjee et al., 2017). From the above studies, it is clear that Pacific water reaches and resides in BoB and since Pacific water has a low pH_T as seen in Figure 8c, it can be concluded that the Pacific water regulates the pH_T within the OMZ of the BoB.

The above studies highlight the possible contribution of ITF waters in the BoB. On the other hand, Sengupta et al. (2006) demonstrated that BoB freshwater reaches remote regions of the tropical Indian Ocean on seasonal time scales. In particular, BoB runoff appears to join the ITF and flow west in the South Equatorial Current. The rain and riverine water lead to an inter-ocean transport tendency of water masses from the BoB to the AS over a depth range from the surface to 500 m as a compound impact of all the freshwater entering the tropical Indian Ocean over 60 years (Valsala 2022). The riverine water was even found to impact the biogeochemical cycles of the Indian Ocean (Valsala et al., 2020).

In the AS, the Indian Central Water is reported to be the primary source of oxygen to the OMZ at depths between 200 and 500 m (Shenoy et al., 2020). It is evident from Figure 7 that relatively higher pH_T water enters the AS from the south, thus indicating that the Indian Central Water apart from being a source of oxygen is also a source of high pH_T waters to the OMZ of AS.

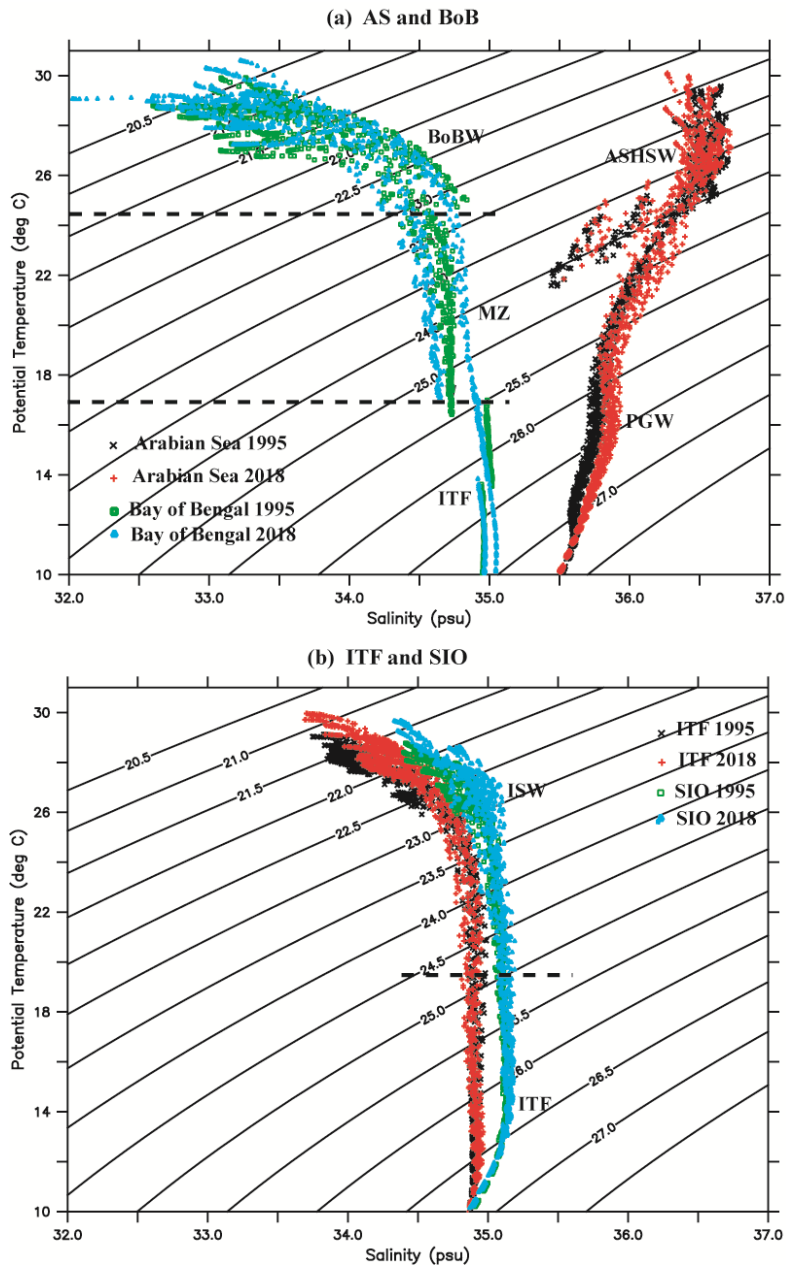


Fig 11 Potential temperature-salinity (T-S) plot of the (a) AS (64°E-70°E and 5°N-20°N), and BoB (85°E-90°E and 5°N-20°N), (b) Southern Indian Ocean (60°E-90°E and Equator-20°S) and Indonesian Through Flow region (80°E-110°E and Equator-15°S).

4.2.2 Role of OMZ on pH_T variation

The AS and BoB are characterized by a well-defined OMZ at sub-surface depths (Naqvi et al., 2006). The AS has a thicker perennial subsurface OMZ than the BoB, which is linked to high plankton production via upwelling and winter convection in the former and river influx and mesoscale eddies in the latter (Vidhya et al., 2022). The vertical profiles of dissolved oxygen in the two basins clearly showing the deeper lower boundary of OMZ in the AS (Figure 4). Also, the core of the OMZ was

deeper in the AS. This is in agreement with Paulmier and Ruiz-Pino (2009) who reported that the core of the AS OMZ is ~20% deeper as compared to other OMZs. They also reported that the thickness of OMZ core is 760 m in the AS, while the closest to it is Eastern Tropical North Pacific (ETNP) with 420 m thickness, while the global average is only 340 m. The interesting highlight is that the core of the OMZ is deeper in the AS and the low pH_T observed coincides with waters having dissolved oxygen around 0.1-0.2 mL/L in both the basins. This indicates that the degradation of organic matter mostly happens at the lower boundary of the OMZ when dissolved oxygen rises above 0.1 mL⁻¹. Therefore, the ongoing ocean deoxygenation events such as intensification and deepening of OMZ in the AS (Lachkar et al., 2018; 2019; 2023) could deepen the depth of low pH_T in the AS. The observed difference in the surface TOC between the two basins (13 μ M) was not reflected at deeper depths, and the BoB TOC was lower by only 5 μ M in deeper waters (Fig 4f). The high TOC in AS also supports high TBC in the AS (Fig 4e). Degradation of organic carbon by heterotrophic bacteria can support increased microbial respiration, which is expected to lower oxygen and pH (Robinson et al., 2019). However, the low TOC and TBC in BoB rule out high respiration and remineralization in the BoB as compared to the AS. Higher temperature can also lead to high heterotrophic processes such as microbial respiration which consume more oxygen than what is generated through autotrophic processes (López-Urrutia et al., 2006; Wohlers et al., 2009). Although the higher temperatures in AS will increase respiration rates (and thus lower pH_T), it will also hold less CO₂ leading to higher pH_T as compared to BoB. An improved understanding of the magnitude and variability of respiration in these basins is needed for better predictions of processes like ocean deoxygenation and ocean acidification as suggested by Robinson et al. (2019).

4.2.3 Difference in remineralization depth

Based on sediment traps deployed in the AS and BoB, lithogenic fluxes were calculated at two depths in these basins. Higher lithogenic flux was seen at the deeper depth in the BoB (60.7 mg/m²/d) as compared to AS (39.3 mg/m²/d). However, the traps at shallow depth show higher lithogenic flux in AS (39 mg/m²/d) than in BoB (15.3 mg/m²/d). The most important finding is that the difference in lithogenic flux between the shallow (600 m) and the deep trap (1200 m) in the AS is less (0.3 mg/m²/d), while the difference in the shallow (430 m) and the deep trap (1020 m) in the BoB was large (45.4 mg/m²/d). Earlier Ramaswamy (1993) also suggested a large difference between lithogenic fluxes in the surface and deeper depths in the BoB as compared to AS. Naqvi et al. (1996) suggested that the rapidly sinking lithogenic matter protects organic matter from oxidation, thus leading to lower oxidation rates in the BoB. Azhar et al. (2017) also suggested deeper remineralization depth in the BoB. It is evident from our sediment trap data and previous studies that the lithogenic flux is high at

deeper depths (>1000 m) in the BoB, and not at the shallow depths (400 to 500 m) indicating that the organic matter gets transported to the deeper depths due to ballast effect. This reflects that the organic matter within the OMZ of the BoB does not get sufficient time to remineralize at shallow depths, thereby ruling out the differences in remineralization depth in these basins for the occurrence of low pH_T in the BoB.

4.2.4 High Temperature and Denitrification in the AS

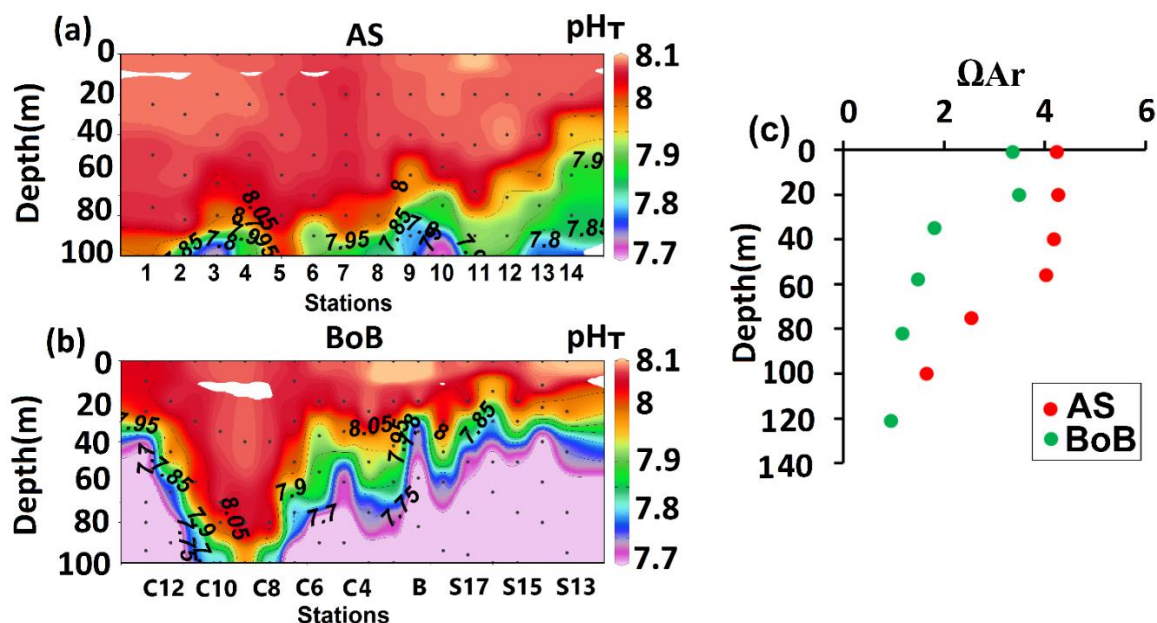
This study highlights the low pH_T waters in the surface and OMZ of the BoB. The average temperature for the water column from 150 to 1000 m is $\sim 12^\circ\text{C}$ and 9.75°C in AS and BoB, respectively indicating a difference of $>2^\circ\text{C}$ in the two basins. Due to high temperature the AS water will hold less CO_2 and thereby have higher pH as compared to the BoB. The AS is also known for denitrification with a prominent secondary nitrite peak (Fig. 4 d) while the non-availability of favorable conditions in the BoB inhibits denitrification (Naqvi, 2006; Bristow et al., 2016; Johnson et al., 2019). However very recently nitrous oxide has been reported in BoB, similar to that of AS (Toyoda et al., 2023). Denitrification is known to increase ocean pH (Almogi-Labin et al., 1986) in OMZs which are otherwise known to lower the pH (Panchang and Ambokar 2021). Almogi-Labin et al. (1986) reported that the good preservation of pteropods at 1050 m depth in the Red Sea was due to denitrification conditions and an increase in pH. Also, in the coastal marine ecosystem, it has been demonstrated that low pH enhances nitrification (Fulweiler et al., 2011). Therefore, the observed differences in the pH_T within the OMZ of AS and BoB could also be due to difference in their denitrification potential.

4.3 Implication of low pH_T in BoB

4.3.1 Effects on marine organisms

The pH_T in the BoB is low as compared to the AS, especially between 30 and 100 m. Figure 12a and b clearly shows a shoaling of the 7.7 pH_T isoline in the BoB as compared to the AS, especially during SWM. Whereas in the AS the high pH_T (~ 7.95) isoline deepened to ~ 100 m. The low pH_T in the BoB waters is reflected in the low Ω_{Ar} values (< 1) that indicate under-saturation of calcium carbonate (Fig 12c). The low Ω_{Ar} and pH_T observed in the BoB at shallow depths (< 100 m) can have an adverse effect on the marine plankton inhabiting these depths, especially the ones with aragonite shells. Pteropods are known to be more susceptible to low pH because of their thin aragonite shells (Mekkes et al., 2021a). Mekkes et al. (2021b) have shown that the pteropod shell thickness declined by $\sim 37\%$ due to low pH in the California Current Ecosystem. Recently, Shetye et al. (2023) have shown the effects of low pH on calcareous marine plankton such as coccolithophores in the coastal waters of the AS. Although historically AS is known for being a source of CO_2 , mostly because of upwelling, the

findings of the present study clearly show that the BoB plankton are at major risk even in the open waters. However, there are no studies on the effects of low pH_T on marine organisms in the BoB which warrants the need for a better assessment of the fate of calcareous plankton/organisms in low pH_T waters.



Fig

12 Vertical profiles of pH_T in (a) AS and (b) BoB during SWM. (c) Aragonite saturation state in the AS and BoB during SWM.

4. Conclusion

Here we present the first report of time-series measurements (based on 19 cruises) of seawater pH_T in open ocean waters of the AS and BoB and the plausible reasons for the low pH_T observed in the BoB. The pH_T within the 30 -100 m in the BoB is lower during SWM by around 0.39 units as compared to AS which could be due to freshwater influx from rivers, mixed layer dynamics, and cold-core eddies. The Ω_{Ar} is < 1 within the top 100 m of BoB indicating that the plankton in the BoB are facing a major risk from OA as compared to the AS. The pH_T within the 150-1500 m layers of these two basins shows lower values by 0.03 (± 0.02) in the BoB. A significant finding is that the lowest pH_T in BoB occurs at 350-500 m, while in AS it occurs at ~ 1000 m depth. Also, the pH_T in the two basins is lower than in the North Atlantic and higher than in the North Pacific waters. The low pH_T in BoB as compared to AS could be due to intrusion of western Pacific water in the BoB, the freshwater influx from rivers, variations in OMZ of the two basins, higher temperature ($\sim 2^\circ\text{C}$) within the OMZ of the AS, denitrification in the AS and also due to the differences in lithogenic material in the two basins. We document a significant long-term decreasing trend of pH_T by 0.0009 yr^{-1} within the 150-1500 m of the AS as compared to the US-JGOFS data, and this rate is lower than the pH_T drop in surface waters. A

continuous monitoring of the rate of pH_T changes is required through precise time-series measurements in the NIO. Apart from this, the role of Pacific waters in governing the pH_T variability in the BoB needs a detailed investigation with a focus on the ITF region.

5. Data availability

The pH data for the Atlantic Ocean, Pacific Ocean, and Arabian Sea JGOFS are taken online (from <https://imars.usf.edu/>, <https://hahana.soest.hawaii.edu/hot/methods/ph.html>, <http://usjgofs.whoi.edu/>). The temperature and salinity data from SIO and ITF for calculation of potential density are taken online (from <https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-oras5?tab=form>). The SSHA data was taken from <https://cds.climate.copernicus.eu/cdsapp#!/dataset/satellite-sea-level-global?tab=form>

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Statements & Declarations

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Competing Interests

Authors declare that they have no financial or competing interest

Author Contributions

Authors contributions: SS: Conceptualization, Methodology, Analysis, Writing; SK: Analysis, Writing—review and editing; DS: Writing—review and editing; MG: Analysis, Writing—review

and editing; AP: Analysis; GS: Analysis; HN: Analysis, MF: Methodology, analysis; VP: Analysis; NK and AS: analysis

Ethical approval

We all authors read and approved the final manuscript.

Consent to participate

All authors agreed to participate in this paper.

Consent for publication

All authors agreed to publish this paper.

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Supplementary Table

Table S1: The sampling details including the cruise number, season, and year

Basin	Cruise No.	Season	Year
AS	SSK 69	SWM	2014
AS	SSK 79	SIM	2015
AS	SSD 12	SWM	2015
AS	SSK 87	NEM	2016
AS	SSD 26	SWM	2016
AS	SSD 33	NEM	2017
AS	SSD 41	SWM	2017
AS	SSD 61	SIM	2019
AS	SSD 72	SWM	2020
AS	SSD74	SWM	2020
AS	SSD 81	SWM	2021
AS	SSD 84	FIM	2021
AS	SSD 76	NEM	2020
BoB	SSD 002	SWM	2014
BoB	SSD 005	FIM	2014
BoB	SSD 15	FIM	2015
BoB	SSD 20	SIM	2016
BoB	SSD 65	SWM	2019
BoB	SSK154	SIM	2023

Supplementary Table 2

Basin	Season/Year	MLD (m)
AS	SWM 2016	49.8
	NEM 2017	69.8
	SIM 2019	45.2
BoB	SWM 2019	13.4
	SIM 2016	23.9
	FIM 2015	10.3

Supplementary figure 1: Bruñt Vaisala frequency in the upper 100 m of the AS and BoB

