Modeling the seasonal variability and the governing factors of Ocean Acidification over the Bay of Bengal region

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Abstract

The Bay of Bengal (BoB) is a high recipient of freshwater flux from rivers and precipitation, making the region strongly stratified. The strong stratification results in a thick barrier layer formation, which inhibits vertical mixing making this region a low-productive zone. In the present study, we attempt to model the pH of the BoB region and understand the role of different governing factors such as sea-surface temperature (SST), sea-surface salinity (SSS), dissolved inorganic carbon (DIC), and total alkalinity (TALK) on the seasonality of sea-surface pH. We run a set of sensitivity experiments to understand the role of each of the governing factors. The results show that the SST, SSS, and DIC are the principal drivers affecting the sea-surface pH, while TALK plays a buffering role. The SST and DIC are consistently found to be opposite to each other. The pre-monsoon season (MAM) has shown to have an almost equal contribution from all the drivers. In the pre-monsoon season, the SST and DIC are balanced by TALK and SSS. The role of SSS is significantly dominant in the second half of the year. Both SST and SSS counter the role of DIC in the southwest monsoon season. The strong stratification plays an essential role in modulating the pH of the BoB region. The thickness of the barrier layer formed in the sub-surface layers positively affects the sea-surface pH. The northern BoB is found to be more alkaline than the southern BoB. Our study highlights the complexity of ocean acidification in the BoB region compared to the other part of the world ocean.

Keywords: Bay of Bengal (BoB), pH, sea-surface Salinity (SSS), Stratification, Barrier Layer Thickness (BLT)

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1. Introduction

The pronounced absorption of the anthropogenic atmospheric carbon dioxide by oceans (2.5 ± 0.6 GtC yr⁻¹, Friedlingstein et al., 2020) causes a drop in oceanic pH referred to as Ocean Acidification. A few studies indicate a drop of 0.1 units in the upper ocean pH, and it is predicted to drop by almost 0.3 to 0.5 units by the end of this century (Sabine et al., 2004; Feely et al., 2009; Kwiatkowski and Orr, 2018). This reduction in sea-surface pH has a major effect on the biological process and production in oceans (Gattuso et al., 2014). These long-term variations in oceanic pH values include short-term fluctuations (diurnal, seasonal, and interannual). The extent of vulnerability of these short-term fluctuations is equal or sometimes greater than the long-term changes (Provoost et al., 2010; Wootton and Pfister, 2012; Sutton et al., 2014). Hence to improve the predictions of ocean pH variability, identification of drivers affecting it is of utmost importance.

The Bay of Bengal (BoB) region chosen in this study is peculiar by reference to its geographical settings (enveloped by land from the three sides and open in the south), the substantial freshwater influx from rivers and precipitation (Unesco, 1969), and the seasonal reversal of coastal currents (Murty et al., 1993; Shetye et al., 1996). The abundance of freshwater in the BoB region makes this region one of the challenging zones for ocean acidification studies. The ocean acidification scenario for the BoB region, as reported by Feely et al. (2009), suggests the pH to be below 8.0 by 2050 and may go below 7.8 in 2095. Since the BoB is a reservoir of an abundance of marine species, especially shells and coral reefs, the ocean acidification scenario presented by Feely et al. (2009) is of great worry to the scientist, environmentalists, and policymakers.

Mukhopadhyay et al. (2002) noted the lowest pH in Mahanadi estuaries (7.02 ± 0.1) and highest in the Ganges estuaries (8.13 ± 0.24). Sarma et al. (2012), based on ship observations in the western coast of BoB, showed that the northwestern coast has higher pH (8.45 ± 0.003) than the southwestern coast (8.12 ± 0.03). The lowest pH was observed in the Godavari estuaries (8.07 ± 0.05), while the Mahanadi estuaries demonstrated the highest pH values (8.41 ± 0.005). The sharp gradient between the northwestern and southwestern pH values are associated with the difference in salinity (31.96 ± 0.88 for Godavari estuaries and 23.24 ± 1.84 for Mahanadi estuaries).

Sarma et al. (2015) compares the measured hydrographic properties and inorganic carbon components of the BoB between March-April of 1991 and 2011, this work also includes a time
series observation of 8 years from 2005 to 2013. All these observations were of the western coast of BoB. The study suggests that the rate of decrease in pH in the southwestern coast was consistent with the global trend, but the northwestern coast experienced a higher decrement rate of pH (3 to 5 times higher). The time series observation at Vishakhapatnam (located on the southwestern coast of BoB) and Paradip (located on the northwestern coast of BoB) demonstrates a decline in the pH at -0.0015 units year\(^{-1}\) and -0.005 units year\(^{-1}\), respectively. The higher declining rate in at the Paradip station is attributed to enhanced aerosol depositions in the BoB.

The seasonal reversing coastal currents are known as the East India Coastal Currents (EICC) or Western Boundary Currents (WBC), play a significant role in controlling the sea-surface salinity (SSS) of the BoB region. From February to May, the northward moving EICC increases the salinity, which weakens the stratification, and enhances coastal upwelling, resulting in a decrease of sea-surface pH \cite{Sarma2018}. The EICC flows southwards during October to December, which brings in low saline, more basic waters from north to the western coast of BoB, that increases the pH of this zone \cite{Sarma2018}. Since the circulation pattern of BoB consists of many eddies, they play a significant role in modulating the sea-surface pH \cite{Sarma2019}. Based on the observation in the western BoB, reports the presence of low pH values in the cyclonic eddy and no-eddy regions (upper 200m) of BoB, whereas the high pH waters extends up to 150 m to 175 m deep in the anticyclonic eddy regions.

Using Multiple Linear Regression (MLR) Sridevi and Sarma \cite{Sridevi2021} shows an increase in pH in the BoB (1998-2015), the only exception being the head bay region. The near proximity of the head bay to the land makes it vulnerable to the atmospheric and river pollutants. The acidification of the head bay is associated with the rise in sea-surface temperature and atmospheric depositions, as well as decrease in freshwater discharge. The SSS plays a major role in controlling the seasonality of the northern BoB (seasonal amplitude of \(\approx 0.18\) units) \cite{Chakraborty2021}. Chakraborty et al. \cite{Chakraborty2021} using interannual model for the North Indian Ocean (NIO) shows the contribution of the various drivers on the seasonality of total pH and \(p\mathrm{CO}_2\) for the Arabian as well as the BoB region.

In this study, we aim to model the seasonality of pH (based on the climatological modeling) for the whole Bay of Bengal region and explore the physical factors driving the seasonality of pH in this region. As the BoB is recipient of large freshwater influx from precipitation and rivers, it affects the stratification dynamics of the BoB. Through this study we attempt to analyze the role
of different drivers influencing the pH variability and the role of stratification in monitoring the pH variability. The remaining paper is structured as follows: Section 2 describes the Data and Methodology used in this study; Results and Discussion are presented in section 3; Conclusion in section 4.

2. Data and Methodology

2.1. Model

In our previous study (Joshi et al., 2020), we exhibit a comparison between two types of model configurations (the first one using the Fairall et al. (1996) bulk formulation to calculate the wind stress and evaporation. In the second configuration, we externally provide the Wind Stress and evaporation minus precipitation (E-P) data). We concluded that the method of externally providing the wind stress and E-P data emulated the carbonate chemistry satisfactorily. We provide an exhaustive evaluation of the different physical (current, sea-surface temperature, sea-surface salinity, barrier layer thickness, mixed layer depth) and carbonate parameters (dissolved inorganic carbon, total alkalinity, pH, and $pCO_2$) in Joshi et al. (2020). Using the same model, we further explore the influence of the freshwater plume spreading and the barrier layer thickness on the sea-surface $pCO_2$ (Joshi et al., 2021). Using this model we also explore the different mechanisms and drivers affecting the BoB (Joshi and Warrior, 2022). Since this study is an extension of our previous work, to avoid recapitulation we provide a summarized model configuration information. We encourage readers to refer to our previous works (Joshi et al., 2020, 2021; Joshi and Warrior, 2022) for a comprehensive model information.

The present study couples the Regional Oceanic Modeling System (ROMS) (physical model) (Shchepetkin and McWilliams, 2005, 2009) to the Pelagic Interaction Scheme for Carbon and Ecosystem Studies (PISCES) (biogeochemical model) (Aumont et al., 2003; Aumont and Bopp, 2006). Fig. 1 reveals the study region, which is considered in this paper. The contours show the bathymetry (extracted from ETOPO2 (Smith and Sandwell, 1997)), and the yellow box indicates the domain of analysis. The rivers with high discharge volume are only included in this study (refer to Fig. 1). The climatological river discharge data is included in the coupled model from Dai et al. (2009, 2013). The horizontal resolution is 1/7° with 32 sigma layers as vertical resolution.

Near the surface, refinement is achieved by assigning 12 vertical layers in the top 100 m at the maximum depth position. The K-profile parameterization turbulence scheme (KPP model)
Fig. 1. The contours represent the model bathymetry. The yellow box marks the study region, while the red star shows the Rama buoy location. The broken black line divides the study region into the Northern Bay of Bengal (N-BoB) and the Southern Bay of Bengal (S-BoB). The rivers included in this study have been demonstrated using green dots.

(Large et al., 1994) is adopted in the present study. We choose the physical tracers for initial and lateral forcing from the World Ocean Atlas, 2009 (WOA09) (Antonov et al., 2010; HE et al., 2010). The Comprehensive Ocean-Atmospheric Data Set (COADS) (Worley et al., 2005) is exploited to provide the heat fluxes, temperature, humidity (both relative and specific), and density of air. The E-P and shortwave radiations are also provided from the COADS data set. The monthly climatology of winds and wind stresses are used from the QuikSCAT satellite scatterometer from 1999-2009 (Liu et al., 1998; Risien and Chelton, 2008).

We provide the nutrients and oxygen for the upper ocean from the WOA09 dataset, while for the interior oceans, oxygen and nutrients from the World Ocean Atlas PISCES (WOAPisces) (Aumont et al., 2003) are used. The carbonate parameters (Dissolved Inorganic Carbon (DIC),
Total Alkalinity (TALK), Dissolved Organic Carbon (DOC), etc.) are taken from the WOAPlises dataset. The recent parameterization scheme of Echevin et al. (2008) is used for the PISCES model.

![Fig. 2. The atmospheric $p$CO$_2$ from the Rama buoy for the year 2014 (panel a), 2015 (panel b), and 2016 (panel c). The reconstructed atmospheric $p$CO$_2$ of the year 2015 is provided in panel d. The black broken line shows the mean atmospheric $p$CO$_2$.](image)

For the atmospheric $p$CO$_2$, we analyze the open ocean Rama buoy values (Fig. 2). As the observations are available for the 2014 full year, whereas January-June for 2015 and March-December for 2016, we reconstruct the atmospheric $p$CO$_2$ for 2015 using the available data. The reconstructed 2015 atmospheric $p$CO$_2$ data contains the original values for the January to June period, while the second half of the year is constructed by averaging the observations from 2014 and 2016. Fig. 2d shows that the fluctuations in the atmospheric $p$CO$_2$ are relatively small, and the mean is 379.79 µatm. Since the previous study Joshi et al. (2020) shows that the carbonate parameters are reasonably emulated by the coupled model using 377 µatm (< 1% from the mean atmospheric $p$CO$_2$ of the reconstructed 2015 year), we continue using 377 µatm for the present study.
We run the ROMS model for 30 years before adding the PISCES model. Then both the models coupled together are further run for another 30 years. Finally, we average the last three years to create the climatology of the BoB region.

2.2. pH calculation and sensitivity of pH seasonality to its governing factors

The sea-surface pH can be factorized as the effect of SST, SSS, DIC, and TALK (Takahashi et al., 2009; Valsala and Murtugudde, 2015; Sreeush et al., 2019). The effect of nutrients and other minor ions (like borate, sulfate, and fluoride) on the sea-surface pH can be neglected and treated as residuals (Hagens and Middelburg, 2016). Hence the sea-surface pH in its linearized form can be written as:

$$\frac{dpH}{dt} = \frac{\partial pH}{\partial DIC} \frac{dDIC}{dt} + \frac{\partial pH}{\partial TALK} \frac{dTALK}{dt} + \frac{\partial pH}{\partial SST} \frac{dSST}{dt} + \frac{\partial pH}{\partial SSS} \frac{dSSS}{dt}$$ (1)

Based on the above equation, we construct the sea-surface pH for the BoB region using the CO2SYS (Lewis et al., 1998; Van Heuven et al., 2011) subroutine. The CO2SYS subroutine abides by the guidelines set by Ocean Carbon-cycle Intercomparison Project (OCIMP). The sea-surface pH calculated using CO2SYS is referred to as control pH ($\text{pH}_{\text{CTRL}}$).

We perform four sensitivity experiments to understand the effect of each of the governing factors (SST, SSS, DIC, and TALK) on the seasonal variability of sea-surface pH. The annual climatological mean is provided to the variable of interest, while other factors are allowed to evolve throughout the year. We refer to the sensitivity cases by pH$_X$, where X represents each governing factor. The difference between pH$_{CTRL}$ and the pH$_X$ divulge the effect of the variable or governing factor X.

2.3. Observational data used for model evaluation

We use two observational datasets to evaluate the modeled pH. The description of these datasets are as follows:

2.3.1. Rama Buoy Data

The only buoy data available in the BoB region is located at 15° N, 90° E (as shown in Fig[1]). This buoy measures the ocean surface pH, $pCO_2$, salinity, and temperature. The data from this Rama buoy is available from 24 November 2013 to 20 November 2018. Though the buoy is
reactivated from January 2020, the data is not available for the public. Joshi et al. (2020) provide a table indicating the three deployments of this buoy within the aforementioned period. The method used for calculating the carbonate and physical variables is described meticulously by Sutton et al. (2014). Unfortunately, pH is the least continuous data measured by the Rama buoy.

Fig. 3 shows the available pH data from Rama buoy. The paucity of real time observation is clearly depicted through Fig. 3. January to May is a common period between these three years, the pH data for December is available for 2013 and 2014, whereas the pH data for June is only present in 2014. However, in spite of the limitations recent studies have utilized the RAMA mooring data for validation purpose (Joshi et al., 2020; Sridevi and Sarma, 2021; Chakraborty et al., 2021). To validate our climatology model, we average the available observation data (Fig. 3).

2.3.2. Takahashi Data

Due to the absence of observational data in the BoB region, we choose the monthly pH climatology of Takahashi et al. (2014) to evaluate the model performance. This data will be referred to as the Takahashi Data in this manuscript and has been used in previous studies for
validating modeled pH (Joshi et al., 2020; Sridevi and Sarma, 2021; Chakraborty et al., 2021). The pH monthly climatology is calculated using the observed $pCO_2$ and calculated potential alkalinity (PALK) data. The PALK is calculated using a PALK-salinity relationship, which has shown an inability to capture the effect of the high freshwater influx in the BoB region (Takahashi et al., 2014). Hence the pH climatology from Takahashi data showed a significant deviation from the observed pH of the BoB region. The horizontal resolution of $4^\circ \times 5^\circ$ does not allow the effect of small-scale activities to be adequately captured. Despite the limitations of the Takahashi data, the paucity of observed pH data justifies its use for evaluating the model performance.
2.4. Statistics used to aid in model evaluation

| Statistical Parameters                       | Description                                                                                           |
|---------------------------------------------|--------------------------------------------------------------------------------------------------------|
| Correlation Coefficient (r)                 | Compares the variability between the model and observation data. It ranges from -1 to +1, where the negative value indicates an inverse relationship while the positive values represent a positive relationship. |
| Average Error (AE)                           | These parameters represent the bias between observation and model data.                                |
| Absolute Average Error (AAE)                |                                                                                                        |
| Root Mean Square Error (RMSE)                |                                                                                                        |
| Reliability Index (RI)                       | The mean deviation between the model and observed value is given by RI. A value near zero indicates the model to be reliable (Stow et al., 2009). |
| Cost Function (CF)                           | It is a measure of ”goodness of fit.” It rates the model in the following manner (Dabrowski et al., 2014):     |
|                                            | $\text{CF} < 1 = \text{excellent}; 1 \leq \text{CF} \leq 2 = \text{good}$                             |
|                                            | $2 \leq \text{CF} \leq 3 = \text{average}; \text{CF} > 3 = \text{poor}$                              |
| Percentage Bias (PB)                         | Similar to CF it is a performance indicator which rates the model in the following manner (Dabrowski et al., 2014): |
|                                            | $\text{PB} < 10 = \text{excellent}; 10 \leq \text{PB} \leq 20 = \text{very good}$                   |
|                                            | $20 \leq \text{PB} \leq 30 = \text{good}; \text{PB} > 30 = \text{poor}$                             |
| Model Efficiency Factor (MEF)               | It determines the predictability of the model concerning mean observed values. MEF quantifies the model efficiency in the following manner (Loague and Green, 1991): |
|                                            | $\text{MEF} \geq 0.65 = \text{excellent}; 0.5 \leq \text{MEF} < 0.65 = \text{very good}$             |
|                                            | $0.2 \leq \text{MEF} < 0.5 = \text{good}; \text{MEF} < 0.2 = \text{poor}$                           |

Table 1: Statistical parameters used in this study.
Table 1 provides a list of statistical indices employed in the present study to evaluate the model performance. The formulation of each of these parameters is provided in Appendix A of Joshi et al. (2020); hence it is not repeated here.

3. Results and discussion

3.1. Model Evaluation

The present modeled pH performance is evaluated against the two observational data described in sec 2.3. When comparing the model and observation data, we interpolate the model data to the grid resolution of the observation data using the “nearest-neighbor interpolation” method.

Fig. 4. In panel (a), the model annual mean pH (color contour) is overlaid with the annual mean pH from the Takahashi data (black lines), and the bias (Model - Observation) is shown in panel (b).

Fig. 4 shows the modeled annual mean pH for the BoB overlaid with the observations from Takahashi data. We observe that the model data shows high spatial variability of the sea-surface pH, while the spatial variability in the Takahashi data is low. The high pH value ($\approx 8.08$ units) observed from the model in the northern region indicates the alkaline nature of the ocean. The nature of the southern region is comparatively acidic ($\approx 8.04$ units). The low salinity in the
northern region due to the high discharge from rivers and the precipitation could contribute to the high pH values in the north (Chakraborty et al., 2021). As mentioned in sec 2.3.2, the Takahashi data fails to account for the freshwater, which may be attributed to the absence of high pH values from the Takahashi data. The model tends to have a positive bias in the north (≈ 0.01 units), as shown in Fig 4b. The pH gradually decreases southwards (Fig 4a), this could be due to the high acidic and less discharge from the peninsular rivers. The high saline waters of the Arabian Sea enters the simulation domain decreasing the pH (≈ 8.02 units) in the southwestern region.

We compare the monthly variability of the modeled sea-surface pH for BoB against the Rama buoy data in Fig 5. The paucity of data in the BoB region adheres as a significant hindrance while evaluating the model. Despite the observational data limitations, the model emulates the monthly cycle variability satisfactorily while comparing with buoy data. The model shows an overestimation in May and June, which could be due to the unavailability of recent years data (since the pH is increasing in the central BoB (Sridevi and Sarma, 2021)) and relatively higher vertical mixing in the model (Joshi et al., 2020, 2021). The high peak pH value from model pH data noted in the December month could be due to the maximum thickness of the barrier layer (Joshi et al., 2021) inhibits mixing of acidic sub-surface waters with the sea-surface. The FPS
distributes the freshwater over the sea-surface of the BoB region, which may affect the solubility and dissociation constants (as they are functions of temperature and salinity). The FPS begins in June, peaking over the post-monsoon season. Thus this freshwater spread may have a significant role in determining the seasonal variability of the pH in BoB. To quantify the model performance, we use different statistical indices as mentioned in sec 2.4 (Table 2).

| Model Vs. RAMA buoy data |
|--------------------------|
| AAE | AE | CF | r | RMSE | PB | RI | MEF |
| 0.006 | 0.005 | 0.316 | 0.94 | 0.0063 | 0.056 | 1.0002 | 0.87 |

Table 2: Statistical comparison of model-simulated pH with the RAMA buoy data. A reasonably low bias (0.006) is found when comparing the modeled pH with the Rama buoy data. A high correlation with buoy data indicates that the model well captures the monthly variability. A low reliability index indicate the model to be reliable for modeling the sea-surface pH. The model is rated “excellent” by all the other statics indices. Especially high MEF shows the model can replicate the sea-surface pH. The good performance of the model against the RAMA buoy data gives us the confidence to continue further analysis of the sea-surface pH of the BoB region. Though we agree that the data is less but this approach to validate modeled pH is popularly adopted in many past studies [Joshi et al., 2020; Sridevi and Sarma, 2021; Chakraborty et al., 2021].

Thus this section highlights the need of more pH observation data in the BoB. We also observe that the SSS may modulate the seasonality of sea-surface pH in the BoB. As the model performance seems fair, we attempt to evaluate and analyze various factors driving the sea-surface pH in the BoB region using this model.

3.2. Effect of individual drivers on the seasonality of pH

Fig.6 presents the effect of individual drivers, viz, SST, DIC, TALK, and SSS on the domain (yellow box, shown in Fig.1) averaged pH. Each panel in Fig.6 shows the effect of these individual drivers on pH, respectively. The pH\textsubscript{CTRL} is shown with a solid black line in each of the Fig.6 panels. The seasonality in pH\textsubscript{CTRL} is shown with a solid black line in each of the Fig.6 panels. The seasonality in pH\textsubscript{CTRL} seems to be driven by 6 and 12 months signal. We observe a maximum peak of pH\textsubscript{CTRL} in October (8.0942) and a minimum crest in May (8.04). It implies...
that the sea-surface of BoB is most acidic in May and most alkaline in October. The annual mean pH of the BoB is 8.0620.

In Fig. 6a, we show the effect of DIC on the pH by replacing it with the annual mean (as described in sec 2.2). The pHDIC is significantly higher than the pHCTRL during the period between December and February. This high value indicates that the role of DIC is to make the BoB acidic. From March-May, the pHDIC is lower than pHCTRL, suggesting that the DIC tends to make the surface of BoB alkaline. Interestingly, from June to November, the effect of DIC is negligible; in fact, the primary variation to the ocean acidification of BoB by DIC is found in the first half of the year. The second half of the year shows an almost negligible effect of DIC on pHCTRL.

Fig. 6b exhibits the effect of temperature (Temp) on the pH. We replace the monthly variation of Temp by annual mean and reproduce pH_SST. From November to February the pH_SST is lower than pH_CTRL, which implies that during this period, the effect of SST is to reduce the acidic nature of BoB. It should be noted that SST tends to drop from November to February (winter months); this may be the reason that during this period, the effect of SST is to make the sea-surface water alkaline. From March to October, the pH_SST is observed to be higher than pH_CTRL:
hence it reveals that the effect of SST is to make the sea-surface acidic in this period. The higher SST during these months may enhance the acidic nature of the BoB.

The BoB region experiences high freshwater influx, and the effect of salinity (SSS) is revealed in Fig.6c. It is clear that for the BoB region, the Sal plays a significant role in modulating the sea-surface pH. From January to August, the pH$_{SSS}$ is higher than pH$_{CTRL}$, which suggests that the role of Sal is to increase the acidic levels during this period. Whereas from August-December, the lower values of pH$_{SSS}$ with respect to pH$_{CTRL}$ reveal that Sal makes BoB alkaline during this period. These results suggest that stratification and the spreading of freshwater plume could possibly heavily modulate the values and dynamics of the sea-surface pH.

Fig.6d presents the effect of TALK on the sea-surface pH. We observe that the TALK effect is only significant in February and March. The effect of TALK is almost negligible for most of the climatological year. So it is evident from Fig.6 that in the BoB region, DIC, SST, and SSS are the major drivers of the seasonality of pH, while TALK has a negligible effect. Further, Fig.7 summarizes the results of Fig.6 to quantify the effect of each driver.

Fig.7 reveals a summary of the effect of each driver on the sea-surface pH of the BoB. Unlike the studies related to the Arabian Sea (Sreeush et al., 2019) and the observational studies from the Pacific ocean, Mediterranean Sea, and North-Atlantic (Hagens and Middelburg, 2016), the BoB pH is significantly affected by SSS, SST and DIC. Whereas SST and DIC were the major drivers of pH in the Arabian Sea, Pacific Ocean, North Atlantic Ocean, and Mediterranean Sea (Hagens and Middelburg, 2016; Sreeush et al., 2019). From Fig.7, we observe that except for February to March period, the effect of TALK is negligible. In February to March period, the effect of DIC and SST is balanced by SSS and TALK. During December and January, the effect of SST is compensated primarily by DIC, whereas both TALK and SSS have negligible effect. The period of August to November is strictly dominated by SSS, making the sea-surface alkaline. The peak in October coincides with the maximum freshwater plume spread (Jana et al., 2015, 2018; Joshi et al., 2021), which indicates that the freshwater which lowers the SSS increases the pH of the BoB region. The acidic peak in May can be attributed to the highest SSS in the BoB region and lowest freshwater plume spread (Jana et al., 2015, 2018; Joshi et al., 2021). This domination of SSS in controlling pH in the BoB region makes this region unique from the rest of the world ocean.

In Fig.8, we demonstrate the seasonal effect of each of the governing factors on the sea-
surface pH of the BoB region. The climatological year is divided based on the monsoon season. The December to February (DJF) is the winter monsoon season, the June to September (JJAS) is the southwest monsoon season, while the phases March to May (MAM) and October-November (ON) are pre-monsoon and post-monsoon seasons, respectively. In the winter monsoon season (DJF), the SST tends to increase the pH, while the DIC averse the rise in pH, but the minor effect of TALK further increments the pH. The effect of SSS is almost zero in the winter monsoon season. The pre-monsoon season (MAM) experiences the combined effect of all the drivers. The DIC tends to increase the pH during MAM, but the combined effect of SSS, SST, and TALK reduces the pH. Thus the pre-monsoon season is the most acidic season among all other seasons.

In the southwest monsoon season (JJAS), pH is dominated by SST. The SST tends to reduce the pH in the southwest monsoon season. The effect of SSS, DIC, and TALK is minimal during
the southwest monsoon season. The SSS dominates the rise in pH during the post-monsoon season. This may be attributed to the freshwater plume spread during this period [Jana et al., 2015, 2018; Joshi et al., 2021]. The DIC effect is countered by the TALK in the post-monsoon season, while the SST has a negligible effect on pH during this season.

Hence, our analysis shows that SSS is a major driver of pH along with SST and DIC in the BoB region. The role of SSS in modulating the pH may be due to the high freshwater flux, strong stratification, and thick barrier layer, inhibiting vertical mixing. The seasonality in SSS and SST also affects the sea-surface pH of BoB. As SSS has a significant role in modulating the seasonality of pH in the BoB region, further analysis on the effect of stratification and barrier layer over the pH becomes highly important.
3.3. Role of stratification on seasonality of pH

Due to the low-density freshwater influx from rivers and precipitation, a shallow layer of freshwater forms in the sub-surface waters, leading to the strong stratification in the BoB region (Shetye et al., 1996; Vinayachandran et al., 2002; Rao and Sivakumar, 2003). The strong stratification leads to a barrier layer formation. Shetye (1993); Shetye et al. (1996) reported observing a low saline strip of freshwater along the western coast in July-August 1989 and December 1991, respectively. The formation and spreading of the freshwater plume in the summer monsoon are meticulously described through observation (Vinayachandran and Kurian, 2007; Jana et al., 2015, 2018; Sandeep and Pant, 2019) explain the role of winds, circulation, and rivers from a modeling perspective, in the formation and spreading of the freshwater plume. The freshwater spread over the surface of BoB, and the barrier layer thickness (BLT) thus formed restricts vertical mixing. This inhibition of vertical mixing does not allow the sub-surface nutrients and chemicals from reaching the surface, which makes the BoB region a low-productive zone (Prasanna Kumar et al., 2002; Gauns et al., 2005). Our previous work (Joshi et al., 2021) shows the extent by which BLT and the freshwater plume influences the sea-surface $pCO_2$. The method to calculate the BLT is same for this study, hence we encourage readers to refer to our previous work for detailed explanation of BLT calculation and its influence on the sea-surface $pCO_2$.

To examine the relationship between the BLT and the sea-surface pH in the BoB region, we make a spatial correlation between the BLT and pH, as shown in Fig.9. The correlation is used as a proxy to highlight the relation between BLT and pH. As we observe from Fig.9, the open oceans of the BoB region shows a high positive correlation between pH and BLT, suggesting that the thicker the barrier layer higher the sea-surface pH. This may suggest that the thick barrier layer impedes the mixing of sub-surface acidic waters with the surface waters of BoB, resulting in higher pH at the sea-surface. The high positive correlation also suggests that the months or seasons having low BLT would result in acidic sea-surface waters.

In Fig.10, we emulate the spatial seasonal pH overlaid with the seasonal BLT. We observe that indeed the areas having thicker barrier layer seem to have higher pH values. The barrier layer is known to be thickest during the winter monsoon period (DJF), and the area having thicker barrier layers is spread the most during this season. Hence the higher pH value area is seen to be more widely spread during the winter monsoon season. The pre-monsoon season (MAM) is observed to have the shallowest barrier layer, and hence the interaction between sub-surface
Fig. 9. Spatial correlation between BLT and sea-surface pH.

and surface waters must be predominant. Thus the pre-monsoon season seems to be the season having the most acidic sea-surface. In Fig.8 we see that the pH in the pre-monsoon season has a significant contribution from all the governing factors, the interaction between sub-surface and surface waters may contribute to the equal roles of all drivers.

The southwest monsoon season (JJAS) in Fig.10 exhibits a strip of low acidic waters slanting from the north BoB along the western coast. The beginning of freshwater plume spread in the southwest monsoon season seems to draw the low-saline waters through the west coast (Shetye, 1993), which increases the pH in this region. The post-monsoon season (ON) marks the largest freshwater plume spread and the lowest SSS, resulting in the highest pH, especially in the northern region. It is observed from Fig.10 that the northern region of BoB (as shown in Fig.1) has higher pH values except for the pre-monsoon season. The freshwater influx from high discharge rivers like Ganga, Brahmaputra, and Irrawaddy, along with the high precipitation, maybe why the high pH and high BLT values in the north. The southern sea-surface is comparatively acidic
in nature. Especially the southwestern region seems to be relatively acidic throughout the climatological year. To further visualize the effect of stratification, we plot the depth-wise annual mean pH (up to 100m) (Fig. 11) for the N-BoB and S-BoB demarcated by the broken black line in Fig. 1.

We observe in Fig. 11 that the pH in the N-BoB is lower than the S-BoB below 20m depth. On the surface, the N-BoB has a pH higher than the S-BoB. We know that the N-BoB is more influenced by the freshwater influx due to the BLT and consequently stronger stratification; hence
the acidic lower pH waters are restricted at around 20 to 22 m depth. The pH in N-BoB is constant till 10 m depth, whereas the S-BoB pH is constant almost till 20 m depth, which indicates that the inhibition of vertical mixing in N-BoB is lower than the S-BoB. Hence the effect of stratification in the N-BoB plays a vital role in making the N-BoB sea-surface alkaline compared to the S-BoB.

4. Conclusion

In the present study, we attempt to explore the climatological ocean acidification state of the BoB region. We use the outputs of a coupled physical-biogeochemical model to reconstruct the pH for the BoB region. The paucity of data from the chemical parameters has been a challenge to establish the model worldwide. The high freshwater influx in the BoB region makes this region...
even more challenging as mathematical relations of chemical parameters based on SSS do not stand valid in this region. Nevertheless, considering these challenges, we try to validate our modeled pH with the limited data available. The modeled pH performs reasonably well when compared with RAMA buoy and Takahashi data. The correlation of 0.94 with RAMA buoy implies that the model well captures the monthly variability. The biases have been reasonably low when comparing with both the observational data. The comparison also highlights the necessity of more real-time chemical parameter observations in the BoB region.

Further, we analyze the role of each governing factor, viz, SST, SSS, DIC, and TALK, in modulating the seasonality of pH. The SSS has emerged as one of the major drivers of pH along with SST and DIC. The first six months of the climatological year have a combined effect of SST, DIC, and SSS on pH, but SSS significantly dominates the last six months. The SST reduces the sea-surface acidity in the winter monsoon season and post-monsoon season, but it increases the acidity in pre-monsoon and southwest monsoon seasons. The winter monsoons experience the maximum effect of SST on pH, and the post-monsoon season has a minimum SST effect on pH. SSS increases the pH significantly in the post-monsoon season. The SSS reduces the pH in the pre-monsoon season. The effect of SSS is minor during the southwest monsoon and winter monsoon seasons. DIC increases acidity in the sea-surface during the winter monsoon season but reduces acidity during the pre-monsoon season. The effect of TALK is lowest among all the governing factors.

As SSS is a major contributor in modulating the seasonality of pH, the role of stratification on sea-surface ocean acidification becomes crucial in the BoB region. The sea-surface pH shows a strong positive correlation with the BLT, which indicates that thicker barrier layer areas have higher sea-surface pH. The N-BoB is found to be more alkaline than the S-BoB. The spreading of the freshwater plume that reduces the SSS may be why the higher pH values in the N-BoB. Below 20 m depth, the N-BoB has more acidic waters than the S-BoB. The stronger stratification and thick barrier layer inhibit these high acidic sub-surface waters from reaching the surface resulting in a more alkaline sea-surface in N-BoB than the S-BoB.

Our study is one of the first climatological modeling approaches for the pH in the BoB region. The study becomes more important as it provides a significant idea of the governing factors of pH in the open ocean part of BoB. With global warming, these roles may change in the future. The relationship between BLT and sea-surface pH gives a different perspective which may be used to
artificially (using a neural network approach) generate pH data. However, interannual modeling has to be done, as cyclones and higher local winds may change the role of stratification on pH. In the next part of this study, we will evaluate the effect of other mechanisms like biological production, solubility, and carbon dioxide flux on the sea-surface pH.

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References

Antonov, J., Seidov, D., Boyer, T., Locarnini, R., Mishonov, A., Garcia, H., Baranova, O., Zweng, M., Johnson, D., 2010. World ocean atlas 2009, volume 2: Salinity, noaa nesdis 68. Washington DC , 39.
Aumont, O., Bopp, L., 2006. Globalizing results from ocean in situ iron fertilization studies. Global Biogeochemical Cycles 20.
Aumont, O., Maier-Reimer, E., Blain, S., Monfray, P., 2003. An ecosystem model of the global ocean including fe, si, p colimitations. Global Biogeochemical Cycles 17.
Chakraborty, K., Valsala, V., Bhattacharya, T., Ghosh, J., 2021. Seasonal cycle of surface ocean pco2 and ph in the northern indian ocean and their controlling factors. Progress in Oceanography 198, 102683.
Dabrowski, T., Lyons, K., Berry, A., Cusack, C., Nolan, G.D., 2014. An operational biogeochemical model of the north-east atlantic: model description and skill assessment. Journal of Marine Systems 129, 350–367.
Dai, A., Qian, T., Trenberth, K.E., Milliman, J.D., 2009. Changes in continental freshwater discharge from 1948 to 2004. Journal of climate 22, 2773–2792.
Dai, M., Cao, Z., Guo, X., Zhai, W., Liu, Z., Yin, Z., Xu, Y., Gan, J., Hu, J., Du, C., 2013. Why are some marginal seas sources of atmospheric co2? Geophysical Research Letters 40, 2154–2158.
Echevin, V., Aumont, O., Ledesma, J., Flores, G., 2008. The seasonal cycle of surface chlorophyll in the peruvian upwelling system: A modelling study. Progress in Oceanography 79, 167–176.
Fairall, C.W., Bradley, E.F., Rogers, D.P., Edson, J.B., Young, G.S., 1996. Bulk parameterization of air-sea fluxes for tropical ocean-global atmosphere coupled-ocean atmosphere response experiment. Journal of Geophysical Research: Oceans 101, 3747–3764.
Feely, R.A., Doney, S.C., Cooley, S.R., 2009. Ocean acidification: Present conditions and future changes in a high-co2 world. Oceanography 22, 36–47.

Friedlingstein, P., O’Sullivan, M., Jones, M.W., Andrew, R.M., Hauck, J., Olsen, A., Peters, G.P., Peters, W., Pongratz, J., Sitch, S., et al., 2020. Global carbon budget 2020. Earth System Science Data 12, 3269–3340.

Gattuso, J.P., Hansson, L., Gazeau, F., 2014. Ocean acidification and its consequences. Ocean in the Earth System, 189–253.

Gauns, M., Madhupratap, M., Ramaiah, N., Jyothibabu, R., Fernandes, V., Paul, J.T., Kumar, S.P., 2005. Comparative accounts of biological productivity characteristics and estimates of carbon fluxes in the arabian sea and the bay of bengal. Deep Sea Research Part II: Topical Studies in Oceanography 52, 2003–2017.

Hagens, M., Middelburg, J., 2016. Attributing seasonal ph variability in surface ocean waters to governing factors. Geophysical Research Letters 43, 12–528.

HE, L., Zweng, M., Johnson, D., 2010. World ocean atlas 2009, volume 1: Temperature. NOAA Atlas NESDIS 68.

Jana, S., Gangopadhyay, A., Chakraborty, A., 2015. Impact of seasonal river input on the bay of bengal simulation. Continental Shelf Research 104, 45–62.

Jana, S., Gangopadhyay, A., Lermusiaux, P.F., Chakraborty, A., Sil, S., Haley Jr, P.J., 2018. Sensitivity of the bay of bengal upper ocean to different winds and river input conditions. Journal of Marine Systems 187, 206–222.

Joshi, A., Chowdhury, R.R., Warrior, H., Kumar, V., 2021. Influence of the freshwater plume dynamics and the barrier layer thickness on the co2 source and sink characteristics of the bay of bengal. Marine Chemistry 236, 104030.

Joshi, A., Roychowdhury, R., Kumar, V., Warrior, H., 2020. Configuration and skill assessment of the coupled biogeochemical model for the carbonate system in the bay of bengal. Marine Chemistry, 103871.

Kwiatkowski, L., Orr, J.C., 2018. Diverging seasonal extremes for ocean acidification during the twenty-first century. Nature Climate Change 8, 141–145.

Large, W.G., McWilliams, J.C., Doney, S.C., 1994. Oceanic vertical mixing: A review and a model with a nonlocal boundary layer parameterization. Reviews of Geophysics 32, 363–403.

Lewis, E., Wallace, D., Allison, L.J., 1998. Program developed for CO (sub 2) system calculations. Technical Report. Brookhaven National Lab., Dept. of Applied Science, Upton, NY (United States . . . . .

Liu, W.T., Tang, W., Polito, P.S., 1998. Nasa scatterometer provides global ocean-surface wind fields with more structures than numerical weather prediction. Geophysical Research Letters 25, 761–764.

Loague, K., Green, R.E., 1991. Statistical and graphical methods for evaluating solute transport models: overview and application. Journal of contaminant hydrology 7, 51–73.

Mukhopadhyay, S., Biswas, H., De, T., Sen, S., Jana, T., 2002. Seasonal effects on the air–water carbon dioxide exchange in the hooghly estuary, ne coast of bay of bengal, india. Journal of Environmental Monitoring 4, 549–552.

Marty, V., Suryanarayana, A., Rao, D., 1993. Current structure and volume transport across 12 n in the bay of bengal. Indian Journal of Marine Sciences 22, 12–16.

Prasanna Kumar, S., Muraleedharan, P., Prasad, T., Gauns, M., Ramaiah, N., De Souza, S., Sardesai, S., Madhupratap, M., 2002. Why is the bay of bengal less productive during summer monsoon compared to the arabian sea? Geophysical Research Letters 29, 88–1.
Provoost, P., Heuven, S.v., Soetaert, K., Laane, R., Middelburg, J., 2010. Seasonal and long-term changes in pH in the Dutch coastal zone. Biogeosciences 7, 3869–3878.

Rao, R., Sivakumar, R., 2003. Seasonal variability of sea surface salinity and salt budget of the mixed layer of the north indian ocean. Journal of Geophysical Research: Oceans 108, 9–1 – 9–14.

Risien, C.M., Chelton, D.B., 2008. A global climatology of surface wind and wind stress fields from eight years of quikscat scatterometer data. Journal of Physical Oceanography 38, 2379–2413.

Sabine, C.L., Feely, R.A., Gruber, N., Key, R.M., Lee, K., Bullister, J.L., Wanninkhof, R., Wong, C., Wallace, D.W., Tilbrook, B., et al., 2004. The oceanic sink for anthropogenic CO2. Science 305, 367–371.

Sandep, K., Pant, V., 2019. Riverine freshwater plume variability in the Bay of Bengal using wind sensitivity experiments. Deep Sea Research Part II: Topical Studies in Oceanography 168, 104649.

Sarma, V., Krishna, M., Paul, Y., Murti, V., 2015. Observed changes in ocean acidity and carbon dioxide exchange in the coastal bay of Bengal—a link to air pollution. Tellus B: Chemical and Physical Meteorology 67, 24638.

Sarma, V., Krishna, M., Rao, V., Viswanadham, R., Kumar, N., Kumari, T., Gawade, L., Ghatkar, S., Tari, A., 2012. Sources and sinks of CO2 in the west coast of bay of Bengal. Tellus B: Chemical and Physical Meteorology 64, 10961.

Sarma, V., Kumar, G.S., Yadav, K., Dalabehera, H., Rao, D., Behera, S., Loganathan, J., 2019. Impact of eddies on dissolved inorganic carbon components in the Bay of Bengal. Deep Sea Research Part I: Oceanographic Research Papers 147, 111–120.

Sarma, V., Kumar, V., Srinivas, T., Krishna, M., Ganapathi, P., Murti, V., 2018. East India coastal current controls the dissolved inorganic carbon in the coastal bay of Bengal. Marine Chemistry 205, 37–47.

Shchepetkin, A.F., McWilliams, J.C., 2005. The regional oceanic modeling system (ROMS): a split-explicit, free-surface, topography-following-coordinate oceanic model. Ocean Modelling 9, 347–404.

Shchepetkin, A.F., McWilliams, J.C., 2009. Correction and commentary for “ocean forecasting in terrain-following coordinates: Formulation and skill assessment of the regional ocean modeling system” by haidvogel et al., j. comp. phys. 227, pp. 3595–3624. Journal of Computational Physics 228, 8985–9000.

Shetye, S., Gouveia, A., Shankar, D., Shenoi, S., Vinayachandran, P., Sundar, D., Michael, G., Namoothiri, G., 1996. Hydrography and circulation in the western Bay of Bengal during the northeast monsoon. Journal of Geophysical Research: Oceans 101, 14011–14025.

Shetye, S.R., 1993. The movement and implications of the ganges—branhaputra runoff on entering the. Current Science 64.

Smith, W.H., Sandwell, D.T., 1997. Global sea floor topography from satellite altimetry and ship depth soundings. Science 277, 1956–1962.

Sreeush, M.G., Rajendran, S., Valsala, V., Pentakota, S., Prasad, K., Murugudde, R., 2019. Variability, trend and controlling factors of ocean acidification over western Arabian sea upwelling region. Marine Chemistry 209, 14–24.

Sridevi, B., Sarma, V., 2021. Role of river discharge and warming on ocean acidification and pCO2 levels in the Bay of Bengal. Tellus B: Chemical and Physical Meteorology 73, 1–20.

Stow, C.A., Joliffe, J., McGillicuddy Jr, D.J., Doney, S.C., Allen, J.L., Friedrichs, M.A., Rose, K.A., Wallhead, P., 2009. Skill assessment for coupled biological/physical models of marine systems. Journal of Marine Systems 76, 4–15.

Sutton, A.J., Sabine, C.L., Maenner-Jones, S., Lawrence-Slavas, N., Meinig, C., Feely, R., Mathis, J., Musielewicz, S., Bott, R., McLain, P., et al., 2014. A high-frequency atmospheric and seawater pCO2 data set from 14 open-ocean sites.
using a moored autonomous system. Earth System Science Data 6, 353–366.
Takahashi, T., Sutherland, S.C., Chipman, D.W., Goddard, J.G., Ho, C., Newberger, T., Sweeney, C., Munro, D., 2014.
Climatological distributions of ph, pco2, total co2, alkalinity, and caco3 saturation in the global surface ocean, and
temporal changes at selected locations. Marine Chemistry 164, 95–125.
Takahashi, T., Sutherland, S.C., Wanninkhof, R., Sweeney, C., Feely, R.A., Chipman, D.W., Hales, B., Friederich, G.,
Chavez, F., Sabine, C., et al., 2009. Climatological mean and decadal change in surface ocean pco2, and net sea-air
co2 flux over the global oceans. Deep Sea Research Part II: Topical Studies in Oceanography 56, 554–577.
Unesco, 1969. Discharge of selected rivers of the world. Unesco.
Valsala, V., Murtugudde, R., 2015. Mesoscale and intraseasonal air–sea co2 exchanges in the western arabian sea during
boreal summer. Deep Sea Research Part I: Oceanographic Research Papers 103, 101–113.
Van Heuven, S., Pierrot, D., Rae, J., Lewis, E., Wallace, D., 2011. Matlab program developed for co2 system calculations.
ORNL/CDIAC-105b 530.
Vinayachandran, P., Kurian, J., 2007. Hydrographic observations and model simulation of the bay of bengal freshwater
plume. Deep Sea Research Part I: Oceanographic Research Papers 54, 471–486.
Vinayachandran, P., Murtu, V., Ramesh Babu, V., 2002. Observations of barrier layer formation in the bay of bengal
during summer monsoon. Journal of Geophysical Research: Oceans 107, SRF 19–1 – SRF 19–9.
Wootton, J.T., Pfister, C.A., 2012. Carbon system measurements and potential climatic drivers at a site of rapidly
declining ocean ph. PloS one 7, e53396.
Worley, S.J., Woodruff, S.D., Reynolds, R.W., Lubker, S.J., Lott, N., 2005. Icoads release 2.1 data and products. Inter-
national Journal of Climatology: A Journal of the Royal Meteorological Society 25, 823–842.