Marine Pollution Bulletin 135 (2018) 540–550

Contents lists available at ScienceDirect

Marine Pollution Bulletin

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

Aragonite saturation state variation and control in the river-dominated marginal BoHai and Yellow seas of China during summer

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ARTICLE INFO

Keywords:
Aragonite saturation state
Marginal sea
Large rivers input
BoHai Sea
Yellow Sea

ABSTRACT

Based on a survey conducted from June to July 2013, aragonite saturation state variation and control in the river-dominated marginal BoHai and Yellow seas were investigated. Surface water Ω_{arag} ranged from 2.0–3.8, whereas subsurface water Ω_{arag} was generally lower than 2.0. Temperature changes had a strong influence on Ω_{arag} through induced CO_2 solubility changes in seawater. Riverine freshwater input decreased Ω_{arag} in the Changjiang and Yalu river estuaries, but induced higher Ω_{arag} in the Yellow River estuary. Biological processes had opposite effects on Ω_{arag}, whereby elevated biological production led to the highest Ω_{arag} in the South Yellow Sea surface water, whereas net community respiration/remineralization induced low Ω_{arag} in subsurface water. Stratification affected the level and scale of low Ω_{arag} in subsurface water. By the year 2100, surface water with $\Omega_{arag} > 2.0$ will disappear except for the Yellow River estuary, and most of the subsurface water will develop substantial aragonite undersaturation.

1. Introduction

As a sink of atmospheric CO_2, the ocean has taken up > 118 billion tons of carbon since 1800, causing changes in ocean water chemistry (Caldeira and Wickett, 2005; Doney et al., 2009) and progressively lowering levels of seawater pH and the carbonate saturation state of aragonite (Ω_{arag}), a phenomenon known as ocean acidification (Feely et al., 2004; Orr et al., 2005; Doney et al., 2009; IPCC, 2014). The carbonate saturation state is essential for many marine organisms for the formation of calcium carbonate ($CaCO_3$) skeletons and shells, and thus Ω_{arag} is a critical parameter for characterizing the influence of acidification on marine organisms (Riebesell, 2004; Waldbusser et al., 2014). It can be defined as:

$$\Omega_{arag} = [CO_3{}^{2-}] \times [Ca^{2+}]/K_{sp} \quad (1)$$

where $[CO_3{}^{2-}]$ and $[Ca^{2+}]$ are the carbonate and calcium concentrations, respectively, and K_{sp} is the apparent solubility product for aragonite. When $\Omega_{arag} = 1$, seawater is in equilibrium with the mineral; when $\Omega_{arag} > 1$, supersaturation favors precipitation and $CaCO_3$ is stable in seawater; and when $\Omega_{arag} < 1$, undersaturation favors dissolution and aragonite is unstable. Currently, mean global surface ocean Ω_{arag} is about 3.0 (Jiang et al., 2015), which has declined by ~16% with increasing atmospheric CO_2 since the preindustrial period and is expected to decrease by a further 50% by the end of this century (Feely et al., 2004 and 2008; Orr et al., 2005).

Increasing attention has been paid to coastal ocean acidification in recent years. In contrast to the open ocean, research has shown that Ω_{arag} in coastal regions demonstrates variability and complexity, and is impacted by multiple biogeochemical processes in addition to increasing atmospheric CO_2, such as upwelling of CO_2-enriched waters (Feely et al., 2008), river discharge (Salisbury et al., 2008; Chou et al., 2013), and metabolic processes led by eutrophication (Cai et al., 2011; Zhai et al., 2014). Of significance, most coastal oceans are also productive marine ecosystems that sustain numerous commercially valuable resources, such as seafood (Gattuso et al., 1998), which are vulnerable to ocean acidification (Ekstrom et al., 2015; Mathis et al., 2015). Recent studies have shown that coastal ecosystems are subject to strong influences of multiple environmental stressors that may act synergistically to exacerbate acidification (Doney, 2010). Therefore, it is critical to gain a better understanding of the Ω_{arag} baseline and how

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https://doi.org/10.1016/j.marpolbul.2018.07.032
Received 19 December 2017; Received in revised form 26 June 2018; Accepted 11 July 2018
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complicated environmental stressors affect it in coastal oceans, particularly in productive marine ecosystems that sustain commercially valuable fisheries.

The BoHai Sea (BHS) and the Yellow Sea (YS) are semi-enclosed shallow seas of the northwest Pacific characterized by large river freshwater input and considerable anthropogenic impact. These areas form a major marine zone in China, and the sensitivity of marine aquaculture industry to ocean acidification is increasing with fast-developing aquacultural activities. Low Ω_{arag} values have been reported in the North Yellow Sea (NYS) subsurface waters in autumn, indicating that this area might represent a system highly vulnerable to the potential negative effects of ocean acidification in Chinese seas (Zhai et al., 2014). Besides net community respiration/remineralization, dilution from the Yalu River contributes to the low Ω_{arag} values detected in the nearshore area of the North Yellow Sea during flood periods (Zhai et al., 2015). Low Ω_{arag} values have also been observed in the South Yellow Sea (SYS) (Xu et al., 2016a), with riverine water discharge from the Changjiang River having important impact on Ω_{arag} distribution in this area. Despite these potentially important consequences, there is only limited understanding of sea water Ω_{arag} and its mechanisms at present, which is important given that excess nutrients have induced serious eutrophication and algal blooms in the BoHai Sea and the Yellow Sea in recent years (State Oceanic Administration of China, 2017).

Based on a field survey conducted from June to July 2013, we investigated the distribution of the carbonate system and ancillary parameters in river-dominated ocean margins of the BoHai Sea and the Yellow Sea, which represent coastal systems with different riverine freshwater inputs and vulnerability to the potential negative effects of ocean acidification. We also investigated the role of temperature change, riverine freshwater input, biological activity, and stratification on regulating Ω_{arag}. Finally, future Ω_{arag} level scenarios in this important marine aquaculture region were explored in the context of increasing atmospheric CO_2 and eutrophication by the year 2100.

2. Materials and methods

2.1. Study area

The study area included the BoHai Sea and the Yellow Sea, both of which form a semi-enclosed shallow marginal sea in the western North Pacific (Fig. 1). The area has a temperate climate and wind is dominated by the East Asian monsoon, with the rain-bearing southwest monsoon dominating in summer and the strong northeast monsoon prevailing in winter (Su, 1998; Su and Yuan, 2005).

With an area of 7.7×10^4 km^2 and mean depth of 18 m, the BoHai Sea is characterized by low salinity and limited water exchange with the Yellow Sea through the narrow BoHai Strait (Fig. 1). Water stratification appears in spring and decreases in autumn (Huang et al., 1999) with seasonal solar insolation and monsoon wind-induced mixing. The Yellow River, with a water discharge of 5.6×10^{10} m^3 yr^{-1} (Hu et al., 1998; Qiao et al., 2010), drains directly into the BoHai Sea, resulting in high dissolved inorganic carbon (DIC) and total alkalinity (TAlk) (Liu et al., 2014). The Yellow Sea is traditionally divided into the North Yellow Sea and the South Yellow Sea. With an area of 7.13×10^4 km^2 and average depth of 38 m, the North Yellow Sea is connected to the BoHai Sea through the BoHai Strait, but is more open to the South Yellow Sea. It receives freshwater from the Yalu River, with a discharge of 3.3×10^{10} m^3 yr^{-1} (Liu and Liu, 1992; Zhang, 1996; Zhang et al., 1997). The South Yellow Sea has an area of 3.09×10^5 km^2 and an average depth of 50 m (Fig. 1), and is fed by freshwater discharge (9.6×10^{11} m^3 yr^{-1}) from the Changjiang River (Mao et al., 1963; Riedlinger and Preller, 1995), which flows through densely populated areas with intensive agricultural and industrial activities. The whole Yellow Sea exhibits pronounced stratification in deep layers during the summer. Furthermore, the area exhibits a cold pool, known as the

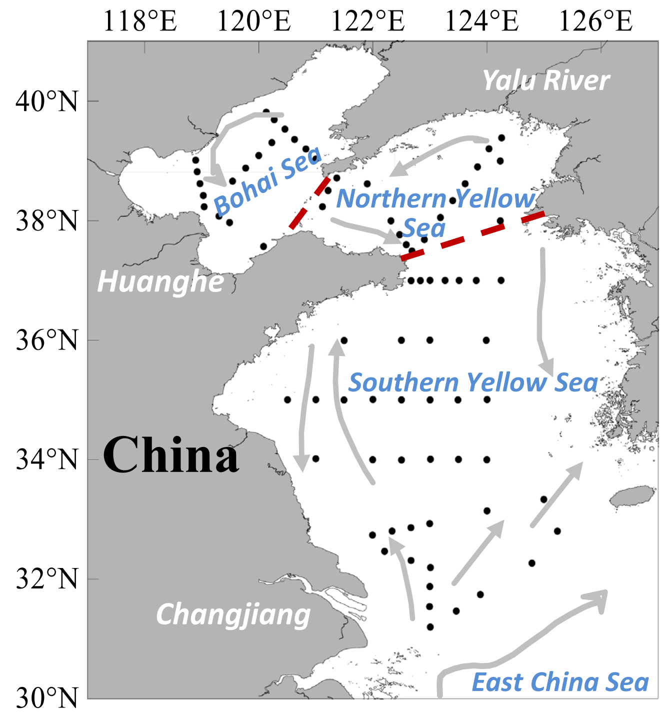

Fig. 1. Study area and sampling stations in the BoHai Sea and the Yellow Sea in summer 2013. Dashed red lines are the traditional boundaries between the Bohai Sea, the North Yellow Sea and the South Yellow Sea. Gray arrows denote major currents in the coastal seas during the southwest monsoon seasons (Zang et al., 2003; Chen, 2009). Black dots denote stations investigated in summer. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Yellow Sea Cold Water Mass (YSCWM), which features low temperatures of 5–11 °C and salinity of 31–33 overlaid by 20 to 25 m of warm water (Miao et al., 1991; Qiao et al., 1998; Chen, 2009). In the summer flood season, freshwater discharge, as well as the YSCWM and other factors, markedly affect the Yellow Sea coastal ecosystem, resulting in complicated variations in the environmental settings between the North Yellow Sea and the South Yellow Sea.

Surrounded by fast-developing economic zones, the whole study area plays an important role in the development of the marine aquaculture industry in the region. For example, the gross marine industrial output of the BoHai Sea accounts for 37% of the total Chinese marine industrial output (State Oceanic Administration of China, 2014). Nonetheless, during the past few years, due to rapid industrialization and increased agricultural production associated with population growth, worsening eutrophication has led to considerable ecological consequences, such as increased algal blooms (Lin et al., 2008; State Oceanic Administration of China, 2017) and altered biogeochemical cycles (Zhai et al., 2012, 2014; Zhao et al., 2017). Thus, changes in the hydrological and biogeophysical properties of the study area are regulated by both oceanic dynamics and human activities (Zhai et al., 2012, 2014; Xu et al., 2016a).

2.2. Sampling and analytical methods

The field survey was carried out aboard the *R/V Dong Fang Hong II* from 23 June to 9 July 2013, and encompassed the summer flood season. The scope of the study area (31–40°N to 118–126°E) and the 80 sampling stations are shown in Fig. 1. During the survey, field measurements of temperature and salinity in the water column were recorded using a calibrated Conductivity-Temperature-Depth/Pressure unit (CTD) recorder (SBE19+, Sea-Bird Co., USA). At each discrete station, seawater samples were collected at three to four different

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