

Hypoxia and nutrient dynamics affected by marine aquaculture in a monsoon-regulated tropical coastal lagoon

Jing Zhang • ZhuoYi Zhu • Wen Yuan Mo • Su Mei Liu • Dao Ru Wang • Guo Sen Zhang

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Abstract The Laoyehai (lagoon) is located at the east coast of Hainan Island in the South China Sea and has been subject to perturbations from human activities, notably marine aquaculture, and has eutrophic surface and hypoxic near-bottom waters. A lack of knowledge of hydrodynamic and biogeochemical processes is a challenge to the sustainable management of lagoon at the ecosystem level in science. Five field campaigns, including three during the southwest monsoon and two in the northeast monsoon periods, were carried out at the Laoyehai in 2008–2011. The aim of this study is to investigate the impacts of dynamic processes of hydrography and human activities on nutrient geochemistry and their relationships to the system eutrophication and hypoxia in the lagoon. In this coastal system, high levels of ammonium relative to nitrate are found, elevated phosphate skews the DIN/DIP relative to the Redfield ratio, and the dissolved silicate concentration

J. Zhang (⊠) · Z. Zhu · G. S. Zhang State Key Laboratory of Estuarine and Coastal Research, East China Normal University, 3663 Zhongshan Road North, Shanghai 200062, China e-mail: jzhang@sklec.ecnu.edu.cn

W. Y. Mo · D. R. Wang

Hainan Provincial Marine Development Plan and Design Research Institute, 15 Longkun Road North, Haikou 570203, China

S. M. Liu

is high because of submarine groundwater discharge. The organic fraction in the Laoyehai accounts for a large proportion of the total nutrients associated with the release of wastes from marine aquaculture. The hypoxia of near-bottom waters in the Laoyehai is created and maintained by heterotrophic processes that are fueled by organic matter, which are exacerbated by poor water exchange as a consequence of the geomorphology and weak tidal circulation.

Keywords Laoyehai · South China Sea · Coastal lagoon · Nutrients · Eutrophication · Hypoxia

Introduction

Coastal hypoxia has become a worldwide problem in the last three to four decades, partly because of landand/or marine-based human activities (Rabalais et al. 2010). Various categories of hypoxia occur in coastal environments, including hypoxia off large river estuaries, in semi-enclosed bays (e.g., fjords), and in areas affected by up-welling (Zhang et al. 2010). Although hypoxic phenomena in marine waters are diverse because of the various forcing factors and feedback mechanisms involved, the consequence is environmental deterioration ranging from deteriorated water quality to changes in function of the ecosystems. Because most of organisms need oxygen to survive, emerging hypoxia is a main threat to the nature of marine ecosystems, including such as species composition, biomass, and

Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, 238 Songling Road, Qingdao 266100, China

abundance as well as the biodiversity (Levin et al. 2009).

Aquaculture in coastal areas is an important technological innovation in marine fisheries. It has considerably increased food production from the marine ecosystem and is considered a crucial factor in terms of future global food security (FAO 2012). In China, the national report of the Ministry of Agriculture has shown that production from marine aquaculture reached $15 \times$ 10^{6} tons/year in 2010 and is comparable in amount to the wild-capture fishery (http://www.moa.gov.cn); wildcapture marine fisheries and aquaculture together account for 20% of the high-quality protein food supply in China. Based on the statistics, the mariculture in China produced an amount of 31.4×10^6 tons of seafood in 2014 (J.H. Zhang, personal communications), which accounted for 66% of the total world production (FAO 2016). It is expected that the experiences and lessons learnt from China, such as the influences of mariculture on the quality of adjacent marine environment and the function of ecosystems, will be beneficial to the adaptive management in a global context, particularly where the coastal planning and regulatory policies are critical in a sustainable human life.

Modern marine aquaculture includes algae cultured in rafts, crab and shrimp culture in ponds, culture of mollusks such as bivalves (e.g., clams) on mud flats and in rafts (e.g., mussels and abalone), and fish culture in cages. Aquaculture can help complement the food supply from the marine ecosystem, which is affected by increasing demand for high-quality protein and depleted wild stocks. However, there are negative effects of the aquaculture of species of economic importance in coastal waters, including pollution from shrimp ponds, the loss of habitat because of reclamation, the release of wastes from fish culture, and changes in biodiversity caused by the introduction of non-native species in culture (Cao et al. 2007). One of the negative consequences of marine aquaculture is the deterioration of water quality in adjacent coastal environments, induced by the input of aquaculture wastes, e.g., from shrimp ponds and fish cages. This results in reduced dissolved oxygen (DO) levels in the water column and in extreme cases the occurrence of hypoxia and anoxia, particularly aquaculture causes increase of organic matter as the waste release into the bottom sediments and oxygen depletion affects marine biodiversity in the vicinity of the cultured area (Diaz and Rosenberg 2008; STAP 2011; Valdemarsen et al. 2015).

In the Laoyehai of South China Sea, fish kill events were reported publicly to have occurred and created serious economic loss, which is attributed to waters of high chemical oxygen demand (e.g., > 5 mg/l) and elevated organic carbon concentrations (5–10%) in bottom sediments. These "fish kill" events all indicate environmental deterioration, but the cause has remained unclear (SCSIO 2006; HNMDDI 2011).

In this study, we investigated hydrographic and nutrient dynamics of Laoyehai in 2008-2011. Our hypothesis was that the input of wastes from the aquaculture of shrimp and fish exceeds the hydrodynamic removal capacity of this system. This results in hypoxia being maintained, induced by heterotrophic decomposition of organic matter, and also affects nutrient cycling. The purpose of this work was to assess if the evolution of hypoxia and/or anoxia in coastal environments is accelerated by adjacent land-based and marine aquaculture activities, using Laoyehai as an example. Although hypoxia and/or anoxia accelerated by aquaculture is well documented in literature, the interactions of driving forcing (e.g., marine agriculture) with hydrodynamics and geomorphology have created different consequences in different areas. This study reports a highly eutrophic system (i.e., Laoyehai) with really high nutrient and sulfide concentrations that is linked to the interactions of nature and human activities in semi-closed system, and groundwater further in combination can worsen oxygen conditions in the area. Lessons learnt from the Laoyehai is relevant to the mitigation strategy for similar problems in other tropical coastal areas where is affected by aquaculture. The data reported here were based on five field observations and combined with numerical simulation approach in 2008-2011.

Materials and methods

Study area

The Laoyehai is a narrow and semi-enclosed coastal bay on the east coast of Hainan Island, in the northern part of the South China Sea, and is classified geomorphologically as a sandbar–coastal lagoon system (Fig. 1). It extends from the Niujiaowei inlet in the west to Niumiaoling at the eastern end of the bay, a length of approximately 13 km, and has a surface area of 5.72 km² (Fig. 1). The lagoon is separated from the open South China Sea by the Niumiaoling hills in the



Fig. 1 The east coast of Hainan Island showing the location of the Laoyehai (a) and the sampling stations in the period 2008-2011 (b). c The initial conditions for the 3-D model simulation using passive tracer tracking techniques

south and east, and the water body is protected by a sandbar extending as far as to Niujiaowei (Fig. 1). The water depth of the Laoyehai is relatively shallow, typically 1–2 m at Niujiaowei and up to 5–6 m in the Niumiaoling area at low tide; the lagoon is connected to the South China Sea through a channel of approximately 50 m width at Niujiaowei.

The area around Laoyehai has a tropical climate, with an annual average temperature of 24.4 °C and annual rainfall of 2141.4 mm/year (SCSIO 2006). It is affected by monsoons dominated by northerly winds in winter and southerly winds in summer. In the offshore marine environment, the surface waters have an average salinity of 34.5, an average temperature of 26.5 °C, and a Secchi disc transparency of 20 m (SCSIO 2006). The Laoyehai is characterized by irregular diurnal tides, with a tidal range up to 0.8–1.9 m at the bay inlet and a tidal current of 0.1–0.5 m/s (HNMDDI 2011).

Adjacent to Laoyehai there are eight counties and towns, including Xinhua, Xincun, Longbao, Lantian, Lenan, Longshan, Fengling, and Qiaohai, with a total population of approximately 40×10^3 in the catchment area. Most of the population in this area relies on agriculture (i.e., rice culture) and fisheries (e.g., aquaculture of shrimp in ponds and fish in cages), and there is limited industrial development. Historically, the Laoyehai was one of the major harbors on the east coast of Hainan Island, but gradual siltation of the water channel made commercial navigation impossible after 1994 (HNMDDI 2011).

Field observations and sample analysis

Five field observations were undertaken in the period 2008-2011. These involved the entire Laoyehai, extending from the eastern end (i.e., Niumiaoling) to the bay inlet at Niujiaowei, and observations covered the northeast and southwest monsoon periods (Table 1 and Fig. 1). During each of the field observations, vertical profiles of depth, temperature, salinity, and epifluorescence were recorded at 10-15 stations using a SeaBird-25 CTD, and turbidity was measured using an OBS-3 optical probe. The hydrographic parameters (i.e., salinity and temperature) and chemical properties (e.g., pH and DO) of discrete water samples were measured in situ using a 350i Multi-parameter probe set; for measurement of the dissolved oxygen, the 350i DO sensor was calibrated using Winkler titration. Bottom sediments were collected using a Van Veen type grab sampler, and short sediment cores of 20–40 cm were collected using a gravity corer launched from the boat.

Surface water samples were collected at the sampling stations in pre-cleaned polyethylene bottles; profile samples were taken in 5-1 Niskin bottles according to CTD readings, and up to 4–5 samples were collected at each station for nutrient measurements. Surface water samples were also collected from catchment rivers and from shrimp ponds around the lagoon, using pre-cleaned plastic buckets. Prior to sampling, bottle and bucket were rinsed 3–5 times using the in situ water, and the water samples were transferred to acid-cleaned 10-1 polyethylene containers and stored in the dark.

Collected sediment samples were stored at -20 °C. Water samples were filtered through acid-cleaned polycarbonate filters (pore size $0.4 \mu m$); the filtrates were fixed with HgCl₂ (1/1000 v/v) for nutrient analysis, and the filters were stored at -20 °C for chemical analysis. Nutrient concentrations in the water samples were determined photometrically using a continuous flow autoanalyzer (model: Skalar SAN^{plus}) having a detection limit of 0.14 μ M for nitrogen, 0.065 μ M for phosphate (DIP) and 0.071 µM for dissolved silicate (DSi), respectively, with precision of less than 5-10% depending on species of interest (Liu et al. 2011). National seawater reference samples were included with each batch of samples analyzed, to evaluate the accuracy of the nutrient analyses. Total dissolved phosphorus (TDP) and total dissolved nitrogen (TDN) were measured photometrically using the auto-analyzer following wet digestion of the samples according to the methods of Grasshoff et al. (1999). The differences between TDP and dissolved inorganic phosphorus (DIP = PO_4^{3-}), and between TDN and DIN (DIN = $NO_3^- + NO_2^- + NH_4^+$), were defined as the dissolved organic phosphorus (DOP) and dissolved organic nitrogen (DON) concentrations, respectively.

Water samples for measurement of phytoplankton pigments (e.g., fucoxanthin: Fuco, and chlorophyll a: *Chl-a*) were immediately filtered through GF/F glass fiber filters (pore size 0.7 μ m) under low vacuum to avoid damage to the cells, and the filters were stored at – 20 °C. In the laboratory, the samples were thawed at 4 °C, immediately extracted twice using acetone and probe-sonication in an ice bath, and then centrifuged at low temperature. We used the analytical protocol for high-performance liquid chromatography (HPLC) analysis of pigments that was developed by Zapata et al. (2000) and modified by Zhu et al. (2015) to improve the

 Table 1
 Filed observations at the Laoyehai from Hainan Island in the period of 2008–2011

Time	Period	Field observations and remarks Field work was carried in the second week of the August 2008, and the hydrographic measurements and water sampling were undertaken from 6 stations inside the Laoyehai; hypoxic water (DO, 21.9 μM) was found at the near-bottom.				
August 2008	SW monsoon					
April 2009	NE monsoon	Field work was carried out twice in the first week and second week of April 2009, respectively, and hydrographic measurements and collection of water samples were made at six stations each time; hypoxia (DO, 18.8 μ M) was found at near-bottom waters.				
August 2009	SW monsoon	Field work was carried out twice in the first week of August 2009, with total 35 stations inside the Laoyehai, the DO minimum of 12.5 μ M was found in the near-bottom waters.				
April 2010	NE monsoon	Field work was carried twice in the last week of April 2010, each time ten stations were occupied for the hydrographic measurements and water sample collections; the minimum DO was found at 6.25μ M.				
August 2011	SW monsoon	Field observations were undertaken at 10 stations in the third week of August 2011, and hydrographic measurements and sample collections were made according to the change in water quality; the DO was found as low as 3.13 µM.				

separation of pigments and the signal to background ratio. For routine analyses, 100 µl extracts were injected into a reversed-phase HPLC (model: Agilent 1100 series) equipped with an online vacuum degasser, quaternary pump, auto-sampler, thermo-state column, and diode array detector (DAD). All procedures were performed under low light, and the extracts were stored at -40 °C until analysis on HPLC. Before sample analysis, the extract was vortexed after addition of Milli-Q water (5:1 v/v), which was to avoid distortion of the shape of early eluting peaks (Zapata and Garrido 1991). The phytoplankton pigments were identified based on the retention time and absorption spectra in comparison to authentic standards (Zhu et al. 2015). The Chl-a standard was obtained from Sigma-Aldrich, and that for Fuco was obtained from DHI.

Numerical simulations

In addition to the field observations, we used a 3-D simulation technique to examine impacts of hydrodynamics on the flushing times and retention of nutrients in the Laoyehai, the ELCIRC hydrodynamic model was used to understand the hydrodynamic processes using numeric simulation for tracking passive tracers (Appendix 1). A Lagrangian particle tracking technique was used to study the pathway of pollutants in the Laoyehai and exchange with the adjacent South China Sea. The ELCIRC hydrodynamic model was used to generate the hydrographic and flow fields that drive the motion of released "passive particles" in Lagrangian mode (Zhang and Baptista 2004). The Lagrangian particle tracking experiment was combined with a random-walk model of horizontal eddy diffusion that enabled a particlebased statistical treatment of turbulent mixing. In the 3-D numerical simulations, position of a given particle was described by Eq. 1:

$$\vec{x}(t + \Delta t) = \vec{x}(t) + \vec{u}\Delta t + z_n \sqrt{2\vec{K}\Delta t}$$
(1)

where \vec{x} is the particle position, Δt is the time step, \vec{u} is the velocity vector, \vec{K} is the eddy diffusion tensor determined in the hydrodynamic model, and z_n is a random vector.

Based on the 3-D numerical simulation of the hydrodynamics (e.g., circulation and tidal structures for salinity and temperature) of the Laoyehai, scenario analyses were used to investigate the water exchange and dispersal of dissolved materials (i.e., nutrients and other pollutants) using the track of passive tracers approach. This included the following:

- (i) Assessing the rate of exchange of the water mass within a typically irregular diurnal tidal cycle in relation to the total water volume of the Laoyehai, which enabled estimation of the flushing time of the system
- (ii) Tracking the trajectories of passive tracers in the Laoyehai, released from the bay inlet and at the end of the water channel, respectively, to enable understanding of the dispersal of pollutants released from aquaculture activities

The model simulation was validated against in situ tidal measurements (e.g., water level change and current) and hydrographic properties, including temperature and salinity at anchor stations (HNMDDI 2011).

Results

The hydrographic and chemical properties of the Laoyehai during the five field observations are shown in Table 2. The data show the seasonal and inter-annual variability of the measured parameters, undertaking in different monsoon periods.

Southwest monsoon period

The field studies undertaken in summer (August 2008, 2009, and 2011) were carried out during the southwest monsoon period of the South China Sea. Analysis of hydrographical properties showed that in this period the Laoyehai was vertically well mixed in the vicinity of the opening to the South China Sea (i.e., the bay inlet). However, there was weak stratification at the eastern end of the water channel (Table 2 and Fig. 2). The salinity reached 32.0-32.5 at the bay inlet, with almost no difference between the surface and near-bottom waters because of exchange with the open South China Sea. At the eastern end of Laoyehai the salinity was as low as 15 at the surface but 20-25 in bottom waters, reflecting the influence of fresh water influx. The temperature in summer was ≥ 30 °C at the bay inlet and

could be 2–4 °C higher at the eastern end of the water channel and hence coastal waters of South China Sea; surface waters were up to 2 °C higher than the nearbottom waters in this period. The DO concentration was up to 280–300 μ M in surface waters (i.e., >125% saturation), and the pH ranged 8.00–8.15, but in nearbottom waters inside the lagoon the DO concentration was 15–30 μ M or less (ca. 10–20% saturation), and the pH was 7.35. This was particularly the case at the eastern end of the channel, where exchange with the South China Sea is mostly restricted because of the narrow water course.

Among the various nutrient species, the DIN concentration was 10 μ M at the inlet of Laoyehai, but increased to \geq 90 μ M inside the lagoon and to the eastern end; previous studies reported eutrophic character in this area (SCSIO 2006). The NO₃⁻ and NO₂⁻ species were both minor components of DIN, and their concentrations were higher at the surface than in the near-bottom waters. The dominant inorganic nitrogen compound was NH₄⁺, which comprised 80–90% or more of DIN in the water column (Table 2 and Fig. 2). High concentrations of NH₄⁺ were also found in the near-bottom waters, which resulted in DIN increasing with water depth. The concentration of DON in the lagoon reached 40–75 μ M, and accounted for 40–50% of the TDN (Table 2).

The DIP concentration in Laoyehai ranged from 0.25 to 10 μ M, with a trend of increase from the opening (west) to the end (east) of the bay. Higher concentrations of PO₄³⁻ occurred in near-bottom samples than in surface waters (i.e., by a factor of 2–4). The DOP concentrations were

Hydrographic and	chemical properti	es					
Time/Period	Temperature (°C)	Salinity		pН	DO (µM)	
August 2008	27.3-29.1		9.1-31.6		7.59-8.14	22–293	
April 2009	24.7-30.4		10.1-33.2		7.53-8.51	19–281	
August 2009	28.6-34.2		12.2–32.2			12-311	
April 2010	27.0-30.1		13.8–33.0		7.68–9.28	5-403	
August 2011	29.9-34.1		13.5–32.3		7.40-8.60	3–284	
Nutrient compositi	ions (unit: μM)						
Time/period	NO_3^-	NO_2^-	$\mathrm{NH_4}^+$	PO_4^{3-}	DSi	DON	DOP
August 2008	0.065-5.81	0.50-4.62	5.63-91.8	0.33-5.18	61.2-86.4	39.9–74.5	0.30-1.50
April 2009	0.10-21.8	0.08-4.20	0.57-47.5	0.26-3.71	6.94–51.0	7.96–34.9	0.17-0.63
August 2009	0.25-11.0	0.065-2.73	5.94-39.7	0.48-4.18	5.95-71.8		
April 2010	0.10-5.35	0.08-5.93	6.90-47.4	0.49–12.4	1.97-25.1		
August 2011	0.10-7.32	0.065–7.68	2.24-44.6	0.24–10.3	2.66-113		

Table 2 Summary of hydrographic parameters and chemical properties of water samples in the Laoyehai in 2008–2011



Fig. 2 Hydrographic properties (i.e., salinity and temperature) and nutrients (e.g., DIN, DIP, and DSi) along a longitudinal section in the Laoyehai in summer, which is dominated by the southwest

typically higher in near-bottom than surface waters and accounted for ca. 20% of the TDP (Table 2).

The Laoyehai was enriched in dissolved silicate, compared with the open South China Sea that is depleted in DSi. The concentration of DSi in surface waters ranged from 1 to 2 μ M to as high as 100 μ M in the lagoon, depending on the sampling site, with a strong gradient of reduction from the end (east) of the bay to the inlet (west). In contrast to the DIN and DIP, which

monsoon over the South China Sea. Note that station L9 is not included in the section

decreased from near-bottom to surface waters, the DSi concentration was 20% higher in surface than nearbottom waters (Fig. 2).

In the summer monsoon period the DIN/DIP molar ratio ranged from 1.5–2 at the end of the lagoon to 15– 20 at the inlet of the bay where DIN/DIP approaches to the Redfield ratio, whereas the DSi/DIN ratio ranged from 1–2 at the inlet of the bay to 30–35 in end of the lagoon, declining towards the open coastal waters.

Northeast monsoon period

Field studies were undertaken during spring (April 2009 and 2010) corresponded to the late stage of the northeast monsoon period in the South China Sea. Over this time, the water samples from the inlet to the Laoyehai had a salinity range of 30.5-33.5 and a temperature range of 27.5-30.5 °C, and the hydrographic parameters indicated a well-mixed vertical profile (Fig. 3). Inside the lagoon the salinity was approximately 20 and the temperature range was 25.0-27.5 °C, demonstrating the

influence of fresh water input from terrestrial sources, and no stratification was evident (Fig. 3). The DO concentration in the water column was approximately 250 μ M at the bay inlet (i.e., 100–105% saturation). Inside the Laoyehai the DO concentration in surface water was 375–410 μ M (i.e., 120% saturation), while the near-bottom waters had DO concentrations as low as 6.0–6.5 μ M, corresponding to an apparent oxygen utilization (AOU) of 220–230 μ M. The pH values of samples were as high as 9.28 at the surface, but could be < 7.50 in the hypoxic zone of near-bottom waters.



Fig. 3 Hydrographic properties (i.e., salinity and temperature) and nutrients (e.g., DIN, DIP, and DSi) along a longitudinal section of the Laoyehai in spring, which is dominated by the northeast

monsoon over the South China Sea. Note that stations L9 and L10 are not included in the section

During the northeast monsoon period the NO₃⁻ concentration was only 1-2 µM at the channel inlet affected by the coastal waters of the South China Sea, but could be as high as 20 µM in surface waters inside the lagoon. The NO₂⁻ concentration was generally low in the coastal waters (i.e., $< 0.2 \mu$ M), but was up to 4–5 μ M at surface inside the Laoyehai. The dominant species of DIN during this period was again NH₄⁺, with concentrations of 1-2 µM for samples associated with open coastal waters and up to 40-45 µM for surface waters inside the lagoon (Fig. 3). The concentrations of phosphate were $< 0.5 \mu$ M in the bay inlet connected to the South China Sea coastal waters, but were 3.0-3.5 µM in surface waters of the lagoon, showing a similar trend for nitrogen species. The DSi concentration was approximately 2.0 µM at the inlet of the Laoyehai, where exchange with the South China Sea is more rapid through, but could be as high as $50-55 \mu$ M inside the lagoon, indicating the influence of terrestrial influx.

During the field studies in the northeast monsoon period the DON concentration was < 10 μ M at the bay inlet but 30–35 μ M inside the Laoyehai; DON represented 80–90% of the TDN for samples at stations affected by the coastal waters and 30–40% within the lagoon. The DOP concentration was 0.2–0.7 μ M at stations sampled inside the Laoyehai, and this accounted for 10–65% of the TDP. The DIN/DIP ratio was as low as 1–2 inside the lagoon, but up to 25–30 at the bay inlet stations, which was similar to values of the South China Sea. The DSi/DIN ratio approached 1.0 at stations affected by the South China Sea coastal waters, but was 5– 10 where terrestrial inputs were significant.

Photosynthetic pigments in surface waters

A spatial gradient of photosynthetic pigments occurred in the Laoyehai during the study. For instance, *Chl-a* at the inlet to the lagoon adjacent to the coastal waters was $0.5-1.0 \ \mu g/l$, indicating the influence of the adjacent oligotrophic South China Sea. However, inside the Laoyehai, the *Chl-a* concentration increased towards the end of the lagoon and reached 70–80 $\mu g/l$ in the surface waters, again indicating nature of eutrophication (Fig. 4).

In terms of phytoplankton community structure, diatoms were abundant in the photosynthetic biomass at the inlet of the Laoyehai, where the Fuco/*Chl-a* ratio was up to 20–30%, whereas for the nutrient enriched waters inside the lagoon the Fuco/*Chl-a* ratio was as low as 4–5%, indicating the dominance of other phytoplankton species, particularly chlorophytes (Zhu et al. 2015).

Vertical profile of DO and nutrients

A remarkable phenomenon in the Laoyehai was the enrichment of phytoplankton pigments in the surface waters, leading to skewed molar ratios for DIN/DIP and DIN/DSi relative to the Redfield ratio. Another possibility is that the skewed nutrient species ratio can be linked to the terrestrial influx that will be discussed later. Furthermore, low DO levels were found in the near-bottom waters, but this was subject to seasonal change and inter-annual variability. Inside the Laoyehai the water depth can be up to 5–6 m, and the DO concentrations at the surface indicated an oxygen over-saturation, but could be as low as almost null near the sea floor (Fig. 5). In this area, the vertical profile for NH₄⁺ showed an increase with water depth, with nearbottom samples having concentrations five- to tenfold higher than in surface waters. The NO₃⁻ and NO₂⁻ concentrations did not show a trend similar to NH4⁺ with water depth, with surface water concentrations being comparable to or higher than those in near-bottom waters (Fig. 5). The concentrations of DIP and DSi could also increase with water depth, depending on the study period, but the difference between surface and near-bottom waters was not as evident as for NH_4^+ (Fig. 5).

Indeed, the water column DO concentration in Fig. 5 decreases rapidly with the increase of reductive to oxidative nitrogen species ratio (e.g., NH_4^+/NO_3^-) as well as DSi/DIP, which can be described using statistics of power



Fig. 4 Distribution of *Chl-a* (μ g/l) in the surface waters of Laoyehai for the period 2008–2011, showing the occurrence of high concentrations inside the lagoon, and oligotrophic condition in the bay inlet area, where is affected by the adjacent South China Sea





Fig. 5 Vertical profiles of hydrographic parameters (**a**) and nutrients (**b**) in the inner Laoyehai, indicating the nature of the hypoxia of near-bottom waters. In the figure, observed DO concentrations

law. Since the ratio of NH_4^+/NO_3^- and DSi/DIP in deep water samples of Laoyehai differ considerably from the nature of adjacent terrestrial sources and drainage from shrimp ponds (Table 3), the observed nutrient ratios in the hypoxia waters can indicate the heterotrophic consumption of DO resulting from the degradation of organic matter and mineralization of nutrient species. The heterotrophic decomposition of organic materials in the Laoyehai can be sustained because of sluggish nature of water exchange that results in a rather long residence time within the system. However, the complex relationship between observed DO and nutrient species ratios (e.g., power law) in the water column indicates that deoxygenation processes in the Laoyehai is a consequence of multidriving forces, rather than a simple/single forcing and response system.

are also plotted again the ratios of NH_4^+/NO_3^- (c) and DSi/DIP (d) for the water samples. Note that decrease of NO_3^- and PO_4^{3-} can be results of impacts from SGD (Ji et al. 2013)

Terrestrial influx and discharges from aquaculture

The surface runoff (i.e., creeks and rivers) emptying into Laoyehai had DO concentrations of 200–250 μ M (ca. 100% saturation) and a pH of 8.3; no other data can be obtained at this time for surface water discharges, either from scientific studies or governmental reports for surface runoff in this area. The concentrations of nutrients in surface runoff were 20–120 μ M for NO₃⁻, 0.5–6.0 μ M for NO₂⁻, 10–35 μ M for NH₄⁺, 0.2–1.5 μ M for DIP, and 100–150 μ M for DSi, respectively (Table 3). This produced a DIN/DIP molar ratio of > 100 and a DSi/DIN ratio in the range 0.5–1 to 4–5 for surface runoff (Table 3).

Samples collected from the outflow channels of shrimp ponds adjacent to the Laoyehai differed from the riverine and coastal waters in terms of the concentration for

River and samples from shi	rimp ponds outflow					
Areas	NO_3^-	$\mathrm{NO_2}^-$	$\mathrm{NH_4}^+$	PO_{4}^{3-}		DSi
Riverine influx	20.3-117	0.67-5.92	11.9–31.1	0.20-1.41		107-147
Shrimp pond outflow	0.24-2.65	0.15-0.83	1.90-38.1	0.08-0.91		5.47-28.8
Samples of groundwater						
Salinity range	NO_2^-	$\mathrm{NH_4}^+$	NO_3^-	PO_4^{3-}	DSi	DO
<1.0	0.1-3.2	0.1-20.1	3.9-489	0.1-28.6	1.2–355	37.5–300
1.0-33.0	0.1–2.1	3.5–25.1	0.14–5.4	0.1–12.0	2.8-53.6	90.6-403

Table 3 Composition (unit: μ M) of water samples from rivers, outflow of shrimp ponds, and groundwaters that empty into the Laoyehai. Data of groundwater composition are from Ji and co-workers (2013)

nutrients (Table 3). Briefly, samples from the outflow of shrimp ponds had concentrations of 1.9–38 μ M for NH₄⁺, 0.14–0.83 μ M for NO₂⁻, and 0.25–2.7 μ M for NO₃⁻, relatively low compared to surface runoff. Shrimp pond outflows had rather low DSi (e.g., 5.5–29 μ M), but comparable DIP concentrations, at 0.08–0.9 μ M relative to surface runoff (e.g., river) in this area. This resulted in different DIN/DIP and DSi/DIN ratios from shrimp pond outflows compared to rivers.

Results of numeric experiments

The application of ecosystem models (e.g., numerical simulation for nutrients, phytoplankton, zooplankton, and detritus) to the Laoyehai is difficult so far, because the external driving forcings (e.g., drainage of wastes from aquaculture) have not been quantified, and knowledge of the structure of the lagoon food web is still poor. However, modeling of the hydrodynamics can shed light on a number of important issues relevant to ecosystem functioning in the Laoyehai (Figs. 1 and 6). With regard to hydrodynamics, the water exchange between the inner part and near the bay inlet is also limited in a typical tidal cycle. The 50% flushing time, estimated by tracking passive tracers (i.e., particles) using numerical techniques, can be ca. 250 days for the water body at Haiji of the lagoon, and the 50% flushing time to the east of Niumiaoling may be up to 700 days (Fig. 6).

Discussion

Comparison with other coastal environments on Hainan Island

Relative to other coastal lagoons and/or bays on the east coast of Hainan Island, the Laoyehai has unique nutrient chemistry, because nutrients are enriched in surface samples and the near-bottom waters can be hypoxic and even anoxic. For instance, compared to Bamenwan (bay) and Xiaohai (lagoon), both of which are coastal lagoon types of water body and further to the north along the east coast of Hainan Island (Fig. 1), the Laoyehai has much higher levels of NH4⁺ and phosphate in surface waters, but lower concentrations of NO_3^- and DSi (Table 4). Similar to Laoyehai, Bamenwan and Xiaohai are both influenced by the aquaculture and reclamation and have rather limited terrestrial influxes (e.g., rivers); however, hypoxic/ anoxic conditions have not yet been documented in their near-bottom waters, presumably because Bamenwan and Xiaohai are more open in nature and so are less impacted by waste drainage (e.g., retention of wastes in the water body). For instance, the water residence time in Bamenwan is approximately 1 week, which is much shorter than that for Laoyehai (INMDDI, 2011; Liu et al. 2011). This implies that organic wastes and nutrients released from mariculture have less chance to accumulate in the system because of the more rapid flushing process, and so the water body remains normoxic in areas such as Bamenwan and Xiaohai. Although previous studies have shown that the scale of aquaculture activities in Bamenwan is five- to sixfold that in Laoyehai (Herbeck et al. 2013), the surface area of Bamenwan is more than fivefold larger and water is shallow, ca. 1 m at low tide. Moreover, information on aquaculture from satellite images (Herbeck et al. 2013) can be misleading because many shrimp and fish ponds of Bamenwan have been abandoned since the late 1990s, leading to an inaccurate assessment of the scale of aquaculture and its impact.

The patterns of nutrient species in the Laoyehai lagoon differ also from those of the coastal waters off Hainan Island, in terms of the concentrations and/or



Fig. 6 Results of the 3-D numerical simulation based on passive tracers, designed to show the impact of tidal circulation on water exchange between the Laoyehai and the South China Sea. **a** The passive tracer distribution 180 days following release under the

initial conditions as shown in Fig. 1c and **b** the time (days) required for a given percentage of the passive tracers that were released at the east end of Laoyehai and monitored at Haiji and Niumiaoling, respectively, in Fig. 1

nutrient species ratios (Table 4). This highlights that, compared with other normoxic coastal lagoons and offshore environments in the east coast of Hainan Island, hypoxic and/or anoxic conditions and the nutrient regimes in the Laoyehai are seasonally affected at local scales by anthropogenic activities, particularly the marine aquaculture.

Impacts of human activities

Historical data for the Laoyehai indicates that there has been a deterioration of water quality in this area. For instance, in 2005–2006 the reported nutrient concentrations in the lagoon were 0.71–9.3 μ M for DIN and < 1.6 μ M for DIP (SCSIO 2006); these values are five- to tenfold lower than in the period 2008–2011 (Table 2).

Human activities over the last 60 years, particularly the construction of dams and water gates in the 1960s to 1980s, have considerably altered the nature of the Laoyehai. These constructions/structures modified the hydrographic conditions and hydrodynamic processes through change in geomorphology by human activities, and the later introduction of marine aquaculture (e.g., shrimp ponds and fish cages) caused further deterioration of the water quality through release of solid and liquid wastes, modification of habitats, and reduction of environmental capacity because of reclamation of wetlands for aquaculture. For instance, in the 1970s, three major dams and/or water gates were constructed in the area of the Laoyehai, giving a combined catchment area of 1920 ha upstream, particularly to the east of Longbao and Niumiaoling (HNMDDI 2011).

Marine aquaculture proliferated in the Laoyehai in the late 1980s, with shrimp culture in ponds occupying 250–300 ha; fish culture commenced in this region in 1986, with the number of fish cages increasing from 1650 in 1992 to 9580 in 2000 (HNMDDI 2011). The fish cages are mainly deployed in the water channel close to the bay inlet, and this has considerably reduced the water exchange rate and modified the pattern of circulation.

Consequently, over the last five decades (i.e., 1962–2010), the surface area of the Laoyehai declined by 40%, from 9.57 to 5.72 km² (Fig. 7). Over the same time, the water volume of tidal prism decreased by 60%, from 7.19×10^6 m³ in 1962 to 3.03×10^6 m³ in 2010, and the width of the channel at the bay inlet decreased from 470 m in 1962 to approximately 50 m in 2010 (HNMDDI 2011). The historical records show also that the tidal current was 0.5–1.0 m/s in the Laoyehai in the 1980s, but measurements at the bay inlet in 2009 showed the current was only 0.1–0.3 m/s. This indicates again that water circulation in the Laoyehai has been substantially reduced in recent years (SCSIO 2006; HNMDDI 2011).

In a summary table, we reviewed and compared different methods used for estimating "residence time" and/or "flushing time" of Laoyehai (Table 5). Recently, there has been an effort to use radio isotopes (e.g., ²²⁴Ra and ²²⁶Ra) as tracer to study the water ages of Laoyehai, and the results showed a residence time of 5.4 days in surface (Wang and Du 2016). This is apparently an underestimate of water residence, because if we consider the amount of 3.0×10^6 m³ of tidal prism (range $1.0 \times$ 10^6 m^3 -4.1 × 10^6 m^3) and an average water depth of 3 m in the Laoyehai (HNMDDI 2011; Wang and Du 2016), the flushing time induced by tidal exchange would be ca. 6 days. Furthermore, given the average tidal range of 0.34 m (HNMDDI 2011) and the water depth at Laoyehai, the time scale of tidal induced exchange can be as short as 9 days. In fact, the slow exchange rate in the lagoon is because of its narrow and semi-enclosed nature with micro-scale tide. Previous work showed that the water mass exchanged in a diurnal tidal cycle was

 $\label{eq:comparison} \mbox{Table 4} \mbox{ Comparison of nutrient concentrations (unit: μM) with other coastal bays of the Hainan Island and coastal waters of the South China Sea$

Areas	NO ₃ ⁻	NO_2^-	$\mathrm{NH_4}^+$	PO ₄ ³⁻	DSi	DIN/DIP	DSi/DIN
Bamenwan	24.3	2.0	15.8	0.5	68.0	84.2	1.6
Xiaohai	12.5	0.5	6.0	0.5	105	40	6.5
East coast of Hainan	4.98	0.52	5.70	0.25	16.1	44.8	1.4

Data of Bamenwan and coastal waters off the east coast off the Hainan Island are from Liu et al. 2011; data from the Xiaohai are from our unpublished results



Fig. 7 Change in the surface area (km^2) and coastline (km) of the Laoyehai since the 1960s and areas used for marine aquaculture, particularly shrimp ponds and fish cages. Refer to Fig. 1 for

only approximately 1% of the total Laoyehai (HNMDDI 2011), which results in an estimate of water residence at scale of hundred days (Table 5).

Deterioration of Laoyehai owing to enrichment of organic materials

Previous studies have shown that among various pollution issues affecting the Laoyehai, marine aquaculture and sewage drainages from adjacent populated areas are important in causing deterioration of the water quality. In the Laoyehai, it has been reported that chemical oxygen demand (COD) is up to 0.9–1.0 tons/day for

 Table 5
 Overview of different methods used for estimations of "flushing time" and/or "residence time" of Laoyehai, and for the reason of simplifying the comparison, we do not distinguish

comparison with human impacts on the evolution of this coastal lagoon through reclamation. Data are from HNMDDI (2011)

the waste drainage from shrimp ponds, 2.0–2.5 tons/day of sewage drainage from urban and rural centers, and 0.5–1.0 tons/day from caged fish culture (SCSIO 2006). In the period 2001–2004, the COD in the channel close to the bay inlet ranged widely from 0.08 to 4.77 mg/l with an average of 2.52 mg/l because of tidal exchange with offshore coastal waters having relatively low COD, while at the eastern end of the Laoyehai the COD was higher but more stable, with range of 4.20–5.83 mg/l and an average of 5.10 mg/l (MEMCHN 2005). The COD data together with elevated nitrogen and phosphorus concentrations indicate that the Laoyehai is eutrophic based on the "nutrient index methods" for

between different "time" concepts and adopt the term of "water age" as an operational approach

Methods	Time scale (days)	References	Remarks
²²⁴ Ra and ²²⁶ Ra as tracers	5.36 ± 3.08	Wang and Du 2016	The work is based on mass balance concept and uses the measurements of radio isotopes in the water column and groundwaters
Estimate based on the tidal exchange	6–9	HNMDDI 2011 and this study	It is assumed that the whole water body is well mixed in a diurnal tide and the characteristic time is the ratio of water volume and tidal prism
Model calculation and validation with hydrographic data	ca. 100	HNMDDI 2011 and this study	Calculation is based on the fact that a portion of tidal prism discharged offshore in ebb period will be back into the system along with nextflood tide, and the net exchange is ca. 1% of whole water body

assessing water quality of freshwater and/or coastal systems in China (Xiao et al. 2007). Our observed chemical properties and biological parameters (e.g., *Chl-a*) also indicate that the Laoyehai has eutrophic characters with enrichments of nutrients and organic matter compared with coastal waters in developed countries, such as in the Europe and North America (cf. Bricker et al. 2003).

In the area affected by marine aquaculture (e.g., shrimp pond outflow and fish cages) of Laoyehai, the bottom sediments had a total organic carbon (TOC) concentration up to 10%, and in the area of oxygen depletion the concentration of sulfides was up to 400–500 mg/kg (SCSIO 2006). This indicates that heterotrophic processes dominate inside the Laoyehai, although the water depth is shallow and the euphotic zone can extend to the benthos in the lagoon (SCSIO 2006). The elevated levels of NH_4^+ in the water column provide with evidence that catabolic processes can dominate the nutrient budgets and mineralization plays an important pool of nutrient recycling, such as nitrogen.

Oxygen depletion in the coastal lagoon has an impact on both the pelagic and benthic communities; for example, in the Laoyehai biomass of benthic fauna affected by depletion of DO was reported to be only 2.5–3.0 g/ m², which is five- to tenfold lower than the values (i.e., 190–200 g/m²) at oxygen replete stations and the area close to the bay inlet (SCSIO 2006). In waters affected by hypoxia/anoxia in the Laoyehai, heterotrophic bacteria abundance can reach up to 20×10^9 cell/l and abundance of virus at 60×10^9 virus/l. Phytoplankton of > 20 µm size fraction is dominated by *Cerataulina pelagica* and *Phizosolenia alata f. gracillima* (Li et al. 2014b). In terms of phytoplankton biomass, it was found that 60% to 98% can be contributed by picoand nano-size fractions (Li et al. 2014b).

The enrichment of nutrients and organic matter in the Laoyehai can be compared to other world coastal areas affected by marine aquaculture, for example, similar and high levels of sulfide, reduced nitrogen (e.g., NH_4^+) and organic materials have been continuously reported from coastal areas affected by aquaculture since 1990s, such as Bay of Fundy (Hargrave et al. 1997), Seto Inland Sea of Japan (Pawar et al. 2001), British Columbia (Sutherland et al. 2007), Tyrrhenian Sea (Mirto et al. 2012), and the Hardanger Fjord of Norway (Valdemarsen et al. 2015). The observed negative impact on ecosystems is not necessarily limited to the water column, but benthic organisms, including such

as composition and structure of communities linked to environmental index (e.g., redox potential, COD and sulfide concentration), species ratio (e.g., nematode to copepod) and richness (Sutherland et al. 2007; Mirto et al. 2012; Valdemarsen et al. 2015).

Hypoxia and the chemistry of nutrients in Laoyehai: short-term variation and seasonality

We investigated the seasonality and short-term variability (i.e., differences between consecutive observations) of hydrographic (i.e., temperature and salinity) and chemical (e.g., nutrients and DO) parameters using statistical software (SPSS 10.0) to process the data. Emphasis was given to the inner part of Laoyehai, where depletion of DO is a recognized consequence of societal and economic activities. In April, during the northeast monsoon period, the water column had lower temperatures and DSi concentrations than in August when the southwest monsoon prevails (one-way ANOVA, p < 0.05), highlighting the effect of greater solar radiation and rainfall in summer. This was followed by stratification and increased input of terrestrial materials including DSi in this area in summer. Statistically, DO concentration in surface waters was significantly lower in April than in August in 2008-2011, providing evidence that vertical mixing is stronger during the northeast monsoon with reduction of phytoplankton biomass (e.g., Chl-a), although the near-bottom waters of the lagoon can be oxygen depleted for most of time in the year.

With regard to short-term variability in the Laoyehai, two consecutive field surveys in April 2009 showed that the concentrations of nutrients and DO differed by twoto fourfold or more over a time scale of 2 weeks (t test, p < 0.05). In the Laoyehai, the depletion of DO leads to hypoxia and/or anoxia in near-bottom waters. The vertical profiles showed the short-term instability of DO in the lagoon. For example, on 2 April 2009, the DO concentration was 160-165 µM in the near-bottom water (e.g., 4.8 m), the salinity was 15.7, and the temperature was 25.7 °C. On 14 April 2009, about 2 weeks later, in the near-bottom waters (i.e., 4.9 m) at the same station, the salinity was 13.1, the temperature was 27.0 °C, and the DO concentration was much reduced, at 18–19 μ M. In the meantime, a reduction in the NO₃⁻ and DSi concentrations, and an increase of 30-40% in the NH4⁺ concentration are also observed. This was presumably related to stratification and eutrophication in surface following freshwater input following rainfall events that occurred between these two observations; this increased the stability of the water column and biomass, and the higher water temperature accelerated heterotrophic consumption of DO in the near-bottom waters.

In the near-bottom waters, the AOU on 14 April 2009 was a factor of three higher than on 2 April (21.0 μ M vs. 70.2 μ M). Although the *Chl-a* concentration was not measured on 2 April, the surface Chl-a on 14 April was 10.8 μ g/l, which is quite comparable to other campaigns and even other coastal bays of Hainan with eutrophic character. The large biomass of phytoplankton may have increased the amount of organic matter that settled into the near-bottom waters, which fuels the heterotrophic degradation of organic matter and drives the oxygen depletion. In a previous study, Ji et al. (2013) reported groundwater discharge (e.g., SGD) in this region. Given that groundwater can be low in DO level, a quantitative evaluation needs to be carried out to investigate the respective contributions of groundwater discharge and heterotrophic activity to the variability of DO in water column.

In the near-bottom layer, the decrease of salinity in the period of 2 weeks (i.e., 2-14 April 2009) suggests the input of groundwater in this area (Ji et al. 2013). Given that the salinity of the groundwater samples around the Laoyehai is in the range of 0-33, the decrease in salinity from 15.7 to 13.1 indicates that the proportional input of fresh groundwater at Longbao can be increased by up to 10–20% with an average of 17% in this period, if we assume the recharge of near-bottom waters by fresh groundwater with a salinity of 0.4 measured from wells nearby. This is consistent with the findings of Ji et al. (2013), who estimated that the groundwater discharge rate was at 0.1 m³/m²/day. The author also reported that the groundwater DO concentration ranged from 37.5 to 400 μ M, with the highest values being found for samples from wells, where the influence of equilibrium with atmosphere cannot be ignored. Moreover, fresh groundwater and recycled saline water samples underground can be different in nutrient levels. For example, in Table 3, fresh groundwater is characterized by elevated NO_3^{-} , PO_4^{3-} , and DSi that can be even higher than riverine concentrations, whereas samples of recycled saline water or saline groundwater, illustrate $NH_4^+ > NO_3^-$, with relatively low concentrations for PO₄³⁻ and DSi. This finding can be used to explain the vertical profiles of nutrients in the Laoyehai, as shown in Fig. 5: the reduction of nitrate and phosphate in near-bottom waters can be induced by recycled saline waters as SGD contribution. It should be noted, however, that SGD can be a major source of nutrients to the marine environment, for example, marine aquaculture area in the northern part of China, e.g., Sanggou Bay (Wang et al. 2014), as well as making a significant contribution to the budget of nutrients at basin-wide scale, e.g., Mediterranean Sea (Rodellas et al. 2015).

In a study of an open ocean beach in the South Atlantic Bight (i.e., the coast from North Carolina to Florida, USA), McCoy et al. (2011) reported that groundwater DO concentration is 29 µM on average, with slightly higher concentrations (e.g., 56 μ M) in summer. For the Laoyehai, based on the average groundwater DO concentration of 215 µM and assume nil for DO as the extreme condition (Ji et al. 2013), the additional 17% groundwater in the near-bottom waters over a period of ca. 2 weeks would have equated to a 5% and 28% reduction, respectively, in the oxygen concentration of water column. Heterotrophic processes in the water column would hence have contributed the other 72-95% of oxygen depletion. McCarthy et al. (2013) indicated that water column processes in the Gulf of Mexico accounted for 75% of total near-bottom water oxygen depletion, with sedimentary process accounting for aother 25%. As a consequence of the release of reductant materials, sedimentary process in the Laoyehai may in total have contributed to an important proportion of the oxygen depletion, for which the quantitative estimate is difficult at this time because of lack of data.

The quasi-simultaneous measurements of hydrographic and chemical properties reveals that in the center and inner part of the Laoyehai the water was oxygen depleted, with DO concentrations of 3 μ M, and in some cases $\sim 0 \ \mu M$ in near-bottom waters (Fig. 8). In this water mass, the salinity was usually low at 10-20 compared with > 30 at the inlet of the bay, and the temperature was lower by 1-2 °C or similar compared with offshore waters. This, in combination with the high concentration of DSi, highlights that the inner part of the lagoon system is considerably affected by terrestrial influxes. Another contribution to the high DSi concentrations in near-bottom waters is from groundwater discharges, and Ji et al. (2013) showed that the DSi concentration ranges from 20 to 355 µM for SGD in this area.



Fig. 8 Quasi-simultaneous measurements (i.e., within half a day) for hydrographic parameters in Laoyehai, showing the center of hypoxia in the near-bottom waters. The black and white circles are

sampling stations, and area for shrimp ponds and/or fish cages associated with marine aquaculture is also shown

The high concentrations of NH_4^+ and elevated NH_4^+/NO_3^- ratio of approximately 20 in the Laoyehai when compared to the riverine influx indicates that dissimilatory nitrate reduction to ammonium (DRNA) can be an important process, along with heterotrophic decomposition of organic matter, that regulates the nitrogen cycle in the lagoon; fixed nitrogen may also be lost as N_2 and/ or N_2O to the atmosphere. Compared with the riverine end-member composition and/or relative to the Redfield ratio, the relatively high DIP/DIN in the water column

indicates that mineralization supported by heterotrophic decomposition of organic matter is important mechanism creating $\rm NH_4^+$ and $\rm PO_4^{3-}$ enrichment of the nearbottom waters of the Laoyehai.

Indeed, the high values of COD in the water column as noted above suggest high organic matter loadings and hence the potential for DO consumption. This again highlights that low DO concentrations in the Laoyehai are sustained by heterotrophic decomposition of organic matter, presumably from



the aquaculture sources and also from eutrophication in the water column. During the field campaigns it was found out that the bottom sediments were dark colored and odorous with smell of H₂S, and a black fluffy layer extended several centimeters above sea floor. The extensive breakdown of organic matter in the Laoyehai is fueled by the influx of wastes from shrimp ponds and caged fish culture (e.g., feeding materials); samples from the shrimp pond outflow had high levels of NH_4^+ relative to NO_3^- and a skewed DIN/DIP molar ratio in comparison to riverine inflows as shown in Table 3. Based on the knowledge obtained from Laoyehai, a conceptual model is developed that links the impact of marine aquaculture as driving force and the responses of ecosystem, e.g., nutrient dynamics in coastal lagoon. This highlights that oxygen depletion and the accumulation of reduced nitrogen (e.g., NH₄⁺) are consequences of various human activities (e.g., reduction of habitats) in combination, and the ecosystem undergoes deterioration. The consequences of this driving force-response interaction loop include unsustainability of economic income and feedbacks to the atmosphere (e.g., emission of greenhouse gases), although the release of gas species and biodiversity aspects were not measured right now (Fig. 9). For instance, in the third week of September 2003, there was a serious fish kill event in the Laoyehai, which resulted in the loss of 0.5×10^6 kg of culture products within 3 days, corresponding to an economic value to the farmers of 20×10^6 yuan (http://www.china.com. RMB cn/chinese/huanjing/4072526.htm).

Given the geographic features and poor water exchange of the Laoyehai, the numerical simulations showed that water flushing process is relatively slow, which highlights that hypoxic water with relative high concentration of nutrients can be retained within the system for a time scale of approximately 100 days or even longer. Based on numerical experiment using passive tracers, the simulation results also illustrate that only one-third of the water in the channel close to the inlet of Laoyehai can be flushed within 100–120 days (HNMDDI 2011).

The vertical density gradient in the Laoyehai is weak relative to other coastal hypoxic ecosystems, such as riverine plume in an estuary (Zhu et al. 2011). In the Laoyehai, the oxygen-depleted water can reach up to the surface, and despite being exposed to the atmosphere, DO concentration in surface waters can be as low as $30-60 \mu$ M.

In summary, the evidence, including that NH₄⁺ dominates DIN and that there is limited exchange with offshore waters, the hypoxic nature of near-bottom samples, and the skewed DIN/DIP molar ratio etc., together underline that DNRA is an important mechanism regulating nitrogen cycling in the Laoyehai, but its relation to the fate of DO remains to be investigated. Groundwater discharge is also an important source of some nutrients into the lagoon; it has a lower DIN/DIP ratio relative to that of coastal waters further offshore, indicating the importance of nitrate reduction and phosphorus input (Li et al. 2014a). Moreover, the high concentrations of sulfides in bottom sediments and near-bottom waters in the Laoyehai can inhibit alternative pathways of nitrogen cycling, including anammox, in coastal environments as illustrated by Jensen et al. (2008, 2009). Again, sulfide is toxic to multicellular organisms and reduces survival times under hypoxia/anoxic conditions, which is presumably one of the reasons for the massive fish kills of Laoyehai.

Perturbations of aquaculture on the coastal ecosystems

The results of Laoyehai Project demonstrates that aquaculture can have a dramatic impact on the structure and function of marine ecosystems. For instance, by introducing cultured species in the coastal environment, aquaculture squeezes the habitats of natural species, and the species under culture occupy the niche of original and natural organisms and modify the energy and material flow of ecosystem, with feedbacks to the biogeochemical cycle. Although eutrophication and hypoxia can be natural in coastal environment (Diaz and Rosenberg 2008) and phytoplankton dominated lagoons tend to have a bottom water-oxygen depletion problem, relative to waters prevailed by macrophytes (Yamamuro 2012), aquaculture does impose an extra-forcing on the already over-stressed ecosystems by other human activities.

In Fig. 10, we compare Laoyehai with a few other coastal bays and lagoons under varies but different human being perturbations (e.g., mariculture), including Xiaohai (lagoon), Xiangshangang (bay), and Sanggou Bay from China; Sacca di Goro Lagoon and Lesina Lagoon from Italy; Louro Lagoon from Spain; and Gamak Bay from South Korea (Daniele et al. 2006; Vignes et al. 2009; Hwang et al. 2010; Cobelo-García



Fig. 10 Comparison of Laoyehai with a few other coastal bays and/or lagoons, including Xiaohai (XH), Xiangshangang, and Sanggou Bay (SGW) from China; Sacca di Goro Lagoon and Lesina Lagoon from Italy; Louro Lagoon from Spain; and Gamak Bay from South Korea (Daniele et al. 2006; Vignes et al., 2008; Hwang et al. 2010; Cobelo-García et al. 2012; Jiang et al. 2012; Jiang et al. 2015; Lee et al. 2016; Mahmood et al. 2016). a The results show the measured low DO increase with higher outlet to length ratio with exception of Xiaohai (XH) and Lesina (LS). b The DO tends to decrease with higher ratio of surface area occupied by aquaculture with exception of Sanggou Bay (SGW). For the purpose of management, DO unit is given as mg/l rather than µM in this figure

et al. 2012; Jiang et al. 2012; Jiang et al. 2015; Lee et al. 2016; Mahmood et al. 2016). In the comparison, we used the ratio of outlet width and length of water body as an index of natural quality, because the narrow mouth and extended length of a bay and/or lagoon usually induce slow exchange with offshore waters and an increase in residence time of natural system. Moreover, we selected the ratio of cultured area to total surface of bay and/or lagoon to distinguish the perturbation of aquaculture on the ecosystem of concern. Laoyehai, Xiaohai, Xiangshangang, and Sacca di Goro Lagoon have aquaculture of shrimp ponds, fish in cages, and bivalves etc.; Sanggou Bay is characterized by implementation of Integrated multi-trophic aquaculture (IMTA) with combination of algae and mollusk (e.g., oysters and abalone); Lesina Lagoon is free of aquaculture, but receives waste water from nearby fish farms as well as sewage from nearby cities; and Louro Lagoon is basically not affected by any activities of aquaculture to a considerable level (cf. Cobelo-García et al. 2012).

As shown in Fig. 10, the observed low DO levels decrease statistically with the reduction of ratio between outlet width and length of bays and/or lagoons, indicating that restriction of water exchange with offshore environment tends to promote the possibility of hypoxia by increasing the water residence time, even though the system is not affected by a large-scale aquaculture. Xiaohai (XH) and Lesina (LS) are exceptional because these two lagoons are both very shallow. Lesina Lagoon is all less than 1.5 m, and more than 90% of surface area of Xiaohai is less than 1.0-1.5 m, with a tidal range of 0.7-2.0 m most part of the bay is exposed to sunshine in low tide, particularly in spring tide period.

However, when considering the perturbations of aquaculture on coastal environment, the observed low DO values tend to decrease with higher proportion of aquaculture area to the surface of semi-enclosed bays and/or lagoons, and it can be expected that hypoxic phenomena take place when the semi-enclosed waters has a nature of restricted exchange and aquaculture is dominated by the fish in cages and shrimp ponds. In this case, Sanggou Bay (SGW) from North China is exceptional that it has an open nature and is more or less normoxic, because of the implementation of IMTA, where the culture of mollusk and fish is mixed with brown algae in the water column and sea cucumber in seabed, which promotes the economic benefits to the local fishing community and improves the water quality (e.g., DO) as well (Fang et al. 2016).

In summary, the comparison between Laoyehai and other coastal waters affected by marine aquaculture reveals that the coastal environment with restricted connection to the offshore waters are more prone to the negative impacts of aquaculture; hence, eutrophication in surface and hypoxia in near-bottom waters are more easily to develop in such systems, because the ability of recovery and the so-called environmental capacity depend largely on the ease of exchange with offshore waters. From this perspective, marine aquaculture of Laoyehai is not sustainable and neither healthy in terms of ecosystem function, and other possibilities have to be explored, such as adaption to RAS and IMTA (Martins et al. 2010; Fang et al. 2016).

Concluding remarks and recommendations

We report here a study of the nutrient dynamics and hypoxic nature of a coastal lagoon in the tropical South China Sea, where the marine ecosystem is under the influence of a monsoonal climate and the biogeochemical cycles are affected by anthropogenic perturbations, notably the reclamation of wetlands and pollution from marine aquaculture.

It was found that elevated concentration of reduced form of nutrient species (e.g., NH4⁺) and a high DIP/DIN ratio in the Laoyehai coincided with hypoxia in the near-bottom waters. Heterotrophic degradation of organic matter and mineralization can be fueled by the release of waste materials from shrimp ponds and fish culture cages; the narrow water channel and the micro-tidal nature of the Laoyehai leads to a limited exchange with the open South China Sea, which results in a prolonged water flushing time in the scale of hundred days. It appears that the hypoxic character of waters inside the Laoyehai is tending to deteriorate. For instance, the observed minimum DO value in near-bottom waters was 20 µM in 2008-2009, and this decreased to 3- $4 \mu M$ in 2011, although such phenomena can be episodic and/or ephemeral. In comparison, in 2005-2006 DO concentration in the Laoyehai was reported at 5.42-7.66 mg/l (i.e., 170-240 µM) and the pH was 8.06-8.36, and no hypoxia had been found (SCSIO 2006).

So far, it is not clear when the lagoon began to be affected by hypoxia and/or anoxia, nor the depletion of DO could be totally alleviated again in the past decades, but anthropogenic perturbations have accelerated the negative impacts of oxygen depletion in this water body. Given that fish kills reported in the public media have been related to the hypoxic and/or anoxic conditions in the Laoyehai since the late 1990s, the results of 2005– 2006 may indicate that waters with low DO might not be necessarily sampled and/or that oxygen depletion in this lagoon was not persistent in that time.

The Laoyehai has a narrow water channel and there is only slow water exchange with the open South China Sea, which suggests that it may be ill-advised to promote large-scale marine aquaculture in this area, either shrimp ponds or caged fish or both, because of the multiple and complicated responses of ecosystem (Fig. 9). Historically, the natural wetlands (e.g., mangroves) would have helped to intercept and retain pollutants from the land sources and served as a nursery for marine fauna having economic values. However, the reclamation of wetlands for shrimp ponds has destroyed this ecosystem and induced loss of economic values in the vision of sustainability. The high-density fish culture in cages has changed the water flow and circulation in this long and narrow water body, further reducing the already sluggish water exchange process with open coastal waters. Moreover, the wastes released from shrimp ponds and fish cages into the Laoyehai have compounded deterioration of the water quality, which is already eutrophic because of land-source pollutants and groundwater discharges. All these problems need to be taken into account if the strategy of environmental remediation and ecosystem-based management, aimed at sustainability of adjacent human society, is to be successful in the Laoyehai as well as other but coastal systems with similar nature. Finally, the comparison of Laoyehai to a few other coastal systems worldwide reveals that the bays and lagoons with a nature of low outlet to length ratio are more prone to the perturbations of aquaculture with consequences of eutrophication in surface and hypoxia/anoxia in near-bottom waters.

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Appendix 1. Numerical model simulation for the Laoyehai

The 3-D ELCIRC hydrodynamic model solves for the free surface elevation, water velocity, and distribution of hydrographic parameters (e.g., salinity and temperature), using a set of six hydrostatic equations based on the Boussinesq approximation, which represent mass conservation in both 3-D and depth-integrated forms (Zhang and Baptista 2004). The governing equations are given below (A1–A4):

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} + \frac{\partial w}{\partial z} = 0 \tag{A1}$$

$$\frac{\partial \eta}{\partial t} + \frac{\partial}{\partial x} \int_{H_{R-\eta}}^{H_{R+\eta}} u dz + \frac{\partial}{\partial y} \int_{H_{R-\eta}}^{H_{R+\eta}} u dz = 0$$
 (A2)

$$\frac{Du}{Dt} = fv - \frac{\partial}{\partial x} \left\{ g(\eta - \alpha \psi) + \frac{P_a}{\rho_0} \right\} - \frac{g}{\rho_0} \int_z^{H_{R+\eta}} \frac{\partial \rho}{\partial x} dz
+ \frac{\partial}{\partial z} \left(K_{mv} \frac{\partial u}{\partial z} \right) + F_{mx}$$
(A3)

$$\frac{Dv}{Dt} = -fu - \frac{\partial}{\partial y} \left\{ g(n - \alpha \psi) + \frac{P_a}{\rho_0} \right\} - \frac{g}{\rho_0} \int_{z}^{H_{R+\eta}} \frac{\partial \rho}{\partial y} dz + \frac{\partial}{\partial z} \left(K_{mv} \frac{\partial v}{\partial z} \right) + F_{my}$$
(A4)

where (x, y) are the horizontal Cartesian coordinates (m), ϕ , λ are the latitude and longitude, z is the vertical coordinate with upward being positive (m), t is time (s), H_R is the z-coordinate at the reference level (i.e., mean sea level, MSL), $\eta(x, y, t)$ is the free surface elevation (m), and h(x, y) is the bathymetric depth (m); $\vec{u}(\vec{x}, t)$ is the water velocity at $\overrightarrow{x}(x, y, z)$, having Cartesian components (u, v, and w), in m/s; f is the Coriolis factor (per s), G is the acceleration under gravity (m/s), $\psi(\phi, \lambda)$ is the tidal potential (m), and α is the effective Earth elasticity factor; $\rho(\vec{x}, t)$ is the water density, for which the default reference value ρ_0 is 1025 kg/m³; and $P_a(x, x)$ y, t) is the atmospheric pressure at the free surface (N/m), K_{mv} is the vertical eddy viscosity (m²/s), and F_{mx} , F_{my} is the horizontal diffusion for momentum and transport equations.

The differential system for the six primary variables (η, u, v, w, T, S) in Eqs. A1, A2, A3, and A4 is closed with the equation of state, the definition of the tidal potential and the Coriolis factor, parameterizations for vertical mixing, and initial and boundary conditions. Initial conditions require problem-dependent specification of pre-simulation fields for all primary variables and for any turbulence parameters required by the vertical mixing parameterization (Zhang and Baptista 2004).

In implementation, the model was firstly validated by time-series data of tidal level, current and hydrographic parameters (e.g., salinity and temperature) from anchor stations of 26 h for both spring and neap tides and the simulation results were compared to the section profiles of hydrography in different seasons (cf. HNMDDI 2011). Then the numerical simulations were applied to understand the hydrodynamic processes of Laoyehai using Lagrangian particles as passive tracer.

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