Graphical Abstract:

During the past 50 years, the **high-intensity** land-based pollutant discharge and land reclamation have been identified as the key control indicators and have a joint contribution on the significant variation of water quality of Jiaozhou Bay, showing synergistic **effect on water quality**.

Note: The C_{DIN} represents dissolved inorganic nitrogen concentration; S_{JZB} is the water area of Jiaozhou Bay (JZB); F_{TN} is the terrestrial total nitrogen flux.

Assessment of long-term water quality variation affected by high-intensity land-based inputs and land reclamation in Jiaozhou Bay, China

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Abstract

With the interactive and accumulative effects of multiple pressures, the dissolved inorganic nitrogen concentration (C_{DIN}) is increasing considerably and is regarded as the dominant chemical pollutant in Jiaozhou Bay (JZB). To efficiently prevent water quality (WQ) deterioration, the relationships among the C_{DIN} in JZB with anthropogenic pressure indicators (APIs) and coastal carrying indicators (CCIs) were identified using long-term series data (1962-2012). First, the control indicators (CIs) were obtained by the application of a conceptual model analysis, and potential sources were identified by a factor analysis. Second, the key CIs were identified by a regression analysis. Finally, the effects of the key CIs on WQ were quantitatively assessed by numerical model experiments. The results showed that the CIs for the C_{DIN} variation contained both APIs, including terrigenous total nitrogen flux (F_{TN}) discharged into JZB, river runoff, sea area of reclamation, and the CCIs, including the water area of JZB (S_{JZB}), chlorophyll-a, dissolved oxygen, pH, salinity and seawater temperature. The APIs on WQ variation were greater than the CCIs. The F_{TN} and S_{JZB} were identified as the key CIs responsible for the variation in the C_{DIN} . The WQ simulation results showed that the increasing trend of the C_{DIN} was caused by the increase in the F_{TN} and the decrease in the S_{JZB} , and the combined effects induced by the two key CIs on the C_{DIN} variation was as high as 79%, higher their separate effects, indicating a synergistic interaction of the two key CIs on the WQ of JZB during the last 50 years. These results illustrated that both terrigenous nitrogen pollutant load control systems and a scientific plan for coastline utilization should be implemented to effectively improve the WQ of JZB.

Keywords: JZB; water quality; land reclamation; land-based pollution; control indicators

1. Introduction

Jiaozhou Bay (JZB), called the mother bay of Qingdao City, is an important spatial land ecological-environmental carrier for the sustainable social-economic development of Qingdao city. More than 60% of land-based pollutants from Qingdao are discharged into JZB (Figure 1) (Lu et al., 2016; Su et al., 2014; Wang et al., 2006). Since the 1960s, especially since the beginning of the 1980s, with the socio-economic development of Qingdao City, human activities such as land-based source pollution, land reclamation and many other indicators showed single, cumulative and synergistic processes. These processes resulted in both the recession of marine resources and the environmental carrying capacity and the deterioration of the water quality (WQ) in JZB, which in turn posed a challenge for the sustainable socio-economic development of Qingdao (Gao et al., 2014; Han et al., 2011; Liang et al., 2015; Shi et al., 2011). Currently, the concentration of dissolved inorganic nitrogen (C_{DIN}) is greatly increasing, and the water area ratio of JZB with WQ exceeding the second grade of the Chinese national seawater quality standard is maintained at approximately 50%; therefore, the improvement of the environmental quality of JZB is centred on improving the WQ (Dong et al., 2010; Lu et al., 2016; Ocean and Fishery Administration of Qingdao, 2015). From the middle of the 2000s, mitigating measures, such as the restoration of adjacent rivers, coastline protection, and moving offshore industry out of the bay area, were implemented in JZB watersheds (Qingdao Environmental Protection Agency, 2012; Qingdao Municipal Government, 2013). However, the WQ of JZB continues to deteriorate (Dong et al., 2010; Liang et al., 2015; Ocean and Fishery Department of Qingdao City, 2015; Qian et al., 2009). The ratio of the areas with the second grade and the inferior second grade of the seawater quality standard increased by approximately 5%, indicating that the deterioration of WQ of JZB is still serious and difficult to restore. An important reason for this difficulty is that the relationships among the WQ and the anthropogenic pressure indicators (APIs) and the coastal carrying indicators (CCIs) have not yet been clearly identified.

Many previous studies explored the relationships of WQ with APIs and CCIs, including the identification of control indicators (CIs) and the assessment of the effect of the key CIs on WQ (Borja et al., 2006; Borja et al., 2016; Jennerjahn and Mitchell, 2013; Skerratt et al., 2013; Wild-Allen et al., 2010; Wu and Wang, 2007; Yuan et al., 2016; Zhou et al., 2007a). However, given the range of factors that can influence WQ

dynamics, and the fact that these have impacts over a range of time scales, some discrete and some continuous WQ dynamics may only be fully investigated when long-term time-series data are available. A long-term investigation may provide a realistic means of establishing variable interactions in ecosystem studies (Lau and Lane, 2002). To investigate the APIs and CIs and their interactions with regard to WQ, a factor analysis (FA), a linear regression analysis (LRA), a cluster analysis and other multivariate statistical analysis methods are widely used to investigate the relationships between WQ and CIs (Cormier et al., 2000; Petersen et al., 2001; Simeonov et al., 2003; Wu et al., 2009). However, the applications of FA in the identification of potential sources based on a conceptual model analysis of selecting indicators has not been conducted, so the indicator selection process is subject to arbitrary decisions (Kim et al., 2015; Niemeijer and de Groot, 2008; Petersen et al., 2001; Simeonov et al., 2003; Wu et al., 2009). In addition, the FA includes not only hydrodynamic indicators but also land-based pollutant discharge of the APIs, resulting in the significant distinction of identifying the potential sources, even contradictory sources (Kano and Harada, 2000; Wu et al., 2009; Zhou et al., 2007b). The key CIs were selected by a LRA on the relationship between the CIs and WQ (Bu et al., 2014; Cormier et al., 2000). The single effects of key CIs on WQ have been quantitatively assessed in coastal waters by a coastal model that coupled three-dimensional hydrodynamic and biogeochemical processes under different scenarios (Lee et al., 2008; Li et al., 2015b; Skerratt et al., 2013; Yuan et al., 2016). Although the previous studies have focused on individual indicators of the effect on WQ in a simulation process, little is known about the effect on WQ induced by multiple key CIs based on variation over a long period of time. In particular, the combined WQ effect of indicators remains unclear in practice. The quantitative effect of key CIs on WQ can respond to the integrative, accumulative and synergistic characteristics in practical WQ management on the basis of identifying key CIs (Nixon and Kristensen, 2003; WFD CIS, 2003). However, there is a gap that is not only reflected in the identification of the CIs and the quantitative assessment of the effect of key CIs on the WQ but also behaves in a causal chain and the inter-relations of indicators in the conceptual model (Borja et al., 2006; Cormier et al., 2000; Niemeijer and de Groot, 2008). The identification of key CIs not only considers the causal chain and a variation analysis but also the independent test of the data of the indicators in the conceptual model. Despite a large body of studies focused on the identification of key CIs and the quantitative impact of single indicators on the WQ of specific compartments (Jia, 2006; Lau and Lane., 2002; Li et al., 2015b; Yuan et al., 2016), the methods of the assessment of integrative, accumulative and synergistic effects of multiple key CIs are poorly studied global issues.

Several studies have focused individually on the selection of indicators or the identification of key CIs, as well as the assessment of individual key CIs on the effect on WQ using one of a conceptual model analysis, FA, LRA and numerical modelling method; however, they were not linked to the combined key CIs on the WQ effect,

leading to considerable uncertainty in identifying and assessing the key CIs that affect WQ. In fact, WQ was controlled by many indicators, which were related and interacted with one another. The advantage of the present study was that arbitrary decisions in indicator selection were avoided to fully investigate the effect on WQ induced by multiple key CIs over long-term variations, and the integration method was adopted to completely understand the effects on WQ in this study. With this in mind, to efficiently promote an improvement in WQ in JZB and provide an approach that can be adapted globally to assess the long-term effects of indicators on WQ, the objective of this study was to investigate the effect of a controlling mechanism on WQ based on the relationships of the CIs using a conceptual model analysis. Furthermore, the study focused on the identification of the CIs and key CIs using a statistical analysis as well as a quantitative assessment of the impact of key CIs on WQ using modelling.

2. Materials and methods

2.1 Dataset

2.1.1 Data sources

The C_{DIN} data were from Wang et al. (2006), Qian et al. (2009) and the Qingdao Environment Monitoring Centre (QDEMC) (2010-2012). The terrigenous total nitrogen flux (F_{TN}) data were from the Qingdao Municipal Statistics Bureau (2013) and Liang et al. (2015). The water area of JZB (S_{JZB}) and sea area of reclamation data were from Zhou et al. (2010) and Ma et al. (2014). The river runoff data were from Sheng et al. (2014). The chlorophyll-a data were from Guo and Yang (1992), Sun et al. (2011a) and the QDEMC (1991-2012). The pH data were from Diao (1984) and the QDEMC (1991-2012). The dissolved oxygen data were from the Aquatic Chemistry Group Institute of Oceanology Academia Sinica (1982), Compiling Committee of Records of China Bays (1993), Zhao (2002) and the QDEMC (1991-2012). The water temperature data were obtained from the Aquatic Chemistry Group Institute of Oceanology Academia Sinica (1982), Diao (1984), Sun et al. (2011a) and the QDEMC (1991-2012). The salinity data were obtained from Diao (1984), Sun et al. (2011b) and the QDEMC (1991-2012).

2.1.2 Data treatment

To concentrate on the long-term effect on WQ, the annual mean data values, based on pooled samples, were determined in the analysis (Li et al., 2015a; McQuatters-Gollop et al., 2007). The time interval between each indicator must

remain consistent with the WQ throughout the time-series due to the presence of many indicators of long-term variation in the study (Halliday et al., 2012; Li et al., 2015a). However, on some occasions, a value for a particular indicator was not obtainable (Lau and Lane, 2002). To acquire the complete indicator dataset that covered past decades, the missing values for the occasional years had to be estimated to fill gaps in the data using statistical methods. To ensure the data quality of the indicators in filling gaps, the LRA was introduced to test the long-term trend of the observed data (Quinn and Keough, 2002). The results showed that the inter-annual patterns of the S_{JZB} , salinity, temperature, sea area of reclamation, chlorophyll-a, dissolved oxygen and the F_{TN} had significant trends ($p < 0.05$). Therefore, the linear interpolation between adjoining observed data was adopted for the S_{JZB} , the sea area of reclamation and the F_{TN} (Little and Rubin, 2002). However, the missing data values of the F_{TN} , chlorophyll-a and pH were estimated by applying the analogy method considering the properties of indicators in the 1960s (Harding and Perry, 1997; Little and Rubin, 2002). At the beginning of the 1960s, the F_{TN} was estimated by the pollutant export coefficient (Wang et al., 2006). In the early 1960s, the initial chlorophyll-a value referred to the value of the same period in the North Sea, which was characterized by similar coastal environmental conditions (McQuatters-Gollop et al., 2007). The pH value in the 1960s referred to values from field data of the adjacent Yellow Sea (Science and Technology Committee of the People's Republic of China, 1963).

With regard to the long-term data and data of different sources, an explanation of sources of error was needed. First, to avoid sources of error in the method of monitoring seawater quality and coastal carrying indicators, the analyses were in accordance with the marine investigation criteria of China and aimed to ensure the consistency of methods as far as possible. Second, for the accuracy of the datasets of all historical indicators, we cited references that had confirmed the comparability of the long-term data (Wang et al., 2006; Liang et al., 2015; Ma et al., 2014). Third, we applied the annual mean value of the long-term data in this investigation, which had the same time series trend and annual variation pattern as in previous studies (Dong et al., 2010; Shen et al., 2001), indicating the possibility of combining the data from different times and the small error in data treatment. Therefore, the aforementioned information regarding the annual mean data of WQ and indicators obtained in the JZB study area were deemed relatively accurate and reliable.

2.2 Methods

2.2.1 Selection and relationships of indicators by conceptual model analysis

Understanding the response of marine systems to human activities and the

resultant pressures requires a good conceptual basis that links the causes and consequences of change (Adams, 2005; Borja et al., 2016; Crain et al., 2008). The responses of seawater quality to indicators are the integrated result of both APIs and CCIs (Borja et al., 2006; Elliott and Quintino, 2007; Reimann and de Caritat, 2005). For efficient management of WQ, the indicators can be defined at two levels according to the degree of impact on WQ: control and key control. The CIs may have effects on WQ in theory, but there is no way to ascertain the qualitative and quantitative effects of the indicators on WQ. The key CIs are quantitatively associated with WQ by a statistical analysis, and their effects on WQ are remarkable. The inter-relation between the APIs and CCIs is interactive and integrative, which is typically presented by the causal chain and inter-relation of indicators (Glibert et al., 2010; Niemeijer and de Groot, 2008). Thus, the relationships between WQ and the indicators should include the identification of CIs and the quantitative assessment of the effects of key CIs on WQ, based on the measurable and available data (Cormier et al., 2000; WFD CIS, 2003). Due to the characteristics of pollutants, the factors that affect the pollutant concentration in seawater are distinct and can be classified as several types of indicators by the causal chain and inter-relation in the conceptual model (Figure 2) (Borja et al., 2006; Cormier et al., 2000). In the conceptual model, the API category includes the groups of the land-based load and the land-ocean boundary. The CCIs category contains the hydrodynamics and biogeochemical process groups. Each group consists of various subgroups, which consist of several types of indicators (Borja, 2005; WFD CIS, 2003). In accordance with the conceptual model, the inter-relation of indicators is the measurable potential CIs at the same level (WFD CIS, 2000)

2.2.2 Annual pattern of change of WQ and identification of potential sources by FA

To examine the interactions of the APIs and CCIs and to identify potential effects on WQ, a FA was applied based on the long-term dataset (Lau and Lane, 2002; Simeonov et al., 2003). Before performing the FA, original indicators data were z-scale standardized (the mean and variance were set to 0 and 1, respectively) to minimize the effects of different units and the variance of variables and to render the data dimensionless (Singh et al. 2004; Zhou et al. 2007a). The Kaiser-Meyer-Olkin and Bartlett's sphericity tests were conducted on the parameter correlation matrix to examine the validity of the FA (Ouyang et al., 2006; Zhou et al., 2007b). To assess the role of indicators (X_j), the FA was used for linear combinations of the original variables, as one subset of FA extracts eigenvalues (eigenvalues > 1) from the covariance matrix of original variables to produce new orthogonal variables (Simeonov et al., 2003). The common F_i information of the most meaningful parameters describes a whole dataset, allowing a reduction in data with minimal loss

of the original information. Furthermore, based on the community of indicators (H^2) at F_i , namely, the factor loading (ϕ_{ij}) square; the absolute value stands for the correlation between F_i and X_j . The sum is the comprehensive loading factor, which assesses the potential relative effects of indicators of WQ (Z_j) on F_i (Lau and Lane, 2002; Reymont and Joreskog, 1993). The potential sources of impacts on WQ were identified by the factor loading in the FA. SPSS 20 (Statistical Package for the Social Sciences Inc. IBM) was used to perform the FA, and the significant P was set to 0.05.

$$H_{ij}^2 = \sum_{j=1}^m \phi_{ij}^2 \quad (2-1)$$

In (2-1), i indicates the number of F , j is the number of control indicators, H_{ij}^2 is the sum of loading of F_i , and ϕ_{ij} is the loading of the j indicators in the F_i .

2.2.3 Identification of key CIs by LRA

To discriminate the key CIs, the regression techniques were used to quantify the relationships between WQ and the CIs. A simple LRA was one of the most widely applied statistical techniques (Cormier et al., 2000; Quinn and Keough, 2002; Valipour, 2014; Valipour and Eslamian, 2014; Valipour, 2015a,b). The LRA described the linear relationship between WQ and the indicators and determined how much of the variation in WQ can be explained by the linear relationship with indicators and how much of this variation remains unexplained. The simple linear regression model is:

$$Y_i = \beta_0 + \beta_1 X_i \quad (2-2)$$

Where Y_i is the C_{DIN} , and X_i is the indicators.

2.2.4 Quantitative assessment effect of key CIs on WQ

To examine and quantify the effects of the single key CIs or joint key CIs on WQ, a series of scenarios were designed based on the identification of key CIs. The details of the model scenarios are shown in Table 1. For one type of scenario, a single key CIs was changed according to its natural variation, while the other key CIs were kept constant in the 1960s. For the other types of scenarios, the WQ effects of different scenarios were assessed by numerical experiments based on the WQ model of JZB. The biogeochemical process model (Li et al., 2007) embedded the hydrodynamic module in the Finite Volume Coastal Ocean Model framework (Gao et al., 2014), which could create a three-dimensional hydrodynamic and biogeochemical model that could run a simulation under a variable boundary. Thus, the biogeochemical module embedded in the modelling framework could simulate the land-based source inputs and the hydrodynamic changes. The tidal elevations derived from the model agreed well with the observations (Gao et al., 2014). The coefficients used for the

biogeochemical processes have been established by field measurements and experiments in JZB (Li et al., 2007). Furthermore, the relative standard deviation between the simulated and observed values was 44.2%. The C_{DIN} spatial distribution between the simulated and observed values was assessed by a kappa coefficient (Fleiss, 1994; Holguin-Gonzalez et al., 2013), which typically is used to measure the spatial distribution relationship between two parameters (Holguin-Gonzalez et al., 2013). Agreements were classified into weak, fair, moderate, good and very good, reflecting the kappa coefficient values of <0.20, 0.21–0.40, 0.41–0.60, 0.61–0.80 and 0.81–1.0, respectively (Brennan and Prediger, 1981). The average kappa coefficient between the simulated and observed values was 0.52. The above analysis indicates that the model simulation results are in agreement with the monitoring results for the temporal and spatial distribution (Brennan and Prediger, 1981; Couto, 2003; Holguin-Gonzalez et al., 2013). The result suggests that the JZB WQ model, coupled with three-dimensional hydrodynamic and biogeochemical processes, could be applied to quantitatively assess the effect of key CIs on the C_{DIN} .

3. Results

3.1 The relationships of indicators on WQ by a conceptual model analysis

According to the analysis results of land-ocean interactions (Figure 2), in the APIs category in the conceptual model of JZB, the F_{TN} discharge may be regarded as one of the main indicators in the land-based pollution subgroup, and the coastline and freshwater discharge into JZB are categorized into the mixed boundary subgroup (Liang et al., 2015; Wang et al., 2006; WFD CIS, 2003). Indeed, the sediment input and river diversions did not change significantly compared to typology, and the water input of the sewage outfall was far less than the river runoff (Sheng et al., 2014; Zhou et al., 2010). The mixed boundary subgroup indicators contain the coastline and freshwater discharge into JZB. The significant changes in the hydrodynamic transport group indicators could be expressed by the S_{JZB} (Shi et al., 2011; Zhou et al., 2010). It is most likely that the types of biological and environmental effects played important roles in the biogeochemical group, the water, suspended particles and the bay bottom sediment media groups, which were far lower than the former (Liu et al., 2010; WFD, 2000; WFD CIS, 2003). According to the analysis results, the potential CIs on the C_{DIN} are the sea area of reclamation, river runoff, F_{TN} , S_{JZB} , dissolved oxygen, pH, salinity, chlorophyll-a and temperature. The land reclamation represents the coastal mixed boundary. The groups of the JZB mixed boundary and biological media are indicated by the S_{JZB} and chlorophyll-a, respectively. Dissolved oxygen, pH, salinity and temperature interactively represent the environmental effects team (Liu et al.,

2010; WFD CIS, 2003). In fact, salinity and temperature are also important indicators of the density current (Lynne et al., 2011). These nine potential CIs are not only at the same level in the conceptual model but also measurable and available. However, the sea area of reclamation is directly related to the S_{JZB} in the conceptual model, and the available data of S_{JZB} are selected as indicators of coastline change, considering the independent assessment of pressures and impact relationships (WFD CIS, 2003). However, the indicators have distinctive differences that do not fully correspond to the time, and data values between the C_{DIN} and the nine potential CIs in the long-term monitoring system are missing. With respect to the time scale, reclamation and large-scale anthropogenic activities began in the early 1960s, and the early 1960s was selected as the starting year in the identification of CIs.

3.2 Annual pattern of change and identification of the potential sources of WQ

During the past 50 years, the variation of the APIs and CCIs showed different annual patterns of change (Figure 3). On one hand, the APIs varied greatly, and the F_{TN} increased significantly by approximately 10 times up until the 2010s ($p < 0.05$). In contrast, river runoff declined and then increased, but no significant change trend was tested ($p > 0.05$), and 2012 had approximately 60% of the freshwater discharge compared to that of the 1960s (Sheng et al., 2014). Due to the land reclamation around JZB, there was an approximately 178 km² of water area reclaimed compared with 1958 (Ma et al., 2014). The annual variations of the CCIs were also diverse. The S_{JZB} dropped from 448 km² to 344 km² during the past few decades, with a significant declining trend ($p < 0.05$), which was much higher than the 4% decrease from the 1930s to the 1960s (Ma et al., 2014). The annual trend of chlorophyll-a showed that it significantly increased ($p < 0.05$), with a peak value of 7.4 µg/L during the 2000s. For the indicators of types of environmental effects, the mean value of temperature significantly increased in the early 1960s ($p < 0.05$), while salinity significantly decreased ($p < 0.05$), with a maximum of 32.4 in the 2000s. The value of pH decreased gradually, with an average of 8.1, showing no significant variation ($p > 0.05$). The dissolved oxygen concentration dropped significantly ($p < 0.05$), and its average value was 7.7 mg/L during the past 50 years.

A FA was applied to the long-term dataset to identify the potential sources of WQ variation (Lau and Lane, 2002; Zhou et al., 2007a). The Kaiser-Meyer-Olkin result for the standardized data was 0.6, and the result for Bartlett's sphericity was 63.8 ($p < 0.05$), indicating that FA may be useful in providing significant reductions in dimensionality (Zhou et al., 2007a; Olsen et al., 2012). Based on the eigenvalues (eigenvalues > 1), two main factors were extracted (Al-Mutairi et al., 2014; Ouyang et al., 2006; Simeonov et al., 2003), and their cumulative contribution to the variance accounted for 58.3%. Factor 1 (F_1) of the variance contribution was 36.0%. The F_{TN} ,

S_{JZB} , dissolved oxygen and pH of the loading values were greater than 0.5 (Figure 4). The F_{TN} and S_{JZB} loading values were greater than 0.9, indicating that the APIs and CCIs played important roles in WQ. Factor 2 (F_2) of the variance contribution was 22.3%, and salinity and river runoff of the loading values of the indicators were greater than 0.5, primarily reflecting the contribution of CCIs. The average loadings of the APIs, including the F_{TN} , S_{JZB} and river runoff, were much greater than those of the CCIs, indicating that anthropogenic influence played a predominant role in WQ variation.

3.3 Identification of key CIs by regression analysis

In terms of the LRA between WQ and indicators (Figure 5), the F_{TN} was significantly positively correlated to the C_{DIN} ($p < 0.05$), and the R^2 value (0.543) indicated that F_{TN} could explain 54.3% of the total variation in the C_{DIN} . The S_{JZB} was significantly negatively correlated to the C_{DIN} ($p < 0.05$), explaining 52.7% of the total variation in the C_{DIN} . In addition, the CCIs, namely, dissolved oxygen and chlorophyll-a, were also significantly negatively associated with the C_{DIN} , explaining 28.2% and 26.6% of the total variation in the C_{DIN} , respectively. However, the pH, temperature, salinity and river runoff were not significantly related to the C_{DIN} , showing no significant linear relationship with the C_{DIN} . Based on the R^2 value, the order of CIs was $F_{TN} > S_{JZB} > \text{dissolved oxygen} > \text{chlorophyll-a} > \text{pH} > \text{temperature} > \text{salinity} > \text{river runoff}$. The mean R^2 of the F_{TN} and S_{JZB} was 2.1 times higher than average values of other indicators, showing a closer association with the C_{DIN} , which meant they were the key CIs contributing to the variation in the C_{DIN} .

3.4 Validation and assessment effects of key CIs on WQ

Three scenarios of model experiments were conducted to reproduce and study the impacts of F_{TN} input and land reclamation in JZB on the variation in the C_{DIN} to explore the effects of the key CIs on the C_{DIN} starting in the early 1960s (Table 1). During the past 50 years, the C_{DIN} of JZB showed a yearly increasing trend, which was from far lower than the first grade of the WQ standard in the early 1960s to the third grade of the WQ standard in the 2010s, showing a degradation in WQ (Figure 6). The annual mean C_{DIN} drastically grew by approximately 4 times from the 1960s to the 1980s. Compared with the C_{DIN} level in the 1960s, the C_{DIN} was 8 times higher in the 1990s. Furthermore, the average C_{DIN} increased approximately 13 times in the 2000s compared with that in the 1960s. In total, the annual mean C_{DIN} has increased sharply by approximately 14 times from the 1960s to the 2010s. Thus, since the beginning of the 1960s, the C_{DIN} in JZB has been increasing with the decrease of the S_{JZB} under the condition of terrestrial F_{TN} input in the 1960s. In contrast, the C_{DIN} in JZB had been increasing with the sharp increase in the F_{TN} input under the condition of the S_{JZB} in the 1960s. Furthermore, the C_{DIN} in JZB had been consistently

increasing under the scenario of the decrease in the S_{JZB} and increase in the F_{TN} input during the same period. At the same time, the model experiments indicated that the effect of the F_{TN} input on WQ was higher than that of land reclamation, showing a synergistic process in WQ during the past 50 years. Referring to the observed C_{DIN} in JZB, the average contributions of the F_{TN} and S_{JZB} were approximately 37% and 29%, respectively. However, the combined effect was approximately 79% in practice, which was higher than the algebraic sum due to the synergistic process. The other CI contributions accounted for approximately 21%, which could be explained by other APIs and CCIs.

4. Discussion

4.1. Selection of indicators of the long-term effect on WQ

This study represents a modelling study of a long-term dataset that concerns the contamination of the JZB by a conceptual model analysis, FA and LRA. To evaluate the long-term effect on WQ of the multiple indicators, it is necessary to consider which indicators need to be incorporated to identify and simulate the processes of APIs and CCIs under a land-ocean interaction framework. Many previous studies focused on the variation in long-term environmental indicators at the regional scale (Lin et al.; 2001; McQuatters-Gollop et al., 2007; Nishikawa et al., 2010; Shen, 2001; Wei et al., 2015), investigating the effect of environmental indicators on WQ, but the selected indicators were not only arbitrary and incomplete but also unrelated to FA due to interactions between multiple indicators. Indeed, the selection indicator played an important role in the identification of key CIs. Nishikawa et al. (2010) explored the long-term monitoring of WQ and phytoplankton relationships in Harima-Nada, eastern Seto Inland Sea, but a decrease in the C_{DIN} was only attributed to nutrient reductions and did not consider the effect of the change in hydrodynamics induced by land reclamation on WQ. However, with respect to the long-term influence of hydrodynamics on WQ, land reclamation should also be considered in the assessment of the effect on WQ, especially in rapidly developing coastal areas (Gao et al., 2014; Hodoki and Murakami, 2006; Koh and de Jonge, 2014; Zarzuelo et al., 2015). Although many studies explored the roles of the quality and quantity of land-based inputs and coastline changes in WQ (Gao et al., 2014; Li et al., 2015b; Liang et al., 2015; Yuan et al., 2016), the results did not give a combined result on the basis of identifying key CIs by the FA and LRA.

4.2 The need for the integration of methods in the assessment of the effect on WQ

According to the results from the conceptual model analysis, the FA, the LRA and modelling experiments, the variation of the C_{DIN} in JZB was not only controlled by the CCIs of dissolved oxygen, pH, chlorophyll-a, temperature, salinity and river runoff but also by the APIs of the F_{TN} and S_{JZB} . Compared with previous studies (Borja, 2005; Borja et al., 2006; Jia, 2006; Li et al., 2015b; Liang et al., 2015; Yuan et al., 2016; Zhou et al., 2007b), the advantages of different methods involved overcoming shortcomings in the process, indicating that the results were credible. Consequently, the merits of the integration method were found in the assessment of the effect on WQ. First, the selection and relationships of indicators by the conceptual model analysis were applied to provide a solid foundation for the identification of key CIs, and incomplete indicators were avoided by experience and arbitrary decisions (Simeonov et al., 2003; Zhou et al., 2007b). Second, previous studies had been conducted on key CIs; however, they were not linked to a quantitative assessment of their effects on WQ by a numerical model (Jia, 2006; Simeonov et al., 2003; Yuan et al., 2016), leading to lack of integrity and accuracy for the quantitative assessment of key CIs on WQ. The land-based pollutant and land reclamation indicators that are directly related to WQ should be incorporated into the identification key CIs of WQ. Third, the effects of the combined key CIs on WQ was not found by the numerical model, which could not fully understand the role of combined key CIs in WQ (Gao et al., 2014; Yuan et al., 2016). Based on above analysis, the integration of methods in the assessment of effects on WQ was conducted due to shortcomings of the individual method application (Jia, 2006; Ouyang et al., 2006; Simeonov et al., 2003; Yuan et al., 2016). In this study, the modelling method result was consistent with the statistical analysis, indicating the two methods were complementary.

Furthermore, the established comprehensive methods framework with the feature of a conceptual model analysis, a statistical analysis and a numerical model included selecting indicators, identifying CIs and key CIs, as well as a quantitative assessment of the effect of key CIs on WQ using the numerical model, which could be readily applied into other rapidly developing coastal areas worldwide. According to the procedures of the integration of methods, other bay and coastal waters that are faced with similar issues could utilize the foundation provided by this study along with available regional data. The first step was to determine the relationship among WQ, APIs, and CCIs with the application of conceptual model analysis on the basis of a long-term series data. Then, the key CIs leading to the variation in WQ were identified by an FA and LRA on the basis of selection indicators. In addition, the WQ model, which could be applicable to the target region, was also developed to assess the effect of the key CIs on WQ quantitatively. In summary, both the integration of methods and results for the assessment of the effect on WQ obtained in the present

study can also be applied to other regions on the basis of available local data.

4.3 Combined influences of land-based pollution input and land reclamation

Due to the combined influences of the input of land-based pollution and land reclamation, the hydrodynamics and biogeochemical transformation processes of DIN in JZB have also changed significantly, resulting in the lasting deterioration of WQ. Contributions of the long-term F_{TN} input and land reclamation revealed a synergistic effect on WQ in JZB, which linked the key CIs in a LRA to the assessment of the effect on WQ. Compared with former studies of the effects land-based inputs and changes in hydrodynamics on WQ (Gao et al., 2014; Li et al., 2015b; Jia, 2006; Yuan et al., 2016), the synergistic effect on WQ was first found in the combined changing process. In addition, the land-based nitrogen input became responsible for a relatively greater level of effort in C_{DIN} pollution controls. The 50-year long-term average contribution of terrestrial TN input and land reclamation to the rise in the C_{DIN} were approximately 37% and 29%, respectively, compared with 79% for their combined effect by the changeable boundary coastal model. In terms of the long-term change in the hydrodynamics in JZB, the decrease in tidal current speed in JZB results from land reclamation within the Bay, and the decreasing water-exchange ability corresponds to an increasing average residence time over the past several decades, which influences the exchangeability of water and biologically and chemically important materials, such as nitrogen (Chen and Chen, 2012; Lin et al., 2015; Shi et al., 2011). At the same time, the increasing inputs of land-based pollution directly affect WQ by biogeochemical process (Lu et al., 2016; Wang et al., 2006). Moreover, due to double synchronous conduction, the individual effects on WQ were amplified, creating a synergistic effect on WQ. However, the total contribution of the other APIs and CCIs was only 21%, which indicated that the other indicators cannot be neglected. Future studies should consider these other indicators in a quantification assessment using the fine modelling method.

4.4 Recommendations for WQ restoration in JZB

To efficiently restore the WQ in JZB, implementations should be consistently undertaken from both sides related to the input of land-based pollution and land reclamation. Even if either the F_{TN} input to the JZB is reduced or the S_{JZB} is restored to the level of the 1960s, the impact on WQ means that it would be hard to meet the national WQ standard. The current WQ status in JZB was primarily the result of a combined synergistic process and long-term variations. During the past 50 years, the annual variation in the C_{DIN} exhibited an increasing pattern, with the concentrations initially increasing sharply from the 1960s to the 1980s and then starting to slightly

decrease at the end of the 1990s, and finally, a slightly fluctuating increase is observed during the 2010s. During the same period, intensive industry and population increases have been aggregated in the coastal zone around JZB, resulting in increased municipal and industrial discharges as well as excessive agricultural fertilizer loss, which were responsible for the dramatic growth of the pollutant load entering JZB (Liang and Zhang, 2005; Liang et al., 2015; Zhang, 2007). In addition, extensive reclamation around JZB was executed to meet the growing demands for land development, leading to the decline in the tidal prism and the weakness of water exchangeability (Gao et al., 2014; Liu, 2010; Shi et al., 2011; Sun et al., 2002; Yuan et al., 2016). Furthermore, the reduction of the self-purification capability resulted in continuous WQ degradation and ecosystem deterioration in JZB. Thus, to alleviate WQ pollution and restore WQ with a practical implementation, effective measures should be taken on land-based pollution and coastline protection based on the clear baseline over a long period of time. In addition, further research is needed to support future assessments, predictions and adaptive management of WQ based on the long-term dataset.

5. Conclusion

The advantages of applying multiple methods to investigate indicator variables monitored in coastal areas were evaluated. The effectiveness of the methodology was demonstrated by applying it to the analysis of the effect of a time series of factors on WQ. Long-term increases in the C_{DIN} were controlled by multiple factors, and the APIs could not be neglected in factor selection by the conceptual model analysis. The increase in the C_{DIN} over the JZB was consistent with the rise of the F_{TN} input and the decrease of the S_{JZB} . Based on long-term data, high-intensity land-based source discharge and land reclamation, which significantly changed the hydrodynamics and biogeochemical processes of the JZB starting in the 1960s, were the two key CIs of long-term variation of the C_{DIN} found by the LRA. The effects of the F_{TN} discharge and land reclamation on the C_{DIN} accounted for approximately 37% and 29%, respectively, indicating land-based nitrogen input became responsible for a relatively greater level of effort in C_{DIN} pollution controls. The combined effect was 79% in practice, showing a synergistic WQ effect. The outcomes of this study showed how the application of long-term monitoring data can be used to examine the effects of indicators on WQ in JZB, which could provide policy makers and management agencies with critical information for incorporation into environmental management frameworks. For the WQ to reach the desired grade standard and to provide scientific support for the sustainable development of JZB, it is necessary to implement a system to control the terrigenous nitrogen pollutant load, and the coastline must be scientifically planned and protected.

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Figures captions

Figure 1. The geographical location and spatio-temporal coastline changes of Jiaozhou Bay (JZB). The coloured lines indicate the coastal boundaries.

Figure 2. The conceptual model of the causal chain and inter-relations of water quality (WQ) and the anthropogenic pressure indicators (APIs) and coastal carrying indicators (CCIs). The solid line rectangles indicate that the control indicators (CIs) are closely related to the dissolved inorganic nitrogen (C_{DIN}) in Jiaozhou Bay (JZB). The dotted line rectangles indicate no significant relationships between the C_{DIN} and indicators. The category of APIs includes land-based sources of pollution and mixed boundary groups. The category of CCIs mainly includes the two groups of hydrodynamic transport and biogeochemical processes. The group of land-based sources consists of quantity and speciation teams of terrigenous pollutants. The mixed boundary group includes mainly the mixed boundary shape, sewage outfall location, river runoff and sediment discharge into the bay. The hydrodynamic transport group includes the type of tide, current, current density, land, sea floor, open sea and gas boundaries. The subgroup of biogeochemical processes includes biology, water, suspended particles and sediments of the four media and types of environmental effects.

Figure 3. The long-term variation of the APIs (left) and CCIs (right). The solid symbols represent the observed values, and the hollow symbols represent the estimated values. The F_{TN} and S_{JZB} represent the terrigenous total nitrogen flux and the water area of Jiaozhou Bay (JZB), respectively.

Figure 4. The anthropogenic pressure indicators (APIs) and coastal carrying indicators (CCIs) factor loading squares and sum of F_1 and F_2 loading squares (F_{sum}). F_{TN} and S_{JZB} represent the terrigenous total nitrogen flux and the water area of Jiaozhou Bay (JZB), respectively.

Figure 5. The R^2 of indicators related to the dissolved inorganic nitrogen concentration (C_{DIN}) by a linear regression analysis. The significance of indicators is specified with a p-value. F_{TN} and S_{JZB} represent the terrigenous total nitrogen flux and the water area of Jiaozhou Bay (JZB), respectively.

Figure 6. The annual variation of dissolved inorganic nitrogen concentrations (C_{DIN}). The solid box symbol represents observed data. The triangle symbol represents the single effect of the F_{TN} and S_{JZB} . The open box symbol is the combined effect on the C_{DIN} . F_{TN} and S_{JZB} represent terrigenous total nitrogen flux and the water area of Jiaozhou Bay (JZB).

Tables captions

818   **Table 1.** Details of the model scenarios

Figure

Figure 1. The geographical location and spatio-temporal coastline changes of Jiaozhou Bay (JZB). The coloured lines indicate the coastal boundaries.

Figure 2. The conceptual model of the causal chain and inter-relations of water quality (WQ) and the anthropogenic pressure indicators (APIs) and coastal carrying indicators (CCIs). The solid line rectangles indicate that the control indicators (CIs) are closely related to the dissolved inorganic nitrogen (C_{DIN}) in Jiaozhou Bay (JZB). The dotted line rectangles indicate no significant relationships between the C_{DIN} and indicators. The category of APIs includes land-based sources of pollution and mixed boundary groups. The category of CCIs mainly includes the two groups of hydrodynamic transport and biogeochemical processes. The group of land-based sources consists of quantity and speciation teams of terrigenous pollutants. The mixed boundary group includes mainly the mixed boundary shape, sewage outfall location, river runoff and sediment discharge into the bay. The hydrodynamic transport group includes the type of tide, current, current density, land, sea floor, open sea and gas boundaries. The subgroup of biogeochemical processes includes biology, water, suspended particles and sediments of the four media and types of environmental effects.

Figure 3. The long-term variation of the APIs (left) and CCI (right). The solid symbols represent the observed values, and the hollow symbols represent the estimated values. The F_{TN} and S_{JZB} represent the terrigenous total nitrogen flux and the water area of Jiaozhou Bay (JZB), respectively.

Figure 4. The anthropogenic pressure indicators (APIs) and coastal carrying indicators (CCIs) factor loading squares and sum of F_1 and F_2 loading squares (F_{sum}). F_{TN} and S_{JZB} represent the terrigenous total nitrogen flux and the water area of Jiaozhou Bay (JZB), respectively.

Figure 5. The R^2 of indicators related to the dissolved inorganic nitrogen concentration (C_{DIN}) by a linear regression analysis. The significance of indicators is specified with a p -value. F_{TN} and S_{JZB} represent the terrigenous total nitrogen flux and the water area of Jiaozhou Bay (JZB), respectively.

Figure 6. The annual variation of dissolved inorganic nitrogen concentrations (C_{DIN}). The solid box symbol represents observed data. The triangle symbol represents the single effect of the F_{TN} and S_{JZB} . The open box symbol is the combined effect on the C_{DIN} . F_{TN} and S_{JZB} represent terrigenous total nitrogen flux and the water area of Jiaozhou Bay (JZB).

Table 1. Details of the model scenarios

Scenarios	F_{TN}	S_{JZB}	Effects on WQ
1	1962	1966	To explore the effect of changes in the S_{JZB}
		1986	
		2000	
		2008	
2	1962	1966	To explore the effect of changes in the F_{TN}
	1986		
	1990		
	1992		
	2000		
	2008		
3	1966	1966	To explore the combined effect
	1986	1986	
	2000	2000	
	2008	2008	