Estimating submarine groundwater discharge in the Jiaozhou Bay using radium isotopes

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Submarine groundwater discharge (SGD) is part of the global water cycles, which delivers a large amount of solutes to the biogeochemical cycle and affects the coastal ecological environment. Natural radium isotopes ($^{224}$Ra and $^{226}$Ra) were used to estimate the SGD in Jiaozhou Bay and the mass balance models were established. Radium sources including river transport, sediment diffusion, desorption from re-suspended particles, open sea flood tide, municipal wastewater, and SGD are considered. Radium output owning to the radioactive decay and ebb tide are also considered. Our results show the SGD fluxes during September to October in 2011 (autumn) and April to May in 2012 (spring) are $1.51 \times 10^6$ m$^3$·d$^{-1}$ and $6.53 \times 10^5$ m$^3$·d$^{-1}$, respectively. In the same period of time, the input of the dissolved inorganic nitrogen (DIN) from SGD is very close to that from river, yet, the soluble silicate and reactive phosphate vary seasonally. SGD is the major source of the nutrient input of Jiaozhou Bay.

[Key words: submarine groundwater discharge, radium isotope, groundwater, nutrient, Jiaozhou bay]

Introduction

Submarine groundwater discharge (SGD) defines as any flow of water on continental margins from the seabed to the coastal ocean, regardless of the composition or driving force$^1$. SGD exchanges water between land and sea, and often occurs in coastal aquifers or estuary where biogeochemical reactions modify the water chemical constituents. Thus, SGD tends to be enriched by groundwater more than seawater$^2$. In recent years, SGD has been recognized as an important source of substances (including dissolved inorganic carbon, trace metals, rare earth elements, nutrients etc.) to the coastal water, which leads to the pollution of coastal marine waters (e.g. Cai et al$^7$, 2003; Moore et al$^8$, 2006; Beck et al$^9$, 2007; Bone et al$^{10}$, 2007; Garcia-Solsona et al$^{11}$, 2010; Santos et al$^{12}$, 2011; Beck et al$^{13}$, 2013; Gonneea et al$^{14}$, 2013; Rodrigo et al$^{15}$, 2014; Kim et al$^{16}$, 2014). Thus, even a small amount of SGD can transport a relative large amount of nutrients and other compounds to the coastal water$^{1,7,8,18}$. In some area, SGD show greater ecological effects than surface runoff. Burnett et al$^{8,9}$ (2006) reported that although submarine fresh groundwater discharges only 6% of the river, it transfers 50% of the dissolved substances as the river transfers. Kim and Kim$^{20}$ (2011) reported that SGD from Jeju Island is the dominant source of REEs in coastal waters, which is comparable to those through major rivers. Johannesson et al$^{21}$ (2011) also reported that the net Nd flux through SGD to the Indian River Lagoon is approximately seven times that from the local surface runoff.

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Many methods are known to estimate SGD. Isotopic measurements such as $^{222}$Rn, $^{226}$Ra, $^{228}$Ra, $^{224}$Ra, $^{233}$Ra, $^{14}$C, $^{234}$U, and $^{238}$U can serve as key indicators of fluxes across the groundwater/marine interface. Radium isotopes have been used most extensively. Ra has four isotopes showing widely varied half-lives ($^{224}$Ra: 3.6 d, $^{223}$Ra: 11.3 d, $^{228}$Ra: 5.8 y, $^{226}$Ra: 1600 y), which are used solely or jointly to explain different characteristics of the SGD phenomenon. Ra is produced by the decay of the parent thorium isotopes on or within the aquifer solids. Radium tends to be highly particle-reactive in fresh groundwater, but low active in the dissolved phase. However, the intruding saltwater result in ion exchanges, and thus, saline groundwater is enriched in Ra with orders of magnitude. The advection and discharge of this high activity interstitial water can support an inventory of dissolved Ra in coastal waters better than other sources.

In this work, dissolved radium in Jiaozhou bay water is from river transport, sediment diffusion, desorption of re-suspended particles, open sea flood tide, municipal wastewater, and SGD. The Ra loss includes radioactive decay and ebb tide. A radium ($^{224}$Ra and $^{226}$Ra) mass balance model (box model) is built to estimate the SGD in Jiaozhou bay over two periods of time (autumn and spring).

**Material and Methods**

Jiaozhou Bay (35°57’–36°18’N, 120°04′–120°23’E) is a typical semi-enclosed water body, situated in the southern part of the Shandong Peninsula and connected to the North Yellow Sea of China (Fig. 1). The average water area is 367 km$^2$, 33 km long from North to South and 28 km wide from East to West; the bay mouth is only about 2.5 km wide. The sediment is mostly sand and silt. The tide of Jiaozhou Bay is typically semi-diurnal with the average tidal range of 2.71 m. Tidal currents in most areas of the Jiaozhou Bay flow back and forth. The four main seasonal rivers include the Yang River, Dagu River, Moshui-Baisha River and Licun River. The flood season usually occurs in July, August and September. Jiaozhou Bay locates in typical East Asian monsoon climate zone, the average precipitation is 680.5 mm, the annual precipitation concentrated in July and August constitutes 45% of the annual precipitation, the precipitation in December is the least one, only 9.8 mm.

Around the Jiaozhou bay, South-Eastern and Southern Hills expose large areas of bed rock that is mainly magmatic rocks. The groundwater is dominated by the weathering fissure water in groundwater is Quaternary unconsolidated
sediments, local outcropping Cretaceous Qingshan group sandstone. Thus, the groundwater around Jiaozhou bay is dominated by pore phreatic-water-in-unconsolidated-sediment-of-the -Quaternary-period, distributed mainly in the lower reaches of the plain of the Yang River, Dagu River, Moshui-baisha River. The aquifer mostly is a double-layer structure with thickness of 5~20m. Lithology is dominated by gravel and sand, with a small amount of clay. Groundwater mainly receives rainfall recharge, and discharges to the sea by evaporation and runoff.

In total 28 groundwater samples, 23 river water samples and 35 seawater samples were collected in September to October, 2011(Fig.1). And 39 groundwater samples, 40 river water samples and 35 seawater samples were collected in April to May, 2012(Fig.2). Groundwater and river water samples were obtained in 15L polyethylene containers, seawater samples were obtained in 30L polyethylene containers at depths of 1 m below the surface. Salinity of water samples was field measured (JENCO Model-3250, accuracy: ±0.5%F.S.). On October in 2011, eight surface sediments were collected (Fig.1) to determine the diffusive Ra flux and desorption flux from re-suspended particles.

**Analytical methods**

Ra isotopes were extracted as soon as the water samples were received in the laboratory onto manganese oxide-impregnated acrylic fiber (hereafter, ‘Mn-fiber’) at flow rates less than 200–250ml per minute. Subsequently, the Mn-fibers were taken out and partially dried and put into the diffusion tubes. \(^{224}\)Ra and \(^{226}\)Ra were analyzed by using FD-125 Radon-Thorium analyzer made in Beijing following the method as reported by Huang et al. (2001) and Xie et al. (1994).

Re-suspended surface sediment particles caused by currents desorb radium into overlying water. Eight sediment samples were used in the laboratory, the surface sediment (1~2cm) were selected and dry naturally. Based on the re-suspended particles concentration in the Jiaozhou bay (30mg·L\(^{-1}\)), we carried out a simple experiment by stirring 450mg of sediment in 15L of filtered, Ra-free seawater (salinity=28.5) for 2h. The slurry was then filtered through a column containing raw acrylic fiber to remove the sediment, and the dissolved Ra was extracted onto the Mn-fiber.

With eight sediment samples, the sediment incubation experiments were carried out according to the methods described by Beck (2007) and Solsona (2008). The radium activity of the overlying water of the sediment cores in each incubation period was measured, then fitting multiple measurement points and theoretical equation were made, the sediment diffusion flux of radium was determined.

Water samples were obtained in 600ml polyethylene seal bottles, and placed in portable ice storage box. As soon as arrived in the lab, samples were immediately filtered with a 0.45 μm membrane and analyzed for dissolved inorganic phosphate(PO\(_4^{3-}\)-P), dissolved inorganic silicate (SiO\(_2^{2-}\)-Si) and dissolved inorganic nitrogen (NH\(_4^+\)-N, NO\(_3^-\)-N, NO\(_2^-\)-N) using the standard spectrophotometric technique. Analytical error was <10% for all nutrients based on duplicate samples analyzed every ten samples.

**Radium mass balance**

Plot 3 showed \(^{224}\)Ra and \(^{226}\)Ra activities versus salinity in surface water of Jiaozhou Bay. If the input of groundwater is not taken into consideration, the seawater in the Jiaozhou bay can be seen as the river water mixed with open seawater, all \(^{224}\)Ra and \(^{226}\)Ra activities should be located on or near the line, which represents the expected conservative mixture between river water and ocean water. Actually, most of \(^{224}\)Ra and \(^{226}\)Ra activities locate upon the line, showing pronounced excess Ra, which means other radium sources in Jiaozhou bay exist. Lee et al. (1977) and Moore et al. (1981) indicated that excess Ra in estuary and bay mainly came from the sources of desorption from river suspension and the diffusion from surface sediment. In recent years, more and more researchers pointed out the
submarine groundwater discharge (SGD) are an important source of excess radium (e.g., Moore, 1996; Kim et al., 2005).

We estimate submarine groundwater discharge in Jiaozhou Bay in September to October 2011 (autumn) and April to May 2012 (spring) by using radium ($^{224}$Ra and $^{226}$Ra) mass-balance model. The main sources of radium to the Jiaozhou Bay include river transport ($F_{river}$), diffusion from sediments ($F_{diffuse}$), desorption from re-suspended particles ($F_{s-desorb}$), open sea flood tide input ($F_{in}$), input from municipal wastewater ($F_{ww}$), and SGD. The source of desorption of riverine suspended particles is ignored because of the low stream flow and low suspended particles concentration. Ra can be removed through radioactive decay ($F_{decay}$) and ebb tide ($F_{out}$).

Based on the source of Radium in Jiaozhou Bay described above, the sources of input and loss are equal when the Ra observed in the water column during any season under at approximately steady state condition. Thus, we can use Ra mass balance to examine Ra cycling in Jiaozhou Bay, the equation is (Bq·d$^{-1}$)

$$F_{out}+F_{decay}=F_{in}+F_{river}+F_{diffuse}+F_{s-desorb}+F_{ww}+F_{SGD}$$

(1)

Radioactive decay

In the environment of Jiaozhou Bay, under which the average water residence time is 40.1d$^{-1}$, radioactive decay of short lived radium ($^{224}$Ra: 3.66d) can be an important losing process. Because of the relatively short residence time of the water, decay is not a significant loss term for the long lived Ra isotopes ($^{226}$Ra: 1600 year). We can estimate the flux of $^{224}$Ra radioactive decay as $F_{decay}=V_{bay}C_{bay}\lambda_{224}$ (2) where $V_{bay}$ is the volume of the Jiaozhou bay, $26.12\times10^8$ m$^3$ (Autumn), $9.552\times10^8$ m$^3$ (Spring); $\lambda_{224}$ is the decay constant of $^{224}$Ra, 0.189d$^{-1}$; $C_{bay}$ is the average activity of $^{224}$Ra in the bay (Bq·m$^{-3}$).

Radium loss with ebb tide

We can assess the loss as $F_{out}=(V_{tide}n_{tide}+Q_{river})C_{BM}$ (3) where $V_{tide}$ is the tidal prism volume in the Jiaozhou bay, $10.713\times10^8$ m$^3$ (Autumn), $9.552\times10^8$ m$^3$ (Spring); $Q_{river}$ is the average river runoff into the sea, $0.28\times10^8$ m$^3$·d$^{-1}$ (2011), $0.13\times10^8$ m$^3$·d$^{-1}$ (2012); $n_{tide}$ is the number of tidal cycles per day, 1.933 d$^{-1}$. $C_{BM}$ is the average activity of Ra in the mouth of Jiaozhou bay.

Ra input from the open sea with flood tide

Jiaozhou Bay is connected to the Yellow sea, seawater enters the Bay at flood tide. We can calculate the Ra input to Jiaozhou bay from Yellow sea as:

$$F_{in}=V_{tide}C_{ocean}n_{tide}$$

(4)

where $C_{ocean}$ is the average Ra activity of Yellow
There are several seasonal rivers including the Baisha-Moshui River, Dagu River, and the Yang River around the Jiaozhou bay, Licun River is almost drying up. The Ra input flux from the rivers transport can be calculated as:

\[ F_{\text{river}} = Q_{\text{river}} \times C_{\text{river}} \]  

where \( C_{\text{river}} \) is the average Ra activity of river (Bq·m\(^{-3}\))

**Diffusion of Ra from sediments**

Ra fluxes from surface sediments in Jiaozhou Bay can be calculated using the following formula:

\[ F_{\text{diffuse}} = J_{\text{diffuse}} \times A_{\text{sed}} \times H_{\text{day}} \]  

where \( J_{\text{diffuse}} \) is the diffusive flux (per unit area per hour) which gets by sediment diffusion experiments (Bq·m\(^{-2}\)·h\(^{-1}\)). \( A_{\text{sed}} \) is the surface area in the Jiaozhou bay, 3.2655×10\(^{8}\) m\(^{2}\). \( H_{\text{day}} \) is the hours per day, 24 h·d\(^{-1}\).

**Ra desorption from re-suspended particles**

Ra fluxes from re-suspended particles of surface sediment can be estimated by:

\[ F_{\text{s-desorb}} = M_{\text{resus}} \times V_{\text{tidal}} \times n_{\text{tidal}} \times C_{\text{resus}} \]  

where \( M_{\text{resus}} \) is the average concentration of suspended particles in the Jiaozhou bay, 30g·m\(^{-3}\). \( C_{\text{resus}} \) is the average Ra activity of desorption from re-suspended particles in the bay which is experimentally determined (Bq·g\(^{-1}\)).

**Ra flux from municipal wastewater input**

We found that municipal wastewater has a high Ra activity around Jiaozhou Bay. This suggests that desorption of Ra from municipal wastewater can’t be negligible. Beck\(^{10}\) (2007) thought that the wastewater was one source of Ra in Jamaica Bay, NY. Wastewater input flux of Ra can be given by

\[ F_{\text{ww}} = V_{\text{ww}} \times C_{\text{ww}} \]  

where \( V_{\text{ww}} \) is the daily wastewater discharge in the bay, 59.97×10\(^{4}\) m\(^{3}\)·d\(^{-1}\). \( C_{\text{ww}} \) is the average Ra activity measured in wastewater (Bq·m\(^{-3}\)).

The values of radioactivity of Ra and diffusion flux in the above equations are given in Table 1—Values of parameters in the radium mass balance model

<table>
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<tbody>
<tr>
<td>( C_{\text{river}} / \text{Bq·m}^{-3} )</td>
<td>2.34±0.23</td>
<td>2.44±0.22</td>
</tr>
<tr>
<td>( C_{\text{BM}} / \text{Bq·m}^{-3} )</td>
<td>2.09±0.17</td>
<td>2.5±0.19</td>
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<tr>
<td>( C_{\text{Bay}} / \text{Bq·m}^{-3} )</td>
<td>1.99±0.16</td>
<td>1.93±0.17</td>
</tr>
<tr>
<td>( C_{\text{ocean}} / \text{Bq·m}^{-3} )</td>
<td>1.87±0.14</td>
<td>2.02±0.12</td>
</tr>
<tr>
<td>( C_{\text{ww}} / \text{Bq·m}^{-3} )</td>
<td>5.84±0.33</td>
<td>10.3±0.41</td>
</tr>
<tr>
<td>( C_{\text{resus}} / \text{Bq·g}^{-1} )</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td>( J_{\text{diffuse}} / \text{Bq·m}^{-2}·\text{h}^{-1} )</td>
<td>0.04</td>
<td>0.04</td>
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### Results and Discussion

**Submarine groundwater discharge**

We quantified all source and loss terms of Ra in Jiaozhou bay except for SGD, a large imbalance in Ra fluxes is found in Jiaozhou bay, which should belong to the SGD flux of Ra, theoretically. By transformation Eq. (1), the flux of Ra from SGD is given by

\[ F_{\text{SGD}} = F_{\text{in}} + F_{\text{river}} + F_{\text{diffuse}} + F_{\text{s-desorb}} + F_{\text{ww}} \]

Calculate by Eq. (9), the results show that the fluxes of \(^{226}\text{Ra}\) and \(^{226}\text{Ra}\) from SGD are 2.49×10\(^{8}\) Bq·d\(^{-1}\) and 0.42×10\(^{8}\) Bq·d\(^{-1}\) in September to October, 2011; the fluxes of \(^{226}\text{Ra}\) and \(^{226}\text{Ra}\) from SGD were 1.34×10\(^{8}\) Bq·d\(^{-1}\) and 0.16×10\(^{8}\) Bq·d\(^{-1}\) in April to May, 2012.

With Ra fluxes of SGD divided by groundwater radium activities, we can get the actual SGD in the Jiaozhou bay. Considering the groundwater...
samples closed to the coastal zone should represent better the groundwater discharged to the Jiaozhou bay, we choose the average Ra activity of the ten groundwater samples (G11,G15,G16, G17,G18,G19,G20,G26,G27,G29) locate in the downstream of Baisha-Moshui river plain, Dagu river plain and Yang river plain as the groundwater end members. The results show that during September to October in 2011, the fluxes of SGD are $21.01 \times 10^6 \text{ m}^3 \cdot \text{d}^{-1}$ (based on $^{224}$Ra mass balance) and $15.17 \times 10^6 \text{ m}^3 \cdot \text{d}^{-1}$ (based on $^{226}$Ra mass balance), and $11.33 \times 10^6 \text{ m}^3 \cdot \text{d}^{-1}$ (based on $^{224}$Ra mass balance) and $6.53 \times 10^6 \text{ m}^3 \cdot \text{d}^{-1}$ (based on $^{226}$Ra mass balance) during April to May in 2012. The SGD fluxes based on $^{224}$Ra mass balance model are higher than that based on $^{226}$Ra mass balance model. The reason may be that the sample seawater end member is closed to the mouth of the Jiaozhou bay, where the measured $^{224}$Ra activity of the Yellow sea is higher than the actual value, and the measured $^{226}$Ra activity is similar to the actual value. So the calculation error based on $^{224}$Ra mass balance model is larger, results derived from $^{226}$Ra mass balance model are reliable.

Derived from $^{226}$Ra mass balance model, the fluxes of SGD are $15.17 \times 10^6 \text{ m}^3 \cdot \text{d}^{-1}$ (Sep-Oct,2011) and $6.53 \times 10^6 \text{ m}^3 \cdot \text{d}^{-1}$ (Apr-May,2012), the SGD fluxes of Sep-Oct, 2011 are 2 times more than the SGD fluxes of Apr-May, 2012 , which is possibly caused by precipitation. In the Jiaozhou Bay, the monsoon (flood season) begins from late June to early September, the groundwater is fully recharged by rainwater because of the plenty of rain in this period, and the groundwater level rises and has high hydraulic gradient from land to sea. A large amount of submarine groundwater discharge happens. From January to May, owing to the minimum precipitation, groundwater will not be recharged and will supply around rivers conversely. Also, groundwater is massively exploited in the dry season in Jiaozhou bay area. All these cause the groundwater level falls, giving a small amount of SGD. Therefore, submarine groundwater discharge is largely dominated by precipitation.

If distributing the fluxes of submarine groundwater discharge into the seabed of Jiaozhou bay, we can get the input SGD fluxes per seabed area. Considering that the Jiaozhou bay area is relatively large, SGD input mainly occurs at the coastal zone and that in the middle of bay maybe small. In addition, the southeast and southwest shores of the Jiaozhou bay are all rocky coast and the range of SGD extending to the seabed is very limited. So we believe that half of seabed area near the shore (mainly in the Northwest, North and Northeast) has SGD seepage and the other half can be negligible. The seabed area of Jiaozhou bay is $326.55 \times 10^6 \text{ m}^2$. Divide the SGD fluxes by half of seabed area, the fluxes of SGD per unit seabed area is $0.09 \text{ m}^3 \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ (Sep-Oct, 2011) and $0.04 \text{ m}^3 \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ (Apr-May, 2012).

**Nutrients from SGD**

During September to October in 2011, the average dissolved inorganic nitrogen (DIN) concentration in groundwater around Jiaozhou bay is $658.3 \mu\text{mol} \cdot \text{L}^{-1}$, soluble silicate concentration is $167.5 \mu\text{mol} \cdot \text{L}^{-1}$, reactive phosphate concentration only is $0.11 \mu\text{mol} \cdot \text{L}^{-1}$ which maybe is relate to the incorporating and consuming of phosphate by summer and autumn seasonal crops and plants. During April to May in 2012, the average dissolved inorganic nitrogen (DIN) concentration is $167.5 \mu\text{mol} \cdot \text{L}^{-1}$, reactive phosphate concentration only is $0.11 \mu\text{mol} \cdot \text{L}^{-1}$, and soluble silicate concentration is $196.1 \mu\text{mol} \cdot \text{L}^{-1}$. Divide the SGD fluxes by half of seabed area, the fluxes of SGD per unit seabed area is $0.09 \text{ m}^3 \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ (Sep-Oct, 2011) and $0.04 \text{ m}^3 \cdot \text{m}^{-2} \cdot \text{d}^{-1}$ (Apr-May, 2012).

The ratios of the average concentrations of N and P in groundwater are $5984.5 (\text{Sep-Oct, 2011})$ and $66.3 (\text{Apr-May, 2012})$, they all exceed Redfield. The seawater environment will turn to phosphorus limitation if the groundwater having high N/P ratio enters into the sea.

Assuming that chemical behavior of nutrients is conservative and nutrient content does not change when it flows through underground estuary, we calculate the nutrient fluxes from SGD by multiplying the SGD fluxes and nutrient
concentration in groundwater. During September to October in 2011, the fluxes of dissolved inorganic nitrogen (DIN), soluble silicate and reactive phosphate are $998.6 \times 10^4 \text{ mol} \cdot \text{d}^{-1}$, $254.1 \times 10^4 \text{ mol} \cdot \text{d}^{-1}$ and $0.2 \times 10^4 \text{ mol} \cdot \text{d}^{-1}$. During April to May in 2012, the fluxes of dissolved inorganic nitrogen (DIN), soluble silicate and reactive phosphate are $385.1 \times 10^4 \text{ mol} \cdot \text{d}^{-1}$, $128.1 \times 10^4 \text{ mol} \cdot \text{d}^{-1}$ and $5.8 \times 10^4 \text{ mol} \cdot \text{d}^{-1}$.

Theoretically, the scale of circulating seawater can’t be large which means that the depth of circulating seawater penetrating into onshore aquifers is limited, so not all circulating seawater is involved in the nutrient transport, especially the transport of the groundwater nutrients through circulating seawater which is caused by waves is limited. Therefore, the nutrients fluxes estimated by SGD fluxes could be higher than the actual fluxes.

Comparing the nutrients inputs of SGD and river, the dissolved inorganic nitrogen inputs are closed in the same period; reactive silicate inputs were closed during September to October in 2011, but different nearly 5 fold during April to May in 2012; reactive phosphate inputs change largely, the inputs of river are dominant during September to October in 2011, but the inputs of SGD are dominant during April to May in 2012. Overall, inputs of SGD were considered to be important for the nutrients inputs into the Jiaozhou Bay.

**Conclusions**

Radium in the Jiaozhou bay water is not merely the conservative mixture of river and open seawater, inputs from groundwater, sediments diffusion, and re-suspended particles desorption, and municipal wastewater and open sea flood tide should be considered as well. Ra loss mainly via radioactive decay ($^{224}\text{Ra}$) and ebb tide. Key of the radium mass balance model (box model) is to quantify the source and loss of radium. Concentration of $^{224}\text{Ra}$ and $^{226}\text{Ra}$ isotopes in different types of water can be easily measured, the box models is an ideal model to estimate the unknown source of SGD. But in this work, the activity of $^{224}\text{Ra}$from open sea is not reasonable, thus, the $^{226}\text{Ra}$ box model is used to calculate the fluxes of SGD to be 15.17 (Sep-Oct, 2011) and 6.53×106 m$^3 \cdot$d$^{-1}$ (Apr-May, 2012), respectively. Submarine groundwater discharge varies seasonally, in the order: monsoon-dry season, indicating the submarine groundwater discharge is dominated to some extent partly by precipitation.

Similar to the river input, the nutrient input from SGD varies seasonally. The dissolved inorganic nitrogen inputs from river and SGD are very closed in the same period of time, thus, the SGD is an important nutrient input in the Jiaozhou Bay.

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**References**


