Dynamics and spatial differentiation of dissolved copper load in the Yangtze estuary

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Abstract: Taking the Yangtze river estuary as the study area and based on the field measured data of the longest sequence from 2004 to 2018, analyzes the Dissolved Copper (DCu) concentration change process of six typical sections in the Yangtze river estuary in the past 15 years, namely, Xuliujiing, Shidongkou, Nangang, Beigang, Qinglonggang and Qidonggang. The characteristics of the evolution trend of each section are quantitatively evaluated by the method of restandard range analysis. The spatial differentiation rules of DCu in the Yangtze river estuary at different stages were revealed and the correlation characteristics of each section load were calculated by Pearson coefficient. The results show that: (1) the evolution characteristics of DCu in the section of Xuliuji section are relatively complex, which can be divided into four stages. The average concentration of DCu in 15 years is about 3.52 \( \mu \text{g/L} \), and the highest monthly average concentration is 17.8 \( \mu \text{g/L} \). (2) Due to the effect of regional sewage discharge, the load of DCu in the mouth section of the south branch stone is generally higher than that of the Nangang and Beigang sections, but the evolution trend of the three sections is basically similar. The oscillation amplitude of DCu concentration in Qidonggang section is the most significant. In 2007, the annual average value of DCu reached 30.3 \( \mu \text{g/L} \), which is the highest annual average value in the existing measured data. (3) Hurst index of Nangang and Qinglonggang reached 0.93 and 1.0 respectively, and the evolution trend of cross section DCu showed a strong persistence. The Hurst index of Qinglonggang section is 0.1515, which indicates that the section DCu has anti-persistence. (4) Except Qidonggang, the DCu in the other five monitored sections all show significant positive correlation, among which the DCu concentration in Beigang and Qinglonggang have extremely similar variation rules. The main reason for this strong correlation should be related to the main driving weight factor of DCu load in the two sections. The research results have some reference value for prevention and control of heavy metal pollution in Yangtze estuary.

1. Introduction

Estuary is a land-sea connection zone with strong interaction between sea and land and complex biogeochemical processes\textsuperscript{[1-3]}. At the same time, the estuary is also the main channel for the transport of terrestrial materials to the ocean, significantly affected by human activities\textsuperscript{[4-6]}. Among the many substances transported by the river to the ocean, heavy metal elements are highly toxic, persistent, and difficult to degrade. As a typical cumulative pollutant, they have become an important factor affecting
the estuary and offshore ecology\cite{7,8}. In recent years, human activities such as urban construction, industrial and agricultural development, and mineral resources development are further increasing the intensity of heavy metal discharge in the basin. Heavy metal pollution in the estuary has gradually become an important hot spot in the global water environment\cite{9,10}. At present, many scholars have carried out a series of studies on the problem of heavy metal pollution in the estuary. For instance, Feng et al.\cite{11} studied the content and distribution of heavy metals in the sediments of the Hudson estuary in New York, USA; Jones et al.\cite{12} used acid simultaneous extraction to determine the content of heavy metals in the Tees estuary in northeast England, and studied its deposit pattern; Hatje et al.\cite{13} conducted a study in Jackson Bay, Australia, and found that high concentrations of particulate heavy metals are mainly from coastal sewage discharges and osmotic water in reclamation areas, and peaks of heavy metals appear in the upper estuary. These studies have laid an important foundation for mastering the temporal and spatial distribution characteristics and migration and transformation of heavy metals in the estuary. However, most of the existing researches are carried out on particulate heavy metals (including suspended and deposited states) in the estuary, and the attention to dissolved heavy metals is weak. Different forms of heavy metals in water can exhibit different biological toxicities and produce different environmental effects. Dissolved heavy metals migrate with biogeochemical processes and exhibit non-conservative behavior; their total amount is lower than that of particulate matter, but its risk factor for exposure to aquatic animals and plants is higher, and it will increase the migration of heavy metals with food chains. Bioavailability. At the same time, dissolved heavy metals directly affect the distribution coefficient of dissolved-suspended-deposited three-phase exchange of water-weight metal. The spatial distribution similarity between the total amount of heavy metals in the water and the dissolved state is studied. The temporal and spatial distribution of dissolved heavy metals in water is analyzed. Heavy metal sources and their migration and transformation mechanisms in water bodies are of great significance. This paper takes China's Yangtze River estuary as the research area, based on the field observation of the old sequence of 2004-2018, and uses the Rescaled Range Analysis (R/S method) to analyze the Xuliujing, Qinglonggang, Qidonggang, Shidongkou in Yangtze estuary, The evolution of DCu content in six sections of Nangang and Beigang was nearly fifteen years; quantitatively revealed the spatial differentiation law and correlation characteristics of DCu in different stages.

![Figure 1. Location of study area and distribution of monitoring sites](image-url)
2. Materials and methods

2.1. Research area

With a total length of 6,397 km, the Yangtze River is the first longest river in Asia and the third longest river in the world. The estuary can be divided into three sections: 1. Anhui Datong (dry season tidal zone boundary) to Jiangyin (Hongji tidal zone boundary) of Jiangsu Province, 400 km long is the near mouth section; 2. Jiangyin to the mouth The gate (the top of the beach) is 240 km long and is the estuary section; 3. from the mouth to the outside of the 30-50 m isobath, the submarine delta development area is outside the mouth, as shown in Figure 1. The Yangtze River estuary area includes Shanghai, Jiangsu Province, Jiangsu Province and Nantong City along the Yangtze River, occupying a pivotal position in the Yangtze River Basin and China's social and economic development. Below the Xuliujing monitoring section, the overall riverbed shape of the Yangtze River estuary is horn-shaped. On the plane of the Yangtze River estuary, there are three levels of tillering and four into the sea. The lower reaches of the Baihe River estuary are separated into the South Branch and the North Branch Waterway by Chongming Island. The South Branch Waterway is divided into Nangang Waterway and Beigang Waterway by Changxing Island and Hengsha Island near Wusongkou. The Nangang Waterway is divided into the South Slot and the North Slot by the Jiuduansha. The north branch, the north port, the north trough and the south trough are the four sea inlets of the Yangtze River estuary. The tidal wave in the sea near the Yangtze River estuary is affected by the tidal wave system of the East China Sea and the tidal wave system of the Yellow Sea. The East China Sea tidal wave system dominates. The Yangtze River estuary is a medium-intensity tidal estuary with a regular half-day tide and a tidal wave deformation in the mouth, which is an irregular half-day shallow sea tide. The Yangtze River estuary is not only the necessary passage for a variety of biological anniversary rivers and rivers, but also the main station for migratory birds migration in the Asia-Pacific region. It plays an important role in maintaining ecological balance, biodiversity, and developing regional ecological economy. Moreover, the water area is also an important water source in Shanghai, with many functions such as industrial and agricultural water intake, navigation, pollution, reclamation and ecology. The Yangtze estuary has become one of the important supports for Shanghai to build a world-class city. The development and utilization of water resources and the protection of water environment have become an important focus. However, for nearly half a century, due to human activities, the concentration and flux of heavy metals in the Yangtze River have increased significantly. Heavy metals are widely present in estuarine waters, suspended solids and sediments through complicated processes of sedimentation, burial, transportation, and transformation, posing serious threats to estuarine water environmental quality and water ecological security. The study on the spatial and temporal distribution and correlation of heavy metals in the Yangtze River estuary has important guiding significance for the water environment and water ecological protection in the region.

2.2. Fieldwork monitoring

From 2004 to 2018, in the Yangtze river estuary, six sections including Xuliujing (120°57′34″, 31°46′17″, 167.3 km from the estuary), Qinglonggang (121°14′01″, 31°51′36″, 137.7 km from the estuary), Qidonggang (121°40′12″, 31°43′12″, 72.7 km from the estuary), Shidongkou (121°24′44″, 31°28′25″, 109.7 km from the estuary), and Nangang (121°33′06″, 31°23′00″, 81.7 km from the estuary) and Beigang (BG, 121°37′00″, 31°30′34″, 82.7 km from the estuary) were continuously monitored monthly in the field (according graph). The Xuliujing section is a continuous monitoring section, and the other five sections are cruise sections. The monitoring frequency of each section differs. The sample data of Xuliujing section and Shidongkou section are the most complete, with 12 samples per year. The survey years of Nangang and Beigang sections are consistent, and the sample data is 4 / year. Qidonggang and Qinglonggang were tested in 2006 and 2010 respectively, with 12 sample data per year. According to the width of each section (whole river section, right river section and left river section), 2-5 water quality monitoring perpendiculars are arranged in each section in the first half of every month. Taking Xuliujing for instance, four monitoring vertical lines are arranged from north to south: 1# starting point is 2760 m away, 2# starting point is 3410 m away, 3# starting point is 4160 m away, 4# starting point is 4830 m away. Two sampling points are arranged for each
vertical line: the water surface (0.50m below the surface) and the bottom (0.50m above the bottom). Collecting water samples with 50kg heavy fish lead and stainless steel sampler. The water sample used for the determination of heavy metal copper content was filtered through a 0.45μm filter membrane after the sample was collected, discarding the initial 50-100ml solution, collecting 500ml filtrate, and the content of nitric acid was adjusted to 1% with nitric acid solution. The content of dissolved copper in water samples was determined by inductively coupled plasma atomic emission spectrometry [24-25].

2.3. Mathematical method
Rescaled Range Analysis (R/S) was used to analyze the variation trend of dissolved copper content at each position in the Yangtze estuary. This method is usually used to analyze the fractal features and long-term memory process of time series, mainly including the following steps [26-27]: (1) divide the time series \{X_i\} into A sub-interval of length n and determine the mean value of each sub-interval (2) calculate the cumulative deviation of each sub-interval corresponding to the corresponding mean, so as to determine the range R of each sub-interval; (3) according to the range difference and standard deviation S of each sub-interval, draw a graph of the pair and carry out a linear regression analysis of them with the least square method, and calculate the slope H of the regression equation, namely, Hurst index. H is an effective statistic of R/S analysis (0≤H≤1), and the trend is judged according to its size. When H = 0.5, it indicates that the time series is a random sequence with independent distribution, that is, the current change has no impact on the future. When 0≤H < 0.5, it indicates that the process has anti-sustainability, and the future change will be contrary to the general trend in the past. The closer H is to 0, the stronger the anti-sustainability will be. When 0.5 < H≤1, it indicates that the time series has a long-term dependence, that is, the future has the same change trend as the past. The closer H is to 1, the stronger the continuity. Pearson coefficient was used to calculate the correlation of DCu content in each section of the Yangtze river estuary. The specific formula is as follows [28-30]:

\[
\begin{align*}
    r &= \frac{l_{xy}}{\sqrt{l_{xx}l_{yy}}} ; l_{xx} = \sum_{i=1}^{n} x_i^2 - \frac{(\sum_{i=1}^{n} x_i)^2}{n} ; l_{yy} = \sum_{i=1}^{n} y_i^2 - \frac{(\sum_{i=1}^{n} y_i)^2}{n} \\
    l_{xy} &= \sum_{i=1}^{n} x_i y_i - \frac{\sum_{i=1}^{n} x_i \cdot \sum_{i=1}^{n} y_i}{n}
\end{align*}
\]

Where, r is the correlation coefficient of two-point potential, xi and yi are the annual mean sequence of dissolved copper content at the research point, respectively; n is the number of years, n=15; LXX and IYY are the sum of mean deviation squares of variables x and y, respectively; Ixy is the mean deviation product sum of variables x and y. The larger the absolute value of correlation coefficient is, the stronger the correlation coefficient is. The closer the correlation coefficient is to 1 or -1, the stronger the correlation degree is. The closer the correlation coefficient is to 0, the weaker the correlation degree is. According to experience, the relevant degree can be divided into the following situations: when 0.8≤ information platform r information platform <1, it is regarded as highly relevant; 0.6≤ information platform (r) ≤ 0.8 shall be deemed to be strongly correlated; 0.4≤ information server (r) ≤ 0.6 shall be regarded as moderately relevant. When information player (r) becomes less than 0.4, it indicates that the degree of correlation between variables is extremely weak, or it is regarded as irrelevant.

3. Results and discussion

3.1. Analysis of evolution trend of dissolved Cu in Yangtze estuary
Based on the change process of six typical sections, it can be seen that the concentration of DCu in the Yangtze estuary changes significantly with time, and the spatial difference between different points is significant (figure 2). The average DCu concentration of Xuliujing section from 2004 to 2018 was about 3.52 μg/L, which can be divided into four stages: (1) From 2004 to 2005, DCu increased significantly from 5.04 μg/L to 7.73 μg/L, with an annual increase of 53.37%. (2) From 2006 to 2008, DCu showed a significant decrease compared with the previous two years, with an average
concentration of about 3.28 μg/L. (3) From 2009 to 2013, the overall concentration of DCu was low and stable, with an average level of 2.73 μg/L. (4) From 2014 to 2018, the concentration of DCu increased to 3.30 μg/L on average, which was the same as the level in the second stage, but the fluctuation amplitude increased. In the past 15 years, the highest monthly DCu concentration in Xuliujing was about 17.8 μg/L (November 2005), which was 5.06 times the average level. The average DCu concentration in the flood season was about 3.80 μg/L, which was higher than that in the dry season by 20.20%. The evolution trend of DCu concentration in the two seasons was basically consistent with the average annual concentration. The evolution characteristics of DCu concentration in the stone mouth section are relatively simple. From 2004 to 2008, DCu concentration basically decreased year by year. The annual DCu concentration decreased from 9.58 μg/L to 3.54 μg/L, with an average decrease of 1.51 μg/L each year. From 2008 to 2016, DCu basically maintained a stable level of 2.35 μg/L, among which DCu increased in 2014 with an average annual concentration of 3.79 μg/L. From 2006 to 2008, the concentration of DCu fluctuated in Qidonggang section. In 2007, the average annual value of DCu reached 30.3 μg/L, which was the highest average annual level in the measured data. In 2008, the average annual DCu concentration dropped to 25.17 μg/L. The concentration of Cu remained at a low level from 2009 to 2013, and the concentration increased instantaneously in February 2011, with no significant fluctuations in other months. In 2014, the concentration showed a slow rising trend, and then basically remained stable. DCu concentration fluctuated slightly around 2.72 μg/L. The measured data of Qinglonggang section is relatively late. The existing results show that the average annual concentration of DCu in 2014 is relatively high, about 4.04 μg/L. From 2015 to 2017, DCu concentration was basically stable with an average concentration of 2.81 μg/L, which was about 69.56% of the load level in 2014. In 2018, the concentration of DCu increased again, with an average annual level of about 3.25 μg/L. From 2004 to 2007, the cross-sections of Nangang and Beigang increased first and then decreased, but the degree of fluctuation was different. The average annual DCu concentration in the Nangang and Beigang sections peaked at 8.02 and 9.33 μg/L in 2005, respectively, and then gradually decreased to 3.67 and 3.33 μg/L in 2007. From 2008 to the end of 2014, both sections maintained a relatively stable level. From 2015 to 2018, the concentration of DCu in both Nangang and Beigang sections increased, and the Nangang section increased significantly, with an average annual level of about 3.86 μg/L and the Beigang section about 2.95 μg/L.

**Figure 2.** Trend curve of Dissolved Cu load variation in typical sections of Yangtze estuary

The measured DCu concentration in different sections of the Yangtze river estuary directly reflects the fluctuation characteristics of DCu in the past 15 years. In order to further reveal the fractal characteristics of DCu time series of each section, the Hurst index of measured data series of each section was calculated by R/S analysis method, n selected 4, 6, 8, 12, 16, 18, 24, 36, 48 and 72
successively for calculation, and used MATLAB to realize the calculation of $\log_{10} \frac{R(n)}{S(n)}$ and $\log_{10} n$, and draw the scatter diagram and fitting curve of $\log_{10} \frac{R(n)}{S(n)}$ and $\log_{10} n$ as shown in figure3. The fitting results showed that except Qinglonggang, the Hurst index of the other five sections from 2004 to 2018 was greater than 0.5. The Hurst index of Nanggang and Qidonggang reached 0.9328 and 1.0 respectively, indicating that these two sections showed strong persistence. The Hurst index of Qinglonggang section is only 0.1515, which indicates that the section DCu has anti-persistence.

Figure 3. The correlation between $\log_{10} \left( \frac{R(n)}{S(n)} \right)$ and $\log_{10} n$ in each monitoring section

3.2. Spatial heterogeneity of dissolved Cu in the Changjiang Estuary

The Yangtze River main stream boundary, the Yangtze River estuary pollution discharge, the complex material exchange between the overlying water and sediment caused by runoff and tidal current, and the adsorption-desorption of the water-sand interface in the overlying water will cause the DCu concentration in the Yangtze River estuary. The spatial distribution is different, and this spatial differentiation also changes with time. Based on the measured data of six typical sections in the Yangtze River estuary, it can be seen that: (1) In 2004-2005, the concentration of DCu in the Xuliutun section was relatively low, with an average of 6.39 $\mu$g/L; the DCu load of the two sections of Nangang and Beigang was equivalent, with an average concentration of 6.50. Gg/L, 6.67 $\mu$g/L, a slight increase of 3.09% compared with Xuliujing; the Shidongkou section is located between the Xuliutun and NanBeigang sections, but its DCu load is nearly 9.25 $\mu$g/L, which is significantly higher than Xu Liuyi 44.86. %, higher than 40.53% of the north-south section; the difference in concentration of DCu in the southern branch should be related to the tailwater discharge of the Shidongkou Sewage Treatment Plant put into operation in 2002 (average daily processing capacity of 170,000 m$^3$/d). (2) From 2006 to 2009, the concentration of DCu in Xuliutun was about 3.24 $\mu$g/L, which was lower than 19.11% in the downstream branch. The average concentration of DCu in the south branch of the Changjiang Estuary was about 4.01 $\mu$g/L. The DCu concentrations in the South and North Port sections were 4.21 $\mu$g/L and 3.46 $\mu$g/L, respectively. The concentration of DCu in the section of Shidongkou was slightly higher than that of the two sections, with an average concentration of 4.34 $\mu$g/L. The DCu concentration of the Qidonggang section of the North Branch is as high as 22.85 $\mu$g/L, which is close to 5.7 times of the concentration of the southern branch. The significant difference of the DCu concentration between the north and the south should be related to three reasons: First, the mainstream
of the Yangtze River is diverted from the head of Chongming Island. The branch flow is significantly lower than that of the south branch. The oscillating water flow caused by runoff and tidal current increases the residence time of pollutants in the north branch. Second, the north branch accepts the second sewage treatment plant of Qidong City in the north bank and the sewage treatment of Chongming Chenjia Town in the south bank. The input of the tail water of the plant increases the DCu load by the pollution source. Thirdly, after the implementation of the narrowing project of the north branch, the ups and downs tidal currents of the north branch are reduced to varying degrees, which affects the convective diffusion efficiency of the pollutants. Taking the Chongtou section as an example, the average flow velocity of the high tide decreased from 1.24 m/s before the project to 1.08 m/s, which decreased by 16 cm/s. The falling tidal current decreased from 1.15 m/s before the project to 1.11 m/s, which decreased by 4 cm/s. (3) During 2010-2018, the spatial difference of DCu concentration in the Yangtze River estuary was weakened. The average concentration of the southern branch was about 2.63 μg/L, and the average concentration of the northern branch was slightly higher than that of the southern branch. The concentrations of Qidonggang and Qinglonggang were 2.98 μg/L and 2.86 μg/L, respectively. This spatial differentiation should be closely related to the Opinions on the Implementation of the Strict Water Resources Management System issued by the State Council in 2012 and the Water Pollution Prevention Action Plan issued in 2015. These important management measures have also reduced the boundary load of the basin's main stream and the regional emissions, thus reducing the DCu concentration in the Yangtze River estuary as a whole.

In order to further quantitatively analyze the correlation characteristics of DCu concentration at various points in the Yangtze River estuary, the Pearson coefficient was calculated by R3.6.0-GGally and the correlation test was performed (Fig. 4). Two significance levels of 0.01 and 0.05 were set, p < 0.05 means statistically significant, and p < 0.01 means significant correlation. The results showed that except for Qidonggang, the DCu between the other monitoring sections showed a significant positive correlation (r=0.447-0.867, p=0.000-0.007), and the DCu concentration between Beigang and Qinglonggang had a very similar variation. (r=0.867, p=0.000). The main cause of this strong correlation should be related to the main driving weighting factor of the two-section DCu load. The main stream of the Yangtze River is divided into the south branch and the north branch at the head of Chongming Island. The south branch is separated into the Nangang waterway and the Beigang waterway by Changxing Island and Hengsha Island. According to the distribution of regional pollution sources, the two sections of Qinglonggang and Beigang are not directly polluted by the upstream. Influence, the cross-section DCu load is mainly affected by the upstream mainstream flow boundary, while the Beigang waterway and the Beizhi Qinglonggang basically undertake upstream and semi-river waters. The same incoming water background leads to a better correlation between the two sections. The Kailu Port DCu showed different correlations with the remaining monitoring sections. For example, there is a significant positive correlation between the DCu concentration of the Qidonggang section and the Shidongkou section (r=0.464, p=0.004), which should be attributed to the relatively high load caused by the external sewage treatment plant discharge; and the Qidonggang section and the Beigang section. The results of DCu concentration test showed that the two did not show correlation (r=0.189, p=0.250). The main reason should be summarized into three aspects: (1) Qidonggang section is directly affected by regional sewage discharge, while Beigang section is mainly The source of DCu in the two sections is different due to the upstream water flow; (2) the significant uneven distribution of water distribution in the north and south branches leads to the difference in the distribution of sediment particles, which directly affects the DCu content of the section; (3) The distance between Qidonggang, Beigang and the estuary gate is about 12km. The material transport under the action of tidal current will also lead to different DCu content.
Figure 4. Correlation of dissolved Cu concentration in typical sections of Yangtze estuary

4. Conclusion and Suggestion
Taking the Yangtze river estuary as the research area, based on the long series of field measured data, the evolution law of DCu content in six typical sections of the Yangtze river estuary in the past 15 years was analyzed by the method of Rescaled Range Analysis (R/S), and the spatial differentiation law and correlation characteristics of DCu in different stages were quantitatively revealed. The results show that the concentration of DCu in the Yangtze river estuary changes obviously with time, and the spatial difference between different sections is significant. The fluctuation characteristics of Xuliujing section are relatively complex. The average DCu concentration is about 3.52 μg/L, while the highest monthly DCu concentration can reach 17.8 μg/L. The evolution characteristics of DCu concentration in Shidongkou, Nangang and Beigang sections are relatively simple. From 2004 to 2008, the concentration of DCu was relatively high and then gradually stabilized. The oscillation amplitude of DCu concentration in Qidonggang section is the most significant. In 2007, the annual average of DCu reached 30.3 μg/L, which is the highest annual average in the measured data. Except Qidonggang, the DCu in the other five monitored sections all show a significant positive correlation, among which the DCu concentration between Beigang and Qinglonggang has an extremely similar change rule. The main reason for this strong correlation should be related to the main driving weight factor of DCu load in the two sections. The results of this paper are of certain reference significance for mastering the spatial and temporal distribution of dissolved heavy metals in the Yangtze estuary, as well as for regional water environment protection and heavy metal pollution prevention and control. However, this paper focuses on the analysis of dissolved heavy metals, and further discussion should be made in the following two aspects: (1) what is the distribution of granular heavy metals under the complex hydrodynamic conditions of runoff and tidal current in the Yangtze river estuary? How does it interact with the concentration of dissolved heavy metals? (2) heavy metal load in the Yangtze estuary is affected by many factors, such as upstream inflow boundary, regional pollution discharge, sediment release, atmospheric dry and wet deposition and biological enrichment, etc. How these factors affect the weight of heavy metal load still needs further study.
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