30+ year evolution of Cu in the surface sediment of Lake Poyang, China

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Abstract: Poyang Lake, the largest freshwater lake in China, is increasingly experiencing Cu crises. Combining field data, laboratory experiments, and long-term simulations, we retrieved 30+ year evolution of Cu in surface sediments (SCu). SCu evolution between 1983-2015 may be separated into two distinguishable periods. During the first period (1983-2003), SCu underwent a continuous increase at a mean rate of 1.80 mg/kg/yr; however, since 2003 it displayed a stepwise reduction tendency. The average SCu concentration of the entire lake in 2015 fell to 54.26 mg/kg, which is approximately 30.01% lower than that in 2003. The operation of Three Gorges Dam (TGD) markedly altered the river-lake relationship, pulled more deposited Cu along with sediment out toward the Yangtze River, and made the regions of high SCu emanate from the southeastern lake extend northwestward between 2003-2015. SCu in the reserves showed significant inter-annual variations, with the exception of the Jiangxi Whitebait Spawning Reserve (JWSR), where SCu generally has not been significantly impacted and has displayed no departure from the 30+year mean of 30.57 mg/kg. The National Germplasm Reserve (NGR) and Nanjishan National Nature

Reserve (NNNR) were detected with the highest SCu, with the peak concentrations, respectively, of 123.15 mg/kg and 103.1 mg/kg.

Key words: Poyang Lake; Heavy metal; Cu; Surface sediment; Numerical simulation

1. Introduction

Heavy metal pollution in lakes has become a global environmental and public health concern (Abdullah and Royle, 1792; Jerome and Henry, 1983; Zahra et al., 2014). Rapidly growing anthropogenic activities, such as city construction, industrial and agricultural development, and exploitation of mineral resources, have made the hazard of heavy metals in lakes a common and far-reaching problem (Guo et al., 2015; Ramamoorthy and Kushner, 1975). Due to the toxicity, abundance, persistence, and subsequent bio-accumulation of these metals, lakes are faced with a serious threat to their roles in freshwater supply, fishery, biodiversity preservation and ecological balance maintenance (Robert et al., 1972; Gunvor et al., 2005; Lee et al., 2000; Miller et al., 2014; John and Barbara, 2003). Examples can be observed in Lake Victoria (the largest of the African Rift lakes) (Makundi, 2001), Lake Winnipeg (Canada) (Torigai et al., 2000), Lake Erie (North America) (Nriagu et al., 1979), Lakes Biwa and Kasimagaura (Japan's largest lakes) (Mito et al., 2004), Lake Taihu (the 3rd largest freshwater lake in China) (Yin et al., 2011), Lake Balaton (the largest lake in Central Europe) (Nguyen et al., 2005), and Lake Moreno Oeste (the mountain lake in Patagonia) (Guevara et al., 2010).

During transport in lakes, heavy metals may undergo numerous changes in their speciation due to dissolution, precipitation, sorption, and complexation phenomena, which affect their behavior and bioavailability (Islam et al., 2015). However, despite these complicated processes, heavy metals entering lakes from diverse sources are finally deposited in the sediments, apart from biological consumption (Linda et al., 2007). However, these particulate phase metals may not be permanently sequestered in sediments. They may be resuspended by wave and current-induced bottom shear stress, biotic and abiotic speciation, and entrance to the trophic web by benthic organism, causing secondary contamination and hazards to overlying water, and critically degrade the aquatic system (Suresh et al., 2012; Akcay et al., 2003; Frederick and Robert, 1981). Hence, sediment as an integral and dynamic part of the lake is both a carrier and a potential source of heavy metal contaminants. Given that most heavy metals eventually accumulated in the surface sediments within 5-10 cm (Zahra et al., 2014; Zhang et al., 2015), which dominate the release process, determining the heavy metal content in sediments is an essential step to understanding their potential toxicity and threat to ecosystems.

Poyang Lake, with an area of 3583 km^2 and a volume of 27.6 km^3 on average, is located at the south bank of the Yangtze River in Jiangxi Province, China (Fig. 1). It is the largest freshwater lake, as well as the most typical river-connected lake, in China. The lake receives water from five rivers (Raohe, Xinjiang, Fuhe, Ganjiang and Xiuhe) and drains into the Yangtze through a narrow outlet to the north (Feng et al., 2013; Wu et al., 2007). Due to the river-lake interaction, Poyang Lake is characterized by marked intra- and inter-annual variations of suspended sediment load (Wang et al., 2014; Wang et al., 2015). The lake is one of the most important ecological regions recognized by the Global Natural Fund and is one of the six wetlands having the most abundant biodiversity, hosting millions of birds from over 300 species (Lu et al., 2012; You et al., 2015; Han et al., 2015; Zhang et al., 2012). In particular, it is vital for conservation of the endangered Siberian crane as more than 95% of its world population congregates here during the winter. However because some mines are located adjacent to the lake, such as Dexing Copper Mine, Asia's largest copper base, the lake has been found to have an increasingly heavy metal load due to expanded exploitation, which contaminates the aquatic environment and damages its ecological function.

A number of studies on heavy metal in Poyang Lake have been conducted since the 1980s. Among the documented works are those of Qian et al. (1985), who evaluated the pollution level of heavy metal in sediment by the root mean square pollution index; Chen et al. (1989), who used an equilibrium adsorption model to describe the heavy metal partitioning in sediment samples from the lake; and Yuan et al. (2011), who estimated the distribution of heavy metals in Poyang Lake based on eight sedimentary cores. These indices provided useful information to local managers and decision makers. However, most of them were traditionally performed by taking ship-borne sediment samples and analyzing these samples in a laboratory. Due to the coarse sampling frequencies and limited sampling points, it is difficult to comprehend the temporal and spatial metal load for an entire lake, which are usually featured by accumulation that is uneven in both time and space scales. Moreover, many studies dealing with particulate metals in the lake suffer from not explicitly considering the different particle-size classes, which have important consequences for entrainment, transport and deposition of heavy metals.

In this work, we selected Cu, which was detected with the highest load in the lake, as the study item. The objectives were to (1) investigate the Cu transport mechanism associated with varied grain-size sediments between the overlying water and the surface deposited sediment, (2) develop and validate an improved metal model that places particular emphasis on Cu transport with size-fractionated sediments, (3) use numerical simulation to quantitatively reveal the spatial and temporal evolution of Cu in the surface sediment during the period 1983-2015 when the surrounding mines were over-exploited, and (4) evaluate the influences of underlying causes, such as anthropogenic discharge, Three Gorges Dam (TGD) emplacement, altered river-lake interaction and atmospheric deposition, on the 30+ year trend in the variation of surface deposited Cu in Poyang Lake. This study may provide insights for policy makers who are attempting to prevent heavy metal pollution and improve the water quality of inland freshwater lakes.

2. Material and methods

2.1. Data Acquisition and Processing

Historical data for a long-term simulation experiment were collected from various sources. The boundary data between 1988-2015, including water quantity, suspended sediment, and Cu concentration, were determined according to the measured data acquired from the Jiangxi Province Hydrology Bureau. Hydrology and suspended sediment data from 1988−2015 were obtained from the “Hydrological Yearbook of the Yangtze River Basin, China”. As the regularly monitored Cu concentrations between 1983 and 1987 were absent, the input data were refined by referring to the irregular field observations at the inlet areas of Raohe, Xinjiang, Fuhe, Ganjiang and Xiuhe, with the help of the Poyang Lake Hydrology Bureau. Information needed for establishing model geometry was determined from a remote sensing image acquired on Oct. 5, 2007 (Lei et al., 2010). The initial Cu contents in surface sediment were derived from the 1983 survey (Qian et al., 1985). Lake bottom data collected in 1980 were used in conjunction with the 1990 data to aid in giving the initial terrain elevation (Wu et al., 2015; Xiong, 1990). Monthly rain falls during the 1983 to 2015 period were collected from five rainfall stations around Poyang Lake (Hukou, Xingzi, Duchang, Poyang, and Jinxian) (Li et al., 2012). The variation in wind data from 1983-2004 was discretized into 12 classes for each month, with their associated frequencies of occurrence, on the basis of the continuous wind data from 2005-2015, obtained from the online database of Weather Underground. The Cu concentrations in surface sediment, which were obtained from the field investigations of 1985, 2003, and 2013, were utilized for model calibration and validation (Huang, 2005).

2.2. Laboratory Experiment

Due to the frequent water exchange between Poyang Lake and the external rivers, the upstream five rivers and the downstream Yangtze River, both the sediment concentration and the particle size were uneven distributed in the lake, and fluctuated within a year and between years. As sediments with varied grain-size have different consequences for Cu transport between the surface bed and the overlying water. The laboratory experiment was conducted to quantitatively explore these processes and provide the deposition and suspension parameters for the subsequent numerical study.

Given the hours required to establish equilibrium in Cu transport between the bed sediment and the overlying water, the annular flume in the Molecular Biology Laboratory of Nanjing Geography and Limnology Institute, Chinese Academy of Sciences, was utilized (Fig. 2). The device is composed of a flume and top lid, which were made of acrylic material and had outer and inner diameters of 120 cm and 80 cm, respectively. With the digital controlling system, the lid can rotate independently and move up or down as required. The annular water channel is 20 cm in width and 41 cm in depth. Several sample outlets are placed at different heights on the external wall of the flume. Driven by the continuously variable-speed motor, the rotation of the flume and top lid in opposite directions can generate water currents under the effect of shear stress. The curvature of the flume induces the centrifugal force on the water current, generating outward secondary flow. However, as the top lid rotates in the direction opposite to that of the flume, it will generate inward secondary flow. As centrifugal force is related to the rotation rate, adjusting the rotation speeds of the top lid and flume allows the centrifugal forces to cancel each other out and thus eliminate the secondary flow. Prior to the experiment, a small amount of sawdust was used as a tracer indicator for calibrating the rotation rates of the top lid and flume to generate expected currents.

Because the sediments of Poyang Lake are marked by a prevalence of the particles in the range of 3.79 -63.0 μm (Cui et al., 2013; Zhang et al., 2014), three sediment size classes, fine-silt (3.79-16.8 μm), medium-silt (16.8-32.57 μm), and coarse-silt (32.57-63.0 μm), were determined for the experiment. The separation of sediment into different sizes was finished with a modified elutriator apparatus (Follmer et al., 1973). Regarding each size class, 6 group experiments were carried out with varied initial suspended sediment concentrations (flood season 0.43 mg/L, dry season 0.78 mg/L) and different initial bed Cu content (low 30.7 mg/kg, medium 62.6 mg/kg, and high 98.5 mg/kg). In each group test, the disturbance intensities were set as 0 $m \cdot s^{-1}$, 0.1 $m \cdot s^{-1}$, 0.2 $m \cdot s^{-1}$, 0.3 $m \cdot s^{-1}$, 0.5 $m \cdot s^{-1}$, and 0.7 $m \cdot s^{-1}$, referring to *in situ* velocities. During tests, water samples were regularly taken from the outlets on the flume wall to examine the processes of dissolved, suspended and deposited Cu. The samples were digested with HCl-HNO_3-HF-$HClO_4$ solution and analyzed by ICP-MS. Considering the gap between the laboratory current and the *in situ* disturbance dominated by the flow current, as well as the wind and wind-induced waves, the bottom shear stress was calculated for results and interpretation.

2.3. Model description

The model was built to yield realistic and accurate simulations of Cu transport in Poyang Lake over a 30+ year period from 1983 to 2015. To reasonably compute the effects of particle size variation, three sediment size classes, fine-silt (3.79-16.8 μm), medium-silt (16.8-32.57 μm), and coarse-silt (32.57-63.0 μm), were simulated in the present study. Poyang Lake can be assumed to be vertically well-mixed, and the general three-dimensional equations were allowed to be approximated by two-dimensional, vertically integrated equations on the basis of the following facts (Periáñez, 2009). First, water depth in the lake experiences a frequent fluctuation in a year, and the mean depths in dry and flood season are respectively 2.8 m and 6.5 m. Second, the huge water surface 3583 km^2 enlarges the width-depth ratio to higher than 1.4×10^4 that prefers 2-D simulation. Thirdly, the close river-lake relationship diminishes the water exchange period to 6.8 d- 22 d in the lake, which hinders stratification. The model developed here consists of four sub-models. First, a hydrodynamic module provides currents over the domain. Second, a size-fractionated sediment transport model provides concentrations of size-diverse suspended sediments over the lake. The third sub-model is the metal transport module, which builds upon a combined description of advection/diffusion plus deposition/resuspension reactions of metals between the surface bed sediments and overlying water. The fourth sub-model is a morphological module that updates the lake bathymetry over time. The metal module is coupled to the hydrodynamic model and sediment transport model, and metal changes associated with each class are summed to yield the concentrations of dissolved, particulate and deposited metal. Hydrodynamic, sediment transport, and morphological model equations are standard and available in the literature (Periáñez, 2005; Wang et al., 2015). The improved metal equations can then be expressed as follows:

$$\text{Dissolved:}\ \frac{\partial hC_d}{\partial t}+\frac{\partial hC_d u}{\partial x}+\frac{\partial hC_d v}{\partial y}=\frac{\partial}{\partial x}\left(E_x\frac{\partial hC_d}{\partial x}\right)+\frac{\partial}{\partial y}\left(E_y\frac{\partial hC_d}{\partial y}\right)-k_{ai}C_d+k_{bi}S_iC_{pi}+P_d$$

$$\text{Suspended:}\ \frac{\partial\left(hS_iC_{pi}\right)}{\partial t}+\frac{\partial\left(huS_iC_{pi}\right)}{\partial x}+\frac{\partial\left(hvS_iC_{pi}\right)}{\partial y}=\frac{\partial}{\partial x}\left(D_{xi}h\frac{\partial(S_iC_{pi})}{\partial x}\right)+\frac{\partial}{\partial y}\left(D_{yi}h\frac{\partial(S_iC_{pi})}{\partial y}\right)+k_{ai}C_d+\lambda_{si}C_b-k_{bi}S_iC_{pi}-\lambda_{di}S_iC_{pi}$$

$$\text{Deposited:}\ H_i\frac{\partial C_b}{\partial t}=\lambda_{di}S_iC_{pi}-\lambda_{si}C_b-\psi C_b$$

where h is water depth; t is time; u and v are the depth-averaged velocity components in x and y dimensions; C_d is the concentration of metal in the dissolved phase; E_x and E_y are the dispersion coefficients of dissolved metal in x and y dimensions; K_{ai} governs the transfer rate from liquid to the particulate phase, associated with the i-class sediment (i=1,2,3, respectively representing fine, medium and coarse-silt); K_{bi} governs the inverse process; and S_i and C_{pi} are, respectively, the concentration of the i-class sediment and the concentration of the metal bounded to it. P_d represents the contribution of atmospheric deposition; D_{xi} and D_{yi} are the dispersion coefficients of particulate metal on the i-class sediment in the x and y dimensions; λ_{si} governs the metal resuspension rate of i-class sediment from the surface bed to the overlying water, while λ_{di} governs the inverse deposition rate, which is obtained from the laboratory results. C_b is the metal concentration of surface deposited sediment. ψ is incorporated to reflect the migration of metals to the deep sediment because the metals deposited on the sediment surface will be buried by particle deposition.

2.4. Numerical Simulation Experiment

Simulation experiments were executed for two purposes as follows: to verify the capability of the model for describing long-term Cu dynamics in Poyang Lake and to extend the numerical simulation experiments for the purpose of replicating the long-term simultaneous fluctuation in Cu concentrations in the surface sediments for the entire lake. As the direct calculation of all different-scale processes on a given grid system poses a formidable computation challenge (large memory and computation time requirements), multi-scale concepts aiming to provide a solution that solves for different scale processes are integrated during simulation, e.g., the hydrodynamic time step was set at 30 s to insure numerical stability, while the morphological time step was increased to 120 s to represent the time required for the bottom change to be sufficiently significant to justify a bathymetry update. The computed area includes that of the entire lake, 3583 km^2, and extends northward to the Yangtze River, 28 km upstream and 15 km downstream. The rivers of Raohe, Xinjiang, Fuhe, Ganjiang, Xiuhe and Yangtze River are recognized as calculation boundaries. The spatial resolution of the computation was set to 700 m×700 m, giving a total of 7533 nodes and 6239 quadrilateral elements for the modelling area. The governing equations were solved in a framework of finite-volume method, and the material fluxes crossing the element interfaces were determined by the flux vector splitting scheme (Wang et al., 2015). The simulations were run from 1983 to 2015 with a display interval of one year.

3. Results

3.1. Laboratory Coefficients

The deposition and resuspension rates, λ_d and λ_s, obtained with varied grain-size sediments are reported in Fig.2(C-E). Grain size has important consequences for Cu exchange flux between the top surface layer and the overlying water. In case of gentle bottom shear stress (<0.05N/m^2), Cu transport is marked by the prevalence of deposition onto the surface sediment, and λ_d generally rises with the increase in grain size and decrease in disturbance intensity. For instance, at 0.04 N/m^2 of the flood season test, λ_d of fine, medium and coarse-silt was, respectively 0.037, 0.056, and 0.067. An enhanced deposition flux was observed in the finest fractions (3.79-16.8 μm) from 0 to 0.01 N/m^2, which should be contributed to by the fine-silt flocculation promoted by suitable dynamic conditions. Comparison of λ_d subjected to different sediment concentrations of the flood and dry season indicated that a higher initial concentration produces a larger Cu deposition flux. Once the bottom shear stress exceeded 0.08 N/m^2, the surface sediment layer was eroded, and Cu was exchanged in the inverse direction. The rising disturbance further led to an increasing λ_s, e.g., from 0.09 to 0.50 N/m^2, and the fraction of medium-silt was strengthened from 0.065 to 0.38. In a given disturbance level, λ_s fell with grain size rising, but it did not seem to be sufficiently significant. Based on the transport flux analyzed in tests with varied initial sediment layers, λ_s shows a smooth increase as the background Cu content increases. λ_s in high pollution sediment was approximately 1.21 times higher than that in low pollution sediment.

3.2. Model Performance

Parameters in the hydrodynamic and sediment module, including the bottom friction factor, water kinematic viscosity, horizontal diffusion coefficient, and the critical shear stress for the deposition of varied grain-size sediments, were adjusted to meet the accuracy requirement. The simulated results could reasonably represent the observed water-sediment conditions in Poyang Lake. The main parameters in the metal model that were adjusted to produce the best model performance were the deposition and resuspension rates, λ_d and λ_s. Cu concentrations at 24 sites from Poyang Lake (Fig.1), collected in 1985, 2003 and 2013, were compared to surface Cu contamination levels calculated by the model for calibration and validation (Fig.3). Good correlation between the computed and the observed data could be achieved by using the values obtained from laboratory experiments. The mean of the absolute value of the relative error (ARE), i.e., ARE=|Calculated- measured|/measured, for the calibration and validation period was 18.6%. The model was capable of scientifically reflecting Cu fluctuation in the surface sediment layer of Poyang Lake.

3.3. Temporal-Spatial Pattern Trend

SCu evolution for 1983-2015 could be separated into two distinguishable periods (Fig.4). From 1983-2003, the mean concentration of the entire lake continuously increased from 31.96 mg/kg to 77.53 mg/kg. Relative to the increase rate of 0.77 mg/kg/yr for the period (1994-2003), the first decade 1983-1993 had a more ambitious accumulation rate of 2.83 mg/kg/yr. However, after 2003, the lake exhibited a general decrease in SCu. The mean SCu in 2013 and 2015 fell to 60.72 mg/kg and 54.26 mg/kg, respectively, 21.69% and 30.01% lower than that in 2003. SCu within the lake shifted with spatial locations, as well as years. SCu in the north lake underwent a rapid increase from 36.62 mg/kg to 59.5 mg/kg in the first decade, smoothly rose to 67.3 mg/kg from 1994-2003, and maintained a stable level since 2004. The central lake was characterized by the most distinct spatial disparity, with a trend toward decreasing SCu from the southeast area to the northern open-lake area. SCu in the near-shore area of Raohe and Xinjiang was featured with the highest content and increasing rate, which reached 102.85 mg/kg in 2003 at a mean rate of 3.12 mg/kg/yr. As SCu decreased with increasing distance from the southeast lakeshore, SCu in the north central lake was approximately 43.7% lower than in the south, but it still exhibited a steady increase in the first 20 years, with rates of increase of 3.49 mg/kg/yr between 1983 and 1993 and 0.99 mg/kg/yr between 1994 and 2003. Since 2003, SCu in the central lake displayed a stepwise reduction tendency. Comparisons with data from 2003, 2008 and 2013 could significantly indicate regions of high SCu emanating from the south central lake and extending northwestward. Until 2015, the mean SCu in central lake fell to 62.3 mg/kg, reduced by 23.5% compared to 2003. The rapid SCu increase in south lake also occurred in the first decade 1983-1993, and in the first half of 1990s, the mean SCu could amount to 36.56 mg/kg. However, during the period of 1994-2015, accumulation of Cu in surface sediment was generally maintained at a stable level, particularly for some tail-lake areas.

Fig.5 provides insight into the 30+ year SCu fluctuations in the 6 reserves. They showed significant inter-annual variations, with the exception of JWSR, where SCu generally had not been significantly impacted and displayed no departure from the 30+year mean of 30.57 mg/kg. As for the entire lake, these reserves experienced enhanced Cu accumulation in the first 2 decades, up to a dividing line found between 2002-2003. NGR and NNNR exhibited the highest SCu, with the peak concentrations, respectively, of 123.15 mg/kg and 103.1 mg/kg. There was also a decelerating trend in SCu in the latter decade, the rate of increase falling from 4.12 mg/kg/yr to 1.51 mg/kg/yr. Although SCu in Yangtze Finless Porpoise Reserve (YFPR), Poyang National Nature Reserve (PNNR) and Poyang National Wetland Reserve (PNWR) did not display an intense rise, the stable increase over 20 years had increased their levels above the background value (Wu et al., 2014). Their peak values ranged from 72.5 to 83.6 mg/kg, with mean rates of increase, respectively, of 3.48, 2.77, and 3.13 mg/kg/yr. Since 2002, SCu in the reserves showed a decreasing trend. The rates of decrease in NGR and NNNR were, respectively, 2.42 and 1.80 mg/kg/yr, while the mean value for YFPR, PNNR, and PNWR was only 0.85 mg/kg/year. Despite the overall decrease in the latest decade, a short increase in SCu could still be detected from 2007 to 2009; e.g., after falling from 76.30 mg/kg in 2003 to 59.43 mg/kg in 2007, SCu in PNNR returned to 73.45 mg/kg in 2009. The variation coefficients in NGR and NNNR were lower than that in YFPR, PNNR and PNWR, with means, respectively, of 23.7% and 33.5%. SCu in JWSR essentially fluctuated slightly over the 30+ years, with a variation coefficient of 20.7%.

4. Discussion

Anthropogenic input was the dominant source of SCu in Poyang Lake. The presence of surrounding mineral deposits and the expansion of mining activities played a key role in Cu increasing in the contributing rivers, as well as in SCu accumulation in Poyang Lake from the 1980s to 2000s. The total Cu transported into the lake was enhanced from 446.88 t to 917.08 t from 1983-1993 and ranged from 722.28 t to 1000.10 t from 1994-2003. Sediments from the contributing rivers were the principal vehicle for Cu transport. In this 2-decade period, the annual averages of sediment feeding Poyang Lake were, respectively, 14.19 Mt and 17.10 Mt. Despite the fact that the amount of sediment transported into the lake during the latter decade was 17.08% higher, Cu input did not correspondingly increase as ambitiously as the past decade but exhibited a decelerating trend because the upstream anthropogenic sources from cooper manufacturing facilities were, to some extent, prevented by the efforts regarding qualified environmental protection measures from the second half of 1990s. The mean annual increase rates of Cu input before and after the middle of 1990s were respectively 47.02 t/a and 27.78 t/a. This may explain the disparities in SCu increase rates in the 20-year period. Influenced by the intense industrial activities in the watersheds of the major tributaries, especially at Yongping and Dexing Copper Mine, Xinjiang and Raohe, which accounted for 22.1% of the total sediment load, contributed more than 35.6% of the Cu input to Poyang Lake due to their high Cu concentrations in both dissolved and suspended forms (Xiong, 1990; Cui et al., 2013). This knowledge suggested a shifting trend in SCu from the southeastern area outward into the main lake.

The emplacement of TGD in 2003 may have the most important consequences for the stepwise reduction trends in SCu during the most recent decade. The means by which it affected SCu evolution in Poyang Lake could be distinguished as follows. First, the dam reduced the sediment flowing into Poyang Lake, which strongly participated in Cu dynamics. Since 2003, less sediment entered the lake, for an annual mean of 12.3 Mt, which was approximately 21.35% lower than that from 1983-2002. Second, the particle size distribution in the lake changed since the dam's inception (Zhang et al., 2014; Mei et al., 2015). The proportion of fine and medium-silt was increased with decreasing coarse-silt, which made Cu remain suspended in the overlying water and decelerated its deposition into the lake bed. Third, the water volume and sediment discharges of the Yangtze River had both fallen after the operation of TGD, which further altered the river-lake interrelationship. The reduced jacking influence and backflow of the Yangtze River, combined with the increasing outflow from Poyang Lake, converted it from a depositional to an erosional system in the most recent decade. More deposited Cu, along with significant amounts of sediment, were pulled out toward the external river. This process supported the high-SCu region evolution from 2003-2013. Moreover, there were two additional explanations for the tendencies in SCu noted during the recent decade. One was reduced precipitation. Relative to the period of 1983-2003, the mean precipitation of 2004-2015 was only 1443 mm/yr, showing a fall from the last 20-year mean of 1693 mm/yr. Although atmospheric sources did not play a dominant role in SCu accumulation, this evident fall may, to some extent, have contributed to the recent deceleration tendency. Another was attributed to the implementation of the "Mountain River Lake Project in the Poyang Lake basin" proposed by the local provincial government. Two activities, soil and water conservation and reservoir construction, were advocated by the project (Huang et al., 2012). The increased forest coverage, combined with the operation of 14 large and more than 200 medium sized reservoirs in the basin, had assisted in cutting down the net Cu flux into Poyang Lake.

Hydrologic conditions interacted with, and often dominated, the spatial SCu evolution within the lake. Water currents in Poyang Lake could be classified by three distinguishable types: (1) Gravity-style current, the major type induced by the lake bottom sloping from south to north. This current permitted and helped the regions of high SCu extending northwestward; (2) Jacking-style current, produced by the essentially flat surface between the north lake and Yangtze River. The mean water velocity sharply reduced in this case, creating a better condition for SCu increase, especially in the central lake, where the sediment-carrying capacity was markedly lower. (3) Backflow-style current, mainly generated by the higher Yangtze River water level between July and September. This current was observed with the lowest frequency but had significant impact on SCu in the north lake. After the operation of TGD, Jacking-style and Backflow-style were both decreased in frequency and intensity in recent years, which pushed more Cu from the lake to the external river. Regarding some tail-lake areas, the hydrodynamic conditions did not vary as much as in the main lake. The older water age consequently resulted in insignificant SCu fluctuation over the 30+ years of data.

These results could show the driving mechanisms for the spatial and temporal SCu evolution in Poyang Lake. However, there are several uncertainties that may need further investigation. First, at the present work, the interactions of Cu with some ecological processes were not considered. The approaches that these processes affect Cu transport can be summed up to the following two aspects. One is that biological consumption and bioaccumulation will reduce Cu load in the overlying water and surface sediment. The other is that some processes, such as biotic and abiotic speciation, and benthic organism activities, will influence Cu exchanging flux between the sediment and the water column. Simplification of these interactions indeed set up a barrier to simulate the field actual Cu transport processes, but it will not affect the general trend of the temporal and spatial SCu evolution in Poyang Lake.

Considering the consumed and bio accumulated Cu, the results gained here may be overestimated. Second, due to the technique used for long-term computation, emphasis was placed on the gross Cu transported with varied grain-size sediments between the overlying water and the surface deposited sediment, and less attention was paid to the microscopic processes, which Cu may experience among the dissolved, particulate and deposited forms, such as complexation, coagulation, adsorption/desorption, formation of hydroxide colloids, and other physicochemical reactions. This may have resulted in bias with respect to understanding SCu evolution. Third, a lack of monitored data made the model suffer by not accurately incorporating the influence of atmospheric sources, which was recognized as a constant at the present work. Despite the discrepancies that may have been be generated by the above uncertainties, our study provided some evidence regarding SCu evolution tendency in Poyang Lake.

5. Conclusions

Combining field data, laboratory experiment and numerical simulation, we explored the SCu evolution in Poyang Lake during the past 30+ years. SCu experienced a continuous increase rate of 1.80 mg/kg/yr between 1983 and 2003, and then displayed a stepwise reduction tendency. Compared to the mean SCu content in 2003, the value in 2015 fell to 54.26 mg/kg, which is approximately reduced by 30.01%. From 2003 to 2015, the operation of TGD had an important consequence for SCu distribution in Poyang Lake. The altered river-lake relationship, pulled more deposited Cu along with sediment out toward the Yangtze River, and made the regions of high SCu emanate from the southeastern lake extend northwestward. Apart from JWSR, SCu in the reserves showed significant inter-annual variations. NGR and NNNR were characterized by the highest SCu, with the peak concentrations, respectively, of 123.15 mg/kg and 103.1 mg/kg. This work may provide insights for policy makers who try to prevent metal pollution and improve the water quality in Poyang Lake and encourage large-scale and long-term heavy metal research on huge inland freshwater lakes.

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Figure 1
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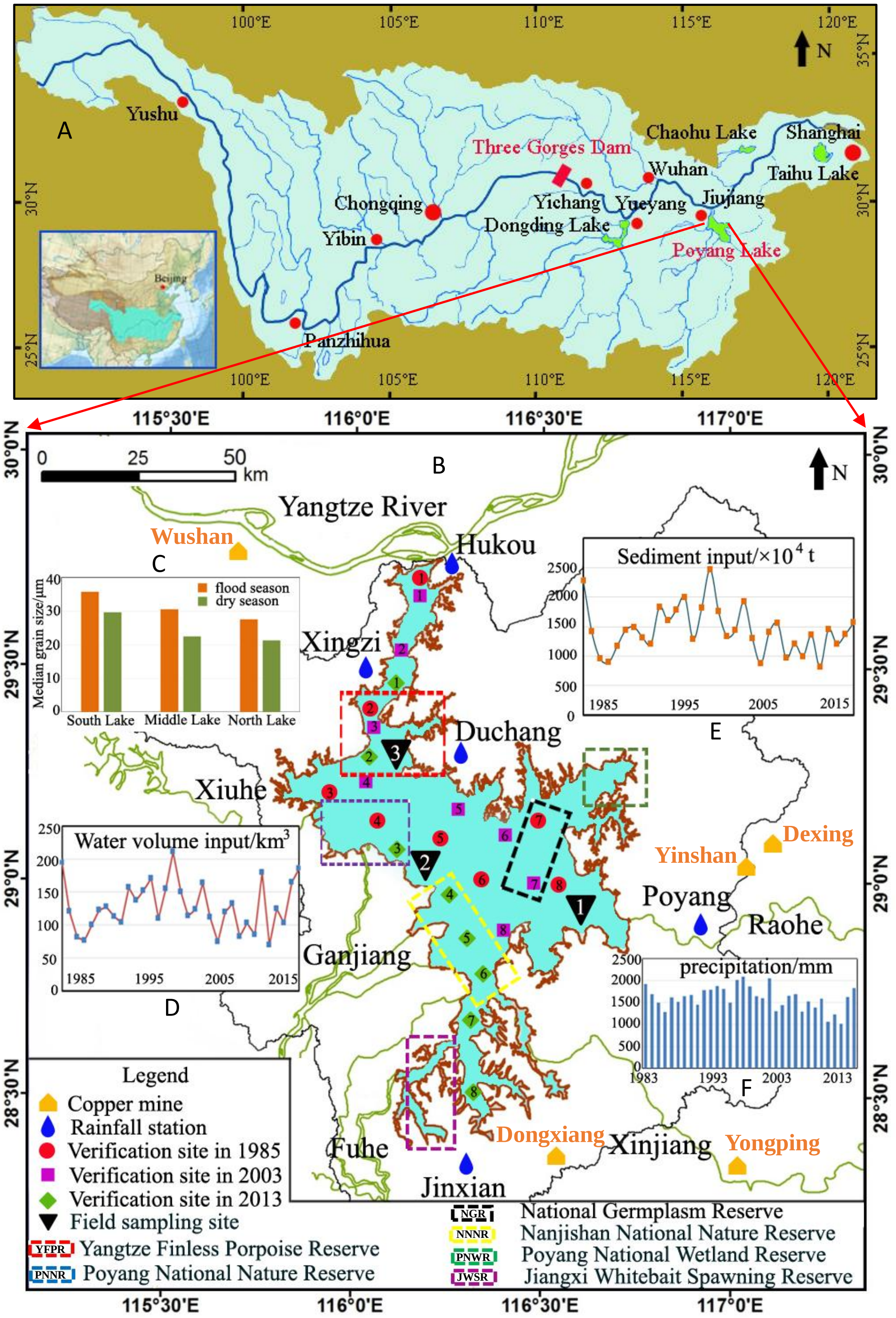

Fig. 1. (A) Location of Poyang Lake in the Yangtze River Basin and PR China. (B) Map of Poyang Lake. Locations of mines, reserves, and field investigation sites are indicated. (C) Grain-size variation in the lake. Inserts (D), (E) and (F), respectively, represent the processes of water volume input, sediment input, and precipitation from 1983-2013

Figure 2

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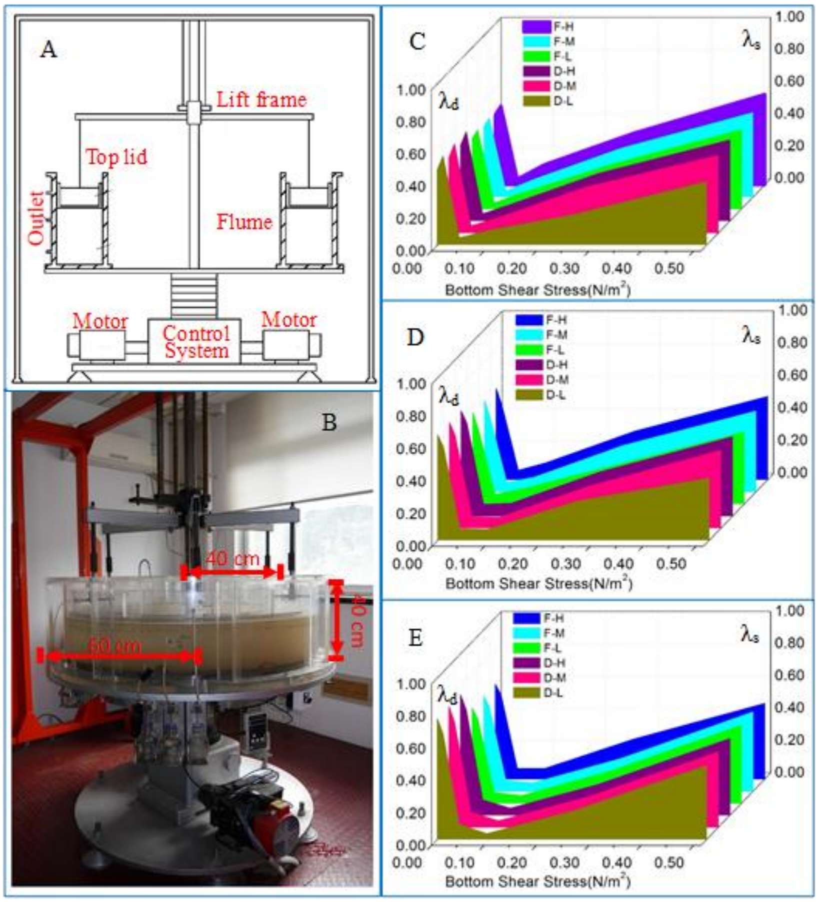

Fig. 2. (A) and (B) are, respectively, the schematic diagram and live photograph of the annular flume. (D), (E) and (F) are, respectively, the deposition and resuspension rates of Cu associated with fine, medium, and coarse-silt under varied shear stress.

Figure 3
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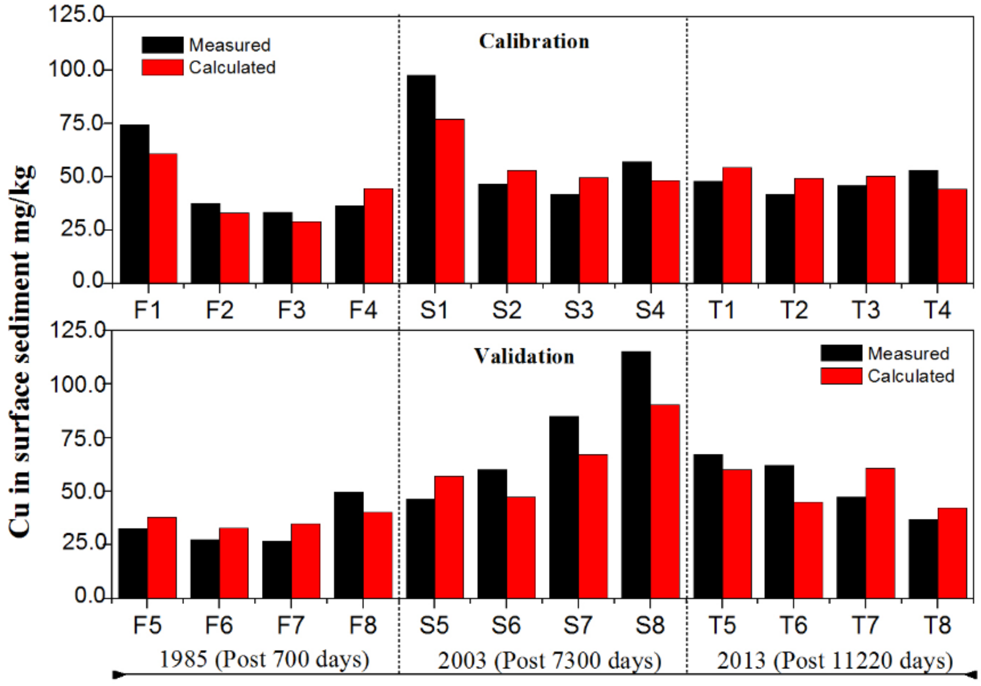

Fig. 3. Comparison between the measured and calculated data

Figure 4
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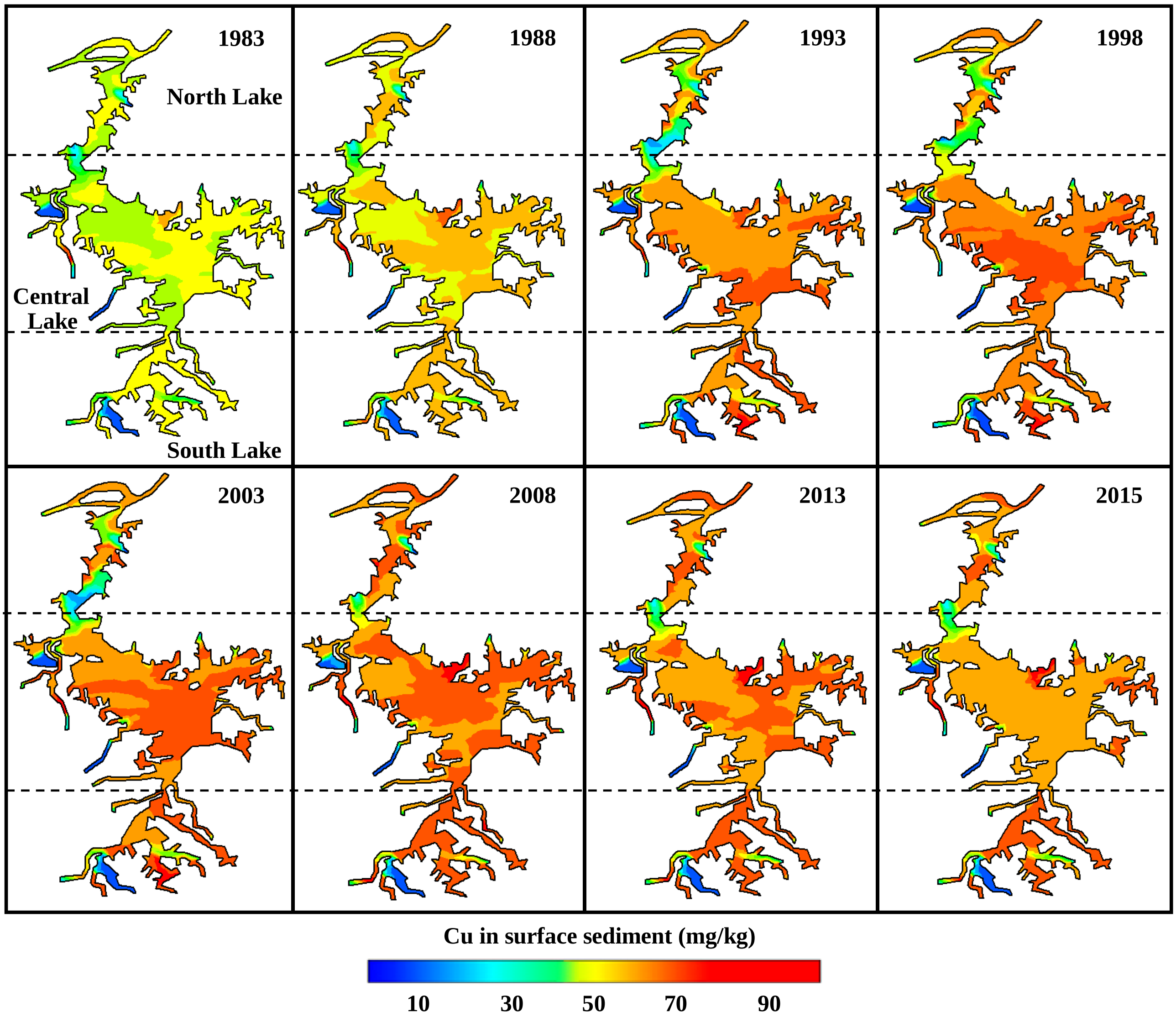

Fig. 4. SCu evolution from 1983 to 2015

Figure 5
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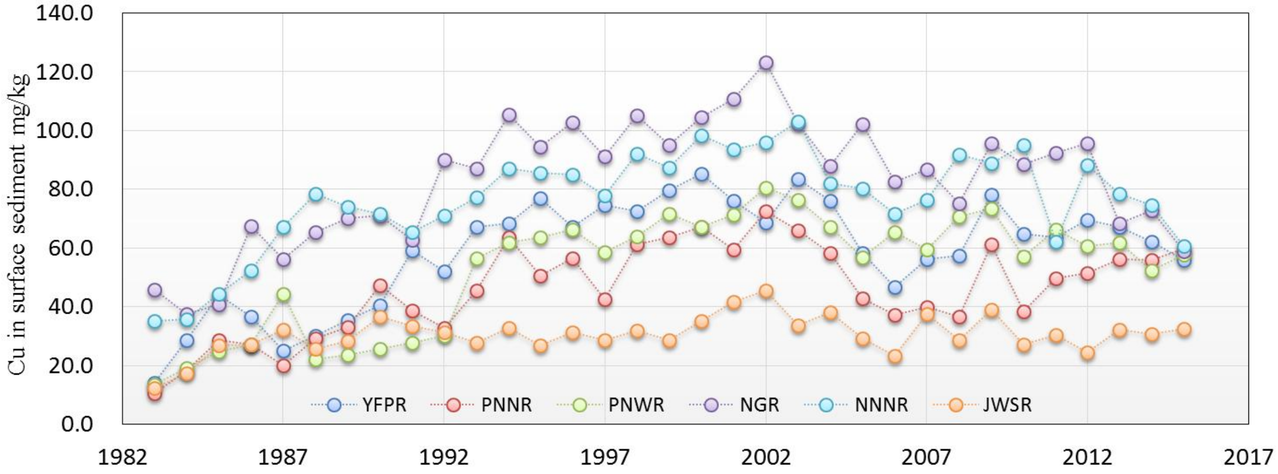

Fig. 5. Fluctuation in SCu in the reserves between 1983 and 2015