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Analysis and assessment of the nutrients, biochemical indexes and heavy metals in the Three Gorges Reservoir, China, from 2008 to 2013

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ABSTRACT

The Three Gorges Reservoir, the world's largest hydropower project, has operated stably for more than five years. To understand its water quality status, the nutrient and biochemical indexes, the total nitrogen (TN), the total phosphorus (TP), the potassium permanganate index (COD_{Mn}), the five-day biochemical oxygen demand (BOD_5) and fecal coliform (F. coli), as well as the heavy metals (Cu, Hg, As, Cd, Zn and Pb) of samples collected from 10 sites during the time period of 2008–2013 were studied via using multiple analysis approaches. For each parameter, pictures of the spatial and temporal distributions were presented, and the reasons behind their variation trends were elaborated. Principal component analysis (PCA) was applied to identify the types of pollution. The Canadian Council of Ministers of the Environment Water Quality Index (CCME-WQI) was calculated to concisely mark the water quality. In addition, a human health risk assessment of the heavy metals in a representative site was conducted. The results showed that the water quality state in the Three Gorges Reservoir was intricate but stable and acceptable from 2008 to 2013. The TN, TP and Pb were considered to be the key pollution indexes. Enforcements to alleviate industrial and urban pollution, along with ship management, have worked. The decrease in heavy metal concentrations from upstream to downstream was associated with the self-purification of the reservoir. However, rural pollution became worse in those years. Improper agricultural activity was an important reason for this trend. For local residents, drinking water was generally safe, but cancer caused by As and Pb is a potential issue.

1. Introduction

With growing populations and rapid industrialization among developing countries, anthropogenic activities, including industrial discharge, city sewage, agricultural and domestic run-off, have increasingly led to water quality deterioration (Liu et al., 2012; Zhai et al., 2014). Dams, a substantial, multifunctional anthropogenic influence that human societies impose upon water basins all around the world, have made water quality regimes more complicated than ever before (Bayram et al., 2014; Zhao et al., 2012). Hence, controversies and debates continue with respect to the adverse environmental and ecological impact of dams (Wu et al., 2004).

The statements above are frequently associated with the Three Gorges Dam in the Yangtze River Basin, China. The Yangtze River Basin feeds more than 450 million people and has suffered from various pollution sources for many years (Yang et al., 2014b). There have been over 50,000 water projects applied to the watershed (Li et al., 2013). Among these, the Three Gorges Reservoir is so far the largest hydropower project in the world (Fu et al., 2010). Therefore, the environmental impact of the reservoir was the focus of various debates during the process of its construction and operation. A large body of research has been conducted to test the water quality,

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access the environmental status and predict the future trend of the water regime in Three Gorges Reservoir over the years (Li et al., 2013). For the reservoir field, researchers prefer to understand the mechanism of pollutants such as phosphorus in terms of their source, sink, interaction and consequence, such as eutrophication in mainstream and its tributaries (Huang et al., 2015). Numerous studies focused on the non-point pollution via simulated models (Ma et al., 2011; Yang et al., 2014a). Additionally, several studies investigated the influence of the Three Gorges Reservoir on the whole watershed. Studies ranging from the headstream of the Yangtze River to the delta in the East Sea, China (Huang et al., 2014; Jiao et al., 2007), were conducted, investigating contaminants including nutrients (Huang et al., 2015), the aquatic ecosystem (Xiao et al., 2013), trace metal elements (Müller et al., 2008) and so on.

Although some useful information about the water quality variation was provided in previous studies, further study is necessary for the following reasons: the studies were mainly based on only one group of parameters, such as the nutrients, aquatic organisms (Xiao et al., 2013) or trace metals (Huang et al., 2008), but there are few combinative analyses of the above-mentioned indexes. For metal elements, reports were usually limited to sediment analysis, and sampling sites were localized (Li and Zhang, 2010; Wu et al., 2009). Many studies focusing on trace metals and relevant health risks in the Three Gorges Reservoir Area were published in Chinese journals, making some progress in this field (Wang et al., 2014a). However, these studies preferred short-term investigations, and their health risk assessments often lacked cancer potential estimation (Yu et al., 2013). A comprehensive water quality analysis and assessment was conducted by (Zhao et al., 2013). However, the Three Gorges Reservoir began running normally from 2008, while the period selected was 2006–2011. The study only chose four parameters (pH, COD_{Mn}, DO and NH_3–N) and just analyzed two sites (inlet and outlet) of the reservoir. The length of the Three Gorges Reservoir exceeds 600 km and flows through both urban and rural areas (Fu et al., 2010). The water quality parameters have different distribution characteristics in different sites of the reservoir areas. Thus, it is necessary to select more appropriate time periods and more representative sites in the Three Gorges Reservoir to analyze its spatial and temporal distribution patterns. To the best of our knowledge, there is still a lack of a complex investigation about the water regime and security of the Three Gorges Reservoir since it has been stably running for more than five years.

Considering the above factors, we selected 11 parameters, including the nutrients, biochemical indexes and heavy metals, from 2008 to 2013 in 10 sampling sites and analyzed them with various analysis methods. The objectives of our study were (1) to illustrate the variation of the spatial and temporal characteristics through the selected parameters, (2) to give a concise evaluation of the water quality with an eligible water quality index, (3) to investigate the sources, trends and associations among each parameter to identify the types of pollution and (4) to assess the human health risk of heavy metals for residents drinking water from the Three Gorges Reservoir. This research could be helpful for knowing the water quality variation after the reservoir began operating normally, thus estimating its security to residents and making contributions to strategic decisions for the Three Gorges Reservoir.

2. Materials and methods

2.1. Study area

As the longest river in Asia, the Yangtze River originates on the Qinghai–Tibet Plateau and flows 6397 km eastward to the East China Sea, in Shanghai, draining an area of approximately 1,800,000 km^2 and raising 450 million people in China(Sun et al., 2013; Yang et al., 2014b). The reach from the source to Yichang is defined as the upstream of the Yangtze River. It is 4300 km long and accounts for about 60% of the total Yangtze River basin. Approximately 44 km upstream of Yichang located the Three Gorges Reservoir.

To date, the Three Gorges Reservoir is the largest reservoir in the world (Table 1). Its function includes controlling flood, generating electricity, and improving navigability. The Three Gorges Reservoir project began in 1990 s and has run at nearly full capacity since 2008. The Three Gorges Reservoir area has a subtropical monsoon humid climate, with an average annual temperature of 18.9 °C (2013) and an annual precipitation between 817.1 and 1360.5 mm (MEP, 2014). Rainfall in the area is abundant but uneven. Thus, meteorological disasters like torrential rain, flood and drought occur commonly, leading to high economic costs. By the end of 2013, the population in the Three Gorges Reservoir area was 16,832,700, 67.1% of which was agricultural population. In 2013, the gross domestic product of the whole region was 82.381 billion U.S. dollars, growing rapidly in recent years.

In this study, 10 sampling sites were selected from upstream to downstream. They were: Yibin, located in upstream of the reservoir, Zhutuo, inlet of the reservoir, Beibei located in the Jialingjiang River and Wulong in the Wujiang River, Wanxian, Xiaojianghekou, Fengjie, Daninghekou and Xiangxi in the mainstream, and Yichang in outlet of the Three Gorges Reservoir (Fig. 1).

2.2. Data source

Data used in this study were all obtained from the Hydrology bureau of Changjiang Water Resources Commission. Monthly data of 11 parameters were collected from 2008 to 2013 (Zn from 2011 to 2013). The parameters, their units and abbreviation, as well as methods used for their determination were illustrated in Table 2.

2.3. Analysis methods

2.3.1. Seasonal Mann–Kendall test

The seasonal Mann–Kendall test has been widely used to detect water quality trends for each user-defined season because it can eliminate the effect of seasonality and avoid missing values in the water quality time series (Chang, 2008). The computation procedure in detail displayed in (Zhai et al., 2014). In this study, the seasonal Mann–Kendall test was used to account for the variation

Table 1
Major indexes to parameters of the Three Gorges Project.

Dam	Height(m)		181.0
	Dam crest above sea level (m)		185.0
	Length (m)		2,335.0
	Bulk volume (10^6 m^3)		26.7
Reservoir	Normal pool	Water level above sea (m)	175.0
		Storage capacity (km^3)	39.3
		Length (km)	663.0
		Area of water surface (km^2)	1,084.0
	Conservation pool (sediment pool)	Water level above sea (m)	145.0
		Storage capacity (km^3)	17.2
Operation stages (water level: m)	Initial stage (begun in June 2003)		135.0
	Transitional stage (begun in October 2006)		156.0
	Quasi normal stage (begun in October 2008)		173.0
	Standard normal stage (begun in October 2010)		175.0

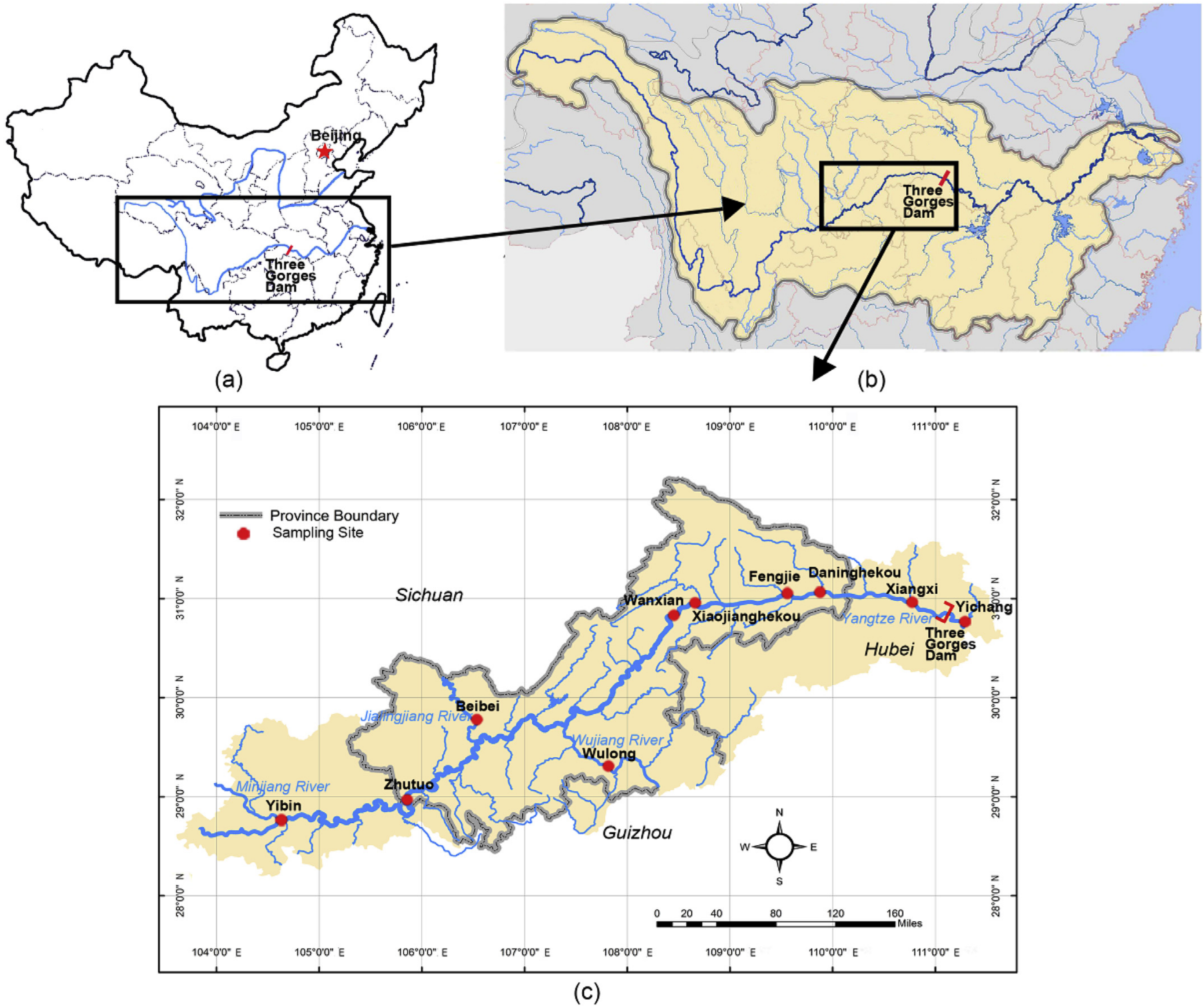

Fig. 1. Study area: (a) Map of China and location of Yangtze River Basin. (b) Profile and water system of Yangtze River Basin and location of Three Gorges Reservoir. (c) Three Gorges Reservoir Area showing 10 sampling sites and the location of the dam.

tendency of the nutrient and biochemical parameters from 2008 to 2013. The process of the seasonal Mann–Kendall test was completed by the PWQTrend software, which was developed by the China Institute of Water Resource and Hydropower Research. (http://www.iwhr.com).

2.3.2. Canadian Council of Ministers of the Environment Water Quality Index (CCME-WQI)

As a concise and convenient tool to express the water quality for different objectives, the water quality index has been widely used all around the world since 1960 s. The water quality index reflects the quality of a water body and can help the management of relevant department and is understandable to non-technical people. Researchers have developed various types of water quality indexes to be suitable for local conditions, and all water quality indexes have advantages and limitations (Lumb et al., 2011). Among them, CCME-WQI has been used in many countries and received great assessments because of its several merits (Dede et al., 2013). In our study, we selected CCME-WQI to evaluate the water quality in the Three Gorges Reservoir by using the parameters above, except Zn. The levels of each parameter are as follows: the TN was 1 mg/L, the TP was .05 mg/L, COD $_{(Mn)}$ was 6 mg/L, BOD$_5$ was 4 mg/L and F. coli was less than 10,000. All values referred to the demand of China's Environmental Quality Standard for Surface Water (GB) Class III(-MEP, 2002). The metal objective values were derived from the WHO (WHO, 2011). Cu was 2000 μg/L, Hg was 6 μg/L, Cd was 3 μg/L, and As and Pb were 10 μg/L. The detailed computation procedure is shown in (Dede et al., 2013). The water quality is scaled to range from 0 to 100 and is ranked in five categories, poor (0–44), marginal (45–59), fair (60–79), good (80–94) and excellent (95–100).

2.3.3. Principal component analysis (PCA)

PCA is a powerful tool widely applied in water quality data analysis (Bengraine and Marhaba, 2003). To identify the underlying factors and further explain their structure and internal relationships when the initial dataset includes a large number of variables and is multifarious, PCA simplifies and clarifies the dataset by though transforming and rearranging correlated variables into a

Table 2
The variables used and their abbreviation, unity and method of determination and origin.

Parameter	Abbreviation	Units	Method	Origin
Total nitrogen	TN	mg/L	GB11894-89	HBOCWRC[b]
Total phosphorus	TP	mg/L	GB11893-89	HBOCWRC
Potassium permanganate index	COD_{Mn}	mg/L	GB11892-89	HBOCWRC
Five-day biochemical oxygen demand	BOD_5	mg/L	GB7488-87	HBOCWRC
Fecal coliform	F.coli	A/L	[a]	HBOCWRC
Copper	Cu	μg/L	GB7475-87	HBOCWRC
Mercury	Hg	μg/L	[a]	HBOCWRC
Arsenic	As	μg/L	[a]	HBOCWRC
Cadmium	Cd	μg/L	GB7475-87	HBOCWRC
Lead	Pb	μg/L	GB7475-87	HBOCWRC
Zinc	Zn	μg/L	GB7475-87	HBOCWRC

[a] Water and wastewater monitoring and analysis method (fourth edition), MEP, 2012.
[b] HBOCWRC:Hydrology bureau of Changjiang Water Resources Commission.

reduced set of new independent, orthogonal components, which are called principal components (PCs). In this study, annual PCA (from 2008 to 2013) of all parameters (except Zn) with the varimax rotation of the standardized component loadings was conducted. The Kaiser–Meyer–Olkin test and Bartlett's test were performed to evaluate the validity of PCA, which required that the Kaiser–Meyer–Olkin values should be more than .5 and Bartlett's test be significant (<.001). PCs with an eigenvalue higher than 1 were retained until the cumulative variance was higher than 85%. In this study, all PCAs reserved same amounts of PCs. The loadings in PCA illustrated the extent to which each original variable contributes to the PCs.

2.3.4. Human health risk assessment

Heavy metals are identified to be an essential environmental factor that can lead to severe human health hazards, including neoplasm via mainly three pathways, oral intake, inhalation and dermal absorption (Li and Zhang, 2010). Mainly based on the concentrations of the parameters (the highest) and the population density (the second), we selected the heavy metal data of Yibin to evaluate the human health risk. Yibin is a prefecture city with 4.46 million people. It is located at the confluence of the mainstream and one of the major tributaries of the Yangtze River (Minjiang River). Before arriving at the confluence, the mainstream flows through the Sichuan mining area, and the Minjiang River flows through several cities, including Chengdu. We made use of the evaluation method, mostly in terms of the USEPA, and some of the equations were modified using other references (Li and Zhang, 2010; Rodriguez-Proteau and Grant, 2005; US-EPA, 2004; Wu et al., 2009).

For exposure assessment, Eqs. (1) and (2) are used to determine the dose through oral intake and dermal pathways, respectively.

$$\mathrm{IDD_{ingestion}} = \frac{C_w \times IR \times EF \times ED \times 10^{-3}}{BW \times AT \times (365 days/year)} \quad [1]$$

$$\mathrm{DAD_{dermal}} = \frac{C_w \times K_p \times SA \times \mathrm{t_{event}} \times EV \times EF \times ED \times 10^{-6}}{BW \times AT \times (365 days/year)} \quad [2]$$

where $\mathrm{IDD}_{\mathbf{ingestion}}$ is the intake daily dose by ingestion in units of mg/kg/day; C_w is the chemical concentration in water in units of μg/L; IR is the ingestion rate in units of L/day and is equal to 2 L/day; EF is the exposure frequency in units of days/year and is equal to 350 days/year; ED is the exposure duration in units of years and is equal to 30 years; BW, is body weight in units of kg and is equal to 70 kg (adults); and AT is the averaging time in units of years. For carcinogens, $AT_c = 70$ years, and for noncarcinogens, $AT_{nc} = 30$ years. $\mathrm{DAD}_{\mathbf{dermal}}$ is the dermal absorbed dose in units of mg/kg/day; K_p is the dermal permeability coefficient in units of cm/h and is taken from the USEPA; SA is the skin surface area available for contact in units of cm^2 and is equal to 18,000 cm^2 (adults); t_{event} is the time spent on an event in units of h and is equal to .58 h/event; and EV is the event frequency in units of events/day and is equal to one event. t_{event} and EV are considered for showering for 35 min per day. ED is nine years (adults), and for a carcinogen, AT is 70 years. For noncarcinogens, the AT value is equal to the ED value.

Risk characterization can be quantified by carcinogenic risk and noncarcinogenic risk. Both of them are based on a dose–response assessment. The carcinogenic risk increases linearly as the dose of an individual chemical increases. We can describe it by Eq. (3), Eq. (4) and Eq. (5).

$$CR_{ingestion} = IDD_{ingestion} \times CSF \quad [3]$$

$$\mathrm{CR_{dermal}} = \mathrm{DAD_{dermal}} \times \mathrm{CSF_{dermal}} \quad [4]$$

$$\mathrm{CR_{total}} = \sum \mathrm{CR_s} \quad [5]$$

where CR is the carcinogenic risk through either ingestion or dermal; CSF is the cancer slope factor in units of $mg/kg/day^{-1}$ and refers to the USEPA; CSF_{dermal}, which is adjusted by ABS_{GI}, is gastrointestinal absorption, derived from the USEPA (Cu, Hg, As, Cd); Pb is cited from Wu et al., 2009; the default Zn value is taken from USEPA; and $CSF_{dermal} = CSF/ABS_{GI}$;

The range of carcinogenic risks acceptable by the USEPA is 10^{-6} to 10^{-4}.

Noncarcinogenic risks are compared to the RfD to form the HQ (hazard quotient). The RfD (reference dose) is the security threshold of a specific chemical. The sum of the HQs is defined as the HI (total hazard index), and $HI > 1$ indicates the potential for an adverse effect on human health or the necessity for further study. These can be calculated by Eq. (6), Eq. (7) and Eq. (8).

$$\mathrm{HQ_{ingestion}} = \frac{\mathrm{IDD_{ingestion}}}{\mathrm{RfD}} \quad [6]$$

$$\mathrm{HQ_{dermal}} = \frac{\mathrm{DAD_{dermal}}}{\mathrm{RfD_{dermal}}} \quad [7]$$

$$\mathrm{HI} = \sum \mathrm{HQs} \quad [8]$$

where RfD_{dermal} is the RfD adjusted by ABS_{GI}, $RfD_{dermal} = RfD \times ABS_{GI}$.

3. Results

3.1. Spatial and temporal variation of the Three Gorges Reservoir from 2008 to 2013

3.1.1. Nutrients (TN, TP)

The spatial and temporal variations of the TN and TP are shown in Fig. 2. For spatial trends, the concentrations of the TN showed fluctuations, and the concentrations of the TP decreased gradually from upstream to downstream (Fig. 2). It was found that an extremely high value of the TP occurred in Wulong (.40 mg/L), the monitoring station in the WuJiang River. This result can be mainly attributed to inappropriate mining in the abundant phosphorate rock resources of the Guizhou province upstream of the Wujiang River.

The temporal trends displayed or showed significant seasonality and similar temporal patterns, higher in the summer and lower in the winter. From the results of the seasonal Mann–Kendall test (Fig. 3), 70 percent of sampling sites showed high-level significant upward trends of the TN and TP. None of them exhibited a downward trend. These results indicated a continuous and rapid deterioration of nutrients in the Three Gorges Reservoir.

3.1.2. Biochemical parameters (COD_{Mn}, BOD_5 and F. coli)

As shown in Fig. 2, the spatial patterns of the biochemical parameters in the reservoir were irregular and complex. The highest concentrations of COD_{Mn}, BOD_5 and F. coli were found in Zhutuo, Xiangxi and Beibei, respectively. The lowest concentrations of the parameters were found in Wulong, Wanxian and Fengjie and Xiangxi, respectively. The specificity of the Three Gorges Reservoir, for instance, large scale, diverse pollution types, probably caused this (Yang et al., 2014b).

The temporal variations of COD_{Mn}, BOD_5 and F. coli also showed seasonality, and their profiles were almost the same as the nutrients. The COD_{Mn} and BOD_5 levels were relatively stable. F. coli showed high variability, ranging from only 2496 A/L in winter to 91,784 A/L in summer. For COD_{Mn}, no trends were found downstream (Daninghekou, Xiangxi and Yichang) (Fig. 3). For BOD_5, the main reservoir areas exhibited significant downward trends (Wanxian, Xiaojianghekou, Fengjie, Daninghekou, Xiangxi and Yichang) (Fig. 3). These trends may indicate the control of the urban pollution. Despite the high variability of F. coli, the seasonal Mann–Kendall test's results showed that it was the most stable parameter, and 70% of the monitor stations showed no trend (Fig. 3). However, some measures must be taken to prevent diseases such as gastroenteritis in view of the extremely high level of F. coli in the summer.

3.1.3. Heavy metal parameters (Cu, Hg, Cd, As, Pb and Zn)

Fig. 4 shows the spatial patterns of the heavy metal indexes in the reservoir area. As observed, all heavy metals remained at low concentrations, except Pb, and showed decreasing trends from upstream to downstream. Such trends appeared to demonstrate the ability of self-purification of the Three Gorges Reservoir and were reported by many studies on dams before (Zhao et al., 2012). Cu, Hg, Cd, As, and Zn were controlled well, and all sites met the requirements of all guideline values (Table 3, Fig. 4). However, the concentrations of Pb exceeded the WHO suggested level, China's drinking water guidelines and the USEPA drinking water guideline (Fig. 4). Therefore, the ecological risk and human health risk is of notable concern in spite of the Pb concentration's decreased trend.

For temporal patterns, the concentrations of all elements showed significant downward tendencies, decreasing by more than 60% for most of them (Table 3). The concentrations of Hg were extremely low, ranging from .01 μg/L to .06 μg/L, and could be considered to remain at a stable state. The concentrations of As showed great stability from 2008 to 2012 but became hazardous (11.30 μg/L) in 2013.

3.2. Application of CCME-WQI and the results of the PCAs

The spatial patterns of the heavy metals showed decreasing trends, but the spatial trends of the other indexes were random. Temporal variations showed that most of parameters had great seasonality, and the state of the nutrients deteriorated persistently in those years. Those preliminary results clearly indicated the complexity and specificity of the Three Gorges Reservoir. As a river-type reservoir, it has length of over 600 km and flows through both urban and rural areas, including the metropolis Chongqing. Its large scale and diverse pollution sources made it is hard to understand the water quality and pollution type just through presentation of the spatial and temporal patterns. To concisely assess the water quality and investigate the source, type and interaction of each pollutant, CCME-WQI and PCA were selected for further analysis and assessment of the reservoir.

3.2.1. Application of CCME-WQI

We calculated both the spatial and temporal variations of CCME-WQI in the Three Gorges Reservoir, and all of the parameters were taken into account. Table 4 lists the results of the spatial pattern, and Fig. 5 shows the temporal profile of the water quality index fluctuation. Regardless of the spatial or temporal assessment, it was found that the water quality indexes remained steady, and their categorizations were all ranked between marginal and fair (Samal et al., 2011). Given the contamination status of the Three Gorges Reservoir, it was an objective and proper assessment.

For the spatial variation, it was observed that CCME-WQI increased from upstream to downstream. Its lowest value was observed in Yibin (52.24 marginal), and the best occurred in Xiangxi (77.73 fair). This phenomenon was inferred to be one of the benefits of dams because its powerful self-purification(Espejo et al., 2012).

For the temporal variation, calculations also demonstrated the stability of the water quality to some extent and simultaneously illustrated seasonality. It was remarkable that the highest CCME-WQI value in one year occurred in winter, and the lowest occurred in summer, particularly in 2008 (50.5 in July, the lowest value in all six years) and 2009 (53.4 in August). Although relatively poor water qualities in summer can be seen as “kind of normal phenomena”, “algae bloom” and infectious disease occurred because the water quality of the Three Gorges Reservoir had already been on the verge of being at the hazardous condition.

According to the results above, CCME-WQI can become an appropriate approach to assess the water quality in the Three Gorges Reservoir at the spatial or temporal dimension. Therefore, CCME-WQI was feasible not only for rivers or lakes (Espejo et al., 2012; Samal et al., 2011) but also for large reservoirs such as the Three Gorges Reservoir.

3.2.2. Principal component analysis (PCA)

The results of the PCA from 2008 to 2013 are presented in Table 5. The ranges of the percentage of the total variance of the PCs in each year were approximately 15 on average, with a maximum of 29.7 (2008, PC1) and a minimum of 5.4 (2011, PC6), which may mean that the importance of each PC was not very distinct. PC1, in 2013, showed 19.6% of the total variance, mainly made up of strong positive loadings on COD_{Mn} (.798), F. coli (.802) and Cu (.734). PC1 seemed to represent a mixed, comprehensive effect of the trace metal, chemical and biologic factors. Similar situations were also observed in 2009 (PC1 consisted of the same parameters and

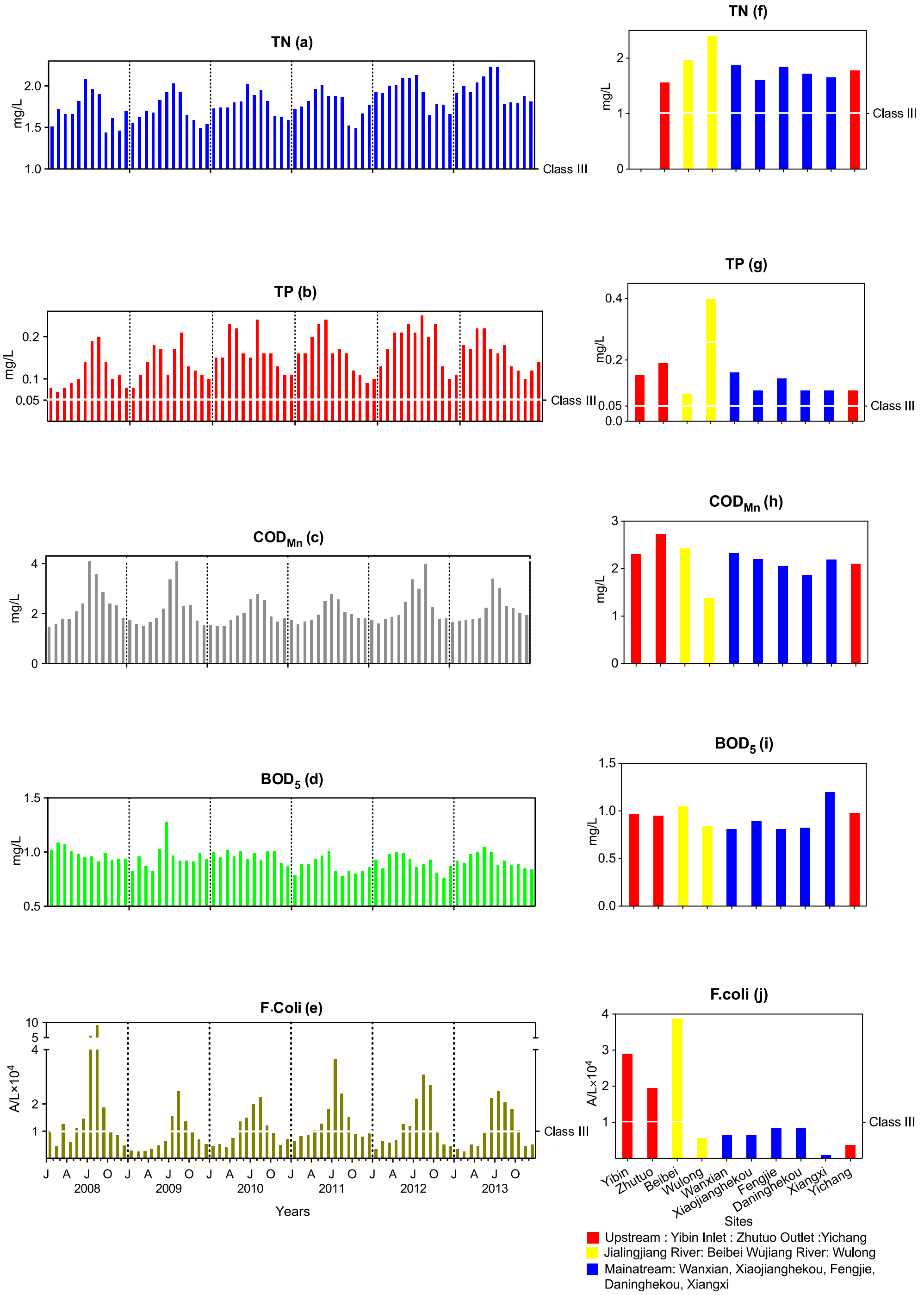

Fig. 2. Left: Nutrient and biochemical parameters of Three Gorges Reservoir with respect to time series from 2008 to 2013 with level of Environmental quality standards for surface water (GB 3838-2002) Class III (white line). Right: Concentrations of above parameters in each sampling locations along with level of standard of GB Class III (white line). Class III applies to: centralized surface water source for domestic and drinking water; fisheries area; swimming area.

contributed to 23.2%), 2010 (PC2 consisted of the same parameters and contributed to 18.4%) and 2012 (PC3 consisted of COD$_{Mn}$ and F. coli and contributed to 14.3%).

Then, PC2 in 2013 had 16.8% of the total variance and strong positive loadings on Cd (.902) and Pb (.778); PC4 explained 12.9% of the total variance, dominated by Hg (.920); and PC6 explained 10.4% of the total variance, dominated by As (.928). These three PCs could be associated with heavy metal pollution. In the remaining PCA results, it was found that one or two PCs was controlled to heavy metals, PC2 (explained 18.1% of the total variance on Hg and As) and PC3 (17.1% on Cd and Pb) in 2009, PC1 (explained 28.0% of the total variance on Hg, Cd and As) and PC3 (12.3% on Cu and Pb) in 2010.

PC3, in 2013, accounted for 15.9% of the total variance, comprised of TN (.882) and TP (.858) with high loadings, which seemed to be attributed to the effect of the nutrient input. Similarly, PC3, in 2011, explained 16.3% of the total variance on the TN and TP, and PC2, in 2012, explained 15.3% of the total variance on the TN

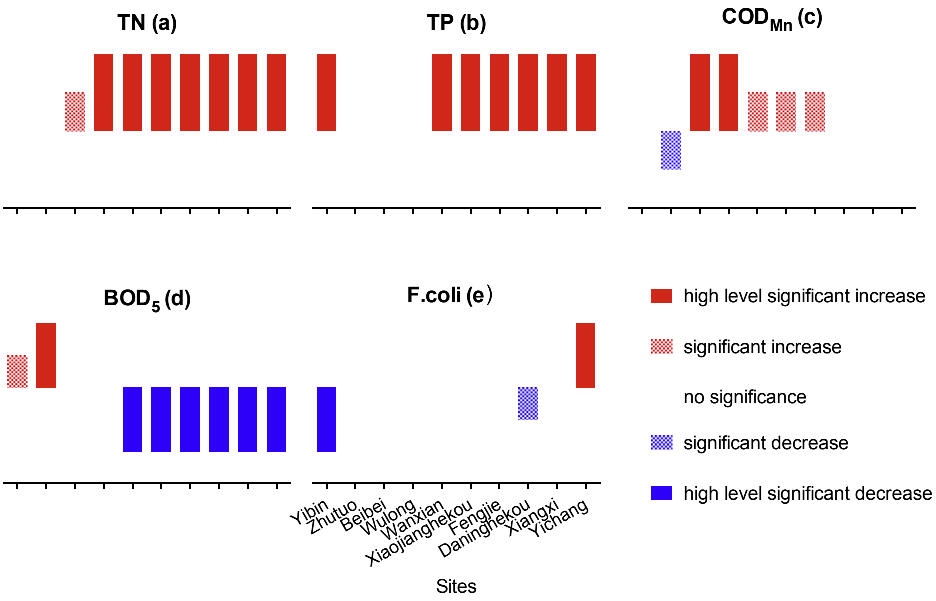

Fig. 3. Seasonal Mann–Kendall test's results of nutrients and biochemical parameters in each sampling site.

and TP, too. The contribution of the nutrient of the TN and TP was expressed separately in PC4 (10.4% of the total variance on the TN) and PC6 (10.1% of the total variance on the TP) in 2009 and PC4 (11.4% of the total variance on the TP) and PC5 (10.5% of the total variance on the TN) in 2010.

Finally, PC5, in 2013, explained 10.4% of the total variance on BOD_5. The possible identification of this principal component was the source of organic pollutants. PC5, in 2008 (10.5% of the total variance), 2009 (10.4% of the total variance) and 2011 (11.0% of the total variance), and PC6, in 2010 (10.4% of the total variance) and 2012 (10.6% of the total variance), also clearly represented the biochemical influence.

The six PCs were ultimately identified as the following four types: mixed pollution, heavy metals, nutrient inputs and organic pollutants in each year from 2008 to 2013. It was found that, although each year included all types of pollution, their orderings changed in different years. For instance, PC1, in 2009, was associated with mixed pollution (explained 23.2% of the total variance) but, in 2011, was linked to heavy metal pollution (explained 20.5% of the total variance). The reasons behind this result were probably the changing environment in the Three Gorges Reservoir. In different years, the situations of the pollution types were different. Some pollution types may increase, whereas some may deteriorate. Those were reflected on the changeable ordering of the PCs and the various loading of the parameters in each PC. This phenomenon demonstrated that pollution in the Three Gorges Reservoir was intricate and complex. Different variables can be important in different circumstances, which agrees with (Li and Zhang, 2010).

3.3. Human health risk assessment of heavy metals

Although over 1.2 million people had been resettled by the end of 2008 (Fu et al., 2010), the population of the Three Gorges Reservoir area has not decreased in the last three years (Fig. 6). Moreover, reports showed that a malignant tumor has been the second leading cause of death for many years (Fig. 6). The results of the PCAs also indicated that heavy metals were a crucial pollution type affecting the water quality in the Three Gorges Reservoir. Therefore, it is necessary to assess the health risk of heavy metals on residents in the Three Gorges Reservoir area.

Table 6 presents the results of the noncarcinogenic and carcinogenic risk assessment via oral and dermal pathways, respectively. For a noncarcinogenic risk, the values of $HQ_{ingestion}$, HQ_{dermal} and HI were all smaller than 1, which revealed a low noncarcinogenic risk of water in Yibin. However, As ($HI = .332$) and Pb ($HI = .524$) were relatively close to the critical level. A warning sign was posted to people that long-term exposure to As and Pb may trigger adverse health conditions. In addition, considering the continuous high concentrations of Pb for many years, its chronic toxicity should be a concern for residents. According to the size of the HI values, it can be concluded that the most harmful element was Pb, followed by As and Cd, while the least harmful elements were Cu and Zn. Therefore, Pb and As were further studied by assessing the carcinogenic risk. The values of CR_{total} of As and Pb were calculated to be 1.72×10^{-4} and 1.55×10^{-4}, respectively. Both of them posed potential risks for developing cancer, which means drinking water for a lifetime may increase the risk of cancer. After human health risk assessment, it was found that Pb and As were the primary and secondary contaminants threatening the residents.

4. Discussion

4.1. Deterioration of nutrients: TN and TP

According to a large amount of research results, non-point source pollution has become an important cause of water deterioration in the Three Gorges Reservoir area (Shen et al., 2015). Non-point source pollution comes from both natural and anthropogenic factors. (Ma et al., 2011) demonstrated that atmospheric deposition was the dominant non-point source pollution of TN, whereas the major part of TP pollution was from land use. Water and soil erosion has also already become a critical source of pollution in the area (Li et al., 2013): Landslides occurred 69 times per year on average from 2008 to 2013. To meet the requirement of rapidly developing agriculture activities, the amount of fertilizer and pesticides, the main causes of TN and TP, continues to grow in the area every year. Fertilizer use increased by an annual average of 5%, from .11 million tons in 2003 to .57 million tons in 2012. It should be explained that, because of the division of the operation stage shown in Table 1, the

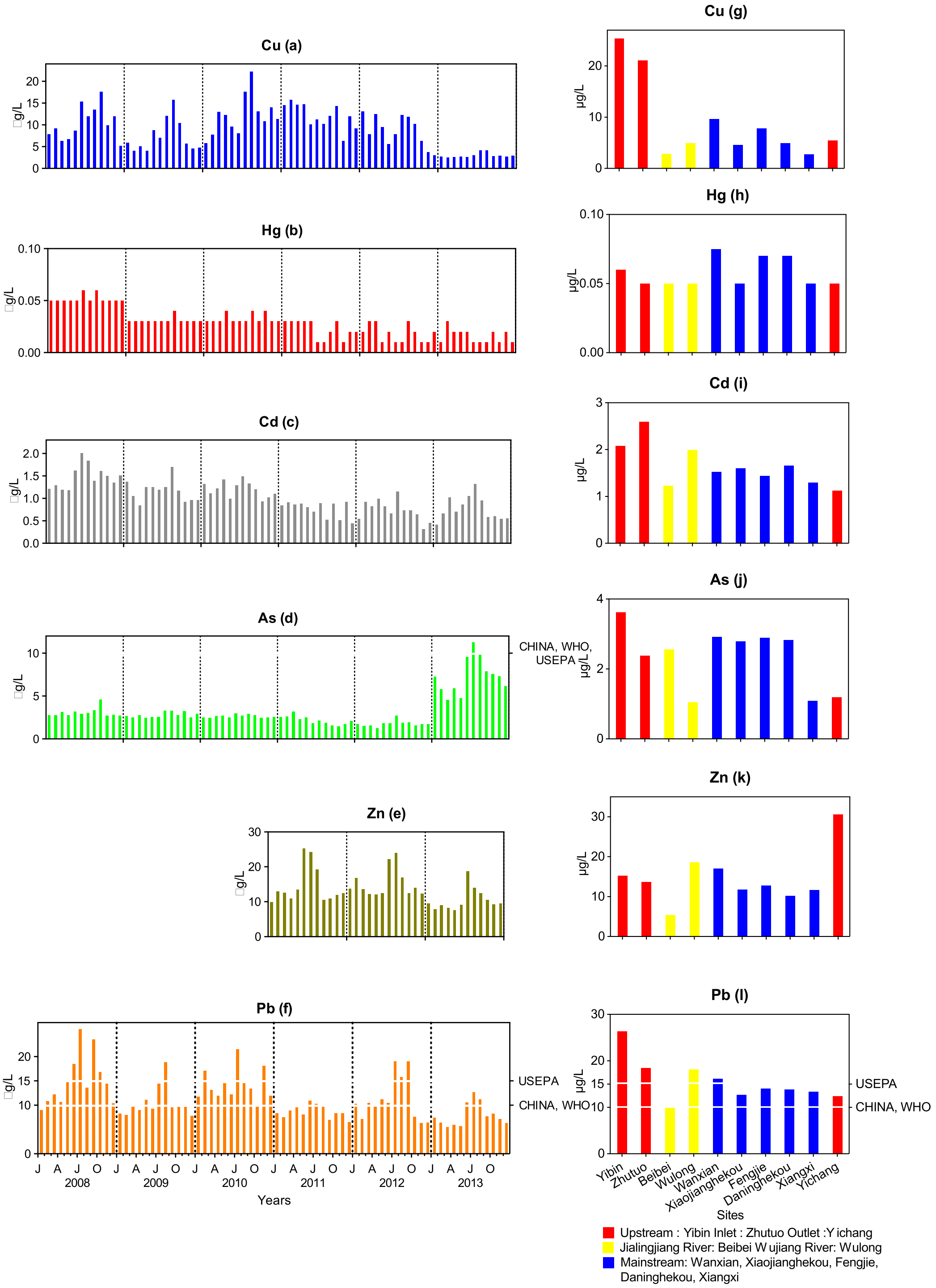

Fig. 4. Left: Heavy metals of Three Gorges Reservoir with respect to time series from 2008 to 2013 with levels of standards of China, WHO and USEPA (white line). Right: Concentrations of heavy metals in each sampling locations along with levels of standards of China, WHO and USEPA (white line). China: Environmental quality standards for surface water (GB 3838-2002) Class III. WHO: Guidelines for drinking-water quality, fourth edition. USEPA: 2012 edition of the drinking water standards and health advisories.

effect of the dam was different during the different operations. Thus, the year 2003 was chosen as the contrast in the Fig. 6, representing the natural state. Nitrogenous fertilizer and phosphate fertilizer were the primary and secondary fertilizers used in the Three Gorges Reservoir areas, accounting for 65% and 28% of the total fertilizer, respectively. Pesticide use also showed an ascending tendency in the past ten years, reaching a maximum of 701.8 tons in 2011. Organophosphorus pesticides were the predominant pesticides. Furthermore, they were underutilized, and approximately 7% of the fertilizer and 9% of the pesticides ran off per year (Fig. 6). A large amount of these products turned to pollutants and were transported to water bodies under the actions of rainfall and irrigation by means such as surface runoff, subsurface flow, farm drainage and seepage. At the same time, the existence of the dam slowed down the flow speed of the mainstream and the tributaries in the reservoir (.07–2.49 m/s on average during 2010–2013 in the

Table 3
Metal concentrations in the Three Gorges Reservoir (Means), and comparison with other studies and guidelines (surface water: unit in μg/L; sediments: unit in μg/g).

Locations		Cu	Hg	Cd	As	Zn	Pb
Three Gorges Reservoir, This study, 2008		10.369	.05	1.475	3.063		15.025
Three Gorges Reservoir, This study, 2013		3.013	.016	.771	1.535	10.431	7.894
Three Gorges Reservoir, This study, 2008–2013		8.94	.03	1.02	2.33	13.039	11.205
Headstream(Tibetan Plateau)[a]		2.504	<.001	.024		2.461	4.148
Midstream(Yanwu)[b]		3.0	.1	.11	3.2	6.1	1.35
Downstream(Nanjing)[c]		10.7		4.7	13.2	9.4	55.1
Tributary(Han River)[d]		26.73		4.59	28.32		18.39
The whole Yangtze river[e]		8.4	.04	.28	7.04	18.75	6.4
Sediments in Water Level Fluctuation Zone of Three Gorges Reservoir[f]		32.74	.095	.445	14.415	82.33	39.15
Sediments in the whole Yangtze river[g]		82.0	.16	2.46	25.4	174.0	60.0
World background[e]		1.0		.02		10.0	.2
Drinking water quality, China[h]		1300.0	1.0	5.0	10.0	–	10.0
Drinking water quality, WHO[i]		2000.0	6.0	3.0	10.0	–	10.0
Drinking water quality, USEPA[j]	MCL	1300.0	2.0	5.0	10.0		15.0
	MCLG	1300.0	2.0	5.0	.0		.0

[a] Huang et al., 2008.
[b] Müller et al., 2008.
[c] Wu et al., 2009.
[d] Li and Zhang, 2010.
[e] Wang et al., 2011.
[f] Ye et al., 2011.
[g] Wang et al., 2014b.
[h] Standards for drinking water quality GB5749-2006, 2007.
[i] Guidelines for drinking water quality, fourth edition, 2011.
[j] Edition of the drinking water standards and health advisories, 2012.

Table 4
The calculated F factors,Water quality index and Categorization values of Canadian Council of Ministers of the Environment Water Quality Index (CCME-WQI) model of each sampling site[a].

Sampling site	F1	F2	F3	Water quality index	Category
Yibin	66.67	30.56	38.27	52.24	marginal
Zhutuo	60	41.53	34.65	53.36	marginal
Beibei	40	26.41	33.40	66.27	fair
Wulong	40	22.18	46.40	62.39	fair
Wanxian	60	29.44	27.66	58.24	marginal
Xiaojianghekou	40	25.71	15.99	71.04	fair
Fengjie	50	29.31	24.76	63.61	fair
Daninghekou	40	27.36	19.14	69.92	fair
Xiangxi	25	24.31	16.49	77.73	fair
Yichang	40	22.92	16.28	71.77	fair

[a] The computation procedure in detail displayed in Dede et al., 2013.

mainstream and some less than .05 m/s in the tributary), and the increase of the water depth caused by impoundment made it harder for water to mix in the vertical direction and result in the enrichment of nutrients in bottom sediments. High TN and TP levels and weak flow were suitable for phytoplankton reproduction and may trigger “algae bloom” (Fu et al., 2010). In 2003, they occurred 10 more times in the reservoir and were observed in more than 20 tributaries in 2013 (Ma et al., 2011). Additionally, the Three Gorges Reservoir formed a 30 m water level fluctuation zone between the elevations of 145–175 m that varied seasonally (Ye et al., 2011). The water level fluctuation zone has been considered an important reason leading to eutrophication because periodically decayed plants and covered soil may release contaminations (Zhao et al., 2013).

4.2. Abatement of biochemical indexes: COD_{Mn}, BOD_5 and F. coli

In contrast to the deteriorations of the TN and TP, levels of COD_{Mn}, BOD_5 and F. coli were under control and showed better trends. These improvements were attributed to more informed policy-making and strict regulations for pollution prevention and abatement. Firstly, industrial wastewater emissions showed a sharp reduction in the area, from 558 million tons in 2008 to 190 million tons in 2013 (Fig. 6). By 2007, over 1500 non-compliant companies were shut down because of heavy pollution (Fu et al., 2010). In 2011, more than 2300 industrial enterprises accepted pollution control and transformation. Secondly, although resettlement and urbanization led to an increase in urban sewage discharge, modern drain piping systems and sewage treatment plants were built to collect and purify rainfall, industrial water and domestic water before discharging into water bodies. There were 56 sewage treatment plants in 2008, and the total number doubled by the end of 2013. In 2011, over 500 km of sewage collection pipe network were constructed in urban areas. Therefore, the domestic sewage treatment ratio was 85.1% in 2008 and has been continuously improving in recent years, reaching 96.9% by 2013 (Fig. 6). In addition, the

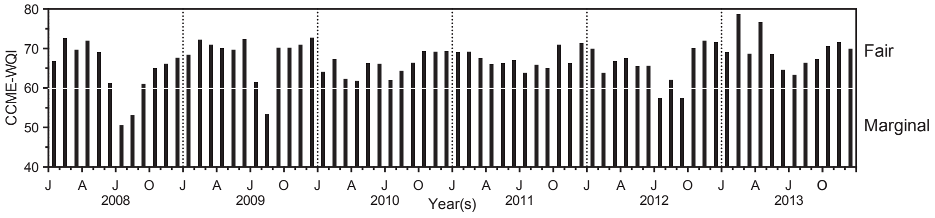

Fig. 5. Temporal variations of Canadian Council of Ministers of the Environment Water Quality Index (CCME-WQI) in Three Gorges Reservoir from 2008 to 2013 with categories.

Table 5
Loadings of 10 variables on Principal Component (PCs) (varimax normalized) from 2008 to 2013. Extraction Method: Principal Component Analysis. Rotation Method: Varimax with Kaiser Normalization. Kaiser–Meyer–Olkin Measure of Sampling Adequacy: .636(2008), .673(2009), .627(2010), .537(2011), .621(2012), .609(2013); Bartlett's Test of Sphericity<.001.

Elements	PC1	PC2	PC3	PC4	PC5	PC6
(a) 2008						
TN	.015	−.111	−.054	**.959**	−.115	.007
TP	**0.725**[a]	.022	.415	.358	.054	.020
BOD	−.178	−.202	.024	−.119	**.942**	−.007
COD(Mn)	.379	.081	**.882**	−.076	.035	.106
F.coli	−.005	.054	.078	.008	−.005	**.995**
Cu	**.892**	.000	.178	−.127	.057	.047
Hg	.085	**.934**	−.145	.015	−.127	−.004
Cd	**.872**	.081	−.086	.074	−.293	−.033
As	−.004	**.862**	.292	−.170	−.104	.082
Pb	**.838**	.045	.313	−.022	−.165	−.036
Eigenvalues	2.965	1.686	1.204	1.120	1.049	1.013
% of Total Variance	29.651	16.858	12.039	11.201	10.486	10.126
Cumulative %	29.651	46.509	58.548	69.749	80.235	90.361
(b)2009						
TN	.149	.128	.150	**.960**	−.029	.074
TP	.190	−.163	.046	.078	−.096	**.953**
BOD	.038	−.035	−.064	−.029	**.985**	−.088
COD(Mn)	**.817**	−.018	.293	.130	.163	.041
F.coli	**.803**	.295	−.177	−.009	−.073	.210
Cu	**.827**	−.192	.316	.127	−.031	.071
Hg	−.073	**.930**	.037	.096	.036	−.053
Cd	.064	.246	**.887**	.051	−.015	.004
As	.146	**.843**	.333	.057	−.108	−.162
Pb	.481	.062	**.752**	.255	−.118	.081
Eigenvalues	2.319	1.809	1.710	1.041	1.041	1.007
% of Total Variance	23.190	18.090	17.099	10.411	10.405	10.068
Cumulative %	23.190	41.280	58.379	68.790	79.195	89.263
(c)2010						
TN	−.028	.014	−.033	.156	**.972**	.032
TP	−.286	−.010	.030	**.820**	.209	−.150
BOD	−.129	.039	−.154	−.113	.032	**.968**
COD(Mn)	−.116	**.844**	.063	−.346	.017	.050
F.coli	.250	**.806**	.010	.398	.061	.072
Cu	−.128	**0.663**[b]	**.611**	.127	−.129	−.133
Hg	**.974**	−.005	.066	−.067	−.047	−.061
Cd	**.721**	−.143	.372	−.356	.165	−.130
As	**.968**	.083	.100	−.081	−.071	−.049
Pb	.458	.094	**.819**	−.019	−.011	−.173
Eigenvalues	2.807	1.840	1.227	1.142	1.045	1.039
% of Total Variance	28.073	18.403	12.270	11.419	10.449	10.389
Cumulative %	28.073	46.476	58.746	70.165	80.614	91.003
(d)2011						
TN	−.161	.093	**.879**	.003	−.118	−.266
TP	.115	−.227	**.889**	−.118	−.015	.226
BOD	−.039	−.108	−.101	.032	**.972**	−.003
COD(Mn)	−.155	.063	−.177	**.872**	.065	.292
F.coli	.121	.071	.048	**.875**	−.019	−.292
Cu	**.908**	.010	−.058	−.134	.029	.184
Hg	.180	**.827**	−.008	.026	−.277	.041
Cd	−.004	**.902**	−.083	.109	.115	−.093
As	**.596**	**.594**	−.085	−.012	−.217	.362
Pb	**.868**	.167	.041	.133	−.061	−.269
Eigenvalues	2.045	1.958	1.626	1.590	1.104	.540
% of Total Variance	20.449	19.584	16.265	15.899	11.045	5.398
Cumulative %	20.449	40.032	56.297	72.197	83.241	88.639
(e)2012						
TN	−.147	**.878**	−.168	.151	.175	−.096
TP	.141	**.848**	.137	−.212	−.252	.026
BOD	−.051	−.055	.099	.035	.019	**.982**
COD(Mn)	.440	−.143	**.779**	−.052	.219	.134
F.coli	.088	.081	**.795**	.471	.005	.046
Cu	**.876**	.052	.017	−.097	−.115	.111
Hg	.019	−.047	.180	**.927**	.207	.031
Cd	.036	−.031	.122	.190	**.951**	.019
As	**.846**	−.016	.236	.123	.016	−.232
Pb	**.891**	−.051	.178	.037	.146	−.009
Eigenvalues	2.524	1.529	1.431	1.216	1.125	1.062
% of Total Variance	25.240	15.292	14.311	12.164	11.250	10.615
Cumulative %	25.240	40.531	54.842	67.006	78.256	88.872
(f)2013						
TN	.002	−.006	**.882**	.202	−.042	−.084
TP	−.053	−.154	**.858**	−.220	−.014	.037
BOD	.028	−.024	−.060	.082	**.930**	−.206
COD(Mn)	**.798**	.404	−.151	.034	−.048	−.076
F.coli	**.802**	.066	.060	.426	−.105	.007
Cu	**.734**	−.033	.024	−.330	.299	.296
Hg	.074	.193	−.010	**.920**	.094	−.003
Cd	.046	**.902**	−.016	.234	.132	.117
As	.070	.185	−.047	.001	−.224	**.928**
Pb	.349	**.778**	−.211	.019	−.285	.151
Eigenvalues	1.956	1.684	1.591	1.288	1.128	1.043
% of Total Variance	19.561	16.840	15.913	12.884	11.275	10.426
Cumulative %	19.561	36.402	52.315	65.199	76.474	86.900

[a] Bold and italic values represent strong loadings (>.75).
[b] Bold values repesent moderate loading (.75–.50).

operation of the reservoir elevated the water level and lengthened the channel for large-tonnage ships. After banning outdated small ships, the total number of vessels in the Three Gorges Reservoir was reduced from 9500 in 2003–7937 in 2013. Ships with power more than 22.5 kW all installed water and oil separators. Thus, though the oil wastewater discharge increased from 421,200 t in 2003–510,200 t in 2013, the improvement of the treatment rate has already maintained the pollution of the sewage of ships at a stable level in the past six years (Fig. 6).

4.3. Analysis of the heavy metals associated with PCA and other research

Previous investigations showed that natural processes (weathering and erosion) and the exploitation of multi-metal minerals were the origin of Cu (Wang et al., 2011, 2014b). From results of the PCA, it was found that Cu was frequently linked to COD_{Mn} and F. coli, which demonstrated that Cu may also be derived from agricultural and urban activities. Random combinations of other elements probably indicated various and interactive sources. As and Cd were generated by domestic sewage and industrial waste, and Hg was more likely associated with traffic exhaust (Ye et al., 2011). The source of Pb, which should be of concern, resulted from industrial discharge and the combustion of gasoline (Zhu et al., 2015).

The improvement of the temporal variations of the heavy metals was attributed to the prevention and alleviation of industrial discharge. Industrial wastewater emission declined more than 60% in the past six years (Fig. 6). The spatial variations of the heavy metals also illustrated downward tendencies (Fig. 4). This phenomenon showed the self-purification of the reservoir and can be explained as the dam significantly decreasing the water flow velocity, increasing the possibility of atoms being absorbed with suspended solids and deposited in sludge or sediment (Wei et al., 2009). Concentrations of heavy metals in river sediments were significantly higher than in the water body (Table 3). Therefore, the persistent low level of the dissolved metals in surface water cannot affirmatively substantiate the healthy state of the water quality because perhaps most pollutants are in the sediment and become a potential pollution source (Yi et al., 2011). Absorbed heavy metals

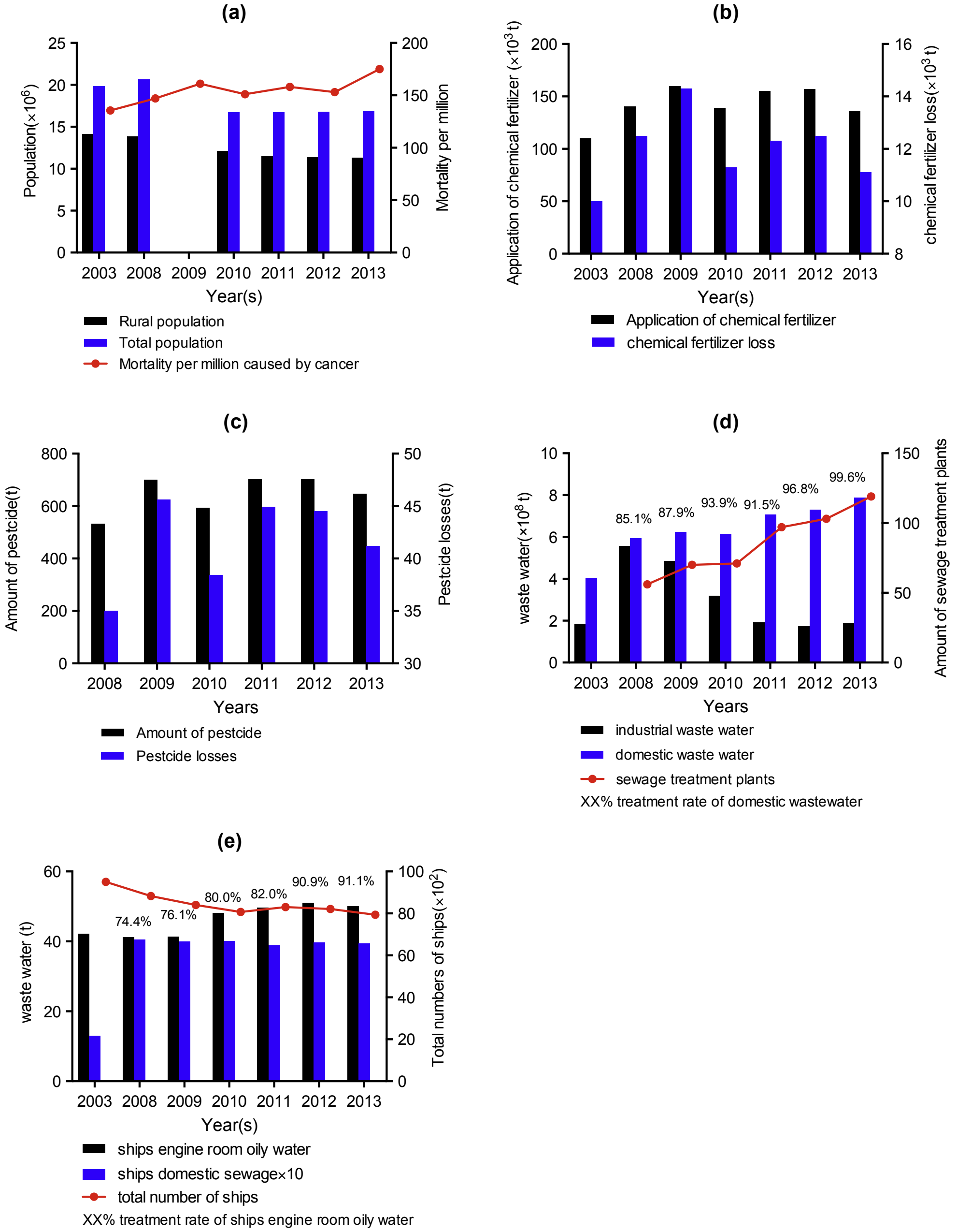

Fig. 6. Population and sources of pollution emissions situation of Three Gorges Reservoir Area from 2008 to 2013 compared with data in 2003.

Table 6
Noncarcinogenic risk results of each element and carcinogenic risk results of As and Pb.

Elements	RfD	$HQ_{ingestion}$	RfD_{dermal}	HQ_{dermal}	HI
Cu	4.00E-02	1.74E-02	2.80E-02	1.34E-04	1.75E-02
Hg	3.00E-04	5.65E-03	2.25E-05	4.06E-04	6.05E-03
Cd	5.00E-04	1.14E-01	2.50E-05	1.23E-02	1.26E-01
As	3.00E-04	3.30E-01	2.85E-04	1.88E-03	3.32E-01
Pb	1.40E-03	5.15E-01	4.20E-04	9.26E-03	5.24E-01
Zn	3.00E-01	1.39E-03	6.00E-02	2.24E-05	1.41E-03
Elements	CSF	$CR_{ingestion}$	CSF_{dermal}	CR_{dermal}	CR_{total}
As	1.5	6.37E-05	1579	1.09E-04	1.72E-04
Pb	.5	1.54E-04	1.667	8.34E-07	1.55E-04

can desorb when environmental conditions such as the pH and temperature change, leading to secondary pollution. (Ye et al., 2011) reported that increased shipping and industrial wastes have deposited large amounts of heavy metals that accumulated in the water level fluctuation zone during the submergence period. Therefore, supervision of those activities could also be beneficial to mitigate heavy metal pollution.

As shown in Table 3, the concentrations of the elements in the Three Gorges Reservoir were much higher than in the headstream at the Tibetan Plateau, which was caused by frequent anthropogenic activities in the province of Sichuan (Wang et al., 2014b). Levels of heavy metals in the Han River, one of major tributaries of the Yangtze Rivers, were much higher than in the reservoir (Li and Zhang, 2010). These levels may indicate that the mid-downstream tributaries made important contributions to the pollution downstream of the Yangtze River. In general, the levels of the heavy metals in the Three Gorges Reservoir were similar to the average of the whole river.

4.4. Influence of the heavy metals to residents' health

From 2008 to 2013, Pb was the only element that exceeded the level of China's and the WHO's drinking water quality guidelines (Table 3). (Zhu et al., 2015) also reported that the concentrations of the heavy metals were low and that the surface water was potable for use by residents living in the Three Gorges Reservoir. However, disturbed by frequent mining, such as in the Panzhihua vanadium–titanium magnetite mining area, human risk assessment results of Pb and As in Yibin were close to hazardous levels. The spatial profile of the preceding report given by Wang et al., 2011, also illustrated the highest concentration of Pb and As of sediments in Yibin. Moreover, other reports on heavy metal contamination covering the whole Yangtze basin (Huang et al., 2008; Wu et al., 2009) all concluded that heavy metal pollution was ubiquitous in the Yangtze Basin, and Pb and As should be of great concern because of their concentration, accumulation, persistence, and toxicity. More studies covering greater ranges, including mainland China and abroad, also clearly indicated that Pb and As pollution have been a global environmental issue (Giri and Singh, 2014; Zhao et al., 2012). The toxicity of As and Pb has been described thoroughly (WHO, 2011). Additionally, although the highest concentrations of Pb and As were observed in Yibin and then decreased as the river flows down, the health hazard caused by Pb and As still deserves high-priority concern because people can be exposed to them by other indirect pathways, such as the consumption of polluted fish (Yi et al., 2011). Aside from fish, livestock and other animals can also accumulate contaminations via the food chain, which will ultimately lead to severe chronic toxic effects to local residents. The risk assessment on drinking water alone cannot account for the real hazard of residents. Additionally, the different chemical property of each element, such as their species transition, absorption and dissolution between sediments and water due to changes of environmental conditions and the individual specificity of each person's age, sex and daily intake, which are hard to quantify, all generate uncertainties in this assessment (Li and Zhang, 2010; Wu et al., 2009). Hence, the human health risk assessment should be further investigated in detail. It is important to formulate an assessment system that is appropriate for inhabitants of the world's largest dam area.

5. Conclusions

A holistic picture of the water quality in the Three Gorges Reservoir since it operated normally was conducted by investigating different types of parameters through various analysis methods. The main conclusions were as follows:

(1) Because of the rapid construction, operation and strict supervision of environmental protection infrastructure, industrial, traffic and urban pollution were under control. Rural pollution, however, deteriorated, as shown by the increasing tendencies of TN and TP and was linked to the abuse of fertilizers and pesticides. The retention of the reservoir played a crucial role in the decreasing concentrations of the heavy metals from upstream to downstream.
(2) Four types of pollution were identified by PCA, mixed, nutrient inputs, organic pollutants and heavy metals, revealing the complexity of pollution in the reservoir area. The results of CCME-WQI, which were applied to the reservoir, showed that the water quality was just fair but slightly improved in those years, suggesting stable and acceptable water quality in the Three Gorges Reservoir.
(3) For the human health risk assessment, drinking water for a lifetime was basically harmless, but the cancer potential of Pb and As should be of concern.

This study captured three key parameters (TN, TP and Pb) and found out that controlling rural pollution should be given priority. The study provided high-quality information and useful methods for helping water quality management efforts. More studies are urged in consideration that the dam will run for many years and has a far-reaching influence.

Acknowledgements

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