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Spatial and Temporal Variations of Water Quality in Songhua River from 2006 to 2015: Implication for Regional Ecological Health and Food Safety

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Abstract: The Songhua River is the largest river in northeastern China; the river's water quality is one of the most important factors that influence regional ecological health and food safety in northeastern China and even the downstream of the Heilong River in Russia. In recent years, the Chinese government implemented several water resource protection policies to improve the river's water quality. In order to evaluate the influence of the new policies on the water quality in the Songhua River, water quality data from 2006 to 2015 were collected monthly from the nine sites along the mainstream of the Songhua River. Results show that the water quality in the Songhua River could be divided into two groups during the last 10 years. Before 2010, water quality in the Songhua River was primarily influenced by regional human activities. Industries were the major pollutant sources in the upstream of the Songhua River. After several new policies were implemented by the local government in 2010, water quality in the Songhua River improved. As a result, the biodiversity of fish and ecological health in the Songhua River improved.

Keywords: Songhua River; water quality; heat map analysis

1. Introduction

From 1949 to 2015, the population in China increased from 0.5 to 1.4 billion [1]. The population growth together with the improving standards of living resulted in an increase in water demand [2,3]. In recent years, with industrial and economical developments, changes in land use types and increasing amounts of wastewater produced by human activities led to water quality deterioration in most of China's river systems [4,5]. In addition, recent climate changes induced many extremes in hydrological variability around the world, ranging from extreme droughts to severe floods [6–8]. Population growth and climate change apply a great pressure to water supply and water quality, especially in China. Many countries started to design and implement related policies to protect and improve water quality and to meet water demand. For instance, the European Union (EU) has designed and implemented the

Water Framework Directive (WFD: 2000/60/EC) to regulate and protect the water resources of each member as national law since 2000. In China, the law of prevention and control of water pollution was implemented in 1984 and then modified in 1996 and 2008 [9,10]. According to the new policies, the integrated discharge standard of water pollutants was implemented in 2012 in order to provide more water resources available for residential utilization [11,12]. However, few studies have evaluated the influence of the new policies on water resources in China, especially in several regions where water resources were seriously polluted by surrounding industries.

Northeastern China was one of the most important industrial centers during the beginning of the People Republic of China in 1950s [13] and is also the most important food production region in China [14,15]. A large amount of pollutants produced by industrial and agricultural discharging have caused serious environmental pollution problems and influenced the water quality of river systems in Northeastern China [16,17]. The Songhua River, an important river in Northeastern China, is the largest tributary on the right side of the Heilong River, which is located at the border of China and Russia [18,19]. The Songhua River flows from the Songnen Plain to the Sanjiang Plain, which are the two most important production regions for corn and soybean in Northeastern China. Therefore, the river's water quality directly influences food safety and production in these two regions [20,21]. Water quality in the Songhua River not only influences the ecological health in Northeastern China, but also affects the downstream of the Heilong River. Changchun and Haerbin are two major cities with nearly ten million people each and many important industries (e.g., Automotive manufacturing and Nonferrous metals) [22,23]. Industrial and residential pollutants from these two upstream cities are the major pollutant sources along the Songhua River.

Although the water quality in the Songhua River is extremely important for food production, safety and sustainability in Northeastern China, few studies have focused on evaluating the water quality and the influence of new water resource protection policies on the water quality in the Songhua River. In order to fill in the research gap, monthly concentrations of water quality related indicators from 2006 to 2015 were collected from nine water monitoring stations along approximately 1000 km of the mainstream of the Songhua River. Based on these data, spatial and temporal variations of water quality in the Songhua River were determined to evaluate the influence of new environment-friendly policies on the water quality of the Songhua River and the implication for regional biological diversity and ecological health.

2. Materials and Methods

2.1. Study Area and Sampling Sites

The Nenjiang River, originating from the Great Hinggan Mountain, and the second Songhua River, originating from the Changbai Mountain, are two major tributaries of the Songhua River and converge at Sanchahe. After the Nenjiang River and the second Songhua River converge at Sanchahe, the downstream of Sanchahe becomes the mainstream of the Songhua River. The water quality monitoring stations are located along the mainstream. Flood seasons in the Songhua River are typically from June to September. The watershed area of the mainstream of the Songhua River is 186,400 km^2 with a length of 939 km from west to east and covers from 124°39′ to 132°31′ E and from 43°01′ to 48°39′ N [24]. In the present study, nine sites, labeled with SH1, SH2, . . . , SH9 from upstream to downstream, were selected as sampling sites (Figure 1).

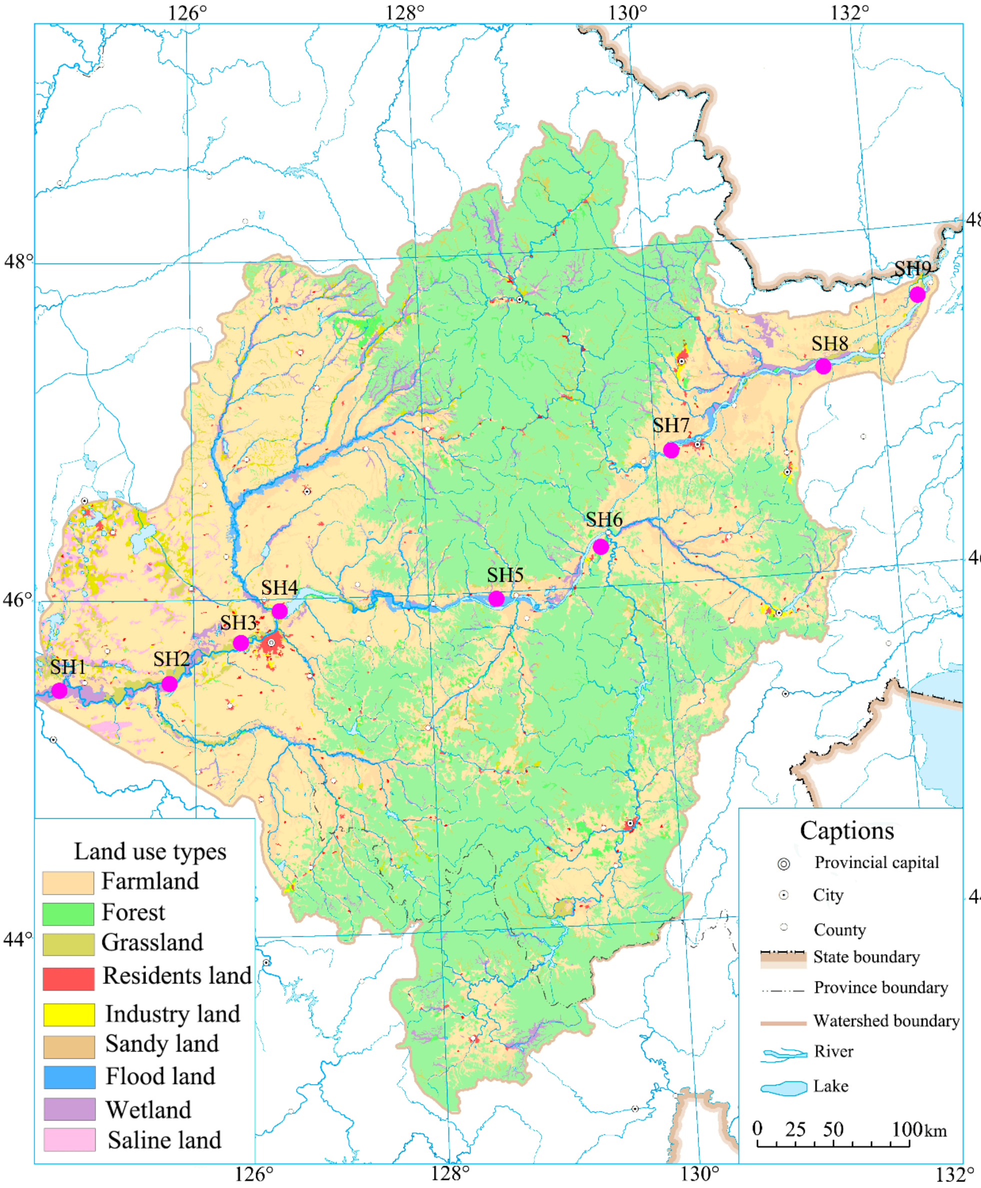


Figure 1. Land use types in the Songhua River basin, location of the mainstream of the Songhua River and the sampling sites in this study.

2.2. Data Sources

From 1956 to 2015, monthly water flows in five hydrological monitoring stations (i.e., SH2, SH4, SH5, SH7 and SH8) were collected from the Songliao Water Resources Commission, Ministry of Water Resources and are shown in Figure S1. Monthly water quality data including pH, NH_3-N, dissolved oxygen (DO), chemical oxygen demand (COD), biological oxygen demand (BOD) and heavy metals (Cu, As and Hg) were collected from hydrological monitoring stations (i.e., SH2, SH4, SH5, SH7, and SH8) and water quality stations (i.e., SH1, SH3, SH6, and SH9) from 2006 to 2015. All water quality indicators were analyzed according to the "regulation for water environmental monitoring SL219-2013" [25]. Briefly, given that there were large differences in concentrations of COD in different stations and samples collecting date, COD were measured by both potassium chromate (COD_{Cr}) and potassium permanganate (COD_{Mn}) methods, thus easily compared to the variation of COD from 2006 to 2010 in the Songhua River. The pH, DO and BOD were measured by the electrode methods. NH_3-N was measured by visible spectrophotometer with a wavelength of 420 nm. As and Hg were measured by Atomic Fluorescence Spectrometer (AFS), and Cu was measured by Atomic Absorption Spectrometer (AAS) [25]. Population and annual gross domestic product (GDP) in the two major cities along Songhua River (Haerbin and Changchun) during 2006 and 2015 were collected from the China Statistical Database [1].

2.3. Statistical Methods

Cluster analysis was used to classify the objects of the system into categories/clusters based on their nearness/similarity, and the dendrogram was used to show the results of the cluster analysis [16,26,27]. Heat map analysis is a false color image with two dendrograms for two different objects and can divide these two objects into several groups. The order of the different influence factors in these two objects were reordered according to their nearness or similarity based on cluster analysis. In this study, heat map analysis was used for evaluating the spatial and temporal variation of water quality in the Songhua River. For evaluating the spatial variation of water quality in the Songhua River, the water quality indicators in each site were used for heat map analysis, and the nine sampling sites along the Songhua River were divided into several groups based on the average of water quality indicators. For evaluating the temporal variation of water quality in each sampling site, annual average water quality indicators and years were regarded as two objects in heat map analysis. Heat map analysis were calculated using the "heatmap.2" function in the R environment for statistical computing with the package "gplots" [28,29].

3. Results

3.1. Variations of pH, NH_3-N and DO

The variation of pH, NH_3-N and DO in different sampling sites from 2006 to 2015 are shown in Figure 2. Before 2010, pH values in SH1 and SH2 were lower than 7.0, although increased gradually from 2006 to 2010. After 2010, pH values in SH1 and SH2 were stable and fluctuating around 7.5. pH values in the other seven sampling sites were similar and between 7.0 to 8.5 for the last 10 years. There exists obvious different of water flows in the Songhua River at between flood seasons and non-flood seasons (Figure S1). In all sampling sites, pH values during non-flood season were higher than those during flood season. Similar to pH values, concentrations of NH_3-N and DO were higher during non-flood season than those during flood season. Differences in NH_3-N between the seasons were obvious; concentrations of NH_3-N during flood season were around 1.5 mg/L, more than two times the non-flood season concentration. In SH2 and SH3, prior to 2008, concentrations of NH_3-N were from 1.0 to 3.0 mg/L, which was much higher than other sampling sites. After 2008, the concentrations of NH_3-N in SH2 and SH3 decreased gradually and became similar to other sampling sites. In addition, in other sampling sites, NH_3-N concentrations were from 0.5 to 2.0 before 2010 and slightly decreased after 2010 (0.3–1.5 mg/L). The concentrations of DO in all sampling sites were between 5 and 12 mg/g, and low concentrations of DO had always appeared during flood seasons for the last 10 years.

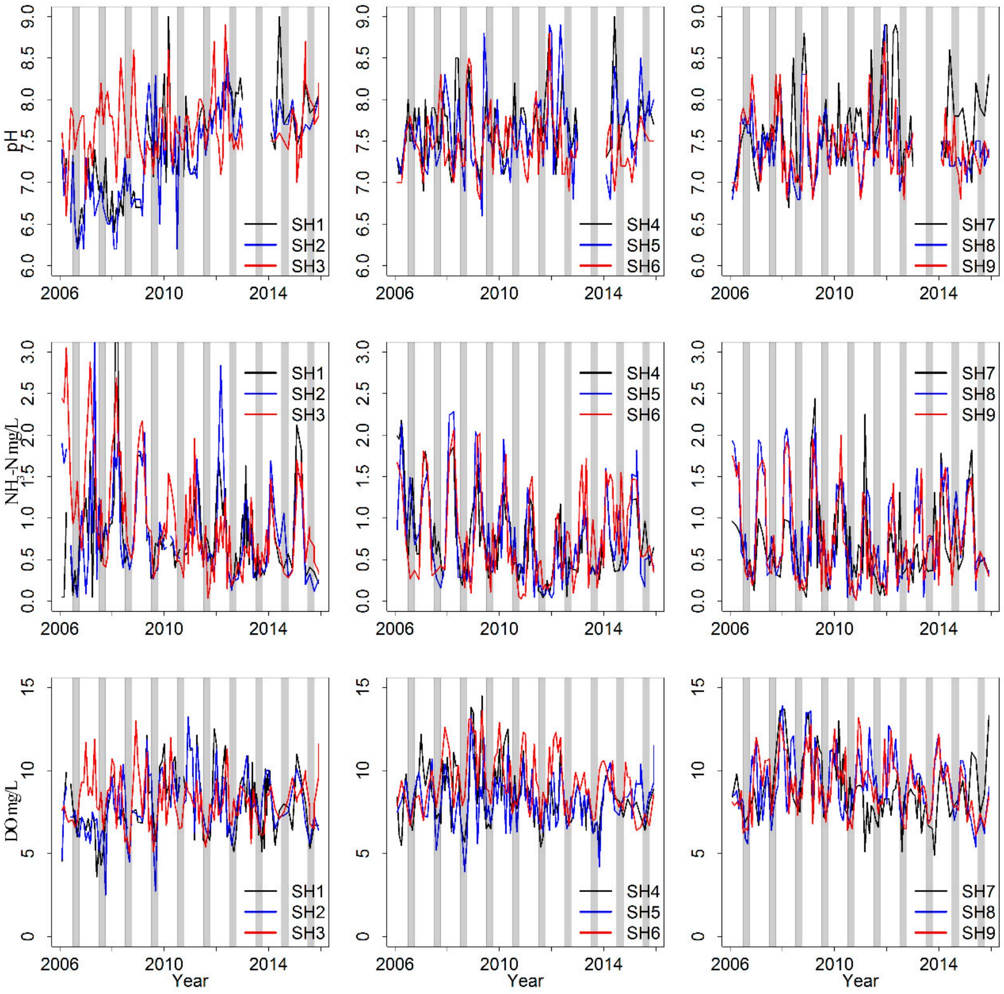


Figure 2. Monthly variation of pH, NH_3-N, and DO in nine sampling sites from 2006 to 2015. The grey shadow means the flood season in the Songhua River.

3.2. Variation of Oxygen Consumed Pollutants Index

Variations of COD measured by the $K_2Cr_2O_7$ and $KMnO_4$ methods and BOD in all sampling sites are shown in Figure 3. In all sampling sites, the concentrations of COD_{Cr} decreased gradually from 2006 to 2015. For SH6, SH8 and SH9, COD_{Cr} concentrations decreased from 35 mg/L in 2006 to 20 mg/L in 2015. Before 2010, COD_{Cr} in SH1, SH2 and SH5 were higher than 30 mg/L for several months. After 2010, the COD_{Cr} concentrations in the upstream (i.e., SH1, SH2, SH3, SH4 and SH5) fluctuated within 10–20 mg/L, which was lower than those downstream (20–30 mg/L). Results of COD_{Mn} in downstream were different from those measured by Cr method; there was no clear decreasing trend of COD_{Mn} before 2010. After 2010, the COD_{Mn} decreased greatly and stabilized from 2012 to the present. Unlike COD concentrations increasing from the upstream to downstream, there were no obvious differences in BOD concentrations from SH3 to SH9; BOD concentrations ranged from 1 to 3 mg/L for the last 10 years. However, before 2010, the BOD in SH1 and SH2 were in the range 4–10 mg/L and much higher than other sampling sites. After 2010, the BOD in these two sampling sites decreased, and the concentrations of BOD in these two sampling sites fluctuated between 1 and 3 mg/L after 2014.

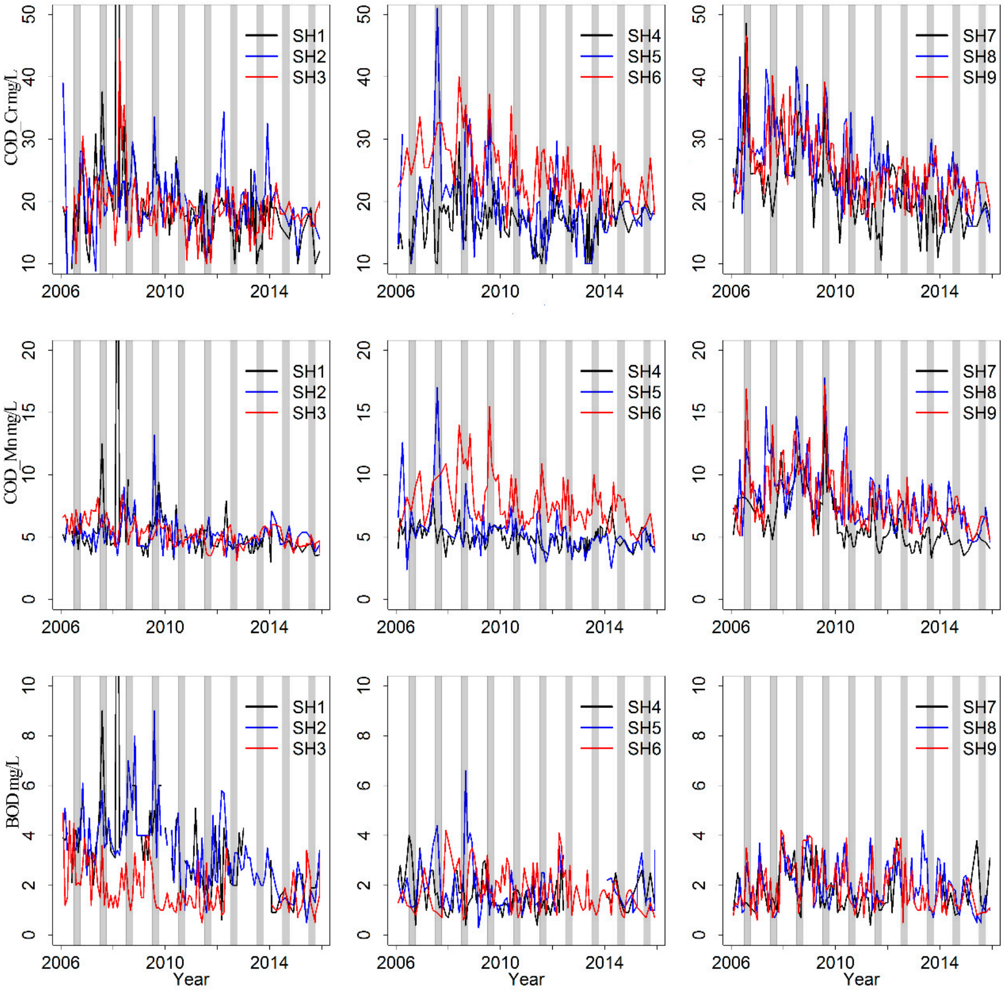


Figure 3. Monthly variation of COD_{Cr}, COD_{Mn} and BOD in nine sampling sites from 2006 to 2015. The grey shadow means the flood season in the Songhua River.

3.3. Variation of Heavy Metals

In this study, we used the concentrations of Cu, As and Hg as indicators to evaluate the degree of heavy metal pollution in the Songhua River (Figure 4). Before 2012, the range of Cu concentrations in SH1 and SH2 were from 5 to 80 μg/L and much higher than those in other sampling sites. In addition, in SH8, high Cu concentrations (higher than 60 μg/L) appeared for several months before 2008. After 2012, the Cu concentrations in SH1 and SH2 decreased greatly and fell below detection limit (4 μg/L). Cu concentrations in SH3, SH4 and SH5 were below detection limit during the last 10 years. In SH7 and SH9, the Cu concentrations fluctuated from 0 to 20 μg/L before 2010, and fell below detection limit in most months from 2010 to the present. Similar to Cu, the concentrations of As in SH2 and SH3 were from 0 to 10 μg/L before 2010 and higher than those after 2010. Three times as high As concentrations (around 10 μg/L) were found in SH2 and two times as high concentrations were found in SH4 during non-flood season. Prior to 2010, concentrations of As in most downstream sites were below the detection limit and fluctuated from 0 to 1 μg/L after 2010. Hg concentrations in all sampling sites except SH1 and SH2 were near the detection limit with fluctuations from 0 to 0.05 μg/L. In SH1 and SH2, Hg concentrations were higher than 0.4 μg/L for a few months before 2008. After 2008, Hg concentrations in SH1 and SH2 decreased greatly and were close to the detection limit and similar to the other sampling sites.

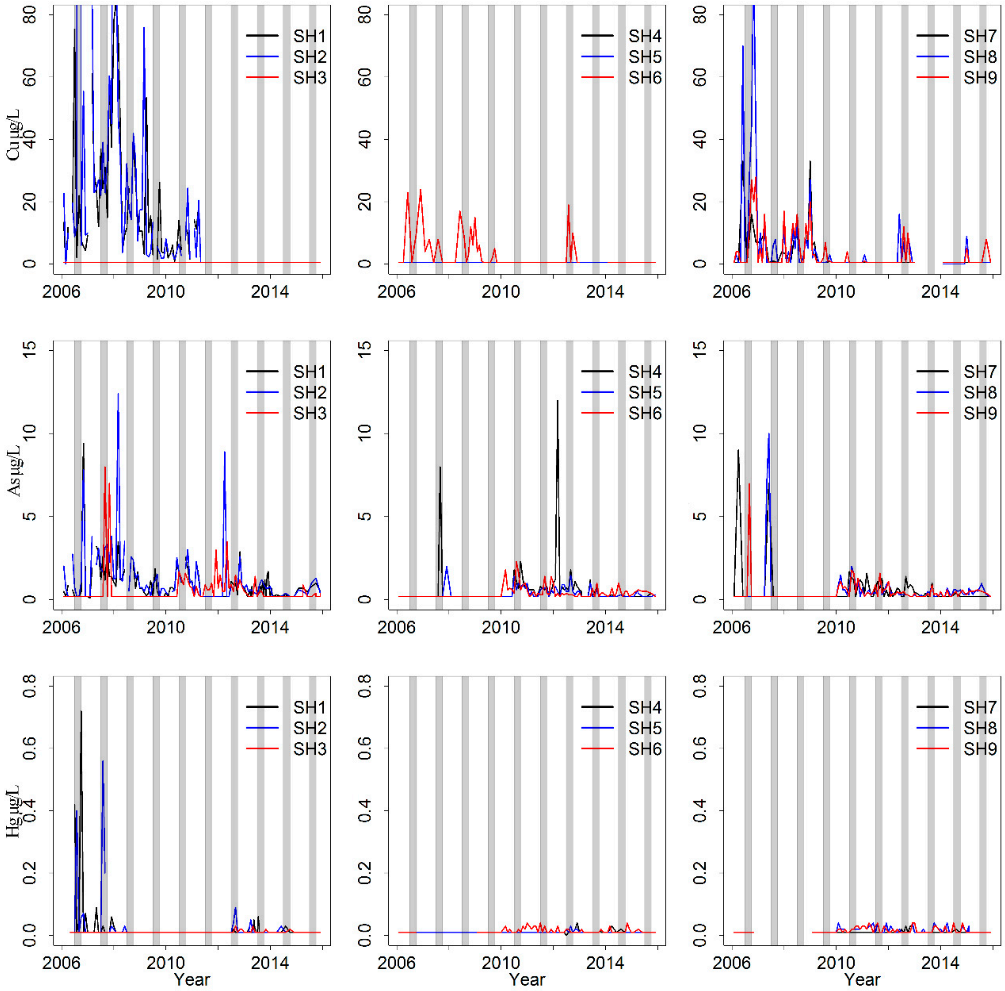


Figure 4. Monthly variation of Cu, As and Hg in nine sampling sites from 2006 to 2015. The grey shadow means the flood season in the Songhua River.

4. Discussion

4.1. *Spatial Variations of Water Quality in Songhua River*

In order to evaluate the spatial variations of water quality in the Songhua River, the average concentrations of each indicator from the nine sampling sites during 2006 and 2015 were used for heat map analysis, and the results are shown in Figure 5. Based on the heat map analysis, the nine sampling sites can be divided into three groups from upstream to downstream. The first group included SH1 and SH2. In these two sites, the scores of heavy metals and BOD were the highest, and the scores of other indicators were the lowest. High scores of heavy metals and BOD indicate high concentrations of heavy metals and BOD. Regional industry development often accompanied increased concentrations of heavy metals in regional ecosystems [30]. In addition, organic pollutants produced from industrial and residental sources could lead to increases in concentrations of BOD in the water system [31,32]. Changchun, a huge city with more than half of its GDP coming from industries and a population greater than seven million, is located at the upstream of these two sites (Figure S2). Fast developing industry in Changchun could be the main cause of higher concentrations of heavy metals and BOD in the upstream of the Songhua River than those in the downsteam, as indicated by the higher scores of related indicators in the SH1 and SH2 sites.

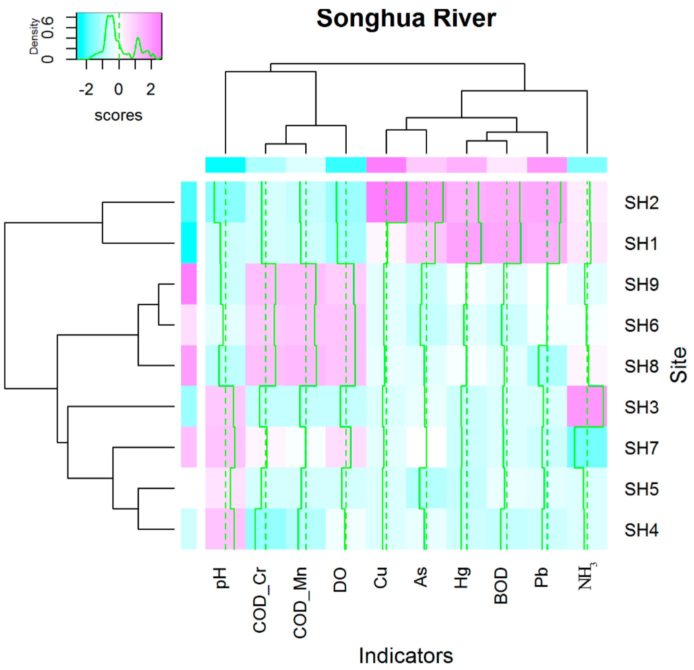


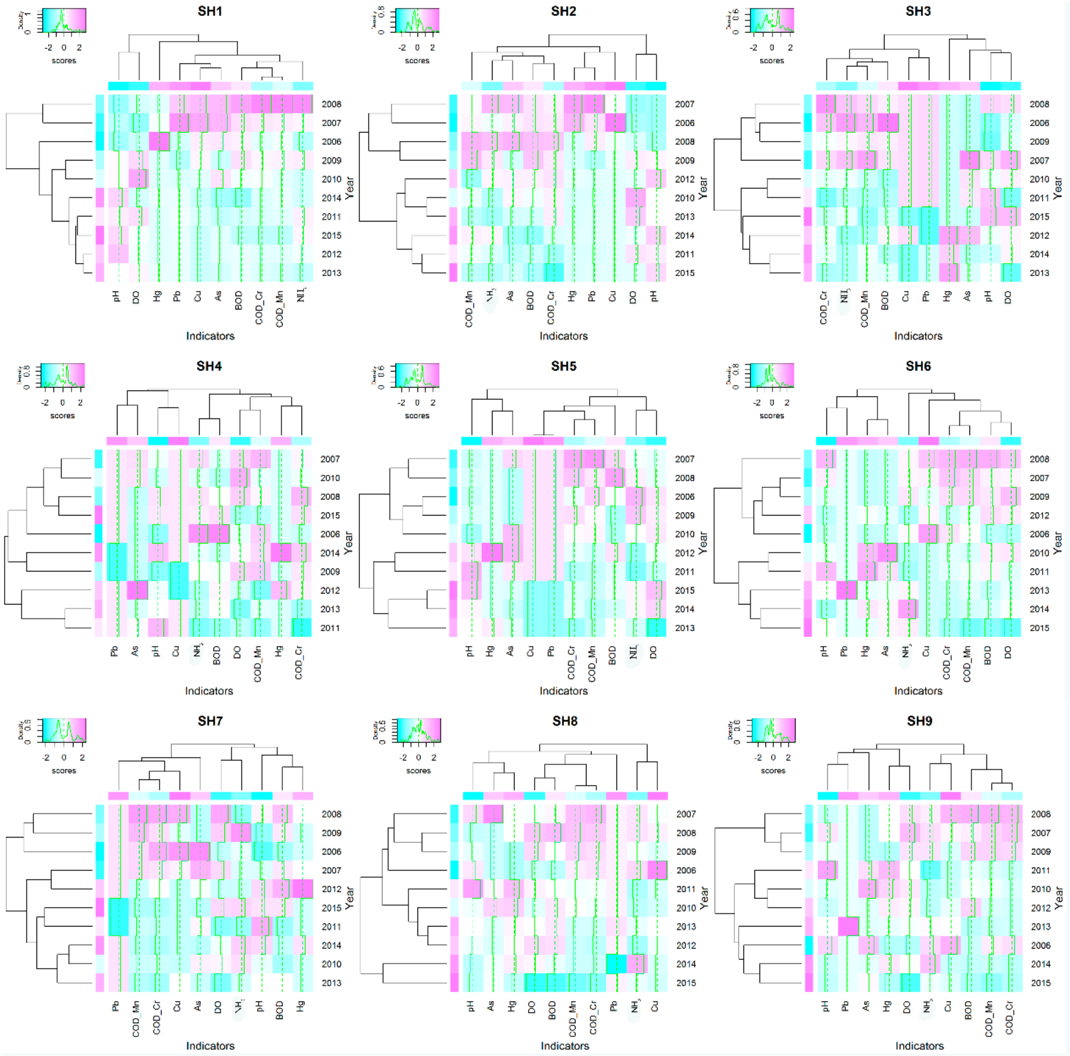
Figure 5. Ten-year average of water quality in nine sampling sites grouped by heat map analysis.

From upstream to downstream, the pollutants in the river ecosystem are generally dilluted and the concentration of pollutants decreases gradually. Sampling sites in the downstream with low concentrations of heavy metals and BOD can be divided into another two groups: sampling sites located at the midstream (SH3 to SH6) in one group and those at the downstraem (SH7 to SH9) in another group. In the midstream of the Songhua River, the values of pH were higher and the values of COD_{Cr}, COD_{Mn} and DO were lower than those in the downstream. The downstream of the Songhua River flows through the Sanjiang Plain, a major farmland and wetland distribution region of China. Wastewater from a large area of farmland discharges into the mainstream of the Songhua River through the tributary rivers; high concentrations of nutrient elements in the agricultural wastewater could lead to concentration increases in the Songhua River [33–35]. As a result, plants and alga in the downstream of the Songhua River could be more abundant and more DO existed in the river [36,37]. In addition to nutrient elements, organic pollutants produced from agriculture also flow into the river system; nutrients and organics both caused COD to be greater in the downstream than the midstream of the Songhua River. High concentrations of organic pollutants and DO might also likely lead to more microbial activities in the downstream, resulting in more CO_2 produced by microbes, thus lowering the values of pH in the downstream compared to that in the midstream.

In general, water quality in the nine sampling sites of the mainstream of the Songhua River can be divided into three groups. From upstream to downstream, the first two sampling sites were grouped into the first group because of their high concentrations of heavy metals at the two sites. With the self-purification of the river and no large sources of heavy metals, concentrations of heavy metals decreased greatly in the midstream and downstream of the Songhua River. pH values and DO concentrations were controlling factors that led the rest of the sampling sites (from SH3 to SH9) to be grouped into second and third groups. Low concentrations of DO and COD in the midstream of the Songhua River resulted in less CO_2 produced by microbial activities and high pH values in the midstream of the Songhua River, therefore SH3 to SH6 sampling sites in the midstream were grouped into the second group. Wastewater from farmland discharging into the downstream of the Songhua River led more nutrient elements and organic pollutants into the downstream of the Songhua River, which caused higher DO and COD concentrations than that of the midstream of the Songhua River. Thus SH7 to SH9 sampling sites were grouped into the third group.

4.2. Temporal Variations of Water Quality in Songhua River

Annual average concentrations of the nine indicators from 2006 to 2015 were used for evaluating the temporal variations of water quality in each sampling site. Similar to the spatial heat map analysis, indicators and years were treated as two objects to classify groups, as shown in Figure 6. Based on the heat map analysis in the previous section, water quality in the nine sampling sites could be divided into three groups. The first group located at the upstream of the Songhua River included SH1 and SH2. In this group, high concentrations of heavy metals and low pH values were seen in the initial years of the past 10 years. In SH1, the high concentrations of heavy metals and low pH values in the first two years (i.e., 2006 and 2007) were different from the other periods and similar trends of heavy metal concentrations were also observed in SH2 in the first four years. In later periods, water quality in the upstream of the Songhua River improved and the values of pH and DO concentrations increased from 2010 to 2015. As mentioned in the previous discussion section, industrial wastewater from two large cities resulted in the high concentrations of heavy metals in the upstream of the Songhua River between 2006 and 2009. With industry wastewater quality standards rising and more stringent environmental monitoring by local governments, less untreated wastewater flowed into the Songhua River [12], thus greatly improving the water quality in the upstream of the Songhua River. For instance, concentrations of DO increased from 2010 to 2015 and the pH also increased gradually.



SH1 SH2 SH3 SH4 SH5 SH6 SH7 SH8 SH9

Figure 6. Annual average of water quality in each sampling site in different years grouped by heat map analysis.

Two different periods of water quality divided in 2010 were also found in the midstream of the Songhua River (i.e., SH3, SH4, SH5 and SH7). Before 2010, concentrations of COD and BOD were consistently higher than those after 2010. The concentrations of Cu and Pb in the midstream of the Songhua River before 2010 were higher than those after 2010. High concentrations of COD and BOD in the midstream of the Songhua River could primarily be produced from residents along the river. The midstream of the Songhua River is located at the downstream of a large city named Harbin, which has a population greater than nine million. In addition, the GDP of tourism in Harbin was a major component of its total GDP during the last 10 years (Figure S2). A large number of tourists in Harbin additionally increased the residential pollutants and concentrations of COD and BOD. With more environment-friendly policies implemented by the Harbin government, the quality of wastewater treated by wastewater treatment plants improved greatly. Therefore, the COD and BOD concentrations in the downstream of Harbin decreased. Concentrations of COD and BOD after 2010 were much lower than those before 2010.

The heat map analysis clearly shows that the annual average values of the indicators in the downstream of the Songhua River can also be divided into two groups and the first group included most of the years before 2010. Similar trends of greater COD and heavy metal concentrations in the downstream when compared to those in the midstream of the Songhua River were found. However, unlike the variation of NH_3-N in the midstream of the Songhua River, the concentrations of NH_3-N in the downstream decreased greatly from the first period to the second period. Wastewater from regional farmland discharging into the mainstream of the Songhua River might be the major causal factor increasing the concentration of NH_3-N in the downstream of the Songhua River in the first several years. However, with local governments regulating and monitoring agricultural wastewater and nutrients, pollutants from the wastewater discharging into Songhua River decreased, thus the concentrations of NH_3-N in downstream of the Songhua River decreased.

In summary, government policies and human activities greatly influenced the water quality in the Songhua River. Before strict environmental protection policies were implemented, wastewater produced from industry in the upstream caused the high concentrations of heavy metals, residential wastewater led the high concentrations of COD and BOD in the midstream, and wastewater from agriculture led to high concentrations of NH_3-N in the downstream. After the implementation of strict environmental protection policies by the local governments in 2010, less wastewater flowed into the Songhua River. Concentrations of pollutants in the mainstream of the Songhua River decreased greatly, and the water quality in the Songhua River greatly improved.

4.3. Water Quality of Songhua River and Regional Ecological Health

Land use types along the Songhua River are one of the most important factors that influence the water quality in the Songhua River. Therefore, they also influence ecological health and sustainability in different ecosystems along the Songhua River. In the upstream of the Songhua River, the main land use types along the river are farmland and grassland (Figure 1). From SH1 to SH3, industrial and transportation lands are also distributed along the Songhua River. Recently, more and more wetland reserves have been established and protected in the upstream and midstream of the Songhua River. Wetland distribution areas could work as natural water purification zones to improve the water quality in the Songhua River [38,39]. In the midstream of the Songhua River (From SH4 to SH6), farmlands are primarily distributed along the mainstream of the Songhua River, and forests are distributed in the tributary region of the Songhua River. In the downstream of the Songhua River, the proportions of forest in the tributary regions decreased while farmland increased. Different types of land and major human activities along the Songhua River influenced the river water quality. Sanjiang plain, one of the most important farmland regions in China, is located at the downstream of the Songhua River. Water quality influenced by pollutant sources in the upstream of the Songhua River also affected the water quality in the downstream, thus directly influencing food safety in regional agriculture.

Water quality of the Songhua River is one of the most important factors that influence the biodiversity of fish in the Songhua River. With the implementation of environment-friendly policies combined with strictly environmental monitoring, water quality in the Songhua River improved greatly, especially after 2010. In 2010, 69 species, 18 families, and eight orders belonging to two different classes (i.e., Cyclostomata and Pisces) existed in the mainstream of the Songhua River. However, nearly 60 and 55 species of fish were found in SH9 and SH7, respectively. From downstream to upstream, the species of fish decreased; only 19 species were found in SH3, and migratory fish could only be found in the downstream of the Songhua River [40]. Interestingly, in 2012, the species in downstream of the Songhua River increased; 68 species of fish were found between SH6 and SH7 [41]. In the present study, we found five more fish species in 2015 than in 2010, and 65 species were found at the SH9 site. Surprisingly, the number of fish species increased greatly in the SH3 site and 50 species were found in 2015, more than double of that (19 species) in 2010. Some fish species (e.g., *Huchotaimen pallas*) disappeared due to bad water quality and overfishing in the Songhua River, but were found for the first time in the Songhua River in recent years. Water quality is one of the important factors that influence the number of fish species in the Songhua River and good water quality in the Songhua River could result in the number of species and amount of fish in the Songhua River to increase. By reducing wastewater along the mainstream of the river and implementing new environmental policies, the better water quality in the Songhua River in recent years led the biodiversity of fish in the Songhua River to increase. The increased biodiversity of fish accompanied with the reappearance of some previously disappeared fish indicates that the ecological health and sustainability in the Songhua River have improved in the recent 10 years.

Water quality in the Songhua River improved recently; concentrations of most pollution elements were below detection limit and the ecological health and sustainability in the Songhua River and other ecosystems along the Songhua River improved in recent years, especially after 2010. However, during non-flood seasons, the major pollutants in the Songhua River were organic pollutants, which led the COD to increase obviously. More environment-friendly policies should be implemented by governments along the Songhua River and governments should pay more attention to the water quality in the Songhua River during non-flood seasons.

5. Conclusions

In this study, we collected the monthly water quality data in nine sites along the mainstream of the Songhua River from 2006 to 2015. Based on the variations of water quality and heat map analyses, we found that the water quality in the upstream of the Songhua River was primarily influenced by industry wastewater and that the water quality of the downstream was majorly affected by residential and agricultural wastewater. With more environment-friendly policies implemented, fewer pollutants in treated wastewater flowed into the Songhua River. As a result, the water quality in Songhua River improved significantly after 2010. Improved water quality in the Songhua River led some fish species that were previously only found in the downstream to be found in the midstream of the Songhua River, thus improving the biodiversity of fish in the Songhua River. Water quality improvement in the Songhua River, decrease in the influence of human activities on regional ecosystems, and more environment-friendly policies designed and implemented by local governments along the Songhua River are important for the sustainability of regional ecosystems, ecological health and food safety in the future.

Supplementary Materials: The following are available online at www.mdpi.com/2071-1050/9/9/1502/s1, Figure S1: Monthly average of water flow in flood season and non-flood season in five hydrological stations of Songhua river from 1956 to 2015, Figure S2: Population, gross domestic product (GDP) of agriculture, industry, tourism and total GDP in Haerbin and Changchun which are two major cities along the Songhua river.

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Author Contributions: C.W. and G.W. conceived and designed the experiments; C.W. and C.G. performed the experiments and collected the data; C.G. contributed analysis tools and prepared figures; C.W., C.G. and G.W. analyzed the data and wrote the paper together with D.H., W.Z., and Q.L.

Conflicts of Interest: The authors declare no conflict of interest.

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