Water Quality Assessment and Apportionment of Pollution Sources of Selected Pollutants in the Min Jiang, a Headwater Tributary of The Yangtze River

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Abstract

This paper analyzed the spatial-temporal variations of surface water quality along the middle and lower reaches of the Min Jiang between 2003 and 2012 and investigated its pollution sources by analyzing the data from 4 water quality monitoring stations. The results showed that surface water quality was higher polluted in the middle reaches of the Min Jiang than that in the lower reaches and its tributary. Seasonal and spatial differences were found for DO, CODmn and NH3-N, whereas for TP the differences were mainly due to the water quality station. The level of organics (CODmn) was higher in summer (high flow period), and the level of NH3-N was higher in winter (low flow period). In the middle reaches of the Min Jiang, point sources (from wastewater treatment plants and industrial effluents) were found to be the dominant inputs of organics (CODmn) and nutrients (NH3-N and TP) to river. In the lower reaches of the Min Jiang, diffuse sources (from agricultural fertilizer, soil erosion, etc.) were the dominant contributor of organics and TP to river, while point sources were the dominant input of NH3-N. In tributary, diffuse sources were the dominant organics and TP input, both point and diffuse sources were dominant NH3-N inputs. Overall, these results reinforced the notion that pollution control by periods and regions was important for effective water quality management, and it is necessary to enhance the treatment of industrial effluent, to strictly carry out the discharge standard for water pollutants and the total amount control system, to incorporate NH3-N in the total amount control system in the Min Jiang.

Introduction

Nowadays, the surface water pollution is of great environmental concern worldwide because of it has hastened the scarce of water resources, and affected sustainable development and human healthy. Rivers are highly vulnerable water bodies to pollution due to their roles in assimilating or carrying off the municipal and industrial wastewater and run-off from agricultural land in their vast drainage basins. Surface water quality is controlled by complex anthropogenic activities and natural factors. The discharge of municipal and industrial wastewater is kind of a constant polluting source. However, the surface run-off is a seasonal phenomenon and highly affected by weather condition. Seasonal variations in precipitation, surface run-off, interflow, groundwater flow and pumped inflow and outflow have strong impacts on river discharge and subsequently on the concentration of pollutants in rivers [1]. Due to the complexity of water environments, water quality specialists and decision-makers are confronted with significant challenges in their efforts to manage surface water resources [2]. Therefore, identifying temporal and spatial changes in water quality in river basins is an imperative task, so as to provide an improved understanding of the environmental conditions and help researchers establish priorities for sustainable water management [3-6].

The temporal pattern of total pollutant load inputs to a river from point and diffuse sources is fundamentally different. Loadings from point sources, such as STWs and industrial effluents, tend to be relatively constant throughout the year, and are generally independent of river flow. Diffuse sources (from agricultural fertilizer, soil erosion, septic tank soak-aways and atmospheric deposition) are principally flow dependent, and should occur intermittently, particularly during the periods of the year with high precipitation. This temporal difference in the mode of pollution delivery results in clear differences in the relationship between pollutant concentration and river discharge [5,7]. Furthermore, the role of pollution source from the point and diffuse

inputs can be identified. In rivers that are point source dominated, the constant rate of input of contaminations means that pollutant concentrations will be highest at low flow, and this concentration will decrease reciprocally with increasing river flow rate, due to dilution. Conversely, rivers that receive pollutant primarily from diffuse sources will tend to show an increase in pollutant load and concentration with increasing river flow [7].

The Min Jiang River has recently been the focus of attention due to recognition of the increasing stress being placed on its water resources and of the resulting environmental degradation in the Yangtze River basin. Since the mid-1970s, the Sichuan Basin area has undergone rapid development. Locally, industry, agriculture, and domestic activities have posed great pressure on the ecological environment, especially the aquatic environment [8]. At the end of the 1990s, the percentage of water quality poorer than Grade III was 30.1% in the Min Jiang based on the Environmental Quality Standards for Surface Water GB3838-2002 in China, see (Table 2). In contrast, the percentage of water quality poorer than Grade III was 41.26% in 2006. In order to suspend the deterioration of water environments and improve the surface water quality, Sichuan province has been urged to take serious actions during the 11th Five Years program (2006-2010). Surface water quality of the Min Jiang has been improved in recent years, but according to

the comprehensive evaluation of 2011, 29.4% of total length was still over grade III. In order to have an effective, long-term management and reduce the constituent concentrations in the Min Jiang we need to acquire understanding of the behavior and the variation of water quality parameters and major pollution sources within the catchments. However, only a few studies on water quality in the Min Jiang have focused on nitrogen contamination [9] and sediment yields [10]. Recent studies of surface water quality in the Min Jiang have reported on the chemical and physical weathering [8,11]. Further investigation of water contamination and pollutant sources is needed.

The objective of this study is to analyze the temporal and spatial variations of the water quality of the Min Jiang and to identify the pollution sources by exploring the concentration-flow relationships. Findings from this study will extend the available information for effective water management for the watershed.

Material and Methods

Study area

In this work, we studied the middle and lower reaches of the Min Jiang, from Dujiangyan to YiBin city, with a length of approximately 370 km (Figure 1). The middle and lower reaches of the Min Jiang has a subtropical climate, with average annual temperatures between 15~18°, and annual precipitation between 1200~1500 mm. Precipitation is concentrated from May to October, during which about 75% of the total annual precipitation occur.

The river serves as a major source of domestic and industrial water supply for nearby cities: Chengdu (population of 11,120,000), Meishan (population of 3,450,000), Leshan (population of 3,530,000) and Yinbin (population of 5,270,000), which are the major urban settlements on the banks of the Min Jiang. Subsequently, the river receives domestic and industrial wastewater from these cities and numerous minor settlements along the river.

Monitoring stations and data

In this study, eight water quality monitoring stations were selected under the river quality monitoring network of Min Jiang basin (Figure
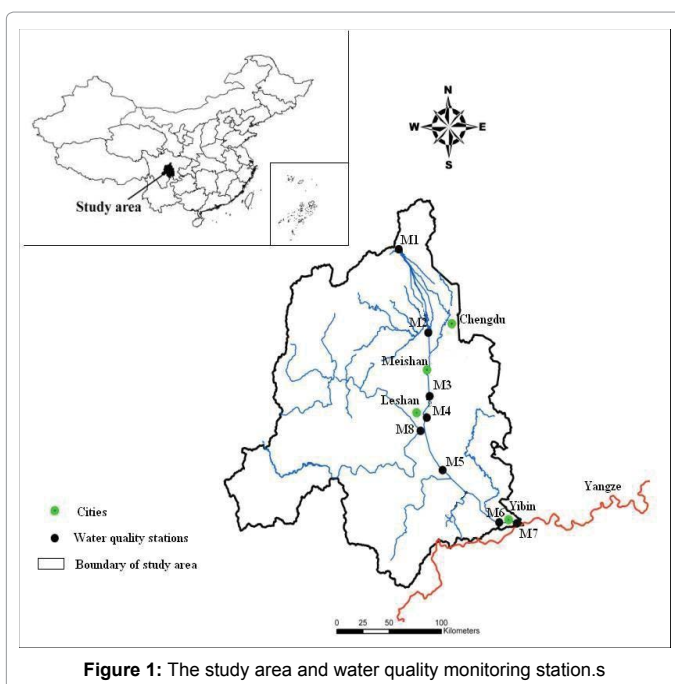

Figure 1: The study area and water quality monitoring station.s

1). Four stations, M1, M2, M3 and M4, on the middle reaches of the main stream of the Min Jiang, three stations M5, M6 and M7, on the lower reaches of the main stream of the Min Jiang, and one station (M8), on the Dadu River, which is the largest tributary of the Min Jiang. These water quality stations were selected because of the completeness of the hydrology data series. Table 1 gives a description of the water quality monitoring stations with their locations and types.

The monthly water quality data for eight water quality monitoring stations between 2003 and 2012 were obtained from Environmental Monitoring Center of Sichuan province. The analyzed water quality parameters include dissolved oxygen (DO), potassium permanganate index (COD_{mn}), ammonia nitrogen (NH_3-N) and total phosphorus (TP). The sampling, preservation, transportation and analysis of the water samples were carried out following standard methods [12]. Daily flow data at four water quality monitoring stations (M4, M5, M7 and M8) between 2006 and 2012 were obtained from Data-sharing network of China hydrology.

Load estimation method

The method used to estimate the pollution loads was based on averaging estimators, also called integration or interpolation methods, use the means of concentrations and flows over a time interval, and has been widely accepted. All the available flow data in the sampled period were used in this method. The equations used were:

$$L_w = \frac{\sum_{i=1}^{n} A_i C_i}{\sum_{i=1}^{n} A_i} \frac{\sum_{i=1}^{n} Q_i}{n} n = \overline{C} \cdot \mu_q \cdot n \qquad (1)$$

With:

$$\mu_q = \frac{\sum_{i=1}^{n} Q_i}{n}$$

where A_i represents the indicator for availability of concentration data (1 if data is available, 0 if not), C_i the concentration on day i, Q_i the average flow on day i, n the total number of days for the period of load estimation. Over bars denote sample arithmetic means, and L_w the resulting load.

Furthermore, to study the point and diffuse source load, L_w were considered for each monthly period. With the average load of low flow months is multiplied by 12 to estimate the annual point source load. Annual load minus point source load is the load of diffuse.

Statistical procedures

Two-way analysis of variance (ANOVA) was performed to estimate the temporal and spatial differences of water quality in the River. Previously, normality and homogeneity of data were checked by means of the Kolmogorov-Smirnov and Levene tests, respectively. The non-parametric Kruskal-wallis'test (K-W) was used for DO, COD_{mn}, NH_3-N and TP, as they were either not homogeneous or not normal distributed [13].

In this study, the relationships between concentration (C) and river flow (Q) were studied by means of regression techniques for different models such as power ($C=aQ^b$), hyperbolic ($C=a+b/Q$), exponential ($C=ae^{bQ}$), linear ($C=a+bQ$) and logarithmic ($C=a+b\ln(Q)$). Different models were proposed to describe the relationship between concentration-flow [14-16] The best regression model was chosen according to maximum correlation coefficient [16].

Results and Discussion

Water quality status

The mean value and standard deviation of the eight water quality parameters from eight water quality monitoring stations in the middle

and lower reaches of the Min Jiang during 2003-2012 were summarized in Table 2 Based on the "Environmental Quality Standards for Surface Water" of China, the surface water environment is divided into five grades, and each grade has its corresponding standard value (Table 2). In the area, the values of pH were within permissible range of surface water (6.0-9.0) at all stations. Both DO and BOD_5 reached grade I at stations M1, M2, M5 M6, M7 and M8, grade III at station M4, while DO reach grade IV at station M3. The COD_{mn} reached grade I at station M1, grade II at stations M2, M5, M6, and M7, grade III at station M4, and grade IV at station M3. The NH_3-N reached grade I at station M6, grade II at four stations (M1, M5, M7 and M8), grade III at stations M2 and M3, while over grade V at station M4. The TP reached grade II at station M8, grade III at stations M1, M5, M6 and M7, and grade IV at station M2, M3 and M4.

It must be emphasized that average concentrations of some variables such as DO, COD_{mn}, NH_3-N and TP in the middle reaches of the Min Jiang are over grade III or V, whereas only water with a grade lower than grade III was allowed in this region, and over grade V is the worst score in the national standard for water quality in China. Therefore, the water resource of the middle reaches of the Min Jiang was not suitable for human consumption or industrial purposes.

Temporal trends of water quality

Two-way ANOVA and K-W test showed significant temporal differences (P<0.05) for all water quality parameters except TP among the 120 sampling times over the nine year period (2003-2012). Temporal variations of the eight variables were illustrated by box-whiskers plots (Figure 2).

The Water temperatures (Figure 2) demonstrated a seasonal pattern. The DO reflected the same temporal patterns showing higher

values in winter and lower values in summer, with a range of 6.42 to 8.70 mg/L and 0.2 to 11.9 mg/L, respectively. In contrast, the COD_{mn} reflected the same temporal patterns showing higher values in summer and lower values in winter, with a range of 0.2 to 13.2 mg/L and 0.4 to 38.6 mg/L, respectively. Regarding nutrients, the concentrations of NH_3-N varied from 0.05 to 9.50 mg/L, with lower values in summer (July and August), and higher values in March. TP was absence significant temporal various, with a range of 0.03-8.23 and 0.01-1.20 mg/L, respectively.

The higher values of COD_{mn} and lower values of DO in summer are influenced by various factors. Higher rainfall and river-flow cause the wash of organic matter into the surface water, which decreases the concentration of dissolved oxygen with biodegradation, whilst the increase in temperature causes a decrease in oxygen solubility, thus causing a further reduces the DO concentrations [17]. In addition, as the amount of available DO decreases, undergoes anaerobic fermentation processes leading to formation of organic acids. Hydrolysis of these acidic materials causes a decrease of water pH values [1]. Furthermore, NH_3-N in river mainly derived from point source, which provides a relatively constant input of constituent to the river throughout the year [14], NH_3-N concentrations will decrease reciprocally with increasing flow rate in summer, due to dilution effect[18].

Spatial distribution of water quality

Results from spatial two-way ANOVA and K-W test displayed significant spatial differences (P<0.05) for all water quality parameters at the eight water quality stations. Spatial variations of eight variables were illustrated by box-whiskers plots (Figure 3).

The lowest water temperature was found at station M1, which located in the mountain area, and other stations (M2-M8) showed less variation (Figure 3). The DO was lower in middle reaches (station M2 to M4) than in lower reaches (station M5 to M7) of the Min Jiang (Figure 3). On the contrary, the COD_{mn}, NH_3-N and TP concentrations were higher in the middle reaches (station M2 to M4) of the Min Jiang, and all of them were lower in tributary (station M8) than in the main stream of the Min Jiang, except NH_3-N.

High values of DO and lower values of COD_{mn} and nutrients (NH_3-N and TP) were found in station M1 indicated that the water from upper reaches of the Min Jiang, relatively unaffected by human activities, was good of quality. However, the values of COD_{mn} and nutrients (NH_3-N and TP) followed the sharply increasing trends from station M2 to M4 (Figure 3), where high population density and main industrial were located. Chengdu city is one of the major cities in this area. The population of Chengdu city account for 73% of

Stations	Locations	Section character	Water quality target
M1	Middle reaches	Aba prefecture boundary section	I
M2	Middle reaches	Chengdu boundary section	
M3	Middle reaches	Meishan boundary section	III
M4	Middle reaches	central section of Leshan city	III
M5	Lower reaches	Leshan boundary section	III
M6	Lower reaches	Cuiping district of YinBin city	III
M7	Lower reaches	Near entry to Yangze River at YinBin city	III
M8	Tributary	central section of Leshan city	III

Table 1: Water quality monitoring stations in the middle and lower reaches of the Min Jiang and its tributary.

Parameters	Middle reaches								Environmental guidelines [a]				
	M1		M2		M3		M4						
	Mean	Std.	Mean	Std.	Mean	Std.	Mean	Std.	I	II	III	IV	V
DO	8.62	1.14	6.55	1.39	4.5	1.49	5	1.7	≥7.5	6	5	3	2
COD_{mn}	1.51	0.72	3.27	1.58	6.94	5.15	5.37	2.09	≤2	4	6	10	15
NH_3-N	0.26	0.19	0.75	0.48	0.98	1.03	2.17	1.93	≤0.15	0.5	1	1.5	2
TP	0.17	0.19	0.25	0.18	0.27	0.14	0.28	0.19	≤0.02	0.1	0.2	0.3	0.4
Parameters	Lower reaches						Tributary		Environmental guidelines [a]				
	M5		M6		M7		M8						
	Mean	Std.	Mean	Std.	Mean	Std.	Mean	Std.	I	II	III	IV	V
DO	8.17	1.12	8.38	1.02	8.48	1.06	8.79	1.12	≥7.5	6	5	3	2
COD_{mn}	3.06	1.13	3.79	1.98	3.55	1.58	2.41	0.92	≤2	4	6	10	15
NH_3-N	0.41	0.2	0.11	0.09	0.16	0.12	0.3	0.16	≤0.15	0.5	1	1.5	2
TP	0.12	0.04	0.14	0.06	0.15	0.06	0.07	0.04	≤0.02	0.1	0.2	0.3	0.4

Table 2: Statistics descriptive of selected water quality parameters in the middle and lower reaches of the Min Jiang during 2003-2012.
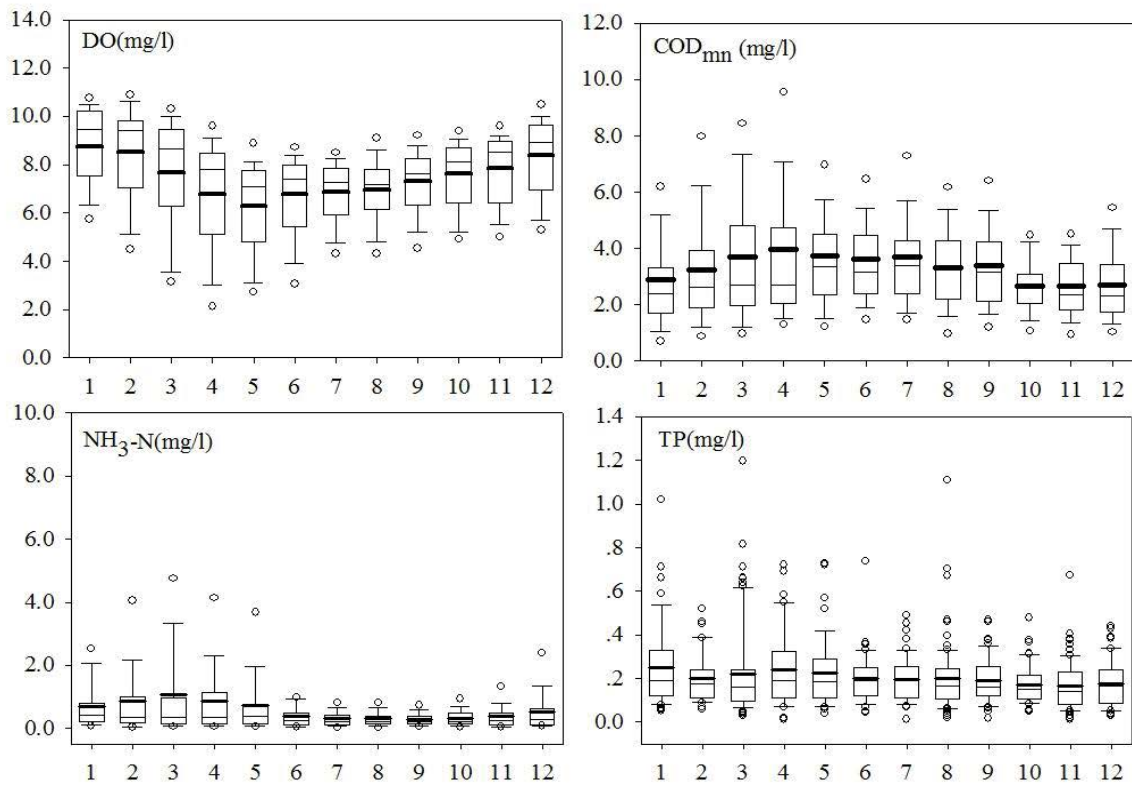
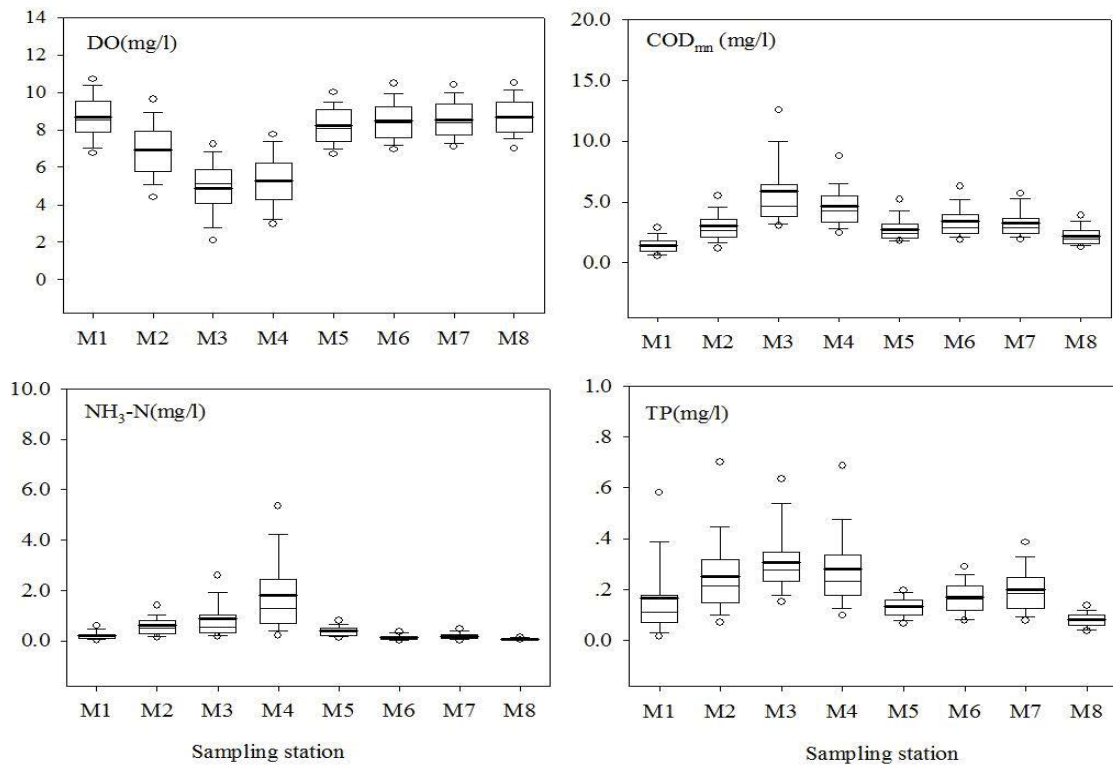**Figure 2:** Seasonal variations of eight variables in the middle and lower reaches of the Min Jiang.

Figure 3: Spatial variations of eight variables in the middle and lower reaches of the Min Jiang.

the Min Jiang Basin, gross industrial output value account for 75%, and pollutant emission account for 73.8%. In addition, Meishan and Leshan city, where stations M3 and M4 were located respectively, have been experiencing a rapid economic development, but the lack of the treatment plants for the produced wastes and inefficient management results in direct contamination of local surface water systems. Less populated water was found in the lower reaches of the Min Jiang and its tributary due to lower anthropogenic activities and the better hydraulic conditions to dilute pollution. Furthermore, differences of surface water quality between the middle reaches and lower reaches due to the influence of tributary (Dadu River) .

Concentration-flow relationships

The relationships between pollutant (COD_{mn}, NH3-N and TP) concentration and river volumetric flow for the stations (M4, M5, M7 and M8), and their associated model solutions, were shown in Fig4. The parameter values produced from the modelling were given in Table 3.

For COD_{mn}, in the lower reaches of the Min Jiang (stations M5 and M7) and its tributary (station M8), COD_{mn} concentration showed an increasing relationship with river flow increasing (Figure 4). This feature implies diffuse source provides the major source controls on river COD_{mn} concentrations. The negative relationship between COD_{mn} and flow for station M4 implies that point source is the dominant input to the middle reaches of the Min Jiang.

The negative relationship between NH_3-N concentration and river flow was observed at all stations, except at station M8 (Figure 4). The power model describes better relationships between the NH_3-N concentration and river flow for stations M4, M7 and M5, with the correlation coefficient values are 0.67, 0.34 and 0.32, respectively (Table 3) No correlation was found between NH_3-N and river flow at station M8 (R^2=0.002). Regarding TP, the better correlation was found between TP and river flow at station M4 (0.461), following by stations M8 (R^2=0.44) and M7 (R^2=0.34), while no correlation was found at station M5 (R^2=0.02).

Station	COD_{mn}				NH_3-N				TP			
	Equation	a	b	R^2	Equation	a	b	R^2	Equation	a	b	R^2
M4	Power	17.52	-0.2497	0.34	Power	171.48	-0.8931	0.59	Power	0.7671	-0.178	0.08
M5	Power	0.7224	0.1911	0.10	Power	11.541	-0.4487	0.32	Linear	0.000003	0.1103	0.02
M7	Power	0.3098	0.3192	0.26	Power	14.337	-0.6304	0.34	Power	0.0115	0.3264	0.34
M8	Power	0.3818	0.2565	0.28	Power	0.3171	-0.0275	0.002	Linear	0.00002	0.0386	0.44

Table 3: Concentration-flow relationships for four stations.
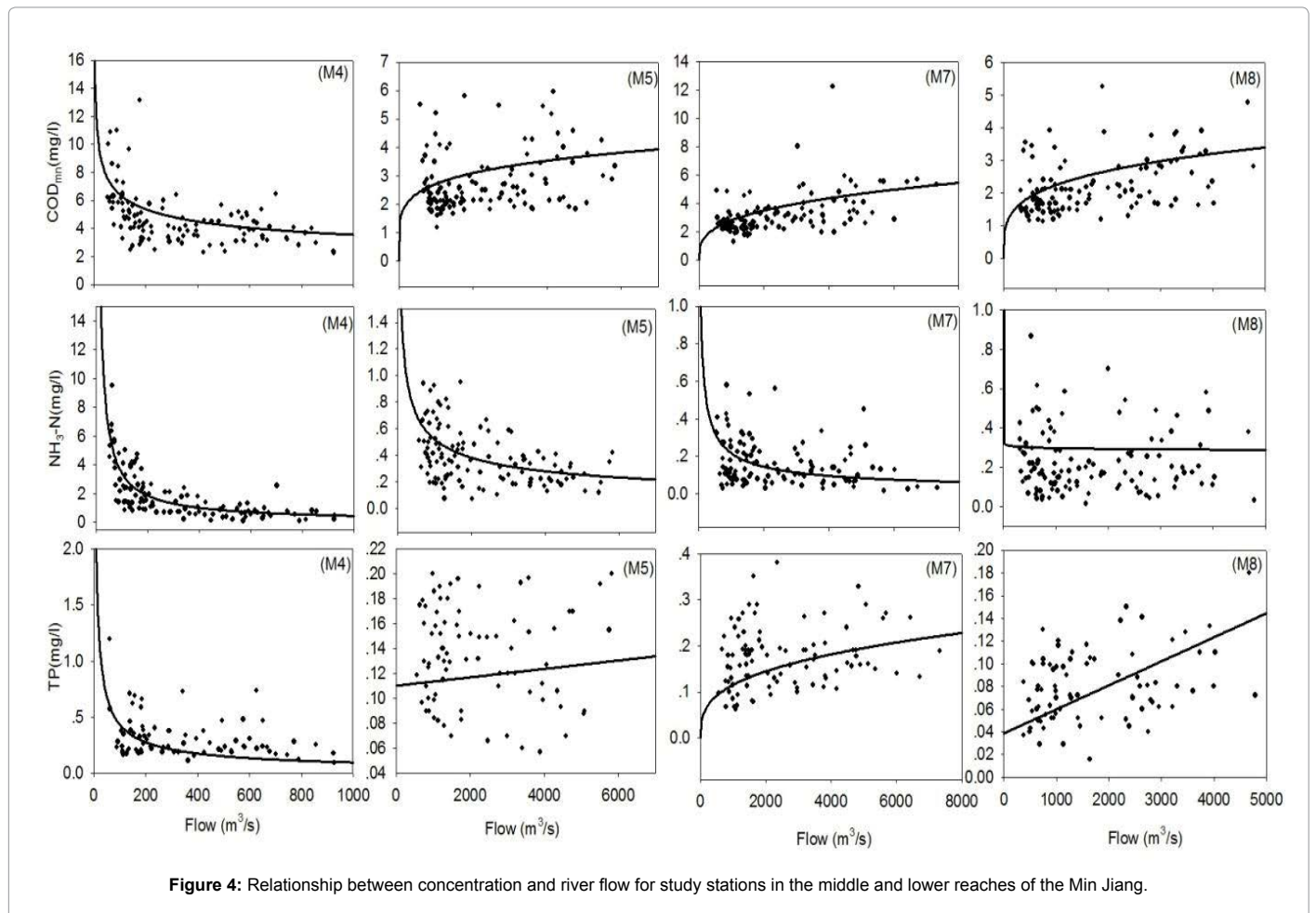

Figure 4: Relationship between concentration and river flow for study stations in the middle and lower reaches of the Min Jiang.

All water quality monitoring stations (M4, M5 and M7) in the main stream of the Min Jiang exhibited a decrease NH_3-N concentration with increasing river flow. This feature implies point source provides the major source controls on river NH_3-N concentrations. In addition, station M4 showed a dilution TP relationship with increasing river flow, indicating contributions from agricultural diffuse sources are minimal and point sources are the overwhelmingly dominant source of TP to rivers [5,19,20]. For stations M7 and M8, TP concentration showed an increasing with river flow increasing, indicating a lack of any significant point source contribution and a dominance of diffuse TP input.

Result from analyses of the concentration-flow relationship showed the organics (COD_{mn}) and nutrients (NH_3-N and TP) pollution in the middle reaches of Min Jiang dominated the point sources input, which attributed to these regions are predominantly domestic and industry activities, and have many wastewater treatment plants (WWTPs) input [21,22]. Furthermore, In the lower reaches of the Min Jiang (stations M5 and M7) and its tributary (station M8), a lack of any significant point source contribution or a dominance of diffuse pollutant input, which attributed to these regions are predominantly agriculture activities and. have no WWTPs input.

Source apportionment of pollution loads

Applying the load estimation model, the loads of COD_{mn}、 NH_3-N and TP entering the Min Jiang, and the percentages of point source and diffuse sources loads to the total loads are shown in Table 4. Table 4 showed that the total diffuse source loads of both COD_{mn} and NH_3-N were less than point source loads in the study basin, and TP loads of point source were nearly equal to diffuse source loads.

The percentages of point source and diffuse sources loads to the total loads at each monitoring station are presented in Figures 5-7, respectively. Diffuse source loads were higher than point source loads at 50% monitoring stations. The point source loads of NH_3-N in M4 and M5 were greater than diffuse source loads, respectively. However, there were some difference between the main stream and tributaries. COD_{mn} loads of diffuse source were greater than those of point source loads M7 did not.

Management Options for Water Quality Improvement

Through the analyses of the spatial-temporal variations of water quality parameters and concentration-flow relationships, a number of control measures can be recommended to mitigate the pollution condition in the middle and lower reaches of the Min Jiang. In summer or high flow period, organics pollution should be as the main controlled target; in winter or low flow period, NH_3-N pollution should be as the main controlled target; TP should be as the main controlled target for the whole year. In the middle reaches of the Min Jiang, organic matters and nutrients pollution from point sources need to be reduced; Nevertheless, in the lower reaches, COD_{mn} and TP pollution control need to focus diffuse source input, while NH_3-N pollution control should target on reducing point source inputs. In tributary, controlling diffuse source inputs is the key to solve the pollution problem caused by COD_{mn} and TP, whilst NH_3-N pollution control needs to be carried out to reduce both point and diffuse source inputs.

Regarding to the quantification of point and diffuse source loads entering to the main stream and tributaries, point source, especially point source NH_3-N is the key pollution source in the middle reaches. It is necessary to enhance the treatment level of industrial effluent, to strictly carry out the discharge standard for water pollutants and the total amount control system, to incorporate NH_3-N in the total amount control system.

Together with the impact of some industrial effluents, agricultural activities and urban sewage caused a clear impact on water quality of the Min Jiang. The proportion of diffuse source pollution to total loads is generally rising, and will become the major source of the pollution loads to the river. Agricultural diffuse source pollution, such as erosion of cropland and the unreasonable application of agrochemicals to cropland, should be controlled and diminished firstly by land use planning and best management practices. Maintaining the natural geomorphologic features, especially the meandering pattern of the river is also compulsory for the good ecological condition of the river, and it is a key factor in preserving the self cleansing capacity of the river [23].

Source	COD_{mn} (t/yr)	Percentage (%)	NH_3-N (t/yr)	Percentage (%)	TP(t/yr)	Percentage (%)
Diffuse	6119	70	749	78	317	49
Point	2640	30	211	22	334	51
Total	8759	100	961	100	651	100

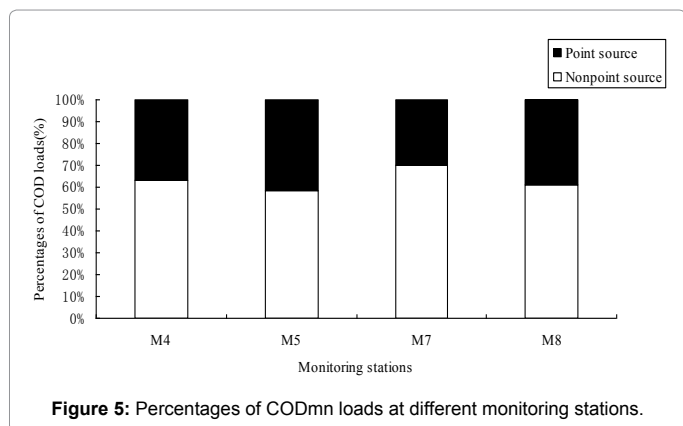**Table 4:** Loads of COD NH3-N and TP entering study area.

Figure 5: Percentages of CODmn loads at different monitoring stations.

Figure 6: Percentages of NH3-N loads at different monitoring stations.
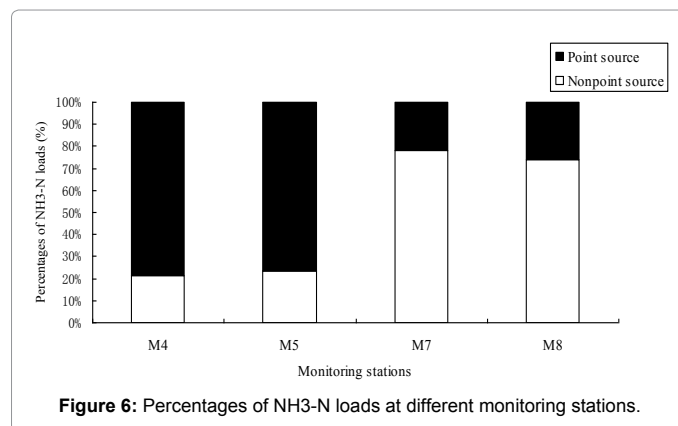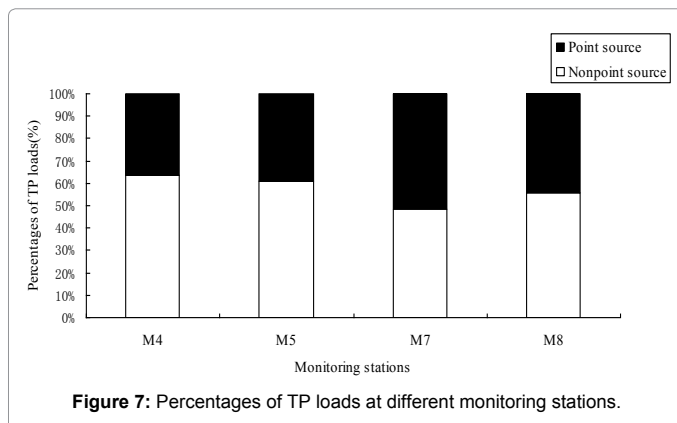
Figure 7: Percentages of TP loads at different monitoring stations.

Conclusions

This paper investigates the spatial-temporal variations and pollution source of surface water quality by analyzing the data from 8 water quality monitoring stations along the middle and lower reaches of the Min Jiang between 2003 and 2011. Seasonal and spatial differences were found for DO, COD_{mn} and NH_3-N, whereas for TP the difference was mainly due to the water quality station. The level of organics (COD_{mn}) was higher in summer (high flow period), and the level of nutrients (NH_3-N) was higher in winter (low flow period). Surface water was highly polluted in the middle reaches of the Min Jiang than that in the lower reaches and tributary. In the middle reaches of the Min Jiang, contributions from point sources (from WWTPs and industrial effluents) were the dominant inputs of organics (COD_{mn}) and nutrients (NH_3-N and TP) to river. In the lower reaches of the Min Jiang, diffuse sources were the dominant contributor of organics and TP to river; point sources were the dominant input of NH_3-N. In tributary, diffuse sources were the dominant organics and TP input, both point and diffuse sources were dominant NH_3-N input. Overall, pollution control by periods and regions is important for effective water management in the Min Jiang. It is necessary to enhance the treatment level of industrial effluent, to strictly carry out the discharge standard for pollutants and the total amount control system, to incorporate NH_3-N in the total amount control system.

Acknowledgements

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