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by Shinta Indah

Spatial variability of ammonium, nitrite and nitrate concentrations in water of Batang Arau River, West Sumatera, Indonesia

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ABSTRACT

The objective of this study is to analyze the spatial variability of ammonia, nitrite, and nitrate concentrations along the Batang Arau River, one of the most important rivers in West Sumatera, Indonesia. The results showed that the ammonium, nitrite, and nitrate concentrations were in the range of 0.180–0.510 mg/L, 0.0–0.178 mg/L, and 0.675–1.165 mg/L, respectively. The ammonium and nitrate concentrations were still below the maximum permissible concentrations for the river water quality standard of class II established by the Regulation Governor of West Sumatera at all of the sampling locations, except at midstream for ammonium, while the nitrite concentration had exceeded the quality standard from midstream to downstream of the river. The increase of the nitrogen concentration was observed from upstream to downstream of the river. Spatial analysis shows significant differences in nitrogen concentrations were obtained between the upstream and the midstream of the Batang Arau River ($p < 0.05$) but were not observed between the midstream and the downstream. This indicates the effect of different anthropogenic activities along the river. Based on the obtained results, for an effective monitoring program of the nitrogen concentration along the Batang Arau River, reducing the sampling locations from eight to five could be implemented.

Key words | ammonium, nitrate, nitrite, river, spatial analysis

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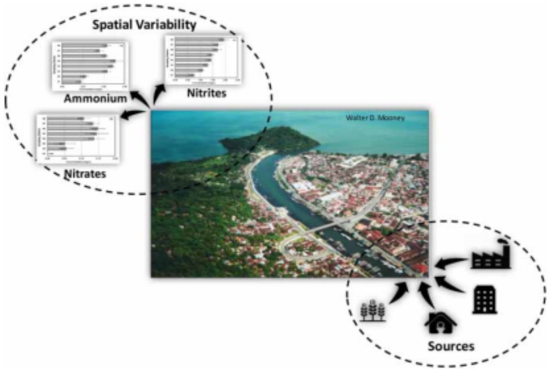
HIGHLIGHTS

- Spatial analysis contributes in evaluation of nutrient pollution in the river.
- Different anthropogenic activities cause the significant spatial variability.
- Result of spatial analysis helps in designing an effective monitoring program.

GRAPHICAL ABSTRACT



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INTRODUCTION

Nitrogen, in the forms of ammonium, nitrite, or nitrate, is a nutrient needed for plant animal growth and nourishment. Many studies indicate that the overland flow of domestic sewage and the direct discharge of untreated industrial sewage are the primary sources of the nitrogenous species pollution in residential and industrial areas, particularly in heavily polluted areas. The application of chemical fertilizers is the primary source of nitrogen species in highly agricultural areas (Ma *et al.* 2015; Mekonnen & Hoekstra 2015; Amin *et al.* 2017; Bu *et al.* 2017). Besides, many factors affect the nitrogenous species concentration, such as the hydrogeological conditions, the topography of the study area, and regional precipitation, particularly in regions that are severely influenced by anthropogenic activities, as well as the distribution of sampling sites. A high load of nitrogen can cause eutrophication of water bodies, which disrupts ecology and causes toxic algal blooms, loss of oxygen, and loss of biodiversity (Tian *et al.* 2007; Lassaletta *et al.* 2009; Mian *et al.* 2010). Eutrophication can also seriously affect our ability to use water for drinking, industry, agriculture, recreation, and other purposes (Zhiwei *et al.* 2014).

As one of the most important rivers in West Sumatera, Indonesia, this river is used by the local communities as a source of fresh water supply for their daily activities like bathing, laundry, recreation, irrigation, and, most important of all, as a source of drinking water. However, the river receives wastewater from different land and water uses such as industrial, agricultural, commercial and domestic, which has generated intense pollution in the river (Hong *et al.* 2012; Indah *et al.* 2018; Helard *et al.* 2019a, 2019b). Previous studies reported that the concentration of nitrogen including ammonium, nitrite, and nitrate was in the range of 0.1 mg/L – 0.26 mg/L, 0.05 mg/L – 0.1 mg/L and 0.6 mg/L – 2.2 mg/L, respectively, from the upstream to the downstream of the river (Bapedalda 2013). Those concentrations, except nitrite at the downstream, had not exceeded the quality standard for river water (class II) based on the Governor Regulation of West Sumatera No. 5/2008 (0.5 mg/L, 0.06 mg/L and 10 mg/L for ammonium, nitrite, and nitrate, respectively). Nevertheless, monitoring of the nitrogen concentration in the river is still necessary to protect freshwater resources from nutrient pollution. So far, a monitoring program and some researches into nutrient pollution in the Batang Arau River are limited in reporting the concentration of nutrients, without any information about their spatial variabilities.

Spatial variation in nitrogenous species has been studied in some catchment areas, and the results indicated that the spatial distribution variability of nitrogenous species is complex (Goss *et al.* 1998; Babiker *et al.* 2004; Mouri *et al.* 2011; Liu *et al.* 2013). For watershed management, the study of the spatial distribution of nitrogenous species in watersheds can be applied in improving the ecological environment of the watershed and reducing the loss of nitrogen in the soil. Getting a better understanding of how the sources of risk, the receptors, and the exposure pathways are distributed in space, spatial analysis of pollutants is essential to be conducted. Besides, a spatial analysis should be performed within a river basin to examine the effects of various anthropogenic activities or land-use types on water quality and to design strategic sampling locations in management of water resources for an effective monitoring program (Delgado *et al.* 2010; Varol *et al.* 2012; Indah *et al.* 2018; Helard *et al.* 2019a, 2019b).

The objective of this study is to describe the spatial variability of nitrogenous species, including ammonium, nitrite, and nitrate, for the identification and evaluation of nutrient pollution in the Batang Arau River. This study was expected to give a better interpretation of nutrient pollution and contribute to promoting water management and conservation strategies, as well as the design of an effective future spatial monitoring network in the Batang Arau River. Eventually, this study would help to provide a reference for insight regarding water management and the safeguarding of water ecology in the river systems.

METHODS

Study area and sample collection

The study area is at the Batang Arau River, which is approximately 19,827 km in length from upstream to downstream and with a catchment area of approximately 172 km^2. At the upstream of the Batang Arau River, the residential human population is relatively rare, and there is a small portion of land used for agriculture (Liu *et al.* 2013). Nevertheless, from the midstream to downstream of the Batang Arau River, intense urbanization occurs that potentially causes water pollution.

Water samples were collected from eight stations along the Batang Arau River, which were divided into one baseline station (S1) and seven impact stations (S2, S3, S4, S5, S6, S7, and S8). The baseline station is located in the upstream of the river, referring to the natural and unpolluted state of the river. The impact stations are located next to the upstream, where some anthropogenic activities have existed. Although without the human activities, the pollutant concentrations can still be different from upstream to downstream, the seven impact stations (S2–S8) reflect the domestic, agricultural, industrial, and commercial activities along the river. Figure 1 displays the locations of sampling, and Table 1 presents a detailed description of sampling points. The grab method was used for five sampling trips at biweekly intervals conducted from March to May 2014. Temperature, pH, and dissolved oxygen (DO) were measured in the field. Water samples were collected in 1 L glass bottles and were placed in cooler boxes with ice at approximately 4°C prior to analysis.

Analytical methods

For pH and temperature, the in-situ measurements were carried out with the corresponding portable meters (Hanna, USA) while for DO a DO-meter (Lutron, Taiwan) was used. The nitrogen parameters were determined using spectrophotometer UV-VIS (Shimadzu UV-2600) and performed according to the standard method (SNI 06-2479-1991 for ammonium, SNI 06-6989.9-2004 for nitrite and SNI 06-2480-1991 for nitrate). Each sample was analyzed in triplicate, and average readings were automatically determined.

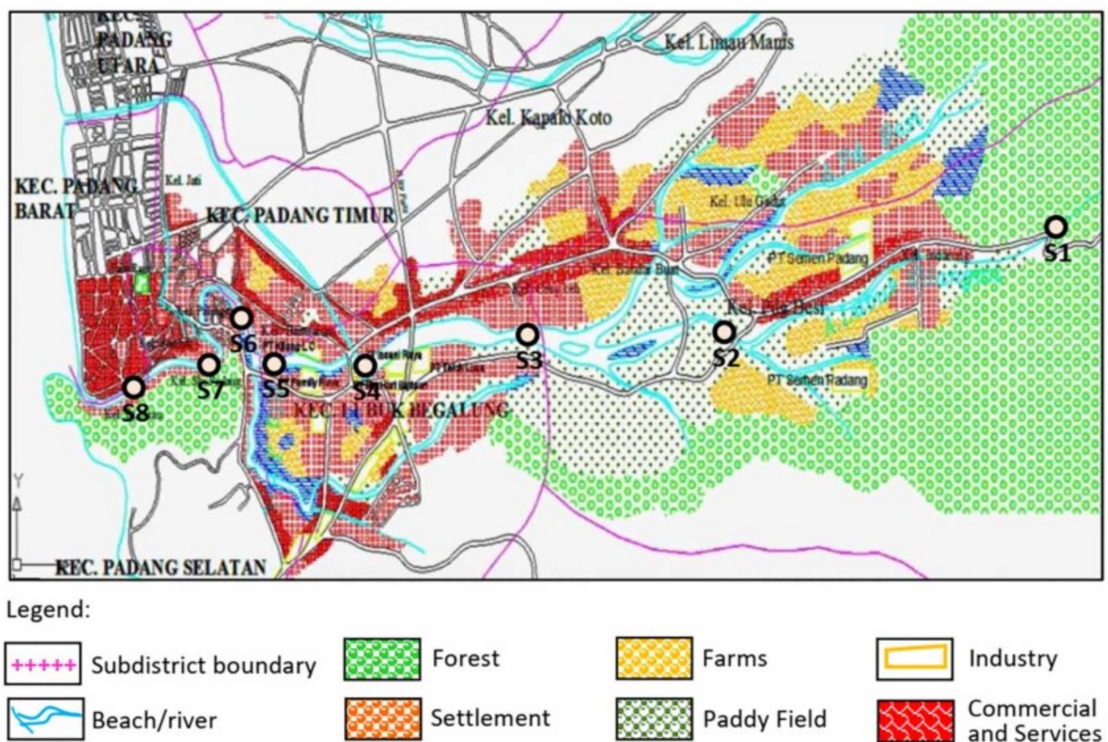

Figure 1 | A map presenting sampling locations on the Batang Arau River.

Table 1 | Description of eight sampling locations

Stations	Latitude	Longitude	Elevation m.a.s.l.[a] (m)	Distance (from S1) (km)	Description
S1	0°56′49.9″	100°30′31.5″	229	0	Located in a forested area that is upstream of the Batang Arau River
S2	0°57′30.4″	100°27′08.0″	124	4.2	Located on a drain that is bringing wastewater out from agricultural activities and a limestone mill
S3	0°57′39.7″	100°25′29.7″	72	10.1	This location has received wastewater from domestic and commercial activities.
S4	0°57′40.8″	100°24′02.3″	18	13.8	The river has passed through agricultural and industrial areas.
S5	0°57′43.3″	100°22′54.1″	7	16.7	The streams have received wastewater from the rubber industry
S6	0°57′26.8″	100°22′41.1″	6	17.6	The stream has received wastewater from domestic and commercial activities
S7	0°57′41.4″	100°22′28.4″	3	18.9	The stream has received wastewater from domestic and commercial activities
S8	0°57′44.8″	100°21′51.5″	1	19.9	Downstream of the river

[a]m a.s.l.: meters above sea level.

Statistical analysis

Data are presented in tables and graphs after analyzing using Microsoft Excel 2013. One-way analysis of variance (ANOVA) was performed to determine the significant spatial variability of nitrogen concentrations. SPSS version 20.0 was employed for statistical analysis.

RESULTS AND DISCUSSION

Overall means, standard deviations, and minimum and maximum values of environmental parameters and nitrogen concentrations, including ammonium, nitrite, and nitrate in the Batang Arau River at the eight stations, are recapitulated in Table 2. A decrease of DO and pH values at eight stations was observed from upstream to downstream of the Batang Arau River, while the temperature increased. The presence of several effluents entering the Batang Arau River and the different conditions of land use from upstream to downstream of the Batang Arau River may be the cause of the finding. Nevertheless, the values of the environmental parameters had not exceeded the quality standard for river water class II based on the Governor Regulation of West Sumatera No. 5/2008.

The spatial variability of nitrogenous species is illustrated in Figure 2. The concentrations of nitrogen including ammonium, nitrite and nitrate were in the range of 0.182–0.510 mg/L, 0.0–0.148 mg/L, and 0.739–1.942 mg/L, respectively. The concentration of ammonia had not exceeded the quality standard for river water (class II) based on the Governor Regulation of West Sumatera, except at S5. For nitrite, the concentration had exceeded the quality standard from midstream to downstream (S4 to S8). However, the nitrate concentration had not exceeded the quality standard at all the stations. The range of concentrations of ammonium and nitrite was higher, while nitrate was lower than those of the previous study conducted in 2013, demonstrating the dynamic fluctuation of the nitrogenous species due to natural and anthropogenic activities along the river (Bapedalda 2013). A similar spatial distribution pattern with a similar increasing trend in the concentration of these three nitrogenous species was observed from upstream to downstream of the river. For example, the mean concentration of nitrate increased from 0.739 ± 0.065 mg/L at the upstream to 1,942 ± 0.201 mg/L at the downstream.

The upstream area, S1, is located in a forested area where relatively no human activities existed, reflecting a natural and unpolluted river condition. This location represents the natural background concentrations of nitrogen and can be the reference stream for the other stations. In general, at the upstream as unaffected environments, the concentration of pollutants in rivers is very low and mostly derived from the weathering of rock and soil

Table 2 | The means, standard deviations, minimum and maximum values of environmental parameters and concentrations of nitrogen at the eight sampling locations on the Batang Arau River ($n = 5$)

Parameters		Stations S1	S2	S3	S4	S5	S6	S7	S8	Water Quality standard[a]
DO (mg/L)	Mean	8.5	7.16	7.42	6.24	6.94	7.18	6.96	6.68	Min. 4
	Std dev	0.3	0.716	0.867	1.82	1.085	0.268	0.537	0.934	
	Min	8	6.4	6.1	3.3	5.2	6.9	6.6	5.5	
	Max	8.9	8.3	8.2	7.9	7.9	7.6	7.9	8	
pH	Mean	8.4	8.72	8.4	7.86	7.76	7.6	7.64	7.54	6–9
	Std dev	0.4	0.449	0.706	0.416	0.568	0.418	0.378	0.577	
	Min	8	8.1	7.8	74	7.1	7.2	7.2	6.9	
	Max	8.9	9.1	9.6	8.5	8.2	8.1	8.2	8.1	
Temperature (°C)	Mean	25.8	28.64	30.8	30.56	30.52	30.5	30.42	30.44	± 3
	Std dev	0.4	1,820	2,477	1,665	1.802	1.735	1.071	1.135	
	Min	25.2	26.4	28.1	29.3	29.2	29.6	29.7	29.5	
	Max	26.2	31.2	34.7	33.4	33.7	33.6	32.3	32.4	
Ammonium (mg/L)	Mean	0.182	0.225	0.433	0.487	0.510	0.423	0.360	0.431	0.5
	Std dev	0.002	0.026	0.037	0.016	0.074	0.025	0.032	0.035	
	Min	0.180	0.180	0.401	0.460	0.410	0.395	0.310	0.390	
	Max	0.185	0.245	0.490	0.499	0.575	0.445	0.390	0.484	
Nitrite (mg/L)	Mean	0	0.055	0.051	0.137	0.144	0.148	0.134	0.107	0.06
	Std dev	0	0.022	0.031	0.030	0.034	0.023	0.020	0.019	
	Min	0	0.039	0.014	0.096	0.108	0.117	0.109	0.088	
	Max	0	0.092	0.091	0.164	0.178	0.172	0.160	0.135	
Nitrate (mg/L)	Mean	0.739	1.092	1.291	1.420	1.456	1.694	1.714	1.942	10
	Std dev	0.065	0.048	0.061	0.030	0.099	0.171	0.187	0.201	
	Min	0.675	1.030	1.221	1.383	1.339	1.559	1.559	1.662	
	Max	0.815	1.162	1.368	1.456	1.560	1.883	1.927	2.165	

[a]Governor Regulation of West Sumatera for river water quality standard of class II.

(Delgado et al. 2010; Varol et al. 2012; Indah et al. 2018; Helard et al. 2019a). It was observed that the concentrations of ammonium, nitrite, and nitrate at station S1 are relatively lower than at the other stations. Moreover, increases in ammonium, nitrite, and nitrate concentrations were found at the next stations until the downstream of the river. As shown in Table 1, there are some anthropogenic sources along the Batang Arau River, such as domestic, agricultural, and industrial. About 4.2 km from the upstream (S1), there is Station 2 (S2) with paddy fields located nearby. This station receives wastewater from a limestone mill and agricultural activities. Station 3 (S3) is located about 5.9 km from S2, and receives wastewater from sand mining, commercial, and household activities. At Station 4 (S4), the river passes through agricultural and industrial areas (i.e. rubber and palm oil processing industries). Next to S4, there is Station 5 (S5), which is about 16.7 km from S1. There, the river still receives wastewater from the rubber industry. Station 6 (S6) is located about 1 km from S5, and gets wastewater from residential and commercial areas such as auto services and public markets. Like S6, Station 7 (S7), about 1.3 km from S6, also receives wastewater from domestic and commercial activities, including a hospital, a hotel, and some restaurants. The last station is Station 8 (S8), the downstream of the Batang Arau River near the estuary that merges with the ocean. At S8, all pollutants along the river, which come from agricultural and domestic as well as industrial wastewater, have accumulated to result in the highest concentration of ammonium, nitrite, and nitrates being observed at this station.

To verify significant spatial variability in nitrogenous species concentrations, a one-way ANOVA was performed. The comparison of p values of ammonium, nitrite, and nitrate concentrations at eight stations is shown in

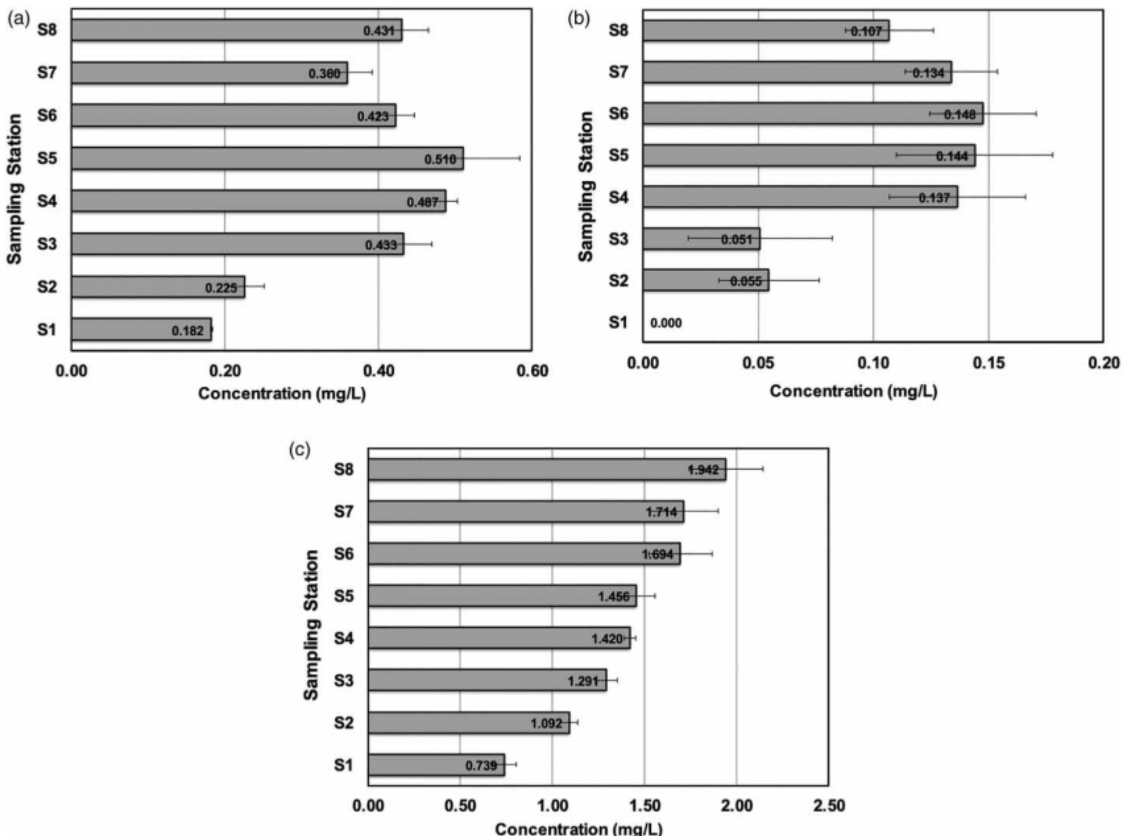

Figure 2 | Spatial variability of (a) ammonium, (b) nitrite, and (c) nitrate in Batang Arau River.

Tables 3–5. Table 3 shows that the ammonium concentration at S1 had no significant difference with S2, while at S3, S4, S5, S6, S7, and S8, significant differences were observed ($p < 0.05$). From S3 to S8, as mentioned above, the river receives wastewater from industrial, agricultural, commercial and domestic sources, which led to the significant variability in the spatial distribution of ammonium concentrations between the S1 and S2 station and the S3 to S8 stations along the river. Since at the S3 to S8 stations, the river receives almost similar sources of wastewater, no significant difference in ammonium concentrations was observed among those stations, except between S3 and S5, S4 and S7, as well as S5 with S6, S7, and S8.

For nitrite concentrations, as presented in Table 4, significant differences were found at S1, S2, and S3 with S4, S5, S6, S7, and S8 ($p > 0.05$), but not between S4, S5, S6, S7, and S8. These results denote that different anthropogenic activities in the upstream to midstream (S1, S2, and S3) compared to the midstream to the downstream (S4, S5, S6, S7, and S8) of the Batang Arau River may lead to a significant difference in the spatial distribution of nitrite concentrations in the river. However, at the midstream to the downstream of the river, there is no significant variability in the nitrite concentrations, indicating similar conditions at these stations. For nitrate concentrations, significant differences were observed at almost all stations except between S6, S7, and S8 stations, as displayed in Table 5. The S6, S7, and S8 stations are located near to the downstream of the river, where they received almost all of the wastewater from anthropogenic activities along the river. The similar conditions and activities at these

Table 3 | *p* values of comparison of ammonium concentrations at eight stations ($\alpha = 0.05$)

	S1	S2	S3	S4	S5	S6	S7	S8
S1								
S2	0.462							
S3	0.000*	0.000*						
S4	0.000*	0.000*	0.317					
S5	0.000*	0.000*	0.048*	0.979				
S6	0.000*	0.000*	1.000	0.173	0.021*			
S7	0.000*	0.000*	0.071	0.000*	0.000*	0.147		
S8	0.000*	0.000*	1.000	0.276	0.039*	1,000	0.086	

Table 4 | *p* values of comparison of nitrite concentrations at eight stations ($\alpha = 0.05$)

	S1	S2	S3	S4	S5	S6	S7	S8
S1								
S2	0.018*							
S3	0.047*	1.000						
S4	0.000*	0.000*	0.000*					
S5	0.000*	0.000*	0.000*	0.999				
S6	0.000*	0.000*	0.000*	0.998	1.000			
S7	0.000*	0.000*	0.000*	1.000	0.993	0.983		
S8	0.000*	0.035*	0.013*	0.525	0.240	0.190	0.690	

Table 5 | *p* values of comparison of nitrate concentrations at eight stations ($\alpha = 0.05$)

	S1	S2	S3	S4	S5	S6	S7	S8
S1								
S2	0.002*							
S3	0.000*	0.213						
S4	0.000*	0.004*	0.714					
S5	0.000*	0.001*	0.443	1.000				
S6	0.000*	0.000*	0.000*	0.031*	0.085			
S7	0.000*	0.000*	0.000*	0.015*	0.045	1.000		
S8	0.000*	0.000*	0.000*	0.000*	0.000*	0.057	0.105	

three stations may occur; therefore, the concentrations of nitrate at those stations also were not significantly different.

Analysis of the spatial variability of ammonium, nitrite and nitrate concentrations along the Batang Arau River reveals that wastewater from industrial, agricultural, commercial and domestic sources that is released into the river from S2 to S8 stations results in a significant difference in nitrogen concentrations to that in the S1 station at the upstream as the natural condition of the river. The ammonium in the wastewater discharged to the river from

S3 to S8, except S5 station, had a similar level of concentration; thus, significant differences were not observed among them. For nitrite, a similar concentration and no significant differences were also found from S4 to S8 stations. However, significant differences in nitrate concentrations were obtained at S1, S2, S3, S4, and S5, but not at S6 to S8, indicating the effect of different conditions in the anthropogenic activities at those stations.

As one of the objectives of spatial analysis, an effective future spatial monitoring program can be designed for evaluation of the nitrogen concentration along the Batang Arau River based on the obtained results. The sampling locations can be defined at the locations where there was a significant difference only from all monitored stations. So, for ammonium and nitrite concentrations, the eight stations can be reduced to three stations only to monitor the concentrations; that is, S1, S2, and S3. While for nitrate, five stations from the eight stations on the Batang Arau River can be chosen to observe the concentration along the river. Reducing the number of locations for sampling can reduce the time and money and, in turn, can be an effective monitoring program for the management of water resources.

Since the nitrogen concentrations have an increasing trend from the upstream to the downstream of the river, with potential to cause nutrient pollution, the monitoring program should be conducted by the local authorities as a part of the management and mitigation of water pollution at the Batang Arau River. Besides, since there is no centralized wastewater treatment in West Sumatera, the local authorities also should obligate the community to treat their wastewater through on-site wastewater treatment before discharging to the water body. Finally, these recommended actions will protect the Batang Arau River as a freshwater resource from water pollution.

CONCLUSIONS

Overall results indicate that the nitrogenous species, including ammonium, nitrite, and nitrate concentrations along the Batang Arau River, had a similar spatial variability pattern, with an increasing trend in concentration from upstream to downstream. This denotes that the variability of the nitrogen concentrations was affected by the natural and anthropogenic activities along the river. The concentrations of nitrogen including ammonium, nitrite and nitrate were in the range of 0.182–0.510 mg/L, 0.0–0.148 mg/L, and 0.739–1.942 mg/L, respectively. The concentrations of ammonium and nitrate at all of the sampling locations were still below the maximum permissible concentrations for the river water quality standard of class II established by the Regulation Governor of West Sumatera, except at S5 for ammonium. However, the nitrite concentration exceeded the quality standard from the midstream to downstream of the river. Significant spatial variability of nitrogen concentrations was obtained between the upstream and the midstream of the Batang Arau River but was not observed between the midstream and the downstream of the river. This reveals that wastewater from industrial, agricultural, commercial, and domestic sources released into the river from S2 to S8 stations resulted in a significant difference in ammonium, nitrite, and nitrate concentrations from that in S1 station, at the upstream, as the natural condition of the river. Based on the results, reducing the number of sampling locations to five stations could be performed for an effective monitoring program of nitrogen concentration along the river. The results of this study can be a reference for local authorities regarding water management and the safeguarding of water ecology in the Batang Arau River systems.

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