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Water quality assessment and pollution threat to safe water supply for three river basins in Malaysia

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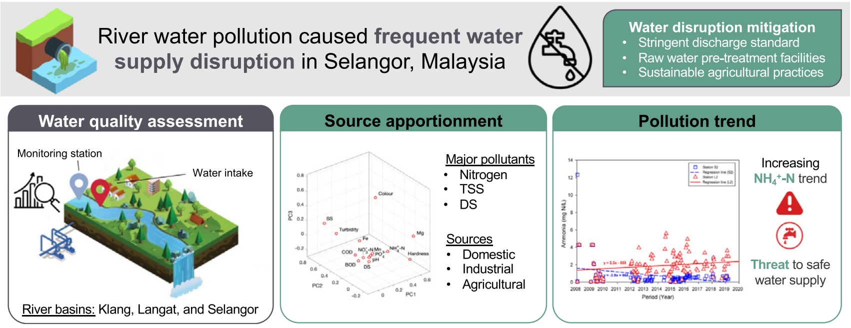
HIGHLIGHTS

- River pollution causes frequent water supply disruptions in Selangor, Malaysia.
- Chemometrics method can assess state-wide and multi-basins water quality.
- PCA/FA identified three factors dominating river quality near water intakes.
- M-K test showed increasing NH_4^+-N trend, threatening smooth water supply.
- Decision makers shall improve mitigation plans to reduce pollution threat.

GRAPHICAL ABSTRACT



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ABSTRACT

Pollution in raw water poses increasing threats to safe water supply in many developing countries. Therefore, a comprehensive water quality assessment is essential to provide various stakeholders the information to deal with this problem. This study applies chemometrics to interpret a recent 10-year water quality data from three major river basins (Selangor River basin, Langat River basin, and Klang River basin) frequented by water supply disruptions in Selangor, Malaysia. We present the application of selected chemometrics approaches, namely agglomerative hierarchical cluster analysis, principal component analysis, factor analysis and Man-Kendall trend analysis. The results showed three spatial groups of monitoring stations with similar land use practices and pollution characteristics. Besides spatial differences, periodic variations were observed when similar pollutants exhibited different pollution loads during rainy and dry periods. We found that nitrogen species, total suspended solids, and dissolved solids represented the major pollution loads in the studied basins. The results further confirmed a significant increasing trend in ammonia pollution. Our study demonstrates how ammonia pollutant is likely to pose a threat to water supply and highlights the vulnerability of Selangor's water resource system to water pollution. The results of this study could facilitate decision making towards more holistic strategies, specifically, incorporating ammonia treatment facilities into the conventional water treatment plant will help achieve smooth water supply operations.

1. Introduction

Surface water pollution is a severe global issue, particularly a pressing challenge in developing countries. United Nations Environment Programme (United Nations Environment Programme, 2019) estimated that serious pollution affected up to one-third of all rivers in developing

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countries. Major pollutants of concern include suspended solids, organic wastes, and nutrients (Mateo-Sagasta et al., 2017; Bashir et al., 2020). Eroded sediments due to stormwater runoffs, land clearing and sand mining activities contribute to high suspended solids. Wastewater discharge from domestic sewage and intensive livestock farms are the key attributes to high organic and nutrient pollutant loads (Malaj et al., 2014). A few impacts caused by the major pollutants include irreversible damage to the aquatic ecosystem, adverse health risk of consumers and decrease in potable water supply (Edokpayi et al., 2017). For example, algal blooms have plagued Changjiang River basin that contains one-third of China's water resources for over three decades (Tang et al., 2020). Yamuna River, which provides drinking water to nearly 20 million population in India, was covered with toxic dense foam caused by high ammonia concentration from industrial effluents (Hussain, 2019). Additionally, public health becomes a social issue. In Vietnam, the Ministry of Natural Resources and Environment reported that polluted water caused almost 80% of the diseases in the nation (Suwal, 2019). As the population increases, the water use will depend heavily on renewable surface water which is providing more than 50% of all drinking water worldwide currently (Smith et al., 2016). However, the river pollution threatens safe water supply due to deteriorating water quality (Chau et al., 2015). This water crisis impacts domestic stability and regional stability. Economically, water crisis means less productivity, deterring a country's credibility and development. Overall, the adverse impact is more pronounced on developing economies than developed economies due to ongoing development (Haddadin, 2001). Hence, Sustainable Development Goal 6 calls to improve ambient water quality by reducing pollution by 2030.

The water crisis is also a national concern in Malaysia, especially in the state of Selangor over the last decade. Selangor including the Federal Territories of Kuala Lumpur and Putrajaya is home to 8.32 million citizens, constituting 26% of Malaysia's population (Department of Statistics Malaysia, 2020a), with an estimated gross domestic production per capita of $ 43,625 (Department of Statistics Malaysia, 2020c). Selangor is the most populous and most developed state in Malaysia and almost 90% of its people live along the state's three main river basins - Selangor River basin, Klang River basin and Langat River basin. However, the three river basins are water stress areas, where data from Selangor Water Management Authority (SWMA) showed 13 cases of water treatment plants (WTPs) shutdown due to raw water quality violation in 2020. Ten out of thirteen cases caused water disruptions which had affected up to 1.14 million consumers (Air Selangor, 2020). Thus, effective water quality management in these river basins is crucial.

Regular river water quality monitoring is imperative for pollution prevention and control. It provides the data to allow a timely response in locating potential sources of pollution and identifies area where quality degradation exists. Once the contaminants exceed the standard limit, emergency strategy is implemented by releasing the water from the dams to flush the contaminants, minimizing water supply disruption. Subsequently, water quality assessment provides the basis for sustainable water resources management. The monitoring data can indicate changes in river water quality on a continuous basis. Hence, policymakers and government should heed the water quality deterioration to impose stricter environmental controls to safeguard and restore the rivers. For a comprehensive evaluation, researchers proposed comprehensive water quality index model, water quality assessment model, trend modelling and detection, pollution source identification and parameter optimisation (Juahir et al., 2011; Calazans et al., 2018; Dutta et al., 2018; Kadam et al., 2019; Camara et al., 2020).

Chemometrics approach proved to be efficient in addressing the challenge of complex environmental monitoring data interpretation (Juahir et al., 2011; Calazans et al., 2018). Chemometrics uses data treatment and multivariate statistical modelling to underline meaningful information and understanding of water quality data. The most common chemometrics methods used for clustering are the agglomerative hierarchical cluster analysis (HCA) and principal component analysis (PCA) with factor analysis (FA) (Kannel et al., 2007). They are often applied to verify spatial and temporal variations in surface water quality under natural and anthropogenic factors (Phung et al., 2015). Juahir et al. (2011) and Tavakol et al. (2017) optimised the number of monitoring locations by considering only representative stations for quick and practical analysis. Calazans et al. (2018) examined the competence of existing water quality monitoring network in Paraopeba River basin, Brazil and concluded that chemometrics was efficient and can be used in different watersheds. Besides spatial variation, Platikanov et al. (2019) proposed to monitor temporal variability of organic and inorganic water quality at drinking water intakes to detect river water pollution events. Many studies further concluded that chemometrics performed data reduction with minimal information loss (Simeonov et al., 2003; Kowalkowski et al., 2006; Tavakol et al., 2017).

Moreover, detection of pollution trends is another important step in water quality assessment. Non-parametric Mann-Kendall (M-K) test is widely used in assessing the significance of monotonic trends; it supports non-normally distributed data compared to parametric methods and has low sensitivity towards outliers and missing values (Camara et al., 2020). Pirnia et al. (2018) proved temporal changes in runoff trend due to dam construction to meet water supply demands for agriculture. Nyikadzino et al. (2020) verified that the M-K test could be applied to both annual and seasonal hydrometeorological data and was central to understanding enhanced disaster risk reduction. The trend result indicates criticalness of an environmental issue and helps to prompt adaptation measures.

A reliable water quality assessment allows policy makers to underpin sustainable water resources management. To date, many existing river water quality assessment studies have raised concerns regarding degrading water quality. Nevertheless, despite a recognizing need and a wide coverage of water quality data, state-wide assessment of water quality as a contributing factor to water disruptions in Selangor has not yet been explored. Little or no studies have investigated the probable water quality parameter that leads to water outages. Addressing this gap, this paper studied the Selangor's rivers water quality over the recent 10 years, by examining the spatial and dry-rainy periodic variations, using the multi-basin datasets from Department of Irrigation and Drainage (DID), Malaysia. In this work, we considered fourteen parameters (pH, colour, hardness, dissolved solid (DS), total suspended solid (TSS), turbidity, Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), ammoniacal nitrogen (NH_4^+-N), nitrate nitrogen (NO_3^--N), ortho-phosphate phosphorus (PO_4^{3-}-P), iron (Fe), manganese (Mn) and magnesium (Mg)) to provide a relatively complete information on the overall water quality compared to only six parameters (pH, COD, BOD, dissolved oxygen (DO), NH_4^+-N and TSS) used in Malaysia water quality index (DOE 2020). We then assessed the condition of water pollution, its threat to water disruptions and its trend based on the major pollutants of concern, which are ammoniacal nitrogen, nitrate nitrogen, biochemical oxygen demand, chemical oxygen demand, turbidity, and total suspended solids. These six parameters were often reported as the type of pollutants which caused WTPs shutdowns in Selangor (PNSB 2012; PNSB 2014; DOE 2020). The results of this study can be used to assist policymakers to understand, utilise the generated information and strategise towards mitigating water supply disruption problem caused by water pollution.

2. Methodology

2.1. Study areas

Selangor (8104 km^2) is located at the west coast of Peninsular Malaysia. It is the most developed state with the largest economy and population in Malaysia. In this study, we selected Klang River basin, Langat River basin, and Selangor River basin, because they are the source of potable water supplies in Selangor; the proportion of water supplies is 7%, 28% and 64%, respectively (Selangor Waters Management Authority, 2015c, 2015d). The coverage of the selected river basins in Selangor is shown in Fig. 1.

Klang River basin is the fourth largest river basin in Selangor with an area of 1288 km^2 and a length of 120 km. The estimated population of the Klang Valley is 4.4 million with 5% annual growth rate (Kandari et al., 2018). However, Klang River basin is not the main source of potable water to the population in Klang Valley, it only provides around 350 million litres per day (MLD) water supply (Selangor Waters Management Authority, 2015c). Langat River basin is the second largest river basin in Selangor, it is also a trans-state river basin with an area of 2423 km^2 and an approximate length of 200 km. The population of the Langat River basin is estimated at approximately 1.98 million as of 2020 (Selangor Waters Management Authority, 2015a). The river provides important water resources, its abstraction capacity is about 1100 MLD of potable water to support economic activities (Selangor Waters Management Authority, 2015d). Selangor River basin is the third largest river basin in Selangor, covering an area of 2200 km^2. It has an abstraction capacity of approximately 3000 MLD (Selangor Waters Management Authority, 2015e). The basin had a population of 451,000 approximately in 2010, the estimated population based on 1.4% average annual growth rate (2016–2019) is 526,000 by 2021 (Department of Statistics Malaysia, 2010, 2020b).

According to the Department of Meteorology Malaysia (MET), the climate in the study area is characterised by high rainfall, high humidity, and uniform temperature. Muhammad et al. (2020) reported minimal rainfall in June and July at Selangor according to 55 years of data supplied by MET. In this study, we collected monthly rainfall data (2009–2019) from 4 rainfall monitoring stations (RF3516025, RF2917112, RF3414032 and RF3118103) under DID, Ministry of Environment and Water Malaysia (see Supplementary Fig. S1). The average rainfall per month was 171 mm, the lowest mean rainfall was 103 mm in February while the highest mean rainfall was 254 mm in November. We considered a wet period when it has the rainfall volume above 150 mm for the month (Meteoblue, 2021). Hence, February – March and June – August are the dry periods. The details of the study area are reported as supplementary information.

2.2. Monitoring stations and data

The water quality monitoring data were obtained from the DID, Malaysia. Fig. 1 and Table S1 illustrate the location of the seven selected stations, covering the key sites that reasonably represent the water intake points. The selected data had a 10 years' time frame (2008–2019); stations S1, S2, L1, and L2 did not have data for 2010 and stations K1, K2, and K3 had missing data for 2013.

Fourteen water quality parameters used in this study included pH, colour (True Colour Unit, TCU), hardness (mg/L), DS (mg/L), TSS (mg/L), turbidity (Nephelometric Turbidity Unit, NTU), BOD (mg/L), COD (mg/L), NH_4^+-N (mg/L), NO_3^--N (mg/L), PO_4^{3-}-P (mg/L), Fe (mg/L), Mn (mg/L) and Mg (mg/L). DOE (2020) reported 99.2% compliance of toxic metals like Mercury, Arsenic, Cadmium, Chromium, Plumbum, and Zinc with Class II limits of National Water Quality Standard (NWQS) for Malaysia rivers. Hence, we considered Mn and Fe as the parameters according to the literature (Basheer et al., 2017b; Othman et al., 2020; Shahbudin and Kamal, 2021). All data used in this study were obtained from the DID database based on the continuity in measurement and the availability of recorded data. Additionally, the parameter selection criterion was based on its ability in limiting river water use for drinking water abstraction. The descriptive statistics of the 10-year dataset are summarized in Table 1.

2.3. Data treatment

The water quality data were prepared and processed using Microsoft Excel (version 16.45). For data pre-treatment, the constituent concentrations were log-transformed to account for the log normal distribution of

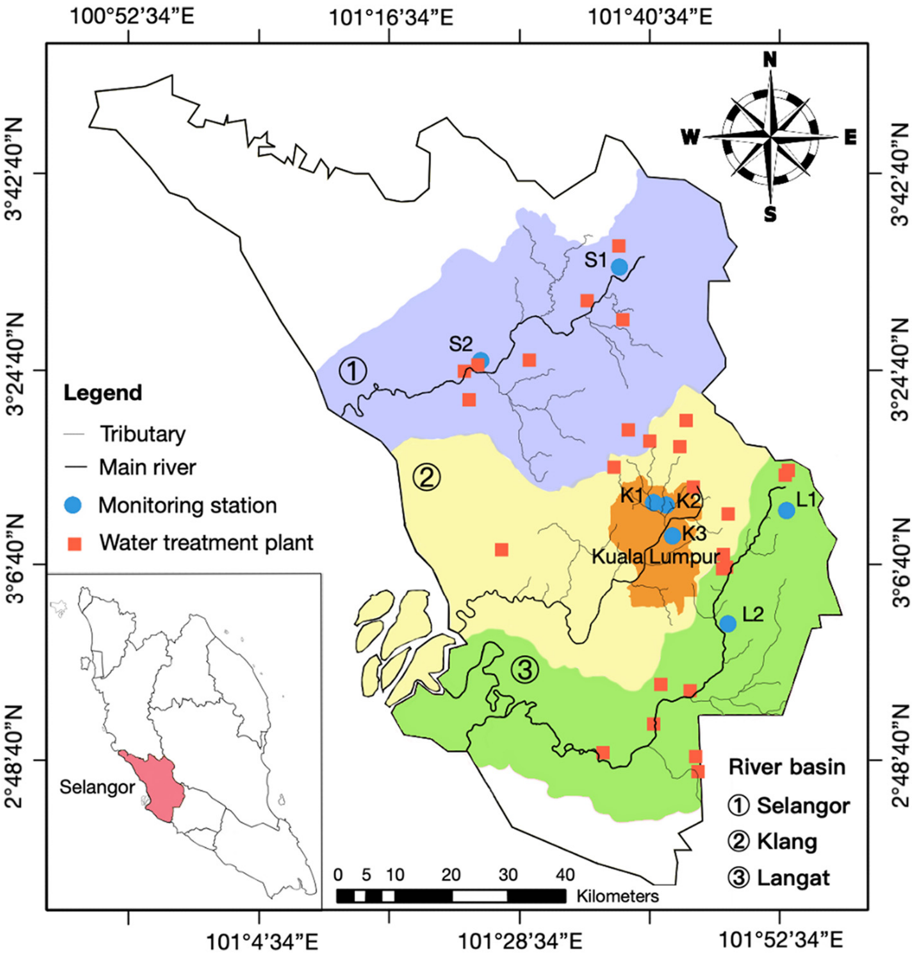

Fig. 1. Three river basins in Selangor and location of river water quality monitoring sites.

Table 1
Descriptive statistics of water quality constituents in Klang River, Selangor River and Langat River basins.

Parameter	Unit	Recommended raw water quality	Klang River basin			Selangor River basin			Langat River basin		
			Range (5th–95th)	Mean (SD)	Median (IQR)	Range (5th–95th)	Mean (SD)	Median (IQR)	Range (5th–95th)	Mean (SD)	Median (IQR)
pH		5.5–9.0	6.2–7.8 (6.5–7.6)	7.1 (0.4)	7.0 (0.7)	2.9–8.7 (6–7.2)	6.6 (0.5)	6.6 (0.6)	5.3–8.6 (6.2–7.2)	6.7 (0.4)	6.7 (0.5)
Colour	TCU	300	10–120 (10–60)	35 (18)	30 (30)	10–560 (20 – 200)	82 (94)	40 (70)	10–500 (20–225)	72 (93)	40 (40)
Hardness	mg/L	500	3–11 (4–8)	6 (1)	6 (2)	<1–8 (1–5)	4 (1)	1 (1)	< 1–69 (1–8)	4 (4)	2 (4)
DS	mg/L	1500	48–25,200 (84–18,000)	3255 (6530)	159 (100)	5–29,100 (5–3570)	745 (2582)	41 (61)	5–88,000 (7–8360)	1559 (7347)	55 (96)
TSS	mg/L	–	5–561 (11 – 122)	62 (60)	41 (51)	2–1085 (5–586)	133 (191)	62 (110)	4–1883 (8–564)	152 (184)	96 (128)
Turbidity	NTU	1000	2–132 (4–91)	31 (26)	22 (24)	2–1400 (3–409)	106 (172)	54 (94)	2–1400 (5–361)	96 (152)	50 (84)
BOD	mg/L	6	0–4 (<1–3)	<1 (1)	<1 (1)	0–<1 (<1)	<1 (<1)	<1 (0)	0–2 (<1)	< 1 (<1)	< 1 (0)
COD	mg/L	10	<1–62 (1–45)	14 (14)	8 (16)	<1–72 (6–44)	16 (12)	13 (12)	6–210 (6–50)	22 (16)	16 (24)
NH_4^+-N	mg/L	1.5	<0.1–9.9 (<0.1–7.6)	2.7 (2.6)	1.4 (3.7)	0–23.0 (<0.1–4.3)	0.8 (2.2)	0.2 (0.4)	< 0.1–18.9 (<0.1–3.8)	1.2 (1.8)	0.2 (1.7)
NO_3^--N	mg/L	10	<1–9 (<1–7)	2 (2)	<1 (2)	<1–38 (<1–2)	1 (3)	1 (1)	0–9 (<1–5)	1 (1)	<1 (2)
PO_4^{3-}-P	mg/L	–	<0.01–43.13 (0.10–5.56)	2.77 (4.10)	3.60 (3.95)	0.03–3.27 (0.03)	0.10 (0.41)	0.03 (0)	0.03–3.27 (0.03–0.09)	0.07 (0.30)	0.03 (0)
Fe	mg/L	1	0–8 (<1–7)	3 (2)	3 (4)	0–120 (<1–15)	5 (9)	3 (4)	0–60 (1 – 11)	4 (5)	2 (3)
Mn	mg/L	0.2	0–240 (0.5–190)	63.2 (73.1)	1.8 (116.9)	0.1–100 (0.4–2.5)	2.2 (10.7)	0.6 (0.7)	< 0.1–100 0.3–2.8)	2.1 (7.5)	0.7 (0.8)
Mg	mg/L	150	1–36 (9–28)	15 (6)	14 (7)	2–34,220 (4–19)	145 (1536)	9 (5)	0–8000 (5–22)	147 (390)	10 (4)

water quality data and to minimise the impact of outliers within the data (Migliaccio et al., 2004; Qian et al., 2007). Log-transformed data were standardised by scaling, so each parameter is considered equally important, using Eq. (1). The mean absolute deviation, s_f was used to minimise the effect of outliers, instead of standard deviation.

$$z_{i_f} = \frac{x_{i_f} - m_f}{s_f} \tag{1}$$

where m_f is the mean of log-transformed parameter f for all stations; s_f is the mean absolute deviation of log-transformed parameter f for all stations; x_{if} represents the original log-transformed parameter at station i; z_{if} is the standardised value of log-transformed parameter f at station i.

Lastly, the missing data and zeros were then replaced by the mean standardised log-transformed concentrations based on their respective parameters.

2.4. Hierarchical clustering analysis

We analysed long-term water quality data using Matlab (R2020b). We adopted HCA to identify distinct clusters within monitoring stations. We used the Euclidean dissimilarity measure and Ward's linkage algorithm which grouped seven monitoring stations based on spatial factor (Singh et al., 2005; Qian et al., 2007; Juahir et al., 2011; Egbueri, 2020). The clustering presents high homogeneity within a cluster and high heterogeneity among different clusters. Mean values of log-transformed constituent concentrations were used in cluster analysis. The dissimilarity (Euclidean distance) between two stations, $d\ (i,j)$, was calculated using Eq. (2):

$$d\ (i,j) = \sqrt{\sum_{f=1}^{r} \left(z_{i_f} - z_{j_f} \right)^2} \tag{2}$$

where z_{if} and z_{jf} are the standardised values of log-transformed parameter f at station i and j respectively; r is the number of parameters.

2.5. Principal component analysis/factor analysis

We also applied PCA/FA to provide information on the most significant parameters according to spatial and temporal variations. PCA is a powerful pattern recognition technique that describes complex data sets by excluding the less significant parameters with minimum information loss, simplifying large variables into meaningful linear combinations for comparison (Mohamed et al., 2015). The set of linear combinations, called the principal components (PCs) explain all the variance of the original data. The PCs (eigenvectors) are the product of original correlated variables with a list of coefficients (loadings). The higher the loading, the more significant the parameter. PCs with eigenvalue greater than 1 explain most of the variation, therefore the remaining PCs are not considered (Iscen et al., 2008; Arslan, 2013). For each clustering group, we applied PCA to both annual and periodic data sets (standardised log-transformed constituent concentrations). PCA based on the decomposition of real data matrices using singular value decomposition (SVD) expressed in Eq. (3) was used due to its robustness and stability.

$$A = USV^T \tag{3}$$

where A is an arbitrary Matrix $A\ (n \times p)$; $U\ (n \times n)$ and $V\ (p \times p)$ are orthogonal and normalised matrices respectively, U columns are the left singular vectors while V^T rows are the right singular vectors; $S\ (n \times p)$ is a diagonal matrix with singular values in decreasing order; n represents observation from river water samples; p represents parameters.

FA approaches data reduction through the measurement model of a latent variable. When PCs generated are not readily interpreted, new groups of variables known as varimax factors (VFs) can be obtained by applying varimax rotation. PCA/FA method helps to understand the correlation among the water quality variables (Qian et al., 2007; Juahir et al., 2011; Ravish et al., 2020). For each clustering group, varimax rotation was used to rotate the PCs with eigenvalue greater than 1. Constituents with loading ≥ 0.3 were considered as principal constituents. Prior to PCA/FA analyses, Kaiser-Meyer-Olkin (KMO) and Bartlett's sphericity test were performed to determine the suitability of the water quality datasets for PCA/FA.

2.6. Man-Kendall trend test

Parametric and non-parametric methods are two main categories in statistical methods for trend analysis. In parametric method, we assume that the data distribute normally, but most of the hydrometeorological data tend to be non-normally distributed (Heo et al., 2013). Thus, our study used non-parametric method on original data to evaluate the pollution trends across a decade at two monitoring station representatives, L2 and S2. We applied M-K test which is robust and widely applied for trend analysis in climatologic and hydrologic time series (Yadav et al., 2014; Ali et al., 2019; de Andrade Costa et al., 2020). For M-K test, the null hypothesis H_0 assumes that there is no trend in the series. H_0 was tested at 95% confidence level to study 6 physio-chemical water quality indicators of concern reported by Department of Environment (2019) and Department of Environment (2020) - NH_4^+-N and NO_3^--N represented nutrient factor; BOD and COD showed organic matter factor; turbidity and TSS referred to the sediment factor. Linear trend lines were then plotted for the selected parameters using Sigmaplot 11.0.

3. Result and discussion

3.1. River water quality

We analysed the river water quality data (2008–2019) from a total of 598 samples measured by DID, the 14 parameters are pH, colour, hardness, DS, TSS, turbidity, BOD, COD, NH_4^+-N, NO_3^--N, PO_4^{3-}-P, Fe, Mn, and Mg. The descriptive statistics includes range of minimum and maximum values, 5th percentile and 95th percentile values (5th – 95th), mean and standard deviation (SD), and median and interquartile range (IQR) of the water quality data as listed in Table 1. The variables were compared with the raw water quality standards specified by the Malaysian Ministry of Health (MOH) and the river water quality standards stipulated by the NWQS for Malaysia rivers (see Supplementary Table S2 and Table S3).

In a freshwater system, the term 'acidic' is reserved for freshwater with alkalinity less than zero, usually with pH less than 5 (Mattson, 1999). In Table 1, the pH values showed distinct minimum values among the 3 river basins, in which the lowest value of 2.9 was recorded once in Selangor River basin, close to the pH value (3.41) reported by Camara et al. (2020) within the same basin. A sudden shift in pH might be due to the accidental input of palm oil mill effluent high in acidity by several palm oil plantations and palm oil mills located in the basin (Baharudin et al., 2021). Acidic river water could contain elevated levels of metal ions such as iron, manganese, copper, lead, and zinc (Nowrouzi et al., 2014). The metal ions react with the piping material and eventually lead to premature damage such as leak or burst, increasing non-revenue water. Nevertheless, all the median pH shown in Table 1 was within the recommended range and fell under Class I according to NWQS. Class I water practically requires no treatment and is suitable for very sensitive aquatic species.

According to Table 1, the median Fe and Mn concentrations in all river basins significantly exceeded the recommended value, 1 mg/L and 0.2 mg/L respectively. Dissolved metal ions in natural water resources, like Fe and Mn, are secondary contaminants. They can alter the taste, odour, and colour of drinking water, and are usually immobilised in the form of particulate matter and co-precipitation as hydroxides and oxides. The contaminated water often contains iron and manganese bacteria, though the bacteria do not pose a health risk, they cause blackish or reddish deposits to build up in the water system. Based on NWQS, the water quality all fell into Class V, fortunately, the median value of colour of the 3 river basins were below 300 TCU and referred to Class II. In contrast, the presence of other dissolved metal ion, Mg that is highly associated with hardness, was found to comply with the standard in all river basins.

Based on Table 1, the median DS concentration in three river basins complied with the standard allowable level of raw water at 1500 mg/L. The DS result was classified as Class I determined by NWQS. However, the DS concentrations were highly skewed, where the highest concentration was 88,000 mg/L recorded at Langat River basin. Prior to our study period, Juahir et al. (2011) reported mean DS of 15,311 mg/L upstream of the Langat River during 1995 to 2002, which was 10 times greater than the recommended value. In this study, the recorded mean DS between 2008 and 2019 in Langat River basin was 1559 mg/L. DS parameter showed considerable quality improvement for over 25 years but extreme anthropogenic activities along the river course could have contributed to higher DS in the rivers.

The BOD concentrations of the three rivers were ranging between 0 and 4 mg/L. The median BOD was well below the recommended limit and within Class I of NWQS. The result was consistent with that reported by Al-Badaii et al. (2013) in the Langat River basin, ranging between 0.63 and 4.56 mg/L in 2012. Nevertheless, the BOD values reported by Camara et al. (2020) in Selangor River basin were between 1.0 and 36.0 mg/L from 2005 to 2015, which were considered high compared to this work. The median BOD (3.0 mg/L) and mean BOD (3.9 mg/L) fell under Class II and Class III of NWQS respectively, reflecting that the high BOD level at 36.0 mg/L could be due to pollutant discharge incidents from the point source pollution within the basin. The incidents might be intermittent and hence the data used in this study did not capture the anomalies. COD was relatively higher than BOD, showing a significant amount of complex organic matter present in the river. The median COD exceeded the suggested limit of 10 mg/L, except for the Klang River basin. Based on NWQS classification, Langat River basin and Selangor River basin were in Class II, where conventional treatment is required for water supply. High COD level points to serious water pollution in the study area.

Turbidity is highly dependent on seasonal variations as high stream flow and/or surface run-off could elevate the turbidity level in water (Yisa and Jimoh, 2010). According to Table 1, the median turbidity values were within the recommended raw water quality, but Selangor River basin and Langat River basin fell into Class III according to NWQS. In addition, both river basins recorded the highest turbidity (1400 NTU) which could be associated with the increased concentrations of suspended solids such as inorganic matter, plankton, silt, and microscopic organisms. Erosion of the riverbanks is the main source of TSS, especially during rainy days. There is no recommended raw water standard for TSS, but the classes in this study ranged between Class II and Class III. In the Langat River basin, the maximum TSS (1883 mg/L) was greater compared to the value (21.27 mg/L) reported by Abidin et al. (2018). Maximum TSS reported in Selangor River basin and Klang River basin were generally lower than those reported in other studies (Camara et al., 2020; Othman et al., 2020).

Natural raw water has low ammonia content around 0.1 mg/L, but Selangor River basin recorded the highest NH_4^+-N concentration at 23.0 mg/L followed by Langat River basin at 18.9 mg/L. High levels of ammonia in water can be from domestic wastewater and manure runoff. In Table 1, the 10-year median ammonia level in Klang River basin was 1.4 mg/L, classifying the basin into Class IV based on NWQS. The water in Class IV is only suitable for irrigation because it can be toxic to aquatic life when the concentration is greater than 0.9 mg/L. High ammonia level in raw water sources also poses a health concern when it enters the water treatment process train whereby it can enhance odorous chloramines formation and the formation of chlorinated disinfection by-products which are carcinogenic and mutagenic (Tian et al., 2013). Besides ammonia, high levels of nitrate can also cause death to aquatic life. Nitrate is associated with an increased risk of human health problem. Maximum value of NO_3^--N was recorded in Selangor River basin at 38 mg/L, nonetheless, the median NO_3^--N values in three river basins were within the maximum permissible limit in raw water set by MOH. Furthermore, PO_4^{3-}-P is often associated with nutrient pollution and eutrophication, but there is no regulation of its level in drinking water supplies. The median PO_4^{3-}-P in Selangor River basin and Langat River basin complied to the NWQS standard, hence they were categorised under Class I. The treatment of boiler water and the use of phosphate-based detergent and fertilizer could lead to high phosphate levels, as reflected in Klang River basin (Ismail, 2011). The median PO_4^{3-}-*P* value exceeded the normal level in the basin, thus the river was grouped into Class V.

Based on the compliance to NWQS, a few water quality parameters such as Fe, Mn, NH_4^+-N, and PO_4^{3-}-P rendered the rivers in Class IV and below.

The distribution of major parameters was high variance with great dispersion around the mean due to large standard deviation, except pH. It could reflect that the pollutant discharge incidents were having high variability.

3.2. Spatial variation among 7 stations

HCA was conducted to examine the correspondence between stations. All seven monitoring stations on three river basins were classified into three statistically significant groups, G1, G2, and G3, as presented in a dendrogram. In Fig. 2, G1 comprised stations K1, K2, and K3 while G2 consisted of stations L1 and S1, whereas stations L2 and S2 formed G3. The stations within the same group had similar constituent characteristics and therefore were likely to be affected by similar land use practices and pollutant sources.

All stations in G1 (K1, K2, and K3) are upstream of Klang River basin, they receive flow from respective tributaries, which eventually merge with Klang River within the Federal Territory of Kuala Lumpur. Therefore, we observed very similar water quality within G1. It is notable that G1 had the greatest Euclidean distance, indicating that G1 had a relatively different water quality and land use practice compared to G2 and G3. G1 is located at the most extensively urbanised area with high human activity which took up 75% of the land use in the basin; the other land use practices were agriculture land, forested areas and water bodies at 14%, 7% and 4% respectively (SWMA 2015c).

Langat River basin and Selangor River basin have similar land use and development history. In G2, stations L1 and S1 are situated at the upstream of the main rivers; station L1 is located at Hulu Langat while station S1 is within Hulu Selangor. The main land use practice of stations L1 and S1 was forest, at 46% and 54% respectively (SWMA 2015d; SWMA 2015e). The other land use practices of station L1 included urban (29%) and agriculture (25%). Station S1's land use practices were urban (10%), agriculture (33%), and wetland (3%). In addition, G3 had a shorter Euclidean distance due to high similarity in constituents corresponding to the pollution background. Stations L2 and S2 are situated at the midstream of the main rivers and the water quality is moderately affected by the land use and anthropogenic activities from both upper and middle reaches. Station L2 is located at the lower reach of Hulu Langat while station S2 is located at Kuala Selangor, where agriculture (48%) was the major land use, followed by forest (34%), urban (16%) and wetlands (3%) (SWMA 2015e). Camara et al. (2020) and Juahir et al. (2011) had classified G2 and G3 as low pollution source area and moderate pollution source area, respectively. The present study was consistent with their findings that the water quality at G2 was relatively good but water quality at G3 was only moderate.

The result shows that HCA provides information which could be useful to the authorities or decision makers to manage the selection of station location for monitoring optimisation, particularly in G1. For example, the study shows that monitoring from one station in G1 is sufficient to represent the water quality in the Klang River basin. By reducing the number of non-representative stations, the cost of maintenance and sampling could be minimised. This clustering technique also proves to offer reliable classification of surface water across different river basins state-wide.

3.3. Pollutant source apportionment

Prior to PCA, the KMO test and Bartlett's sphericity test were performed on the water quality datasets. The KMO value was 0.7557 (>0.6), and Bartlett's test value was 1134.83 ($p = 0.00 < 0.05$). The results implied that PCA/FA will be effective in reducing the dataset's dimensionality. The fourteen variables were reduced to a smaller dimension which contains important information for pollution source identification. PCs with eigenvalues larger than 1 were extracted: seven PCs each for G1, G2, and G3, summing approximately 89.2%, 90.4% and 85.9% of the total variance in the datasets, respectively. Varimax rotations were applied on the extracted PCs to assess the relative importance of water quality parameters in identifying water quality variations spatially. Liu et al. (2003) defined the absolute factor loading values as "strong", "moderate", and "weak" for values >0.75, >0.5–0.75, and 0.3–0.5, respectively. Table S5 presents the principal constituents (loading >0.5) involved in the construction of VFs. The VFs with moderate and strong loading values were considered significant due to large study area. The biplots presenting all loadings of VF1 to VF4 were shown in Fig. 3, Fig. 4, and Fig. 5 for G1, G2, and G3 respectively; close-up biplots were displayed in supplementary information (Fig. S2, Fig. S3, and Fig. S4).

For G1, among the seven VFs, VF1 presented 44.4% of the total variance. According to Fig. 3 (a), Mn and NO_3^--N showed moderate loadings.

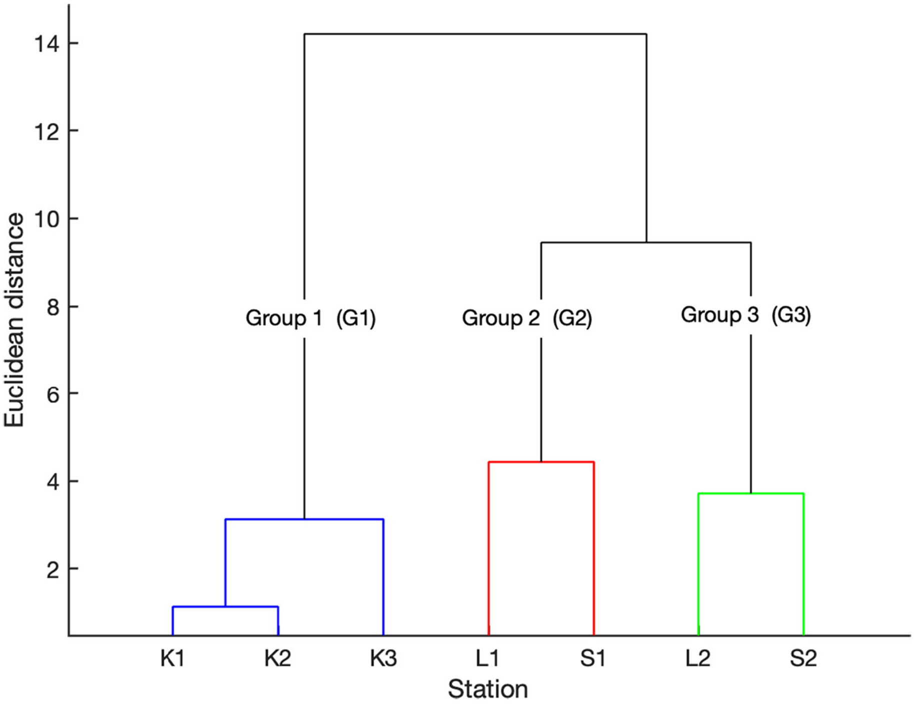

Fig. 2. Dendrogram showing clustering of river quality monitoring stations in Langat River (L), Selangor River (S) and Klang River (K) basins.

High level of Mn could be due to industrial effluent and earthworks along the river while high NO_3^--N implied nutrient pollutants of the domestic waste discharge (Samsudin et al., 2018; Rudi et al., 2020). The domestic waste discharge, which is known as sullage, has pollutants concentration that is lower than sewage, but its annual loading in rivers is higher than that of sewage at urban areas (Jaron, 2013). Hence, sullage could be the major pollution source to the rivers in G1 area due to high population density. VF2 in Fig. 3 (a) showed the sediment factor, where TSS exhibited strong positive loading. The plausible causes for high TSS are sand mining and illegal sand washing activities (Tan and Rohasliney, 2013). The illegal sand washing activities at the city centre rendered the Klang River polluted at Class III or Class IV and challenged the river rehabilitation operations under River of Life project (Bavani, 2016, 2019). The project aims to revitalise the Klang River by 2024 through three transformation programme, which are river cleaning, river beautification, and property development. Besides, cases of quarry sediment ponds overflowing during storm events resulted in shutdown of WTPs due to choking of the filters (Selangor Waters Management Authority, 2015c). VF3 and VF7 had strong positive loadings on COD and BOD respectively, reflecting the organic factor. VF4 (Fig. 3 (b)) had a strong positive loading on NH_4^+-N, thus VF1 and VF4 could represent nutrient factor. Untreated sewage or partially treated effluents and industrial discharge were the key attributes (Selangor Waters Management Authority, 2015c). Rapid population growth has resulted in increasing sewage production and deemed sewerage treatment less ineffective due to operational strain at sewerage treatment plants (Van Drecht et al., 2009; Zhang et al., 2015). Particularly, a report published by Selangor Waters Management Authority (2016) recorded the discharge of untreated sewage at Rumput River, upstream of the study area. Substantial nutrient, organic, and sediment loads were also recently reported within this region (Samsudin et al., 2018; Mohtar et al., 2019). Besides nutrient and organic factors, VF5 and VF6 had a moderate loading on Mg and a strong loading on DS respectively, which could be linked to VF1. Overall, stations in G1 were exposed to different point sources, especially municipal and industrial sewage (Mohamed et al., 2015).

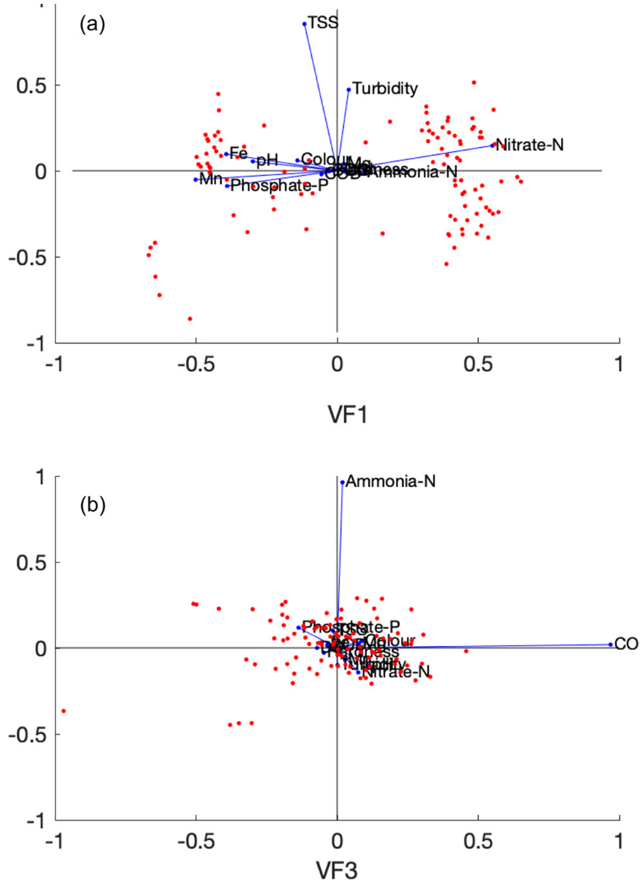

Fig. 3. Biplot for (a) VF1 and VF2 and (b) VF3 and VF4 showing loadings and normalised score of G1 stations.

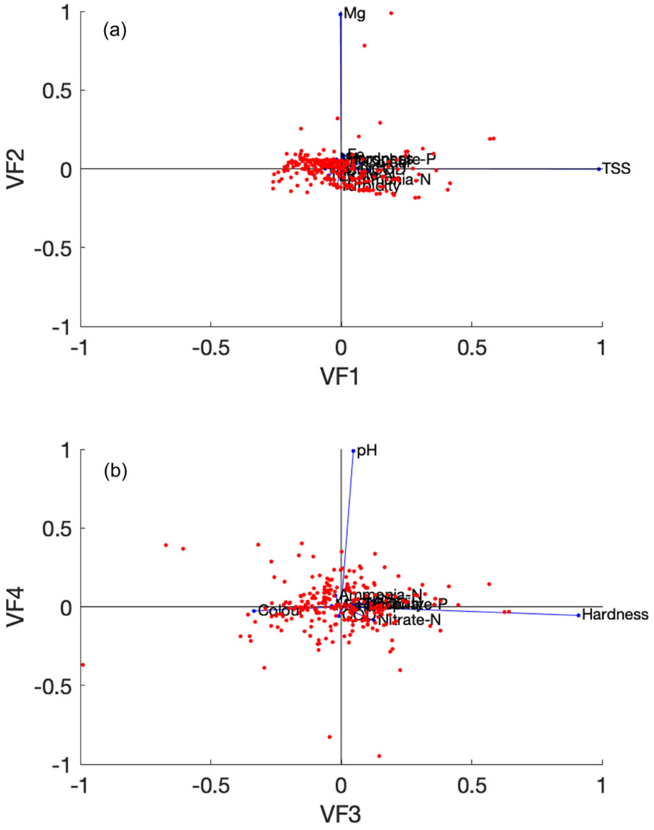

Fig. 4. Biplot for (a) VF1 and VF2 and (b) VF3 and VF4 showing loadings and normalised score of G2 stations.

In G2, VF1 (Fig. 4 (a)) and VF7 had strong positive loadings on TSS and turbidity respectively. Clearing of the forest because of urban developments caused increased surface erosion and runoff, hence increasing the turbidity within the G2 region. The composition of sediments, predominantly controlled by the lithology, had also caused high concentration of DS (VF5) in the rivers during the rainy days (Lim et al., 2012). VF2 explained 20.2% of the total variance and showed a strong positive loading on Mg based on Fig. 4. (a). Magnesium salts are found naturally in high concentrations in surface water. Additionally, VF3 in Fig. 4 (b) had a strong positive loading on hardness. VF3 and VF5 were highly intertwined and could be attributed to geological structure of the catchment area, weather conditions and other factors like intensity and type of water supply (Kuriata-Potasznik and Szymczyk, 2015). Besides, suspended solid acts as nutrient carriers in rivers, when present for biological uptake, it could accelerate eutrophication of streams (Sorensen et al., 1977; Paudel et al., 2019; Villa et al., 2019). VF4 shown in Fig. 4 (b) had a strong positive loading on pH, which could be linked to VF6 that had a strong positive loading on NH_4^+-N. The nutrient factor was related to agricultural runoff. The dominant agriculture activity like oil palm plantations was more commonly practiced in the vicinity of stream areas, hence increasing the entrainment of nutrient and polluting the aquaculture farms downstream (Selangor Waters Management Authority, 2015e). Previous studies had also revealed high level of ammonia in raw water at the study area (Abu Hasan et al., 2011; Kawasaki and Ramachandran, 2016).

Lastly, in G3, VF1 had a strong negative loading on colour and a moderate positive loading on hardness according to Fig. 5 (a). The local

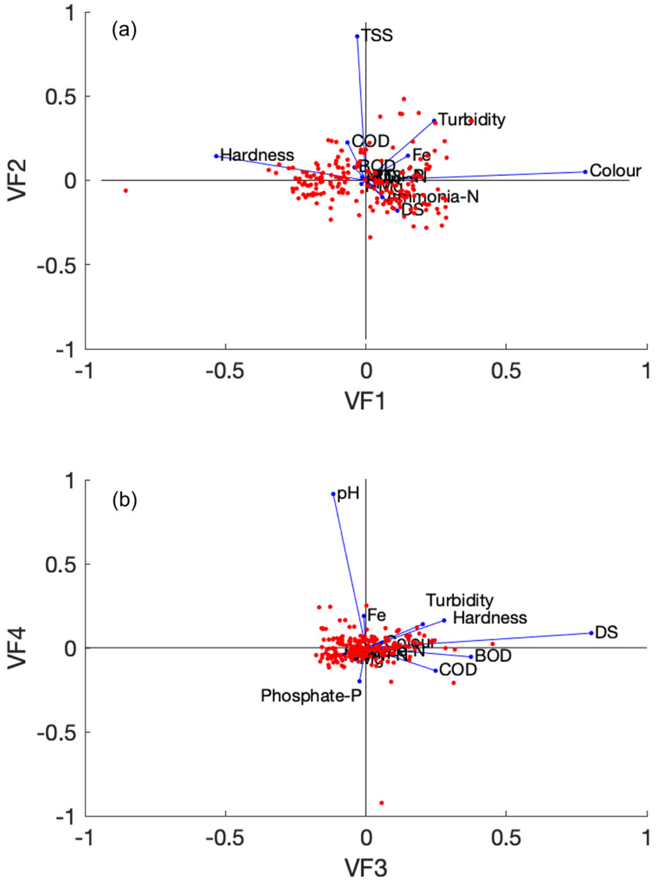

Fig. 5. Biplot for (a) VF1 and VF2 and (b) VF3 and VF4 showing loadings and normalised score of G3 stations.

authorities reported the colour of the river water changed due to large amounts of chemical runoffs from impermeable land surfaces into the river (Selangor Waters Management Authority, 2015e). Additionally, VF3 (Fig. 5 (b)) and VF5 had strong positive loadings on DS and Mg respectively, which were highly relevant to VF1. Mg concentration was higher in the catchment featuring agricultural land than in the catchment occupied by forest (Kuriata-Potasznik and Szymczyk, 2015). Besides, VF2 exhibited a strong positive loading on TSS, which was connected to land clearing and illegal sand mining (Nurhidayu et al., 2015; Selangor Waters Management Authority, 2015e). Memarian et al. (2013) found that sediment discharge into Langat River basin increased because of land use conversion from rubber plantation in 1984 to urbanised area in the recent decades. Past study showed the correlation of suspended solid with the increase in metals such as Fe and Mn, which could affect the water colour (Bayram et al., 2014). VF4 (pH) indicated the acidity of water while VF6 (NO_3^--N) and VF7 (NH_4^+-N) were the nutrient factor. The result was in conformity to Al-Badaii et al. (2013) and the probable sources included industrial, commercial, domestic, and agricultural activities. Agricultural practices that heavily polluted the river water included direct discharge of livestock sewage into the river and uncoordinated major cleaning-up of farms (Grace, 2017; Zakiah, 2020). Besides, Langat River water in G3 area was also prone to the pollution from leachate leak because the present unsanitary landfills lacked impervious lining and leachate treatment (Selangor Waters Management Authority, 2015a).

This study shows that PCA/FA results in significant data reduction by integrating several constituents into significant factors. Nutrient factor, dissolved solid factor, and sediment factor were the major parameters involved in river water pollution for all three river basins. We identified that domestic, industrial, and agricultural activities all contributed to the pollution sources in the study area.

3.4. Dry and wet periodic variations

Periodic variations were observed for all the clustering groups with principal constituent of loading >0.5 shown in Table 2. Though the principal constituents may be similar for dry and wet periods, they could appear in various factors with different eigenvalues and hence contributing different amounts of the total variance. In G1, for instance, COD had strong loading in VF2 and accounted for 15.5% of the variance for wet period compared to dry period, COD was found in VF3 which represented 9.2% of the variance. BOD showed strong loading during the dry period in VF5. This shows that complex organic compounds predominate over the year in the Klang River basin, unlike biodegradable organic compounds. In addition, TSS had comparable strong loadings during both dry (VF2) and wet (VF3) periods. Slightly higher loading in wet and rainy period may be associated with the variation of build-up and run-off processes (Salim et al., 2019). Furthermore, moderate PO_4^{3-}-P was reflected in VF1 during the wet period but not in the dry period. NH_4^+-N and NO_3^--N both appeared in dry and wet periods, but they contributed to a higher variance proportion during the wet period. The analysis shows that nutrient pollution is serious during the event of heavy rain in the Klang River basin. The result matched with a previous study by Kawasaki et al. (2016), who reported higher concentration of nutrients as a result of soil erosion in the wet period. The other principal constituents during the dry season were Mn in VF1, DS in VF4 and Mg and hardness in VF7, which were mainly attributed to natural processes (lithology) and anthropogenic activities (industrial effluent and earthworks). The results were aligned to that shown in Fig. 3 (a), whereby two significant groups of scores correlated with the observed loadings of NO_3^--N and Mn respectively. The observation implies that NO_3^--N leaching during the wet and rainy period is potent. During the wet period, the high surface runoff can drastically increase the NO_3^--N level, the effect is considerable compared to the dilution from a high river flow. Mn loading was significant during the dry period, suggesting earthworks as the most prominent activity and that anthropogenic impact is critical. Similar pattern of periodic variations was also observed for G2 and G3 stations.

Overall, PCA/FA indicates periodic variations in water quality and shows that TSS was the main factor that showed up at the study area in both periods. Hence, addition of pre-treatment units in WTP might solve the issue of filter choking due to high TSS. There were higher loadings of certain constituents observed during the dry period, like Mg, NO_3^--N, and so on. One plausible explanation is the prolonged wash-off of sediment and pollutants with extended dry conditions (United States Geological Survey, n.d.). Besides TSS, more attention should be given to reduce nutrient pollution in dry period because decreased level and stagnant water could cause entropic conditions in the river (Basheer et al., 2017a). Fortunately, high concentration of pollutants in surface water may be diluted by regulating the releases from reservoir (Hong and Hong, 2016). However, at times of seasonal extremes such as El Niño in 2014, the water level at the reservoir was critically low, thus water disruptions occurred frequently as there was no dilution (The Sun Daily, 2014). Contrarily, high precipitation during the wet period could either dilute the pollutant concentration or deteriorate the river water quality due to increased surface runoff. The relative importance of numerous water quality parameters during two different periods indicated the relative importance of point and non-point sources of constituents in the three river basins. Presumably, the hydrology, biological, and physiochemical factors in the environment were responsible for the periodic variations.

3.5. Water pollution threat to safe water supply and its trend

River pollution is the main factor causing frequent water disruptions in Selangor. Between 2009 and 2011, 67 cases of WTP shutdown were reported in the Selangor River basin (Selangor Waters Management Authority, 2011b). In 2009, Selangor River Phase 2 (SSP2) WTP and Selangor River Phase 3 (SSP3) WTP suffered two shutdowns and thirteen shutdowns, respectively due to high turbidity. In 2010, Selangor River Phase 1 (SSP1) WTP experienced seven shutdowns because of high

Table 2

Loadings of principal constituents on varimax rotated periodic matrix (dry and wet periods) for three clustering groups[a].

Clustering group	Wet				Dry		
	Factor	Principal constituent	Loading	Proportion variance (%)	Principal constituent	Loading	Proportion variance (%)
G1	VF1	PO_4^{3-}-P	0.606	45.4	Mn	0.622	43.6
	VF2	COD	0.964	15.5	TSS	0.826	14.9
	VF3	TSS	0.912	11.3	COD	0.925	9.2
	VF4	NH_4^+-N	0.956	7.4	DS	0.818	7.4
	VF5	DS	0.681	5.3	BOD	0.981	6.6
	VF6	NO_3^--N	0.707	4.5	NO_3^--N	0.955	4.3
	VF7	–	–	–	Mg	−0.688	3.6
					Hardness	−0.616	
	VF8	–	–	–	NH_4^+-N	0.899	3.3
G2	VF1	TSS	0.969	38.0	TSS	0.917	41.9
	VF2	Mg	0.958	17.6	Mg	0.983	20.9
	VF3	Hardness	0.869	11.2	Hardness	0.874	11.2
	VF4	pH	0.968	8.5	DS	0.574	5.9
					Turbidity	0.607	
	VF5	DS	0.879	7.0	NH_4^+-N	−0.866	5.2
	VF6	NH_4^+-N	0.877	4.3	pH	0.964	4.6
	VF7	Fe	−0.520	4.0	NO_3^--N	−0.552	2.7
		Turbidity	−0.770		PO_4^{3-}-P	−0.573	
G3	VF1	Hardness	0.832	30.4	Hardness	0.814	29.1
	VF2	TSS	0.724	16.6	TSS	0.888	19.3
	VF3	Colour	−0.526	14.4	pH	0.935	13.9
		DS	−0.785				
	VF4	NH_4^+-N	0.858	8.8	DS	0.794	8.7
	VF5	NO_3^--N	0.941	6.4	NO_3^--N	−0.983	7.4
	VF6	Mg	−0.888	5.5	Mg	0.927	6.4
	VF7	pH	0.813	4.6	Colour	0.959	5.0

[a] Only principal constituents with loading >0.5 (moderate and strong) are presented.

turbidity, fluoride, and ammonia, whereby similar causes were observed in 2011 with an increased number of shutdowns to eighteen. In the Langat River basin, Semenyih River WTP was shut down due to ammonia pollution in September 2010. In the following year, Cheras Batu 11 WTP and Semenyih River WTP experienced five shutdowns during 2011 due to diesel fuel pollution from industrial effluent (Selangor Waters Management Authority, 2011a). High turbidity also contributed to several shutdowns of Langat River WTP in 2012 (Ahmed et al., 2018).

The Selangor water crisis in 2014 was the most adverse water issue in Selangor since the Klang Valley water crisis in 1998. The dam's water level was critical due to dry weather from February to August. In 2014, WTPs in Selangor experienced 42 times shutdown for 2838 h in total due to raw water quality violation, mainly because of high ammonia levels (Permodalan Negeri Selangor Berhad, 2014; The Star, 2016). In 2016, odour pollution in Langat River basin caused the shutdown of Semenyih River WTP (Ahmed et al., 2018). Routine water disruptions in 2019 and 2020 were traced to odour pollution and diesel fuel pollution at SSP1, SSP2, SSP3, Rantau Panjang and Semenyih River WTPs. The water supply recovery was lengthy and had affected up to 1.2 million account holders (Christy, 2020). Very low water reserve margin (1.5%) only exacerbated the problem (Rajvinder, 2020). Ideally, the margin should be above 20%, when it falls to 5% and below, it enters the critical zone. To date, Selangor has a reserve margin at 12.8% and is expected to increase in the near future through complete construction of the Rasau Water Supply Scheme Project in 2028 (Air Selangor, 2021).

The recurring water supply disruption incidents have become a norm that is too costly to the citizens and government. Emergency Response Plan activated by Air Selangor, a water works company owned by Selangor, would cost approximately $ 1.63 million each time the WTPs are shut down (Azim, 2020). An estimated total of $ 4.96 million was spent for the cleaning works at the premises with four shutdowns, caused by Semenyih River pollution during 2016.

Consequently, water quality monitoring data collected in this study essentially convey a 10-year trend, serving as a performance indicator of the collective effort by various stakeholders over the time. The M-K test was applied to detect trends in the water quality parameters of concerns from three perspectives: nutrient, organic, and sediment factors. We studied the trends at G3 stations (Station S2 and L2) upstream of the main water intake points where repeated shutdown incidents had occurred. Station S2 is upstream of SSP1, SSP2, SSP3, and Rantau Panjang WTPs while station L2 is upstream of Semenyih River WTP and Bukit Tampoi WTP. The trend results are presented in Table S5.

3.5.1. Ammonia and nitrate

The M-K test showed no statistically significant trend for ammonia at Station S2 (midstream of Selangor River basin) but a statistically significant positive trend at Station L2 (midstream of Langat River basin). Based on Fig. 6 (a), the long-term trend was increasing, indicating the severity of ammonia pollution. Though SWMA embarked on Langat River Basin Management Plan to develop Integrated River Basin Management, the result suggested that the effort seemed inadequate, such as the lack of impervious lining and leachate treatment in sanitary landfills and lack of waste treatment in animal farming (Selangor Waters Management Authority, 2015a). Corrective measures are imperative to address ammonia pollution: semi-aerobic landfill and mandatory animal wastewater treatment pond which is well-maintained (Kamaruddin et al., 2017). Nitrate in Fig. 6 (b) was generally below the recommended standard for raw water quality, but there was insufficient evidence to prove that a trend exists with 95% confidence level at both stations.

3.5.2. BOD and COD

Fig. 7 (a) shows that the BOD level is low and M-K test presented no trend for BOD at both stations. Statistically significant negative trend was observed for COD at station S2 as shown in Fig. 7 (b), while there was no trend observed at station L2. This signifies COD trend as the primary indicator for organic factor in G3 stations. The decreasing trend in COD mirrored the improvement in water quality due to pollution abatement measure: rationalization and upgrade of existing sewage treatment plants (STP). However, it was found that 203 out of 278 STP in Selangor River basin were classified under Category 3 according to Sewage Effluent Regulation, which has a more lenient discharge limit compared to those under Category 1 and Category 2 (Selangor Waters Management Authority,

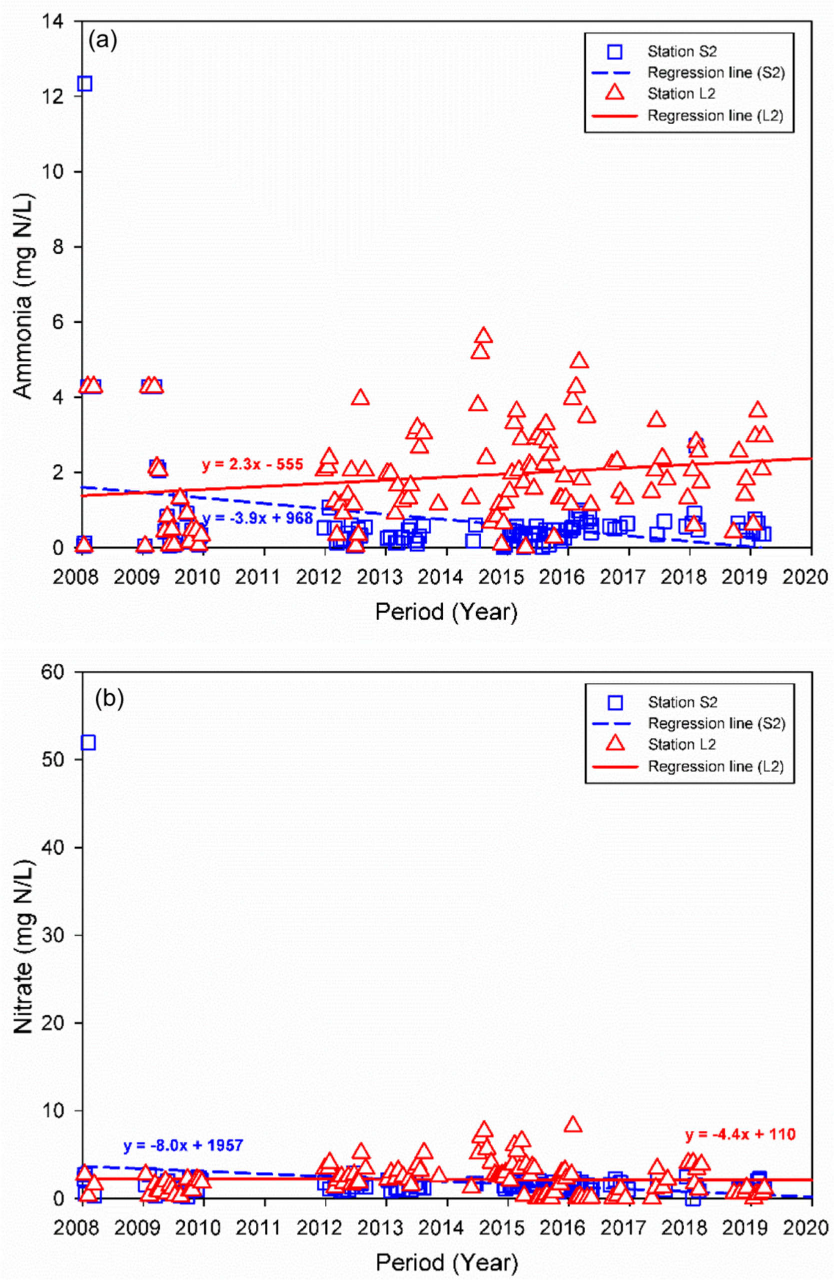

Fig. 6. Time-series trend of (a) ammonia and (b) nitrate at stations S2 (□) and L2 (△).

2015b). Consequently, the focus of sewerage master plan is to ensure that all plants achieve Category 1 through continuous upgrade.

3.5.3. Total suspended solid and turbidity

Statistically significant negative trends were observed for both TSS and turbidity at station S2 and L2. Fig. 8 (a) and (b) shows that TSS and turbidity trends were comparable and decreasing, both parameters are similar representative of sediment factor. Since late 2015, Hybrid Off River Augmentation Storage (HORAS), a water collection system built at former mining lands, has been operating to supply raw water in Selangor and Langat River basins during dry period. The result proves that HORAS not only provides emergency water supply, it facilitates sediment settling and improves the raw water quality (Selangor Waters Management Authority, 2015a). HORAS could be utilised as a pre-sedimentation pond to treat sediment pollution. Therefore, sediment pollution no longer contributes threat to water supply in the second half of the study period.

3.6. Effective water resource management

An effective water resource governance encompasses the interactions among all the socio-economic, political, and administrative systems. Yet, there remains resistance to widespread stakeholder engagement in devising water-related policies in Malaysia. There is a need to recognise water as a commodity that everyone has a stake, hence participatory approach in decision making requires institutions at all governance levels (Morrison, 2003). The stakeholder inclusiveness and collaboration will successfully address various water resources governance concerns and ultimately achieve the common goals towards ensuring sustainability and security of water resources. Nevertheless, there are cases in which overlapping responsibility or responsibility gaps create uncertainties and result in an unintegrated pollution control and enforcement. For instance, DID's engagement both at the federal and state level overlaps the state authority's jurisdiction, SWMA in Selangor. Enforcement of federal environmental law in states is

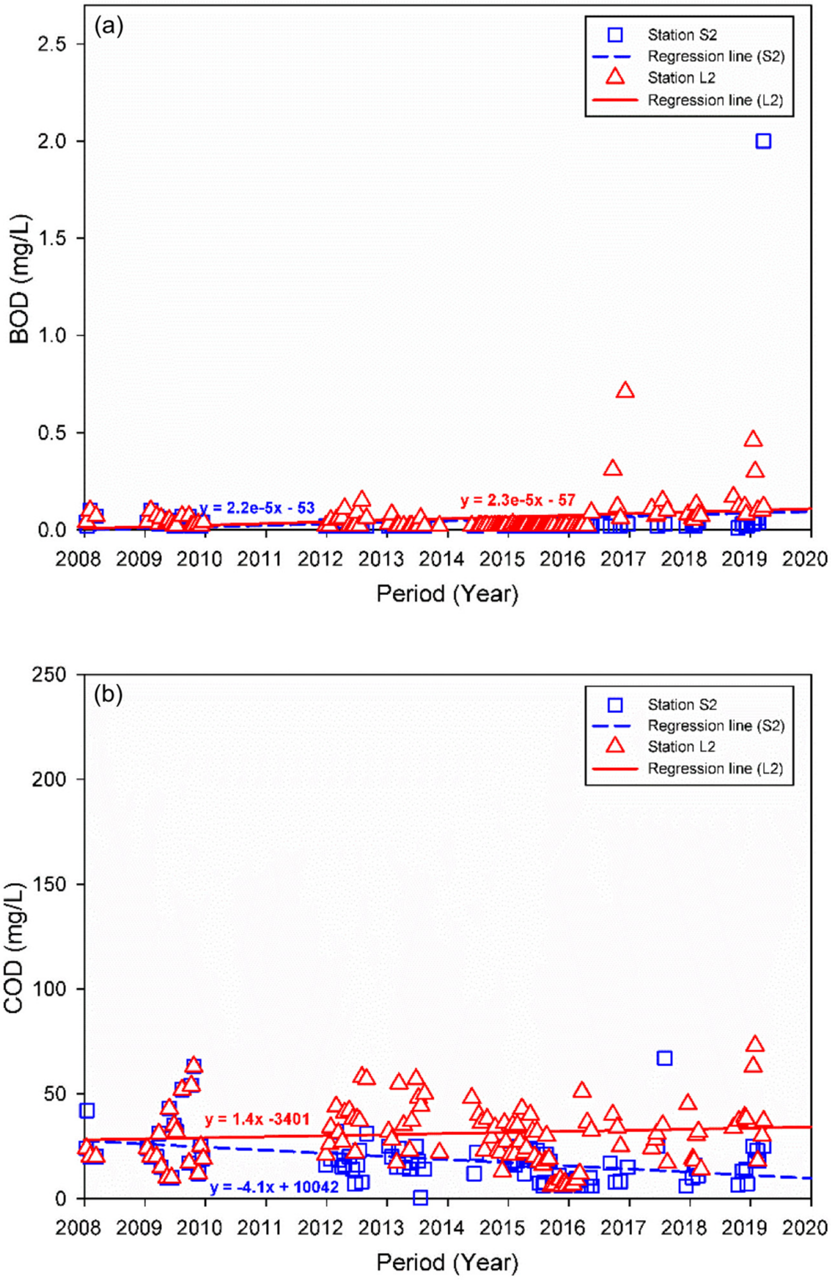

Fig. 7. Time series trend of (a) BOD and (b) COD at stations S2 (□) and L2 (△).

also hindered due to overlapping laws. For example, Environmental Quality Act 1974 does include provisions in banning inland pollution and industrial water pollution, but every state has their respective enforcement and policies. Thus, an effective water resources governance requires a clearer, formal delineation of its enforcement power to facilitate decision making process.

This study highlights that the decision making will not be focused only on one parameter, but all principal constituents included in the PCA/FA model. With the major constituents pinpointed, decision-makers could establish holistic strategies and effective legislation. For example, industrial zoning strategy could ensure lower risk of pollution incidents at potable supply water intakes. Though implementing industrial zoning is costly and challenging, but with proper planning, the benefits it offered have proven to outweigh the disadvantages. In China, Binhai Industrial Park shows effective control in water pollution while promoting the circular economy (United Nations Economic and Social Commission for Asia and the Pacific, 2019). Given the success of Binhai Industrial Park, the Selangor government should pursue a similar approach to accommodate more industrial zones at strategic locations far from water intakes. In response to accidental pollution, the governance bodies need to identify potential risk spots and establish an Accident Emergency Warning System to warn the potentially impacted water intakes downstream (International Commission for the Protection of the Danube River, 2015). This strategy is implemented in the Danube River Basin Management Plan and is deemed to be of benefit to Selangor state. Besides, increasingly serious ammonia pollution shown in the M-K trend analysis indicated that existing sewage treatment plants in Malaysia are inefficient in treating domestic wastewater (Zhanga et al., 2019). Therefore, further research could direct a path on continuous improvement of water and wastewater technology against nutrient pollution. One example is the application of mainstream and side-stream deammonification in wastewater treatment. The technology features energy-efficient biological nitrogen removal and has been practiced in a

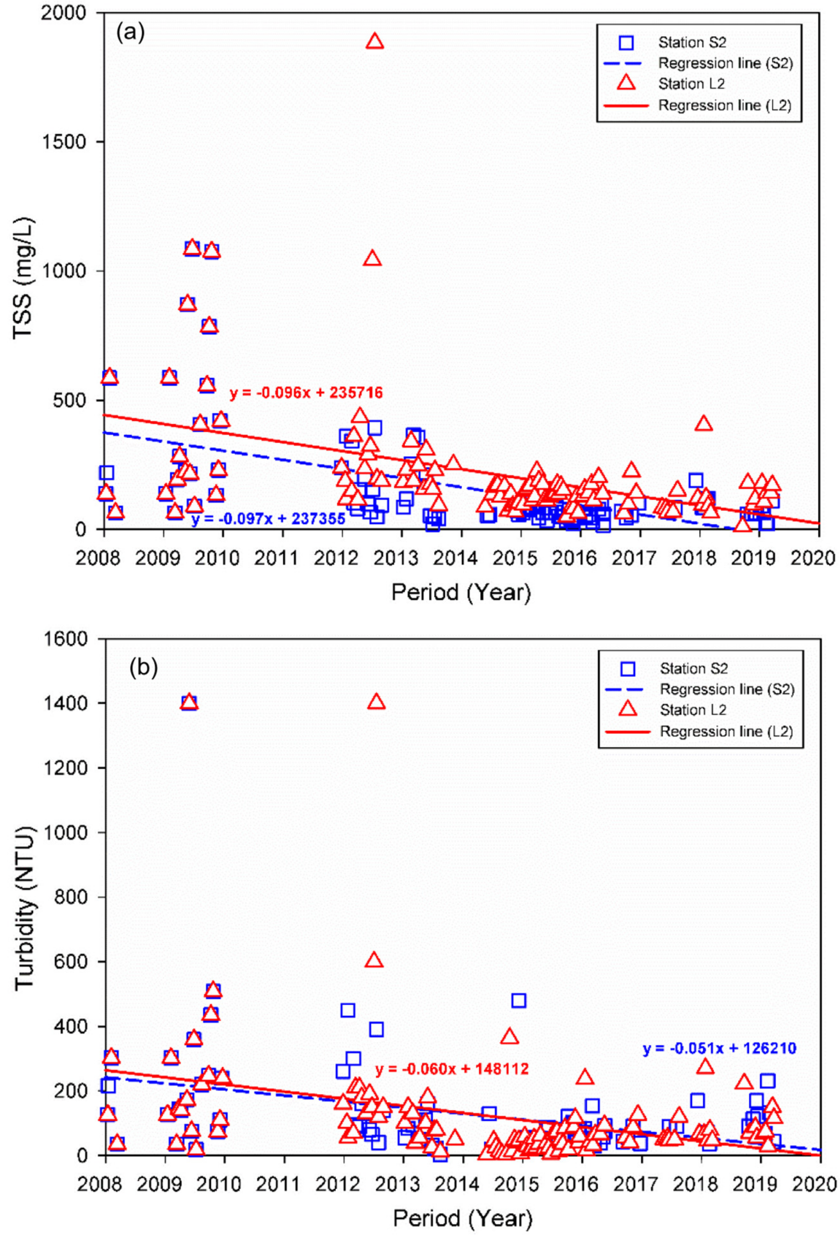

Fig. 8. Time series trend of (a) TSS and (b) turbidity at stations S2 (□) and L2 (△).

wastewater treatment demonstration plant at Singapore, paving the way for full-scale implementation in the near future (Singapore's National Water Agency, 2015, 2018). Similarly in Malaysia, How et al. (2018) proposed an energy-saving low-DO nitrification (<0.5 mg/L) suitable for the treatment of tropical wastewater which is low in COD to nitrogen ratio. The incorporation of low-DO anoxic-oxic-anoxic (AOA) reactor can enhance the nitrogen removal in tropical wastewater significantly (How et al., 2019). The simplification in reactor configuration from AOA to oxic-anoxic was also shown to improve nitrogen removal, befitting that of the energy sustainability operation by Indah Water Konsortium, a national sewerage company in Malaysia (How et al., 2020).

Eradicating ammonia pollution in raw water is a long-running battle, therefore the policy makers shall consider retrofitting the current water treatment facilities with technology effective in removing ammonia. Aside from wastewater treatment technology, biological nitrogen removal is also widely applied in polluted raw water pretreatment. Wu et al. (2021) demonstrated high-rate NH_4^+-N removal (hydraulic retention time: 0.5 h) from a major surface water source in Taiwan using a pilot-scale submerged biofilm reactor filled with porous polyurethane carriers. However, submerged biofilm reactor requires aeration for efficient nitrification, which can be energy-intensive. Alternatively, Maharjan et al. (2020) proposed an energy-saving downflow hanging sponge reactor (non-submerged biofilm reactor) for the removal of NH_4^+-N from contaminated groundwater source. The water is exposed to oxygen when it trickles down from the top of the reactor, providing an oxic environment naturally. As biological ammonia removal produces NO_3^--N, NO_3^--N removal is crucial to prevent negative health impacts such as methemoglobinemia and thyroid disease. Biological NO_3^--N removal is possible with the addition of external carbon source as electron donor reported by How et al. (2021). The biofilm technologies are advantageous because they retain abundant microorganisms and provide protection against hostile environmental changes. Hence, uninterrupted supply of safe drinking water in Selangor will necessitate the use of such promising pre-treatment technologies. However, the holistic effort of water resources governance should balance out the reactive phase of curing water pollution and the proactive phase of preventing water pollution.

4. Conclusion

This study investigated the distribution of pollutants in three major river basins in Selangor, Malaysia that serve as main sources for raw water supply. Agglomerative hierarchical cluster analysis grouped seven water monitoring stations into three groups of similar physicochemical characteristics and land use patterns. Principal component analysis/factor analysis further revealed nutrient factor (ammonia and nitrate), sediment factor (TSS), and DS factor as the significant water quality characteristics in the studied groups. Besides spatial variations, the results showed certain periodic variations (magnesium and nitrate), which were likely caused by seasonality of flows due to rainfall and runoffs from anthropogenic activities. In addition, the recurring water rationing had highlighted the vulnerability of Selangor's water resource system to water pollution, specifically, our result demonstrated a statistically significant increasing trend for ammonia pollution in midstream Langat River basin, upstream of the water intakes for at least four water treatment plants. Contradictorily, organic (COD) and sediment pollution showed signs of improvement with statistically decreasing trends observed. This work is important because it provides a direction to the environmental managers to devise action plans in reducing the impact of ammonia pollution. This might be succeeded by adopting sustainable agricultural practices and imposing a more stringent standard for sewage and industrial effluent discharge, hence, reducing the accumulation of ammonia in water and allowing ample time for dilution of pollution loads by natural means. This study also calls for immediate action by Selangor water works company to establish treatment facilities for ammonia-polluted raw water as key to uninterrupted water supply operation. Nevertheless, regular monitoring and continuous evaluation of raw water quality would be warranted.

CRediT authorship contribution statement

Jia Xing Loi: Conceptualization, Methodology, Formal analysis, Writing- Original draft preparation; **Adeline Seak May Chua**: Supervision, Project Administration, Funding acquisition; **Chee Keong Tan:** Supervision, Methodology; **Mohamad Fairus Rabuni:** Supervision, Conceptualization; **Sai Hin Lai:** Conceptualization; **Yasuyuki Takemura:** Visualization; **Kazuaki Syutsubo:** Funding acquisition, Project Administration. All authors have reviewed and approved the final manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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