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Environmental forensic study: tracing of pollution sources using environmetric technique in Balok and Tunggak Rivers near Gebeng industrial area, Kuantan, Pahang, Malaysia

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ABSTRACT

Extensive deterioration of water quality caused by the intensive land use activities in rivers and rapid response of pollutants from different sources may harm the aquatic organisms, humans, and the environment. The water quality of the Balok River is believed has been deteriorated by anthropogenic impact as a result of the industrial activities in the Gebeng industrial area. This alarming occurrence has initiated this study to investigate the main sources of pollution in two rivers near the Gebeng industrial zone. Data collection was done from two rivers namely Balok and Tunggak Rivers from 2018–2019 near Gebeng industrial area. The physicochemical parameters measured in this study were temperature, specific conductivity, pH, turbidity, dissolved oxygen, and salinity. The water samples were collected for tracing the radioactive elements and heavy metals by using inductively coupled plasma-mass spectrometry (ICP–MS). Then, the data were compared and analyzed by using environmetric analysis (cluster analysis and principal component analysis) based on the sampling site and source point. The finding from the environmetric analysis showed that the main pollutants loading in Balok and Tunggak River were Lead (first component), specific conductivity (second component), Thorium (third component), and pH (fourth component). It can be concluded that the presence of the pollutants in these two rivers were originated from both point and non-point sources which could be disquieting for the sustainable development of fisheries resources in the near future.

Keywords: Sources of pollution; Rivers; Environmetric analysis; Balok; Tunggak River

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1. Introduction

Malaysia is one of the countries that rich with its bounty of water resources and these resources are contributing to the economic and development of the country. Currently, one of the activities that gave beneficial economic development for countries is mining exploitation and also industrial activities. Therefore, water is becoming the natural resources that are very important to the earth and its residents for domestic and also industry user.

However, the conditions of water resources in Malaysia have been changing with urbanization and industrialization growth [1]. The river quality monitoring program was conducted by Department of Environment in 2014 found that 52% of the river in the country was found to be clean, while, 39% slightly contaminated and 9% are severely contaminated, where the major sources of pollution were contributed by the beverage industries [2].

Generally, industrial activities are generating a lot of pollutants through their production and manufacturing process [3]. Hence, these activities may cause extensive damages to the environment, river ecosystem, and human health if the concentration of toxic pollutants such as heavy metals and radioactive elements in the rivers is higher than the permissible limit. Since those toxic pollutants tend to dilute in the rivers and sea, their prescribed average safety levels in surface water are often misleadingly high [4,5].

Besides, the leachate, industrial effluents, and wastewater originated from those areas that will flow to the rivers and causes the deterioration of surface water quality [6–8]. Besides, the agricultural activities tend to increase pesticide load and nutrient deposition in the river due to land runoff [9,10]. The release of these pollutants into the lakes, streams, rivers, estuaries, and oceans may cause interference with the beneficial use of the water or with the natural functioning of ecosystems, thus affected the quality of the water body [11].

In addition to the release of substances, such as chemicals or microorganisms, water pollution may also include the release of energy, in the form of radioactivity or heat, into the water bodies. In short, river water bodies can be polluted by several pollutants, including the pathogenic microorganisms, organic waste, plant nutrients, toxic chemicals, sediments, heat, petroleum, and radioactive substances.

Hence, the rapid growth of industrial development may impose severe stress on the available water resources in terms of clean water supply. The deterioration of water quality and the depletion in the availability of water resources due to the radioactive element and heavy metals pollution may harm public health [12]. The previous study showed that the toxicity and accumulation of heavy metals and radioactive elements do not only depend on the metals concentrations but other factors. These factors include the form in which the metals are present, the type and concentration of other materials present and the integration of physicochemical water quality parameters [6,13].

This phenomenon has stimulated the search for prevention and treatment strategies against the deteriorating of water quality and the pollution of radioactive elements and heavy metals in rivers such as bioremediation and phytoremediation technology. However, to achieve this, a clear picture of the continuous monitoring of surface water quality and the isotopes such as heavy metals and radioactive elements present in it should be identified, as it will provide keywords or primary data for easier detection of main sources of pollutant in the rivers. The isotope of heavy metals and radioactive elements are the special value for detecting the residence time and monitoring its concentration in rivers that assuming that there is no contamination of the water has occurred.

Several studies were investigated about water quality and heavy metals [14,15], but no research focuses on the radioactive elements in surface water for tracing the pollution sources by using environmetric techniques had been attempted. In these contexts, investigation on the present status of rivers by applying the environmental forensic concept in tracing the main sources of pollutant loading in rivers within an industrial area by using the environmetric technique such as cluster analysis (CA) and principal component analysis (PCA) was carried out in this study. This study was focusing on two selected rivers namely the Balok river and Tunggak river near Gebeng industrial areas in Kuantan, Pahang, Malaysia.

2. Materials and methods

2.1. Selection of sampling area

The flow chart and the overall research methodology of this study is depicted in Fig. 1. The water samples were collected from water resources at Gebeng industrial area which is the Balok River and Tunggak River, Pahang and both of the rivers were located in the East of Peninsular Malaysia. Balok River is the main river that flows originally from Batang Panjang River from the hill to the northwest of Gebeng industrial area [16]. According to the data received from the Department of Irrigation and Drainage Kuantan, Balok River was ~45 m width and 10.5 km long from the upstream area to the downstream area. For Tunggak River, it is originated from Tanah Merah peat swamp forest and flows south along the eastern boundary of Gebeng industrial area. Based on the data received from the Department of Irrigation and Drainage Kuantan in the year 2018, Tunggak River was ~21 m width and 2.4 km long from the upstream area to the downstream area.

The map of Gebeng industrial area is shown in Fig. 2 to illustrate the sampling points selected for this study, while Table 1 summarized all the coordinates of the sampling points for both the Balok River and Tunggak River. This table explained in brief on the land use activities and the coordinates for each sampling point selected for this research study.

2.2. Sample collection and analysis

2.2.1. Water sample collection

The method for water sample collection was adapted from [17,18]. The water samples for *ex-situ* analysis were collected by using Van Dorn water sampler and there were three replicates of water samples were taken for each of the sampling points. The physicochemical parameters of river water quality were taken on-site (*in-situ*) using Hydrolab. Parameters such as temperature, specific conductivity, pH, turbidity, dissolved oxygen (DO), and salinity were recorded during the three times sampling in the wet season

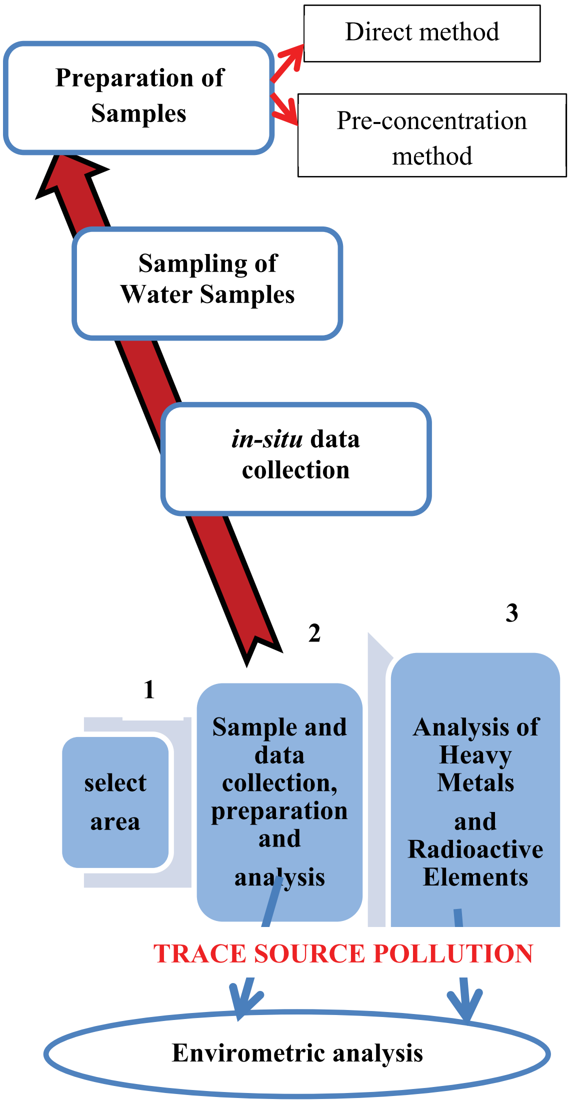

Fig. 1. Overall research methodology.

(September–December) in the year 2018 and dry season (March–June) in the year 2019.

2.2.2. Analysis of water (direct method)

The water samples for the direct method were analyzed following the previously published method [19]. This method was applied to the samples from the upstream and middle stream area because this area had salinity lower than 4.35% [20]. First, the samples were filtered (0.45 µm filter paper) on a vacuum filtration system. Then, 20 mL of water samples were topped up to 40 mL with 2% nitric acid. Then, the prepared samples were stored in the refrigerator (–20°C) before analyzed with the inductively coupled plasma mass spectrometry (ICP-MS).

2.2.3. Analysis of water (pre-concentration method)

The method for pre-concentration water analysis was modified from the previous method [21]. First, the surface

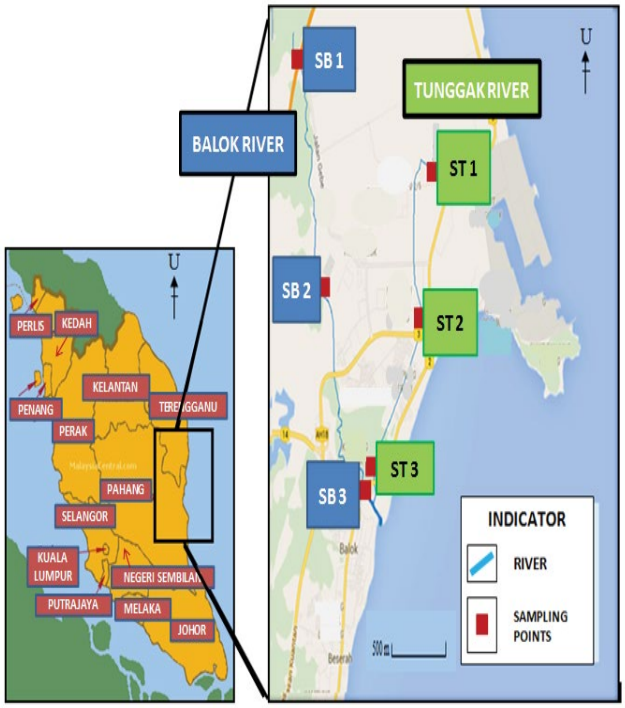

Fig. 2. Map of sampling locations at Gebeng industrial area.

water samples were filtered by using a low-pressure vacuum pump (0.45 µm filter paper) to remove any impurities. The Chelex-100 columns were packed in class 100 laminar flow hood. The Chelex-100 was weighed to 2.0 g (mesh size 100–200, powder form, Bio-Rad Laboratories). Then, it was soaked in 5 M HNO_3 for 72 h and loaded carefully into the column that blocked with glass wool at the end of it.

For this study, Masterflex Peristaltic Pump and C-flex tubing were used at the flow rate of 2 mL min^{-1}. About 15 mL of 2.5 M HNO_3 was added into the column, followed by 10 mL of distilled water. After that, 10 mL of 2 M ammonia was added into the column, followed by 20 mL of distilled water.

Next, 2 mL of 1 M ammonium acetate was added to buffer the column to pH 5.5, following the standard method [22]. A hundred milliliters of water samples were then added into the column. The water samples were eluted with 10 mL of 2 M HNO_3. The eluted solution was stored in the refrigerator (–20°C) for further heavy metals and radioactive elements analysis with ICP-MS.

At the end of the experiment, the column was rinsed with 10 mL of distilled water, followed by 10 mL of 1 M ammonium acetate, and 10 mL of distilled water, following the modified method by [22,23].

2.3. Analysis of heavy metals and radioactive elements

Heavy metals and radioactive elements analysis were performed by injecting the prepared samples and run in ICP-MS as described previously in [17,24] with some modification. In this study, Perkin Elmer ELAN 9000 system ICP-MS was used by referring its standard configuration as stated in Table 2.

Table 1
Coordinates and nearby industrial activities in Balok and Tunggak River

Sampling points	Location	Nearby activities
Balok River		
SB1	N 4°00′29.3″ E 103°20′59.50″	Steel factories, construction site, highway for car, lorry, and the other transportations
SB2	N 3°59′34.10″ E 103°21′28.50″	Petrochemical factories, Rare Earth Processing Plant, small road for car, lorry, and other transportations
SB3	N 3°56′31.30″ E 103°22′20.30″	Big road for car, lorry, other transportations, fishing area and jetty for boat, residential area, restaurants and shops, confluence of Balok River, and Tunggak River
Tunggak River		
ST1	N 3°59′14.40″ E 103°23′17.20″	Small road for car, construction site, lorries and other transportations, and mining activities
ST2	N 3°57′54.20″ E 103°23′23.10″	Wood factories, the industrial activities, big road for car, lorry and other transportations, shops, restaurants, agriculture waste, domestic waste
ST3	N 3°56′34.80″ E 103°22′33.10″	Big road for car, lorry, motorcycles and other transportations, fishing, the residential area, restaurants and shops, agriculture waste, and domestic waste

Table 2
Standard running conditions of ICP-MS

No.	Characteristics	Condition
1	Carrier gas flow rate	1.15 L min^{-1}
2	Coolant gas flow rate	16 L min^{-1}
3	RF power	1,340 W
4	Integration time	18 s cycle^{-1} isotope^{-1}
5	Number of cycles	3 cycle
6	Nebulizer	Standard
7	Sample flow rate	0.3 mL min^{-1}
8	Spray chamber temperature	2°C
9	Measurement mode	Peak hopping
10	Sweeps/reading	3
11	Reading/replicate	3
12	Replicates	3
13	Scanning mode	Peak hopping
14	Sampler and skimmer cones	Platinum

2.4. Preparation of standard solution for the calibration curve

The results obtained were quantified via the calibration curve generated from serial dilutions of multi-element calibration standards (Merck, US) before the analysis process takes place. A standard calibration curve was produced by diluting the stock solution to 1.0, 2.0, 5.0, 10.0, 50.0, 100.0, 300.0, and 500 ppb, respectively.

The multi-element calibration standard that was used for heavy metals and radioactive elements were different according to the metals and elements measured. In this study, both of multi-element calibration standard 2 and 3 were used. The multi-element calibration standard 2 was used for the determination of lead (Pb), cadmium (Cd), Arsenic (as), iron (Fe), and uranium (U). On the other hand, while the multi-element calibration standard 3 was used for the determination of thorium (Th).

2.5. Environmetric technique analysis

For this study, the environmetric technique analysis of physical water quality parameters including the heavy metals and also radioactive elements was undertaken to determine the interrelationship between different variables and also to identify probable sources of pollution to explain the pollution status [25]. This technique involves the analysis of cluster (CA) and also the principal component analysis (PCA).

In recent years, CA and PCA techniques had been applied to a variety of environmental applications [26] and this analysis was revered to be the best approach to periphrasis the misinterpretation of large amounts of complex environmental monitoring data. For this study, the environmetric technique of analysis was carried out using SPSS software 20 v (SPSS Inc., Chicago, USA).

2.5.1. Cluster analysis

The CA was practical to detect the similarities between sampling points in different sampling areas for different sampling times [17,27]. For this study, it was applied to the water quality, heavy metals, and also radioactive elements parameters data set with a view to group the parallel sampling points (spatial variability) spread over the river of Balok and Tunggak. This analysis was forming the higher clusters step by step and the result obtained was illustrated by a dendrogram.

2.5.2. Principal component analysis

In PCA used in this study, the eigen analysis of the water quality, heavy metals, and radioactive elements data were performed to extract the principal components (PC) of the measured data by using two selection criteria which are the scree plot test and the corrected average eigenvalue.

The characteristic roots or eigenvalues of the PC were a measure of their linked variances, and the sum of eigenvalues matches the total number of variables [27]. The correlation between PC and the original variables was given by loadings [17]. This analysis was useful for tracing the most important pollutant loading in Balok and Tunggak River.

3. Results and discussion

The PCA and CA were utilized to identify the effects caused by human activities on water quality for the rivers in Gebeng area. The PCA was applied to identify the main pollutant sources and the possible sources of pollution in this area whereas CA was done to group the monitoring sites according to the pollutant level [28] to identify the most polluted area. In this study, there is only spatial pattern was studied and this pattern was mainly related to the anthropogenic activities especially land input of pollutions. To analyze the data using CA and PCA, water quality parameters reading were taken on-site and the data were shown in Table 3 for both the year 2018 and 2019. Radioactive elements and heavy metals data also included in this analysis. However, the data collected for radioactive elements and heavy metals involved in this study for the year 2019 were not disclosed.

3.1. Cluster analysis

Fig. 3 illustrates the dendrogram on surface water from hierarchical CA that showed spatial clusters of six sampling sites at Balok and Tunggak River. CA is widely used for monitoring of both temporal and spatial variations and the interpretation of large and complex data sets [29]. CA was used to classify sampling points based on their level of similarity [30]. Hence, it was applied to give spatial variation among sampling points [31]. In this study, CA was performed on the water quality, heavy metals, and also radioactive elements data set to examine the spatial variation among the sampling point. The classification procedure procreated three cluster groups in a very definite way because the stations in these groups have similar characteristics.

The three clusters that formed according to their resemblance of variable properties were Cluster 1 (SB1, SB2, and ST2), Cluster 2 (ST1 and ST3), and Cluster 3 (SB3). Based on Fig. 3, Cluster 1 assigned as high pollution source (HPS), Cluster 2 assigned as a moderate pollution source (MPS), and Cluster 3 assigned as low pollution source (LPS). The distance cluster represents the degree of association between sampling points. The more significant association was resolute by the lower the value of the distance cluster.

The result obtained from this study showed that only one station for each cluster was needed to represent an accurate spatial assessment of the water quality, heavy metals, and radioactive elements for the whole network of rivers in the Gebeng industrial zone. This result indicates for rapid assessment of water quality, heavy metals, and radioactive elements in surface water and helps to improve the efficiency of surface water quality management [32].

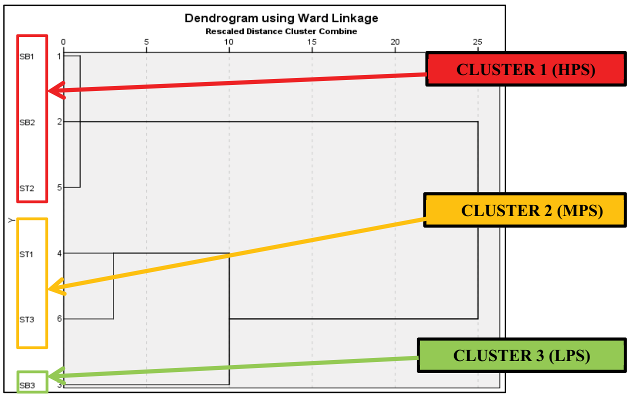

Fig. 3. A dendrogram from hierarchical CA that showed spatial clusters of six sampling sites at Balok and Tunggak River.

Table 3
In-situ water quality parameters analysis during the sampling period. Data expressed as mean

Parameters	Temperature (°C)	Specific conductivity (mS cm^{-1})	pH	Salinity (PSS)	Turbidity (NTU)	DO (mg L^{-1})
Year 2018						
SB1	30.36667	0.233333	7.851111	0.04	833.5556	6.406667
SB2	29.36111	11.96667	7.76	0.594167	829.6311	7.058889
SB3	27.84889	7.301111	7.587778	4.938889	326.0222	7.293889
ST1	28.17556	0.477111	7.247778	0.108889	621.8444	6.676667
ST2	28.28111	0.504556	8.476667	0.243333	812.3111	6.743333
ST3	28.16667	13.46722	7.546667	7.747456	524.5878	6.885556
Year 2019						
SB1	28.46556	0.847778	7.174444	0.417778	137.8333	3.568889
SB2	29.495	0.095	5.383333	0.051667	114.5	3.752222
SB3	30.45667	50.85	8.158333	33.69333	55.06667	4.931111
ST1	28.64833	5.553333	7.44	2.931667	112.8333	0.536667
ST2	28.29833	1.472333	8.158333	0.674444	57.53333	4.658889
ST3	30.265	52.31667	8.157778	34.78667	42.2	5.25

Therefore, the number of sampling points in the monitoring network can be reduced without losing any significance for the results [33]. This method will offer reliable clustering of water quality, heavy metals, and radioactive elements variables and can design the future sampling strategies in an optimum manner [32].

3.2. Principle component analysis

PCA was functional to the normalized data to compare the compositional patterns between the analyzed surface water samples in Balok and Tunggak River, and also to identify the factors that influenced each other. The PCA clarified most of the whole variations with some significant PC [34]. For further analysis of the PCA test, only four PC in the scree plot which were larger than one was retained.

Fig. 4 illustrates the scree plot found for this study. This scree plot was built based on 12 variables of water quality, heavy metals and also radioactive elements present in surface water to obtain the eigenvalues for further analysis by PCA.

In this study, these four PC had eigenvalues greater than one or close to unity and it explained 49.8%, 20.0%, 14.1%, and 11.9% of the total variances of the information contained in the original surface water data set. These values were obtained from the extraction sums of squared loadings in Table 4.

An eigenvalue gives a measure of the significance of the factor components by reducing the noise in data. It refers to the scalar that is used to transform or stretch an Eigenvector or the factor components. It eliminates features that have a strong correlation between them and helps in reducing over-fitting. The factor components with the highest eigenvalues were the most significant factors to be used. Eigenvalues of one or greater are considered significant [35].

PCA is one of the key strategies that are utilized to reduce dimension space without losing valuable information. The core components of PCA are built on the concept of the eigenvalues and factor components. In this study, the pollutants can be categorized into several groups by PCA according to their loading, where the component with higher loading described the characteristic of the total data set [36]. Projections of the original variables on the subspace of the PC were termed as loadings and overlap with the alternation coefficients between PC and variable.

Table 5 presented the rotated component matrix for water quality, heavy metals, and radioactive elements parameters from the PCA. The rotation of the axis created by PCA produced a new set of factors, which is each one of it involves primarily a subset of the original variables were divided into groups that independent towards each other [33].

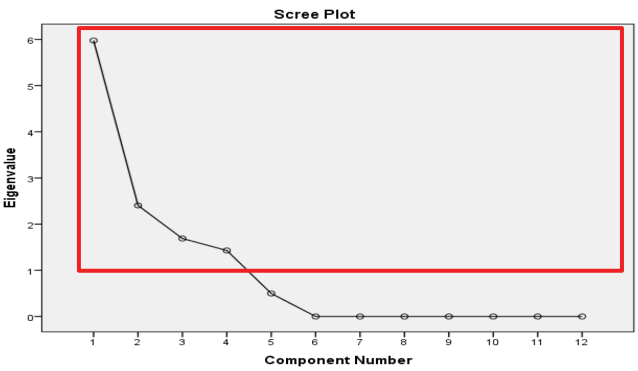

Fig. 4. Scree plot for variables studied.

Table 5
Rotated component matrix for water quality, heavy metals, and radioactive elements parameters

Parameters	Component			
	1	2	3	4
Temperature	–0.859		0.446	
Specific conductivity	0.296	0.914		0.103
pH	–0.407	–0.182	–0.521	0.721
Salinity	0.757	0.392	0.303	0.338
Turbidity	–0.964		–0.175	
DO	0.619	0.629	–0.360	
Ferum	–0.150		–0.149	–0.966
Lead	0.979		0.181	
Cadmium	0.750	0.577		–0.128
Arsenic	–0.471	0.766	–0.345	–0.264
Thorium		–0.162	0.963	
Uranium	0.789	0.304	0.411	0.275

Note: Extraction method: PCA. Rotation Method: Varimax with Kaiser Normalization. Rotation converged in 10 iterations.

Table 4
Extraction values of the total variance for water quality, heavy metals, and radioactive elements parameters

Component	Extraction sums of squared loadings			Rotation sums of squared loadings		
	Total	Percentage of variance (%)	Cumulative %	Total	Percentage of variance (%)	Cumulative %
1	5.978	49.814	49.814	5.273	43.945	43.945
2	2.405	20.045	69.859	2.472	20.600	64.545
3	1.689	14.072	83.931	2.000	16.668	81.212
4	1.430	11.918	95.849	1.756	14.637	95.849

Note: Only the first four components were shown since their eigenvalues >1.0

Based on Table 5, some of the parameters which loaded highly in the first component accounted for 49.81% of the total variance was lead (0.979), uranium (0.789), salinity (0.757), cadmium (0.750), DO (0.619), and specific conductivity (0.296). This result indicated that lead and uranium were the major pollutants in Balok and Tunggak River, followed by cadmium.

The sources of higher concentration of lead in river water might be coming from road runoff such as petrol and oil spill, paint, coating agent, and also batteries [37–40].

However, the second component had specific conductivity, arsenic, DO, cadmium, salinity, and uranium as the main parameters with a value of 0.914, 0.766, 0.629, 0.577, 0.392, and 0.304, respectively. Higher specific conductivity might be coming from natural sources, the inflow of saline water, and the mineral component of river water [18,41].

Conversely, the third component consisted of thorium (0.963), temperature (0.446), uranium (0.411), salinity (0.303), and lead (0.181). The higher thorium content in river water might be coming from anthropogenic activities such as rare earth treatment plants, factories, and industrial activities [42]. Our previous reports [4–6] identified major industries operated in Gebeng were the petrochemical processing, manufacturing, and the rare-earth elements processing and refining plant. Treated waters from other industries such as steel industries, palm oil mills, pipe coating companies were directly discharged into the Balok and Tunggak river which will then finally flowed into the South China Sea [6,30]. The fourth component, the main pollutants detected were pH (0.721), salinity (0.338), uranium (0.275), and specific conductivity (0.103). Higher pH value in the river might be coming from photosynthesis activities, microbial respiration, and decomposed activities of sewage and domestic waste [17,43,44].

4. Conclusion

This study showed that the CA helped to cluster the six sampling stations for both Balok and Tunggak River into three clusters (HPS, MPS, and LPS) of a similar characteristic based on water quality, heavy metals, and radioactive elements characteristics. Therefore, the few places with high loadings pollution sources of heavy metals and radioactive elements associated with hostile levels of physical water quality parameters such as SB1, SB2, and ST2 need more attention for effective pollution control mitigation. From the results obtained, the PCA showed significant data for variations in rivers water quality with strong positive loading of different pollutants at six sampling points. In short, the four varifactors or components obtained from the principal component point out that the parameters responsible for Balok and Tunggak Rivers quality are mainly contaminated by both point and non-point sources of pollution. Strong legislative support from authorities together with continuous monitoring of water bodies in the Gebeng industrial zone would pave a way in sustainable utilization of water resources in this area.

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