# Natural and Anthropogenic Determinants of Freshwater Ecosystem Deterioration: An Environmental Forensic Study of the Langat River Basin, Malaysia

**Ahmad Zaharin Aris, Wan Ying Lim and Ley Juen Looi**

**Abstract** Freshwater ecosystems face numerous threats that challenge the local authorities' ability on tackling down the water security (quantity and quality) issues and their management. The quality of surface water is an essential component of the natural environment and is considered as the main factor for controlling ecosystem health and potential hazard to the surrounding environment. The Langat River Basin in Selangor, Malaysia is exposed to natural and anthropogenic activities. A forensic investigation via the use of geostatistical and geochemical approaches and different standard criteria revealed two sources controlling the evolution of Langat River Basin water chemistry: (i) anthropogenic (agricultural and industrial activities) and (ii) natural processes (seawater intrusion and geological weathering). In addition, the suitability of river water for various purposes was determined based on the application of selected indicators and indices. The findings serve as an essential platform for the protection of water resources.

**Keywords** Environmental forensics · Geostatistics · Geochemistry · Heavy metals · Pollution indices · Indicators

## 1 Introduction

The freshwater ecosystem contributes to both human welfare and aquatic ecosystem by providing water for various purposes throughout the world. Therefore, the river water quality is an essential component of the natural environment and is considered as the main factor for controlling environmental health and potential hazard to the surrounding ecosystem. However, rivers may also carry a significant load of pollutants from different sources and affect not only the regions in the vicinity, but

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also the downstream reaches and possibly, the coastal and marine regions to which the river debauches (Shrestha and Kazama 2007; Alkarkhi et al. 2009). The land area within the basin, especially from the middle to lower reaches of the river always have been the preferred locales of housing, industrial, and agricultural activities. Since the waste discharges emanating from these activities are commonly not regulated and effectively controlled, it is more likely that levels of pollutants will increase in line with the scale of development (Suratman et al. 2009). These contaminants eventually end up into the runoff, land based area, and mangroves (Mokhtar et al. 2009a; Yu et al. 2010) and pollute the aquatic ecosystems. Therefore, the river quality has become a critical issue in many countries due to the concern that freshwater will be a scarce resource in the future.

Several workers attempted identification of metal pollution status in the river waters (Mokhtar et al. 1994, 2009a; Lim and Kiu 1995; Ahmad et al. 2009; Naji et al. 2010; Nasrabadi et al. 2010; Sultan et al. 2011; Eneji et al. 2012; Sultan 2012). Elevated concentrations of As, Pb, Cu, Cd, Cr, Ni, and Zn have been reported in various environmental media, especially in water, biota, and sediments. These metals were the most common pollutants found in the Strait of Malacca (Sarmani 1989; Gadgil 1998; Abdullah et al. 1999; Shazili et al. 1999, 2006; Yap et al. 2002). The Pb, Cd, Cr, Hg, and As are notorious contaminants, and are listed as the most hazardous inorganic contaminants in the EPA Hazardous Substance Priority List (EPA 2012). These elements are dissolved in water, highly toxic, and can cause significant health effects towards human and organism (Awofolu et al. 2005). They could exhibit extreme toxicity even at trace amounts and cause deadly diseases such as like edema of eyelids, tumor, neurological and genetic malfunctions (Hem 1970; Tsuji and Karagatzides 2001; WHO 2011). The Department of Environment recorded that areas with high anthropogenic pressure, particularly in Johor and Selangor states, have the highest number of water pollution sources. Sources of these pollutions are mostly attributed to the manufacturing industries, sewage treatment plants, agro-based industries, and animal husbandries (Ismail et al. 1993; Shazili et al. 2006; DOE 2010; Naji et al. 2010; Zulkifli et al. 2010b). Rahman and Surif (1993) and Zulkifli et al. (2010b) found that metal processing and petroleum-related industries produce e-waste which contain elevated levels of metals, including Zn, Cu, Ni, Fe, Al, Pb, Mn, Cr, and Sn. The by-product derived from nutrient supplements in animal feeds (such as copper sulfate pentahydrate) contributed elevated levels of Cu and has been correlated to contamination of sediments and molluscs (Sarmani et al. 1992; Ismail and Ramli 1997; UPUM 2002; JICA and MGDM 2002; Lee et al. 2006). Port and shipping activities are the complementary pollution sources contributing tributyltin, Pb, Cu, and As (Abdullah et al. 1999; Zulkifli et al. 2010b). Mining of Cu, Sn, Fe, and Au also increase the prevalence and occurrence of metal contamination in the aquatic ecosystems (Yusuf 2001; Ali et al. 2004).

Considerable efforts have been made in the past 2 decades to understand the environmental conditions of the river ecosystems in Malaysia. Notable among them are: river basin management (Mokhtar et al. 2011), hydrological properties and water quality (Alkarkhi et al. 2009; Suratman et al. 2009; Fulazzaky et al. 2010),

the distributions of heavy metals in soil, sediment and surface water (Sarmani 1989; Lim and Kiu 1995; Ali et al. 2004; Mokhtar et al. 2009a; Suratman et al. 2009; Sultan et al. 2011), the distribution and biodiversity of benthic macroinvertebrates (Azrina et al. 2006; Al-Shami et al. 2010), and the compositional patterns and occurrence of organic pollutants (Osman et al. 2012; Zainuddin et al. 2012). A common inference from all these diverse studies is that, the freshwater ecosystems are increasingly facing multiple pressures from a variety of contaminants.

The Langat River Basin is one of the most important freshwater ecosystems in the tropical west coast of Peninsular Malaysia. This tropical catchment area is now experiencing rapid urban expansion (Azrina et al. 2006; Amini et al. 2009; Ali et al. 2012; Zainuddin et al. 2012). The rapid development of this catchment area poses a threat to the river quality from manufacturing and agro-based industries (palm oil mills and rubber processing plants), sewage treatment plants, wastewater from animal farms (pig farms), ship waste and domestic activities (JICA and MGDM 2002; Lee et al. 2006; Bahaa-Eldin et al. 2008; Mokhtar et al. 2009b; Juahir et al. 2010; Zainuddin et al. 2012). The inadequate water management, uncontrolled contaminant discharge from industrial, domestic and economic activities bear a direct effect on the river ecosystem (Mokhtar et al. 2009b). Besides, the tropical climates, with warm temperature and high rainfall intensity enhance both physical and chemical weathering of rocks (Sultan et al. 2011), thus enhancing the suspended and solute fluxes.

Sarmani (1989), Yap et al. (2003), Bahaa-Eldin et al. (2008) and Mokhtar et al. (2009b) studied the heavy metal contamination in the Langat River. However, these studies have not attempted documenting the potential toxicity and suitability of the river waters for domestic and agricultural purposes, as is being studied elsewhere (Sultan et al. 2011). The present study attempts identification of the determinant factors that influence the hydrochemistry of the Langat River, Malaysia.

## 2 Regional Setting

The Langat River Basin is a trans-state river basin of the Peninsular Malaysia and has been recognized by the United Nations Educational Scientific Organization (UNESCO) as an Evolving HELP basin (Mokhtar et al. 2011; Fig. 1). The river flows from the western slope of Titiwangsa Mountains and draining into the Strait of Malacca. The Langat River Basin is located close to the equator, and thus is under the influences of northeast (November–March) and southwest (May–September) monsoons. There are two inter-monsoonal periods in between these monsoons, which are characterized by variable winds and thunderstorms (JICA and MGDM 2002). The spatial extent of the Langat River Basin and its hydrological characteristics are given in the Table 1. The topography of this basin can be separated into three distinct zones, namely, mountainous area, hilly area and lowland area (DOA 1995; Lim et al. 2012). The bedrock in mountainous area includes Permian igneous rocks, Pre-Devonian schist, and phyllite of the Howthornden Formation

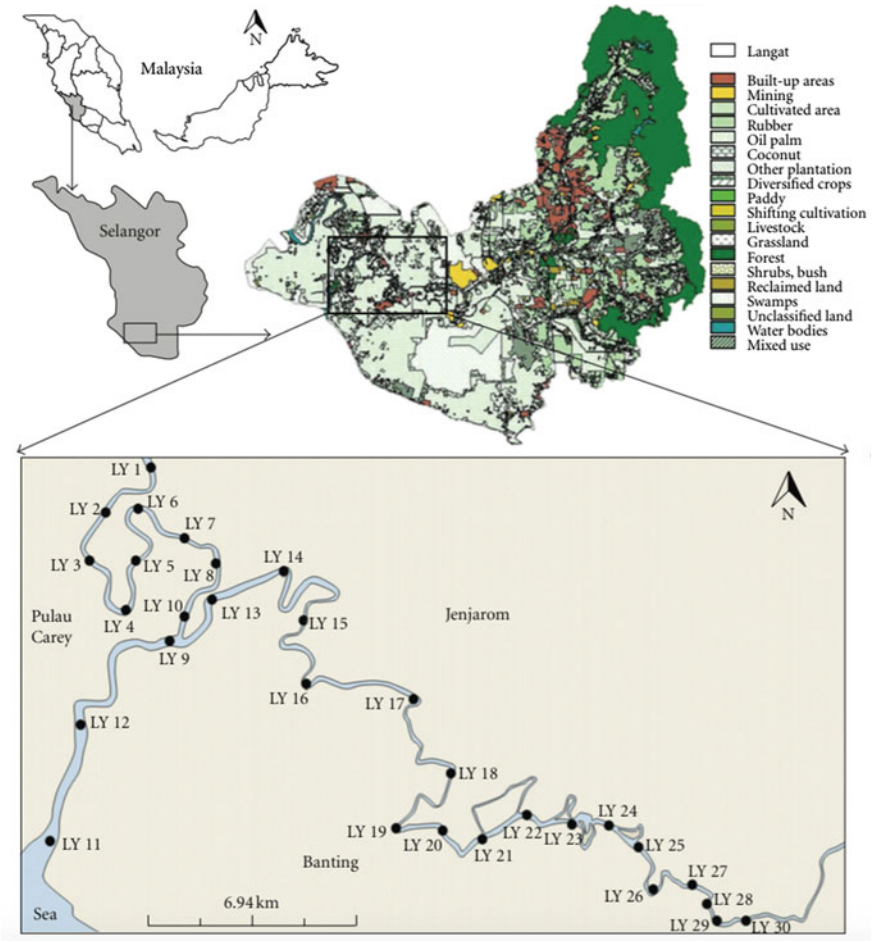


**Fig. 1** Map of sampling points in Langat River

(Gobbett and Hutchison 1973). The Permo-Carboniferous meta-sandstone, Kajang Formation, and Kenny Hill Formation are predominated in the hilly area while the lowland area is occupied by the Quaternary deposits of Beruas, Gula, and Simpang Formations (Gobbett and Hutchison 1973; Taha 2003). A downstream trend of change of river bed sediments from gravel in the catchments to clayey in the lowland area is conspicuous (JICA and MGDM 2002) as could be observed elsewhere.

There are three constructed dams, namely the Semenyih Dam, the Langat Dam, and the Putrajaya Dam, which supply water to the entire basin. The basin, especially its lower region has been experiencing massive development since the past few decades (Ali et al. 2012). On a regional scale, the upper region of the basin is predominated by natural forests, the middle region by built-up area and mining activities and the lower region by agricultural activities including oil palm

**Table 1** Spatial extent and hydrological characteristics of the Langat River Basin

| Parameters | Characteristics |
|---|---|
| Coordinate | Latitudes 2° 40′ 152″ N–3° 16′ 15″ N<br>Longitudes 101° 19′ 20″ E–102° 1′ 10″ E |
| Basin area | 1,815 $km^2$ |
| Main river course | 141 km |
| Total length | 120 km |
| Total number of tributaries | 39 |
| Annual average rain fall | 2,469 mm |
| Minimum and maximum range of rainfall | 1,521 and 2,883 mm, respectively |
| Average rainy day per year | 190 days/year |
| Basin level water production capacity | 1,052 mL/day |
| Average annual flow | 35 $m^3/s$ |
| Annual flood | 300 $m^3/s$ |
| Water abstraction range | 3.41–5.86 $m^3/s$ |
| Average temperature | 32 °C |
| Humidity | 80 % |

*Sources* Modified from DOA (1995), Yusuf (2001), DID (2010), Juahir et al. (2011), Mokhtar et al. (2011), Lim et al. (2012)

plantation, rubber, mixed horticulture and livestock (Fig. 1; DOA 1995; JICA and MGDM 2002; Ali et al. 2012). It is quite understandable from these that the demand for water for all these activities has been on the rise concomitant with deterioration of quality of the available water.

## 3 Materials and Methods

### *3.1 Sample Collection and Preservation*

During July 2011, a total of 90 water samples (3 replicates of 30 samples) were collected at 30 different sampling stations, which were labeled as LY 1–LY 30 (Fig. 1). All the variables were measured in triplicates to estimate the variability resulting from the sampling and analytical procedure. The sampling, preservation, transportation and storage of samples were based on the Standard Method for Water and Wastewater Analysis (APHA 2005). An appropriate quality control and quality assurance were practiced to provide and maintain a degree of confidence in data integrity and accuracy. In addition, all the chemical and reagents used were of analytical grade or equivalent and free from any contaminants. The collected samples were filtered with 0.45 μm cellulose acetate membrane filter (Whatman Millipore, Clifton, NJ, USA) to prevent clogging during analysis and to obtain the dissolved ions for metal analysis (APHA 2005). Each sample was then separated into two

pre-cleaned polyethylene bottles. The first bottle was utilized for determination of sulfate ($SO_4^{2-}$) and nitrate ($NO_3^-$), and the second bottle was for the determination of cations and heavy metals. Samples taken for metal analysis were acidified with concentrated $HNO_3^-$ (pH < 2) to prevent adsorption onto the walls of the bottles, to prevent precipitation and to retard any biological activities (APHA 2005).

## 3.2 In Situ Parameters Measurement

The in situ variables (temperature, dissolved oxygen (DO), pH, redox potential (*Eh*), electrical conductivity (EC), total dissolved solids (TDS), and salinity) were measured immediately at the sampling locations. This was to acquire representative value of water quality and to avoid biochemical changes in the samples (APHA 2005; Radojević and Bashkin 2007). The DO and temperature were measured using YSI 52-dissolved oxygen meter (YSI incorporated, Yellow Spring OH, USA). pH and Eh were determined using the SevenGo Pro-SG78 probe meter while EC, TDS and salinity were determined using the SevenGo Pro-SG7 probe meter (Mettler Toledo AG, Schwerzenbach, Switzerland). Before measurements, each meter was calibrated with freshly prepared buffer solutions to ensure flawless functioning and accurate reading.

## 3.3 Major Ions and Heavy Metals Analyses

The $HCO_3^-$ and $Cl^-$ concentrations were measured on site based on titration and Argentometric methods, respectively, by using raw water samples collected (APHA 2005). Filtered un-acidified samples were used for determination of $SO_4^{2-}$ (SurfaVer 4 method) and $NO_3^-$ (NitraVer 5 method) through UV-spectrophotometry (DR/2500 Spectrophotometer, HACH Odyssey, Loveland, Colorado, USA). The cations ($Ca^{2+}$, $Na^+$, $Mg^{2+}$, and $K^+$) and heavy metals ($^{27}Al$, $^{75}As$, $^{138}Ba$, $^{111}Cd$, $^{59}Co$, $^{63}Cu$, $^{52}Cr$, $^{57}Fe$, $^{55}Mn$, $^{60}Ni$, $^{208}Pb$, and $^{66}Zn$) were measured from filtered and acidified samples by flame atomic absorption spectrometry (FAAS, AA6800, Shimadzu Scientific Instruments, Kyoto, Japan) and by inductively coupled plasma mass spectrometry (ICP-MS, ELAN DRC-e, Perkin Elmer, Massachusetts, USA), respectively. The operating conditions of ICP-MS are given in Table 2. Several isobaric interferences were programmed (Table 3), and the interference corrections were automatically applied (if used) for standard-mode analysis by the ELAN software. The cations concentrations are expressed as milligrams per liter (mg/L) while the heavy metals concentrations are expressed as micrograms per liter (μg/L). The accuracy of metal analysis was assessed by external standards, which were prepared by diluting the ICP Multi-Element Mixed Standard III (Perkin Elmer, Massachusetts, USA) into series of concentrations with the same acid mixture used for sample dissolution. Recovery rates of 97–103 % (±5 %) were achieved.

**Table 2** The instrument setting of ICP-MS

| | Condition used |
|---|---|
| Instrument | Perkin Elmer SCIEX, ELAN DRC-e |
| RF power | 1,100 W |
| Gas | Argon gas-flow |
| Nebulizer gas flow rate | 0.72 L/min |
| Auxiliary gas flow rate | 1.2 L/min |
| Nebulizer | Cross-flow Gem-tip |
| Spray chamber | Cyclonic |
| Detector mode | Dual (electron multiplier operating in both pulse counting and analog modes) |
| Sampler/skimmer cones | Nickel |
| Dwell time per amu | 50 ms |
| Points per peak | 1 |
| Sweeps per reading | 10 |
| Readings per replicate | 3 |
| Replicates | 3 |
| Sample flush | 40 s (48 rpm) |
| Sample reading delay | 20 s (20 rpm) |
| Sample wash | 45 s (48 rpm) |
| Isotopes monitored | $^{27}$Al, $^{75}$As, $^{138}$Ba, $^{111}$Cd, $^{59}$Co, $^{63}$Cu, $^{52}$Cr, $^{57}$Fe, $^{55}$Mn, $^{60}$Ni, $^{208}$Pb and $^{66}$Zn |

**Table 3** Isotopes monitored and applicable parameters in standard mode

| Analyte | Mass | Potential interferences | Correction equation |
|---|---|---|---|
| Al | 26.9815 | BO, CN, BeO | – |
| As | 74.9216 | ArCl, $Sm^{2+}$, $Nd^{2+}$, $Eu^{2+}$ | −3.127 * [ArCl77 − (0.815 * Se82)] |
| Ba | 137.9050 | La, Ce | −0.000901 * La139 − 0.002838 * Ce140 |
| Cd | 110.9040 | MnO | – |
| Co | 58.9332 | CaO | – |
| Cu | 62.9298 | $PO_2$, TiO | – |
| Cr | 51.9405 | ArN, ClO, ArO, SO, ArC, HClO | – |
| Fe | 56.9354 | CaO, ArO | – |
| Mn | 54.9381 | ArN, HClO, ClO | – |
| Ni | 59.9332 | CaO | – |
| Pb | 207.9770 | – | – |
| Zn | 65.9260 | TiO, VO, $SO_2$, $Ba^{2+}$ | – |

### 3.4 Data Analyses

Descriptive analysis and skewness were performed using the PASW Statistics 18 (formerly known as SPSS Statistics 18, or SPSS Base). The descriptive statistic was performed to calculate maximum, minimum, mean, standard deviation (SD), and coefficient of variance (CV). SD was used as an indication of the precision of each parameter while CV was calculated based on the sum value of standard deviation from each parameter divided by its mean value. One-way ANOVA was applied to test the significant difference for all water quality variables among stations. Principal component analysis (PCA) was applied to apportion pollution sources and the contribution of each variable to the study area. The ionic ratios including ($Cl^-/HCO_3^-$) versus $Cl^-$, $Na^+$ versus $Cl^-$, $Ca^{2+}$ versus ($Ca^{2+} + SO_4^{2-}$) and ($SO_4^{2-} + HCO_3^-$) versus ($Ca^{2+} + Mg^{2+}$) were computed to delineate the possible mechanisms, which contribute to the river constituents. Meanwhile, the suitability of river water for irrigation purpose is evaluated based upon the estimation of the parameters like sodium percentage, magnesium hazard, Kelly's ratio and Wilcox diagram. The results were also compared with the different drinking water quality standards stipulated by the Malaysian Ministry of Health (MOH 2004) and World Health Organization (WHO 2011).

## 4 Results and Discussion

### 4.1 Source Apportionment of River Pollution in Relation to Natural and Anthropogenic Activities

Source apportionment approach improves the knowledge on the natural and human impacts on the aquatic environment. This explanatory analysis estimates the contribution of each variable and affords data reduction with a minimum loss of the original information (Shrestha and Kazama 2007; Mustapha et al. 2012). The PCA was applied on the water quality data set to identify the spatial sources of pollution in the Langat River. To ensure that there is no violation, preliminary analysis on the assumptions of Kaiser–Meyer–Olkin (KMO) measure of the sampling adequacy and Bartlett's test of sphericity were conducted. The KMO result was 0.916, and the Bartlett's test of sphericity was significant ($p < 0.001$). They proved that PCA can be considered as appropriate to provide significant reduction in the data dimensionality (Mustapha et al. 2012). Figure 2 and Table 4 display the component loadings after varimax rotation. Four components with eigenvalues greater than 1.0 were extracted that explained 89 % of total variance (Table 4). More than 70 % of the data variance was explained by the first two components (Fig. 2; Table 4). The first PC explains the highest variation among the variables; while the remaining PCs explain the variations among the variables in order (Civan et al. 2011).

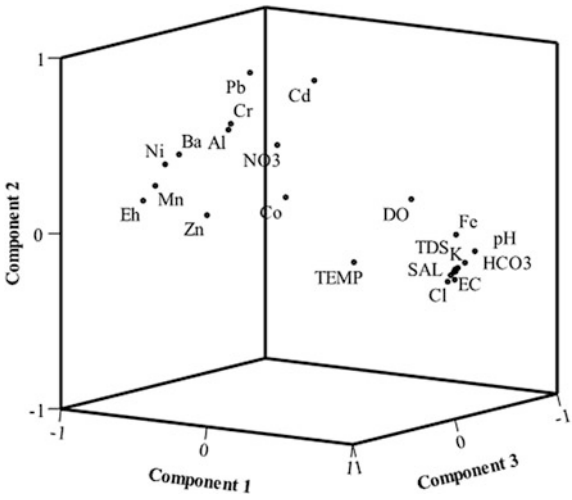


**Fig. 2** Component plot of PCA in rotated space

The components expose the potential factors responsible for variation in river water quality and eventually lead to sources identification of river pollution.

PC 1 with total variance of 64 % has a strong loadings of $SO_4^{2-}$, salinity, EC, TDS, $Ca^{2+}$, $K^+$, $Mg^{2+}$, Cu, As, $Na^+$, $HCO_3^-$, Fe, Mn, $Cl^-$, pH, Eh, Ba, temperature, Cr, Ni, and $NO_3^-$. The presence of dissolved ions including major ions ($SO_4^{2-}$, $Ca^{2+}$, $K^+$, $Mg^{2+}$, $Na^+$, $HCO_3^-$, $Cl^-$, $NO_3^-$) and heavy metals (Cu, As, Fe, Mn, Ba, Cr, Ni) may trigger the values of EC, salinity and TDS (Radojević and Bashkin 2007; Lim et al. 2012; Shafie et al. 2013a). In addition, the strong positive loadings on EC, salinity, TDS, and major ions explained that the ion-exchange reactions between freshwater (river) and seawater corresponded to seawater intrusion into the Langat River (Aris et al. 2012; Lim et al. 2012). Salinity portrays a vital role in the adsorption and desorption mechanism as it gives strong correlation with all the exchangeable cations (Shafie et al. 2013a). Therefore, the mixing of seawater and river water caused the competition between cations and heavy metals for binding sites in particulates (Connell and Miller 1984; Elder 1988; Lim et al. 2012). The cations being more prominent than other ions, especially the heavy metals and thus caused these ions desorbed from sediment. Thus, it will increase the concentration of heavy metals in river water (Connell and Miller 1984; Elder 1988). The presence of these metals also indicated that the quality of river water was affected by the inorganic compounds perhaps from anthropogenic activities in the study area. Heavy metals including Cu, As, Fe, Mn, Ba, Cr, and Ni were prominent in PC 1. These elements could be characterized by their natural availability in the river basin. Their presence is likely due to the oxisols and ultisols, which are the dominant soil in Peninsular Malaysia (Nieuwolt et al. 1982; Tessens and Jusop 1983; JICA and MGDM 2002). In addition, the oxides of Fe and Mn (goethite and hematite) are commonly found in tropical soils (Nieuwolt et al. 1982; Tessens and Jusop 1983) and have a great effect on the chemical behaviour of metals in sediment

**Table 4** Component loadings of river water quality variables on varimax rotated matrix

| | PC 1 | PC 2 | PC 3 | PC 4 |
|---|---|---|---|---|
| $SO_4^{2-}$ | **0.98** | −0.15 | −0.03 | 0.09 |
| Salinity | **0.98** | −0.15 | −0.04 | 0.11 |
| EC | **0.98** | −0.16 | −0.03 | 0.11 |
| TDS | **0.98** | −0.16 | −0.03 | 0.11 |
| $Ca^{2+}$ | **0.98** | −0.17 | −0.04 | 0.11 |
| $K^+$ | **0.98** | −0.15 | −0.06 | 0.12 |
| $Mg^{2+}$ | **0.97** | −0.17 | −0.02 | 0.11 |
| Cu | **0.97** | −0.18 | 0.00 | 0.10 |
| As | **0.97** | −0.21 | −0.04 | 0.05 |
| $Na^+$ | **0.97** | −0.16 | −0.05 | 0.15 |
| $HCO_3^-$ | **0.96** | −0.13 | −0.15 | −0.07 |
| Fe | **0.95** | 0.03 | −0.09 | −0.16 |
| Mn | **−0.94** | 0.16 | 0.16 | 0.19 |
| $Cl^-$ | **0.93** | −0.23 | −0.03 | 0.21 |
| pH | **0.89** | −0.10 | −0.35 | −0.14 |
| Eh | **−0.89** | 0.10 | 0.35 | 0.14 |
| Ba | **−0.88** | 0.32 | 0.02 | 0.13 |
| Temperature | **0.61** | −0.08 | 0.43 | 0.48 |
| Cr | **−0.61** | 0.51 | −0.10 | −0.39 |
| Ni | **−0.60** | 0.37 | 0.55 | 0.22 |
| $NO_3^-$ | **−0.56** | 0.33 | −0.49 | 0.04 |
| Pb | −0.28 | **0.87** | 0.18 | 0.08 |
| Cd | 0.00 | **0.82** | −0.05 | 0.15 |
| Al | −0.48 | **0.51** | 0.10 | −0.40 |
| Zn | −0.12 | 0.17 | **0.82** | 0.18 |
| DO | 0.14 | 0.05 | **−0.79** | 0.03 |
| Co | −0.07 | 0.17 | 0.13 | **0.76** |
| Initial eigenvalue | 17.21 | 2.77 | 2.44 | 1.51 |
| Percent of variance | 63.73 | 10.27 | 9.02 | 5.60 |
| Cumulative percent | 63.73 | 74.01 | 83.03 | 88.62 |

*EC* Electrical conductivity; *Eh* Redox potential; *DO* Dissolved oxygen; *TDS* Total dissolved solids. The values in bold are factor loadings above 0.50 that were taken after varimax rotation was performed

(Alloway 1995). However, Cu, As, Cr and Ni may be attributed to anthropogenic pollution. Agriculture is the dominant activity in the study area. The use of inorganic fertilizers such as arsenical herbicides or ammonium nitrate is frequent and widespread. Therefore, high loadings of As and $NO_3^-$ could have been caused by agricultural activities. Besides, another source of contamination might come from pig farming activities. The by-products from pig food (copper sulfate) can contribute to Cu concentration via effluent discharged into the river (Sarmani et al. 1992;

UPUM 2002; Lee et al. 2006). This inference is also supported by several previous studies, which explained the effects of anthropogenic activity toward this river (Sarmani 1989; Sarmani et al. 1992; Lee et al. 2006; Mokhtar et al. 2009b; Juahir et al. 2011). Thus, PC 1 is believed to be interpreted by two contributions, the first is related to seawater intrusion; and the latter, related to the agricultural and farming activities.

PC 2 with total variance of 10 % consists of Pb, Cd, and Al. Relatively high loadings of these elements indicate that the river waters suffer from metal pollution, possibly by the heavy shipping traffic (Shazili et al. 2006; Mokhtar et al. 2009b; Berandah et al. 2010). The steel manufacturing and metal finishing factories located in the upstream (Dengkil) of the sampling location (Mokhtar et al. 2009b) could also have contributed. Yet another reason, namely the ongoing intensive dredging, reclamation, and construction activities in the river channel and in the adjoining regions might resuspend the stream-bed sediments and reintroduce the trace metals bound in the sediments into the river water (Nayar et al. 2004; Zulkifli et al. 2010a). Similar inferences were reported in previous studies, which explained the effects of anthropogenic inputs (Sarmani 1989; Shafie et al. 2013a) into this river. PC 3 and PC 4 have strong loadings of Zn, DO, and Co which illustrate that the DO plays a role in controlling the behavior and distribution of metals in the river water. The Co was possibly derived from lithogenic or anthropogenic sources, especially from agricultural activities (Hooda 2010). However, Co was observed to be present in low concentration compared with other metals. Hence, lithogenic source to Co is attributed.

## *4.2 Ionic Ratio*

As discussed in the PCA section, it was deduced that the mixing of seawater is an influential hydrochemical process in the study area. To ascertain the origin and chemical behavior of the river water, several ionic ratios were attempted. The relationships of ($Cl^-/HCO_3^-$) versus $Cl^-$, $Na^+$ versus $Cl^-$, $Ca^{2+}$ versus ($Ca^{2+} + SO_4^{2-}$) and ($SO_4^{2-} + HCO_3^-$) versus ($Ca^{2+} + Mg^{2+}$) were computed to delineate the possible mechanisms that may contribute to the river water constituents (Hounslow 1995; Moujabber et al. 2006; Aris et al. 2012; Isa et al. 2012; Lim et al. 2012). Typically, $Ca^{2+}$ and $HCO_3^-$ are the dominant constituents found in freshwater whereas $Na^+$ and $Cl^-$ are the most abundant constituents in seawater and the enrichment of these constituents could be an indicator of saline water intrusion to river water (Aris et al. 2012). From the Fig. 3a, it follows that sampling stations located near coastal area (LY 1–LY 15), are saline than other stations, resulting in a higher ($Cl^-/HCO_3^-$) versus $Cl^-$ ionic ratio. The result represents a greater contribution of saline water to the river water relative to the amount of $Cl^-$ that might be present. In addition, 50 % of the samples with low ratio of ($Cl^-/HCO_3^-$) versus $Cl^-$ indicate freshening of river water. A $Na^+/Cl^-$ ratio that is greater than 1 describes silicate weathering and about 44 % of the samples fall within this range (Fig. 3b). Around 50 % of the samples were below 1, and represent the occurrence of ionic exchange process while 6 % of the samples were equal to 1, representing halite dissolution. The $Na^+/Cl^-$ ratio of the

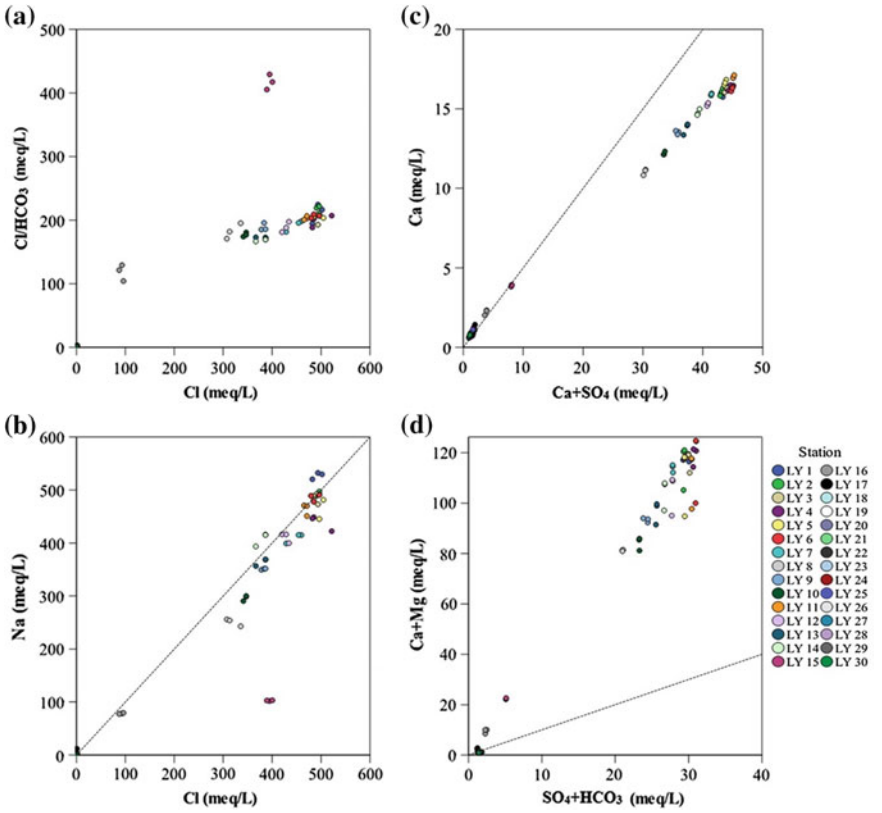


**Fig. 3** Distribution of ionic ratio for major ions describing mechanism involve

samples collected from stations located near coastal area (LY 15–LY 30) indicate salinization due to the result of mixing with seawater. The stations LY 16–LY 30, show heightened levels of the $Na^+$ and the same may have been resulted by irrigation return flow or due to evaporation (Jeevanandam et al. 2007). The scatter plot $Ca^{2+}/Ca^{2+} + SO_4^{2-}$ (Fig. 3c) is another example to illustrate the ionic exchange process. It was deduced that 52 % of samples were below 0.5, which indicates the occurrence of Ca depletion, either through ion exchange or calcite precipitation. The order of cation affinity tends to be $Na^+ > K^+ > Ca^{2+} > Mg^{2+}$ if there is mixing with seawater, where Ca being displaced from the exchanger in the first order and Na eventually dominating the water and the exchanger (Aris et al. 2007, 2012). 48 % of the samples contain Ca > 0.5, and indicate that the Ca may have been drawn from carbonate ($CaCO_3$) or silicate ($Ca_2SiO_4$) sources. This inference is affirmed by $(Ca^{2+} + Mg^{2+})$ versus $(SO_4^{2-} + HCO_3^-)$ ratio (Fig. 3d), as 43 % of the samples fall below the equiline which indicates weathering of minerals. The rest of samples lie above equiline, and suggest that carbonate weathering might have contributed ions to the samples under study.

## 4.3 Suitability for Drinking and Domestic Purposes

The analytical results of in situ variables, major ions and heavy metals are presented in the Table 5. Significant differences of all the studied variables among sampling stations ($p < 0.05$) are observed. All the variables were compared with the drinking water quality standards stipulated by the Malaysian Ministry of Health (MOH 2004) and the World Health Organization (WHO 2011). As depicted in the table,

**Table 5** Descriptive analysis of in situ variables, major ions and heavy metals (n = 90)

| | Unit | Range | Mean | SD | CV | MOH | WHO |
|---|---|---|---|---|---|---|---|
| *In situ* | | | | | | | |
| pH | – | 6.20–7.61 | 6.82 | 0.34 | 4.94 | 6.5–9.0 | NA |
| *Eh* | mV | −25.90–61.90 | 23.30 | 21.04 | 90.27 | NA | NA |
| Temp | °C | 28.10–30.20 | 29.40 | 0.68 | 2.31 | NA | NA |
| EC | µS/cm | 173.10–39500.00 | 17024.69 | 17665.66 | 103.76 | NA | NA |
| Salinity | ppt | 0.09–25.10 | 10.68 | 11.21 | 104.94 | NA | NA |
| DO | mg/L | 0.72–3.17 | 1.55 | 0.51 | 32.88 | NA | NA |
| TDS | mg/L | 86.50–19740.00 | 8511.26 | 8831.83 | 103.77 | 1000 | NA |
| *Major ions* | | | | | | | |
| $HCO_3^-$ | mg/L | 26.84–156.16 | 95.02 | 41.36 | 43.53 | NA | NA |
| $Cl^-$ | mg/L | 16.00–18494.27 | 7802.55 | 7773.31 | 99.63 | NA | NA |
| $NO_3^-$ | mg/L | 0.06–3.50 | 1.32 | 0.79 | 60.05 | NA | 50 |
| $SO_4^{2-}$ | mg/L | 15.00–1375.00 | 590.18 | 600.65 | 101.77 | 250 | NA |
| $Na^+$ | mg/L | 11.63–12240.00 | 4645.45 | 4857.57 | 104.57 | 200 | NA |
| $Ca^{2+}$ | mg/L | 11.92–343.09 | 153.40 | 142.34 | 92.79 | NA | NA |
| $K^+$ | mg/L | 5.53–363.41 | 135.22 | 133.96 | 99.07 | NA | NA |
| $Mg^{2+}$ | mg/L | 0.98–1319.30 | 528.28 | 553.90 | 104.85 | 150 | NA |
| *Heavy metal* | | | | | | | |
| Al | µg/L | 11.72–122.90 | 46.28 | 32.71 | 70.67 | 200 | NA |
| As | µg/L | 1.79–21.48 | 11.18 | 8.29 | 74.13 | 10 | 10 |
| Ba | µg/L | 3.42–27.40 | 12.18 | 8.14 | 66.81 | 700 | 700 |
| Cd | µg/L | <0.01–0.38 | 0.07 | 0.09 | 126.84 | 3 | 3 |
| Co | µg/L | 0.10–0.56 | 0.21 | 0.07 | 33.13 | NA | NA |
| Cu | µg/L | 0.40–58.82 | 26.02 | 25.45 | 97.79 | 1,000 | 2,000 |
| Cr | µg/L | <0.005–3.09 | 0.67 | 0.90 | 133.22 | 50 | 50 |
| Fe | µg/L | 61.90–162.53 | 116.23 | 28.68 | 24.67 | 300 | NA |
| Mn | µg/L | <0.0005–504.46 | 200.54 | 190.71 | 95.10 | 100 | NA |
| Ni | µg/L | <0.005–3.07 | 0.46 | 0.66 | 142.80 | 20 | 70 |
| Pb | µg/L | <0.005–1.34 | 0.16 | 0.23 | 145.91 | 10 | 10 |
| Zn | µg/L | 0.34–19.21 | 3.10 | 3.29 | 106.26 | 3,000 | NA |

*SD* Standard deviation; *CV* Coefficient of variance; *Eh* Redox potential; *Temp* Temperature; *EC* Electrical conductivity; *DO* Dissolved oxygen; *TDS* Total dissolved solids

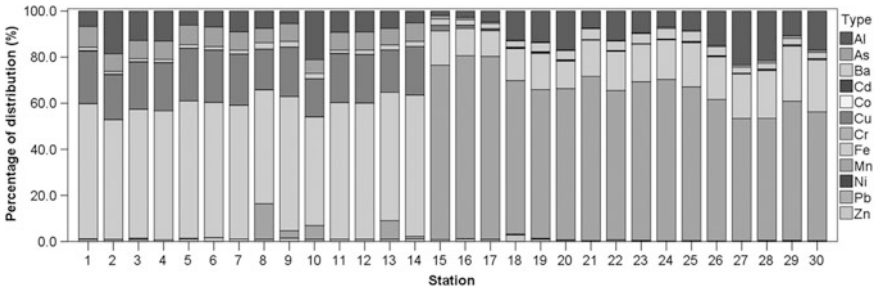


**Fig. 4** Distribution of heavy metals according to its sampling stations

the pH value of the water samples is weakly acidic to circum-neutral ranging from 6.20 to 7.61. Most of the stations were well within the limit prescribed by MOH (6.50–9.0). The TDS values ranged from 86.50 to 19740.00 mg/L and 53 % of them exhibited a value greater than 1,000 mg/L recommended by MOH. For major ions, it was found that 57, 50, and 50 % of samples exceeded the permissible limits of Na, Mg and $SO_4$ respectively (MOH 2004; WHO 2011; Table 5). The Fig. 4 depicts the spatial distribution of metals.

The mean concentration of heavy metals follows the order of: Mn > Fe > Al > Cu > Ba > As > Zn > Cr > Ni > Co > Pb > Cd. Except As and Mn at certain locations, all the heavy metals in the studied water samples are within the permissible limits (MOH 2004; WHO 2011; Table 5). 46 % of the samples exceed the permissible As content (10 µg/L; MOH 2004; WHO 2011) For As, 46 % of samples were found to exceeded the MOH and WHO permissible limits. Several studies stated that the As pollution in the Langat River basin result by agricultural application (arsenical herbicides) and tin mining (Sarmani 1989; Shafie et al. 2013a, b). 53 % of stations exhibited Mn concentration higher than the MOH standard (100 µg/L; MOH 2004). The direct reduction of particulate manganese oxides in aerobic environments by organic matters, the natural weathering of Mn-bearing minerals and acid drainage are the possible factors that control Mn mobility and bioavailability besides anthropogenic inputs (Heal 2001; Howe et al. 2005; WHO 2011). Mn does not occur naturally as a base metal but is a component of various minerals (WHO 2011). In the aquatic environment, Mn exists in two main forms: $Mn^{2+}$ and $Mn^{4+}$, while the solubility of $Mn^{2+}$ is higher than $Mn^{4+}$. Mn tends to become more bioavailable with decreasing pH and redox potential (Heal 2001; Eq. 1).

$$Mn^{2+} + 2HCO_3^- + H_2O + \frac{1}{2}O_2 \rightarrow Mn(OH)_4 + 2CO_2 \quad (1)$$

Considering the pH value of the Langat River, it ranged between 6.20 and 7.61. The acidic conditions observed at certain stations might have caused the release of Mn from Mn-bearing minerals or Fe–Mn oxide minerals. The guideline values for variables such as *Eh*, temperature, EC, salinity, DO, K, $Na^+$, $Cl^-$, and $HCO_3^-$, have not been established since they are not of health concern at levels found in drinking

water. However, some substances may affect the color, taste, odour or appearance of drinking water, which indirectly affect the acceptability of water for drinking and domestic purposes (WHO 2011). The application of these standards has shown that most of samples collected from the Langat River are suitable for domestic use, whereas only few samples were found to be unsuitable based on elevated contents of certain elements.

## *4.4 Suitability for Irrigation Uses*

The suitability of irrigation water depends upon the chemical constituents of water. Major ions such as $Ca^{2+}$, $Mg^{2+}$, $Na^{+}$, $Cl^{-}$, $HCO_3^{-}$ and $SO_4^{2-}$ are the main dissolved constituents which determine the suitability of water for irrigation purposes. Several indicators and indices including sodium percentage, magnesium hazard, Kelly's ratio and Wilcox diagram were employed to evaluate the usefulness of river water for irrigation (US Salinity Laboratory Staff 1954; Wilcox 1955).

The sodium percentage was calculated and the quantities of all cations are expressed in meq/L (Eq. 2). Based on the sodium percentage present in the samples under study, the water can be categorized as excellent (<2 %), good (2–40 %), permissible (40–60 %), doubtful (60–80 %) and unsuitable (>80 %). As higher $Na^{+}$ increases the hardness and reduces its hydraulic conductivity of soil or permeability to water (Glover 1996; Jeevanandam et al. 2007), waters with high concentration of $Na^{+}$ are considered to be undesirable for irrigation purposes (Glover 1996). In the study area, the sodium percentage varied from 27 to 90 % and their classification varied from 'good' to 'unsuitable' classes (Fig. 5a). The magnesium hazard (MH) is used to evaluate the potential of $Mg^{2+}$ hazard to irrigation water where the value of magnesium hazard above 50 is considered as harmful and unsuitable for irrigation purposes (Szabolcs and Darab 1964; Eq. 3). From the calculated MH values, 57 % of the water samples (LY 1–LY 17) are classified as unsuitable for irrigation use (Fig. 5b). The Kelly's ratio is another index employed to identify the suitability of water for irrigation use. The Kelly's ratio is calculated by the level of $Na^{+}$ measured against $Ca^{2+}$ and $Mg^{2+}$ (Kelly 1951; Eq. 4). From the calculated value, 65 % of water samples are classified as unsuitable for irrigation due to the excess level of $Na^{+}$ in the water, where the Kelly's ratio were above unity (Fig. 5c). The rest of samples were classified as good quality for irrigation as these fall along the equiline or below the unity.

$$\text{Sodium percentage} = \frac{[Na^{+}]}{\left([Ca^{2+}] + [Mg^{2+}] + [K^{+}] + [Na^{+}]\right)} \times 100 \tag{2}$$

$$\text{Magnesium hazard} = \frac{[Mg^{2+}]}{[Ca^{2+}] + [Mg^{2+}]} \times 100 \tag{3}$$

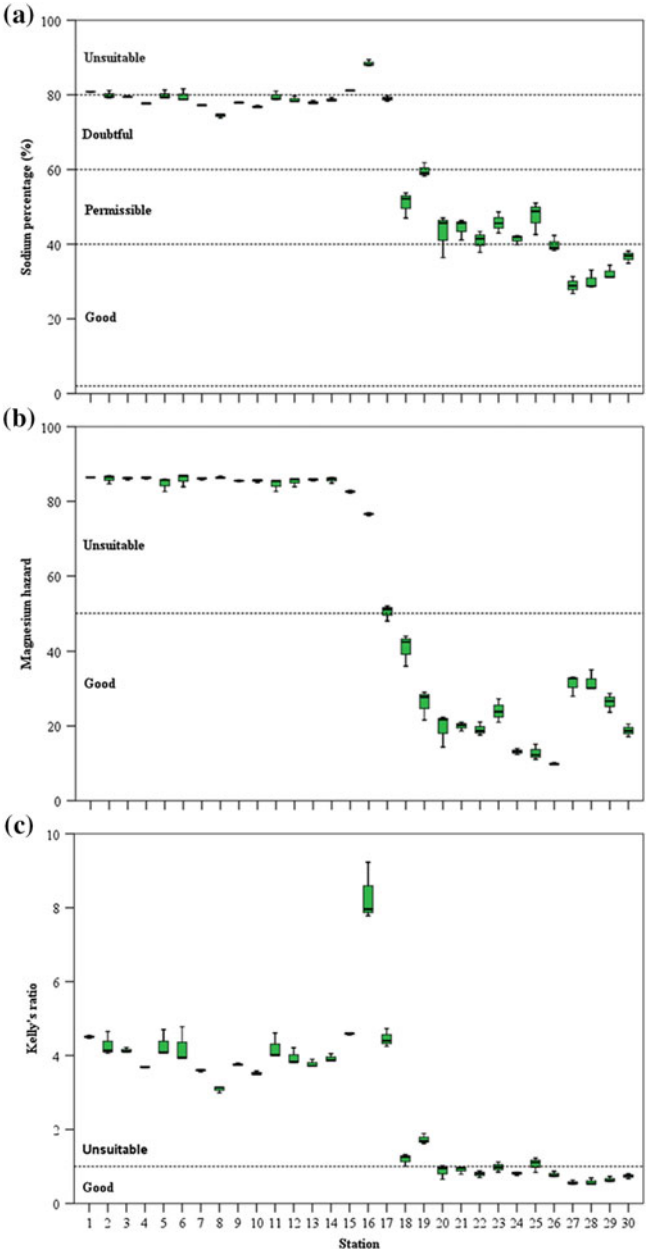


**Fig. 5** **a** Sodium percentage, **b** magnesium hazard and **c** Kelly's ratio for overall study location

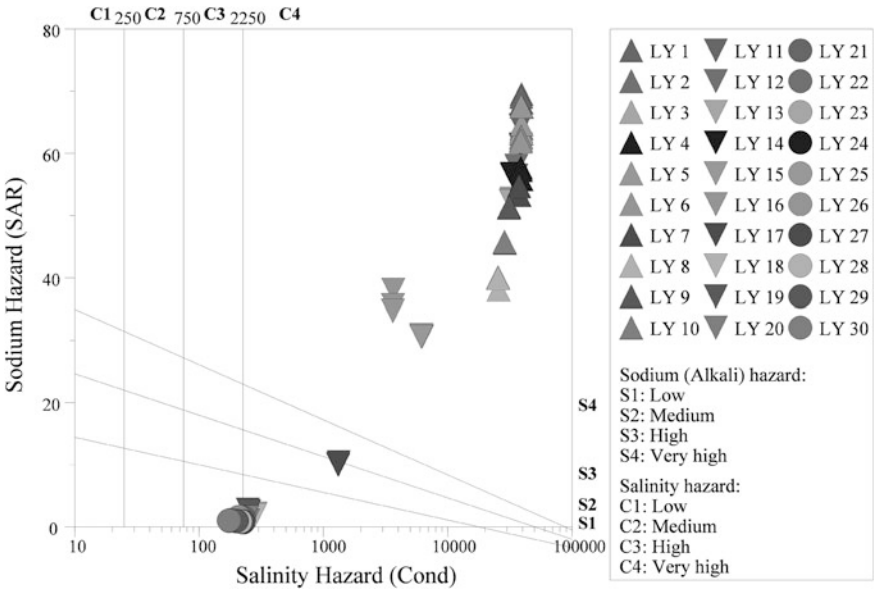


**Fig. 6** Wilcox diagram of irrigation water accordingly to its sampling stations

$$\text{Kelly's ratio} = \frac{[Na^+]}{[Ca^{2+}] + [Mg^{2+}]} \tag{4}$$

$$\text{SAR} = \frac{[Na^+]}{\sqrt{\frac{1}{2}\left([Ca^{2+}] + [Mg^{2+}]\right)}} \tag{5}$$

Wilcox's diagram comprises the sodium adsorption ratio (SAR, or known as sodium hazard) and salinity hazard (with respect to EC). The SAR is the ratio of the $Na^+$ to the combination of $Ca^{2+}$ and $Mg^{2+}$ in relation to the recognized effects on soil dispersibility (Eq. 5). It measures the degree to which cation exchange between $Na^+$ with $Ca^{2+}$ or $Mg^{2+}$ can occur in the soil, which influence the soil structure (Glover 1996; Radojević and Bashkin 2007). Based on the SAR value, the water can be categorized into four classes as S1 (<10), S2 (10–18), S3 (18–26) and S4 (>26). For salinity hazard, water with EC value less than 250 µS/cm is considered as low salinity water (C1), 250–750 µS/cm as medium salinity water (C2), 750–2,250 µS/cm as high salinity water (C3) and above 2,250 µS/cm as very high salinity water (C4). The Wilcox diagram (Fig. 6) shows that 53 % of samples (LY 1–LY 16) were unsuitable for irrigation purposes, while the rest of the samples can be used for irrigation on most crops in most soils. High SAR and salinity hazard for water is inappropriate for irrigation purpose as this will reduce the osmotic activity of plants and restrict the roots of plants from absorbing water from the soil (Hiscock 2005).

## 5 Conclusions

- The PCA apportioned that the major sources controlling the hydrochemistry of the Langat River are from both natural and anthropogenic activities.
- Employing various ratios suggested that weathering in the catchments and seawater intrusion contribute to the hydrochemistry of the river.
- Present study found that selected water samples were unsuitable for drinking purposes directly without treatment because their water quality variables such as pH, TDS, $SO_4^{2-}$, $Na^+$, $Mg^{2+}$, As, or Mn values were beyond the permissible limits suggested by MOH and WHO.
- Based on different indicators and indices calculated, it was found that the water samples collected from sampling stations close to coastal area were not suitable for irrigation.
- The Langat River faces pollution from multiple sources, primarily associated with agriculture, forming, ship traffic and manufacturing units. These activities also reintroduce pollutants such as Mn into the river water by resuspending channel-bed sediments and accelerated erosion of flood-plain regions. A basin-scale multi-disciplinary study based restoration scheme has to be undertaken in this basin.

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