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Evaluation of groundwater in aluminium slag disposal area

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Abstract: Aluminium slag waste is a residue from aluminium recycling activities, classified as hazardous waste so its disposal into the environment without processing can cause environmental problems, including groundwater pollution. There are 90 illegal dumping areas for aluminium slag waste spread in the Sumobito District, Jombang Regency. This study aims to evaluate the quality of shallow groundwater surrounding aluminium slag disposal in the Sumobito District for drinking water. The methods applied an integrated water quality index (*WQI*) and heavy metal pollution index (*HPI*), multivariate analysis (principal component analysis (PCA) and hierarchical clustering analysis (HCA)), and geospatial analysis for assessing groundwater quality. The field campaign conducted 40 groundwater samples of the dug wells for measuring the groundwater level and 30 of them were analysed for the chemical contents. The results showed that some locations exceeded the quality standards for total dissolved solids (*TDS*), electrical conductivity (*EC*), and Al^{2+}. The *WQI* shows that 7% of dug well samples are in poor drinking water condition, 73% are in good condition, and 20% are in excellent condition. The level of heavy metal contamination based on *HPI* is below the standard limit, but 13.3% of the water samples are classified as high contamination. The multivariate analysis shows that anthropogenic factors and natural sources/geogenic factors contributed to shallow groundwater quality in the study area. The geospatial map shows that the distribution of poor groundwater quality is in the northern area, following the direction of groundwater flow, and is a downstream area of aluminium slag waste contaminants.

Keywords: aluminium slag waste, geospatial analysis, heavy metal pollution index, multivariate analysis, water quality index

INTRODUCTION

Aluminium is the third most abundant element in the Earth's crust and is the second most used metal after iron (Tsakiridis, 2012). Due to its lightweight, aluminium is widely used as a raw material for aircraft manufacture, architectural construction, the marine industry, electronic devices, and various household appliances. Aluminium is produced in two ways, the primary production process and secondary production. Primary production is carried out by extracting alumina from bauxite ore. Meanwhile, secondary production or aluminium recycling is carried out by purifying aluminium from primary production process waste and aluminium scrap from used drink cans, foil and metal scraps to be reprocessed into aluminium ingots (Xiao *et al.*, 2005; Tsakiridis, 2012).

The primary production process will produce a residue of primary aluminium dross or white dross in the form of blocks with a high aluminium content so that it can still be extracted by smelting. Meanwhile, the secondary production process or aluminium recycling will produce secondary aluminium dross or black dross residue in the form of granules with lower aluminium content, which is also known as salt cake or salt slag or aluminium slag (Tsakiridis, 2012; Mahinroosta and Allahverdi, 2018; Shen *et al.*, 2021). In the production process of one ton of secondary aluminium, the residual slag from the smelting process will reach 200–500 kg, depending on the mixture of raw materials (Tsakiridis, 2012). The aluminium slag from aluminium smelting still contains 5–30% of aluminium oxide, 30–55% of sodium chloride, 15–30% of potassium chloride, 5–7% of aluminium metal, oxides of alloying elements (Si, Cu, Fe, Zn, etc.) and impurities of raw materials (carbides, nitrides, sulfides, and phosphides) (Tsakiridis, 2012).

Aluminium slag is classified as hazardous waste because of its high toxic concentration, flammability, irritant with detrimental effects on human skin and organs, and its tendency to produce leachate in water (Huang *et al.*, 2014; Samara *et al.*, 2020). Furthermore, aluminium slag waste in contact with water and air tends to produce harmful gases such as phosphine, hydrogen sulfide, ammonia, and methane which are explosive, toxic, and have a terrible smell. In addition, the dissolution of dross waste and aluminium slag in water results in the release of many elements or heavy metal components in the waste, potentially contaminating surface water and groundwater (Shinzato and Hypolito, 2016; Samara *et al.*, 2020).

The aluminium recycling industry in Sumobito District, Jombang Regency, East Java, has been operating since 1970 (Jombangkab, 2015). However, the large volume of waste generated and the enormous costs required to manage waste trigger illegal waste dumping activities. There are 90 locations for dumping aluminium slag waste in the Sumobito District, which local people use as river embankments and road-filling materials without any processing. Toxicity characteristic leaching procedure (TCLP) test and total concentration (TK) test on sediments showed high concentrations of heavy metals Al^{3+}, Zn^{2+} and Cu^{2+} (TK-A); Pb^{2+} (TK-B) (Ministry of Environment and Forestry, 2019). Meanwhile, most of the population in the Sumobito District uses groundwater, either hand-dug or deep wells, as the primary source of freshwater, including drinking water (BPS, 2021). Therefore, it is necessary to evaluate the groundwater quality of the residents around the location to ensure it is safe to use.

The water quality can be assessed and evaluated through the water quality indices method, which is a single number that indicates water quality overall at a given location based on the combination of several individual water quality parameters with a given calculation method (Vasanthavigar *et al.*, 2010; Tiwari *et al.*, 2015). Its values provide information on water quality status and water adaptation for specific uses (drinking, bathing, irrigation, recreation, industry) (Islam *et al.*, 2018). It is effective in assessing water quality because it is relatively easy to use and can simplify a complex water quality data set into a measure of water quality (Lumb, Sharma and Bibeault, 2011). There are many models of indices, but appropriate indexes should be selected and applied depending on the complexity of the ecosystem, the type of pollutant source, and the objectives of the water quality monitoring activity (Calmuc *et al.*, 2020). In this study, the water quality indices method used is the *WQI*, and *HPI* for heavy metals is used to evaluate the groundwater quality suitability for drinking.

Several studies have integrated water quality indices with geographic information systems (GIS) to monitor groundwater quality status and visualise its distribution (Shanmugam and Velappan, 2015; Mahapatra *et al.*, 2020; Silva *et al.*, 2021). Both can be used to synthesise the various available water quality data into an easy-to-understand format, providing a way to summarise overall water quality conditions in a way that can be communicated to policymakers (Singh *et al.*, 2014–2015). Several studies also applied multivariate statistics as a follow-up analysis of water quality assessment. Multivariate statistical techniques such as principal component analysis (PCA) and hierarchical clustering analysis (HCA) can be helpful tools for identifying potential pollutant pathways and sources of heavy metals and physicochemical variables (Boateng, Opoku and Akoto, 2019; Amano *et al.*, 2021).

The main objective of this study is to evaluate the physicochemical parameters of groundwater, such as pH, total dissolved solids (*TDS*), electrical conductivity (*EC*), and several heavy metals such as aluminium (Al^{2+}), lead (Pb^{2+}), cooper (Cu^{2+}), and zinc (Zn^{2+}) in Sumobito District, especially around the slag aluminium dumping sites. In addition, the study also generates an assessment of shallow groundwater quality and its use for drinking purposes. The *WQI* and *HPI* are applied to evaluate shallow groundwater's status and heavy metal pollution levels. To make the results more robust geospatial analysis and multivariate statistical techniques (PCA and HCA) are applied to develop distribution models of water quality and identify probable sources of pollutants that contribute to groundwater quality. This study will become material consideration for policymakers in making policies related to handling contaminated land and managing groundwater contamination by identifying the primary sources of pollution and their distribution.

MATERIALS AND METHODS

STUDY AREA

The study area is located in Sumobito District, a part of Jombang Regency in East Java Province, Indonesia, which is geographically located between 112°16'0"–112°23'0" E and 7°29'0"–7°34'0" S (Fig. 1a). It has an administrative area of 47.64 km^2 comprising 21 village with a population density of 1,813 people per square kilometer. The altitude ranges between 22 and 42 meters above sea level (m a.s.l.), with higher areas in the south and descending to the north. The average precipitation recorded is between 1,400 and 1,900 $mm \cdot y^{-1}$, mainly observed during the monsoon season (November–April). The alluvium is the primary geological formation in the study area (Fig. 1b) (Santosa and Atmawinata, 1992). Soil type is dominated by alluvial soil at 74% in the middle and northern part of the study area, while the rest is Andosol soil at 26%, which is spread in the southern region. As shown in Figure 1a, land use in the study area is dominated by rice fields at 63.03%, settlements at 22.63%, mixed gardens at 9.41%, and the rest are sugarcane plantations, fields/moorlands with secondary crops, rivers, and another open land. Moreover following Nuzulliyantoro *et al.* (2020), the study area has a high potential for groundwater potency with a discharge of 5 $dm^3 \cdot s^{-1}$ (Fig. 1c).

The location of aluminium slag dumping spread in the central and northern parts of the study area, which are located in the residential areas, the rice fields, and the riverbanks.

SAMPLE COLLECTION AND ANALYTICAL PROCEDURE

A purposive sampling design was chosen, with the sampling point location as shown in Figure 1a. The field campaign for hydrogeological mapping was carried out on 40 dug wells, representing the shallow groundwater in the study area. Measurements were carried out at the beginning of the rainy season, November 2021. The measurements included recording the coordinates of shallow wells, recording soil elevation, and measuring groundwater depth. While 30 of them were analysed the chemical contents were taken using glass bottles and then collected in containers made of high-density polyethene with a volume of 2 dm^3. The containers had previously been washed

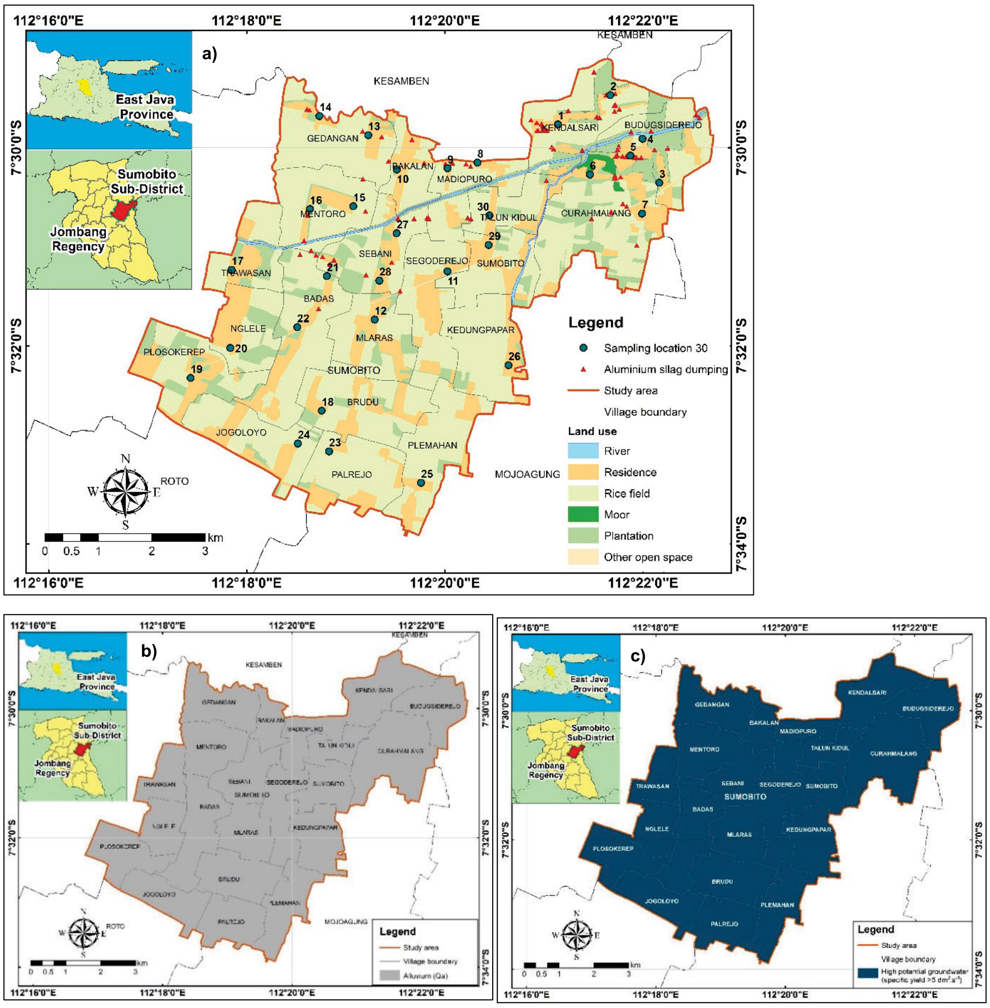


Fig. 1. Map of the study area in Sumobito District: a) dug wells' sampling site locations and land use, b) regional geology, c) potential of groundwater; source: own elaboration based on Santosa and Atmawinata (1992), and Nuzulliyantoro *et al.* (2020)

with detergent, rinsed with clean water, rinsed with 1:1 HNO_3, rinsed three times with distilled water, and dried. Preservation of samples was carried out by adding HNO_3 to pH < 2 to minimise adsorption on the container's walls and changes in the metal content of the samples (Baird and Bridgewater, 2017). Heavy metals such as Al^{2+}, Pb^{2+}, Cu^{2+}, and Zn^{2+} were determined in the laboratory using atomic absorption spectrophotometer following the procedures described in "Standard Methods for the Examination of Water and Wastewater" (Baird and Bridgewater, 2017). During sampling in the sampling sites, we measured pH, total dissolved solids (*TDS*), and electrical conductivity (*EC*) by using a portable pH meter (Eutech pH Test 20), TDS meter (Eutech TN100), and conductivity meter (HACH Sension 5), respectively.

THE WATER QUALITY INDEX

The water quality index (*WQI*) aims to reduce a large number of water quality parameter measurements to a single value indicating the ecological status of a particular watercourse. The general algorithm for calculating the *WQI* consists of converting all parameter values to a common scale (sub-index) and combining them into one final value (index) (Calmuc *et al.*, 2020). There are two dominant *WQI* calculation models used to assess groundwater quality and evaluate the feasibility of drinking water, including the weighted arithmetic of *WQI* (Rahman *et al.*, 2020; Zakir *et al.*, 2020) and the weighted of *WQI* (Vasanthavigar *et al.*, 2010; Putranto and Ginting, 2020; Amano *et al.*, 2021; Iwar, Utsev and Hassan, 2021). The weighted arithmetic of *WQI* applied the weighting based on the maximum permissible concentration standard so that the *WQI* value will be strongly influenced by water quality parameters with a low maximum permissible concentration, such as heavy metals (Calmuc *et al.*, 2020). Meanwhile, the weighted *WQI* assigned a weighted number of 1–5, which was determined by considering the level of importance of water quality and the health risks.

The *WQI* weighted model was carried out to evaluate the water quality in the study area. The water quality parameters used for determining the *WQI* were a combination of physicochemical parameters, including heavy metals, adapting the research of Iwar, Utsev and Hassan (2021), namely *TDS*, *EC*, pH, Al^{2+}, Pb^{2+}, Cu^{2+}, and Zn^{2+}. Meanwhile, the weighted *WQI* applied a weighted number of 1–5, which was determined by considering the level of importance of water quality and the health risks. Therefore, the *WQI* weighted model will be applied to evaluate the water quality in the study area, using the formulation which was originally proposed by Horton (1965) and applied by some researchers (Xiao *et al.*, 2014; Tavassoli and Mohammadi, 2017; Adimalla, 2020):

$$q_i = \frac{C_i}{S_i} 100 \quad (1)$$

$$W_i = \frac{w_i}{\sum_{i=1}^{n} w_i} \quad (2)$$

$$WQI = \sum_{i=1}^{n} q_i \cdot W_i \quad (3)$$

where: q_i = the quality rating scale for each parameter i; C_i = the concentration of each parameter based on the measurement results; S_i = the standard or quality standard used, uses the Indonesian Ministry of Health Regulation concerning drinking water quality requirements, except the *EC*, which uses the WHO guideline value (WHO, 2011); W_i = the relative weight of each parameter; w_i = the weight of each parameter, a value between 1–5; n = the number of parameter.

For example, the heavy metal parameters Al^{2+}, Pb^{2+}, Cu^{2+}, and Zn^{2+} were assigned a weight of 5, the *TDS* parameter was set at a value of 4, while the pH and *EC* parameters were given a weight of 3. The formula for the relative weight (W_i) used is listed in Table 1. The *WQI* values that have been obtained were then classified into five classes referring to the classification of Vasanthavigar *et al.* (2010), namely excellent (<50), good (50–100), poor (100–200), very poor (200–300), and unfeasible for drinking water (>300).

Table 1. Relative weight formula for the water quality index

Parameter	Weight (w_i)	Relative weight (W_i)
pH	3	0.10
TDS	4	0.13
EC	3	0.10
Al^{3+}	5	0.17
Cu^{2+}	5	0.17
Pb^{2+}	5	0.17
Zn^{2+}	5	0.17
Σ	**30**	**1.00**

Explanations: *TDS* = total dissolved solids, *EC* = electrical conductivity.
Source: own elaboration based on Iwar, Utsev and Hassan (2021).

THE HEAVY METAL POLLUTION INDEX

The heavy metal pollution index (*HPI*) was employed to evaluate water quality based on heavy metal parameters for drinking water. In this study, the parameter used for the *HPI* calculation was heavy metals i.e. Al^{2+}, Pb^{2+}, Cu^{2+}, and Zn^{2+}. The *HPI* was then calculated by the weighted arithmetic method adopted from the research of Mohan, Nithila and Reddy (1996) and Zakir *et al.* (2020). The *HPI* calculation uses the following equation:

$$HPI = \frac{\sum_{i=1}^{n} (Q_i \cdot W_i)}{\sum_{i=1}^{n} W_i} \quad (4)$$

$$W_i = \frac{k}{S_i} \quad (5)$$

$$k = \frac{1}{\sum \frac{1}{S_i}} \quad (6)$$

$$Q_i = \sum_{i=1}^{n} \frac{|M_i - I_i|}{(S_i - I_i)} 100 \quad (7)$$

where: W_i = the relative weight, Q_i = the sub-index for heavy metal parameters, k = the proportionality constant, S_i = the maximum permissible concentration according to the standard, M_i = the heavy metal concentration of the analysis results, I_i = the ideal value or maximum desired value of heavy metal parameters for drinking water.

In this study I_i value for all heavy metals' parameters was assumed to be zero. In this study, the formula for the relative weight (W_i) for each of the heavy metal parameters used was calculated using Equation (5) and Equation (6) to obtain the weight formula as listed in Table 2. The results of the calculation of the *HPI* value were then classified into three classes of contamination levels referring to Zakir *et al.* (2020), <20 was low, 20–30 and >30 were medium to high, respectively.

Table 2. Relative weight formula used for the heavy metal pollution index

Parameter	Standard (S_i; mg·dm^{-3})	Relative weight (W_i)
Al^{3+}	0.20	0.047
Cu^{2+}	2.00	0.005
Pb^{2+}	0.01	0.945
Zn^{2+}	3.00	0.003
Σ	–	**1**

Explanations: S_i = standard of Ministry of Health of the Republic of Indonesia.
Source: own elaboration based on Regulation (2010) and Zakir *et al.* (2020).

MULTIVARIATE STATISTICS

Multivariate statistics were applied to identify the relationship between heavy metals and physicochemical parameters in the study area. The application of multivariate statistical techniques facilitates the interpretation of complex data matrices to understand better various environmental factors (Rezaei *et al.*, 2019). Principal component analysis (PCA) was used to identify the sources of pollutants and the most contributing factors to shallow groundwater quality at the study site (Boateng, Opoku and Akoto, 2019). In this study, Kaiser–Meyer–Olkin (KMO) and Bartlett's tests were used to test the feasibility of PCA. A significant p-value (<0.05) and KMO value (>0.5) could be accepted as suitable for PCA (Boateng, Opoku and Akoto, 2019). The number of components was determined based on the eigenvalue. Components with eigenvalues >1 were retained. Meanwhile, hierarchical clustering analysis (HCA) was used to identify water samples with similar quality or content groups, where groupings in the same cluster may have the same source (Khadija *et al.*, 2021). The clustering analysis was accomplished by Ward's linkage with squared Euclidean distance. PCA with varimax normalised rotation and HCA were performed using the statistical software SPSS version 25.0.

GEOSPATIAL ANALYSIS

The water quality data that have been obtained were then spatially extracted and analysed using ArcGIS version 10.4. The interpolation method was used to estimate values at locations where data were unavailable by identifying spatial patterns and interpolation values at unallocated locations (Arslan, 2012). Interpolation techniques were performed by a Geostatistical Analyst on ArcGIS software. The interpolation was used with several deterministic and geostatistical methods and then compared for visualising the spatial distribution. The deterministic interpolation techniques depicted the surfaces from a measured point, either based on the level of equivalence (e.g., inverse distance weighted (*IDW*)) or the level of smoothing (e.g., radial basis functions (*RBF*)). Hartmann, Krois and Waske (2018) stated that the *IDW* approach was an inverse distance-based weighted interpolation for estimating the value z at location x, it was a weighted mean of nearby observations. To expect a cost for any unmeasured area, *IDW* made use of the measured values surrounding the prediction area. The measured values closest to the prediction area had a greater impact on the expected cost than the ones further away. *IDW* assumed that every measured factor had a nearby impact that diminishes with distance. It offered more weights to factors closest to the prediction area, and the weights decrease as a feature of distance, consequently, the call inverse distance was weighted. The *RBF* was an interpolation whose value was determined solely by the distance from the origin. In practice, the function must only contain real values. The distance from another point was applied to define alternative forms of *RBF*, while geostatistical interpolation techniques (e.g., ordinary kriging (OK) and empirical Bayesian kriging (EBK)) employed statistical properties from measured points (Elubid *et al.*, 2019). Due to the limited sample size, the cross-validation to determine the most accurate interpolation techniques was performed with leave-one-out cross-validation (LOOCV) (Mirzaei and Sakizadeh, 2016). The best interpolation technique was selected based on the accuracy level of data interpolation determined by root mean square error (*RMSE*) values, where the smaller the *RMSE* values, the better the interpolation (Simpson and Wu, 2014; Putranto and Alexander, 2017). This *RSME* value was known using the Geostatistical Wizard.

RESULTS AND DISCUSSION

GROUNDWATER FLOW DIRECTION

Based on the field campaign, the shallow groundwater table at the study site ranges from 23.6 to 31.9 m a.s.l. with an average height of 26.9 m a.s.l. The distribution of shallow groundwater levels is the highest on the south side of the Palrejo Village. It decreases towards the lowest side in the north and northeast, namely in the Gedangan Village and Budugsidorejo Village (Fig. 2). The height difference can trigger groundwater movement, forming a shallow groundwater flow pattern from the south to the north and northeast. The groundwater flow pattern's direction follows the study site's topography, which tends to be lower towards the north.

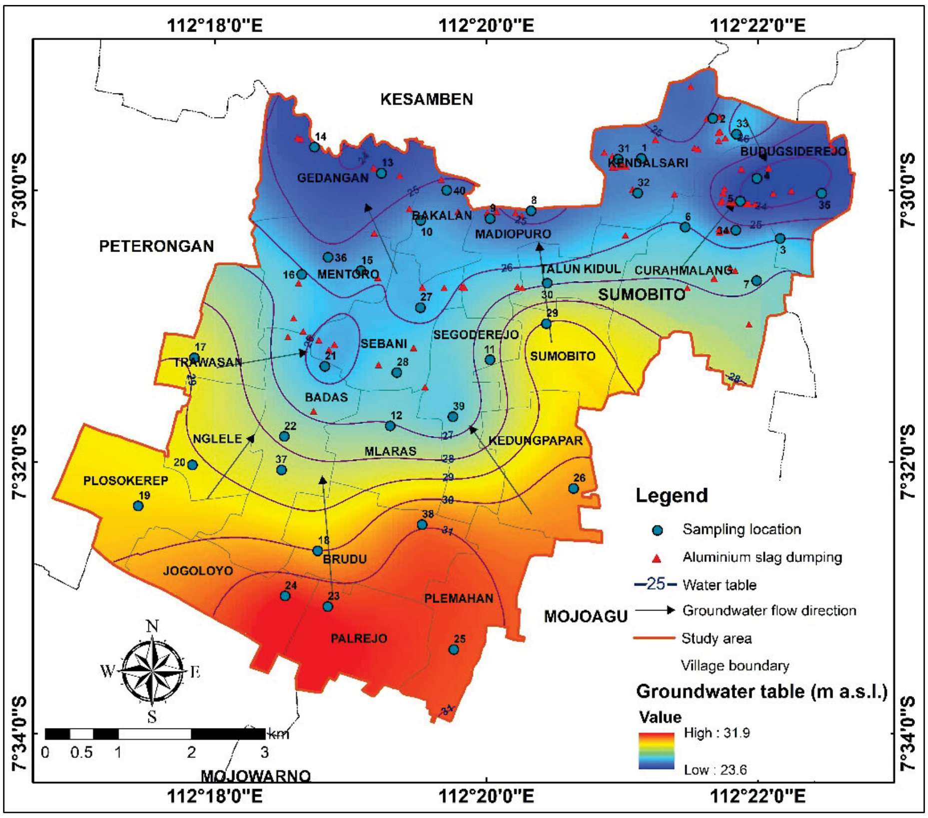


Fig. 2. Map of groundwater table and its flow direction; source: own study

CHARACTERISTICS OF GROUNDWATER QUALITY

Descriptive statistics of physicochemical parameters and heavy metals concentration of groundwater samples consist of minimum, maximum, and mean values of each parameter, which are compared with the drinking water standards limit, and are summarised in Table 3.

Meanwhile, Figure 3 shows a box plot illustrating the groundwater quality's statistical parameters.

In-situ measurements in the study area show that the pH values of groundwater samples have a range of 6.8–7.36, with an average value of 7.03, indicating neutral. The pH value is below the drinking water quality standard (6.5–8.5). The *TDS* values obtain 271–1,040 mg·dm^{-3} with an average concentration of 538.7 mg·dm^{-3}.

The highest *TDS* concentration is found at sample ID 13 in Gedangan Village. Compared with the standard limit for drinking water, only 56.7% of the well water samples met the required quality standard for the *TDS* (<500 mg·dm^{-3}). The high *TDS* content in water will not be lost through the boiling process. Inorganic minerals that settle in the body for a long time can cause disturbances in various channels in the body, triggering the emergence of diseases such as kidney stones (Setioningrum, Sulistyorini and Rahayu, 2020).

Meanwhile, the *EC* value of water samples at the study site is 416–1,390 μS·cm^{-1}, with an average value of 729.1 μS·cm^{-1}. The highest *EC* concentration was also found in sample ID 13. Compared with the drinking water standard value, around 70% of samples are still below the standards. Consumption of water with high *EC* can cause health problems, including disturbances in salt

Table 3. Statistical summary of groundwater quality data and comparison with standards limit for drinking water

Parameter	Min.	Max.	Mean	Standard limit[1)]	Sum of the sample exceeding the standard limit	Sample exceeding the standard limit
pH	6.8	7.36	7.03	6.5–8.5	0 (0%)	–
TDS (mg·dm^{-3})	271	1,140	538.7	500	13 (43.3%)	1–4, 6, 8–9, 13–14, 21–22, 25, 27
EC (μS·cm^{-1})	416	1,390	729.1	750	8 (30%)	4, 8–9, 13–14, 21, 27–28
Cu^{2+} (mg·dm^{-3})	0.001	0.038	0.013	2	0 (0%)	–
Zn^{2+} (mg·dm^{-3})	0.001	0.024	0.006	3	0 (0%)	–
Pb^{2+} (μg·dm^{-3})	0.100	5.449	0.966	10	0 (0%)	–
Al^{3+} (mg·dm^{-3})	0.102	0.837	0.328	0.2	23 (76.7%)	2–10, 12–16, 19–23, 26–28, 30

[1)] Standard value acc. to Ministry of Health of the Republic of Indonesia, excluding *EC* value (acc. to WHO).
Explanations: *TDS* = total dissolved solids, *EC* = electrical conductivity.
Source: own study based on Regulation (2010), WHO (2011).

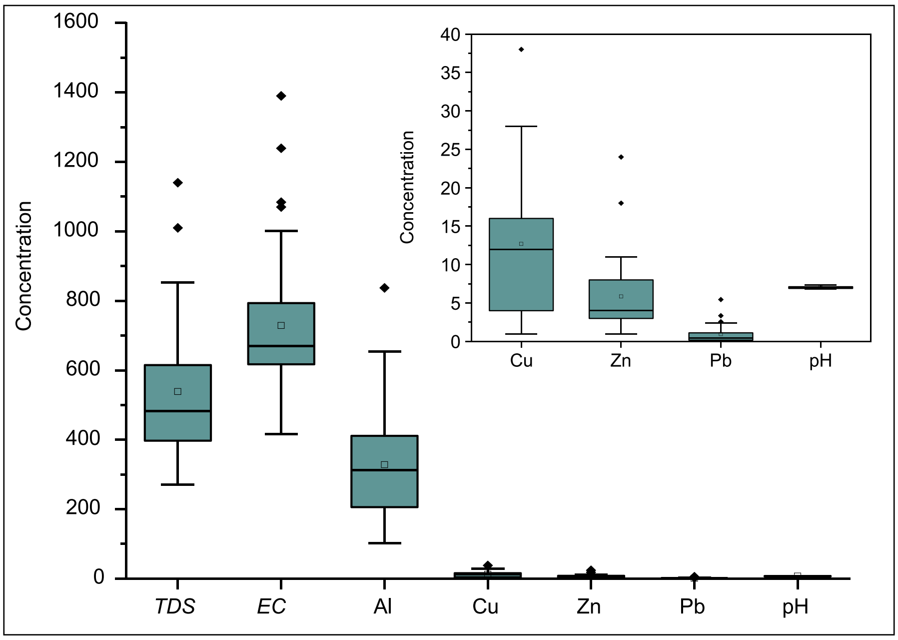


Fig. 3. Boxplot of shallow groundwater quality at Sumobito District; parameter units: *TDS* = mg·dm^{-3}; *EC* = μS·cm^{-1}; Al, Cu, Zn, Pb = μg·dm^{-3}; source: own study

and water balance, negative effects on heart patients, individuals with high blood pressure, individuals with kidney disease, and a laxative effect due to increased sulfate concentrations (Fatoki and Awofolu, 2003).

The dissolved heavy metals parameter results in the groundwater samples show the concentrations of Cu^{2+}, Zn^{2+}, Pb^{2+}, and Al^{3+}, which are (0.001–0.038] mg·dm^{-3}, (0.001–0.024] mg·dm^{-3}, (0.1–5.45] μg·dm^{-3}, and [0.102–0.837] mg·dm^{-3}, respectively. The average concentration of each parameter of heavy metals is 0.013 mg·dm^{-3} for Cu^{2+}, 0.006 mg·dm^{-3} for Zn^{2+}, 0.966 μg·dm^{-3} for Pb^{2+}, and 0.331 mg·dm^{-3} for Al^{3+}. Referring to the drinking water standard, all samples are below the drinking water quality standard for Pb^{2+}, Cu^{2+}, and Zn^{2+} parameters. Only one dug well sample with a Pb^{2+} concentration of 5.45 μg·dm^{-3}, closest to the maximum permissible level, is located in Budugsidorejo Village. In contrast to the Al^{3+} parameter, only 23.3% of the samples meet the required drinking water quality standard of 0.2 mg·dm^{-3}. The highest Al^{3+} concentration is 0.84 mg·dm^{-3}, also found in sample ID 5 in Budugsidorejo Village, 25 m from the dumping site for aluminium slag waste. Al^{3+} in drinking water with a concentration of >0.2 mg·dm^{-3} can cause health problems (WHO, 2011). High content of Al^{3+} in water causes neurodegenerative disorders and bone osteomalacia. In addition, Al^{3+} accumulation is associated with Alzheimer's disease (Bignucolo *et al.*, 2012).

When aluminium slag waste is dumped on the ground without any protection and exposed to rain, the components in the effluent will react with the water. This reaction can cause many components in the waste to be extracted and dissolved in water, including heavy metals. The contaminant components in the waste, along with water, will enter the soil pores through the infiltration mechanism and then move through the soil layer through the percolation mechanism and enter the dug wells through the flow of water in the soil. The composition of aluminium slag waste mostly consists of aluminium, sodium, and chloride (Tsakiridis, 2012). Moreover, the addition of salt flux, which contains sodium chloride (NaCl) and potassium chloride (KCl), in the aluminium recycling process to increase the extraction of aluminium from dross results in high chlorides, thereby increasing the concentration of inorganic ions in groundwater and increasing the *EC* value in groundwater (Shinzato and Hipolito, 2016; Attia, Hassan and Hassan, 2018). Meanwhile, the inorganic salt content in the aluminium slag waste, which dissolved in water, is estimated to have contributed to the increase in the *TDS* value at the study site.

The shallow groundwater quality found in this study is better than in the research by Shinzato and Hypolito (2016). They also evaluated the quality of monitoring well water in the dross waste storage area in the aluminium recycling industry in Sao Paolo, Brazil, where the pH of groundwater was found to tend to be low acid (pH 4) with an average concentration of Al^{3+} 15.75 mg·dm^{-3}, Pb^{2+} 0.36 mg·dm^{-3}, Cu^{2+} 0.14 mg·dm^{-3} and Zn^{2+} 0.5 mg·dm^{-3}. The different results can be influenced by the distance between the well and the contaminants, the concentration of contaminants, the contaminant dumping age, and the rock and soil conditions at the study site. In addition, the time of sampling, which was carried out in November, which is the rainy season, could also contribute. Research by Singh *et al.* (2014–2015), Mahapatra *et al.* (2020), and Iwar, Utsev and Hassan (2021) showed that the concentration of heavy metals in the dry season was found to be higher than during the rainy season. The decrease in relative concentrations of metals from summer to the rainy season is due to high rainfall, seepage, and groundwater recharge, which not only dilutes metals but also aids in their migration (Singh *et al.*, 2014–2015).

EVALUATION OF WATER QUALITY BASED ON INDICES

Table 4 shows the water quality index (*WQI*) value of the dug wells in the study area ranging from 38.20 to 110.31, with an average of 63.03. The highest *WQI* value is found in sample ID 5, which is 25 m from the dumping location of aluminium slag waste. Referring to the *WQI* classification for drinking water, according to Vasanthavigar *et al.* (2010), 7% of dug well water

Table 4. Evaluation of groundwater quality based on the water quality index (*WQI*) and heavy metal pollution index (*HPI*)

Sample ID	Village	Distance from dumping area (m)	*WQI*	Water class	*HPI*	Degree of pollution
1	Kendalsari	230	50.04	good	6.05	low
2	Kendalsari	78	63.58	good	9.31	low
3	Budugsidorejo	475	71.36	good	11.91	low
4	Budugsidorejo	150	105.36	poor	49.77	high
5	Budugsidorejo	25	107.11	poor	71.26	high
6	Curahmalang	470	55.18	good	7.57	low
7	Curahmalang	318	62.10	good	30.83	high
8	Madiopuro	145	85.15	good	43.91	high
9	Madiopuro	90	83.82	good	21.56	low
10	Bakalan	220	73.04	good	21.87	medium
11	Segodorejo	970	40.95	excellent	4.93	low
12	Mlaras	700	51.78	good	9.64	low
13	Gedangan	125	75.77	good	5.92	low
14	Gedangan	30	72.98	good	8.97	low

cont. Tab. 4

Sample ID	Village	Distance from dumping area (m)	*WQI*	Water class	*HPI*	Degree of pollution
15	Menturo	250	50.31	good	7.07	low
16	Menturo	127	56.54	good	8.50	low
17	Trawasan	1,280	38.20	excellent	5.16	low
18	Brudu	1,790	40.40	excellent	8.10	low
19	Plosokerep	2,692	54.51	good	15.62	medium
20	Nglele	1,905	52.06	good	15.79	medium
21	Badas	230	90.10	good	28.79	medium
22	Badas	543	64.26	good	26.66	medium
23	Palrejo	2,523	43.45	excellent	7.52	low
24	Jogoloyo	2,752	38.49	excellent	4.22	low
25	Plemahan	3,290	47.18	good	9.60	low
26	Kedungpapar	589	69.16	good	28.43	medium
27	Sebani	280	71.47	good	14.02	low
28	Sebani	270	77.36	good	14.69	low
29	Sumobito	600	45.42	excellent	4.89	low
30	Talun Kidul	351	50.30	good	13.86	low
Mean			63.03	–	16.88	–
Minimum			38.20	–	4.22	–
Maximum			110.31	–	71.26	–

Source: own study.

samples in the study area are in poor condition for drinking water, 73% of samples are in good condition, while the remaining 20% of samples are in excellent condition.

The *HPI* calculations range from 4.22 to 71.26, averaging 16.88 (Tab. 5). The highest *HPI* value was also found in sample ID 5. The *HPI* values for all samples below the critical limit of 100 (Mohan, Nithila and Reddy, 1996). Based on the results of the *HPI* calculations and the classification levels of heavy metal contamination referring to Zakir *et al*. (2020), 13.3% of dug well water samples are classified as high pollution levels, 20% of samples were medium, and 66.7% are low.

Table 5. Groundwater classification based on the water quality index (*WQI*) and heavy metal pollution index (*HPI*) for drinking water

Index method	Category	Degree of pollution/ water class	Number of samples	Distribu-tion (%)
WQI	<50	excellent	6	20
	(50–100]	good	22	73
	(100–200]	poor	2	7
	(200–300]	very poor	–	–
	≥300	unfeasible for drinking	–	–
HPI	<15	low	20	66.7
	(15–30]	medium	6	20
	(30–71]	high	4	13.3

Source: own study.

Poor groundwater quality and high contamination levels are found in Budugsidorejo Village, Madiopuro Village, and Curahmalang Village, with a radius of <500 m from the waste disposal point. The results of mapping the groundwater level and the direction of groundwater flow indicate that the groundwater level in these three areas is lower than in other areas following the area's topography and is located downstream of the dumping location for aluminium slag waste. As a result, surface runoff and groundwater flow through percolation may accumulate in that area, so it becomes more contaminated than others.

STATISTICAL ANALYSIS

Understanding the main factors controlling hydrochemistry and groundwater quality is critical for sustainable groundwater management (Rezaei *et al*., 2019). In this study, PCA analysis was used to identify the main factors or sources of pollution that contributed to the water quality of dug wells at the study site. In this study, the Kaiser–Meyer–Olkin (KMO) value test result was higher than 0.5 (KMO = 0.585), and the results of the Barlett test show that the correlation matrix is significant (sig. 0.000 < 0.05). Thus, PCA can be used to analyse the data set. The results of extracting the water quality data set obtain three components with an eigenvalue >1, which could explain 82.44% of the total variance (Tab. 6). Liu *et al*. (2003) classify component loadings according to absolute loading values as strong (<0.75), moderate (0.75–0.50), and weak (0.50–0.30). Varimax rotation results show that the component explains 40.13% of the total variance consisting of heavy metals Pb^{2+} and Al^{3+} with strong positive loading values and pH with moderate negative loading values.

Principal component in 2nd grup (PC2) explains 25.05% of the total variance consisting of *TDS* and *EC* with a strong loading value. Component 3 explains 17.25% of the total variance consisting of Cu^{2+} and Zn^{2+} with a strong positive loading value.

Table 6. Principal component analysis (Varimax rotated with Kaiser normalisation)

Parameter		Component		
		1	2	3
pH		**–0.665**	0.176	–0.277
TDS		–0.129	**0.963**	0.003
EC		–0.073	**0.961**	–0.113
Cu		0.041	–0.286	**0.876**
Zn		0.282	0.170	**0.837**
Pb		**0.940**	–0.076	0.065
Al		**0.880**	–0.016	0.080
Eigenvalue		2.81	1.75	1.21
Variance	%	40.13	25.05	17.25
Cumulative	%	40.13	65.18	82.44

Explanations: *TDS* = total dissolved solids; *EC* = electrical conductivity; values in bold indicate that the parameter affects the component in that group.
Source: own study.

The presence of the components in component 1 is estimated from anthropogenic activities and geogenic factors (rock and soil mineral dissolution). High loading of Al may be due to slag aluminium dumping site leachate and geogenic factors (rock and soil mineral dissolution). It is because the distribution of Al^{3+} that exceeds the quality standard is also found in areas quite far from the slag waste dumping site. According to Buragohain, Bhuyan and Sarma (2010), Al^{3+} in groundwater can be sourced from industrial waste, dissolving clay elements, and alumino-silicate minerals in soil and rock. Meanwhile, Pb^{2+} in groundwater can generally be sourced from industrial metal waste, batteries, paints, and leachate disposal from landfills (Boateng, Opoku and Akoto, 2019). Component 2 is estimated to be derived from anthropogenic activities, namely the aluminium slag dumping and domestic waste runoff. According to Rezaei *et al*. (2019), high *TDS* values in groundwater can be caused by industrial waste disposal, domestic household waste, percolation of canal water containing solids and agricultural waste, runoff from the soil, and weathering of rocks. Component 3 is estimated to be derived from anthropogenic activities, namely agricultural activities. Fertilisers and agricultural chemicals are a source of Cu^{2+} and Zn^{2+} in groundwater (Rezaei *et al*., 2019). It is possible because the land use in the study area is dominated by rice fields (63.03%).

Moreover, HCA is used to classify 7 parameters into 4 clusters, as presented in Figure 4. Cluster 1 consists of *TDS* and *EC*, cluster 2 only consists of pH, cluster 3 consists of Pb^{2+} and Al^{3+}, and cluster 4 consists of Cu^{2+} and Zn^{2+}. These results mostly agreed with the PCA.

SPATIAL DISTRIBUTION MAP

Water quality data and indices are then processed into a map representing the spatial distribution of groundwater quality and the suitability of shallow groundwater for drinking. Several interpolation methods are used and compared to get the best distribution model, as shown in the distribution map in Figure 5. Based on the comparison shown in Table 7, it conducts the radial

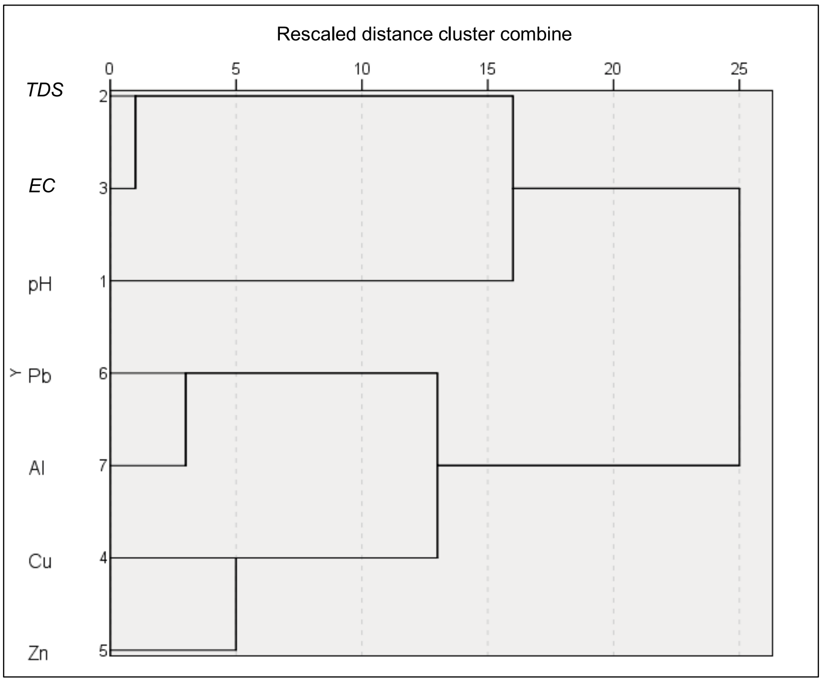
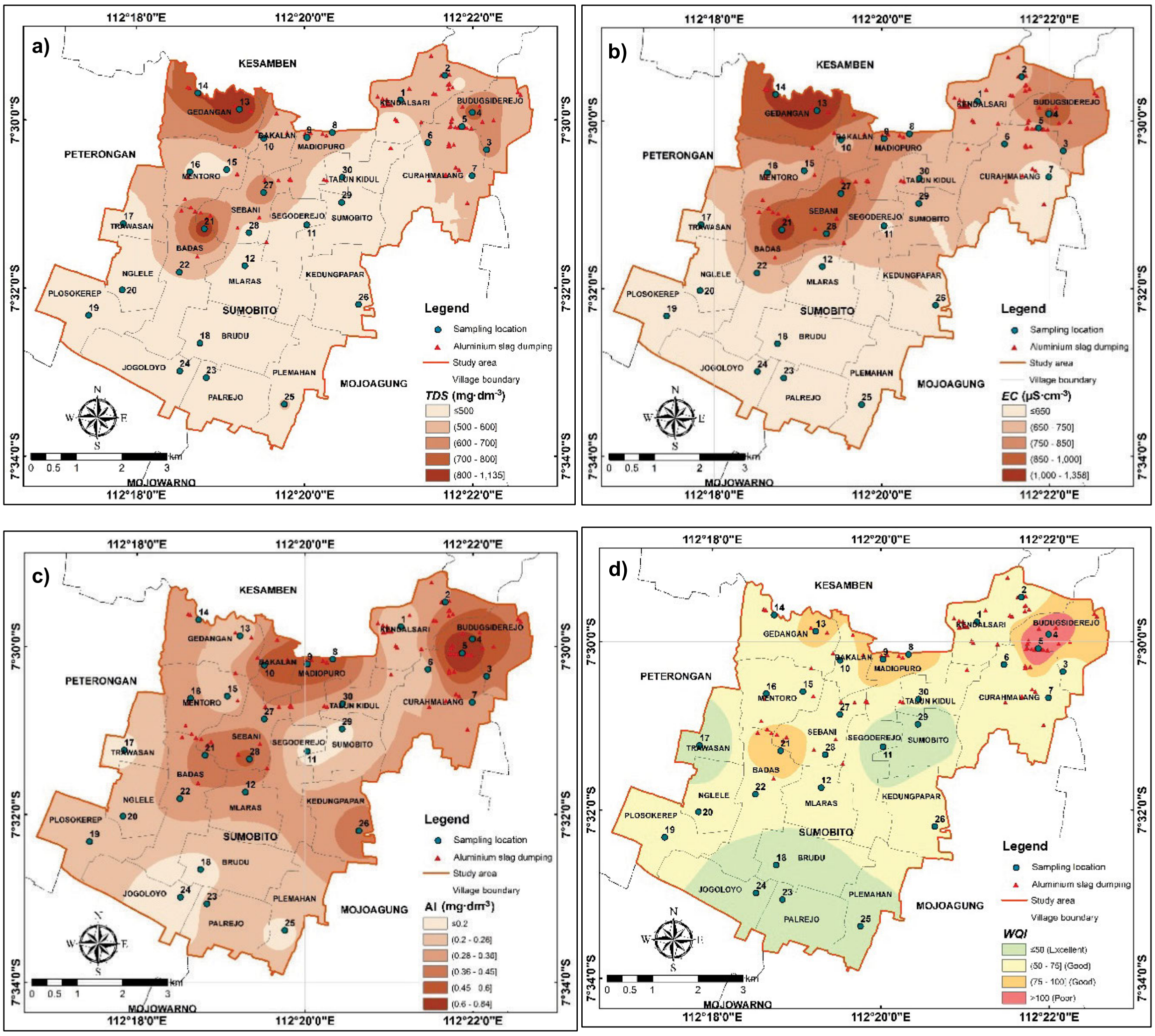


Fig. 4. Dendogram cluster analysis using Ward's linkage; *TDS* = total dissolved solids, *EC* = electrical conductivity; source: own study



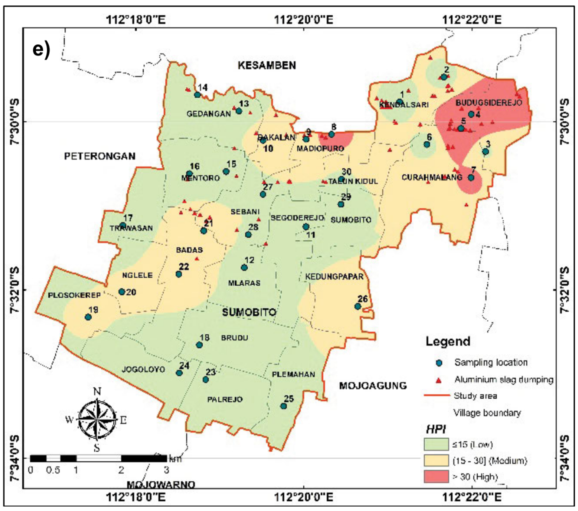


Fig. 5. Spatial distribution maps based on inverse distance weighted (*IDW*) at Sumobito District for: a) total dissolved solids (*TDS*) and radial basis functions (*RBF*), b) electrical conductivity (*EC*), c) Al^{3+}, d) water quality index (*WQI*), e) heavy metal pollution index (*HPI*); source: own study

basis functions (*RBF*) produced a better model (lower *RMSE* value) than other methods, except for the parameter *TDS*. The empirical Bayesian kriging (EBK) method produces a smaller *RMSE* value for the *TDS* parameter, but the minimum and maximum values produced are quite far from the actual value. Thus, the inverse distance weighted (*IDW*) method is applied with the second smallest *RMSE* value but produces a range of values close to the actual value in the field, as presented in Figure 5. The result is in line with the research of Arslan and Turan (2015), which showed that the *IDW* and *RBF* interpolation methods show better results for mapping heavy metals in groundwater than the ordinary kriging (OK) method. In addition, research by Putranto and Alexander (2017) also indicates that the *RBF* interpolation method is the most balanced compared to the *IDW* and EBK methods in mapping the electrical conductivity (*EC*) value in groundwater basins. However, these results are different from research by Gunarathna, Kumari and Nirmanee (2016), which shows that the EBK method provided better results

Table 7. Comparison of interpolation results for water quality mapping

Parameter		Interpolation method			
		IDW	*RBF*	OK	EBK
TDS ($mg \cdot dm^{-3}$)	*RMSE*	177.23	179.44	177.90	177.03
	value	270.9–1,139.8	272.3–1,129.3	420.2–645.7	432.6–634.5
EC ($\mu S \cdot cm^{-1}$)	*RMSE*	204.42	201.93	206.75	203.42
	value	416–1,389.7	409.5–1,397.4	486.3–1,203.4	591.9–870.2
Al^{3+} ($mg \cdot dm^{-3}$)	*RMSE*	0,156	0.149	0.158	0.157
	value	0.102–0.834	0.097–0.854	0.184–0.615	0.219–0.475
WQI	*RMSE*	13.99	13.12	14.56	14.67
	value	38.49–107.05	37.88–111.89	47.24–86.85	49.6–78.15
HPI	*RMSE*	14.34	14.05	14.56	14.57
	value	4.25–70.95	3.98–71.52	8.38–38.44	11.77–33.18

Explanations: OK = ordinary kriging, EBK = empirical Bayesian kriging, *RMSE* = root mean square error, *EC*, *TDS*, *RBF*, *IDW*, *WQI*, *HPI* as in Fig. 5.
Source: own study.

for mapping pH in groundwater in Sri Lanka than the *IDW* and *RBF* methods. The difference in the best interpolation method can be influenced by the number of samples, the distance between the sampling locations, and the sample density (Mirzaei and Sakizadeh, 2016).

CONCLUSIONS

The investigation of 30 groundwater samples around the disposal of aluminium slag waste in Sumobito District showed that at some points, the dug wells exceeded the drinking water quality standards for the *TDS* parameter by 43.7% of samples, *EC* – by 30% of samples, and Al^{3+} – by 67.7% of samples. Meanwhile, for the Cu^{2+}, Pb^{2+}, and Zn^{2+} parameters, all dug well water samples are within the drinking water quality standards. The evaluation results based on the *WQI* for drinking water show that 7% of the dug well samples are in poor condition, and 73% are in good condition. The remaining 20% of the samples were in excellent condition. Meanwhile, based on the *HPI*, 13.3% of dug well samples are classified as having high contamination levels but had not yet reached the standard limit.

Multivariate statistical analysis indicates that three main factors influenced the decline in groundwater quality in the study area. The first factor entails Al, Pb, and pH estimated to be derived from a combination of anthropogenic factors, namely the disposal of aluminium slag waste and natural/geogenic factors (dissolution of soil and rock). The second factor entails *EC* and *TDS*, which are thought to have originated from anthropogenic factors, namely the disposal of aluminium slag waste and domestic waste runoff. While the third factor entails Cu^{2+} and Zn^{2+}, which are thought to be derived from anthropogenic factors, namely agricultural activities.

The deterministic interpolation methods, *IDW* and *RBF*, are validated as the most suitable methods for the modelling of groundwater quality distribution in the study area. The mapping of the *WQI* results shows poor conditions and high levels of metal contamination in the northern part, namely Budugsidorejo Village, Madiopuro Village, and Curahmalang Village, in the direction of the groundwater flow pattern that leads from the south to the north. Hence aluminium slag waste cleaning up and contaminated soil remediation are immediately necessary, especially in those three areas. However, utilising dug wells in those areas for consumption is unrecommended, except with additional processing. To obtain a more comprehensive overview of shallow groundwater quality in the study area, further research is needed on the quality of dug wells in the dry season, the major anion-cations parameters and other heavy metals.

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