Hydrogeology And Hydrochemistry Of Shallow Groundwater And Surface Water And Potential Water Pollution In Gadung District, Buol, Central Sulawesi

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Abstract. Inhabitants of Gadung District depend on Labuton River and shallow groundwater as their water resources. But issue of river pollution caused by gold mining activities operated for decades around upstream area of Labuton River becomes challenge for their water demands. In the other hand, shallow groundwater from dug wells has low resistance for contaminants from urban activities around downstream area. This study aims to analyze characteristics of hydrochemistry and to identify pollution cases in research area. In order to recognize flow path of shallow groundwater water table in research area have been measured. EC, pH, and TDS from water samples (13 samples from Labuton River and 16 samples from groundwater) has been measured. Na⁺, K⁺, Ca²⁺, Mg²⁺, F⁻, Cl⁻, NO₃⁻, SO₄²⁻ concentrations of the samples measured using Ion Chromatograph. HCO₃⁻ concentrations of the samples measured using Alkalinity Checker. Hydrochemistry Labuton River which is dominantly associated with Dolokapa Formation is controlled by rock weathering process. It is indicated by TDS 50 - 200 mg/L and total cationic charge 428.7 - 2973.8 μeq/L. Labuton River and shallow groundwater in research area has hydrofacies Mg+HCO₃ type generally, but Mg+SO₄ type is shown along ± 5.2 km of water flow of Labuton River from mining area to the downstream zone which is categorized as MIW (Mining Influenced Water). MIW in Labuton River has high concentration of SO₄²⁻ (9.27 to 35.12 - 36.03 mg/L), high TDS value (100 to 140 - 160 mg/L), and low pH (6.3 to 5.4 - 5.6) from its original water. Labuton River water is polluted by contaminant from mining activity along ± 2.8 km water flow indicated by pH 5.4 - 5.6. Groundwater in some part of study area is polluted by urban contaminant indicated by concentration NO₃⁻ > 3 mg/L (3.33 - 19.43 mg/L).

1. Introduction

Gadung is a district part of Buol region located in Central Sulawesi Province. Its territory covers 160,38 km² with 11,789 inhabitants [1]. Inhabitants are heavily dependent on Labuton River and shallow groundwater to meet their water needs. However, the challenge they face is the issue of river water pollution due to gold mining activities operated since 1896 around upstream zone of Labuton River [2][3]. Meanwhile, shallow groundwater is vulnerable to pollution caused by contaminants from anthropogenic and agricultural activities [4–6]. For groundwater study, study area relies on citizen’s well which is concentrated in coastal area, northern part of Gadung. Bulagidun Watershed, eastern part of Gadung, which is associates with Labuton River is study area selected for surface water study. Study areas devide conveniently in order to analyze characteristics of hydrochemistry and to identify pollution cases in research area which is the purposes of this study. It will be the first report.

Study area covered by Diorit Boliohuto, Dolokapa Formation, Breksi Wobudu, Lokididi Formation, and Alluvium (see Figure 1). Diorit boliohuto, Middle Miocene - Late Miocene, consists
diorite, granodiorite. Dolokapa Formation, Middle Miocene - Late Miocene, consists greywacke, siltstone, mudstone, conglomerate, tuff, lapili tuff, agglomerate, volcanic breccia, andesitic to basaltic lava, and schist [7][8]. Dolokapa Formation thickness is ± 1000-2000 m. Breccia Wobudu, Middle Pliocene - Late Pliocene, consists volcanic breccia, agglomerate, tuff, lapili tuff and lava, with andesitic to basaltic composition. Breccia Wobudu thickness is ± 1000 m. Lokodidi Formation, Late Pliocene - Early Pleistocene, consists conglomerate, sandstone, conglomeratic sandstone, tuffaceous sandstone, tuff, claystone, black shale. Alluvium, Holocene, consists sand, clay, silt, mud, gravel and pebble. Its thickness is ± 120 m [8].

Groundwater and productivity of aquifers in study area occur, i.e, (1) aquifer which flow is intergranular, locally, moderately productive aquifers which is mostly incoherent aquifers of low thickness and transmissivity, wells yield < 5 L/sec, (2) aquifer (fissured or porous) of poor productivity, generally low transmissivity; locally limited shallow groundwater can be obtained in the valleys and weathered zones, and (3) regions without exploitable groundwater (see Figure 1) [7].

Climate of the study area classified as tropical rain forest climate which has $T_{min} > 27 \, ^\circ C$, $P_{min} > 126.77$ mm, and annual precipitation $P_{ann} 1800 - 2600$ mm [9–11]. Land cover of the research area consists of primary dryland forests, secondary dryland forests, primary mangrove forests, shrubs, dryland agriculture, swamps, rice fields, settlements and roads (see Figure 2) [12–14].
Land use can be in the form of settlements and agriculture that can produce contaminants from anthropogenic and agricultural activities such as nitrate (NO$_3^-$) as a nitrification product that can degrade the quantity and/or quality of groundwater [15–17]. Studies of NO$_3^-$ and its distribution as contaminants from agricultural and urban activities is common [4,6]. The concentration of NO$_3^-$ in groundwater represents natural water background with concentration <0.2 mg/L, transitional representation between the presence or absence of anthropogenic influences with concentration 0.21 - 3.0 mg/L, and anthropogenic effects with concentration 3.1 - 10 mg/L (Zapata et al., 2014).

Labuton River might be contaminated by ARD (Acid Rock Drainage) from mining activity operating around upstream area. Water contaminated by mining activity or MIW (Mining-Influenced Water) has higher TDS, higher SO$_4^{2-}$ concentration, and lower pH than its original water [17–20]. Lower pH and higher SO$_4^{2-}$ concentrations are attributed to the oxidation of pyrite minerals (FeS$_2$) from high sulphide-containing rocks [18]. If water has > 400 mg/L concentration of SO$_4^{2-}$ or < 6 pH value, it is classified as polluted [21,22]. MIW is a particular case. MIW can be found with pH around 7 and SO$_4^{2-}$ concentration < 400 mg/L due to the activity of SRB (Sulphate-Reducing Bacteria) agents, but MIW may contain heavy metals, e.g., Pb, Cu, Zn, and Mn with concentrations exceeding the standard water quality (see Table 1) [23–26]

| Table 1. Concentration in mg/L of constituents in MIW |
|-------------------------------------------------------|
| pH          | Westfork       | Ferris Haggarty | MSF     | Rico     | Aquatic Standards |
|-------------|----------------|-----------------|---------|----------|-------------------|
| Mn (mg/l)   | 0.01           | 0.03            | 0.37    | 0.06     | 0.20              |
| Ni (mg/l)   | 0.05           | 7.5             | 0.01    | 0.20     |                   |
| Cu (mg/l)   | 0.02           | 20.6            | 0.015   | 0.01     |                   |
| Zn (mg/l)   | 0.21           | 0.07            | 0.20    | 11.4     | 0.10              |
| Cd (mg/l)   | 0.002          | 0.003           | 0.009   | 0.03     | 0.005             |
| Pb (mg/l)   | 0.7            |                 |         |          | 0.020             |
| SO$_4^{2-}$ (mg/l) | 63            | 48              | 1005   | 450      |                   |
| Alkalinity  | 156            |                 | 33     | 595      |                   |

In order to identify pollution cases, understanding of natural water background is needed. It can be understood through understanding of hydrofasies, hydrochemical characteristics of water, and predominant source-rock types. Gibbs classification and its diagrams modified by Gibbs in 1992 represent hydrochemical characteristics of water [17,27–30]. Based on dominant factors that affect the hydrochemical characteristics of river water including gibbs category, Stallard and Edmon, 1983, classified four type of water river and its predominant source-rock types [17]. Piper diagram represents groundwater and river water hydrofasies [17,31,32].

2. Data and Method

The data used consist of secondary data and primary data. Secondary data consist of geological map, hydrogeological map, watershed map, and land cover map. Primary data consist morphological map, water table measurements on 34 wells and 9 river points, and hydrochemical data from 13 Labuton River samples and 16 shallow groundwater samples. The collection and quality of water samples from the field of research to the Geochemistry Laboratory of Geological Engineering Department of Gadjah Mada University are based on the provisions of APHA, 1999 [33]. Hydrochemical data consist of pH, EC and TDS measured in the field using Hanna HI-98115, concentration of HCO$_3^-$ using alkalinity checker Hanna HI-775, ion concentration, i.e., Na$^+$, K$^+$, Ca$^{2+}$, Mg$^{2+}$, F$^-$, Cl$^-$, NO$_3^-$, SO$_4^{2-}$ in water measured using Metrohm 850 professional IC (Ion Chromatograph) with column; Metrosep A sup 7-250/4.0 (6.1006.630) for anion and Metrosep C.4-250/4.0 (6.1050.430) for kation. Hydrochemical data from IC processed by calculating BCE (Balance-Charge Error) (value< 5% not used), plotting data on piper diagram using Rockworks 15 software, calculating total cation and classifying river type based on river classification, plotting river’s data in Gibbs diagram, calculation of anhydrite and gypsum SI using PHREEQC 3.4 software, modeling the direction of groundwater flow, and
identifying the source and distribution of contaminants, analyzing of hydrochemical characteristics of river water and groundwater, analyzing the sources and distributions water contaminants to identify pollution cases in the study area.

3. Result and discussion
Labuton River has 80 - 200 mg/L TDS, 889 - 1538 μeq/L total cation, and 0.7 - 1.1 ratio of (Ca$^{2+}$ + Mg$^{2+}$) / (0.5 HCO$_3^-$ + SO$_4^{2-}$) (see Table 2). Based on Stallard and Edmon river classification, Labuton River is classified as type 3 river. It indicates that water chemical characteristics of Labuton River are predominantly controlled by the rock weathering activity that occurs in Bulagidun Basin. Furthermore, it defines also by ratio Na$^+$/Na$^+$/Ca$^{2+}$ and Cl$^-$/(Cl$^-$+HCO$_3^-$) in boomerang / gibbs diagram (see Figure 3). Predominant source-rocks in the basin are high pyrite-containing rocks.

Figure 3. Ploting hyrochemistry data of Labuton River in gibbs/boomerang diagram

Hydrofases of groundwater and Labuton River water are generally classified as Mg$^+$HCO$_3^-$ type except in Labuton River water from mining area to ± 4.1 km its flow direction (represented RL11 - RI7) which has hydrofases type Mg$^+$SO$_4$ (see Figure 4). Meteoric water is the main origin of HCO$_3^-$ in groundwater and Labuton River water. Mg$^{2+}$ cations in water might be originated from weathering rocks which contain high magnesium in its mineral constituents, e.g., olivine (forsterite / Mg$_2$SiO$_4$) minerals, pyroxene (clinoenstatite / MgSiO$_3$), and amphibole (anthophyllite / Mg$_7$Si$_8$O$_{22}$(OH)$_2$ and tremolite / Ca$_2$Mg$_5$Si$_8$O$_{22}$(OH)$_2$). The weathering reaction of these minerals is written as follows:

\[
\begin{align*}
\text{Mg}_2\text{SiO}_4\text{(forsterite)} + 4\text{H}^+ & \rightarrow 2\text{Mg}^{2+} + 2\text{H}_2\text{O} + \text{SiO}_2 \\
\text{MgSiO}_3\text{(clinoenstatite)} + 2\text{H}^+ & \rightarrow \text{Mg}^{2+} + \text{H}_2\text{O} + \text{SiO}_2 \\
\text{Mg_7Si_8O_22(OH)_2\text{(anthophyllite)} + 14\text{H}^+} & \rightarrow 7\text{Mg}^{2+} + 8\text{H}_2\text{O} + 2\text{SiO}_2 \\
\text{Ca}_2\text{Mg}_5\text{Si}_8\text{O}_{22}\text{(OH)_2\text{(tremolite) + 14H}^+} & \rightarrow 5\text{Mg}^{2+} + 2\text{Ca}^{2+} + 8\text{H}_2\text{O} + 8\text{SiO}_2
\end{align*}
\]
These minerals have lower resistance to weathering process. Olivine minerals are generally associated with basaltic igneous rocks and metamorphic rocks such as schist. Acidic to alkaline igneous rocks contain pyroxene and amphibole. These rocks are found in Dolokapa Formation which is the oldest formation in the research area (Middle Miocene - Late Miocene) and covers most of the study area. High levels concentration of Labuton River water occurs around gold mining area. It is caused by mining activity involving exposure of sulphide-containing volcanic rocks followed by increasing oxidation of pyrite minerals which releases $\text{SO}_4^{2-}$ and $\text{H}^+$. 

Upstream Labuton River water along ± 2.8 km stream flow (from RL11 to RL9) has 5.4 - 5.9 pH (see Figure 6 and 7). It is caused by the high concentration of $\text{H}^+$ in water as a pyrite oxidation product due to mining activity. From upstream Labuton River around mining area to ± 5.2 km stream:

![Figure 5. Shallow Groundwater Flow Map in Gaung District, Buol Region, Central Sulawesi](image1)

![Figure 6. Land Cover Map on Bulagidun Watershade and Labuton River sampling point.](image2)
Figure 7. Labuton River's change of concentration SO$_4^{2-}$, SI$_{anhydrite}$, SI$_{gypsum}$, TDS, and pH diagram

Flow (RL11 - RL7), hydrochemistry of Labuton River sustains increasing concentration of SO$_4^{2-}$ up to 37.35 mg/L, decreasing pH up to 5.4, and increasing TDS up to 160 from its original water. Original water represents by RL13 which has 100 mg/L TDS, 6.3 pH, and 9.27 mg/L SO$_4^{2-}$ concentration. Labuton River water from RL11 to RL7 categorized as MIW (Mining Influenced Water). MIW has SO$_4^{2-}$ concentrations with range 35.12 - 36.03 mg/L, TDS 140 - 160 mg/L, and pH 5.4 - 6.2. The gold mining contaminant are gradually reduced from the mining site (around RL11 and RL10 area to RL5). Hydrochemistry of RL5 shows the similar characteristic of hydrochemistry with original water of Labuton River. RL5 has 100 mg/L TDS, 6.2 pH, and 9.86 mg/L SO$_4^{2-}$ concentration. RL5 does not show of mining effect based on pH and SO$_4^{2-}$ concentration but it is still possible that water stream still contains heavy metal above water quality standards. Intercalation of pH value can be caused by mixing between stream water and tributary water. Reduction of SO$_4^{2-}$ concentration can be caused by SBR activity. Intercalation of SO$_4^{2-}$ at RL4 - RL1 occurred by pyrite oxidation on rocks containing pyrite, e.g., volcanic rocks, high organic matter of sediment rocks and sediment material, that can be found along the Labuton stream through dryland agriculture, shrubs, and swamp areas. Fluctuation of SO$_4^{2-}$ concentration along Labuton Stream is similar with the fluctuation of SI$_{anhydrite}$ and SI$_{gypsum}$. SI$_{anhydrite}$ value is around -4.46 to -3.63 (See Figure 7). SI$_{gypsum}$ value is around 4.16 to -3.40. Anhydrite and Gypsum in Labuton River water are in under saturated condition (< 1). Cl$^-$ concentration in RL1 is higher than other sample it is because of sea water effect at estuary zone of Labuton River.

The original groundwater in study area represents by WL2. WL2 water lessened before its sample was taken. NO$_3^-$ concentration of WL2 is 0. For its ion major, WL2 has no dominant type between Mg$^{2+}$, Ca$^{2+}$, and Na$^+$+K$^+$ and its HCO$_3^-$ concentration most higher than the other wells (Figure 4). In groundwater from 16 wells at the study area, NO$_3^-$ was found to be at concentrations of 0 - 19.43 mg/L (see Table 2). Polluted groundwater is found in wells which have NO$_3^-> 3$ mg/L in its water, i.e., WD1.
(3.33 mg/L), WL1 (9.35 mg/L), WL7 (19.43 mg/L), WM3 (10.37 mg/L), WM5 (3.51 mg/L). Land covers surrounding wells are dryland agriculture and settlements. Most of dryland agriculture area are planted with annual crops, i.e. coconut, cacao, and coffee. Fertilizer is rarely used. So that, the main contaminant origin is contaminant from urban activity.

Based on groundwater map (see Figure 5), in the Matinan Watershed, groundwater flows from WM4 to WM3 through WM2. It tends to flow from the highest to the lowest terrain. In Bulagidun Watershed, groundwater flows from WM4 to WL8, from WL6 to WL7, from WL6 to WL4, from WL1 to WL2. Flow path of hydrochemical groundwater is too erratic. It does not have particular path which might be able to confirm that shallow groundwater in dug well easily to be affected by activity around. Groundwater flows from WL1 to WL2 which is ± 100 m apart but NO3− concentration in both wells are too contrast, i.e., 9.35 mg/L for WL1 and 1.62 mg/L for WL2. The contrast can be indicated that NO3− in wells are locally distributed and the contaminant is from urban activity around the well. Poor built well and poor waste water irrigation around the study area trigger the increasing nitrification process.

4. Conclusion
Based on the data analysis, the research has the following conclusions: Hydrochemical characteristics of Labuton River is controlled by rock weathering activity. Labuton River water and groundwater hydrofiasies are Mg++HCO3 except Labuton River water around mining area along ± 5.2 km of the flow which has Mg++SO4 type. Labuton River is polluted by contaminant from gold mining activities around upstream area along ± 2.8 km flow stream from mining location, indicated by 5.4 - 5.9 of pH. Labuton River categorized as MIW from mining location to ± 5.2 km flow stream. Groundwater is polluted by contaminant from urban activity in some areas indicated by the occurrence > 3 mg/L of NO3− concentrations.

Table 2. Hydrochemistry data of Labuton River and Groundwater

| No. Sample | Cation | Anion | EC N | pH | TDS | Water Temperature (°C) | Cation Total (mg/L) | [Ca++Mg]/(0.5HCO3+SO4) (mol/mol) | SI Activity | SI Gypsum |
|------------|--------|-------|------|----|-----|-------------------------|---------------------|-----------------------------------|-------------|----------|
| WL1        | 1.1    | 8.02  | 1.10 | 3.95 | 11.73 | 7.27 | 7.21 | 49.95 | 0.71 | 200 | 31.5 | 1338.15 | 3.1 | 3.4 |
| WL2        | 0.32  | 3.35  | 0.63 | 2.77 | 11.47 | 0.50 | 0.94 | 19.91 | 6.72 | 332 | 32.1 | 529.15 | 3.1 | 3.4 |
| WM1        | 3.31  | 1.03  | 2.42 | 11.44 | 0.42 | 0.17 | 17.04 | 53.64 | 6.98 | 100 | 30.9 | 1275.39 | 1.0 | 3.7 |
| WL4        | 1.36  | 0.83  | 1.09 | 10.74 | 0.42 | 0.12 | 12.68 | 54.91 | 6.93 | 120 | 30.7 | 1196.66 | 1.0 | 3.8 |
| WM3        | 1.34  | 0.71  | 1.05 | 10.58 | 0.51 | 0.90 | 56.77 | 1.1 | 100 | 30 | 1185.26 | 0.7 | 3.9 |
| WL5        | 1.61  | 0.90  | 1.76 | 8.24 | 0.39 | 0.29 | 20.21 | 24.47 | 6.12 | 90 | 20.7 | 928.18 | 1.0 | 3.6 |
| LM4        | 1.21  | 0.94  | 1.01 | 0.49 | 0.46 | 0.12 | 30.10 | 18.50 | 6.72 | 120 | 30.7 | 1185.26 | 1.0 | 3.9 |
| LM5        | 2.80  | 0.91  | 1.35 | 9.15 | 0.45 | 0.37 | 35.13 | 13.42 | 6.62 | 120 | 28.4 | 905.31 | 1.0 | 3.7 |
| RL9        | 3.47  | 1.20  | 1.65 | 10.49 | 0.46 | 0.08 | 64.54 | 1.22 | 11 | 160 | 27.9 | 880.75 | 1.0 | 3.5 |
| WL2        | 2.87  | 0.98  | 1.60 | 9.88 | 0.36 | 0.77 | 35.12 | 10.69 | 5.99 | 240 | 26 | 100.40 | 1.1 | 3.1 |
| RL10       | 2.10  | 0.91  | 1.40 | 9.86 | 0.36 | 0.77 | 35.12 | 10.69 | 5.99 | 240 | 26 | 100.40 | 1.1 | 3.1 |
| RL11       | 1.43  | 0.54  | 1.22 | 9.45 | 0.37 | 0.01 | 12.62 | 6.70 | 6.19 | 110 | 25.8 | 880.75 | 1.0 | 3.5 |
| RL12       | 2.45  | 1.04  | 0.64 | 5.86 | 0.63 | 0.03 | 34.92 | 1.22 | 15 | 100 | 25.8 | 880.75 | 1.0 | 3.5 |
| WI1        | 3.22  | 1.45  | 3.16 | 9.15 | 0.96 | 3.51 | 52.77 | 42.68 | 6.23 | 100 | 25.7 | 903.54 | 1.1 | 4.4 |

4.1 Shallow Groundwater

4.2 Wetland Groundwater

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