Aquifer connectivity assessment using stable isotope and hydrochemical analysis in the Raimanuk and its surrounding area on the Timor island

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Abstract. Raimanuk and its surroundings are a potential agricultural area in Indonesia. Sufficient groundwater in this area was a significant factor for supporting agricultural activities and household needs. Therefore, identifying the aquifer connectivity is essential to determine the groundwater utilization pattern in this area. This research aims to determine the aquifer connectivity based on stable isotopes and hydrochemical from the groundwater and the meteoric water (rainwater). The groundwater samples were taken from ten deep wells, six dug wells, and four springs. While the meteoric water samples were taken from four rainfed locations, samples were analyzed using Los Gatos Research DLT-100, Ion Chromatography Metrohm 830IC, and acid-base titration. The result shows there are four groups of groundwater. Group I is the springs that come from local meteoric water. Group II consists of the dug wells that evaporated and showed a relationship to the meteoric water that falls in higher elevations. Group III was the drilled well groundwater from the local flow system. Furthermore, Group IV is the drilled well groundwater with a more enriched δ 18O isotope ratio due to water-rock interaction. Even though they are in a different group, there is a hydraulic connection between Group III and IV.

Keywords: aquifer connectivity, stable isotope, hydrochemistry, Raimanuk Area

1. Introduction
Raimanuk and its surroundings are categorized as the excellent food security areas on Timor island, even in East Nusa Tenggara province of Indonesia [1]. The availability of water for irrigation is one of the significant factors to support agriculture activities in this area. With surface water, groundwater becomes the primary source of irrigation in the Raimanuk and its surrounding area. The community utilizes groundwater from drilled wells built by the government as a groundwater source for their agricultural activities. Groundwater for household needs is from dug wells and springs in several locations. The continuous groundwater pumping from drilled well for an extended period is feared can affect the groundwater in dug wells and springs. The study on the connectivity between these groundwater sources and how they affect each other needs to conduct as the primary analysis to determine the pattern of groundwater utilization in this area.

2. Location and Geological Setting
The study area is located on UTM coordinates (WGS 1984) Zone 51S lines 694936-713375 and 8963437-8975645, and it covers 146.352 km2 consisting of 7 sub-districts in 3 different regencies. The first regency is
Belu, with Raimanuk TasiFeto Barat and Nanaet Duabesi as the sub-districts. The second regency is Timur Tengah Utara with Biboki Utara and Biboki Tan Pah as the sub-districts. The third regency is Malaka, with Laenmanen and Malaka Timur as the sub-districts. The study area is a lowland on the west and hills on the east. The sedimentary rocks dominated the geological condition of the study area with a structure in the form of layers, laminates, and massive. The distribution of rocks is irregular, caused by the Bobonaro complex [2]; according to field investigation, the geology of the study area is divided into seven units (Figure 1). The unit is: The carbonate siltstone 1 unit, part of the pre-Perm Bisane Formation, this is the oldest unit in the study area [2] and is dominated by carbonate siltstone. In addition, crystalline limestone, carbonated claystone, and coral limestone were also seen as inserts. The crystalline limestone is part of the Perm to Triassic Maubisse Formation [2] and is dominated by crystalline limestone and wackestone. The carbonate siltstone 2 unit belongs to the Bobonaro Complex [2], a mixed rock with rock fragments up to the size of a hill on a scaly clay matrix [3]. The porphyry basalt unit. It is part of the Maubisse Formation but is interpreted as a fragment of the Bobonaro Complex. The general condition of the outcrop of this rock unit tends to be weathered, the composition in the form of plagioclase and mafic minerals. In some places, the rocks have been altered to appear green minerals, considered zeolite minerals. The coral limestones unit, part of the Tertiary-Quaternary Noele Formation [2], dominates by Coral limestones. In addition, wackestone limestone was also found. The wackestone unit is part of the Quaternary Coral Limestone [2]. Wackestone and coral limestone are dominated in this unit. The last unit is The Alluvium unit. This unit is spread in the western part of the study area, plain covering most of the lowlands area in the west, originates from the surrounding hills, which transport sand, gravel, and boulder-sized materials through rivers.

Figure 1. The study area and geological setting of The Raimanuk and its surrounding, Basemap taken from DEMNAS (BIG, 2018)

3. Theoretical Background
3.1. Isotope analysis
The isotopic ratio of oxygen (δ¹⁸O) and hydrogen (δD) on groundwater are powerful tools for understanding the isotopic fractionation and mixing processes of isotopic composition in different water bodies [4]. Furthermore, hydrogeology is used to determine the groundwater recharge area, predict flow direction, and the relationship between two or more different hydrogeological systems [6]. Isotope ratios vary in each region due to various conditions, such as temperature conditions, topography, rainfall, and the geographical location of an area [5,6,7]. The isotope ratio will increase (enriched) in line with the temperature increase in temperature effects. However, in terms of the
amount (rainfall) effect and altitude (elevation) effect, the isotope ratio will be decreased (Depleted) when the elevation and the rain are getting higher. Since the amount of rainfall, temperature, humidity varies, the values of $\delta^{18}O$ and $\deltaD$ at each rainwater sampling location must be averaged using the Equation [7,8]:

$$\delta_i \%o = \frac{\sum_{i=1}^{n} P_i \delta_i}{\sum_{i=1}^{n} P_i}$$  \hspace{1cm} (1)

Where $\delta$ is the mean isotopic ratio in meteoric water (rainwater), $\delta_i$ is the isotopic ratio in a meteoric water sample at a particular time (i), $P_i$ is the rainfall amount between sample $i-1$ and $i$. The average isotopic ratio is used to create the Local Meteoric Waterline (LMWL) graph. Later, the isotopic ratio from groundwater samples will be plotted to determine the relationship between the groundwater system and its origin.

3.2. Hydrochemical analysis

Groundwater hydrochemical analysis is carried out by measuring the amount of major ions content dissolved in groundwater. The major ions consist of Na+, K+, Ca+, Mg+ for the cations and $\text{HCO}_3^-$, $\text{Cl}^-$, $\text{SO}_4^{2-}$ for the anions. The major ions can represent the aquifer lithology and chemical process and determine connectivity on the groundwater system. Reaction Error (RE) calculation is usually performed as a check tool for hydrochemical analysis. RE is presented as a percentage of the total ion concentration. The Reaction Error equation is:

$$RE\% = \frac{\Sigma \text{Cation} - \Sigma \text{Anion}}{\Sigma \text{ion}}$$  \hspace{1cm} (2)

3.2.1. Groundwater facies. Piper trilinear diagrams (1944) are commonly used to determine the facies of groundwater. Groundwater facies describe lithology, solution kinetics, and the direction of groundwater flow [8]. The mechanism plots the points on the diagram according to major groundwater ions. Both anions and cations will be plotted separately on the triangular diagram on the left and right, then drawn perpendicular to the main diamond diagram in the middle [7,9], the classification of Piper Trilinear Diagram later modified by Furtak & Langguth (1965) as shown in Figure.2.

3.2.2. Groundwater connectivity. The Schoeller diagram is one of the hydrochemical methods to determine connectivity between groundwater systems (Figure.3). This is a semi-logarithmic diagram of the concentrations of the study area's groundwater samples. Each line is the compositional imprint of a water sample, and a similar pattern means a relatively similar water type and sources [7,10,11].

4. Methodology

4.1 Water sampling

Groundwater sampling for isotopic and hydrochemical analysis was collected from 10 Drilled Wells (SB), 6 Dug Wells (SG), and 4 Springs (MA). The meteoric water sampling for isotopic analysis was collected from 4 rainfed locations, each location representing the elevation from lowland to higher area in the eastern (Figure. 4). The SB groundwater sampling was conducted in the rainy period (March and April 2021), the same as the meteoric water sample. The SG and MA groundwater sampling was carried out in the dry period (July 2021) to avoid meteoric water mixing.

4.2 Sample analysis

The isotopic and hydrochemical analysis was conducted in the hydrology and geothermal lab, Center for Isotope and Radiation Application, National Nuclear Energy Agency – Jakarta. Analysis of stable isotopes was done using LGR (Los Gatos Research) DLT-100 Liquid Water Isotope Analyzer. Then two different ways carried out the hydrochemical analysis, acid-base titration for $\text{HCO}_3^-$ using HCl as titrant and Ion Chromatography Metrohm 830IC for $\text{Cl}^-$, $\text{SO}_4^{2-}$ and Na+, K+, Ca++, Mg++.
5. Result and discussion

5.1. Isotope Analysis

The result from isotope data from the laboratory (Table.1) was relative to Standard Mean Ocean Water (SMOW). The isotope data of meteoric water need to be averaged by considering the amount of rainfall each month when the sample is taken using Equation (1). The average meteoric water isotope data is plotted into a graph. We get the Local Meteoric Waterline (LMWL) equation from the graph as shown in Equation.3. this LMWL named as Raimanuk Meteoric Water Line (RMWL)
Table 1. Groundwater and meteoric water sampling location in the study area

| No. | Sampling Location | Elevation (masl) | δ¹⁸O (‰) | δD (‰) | Rainfall (mm) | Average |
|-----|------------------|-----------------|----------|--------|--------------|---------|
|     |                  | March 2021      | April 2021 | March 2021 | April 2021 | March 2021 | April 2021 | δ¹⁸O | δD |
| 1   | TH-01            | 746             | -7.43     | -7.12   | -47.40      | -47.20   | 252        | 263.5 | -7.27 | -47.30 |
| 2   | TH-02            | 668             | -6.72     | -6.84   | -45.60      | -43.80   | 252        | 263.5 | -6.78 | -44.68 |
| 3   | TH-03            | 417             | -5.01     | -6.39   | -30.90      | -40.50   | 252        | 263.5 | -5.72 | -35.81 |
| 4   | TH-04            | 352             | -4.89     | -6.24   | -30.80      | -35.90   | 252        | 263.5 | -5.58 | -33.41 |

δD = 8.1257 δ¹⁸O + 11.195  \quad (3)

R² = 0.9867

The groundwater isotope data (Table 3) was then plotted to the RMWL Graph (Figure 5) to determine the relationship between groundwater and meteoric water. Furthermore, the groundwater origin could be defined by using Equation (4) and Equation (5) obtained from the diagram of the relationship between isotope values and elevation (Figure 6 and Figure 7).

Table 2. Groundwater and meteoric water sampling location in the study area

| Sampling Location | Elevation (masl) | δ¹⁸O (‰) | δD (‰) |
|------------------|-----------------|----------|--------|
| SB-1             | 411             | -5.02    | -34.40 |
| SB-2             | 401             | -5.77    | -36.90 |
| SB-3             | 375             | -4.05    | -23.80 |
| SB-4             | 372             | -5.68    | -36.30 |
| SB-5             | 353             | -5.68    | -35.60 |
| SB-6             | 338             | -5.28    | -34.40 |
| SB-7             | 330             | -5.38    | -33.90 |
| SB-8             | 368             | -5.15    | -34.20 |
| SB-9             | 356             | -5.34    | -34.70 |
| SB-10            | 363             | -5.80    | -36.50 |
| SG-1             | 369             | -3.68    | -24.90 |
| SG-2             | 388             | -4.52    | -28.30 |
| SG-3             | 348             | -4.30    | -28.00 |
| SG-4             | 365             | -4.41    | -26.90 |
| SG-6             | 343             | -4.08    | -26.80 |
| SG-7             | 369             | -4.06    | -27.80 |
| MA-1             | 676             | -4.84    | -28.50 |
| MA-2             | 534             | -4.74    | -27.90 |
| MA-3             | 392             | -4.45    | -26.30 |
| MA-4             | 413             | -5.25    | -30.00 |

Elevation = -230.71 δ¹⁸O - 916.39  \quad (4)

R² = 0.9893

Elevation = -230.71 δ¹⁸O - 916.39  \quad (5)

R² = 0.9893
The graph shows there is at least four groundwater system. Group I is the groundwater in the springs (MA-1, MA-2, MA-3, MA-4) that are attached to the RMWL, which indicated that the groundwater in springs is from local meteoric water that falls on their elevation [4, 8,13] therefore, there should be no hydraulic connectivity between the springs. Group II is the groundwater attached to the evaporation line. This condition indicates that the groundwater in this group is affected by the evaporation during flowing in the unsaturated zone or indicating surface water influence [8]. The intersect between the evaporation line with RMWL shows the original composition of the groundwater on this group is slightly more depleted than the MA-1 isotopic ratio, and it shows the possibility of groundwater origin is coming from around MA-1 or above. The connectivity analysis will support this condition. This group consists of (SG-1, SG-2, SG-3, SG-4, SG-6, SG-7, and SB-3). When checked by using Equation. 4 and Equation. 5, Group I and Group II groundwater origin elevation is below the sampling elevation. The condition is assumed to be caused by the different sampling periods with rainwater samples and indicates the rainfall amount effect. Group III is the groundwater from meteoric water at 394-440 masl elevation. This elevation is slightly higher than the sampling location elevation,
indicating the groundwater is in the local flow groundwater system, the member of this Group is SB-2, SB-4, SB-5, SB-10. The last Group is Group IV, the groundwater plotted still near the RMWL, but the $\delta^{18}$O value is more enriched than groundwater in Group III. This indicates the fractionation on $\delta^{18}$O affected by oxygen shifting from the water (H$_2$O) into carbonate minerals (the dissolved of CaCO$_3$) due to water-rock interaction [4,14]. This group consists of SB-1, SB-6, SB-7, SB-8, SB-9.

5.2. Hydrochemical Analysis

5.2.1. Groundwater facies. Before the analysis is conducted, the major ions unit from the laboratory needs to be converted from mg/L to meq/L by multiplying the mg/l with the conversion factors [11]. The major ions in meq/L. After being converted to meq/L, the Reaction Error (RE) on groundwater was checked by using Equation (2), as shown in Table 4. The calculation RE value less than 10% means that all samples can be used for hydrochemical analysis [15]. The major ions data must be plotted into a piper diagram to determine the groundwater facies.

| Sampling Location | Na$^+$ | K$^+$ | Ca$^{2+}$ | Mg$^{2+}$ | Cl$^-$ | SO$_4^{2-}$ | HCO$_3^-$ | RE (%) |
|------------------|--------|-------|-----------|-----------|--------|------------|-----------|--------|
| SB-1             | 2.83   | 0.06  | 5.35      | 2.62      | 0.70   | 1.34       | 7.96      | 4.12   |
| SB-2             | 1.61   | 0.14  | 5.92      | 2.97      | 0.76   | 0.97       | 8.06      | 4.20   |
| SB-3             | 2.80   | 0.03  | 2.14      | 1.08      | 0.36   | 0.87       | 5.28      | -3.64  |
| SB-4             | 1.57   | 0.06  | 5.34      | 1.26      | 0.22   | 0.91       | 7.80      | -4.04  |
| SB-5             | 1.82   | 0.07  | 7.20      | 0.78      | 0.26   | 0.93       | 7.96      | 3.84   |
| SB-6             | 9.59   | 0.17  | 3.55      | 1.02      | 3.70   | 0.62       | 10.63     | -2.14  |
| SB-7             | 1.55   | 0.09  | 5.11      | 1.30      | 0.42   | 0.43       | 6.71      | 3.17   |
| SB-8             | 14.96  | 0.06  | 2.81      | 2.30      | 0.34   | 3.54       | 5.83      | -5.44  |
| SB-9             | 2.34   | 0.09  | 4.38      | 1.74      | 0.65   | 0.23       | 7.57      | 0.63   |
| SB-10            | 1.52   | 0.06  | 4.44      | 2.54      | 0.44   | 0.64       | 8.18      | -3.89  |
| SG-1             | 3.51   | 0.08  | 2.81      | 2.30      | 0.34   | 3.54       | 5.83      | -5.44  |
| SG-2             | 1.68   | 0.04  | 2.24      | 0.91      | 0.19   | 0.31       | 4.77      | -3.96  |
| SG-3             | 4.41   | 0.10  | 2.55      | 0.99      | 0.53   | 1.36       | 6.80      | -3.86  |
| SG-4             | 1.62   | 0.04  | 2.36      | 0.86      | 0.10   | 0.17       | 5.13      | -5.07  |
| SG-6             | 2.68   | 0.05  | 2.56      | 1.22      | 0.68   | 0.66       | 5.56      | -2.96  |
| SG-7             | 2.28   | 0.03  | 2.14      | 2.62      | 0.34   | 0.58       | 6.80      | -4.41  |
| MA-1             | 2.02   | 0.06  | 1.75      | 1.38      | 0.28   | 0.33       | 5.07      | -4.27  |
| MA-2             | 0.58   | 0.03  | 2.59      | 1.56      | 0.19   | 0.12       | 4.97      | -5.01  |
| MA-3             | 8.10   | 0.29  | 2.68      | 5.28      | 3.56   | 3.61       | 10.29     | -3.28  |
| MA-4             | 1.70   | 0.05  | 1.46      | 1.87      | 0.21   | 0.33       | 4.94      | -3.82  |

Based on Furtak & Langguth classification (Figure. 8), groundwater facies in Raimank and its surroundings consist of 3 groups. Group A is alkaline-earth water bicarbonate predominated, consisting of SB-2, SB-4, SB-5, SB-7, SB-10, and MA-2. This condition indicates that groundwater comes from local recharge areas with a tendency dominated by clay minerals [9,16]. The isotope analysis also confirms this result that shows their relationship with the RMWL. Group D is Alkaline-earth water with higher alkali content bicarbonate predominated, consisting of SB-1, SB-3, SB-9, SG-1, SG-2, SG-4, SG-6, SG-7, MA-1, MA-3, and MA-4. This group indicates the groundwater has flowed away from the recharge area [9]. The result supports the isotopic analysis, which shows that most groundwater in this group is affected by evaporation and oxygen shifting during flowing and infiltration. Grup F is Alkaline water bicarbonate predominated the groundwater on this group is
indicated comes from the discharge area and experienced interaction with rocks containing high \( \text{Na}^+ \) [12], this group consist of SB-6, SB-8, and SG-3.

![Diagram showing Furtak & Langguth classification in Piper Diagram result]

**Figure 8.** The Furtak & Langguth classification in Piper Diagram result

5.2.2. **Aquifer Connectivity.** Schoeller diagram shows the groundwater cations in the study area dominated by \( \text{Ca}^+ \), while the anions are dominated by \( \text{HCO}_3^- \) (Figure 9). The \( \text{Ca}^+ \) domination is caused by interaction with carbonate rocks. On the other hand, the \( \text{HCO}_3^- \) dominance shows that most groundwater comes from shallow groundwater systems [17,18]. The result shows four major patterns in the Schoeller diagram, as shown in Figure 10. The groundwater, which has a similar pattern, has hydraulic connectivity. Group 1 in the Schoeller diagram plotting consists of SB-1, SB-4, SB-5, SB-7, SB-9. This result is slightly different with isotopic analysis, SB-1, SB-7, SB-9 are not in the same group with SB-4 and SB-5 it shows the possibility of mixed water between those groups. Group 2 is the groundwater from SB-2 and SB-10. This result supported the isotope and piper diagram analysis that shows that groundwater comes from rain infiltration at the same elevation. Group 3 members are SB-6 and SB-8. This group has more \( \text{Na}^+ \) and fewer \( \text{Ca}^+ \) and \( \text{Mg}^+ \) than the others. This condition shows the characteristic of the cation exchange that indicated there is water rock-rock interaction during groundwater evolution [18]. Group 4 consists of the SG-1, which has slightly more \( \text{SO}_4^{2-} \) than other groundwater of this group, SG-2, SG-3, SG-4, SG-6, SG-7, MA-1, MA-3, and SB-3. It confirmed that SB-3 is on the same groundwater system as Dug Well (SG). This result also indicates groundwater on Dug Well is coming from around MA-1 or above. In addition, there is groundwater with different patterns (MA-2 and MA-4), so it cannot be grouped and shows no hydraulic connectivity with other groundwater sources (Figure 11).
Figure 9. General description of Schoeller Diagram of groundwater in the study area

Figure 10. (a,b,c,d). Major patterns of Schoeller Diagram of groundwater in the study area

Figure 11. The groundwater with a non-similar pattern of Schoeller Diagram of groundwater in the study area
6. Conclusion

Based on isotope and hydrochemical analysis, groundwater in Raimanuk and its Surroundings consist of at least 4 groups, Group I (MA-2 and MA-4). The source groundwater on this group is meteoric water that falls on each location. Therefore groundwater in this group has different isotopic compositions and groundwater facies. There is no indication of hydraulic connectivity with other groups since the results of the Schoeller diagram analysis show a different pattern from the other groups. Group II (SG-1, SG-2, SG-3, SG-4, SG-6, SG-7, SB-3, MA-1, and MA-3) is the groundwater from dug wells that are affected by evaporation. This indicates a shallower aquifer. The groundwater facies and connectivity analysis shows that the groundwater in the dug well is influenced by meteoric water that falls around MA-1, which is flowed away into lowlands through the unsaturated zone (or rivers), and the evaporation occurs during this period. Group III (SB-2, SB-4, SB-5, SB-10) is the groundwater that originated from meteoric water at 394-440 masl. This elevation is not far from the sampling location, and it shows that the groundwater has come from the local recharge area. The groundwater facies analysis also indicates the groundwater in this group is affected by the meteoric water composition. Group IV (SB-1, SB-6, SB-7, SB-8, SB-9) is the groundwater with δ¹⁸O enriching compared to groundwater in Group III. This condition is affected by oxygen shifting from the water (H₂O) with the carbonate minerals (CaCO₃). The Scholler Diagram indicates hydraulic connectivity between this Group and Group III. It shows that the groundwater utilization in this group can affect groundwater conditions in Group III and vice versa. Although it consists of several groundwater groups, the groundwater system in Raimanuk and its surroundings is indicated as young groundwater from shallow aquifers.

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