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# Origin and Recharge Estimation of Groundwater using Chemistry and Environmental Isotopes in the Mahafaly Sedimentary Aquifer, District of Betioky Southwestern Madagascar

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### Abstract

Hydrochemical and environmental isotopes studies are used to determine the groundwater origin and recharge estimation in the sedimentary aquifer of Mahafaly, southwest of Madagascar. The groundwater chemical type in the study zone varies according to the geological formation and the altitude of the sampling site. Groundwater at high altitude of the Isalo formation represents freshwater with Na-Ca-HCO<sub>3</sub>-Cl type and it changes to more saline downwards while following the direction along the flow paths, where in the border of the Onilahy River the water type becomes Na-Ca-Mg-SO<sub>4</sub>-Cl-HCO<sub>3</sub>. Highly mineralized deep groundwater mixed with local rainwater occurs in the Sakamena basin except for the groundwater samples near the Onilahy River, which represents a mixing with the river. Groundwater isotopic composition results in the study area show that the aquifer is partially recharged by local rainfall during the summer season. Mixing and evaporation are the processes dominant which control the groundwater mechanism recharge. Besides, the tritium results have a mean value of around 0.83TU, which confirms that the majority of the samples, around 60%, are produced by mixing of recent and old groundwater from different areas. Hot springs have a tritium value less than 0.2TU indicating a paleo-recharge from a distant recharge zone. We propose a mean groundwater residence time in the study zone of around 75 years, which corresponds to a mean recharge rate of 60mm/year giving a total recharge of  $2.7*10^8 \text{ m}^3/year over the total study area.$ 

Keywords: Hydrochemistry, Isotopes, Recharge, Groundwater, Residence time, Madagascar.

## Introduction

Water is the source of life. Since 2007, the Malagasy government attempts to increase the access to drinking water in the rural zones of the country, by installing improved drinking water sources. Despite these installations, the access to safe drinking water remains very limited both in space and time. Only limited areas had been supplied with drinking water sources, yet, in most of the regions, which had benefited from water source infrastructures, a long term availability of safe drinking water is still difficult to meet. Technical and/or mainly hydrogeological issues are often the causes of those limitations, which are linked to both the groundwater quality and quantity. In the southwestern region of Madagascar, the access rate to safe drinking water is less than 38%, one of the lowest rates in the country, making the region one of the least developed of the Island<sup>1</sup>. The water resources problem in the region is of twofold: water quality due to agricultural or mining pollution and water quantity resulting from well water exhaustion due to decreasing water table during the dry winter season<sup>2</sup>. The present study aims at helping the NGOs working in the District of Betioky to supply safe drinking water while determining the source of groundwater recharge and their rate infiltration in the sedimentary aquifer of Mahafaly plateau using environmental isotopes and chemical investigation. Isotopic and hydrochemical indicators often serve as effective methods for resolving multiple problems in hydrology and hydrogeology, especially in semi-arid and arid regions<sup>3</sup>. Hydrochemical and isotopic data are powerful used to identify the sources of groundwater recharge systems and to trace the groundwater flow<sup>4</sup>. Temperature, <sup>2</sup>H, <sup>18</sup>O, <sup>14</sup>C, <sup>3</sup>H, <sup>3</sup>H/<sup>3</sup>He and dissolved gas are environmental tracers which help to give better estimation of recharge and flow time scales<sup>5</sup>.

**Study zone:** The study zone is situated in the middle of Betioky district, between the 23°15'S and 24°10'S latitudes and the 44°10'E and 45°00' E longitudes. The district of Betioky is surrounded in the north by the district of Toliara, the district of Benenitra in the eastern part and the district of Ampanihy in the south. The district consists of wide plains through which the Onilahy River flows westwards through the Mahafaly kartsic plateau. The total geographical area of the study zone is 4,500 km<sup>2</sup>. The altitude of the plains varies between 100 m to 400 m. Globally, the district is completed of large peneplains. It is bounded in its eastern border by the Sakoa basin and in the west by the Mahafaly plateau. Thus, the whole area can be divided into three categories, the Betioky hill, the alluvial plains and the east valley in Sakamena and Sakoa basins. The district is

characterized by a semi-arid climate zone with temperatures of 25°C. Average rainfall precipitation is close to 500 mm/yr and it rains only during the summer season.

Geological setting of study area: The study zone is a sedimentary basement, mainly formed by Neocene continental with sandstone and sandy clay extending below a shell of red sands<sup>6</sup>. The zone belongs to the Karroo group which distinguishes three series of geological formation namely Sakoa, Sakamena and Isalo. The Sakoa formation on the east part of study zone is a glacial series in the base attributed in the Upper Carboniferous, following the Permian coal series and red series. The Sakamena formation involves in the base a powerful schist sandstone series of upper Permian following clay with septaria corresponding at the upper Triassic. The Isalo formation corresponds to the large group of continental wastes. The Isalo group is divided into three subgroups. Isalo I consists of a massive sandstone but soft poorly cemented, cross-bedded without fossil. In the middle southern part of the island, Isalo II presents a continental facies, with soft sandstone cross-bedded,

alternating with green or red clays and argillite. Sometimes, a silicified wood fossil which is very large (Araucarioxylon, Cedroxylon) is the rest of fish (Ceratodus) and Reptiles (Précrocodiliens).

In the western part of the study zone, middle Jurassic ends definitively the Karroo system. The upper Jurassic is characterized by generalized marine transgression, which affects the deposits along the western coast of Madagascar, represented by a calcareous limestone, marls and clays containing in abundance of Ammonites and Belemnites. The upper Jurassic forms in the western coastal area a narrow strip of around 10 to 20km. These layers, which are enriched in glauconite can form an impermeable cover protecting the aquifer. Clays always contain crystals of gypsum.

The Lower Cretaceous is quite predominantly continental and epi-continental. Again, it is reported as gypsiferous at several levels. However, alluvium formed with clayey sands are found along the river<sup>7</sup>.



Figure-1 Sampling sites of the study area



(Source: FTM BD 500, 2000)

#### Figure-2 Geological map of study zone

**Hydrogeological setting of study zone:** Groundwater in the sedimentary aquifer at Mahafaly region drains a vast area starting at the upstream on the crystalline peneplains of Androy and mahafaly. It flows westwards up to the Onilahy River across various geological formations. All the way, the main recharge zone area is located at level of crystalline basement where precipitation is more important<sup>9</sup>. In this zone, runoff acts only during heavy rains created a flood river and then can temporary supply the main aquifer. The main aquifer is fresh water in upstream except a few points presenting a high salinity. Groundwater is more mineralized while following the direction in downstream through the different geological formation. Two main faults are observed in the study area namely, the fault of Manalombe at the East part of the Betioky hill and the Ampakabo fault at the western part of the Betioky hill.

# Methodology

Field works are performed for sampling water during August 2012 and July 2013. Forty six water samples were collected from different water sites. The sites of water sampling for isotopes and chemical analysis are dug wells, boreholes, springs

and rivers. Dug wells are installed in different geological zones while the two boreholes are not distant which drains the shallow aquifer. Field measurements include pH, electrical conductivity, temperature, dissolved oxygen and alkalinity. Samples for chemical analysis are filtered with whatman paper of 0.45µm diameter and collected in a polyethylene bottle of 50ml. Samples for cation analysis are added a few drops of sulfuric acid 0.16N. Cation and anion analyses were performed by Ion chromatography (DX-120) at X-ray fluorescence and Environment laboratory of INSTN-Madagascar. Samples for stable isotopes analysis were collected in polyethylene bottle 50ml and kept at constant temperature of 4°C. The stable isotopes analyses were carried out using laser spectrometry PICARRO L1102-i in the Geochemistry and Geochronology Department at CEREGE, Aix-en-Provence. The accuracy of the measurements is respectively  $\pm 0.15\%$  and  $\pm 2\%$  for the <sup>18</sup>O and <sup>2</sup>H. <sup>3</sup>H analyses were carried out at laboratory of Isotope Hydrology Department of INSTN-Madagascar. Water samples were enriched by electrolytic method before using liquid scintillation counting Tri-carb 3170 TR/SL. Tritium concentration is expressed in tritium unit (TU) and the accuracy of measurement is 0.25TU.



(Source: Hydromad, 1999)

Figure-3 Piezometric map of the study area

#### **Results and Discussion**

Hydrochemistry: The Electrical Conductivity of groundwater samples varies from 150µS/cm to 5000µS/cm. This indicates that the geochemical evolution starts from fresh to saline water depending on the geological formation of the study zone. Low electrical conductivity corresponds to the samples collected in high altitude, which is related to fresh water on sandstone and clay formation in the Isalo group. The relative abundancy is presented in decreasing order by following Na>Ca>Mg>K for cations and Cl>SO<sub>4</sub>>HCO<sub>3</sub>>NO<sub>3</sub> for anions. Therefore, groundwater in the Isalo formation has a Na-Ca-HCO<sub>3</sub>-Cl type in high altitude and move to Na-Ca-Cl-HCO<sub>3</sub> type in low altitude because the chloride ion concentration increases along the flow path direction. Piper diagram (Figure-4) shows that mixing of fresh and mature groundwater is observed in this zone. Therefore, halite dissolution and cation exchange give high concentration of sodium because Na is released by the clays sandstone in the Isalo group formation. This occurs in the normal process of cation exchange because sodium ion released to water is replaced by calcium ion in the aquifer material<sup>10</sup>. This is confirmed by the high ratio of Na/Cl. Moreover, calcite dissolution from the calcareous sandstone formation is probably the source of calcium and bicarbonate. On the other hand, high electrical conductivity is observed in the groundwater near the river in the Jurassic and cretaceous formation meaning the groundwater is more mature with a Na-Ca-Mg-SO<sub>4</sub>-Cl-HCO<sub>3</sub> facies due to different geochemical process mainly gypsum dissolution and cation exchange with Na/Ca ratio more than 1.75 suggesting a decrease of Ca concentration with respect to Na<sup>11</sup>. The groundwater is more saline marked by high mineralized groundwater with electrical conductivity up to 5000µS/cm in the southern part of the study zone which presents a Na-Cl type suggesting that dissolution of halite is occurring from salt clays of upper Sakamena formation<sup>7</sup>. Another assumption would be a high mineralized deep groundwater flowing up to the shallow aquifer. Combined tectonic dip in the monoclonal plays a very important role conducting the upwelling of high mineralized deep groundwater<sup>2</sup>. For the groundwater in the Sakamena basin at the east valley, the high mineralization refers to the high sulphate concentration from dissolution of halite and dissolution of gypsum giving a Na-SO<sub>4</sub>-Cl water type. On the contrary, the groundwater is less mineralized for the samples collected near the river because of a mixing process between river water of Onilahy, groundwater with distant recharge and groundwater from direct infiltration of local rainfall which transforms the water chemical type to Na-Ca-SO<sub>4</sub>-HCO<sub>3</sub>.

**Isotopes composition** ( $\delta^{18}$ **O**,  $\delta^{2}$ **H and tritium**): The stable isotopes composition of the groundwater samples from boreholes and wells in the study zone vary from -5.82% to - 3.72% for  $\delta^{18}$ O and from -38.28% to -26.58% for  $\delta^{2}$ H.

For the rivers, the  $\delta^{18}$ O and  $\delta^2$ H composition varies respectively from -4.13% to -4.63% and from -29.2% to -32.6%. Then, the stable isotope composition of the hot springs varies from -6.63% to -7.45% for  $\delta^{18}$ O and from -45.5% to -49.3% for  $\delta^2$ H. The deuterium excess values for the groundwater samples in Isalo formation, Jurassic formation and Sakamena formation are 4.1%, 5.7% and 5.9% respectively. For the river water, this value is 3.9% due to evaporation, while deuterium excess for the springs, is 9.34%.

Generally, the tritium concentration varies from 0.05TU to 1.25TU in the groundwater samples and the mean value is 0.74TU. The smallest value corresponds to the borehole water and the highest value was obtained from the groundwater sample in high altitude. The mean tritium concentration of the rivers is close to 1TU; on the contrary this value is less than 0.02TU for the hot springs. The mean tritium content and stable isotopes concentration are classified by geological formations in the Table-1.



Isotope composition  $\delta^{18}$ O and  $\delta^{2}$ H plot for all samples

| Zone    | Geological formation     | δ <sup>18</sup> Ο (‰) | δ <sup>2</sup> H (‰) | d_excess (%) | <sup>3</sup> H (TU) |
|---------|--------------------------|-----------------------|----------------------|--------------|---------------------|
| А       | Isalo group              | -3.725.39             | -26.5837.83          | 4.13         | 0.83                |
| B-C     | Sakoa and Sakamena group | -3.815.15             | -29.4232.88          | 5.70         | 0.89                |
| D       | Jurassic and cretaceous  | -4.255.82             | -26.83 38.28         | 5.85         | 0.89                |
| Springs | Isalo group              | -6.637.45             | -45.4849.28          | 9.34         | 0.2                 |
| River   | Alluvium                 | -4.134.63             | -29.1632.64          | 3.88         | 0.99                |

Table-1 Isotopic composition of all samples

Origin of the groundwater: The equation of local meteoric water line for the precipitation in Antananarivo (Madagascar) is  $\delta^{2}H = 8.05*\delta^{18}O + 14$  (r<sup>2</sup>=0.969) which is close, but slightly above the Global Meteoric Water Line  $\delta^2 H = 8.13 * \delta^{18} O + 10.8$ <sup>12, 13</sup>. The regression line for the groundwater in the sedimentary basement at the Betioky hill (Zone A) has an equation  $\delta^2 H =$  $5.71*\delta^{18}O - 6.5$  (r<sup>2</sup>=0.848) indicating a slight influence of evaporation during or after infiltration. The isotopic composition of  $\delta^{18}$ O of the rainfall collected from December 2000 to march 2001 ranges from -4.25% to -1.21% with a mean value of -3.06% in Taolagnaro station 150km away from the study area<sup>14</sup>. Groundwater composition more depleted that local rainfall suggest that groundwater is only partially recharged by local precipitation during summer rainfall. The mean tritium value of all groundwater samples is 0.83TU indicating that the groundwater is a mixing of old and recent precipitation in the study zone.

Groundwater samples near the river on the Jurassic and Cretaceous formation (zone D) display a linear regression line equation  $\delta^2 H = 1.9 * \delta^{18} O - 22.86$  (r<sup>2</sup>=0.75). This slope is too low for reflecting a continuous evaporation enrichment process. Therefore, it more probably reflects a mixing between different water origins. The similarity between the isotopic composition ( $\delta^{18}O/\delta^{2}H$ ) of the groundwater in the Jurassic Cretaceous formation (zone D) and that of the river (Table-1, Figure-5) suggests that a mixing of non evaporated groundwater with the river water is observed in the samples from this zone. Similarly to groundwater in the east valley to Sakamena basin, the groundwater isotope contents in the south are depleted compared to the isotope composition of the groundwater in the alluvial plain.

The groundwater with more depleted values could indicate a mixing between surface recharge and deep mineralized groundwater. Groundwater in the alluvial plain represents a mixing of local precipitation with the river. Beside, the river has a similar isotope signature with that of the wells. Regarding the thermal springs, the samples isotope signatures are more depleted indicating a probable paleo-recharge. Consistently, the tritium contents in the spring water are less than 0.02TU representing old groundwater.

**Groundwater residence time and recharge estimation:** Tritium was widely used to estimate modern groundwater residence time because it is a part of the water molecule constituent and thus its behaviour is similarly whatever their type<sup>15,16</sup>. The mathematical model based on exponential distribution function developed by Malowszewski was applied in this zone.

For the exponential model, the flow lines have approximately a distribution of exponential transit times<sup>17</sup>. Thus, the tritium data in Antananarivo from 1962 to 1975 and some samples from the rainfall collected between 2009 and 2012 which were analysed at the Isotope Hydrology Department of INSTN-Madagascar were taken as required input function. The earlier missing data was interpolated using a good logarithmic correlation with Antananarivo and IAEA station (Vienna, Austria)<sup>18</sup>. The simulated tritium response curves were calculated using the convolution integral expressed by:

$$c_o(t) = \int_0^\infty c(t-\tau) * f(\tau) * \exp(-\lambda) dt$$
(1)  
where  $f(\tau) = \frac{1}{\tau_m} * e^{-\left(\frac{1}{\tau_m}\right)t}$  and  $\tau_m = \frac{V}{Q}$ 

Where  $c_o(t)$  is the outlet tracer concentration,  $c(t-\tau)$  is the concentration of tracer at the time t- $\tau$ , t is the time of sampling,  $\tau$  is the date at which parcel entered the aquifer system,  $\lambda$  is constant decay,  $\tau_m$  is the mean age of parcel water, V is the volume of reservoir system and Q is the outflow flux into the system. The calculation of curve response was established using the FLOWPC programme developed by the International Atomic Energy Agancy<sup>19</sup>.

The results of the simulation obtained show that the groundwater residence time in the region varies from 35 years to 150 years depending on the location of the sampling site; however the mean residence time of all groundwater samples is around 75 years generally. The groundwater located at high altitude of the Isalo formation is recently recharged with tritium concentration greater than 1.25 TU by the local precipitation with a residence time starting at 30 years and it increases while following the direction along the flow path.



Groundwater flow direction according to tritium concentration of the samples

n \* H

For the boreholes water, the mean residence time is up to 150 years due to their contact with the deep groundwater. In the study zone, more mineralized groundwater is older than others, as the dissolved constituents' concentration increases with longer water- rock interaction time <sup>20</sup>. For the hot springs, they have an apparent age of more than 200 years indicating a remote recharge. The groundwater in the alluvial plain has a longer residence time than that of the samples in the Betioky hill because the river drains the aquifer in the latter area and increases the tritium concentration of the groundwater samples.

Groundwater recharge estimation is given by the following relationship <sup>21</sup>:

$$R = \frac{r}{T}$$
(2)

p is the effective porosity, H is the thickness of the aquifer and T is the mean groundwater residence time. We assume that the effective porosity is 6% in the sedimentary basement of Mahafaly region and the thickness of the aquifer includes between 20m and 60m according to the boreholes depth<sup>22</sup>.

Considering a groundwater residence time calculated above, the annual recharge rate in the study zone varies from 20 mm/yr to 120 mm/yr with mean of 60 mm/yr giving a mean recharge of  $2.7 \ 10^8 \text{m}^3$ /yr dispersed on  $4500 \text{km}^2$  of the total area.

| Sample code | X (m)     | Y (m)      | Geological formation    | 3H value(UT) | Mean residence time | Recharge<br>(m/yr) |
|-------------|-----------|------------|-------------------------|--------------|---------------------|--------------------|
| JIR01       | 190359.82 | 264811.55  | Isalo                   | 0.03         | 125                 | 0.02               |
| MAS01       | 189376.46 | 266154.11  | Isalo                   | 0.61         | 80                  | 0.04               |
| MAS02       | 189560.26 | 265960.65  | Isalo                   | 0.89         | 65                  | 0.05               |
| P01         | 190741.38 | 265721.04  | Isalo                   | 0.79         | 75                  | 0.04               |
| P04         | 193779.72 | 250258.13  | Isalo                   | 1.11         | 30                  | 0.10               |
| P05         | 194753.79 | 261114.36  | Isalo                   | 1.00         | 35                  | 0.09               |
| P06         | 194733.57 | 260926.51  | Isalo                   | 1.26         | 25                  | 0.12               |
| P07         | 217478.17 | 273048.41  | Sakamena                | 1.25         | 25                  | 0.12               |
| P08         | 215919.93 | 275536.92  | Sakamena                | 1.22         | 25                  | 0.12               |
| P017        | 191236.33 | 246439.05  | Isalo                   | 0.42         | 95                  | 0.03               |
| P018        | 192299.73 | 240825.11  | Isalo                   | 0.89         | 65                  | 0.05               |
| P020        | 193168.08 | 239268.03  | Isalo                   | 0.77         | 75                  | 0.04               |
| P022        | 180141.45 | 281685.88  | Jurassic and Cretateous | 1.22         | 35                  | 0.09               |
| P023        | 179360.37 | 280783.16  | Jurassic and Cretateous | 0.81         | 65                  | 0.05               |
| P024        | 178933.49 | 280900.21  | Jurassic and Cretateous | 0.64         | 85                  | 0.04               |
| P026        | 181135.98 | 281515.16  | Jurassic and Cretateous | 1.05         | 35                  | 0.09               |
| P027        | 181723.47 | 284137.28  | Jurassic and Cretateous | 0.93         | 55                  | 0.05               |
| P028        | 181946.42 | 284417.13  | Jurassic and Cretateous | 0.74         | 75                  | 0.04               |
| P029        | 188152.74 | 263435.502 | Isalo                   | 0.44         | 120                 | 0.03               |
| P030        | 189499.48 | 262223.33  | Isalo                   | 0.55         | 125                 | 0.02               |
| P035        | 201147.94 | 259867.62  | Sakamena                | 0.87         | 70                  | 0.04               |
| P036        | 202189.65 | 261586.83  | Sakamena                | 0.36         | 130                 | 0.02               |

 Table-2

 Recharge values of different geological formation in the study area

### Conclusion

The data obtained from the chemical and isotopes analyses were used for identifying the groundwater origin and estimating the groundwater recharge in the sedimentary aquifer in the Betioky district, southwestern part of Madagascar. We conclude that hydrochemical results show mainly a mixing of two different water types in the Isalo formation and their chemical facies type varies according to the electrical conductivity of the water sample from fresh to saline. Chloride concentration and total dissolved solute increase along the flow path while moving downwards from the recharge zone area. High mineralized groundwater in the Jurassic and cretaceous formation is observed conducting to mature water type. For the isotopes results, the isotope compositions of  $\delta^{18}O$  and  $\delta^{2}H$  show a partial recharge from the local precipitation and different mixing process occurs in groundwaters according to the geological

formation and geographical position of the aquifer. This assumption is confirmed by the tritium results, which show that most of the groundwater samples have a tritium concentration between 0.7TU and 1TU with a mean value of 0.85TU which indicates that groundwater comes from modern and old mixed. The groundwater residence time in the study zone varies from 35 years to more 150 years. The most recent groundwater samples are localized in the alimentation zone; on the contrary the water samples for the hot spring have a residence time greater than 200 years. Generally, the annual total recharge of groundwater in the study zone is estimated to  $2.7*10^8 \text{m}^3$ /year corresponding to the 4500km<sup>2</sup> of the total site area.

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