

Radon Progenies as a Source of Gross Alpha-beta Activities in Drinking Water in Vinaninkarena, Antsirabe-Madagascar.

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The village of Vinaninkarena, Antsirabe, Madagascar (47°02'40''E, 19°57'17''S) is located in a high radioactivity area. With the aim of assessing the health impact of radioactivity pollution, a survey was conducted around an abandoned radium mining. Waters from 24 sampling points, including 5 wells and 13 springs were sampled. The water radioactivity was measured using a portable Triathler LSC, model 425-034 from Hidex Turku. Each sample is counted more than 3 times: less than 2 h after sampling, 4 h to few days and more than 3 weeks after when ^{226}Ra attains equilibrium with ^{222}Rn and its short-lived daughters. Combination of water non miscible, radon extractive LS cocktail and water miscible one is used to reveal contribution of radium, radon and its short lived progenies to the gross alpha-beta activities. Maximum activity values found are : $3.6 \pm 0.3 \text{ Bq.L}^{-1}$ for ^{226}Ra , $554 \pm 9 \text{ Bq.L}^{-1}$ for the excess of radon, and $408 \pm 8 \text{ Bq.L}^{-1}$ for the excess of radon progenies. Based on these data, a daily consumption of 2 L of these waters by a standard person would produce an annual ^{226}Ra ingestion well above the annual intake limit for ^{226}Ra . Moreover, the increasing gross alpha-beta activity resulting from radon progenies raises the issue of health effects. The potential for rapid spring-to-mouth delivery, which is frequent in rural area, may expose the water consumer to the short-lived alpha particle emitters.

1. INTRODUCTION

The measurement of the activity of radium and its progenies in drinking water is crucial from the radiological point of view. So far, there has been a clear lack of data concerning those radionuclides in Madagascar. However, the region of Vinaninkarena, Antsirabe, Madagascar (47°02'40''E, 19°57'17''S), is known to be a high natural radioactivity area (see Figure 1). Uranium ore was extracted in the region during the fifties and early sixties by the French CEA (Commissariat à l'Énergie Atomique). But in the mid sixties, mining activities were abandoned for economical and political reasons. As a rural area, drinking waters are collected exclusively from wells and natural springs.

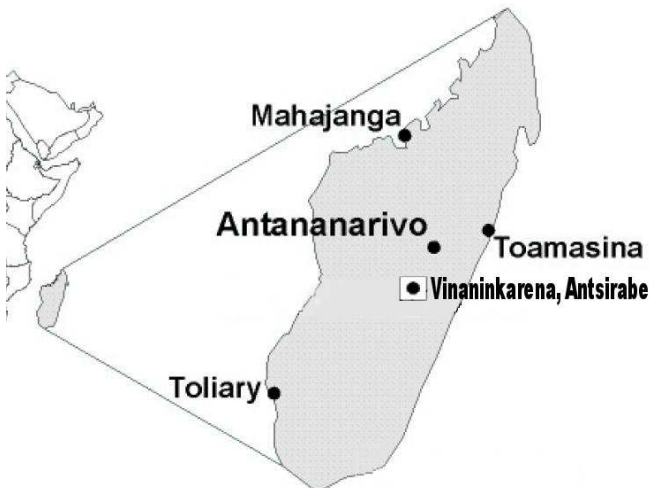
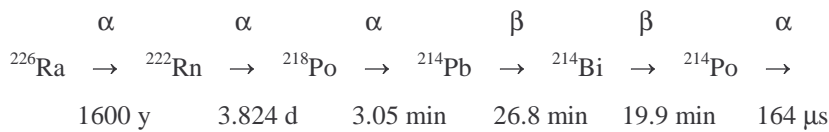


Figure 1 : Geographical location of the village of Vinaninkarena, Antsirabe, Madagascar

Previous soil measurement conducted by a team from Madagascar-INSTN (Madagascar Institut National des Sciences et Techniques Nucléaires), using gamma spectrometry, showed that high radioactivity in the region originates mainly from the uranium series, especially from ^{226}Ra [1]. This work is aimed to measure the level of radioactivity in drinking waters used by villagers in order to assess the health impact of radioactivity pollution.

2. MATERIALS AND METHOD

Field campaign was conducted from 13 February to 22 February 2004. Waters from 24 sampling points, including 5 wells and 13 springs were sampled. Water radioactivity was measured using a portable Triathler LSC (model 425-034 from Hidex Oy, Turku) which makes possible field measurement of radon progenies immediately after collection. Moreover, the Triathler LSC is equipped with pulse length discriminator, which allows separation of alpha from beta particle signal in a 2-D (2 dimension) graph. Successive counting is done until equilibrium between ^{226}Ra and its progenies is established according to [2] :



Glass mini-vials were used, filled with 4 mL of aqueous phase and 3 mL of organic phase. Water non miscible, radon extractive LS cocktail Toluene scint, containing 0.4 mg of PPO and 0.05 mg of POPOP in 1 L of toluene was used as cocktail. Each sample is counted 5 times : less than 5 mn after sampling, after 15 mn, 3.5 h, 3 days and more than 4 weeks. Counting time is 10 mn for the first four measurements, and 60 mn for the last measurement. Result consists of 1-D (1-dimension) alpha-beta spectra, with the protocol P-32, and 2-D spectra, using α/β discrimination protocol. If the alpha spectrum is well separated, and window parameters optimized, the efficiency for alpha particles equals approximately 100% and there is no need for calibration. Moreover, alpha background is low, resulting in lower limit of detection.

Net count rate, R, in a sample is calculated as:

$$R = R' - B$$

where R' is the gross count rate and B the background count rate. The detection limit L_D , is usually defined as [3]:

$$L_D = B + 3 \sigma_B$$

where σ_B is the standard deviation of B. Based on use of statistical theory, Currie [4] defines detection limits for radioactivity, at 95 % confidence, as:

$$L_D = B + 4.65 \sigma_B$$

For 10 mn counting time, the L_D is 0.019 cps, or 4.8 Bq.L^{-1} in term of alpha activity. For 60 mn counting time, the L_D is 0.012 cps, or 3 Bq.L^{-1} which means 1 Bq.L^{-1} for ^{220}Rn or ^{226}Ra at equilibrium with their daughters.

According to Manjón G. et al. [5], ^{226}Ra activity can be deduced from gross alpha/beta activity, considering that at equilibrium, i.e. after 4 weeks, one ^{226}Ra disintegration produces 4 alpha and 2 beta particles. In the case of ^{222}Rn , it consists of 3 alpha and 2 beta particles and equilibrium is attained only after 3.5 hours.

With a 2-D graph, alpha/beta pulse shape discrimination for ^{226}Ra provides sufficient separation from all low and medium energy betas, giving more precise results [6].

3. RESULTS

Typical 1-D LS spectrum (see Figure 2), especially for measurement done less than 3 hours after sample collection, shows evidence of alpha and beta radioactivity. We can see the difference when comparing it with the background spectrum.

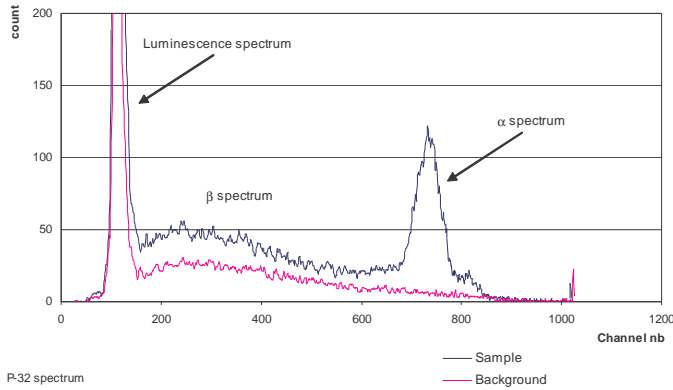


Figure 2 : typical sample 1-D LS spectrum

Typical LS spectrum in 2-D graph (see figure 3) shows also evidence of alpha and beta radioactivity.

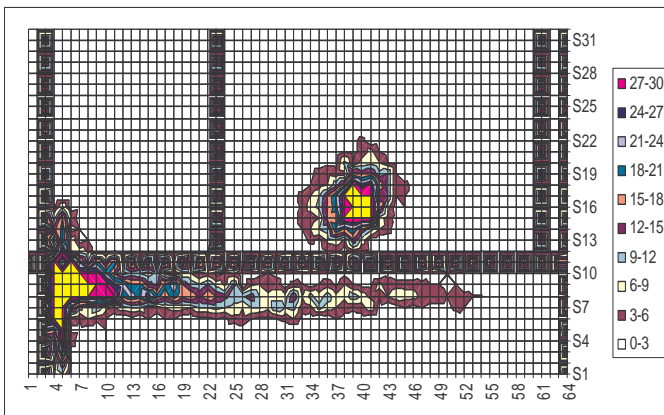


Figure 3 : Typical sample LS spectrum in 2-D graph. Alpha spectrum is in the upper right window.

Sample activities vary according to sample location and measurement time. Table 1 shows the range of activity concentrations of ²²⁶Ra, ²²²Rn and radon progenies measured in both surface, well and spring water supplies in the region of Vinaninkarena.

Table 1 : Alpha and beta emitters concentrations range in drinking water samples

	Activity (Bq.L ⁻¹) (*)	
	Min	Max
Excess of radon progenies	1.4 ± 0.6	408 ± 8
Excess of radon	< LLD (1 Bq.L ⁻¹)	554 ± 9
²²⁶ Ra	< LLD (1 Bq.L ⁻¹)	3.6 ± 0.3

(*) : with only statistical uncertainties, quoted at 1 σ

We have found that activity concentrations were higher in spring source samples than in well or superficial waters. This is due to the fact that groundwater flows through radioactive ores carrying radioactive materials and other elements from the solid to the liquid phase. And in the case of spring water, sampling is done right after emergence. Predicting

the degree of mobility of the radionuclides however is complex, as it is affected by factors like solubility and chemical affinity, as well as some aquifer characteristics like porosity, surface area exposed to water and residence time [7].

According to Manjón G. et al. [4], if the water sample contains only ^{226}Ra without its daughters, activity would steadily increase until equilibrium is reached. In that case, the next relationship is always verified:

$$A(1) < A(2) < A(3)$$

where $A(i)$ is the i^{th} measurement done after sample collection.

In our case, the general relationships are:

$$A(1) > A(2) > A(3), \text{ or}$$

$$A(1) < A(2) > A(3)$$

Both relationships show evidence that in the sampled drinking waters, progenies are manifold in excess of ^{226}Ra . Nevertheless, it is remarkable that the activity values are quite high, compared with the world quoted mean value of 0.5 mBq.L^{-1} for ^{226}Ra in drinking water [8].

Estimation of additional lifetime risk of cancer associated with drinking water that has a combined concentration of ^{226}Ra and ^{228}Ra of 5 pCi.L^{-1} of water (0.19 Bq.L^{-1}), is about 1 in 10,000 [9]. The risk doubles to 2 in 10,000 at 10 pCi.L^{-1} (0.37 Bq.L^{-1}) and triples to 3 in 10,000 at 15 pCi.L^{-1} (0.56 Bq.L^{-1}). This analysis means that if 10,000 people were to consume 2 liters of water containing 5 pCi.L^{-1} (0.19 Bq.L^{-1}) of radium every day for 70 years, one additional fatal cancer would be expected in the 10,000 people exposed. Consequently, USEPA (U.S. Environmental Protection Agency) regulations set the drinking water standard at an average annual combined concentration of ^{226}Ra and ^{228}Ra to 5 pCi.L^{-1} (0.19 Bq.L^{-1}) [10-11]. In order to assess the risk associated to the consumption of drinking water in the region of study, we have plotted the ^{226}Ra concentration level map (see figure 4). It shows that for most part of the region of study, ^{226}Ra activities are well above the USEPA acceptable limit.

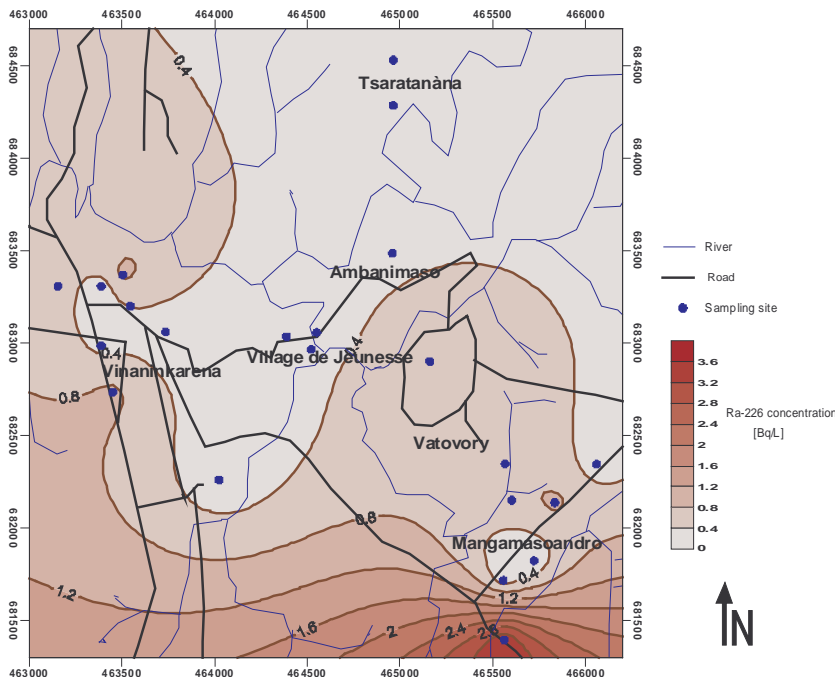


Figure 4 : Map of ^{226}Ra concentration level

4. CONCLUSION

Radium-226, radon-222 and their progenies were measured in-situ in drinking water samples, from a high radioactivity area of Madagascar. The activities found are well above the world mean activity values, as well as the acceptable limits. As these values are from the sole ^{226}Ra , contribution of other radionuclides, to the absorbed dose raises concern to the use of these waters by the local population. Moreover, the increased gross alpha-beta activity resulting from radon progenies raises the issue of health effects. The potential for rapid spring-to-mouth delivery, which is frequent in rural area, may expose the water consumer to internal irradiation by short-lived alpha particle emitters. Legislation must take into account such eventuality.

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