Investigating the spatial anisotropy of soil radioactivity in the region of Vinaninkarena, Antsirabe—Madagascar

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A study was conducted in the region of Vinaninkarena-Antsirabe, to investigate the spatial distribution of the environmental radioactivity. Forty-two (42) top soil samples were collected from two different sampling points. They were analyzed for uranium and thorium series by gamma spectrometry. In order to determine radioactivity spatial structure, semi-variance analysis was used. From 82 samples and 840 paired data, semi-variances were computed, variograms charted and modelled. Results showed that spatial dependence ranges vary from 100 m to 300 m. Moreover, spatial anisotropy is also detected. Such result allows optimizing sampling design for future mapping of the environmental radioactivity of the region.

1. INTRODUCTION

The region of Vinaninkarena, a few kilometres south of Antsirabe, in central Madagascar (47º02’40”E, 19º57’17”S), is known to be a high natural radioactivity area. Uranium ore was extracted in the region during the 1950s and early 1960s by the French Commissariat à l’Energie Atomique (CEA). In the mid-1960s, the rich Katanga uranium deposits in Zaire and in Gabon provided a far cheaper and more reliable source of uranium, resulting in closing of the Madagascar uranium operation. But recently, due to worldwide concern on shortage of traditional energy sources, there is a renewed interest in the uranium-thorium deposits of the region.

In the meantime, continuous studies of the environmental radioactivity of the region were conducted by teams from Madagascar-INSTN (Institut National des Sciences et Techniques Nucléaires). Raoelina Andriambololona et al. showed that the soil of the area is a major source of radon escaping into the atmosphere [1]. Soil measurements conducted in 1999 using gamma spectrometry, showed that high radioactivity in the region originates mainly from the uranium series, with activity concentrations up to 200 $kBq.kg^{-1}$ [2].

For radioactivity baseline study, the soil of a studied area is usually sampled using a transect or grid pattern. For this purpose, it must be ensured that an adequate number of samples is taken to allow a reliable interpolation for areas not exhaustively sampled. Geostatistics can be used to characterize the spatial behaviour and spatial distribution of a parameter and to use this information to predict the value of this variable between sampled points and to minimise estimation error [3]. Elaboration and development of variogram is an important and necessary step for finding and modelling spatial structures in the data. The objective of this investigation is to determine the spatial structure of soil radioactivity in the region of Vinaninkarena, Antsirabe—Madagascar, by means of the variogram.

2. MATERIALS AND METHODS

2.1. Sampling

A field campaign was conducted in the year 2004. The area is geomorphologically divided in two parts: the eastern side is hilly with gentle to steep slope, whereas the western side is rather level. To represent the two landscapes, two sites, centred at P1 and P2, were sampled. P1 and P2 are distant of c.a. 2,380 km from each other. Sampling was done following two cross-shaped transects. The first one is oriented in E-W direction, the second one in N-S direction. Soil was sampled at 10–15 cm depth. A Magellan GPS 315 set was used to determine geographical coordinates. The step distance was maintained at c.a. 1" (30 m). Apart from P1 and P2, twenty samples were sampled per transect, giving a transect length of 600 m. In all, 82 samples were collected for analysis.

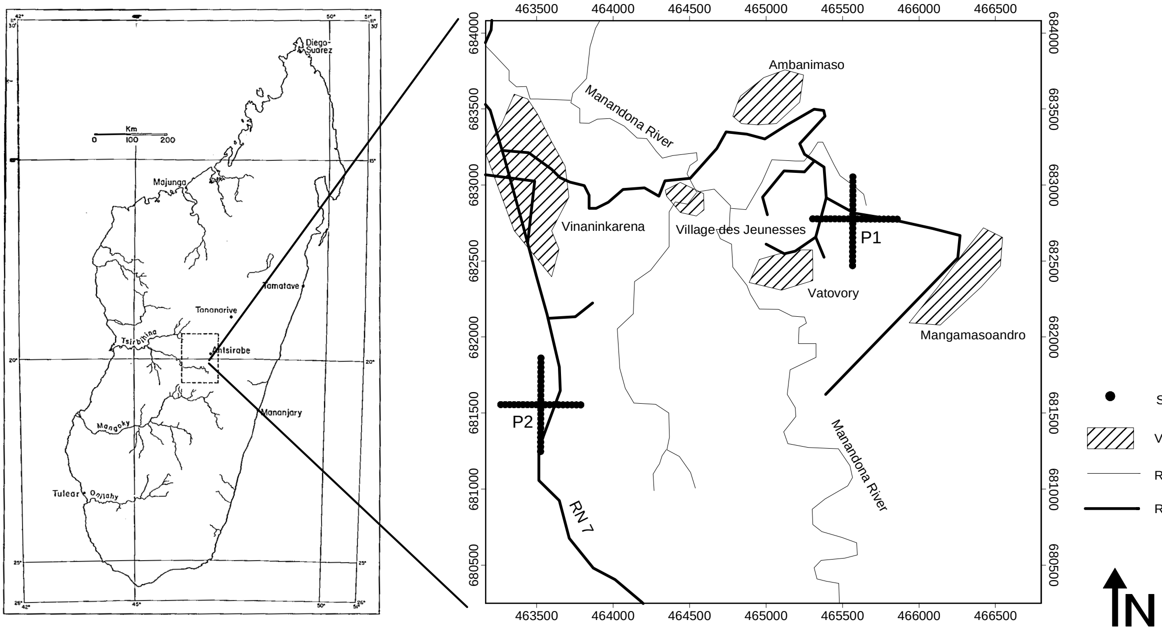

Figure 1: Map of the study region and sampling pattern for at P1 and P2

2.2. Measurement

After humidity correction, the samples were sealed in 100 cm^3, cylindrical, air-tight plastic containers, and stored to allow secular equilibrium between parents and short-lived progenies. Measurements were performed using a high-resolution HPGe γ-spectrometry, with 30% relative efficiency and energy resolution of 1.8 keV at 1332.5 keV, allowing the determination of the uranium and thorium series radionuclides. Depending on the sample activity, counting time varied from 3 to 15 hours. Data acquisition was controlled by Genie-PC Basic spectroscopy software (Canberra, version for OS/2). Finally, activity data were processed to investigate for spatial variability.

2.3. Geostatistics

The geostatistical properties of the data are determined based on the geostatistical model of regionalized variables [4] [5]. To characterize data correlation of paired data, covariance is usually used. For a paired variable (z_1, z_2), it is defined as:

$$C_{1,2} = \frac{1}{N}\sum_{i=1}^{N}\left\{(z_1-\bar{z}_1)(z_2-\bar{z}_2)\right\} = E\left[(z_1-\bar{z}_1)(z_2-\bar{z}_2)\right] \tag{1}$$

To be relevant to spatial correlation study, a weak (second order) stationarity is requested. It means the constancy of the mean, variance and covariances that depend only on separation (lag *h*) and not on absolute position. In that case, we can define the auto-covariance function *C(h)* :

$$\begin{aligned} C_{x,x+h} &= E\left[(z(x)-\bar{z})(z(x+h)-\bar{z})\right] \\ &= E\left[\left(z(x)z(x+h)-\bar{z}^2\right)\right] = C(h) \end{aligned} \tag{2}$$

Furthermore, to overcome departure from weak stationarity, Matheron proposed to use the variances of differences instead of correlation. The semi-variance at lag h, $\gamma(h)$ is then defined as :

$$\begin{aligned} Var\left[z(x)-z(x+h)\right] &= E\left[\left(z(x)-z(x+h)\right)^2\right] \\ &= 2\gamma(h) \end{aligned} \tag{3}$$

Variogram is the plot of the semi-variance as a function of the lag. As the variogram is the cornerstone of geostatistics, it is necessary to estimate and model it correctly. It is mainly described by three parameters:

- the range is the distance beyond which the variogram becomes constant. The correlation range marks the limit of spatial dependence,
- the nugget variance is the variogram value at near zero lag,
- the sill is the variogram value for distance beyond the range of the variogram (see figure 2).

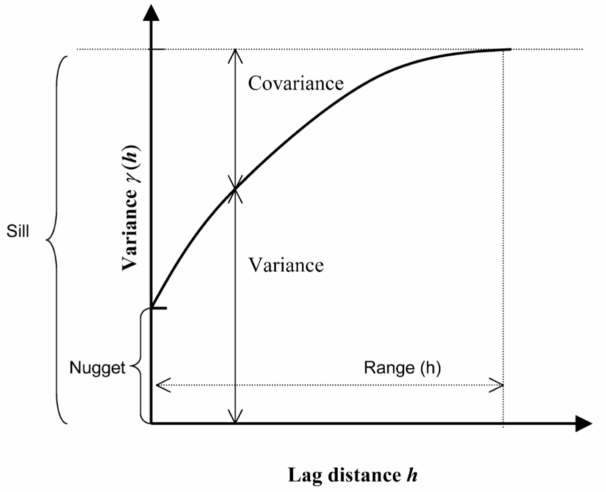

Figure 2 : Typical variogram

3. RESULTS.

For variogram construction, minimum sampling lag distance is 30 m, and maximum lag is 300 m. From 82 samples, 840 paired data were computed for variogram determination and plotting.

Data distribution from uranium series is right skewed. Therefore, before computing, log-transformation was done to normalize the distribution. The uranium directional variograms are shown in figure 3.

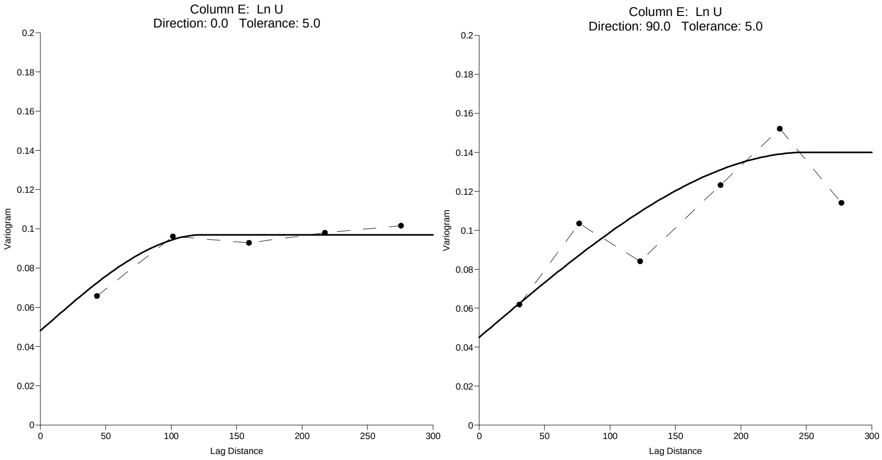

Figure 3 : Variogram plots for uranium series

Along the east-west direction, we obtain a smooth curve, whereas it is erratic along the north-south direction. But we find that the variograms are bounded for both directions. Limits of spatial dependence, as expressed by the range, are 100-125 m along the E-W direction and 200-250 m along the N-S direction.

In the case of the thorium series, data are normally distributed. So there is no need for data transformation.

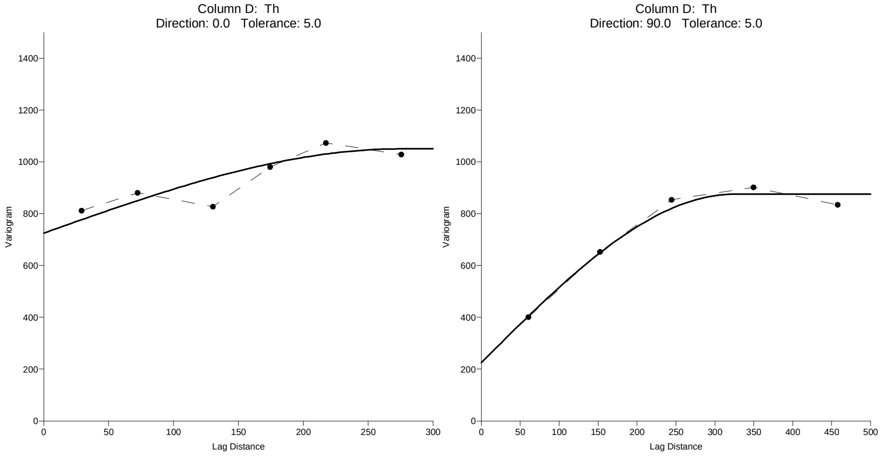

Figure 4 : Variogram plots for thorium series

Thorium directional variograms are shown in figure 4. Unlike the uranium series, we obtain an erratic curve along the east-west direction, and a smooth one along the north-south direction. But like the uranium series, variograms are bounded for both directions. Limits of spatial dependence are 200-250 m along the E-W direction and 250-300 m along the N-S direction.

Table 1 summarizes the results. Whereas all variograms are bounded, spatial anisotropy is detected. Spatial dependence depends on the direction. Spatial dependance limit (range) along E-W direction is shorter than along N-S direction. Moreover, micro-scale variabilities, as expressed by the nuggets, are higher along E-W direction than along N-S direction.

Table I: Experimental variogram parameters

Radionuclides	**Direction**	**Range (m)**	**Nugget ($(Bq.kg^{-1})^2$)**	**Sill ($(Bq.kg^{-1})^2$)**
Uranium	E-W	100-125	0.05*	0.09*
	N-S	200-250	0.045*	0.13*
Thorium	E-W	200-250	725	1000
	N-S	250-300	225	825

(*) from log transformed data

4. CONCLUSION

The variogram model allows one to interpolate the data, i.e. to predict the values at unsampled locations and to estimate spatial interpolation uncertainty. For efficient sampling and mapping, sampling interval should be smaller than the spatial dependance range. Usually, half-range sampling interval is used by practitioners. In our case, incrementation should be in the order of 100 m. Moreover, it should be lower for E-W direction and higher for N-S direction.

Finally, this study shows the necessity of a two stage field work. The first stage is aimed to determine the spatial structure. Main investigation should be done during the second stage, according to results obtained from the first stage.

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