FALLOUT OF ^{238}Pu OVER MADAGASCAR FOLLOWING THE SNAP 9A SATELLITE FAILURE

E. Holm¹, C. Rääf², N. Rabesiranana³, R. Garcia-Tenorio⁴ and E. Chamizo⁴

¹Department of Radiation Physics, Sahlgren Academy at the University of Gothenburg Gothenburg, Sweden

²Medical Radiation Physics, IKVL, Malmö, Lund University, Sweden

³Department of Nuclear Analyses and Techniques Institut National des Sciences et Techniques Nucléaires, INSTN, Antananarivo, Madagascar

⁴Centro Nacional de Aceleradores CNA, University of Seville, Seville, Spain

1 INTRODUCTION

In 1964, a SNAP 9A satellite fuelled by radiothermic generator (RTG) failed to achieve orbit, re-entered the upper atmosphere over Madagascar and burned up. Its RTG released its full ^{238}Pu contents. It was not designed to survive re-entry. The SNAP-9A RTG dispersed plutonium worldwide, nearly tripling the amount of that isotope which became detectable globally in the environment.

After the accident the dispersion was investigated as a function of latitude North and South^{1,2}. It was obvious that most of the fallout of ^{238}Pu was on the southern hemisphere. However no data from Madagascar, where the accident happened, were reported.

The activity ratio $^{238}\text{Pu}/^{239+240}\text{Pu}$ varies depending on source term such as weapon plutonium, fallout from nuclear tests, the Chernobyl accident and releases from nuclear fuel reprocessing plants. The objective of this project was to investigate if there was any close fallout over Madagascar even if it was claimed that all ^{238}Pu was evaporated in the upper atmosphere.

Peat bogs constitute historical archives for atmospheric deposition and have been used in this study to investigate the local deposition from the satellite failure. Several vertical profiles were taken at different sites at Madagascar at about 18°S, 47°E. The samples were sliced in 1-2 cm the first 20 cm and then deeper down in 5 cm slices. Samples were dried, homogenized and analyzed for Pu-isotopes. The dating of the profiles was made by analysis of ^{210}Pb . Plutonium isotopes were measured by alpha spectrometry after radiochemical separation.

The RTG generator might have contained also ^{239}Pu . The $^{240}\text{Pu}/^{239}\text{Pu}$ ratio depends also on the source term. Since the aim was to produce mainly ^{238}Pu the activity ratio $^{240}\text{Pu}/^{239}\text{Pu}$ should be low from the satellite compared to other sources. In order to investigate especially the presence of ^{239}Pu , selected samples were analyzed for ^{239}Pu and ^{240}Pu using mass spectrometric analyses, AMS.

2 METHODS AND RESULTS

2.1 Sampling and Analysis

Two sampling expeditions were performed during July and December 2012. Sixteen cores of peat were collected at different sites. Of those 8 have been subject to analysis. A picture of the sampling in July 2012 is shown in Figure 1. The sampling coordinates are given in Table 1. A steel corer with a diameter of 7 cm was used. The first 20 cm were sliced in 1-2 cm and deeper down in 5 cm sections. The samples were dried and brought to Sweden from Madagascar for further treatment.

Carte 1 : Localisation des sites visités (Le Marais d'Ifanja et le Lac Itasy)

Figure 1 Sampling sites in July 2012.

Table 1 *Sampling sites at Madagascar*

Sample code	Location	Sampling site	Coordinates		
			Latitude	Longitude	Altitude
AI-01	Ifanja marshland	Amparihikely	18°53'33.9''S	46°42'10.5''E	1064 m ± 3 m
AI-02	Ifanja marshland	Amparihikely	18°53'33.8''S	46°42'10.3''E	1064 m ± 3 m
AM-01	Lake Itasi	Ambohimitombo	19°04'00.5''S	46°45'22.8''E	1230 m ± 3 m
AM-02	Lake Itasi	Ambohimitombo	19°04'00.9''S	46°45'22.1''E	1230 m ± 4 m
AM-03	Lake Itasi	Antanimanga	19°03'52.6''S	46°45'02.1''E	1240 m ± 3 m
KH-01	Lake Itasi	Kaihy	19°03'16.2''S	46°48'42.4''E	1240 m ± 3 m
SH-01	Lake Itasi	Sahapetraka	19°05'17.5''S	46°48'09.9''E	1236 m ± 3 m
SH-02	Lake Itasi	Sahapetraka	19°05'14.7''S	46°48'12.1''E	1240 m ± 3 m
T-01	Tampoketsa plateau	Vatoambo	18°09'35.3''S	47°14'03.8''E	1535 m ± 3 m
T-02	Tampoketsa plateau	Vatoambo	18°09'35.2''S	47°14'03.7''E	1535 m ± 3 m
T-03	Tampoketsa plateau	Versité	18°06'35.4''S	47°13'00.3''E	1505 m ± 3 m
T-04	Tampoketsa plateau	Inconnu	18°06'22.3''S	47°14'07.6''E	1546 m ± 3 m
T-05	Tampoketsa plateau	Analatanety	18°06'48.4''S	47°12'10.3''E	Alt.: 1520 m ± 3 m
T-06	Tampoketsa plateau	Analatanety	18°06'51.1''S	47°12'10.9''E	1521 m ± 3 m
T-07	Tampoketsa plateau	Ambohitantely	18°11'50.7''S	47°16'56.4''E	1589 m ± 3 m
T-08	Tampoketsa plateau	Ambohitantely	À 2 m de T07		

The growth rate was determined by analysis of ^{210}Po ($T_{1/2} = 138$ days) in the different layers. ^{210}Po is the decay product of ^{210}Pb ($T_{1/2} = 21$ years). Radiochemical separation and alpha spectrometry were used with ^{209}Po as the radiochemical yield determinant.

^{238}Pu and $^{239+240}\text{Pu}$ were determined by alpha spectrometry after radiochemical separation using ^{242}Pu as the radiochemical yield determinant. Mass spectrometry by AMS (Accelerator Mass Spectrometry) was used to distinguish between ^{240}Pu and ^{239}Pu after a similar radiochemical separation.

2.2 Results and Discussion

The deposition of ^{210}Pb depends on precipitation, surrounding land mass and emanation of ^{222}Rn from soil. The emanation depends on uranium content in soil and rock. Even though Madagascar is a large island, the deposition of ^{210}Pb is lower than for example on the European continent.

The growth rate of the peats using the ^{210}Pb method was determined to about 3 mm per year. This implies that layers around 15 cm depth would correspond to 1964.

The fallout of radioactive elements following nuclear tests is lower at the southern hemisphere due to fewer tests there¹. The maximum deposition is at 45 degrees N and S and is lower at the Madagascar latitude¹.

The activity ratio $^{238}\text{Pu}/^{239+240}\text{Pu}$ in fallout during the nuclear tests in the 1960s was 0.026^2 and would today be 0.017 after decay of ^{238}Pu . By analysing soil samples Hardy *et al.* reported in 1970 a ratio of 0.18 ± 0.04 in the latitude bands, 20-30 and 30- 40 °S, and a ratio of 0.22 ± 0.06 in the 10-20 °S latitude band^{3,4}.

Figure2 Vertical concentration of $^{239+240}\text{Pu}$, ^{238}Pu and ^{210}Pb in in a peat profile from Madagascar as a function of depth.

The major nuclear tests were conducted during the early 1960s and the mean residence time in the stratosphere is 11 months and the peak is quite broad. The fallout of $^{239+240}\text{Pu}$ from the nuclear test period coincides more or less with fallout from SNAP-9A and cannot be resolved in the peat layers. The peak from the satellite might be narrower since it was a single event.

We found a $^{238}\text{Pu}/^{239+240}\text{Pu}$ around 0.12 in 2014 with large variations, corresponding to 0.18 at the time of the accident since the physical life of ^{238}Pu is 89 years (Figure 3). The deposition was slightly higher at Mocambique west of Madagascar showing an activity ratio of 0.20-0.24⁴ in 1970. Even far from Madagascar, at the Antarctic peninsula, values about 0.25 were reported for 1989, using carpets of lichens as bioindicator⁵.

Figure 3 *The activity ratio $^{238}\text{Pu}/^{239+240}\text{Pu}$ in some peat profiles as function of depth.*

In Figure 2 the activity concentrations of ^{210}Pb , $^{239+240}\text{Pu}$ and the activity ratio $^{238}\text{Pu}/^{239+240}\text{Pu}$ as a function of depth in a typical peat core are shown. ^{238}Pu is produced by neutron irradiation of ^{237}Np ($^{237}\text{Np} (n, \gamma) ^{238}\text{Np}, \text{B} \rightarrow ^{238}\text{Pu}$). By multiple neutron capture ^{239}Pu might be formed and present in the RTG. This would give a lower $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio than from nuclear test fallout. Figure 4 shows the activity concentrations of ^{239}Pu , ^{240}Pu and the ratio $^{240}\text{Pu}/^{239}\text{Pu}$ in a peat profile. The mass ratio in layers between 12/20 cm was about 0.15 ± 0.03 at the depth of 15 cm. The isotopic ratio has not been reported to be different at the southern hemisphere compared to the northern one, even if anomalies were found in soils from Chile⁶. The value is lower but not significantly than that from nuclear tests, i.e. 0.18^1 . Therefore we cannot state that there was any significant presence of ^{239}Pu in the RTG.

Figure 4. Activity concentrations of ^{239}Pu , ^{240}Pu and the ratio $^{240}\text{Pu}/^{239}\text{Pu}$ in a peat profile as function of depth.

3 CONCLUSION

Peat bogs are excellent historical archives for atmospheric fallout and for certain radioactive elements such as ^{210}Pb and plutonium isotopes. It was mainly a stratospheric injection and accordingly, the activity ratio $^{238}\text{Pu}/^{239+240}\text{Pu}$ at Madagascar is not much different from other areas at the Southern Hemisphere. The satellite failure in 1964 over Madagascar had no significant local fallout of plutonium from the event. The mass atomic ratio $^{240}\text{Pu}/^{239}\text{Pu}$ is not significantly different from other areas and there is no significant evidence of the presence of ^{239}Pu in the RTG.

References

- 1 UNSCEAR, 1982
- 2 E. Holm, R.B.R. Persson, *Health Physics*, 1975, **29**, 43
- 3 E.P. Hardy, P.W. Krey, H.L. Wolchok, *Nature*, 1973, **241**, 444
- 4 E.P. Hardy, P.W. Krey, H.L. Volchok, *HASL-250, Health and Safety Laboratory, US Atomic Energy Commission*, 1972
- 5 P. Roos, E. Holm, R.B.R. Persson, A. Aarkrog, S.P. Nielsen, *J. Environm. Radioactivity*, 1994, **24**, 235.
- 6 E. Chamizo, M. Garcia-Leon, J.I. Peruchena, F. Cerceda, V. Vidal, E. Pinilla, C. Miro, *Nucl. Instr. Method Phys. Res*, 2011, **269**, 3163.