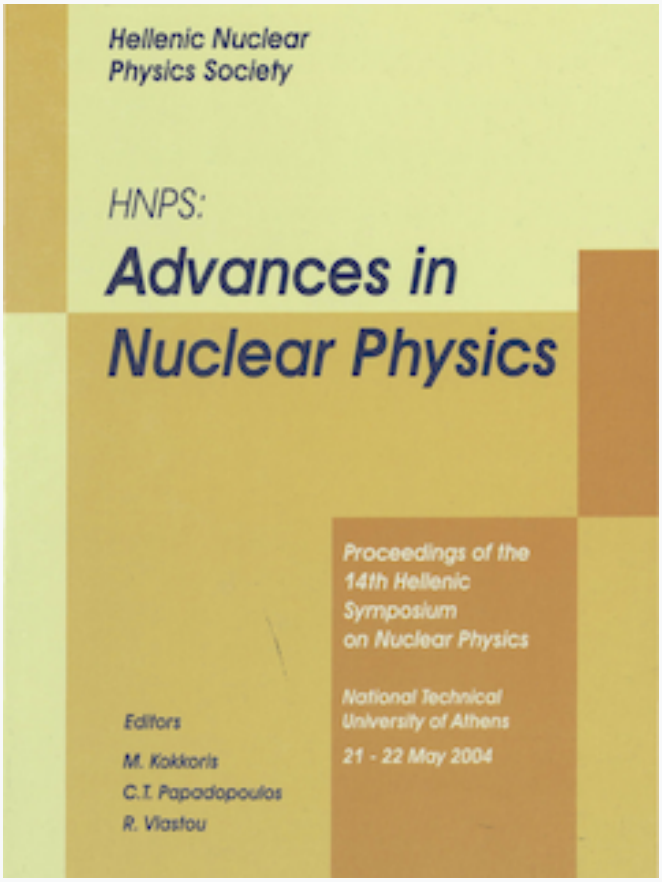
Annual Symposium of the Hellenic Nuclear Physics Society

Τόμ. 13 (2004)

HNPS2004



Radioactive Contamination due to 241Am Lightning Rod Failure

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doi: 10.12681/hnps.2976

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K.L. Karfopoulos, G.N. Papadakos, D.J. Karangelos,
P.K. Rouni, N.P. Petropoulos, M.J. Anagnostakis
E.P. Hinis, S.E. Simopoulos

Nuclear Engineering Department - National Technical University of Athens, National Technical University of Athens, 15780 Athens, Greece

Abstract

It is estimated that until 1978 about 200000 lightning conductor rods with $-\alpha$ emitting sources attached to their end were installed worldwide. The sources were supposed to increase the lighting collection efficiency of these rods through the ionization of the surrounding air. Nevertheless, this improvement has never been established conclusively. Such devices are, in most cases, not accessible by the public; therefore, the dose to the population is considered insignificant. However, the possibility of radioactive material leakage, due to the source attachment failure, and the subsequent contamination of the surroundings that could lead to possible health risk of the public cannot be excluded. In this work, the case of ^{241}Am contamination due to a lightning rod conductor failure is investigated. This contamination was accidentally detected on the surface soil around a laboratory building in the National Technical University of Athens Campus, during a routine in-situ gamma-ray measurement campaign that took place in 2003. A detailed survey revealed that this ^{241}Am contamination was due to the leakage from two lightning rods on the building roof. Consequently the rods were removed from the building and the contamination pattern on the roof and on the surface soil around the building was examined in detail. From the results obtained so far it may be concluded that there exists well localized contamination on the roof and also around the building. It was established that the pathway through which contamination reached the ground was the rainwater drainage system of the building. The gamma ray dose rate due to ^{241}Am contamination found on the roof and on the surface soil is low compared to that due to its natural radioactivity and does not seem to pose any health risk to the people working in the building or to the public.

1 Introduction.

The attachment of radioactive sources to the end of lightning conductor rods, as a means of increasing their efficiency to attract a lightning by ionising the surrounding air, has been used by several lightning conductor rods manufactures in the past. Though the efficiency improvement has never been conclusively proved, it is estimated that until 1978 about 200000 such lightning conductor rods were installed worldwide. Given that such devices are in most cases not accessible by the public the dose to the population is considered insignificant. However, the possibility of radioactive material leakage, due to the source attachment failure, and the subsequent contamination of the surroundings that could lead to possible health risk of the public, cannot be excluded, since the source is open and exposed to weather conditions, and especially rain, wind and any other mechanical stress including those of the lightnings themselves. It should be noted that the situation is not the same with lightning rod installers, where occupational exposure may be significant and doses as high as $9mSvy^{-1}$are reported [Schmitt-Hannig, 1995].

Today, the use of such radioactive lightning rods is decreasing and it has been actually banned in several countries. In Greece import and installation of such lightning rods has been forbidden since 1986. However, between 1960 and 1986 approximately 2500 radioactive lightning rods were installed. Most of them ($\sim$2000) contained ^{241}Am sources and a small part ($\sim$200) ^{226}Ra [Schmitt-Hannig, 1995]. The activity of these lightning rods ranged between 3.7-111 MBq; the most frequent value being 18.5 MBq. The total activity of all the radioactive lightning rods installed in Greece is estimated to about 3GBq of ^{226}Ra and 185GBq of ^{241}Am.

In this research the case of ^{241}Am contamination due to a lightning rod conductor failure is investigated. The ^{241}Am contamination was accidentally detected around a laboratory building in the National Technical University of Athens (NTUA) Campus, during a routine gamma-ray in-situ measurement campaign in the Campus. The campaign was aiming to the calibration of an in-situ Broad Energy Germanium (BEGe) detector. Since ^{241}Am is not a natural radionuclide, it may be found in the environment only as a result of contamination due to: (a) nuclear tests or nuclear accidents, either directly as ^{241}Am or as a ^{241}Pu daughter product, (b) accidents involving consumer products or industrial sources containing ^{241}Am. Following the detection of ^{241}Am a detailed investigation was started in order to determine the origin and the extent of the contamination and its possible health effects –if any.

2 Materials and methods

The Nuclear Engineering Laboratory of NTUA (NEL-NTUA) has been working in the field of gamma spectroscopy since late 70's. Today, NEL-NTUA is equipped with four HPGe detectors of various types, including a Low Energy Ge (LEGe) detector and an Extended Range Ge (XtRa) detector with very high efficiency (relative efficiency 105%). Both detectors are capable of detecting and analyzing low energy photons, such as those emitted by ^{234}Th (63.29keV), ^{210}Pb (46.52keV) and ^{241}Am (59.54keV). For the γ-spectroscopic analysis the in-house developed UNIX software SPUNAL is used. The detection and quantitative determination of ^{241}Am in bulk environmental samples using γ-spectroscopy is not an easy task, since this isotope emits very low energy photons, the most important being those of 59.54keV, with emission probability 35.9%. Self-attenuation of such low energy photons is significant and should be taken into consideration. Self-attenuation highly depends on the sample geometry, the type of material, the packing density and the photon energy and is generally different than that of the efficiency calibration standard. A method has been developed in NEL-NTUA [Anagnostakis, 1995a] in order to take into account the difference in the self-absorbing properties between the calibration source material and the sample material, introducing an efficiency correction factor. The detection limits for ^{241}Am with the instrumentation and the techniques used is in the order of 0.2 Bqkg^{-1}. Using γ-spectrometry for the determination of ^{241}Am has the advantage of a fast, non-destructive technique, while at the same time no sample preparation is needed, contrary to α-spectroscopy, which gives much lower limits of detection but is a destructive, expensive and time-consuming technique, requiring radiochemical separation of ^{241}Am. Among the research activities completed so far at NEL-NTUA is the mapping of the natural radioactivity (^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th, ^{40}K) and the mapping of ten radionuclides detected in the Chernobyl fallout in Greece. During this campaign, where more than 2000 samples from allover Greece were analysed, ^{241}Am was detected in only very few soil samples. The origin of ^{241}Am in these samples is reasonably attributed to be the Chernobyl fallout, since ^{241}Am was also detected in air filters collected during the Chernobyl accident in May 1986 [Simopoulos, 1989]. The maximum ^{241}Am air concentration at NTUA Campus was estimated to 8 mBq/m^3.

Since the mid 80's, NEL-NTUA has been using portable NaI detectors, initially for the in-situ assessment of the gamma-ray dose rate, which in Greece ranges between 20–120nGy/hr, with a mean value ~40nGy/hr [Anagnostakis, 1995b], and later on for the rough in-situ qualitative determination of the soil natural radioactivity. However, NaI detectors, due to their poor resolution, cannot distinguish between photons with similar energies and this poses significant limitations in this application. Furthermore, NaI detectors, due to their aluminum cover, are not capable to efficiently detect low energy photons.

For the in-situ determination of natural radioactivity, especially if the detection of low energy photons is needed, special type Ge detector systems are required, such as an in-situ Broad Energy Ge detector (BEGe). NEL-NTUA was equiped with such a BEGe detector in 2003. It has a rather large surface of 96cm^2, resolution of 1.7keV @1.332MeV, and a carbon-fiber window, which makes it very efficient for the detection of low energy photons, such as those emitted by ^{234}Th (63.29keV), ^{210}Pb (46.52keV) and ^{241}Am (59.54keV). It was during the initial testing and calibration of this detector inside the NTUA Campus, when ^{241}Am was unexpectedly detected in the soil around a certain three-storey Laboratory Building. A visit paid at the building roof showed that there were two ^{241}Am lightning rods installed, indicating that the soil contamination was most probably due to the rod sources. After that, both lightning rods were removed and replaced with non-radioactive ones.

3 Sampling and analysis

In order to investigate the extent of the ^{241}Am contamination in the soil around the building, an in-situ measurement and sampling campaign inside the NTUA Campus was conducted. The following in-situ measurement and sampling locations were selected:

i. Four reference locations inside the NTUA Campus, away from any building, in apparently undisturbed soils.

ii. Seventeen locations around the building where contamination was detected.

In each sampling location a 10000s in-situ measurement with the BEGe detector was conducted and at least one surface soil sample was collected from the top 1 cm surface soil layer (in some cases depth soil was collected as well). Particular care was taken to avoid soil sampling at sites where undulations might have made control of 1cm sampling depth difficult, or where soil was covered with grass or any other kind of vegetation. In Figures 1 and 2 the reference in-situ measurement and sampling locations at the NTUA Campus as well as the in-situ measurements and sampling locations around the building in question are presented respectively. In sampling point #27 where high ^{241}Am contamination was detected, depth soil samples were also collected in order to study the vertical migration of ^{241}Am. A total of 7 soil samples were collected from the reference locations and 25 soil samples were collected around the building. In order to investigate the possible transfer of ^{241}Am to vegetation, mushrooms were also collected around the building. At each in-situ measurement and sampling location gamma-ray dose rate measurements were conducted by means of a gamma dose-rate calibrated NaI detector (Victoreen 190Y). In-situ measurements and gamma dose-rate measurements were also

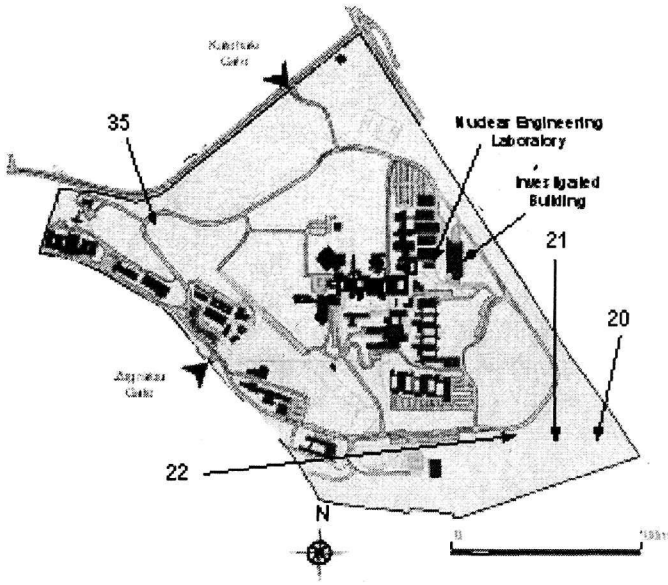

Fig. 1. Mapping of the NTUA Campus with the investigated building and the reference locations.

conducted on the building roof, close to the lightning rods, before and after their removal. The spectra collected from the BEGe detector were analysed using the computer code SPUNAL and the counts per second (cps) for the ^{241}Am 59.54keV photopeak was determined, as a ratio of the area under the photopeak divided by the collect time. The soil samples that were collected were brought to the NEL-NTUA, air-dried under ambient temperature and after all foreign particles, such as glass pebbles, rubbish or grass were removed, their water content was determined. An amble quantity of carefully cleaned soil was used to fill a 0.282cc plastic cylindrical box, of 70mm height. The samples were then analysed using the LEGe and the XtRa detectors. Depending on the detector used and the level of activity of ^{241}Am, the spectrum collect time ranged from 20000 to 200000s.

In order to investigate the pathway through which ^{241}Am was transferred to the soil, a rainwater sampling campaign was conducted. The sampling scheme that was adopted required the sampling of 7L of rainwater through the rain drainage system of the building during heavy rains. A total of 4 rain drainage exits around the building served as sampling points. It is clear that no reproducibility can be obtained during this sampling, since – obviously – the raining conditions are not reproducible. The water that was collected was filtered through 4" glass fibre filters to collect the suspended particulate matter with size greater than approx. 4μm. The filters were then weighted and analysed using gamma-spectroscopic techniques thus resulting to the specific ^{241}Am ac-

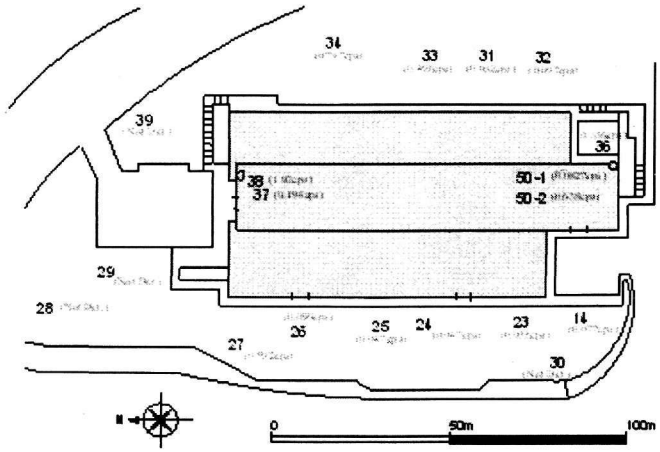

Fig. 2. Mapping of the in situ measurements and sampling locations around the building.

tivity of the water (mgL^{-1}) due to this particulate mater. The filtered water was also analysed by gamma-spectroscopy but no ^{241}Am was detected. Wipe-off tests conducted on the lightning rod antenna were positive for ^{241}Am. The detection of ^{241}Am in water and the wipe-off tests confirmed that the ^{241}Am in the soil around the building was a result of the rainwater agency and the washout of ^{241}Am from the sources at the lightning rods. However, the possibility of ^{241}Am removal through the air could not by overlooked and it was also investigated.

Systematic air sampling is conducted at NEL-NTUA building roof, which is very close (~50m) to the contaminated area since 1999. Air-filters are collected 1-2 times per month, using high volume air samplers ($1000m^3$ in 12h). The air filters are then analyzed using gamma spectroscopic techniques. Americium-241 has never been detected in these air filters.

4 Results

Table 1 presents the results of the in-situ measurements and the samples laboratory analysis for the determination of the radionuclides existing in the soil, for all the investigated locations around the building and also for all the reference locations. It may be concluded that ^{241}Am contamination around the building is well localized and very inhomogeneous. Furthermore, even in the most highly contaminated locations, the ^{241}Am activity is low compared to the natural radioactivity of the soil (especially that of ^{40}K). Figure 3 presents the vertical distribution of ^{241}Am in a relatively highly contaminated location (#27).

Table 1
Soil sample analysis around the contaminated building and at the reference locations

Location	^{241}Am (in-situ)	Surface soil activity in Bq/kg (mean ± combined standard uncertainty %)				
		^{241}Am	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
14	0.08 ± 28%	10.5 ± 6.1	23.5 ± 1.7	23.1 ± 1.9	374 ± 1.6	12 ± 16
23	0.06 ± 41%	7.6 ± 4.4	32.5 ± 1.9	30.4 ± 2.7	415 ± 2.1	8 ± 56
24	0.05 ± 45%	2.8 ± 5.7	22.7 ± 1.4	30.6 ± 1.5	454 ± 1.4	6 ± 36
25	0.05 ± 45%	6.3 ± 4.3	20.0 ± 1.4	29.5 ± 1.5	432 ± 1.4	6 ± 26
26	0.10 ± 22%	6.3 ± 9.5	33.7 ± 1.6	33.8 ± 1.8	447 ± 1.6	9 ± 32
27	0.59 ± 3.9%	17.2 ± 3.2	22.3 ± 1.6	29.4 ± 1.6	399 ± 1.5	5 ± 62
28	ND	ND	17.7 ± 1.6	20.9 ± 1.6	235 ± 1.5	9 ± 23
29	ND	ND	21.2 ± 1.7	31.8 ± 2.0	351 ± 1.7	ND
30	ND	1.9 ± 15	18.2 ± 2.0	21.3 ± 2.0	341 ± 1.7	10 ± 23
31	0.08 ± 26%	8.2 ± 10	20.5 ± 2.1	27.2 ± 2.0	375 ± 1.7	18 ± 10
32	0.10 ± 24%	20.6 ± 3.5	24.1 ± 2.5	31.4 ± 3.0	430 ± 2.4	26 ± 9.5
33	0.50 ± 4.8%	60.3 ± 1.8	23.0 ± 1.7	30.4 ± 1.8	408 ± 1.5	25 ± 4.5
34	0.79 ± 3.2%	71.9 ± 2.0	26.8 ± 1.6	32.2 ± 1.6	420 ± 1.5	15 ± 9.2
36	0.34 ± 6.5%	42.9 ± 2.6	27.0 ± 1.6	31.3 ± 1.7	507 ± 1.5	ND
39	ND	0.5 ± 17	15.1 ± 1.7	18.4 ± 1.7	233 ± 1.6	7 ± 28
Reference locations at NTUA Campus						
20	ND	ND	37.9 ±1.6	30.9 ±1.9	324 ±1.7	59.9 ±1.6
21	ND	ND	20.9 ±1.6	18.7 ±1.8	270 ±1.5	2.9 ±6.0
22	ND	ND	12.1 ±1.5	12.3 ±1.6	181 ±1.5	19.4 ±1.4
35	ND	ND	18.4 ±1.6	22.2 ±1.7	305 ±1.5	3.3 ±5.5

* ND: Not detected (below 0.2Bqkg^{-1})

Table 2 presents the results of the gamma dose-rate measurements for each sampling location as measured using the dose calibrated portable NaI detector (Victoreen 190), and calculated using dose conversion factors for (a) the natural radionuclides existing in the soil, (b) ^{137}Cs that was also detected in the soil and (c) ^{241}Am due to the rods. For the natural radionuclides the dose conversion factors were taken from [UNSCEAR, 1993]. Americium-241 and

^{137}Cs dose conversion factors were taken from [IAEA, SAFETY REPORTS SERIES No. 44]. The gamma dose-rate measured is in satisfactory agreement to the gamma dose-rate calculated according to the dose conversion factors used. From the gamma dose-rate calculations in Table 2 it may be concluded that:

i. On the surrounding ground the percentage of gamma dose-rate due to ^{241}Am was in most cases of the order of 1% of the gamma dose-rates measured.

ii. On the building roof, close to the lightning rod (radius 2m, location 38,) this percentage is estimated to 64% of the gamma dose-rate measured. It should be noted that gamma dose-rate measured in the building roof was significantly lower than that measured in ground level. This is expected since the contribution of the natural radionuclides existing in the soil is reduced due to the distance from the ground and the building materials shielding.

iii. As it was expected, the gamma dose-rate at the building roof was reduced after the lightning rods removal in 2003. However, due to local contamination the dose rate remained relatively high. It is expected that the washout of the contamination with rain water will reduce gradually the dose rate due to ^{241}Am.

For the most heavily contaminated water sample that was collected before the rods removal, the particulate matter was determined to 62mgL^{-1} and its ^{241}Am activity was estimated from the gamma-spectroscopic analysis of the filter to 21mBqL^{-1}. It should be noted here that according to US-EPA regulations the ^{241}Am concentration limits for drinking water is 600 mBqL^{-1}. The analysis of the vegetation collected around the building did not show any ^{241}Am activity present.

5 Discussion

Given that lightning rods are in most cases not accessible by the public, and since, both the ^{241}Am α-particles range in air is a few centimeters and its low energy photons (up to 59.54keV) are easily shielded, it is expected, under normal circumstances, that the dose to the public due to ^{241}Am sources used in lightning rods would be insignificant. Therefore, the only possible significant exposure pathway to the humans is inhalation or ingestion of ^{241}Am. In the case of leakage and subsequent contamination of the surrounding environment, it is deduced from Tables 1 and 2 that any dose due to ^{241}Am existing in soil or contaminated building materials in the roof is much lower than the dose due to the naturally occurring radionuclides, for the same levels of specific activity (Bq/kg).

Table 2
Dose measurements and calculations around the contaminated building, on the building roof and at the reference locations

Location	External gamma dose-rate [nSv/h]			
	NaI measurement	Calculated according to UNSCEAR 1993	Calculated ^{241}Am (*)	Calculated ^{137}Cs (*)
14	48	41	0.6	1.2
23	48	51	0.3	0.8
24	45	48	0.1	0.6
25	45	45	0.2	0.6
26	45	55	0.5	0.9
27	48	45	0.5	0.5
28	38	30	-	0.9
29	50	44	-	-
30	53	36	0.3	1.0
31	40	42	0.7	1.8
32	53	49	0.6	2.6
33	55	46	1.0	2.6
34	53	50	1.3	1.5
36	45	53	1.0	-
39	40	28	0.1	0.7
Building roof				
37[b]	31	-	8.4	-
38[a]	36	-	23	-
50-1[a]	32	-	1	-
50-2[a]	32	-	8	-
50-2[b]	30	-	4	-
Reference locations at NTUA Campus				
20	45	50	-	6.1
21	43	33	-	0.3
22	30	21	-	2.0
35	40	35	-	0.3

*: IAEA, SAFETY REPORTS SERIES No. 44, [a,b]: before and after the lightning rod removal respectively

In order to investigate any possible health effects due to inhalation and ingestion of ^{241}Am, the following worst case scenarios were evaluated, using all data available and making reasonable assumptions:

i. It is reasonable to assume that, leaking ^{241}Am was transferred from the lightning rods to the surrounding building soil only due to rainwater. Therefore, the only possibility to detect it in air is due to re-suspension from the ground. The mean activity of ^{241}Am in the contaminated soil was estimated to ~20 Bqkg^{-1}. From the routine measurements conducted at NEL-NTUA, the maximum concentration of suspended particulate in the air is 140μgrm^{-3}. Even in the most unlikely case that 10% of the suspended particulate matter was ^{241}Am, then the air concentration would be 0.3 mBq/m^3.This value should be compared with the maximum permitted air concentration for **continuous** working (8hrs) or public (24 hrs) daily inhalation: 0.27 mBq/m^3 according to the Greek Regulation for Radiation Protection.

ii. A mean rain height of 15mm per week during wintertime, gives a mean total rainwater volume discharges from the building roof of 8m^3 per day. For the maximum ^{241}Am concentration in water measured 21mBqL^{-1} the total daily discharges of ^{241}Am is 170 Bq/d, which should be compared to the limit for continuous daily discharge for ^{241}Am which, according to the Greek Regulation for Radiation Protection, is 200 Bq/d.

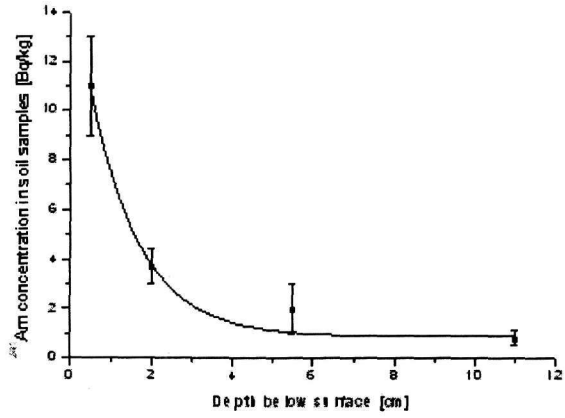

Fig. 3. Vertical distribution of ^{241}Am in sampling location #27.

6 Conclusions

From the results obtained so far it may be concluded that there exists well localized contamination around the building, which is a result of the leakage

of ^{241}Am from the two lightning rods. It was established that the pathway through which contamination reached the ground was the rainwater drainage system of the building. The dose due to ^{241}Am contamination in the soil is low compared to that due to its natural radioactivity and does not pose any health risk to the people working in the building or to the public, however, the lightning rods were removed from the building to fulfill the ALARA (As Low As Reasonably Achievable) principle. It is expected that ^{241}Am will be detected in the rainwater for some time after the removal of the lightning rods due to the washout of contamination from the building roof. The transfer of ^{241}Am in the soil and its migration is of scientific interest and therefore samplings and measurements should continue.

Acknowledgements

The Project is co-funded by the European Social Fund (75%) and National Resources (25%) - (EPEAEK II)- PYTHAGORAS.

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