ASSESSING THE ENVIRONMENTAL POLLUTION USING NEUTRON ACTIVATION ANALYSIS – K_0 METHOD ON MOSSES

Bucsa Adrian Florinel

Institute for Nuclear Research Pitesti – No.1, Campului Street, 115400, Mioveni, Romania

ABSTRACT

The problem of toxic pollutant agents that exists in environment was and still is a main interest subject for nowadays health protection issues. Evaluating the impact of these agents on human health can influence major decisions that are to be taken by the authorities concerning the industrial activities developed in the inhabited areas or from the surrounding areas that directly impact the environment and inferentially the man. Precise measurements of toxic particles in the environment at the level of microelements are essential for exact and correct evaluations of the pollution degree. In order to determine the chemical elements that have negative effects on human health, we used as vegetal indicator, the ground moss. This plant has the ability to retain in the tissue the chemical elements precipitated from the atmosphere, because it is missing the cuticle that would normally prevent the elements from penetrating the cell interior. In order to determine the elements' concentration in the samples, it was employed as analysis method the neutron activation (NAA-k_0) using k_0 standardization. The neutron activation analysis is an analytic technique based on measuring the number and energy of gamma radiation emitted by the radioactive isotopes produced in the sample matrix by irradiation with thermal neutrons in a nuclear reactor. After the irradiation and the specific radioactive decay, the energy spectrum of gamma rays is obtained by measuring the sample with a detection system for high-resolution gamma spectrometry. The sample irradiation was processed in the TRIGA ACPR reactor in the rabbit (D10 location). The study was conducted on a total of four samples of ground moss from the environment (rough samples) and these were processed, irradiated and studied and the results obtained were recorded in a database. The thermal neutron flux supplied by the ACPR reactor in the rabbit location was fair enough for activating elements like manganese, potassium, bromine, europium, lanthanum, arsenic, scandium, antimony, iron. During the entire irradiation period, the presence of the sample in the irradiation location did not disturb the average value of the thermal neutron flux. The differences showed between the elements concentrations in the samples are actually because of the pollution more or less intense in the areas from which they were taken. Hence the quality of the ground moss as a good monitor. From the analysis of the results obtained on these samples and following the comparison with the reference values, it was noticed a slight excess concentration for arsenic and antimony. These overhauls are smaller than 2 ppm. Nevertheless, the alert value is not reached.

Key words: Neutron activation analysis, K_0 method, moss, ACPR

Introduction

The purpose of this paper is to determine the concentrations of the chemical elements that affect the human health, toxic agents that are present in the air and that are retained through fallout in the earth moss used as a genuine pollution indicator.In order to determine these chemical elements that negatively affect the human health, we used the moss as

indicator. This plant has the ability to retain in the vegetative tissue the chemical elements precipitated from the atmosphere, because it lacks the cuticle that would stop the penetration of elements in the cells.In the urban areas, the air's quality is strongly influenced by many human activities. The high population density, the heavy traffic and the house heating system in winter and various industrial activities in the outskirts, influence the concentration of trace elements and radionuclide in the atmosphere. Consequently, the population is exposed to potential adverse effects of changes in the composition of urban air. Thus, monitoring of air quality has become a standard of quality control procedures in the urban environment.This plant has the ability to retain in the tissue the chemical elements precipitated from the atmosphere, because it is missing the cuticle that would normally prevent the elements from penetrating the cell interior.

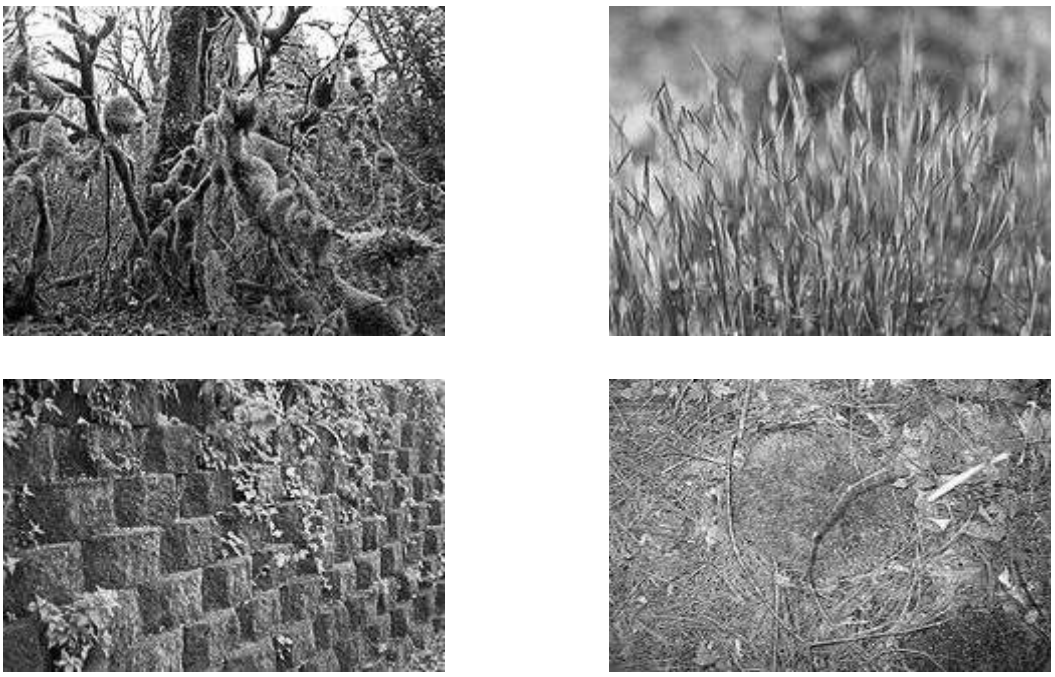

Figure 1. Samples of mosses

Moss samples were picked-up from certain locations (the locations considered with an upper degree regarding environment pollution), and then these were processed (burned in an industrial oven) and finally their residual solid state was inserted in polypropylene vial. These vials were exposed to a neutron flux in TRIGA ACPR rabbit. After been activated in the neutron field, the samples were measured via high resolution gamma ray detection system. The system consists of hyper-pure germanium detector crystal, electronic system (preamplifier, pulsating source, detector polarization unit and amplifier) and a multichannel analyser which was delivered with software specially used for radiation spectrum analysis. This system was used in order to obtain the qualitative analysis of the sample (which are the elements contained in the samples) and the quantitative analysis (determination of the concentrations of the elements discovered) using NAA-k0 standardization method. The results can be used to make a graphical representation of the areas with a high degree of pollution and based on this map decisions can be taken to reduce environment pollution.

Sampling

For the experimental part of this research work there have taken a total of 9 samples from different locations of the Pitesti city.Out of the total samples we choose to analyse only 4 samples due to the lack of time.

Table 1. Samples selected for irradiation in the TRIGA ACPR

Sample no.	Sampling location	Sample weight (g)	Mapping indicative
1	TRIGA Reactor stack	4470E-5	P1
2	Pitesti South train station	6715E-5	P4
3	ARPECHIM Petrochemical Factory	10884E-5	P6
4	ROLAST S.A. (Gavananeighbourhood)	12113E-5	P7

The samples and the flux monitor preparation. Irradiation experiment

After collecting the samples, they were stored in polyethylene bags at room temperature for 10 days after which they were weighed and then placed in an oven at a temperature of 40^{o}C. There were made regular measurements of the masses and then the work continued with drying them in the oven until the land moss's mass remained constant.

The samples burning took place in CTD 2 oven type at a temperature of 450^{o} C until a white coloured ash was obtained. This step was made for the following reasons:

- Water evaporation from the vegetative tissue of mosses; and
- Chemical elements concentration; thru this method it will be easier to determine the chemical elements resulted from neutron activation in TRIGA ACPR rabbit.

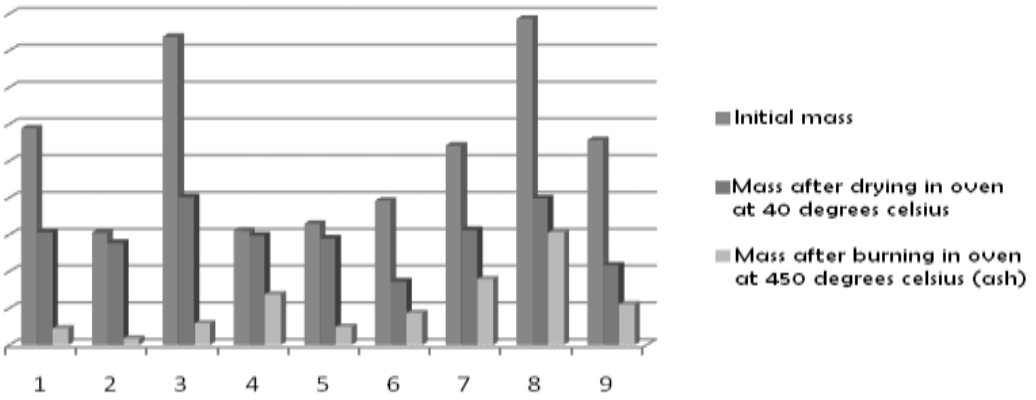

Figure 2. Moss mass evolution from initial sampling until burning (ready to be irradiated in reactor core)

For irradiation experiment, we have chosen a quantity approximately equal from the all four samples; these were weighted in polypropylene vials with small dimensions and then were sealed. The samples irradiation was processed in the TRIGA ACPR reactor in the rabbit (D10 location). The study was conducted on a total of four samples of ground moss from the environment (rough samples) and these were processed, irradiated and studied and the results obtained were recorded in a database.The samples together with flux monitor (foil of Au-197) were irradiated in ACPR rabbit for 4142 seconds. During the entire irradiation period, the presence of the sample in the irradiation location did not disturb the average value of the thermal neutron flux.The calculus made after irradiation showed that the mean value of neutron thermal flux was about 9.47E+11 neutrons / (cm^{2}·sec).It should be mentioned that the

short irradiation period of time did not allow the activation of heavy metal elements. Hence, in 4142 seconds of irradiation we succeed in activation of short life-time isotopes: ^{55}Mn, ^{41}K, ^{45}Sc, ^{121}Sb, ^{139}La, ^{151}Eu, ^{75}As, ^{81}Br, ^{58}Fe.

Qualitative-quantitative measurements of the elements retained in mosses; NAA-k_0 standardization method

Accurate measurements of toxic particles in the environment at the level of microelements are essential for exact and correct evaluations of the pollution degree.In order to determine the elements' concentration in the samples, it was employed as analysis method the neutron activation (NAA-k_0) using k_0 standardization.The neutron activation analysis is an analytic technique based on measuring the number and energy of gamma radiation emitted by the radioactive isotopes produced in the sample matrix by irradiation with thermal neutrons in a nuclear reactor. Usually, the sample matrix together with specific flow monitors, duplicates and standards for items of interest are irradiated for a selected period of time in a neutron flux in the core of the research reactor. After the irradiation and the specific radioactive decay, the energy spectrum of gamma rays is obtained by measuring the sample with a detection system for high-resolution gamma spectrometry.It was made a set of eight measurements on gamma ray HPGe detector for each one of the four samples irradiated. For chemical elements concentrations determination it was used NAA-k_0 standardization method.The high resolution gamma ray detector is shown in the following figure.

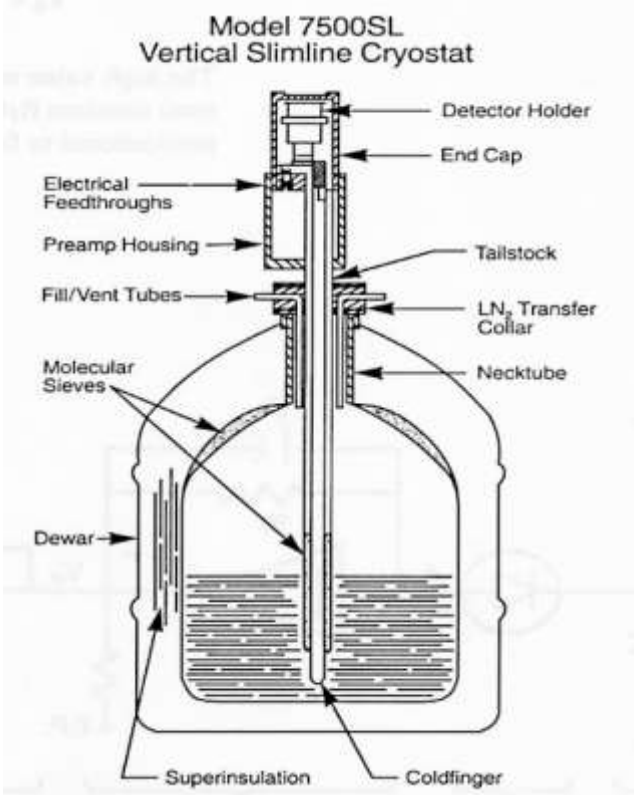

Figure 3. HPGe Gamma ray detector scheme

For this experiment it were used the following samples, apparatus and installations: ground mosses samples, polypropylene vials, TRIGA ACPR reactor (rabbit) and detection system consisted of bin and power supply, high resolution germanium γ-rays detector, amplifier, detector bias supply, pulsating source, multichannel analyser (MCA) and personal computer.

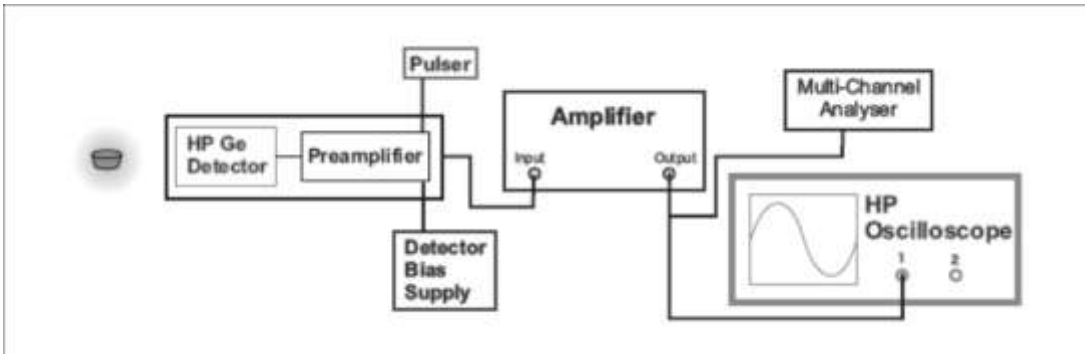

Figure 4: Block diagram for high resolution gamma ray measurement chain

According to NAA-k_0 method, element concentration in a sample is obtained as follows:

$$\rho_a(\mu g/g) = \frac{\left(\frac{N_p/t_c}{SDCW}\right)_a}{A_{sp,m}} \cdot \frac{1}{k_{0,m}(a)} \cdot \frac{G_{th,m}\cdot f + G_{e,m}\cdot Q_{0,m}(\alpha)}{G_{th,a}\cdot f + G_{e,a}\cdot Q_{0,a}(\alpha)} \cdot \frac{\varepsilon_{p,m}}{\varepsilon_{p,a}} \cdot 10^6$$

where,

- ρ_a – analysed element concentration - "a" (μg/g);
- m – co-irradiated monitor for neutron thermal flux value determination;
- N_p – measured net peak aria;
- t_c – counting time [s];
- S – saturation factor; = 1 – exp(-λt_{ir}), where t_{ir}– irradiation time and λ= (ln2)/$T_{1/2}$ with $T_{1/2}$ – half-life time;
- D – decay factor; = 1-exp(-λt_d), where t_d– decay time (starting from the end of irradiation until the measuring start);
- C – counting factor; = [1-exp(-λt_c)]/λt_c;
- W – sample mass [g];
- A_{sp} – (N_p/t_c)/SDCw, specific counting rate, w – monitor element mass [g];
- $k_{0,m}(a)$– k_0 factor of analysed element "*a*" experimental determined relative to mmonitor, defined as: $k_{0,m}(a) = (M_m\theta_a\sigma_{0,a}\gamma_a)/(M_a\theta_m\sigma_{0,m}\gamma_m)$, with M – molar mass, θ – isotopic abundance, σ_0– cross section of (n,γ) reaction at 2200 m·s^{-1}, γ – absolute intensity of gamma ray;
- G_{th} – correction factor for thermal neutrons self-shielding;
- G_e– correction factor for epithermal neutrons self-shielding;
- f – Φ_{th}/Φ_e, thermal neutron fluency rate and epithermal neutron fluency rate ratio; (for ACPR reactor the *f* parameter value at irradiation date was 17.12);
- $Q_0(\alpha)$ – $\{(Q_0 - 0.429)\bar{E}_r^{-\alpha} + 0.429/[(0.55)^\alpha(2\alpha+1)]\}(1eV)^\alpha$, where $Q_0 = I_0/\sigma_0$, with I_0- resonance integral, defined as $I_0 = \int_{0.55\,eV}^{\infty}\sigma(E)dE/E$
- $\bar{E}_r$– resonance effective energy in eV;
- A – the value for the epithermal neutrons fluency rate distribution deviation from ideal shape 1/E, approximated as $1/E^{1+\alpha}$; (for ACPR reactor the α parameter value at irradiation date was 0.01159);
- ε_p – Full-energy peak detection efficiency.

Figure 5 shows the concentration of each detected element by samples, resulted after making the calculus using the NAA-k_0 standardization method.

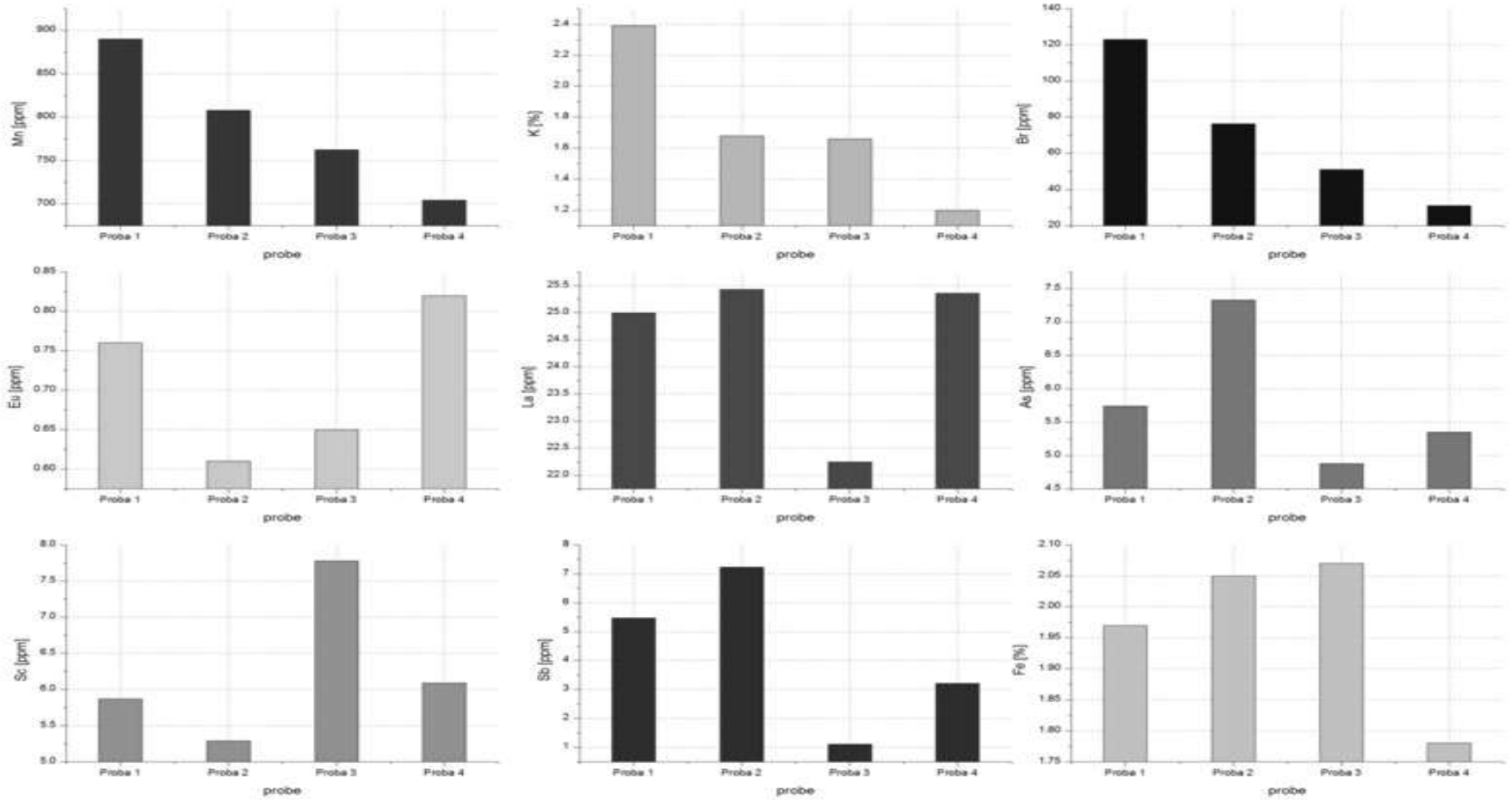

Figure 5. Chemical elements concentrations from each analyzed sample

Conclusion

Thermal neutron flux provided by ACPR reactor in rabbit point had an experimental determined value of 9.47E+11 n/(cm^2·sec). During irradiation period, the presence of samples in irradiation location did not influence the mean flux value. The results indicated the presence of the following elements contained in samples: manganese, potassium, bromine, europium, lanthanum, arsenic, scandium, antimony and iron. The differences showed between the elements concentrations in the samples are actually because of the pollution more or less intense in the areas from which they were taken. Hence the quality of the ground moss as a good bio-monitor.After analysing the results of the samples and after comparing them with reference values from Environmental Protection Agency database, it is noticed a slight excess concentration for arsenic (accepted reference value = 5 ppm) in P1 (TRIGA Reactor stack), P2 (Pitesti South train station) and P4 (ROLAST S.A.) samples. These exceeding are less than 2 ppm. Nevertheless, the alert value is not reached.Another identified element in the samples and for which there is a reference value from EPA is antimony (accepted reference value = 5ppm). In two of four analysed samples there is an exceeding for antimony in P1 (TRIGA Reactor stack) and P2 (Pitesti South train station). For manganese the threshold is 900 ppm but it is not reached. For the other elements found in samples there is no exceeding. It is worth mentioning the fact that no sample showed elements like cadmium, cobalt, mercury, selenium, or zinc.

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