INTRODUCTORY TO NEUTRON ACTIVATION ANALYSIS IN LIFE SCIENCES

FINAL REPORT

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1. INTRODUCTION TO NEUTRON ACTIVATION ANALYSIS (NAA)

Analysis of samples is currently carried out using not only classical methods of analytical chemistry, but also nuclear physics techniques, such as atomic absorption spectrometry (AAS), X-ray fluorescence analysis (XFA), inductively coupled plasma–atomic emission spectrometry (ICP-AES), inductively coupled plasma–mass spectrometry (ICMP-MS), synchrotron radiation, neutron activation analysis (NAA), etc. in this work we will deal with details the NAA. Figure (1) shows the detection limits for different analytical methods.
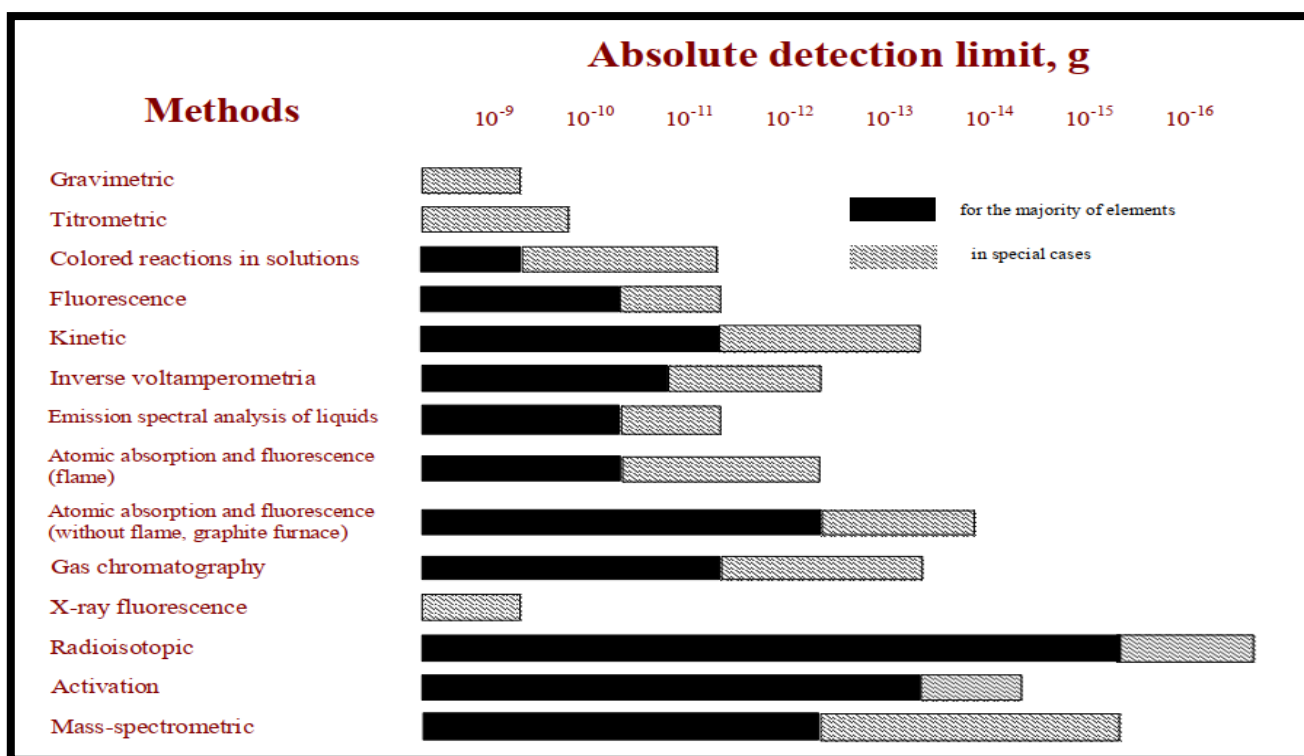

Fig 1 Absolute detection limit in gram for different analytical methods.

1-1 Neutron Activation Analysis (NAA):

NAA is an analytical method, which allows us to do qualitative and quantitative analysis. NAA is based on the transformation of stable atomic nuclei to radioactive nuclei by exposure to a flux of neutrons. After that we can measure the radiation released to know the composition and may be concentration.

The artificial radioactivity was discovered by **Herbert George Wells** in 1933. After that in 1934, Frederic and **Irene Joliot-Curie** succeeded in producing phosphorus isotope P^{30} by bombarding Aluminum with alpha particles.

After that, **Enrico Fermi** noticed that, neutrons could produce artificial radionuclides efficiently more than alpha particles, and here was the beginning where **George Charles de Hevesy** used that to introduce the **N**eutron **A**ctivation **A**nalysis in 1936, by performing rare earth element analysis using neutrons from a Ra–Be source.

The next step in the development of the technique is associated with high-resolution Ge (LI) semiconductor γ detectors and improved multichannel (2048, 4096) pulse analyzers, which appeared in 1965. It became possible to carry out multi element INAA of various samples with small concentrations of elements to be determined. Mathematical and statistical methods for processing of spectrometric information and interpretation of analytical results also made a large contribution to the development of INAA.

1-2 NAA in JINR - REGATA:

As mentioned, the NAA based on the neutrons, and neurons can produce from many sources, but the most powerful source is the reactors. Since the early 1990s, too wide use of neutron activation analysis based on the IBR2 reactor (10^{12}–10^{14} neutrons $cm^{-2} s^{-1}$) (Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research

(FLNP, JINR)) achieved in different fields of science research mainly in biology, medicine, and ecology. Neutron activation analysis workplace at IBR-2 is called "REGATA" Acronym stands for Russian Europe GAte to Asia as shown in figure 2. For example, concentration control of heavy metals and other toxic elements in living organisms, atmosphere, soil, and vegetation and monitoring and fore casting of an increase in the pollution level over time. Pneumatic sample transport systems in reactor NAA laboratories allowed determination of short-lived radionuclides and created prerequisites for automation of the analysis process. Because of developing special neutrons, filters made of substances that have larger cross sections for absorption of neutrons with energies below some specified values. It is possible to use Epithermal neutrons, which employed to determine very small

Concentrations of trace elements in naturally occurring samples. With the aim of increasing accuracy and decreasing the error, certified international standard samples and reference materials used for calibration.
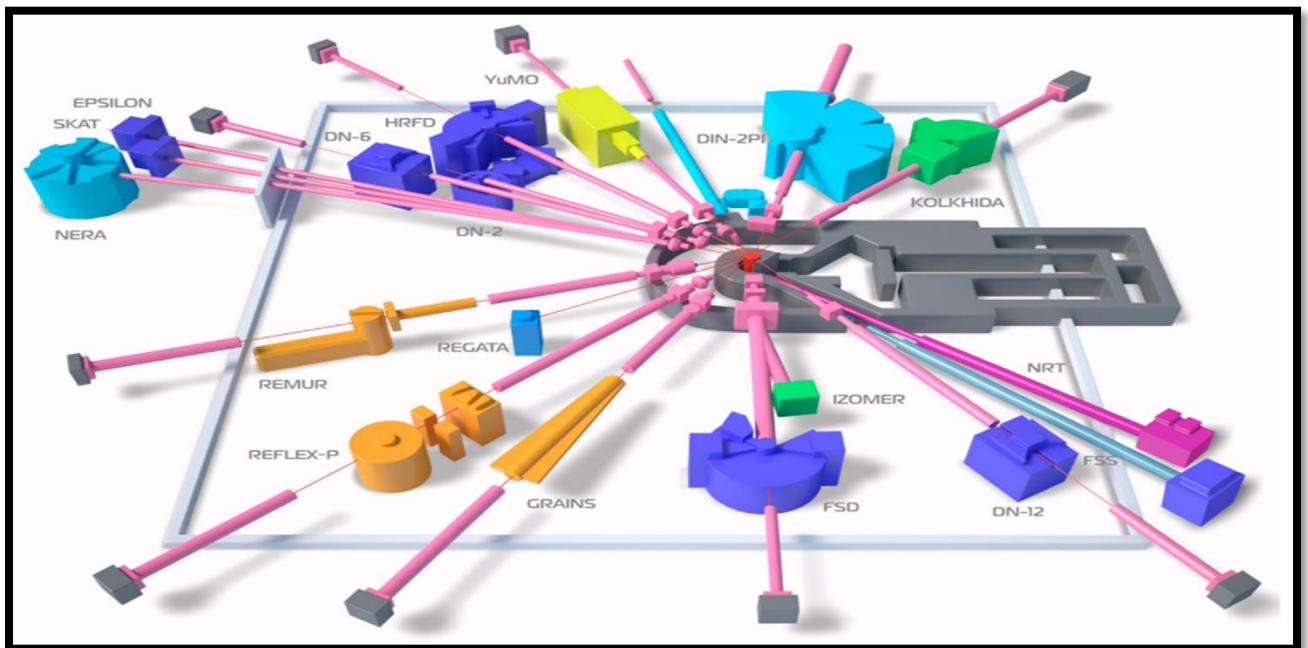
Fig 2 IBR-2 Experimental hall.

There are many types of radiation that can be measured, and is considered the best "gamma-radiation" (Gamma radiation is a high-energy electromagnetic radiation that originates in the nucleus; It is emitted in the form of photons, discrete bundles of energy that have both wave and particle properties). Because of the high selectivity of gamma ray, spectrometry allows us to determine many of radionuclides.

NAA do not need to undergo any chemical treatment, neither during, nor after the activation, so we can call it "non-destructive" (non-destructive is desirable in many applications like nuclear and geochemistry applications). NAA enabling us to detect the traces of light elements in sensitivity up to mg/Kg to µg/Kg. in addition light elements such as H, C, N, O, and Si (the major components in any sample) do not produce any radioactive products so they do not interfere the determination of other elements. In addition, because of the dependence of results in the radiation, which is related to the nucleus properties, the physical or chemical state do not affect the results from NAA.

1-3 Types of NAA:

There are two types of NAA:

I. **Nondestructive NAA**, i.e. The resulting radioactive sample is kept intact. Nevertheless, there will be changes at the nucleus level. Nondestructive NAA contains the following types:

- Instrumental Neutron Activation Analysis Activation INAA
- Epithermal Neutron Activation Analysis Activation ENAA
- Fast Neutron Activation Analysis Activation FNAA
- Cyclic Neutron Activation Analysis Activation CNAA
- In Vivo - Neutron Activation Analysis Activation In-vivo NAA

II. **Destructive NAA**, in destructive NAA the radioactive sample is decomposed or in other wards chemically processed after irradiation.

  This kind including only the following type:

- Radiochemical or destructive neutron activation analysis RNAA or DNNA

1-4 Standardization:

The main product from the NAA process is gamma ray spectrum, and the standardization can be definite the determination of the proportionality factor F (g^{-1}) that relates the net peak areas in the gamma ray spectrum to the amounts of the elements present in the sample under the given conditions of irradiation:

$$F = \frac{A}{W}$$

1. **Absolute standardization**: Here, the values of the physical parameter determining the proportionality factor, $\sigma(E)$, $\Phi(E)$ and ε are taken from the literature and accurate measurement.

$$m = \frac{A_{nc} \cdot W}{0{,}6023 \cdot \theta \cdot \Phi(E)\sigma(E)s.\varepsilon_p .S.D.C}$$

2. **Relative Standardization**: Here, the unknown sample is irradiated together with a calibration sample containing a known amount of the element(s) of interest. The calibration sample is treated under the same conditions as the sample (e.g., sample-to-detector distance, sample size, and if possible composition). It requires standards to have all the elements, and the error mainly depends on m_s and A_{nc}.

$$m = \frac{A_{nc} \; e^{-\lambda t_{e,s}} \; m_s}{A_{nc,s} \; e^{-\lambda t_e}}$$

1-5 Advantages and limitations of NAA:

Advantages of NAA:

1- High sensitivity to a majority of elements, especially to heavy metals with $Z >$ 10.

2- Good selectivity due to specific nuclear physics characteristics of elements.

3- A possibility of simultaneously determining a large number of elements.

4- Independence of the results on the form of chemical compounds.

5- A nondestructive nature, which allows avoiding the risk of contamination of samples with reagents or their incomplete dissolution.

6- Easy procedure for preparation of samples for analysis.

7- Good accuracy, about ±10–15%, in determination of concentrations of the order of ppm (10^{-6} g).

8- A possibility of eliminating systematic errors.

9- A possibility of minimizing the effect of matrix elements of samples.

10 - Ease of preparation of standards for comparison.

Limitations of NAA:
1- Need for nuclear reactor.

2- Work with radioactive materials.

3- Time of analysis.

4- Sample preference.

<h1 style="text-align:center">Section 2</h1>

<h1 style="text-align:center">THEORITICAL CONSIDERATIONS OF NAA</h1>

2-1 Radioactivity:

Radioactivity: Radioactivity may be happen naturally to those unstable naturally occurring nuclides, or artificially after activation.

Radioactive decay: Nucleus may have an excess of energy, which makes the nucleus unstable. In such a case, it releases the excess energy by radioactive decay (called, nuclear disintegration) under emission of nuclear radiation. Which mainly which can summarized to:

1- α - Radiation.

2- β^- - Radiation.

3- β^+ - Radiation.

4- γ -Radiation.

γ -Radioactivity can take place via emission one Gamma, and then the nucleus will reach directly to the ground state. Alternatively, emit two or more Gamma; this is called a Gamma-cascade. This may take place in very short time (µs) or the excited level has a measurable life time from seconds (s) to hours (h) or even longer (The last type denoted as m (metastable)),a simple example is shown in figure 3 .
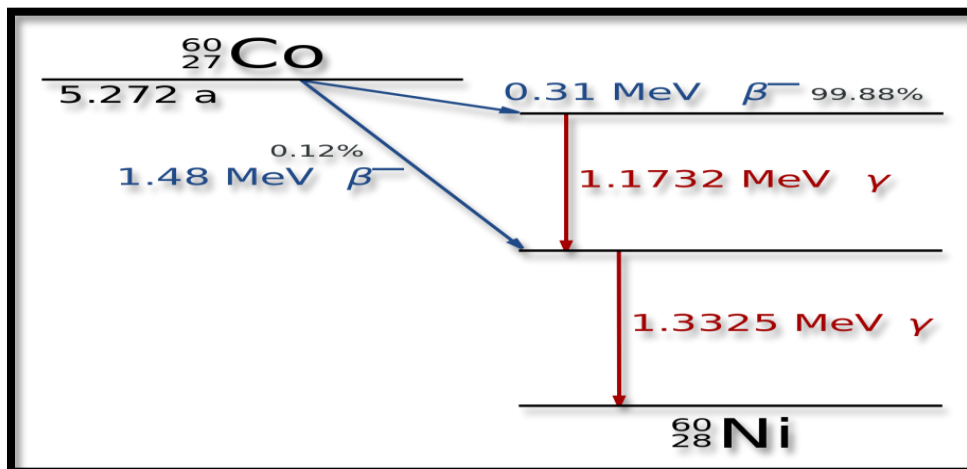

Fig 3 Simplified decay scheme of the radionuclide ^{60}Co.

This leads to a very important definition.

γ- yield: is defined as the fraction of the number of disintegrations that results in γ-radiation. Because of the fact that not every transition in γ- decay results in emission of γ- radiation, sometimes the Gamma is absorbed by the electrons and electron are thrown out of the shell.

Gamma- radiation is chosen for NAA due to the following advantages:

1- Monoenergetic radiation.
2- Characteristic for the emitting nucleus (In most cases).
3- Has a high penetration power (so it will not suffer from the absorption inside the radiative material itself).

As mentioned, the NAA is qualitative and quantitative analysis. So how the NAA can be qualitative? The answer is summarized in the following procedures:

1- Suppose that we have a reaction A' (x,y) B,
2- The produced radionuclide B decays to a stable atomic nucleus C under emission of characteristic γ radiation.
3- Often, characteristic γ-radiation is emitted. By determining the energy of the γ-radiation and using the decay schemes, the radionuclide B may be identified.
4- Eventually, this identification may be even more selective by examining the half-life of radionuclide B.
5- When B has been identified, the nature of the target isotope A' can be derived from knowledge of the nuclear reaction that has taken place.

In addition, how the NAA can be quantitative?

By measuring the intensity of the radiation, i.e., measurement of the activity of radionuclide B. This radioactivity is proportional to the number of target nuclei A' of element A in the irradiated sample. Which follow the simple equation of activity:

$$A = \frac{-dN}{dt} = \lambda N$$

Where, A is the activity, λ is the decay constant and N the number of radioactive nuclei.

2-2 Reactions:

2-2-1 neutrons reactions:

Activation is a process in which the conversion of stable nuclei into radioactive one via nuclear reaction results from bombarding these nuclei with neutrons, energetic charged particles (e.g., protons), or high energy photons. From the name (Neutron Activation Analysis), it is clear that neutron will used for activation. The main product from the activation process (more interest to NAA), will be the characteristic Gamma radiation to be measured.

The main types of reactions based on the neutrons listed below:

1) Scattering
    1- Elastic Scattering (n, n).
    2- Inelastic scattering (n, n').
2) Absorption Reactions.
    1- Radiative Capture (n, γ).
    2- Particle Ejection (n, α), (n, p), and (n, 2n).
    3- Fission (n, f).

In IBR-2 the samples are irradiating in the reactor flux of 5×10^{16} neutrons m^{-2} s^{-1} with the energy 0.5×10^4 eV (epithermal neutrons).

The cross section of thermal neutrons inversely proportional with neutrons energy, and for Epithermal neutrons, the cross section is strongly change in a discrete energy and shows resonance peaks. So, sometimes activation with epithermal reactor neutrons is preferred to enhance the activation of elements with a high ratio of resonance neutron cross section over thermal neutron cross section relative to the activation of elements with a lower such ratio.

The neutron activation of a number of low Z-elements (H, He, Be, Li, B, C, N, 0) and of a few high Z-elements (Bi, Tl, Pb) is characterized by:

1- Very low activation cross sections,
2- Activation products with very short half-lives (in the order of seconds),
3- The emission of radiation, which does not interfere with the measurement of the radiation emitted by the other activation products.

2-2-2 Gamma reactions:

In activation analysis, gamma-radiation is almost exclusively measured. In order to understand the measuring of Gamma-radiation, the interactions of Gamma-radiation with matter must be well understood. Reaction type of incident gamma rays depends on the energy of incident gamma.

The main types of reactions based on the Gamma-radiation listed below:

1- **Photoelectric effect:** This reaction is most predominant with low energy gammas.

Effect. In the detector material, the Gamma-ray causes ejection of an electron from one of the inner electron shells of the atoms. The energy of the photon, minus the binding energy of the electron, is transferred to this electron (photo-electron). The resulting vacancy in the electron shell is filled up by the emission of x-radiation or Auger electrons. The energy of the ejected electron, x-radiation or energy of the Auger electrons is absorbed in the detector, which implies that the total photon energy is transferred to the detector. It results in a photo-peak or full-energy peak in the Gamma-ray spectrum, which reflects the number of recorded events as a function of their energies.

Due to the detection and pulse processing, the shape of photon peak does not appear as a line, but can approximated by a Gaussian broadening or Bell shape (In spite of the monoenergetic nature of Gamma-radiation). The smaller the width of the peak, the better the resolution of the detector (full width on half maximum (FWHM)).

The probability of photoelectric effect increases with atomic number Z and decreases with Gamma-radiation energy.

2- **The Compton effect:** This reaction becomes important for gamma energies of about 0.1 MeV and higher.

In Compton Effect the incident Gamma-radiation will partially absorbed by electron followed by ejection of electron (Compton electron) and low energy Gamma-radiation (Compton gamma). The energy of the Compton electron is absorbed in the detector, and the Compton gamma may escape from the detector. In that case, the gamma-ray spectrum shows a Compton continuum at the lower energy side of the photo-peak. However, when Compton gamma is also

absorbed, summation of all deposited energies results 1n a contribution to the full energy photo-peak.

The probability of Compton Effect is not dependent on atomic number Z, and slightly dependent on gamma energy.

Compton continua are a complication in gamma-ray spectrometry since photo-peaks of low energy gammas are often located on a continuum of a high-energy gamma ray. Thus, detection of a peak on a continuum depends on the signal-to-noise ratio.

3- **The Pair Production:** The incident gamma must have at least 1.02 MeV energy.

Gamma rays with energies higher than 1022 KeV cause the formation of an electron positron pair when the Gamma ray approaching the electromagnetic field of the nucleus. The rest of the energy (more than 1022 KeV) will be transferred to the particles in the form of kinetic energy and deposited in the detector resulting in the first pair-peak at an energy 1022 keV lower than the energy of the original gamma.

The positrons annihilate with electrons in the detector, which is characterized by the emission of two annihilation quanta, each with an energy of 511 keV.

When both quanta escape from the detector, only the first pair-peak is measured. If one of the quanta is absorbed via photoelectric effect in the detector, a peak at an energy 511 keV lower than the energy of the original gamma results. When both annihilation quanta are absorbed in the detector via photoelectric effect, they contribute to the photo peak.

The probability of the pair production increases with the atom number of the detector material (roughly proportional to Z), and increases with the gamma energy.

Sometimes the radionuclide decays via gamma cascades. Coincident gammas may be detected simultaneously, resulting in a peak in the spectrum at an energy equal to the sum of the energies of the coincident gammas, the sum-peak.

Figures 4 shows the three types of gamma interactions:
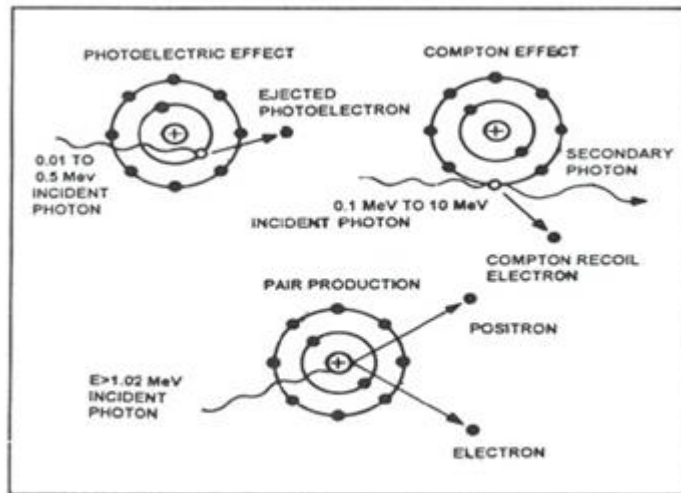
Fig 4 Interactions of gamma radiation with matter.

2-3 Gamma Detection system setup:

The measurement of Gamma-radiation in NAA as a spectroscopic method. The differences with other spectroscopic methods for elemental analysis in:

1- In activation analysis, the Gamma-radiation originates from the atomic nucleus, not from election shells. So its energy in order of MeV.
2- The radiation is measured not during the excitation, as the nuclear reaction, but at a given time after the end of activation (short lived – long-lived 1 – long-lived 2, depending on the decay time).
3- The method is isotope-characteristic, in contrast to other methods in which the total of all isotopes of the element is assessed. Because isotopic abundance is well-known fraction of every element (with only a few also well-known exceptions), determination of isotope implicitly means determination of elements.

Gamma-radiation detectors are based on the principle that, upon interaction with matter, the energy quantum of a photon is converted into a product that can be detected by physical or electronic means.

The radionuclide activity concentrations in JINR are measured using a High-Purity Germanium Detector (HPGD). The gamma spectrometry system consists of an N-type HPGD (CANBERRA), mounted in a cylindrical lead shield (100 mm thick) and cooled in liquid nitrogen, and coupled to a computer based Multi-Channel Analyzer(MCA).

The relative efficiency of the detector was 40 %, with an energy resolution of 2 keV at a gamma ray energy of 1332 keV of ^{60}Co. LABSOCS efficiency calibration software, mathematical efficiency calibration software and software program (Genie 2000) were used to calculate the activity concentrations of the samples, taking care to subtract the natural background level.

Some problems appear during the analysis. One of them is, there are more than one reaction induced by neutrons producing Al^{28} & Mg^{27}: ^{27}Al(n, γ)^{28}Al; ^{28}Si(n, p)^{28}Al, ^{31}P(n, α)^{28}Al & ^{26}Mg(n, γ)^{27}Mg; and ^{27}Al(n, p)^{27}Mg. Which leads to less accuracy and errors in determination of concentrations of the corresponding elements.

Another problem is the high background from Compton effect, which can be overcome by measurements are sometimes carried out after an appropriate delay time which is beneficial if the main interfering radionuclides have relatively short half lives.

Another problem in IBR-2 is the limitation to detect (Cu, Cd, Pb & some light elements Z < 10) which are permanently installed in the reactor irradiation channel or placed into transport containers. So they can detect by another technique like atomic absorption spectrometry (AAS).

The data in Table 1 are for the elements determined from short-lived products of (n, γ) reactions with half-lives below 15 h, and the data in Table 2 are for the elements determined from long-lived products of those reactions. These tables present radionuclides and their associated γ lines most often used to determine the corresponding element and also fractions of decays where each γ ray is emitted in coincidence with the one or more than one γ ray.

Table 1. Some nuclear data for the short-lived nuclides ($T_{1/2} < 15$ h).

Element	Radionuclide	$T_{1/2}$	I_0/σ_0^*	γ ray energy, keV	% of coincidences
Na	^{24}Na	15.0 h	0.59	1368	100
				2754	100
Mg	^{27}Mg	9.5 min	0.68	842	2
			1013	0	
Al	^{28}Al	2.3 min	0.74	1780	0
S	^{37}S	5.1 min	1.1	3090	0
Cl	^{38}Cl	37.3 min	0.69	1600	100
				2170	100
K	^{42}K	12.4 h	0.97	1524	0
Ca	^{49}Ca	8.8 min	–	3100	0
Ti	^{51}Ti	5.8 min	0.66	928	1
V	^{52}V	3.8 min	0.55	1434	1
Mn	^{56}Mn	2.58 h	1.05	847	48
Ni	^{65}Ni	2.56 h	0.64	1115	40
				1481	0
Cu	^{64}Cu	12.8 h	1.1	511	100
	^{66}Cu	5.1 min	1.01	1039	3
Zn	^{69m}Zn	13.8 h	2.9	439	0
Ga	^{72}Ga	14.1 h	6.6	835	90
Sr	^{87m}Sr	2.83 h	5.7	388	0
Zr	^{97}Zr	17.0 h	231	747	7
	^{97}Nb	72 min		665	0
Mo	^{101}Mo	14.6 min	18.8	2080	0
	^{101}Tc	14.0 min		307	10
In	^{116}In	54.0 min	16.3	417	100
				1097	100
				1293	100
I	^{128}I	25.0 min	23.7	441	15
Cs	^{134m}Cs	2.90 h	13.2**	127	0
Ba	^{139}Ba	82.9 min	0.89	1430	0
Sm	^{155}Sm	22.4 min	6	104	0
Eu	^{152}Eu	9.3 h	3.9**	122	100
				963	0
Dy	^{165}Dy	139.2 min	<0.5**	94	0
Er	^{171}Er	7.5 h	5.3**	124	100
				308	100
U	^{239}U	23.5 min	100	75	0

Table 2. Some nuclear data for the long-lived nuclides ($T_{1/2} > 15$ h)

Element	Radionuclide	$T_{1/2}$	I_0/σ_0^*	γ ray energy, keV	% of coincidences
Na	^{24}Na	15.0 h	0.59	1368	100
				2754	100
Ca	^{47}Ca	4.53 d	1.28**	1300	0
	^{47}Sc	3.43 d		160	0
Sc	^{46}Sc	83.9 d	0.44	889	100
				1121	100
Cr	^{51}Cr	27.8 d	0.49	320	0
Fe	^{59}Fe	44.6 d	1.3	1095	0
				1292	5
Co	^{60}Co	5.26 y	1.99	1173	100
				1332	100
Zn	^{65}Zn	244 d	1.91	1115	0
Ga	^{72}Ga	14.1 d	6.62	630	100
				835	100
As	^{76}As	26.5 d	13.6	559	20
Se	^{75}Se	120.4 d	10	122	100
				136	100
				265	100
				280	100
				401	87
Br	^{82}Br	35.3 h	18.5	554	100
				777	100
				1044	100
				1475	61
Rb	^{86}Rb	18.7 d	11.3	1078	0
Sr	^{85}Sr	64.9 d	13.3**	514	0
Zr	^{95}Zr	64.0 d	4.61	724	0
				756	0
	^{95}Nb	35.2 d		765	0
Mo	^{99}Mo	66.0 h	53.1	181	33
				740	100
	^{99m}Tc	6.0 h		143	0
Ag	^{110m}Ag	250 d	15.4	657	100
				764	100
				884	100
				937	100
				1387	100
Cd	^{115}Cd	53.5 h	78**	530	2
	^{115m}In	4.50 h		335	0
In	^{114m}In	50.0 d	131**	192	0
Sn	^{113}Sn	115 d	30	393	2
	^{117m}Sn	14.0 d	82	158	100
Sb	^{122}Sb	2.80 d	33.9	564	5
	^{124}Sb	60.3 d	30.2	603	78
				1692	100
Cs	^{134}Cs	2.05 y	12.4	605	100
				796	100
Ba	^{131}Ba	11.5 d	25**	123	100
				216	15
				373	35
				493	100
La	^{140}La	40.2 h	1.32	326	100

Element	Radionuclide	$T_{1/2}$	I_0/σ_0^*	γ ray energy, keV	% of coincidences
				487	100
				815	100
				1596	73
Ce	^{141}Ce	32.5 d	0.82	145	0
Nd	^{147}Nd	11.1 d	2.3**	91	6
				531	0
Sm	^{153}Sm	46.8 h	14.4	103	40
Eu	^{152}Eu	12.4 y	1.15**	122	100
				965	100
				1087	0
				1408	100
	^{154}Eu	8.5 y	4.21	123	100
				724	100
				1278	100
Gd	^{159}Gd	18.0 h	24**	363	0
Tb	^{160}Tb	72.1 d	17	87	100
				298	100
				879	100
				966	50
				1178	100
Ho	^{166}Ho	26.9 h	10.3	81	0
Tm	^{170}Tm	128.6 d	13.5**	84	0
Yb	^{169}Yb	30.7 d	4.6**	63	100
				94	100
	^{175}Yb	4.2 d	0.46	283	100
				396	0
Lu	^{177}Lu	6.7 d	<0.5**	113	50
				208	100
Hf	^{181}Hf	42.4 d	2.62	133	100
				482	100
Ta	^{182}Ta	115.0 d	33.5	100	100
				179	100
				222	100
				264	100
				1189	100
W	^{187}W	23.9 h	13.2	134	100
				478	100
				686	0
Ir	^{192}Ir	74.2 d	3.8**	308	84
				316	100
				468	100
Au	^{198}Au	2.70 d	15.7	412	0
Hg	^{197}Hg	64.1 h	0.49	77	0
	^{203}Hg	46.9 d	0.98	279	0
Th	^{233}Pa	27.0 d	11.5	300	100
				312	0
				416	0
U	^{239}Np	2.35 d	100	106	100
				228	100
				278	0#

The data from [2].

Section 3

3-Experimental work.

To perform the NAA the following steps should be followed:

3-1 Sample collection:

Most samples are plant-type (as Moss), soil and sedimentation. The collected plant should be wrapped and isolated from the air in order not to further absorb pollutants from the air.

Soil or sedimentation samples should not be collected with metal tools in order to avoid contaminate the samples with the metal. As shown in figure 5.

Fig 5 The right way to collect moss

Table 3 shows the detection limits for elements in materials of plant and soil origin, mg/kg.

Table 3: the detection limits for elements in materials of plant and soil origin, mg/kg

Element	Plant	Soil	Element	Plant	Soil
Na	2	10	K	200	1500
Ca	700	4000	Sc	0.001	0.02
Cr	1	1	Fe	8	100
Co	0.02	0.3	Ni	2	30
Zn	0.4	6	Ga	2	10
As	0.2	0.8	Se	0.1	1
Br	0.3	0.8	Rb	0.4	6
Sr	5	60	Zr	5	80
Mo	4	10	Ag	0.2	2
Cd	3	8	Sn	10	20
Sb	0.02	0.2	Te	0.3	3
Cs	0.02	0.3	Ba	10	40
La	0.1	0.3	Ce	0.2	1
Nd	0.7	8	Sm	0.01	0.03
Eu	0.006	0.05	Tb	0.008	0.1
Yb	0.03	0.2	Lu	0.004	0.02
Hf	0.01	0.1	Ta	0.01	0.2
W	0.3	1	Re	0.08	0.2
Os	0.1	0.6	Ir	0.0006	0.004
Au	0.003	0.01	Hg	0.5	0.4
Th	0.01	0.1	U	0.2	2

3-2 Sample preparation:

First, the samples are dried in special dryers, and the samples are grinded and pelletized in press-forms and precisely weighed. Figure 6 shows the sample processing.

If it is already known that there are high concentrations of heavy materials in the samples, 0.1 g is taken and pelletized. Other wise 0.3 g are taken.

Fig 6. Photos showing sample processing (drying → grinding → weighting → pelletizing → packaging)

3-3 Packaging samples:

There are two types of packaging, Short-term irradiation samples are heat-sealed in polyethylene foil bags, while the samples for long-term irradiation ones are packed in aluminum cups.

Also samples of certified reference materials (CRMs) are pelletized and packed in the same way to be irradiated and measured with the samples. It is used later for the calculation of the concentrations using the comparative method. These samples are placed in the top, middle and bottom of transport containers to make sure the regular irradiation of all samples.

3-4 Transport containers:

Samples are packed in transport containers made of polystyrene for short time irradiation and aluminum for long time irradiation, see figure 7. Transport containers containing the samples are irradiated at REGATA (the pneumatic system used for irradiation at Pulsed fast reactor IBR-2).

Fig 7 Transport containers used for irradiation: aluminum at the left and polystyrene at the right.

3-5 REGATA experimental setup & irradiation:

The REGATA experimental setup for INAA at the reactor IBR-2M consists of four channels for irradiation (Ch1-Ch4), the pneumatic transport system (PTS) and gamma-spectrometers.

The main parameters of the irradiation channels are presented in Table 4. The channels Ch3, Ch4 are cooled by water, and the channels Ch1, Ch2, connected with the pneumatic transport system, are cooled by air. That is why the temperature in channels Ch3 and Ch4 is lower than the temperature in channels Ch1 and Ch2 in spite of the greater neutron flux density. The time of sample irradiation in channels Ch3, Ch4 depends on the operation cycle duration of the reactor and is equal to 10-12 days as a rule. The irradiation channels Ch1 and Ch2 are the same, but Ch1 is Cd-coated.
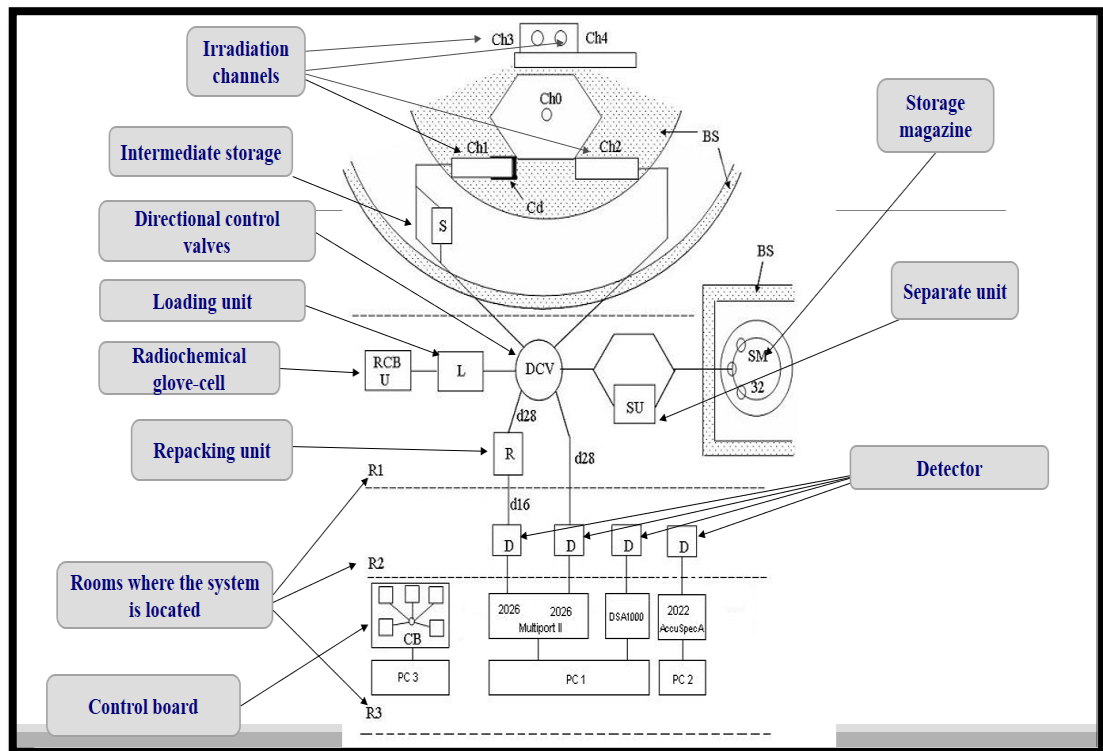

Fig 8. REGATA experimental setup

Up to 7 containers can be simultaneously loaded in each channel for long irradiation. The neutron flux density is controlled by monitors (Au, Zr, etc.). Containers are transported by compressed air. Acoustic detectors, placed on the «flight pipe» of irradiation channels behind the first ring of a biological shield, allow one to determine the time of container arrival and departure accurately.

The intermediate storage (S) is used to reduce the activity of aluminum containers after long irradiation. It is located between two rings of the biological shield of the reactor. There is also a magazine to store highly activated samples with 32 cells (SM) (Fig.8). It is surrounded with the biological shield made of lead and concrete blocks.

PTS has loading (L) and unloading (U) units (Fig.11) to load and to extract containers from the system. To provide radiation safety the unloading unit is placed into a glove-cell. All devices of the pneumatic system are equipped with photo sensors and end-switches for indication of the container position in the system and for correct operation of all mechanisms.

Table.4. Irradiation channel's characristics.

Irradiation site	Neutron flux density (n/cm^2 s)×10^{12}			T (°C)	Channel Diam. (mm)	Channel length (mm)
	Thermal	*Resonance*	*Fast*			
Ch1	Cd-coated	3.31	4.32	70	28	260
Ch2	1.23	2.96	4.1	60	28	260
Ch3	Gd-coated	7.5	7.7	30-40	30	400
Ch4	4.2	7.6	7.7	30-40	30	400

3-6 Measurement sequences:

The system of detection consists of three HPGe detectors with spectrometric electronics, three sample changers and the original control software. Each sample changer consists of two-axis linear positioning module M 202A by DriveSet (DriveSet.de) company and disk with 45 slots for containers with samples. As shown in figure 9.
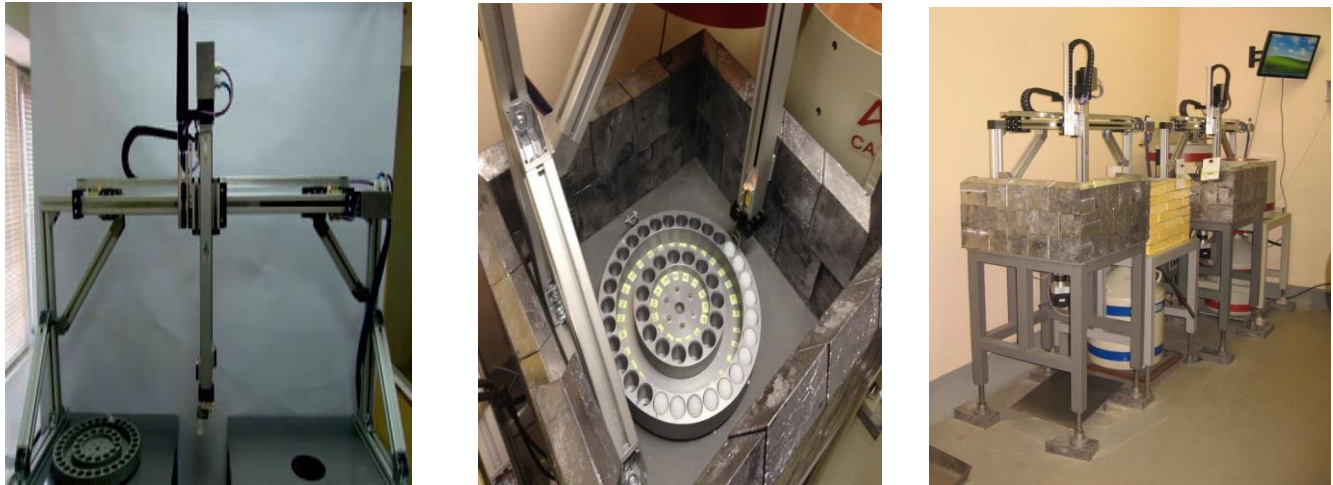

Fig 9. Sample changer

3-7 Analysis of the results:

3-7-1 Analysis spectra from NAA

The spectra obtained is analyzed by a program Genie-2000 which developed by Canberra with an additional interactive peak fiting module S506 is used. Calculation of elemental concentrations in the samples is performed by a relative method using the Concentration program.

In Genie-2000, one can separate manually two peaks are overlapped or close together.

Before start the analysis, the analyst should make sure the following:

1- Calibrate the system to change the channel numbers to Energy lines: (Energy calibration) by using standard source.

2- Perform an efficiency-energy calibration by standard source with a certificate (gamma emissions and corresponding activity).

Finally, the results must be processed by program CalcCON to obtain the concentrations of elements. See figures 10 & 11.
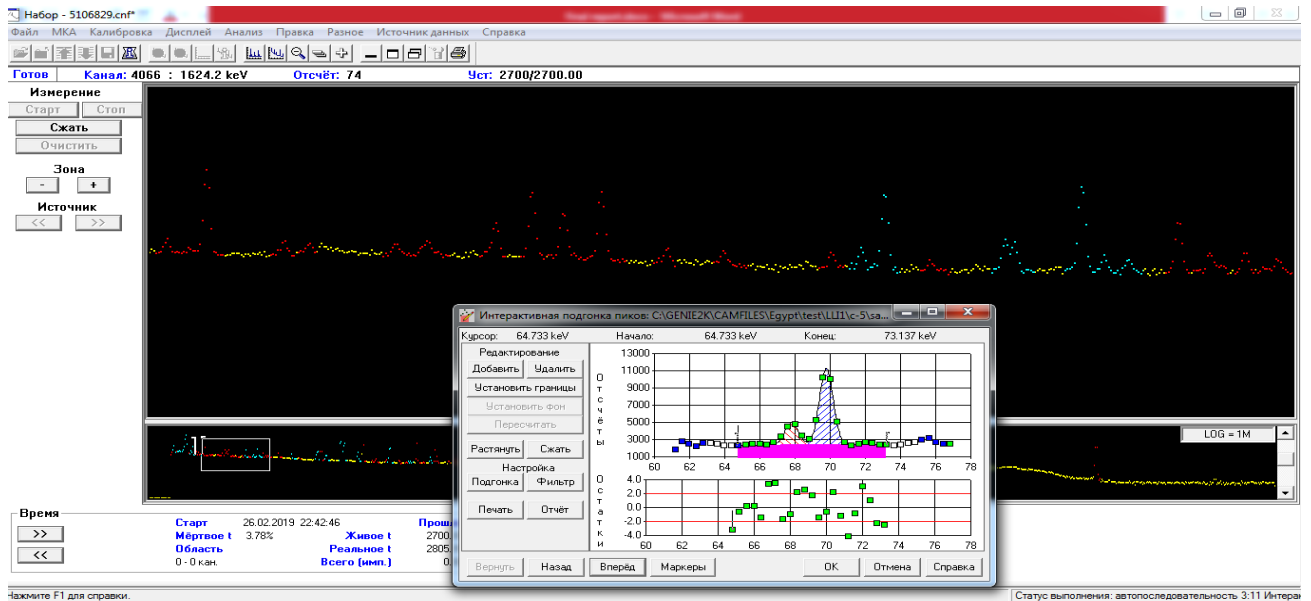

Fig 10 Program Genie-2000 during the analysis of long-lived sample from Egypt.

Fig 11 The results with the concentrations for long-lived samples from Egypt.

3-7-2 Atomic Absorption spectrometry

As a complimentary analysis, AAS used in JINR in order to detect and measure the concentrations of those elements which difficult to detect by NAA as Cu, Cd and Pb.

The samples first decomposed in (2 M) HNO_3 and H_2O_2 solution at temperature of 200 °c. Then injected to the analysis device. In a few minutes, the results are obtained.

4- OUTCOMES

The summer student program was been very fruitful, useful and helped me to make a decision for my future scientific plans. The results of the training can be summarized below:

1- In general recognition of the Institute laboratories and its potential.
2- Deeply know about the frank laboratory and neutron activation analysis and their contribution in daily life and environment.
3- Gained the theoretical and practical knowledge in NEUTRON ACTIVATION ANALYSIS.
4- Perform proper collection of samples.
5- Processing samples in the chemistry lab.
6- Performing analysis of Gamma-spectroscopies for sediment's samples collected from Egypt and successfully determination of concentrations for the radionuclides of interest using the program GENIE-2000 and concentration program.

REREFENCES

1- Harb, S., Khalifa, N., Badawy, W., Saad, N., 2016. ESTIMATION OF RADIOECOLOGICAL PARAMETERS OF SOIL SAMPLES FROM A PHOSPHATIC AREA. Nuclear Technology and Radiation Protection 31, 165-172,10.2298/NTRP1602165H.

2- Marina Frontasyeva , 2011,NEUTRON ACTIVATION ANALYSIS IN THE LIFE SCIENCES, DOI: 10.1134/S1063779611020043.

3- Robert. A. Meyers.ENCYCLOPEDIA OF ENVIRONMENTAL ANALYSIS AND REMEDIATION.

4- Peter Bode, INSTRUMENTAL AND ORGANIZATIONAL ASPECTS OF A NEUTRON ACTIVATION ANALYSIS LABORATORY.

5- Mahmoud. Bady, Hassanien M. Hassanien, Adel M. Kamal El Deen and A. M Hussein, STUDY OF SOLID WASTE AND ASHES CONTENT OF RADIOACTIVE AND HEAVY METALS IN ASSIUT THERMAL POWER PLANT, Journal of Engineering Sciences, Assiut University, Vol. 39, No 6, pp.1335-1342, November 2011.

6- W.M. Badawy, E.H. Ghanim , O.G. Duliu , H. El Samman , M.V. Frontasyeva, Major and trace element distribution in soil and sediments from the Egyptian central Nile Valley, 2017, Elsevier Ltd.

7- WAEL MAHMOUD BADAWY, HASSAN SOLIMAN EISSA and TAREK MAHMOUD MORSI, RADIONUCLIDES CONTENT AND THE RADIATION DOSE IN SOME IMPORTED TOBACCO SAMPLES – EGYPT, 2015, Journal of Nuclear Research and Development.

PROPOSAL FOR FUTURE PLANS

Title

"NEUTRON ACTIVATION ANALYSIS IN STUDYING THE EFFECT OF OIL FUELED POWER PLANT ON THE SURROUNDING ENVIRONMENT"

1- Introduction

The knowledge I gained will be used to conduct aware of analysis of soil's filters in rural and urban areas in Egypt to study the impact of power plant on the surrounding agricultural areas.

A good knowledge of the local radioactivity level including the content of main heavy and radioactive elements is an essential task in assessing any risk associated with the daily life of man. To produce electricity in Egypt, coal, oil and natural gas are used as fuel. Fuel combustion results in ashes, a portion of the ash carries with the flowing with the flue gas; this portion is called fly ash. The remainder of the ash settles to the bottom of the furnace and is called bottom ash.

The amount and characteristics of each type of ash produced depends on the type of fuel and boiler. Coal produces a relatively large amount of both ashes, while oil produces little bottom ash but more fly ash, and gas produces little of either ashes.

Fly ashes may contain heavy and radioactive metals and other noncombustible and potentially toxic chemicals that may have been in the oil. Fossil fuel combustion results in concentration of most these elements in ash by approximately 10 times the concentration in the original fuel. A 1997 analysis by the U.S. Geological Survey (USGS) found that fly ash typically contained 10 to 30 ppm of uranium, comparable to

the levels found in some granitic rocks, phosphate rocks, and black shale. Some of these elements can seep into the surrounding environment by different pathways.

This study aims to carry out a scan around (Asyut) power plant in Egypt and North- Cairo power station (which uses the oil as fuel) to understand the behavior of airborne of fly ash in aimed elements to be detected, Also assess in principle the possibility whether the quantities are enough to extract these elements from the ashes or not.

2- Studied area

To evaluate the influence of fly ash on human life, which produced during electricity production, two sources (two electrical power stations) are selected to implement the study. The first is North Cairo electrical power station, and the second is Assiut electrical power station. For studying the cycle of heavy elements, which involved in fly ash, two agricultural areas. These areas are located in the southwest direction from the sources, and selected in the direction of the wind spread most of the year in Egypt.

For each area selected 6 points, from which samples collected. From those areas, collected two kinds of samples. The first kind was from soil with 37 samples at different depths. The depths of sampling are 0 (i.e. at surface of earth), 10 and 20 cm from the surface. The difference in depth of sampling to build 3D map for concentrations of heavy elements, which are deposited from fly ash. This map will help us to study the diffusion mechanism of heavy elements through the soil physically or chemically. The second kind of samples collected from plants to study the cycle of heavy elements, which can affect on human life directly. It was 14 samples with two kind's short term plants and long term plants (trees).

The samples collected from the corners of agriculture basin, which have the minimum limit of mixing. The samples were collected manually. A sample was

collected from ash, which stilled in the burning furnace. This sample was separated by magnetic separation into three samples to study the ash composition. Table 1. Shows the kind of sample, and where the samples were collected. And Figure 1. shows the sampling locations around Assiut power Station, and figure 2. Shows North- Cairo power station.

3- Aim of the work

1- The present study aims to introduce comparison between different concentrations of A total of 43 elements viz., Li, Na, Mg, K, Ca, Sc, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Br, Rb, Y, Zr, Nb, Mo, Cs, Ba, La, Ce, Pr, Nd, Sm, Eu, Ga, Gd,Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf, Tl, Pb, Th, and U contained in ashes and in sediments around (Asyut) power plants and North- Cairo power plant, collecting from a grid 1*3 km^2 and depth 1 m and analyzed by using NAA.

2- To construct the spatial distribution maps of the selected pollutants around power plants.

3- To evaluate the movement mechanism of heavy materials from soil.

4- To achieve recommendations decreasing the effect of heavy elements on human life.

5- Search in principle the possibility of extracting some of these rare metals from the ashes.

6- Comparison of new results obtained NAA with previous results obtained chemical analysis by another authors [3].

7- Using Moss technique to monitor the heavy metals around the power stations for long time monitoring, up to 3 kilometers from the stations.

Table 1. The kind of sample, and where the samples were collected.

Profile	Sample number	Point number	Type	Coordinates		Notice

1	1	1	Soil			Surface
	2		Plants			Surface
	3		Soil			At depth 20 cm
	4		Soil			At depth 10 cm
	5		Plants			Plants
2	6	2	Soil			Surface
	7		Soil			Surface
	8		Soil			At depth 10 cm
	9		Soil			At depth 20 cm
	10		Plants			Plants
	11		Plants (baklashan)			Plants (baklashan)
	12		Plants (baklashan)			Plants (baklashan)
	13		Tree			Tree
	14		Tree fruits			Tree fruits
3	15	3	Soil			Surface
	16		Soil			At depth 10 cm
	17		Soil			At depth 20 cm
	18		Plants			Plants
4	19	4	Soil			Surface
	20		Soil			At depth 10 cm
	21		Soil			At depth 20 cm
	22		Plants			Plants
5	23	5	Soil			Surface
	24		Soil			At depth 10 cm
	25		Soil			At depth 20 cm
	26		Plants			Plants
	27		Tree			Tree
6	28	6	Soil			Surface
	29		Soil			At depth 10 cm
	30		Soil			At depth 20 cm
	31		Plants			Plants
____	32	-	Soil			High way
	33	-	Soil			High way
	34	-	Soil			Desert sample
7	35	7	Soil			Surface
	36		Soil			At depth 10 cm
	37		Soil			At depth 20 cm
	38		Plants			Plants
8	39	8	Soil			Surface
	40		Soil			At depth 10 cm
	41		Soil			At depth 20 cm
	42		Plants			Plants
9	43	9	Soil			Surface
	44		Soil			At depth 10 cm
	45		Soil			At depth 20 cm
	46		Plants			Plants
10	47	10	Soil			Surface
	48		Soil			At depth 10 cm
	49		Soil			At depth 20 cm
	50		Plants			Plants
11	51	11	Soil			Surface
	52		Soil			At depth 10 cm
	53		Soil			At depth 20 cm
	54		Plants			Plants

12	55	12	Soil			Surface
	56		Soil			At depth 10 cm
	57		Soil			At depth 20 cm
	58		Plants			Plants
13	59	Magnetic	Soil			0.05 amp
	60	Magnetic	Soil			0.3 amp
	61	Non mag	Soil			0.3 amp
___	62	Water residue				

Fig 1 The sampling locations around Assiut power Station.

Fig 2 The sampling locations around Shows North- Cairo power Station.

References:

1- Clean Air Task Force, the Land and Water Fund of the Rockies ''the Last Straw, Water Use by Power Plants in the Arid,'' the Energy Foundation, the Hewlett Foundation, USA, April 2003, pp.5, Internet Available: http://www.lawfund.org/ and http://www.catf.us/ .

2- U.S. Geological Survey (October, 1997), " Radioactive Elements in Coal and Fly Ash: Abundance, Forms, and Environmental Significance," (PDF), U.S. Geological Survey Fact Sheet FS-163-97, Internet Available: http://pubs.usgs.gov/fs/1997/fs163-97/FS-163-97.pdf .

3- Mahmoud. Bady, Hassanien M. Hassanien, Adel M. Kamal El Deen and A. M Hussein, STUDY OF SOLID WASTE AND ASHES CONTENT OF RADIOACTIVE AND HEAVY METALS IN ASSIUT THERMAL POWER PLANT, Journal of Engineering Sciences, Assiut University, Vol. 39, No 6, pp.1335-1342, November 2011.

4- "Geochemistry of the Egyptian Mediterranean Coastal Sediments and Their Contamination with Heavy Metals Using Instrumental Neutron Activation Analysis", FINAL REPORT ON THE SUMMER STUDENT PROGRAM, JOINT INSTITUTE FOR NUCLEAR RESEARCH Frank Laboratory of Neutron Physics Sector of Neutron Activation Analysis and Applied Research.