

APPLICATIONS OF AMS IN BUCHAREST FOR DETECTING NUCLEAR POLLUTION

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The Accelerator Mass Spectrometry (AMS) is an analyzing method of highest sensitivity. Applications will be presented for providing a rapid response to the aims of the Romanian – Bulgarian cross-border cooperation program concerning chemical and nuclear disasters. AMS machineries and their broad range of applications are described such as the detection of produced and existing nuclear pollution, mapping of current and past nuclear contamination, forensic science and nuclear activity surveillance.

Key words: AMS, ^{129}I , ^{239}Pu , ^{240}Pu

1. INTRODUCTION

Today, more than ever, the exploding progress of technology and research has generated a rising risk of negative impact on the environment and human health. The chemical and the nuclear pollution, are some times produced by hazard or disasters but could be also be produced by criminal actions. These are responsible for sever perturbations on our planet with implications everywhere, from the level of small entities like genomes up to the level of worldwide large scale climatic changes and perturbations.

Large efforts are made by International and National Commissions and ecological organizations to ensure rapid and highly qualified response actions for preventing, evaluating, monitoring and reducing or eliminating the consequences of chemical and nuclear disasters. In this sense is focused the Romanian – Bulgarian cross-border cooperation program.

Our paper addresses these issues specifically. We present our experimental systems, scientific procedures and skills, able to respond to part of these aims by using the most sensitive method of analyses, the *Accelerator Mass Spectrometry* (AMS). This method is able to count atoms one by one. Detecting and analyzing radioisotopes like ^{129}I and ^{239}Pu , the AMS is able to detect and monitor rapid nuclear criminality.

In the next section, the AMS method is briefly presented. Then, next sections will present the applications with the ^{129}I radioisotope, for detecting past and present nuclear accidents, and the forensic method based on $^{240}\text{Pu}/^{239}\text{Pu}$ detection, able to distinguish between producers of nuclear bombs or of nuclear electric power.

2. THE ACCELERATOR MASS SPECTROMETRY ANALYZING METHOD AND FACILITIES AT IFIN-HH

Long lived radionuclides are detected by AMS, free of molecular interference, and with the detector background at isotopic ratio as low as 10^{-16} .

In our institute, AMS research started many years ago (1996). At the beginning, a facility was constructed based strictly on in house made components [1, 2]. About ten years later it was upgraded with a new ion source of MC - SNICS type, purchased from NEC and a complete automation of the entire facility [3]. Finally, in 2013 a compact 1MV AMS - HVEE dedicated machine, fully automated was purchased and commissioned [4].

Fig. 1 presents the general layout of our first AMS facility constructed based on the 9 MV multipurpose tandem accelerator at IFIN-HH that is still in operation, specially for light isotopes like ^3H , ^2H , ^{14}C and ^{26}Al .

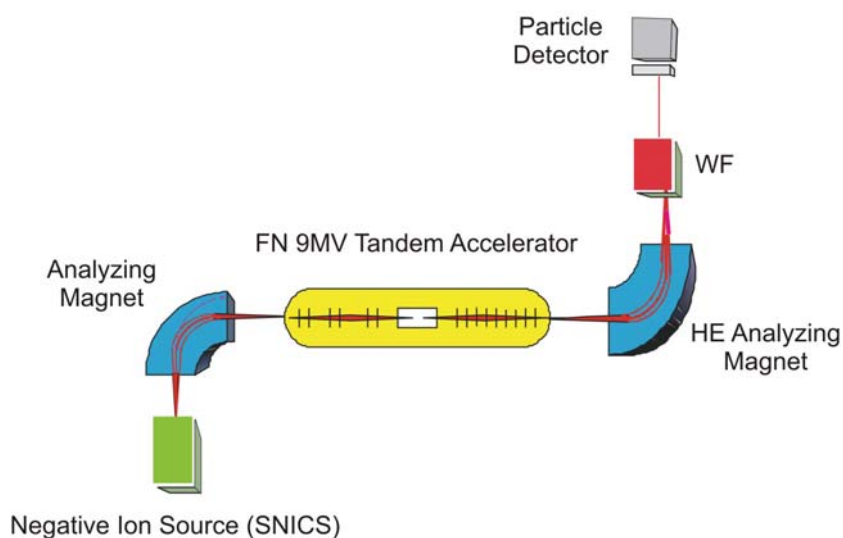


Fig. 1 – Schematic view of the AMS machine at the 9 MV tandem accelerator at IFIN-HH, Bucharest.

The sample material is sputtered in the ion source and is transformed into an ion beam that has to pass a long way through the accelerator and the different

filtering and analyzing devices. The transports of an extremely low intensity ion beam (few atoms/second), on its way from the sputter ion source all the way to the detector has to be optimized. AMS solves this problem by using the so called “pilot beam”. It is formed by ions with measurable current beam intensities (approx. 100 nA) and with properties (energy, mass, charge) chosen in such a way that ions experience the same electromagnetic forces during the ion optic transport as the rare and “invisible” ion beam.

An important breakthrough in our experimental capabilities was accomplished by purchasing and installing a compact 1MV AMS - HVEE dedicated machine, fully automated.

Our machine is able to perform AMS analyses with the following isotopes: ^{14}C , ^{10}Be , ^{26}Al , ^{41}Ca , ^{129}I , ^{239}Pu , and ^{240}Pu . Fig. 2 presents the general view of the new compact AMS machine.

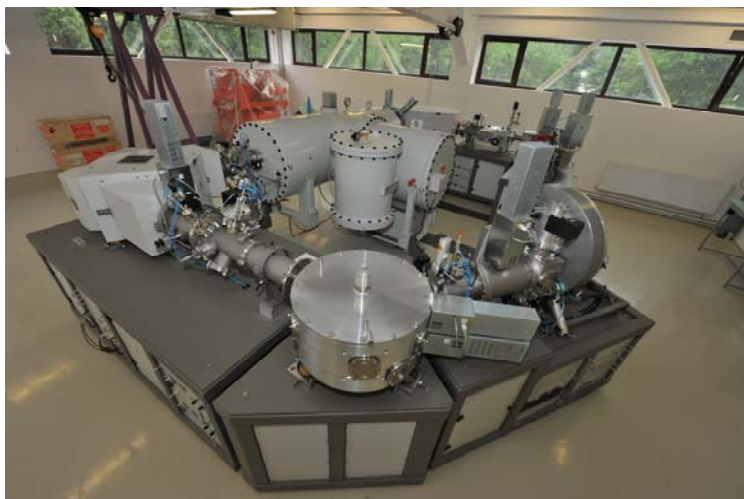


Fig. 2 – The 1 MV HVEE AMS COCKCROFT WALTON type tandemtron at IFIN-HH.

Samples required for AMS experiments are less than 2 mg in weight, but must be in a solid form, are placed in a carousel type target changer that makes the experiment easy and rapid to handle.

The new 1MV AMS machinery allows a continuing monitoring of an experiment and the complete automation makes it possible to realize a perfect tuning of the radioactive beam. Therefore, the experimental precision measured with our new AMS machine was optimized to the following values of relative standard deviation and background: (0,25%, 10^{-15}) for ^{14}C , (1,2%, 10^{-14}) for ^{10}Be , (0,8%, 10^{-14}) for ^{26}Al and (1,7%, 10^{-14}) for ^{129}I . The average background level for most of the radio isotopes was measured to be 2×10^{-15} .

3. AMS APPLICATIONS FOR PROTECTION AGAINST NUCLEAR POLLUTION

3.1. DETECTION AND MONITORING OF NUCLEAR EVENTS FROM THE PAST OR PRESENT

After many years of open nuclear explosion and experiments, starting with the 60's, the ^{129}I ($T_{1/2}=15.7$ Ma) fission product became the highest source of nuclear pollution for the environment. Emitted by the nuclear installations, either in the liquid or in the gas form, ^{129}I has travelled all around the world. Between 1966 and 1994 the biggest Western Europe nuclear reprocessing plants from La Hague (France) and Sellafield (Great Britain) contributed to the nuclear pollution with an input of 906 kg of ^{129}I , discarded in the English Channel and an input of 534 kg of ^{129}I , discarded in the Ireland Sea.

It is to be emphasized that due to its high volatility, when entering into the atmosphere (nuclear explosions or releases), the ^{129}I is transported over very large distances. The releases of nuclear installations will make iodine dispersing rapidly over rivers and soil and increase the normal values in the environment.

As already presented before, with our 1MV AMS facility we are able to perform ^{129}I determinations with a precision better than 2% and with a sensitivity of 10^{-14} for the $^{129}\text{I}/^{127}\text{I}$ ratio.

In the light of the ultrasensitive measurements of the ^{129}I , the monitoring possibilities of the AMS were evaluated by the International of Atomic Energy Agency, Vienna (IAEA). As a result, IAEA has identified and recommended the measurement of the ^{129}I as a potential signature mark of the reactor or reprocessing operations, or for the detection of undeclared nuclear activities [5].

Therefore, AMS can be applied in the framework of the Romanian – Bulgarian cross-border cooperation program for measuring and monitoring nuclear pollution in the environment [6, 7, 8, 9], for controlling the level of radionuclides in the ecosystems and in the vicinity of the nuclear plants (water, air, earth), for the supervision of the nuclear power plants activities, for controlling the integrity of nuclear installations etc.

In case of emergency situations, rapid analyzes can be performed by collecting water from big rivers like the Danube River, from the Black Sea, from precipitations like rain or snow, the dust from air (filter absorption) or from the grass, or tree leaves, or soil samples.

Such monitoring was performed in the past by our laboratory for the Danube River. Water samples were monthly collected from up-stream and down-stream locations by the two nuclear power plants on the Danube river: the VVER type reactor at Kosloduy, Bulgaria and the CANDU reactor at Cernavoda, Romania and from the Black Sea. Our results for the samples collected in 2005, are presented in Table 1. The measured $^{129}\text{I}/^{127}\text{I}$ values do not show differences between up and down-stream locations and on the average they have the concentration value in water of 6×10^7 at/L. This value is not exceeding the normal concentration values of

about 10^8 at/L, currently measured for rivers in Europe and it can be considered as a current normal value.

Table 1

$^{129}\text{I}/^{127}\text{I}$ and ^{129}I atoms/L values measured by AMS up- and down-stream from the two nuclear power plants Kosloduy and Cernavoda, and from the Black Sea

	Location	$^{129}\text{I}/^{127}\text{I}$ (10^{-9})	Average value (atoms/L x 10^7)
1	Giurgiu (up-stream)	4.3 ± 0.3	6 ± 0.4
2	Oltenita (up-stream)	4.2 ± 0.7	
3	Cernavoda (up-stream)	4.4 ± 0.7	
4	Cernavoda (down-stream)	3.5 ± 0.4	
5	Galati (down-stream)	3.1 ± 1.5	
6	Black Sea	5.2 ± 1.8	14 ± 6

To further demonstrate the high ability of AMS with ^{129}I to detect any nuclear activities on earth, we give two examples of how it can reconstruct the history of pollution many years back and of how it can discover accidents that happened many years but remained undeclared. For all the mentioned goals, AMS research uses the accumulation of radioisotopes in the natural archives like glaciers, lakes and centennial tree rings. The outspreading or nuclear pollution is then calculated according to the Global Transport Model (GTM) [10]. This is a computer code that permits a detailed description of the troposphere propagation and of the biosphere exchanges.

The first example concerns the nuclear pollution in Central Europe produced by the Nuclear Reprocessing Plants. Besides Sellafield (GB) and La Hague, mentioned before, another reprocessing plant at Marcoule (F) produced large pollution of the earth's atmosphere. Therefore, a high pollution of the earth's atmosphere was also measured.

Fig. 3 shows the declared reactor calculated emission radioactivity expressed in GBq/a, during 40 years of operation.

The striking effect is that the overall produced pollution from these three sources was and still is visible, even far away from them, in the natural archive of the Fisher Horn Glacier in Switzerland. AMS measurements on core drillings from the Fisher Horn Glacier revealed the time evolution of ^{129}I nuclear pollution at high altitudes in Central Europe. Figure 4 shows the AMS results. A steady increase of the concentration was registered until 1990 when the activity of these nuclear facilities was either diminished or stopped. At a first glance, the experimental results are in good agreement with the declared values for nuclear releases by Marcoule. Furthermore, a nuclear pollution input in the Fisher Horn comes also from the nuclear weapon experiments. The fission products ^{129}I and ^{137}Cs are produced from ^{238}U bombarded by fast neutrons, with yields of 5.57% and 1.58%, respectively. Therefore, after about 30 years from such events, the deposition ratio $^{137}\text{Cs}/^{129}\text{I}$ is estimated to be 1.6 in the environment.

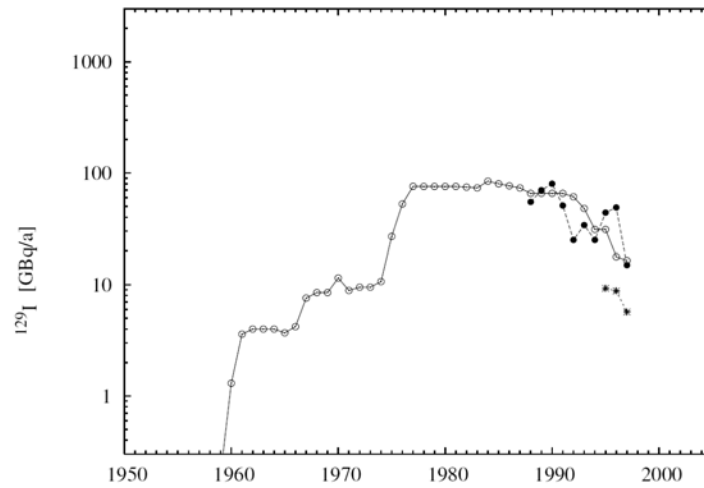


Fig. 3 – Declared ^{129}I radioactivity expelled from Marcoule reprocessing power plant into the atmosphere [11].

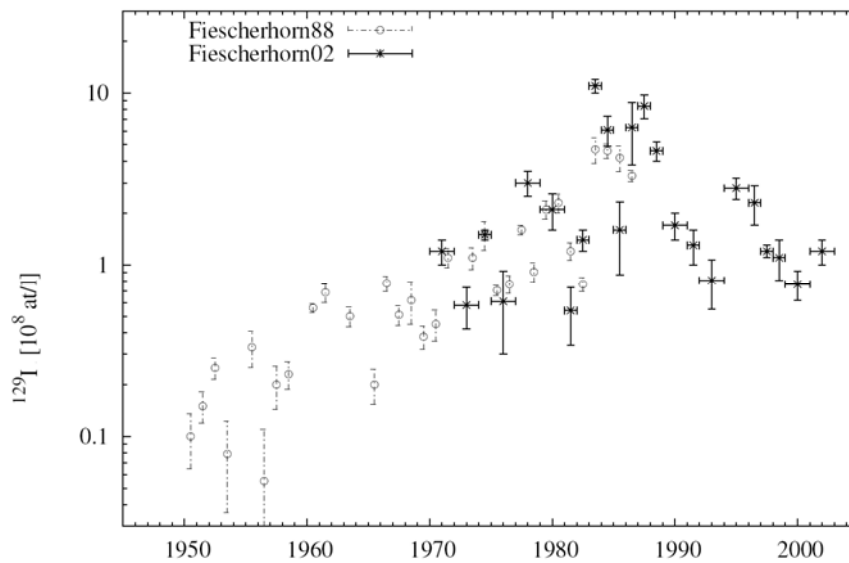


Fig. 4 – Timely ^{129}I deposition (fall out) on the Fisher Horn Glacier [11].

The second example refers to the region of Eastern Europe and Asia [12]. Similar to Europe, nuclear installations from the former USSR have produced important nuclear pollution to the environment. The most important Nuclear Power Stations, from Tschelniabisk (Majak), Tomsk and Krasnoyarsk, can be seen in the map below (see Fig. 5). All these large power plants are situated in a reasonable distance from important natural archives like the glacier from Belucha in the Altai Mountains.



Fig. 5 – The map of the former USSR with the main nuclear installations and with natural archives for the AMS research, like the Belucha glacier and the Baikall Lake.

In 2001, a core drilling of 140 m length in the glacier was performed by an international expedition [13]. As shown in Fig. 6, the AMS measurements showed a fast increase of ^{129}I fall-out starting in 1950, followed by a constant value between 1958 and 1976.

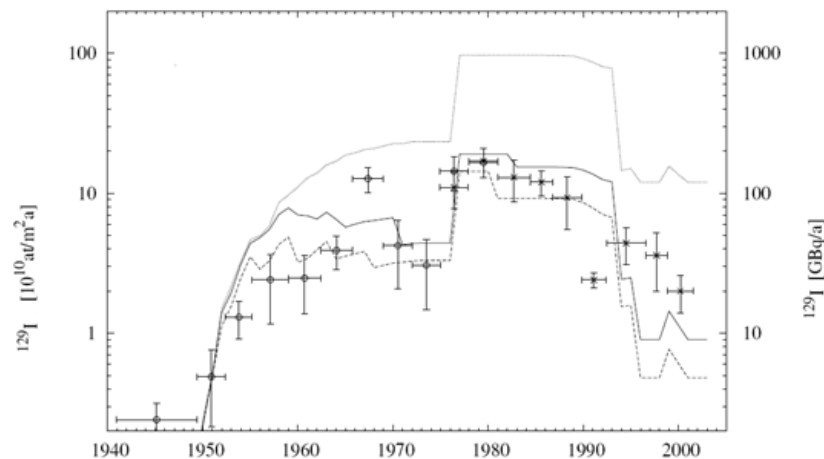


Fig. 6 – ^{129}I fall out measured by AMS at the Belucha glacier. The equivalent radioactivity deposition is shown on the right hand side [12].

In 1977 a sudden increase of the fall-out is to be noticed. It is correlated with an unreported nuclear accident that happened that year at the Majak reactor. Based on the AMS and on the GTM, the measured data from the Belucha glacier also

determined the separate contribution to the environmental pollution of the mentioned region: 44% by Majak, 33% by Tomsk, 15% by Krasnojarsk and 8% contribution from Europe.

3.2. TRITIUM IN THE ENVIRONMENT

In the mid-50's and in the early 60's, tritium was widely dispersed during the atmospheric testing of nuclear weapons. The quantity of tritium, existing in the atmosphere from the weapons testing, peaked in 1963 and it has been decreasing ever since. Today, sources of tritium include electric power nuclear reactors, research reactors, reprocessing plants and weapons production plants. Tritium may be released from these facilities as steam or it may leak into the underlying soil and ground water. Such releases are usually small and are required to meet international environmental standards. There is a special interest for tritium research for health and environment protection. Since tritium is almost always found in water, it goes directly into soft tissues and organs. The associated dose to these tissues is generally uniform and dependent on the tissues water content. Recent studies have shown the correlation between this kind of infestation and the appearance of carcinomas. Our laboratory is prepared and has the experimental means to detect by AMS tritium in all kinds of samples collected from the environment or materials that were penetrated by tritium [14, 15, 16]. Figure 7 presents tritium measured from the accumulation in carbon foils. The AMS analyze is able to provide a rapid response to the need of detection of a tritium contamination.

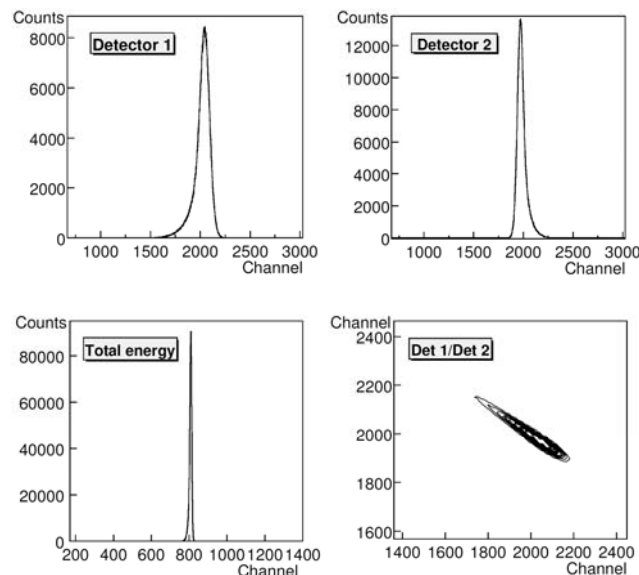


Fig. 7 – Tritium peaks from energy releases of energetic particles (14 MeV) in a diode array of two thin surface barrier detectors Si (Au).

3.3. FORENSIC RESEARCH

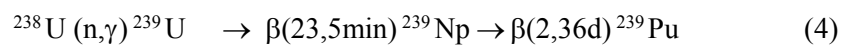
The non-proliferation of nuclear weapons or the halt of nuclear weapon programs are subjects of the most difficult problems of our contemporary international politics to stop this nightmare. Whatever the declarations will be, or whatever satellites or institutional inspections will discover, AMS can distinguish firmly and from far away distances the differences between good and evil.



Fig. 8 – Open nuclear explosion.

AMS is the only analyzing method able to measure accurate and with high sensitivity the isotopic ratio $^{240}\text{Pu}/^{239}\text{Pu}$ somewhere in the environment, close or far away from the nuclear plant (on leaves, grass, soil, water or air). The ratio value is striking different for the pollution produced by the Electric Nuclear Power Plants and that produced by the Weapon Nuclear Reactors [12].

Nuclear weapons use as main fuel ^{239}Pu that is produced from ^{238}U , after (n,γ) , followed by a two β -decay:



Depending on the burning time in the reactor ^{240}Pu will also be produced. It is formed when ^{239}Pu captures a neutron. About 62% to 73% of the time when ^{239}Pu captures a neutron it undergoes fission, the rest of the time it forms ^{240}Pu . The longer a nuclear fuel element remains in a nuclear reactor, the greater the relative percentage of ^{240}Pu in the fuel becomes. For weapons use, the fuel needs to be as low in ^{240}Pu as possible, usually less than 6% of the total plutonium. This is because ^{240}Pu sometimes undergoes spontaneous fission, causing the weapon to detonate prematurely.

Table 2

Plutonium grades

Plutonium-grade	Plutonium-240
Super-grade	< 3%
Weapon-grade	< 7%
Fuel-grade	between 7% and 18% incl.
Reactor-grade	> 18%

The weapon-grade can be achieved by reprocessing the fuel after just 90 days of use and about 36 days of letting cooling down the fuel. At this moment of time the reactor core must be opened and downloaded.

However, the AMS analyzing measurement is rather straight forward. It does not need any standard samples and it gives firmly the ratio $^{240}\text{Pu}/^{239}\text{Pu}$, larger or smaller than 7%.

Sensitive AMS plutonium measurements are important for the environmental protection too. Plutonium concentration in sediments and in water can be used to identify the sources of plutonium and to monitor its dispersal away from nuclear installation. As a result of the atmospheric weapon testing and of the reprocessing operations of the spent nuclear fuel, these isotopes are widely dispersed in the environment. Because the long half-lives ($T_{1/2}^{240}\text{Pu} = 6550$ a, $T_{1/2}^{239}\text{Pu} = 24000$ a, $T_{1/2}^{236}\text{U} = 2.3 \times 10^7$ a), these isotopes will persist in the environment for periods of time much longer than our lifetime and will be a very negative heritage for our successors.

4. SUMMARY

AMS is a powerful means of measuring the isotopic abundance of long-lived radionuclides and applications were presented as examples for a highly efficient and rapid response to the aims of the Romanian – Bulgarian cross-border cooperation program.

The experiments were focused on environmental detection and protection against nuclear pollution. In this respect, AMS measurements were performed on samples collected in the vicinity of nuclear power plants and of nuclear reprocessing plants. Some of researches were performed in large international collaboration with laboratories from ETH–Zurich, Switzerland, Technical University Munich – Germany and The Technical University of Sankt Petersburg – Russia.

^{129}I , used in the most presented results, is very useful for the reconstruction of the nuclear releases from the past and for the monitoring of the nuclear activities in the present. The IAEA has identified and recommended the AMS measurement of ^{129}I as a potential signature of the reactor or reprocessing operations, or for the detection of any pollution produced by nuclear activities. The ratio $^{129}\text{I}/^{131}\text{I}$ is about 50. ^{129}I being high volatility is transported over large distances, providing a sensitive fingerprint for the detection of the smallest nuclear releases.

The new compact 1MV AMS-HVEE dedicated machine installed and commissioned in 2013 in our institute, opened new and major improved experimental conditions. Precision and rapidity of performing analyses were considerably improved. Heavy isotopes, like plutonium, are now possible to be analyzed and could contribute with very important information concerning the discovery of undeclared nuclear activities, responding to the goal of the Romanian – Bulgarian cross-border cooperation program. In the framework of this program AMS could be applied for measuring, monitoring and investigation of the concentration of ^{129}I , ^{239}Pu , ^{240}Pu and ^3H in the environment, for controlling the level of these nuclides in the ecosystems and in the vicinity of the nuclear plants (water, air, earth), for the supervision of the nuclear power plants activities, for controlling the integrity of nuclear installations, of water quality and of life.

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