Search for Non-registered Radioactive Sources - an Important Part of the National Inspection Programme

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Abstract: The non-registered sources pose a threat because as a rule no safety measures are in place when handling them. Sources outside the regulatory control can be easily lost or abandoned and as such they can be used in malevolent acts causing exposure to humans or contamination of the environment. Numerous national or international defence systems were put in place in order to identify them in a due time The HASS directive of the EU demands from the countries to conduct campaigns to identify orphans sources and to have in place emergency preparedness system. According to experiences of the Slovenian Nuclear Safety Administration which conducted a comprehensive programme related to non-registered sources or waste in the period 2004-2010, majority of sources or waste found have never been under the control in the past. Around one thousand of various non-registered sealed, unsealed sources and radioactive waste were identified at research, educational, military institutions as well as in collections. None of the sealed source found can be defined as a HASS source according to the directive. The inspection programme resulted not only in identification of sources or radioactive waste. The safety assessment and safety measures followed. A registration or a full licensing procedure related to sources was put in place. In cases when owner identified sources as radioactive waste such waste was stored in the central storage for radioactive waste. Non-registered sources, especially sources used in the past, pose a threat not only to a specific country but also internationally due to the international trade. National campaign conducted in all countries on their own territories can contribute to decrease a level of such threat.

Key Words: radioactive sources, contamination, orphan sources, inspection, threat

1. Introduction

The complete and detailed inventory of sources and radioactive waste in a state is one of the main milestones of radiation and nuclear safety. It enables to follow the lifetime phases of a specific source or radioactive waste from its cradle to grave. The quality of such a register is a parameter determining everyday regulatory activities. Moreover, it also enables long term planning of specific nuclear and radiation safety activities, e.g. planning storage for radioactive waste or planning a disposal site. The complete national inventory of sources became even more important due to the international industry of scrap materials.

An insufficient control over radiation sources can result in severe accidents causing exposure of humans as well as substantial contamination of the environment. In the last decade numerous cases were reported when orphan sources were melted or abandoned causing exposure of people or contamination of the environment. Handling such radiological emergency can be a comprehensive task which requires an involvement of many institutions including among others also international organisations. This task can also require a substantial financial support. One of such event, namely melting of Am-241 source, was lastly reported in March 2012 [1]. The estimated activity was in the range of 0.6 - 60 GBq, i.e. it was a source of the Category 4 of the IAEA categorisation scheme. Moreover, abandoned or lost sources also pose a specific threat regarding malevolent use of ionising radiation, e.g. a dispersion of a typical Cs-137 source of 10¹² Bq could cause a contamination of an area of about 10⁴ football playgrounds taking into account the allowed limit of contamination (40 $Bq/100 \text{ cm}^2$). This threat became more evident in the last decade because neither sources used in the past nor a whole radiation nor nuclear safety systems were not designed to cope with such threat. Furthermore, the waste management of sources from the past which are not used nowadays can pose a substantial financial burden to present owners. While in the past countries very often established an efficient state control using different mechanisms, e.g. state control of equipment with sources in military activities, today after huge changes of the state control the vulnerability of safety became evident. As a result abandoned or lost sources or radioactive waste can be easily acquired and used in malevolent actions.

The international community as well as countries put a lot of effort to identify non-registered sources, e.g. the HASS directive of the EU [2] from 2003 specifically requires to conduct campaigns to identify orphans sources. Furthermore, it requires an emergency preparedness system to be in place which can be a substantial organisational and financial burden of a state. The directive does not require portal monitors or other sophisticated equipment at each border crossings or at companies premises where lost sources or radioactive waste can be found, but it requires a state system to identify such sources and to act in a due time in order to prevent exposure of humans and contamination of the environment. Many countries all over the world decided to have an elaborate system to identify such sources at the borders as well as inside the state, as for example the USA as given in [3] or Spain [4].

Already in the middle of nineties of the last century and especially after the IAEA Dijon conference held in 1998 [5] the IAEA started with a programme to analyse the urgent need for international cooperation. The IAEA database of typical radioactive sources was prepared [6] as well as database of all reported events with illicit trafficking [7]. All incidents with orphan sources are reported in the IAEA NEWS database [8]. Numerous documents related to border control were prepared by the IAEA, e.g. [9, 10], and a new code of conduct is under way [11].

Slovenia started to develop a control of orphan and abandoned sources as well as the system of controlling scrap metals more than a decade ago when it was faced by a melting of some medium activity radioactive Co-60 sources, i.e. of around 70 MBq each, of unknown origin. Due to its specific geographical location, Slovenia was also faced with specific threats related to orphan sources from abroad. Namely, Slovenian is located close to the region where big political and military changes took place. In addition, it was close to the war region where many facilities with radioactive sources were demolished. Till now the defence system has been established at the borders as well as inside the country. It involves regulatory authorities, customs, owners of a scrap yard facilities and steel factories, Agency for Radioactive Waste Management, qualified experts etc. The regulatory authority, i.e. Slovenian Nuclear Safety Administration (SNSA), started to strictly follow the lifetime of all registered sources, firstly the sources which were known from the databases being available to the regulatory authorities. As a rule basic physical characteristics of a source as well as of equipment related to the specific source were available in the databases mentioned. If the status of the source was not known, an inspection procedure followed based on investigation of all possible lifetime phases of a typical source, e.g. export, dismantling. During such investigation processes weak parts of the controlling system in the past were identified, among them also a fact that for some sources found during investigation procedure no information was available in the state registers. Such sources or waste were not considered by the owner as a source or radioactive waste, as appropriate, and were very often even not documented in the databases of the owner. The terms "non-registered sources" or "non-registered waste" in this paper mean such sources or waste, i.e. objects which have never entered into a regulatory authority system in the past but they are still used or stored at industrial or research facilities. The issue of non-registered sources or waste emerged in 2004. As a result and in parallel with other activities mentioned the SNSA conducted from 2004 till 2010 a comprehensive and systematic campaign in order to find a great majority of so-called non-registered sources or radioactive waste on whole territory.

2. Campaign to Identify Non-registered Sources in a State

Non-registered sources or non-registered waste identified in the campaign have all characteristics of a source or radioactive waste by definition given in the national legislation. Many reasons of skipping such sources or waste from a regulatory control in the past exist. For example, the former legislation did not require such a control for military sources which were controlled differently as nowadays or a definition of a source producing ionising radiation was different in a past. In several cases at the time of acquiring a source or radioactive waste its owner was not aware of basic characteristics of the object and did not have enough knowledge in order to properly handle it. The owner was not aware of radiological risks. Also other stakeholders were involve in many inspections. The stakeholders in many cases had very little knowledge about radiation safety.

In addition, at specific facilities, e.g. related to a use of thorium or uranium, handling of radioactive materials causes the occurrence of additional sources or radioactive waste which were not registered, sometimes even not in the database of the owner. As a rule, handling non-registered sources or waste was mostly done without using all safety measures which are necessary and are given for example in the IAEA BSS [12], EURATOM BSS [13] and related directives.

The campaign of the SNSA was based on the development of special methodology using ten steps altogether. It involves among others also a use of field measurements or a study of scientific literature and documentation from which it was evident that radioactive sources were used for a specific research or project. Data on radiation sources were obtained also from the website information, advertisements and personal communications. The methodology for searching of non-registered sources in a state is given in details in [14]. Till 2011 nearly one hundred inspections were conducted based on this methodology and around thousand sources were found, i.e. sources whose activities are above exemption levels given in [13]. Table 1 shows the distribution of inspections performed in the period 2004-2010. Majority of inspections, i.e. 47% were related to research institutions where handling of non-registered sources was a practice for decades. The number of sources identified in the research institutions was a few hundreds and they span from calibration sources just above exemption levels to the sources with moderate activity for which a full scope licence is required. One of the main reasons for such a huge number of non-registered sources lies in the fact that research projects related to radiation sources were very often conducted in a campaign followed by a substantial reorganisation of laboratories. Such reorganisations which were not fully included in a quality management system did not allow sufficient tracking lifetime phases of each individual source or radioactive waste as required by the legislation. In addition, some research institutions were for decades also involved in uranium mining and processing, especially after the discovery of uranium deposits at Žirovski Vrh in 1961, and nuclear fuel cycle in general.

Table 1. The number of inspections performed in the campaign to identify non-registered sources in
the state in the period 2004-2010

Type of Institution	No. of Inspections
Research institutions	43
Educational institutions	21
Defence and civil protection institutions	19
Museum collections	8

The second important group of inspections was performed at universities and other educational institutions. Altogether 23 % of inspections were conducted in such institutions where sources were not related only to educational process. It should be also taken into account that educational institutions are very often related to research and inspecting such educational institutions was a natural step of the inspection project.

During the inspection programme a whole spectrum of well known radioisotopes was identified from H-3 to various uranium compounds. As a result the inspection was confronted by very different radio-toxicities ranging from the group I to the group V [15]. The physical state of sources or waste included all possible states i.e. gaseous, solid or liquid. So a risk for a possible contamination was identified during some of inspections and safety measures were immediately put in place. Furthermore, in some institutions inspected an accumulation of sources was identified, as for example in the storage of the military facility.

Some of the man-made non-registered sources identified are not available on the market today. It is even very difficult to get any information about their producers or about the producers of the related equipment, as for example a shielding of a source or transport packages for the source transport. In this paper such sources are called unconventional sources. They were produced in a research laboratory for a unique occasion, e.g. during a specific projects or in a so called "strategic facility" for a specific purpose related to military use decades ago.

Figure 1 shows the oldest man-made source in Slovenia which was actually found during the inspection campaign in 2007. It was produced in 1902 and contains Ra-226 of 0.15 MBq. The source was used in ophthalmology and later just stored in an abandoned storage as a non-registered source. Finally it was found provisionally shielded in the storage at the Institute of the history of medicine.



Figure 1. The oldest man-made source used in Slovenia. The Ra-226 source of 0.15 MBq was produced in 1902 and it was used in ophthalmology. Later it was lost and in 2007 it was found in an abandoned storage during an inspection programme of finding non-registered sources in the state.

As a rule and in accordance with expectations no short lived radioisotopes were found originating from the past activities. Nevertheless, in some specific cases sources were actually acquired only a year or two before the inspection campaign of non-registered sources started. In such cases the inspection identified that either the producer of the source was located in the state outside of the EU or the final seller of the source was located outside of the EU. In both cases the legislation of a producers' or seller's state was not harmonised with the EURATOM directive.

The inspection programme resulted not only in an identification of numerous non-registered sources or radioactive waste but also other actions followed in accordance with present legislation. The safety assessment was performed. All sealed sources identified contained a radionuclide whose activity was bellow the activity of corresponding so-called "high-activity source" given in the HASS directive. An assessment of possible contamination was performed if unsealed sources were found and safety measures were put in place if necessary. A registration of sources or their full scope licensing followed. The owners of the sources became aware of the radiation safety or nuclear safety issues, as appropriate. In cases when owner identified sources as radioactive waste such waste was stored in the central national storage for radioactive waste according to the procedure required, e.g. the liquids were processed and only radioactive material in a solid form was stored.

Research institutions

Several research institutions were inspected during the campaign. The first of them was the institute dedicated to physics in the past, followed by the chemical institute, geological institute etc. Such sources or radioactive waste were identified in laboratories where activities with sources have been still going on as well as in local owner's storages. Very seldom such storages were actually abandoned storages. Many sources were related to past activities with uranium and termination of such activities, among them also about 500 litres of contaminated liquids and several hundreds kilograms of yellow cake was found not to mention contaminated equipment. Details of termination of research activities with uranium compound are given in [16, 17]. Due to the termination of activities a special precaution was taken in order to identify contaminated equipment which was actually or potentially present in laboratories, e.g. pipes, ventilation, liquid storages, fume hood. In addition, the inspection also identified that very often technical data for the sophisticated equipment, e.g. gas chromatographs,

liquid scintillation counters (LSC), etc. does not include enough information about a radiation source which is installed in the equipment. Such sources can be purchased by a research laboratory not knowing that special precautions are necessary. A list of such laboratory equipment includes so-called chemical detectors, e.g. ion mobility spectrometers – IMS with Ni-63 or Am-241, liquid scintillation counter with Cs-137, Eu –152 or Ra-226 and electron capture detectors with H-3 or Ni-63. Details including are given in [18]. The inspection also identified a new non-registered uranium source used in the laboratory which was imported from the county that does not require the same level of protection for export of such material as required in the EU. Figure 2 shows storage of uranium compounds found at the research institute as well as a new non-registered source identified at an inspection.



Figure 2. Uranium compounds found at the research institute (left) as well as a new non-registered source identified at an inspection of a research laboratory (right).

Educational institutions

The educational institutions being inspected include a large spectrum of faculties among them medical faculties, faculties for chemistry, pharmacy, geology, physics, civil engineering, biology, agriculture. The laboratories used in an educational process were inspected as well as storages used in everyday life of such institutions. In addition, collections of valuable items used for research and educational purposed were inspected and many items which were used in the past, being later stored in dislocated storages. The educational institutions used equipment with radiation sources of the same type as the research institutes, e.g. IMS, ECD. In 2007 the long-lived Tc-99 source of 2.5 GBq was found in the chemical laboratory fully occupied by students. Due to a very specific educational programme radioactive sources used in education may be also installed in a unique apparatus as for example Am-241 source of activity of 3.8 GBq in a prototype apparatus for measurement wood density. In addition, in some cases educational institutions stored unconventional sources acquired decades ago that were not used in the recent years. Figure 3 shows such unconventional sources of unknown origin identified at the faculty of physics during inspection which were acquired decades ago and have not being used for a long period. Figure also shows the abandoned LSC at the faculty of medicine which contained Ra-226 and was not in a use for years. The educational institutions also used solid and liquid uranium and thorium compounds which can easily contaminate school laboratories. Such contamination was actually identified during inspections. In addition, when inspecting faculty for geology beside the man-made sources a numerous radioactive geological samples and mineral collections were identified causing a quite large dose rate at laboratories, offices as well as collections of minerals at corridors and halls. Moreover, such inspections found that two additional issues must be taken into account, namely possible contamination as well as radon issues. Details are given in [19, 20].



Figure 3. Unconventional non-registered sources of unknown origin identified at the faculty of physics which were not used for years (left). An abandoned LSC used the faculty of medicine in the past which include Ra-226 (right).

Defence and civil protection institutions

Altogether 19 inspections were performed in the defence and civil protection institutions. At the time of inspection some sources built in modern inspection instruments were already under the regulatory control, e.g. sources installed in apparatus for tracing chemicals (e.g. M90, ChemPro, Vapour Tracer). A use of tritium in defence activities is also very frequent and well known. Such a use was licensed as appropriate. Other users of radiation sources in civil protection and defence institutions were not identified prior the inspections and hundreds of non-registered source of various origin were identified. As a rule the equipment which was acquired decades ago was not labelled with a label showing that a radioactive source was installed in it. Moreover a database of the equipment did not contain information about the radioactive source. In the past the radioactive paint with Ra-226 was extensively used in equipments such as compasses, altimeters and other measuring gauges in vehicles, tanks and airplanes. Special precautions are necessary because of possible contamination during reparatory and maintenance works. Contaminated areas within the military workshops were actually identified. Elevated radiation fields were measured at workplaces where workers were in a close vicinity of a circularly located set of instruments with radium paint.

A list of equipment with non-registered sources includes also unconventional sources:

- parts of some radar systems, e.g. electron gun "Twystron" causing increase dose rate up to few micro Sv/h at the workplace of an operator

- H-3 or Ra-226 weapon aiming devices
- H-3 in military telescopes and binoculars
- radioactive source as a part of TLD calibration systems
- optical lens containing Th-232

- calibration sources used for field instrument measurements of radiation with Sr-90 or Cs-137 with levels exceeding several times exemption levels.

The last type of sources found can pose specific issues because they can be a part of the instrument or a part of the transport package, e.g. a plastic housing and as such a control over these sources can be easily lost. In addition, such instruments were widely distributed in a state for civil protection purposes and as a rule never used. Figure 4 shows two types of non-registered sources, namely Ra-226 radioactive paint used in an old instrument causing s dose rate more than 9 μ Sv/h (left) and a new instrument with 6 MBq Am-241 installed in a ChemPro gas analyser (right).



Figure 4. Two types of non-registered sources, namely Ra-226 radioactive paint used in an old instrument causing s dose rate more than 9 µSv/h (left) and a new instrument with 6 MBq Am-241 installed in a ChemPro gas analyser (right).

The collectors of military items can easily acquire sources installed in some of the instruments or items which contain the Ra-226 paint. Inspections were also performed in a military museum where such sources were identified. Figure 5 shows a typical military aiming device with non-registered Ra-226 sources inside it.



Figure 5. A typical military item with non-registered Ra-226 source inside it. No label related to radioactivity was used at the time the equipment was acquired.

Museums and collectors

The non-registered radiation sources can be also present in the museum collections. Two types of enhanced risks related to exposure to ionising radiation were identified. Most frequently the sources are radioactive minerals or other items in geological collections, e.g. radioactive mastodon tusks, but beside those also radioactive materials can be applied or installed in the instruments. Geological samples can cause very high dose rate in cases where hundreds of radioactive items are stored in a small place. Radioactive geological items were present in showrooms, offices as well as in storages. Details are given in [20]. A special issue is the existence of private collectors who handle such items without proper safety measures, e.g. in one case such items were sent by a post as a gift of a private collector to a museum. Due to the uranium mine which operated in the past in Slovenia, radioactive items causing elevated radiation levels are easily present also nowadays in museum and private collections.

In addition, a search for man-made items with radiation sources in museums was performed in order to identify them since numerous such items are known from the literature [21]. Till now the SNSA

identified items with Ra-226 paint or a use of uranium material in ceramics, but only a limited number of such items is a matter of concern related to elevated high dose rates or high radon emanation. In 2010 portal monitors were already triggered two times by museum items, i.e. by Ra-226 emanators produced before Second Word War which were found in a scrap metal.

3. Conclusions

A strict control of all radiation sources and radioactive waste in the state is one of the main tasks of the regulatory authority in order to protect humans and the environment from accidents or malevolent acts. A focused state campaign in order to identify all non-register sources is a first step in order to clean the territory of a state and to put the safety measures in place. In parallel, a strict control over new sources being produced, imported or otherwise transferred in the state should be enhanced.

In Slovenia around hundred inspections were focused on identification of non-registered sources and waste in the period 2004-2010. Around thousand of various sources or radioactive waste items were identified at research, educational, military institutions as well as in collections. None of the sealed source can be defined as a HASS source according to the HASS directive of the EU. The inspection programme resulted not only in identification of sources or radioactive waste but also other actions followed in accordance with the present legislation. The safety assessment were performed, safety measures were put in place if necessary, e.g. decontamination, as well as a registration of sources or their full scope licensing. The owners of the sources became aware of the radiation safety or nuclear safety issues, as appropriate. In cases when owner identified sources as radioactive waste such waste was stored in the storage for radioactive waste according to the procedure required, e.g. the liquids were processed and radioactive material in a solid form was stored.

Due to numerous reasons non-registered sources, especially sources used in the past, still pose a threat. The treat is not limited only to a specific country but due to the global trade it is international. National campaign conducted in all countries on their own territories can contribute to decrease a level of this threat.

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