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Environmental radioactivity aspects of recent nuclear accidents associated with undeclared nuclear activities and suggestion for new monitoring strategies

Jerzy W. Mietelski[a,*], Pavel P. Povinec[b]

[a] Institute of Nuclear Physics, Polish Academy of Sciences, 31-342, Krakow, Poland
[b] Faculty of Mathematics, Physics and Informatics, Comenius University, 84248, Bratislava, Slovakia

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ABSTRACT

Recently environmental radionuclide signals were observed in the atmosphere which could be associated with undeclared nuclear activities, not directly connected with development of nuclear weapons. Large-scale contamination of European air with Ru-106 observed in 2017 may represent such an accident, which was probably associated with the Mayak nuclear fuel reprocessing facility in the Chelyabinsk region of Russia. A recently announced nuclear accident at Nyonoska in the Archangelsk region may represent an undeclared nuclear activity associated with the development of a nuclear jet engine which could be based on radionuclide energy generator or a small nuclear reactor. It is concluded that environmental radioactivity impacts associated with recent nuclear activities create new challenges for fast and reliable national and international monitoring systems which would require development of new monitoring strategies.

1. Introduction

Environmental radioactivity impacts of recently discussed nuclear accidents which have not been directly associated with developments of nuclear weapons present new challenges in monitoring of radionuclides in the atmosphere (Masson et al., 2018, 2019). Some of these accidents were associated with undeclared nuclear activities, the others were associated with radionuclide production for scientific and medical applications. It has been interesting that some of the accidents have been observable even on the global scale (Masson et al., 2019), although some of the accidents had only a regional character.

The most widely discussed accident was associated with elevated levels of radioactive ruthenium-106 (Ru-106) and its daughter rhodium-106 (Rh-106) which were observed on atmospheric aerosols in many European countries at the beginning of October 2017, and at much lower levels also in America (Masson et al., 2019). This accident probably happened at the Mayak nuclear fuel reprocessing facility in the Chelyabinsk region (east of Ural Mountains in Russia), although we cannot rule out other possibilities associated with the development of a nuclear engine for propulsion of missiles.

The second undeclared nuclear accident, which has recently been discussed, occurred in Nyonoksa in the Archangelsk region on 8th August 2019. Although this accident did not cause any measurable radionuclide levels in the European air, it is expected that it could be associated with tests of nuclear fuel engine for propulsion of missiles.

The aim of the present paper has been to discuss possible environmental aspects and potential explanation of recent undeclared nuclear accidents at Chelyabinsk and Archangelsk regions, to point out on new challenges and consequences of such nuclear tests on radioactive contamination of the environment and to propose new radionuclide monitoring strategies for the future.

2. Ruthenium-106 accident

2.1. Ruthenium-106 levels in Europe

At the beginning of October 2017 Ru-106 and its daughter Rh-106 were observed on atmospheric aerosols in many locations in Europe and at much lower levels also in America. Thanks to Ring of Five (Ro5) network and coordination of European radionuclide monitoring activities, detailed sampling and evaluation programs have been going on which enabled to draw a map of Ru-106 distribution in the European air (Masson et al., 2019).

Fig. 1 shows the main European sampling sites where elevated Ru-

* Corresponding author.
E-mail address: jerzy.mietelski@ifj.edu.pl (J.W. Mietelski).

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106 concentrations on aerosols were recorded. The maximum Ru-106 levels in the air (about 150 mBq/m^3) were observed in Romania (IRSN, 2018; Masson et al., 2019), followed by close to 60 mBq/m^3 levels in certain locations of Bulgaria (Penev et al., 2018), Hungary (Jakab et al., 2018) and Czech Republic. Lower levels were observed at many other European monitoring sites, down to about 0.01 mBq/m^3 (IRSN, 2018). Some very small traces of Ru-103 (0.001–0.007 mBq/m^3) were found at some Austrian, Czech, Polish and Swedish stations as well. The observed levels were too low to pose any problems on humans and the environment, they were interesting just from scientific point of view of their sources and observed variations in the atmosphere (IAEA, 2017).

It should be noticed that there are two types of Ru-106 data presented in Fig. 1. The circles represent maximum Ru-106 values observed at monitoring stations, mostly only for one day. As the movement of the radioactive cloud over the sampling regions was within one-two days, a significant fraction of uncontaminated air was sampled on the aerosol filters if the sampling was carried out during traditional one-week sampling. The long-term sampling thus caused that reported Ru-106 activity concentrations in the air.

More than a month later after the observed large scale contamination in Europe, in mid-November 2017, it was revealed that a Ru-106/Rh-106 contamination was found also on the ground in the Chelyabinsk region, close to the Mayak facility, where reprocessing of spent nuclear fuel has been taking place (TRV, 2018; IRSN, 2018). The deposition rates of Ru-106 (shown in Fig. 1 in boxes) observed from 23 to 29 September 2017 at the Mayak area (Kyshtym, Argayash, Novogornyy, Khudaiberdinski) were from about 10 to 340 mBq/m^2 (Roshydromet, 2018).

The observed Ru-106 distribution has been well fitting the mountain topography of the Europe (Fig. 1). Detailed modelling studies suggest south-west transport of air masses from the east of Ural Mountains to the Central Europe, continuing then to Scandinavia (IRSN, 2018; Masson et al., 2019; Povinec et al., 2019). The site of the Ru-106 release could be therefore in the Chelyabinsk area. It has been estimated that the release occurred during one day between 25 and 28 September. The required source term for the observed Ru-106 contamination was estimated to be 0.1–0.3 PBq (Masson et al., 2019). The maximum Ru-106 deposition rates observed around the Mayak reprocessing facility (340 mBq/m^2) for this source term looks low. For comparison, Ru-106 deposition rates observed in Krakow between 2 and 4 October were 78 Bq/m^2 (Masson et al., 2019).

2.2. Ruthenium-106 characteristics

Ru-106 is a typical fission product with a half-life of 371.8 day, present in spent nuclear fuel. In routinely reprocessed nuclear fuel, the cooling time is in the range of at least 10 years, so the Ru-106 activity would decrease by a factor of greater than 2^{10} (i.e., more than 1000 times). There is another Ru radioisotope produced in fuel, Ru-103, which has a shorter half-life time of 39 days only. The probability of production of the mass 103 in a fission of U-235 is 3.10%, smaller than for the mass 106 (4.20%). To get pure Ru-106 one has to wait for enough Ru-103 half-life times – let say about two years. In the observed air contamination of Europe in October–November 2017 the activity of Ru-103 was by a factor of about 3500 lower than that of Ru-106/Rh-106 (Masson et al., 2019), which in general supports the concept of about two years of cooling time of the spent fuel from which ruthenium was separated.

Ru-106 is a pure β-emitter with a very low energy of the emitted β-particles (maximum at 39.4 keV). However, its decay product, Rh-106, is also a β-particle emitting radionuclide, with a half-life of 29.8 s only, and a high maximum energy of the β-particles of 3541 keV. Rh-106 emits several γ-rays of which the most intense have energies of 511.8 keV (the emission intensity is 20%), 621.9 keV (9.93%) and 1050.4 keV (1.56%) (Nucleardata, 2019). The short half-life of the daughter radionuclide results in the establishment of a secular equilibrium between them within a few minutes.

The main problem with separation of Ru from nuclear fuel is, however, that besides Ru-103 and Ru-106 also stable Ru isotopes (Ru-101, Ru-102 and Ru-104) are produced, which cause that Ru-106 is isotopically diluted in stable ruthenium. Ruthenium forms many compounds. In metallic form poses large density of 12.1 g/cm^3 (enabling effective self-absorption of the emitted β-radiation), and the high melting point of 2606 K (RSC, 2019).

2.3. Mayak reprocessing plant hypothesis

The Mayak nuclear fuel reprocessing facility has been considered as possible site of Ru-106 release with consequences on Ru-106

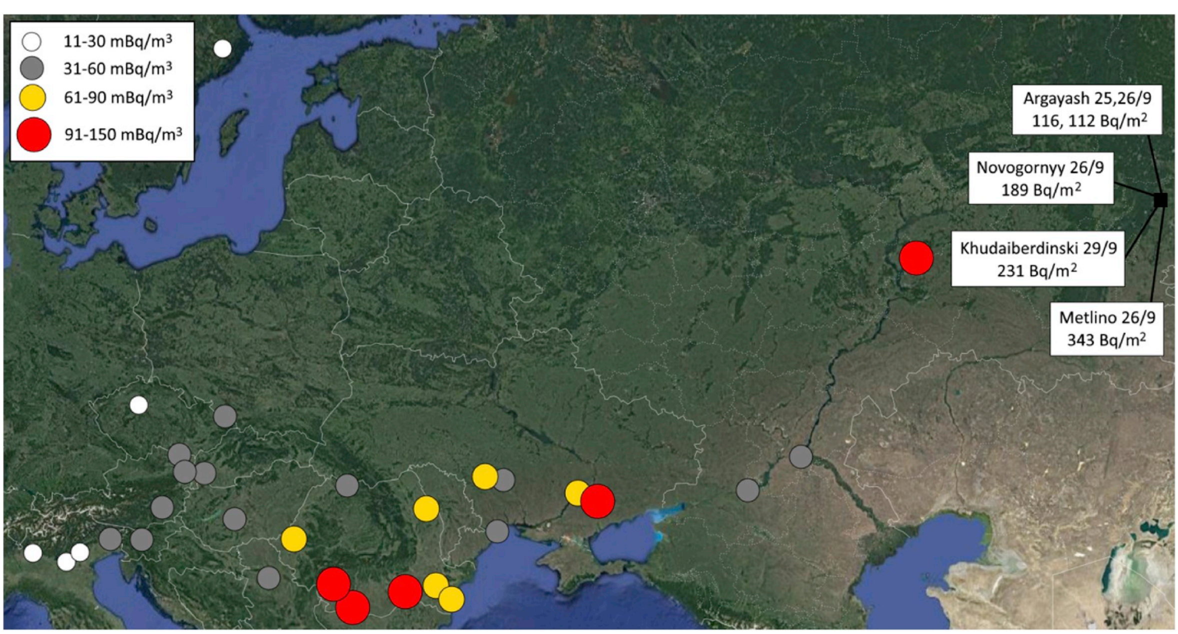

Fig. 1. Activity concentration of Ru-106 on aerosol filters at the beginning of October 2017. The circles represent maximum observed activities during 1-2-day sampling, the boxes contain maximum deposition rates observed in the Chelyabinsk area (data from IRSN, 2018).

contamination of European air (Ramebäck et al., 2018; Masson et al., 2019), which could be associated with reprocessing of relatively fresh nuclear fuel containing high Ru-106 levels. A need of such early reprocessing was supposed to be related with a production at the Mayak reprocessing facility of the Cerium-144 SOX source (Short Distance Neutrino Oscillation Source for BoreXino), within a contract signed with Borexino experiment conducted in the Gran Sasso National Laboratory in Italy (Durero et al., 2016). It should be noted that the Ce-144 has a half-life of 284 days, similar to that of Ru-106. The probability of production of the mass 144 in a fission of U-235 is 3.76%, a bit smaller than for the mass 106 which is 4.20%. The Ce-144 was chosen by the Borexino collaboration to produce the SOX neutrino source with activity of about 5 PBq. The preparation of the Ce-144 source requires work with briefly cooled nuclear fuel (Gerasimov et al., 2014), and it was very likely that both Ce-144 and Ru-106 were obtained during the same campaign in the Mayak facility. Recently, the Ce-144 production was, however, cancelled by the Mayak institution (Sciencemag, 2019), which would indicate unsuccessful operations in the Mayak facility.

(i) why there was an accident with production of Ru-106 when Ce-144 should be produced for the SOX neutrino source?
(ii) The estimated accidental release of Ru-106 (0.1–0.3 PBq) is too small, when compared with the expected activity of the Ce-144 SOX neutrino source (about 5 PBq).

The appearance of the Ru-106 contamination created general confusion in many agencies responsible for nuclear safety since nobody admitted to the site or circumstances of a release. Finally, in late November an official Mayak spokesman admitted that the Mayak was the place of the release of Ru-106, after initially denying it (Cartlidge, 2018), later, however, possible releases from the Mayak were denied again.

The whole affair remains mysterious and this announcement raises doubts since no clear effect related to a decreasing concentration of the contamination with distance from the Mayak has been observed. Such a late and dubious announcement has been posing questions about the true origin of the contamination. Therefore, up to now, there is no reliable information on the potential source of the Ru-106, and we should not rule out possible explanation that the accident could be associated with a test for separation of larger Ru-106 activities which could be used as nuclear engines for the propulsion of rockets.

2.4. Nuclear jet engine hypothesis

The concept of using nuclear energy for the propulsion of aircrafts and rockets originated already from the time of WW II. The reason is obvious – a nuclear engine would represent almost unlimited range of any kind of aircraft or rocket driven by such propulsion. In principle, the idea is simple: the thermal energy needed to drive the engine comes from a nuclear process instead of the combustion of a traditional fuel. In the mid-1950s some test installations were constructed when the heavy US bomber Convair B-36 was modified to carry a nuclear reactor Cortright (1995). It made several flights, but no actual in-flight tests of the nuclear engine were carried out. The USA also tested a nuclear ram-jet on ground within the so-called Pluto project. Far less is known about the development of similar ideas in the former Soviet Union during the Cold War era. One can expect that similar attempts were done there as well. For example, it is known that a nuclear reactor was installed on a Tupolev Tu-95 LAL Bear (NATO code name) bomber and tested in 1961–65, followed by an experimental unfinished project using Tu-119 aircraft (Colon, 2009).

Despite the obvious advantages of nuclear propulsion, primarily with unlimited range of an airplane, this concept was discontinued under pressure of public opinion due to safety reasons. However, nuclear propulsion has still been under development for spacecrafts as modifications of rocket engines. A review of current technical projects of nuclear engines was recently presented by Gabrielli and Herdich (2015). There are new ideas coming up continuously in this field, however, in open scientific debates only spacecraft propulsion has been discussed recently (Nam et al., 2015).

2.4.1. Ruthenium-106 as an energy source

Modifications of any kind of a jet engine into a nuclear engine mean that in place of the combustion chamber, the heat source made of a nuclear reactor or a strong radioactive source is installed instead. The advantage of using strong radioactive sources is obvious in its relative simplicity. The radionuclides which can be used here are maybe the same which have already been used in Radioisotope Thermoelectric Generators (RTG) (Gabrielli and Herdich (2015), which are known from much more than 50 years now. They are based on the transfer of energy from radioactive decay into e.g. electric energy through conversion of the thermal energy (heat) produced in radioactive decay (Fig. 2).

For example, the Ru-106/Rh-106 generator may be a very good choice because of relatively long-lived parent and short-lived daughter of a high decay energy. The nuclear energetic aspects of Ru-106 properties were previously underestimated (Gabrielli and Herdich (2015), as the radioactive equilibrium with its daughter nuclide, Rh-106, had not been taken into account.

There are, except of the Ru-106/Rh-106 system, a few similar high energy daughter generators, e.g. Ar-42/K-42 and Sr-90/Y-90. Especially, the Sr-90/Y-90 system had been frequently used in the former Soviet Union for energy production, e.g. for lightning sources at sea, which because of the long half-life of Sr-90 (28.81 years) could operate tens of years without any surveyance. Some of these sources were lost in the Okhotsk Sea and in the north-west Pacific Ocean, having Sr-90 activities from 1.3 to 25 PBq (Ikeuchi et al., 1999; Livingston and Povinec, 2000; Hirose and Povinec, 2019).

The generated thermal power P of an RTG is a product of its total activity A (which is a function of time t) and the mean energy of the radiation deposited inside, E_d

$$P = A(t)E_d \tag{1}$$

The relatively short half-lives of Ru-106 and Rh-106, and the high energy of the β-radiation of the daughter nuclide make the Ru-106/Rh-106 generator a strong candidate for a nuclear engine. However, a presence of stable ruthenium in the system spoils high energy density produced in Ru-106/Rh-106 decay. Very efficient decay energy production could be brought back if the Rh-106 would be on-line separated from ruthenium (since they are isotope generator systems), and then rhodium is trapped inside a heat exchanger, located inside the jet engine.

We expect that nuclear engines based on the RTG will drive small

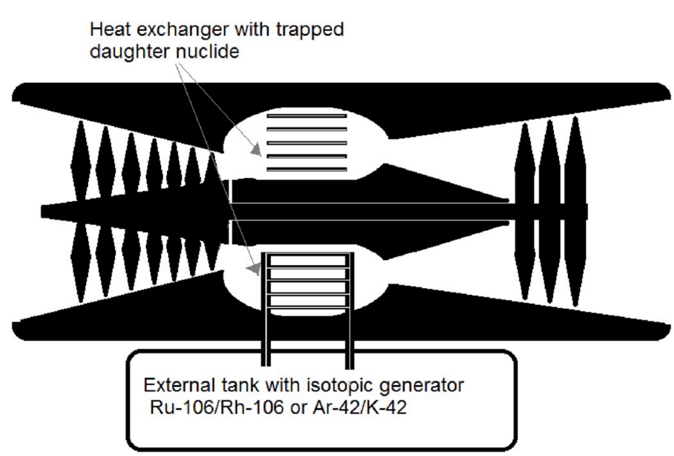

Fig. 2. Schematic idea of a simple turbo-jet engine driven by isotopic generator sources.

missiles, only a few meters long. For example, a typical Russian cruise missile of type KH-101 (a conventional warhead) or KH-102 (a nuclear warhead) is driven by a turbofan engine with a maximum thrust of 4.5 kN, moving with a speed of about 200 m/s (MDAA, 2018). Multiplication of those two numbers gives power requirement of 900 kW. The maximum thrust is used, however, only during take-off, the power required to drive such a big missile would be much lower if for take-off an auxiliary jet booster is used. The power obtained from the radioactive decay of Ru-106 using equation (1) would require an activity of the Ru-106 source at the level of about 500 PBq per each 100 kW (or about 10% less if the energy deposited by gamma-rays would also be efficiently absorbed).

Such high active source (i.e. of level of 10^{18} Bq) of Ru-106 in case of accidental crash in accident would do serious radioactive contamination. If it is a local contamination on 1 square km, the average activity would be at level of TBq/m^2. Such a high active source is from the dosimetry point of view also a very difficult material for handling. The gamma dose rate for Ru-106 in equilibrium with Rh-106 constant is ca. $3.7{\cdot}10^{-5}$ mSv m^2/h MBq (one tenth of the value for Co-60) (Umger and Trubey, 1982), resulting in $0.35{\cdot}10^5$ Sv/h at 1 m distance for a 1 EBq source and still 3.5 Sv/h at 100 m distance. The gamma radiation quality is similar to that of Cs-137, giving a one-tenth layer thickness of 21 mm for lead (Smith and Stabin, 2012) (to be compared with 21.8 mm in the case of Cs-137 (Smith and Stabin, 2012)). If one will consider tungsten as an appropriate shielding material – for 0.6 MeV gamma radiation the one-tenth layer thickness is 15 mm, a 10^{-4} layer thickness is 51 mm and attenuation to 10^{-6} requires 78 mm (Sauermann, 1976)). With a mass attenuation coefficient of 0.0793 for ruthenium (Hubbell and Seltzer, 1995), gamma self-absorption would reduce the external dose rate to 30% of the initial value, resulting in $5.4{\cdot}10^4$ Sv/h. A 50 mm tungsten shielding, making the handling of the source more manageable, would add another 150 kg to the mass and reduce the dose rate to ca. 5 Sv/h at 1 m distance.

For a small missile (or rather a drone) with a nuclear engine, the power requirements should be smaller at least by a factor of 10 or even 100, therefore 5–50 PBq of Ru-106 would be enough to drive such small missiles (drones). The predicted Ru-106 activities would be thus comparable with Sr-90 activities already used in the RTG for lightning sources. The required Ru-106 activity is smaller than for example the activity of Ru-106 present in the core of the RBMK reactor in Chernobyl at the moment of the accident (2 EBq, IAEA, 1986). This means, that to separate such an activity of Ru-106 one has to process fuel rods from less than one nuclear reactor (of $\sim$1 MW$_e$), taking into account a fuel that was cooled for about two years to eliminate Ru-103.

2.4.2. Possible mechanisms of environmental contamination by Ru-106 from nuclear engines

The release of radioactive ruthenium could happen during preparation tests or as in-flight release (a leakage) from a ruthenium jet engine. One can consider as a rocket testing ground to be the Makayev Rocket Design Bureau located at Miass (which is only about 100 km westward from Mayak), as the starting place (and ground test) and that the flight took part northwards. This assumption would explain why a radioactive contamination was observed in the Mayak area. The choice of this test site could be convenient for a short-range transport of highly radioactive Ru-106 sources. A leakage of Ru-106 from a jet engine is proposed as a mechanism for the production of the detected airborne contamination, as the estimated Ru-106 source term for the contamination of the Europe was below 0.3 PBq.

3. Nyonoksa nuclear accident

The second undeclared nuclear accident recently discussed is the Nyonoksa accident which happened on 8th August 2019 in the Archangelsk region. It is quite possible that this accident could happened during tests of Ar-42/K-42 generator system, which could be similar to already discussed Ru-106/Rh-106 system. If radioactive argon was used as a fuel, this would explain why there was a lack of serious radioactive contamination on site after the accident. Moreover, it fits to first official announcement on accident which happened during "test with liquid isotopic source", as we expect that the fuel was in the form of supercritical (in thermodynamic, not nuclear sense of word "critical") liquid argon.

The Ar-42/K-42 generator would be more practical to use in nuclear jet engine due to long half-time (32.9 years) of the parent nuclide, and much different chemical and physical properties of Ar and K which makes separation in heat exchanger much easier. The production of required activity of Ar-42 is more difficult when compared to Ru-106, however, it is technically feasible. It is quite possible that in such case the Ru-106/Rh-106 system could be treated as a feasibility study for future development of the Ar-42/K-42 isotopic system for jet propulsion. The test of Ru-106/Rh-106 model system could be useful to motivate that efforts needed to produce Ar-42. However, in both cases the needed activity to drive even a small missile would be probably above 10 PBq.

There have been ongoing experimental efforts to look for increased Ar-42 levels in the European air, however, till now no clear excess of Ar-42 was found. Modelling exercises are in progress as well, which should indicate possible transport of air masses from Archangelsk region to central Europe. Therefore, it has not been possible yet to estimate Ar-42 activity which could be released during the Nyonoksa accident.

The other possible explanation is that a small nuclear reactor is an energy source for the jet engine and in Nyonoksa accident it became critical on prompt neutrons. However, such an accident under normal circumstances should produce wide range of radionuclides including also long-lived fission products, which apparently were not discovered in air so far.

The alternate to Ar-42/K-42 isotopic generator as source of energy to drive nuclear engine is use of unspecified kind of nuclear reactor. Since the missile is unmanned, there is no need of carrying heavy biological shields on board, however some light shields to protect electronics in control systems will still be needed. So, the alternate hypothesis on what really happened in Nyonoksa is the scenario with a criticality event of unshielded reactor, maybe even related with a loss of control –with criticality on prompt neutrons followed by destruction of the reactor. This could explain lethal doses to victims of accident and it is in some way suggested by official announcements about presence of short lived fission products identified on site. However it does not explain why there is no even tiny traces of long lived contamination (fission and activation products, perhaps also traces of a nuclear fuel) and it does not explain why the isotopic signature of event was that much characteristic, that five CTBTO (Comprehensive Test-Ban Treaty Organization) station in Russia had to be turned down.

However, the Nyonoksa accident may confirm the official statement of the Russian Federation President V.V. Putin in March 2018 at the Federal Assembly about on-going developments, in-flight and ground tests of nuclear-powered engines as a new propulsion system of cruise missiles (Putin, 2018).

4. Consequences for monitoring strategies

Undeclared nuclear activities associated not only with development and explosions of nuclear weapons, but also with development and tests of nuclear engines for missiles should be expected in the near future, both in the atmosphere and the marine environment (nuclear torpedoes). A development of underwater systems for detection of reactor neutrinos for localization of nuclear submarines could be another example of nuclear activities which are surely on going in military centers.

The possibility of conducting tests of different kinds of nuclear engines in the environment (air or sea) opens new challenges for monitoring of radionuclides in the environment. This has also been great satisfaction for successful operation of the Ro5 atmospheric radionuclide monitoring network, which is based only on unformal, purely scientific initiative to carry out regular monitoring activities with well-organized exchanges and discussions of results (Masson et al., 2018, 2019). This has also been a challenge for the CTBTO network established for monitoring of nuclear weapons tests (CTBTO, 2018).

Currently, the worldwide environmental radioactivity monitoring systems are mostly based on γ-spectrometry since they are oriented on radionuclide releases related to fission products from nuclear reactors and from nuclear weapons tests. A wide range of different radionuclides can be released from nuclear installations depending on the thermal conditions of the release. The easiest radionuclides to be released are the radioactive noble gases, followed by radioisotopes of very volatile elements like iodine and tellurium, and then by the less volatile ones like cesium, strontium, etc.

However, it seems to be possible to have a large environmental contamination due to radionuclides which are not γ-emitters attached on aerosols (e.g. such as Ar-42), which need not be easily registered, so a serious radioactive contamination could be overlooked or discovered too late. For example, if we consider the case of strong RTG sources from which, in the case of accidents, large quantities (>1 PBq) of radionuclides like Sr-90/Y-90, Ar-42/K-42, Ru-106/Rh-106, Pm-147, Po-210, Pu-238, Cm-242 and Cm-244 could be released to the environment. Some of these radionuclides are only pure β or α-emitters so presently operating monitoring systems will not register them, as both the laboratory and field instruments are mostly based only on γ-spectrometry. It has been established from previous nuclear accidents that already releases above 1 PBq may represent a radioecological stress for the terrestrial and marine environments with possible impacts on the population (Livingston and Povinec, 2000; Povinec et al., 2013).

These have been new challenges which require serious debates in European countries and responsible international organizations about the methods of environmental radioactivity monitoring as a response to new types of undeclared nuclear activities, which in terms of released of some of the radionuclide activities could be comparable with Chernobyl and Fukushima accidents. Development of new monitoring strategies could also be important in connection with possible terroristic attacks accompanied with “dirty bomb” explosions (SSK, 2015).

5. Conclusions

Two undeclared nuclear accidents observed in 2017 and 2019 discussed in this paper could be directly or un-directly associated with developments of new nuclear engines for small missiles. The contamination of European air by traces of Ru-106/Rh-106 observed in autumn 2017 could be the result of an accident in the Mayak nuclear fuel reprocessing facility during separation of large Ru-106 quantities from spent nuclear reactor fuel rods. However, we cannot rule out that this accident could be associated with development of a nuclear engine to drive a small missile or a drone. As this has been an undeclared nuclear activity, the exact scenario of the accident which produced large-scale airborne radioactive contamination of Europe remains unclear.

The recent Nyonoksa accident (which is in line with official statements about developments of new nuclear-powered missiles in Russian Federation) could happened during preparation for tests of a nuclear engine driven either by a liquid radioisotope generator Ar-42/K-42, or by a small nuclear reactor, although till now we do not have any signals of possible radioactive contamination of the European air.

The very disturbing consequence of such tests is that a severe radioactive contamination on a scale comparable to radionuclide releases during the Chernobyl and Fukushima accidents could happen. The tests of new nuclear propulsion systems in the open environment may have serious consequences but also challenges for the world’s nuclear safety national and international monitoring networks (e.g. the CTBTO network, the Ro5 network) as new monitoring strategies should be developed, covering also presently not registered pure β and α-emitters in the atmosphere.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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