

Investigations on the source term of the detection of radionuclides in North of Europe in June 2020

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Abstract. During the second half of June 2020, small quantities of artificial radionuclides (^{60}Co , ^{134}Cs , ^{137}Cs , ^{103}Ru , ^{106}Ru , ^{141}Ce , ^{95}Nb , ^{95}Zr) have been detected in northern Europe (Finland, Sweden, Estonia), the source of the release being unknown. The measured values were close to detection limits and didn't represent any health issue. This paper presents the investigations carried out at IRSN in order to identify the release origin. The most probable source location and the release magnitude estimation are briefly presented. This recent set of detection is also compared to previous similar ones. This paper mainly focuses on the investigations which have been performed in order to answer two main questions. First "from which type and part of a nuclear installation the release could come from?". Although no certainty is achievable, the most probable source is found to be a spent primary ion exchange resin. The second question addressed was "how this radiological inventory could have been released into the atmosphere?". But, mainly due to the lack of information, no satisfying answer has been found to that question and what really happened remains unknown.

1 Introduction

In June 2020, several northern European countries have reported an increase of air concentration levels on aerosol filters sampled at stations belonging to their national monitoring networks. Various artificial radionuclides were detected. The Swedish and Finnish authorities reported that cobalt 60, caesium 134, caesium 137, and ruthenium 103 had been detected by their stations from June 8, 2020. Moreover, the same radionuclides were also detected in Estonia the last week of June. IRSN published an information report on that event [1].

The concentration levels were very small within the order of several $\mu\text{Bq}/\text{m}^3$ and did not pose any health or environmental effects. Maximum air concentration measurements (Fig. 1) were reported in Finland at Helsinki, from June 16 to 17, 2020. The limited number of measurements above detection limits makes it very difficult to pinpoint precisely the geographical origin of the detections.

In Section 2, the results about the possible locations of the origin of the release and its magnitude are presented. In Section 3, the characteristics and the estimated magnitude of each isotope are analysed in order to determine from which type and part of a nuclear installation the release could come from. Section 4 focuses on comparison with previous releases, and Section 5 presents some hypothesis

to understand how this radiological inventory has been released into the atmosphere.

2 Release location and magnitude

Inverse modelling techniques which combine atmospheric transport model and observations in the environment are applied to reconstruct the source location and its magnitude. This approach is described in detail in [2].

^{134}Cs air concentration measurements are the most numerous and are therefore exploited for source identification. It is first assumed that the release occurred somewhere between Germany and Russian Federation. For computation time reasons, the domain likely to contain the source is divided into a set of 348 grid points. Each centre of cell is then considered to be a potential source of release. Dimensions of the domain including the potential sources are [14E, 42E], [55N, 66N] with $1^\circ \times 1^\circ$ resolution.

For each potential source, a source term which represents the temporal evolution of the ^{134}Cs release rates is assessed by inverse modelling. The approach consists in the minimization of a cost function which measures the mismatch between modelled and observed ^{134}Cs air concentration. Regularization techniques are applied to ensure the existence and the unicity of the solution [3]. Then, for each optimal inverted source related to each grid point, the agreement between modelled and observed air concentration measurements is assessed using

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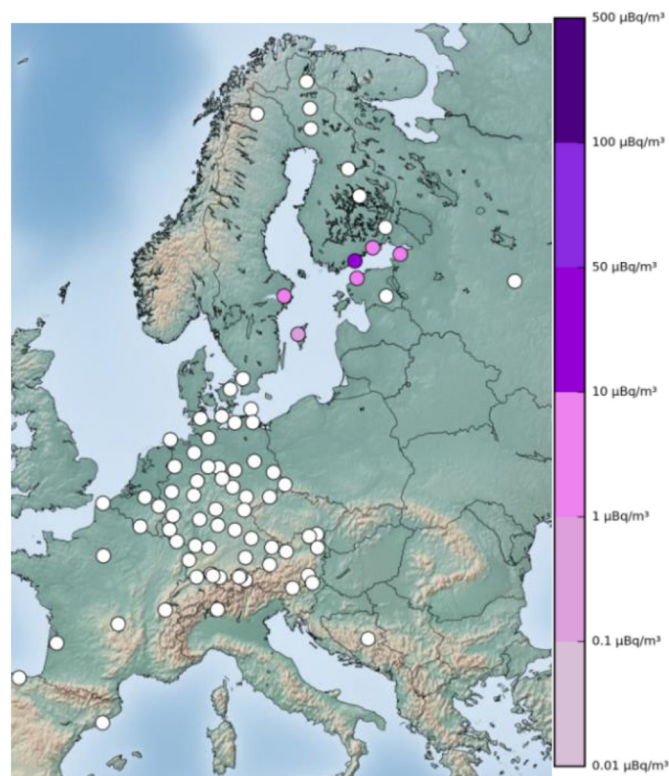


Fig. 1. Maximum ^{134}Cs air concentration values for each monitoring station ($\mu\text{Bq}/\text{m}^3$). White dots are air concentration measurements below the detection limit.

the factor 5 indicator (FAC5). FAC5 is the proportion of the simulated activity concentrations calculated using the reconstructed release that are within a factor of 5 of the observed values.

The interpolated values of FAC5 from each grid point are shown in **Figure 2**. Despite the small number of measurements used to evaluate each source term, the area where the FAC5 values exceeds 80% is small. This small area is identified as the most reliable source location according to calculations. It extends from eastern Estonia to western Russian Federation within which the FAC5 values reach 100% in an area restricted to the Leningrad region. Moving away from this area, the FAC5 values decrease rapidly indicating that the hypothesis of a release from Western Europe and Scandinavia is unlikely.

For the most plausible source location, the released activity of ^{134}Cs is around a few gigabecquerels (GBq). Based on the isotopic ratios obtained from the measurements of radionuclides detected in the environment (^{106}Ru , ^{103}Ru , ^{60}Co , ^{137}Cs), it can be deduced that several tens of gigabecquerels were released during the period considered.

Considering all the measurements, it can also be estimated that the release occurred from June 13 to June 22.

3 Investigations about the source term origin

The objective of this part is to determine from which type and part of a nuclear installation the release measured in June

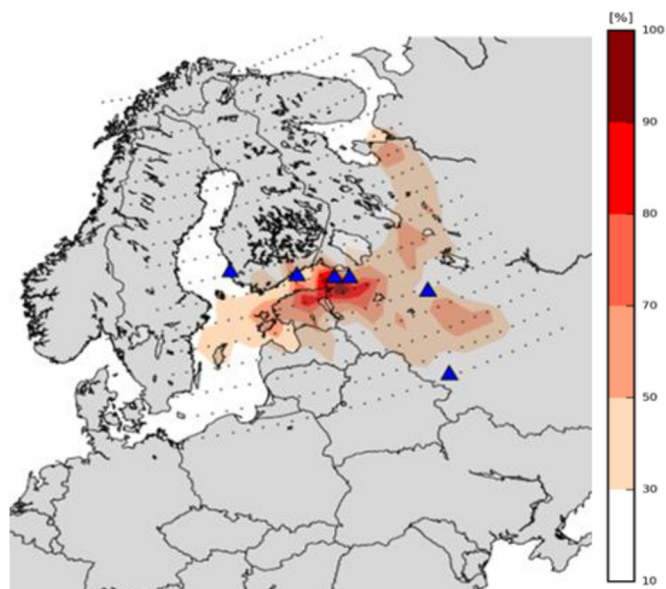


Fig. 2. Percent of the simulated air concentrations that is within a factor of 5 of the observed values. Blue triangles are nuclear plants located in the area of interest: Leningrad NPP, Petersburg Nuclear physics Institute (Gatchina), Olkiluoto NPP, Loviisa NPP and Smolensk NPP.

2020 could come from. For that purpose, the characteristics and the estimated magnitude of each isotope are analysed.

3.1 General information on the source term

Considering the ^{134}Cs release magnitude at the most probable release location, and the various activity ratios observed in the different measurements, the order of magnitude of each isotope of the source term has been evaluated and is presented in **Table 1**.

Some general comments can be drawn on that source term: – first, it must be noticed that it doesn't contain any short-lived isotopes (such as iodine). This tends to exclude an event concerning very recently irradiated material (less than a few weeks).

– Second, it contains some “intermediate half-life” isotopes (meaning about a few months) such as ^{103}Ru , ^{141}Ce , ^{95}Nb , ^{95}Zr . This implies that the event concerns relatively recent irradiated materials, with less than a year of decay. The presence of long-lived isotopes (^{60}Co , ^{134}Cs , ^{137}Cs , ^{106}Ru) is coherent with that assumption.

– Third, these isotopes come from various production sources;

- ^{60}Co is an activated corrosion product,
- ^{103}Ru , ^{134}Cs are fission products, caesium being volatile, ruthenium being non-volatile.

As a result, the origin of the release must be something mixing activated corrosion products and fission products, with probably a few months of decay.

3.2 ^{60}Co – presence of activated corrosion products

The evaluated source term contains a few GBq of ^{60}Co . This isotope is an activated corrosion product, produced by neutron capture on ^{60}Ni or ^{59}Co .

Table 1. Type and estimated order of magnitude of the source term.

Isotope	Half-life	Estimated Source Term	Origin
^{60}Co	5.26 y	~GBq	Activated corrosion product
^{134}Cs	2.07 y	~10 GBq	Volatile fission product
^{137}Cs	30.1 y	~10 GBq	
^{103}Ru	39.2 d	~GBq	Low-volatile fission product
^{106}Ru	371.8 d	~GBq	
^{141}Ce	32.5 d	~100 MBq	
^{95}Nb	35 d	~GBq	Low-volatile fission product or/and activated corrosion product
^{95}Zr	64 d	~GBq	

Such amount of ^{60}Co is expected to be found in only 4 places:

- on internal structures (steel) of a nuclear reactor. But this source can't lead to massive atmospheric release, and as a result is not an option;
- in the primary water of a nuclear reactor, due to corrosion of activated steel, or activation of corrosion products due to the neutron flux in the core. That inventory should be partially released into the atmosphere in case of primary water leakage outside of the containment. But, in such a case, a lot of other short-lived isotopes would be expected (such as ^{58}Co or iodine in case of fuel cladding leakage or failure). In addition, the amount of ^{60}Co that could be released into the atmosphere during such event is expected to be a few orders of magnitude too low to explain the measurements. So, this hypothesis isn't consistent with the measured data;
- in a ^{60}Co gamma radiation source. In common gamma radiation sources, the ^{60}Co activity can vary from a few GBq up to millions of GBq. But, due to the high chemical stability of cobalt, it's very unlikely to release a large part of that activity in the atmosphere, except in case of fire or mechanical cutting in the open air generating fine dust. In addition, in case of an accident on a ^{60}Co source, none other isotope would be expected, in contradiction with the presence of ^{103}Ru or ^{134}Cs . As a result, this hypothesis is not either an option;
- in primary ion exchange resin. These resins are commonly used to clean the primary water. They accumulate during several years the activated corrosion products of the primary water, and fission products in case of fuel cladding leakage or failure. The typical ^{60}Co inventory in a spent primary resin is about hundreds to thousands of GBq, which is fully compatible with a few GBq atmospheric releases. Some primary filter may also have significant amounts of ^{60}Co in particle form.

In conclusion, the primary ion exchange resin, or other component of the primary loop cleaning system (such as filter), are the best candidate to explain the presence of such amount of ^{60}Co .

3.3 ^{134}Cs and ^{137}Cs – presence of volatile fission products

^{134}Cs and ^{137}Cs , which were evaluated to a few tens of GBq each, are volatile fission products[†]. They are expected to be found only in spent nuclear fuel, or in the facilities processing them, except in case of fuel cladding leakage or failure during an operating cycle. In the June 2020 release, the hypothesis of a spent fuel assembly accident, or an event in a spent fuel facility can be excluded, due to the presence of ^{60}Co (not found in such quantities in a fuel assembly), and the presence of intermediate half-life isotopes (such as ^{103}Ru), despite the fact that spent fuel assembly generally decays during few years in the spent fuel pool before being evacuated.

In case of fuel cladding leakage or failure in an operating reactor, the caesium isotopes would easily migrate to the primary water. But again, in case of a primary water leakage, the amount of caesium released in the atmosphere would be too low to explain the June 2020 measurements. Besides, in the primary water, they will be cleaned by the primary ion exchange resin and accumulated in it. Considering their chemical form (ionic), the main part of caesium is expected to be found in the ion exchange resin, and not in the filters. Typical caesium activities in primary resin in case of fuel leakage or cladding failure during operation are from tens to thousands of GBq. Such amount could explain the June 2020 measurements.

The $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio is also a good indicator of the type of nuclear fuel involved. It has been measured from 1.2 to 1.7 (Helsinki station). That ratio could be obtained in almost any spent fuel of a Pressurized Water Reactor (PWR) (including WWER) after a few months of decay, depending of the enrichment and burn-up. But this value seems very unlikely to be obtained in a RBMK reactor. Indeed, the typical values at end of cycle are close to 0.5 and decrease after shutdown [4].

[†] ^{134}Cs is not strictly a fission product as it is mainly produced by the neutron capture on the ^{133}Cs , which is a “direct” fission product. Nevertheless, it is generally associated with the fission products, because it requires fission to be produced.

As a result, the best hypothesis for the caesium origin in the June 2020 release is a primary resin from a PWR, with a fuel cladding leakage or failure.

3.4 ^{103}Ru and ^{106}Ru – presence of low volatile fission products

The presence of ^{103}Ru and ^{106}Ru in an atmospheric release is particularly interesting. Indeed, ruthenium is a low volatile fission product, which mainly remains in the fuel pellet. It is very stable and only a very small fraction will be released out of the pellet, even in case of fuel cladding failure, except in case of high temperature fire ($>2000\text{ }^{\circ}\text{C}$) in oxidizing conditions. Moreover, the presence of the ^{103}Ru isotope, with a half-life of 39 days, is incompatible with the usual decay of spent fuel in the pool for several years.

Nevertheless, in case of fuel failure, some significant quantities of ^{106}Ru (from tens to thousands of GBq) can be released from the accumulated inventory in the fuel pellets. But, this migration from the UO_2 matrix to the gap is so slow, that the ^{103}Ru decays before reaching the primary water. It is mainly ^{106}Ru which is released by this way.

The only way to get a few GBq of ^{103}Ru outside of a fuel pellet is to have a fuel dispersion in the primary water. Indeed, some fuel cladding failure may lead to the dispersion of small amounts of uranium oxide outside of the pin, in the primary water. Exposed to the neutron flux, that fissile material will fission and release its fission products directly in the primary coolant, in an ionic form. They will then accumulate in the primary ion exchange resin (and not in the filters). Such events are easy to detect by an increase of short-lived isotopes in the coolant, such as ^{135}Xe , ^{138}Xe or ^{134}I [5]. They are quite rare, but considering a fleet of tens of nuclear reactors, they happened time to time. In that case, significant amounts of ruthenium could be released in the primary water and accumulated in the primary resin. It can be estimated that, with respect to the French operating limits, the amount of ^{103}Ru in a resin in such a case can reach hundreds to thousands of GBq. Considering a few months of decay, the inventory will remain of tens to thousands of GBq, consistent with a release of a few GBq.

Similarly, to the caesium isotopes, the $^{106}\text{Ru}/^{103}\text{Ru}$ ratio can provide some additional information. The Visby (Sweden) measurements, the only one to detect some ^{106}Ru activity, enable to evaluate it to 0.85–3.1. Assuming that ruthenium came from fission from dispersed fuel in the primary water (and not from the accumulated inventory in the fuel pellets), it can be evaluated that the decay time of the resin should be in the 4–9 months range, which is consistent with the previous conclusions. Nevertheless, considering a part of the ^{106}Ru inventory may come from the pellet inventory (and not the fission in the water), the ruthenium ratio is over-estimated. So, the only reliable information coming from this ratio is that the decay time is less than 9 months.

As a result, the best hypothesis to explain the ^{103}Ru origin of the June 2020 release is a primary resin from a PWR, after a few months to 9 months of decay, with a fuel cladding failure leading to a fuel dispersion in the primary loop.

3.5 Other isotopes (^{141}Ce , ^{95}Nb , ^{95}Zr)

The same analysis as for the ^{103}Ru can be drawn for the ^{141}Ce isotope. It is a low volatile fission product, with an intermediate half-life (32 days) and which is not expected to move in significant amounts out of the pellet. Its presence also argues to an origin in a primary resin after a cycle with a fuel cladding failure with dispersion in the primary loop.

The couple ^{95}Zr (parent) and ^{95}Nb (decay product), can have two main origins:

- corrosion of activated ^{95}Zr coming from the pin cladding under irradiation (neutron capture on ^{94}Zr);
- fission of dispersed fuel in the primary water. Indeed, ^{95}Zr is also a low-volatile fission product, which is not expected to move out of the pellet, except in case of fission directly in the water.

In both cases, the ^{95}Zr will accumulate in the primary resin. The measured $^{95}\text{Zr}/^{95}\text{Nb}$ ratio is close to 1, consistent with the secular equilibrium expected for this couple after several months of decay. Both production hypotheses can explain the measurements and are consistent with earlier conclusions from other radionuclides.

3.6 Lack of other fission products

As explain above the most probable source seems to be of a primary resin, after a few months of decay, of a pressurized water reactor with fuel cladding failure and dispersion of fissile material in the primary. In order to check this hypothesis, the radiological inventory of fission products that could be obtained after direct fissions in the main coolant, and six months of decay, has been evaluated. The list of isotopes that are responsible of at least 0.1% of the total activity, is the same that which have been detected (^{134}Cs , ^{137}Cs , ^{103}Ru , ^{106}Ru , ^{141}Ce , ^{95}Nb , ^{95}Zr), except:

- ^{89}Sr , ^{90}Sr , ^{90}Y , ^{91}Y , which are mainly β emitters and thus couldn't have been detected by usual gamma spectrometry techniques because of their tiny gamma decay branching ratio;
- ^{144}Ce , which is an isotope similar to ^{106}Ru , with relatively long half-life (285 days), and comparable fission yield;
- ^{147}Pm , which also has the same range of fission yield and relatively long half-life (2.6 years).

Hence, considering a primary resin inventory with fuel dispersion in the primary loop, ^{144}Ce and ^{147}Pm could potentially have been detected. Nevertheless, one can remark that the ^{106}Ru has been detected in only one station. A lower detection limit, or different atmospheric behaviour of cerium or promethium compare to ruthenium, could easily explain why they were not detected.

As a result, the detected isotopes are coherent with the hypothesis, and no additional isotope should have mandatory been detected.

3.7 Conclusion

The analysis of the origin of each detected isotope, taking into account the cumulated presence of activated corrosion products and fission products indicates that the release

Table 3. Various kind of event at Finland border since 2012.

Release	Composition FP: Fission Products CP&AP: Corrosion Products and Activation Products	Possible origin	Decay before release
May 2015 April 2018 June 2020	CP&AP and low volatile FP Presence of intermediate half-life	Primary resin with fuel cladding failure with fuel dispersion	Few months
May 2017	CP&AP and volatile FP Presence of intermediate half-life	Primary resin with fuel cladding failure without fuel dispersion	Few months
August 2014 April 2019 July 2019 March 2020	CP&AP Presence of intermediate half-life	Primary resin without fuel cladding failure Primary cleaning system filter	Few months
October 2015	CP&AP and volatile FP No intermediate half-life	Primary resin with fuel cladding failure	Few years

- it shouldn't be so frequent (almost one uncontrolled fire in an auxiliary building per year);
- the release duration shouldn't be so long as observed in June 2020 release (2–3 weeks).

One could also imagine that the release came from the deliberate incineration of solid waste, being contaminated with a few amount of a primary resin (for example, after an intervention concerning a resin, as presented in §5.3). This hypothesis could match the release characteristics, except the activity levels (Tab. A1). Indeed, in such case of deliberate incineration, the smokes are expected to be filtered, which strongly decreases the activity released at non-detectable levels.

Some additional considerations about these hypotheses are presented in Table A2, of Appendix A.

In conclusion, despite a large set of hypotheses, none of the fires considered matches with the release characteristics. Assuming an uncontrolled fire, several additional hypotheses are required. As a result, this assumption is unlikely.

5.2 Resin thermal decomposition?

Another unexpected event that could make the inventory of a primary resin volatile, could be its thermal decomposition. Indeed, above 150 °C the resin decomposes, and produces some aerosols or organic gases. In case of human error leading to a primary resin injection in the primary loop, as it occurs, for example, in Olkiluoto Boiling Water Reactor plant in December 2020, the resin will be heated up and decomposed. The radiological inventory will end in the gaseous effluent treatment system. Then, after some month of decay, it will be slowly released into the atmosphere.

This hypothesis enables to obtain a “volatile” inventory of a primary resin, with a few months of decay, and possibly a consistent release duration. But it suffers some major limitations:

- the release would be filtered, and the activity would not be sufficient to be detected;
- aerosols will stay in the gaseous effluent treatment system, and it is very unlikely to keep any volatile organic compound of isotopes such as cobalt, ruthenium and caesium.

As a result, this hypothesis doesn't provide satisfying explanation of the release measured in June 2020. Some additional considerations about these hypotheses are presented in Table A3, of Appendix A.

5.3 Issue during the resin transfer to the waste management building?

The last investigated hypothesis concerns an issue during the transfer of the resin to the waste management building. Indeed, this transfer is generally performed by the hydraulic transfer (use of pressurized water to pull the resin) through some pipes outside of the plant buildings. In case of a break of such a pipe during a transfer, or an intervention on that pipe after a resin blocking, an atmospheric release could occur.

But again, this assumption suffers some limitations. First, the resin transfer is supposed to occur during the reactor shutdown. So, the decay time should be of some weeks and not months, which is incoherent with the lack of short-life isotopes and the ruthenium ratio. Second, mainly depending on the operations and radiological precautions taken in case of such event, the released activity might be too low to be detected. As a result, this explanation seems to be unlikely.

Some additional considerations about these hypotheses are presented in Table A4, of Appendix A.

6. Conclusion

In conclusion, the detection of artificial radionuclides (^{60}Co , ^{134}Cs , ^{137}Cs , ^{103}Ru , ^{106}Ru , ^{141}Ce , ^{95}Nb , ^{95}Zr) in various countries of Northern Europe (Finland, Sweden, Estonia), reveals that a release occurred in 2020 from June 13 to June 22. The measured values were close to detection limits and didn't represent any health issue. Using inverse modelling techniques, the most probable release location has been evaluated in a zone from eastern Estonia to western Russian Federation. An estimation of the release magnitude, at the most probable location, estimates the activity at a few GBq of ^{60}Co and tens of GBq of caesium isotopes.

The analysis of the isotopic composition of the release establishes that its most probable source is a primary ion exchange resin of a pressurized water reactor with fuel cladding failure and dispersion of fissile material in the main coolant, after a few months to 9 months of decay, even if no certainty is achievable with so little information.

This event is finally quite common since at least three similar events were observed at Finland border in the 2012–2020 period, and potentially up to nine.

Three main phenomena have been investigated in order to imagine what event could explain this release: a resin fire, a thermal decomposition of such resin in a primary loop, or an issue during the resin transfer to the waste management building. Nevertheless, mainly due to the lack of information, no answer has been found. None of the hypothesis considered matches with all the characteristics of the release.

Conflict of interests

The authors declare that they have no competing interests to report.

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Data availability statement

Data associated with this article cannot be disclosed due to legal reason.

Author contribution statement

All the authors were involved in the preparation of the manuscript. All the authors have read and approved the final manuscript. J.J. Ingremeau wrote and conducted the analysis of the Sections 3–5. O. Saunier wrote and conducted the analysis of the Section 2.

Appendix A Analysis of the various scenarios

Table A1. Legend.

Table 4. Legend

	Likely hypothesis	Expected result for this hypothesis
	Possible	No incompatibility with the hypothesis
	Unlikely hypothesis	Unexpected result for this hypothesis
	Excluded hypothesis	This result exclude this hypothesis

Table A2. Analysis of resin fire to explain the June 2020 release.

<i>Resin fire</i>			Release duration (2-3 weeks)	Presence of ¹⁰³ Ru and ¹⁴¹ Ce	Activity Levels	Likelihood of initiating event / operation	Recurrent nature of the release
<i>Waste treatment installation of Leningradskaya</i>	Deliberate burning of large amount of resin	With smokes filtration					
		Without smokes filtration					
		Without smokes filtration and with human error leading to short decay resin burning					
	Deliberate burning of solid waste contaminated with a resin	With smokes filtration					
	Uncontrolled fire during treatment	After long decay					
		With human error leading to short decay resin treatment					
	Uncontrolled fire during resin storage of drying operation						
<i>Research center of Sosnoviy Bor</i>	Uncontrolled fire during a research experiment						

Table A3. Analysis of resin thermal decomposition to explain the June 2020 release.

	Release duration (2-3 weeks)	Isotopes			Activity Levels	Likelihood of initiating event / operation	Recurrent nature of the release
		Lack of short-lived isotopes	^{60}Co	^{103}Ru , ^{134}Cs ,			
<i>Resin thermal decomposition in the primary loop</i>	✓	✓	?	??	?	✓	?

Table A4. Analysis of an issue during the resin transfer to explain the June 2020 release.

	Release duration (2-3 weeks)	Isotopes		Activity Levels	Likelihood of initiating event / operation	Recurrent nature of the release
		Lack of short-lived isotopes	^{103}Ru , ^{134}Cs , ^{60}Co			
<i>Issue during the resin transfer to the waste management building</i>	?	?	✓	?	✓	✓

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