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Detection of radionuclides in northern Europe in June 2020: investigations on possible source term location and origin

JJ. Ingremeau, O. Saunier,

Institut de Radioprotection et de Sûreté Nucléaire (IRSN), Fontenay-aux-Roses, France

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 performed at IRSN in order to identify the release location and its origin. Using inverse modelling techniques, the most probable source location has been determined together with an estimation of the release magnitude. Additional investigations have been performed in order to determine from which type and part of a nuclear installation the release could come from. Although no certainty is achievable, the most probable source is a spent primary ion exchange resin. This detection has also been compared to previous similar ones. Finally, some hypothesis to understand how this radiological inventory has been released into the atmosphere are presented. 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 indeed detected. The Swedish and Finnish authorities reported that cobalt 60, cesium 134, cesium 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 concentrations levels were very small in the order of several $\mu\text{Bq}/\text{m}^3$ and do not pose any health or environmental effects. Maximum air concentrations measurements (Figure 1) were reported in Finland at Helsinki, from June 16 to 17, 2020, of the order of ten microbecquerels/ m^3 : Cs-137 = 16,4 $\mu\text{Bq}/\text{m}^3$; Cs-134 = 21,5 $\mu\text{Bq}/\text{m}^3$; Co-60 : 7,6 $\mu\text{Bq}/\text{m}^3$; Ru-103 : 4,8 $\mu\text{Bq}/\text{m}^3$. The limited number of measurements above detection limits makes it very difficult to pinpoint the geographical origin of the detections.

In Section 2, atmospheric transport modeling combined with measurements was therefore applied to both narrow down the possible locations of the origin of the release and to estimate its magnitude. In Section 3, the characteristics and the estimated magnitude of each isotope are analyzed 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.

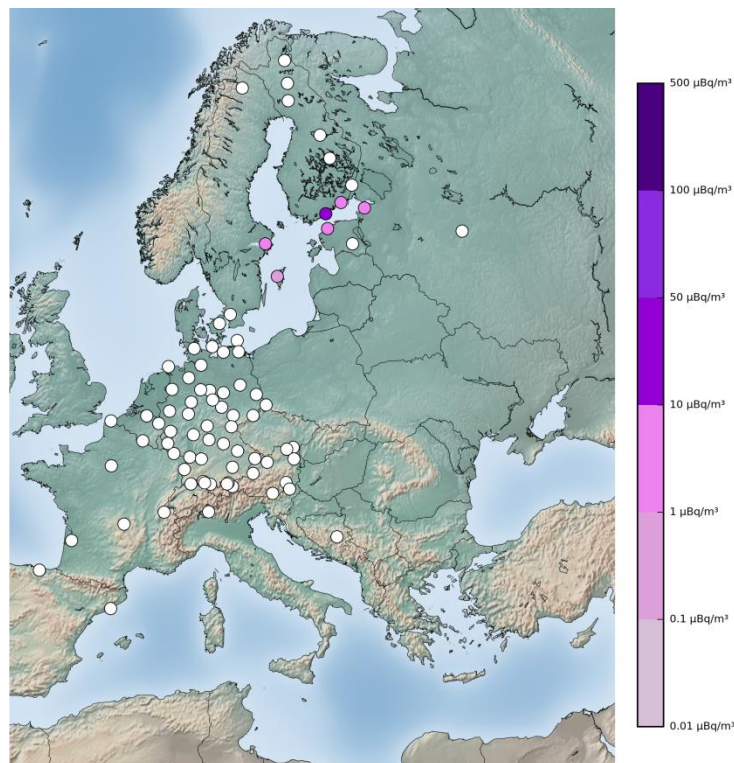


Figure 1: Maximum ^{134}Cs air concentration for each monitoring station ($\mu\text{Bq}/\text{m}^3$). With dots are air concentration measurements below the detection limit.

2 INVESTIGATIONS ABOUT THE RELEASE LOCATION AND MAGNITUDE

2.1 Source location

Inverse modelling techniques which combine atmospheric transport model and observations in the environment are applied to reconstruct the source location and its magnitude. ^{134}Cs air concentrations measurements are the most numerous and are therefore exploited for source identification. The approach used is described in details in [2]. 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 is assessed by inverse modelling using a variational approach assuming that the measurement vector can be described as a linear model with a source-receptor matrix and unknown source term vector σ :

$$\mu = \mathbf{H}\sigma + \epsilon \quad (1)$$

The \mathbf{H} source-receptor matrix holds modelled ^{134}Cs air concentrations using the IdX atmospheric transport model and in response to a unit source. IdX is used to simulate the ^{134}Cs dispersion over the domain of dimensions [4W, 44E], [40N, 71N], it is part of IRSN's C3X operational platform [3]. It has been applied to deal with massive accidental releases into the environment [4], [5] and minor radionuclide detection events [6]. IdX is forced with hourly meteorological data from ARPEGE model provided by Météo-France. Spatial resolution of the data is $0.1^\circ \times 0.1^\circ$.

The vector ϵ represents a combined model-representation-instrumental error. Assuming that the errors ϵ follow a log-normal distribution, the source term can be assessed by minimizing

the cost function J_{LN} which measures the differences between the model predictions $H\sigma$ and the real measurements μ :

$$J_{LN}(\sigma) = \frac{1}{2}(\ln(\mu) - \ln(H\sigma))^T R^{-1}(\ln(\mu) - \ln(H\sigma)) \quad (2)$$

where $R = E[\epsilon\epsilon^T]$ is the error covariance matrix associated with the error. It is well known that the source-receptor relationship (1) constitutes an ill-posed inverse problem and its resolution may fail particularly when the number of observations is limited. That is why a background term is usually added in the cost function (2) to stabilize the inverse problem and to ensure the unicity of the solution. This term measures the differences between a priori (or background) source term σ_b and the updated source term σ :

$$\frac{1}{2}(\ln(\mu) - \ln(H\sigma))^T R^{-1}(\ln(\mu) - \ln(H\sigma)) + (\sigma - \sigma_b)^T B^{-1}(\sigma - \sigma_b)$$

In order to mitigate the influence of very small concentration values, a threshold θ ($\theta = 1 \mu\text{Bq}/\text{m}^3$) is introduced in the cost function:

$$J_{LN}(\sigma) = \frac{1}{2}(\ln(\mu + \theta) - \ln(H\sigma + \theta))^T R^{-1}(\ln(\mu + \theta) - \ln(H\sigma + \theta))$$

In this study, it is assumed that R and B are diagonal and the error variance is the same for all diagonal elements of the matrix. The minimization of $J_{LN}(\sigma)$ is therefore equivalent to minimizing:

$$J_{LN}(\sigma) = \frac{1}{2}(\ln(\mu + \theta) - \ln(H\sigma + \theta))^T (\ln(\mu + \theta) - \ln(H\sigma + \theta)) + \lambda^2(\sigma - \sigma_b)^2$$

The λ parameter acts on the magnitude of the source and has to be determined outside the minimization procedure. Usually, when the number of observations used to reconstruct a release event is low, the source term magnitude may be very sensitive to the value of λ parameter. In that case, strong values of λ lead to source term tempered too much whereas small values of λ yield to unrealistic source term in which the norm may tend to infinity. In practice, the choice of the λ parameter therefore involves a compromise between reducing the error value related to the cost function and increasing the amount of radionuclides emitted. In this study, the optimal choice of λ is determined using L-curve method [7]. It consists in plotting the source term norm versus the norm of the first term of $J(\sigma)$ (residues) and to identify the maximum curve point in the graph. For every source term assessment associated with each grid point, an optimal value of λ is determined applying the L-curve technique. The cost function is directly minimized using the L-BFGS-B limited-memory quasi-Newton minimizer by enforcing the positivity of the source. The source reconstruction period is between June 5 and June 28 with daily time intervals. Only ^{134}Cs air concentration measurements have been exploited since they are most numerous. A total of 250 ^{134}Cs air concentration measurements have been taken into account to assess the origin and the magnitude of the release, of which 10 are non-zero measurements and the remaining are non-detections, i.e. measurements below the detection limit.

Then, for each inverted source related to each grid point, the agreement between modelled and observed air concentration measurements is assessed using 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.

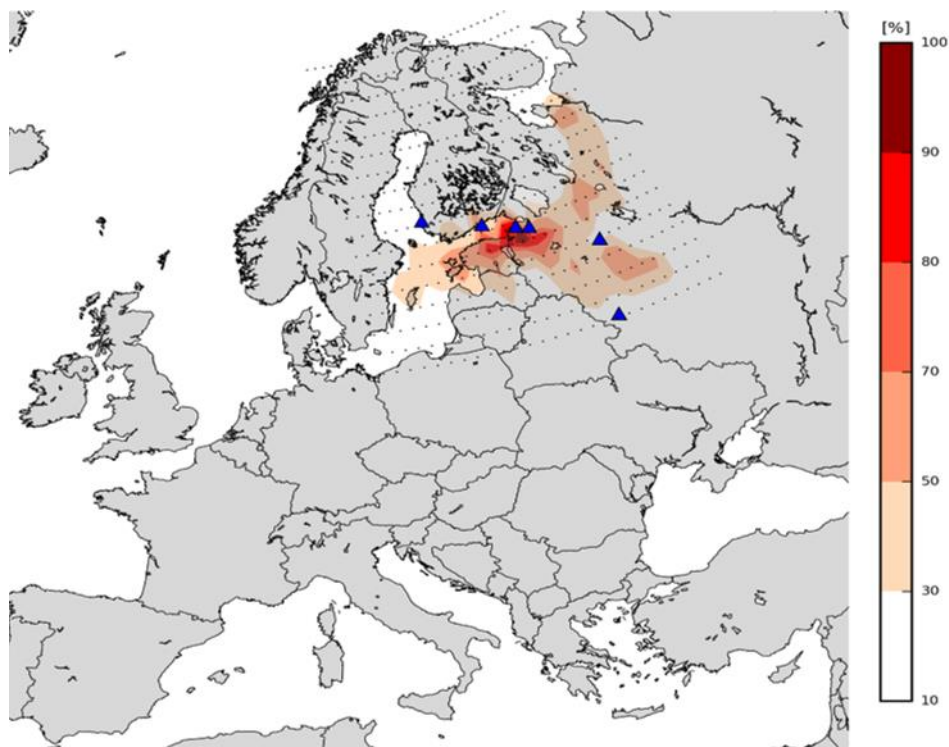


Figure 2: Percent of the simulated air concentrations that is within a factor of 5 of the observed values. Blue triangles are nuclear plants located in the area of interest: Leningrad NPP, Petersburg Nuclear physics Institute (Gatchina), Olkiluoto NPP, Loviisa NPP and Smolensk NPP

The interpolated values of FAC5 from each grid point are shown in Figure 2. Despite the small number of measurements used to reconstruct each source term, the area where the FAC5 values exceeds 80% is small. This region extends from eastern Estonia to western Russian Federation whereas 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.

2.2 ^{134}Cs Release magnitude

In the most plausible release location, we identified two facilities that may be responsible for the emissions namely the Leningrad Nuclear Power Plant and the Gatchina Institute of Nuclear Physics. According to our calculations, due to the proximity of the two nuclear sites, it is not possible to favour one facility over the other.

Nevertheless, considering the source term characteristic the Gatchina location can be excluded. Indeed the Gatchina site had only one nuclear reactor in operation in June 2020, which had only worked at low power at that time. It's impossible that such low power operation can generate such amount of ^{60}Co . Moreover, it is very unlikely that such reactor lead to the measured cesium ratio (§3.3). Gatchina has also some cyclotron, that cannot produce this type of source term.

We therefore estimated the source term from the Leningrad Nuclear Power Plant. As the minimization of the cost function \mathbf{J}_{LN} only provides an optimal solution without quantifying the model-representation-instrumental error, we use a Monte-Carlo sensitivity analysis [2] to partially quantify that error. It consists of generating a set of source terms where, for each sample n , the observation vector μ is perturbed such as $\tilde{\mu}_i = r\mu_i$, $1 \leq i \leq d$, where d is the measurement number and r follows a log-normal law $r \sim \exp(N(0, \zeta^2))$ where ζ is the standard

deviation of the related Gaussian law $N(0, \zeta)$. Then, for each sample n , an optimal source term $\hat{\sigma}_n$ is obtained by minimizing the cost function $J_{LN}(\hat{\sigma}_n) \cdot J(\hat{\sigma}_n)$. The Monte-Carlo analysis was performed using $n=20,000$ samples with a realistic value of the standard deviation $\zeta=0.8$. This analysis indicates that the estimated ^{134}Cs source term for the entire period lies between 2 and 20 GBq. This is a relatively wide range and reflects the uncertainties associated with the source term assessment. Figure 3 shows the temporal evolution of the releases between June 4 and June 28. It can be seen that the first releases started on June 13 and continue until June 22. During this period, the magnitude of the peaks varies significantly and the standard deviation associated with each peak is very large. This emphasizes the uncertainty in our estimates, which may be partially attributed to the use of a limited number of observations. After June 23, the calculations indicate that the occurrence of a release is not consistent with the observations. Indeed, if the release had continued after June 22, the stations would have reported an increase of ^{134}Cs air concentrations. It is therefore likely that the release ended on June 23 or else, it was too small to be detected.

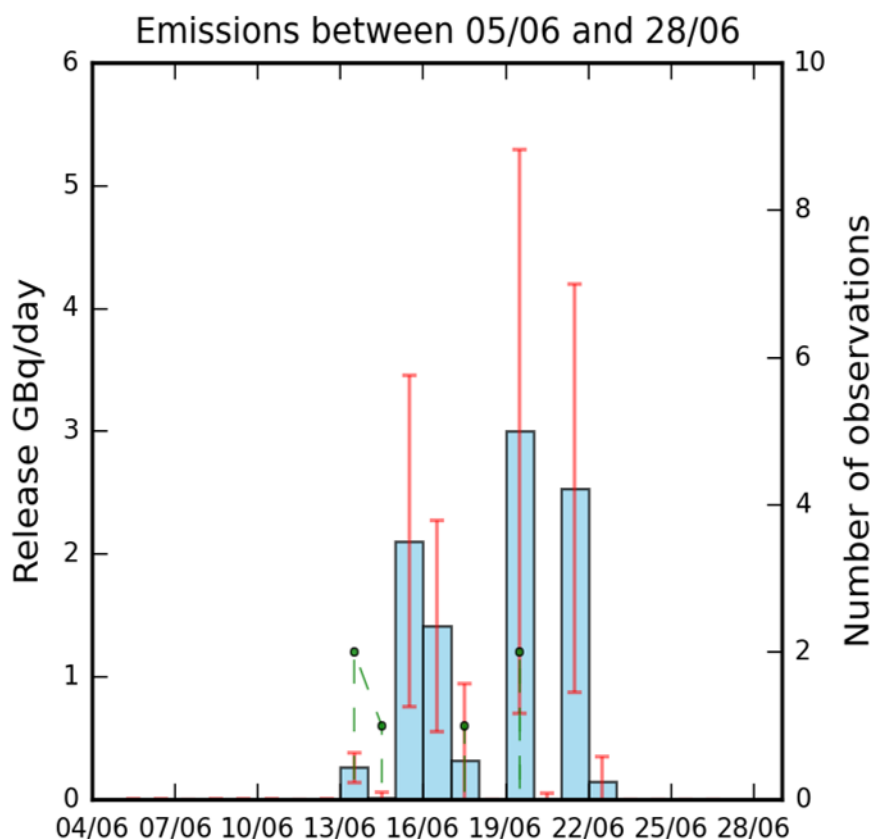


Figure 3: Mean and standard deviation range of the reconstructed source term (blue) using Monte-carlo sample. Green dots are the number of observations used in the inversion procedure.

2.3 Plume dispersion analysis

The ^{134}Cs plume dispersion was performed using the reconstructed source term from Leningrad NPP. The Figure 4 shows the dispersion of the plume at different dates between June 16 and June 25. During the period of the release, it can be seen that the plume mainly affected the northern European countries located between Scandinavia and western Russian Federation. Driven by east to north-east winds, the plume temporarily reached the regions between central Europe and Germany, particularly between June 16 and June 18 then between June 24 and June 26. However, the modelled air concentration levels are extremely low, usually undetectable by the monitoring networks. Nevertheless, the station of Braunschweig in Germany was able to detect tiny traces of ^{60}Co between June 15 and 22, which tends to confirm the results obtained by modelling.

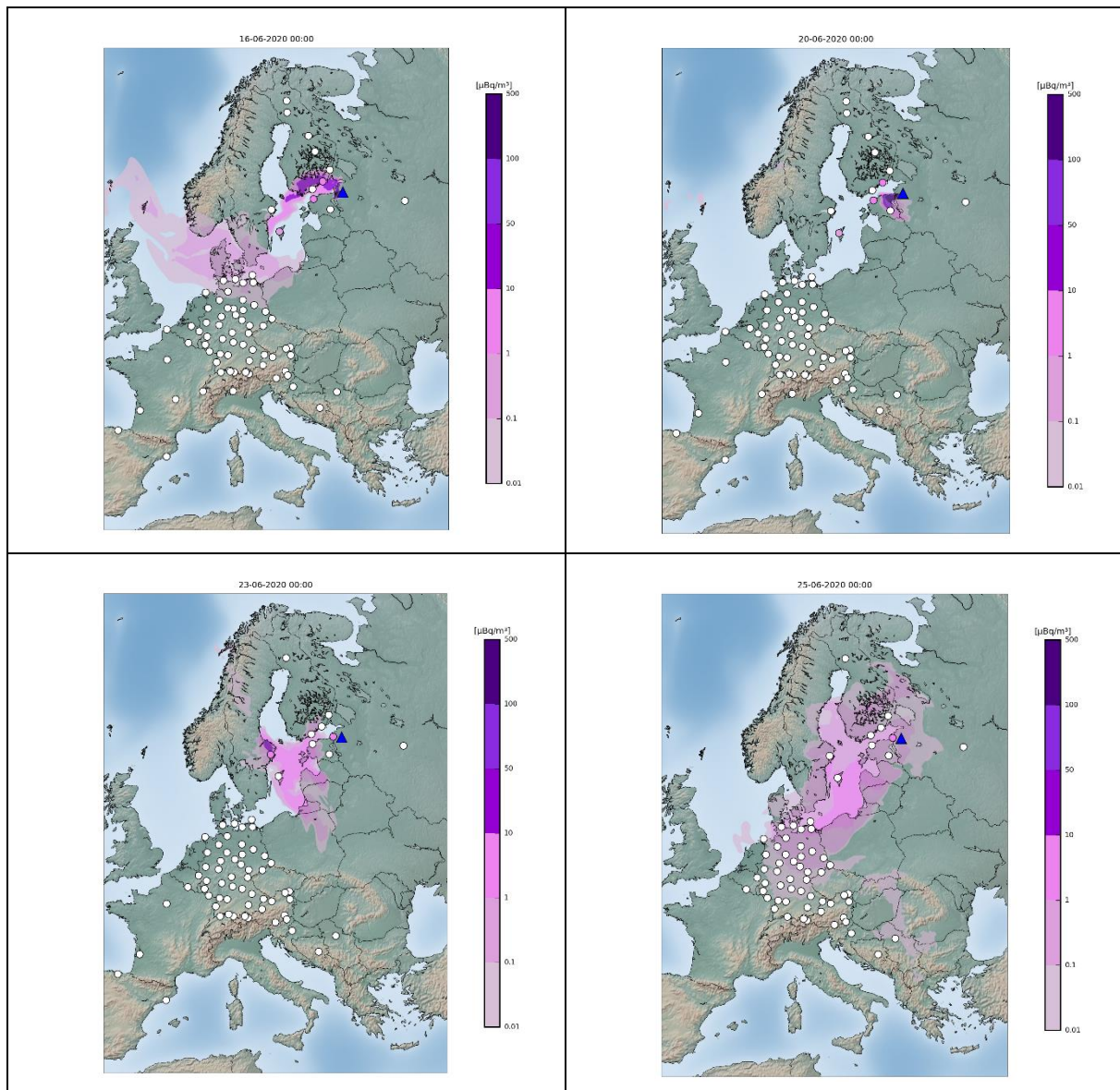


Figure 4: Modelled ^{134}Cs air concentration at different times (UTC) assuming a release location at Leningrad NPP. Blue triangle is Leningrad nuclear power plant. Circles are ^{134}Cs observed air concentrations.

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

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 the Table 1.

<i>Isotope</i>	<i>Half-life</i>	<i>Estimated Source Term</i>	<i>Origin</i>
^{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	

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

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, cesium 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 product

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 .

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 massively 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 source, 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 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, is the best candidate to explain the presence of such amount of ^{60}Co .

3.3 ^{134}Cs & ^{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 products¹. 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 few years in the spent fuel pool before being evacuated.

In case of fuel cladding leakage or failure in an operating reactor, the cesium isotopes would easily migrate to the primary water. But again, in case of a primary water leakage, the amount of cesium 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. Typical cesium 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}Cs / ^{137}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 [8].

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

3.4 ^{103}Ru & ^{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 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°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.

¹ ^{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.

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. 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 [9]. 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 cesium 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 to 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 or WWER, 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 than 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 amount 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 (the father) and ^{95}Nb (the son), 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. The two types of production can explain the measurements, but whatever, these isotopes lead to similar conclusions than previous ones.

3.6 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 measured in June 2020 might be issued from a primary resin after a few months of decay.

No certainty is achievable with so little information, but the most probable hypothesis seems to be of a primary resin, after a few months to 9 months of decay, of a pressurized water reactor with fuel cladding failure and dispersion of fissile material in the primary.

4 COMPARISON WITH PREVIOUS RELEASES

The June 2020 event is not the only detection of such isotopes in the past decades. At least 9 similar detections have been recorded since 2012 at Finland border. They are provided in Table 2. It must be noticed that, due to Chernobyl contamination, a few $\mu\text{Bq}/\text{m}^3$ of ^{137}Cs is always detected. So, in the table, the detections compatible with the Chernobyl background are marked with a “x” and the larger detections, clearly suggesting a fission products release, are identified with a “X”. Besides, in May 2017 and March 2020, no ^{95}Zr have been detected, despite its son, ^{95}Nb , have been recorded. It might be interpreted as the fact that ^{95}Zr were present, but below detection limit.

Release date	Activated corrosion product					Fission product					Release localization
						Low volatile		Volatile		Low volatile	
	^{60}Co	^{58}Co	^{59}Fe	^{54}Mn	^{95}Nb	^{95}Zr	^{134}Cs	^{137}Cs	^{103}Ru	^{141}Ce	
	Half-life	5,3 y	71 d	45 d	0,86 y	35 d	64 d	2 y	30 y	39 d	
August 2014	X	X	X	X	X	X		x			Not evaluated
May 2015	X				X	X	X	x	X	X	Compatible with reference
October 2015	X			X			X	X			From the North
May 2017	X		X	X	X	?	X	x			Not evaluated
April 2018	X	X	X	X	X	X	X	x	X	X	Not evaluated
April 2019	X	X	X	X	X	X		x			Not evaluated
July 2019	X		X	X				x			Not evaluated
March 2020	X		X	X	X	?		x			Not evaluated
June 2020	X				X	X	X	X	X	X	Reference

Table 2: All comparable detections at Finland border since 2012

All these detections can be gathered in four categories, described in Table 3.

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

Table 3: Various kind of detection at Finland border since 2012

First, three release measurements have the same characteristics than the June 2020 ones. Five additional detections might also be interpreted as primary resin with various fuel cladding failure states (significant failure in May 2017 but without fuel dispersion, limited in the other cases), with a few months of decay. And one last detection that occurred in October 2015, might also be attributed to a primary resin with fuel cladding failure but with years of decay.

As a conclusion, all these detections are consistent with from 3 to 9 atmospheric releases of primary resin inventory at Finland border, since 2012, such event occurring subsequently quite often.

5 WHAT HAPPENED?

One major issue remains unknown in that state: how the inventory of a primary resin could have been released into the atmosphere?

Indeed, after generally few years of operation, the spent primary resins are usually moved to the waste management plant for dry storage during some years of decay. Finally, they are conditioned for long-term disposal. And no atmospheric release of their inventory is expected in all these operations.

5.1 Resin Fire?

A first hypothesis could be an uncontrolled resin fire. Indeed, resins are burnable and such event could release a significant part of their inventory in the atmosphere. But:

- Assuming it happens during the long-term conditioning, it should occur after some years of decay, and so, no intermediate half-life should be expected;
- Assuming it happens during the dry storage:
 - 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. 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 4.

	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

<i>Resin fire</i>		Release duration (2-3 weeks)	Présence 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 4: Hypothesis matrix of resin fire to explain the June 2020 release

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 reject 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 cesium.

	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 5: Analysis of resin thermal decomposition in the primary to explain the June 2020 release

As a result, this hypothesis doesn't provide satisfying explanation of the release measured in June 2020.

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. Moreover, 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.

	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>	?	?		?			

Table 6: Analysis of an issue during the resin transfer to explain the June 2020 release

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 cesium isotopes.

The analyze 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 primary, 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 hypothesis considered matches with all the characteristics of the release.

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