



HAL
open science

Methodology for the investigation of undeclared atmospheric releases of radionuclides: Application to recent radionuclide detections in Northern Europe from 2019 to 2022

Olivier Saunier, J J Ingremeau, Ian Hoffman, Pawel Mekarski, Jing Yi, Adrian Botti

► To cite this version:

Olivier Saunier, J J Ingremeau, Ian Hoffman, Pawel Mekarski, Jing Yi, et al.. Methodology for the investigation of undeclared atmospheric releases of radionuclides: Application to recent radionuclide detections in Northern Europe from 2019 to 2022. *Annals of Nuclear Energy*, 2023, 192, pp.109907. 10.1016/j.anucene.2023.109907 . irsn-04119406

HAL Id: irsn-04119406

<https://irsn.hal.science/irsn-04119406>

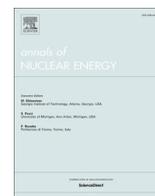
Submitted on 6 Jun 2023

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial - NoDerivatives 4.0 International License



Methodology for the investigation of undeclared atmospheric releases of radionuclides: Application to recent radionuclide detections in Northern Europe from 2019 to 2022

O. Saunier^{a,*}, J.J. Ingremeau^{a,*}, Ian Hoffman^b, Pawel Mekarski^b, Jing Yi^b, Adrian Botti^b

^a Institut de Radioprotection et de Sûreté Nucléaire (IRSN), 31 Avenue de la Division Leclerc, 92260, Fontenay-aux-Roses, France

^b Health Canada, Ottawa, Ontario, Canada

ARTICLE INFO

Keywords:

Artificial radionuclide detection
Atmospheric transport modelling
Source identification
Source term analysis
Inverse modelling

ABSTRACT

Traces of radionuclides have been frequently detected in the European atmosphere for several years. The measured concentrations are usually very low, ranging from 0.1 to 10 $\mu\text{Bq m}^{-3}$, and do not pose any health or environmental problems.

This study aims to diagnose the origin of small undeclared radionuclide releases into the atmosphere. An inverse modelling approach that combines environmental measurements and atmospheric transport modelling is first used to assess the source location of the release. In addition, the type and process of the nuclear facility from which the release could originate are investigated by identifying the isotope production pathways and comparing them with known typical inventories. These two parts of the proposed method are complementary and allow us to extract as much information as possible from a set of radionuclide measurement data.

In a previous study, the origins of detections of various radionuclides (^{60}Co , ^{134}Cs , ^{137}Cs , ^{103}Ru , ^{106}Ru , ^{141}Ce , ^{95}Nb , ^{95}Zr) in Finland, Sweden and Estonia in June 2020 have been investigated.

In this paper, the previous investigation is extended by analysing two additional events that occurred in northern Europe in July 2019 and May 2022, as well an overview of other unknown releases detected in Finland over the last decade. A more detailed analysis of the 2020 event is also provided by analysing new available environmental measurements.

The calculations indicate that the source location of the three events appears to be in the same region, in Russian Federation. The most probable origin of the June 2020 release seems to be a primary ion exchange resin, after 2 to 5 months of decay, of a pressurized water reactor with fuel cladding failure, and dispersion of fissile material in the primary.

The July 2019 and May 2022 events are of particularly noteworthy due to the simultaneous presence of ^{46}Sc , which is neither produced nor in the fuel, nor in the primary loop of PWR or RBMK nuclear power plants, and typical corrosion-activated products from power plants (^{60}Co). Two hypotheses are proposed to explain this source term: a mixture of various solid wastes or recently irradiated graphite from a RBMK reactor.

The reliability of the methodology is demonstrated, in particular in the section dedicated to atmospheric transport modelling, and the successful association with source term analysis provides a valuable tool for future studies and assessments of both minor and major radionuclide releases.

1. Introduction

In recent years, European monitoring networks responsible for measuring radioactivity in the environment have repeatedly detected traces of radionuclides in the atmosphere. For some of these events, the geographical origin of the detections remained unknown. Radiological

emergency preparedness requires vigilance for low-level signals from environmental monitoring systems, even if the event has no impact on health and the environment. Diagnosis of undeclared atmospheric releases offers an opportunity to assess and improve emergency preparedness and response strategies. IRSN is therefore in the objective to predict the possible impact over the French territory using atmospheric

* Corresponding authors.

E-mail addresses: olivier.saunier@irsn.fr (O. Saunier), Jean-jacques.ingremeau@irsn.fr (J.J. Ingremeau).

<https://doi.org/10.1016/j.anucene.2023.109907>

Received 29 January 2023; Received in revised form 21 April 2023; Accepted 2 May 2023

0306-4549/© 2023 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

transport models. In the case of an event of unknown origin, it is essential to first identify the source location before simulating the dispersion of radionuclides in the atmosphere (Dumont Le Brazidec et al., 2020); (Lucas et al., 2017); (Tichý et al., 2017); (Yee et al., 2014). In recent years, the most notable event was the widespread detection of Ruthenium-106 across Europe during the fall of 2017 (Masson et al., 2019). This release, which has not been officially acknowledged was significant enough for the majority of European countries to detect the presence of ^{106}Ru on their territory. Efforts to pinpoint the source location relied primarily on inverse modelling techniques which combine atmospheric transport modelling with environmental measurements. The results pointed to the Southern Urals in the Russian Federation (Kovalets et al., 2020); (Saunier et al., 2019); (Dumont Le Brazidec et al., 2020); (Cooke et al., 2020) as the likely origin of the detections.

Between 2019 and 2022, more recent detection events with smaller geographical extent were reported in Europe (De Meutter and Hoffman, 2020); (Masson et al., 2021); (De Meutter et al., 2021). As in the case of ruthenium-106, the source location was sometimes not known at the time the detections were reported. In June 2020, various radionuclides were measured by European monitoring networks, most of which are connected through a network called “Ring of Five” (Ro5). The Ro5 network, established in 1983, is an informal information group for the purpose of rapidly exchanging data on occasional enhanced concentrations of radionuclides at trace levels in the atmosphere. In addition to the “measuring laboratories”, the network includes other partners such as national meteorological services or staff members of universities or of the International Atomic Energy Agency (IAEA). Radionuclides detected in June 2020 included nuclear activated corrosion products and fission products characteristic of irradiated nuclear fuel in a power plant. Seeking to help identifying their possible origin, the IAEA inquired with their counterparts in the European region and requested information on whether the radionuclides were detected in their countries, and if any event may have been associated with the possible atmospheric release (IAEA 2020a); (IAEA 2020b). A database of the available measurements was thus constructed and was useful for the understanding of the event using atmospheric transport modelling. Two quite similar events occurred in July 2019 and May 2022 with the detections of ^{60}Co , ^{59}Fe , and ^{46}Sc , at very low levels in Finland, similar in order of magnitude to the June 2020 event. In contrast to the June 2020 detection event, which involved several countries, these two events were restricted to southern Finland. Indeed, several air sampling stations in the Finnish network are equipped with high-volume samplers that can detect extremely low levels of radioactivity. The filters of the samplers are changed once a week and the data are published weekly by the Finnish authorities (STUK) on their website.

In a previous publication (Ingremeau and Saunier, 2022), the focus was on the 2020 event, where an analysis of the source term origin was proposed, and hypotheses were made to explain how the radiological inventory could have been released into the atmosphere. This paper extends the previous work to include an analysis of the July 2019 and May 2022 release events, with an update of the June 2020 analysis made possible using newly available airborne concentration data.

In Section 2, an overview of the detections is provided for each event. Section 3 describes the inverse modelling techniques used for source identification and their application to the events of July 2019, June 2020 and May 2022. The results obtained are then validated by comparison with environmental measurements. In Section 4, the characteristics and the estimated magnitude of each isotope are analysed in order to determine the type and part of a nuclear installation from which these releases could have originated. This section also includes a comparison with other previous releases which occurred in the same geographical area.

2. Radionuclide detection events and measurements overview

2.1. June 2020

In June 2020, several northern European countries reported a slight increase of airborne concentration radioactivity levels at stations belonging to their monitoring networks. The Swedish and Finnish authorities reported that ^{60}Co , ^{134}Cs , ^{137}Cs , ^{106}Ru , ^{141}Ce , ^{95}Nb , ^{95}Zr , and ^{103}Ru were detected from June 8 to June 22. The same radionuclides were also detected in Estonia the last week of June. The three countries declared that there had been no release events on their territory that could explain the presence of the radionuclides. Many other European countries had also voluntarily reported to the IAEA that there were no events on their territory that could explain the release.

The concentrations measured were very small in the order of several $\mu\text{Bq}/\text{m}^3$ and did not pose any health or environmental risks. Maximum ^{134}Cs air concentrations measurements (see Fig. 1) were reported in Finland at Helsinki, with around $15 \mu\text{Bq}/\text{m}^3$ between 16 and 17 June. IRSN published an information report on that event (IRSN Report, 2020), and a publication on the analysis of the release origin (Ingremeau and Saunier, 2022).

With the exception of Scandinavia and Estonia, the airborne concentration measured did not exceed the detection limits of the instruments which are typically in the order of $\mu\text{Bq}/\text{m}^3$ or less. However, detection limits can vary depending on factors such as the type of instrument, the duration of air sampling and the method of sampling. The very small number of measurements above detection limits makes it very difficult to pinpoint the geographical origin of the detections.

2.2. July 2019

In mid-July 2019, the Finnish authorities (STUK) reported the detection of particulate radioactive ^{60}Co , ^{46}Sc , ^{59}Fe and ^{54}Mn without knowing the source location. ^{60}Co and ^{46}Sc were detected at the Helsinki, Imatra and Kotka stations, while ^{59}Fe and ^{54}Mn were detected only

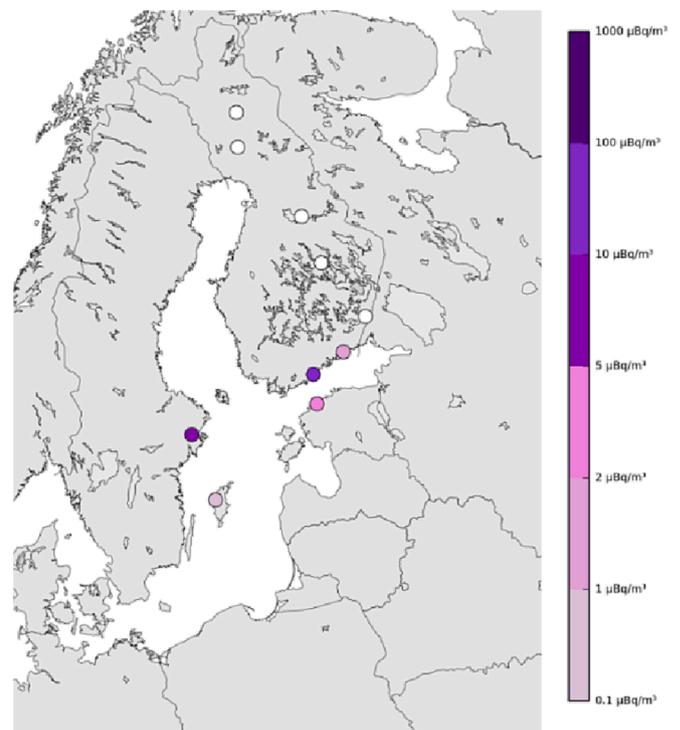


Fig. 1. Maximum ^{134}Cs air concentration measured by monitoring networks in Europe ($\mu\text{Bq}/\text{m}^3$) in June 2020. Blank dots are air concentration measurements below the detection limit.

at the Kotka station. The extremely low air concentration measured are listed in Table 1. Only measurement data with concentrations above the detection limits of the instruments are mentioned.

2.3. May 2022

In May 2022, a similar event was reported by the Finnish authorities. Traces of several radionuclides were detected at the Imatra and Kotka stations in southern Finland. The observed air concentration levels are given in Table 2.

In the remainder of the paper, the objective is to apply inverse modelling techniques to identify the most plausible geographical origin of the 2019, 2020 and 2022 detection events and the magnitude of the associated source term. In contrast to the 2017 ruthenium-106 event, the small number of measurements showing an increase in air concentrations makes the reconstruction of the source term by inverse modelling more challenging.

3. Investigations about the release location and magnitude

3.1. Methodology for source identification

The approach used in this study is described in detail in (Saunier et al., 2019) and is divided into three steps:

1. Definition of a domain containing a set of grid points with regular spacing that are considered potential source locations.

First, the spacing between two grid points is chosen to be large enough to reduce the computation time. With no information on the geographical origin of the detections, the probability that the source of the release comes from one particular grid point is the same for all grids points included in the domain.

2. Source term assessment by inverse modelling.

The source term related to each grid point is assessed by variational inverse modelling approach (Stohl et al., 2012); (Stohl et al., 2009); (Winiarek et al., 2011), which consists of the minimisation of the following cost function:

$$\mathbf{J}(\mathbf{x}) = \frac{1}{2} \sum_{i=1}^d (\ln(y_i + \theta) - \ln(\mathbf{H}\mathbf{x} + \theta))_i^2 + \lambda^2 \sum_{j=1}^N (x_j - x_{b_j})^2 \quad (1)$$

Where \mathbf{y} is the vector of measurements, \mathbf{x} is the unknown source term vector, x_b is the a priori of the source term, \mathbf{N} is the size of the source term vector and d is the number of measurements. As no information is available about the source, a zero a priori ($x_b = 0$) is chosen. \mathbf{H} is the source-receptor matrix computed using an atmospheric transport model

Table 1
Summary of radionuclide detections reported by STUK in July 2019.

Station location	Latitude / longitude	Air sampling period	Radionuclide	Air concentration ($\mu\text{Bq}/\text{m}^3$)
Kotka	60.48; 26.92	15/07-- 22/07	^{46}Sc	0.7
Kotka	60.48; 26.92	15/07-- 22/07	^{60}Co	2
Kotka	60.48; 26.92	15/07-- 22/07	^{59}Fe	1.1
Kotka	60.48; 26.92	15/07-- 22/07	^{54}Mn	0.4
Helsinki	60.17; 24.94	16/07-- 17/07	^{46}Sc	1.8
Imatra	61.19; 28.76	15/07-- 22/07	^{46}Sc	0.7
Imatra	61.19; 28.76	15/07-- 22/07	^{60}Co	1.9

Table 2
Summary of radionuclide detections reported by STUK in May 2022.

Station location	Latitude / longitude	Air sampling period	Radionuclide	Air concentration ($\mu\text{Bq}/\text{m}^3$)
Imatra	61.19; 28.76	19/05-- 26/05	^{46}Sc	17.3
Kotka	60.48; 26.92	23/05-- 30/05	^{60}Co	0.6
Kotka	60.48; 26.92	23/05-- 30/05	^{59}Fe	0.3
Kotka	60.48; 26.92	23/05-- 30/05	^{46}Sc	0.8

and describing the sensitivity of each measurement to a unitary release. In order to mitigate the influence of very small concentration values, a threshold θ ($\theta = 1 \mu\text{Bq}/\text{m}^3$) is added to the cost function.

As the inverse problem to be solved is usually ill-posed, a second term is added to the cost function to ensure uniqueness and the stability of the solution. The choice of the parameter λ plays a very important role by affecting on the magnitude of the source term especially when the number of observations is small. In this study, the optimal choice of λ is determined outside the minimisation procedure using the L-curve method (Hansen, 1992). It consists in plotting, for several values of λ , the norm of the source term against the norm of the first term of $\mathbf{J}(\mathbf{x})$ obtained after the minimisation. The point of maximum curvature on the graph is then taken as the optimal value of λ . Therefore, for each source term assessment associated with each grid point, an optimal value of λ is therefore determined by applying the L-curve technique. The minimisation of the $\mathbf{J}(\mathbf{x})$ is carried out using the L-BFGS-B algorithm (Liu and J. Nocedal J., 1989).

3. Use of statistical indicators to determine the relevance of each grid point as a source location

For each hypothetical source related to a grid point, the agreement between modelled and observed air concentration measurements is assessed using two indicators:

- the factor 5 indicator (FAC5): this 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 reduction factor of the cost function, which is the ratio between the initial value of the cost function and that obtained after minimisation. This is a more appropriate indicator for handling detection events with a very small number of measurements.

An iterative process of refining the spacing between grid points in the search domain is then carried out to more accurately determine the release location.

3.2. Application to June 2020 detection event

The domain likely to contain the source is divided into a set of 348 grid points, assuming that the release occurred somewhere between Germany and the Russian Federation. The dimensions of the domain containing potential sources are [14E, 42E], [55 N, 66 N] with a resolution of $1^\circ \times 1^\circ$. Among the air concentration measurements, the ^{134}Cs data are the most numerous and are therefore used for source reconstruction. A total of 250 ^{134}Cs air concentration measurements have been taken into account to assess the origin and the magnitude of the release, including 10 non-zero measurements and the remaining are measurements below the detection limit of the instruments. The \mathbf{H} matrix contains modelled ^{134}Cs air concentrations using the IdX atmospheric transport model which is part of the IRSN C3X operational platform (Tombette et al., 2014). The IdX model has been used to handle massive accidental releases to the environment (Saunier et al., 2013) as

well as smaller radionuclide detection events (Saunier et al., 2019); (Dumont Le Brazidec et al., 2020). The computational domain has dimensions of [4 W, 44E], [40 N, 71 N] and IdX is driven by hourly meteorological data from ARPEGE model provided by Météo-France with a spatial resolution of $0.1^\circ \times 0.1^\circ$. The source reconstruction period is from June 5 to June 28, with daily intervals. Due to the small number of non-zero measurements, the influence of λ can be significant, requiring a proper determination of this parameter. Initial values of λ range from 10^{-12} to 10^{-3} and by applying the L-curve technique, the optimal value is reached for $\lambda = 10^{-6}$.

The interpolated values of FAC5 from each grid point, obtained after applying the inverse procedure, are shown in Fig. 2. Despite the small number of measurements used to strongly constrain the source reconstruction, the geographical area where the FAC5 values exceed 90% is relatively small. The most reliable region extends from eastern Estonia to western Russia. Furthermore, only the area in the Leningrad region has FAC5 values close to 100%. Moving away from this area, the FAC5 values decrease rapidly indicating that the hypothesis of a release from Western Europe or Scandinavia is highly unlikely. To confirm the source location, a sub-domain restricted to the release area identified as most relevant, was considered. This sub-domain, with dimensions of [25E, 32E], [58 N, 60 N] is divided into a set of 322 grid points that are assumed to be source locations. The spatial resolution between two potential source locations is $0.2^\circ \times 0.2^\circ$. The inverse method was then applied to estimate the source term for each of the 396 potential source locations. The Fig. 2 shows the agreement between observed and simulated concentrations assessed for each potential source location in the domain of $0.2^\circ \times 0.2^\circ$. The results confirm the conclusions drawn for a resolution of $1^\circ \times 1^\circ$, as the Leningrad region well reproduce the measurements. In addition, regions in northern Estonia also appear as potential release locations.

Assuming the most plausible source location, the released activity of ^{134}Cs is in the order of a few gigabecquerels (GBq). On the basis of the isotopic ratios obtained from the measurements of radionuclides detected in the environment (^{106}Ru , ^{103}Ru , ^{60}Co , ^{137}Cs), it can be concluded that several tens of gigabecquerels were released over the whole period.

3.2.1. ^{134}Cs release magnitude

The Leningrad nuclear power plant and the Petersburg Nuclear Physics Institute (Gatchina) are located close to the area with the highest FAC5 values. Errors in the modelling (transport model and meteorological fields) may explain the fact that the maximum FAC5 values are not exactly located at one of the two facilities. In addition, the reconstruction of the source terms assuming daily increments may also lead to additional errors, especially in the case of releases with large variability on a single day. In contrast, no facilities could be identified in Estonia. For all these reasons, it is assumed that the most likely source is assumed to be the Leningrad region. According to our calculations, due to the proximity of the Leningrad NPP and the Petersburg Nuclear Physics Institute, it is not possible to favour one facility over the other. Nevertheless, considering the source term characteristics, the Gatchina site can be excluded. In fact, the Gatchina site had only one nuclear reactor in operation in June 2020, and it was operating at low power. It is impossible that such a low power operation can produce such amounts of ^{60}Co . Moreover, it is very unlikely that this reactor would have led to the measured caesium ratio (Section 4). Gatchina also has some cyclotrons, that cannot produce source terms of this magnitude.

Monte-Carlo sensitivity analysis (Saunier et al., 2019) coupled with inverse modelling is therefore applied to the reconstructed source term from the Leningrad NPP. It allows partial quantification of modelling and measurement errors. The total quantities assessed range from 2 to 20 GBq. The relatively wide range of the total quantities highlights the uncertainties associated with the source term assessment, which are partly due to the use of a small number of observations. The releases would have started on June 13 and continued until June 22. After June 23, the calculations indicate that the occurrence of a release is not consistent with the observations. In fact, if the release had continued after June 22, the stations would have reported an increase in ^{134}Cs air concentrations. It is therefore likely that the release stopped on June 23 or became too small to be detected.

3.2.2. Model-to-data comparison

Due to the high magnitude of the estimated source term, the dispersion of the ^{134}Cs plume was calculated in order to determine whether the plume could reach the French territory. The reconstructed source term from the location $(x,y) = (29.2; 59.2)$ which maximizes the

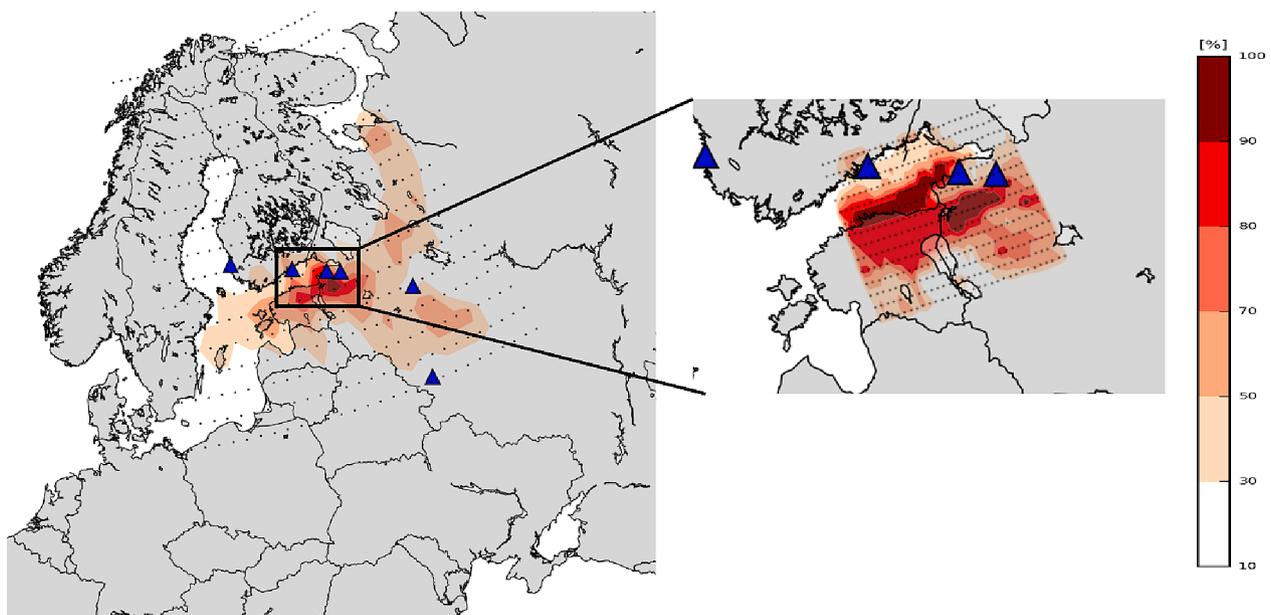


Fig. 2. June 2020 event : percent of the simulated air concentrations that is within a factor of 5 of the observed values for grid resolution of $1^\circ \times 1^\circ$ (left) and $0.2^\circ \times 0.2^\circ$ (right). Blue triangles are nuclear plants located in the area of interest: Leningrad NPP, Petersburg Nuclear physics Institute (Gatchina), Olkiluoto NPP, Loviisa NPP and Smolensk NPP.

FAC5 values is used for the calculation. Fig. 3 shows the time-integrated ^{134}Cs air concentration between June 13 and June 29.

During the period of the release, the plume mainly affected the northern European countries located between Scandinavia and the western part of the Russian Federation. Driven by east to north-east winds, the plume temporarily reached the regions between Central Europe and Germany. However, the modelled air concentrations are extremely low and usually not detectable by monitoring networks. Fig. 3 shows that the highest integrated concentrations are located between south-eastern Norway and the Leningrad region, passing through the extreme south of Finland. The simulation reproduces the observations very well. There is only a slight overestimation of the observed concentrations at Kotka, which is located at the edge of the highest simulated values.

Fig. 4 provides comparison of the time-series of ^{134}Cs observed and modelled air concentration for monitoring stations located in Finland and Sweden. The stations in Helsinki and Stockholm have a 24-hour sampling period, which allows a more accurate identification of the time of plume passage. According to Fig. 4, the plume passed through these two stations in less than a day, reaching Helsinki on June 16 and Stockholm on June 23. The simulation predicts the arrival of the plume in Helsinki on June 15 and the observed concentration is significantly underestimated on June 16. In Stockholm, however, the agreement between the simulated and observed values is very satisfactory. At the other two stations, the simulation also reproduces the measurements quite well. Two plumes would have reached the Visby station, which is

consistent with the assessment of a release with a duration of several days.

3.3. July 2019 and May 2022 detection events

The above approach is applied to identify the source location of the detections of July 2019 and May 2022. In both cases, the search domain extends between Germany and the Russian Federation with 1161 grid points. The dimensions of the search domain including the sources are $[15\text{E}, 57\text{E}]$, $[44\text{ N}, 70\text{ N}]$ with a resolution of $1^\circ \times 1^\circ$. The ^{46}Sc measurement data are used for source reconstruction as this is the only radionuclide for which at least two measurements are available. In addition to the non-zero measurements, measurements below the detection limit in Finland have also been accounted for in the calculations. To simulate the atmospheric dispersion of the ^{46}Sc , the IdX model is first forced with three-hourly meteorological data from the ARPEGE model with $0.5^\circ \times 0.5^\circ$ spatial resolution. In fact, the 0.1° spatial resolution of ARPEGE is not available for the whole domain including the potential sources. However, the meteorological fields were interpolated with a resolution of $0.25^\circ \times 0.25^\circ$ in order to reduce model dilution effects in the vicinity of the release point. The source reconstruction periods are between July 12 and July 23, 2019 and between 14 May and 1st June 2022 with daily time intervals. Using the L-curve technique, the optimal values of λ are $\lambda_{2019} = 10^{-6}$ and $\lambda_{2022} = 2.10^{-6}$ respectively.

As the number of observations above the detection limits was at most

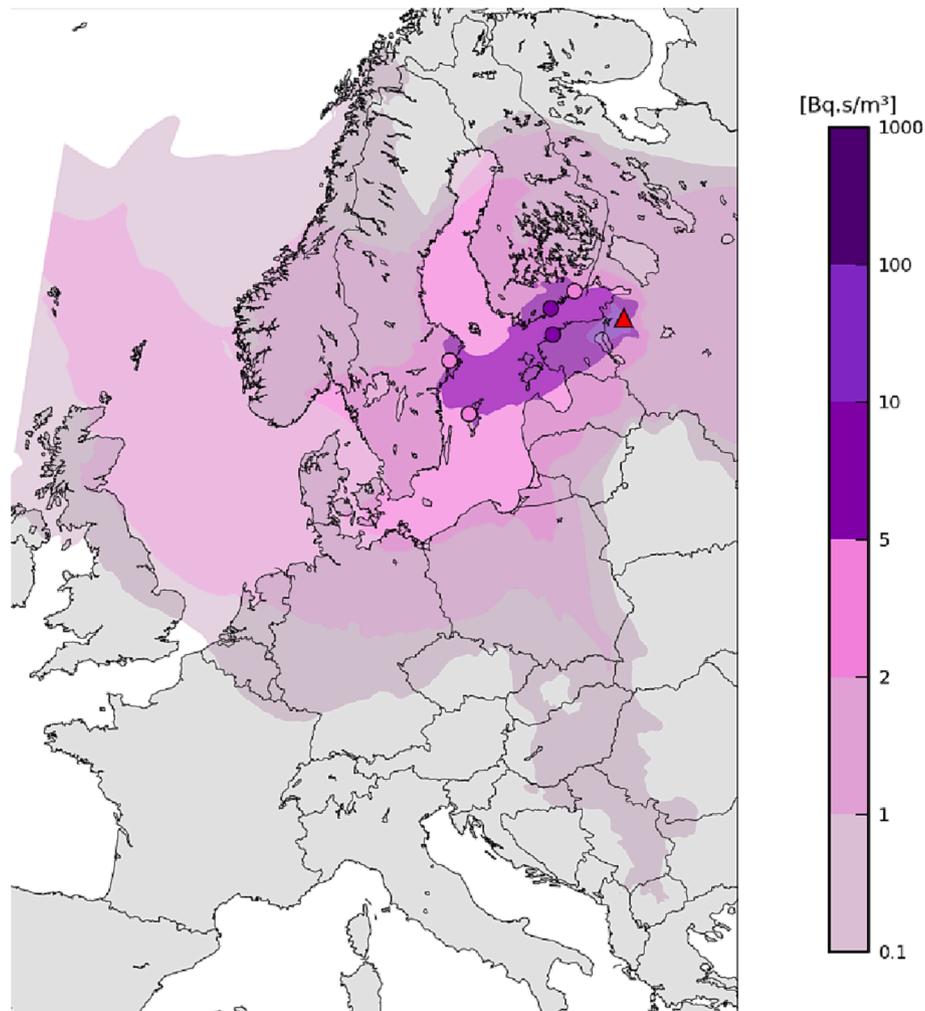


Fig. 3. June 2020 event: modelled ^{134}Cs time-integrated air concentration assuming a release location at the coordinates (29.2; 59.2). Red triangle is the release location. Circles are ^{134}Cs observed air concentrations in Bq.s/m³.

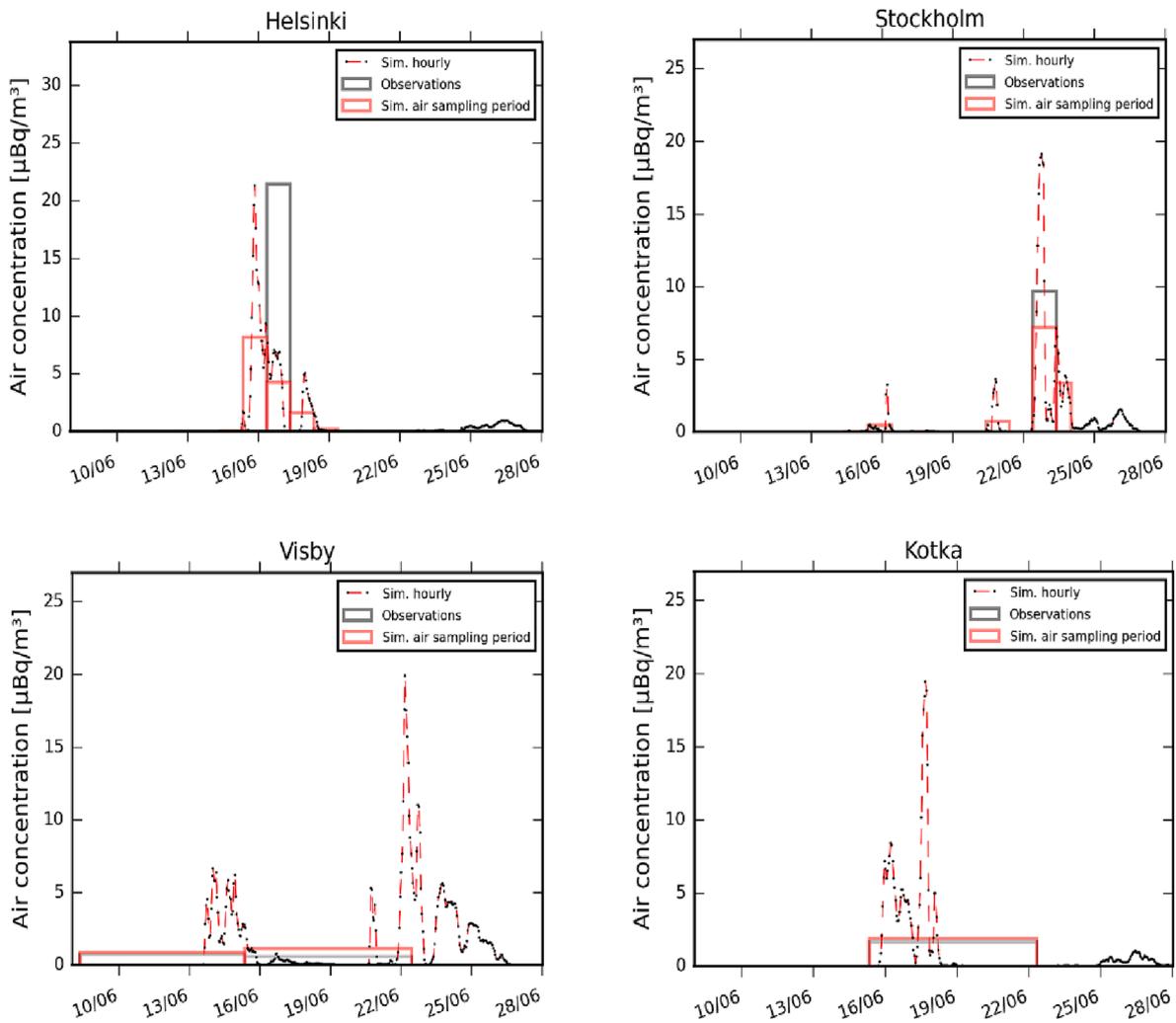


Fig. 4. Time series of observed and modelled ^{134}Cs air concentration at different locations in Sweden and Finland in June 2020.

three (two for the 2022 event), the more discriminating statistical indicator based on the cost function reduction factor was chosen to identify the most plausible release area. According to Fig. 5a, the geographical area from which a release could have occurred in July 2019 is located along the border between Estonia and the Russian Federation. In this area, the values of the reduction factor of the cost function are higher than 10000. The area around Southern Finland also has high values while outside this area and everywhere else in Europe, the values of the indicator decrease very quickly.

For the May 2022 event, the most plausible area identified is much larger (Fig. 5b), located between eastern Finland and western Russia. In addition, the release areas identified as most plausible for the July 2019 event are consistent with those for 2022.

The Leningrad region is on the edge of the highest values of the reduction factor of the cost function. The very low number of measurement data and modelling errors could explain this phenomenon.

In order to partially account for the errors associated with the modelling, in particular those from the meteorological fields, new simulations were performed using the hourly ERA5 reanalysis meteorological data from the European Center for Medium-Range Weather Forecasts (ECMWF) with $0.25^\circ \times 0.25^\circ$ spatial resolution. All other parameters for source reconstruction are similar.

Fig. 5c and Fig. 5d clearly show that the highest values of the reduction factor of the cost function are in the same areas as those identified in the first evaluation from the ARPEGE meteorological fields. These are the regions along the Finnish-Russian border. In addition, it

emerges that the Gatchina and Leningrad NPP facilities are now included in the most plausible release location. Finally, for the grid points identified as the most likely, the cost function values obtained are reduced by about 20% when the ECMWF meteorological fields are used. However, these results could be confirmed by using other numerical weather prediction models.

The uncertainties in estimating the magnitude are large due to the limited number of observations. For the most plausible release location, the total quantities released in ^{46}Sc are 1-10 GBq for the 2019 event and 5-50 GBq for the 2022 event.

4. Investigations about the source terms origins

The objective of this part is to determine from which type and part of a nuclear installation, a release could be originated. 4.1 section presents a general overview of the method used for this analysis. 4.2 section focuses on the analysis of the June 2020 event while Section 4.3 gives an overview of the detections in Finland over the last ten years. Finally, 4.4 section analyses the events containing ^{46}Sc that occurred in July 2019 and May 2022.

4.1. Source term origin analysis method

In case of detection of artificial radionuclides into the environment that cannot be linked to an event declared by a nuclear operator, many information can be deduced from the analysis of the involved

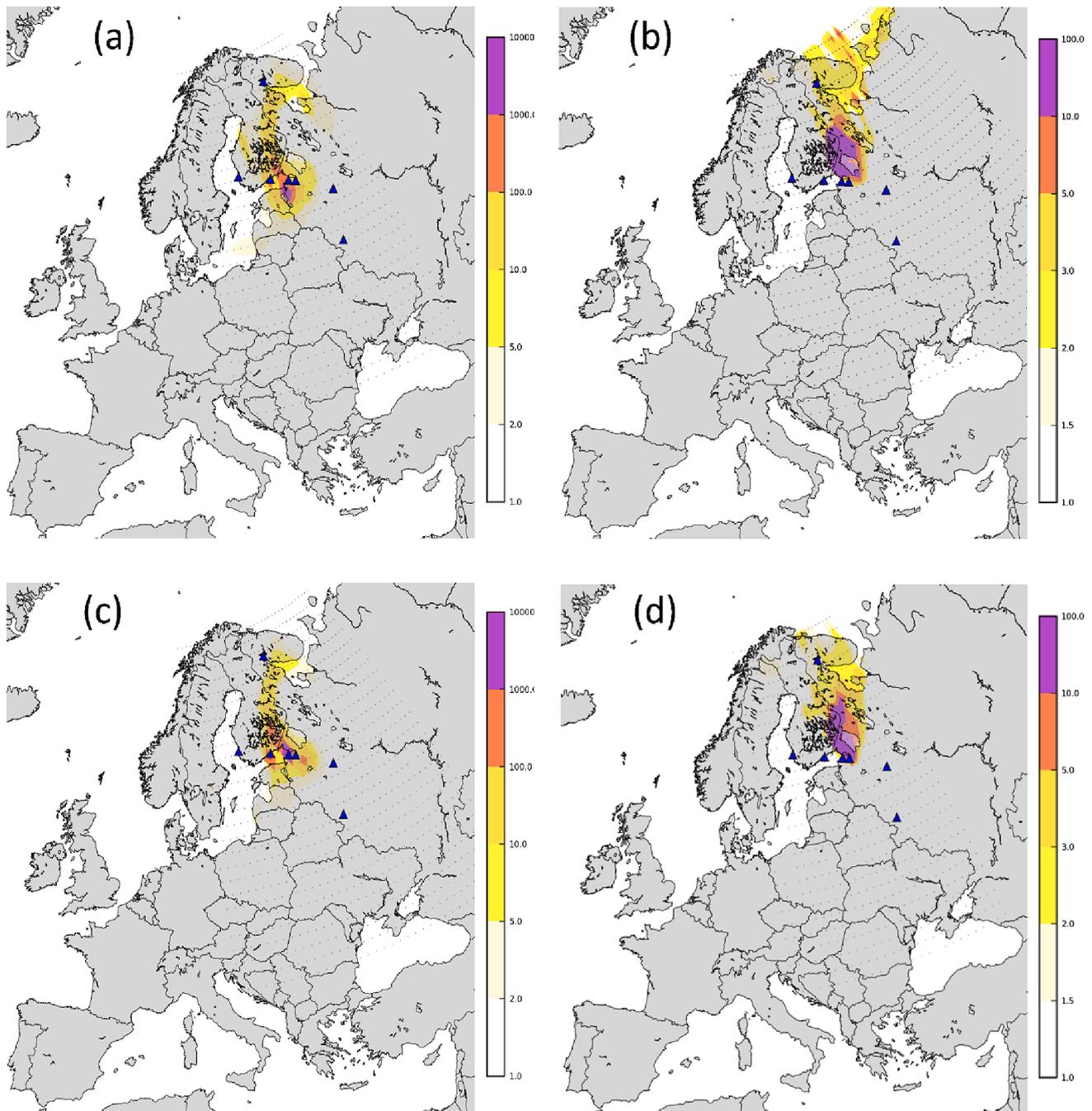


Fig. 5. Factor reduction of the cost function: (a) 2019 event and ARPEGE meteorological fields, (b) 2022 event and ARPEGE meteorological fields (c) 2019 event and ECMWF meteorological fields. (d) 2022 event and ECMWF meteorological fields. Blue triangles are nuclear plants located in the area of interest: Leningrad NPP, Petersburg Nuclear physics Institute (Gatchina), Olkiluoto NPP, Loviisa NPP and Smolensk NPP.

radionuclides.

First, radionuclides can be divided into three categories:

- **Fission Products.** Their presence implies a leak from nuclear fuel or fuel reprocessing facilities, or fissions outside of the nuclear fuel.
- **Activated Corrosion Products.** Their presence implies a release linked to the primary coolant of a reactor, the cleaning associated installation and its wastes, or eventually the decommissioning of a primary or irradiated structures.
- **Others: Sources and tracers.** Some isotopes can be produced on purpose for industrial or medical uses (^{60}Co , ^{47}Sc , ...), and may not be linked with a nuclear reactor installation.

In each case, the distinction between short-lived and long-lived isotopes give precious information about the time since the last neutron exposure (reactor operation, irradiation or criticality event), and may aid in the determination of which type of facility (e.g. nuclear power plant) or activity (e.g. fuel reprocessing) was responsible for the release.

Sometimes, some isotopes may belong to two of these categories (e.g. ^{95}Zr is a fission product and an activated corrosion product, and ^{60}Co is an activated corrosion product and also used as a gamma source). In that case, the category of the other detected isotopes is an important indicator that may suggest a similar origin to the other isotopes. Sometimes two possible origins are involved simultaneously (e.g. ^{95}Zr) but in any case, these isotopes with two possible origins require a more detailed analysis.

The main part of the source term origin analysis consists of an isotope-by-isotope comparison of the estimated source term magnitude of the release to classical radiological inventories such as those for primary coolant or wastes in order to check the capability of these inventories to generate that release. The fact that only a fraction of the radiological inventory is transferred in an atmospheric release also needs to be taken into account in the analysis. A very wide range of initial origin hypotheses has to be considered, and progressively refuted by comparing with the expected source term in the atmosphere. Every other available information about the release, such as its duration, which is sometime known, can be used in the analysis.

Performing such analysis requires a minimum number of detected isotopes to obtain enough information. With only one or two isotopes detected, it is very difficult to deduce anything about a release, unless it is very specific or with a high source term (for example (Masson et al., 2019)). Some recurrent releases involving three or four different isotopes, are an acceptable basis for a small investigation, as shown in Section 4.4. Ideally, tens of isotopes or more may lead to conclusions about the source term origin, as illustrated in Section 4.2.

Additionally, the absence of some isotopes, especially the most radioactive and volatile ones (such as ^{131}I for nuclear fuel or ^{60}Co for activated corrosion products), can provide valuable information. For example, the detection of fission products, without any ^{131}I , or any short-lived isotopes, can provide bounds on the minimum decay time. However, it is important to note that other isotopes may be present in the release, but not detectable due to instrument detection limits. Several elements may also have different physical or chemical interactions in the atmosphere, leading to differences in their spread. Additionally, each isotope has its own unique detection limits.

When two radioactive isotopes of the same element (such as Cs, Ru, Ce, ...) are detected, their ratio can provide useful information. Assuming an origin of the release (reactor type and fuel, ...), the ratio enables to evaluate the decay time since the end of irradiation. However, the evaluation of the initial ratio before the decay is an issue to be treated with caution. Indeed, depending on the reactor type (PWR, RBMK, CANDU, ...), and the specific fuel considered (burn-up), the isotopic ratio of a spent fuel (Cs for example) can vary significantly (this can also be used as an indication about the reactor origin of the release). Moreover, if the release did not come directly from a spent fuel, the spent fuel isotopic ratio cannot be used for the initial ratio. For example, in the Section 4.2, the decay time is estimated from the ratio of Ru and Ce, assuming that the release came from a spent primary ion exchange resin, with a dispersion of fissile material in the primary. In this case, the isotopic ratio accumulated in the primary resin is quite different from that present in the spent fuel, and must be specifically estimated, before evaluating a decay time.

Due to the different behaviour of the various elements in the atmosphere, the ratio between different elements should not be used, except for broad order of magnitude comparisons.

Such an approach cannot be conclusive and may not be sufficient to clearly identify the event causing the release, but it helps to greatly reduce the possible sources of the release in terms of installations and modalities.

4.2. June 2020 event

A detailed analysis of this event has been presented in a previous publication (Ingremau and Saunier, 2022), investigating from which type and part of a nuclear installation the release could have originated, and how these radiological inventories could have been released into the atmosphere.

This part summarises the main results and arguments concerning the origin of the radionuclides (section 4.2.1), and presents the analysis of new data on this event (Hoffman and Mekarski, 2021) (section 4.2.2).

4.2.1. Synthesis of the previous analysis on the origin of source term the June 2020

This event implied some activated corrosion products and some fission products, without any short-lived isotopes (suggesting a few months of decay before the release). Comparison with known possible sources of ^{60}Co , and other activated corrosion products, suggests that a spent primary ion exchange resin, or other component of the primary loop cleaning system, is the best candidate to explain the presence of such an amount of ^{60}Co , especially when measured in association with fission products. The presence of ^{134}Cs and ^{137}Cs , is expected in a spent primary ion exchange resin, in the event of cladding leakage or failure. The $^{134}\text{Cs} / ^{137}\text{Cs}$ activity ratio is consistent with a PWR origin and tends to exclude the RMBK reactor type.

The presence of ^{103}Ru and ^{106}Ru in the release is particularly interesting. Indeed, ruthenium is a low volatile fission product, that normally remains in the fuel pellet. It is very stable and only very small fractions are released out of the pellet, even in case of fuel cladding failure. So, the only identified way to get such amount of ^{103}Ru outside of a fuel pellet is to have fuel dispersed in the primary water. Indeed, some fuel cladding failures can result in the dispersion of small amounts of uranium oxide outside of the pin, in the primary water. When exposed to the neutron flux, the fissile material will fission and release its fission products directly into the primary coolant. Such events are quite rare, but considering a fleet of tens of nuclear reactors, it can occasionally happen. In this case, significant amounts of ruthenium could be released into the primary water and accumulate in the primary resin. In view of the French operating limits, it can be estimated that the amount of ^{103}Ru in a resin in such a case could be consistent with the release, taking into account a few months of decay,

In addition, the $^{103}\text{Ru}/^{106}\text{Ru}$ ratio provided some information about the decay time. Assuming that the ruthenium came from fission of the 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 range of 4 to 9 months, which is consistent with the previous conclusions. However, considering that a part of the ^{106}Ru inventory may come from the pellet inventory (and not from fission in primary water), the ruthenium ratio is overestimated. Therefore, the only reliable information from this ratio is that the decay time is less than 9 months.

In conclusion, the analysis of the origin of each detected isotope, taking into account the cumulative presence of activated corrosion products and fission products, indicates that the release measured in June 2020 could have been released from a primary resin after a few months, but less than 9 months of decay from a pressurised water reactor with fuel cladding failure and dispersion of fissile material in the primary. Nevertheless, no certainty can be achievable with so little information.

4.2.2. Analysis of additional data

A re-analysis of the samples from that event was conducted by Hoffman and Mekarski of Health Canada (Hoffman and Mekarski, 2021), which led to the detection of three additional isotopes: ^{144}Ce , $^{110\text{m}}\text{Ag}$ and ^{46}Sc . In addition, their assessment of the release location yielded results consistent with those of the IRSN (Ingremau and Saunier, 2022).

The detection of ^{144}Ce , a fission product, is fully consistent with the presence of ^{141}Ce , ^{106}Ru and ^{103}Ru , and confirms the previous analysis. Moreover, in the previous publication (Ingremau and Saunier, 2022); ^{144}Ce was already identified as an isotope that "could potentially have been detected" with lower instrumental detection limits, taking into account its fission yields and half-life. This analysis is therefore confirmed.

This measurement also allows the evaluation of the $^{141}\text{Ce}/^{144}\text{Ce}$ ratio and provides some additional information on the decay time. Assuming that the cerium originates from the fission of the dispersed fuel in the primary water (and not from the accumulated inventory in the fuel

pellets), it can be estimated that the decay time of the resin should be in the range of 2 to 5 months depending on the measurement uncertainty and the accumulation period on the resin. This result narrows the range of the ¹⁰³Ru/¹⁰⁶Ru ratio (less than 9 months). However, as with ruthenium, the cerium ratio may be under-estimated, considering that part of the ¹⁴⁴Ce inventory may come from the pellet inventory (and not the fission in the water). Therefore, the only reliable information from this ratio is that the decay time is less than 5 months. Given the absence of ¹³¹I, it can be estimated that the decay time is at least 2 months. Therefore, using these new data, the decay time of the primary resin inventory can be estimated between 2 and 5 months.

Regarding the presence of ^{110m}Ag, Neeb (Neeb, 1997) points out that for some VVER plants, high levels of ^{110m}Ag have been reported in the primary coolant and therefore in a primary ion exchange resin. The main source of this silver contamination is the silver-containing seals, of which traces are released “by corrosion or mechanical wear”. High ^{110m}Ag concentrations have also been measured at some other VVER plants due to silver impurities in the KOH used to alkalize the coolant. As a result, the presence of ^{110m}Ag is another piece of evidence, consistent with the hypothesis that the release originated from a spent VVER primary ion exchange resin. However, detailed data on the radiochemistry of the Leningrad VVER would be required to confirm this interpretation.

On the contrary, the presence of ⁴⁶Sc in a primary ion exchange resin is unexpected, as explained in Section 4.4.1. However, as described below, ⁴⁶Sc can be used and released in contexts very different from nuclear power generation. Furthermore, ⁴⁶Sc has only been detected at the Visby station, far from the estimated release location of ¹³⁴Cs. Hoffman and Mekarski in (Hoffman and Mekarski, 2021) suggest that the ⁴⁶Sc could originate from another independent release. They estimate that a ⁴⁶Sc release in Poland at the appropriate time, taking into account wind patterns, could lead to a detection at Visby during this period. Moreover, ⁴⁶Sc can be used as tracer for hydro-fracking, which is authorised in Poland. ⁴⁶Sc sellers can also be found in Poland (Polatom website). At current state of the analysis, this hypothesis is the most likely to explain the unique ⁴⁶Sc detection during this event.

In conclusion, these new data supports the interpretation that the source is a primary resin from a VVER and refines the estimated decay time after resin unloading to a range of 2 to 5 months.

4.3. Overview of comparable detection in Finland since 2012

These isotopes were not only detected in June 2020, but also several times during the last decade. At least 12 similar detections have been recorded in Finland since 2012, as shown in Table 3. It should be noted that, due to the Chernobyl accident in 1986, traces of ¹³⁷Cs are always detected in Finland. In the table, detections that are consistent with the usual ¹³⁷Cs background levels are marked with a “x” and larger

detections, that clearly indicate a fission product release, are marked with a “X”. Besides, no ⁹⁵Zr was detected in May 2017 and March 2020, although its daughter, ⁹⁵Nb, was recorded. This might be interpreted as ⁹⁵Zr being present but in amounts too small to be measured. The ⁴⁶Sc detection in June 2020 is suspected to come from another independent event.

All these detection events can be gathered into five categories, which are described in Table 4.

First, three release events have the same characteristics as the one that occurred in June 2020. Four additional detections could be interpreted as primary resin with different fuel cladding failure states (significant failure or leakage in May 2017, but without fuel dispersion, limited in other cases), and a few months of decay. They could also be a filter from the primary water cleaning system, or operational waste linked to this system. Four other detection are noteworthy for the presence of activated corrosion products and ⁴⁶Sc. These are analysed in the section §3.4. Finally, another detection, that occurred in October 2015, could also be attributed to a primary resin, a filter from the cleaning system, or operational waste, with fuel cladding failure, but with years of decay.

It can also be noticed that almost all the detections occurred in spring and summer (between March and August), with the exception of one in October. This seasonality does not seem to be explained by the prevailing wind direction in this season, which is not significantly more favourable to a detection in that period than in autumn and winter. It has therefore to be linked with the seasonality of the industrial activity leading to that release.

In conclusion, twelve atmospheric releases involving several isotopes have been detected in Finland since 2012, with subsequent events occurring quite frequently. Between three and twelve atmospheric releases could correspond to a primary resin inventory. Of the twelve, nine could be associated with a filter or operational waste from the primary of a reactor.

4.4. Analysis of the ⁴⁶Sc detection events (May 2022, July 2019 and others)

The objective of this section is to focus on the four events including a ⁴⁶Sc detection (five including the June 2020 one), especially the May 2022 and July 2019 events. Indeed, these two events are specific because they resulted in ⁴⁶Sc detections at two and three stations, respectively, enabling an assessment of the most plausible source location by inverse modelling (the other detections were limited to a single monitoring station).

4.4.1. ⁴⁶Sc identity card

This section focuses on the ⁴⁶Sc detections, as it is neither a fission

Table 3
All major detections in Finland since 2012.

Release date	Activated corrosion product				Fission product and Activated corrosion products			Fission product				Other isotopes	Release localization
	⁶⁰ Co	⁵⁸ Co	⁵⁹ Fe	⁵⁴ Mn	⁹⁵ Nb	⁹⁵ Zr	Volatile		Low volatile				
Half-life	5,3 y	71 d	45 d	0,86 y	35 d	64 d	¹³⁴ Cs	¹³⁷ Cs	¹⁰³ Ru	¹⁴¹ Ce	⁴⁶ Sc		
August 2014	X	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	
August 2016	X			X				x		X	X	Not evaluated	
May 2017	X		X	X	X	?	X	x				Not evaluated	
April 2018	X	X	X	X	X	X	X	x	x	X	X	Not evaluated	
May 2018	X		X					x			X	Not evaluated	
April 2019	X	X	X	X	X	X		x				Not evaluated	
July 2019	X		X	X				x			X	Compatible with reference	
March 2020	X		X	X	X	?		x				Not evaluated	
June 2020	X				X	X	X	X	X	X	X?	Reference	
May 2022	X		X								X	Compatible with reference	

Table 4

Various kind of detection in Finland since 2012.

Release	Fission Products				Composition FP: Fission Products CP&AP: Corrosion Products and Activation Products	Possible origin	Decay before release
	Low volatile	Volatile					
May 2015 April 2018 June 2020	X	X	X	X		Primary resin with fuel cladding failure with fuel dispersion	Few months
May 2017		X	X	X		Primary resin, cleaning system filter or operational wastes, with fuel cladding failure or leakage without fuel dispersion	Few months
August 2014			X	X		Primary resin, cleaning system filter or operational wastes, without fuel cladding failure or leakage	Few months
April 2019 March 2020							
August 2016			X	X	X	See §4.4	Few months
May 2018 July 2019 May 2022							
October 2015		X	X			Primary resin, cleaning system filter, or operational wastes, with fuel cladding failure or leakage	Few years

product nor an activated corrosion product in PWR. It must therefore have a different origin from the releases discussed in section 4.2. This isotope is an intermediate half-life isotope (84 days) that can be produced mainly by two different nuclear reactions:

- by irradiation of a natural titanium target with a fast neutron flux through the $^{46}\text{Ti}(n, p)^{46}\text{Sc}$, in a research nuclear reactor (Mikolajczak et al., 2021), or in very small quantities in the case of the presence of ^{46}Ti impurities in the internal structures of a power nuclear reactor (Ancius et al. 2005).
- by thermal neutron capture on ^{45}Sc , which is the only natural isotope of scandium: $^{45}\text{Sc}(n, \gamma)^{46}\text{Sc}$. This reaction can take place in two different contexts in a nuclear reactor:
 - Production of ^{45}Ca for medical purposes: natural ^{44}Ca targets are irradiated in a research nuclear reactor. This reaction produces ^{45}Ca ($^{44}\text{Ca}(n, \gamma)^{45}\text{Ca}$), which decays to ^{45}Sc , which can also capture a neutron to obtain ^{46}Sc (Chakravarty et al., 2022). ^{46}Sc is therefore a radionuclidic impurity.
 - Accidental neutron irradiation of internal structures of a nuclear reactor containing natural scandium (^{45}Sc). In this case, ^{46}Sc is an activated corrosion product. PWR do not contain scandium in their internal structures, but this can be the case of some research reactor, or the graphite of some RBMK (Plukiené et al., 2018).

^{46}Sc , and especially ^{47}Sc , are used in medicine radiotherapeutic or diagnostic applications (Mikolajczak et al., 2021); (Chakravarty et al., 2022). ^{46}Sc is also sometimes used in the oil industry as a tracer (Tingvoll), typically used in hydraulic fracking (Hoffman and Mekarski). Companies supplying ^{46}Sc can be found in Poland for example (Polatom website).

4.4.2. Detections including ^{46}Sc

Table 5 shows the four detection events with ^{46}Sc in Finland since 2012 (excluding the one in June 2020). First, it can be noted that ^{46}Sc has always been detected in association with ^{60}Co . In the four detections, another activated corrosion product ^{59}Fe or ^{54}Mn is also present (both for the July 2019 event). Neither short-lived isotopes nor fission product are detected in these events (except for ^{141}Ce detection in August 2016). ^{46}Sc therefore appears to be associated with common activated corrosion product.

Table 5Detections containing ^{46}Sc in Finland since 2012.

Release date	^{46}Sc	^{60}Co	^{59}Fe	^{54}Mn	Others	Release location
Half-life	84 d	5,3 y	45 d	0,86 y		
August 2016	X	X		X	^{141}Ce	Not evaluated
May 2018	X	X	X			Not evaluated
July 2019	X	X	X	X		Leningrad region
May 2022	X	X	X			Leningrad region

^{141}Ce was also detected during the August 2016 event. The detection of this low volatile fission product is quite surprising given the presence of only activated corrosion products and the absence of other fission products. A possible explanation is provided in Section 4.4.5. However, it could also have originated from another release source during the same period.

The June 2020 event is not presented in Table 4 because the ^{46}Sc was hypothesized to have a different source. Furthermore, the detection occurred only in Visby in Sweden, and not in Finland.

4.4.3. Analysis of the July 2019 and May 2022 events

A dedicated atmospheric transport analysis has been conducted (see section 2), enabling an evaluation of the source term magnitude. ^{46}Sc is a significant component of the release as it was detected at all stations measuring the event (unlike the June 2020 event). Assuming the ^{46}Sc release magnitude at the most probable release location, and the various activity ratios observed in the measurements, the order of magnitude of each isotope of the source term of the two events is from a few GBq up to one hundred GBq.

Some general comments can be drawn on these source terms:

First, it must be noted that, similarly to the twelve events described in section 3.3, it does not contain any short-lived isotopes (such as iodine). This tends to exclude an event involving very recently irradiated material (less than a few weeks old).

Second, ^{59}Fe is an isotope with “intermediate half-life” (meaning around a few months). This implies that the event concerns relatively recent irradiated materials, with less than a year of decay.

Third, in contrast to the June 2020 release, and other similar events, no fission products have been detected.

The amounts of ^{60}Co , ^{59}Fe , and ^{54}Mn involved are quite common in

primary resin, but can also be found in some solid wastes generated during the daily operation of a nuclear power plant (with a decay time of only a few months): filters of the primary water purification system, together with gloves, clothes, tools, replaced metal parts of primary components, irradiated graphite of RBMK, ... For the June 2020 event, the presence of fission products, and especially non-volatile ones, suggests a primary resin source. However, without any fission products, it could be any "primary" solid waste.

Another characteristic of these events, as well as those of the August 2016 and May 2018, is the presence of ^{46}Sc associated with these activated corrosion products. As previously explained, ^{46}Sc is not expected to be found in a PWR nuclear power plant or in the primary loop of a RBMK, and is generally intentionally produced by specific facilities. The simultaneous detection of these isotopes is therefore a puzzle, especially as no other isolated ^{46}Sc detections have been reported in the last decade (except possibly the ^{46}Sc detection in June 2020, but as it is mixed with the main release it is impossible to distinguish). ^{46}Sc and these activated corrosion products have always been detected together.

4.4.4. The mixing hypothesis

One hypothesis to explain this synchronicity, could be to consider that ^{46}Sc and the activated corrosion products are waste products of different origin, and were incinerated together in a solid waste incineration center. This would be consistent with a hypothesis proposed in (Ingremau and Saunier, 2022) to explain the June 2020 detection event.

However, according to this interpretation, ^{46}Sc and activated corrosion products should not always be detected together. Instead, there should be occasional detections of either ^{46}Sc alone or activated corrosion products alone. Cases of isolated activated corrosion products have been observed several times (e.g. some of the events listed in Table 3, or detection of only ^{60}Co and ^{54}Mn in April and July 2012). However, no isolated detections of ^{46}Sc have not been reported in Finland during the last ten years. The absence of such detections is evidence against this hypothesis.

In addition, in such a case of deliberate incineration, it is expected that the emissions will be filtered, greatly reducing the activity released to non-detectable levels.

4.4.5. The RBMK graphite hypothesis

^{46}Sc is also unintentionally produced in the graphite of RBMK reactors, in the core, outside the primary loop. In fact, graphite (used as a moderator in RBMK reactor cores) always contains some impurities. These impurities are highly variable, depending on the industrial purity required (Toyo Tanso) and the origin of the graphite (Ancius et al. 2005); (Plukienė et al., 2018); (Plukienė et al., 2011).

If ^{46}Ti is present in the graphite, ^{46}Sc can be produced by the fast neutron reaction $^{46}\text{Ti}(n, p)^{46}\text{Sc}$, as evaluated in (Plukienė et al., 2011). However, in this case, the amount of ^{46}Sc is quite small, around 1 kBq/g of graphite in operation. Considering that the source term of ^{46}Sc in the various detected releases is of the order of GBq, it would require several tonnes of recently irradiated graphite to obtain this amount of ^{46}Sc , which seems rather unrealistic.

However, if there is ^{43}Sc impurity in the graphite, the amount of ^{46}Sc can be much higher, as evaluated in (Ancius et al. 2005), reaching tens of kBq/g after one year of decay. With a shorter decay time of few months, the activity of ^{46}Sc could even be around a hundred of kBq/g. With this hypothesis of the Scandium impurity content and decay time, a few tens of kilograms of irradiated graphite could contain one GBq of ^{46}Sc .

Another interesting aspect of this hypothesis is that ^{46}Sc is associated with ^{60}Co (detected) and ^{54}Mn (detected in August 2016 and July 2019) in the irradiated graphite. The two publications (Ancius et al. 2005); (Plukienė et al., 2011) focused mainly on long term waste management, and therefore did not evaluate the activity of short-lived isotopes such as ^{59}Fe (45 days). However, the high activity estimated for ^{59}Fe (long lived,

with 2.7 years of half-life), indicates that ^{59}Fe would be present with significant activity for few months of decay, consistent with its frequent detection with ^{46}Sc . Other isotopes would also be present with high activities, such ^{14}C , which could not be detected because it would be in a gaseous form (CO_2), ^3H , also undetectable (H_2O), ^{55}Fe or ^{45}Ca , both undetectable with gamma spectroscopy. **As a result, the inventory of a few tens of kilograms of irradiated RBMK graphite after a few months of decay, assuming a significant scandium impurity content, is fully consistent with the source term of the detections from Table 5.**

The ^{141}Ce detection of August 2016 could also be explained with this hypothesis. Indeed, graphite also contains some Uranium impurities, which produce fission products and minor actinides, as seen in (Ancius et al. 2005); (Plukienė et al., 2011). And ^{141}Ce is one of the most active fission products after few months of decay. A dedicated short-lived isotope simulation of graphite irradiation would be required to confirm this explanation, but the orders of magnitude are compatible with this hypothesis.

Russia and others countries with large amounts of irradiated graphite, such as the UK or Ukraine, have not yet defined the future of the graphite from decommissioned nuclear power plants that is currently decaying in reactors (Dorofeev et al., 2019). However, this inventory is not of interest because after one year of decay, it would not contain enough ^{46}Sc to explain the detections events. To explain the ^{46}Sc detections, an intervention on recently irradiated graphite must be considered. At the time of the first ^{46}Sc release event in 2016, four RBMK reactors were in operation at the Leningrad power plant in 2016, and two are still in operation in 2023.

It is possible that a graphite block in an operating RBMK may have been damaged and the cleaning process may have produced some debris of irradiated graphite which, after few months of decay, ended up in a solid waste incinerator. However, in such case of intentional incineration, the emissions are expected to be filtered, which would significantly reduce the released activity to undetectable levels. Despite this major release magnitude issue, that assumption of irradiated graphite could explain the characteristics of the ^{46}Sc events.

4.4.6. The PIK hypothesis for the May 2022 event

One of the main industrial uses of scandium is in aluminum alloys (Kulikov et al., 2018; Zakharov, 2003). In particular, the Russian Federation developed an aluminum-scandium alloy during the Cold War, which was used mainly in military aircraft (AZO, 2021). The use of such an alloy in the primary loop of a nuclear reactor could produce significant amounts of ^{46}Sc , together with other activated corrosion products. In support of this idea, ^{46}Sc was one of the isotopes released by the effluent treatment plant at the Hanford nuclear research center in the 1960s, and neutron capture on the scandium content of the aluminum alloy in the reactor was one of the sources investigated, although not the main source according to (Hanford, 1970).

PWR, such as VVER, or RBMK do not use the aluminum alloy in their primary loop. However, the estimated area from which the release could have originated includes the Gatchina Research Center. This site is home to the PIK reactor, a high-flux research nuclear reactor (Reactor PIK), which uses aluminum for the core tank (Konoplev, 1995). Although it is not stated whether the core tank uses a scandium-aluminum alloy, this possibility that cannot be ruled out.

It should be noted that the PIK reactor started operating at high power on 8 February 2021 (Russia launches PIK research reactor, 18 February, 2021), and has been upgraded in February 2022 (Russian PIK research reactor upgraded, 2022). Therefore, solid waste linked to operational activities on the primary loop of the PIK reactor, could be expected to contain ^{60}Co and ^{59}Fe together with ^{46}Sc and could be a credible origin of the May 2022 release. This detection occurred one and half year after the start of high-power operation of the reactor, which is a credible delay (albeit short) for solid waste production and incineration.

However, this hypothesis could not explain the three ^{46}Sc detections

occurred before 2021 and is therefore not the most likely source.

4.4.7. Synthesis on ^{46}Sc detections

^{46}Sc , which is not produced in PWR reactor, was unexpectedly found to be consistently associated with activated corrosion products from nuclear reactors in the four detection events considered.

One hypothesis to explain this synchronicity is that ^{46}Sc and the activated corrosion products are waste products of different origin that were incinerated together in a solid waste incineration center. In this scenario, ^{60}Co , ^{59}Fe , and ^{54}Mn would be expected to originate from operational solid waste from the primary loop, such as resins, filters or other materials, after a few months of decay, while ^{46}Sc would originate from medical or industrial sources. However, according to this interpretation, ^{46}Sc and the activated corrosion products should not always be detected together, and occasional detections of ^{46}Sc alone should occur. The absence of such detections in Finland during the last decade is evidence against this hypothesis.

An alternative hypothesis is that the radiological inventory originated from tens of kilograms or more of irradiated graphite from an RBMK after a few months of decay, assuming a minimum scandium impurity content in the graphite. This assumption could explain all the characteristics observed in the ^{46}Sc events.

However, for both hypotheses (mixed waste in the incinerator or RBMK graphite), the intentional incineration of solid waste is expected to involve emission filtering, which would strongly reduce the released activity to undetectable levels. At this stage of the analysis, a satisfactory solution to this contradiction remains elusive.

Finally, an alternative explanation could involve the PIK reactor, which has an aluminum core tank and could use a scandium-aluminum alloy. In this scenario, irradiation would produce ^{46}Sc as an activated corrosion product, which could be present in some solid waste. However, this hypothesis has a limitation: it can only explain the May 2022 event and not the three previous events, making it a less likely explanation.

Further investigation is therefore needed to determine the exact cause of these events, which could include more detailed measurements and increased international cooperation and information sharing between nuclear facilities.

5. Conclusions

In conclusion, this study demonstrates the value of the developed methodology in investigating small undeclared releases of radionuclides to the atmosphere, while acknowledging the challenges in definitively identifying the event causing the release. The results provide valuable insights into the potential sources and origins of these releases and contribute to a better understanding of radionuclide release events.

The methodology consists of two main parts: first, an inverse modelling approach that combines environmental measurements and atmospheric transport modelling to determine the most relevant source location; and second, an analysis of isotope production pathways and comparison with known typical inventories to identify the type and part of a nuclear facility potentially responsible for the release.

This study provides a detailed analysis of three detection events: July 2019, June 2020 and May 2022. Source identification based on inverse modelling indicates that releases from the Leningrad region of the Russian Federation could explain the detection of artificial radionuclides in several northern European countries during these events. The calculations suggest that only a release from this geographical area is suitable to reproduce the set of measurements. The use of ECMWF meteorological fields improves the agreement between modelled and observed concentrations, thereby increasing confidence in the estimation of the source term. The total amounts released to the atmosphere during these three events are generally of similar magnitude, ranging from a few to tens of gigabecquerels.

The previous analysis of the June 2020 (Ingremeau and Saunier,

2022) event has been re-examined, using new measurement information obtained from complementary air filter analysis. The updated interpretation suggests that the most likely origin of this release is a primary ion exchange resin, after a decay period of 2 to 5 months, from a pressurised water reactor that experienced a fuel cladding failure and dispersion of fissile material in the primary.

For the July 2019 and May 2022 releases, the presence of ^{46}Sc was also found in association with activated corrosion products. This is quite unexpected, as ^{46}Sc is not produced during the operation of PWR or in primary loop of RBMK. Two hypotheses are proposed to explain the origin of their source term:

One possibility is that ^{46}Sc and the activated corrosion products are waste products of different origin that were co-incinerated in a solid waste incineration center.

Another hypothesis suggests that the source could be tens of kilograms or more of irradiated graphite from an RBMK reactor, after a few months of decay and assuming a minimal scandium impurity content in the graphite.

Both hypotheses can explain the characteristics of the ^{46}Sc events, but suffers from the fact that, in a case of deliberate incineration, emissions are expected to be filtered, which should strongly decrease the released activity released to undetectable levels. Consequently, the origin of these two releases remains uncertain and requires further investigation.

CRedit authorship contribution statement

O. Saunier: Formal analysis, Validation, Writing – original draft. **J.J. Ingremeau:** Investigation, Validation, Writing – original draft. **Ian Hoffman:** Resources, Writing – review & editing. **Pawel Mekariski:** Resources, Writing – review & editing. **Jing Yi:** Resources, Writing – review & editing. **Adrian Botti:** Resources, Writing – review & editing.

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.

Data availability

The data that has been used is confidential.

Acknowledgments

The authors express their gratitude to all members of the ro5 network and the STUK for providing measurement data. They also acknowledge the contribution of IRSN colleagues involved in the analysis of the event. Additionally, they extend thanks to Tero Karhunen from STUK for engaging in productive discussions.

References

- Advantages of Aluminum-Scandium Alloy in Shipping, November 2021, n.d.. AZO Materials website. <https://www.azom.com/article.aspx?ArticleID=20947>.
- Chakravarty, R., Ram, R., Patra, S., Sarma, H.D., Chakraborty, S., 2022. A solvent extraction-based procedure for removal of ^{46}Sc impurity from reactor produced ^{45}Ca CaCl₂ for its potential use in bone pain palliation [WWW Document]. Applied radiation and isotopes. In: including data, instrumentation and methods for use in agriculture, industry and medicine. <https://doi.org/10.1016/j.apradiso.2022.110352>.
- Ancius, D., Ridikas, D., Remeikis, V., Plukis, A., Plukiene, R., Cometto, M., 2005. Evaluation of the activity of irradiated graphite in the Ignalina Nuclear Power Plant RBMK-1500 reactor. *Nukleonika* 50, 113–120.
- Cooke, M.W., Botti, A., Zok, D., Steinhauser, G., Ungar, K.R., 2020. Identification of a chemical fingerprint linking the undeclared 2017 release of ^{106}Ru to advanced nuclear fuel reprocessing. *Proc. Natl. Acad. Sci. U.S.A.* 117 (26), 14703–14711.
- De Meutter, P., Gueibe, C., Tomas, J., den Outer, P., Apituley, A., Bruggeman, M., Camps, J., Delcloo, A., Knetsch, G.-J., Roobol, L., Verheyen, L., 2021. The assessment of the April 2020 chernobyl wildfires and their impact on Cs-137 levels in Belgium

- and The Netherlands. *J. Environ. Radioact.* 237, 106688 <https://doi.org/10.1016/j.jenvrad.2021.106688>.
- De Meutter, P., Hoffman, I., 2020. Bayesian source reconstruction of an anomalous Selenium-75 release at a nuclear research institute. *J. Environ. Radioact.* 218, 106225 <https://doi.org/10.1016/j.jenvrad.2020.106225>.
- Dorofeev, A.N., Komarov, E.A., Zakharova, E.V., Volkova, A.G., Linge, I.I., Ivanov, A.Y., Utkin, S.S., Pavlyuk, A.O., Kotlyarevsky, S.G., 2019. On reactor graphite. *RW* (2), 18–30.
- Dumont Le Brazidec, J., Bocquet, M., Saunier, O., Roustan, Y., 2020. MCMC methods applied to the reconstruction of the autumn 2017 Ruthenium-106 atmospheric contamination source. *Atmospheric Environment: X* 6, 100071.
- Dumont Le Brazidec, J., Bocquet, M., Saunier, O., Roustan, Y., 2020. MCMC methods applied to the reconstruction of the autumn 2017 ruthenium 106 atmospheric contamination source. *Atmos. Environ.* 6, 100071.
- Declassified Hanford historical document, "Source of process water scandium", June 1970.
- Hansen, C., 1992. Analysis of discrete ill-posed problems by means of the L-curve. *SIAM Revue* 34 (4), 561–580. <https://doi.org/10.1137/1034115>.
- Ingremau, J.J., Saunier, O., 2022. Investigations on the source term of the detection of radionuclides in North of Europe in June 2020. *EPJ Nuclear Sci. Technol.* 8, 10. <https://doi.org/10.1051/epjn/2022003>.
- Information Report from IRSN "Detection of an increase in airborne radioactivity levels in Northern Europe – Update of July 22, 2020".
- Hoffman, I., Mekarski, P., n.d. CTBT Science and Technology Conference 2021 (SnT2021). CTBT Conferences and Workshops (Indico). URL <https://conferences.ctbto.org/event/7/contributions/904/> (accessed 6.2.23).
- Konoplev, K.A., 1995. The state of the PIK reactor construction. Petersburg Nuclear Physics Institute Russia. <https://www.osti.gov/etdeweb/servlets/purl/20571819>.
- Kovalets, I.V., Romanenko, O., Synkeyvych, R., 2020. Adaptation of the RODOS system for analysis of possible sources of Ru-106 detected in 2017. *J Environ Radioact.* 220–221, 106302 <https://doi.org/10.1016/j.jenvrad.2020.106302>. Epub 2020 May 16 PMID: 32560890.
- Kulikov, B.P., Baranov, V.N., Bezrukih, A.I., Deev, V.B., Motkov, M.M., 2018. Preparation of Aluminum-Scandium Master Alloys by Aluminothermal Reduction of Scandium Fluoride Extracted from Sc2O3. *Metallurgist* 61, 1115–1121. <https://doi.org/10.1007/s11015-018-0614-1>.
- Liu, D.C., 1989. On the limited memory method for large scale optimization. *Math. Program. B.* 45 (3), 503–528.
- Lucas, D.D., Simpson, M., Cameron-Smith, P., Baskett, R.L., 2017. Bayesian inverse modeling of the atmospheric transport and emissions of a controlled tracer release from a nuclear power plant. *Atmos. Chem. Phys.* 17, 13521–13543. <https://doi.org/10.5194/acp-17-13521-2017>.
- Masson, O., Steinhäuser, G., Zok, D., Saunier, O., Angelov, H., Babić, D., Bečková, V., Bieringer, J., Bruggeman, M., Burbidge, C.L., Conil, S., Dalheimer, A., De Geer, L.-E., de Vismes Ott, A., Eleftheriadis, K., Estier, S., Fischer, H., Garavaglia, M.G., Gasco Leonarte, C., Gorzkiewicz, K., Hainz, D., Hoffman, I., Hýža, M., Isajenko, K., Karhunen, T., Kastlander, J., Kätzberger, C., Kierepko, R., Knetsch, G.-J., Kövendiné Kónyi, J., Lecomte, M., Mielteński, J.W., Min, P., Möller, B., Nielsen, S.P., Nikolic, J., Nikolovska, L., Penev, I., Petrínek, B., Povínek, P.P., Querfeld, R., Raimondi, O., Ransby, D., Ringer, W., Romanenko, O., Rusconi, R., Saey, P.R.J., Samsonov, V., Šilobritienė, B., Simion, E., Söderström, C., Šostarić, M., Steinkopff, T., Steinmann, P., Sýkora, I., Tabachnyi, L., Todorovic, D., Tomankiewicz, E., Tschiersch, J., Tšibranski, R., Tzortzis, M., Ungar, K., Vidic, A., Weller, A., Wershofen, H., Zagvyai, P., Zalewska, T., Zapata García, D., Zorko, B., 2019. Airborne concentrations and chemical considerations of radioactive ruthenium from an undeclared major nuclear release in 2017. *Proc Natl Acad Sci USA* 116 (34), 16750–16759.
- Masson, O., Romanenko, O., Saunier, O., Kirieiev, S., Protsak, V., Laptev, G., Voitsekhovaly, O., Durand, V., Coppin, F., Steinhäuser, G., de Vismes Ott, A., Renaud, P., Didier, D., Boulet, B., Morin, M., Hýža, M., Camps, J., Belyaeva, O., Dalheimer, A., Eleftheriadis, K., Gascó-Leonarte, C., Ioannidou, A., Isajenko, K., Karhunen, T., Kastlander, J., Kätzberger, C., Kierepko, R., Knetsch, G.-J., Kónyi, J. K., Mielteński, J.W., Mirsch, M., Möller, B., Nikolić, J.K., Povínek, P.P., Rusconi, R., Samsonov, V., Sýkora, I., Simion, E., Steinmann, P., Stoulos, S., Suarez-Navarro, J.A., Wershofen, H., Zapata-García, D., Zorko, B., 2021. Europe-wide atmospheric radionuclide dispersion by unprecedented wildfires in the chernobyl exclusion zone, April 2020. *Environ. Sci. Technol.* 55, 13834–13848. <https://doi.org/10.1021/acs.est.1c03314>.
- Mikolajczak, R., Huclier-Markai, S., Alliot, C., Haddad, F., Szikra, D., Forgacs, V., Garmuszek, P., 2021 May 25. Production of scandium radionuclides for theranostic applications: towards standardization of quality requirements. *EJNMMI Radiopharm Chem.* 6 (1), 19. <https://doi.org/10.1186/s41181-021-00131-2>. PMID: 34036449; PMCID: PMC8149571.
- Neeb, K.H., 1997. The radiochemistry of nuclear power plants with light water reactors, ISBN 3-11-013242-7.
- Plukienė, R., Plukis, A., Puzas, A., Remeikis, V., Duškesas, G., Germanas, D., 2011. Modelling of impurity activation in the RBMK reactor graphite using MCNPX. *Progress in Nuclear Science and Technology* 2 (0), 421–426.
- Plukienė, R., Lagzdina, E., Juodis, L., Plukis, A., Puzas, A., Gvozdaitė, R., Remeikis, V., Révay, Z., Kučera, J., Ancius, D., Ridikas, D., 2018. Investigation of impurities of RBMK graphite by different methods. *Radiocarbon* 60 (6), 1861–1870.
- Polatom website, <https://www.polatom.pl/produkt/skand-46sc-3-2/>.
- IAEA: Aware of Information on Detections of Radioisotopes in Air, Seeking Information from IAEA Member States [WWW Document], 2020a. URL <https://www.iaea.org/newscenter/pressreleases/iaea-aware-of-information-on-detections-of-radioisotopes-in-air-seeking-information-from-iaea-member-states> (accessed 4.4.23).
- Reactor PIK, website of the Petersburg nuclear physics institute, <http://www.pnpi.spb.ru/en/facilities/reactor-pik>.
- Russia launches PIK research reactor, 18 February 2021, Nuclear Engineering International, <https://www.neimagazine.com/news/newsrussia-launches-pik-research-reactor-8529400>.
- Russian PIK research reactor upgraded, 7 February 2022, World Nuclear News, <https://www.world-nuclear-news.org/Articles/Russian-PIK-research-reactor-upgraded>.
- Saunier, O., Didier, D., Mathieu, A., Masson, O., Dumont Le Brazidec, J., 2019. Atmospheric modeling and source reconstruction of radioactive ruthenium from an undeclared major release in 2017. *Proc. Natl. Acad. Sci.* 116 (50), 24991–25000. <https://doi.org/10.1073/pnas.1907823116>.
- Saunier, O., Mathieu, A., Didier, D., Tombette, M., Quélo, D., Winiarek, V., Bocquet, M., 2013. An inverse modelling method to assess the source term of the Fukushima Nuclear Power Plant accident using gamma dose rate observations. *Atmos. Chem. Phys.* 13, 11403–11421.
- Stohl, A., Seibert, P., Arduini, J., Eckhardt, S., Fraser, P., Grealley, B.R., Lunder, C., Maione, M., Mühle, J., O'Doherty, S., Prinn, R.G., Reimann, S., Saito, T., Schmidbauer, N., Simmonds, P.G., Vollmer, M.K., Weiss, R.F., Yokouchi, Y., 2009. An analytical inversion method for determining regional and global emissions of greenhouse gases: Sensitivity studies and application to halocarbons. *Atmos. Chem. Phys.* 9, 1597–1620. <https://doi.org/10.5194/acp-9-1597-2009>.
- Stohl, A., Seibert, P., Wotawa, G., Arnold, D., Burkhardt, J.F., Eckhardt, S., Tapia, C., Vargas, A., Yasunari, T.J., 2012. Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition. *Atmos. Chem. Phys.* 12, 2313–2343. <https://doi.org/10.5194/acp-12-2313-2012>.
- Tichý, O., Šmíd, V., Hofman, R., Sindelářová, K., Hýža, M., Stohl, A., 2017. Bayesian inverse modeling and source location of an unintended ¹³¹I release in Europe in the fall of 2011. *Atmos. Chem. Phys.* 17, 12677–12696. <https://doi.org/10.5194/acp-17-12677-2017>.
- Tingvoll, J. T. Radioactivity in the Oil Industry – How Much is it and is it of any Concern?, Oil Field Chemicals, 7th international symposium, 17-20 march 1996 <https://www.osti.gov/etdeweb/servlets/purl/491903>.
- IAEA: Receives Member State Data on Radioisotopes Detected in the Air, Sees No Human Health Risk [WWW Document], 2020b. URL <https://www.iaea.org/newscenter/pressreleases/iaea-receives-member-state-data-on-radioisotopes-detected-in-the-air-sees-no-human-health-risk> (accessed 4.4.23).
- Toyo Tanso, "Propriétés des produits en graphite special" https://www.toyotanso.france.com/wa_files/1_Special_graphite_fr_0421_web.pdf.
- Tombette, M., Quentric, E., Quélo, D., Benoit, J.P., Mathieu, A., Korsakissok, I., Didier, D., 2014. A software platform for assessing the consequences of an accidental release of radioactivity into the atmosphere. <http://venus.iis.u-tokyo.ac.jp/english/workshop/Poster/3rd%20March/Damien%20Didier.pdf>.
- Winiarek, V., Vira, J., Bocquet, M., Sofiev, M., Saunier, O., 2011. Towards the operational estimation of a radiological plume using data assimilation after a radiological accidental atmospheric release. *Atmos. Environ.* 45, 2944–2955. <https://doi.org/10.1016/j.atmosenv.2010.12.025>.
- Yee, E., Hoffman, I., Ungar, K., 2014. Bayesian Inference for Source Reconstruction: A Real-World Application. *International Scholarly Research Notices* 2014, e507634. <https://doi.org/10.1155/2014/507634>.
- Zakharov, V.V., 2003. Effect of scandium on the structure and properties of aluminum alloys. *Met. Sci. Heat Treat.* 45, 246–253. <https://doi.org/10.1023/A:1027368032062>.