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Possible impact on IMS observations of radioiodine and radioxenon emission produced by spontaneous fission of Cm-244 and Pu-240 and released during spent nuclear fuel (SNF) reprocessing plants

Tatiana Boitsova, Martin Kalinowski, Carla Pires, Jolanta Kuśmierczyk-Michulec
OSI&IDC/CTBTO, Vienna, Austria

***Disclaimer**: The views expressed on this presentation are those of the author and do not necessarily reflect the view of the CTBTO.*

Introduction

It is commonly recognised that only nuclear facilities such as Medical Isotope Production Facilities (MIPF) and Nuclear Power Plants (NPP) discharge significant amounts of radioxenon radionuclides into the environment, whereas the release from other ones such as SNF reprocessing and HLW vitrification plants, SNF and waste storage facilities is negligible. As a result, most of the amount of radioxenon created during irradiation decays during the years of cooling or processing.

However, due to spontaneous fission of Cm-244 and Pu-240 isotopes created in fuel during irradiation, some radioxenon can still be present in the fuel even after extensive cooling. There has been no investigation to ascertain whether the IMS observations are also caused by the operation of a Spent Nuclear Fuel (SNF) reprocessing plant. SNF reprocessing can swiftly consume the whole radioxenon stockpile in the absence of a reliable gas-off system.

The goal of this initiative is research into potential radioxenon emissions from heavy element spontaneous fission during spent nuclear fuel reprocessing.

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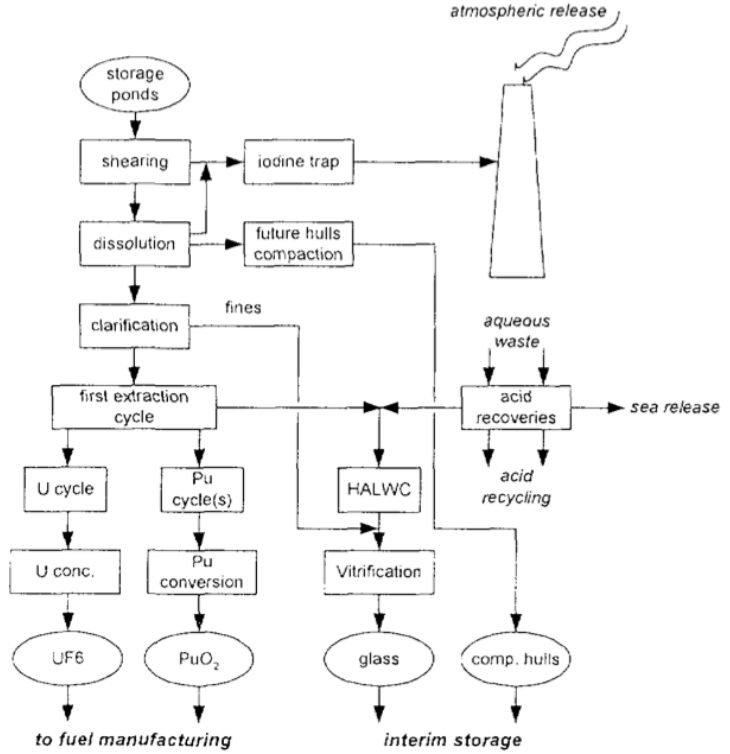
SNF reprocessing facilities and technology overview

There is currently 3 commercial reprocessing plant in operation: 2 in France (UP-2-800, UP-3, 1000 t HM/year each) and 1 in Russia (RT-1, 400 t U/year). Commercial reprocessing plant in Sellafield ends its operations in 2020 and there are 3 more commercial plants under construction in Russia and Japan [1]

Reprocessing at the La Hague complex (UP2-800 and UP3 plants), steps:

1. Fuel *transportation*
2. *Shearing and dissolution* of the fuel (contains **Cm-244 and Pu-240**)
3. Uranium and plutonium *separation and purification* from fission products (FP) by a liquid-liquid extraction process (PUREX process).
   Solution generated:
   - U containing solution
   - Pu containing solution (contains **Pu-240)**
   - FP and MA containing raffinates (contains **Cm-244**)
   - the recycled solvent
4. *Final conversion* of uranium and plutonium solutions to end-products (oxides) (contains **Pu-240**)
5. *Management and treatment* of process waste:
   - *Compacting* solid waste (The hulls, provided during shearing and dissolution operations), for final disposal.
   - *Storage of HALW* concentrates and insoluble particles in large vessels fitted with cooling and pulsation devices *and vitrification* (contains FM and MA including **Cm-244**).
   - Nitric acid *regeneration.*



LWR reprocessing Scheme at La Hague [2]

[1] Nuclear fuel cycle facilities database (NFCIS) Available at: https://infcis.iaea.org/NFCFDB/facilities
[2] Reprocessing rtr fuel in the La Hague Plants https://inis.iaea.org/collection/NCLCollectionStore/_Public/32/027/32027374.pdf

Methodology overview

The following strategy was used to identify the radioxenon emissions from SNF reprocessing facilities and its influence on IMS observations:

Step (approach) 1:

a) Identify the characteristics of the fuel reprocessed during defined period (type, burn up, enrichment, cooling time) and days of plant operations.
b) Calculate the amount of fuel and the content of Cm-244 and Pu-240 reprocessed daily.
c) Calculate amount of radioxenon and radioiodine from the spontaneous fission of Cm-244 and Pu-240.

Step (approach) 2:

a) Collect available published data on I-131 and I-133 releases (liquid, gaseous) from SNF reprocessing facilities of interest during defined period.
b) Calculate radioxenon using I-131 as a proxy.

Step 3. Compare the amount of radioxenon and radioiodine released during the reprocessing calculated using both approaches.

Step 4. Perform ATM simulation to identify the possible effect from radioxenon release created by the SNF reprocessing facilities on IMS observations

Part 1. Methodology

SNF reprocessing plant:

- European Nuclear Fuel Reprocessing Plants from La Hague in France

Main sources of information:

1. Orano Annual reports and website, La Hague operational reports, SFCOMPO 2.0 database [3], scientific publications, etc.

Extracted data:

1. Number of non-operational day, such as maintenance, equipment replacement, paned checks of operation capabilities, of separate units, etc., during the period 2018 - 2022
2. Cm-244 and Pu-240 content in PWR and BWR SNF UO2 fuel with 45 GWdIt burnup, enrichment 3.7- 4.5% 235U (SFCOMPO 2.0, 2017)

Assumptions:

1. The selection of the fuel samples from SFCOMPO data base was done based on the fuel parameters
2. 5 years of cooling after uploading before reprocessing

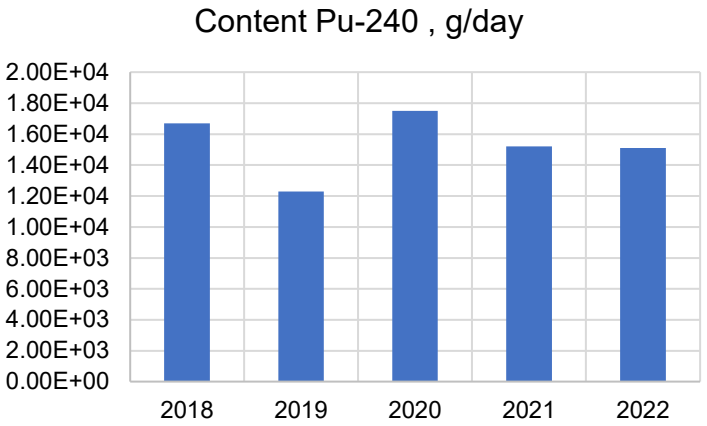
[3] SFCOMPO 2.0 (2017). Spent Fuel Isotopic Composition. OECD, Nuclear Energy Agency. Available at: https://www.oecd-nea.org/jcms/pl_21515/sfcompo-2-0-spent-fuel-isotopic-composition,

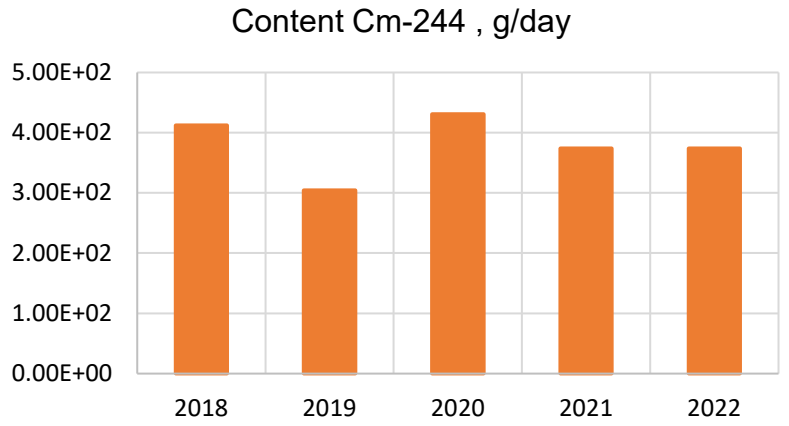
Part 1. Intermediate Results

- 12 samples of PWR and BWR SNF was selected from SFCOMPO
- The approximate deviation of the enrichment and burn-up of the chosen samples from the reference did not exceed 4,5-8%.
- The average content of the Cm-244 and Pu-240 in the SNF totalled 0.06 mg/gUi and 2,24 mg/gUi, respectively.

Parameters	Years of operation, years				
	2018	2019	2020	2021	2022
Total number of working days, days	252	251	253	254	253
Total number of non-operational days, days	104	60	120	103	90
Total number of operational day, days	148	221	133	151	163
Amount of SNF reprocessed (dissolved) per year, tHM	1100	1213	1035	1021	1100
Amount of fuel reprocessed (dissolved) per day, tHM/day	5,489	5,489	7,782	6,762	6,748

Content of isotopes in the SNF reprocessed per day





Part 1. Methodology.

Calculations of radioxenon and radioiodine isotopes from Spontaneous fission of Cm-244 and Pu-240

The calculation of radioxenon and radioiodine isotopes release per day from Pu-240 and Cm-244 present in the SNF after cooling were done using I-131, I-133, Xe-131, Xe-133m, Xe-133, Xe-135 cumulative yields and decay constants as well as Cm-244 and Pu-240 spontaneous fission rates.

Step1. The equation to calculate effective decay constants of ^{131}I, ^{133}I, ^{131}Xe, ^{133m}Xe, ^{133}Xe, ^{135}Xe:

$$R_{d.ef.} = Y_{cum.} \times \lambda_{s.f.},$$

- $R_{d.ef.}$ - effective decay constant of fission product (i.e. ^{131}I, ^{133}I, ^{131}Xe, ^{133m}Xe, ^{133}Xe, ^{135}Xe), 1/s;
- $Y_{cum.}$ - spontaneous fission cumulative yield of fission product (i.e. ^{131}I, ^{133}I, ^{131}Xe, ^{133m}Xe, ^{133}Xe, ^{135}Xe), $\frac{\%}{100}$;
- $\lambda_{s.f.}$ - lambda spontaneous fission, 1/s.

Step 2. The equation to calculate the activity of radioiodine and radioxenon released from Cm-244 and Pu-240 spontaneous fission:

$$R_{dacay} = (C \times R_{d.ef.}) / (M \times N_A),$$

- R_{dacay} - decay rate of the fission product (in secular equilibrium), Bq/day;
- C - Cm-244 or Pu-240 content in the SNF after 10 years of cooling, g/day; $R_{d.ef.}$ - effective decay constant of fission product (i.e. ^{131}I, ^{133}I, ^{131}Xe, ^{133m}Xe, ^{133}Xe, ^{135}Xe), 1/s;
- M - molar mass of Cm-244 or Pu-240 isotopes, g/mol;
- N_A - Avogadro's number $6{,}022 \cdot 10^{23}$ atoms/mol.

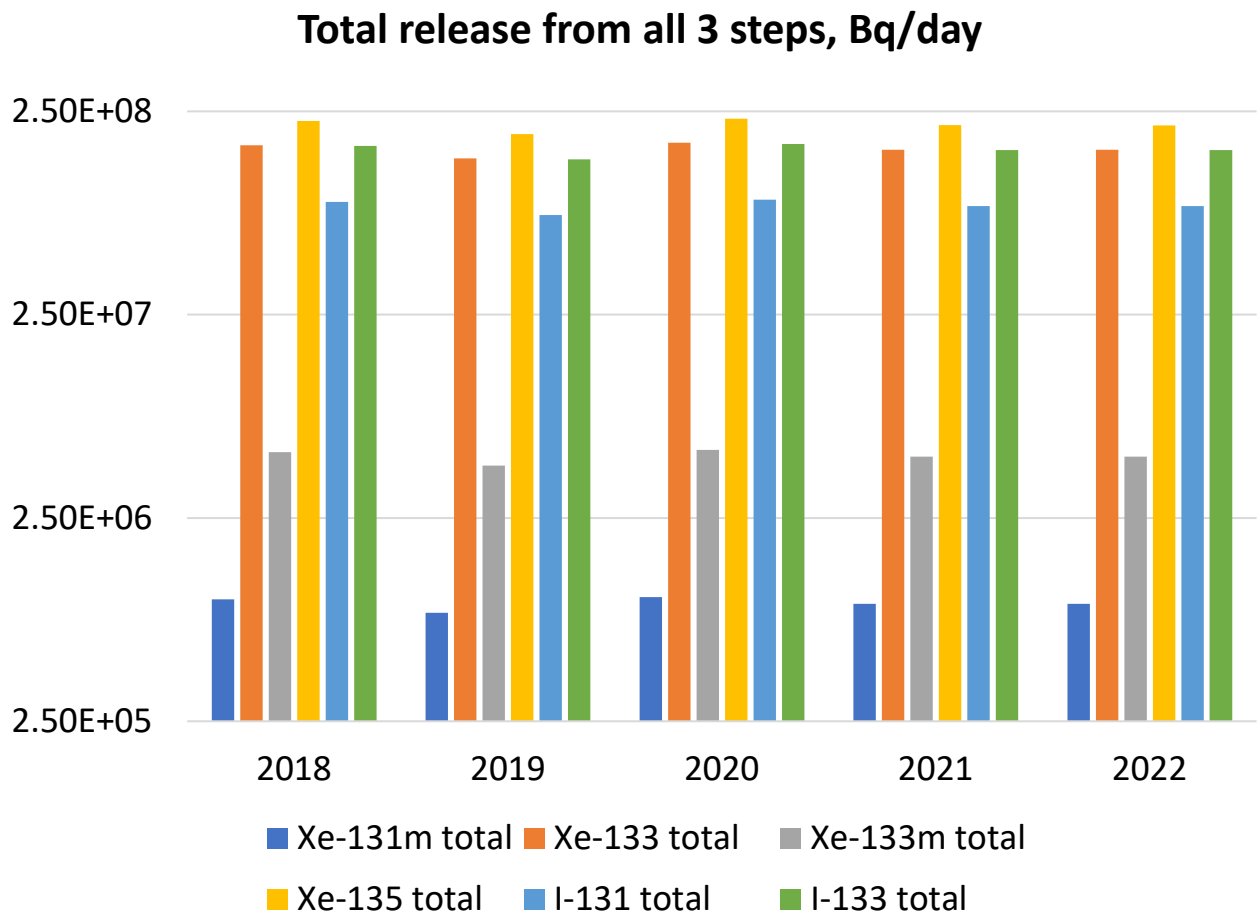
Part 1. Results

The following steps of reprocessing were researched:

1. Dissolution
2. First extraction cycle (PUREX process)
3. HALW vitrification

Assumption made for calculations:

1. Dissolution (all generated radioxenon and radioiodine released immediately and fully)
2. First extraction cycle (PUREX process) (calculation take in consideration the process parameters such as feed solution composition: U ~ 250 g/l, [HNO3] - 4,5 mol/l, Feed solution flow rate - 637 l/h, Organic phase flow rate - 1272 l/h, dissolution factor of dissolved SNF solution - 1,4)
3. HALW vitrification (1 canister -16h, 1,8 tU)



Part 2. Methodology

The source of data of I-131 and I-133 releases:

- European Commission RAdioactive Discharges Database (RAAA). Radioactive discharges [4].

SNF reprocessing plants:

- European Nuclear Fuel Reprocessing Plants from La Hague in France
- Sellafield in the United Kingdom.

[4] RADD – the European Commission RAdioactive Discharges Database https://europa.eu/radd/

Extracted data:

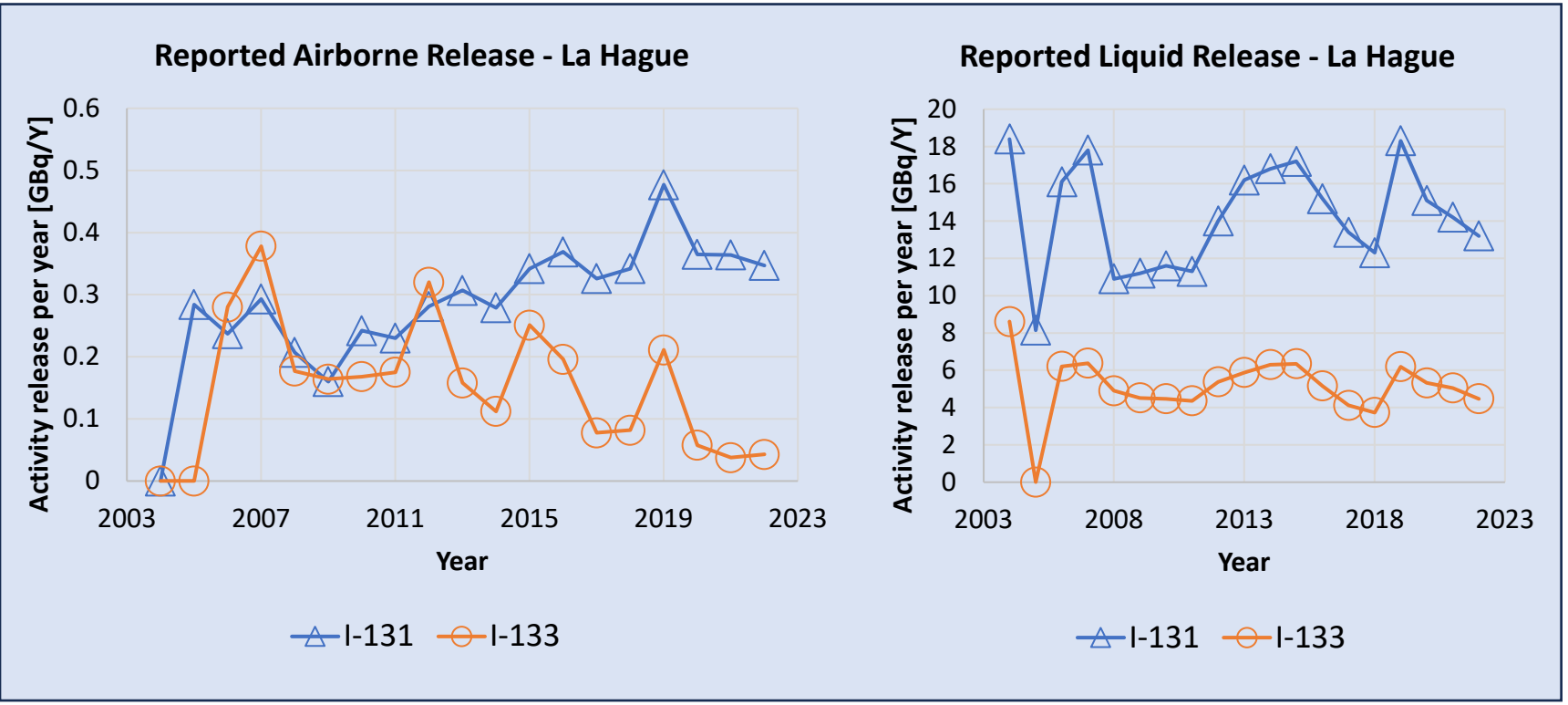
- airborne and liquid discharge data,
- 1993 – 2023
- Discharge values are expressed in GBq/year.

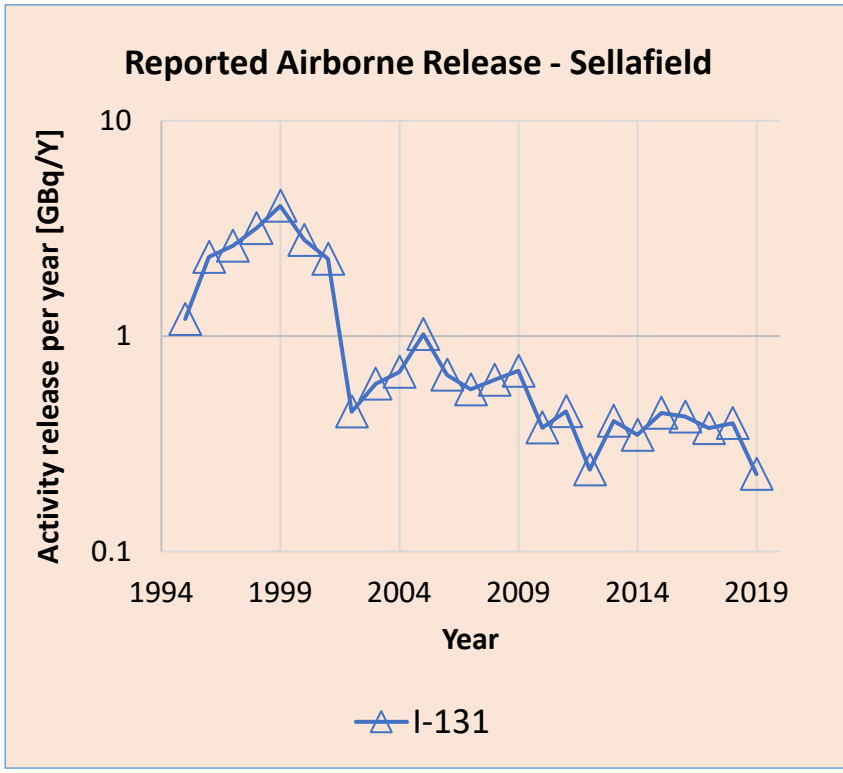
Assumptions:

1. The isotopic activity ratio of I-131 and I -133 releases was used to determine the delay since fuel dissolution and decay correction was applied.
2. Decay corrected I-131 inventories were used as a proxy for the estimation of radioxenon releases

Part 2. Results

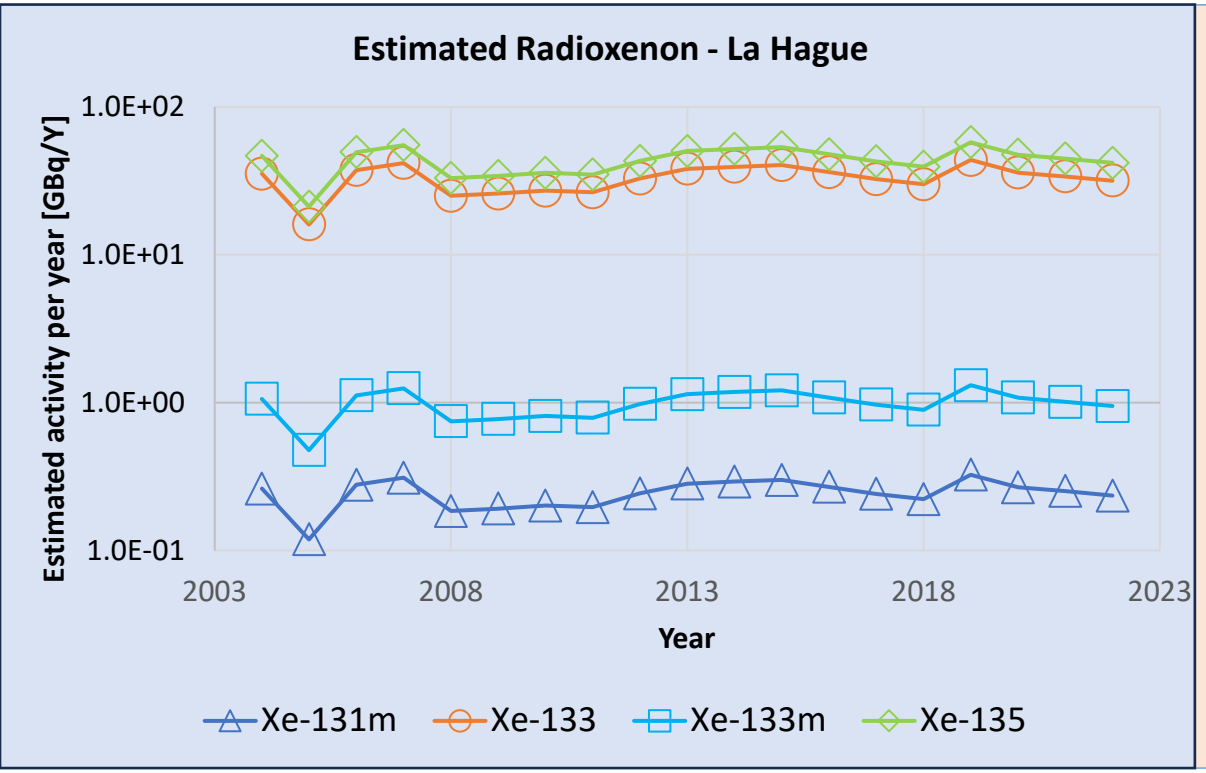
Timeline airborne and liquid inventory of I-131 and I-133 from La Hague and Sellafield Nuclear Fuel Reprocessing Plant according to RADD database.

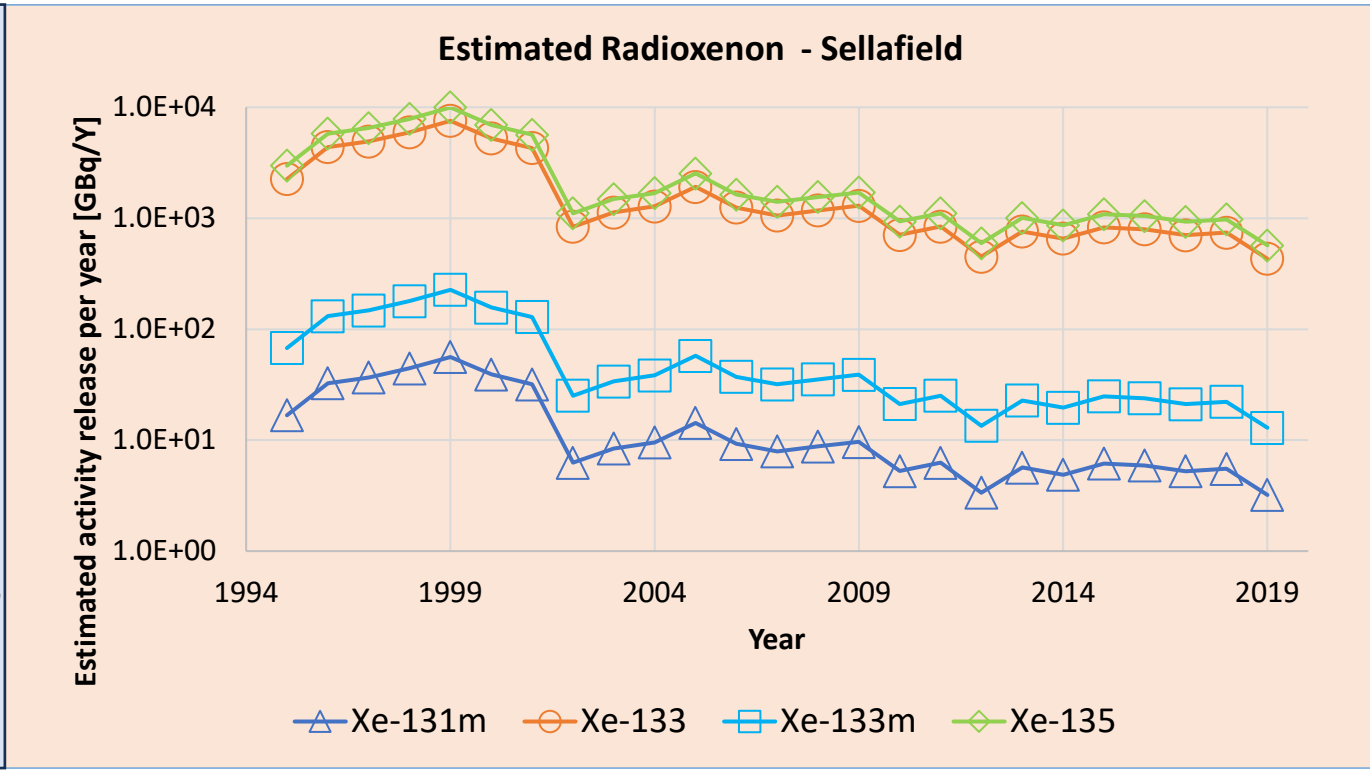




Part 2. Results

Timeline radioxenon emission estimates from La Hague and Sellafield Nuclear Fuel Reprocessing Plant using I-131 reports in the RADD database as proxy.





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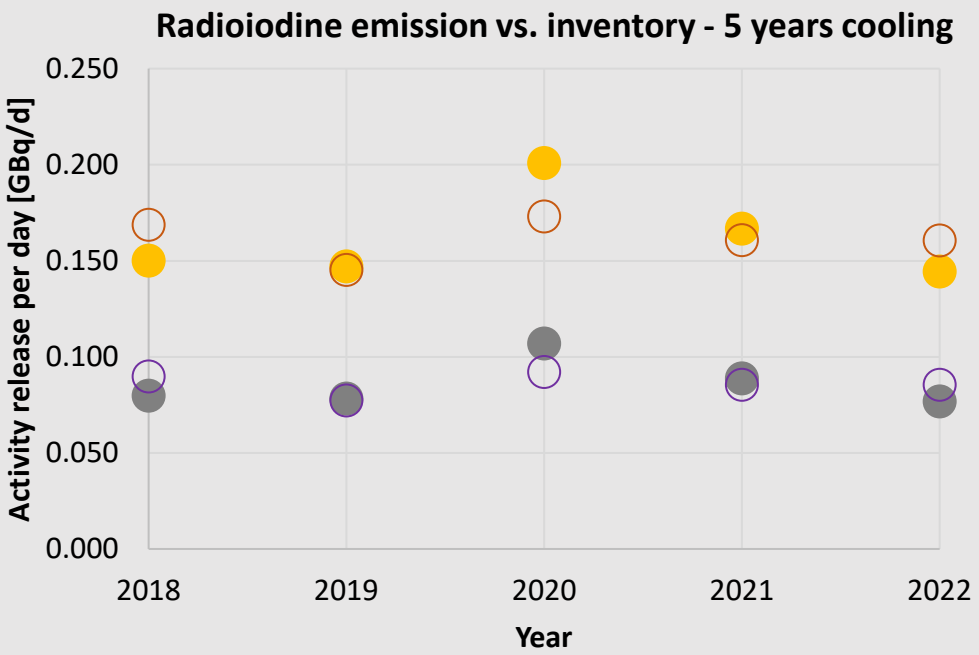
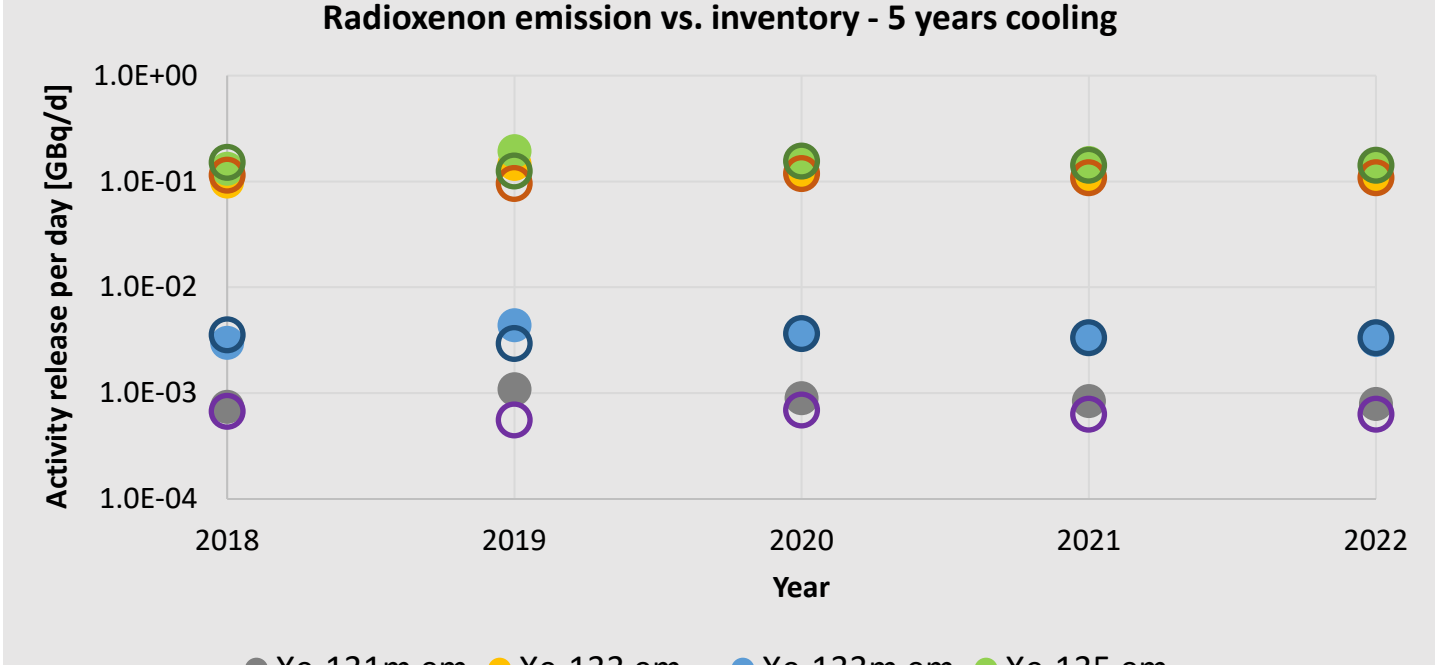
Comparison of radioxenon and radioiodine releases based on SNF inventory and RADD data

Comparison of the radioxenon and radioiodine calculated by 2 approaches. In average over the five years, the results based on Cm-244 and Pu-240 inventories were underestimated by a factor of 1.34. This is a good match with small variations over the 5 years. Validation successful.





Part 3. Methodology

- The operational ATM system deployed and used at CTBTO [5,6] is based on the Lagrangian Particle Dispersion Model FLEXPART [7] driven by the global meteorological fields provided by the European Centre for Medium-Range Weather Forecasts (ECMWF) and the US National Centres for Environmental Prediction (NCEP).
- The ATM system is used in a backward mode to compute SRS (Source-Receptor-Sensitivity) fields for each sample at all radionuclide measurement locations, for a period of 14 days.
- Simulations are done for air tracers and during post-processing are adjusted for a given radionuclide by applying the decay correction.
- To enable visualization of the ATM outputs, and consequently analysis of the radionuclide detections observed at the IMS stations from the angle of their possible origin, the Web-connected graphics engine (WEB-GRAPE) software has been developed.
- The continuously emitting sources (CES) functionality was used to predict the measured concentrations for a set of stations and dates.
- Input data: set of sources, source location, individual daily release rates for a certain time range, the continuously emitting sources functionality.
- Reprocessing facilities are assumed to be hypothetical sources of emissions, working continuously.

[5] Wotawa, G., et al. (2003). Atmospheric transport modelling in support of CTBT verification – Overview and basic concepts. Atmospheric Environment 37 (18) 2529-2537.
[6] Kuśmierczyk-Michulec, et al. (2021). "Advancements in atmospheric transport modelling (ATM) at the CTBTO PTS during the past two decades and plans for the future". (Presentation at the CTBT: Science and Technology Conference, Vienna, Austria 2021).
[7] FLEXible PARTicle dispersion model (FLEXPAT) Available at: http://flexpart.eu

Part 3. Results I

Radioactive measurements at IMS stations located nearby La Hague and Sellafield. Emission estimates are based on I-131 emission data published at RADD. The concentrations at IMS stations are estimated using the ATM backward simulations.

Reprocessing facility: La Hague					
IMS station	I-131 [Bq/m3]	Xe-131m [Bq/m3]	Xe-133 [Bq/m3]	Xe-133m [Bq/m3]	Xe-135 [Bq/m3]
RN33	4.76E-09	5.35E-09	5.93E-07	1.22E-08	5.99E-08
RN63	3.21E-09	6.09E-09	5.13E-07	8.09E-09	3.46E-08
RN49	5.02E-10	1.36E-09	1.18E-07	1.25E-09	1.98E-10
RN61*	1.08E-09	NA	NA	NA	NA
RN34*	1.51E-09	NA	NA	NA	NA

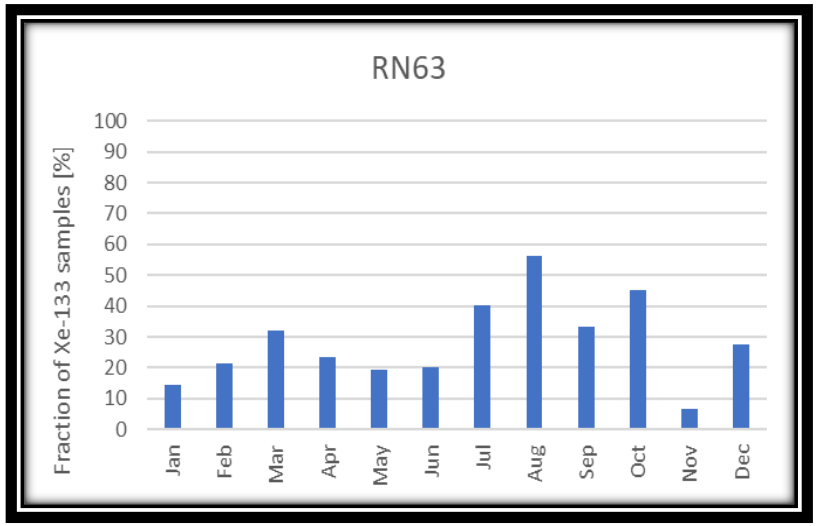
Reprocessing facility: Sellafield					
IMS station	I-131 [Bq/m3]	Xe-131m [Bq/m3]	Xe-133 [Bq/m3]	Xe-133m [Bq/m3]	Xe-135 [Bq/m3]
RN33	1.22E-09	4.21E-08	4.65E-06	8.51E-08	3.90E-07
RN63	1.62E-09	1.14E-07	1.31E-05	2.70E-07	6.85E-07
RN49	3.71E-10	4.54E-08	3.96E-06	4.39E-08	1.02E-08
RN61*	7.49E-10	NA	NA	NA	NA
RN34*	1.23E-09	NA	NA	NA	NA

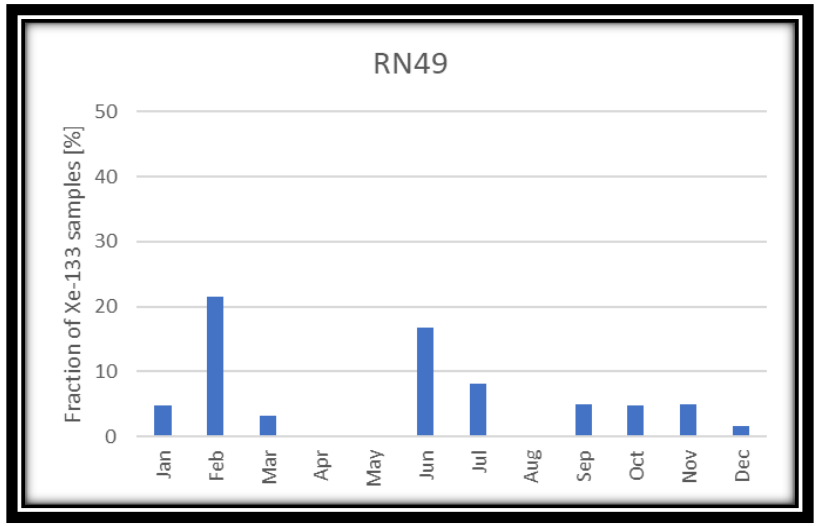
The samples with simulated activity concentration above 1E-07 Bq/m3 during a given month could potentially contribute to the IMS detections (marked green).

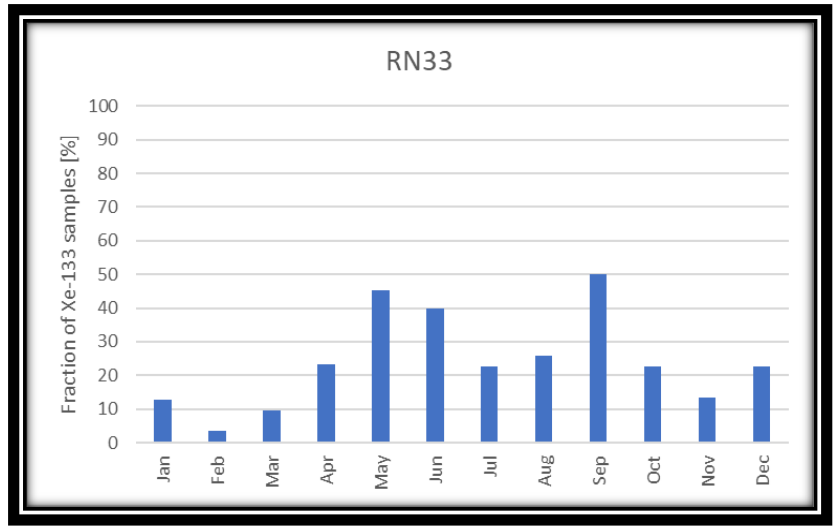
Part 3. Results II

To demonstrate the effect of atmospheric circulation, the reprocessing facility in Sellafield was selected.

- Fraction of Xe-133 samples in a given month that could be potentially influenced by the emissions from the reprocessing facility in Sellafield.
- Collection time is 12 hours for RN63 and RN49.
- Collection time is 24hours for RN33.







Summary 1/2

- Spent nuclear fuel (SNF) reprocessing plants were not previously considered of relevance to nuclear explosion monitoring.
- CTBT-relevant radioxenon isotopes are generated by spontaneous fission and released during dissolution of fuel, extraction and vitrification.
- These emissions are not reported. To estimate them, two independent methods are applied:
  - Determine radioxenon inventories from the SNF inventories processed per day.
  - Use annual I-131 emissions as proxy.
- Both methods compare well for the 5 years studied. This validates the I-131 proxy method.

Result

- The radioxenon emission inventory for Sellafield (1995-2019) and La Hague (2004-2022) is determined for all years with reported I-131 emissions.

Summary 2/2

- Using the estimated releases of 2014, the continuously emitting sources (CES) functionality was used to predict the measured concentrations for closest IMS NG systems.
- The fraction of emission releases from reprocessing facilities being transported to the IMS stations depends on the distance between the source and the station, on the strength of the source and on the atmospheric circulation.
- The ATM simulation showed that Xe-133 emitted from Sellafield is most likely to contribute to IMS detections observed at RN33, RN63 and RN49 and the samples with simulated activity concentration above 1E-07 Bq/m3 during a given month could potentially contribute to the IMS detections.

Conclusion

- Though SNF reprocessing facilities are a comparatively weak source and only few facilities of this type exist, they cannot be neglected in nuclear explosion monitoring.

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Vienna International Centre, PO Box 1200
1400 Vienna, Austria
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