

Contribution of all nuclear research reactors to the global radionuclide emission inventory

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Introduction

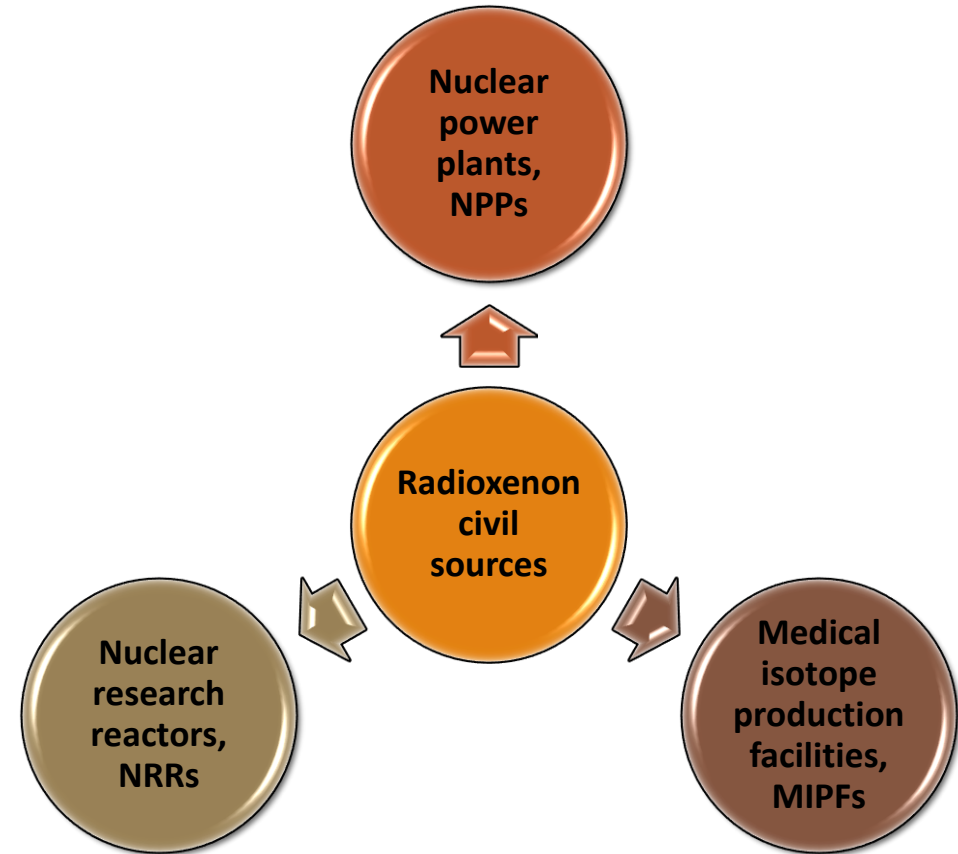
Introduction

Annual release (Bq/y) in 2014

Facility	^{131m} Xe	¹³³ Xe	^{133m} Xe	¹³⁵ Xe	Ref
174 NPPs' sites	3.9E+13	6.7E+14	1.5E+13	4.8E14	1

Facility	^{131m} Xe	¹³³ Xe	^{133m} Xe	¹³⁵ Xe	Ref
7 MIPFs	5.4E+13	4.1E+16	1.4E+15	2.2E+15	2

Facility	^{131m} Xe	¹³³ Xe	^{133m} Xe	¹³⁵ Xe	Ref
160 NRRs	?	?	?	?	?



Overview of the world-wide fleet of NRRs

Reactor category	Thermal power range	Number of Reactors		
		31 Dec. 2013	22 Apr. 2018	23 Jan. 2019
Large	Higher than 1 MW	88	87	90
Medium	Between 1 kW and 1 MW	55	59	64
Small	Less than 1 kW	17	53	48
Total	Any	160	199	202

- Not included all NRRs with zero power

Annual radioxenon emissions (Bq/y) from NRRs (measured)

- Very few data are available on NRRs' emission

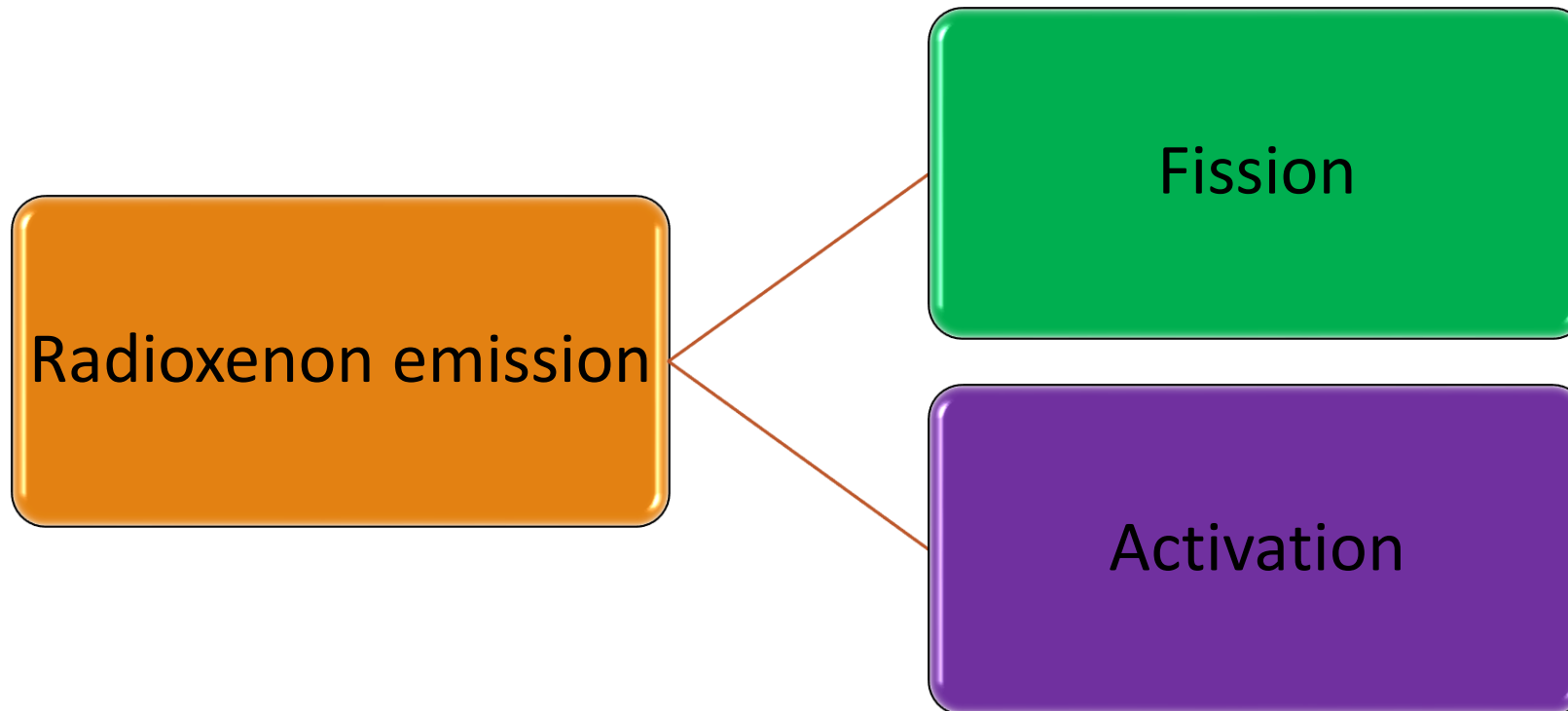
Reactor	Country	Type	Power (MW)	¹³⁵ Xe	^{133m} Xe	¹³³ Xe	^{131m} Xe	Note
HFIR	USA	Tank	85	1.07E+12	7.46E+11	1.92E+11	5.16E+12	Median of 10-year emission
HWPWR	USA		40			5.18E+09		
HANARO	Korea	Pool	30	5.63E+09	6.12E+08	3.00E+10	1.64E+08	
FRM II	Germany	Pool	20	8.32E+08	7.24E+07	1.20E+09	7.82E+06	
OPAL	Australia	Pool	20	2.78E+10		5.60E+09		
RA3	Argentina	Pool	10	1.68E+09		6.86E+08		Median of 10-year emission
NETL TRIGA Mark II	Texas	Homog (s)	1.1	3.16E+07	3.57E+06	1.40E+07	9.44E+05	
TRIGA Mark II	Vienna	Pool	0.250	5.49E+06	1.40E+05	2.68E+06	3.29E+04	

Operational modes and capacity factors (CF) of NRRs

- The key parameters in production rate are the **power** of the reactor and the **capacity factor**.
- NRRs have a large range of different operational working schedules from a few hours to several days that make the production and release patterns versatile. The working hours of all the research reactors can be found in the IAEA Research Reactor Database RRDB [3]. With that database, one can calculate the capacity factor, i.e. **the fraction of time that the power is up during a year**.

Facility Name	Hours per Day	Days per Week	Weeks per Year	Hours per Year	Capacity factor (%)
RA-3	24	5	46	5520	63.00
OPAL	24	7	52	8736	100
TRIGA II VIENNA	7	5	48	1680	19.20
FRM II	24	7	34	5712	65.20
HANARO	24	7	28	4704	53.71
TRIGA II UNIV. TEXAS	6	5	45	1350	15.40
HWPWR					13.20

Radioxenon generation in NRRs



- For most reactors, one of the two production processes is the dominating source of the emissions.



Contribution of *fission* to
radioxenon production

Scaling the radioxenon fission production rate by thermal power

The birth rate in terms of generation of activity inventory per time interval for a certain fission product depends on the power of the reactor. This can be calculated from the following equation:

- B (Bq/y): Fission birth rate
- λ (s^{-1}): Decay constant of the radioisotope of interest
- Y : Cumulative fission yield of the radioisotope of interest
- CF : Capacity factor
- P (MW): Thermal power of the reactor
- E_f (MeV): Typical fission energy release deposited in the reactor per fission event, it is about 200 MeV
- $1.6E-19$: Factor that converts MeV to MJ
- $3.1536E+07$: Number of seconds per year

$$B = \lambda \times \frac{P}{E_f \times 1.6 \times 10^{-19}} \times Y \times CF \times 3.1536 \times 10^7$$

Scaling radioactive noble gas emissions from production rate

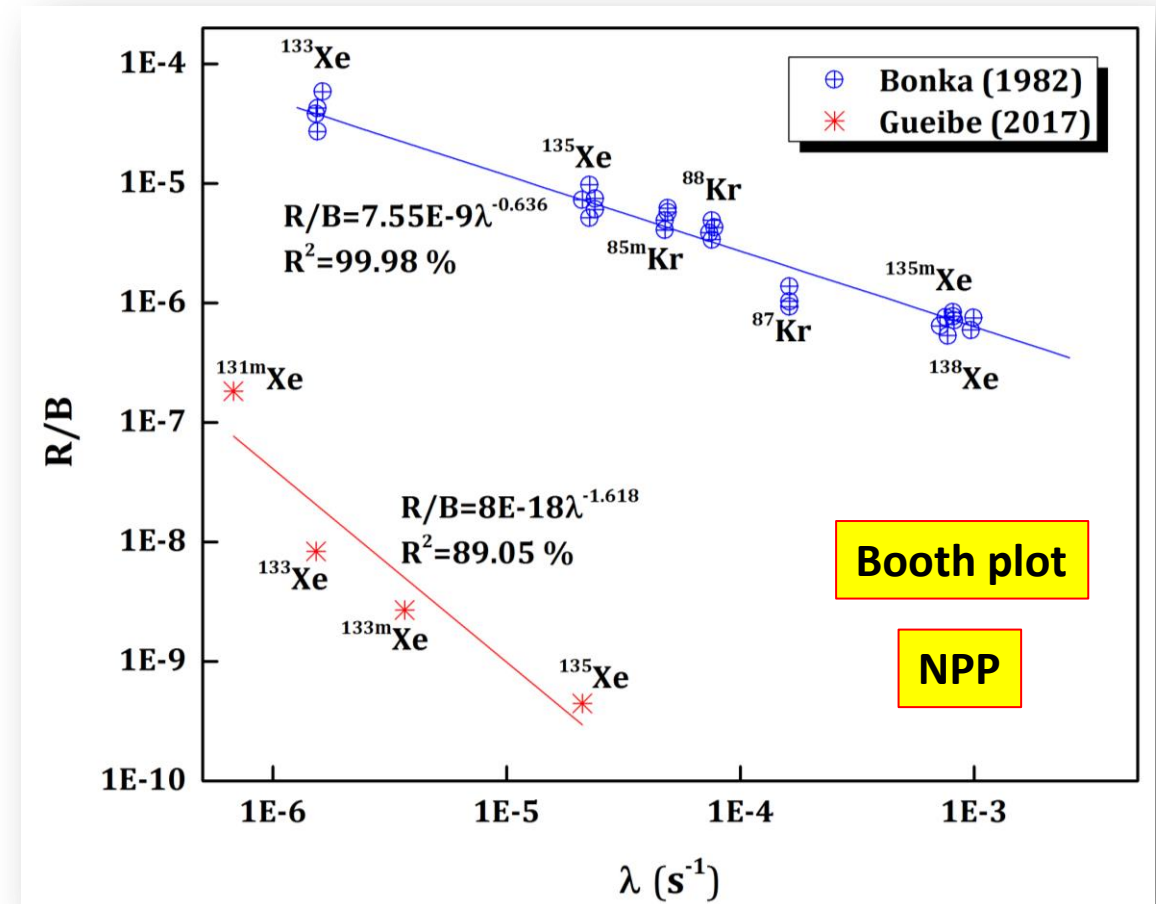
Booth equation

The release fractions are divided by B depend on the half life.

- R (Bq/y): release rate
- B (Bq/y): birth rate
- k: emission rate constant
- λ (s^{-1}): decay constant
- α : exponential

$$\frac{R}{B} = k \cdot \lambda^{-\alpha}$$

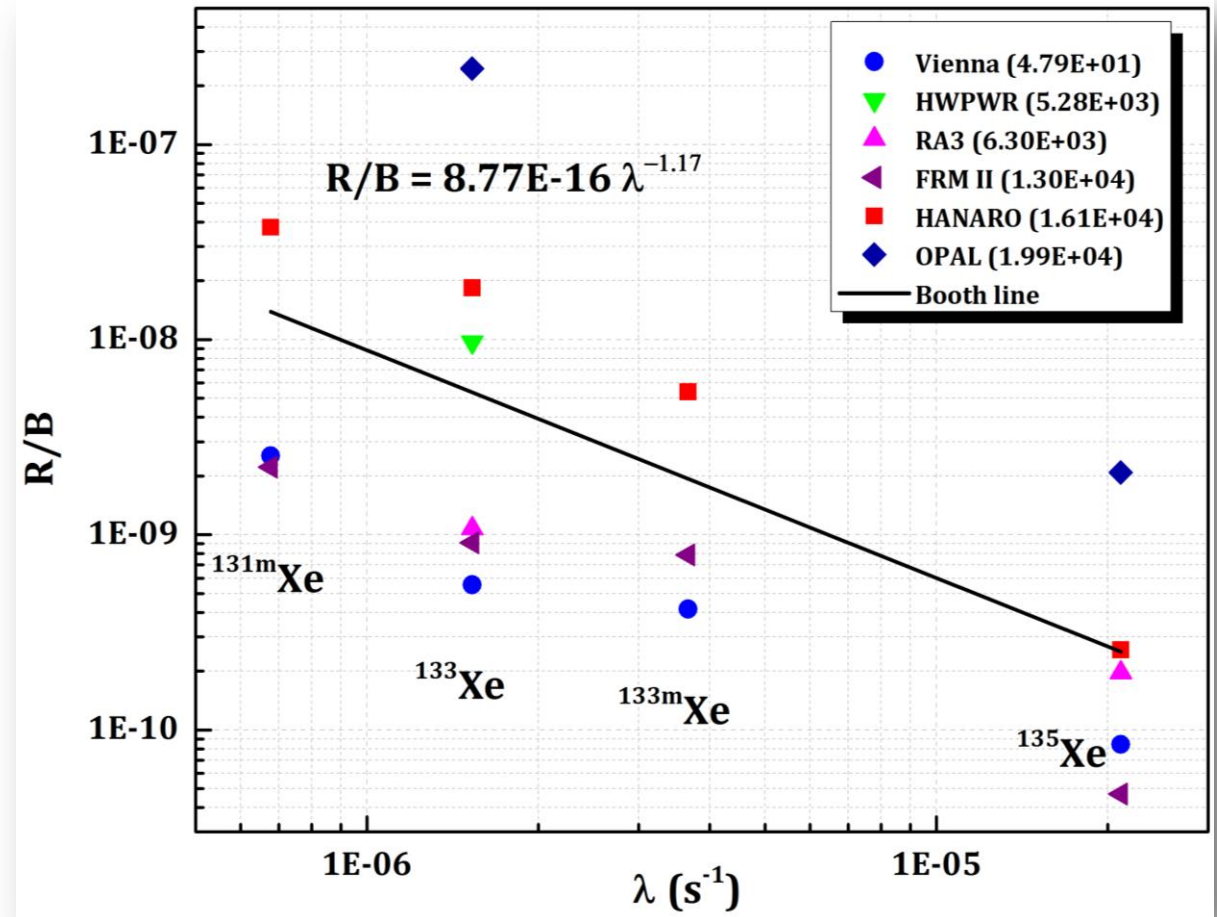
- A.H. Booth, Chalk river, 1957



Booth plot for NRRs

- Each reactor agrees well with the slope of Booth line but spreading over almost 3 orders of magnitude.
- The release data from **FRM II**, **HANARO** and **TRIGA Mark II Vienna** reactors are available for all four isotopes and can therefore be used to obtain the parameter α .
- The parameter k is calculated from ^{133}Xe release data of **RA3**, **HWPWR**, **OPAL**, **HANARO**, **FRM II** and **TRIGA Mark II Vienna**.

$$\frac{R}{B} = 8.77 \times 10^{-16} \cdot \lambda^{-1.17}$$



Contribution of *activation* to
radioxenon production

Scaling the radioxenon release rate originating from activation

TRIGA reactor at University of Texas at Austin

- Argon purge stack
 - Flow: 0.52 m³/s
 - ¹³³Xe: 2.22E+03 mBq/m³
 - ¹³³Xe: 1.15E+03 mBq/s
- HVAC stack
 - Flow: 3.4 m³/s
 - ¹³³Xe: 5.08E+02 mBq/m³
 - ¹³³Xe: 1.73E+03 mBq/s



¹³³Xe = 2.88E+03 mBq/s

CF = 15.4 %

P = 950 kW

¹³³Xe = 1.09E+01 Bq/kWh

HFIR in Oak Ridge National Laboratory

- 10-year median release rate of ¹³³Xe = 1.92E+11 Bq/y
- CF = 46.0 %
- P = 85000 kW

¹³³Xe = 5.59E+02 Bq/kWh

SF for ¹³³Xe = 2.85E+02 Bq/kWh

Scaling the radioxenon release rate originating from activation

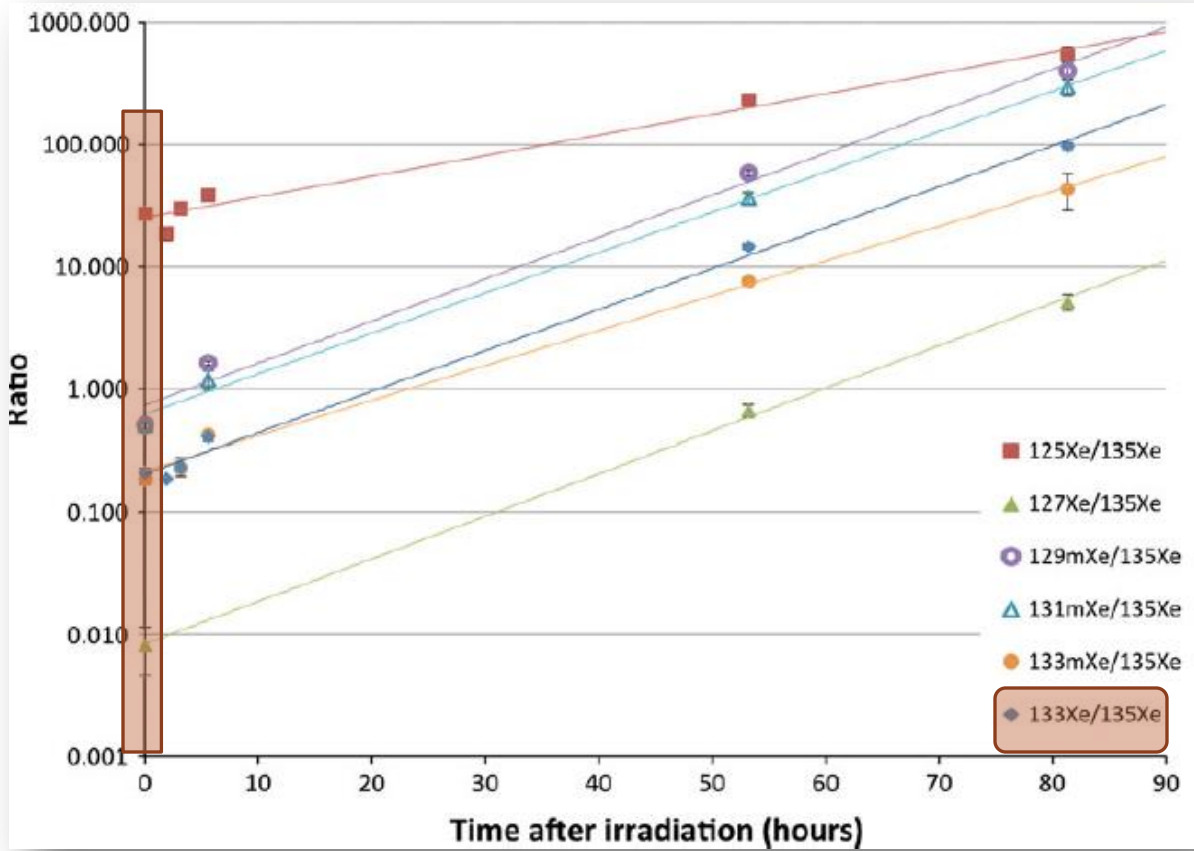


Fig. 3 Xenon activity concentrations after irradiation of natural xenon gas with epithermal neutrons

The scaling factors for 3 other isotopes are derived based on the isotopic ratios from air activation and the scaling factor of ¹³³Xe.

SF for ¹³³Xe = 2.85E+02 Bq/kWh

Isotope	Scaling factor (Bq/kw.h)
¹³⁵ Xe	1.30E+03
^{133m} Xe	2.51E+02
^{131m} Xe	6.31E+02

Plot from ref. 4

Scaling the radionuclide release rate originating from activation

$$\text{Release rate (Bq/y)} = \text{CF} \times \text{SF} \times \text{P} \times 365 \times 24$$

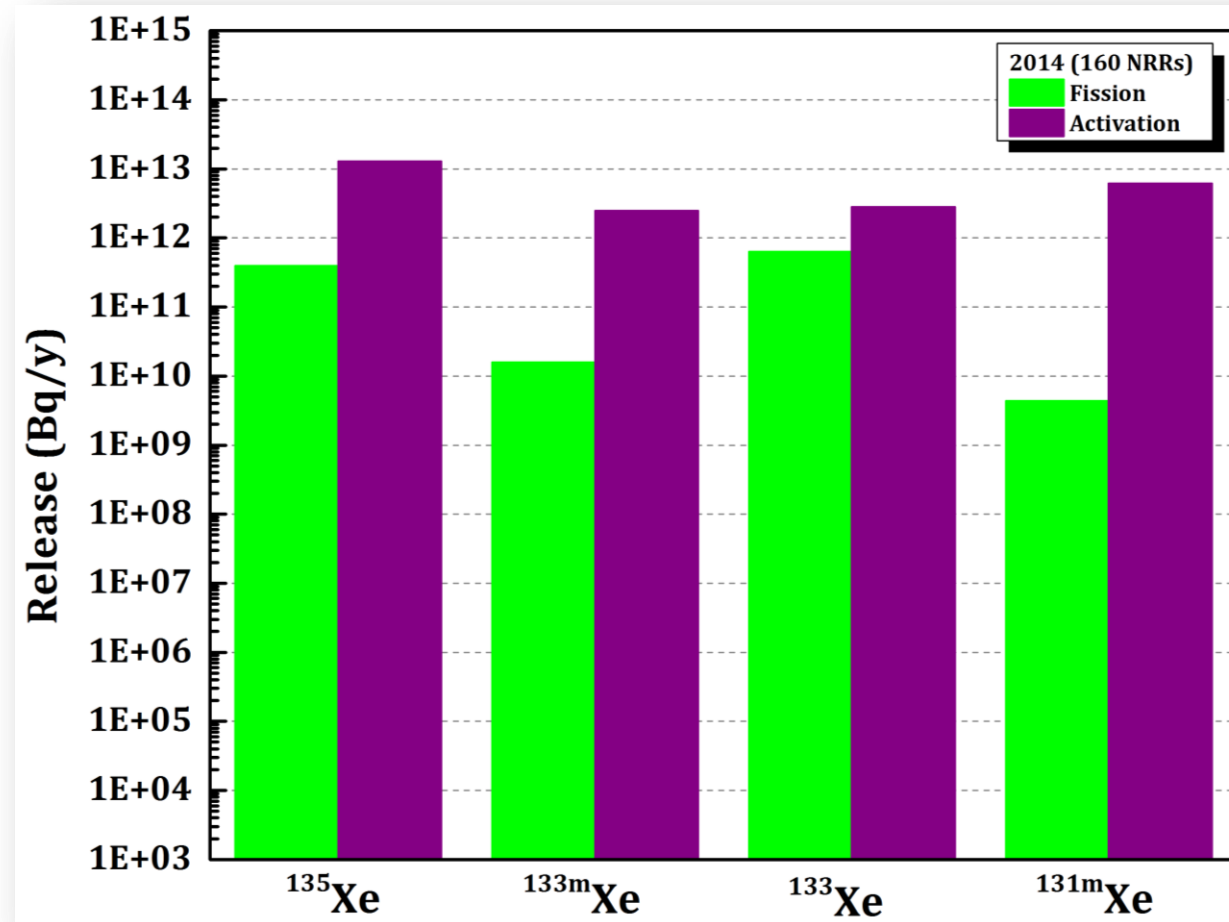
- CF: capacity factor
- SF (Bq/kW.h): scaling factor
- P (kW): power
- 365 × 24: Number of hours per year



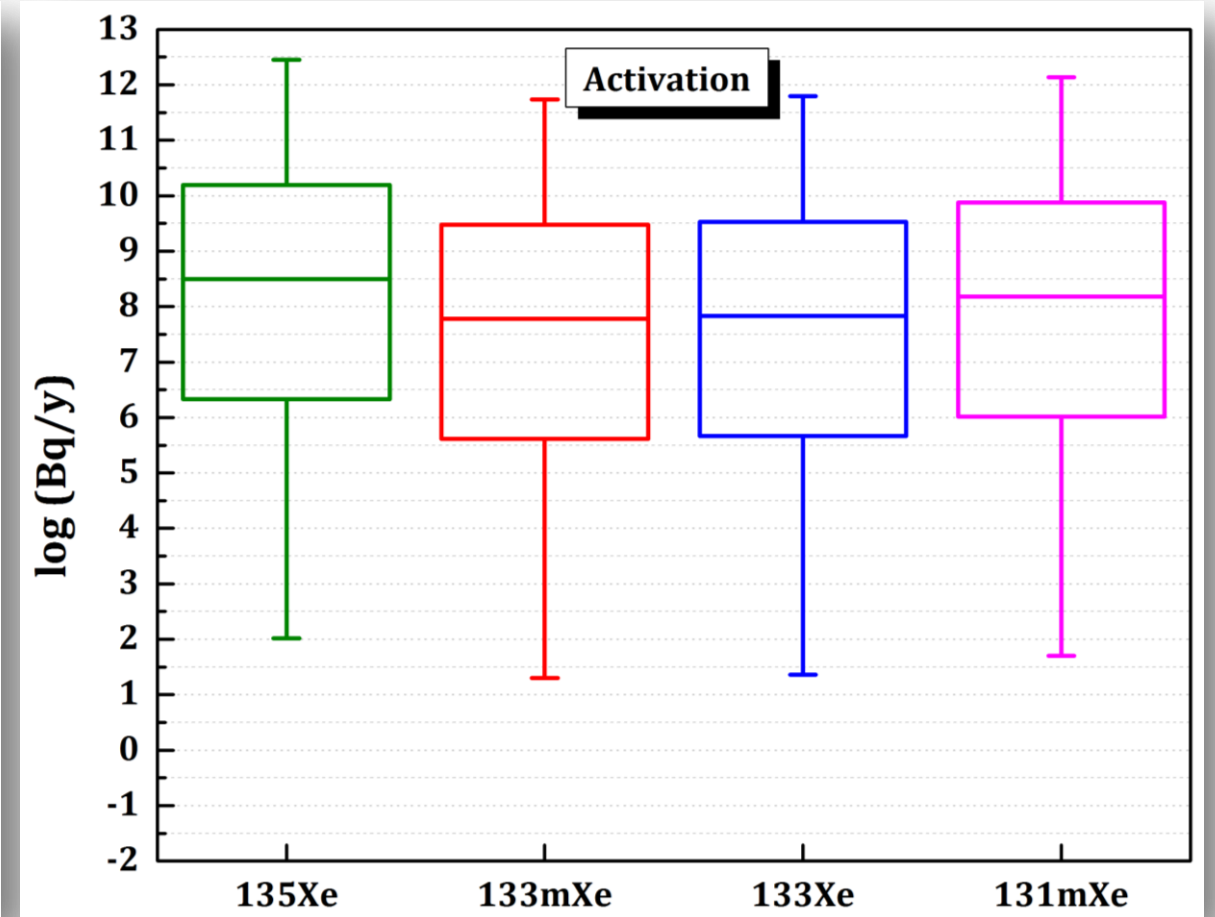
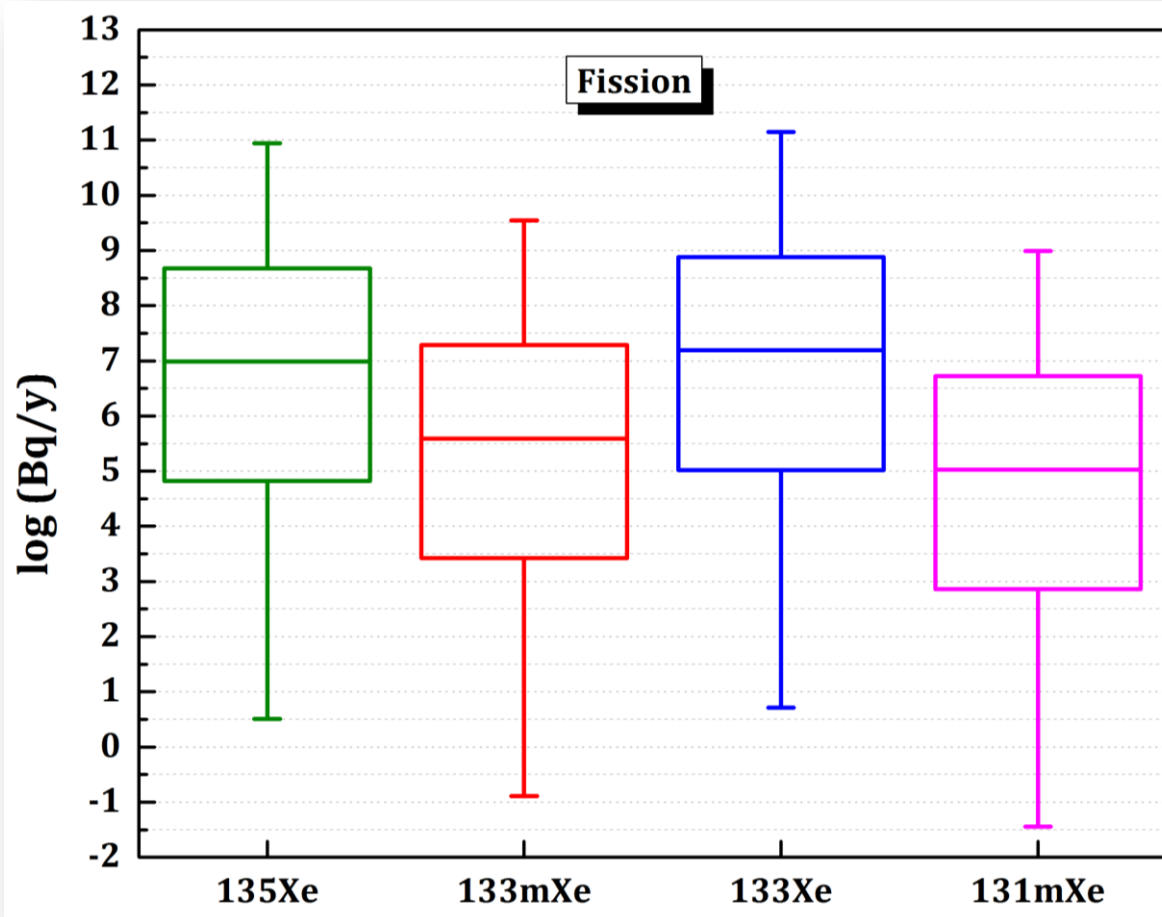
Results

Results

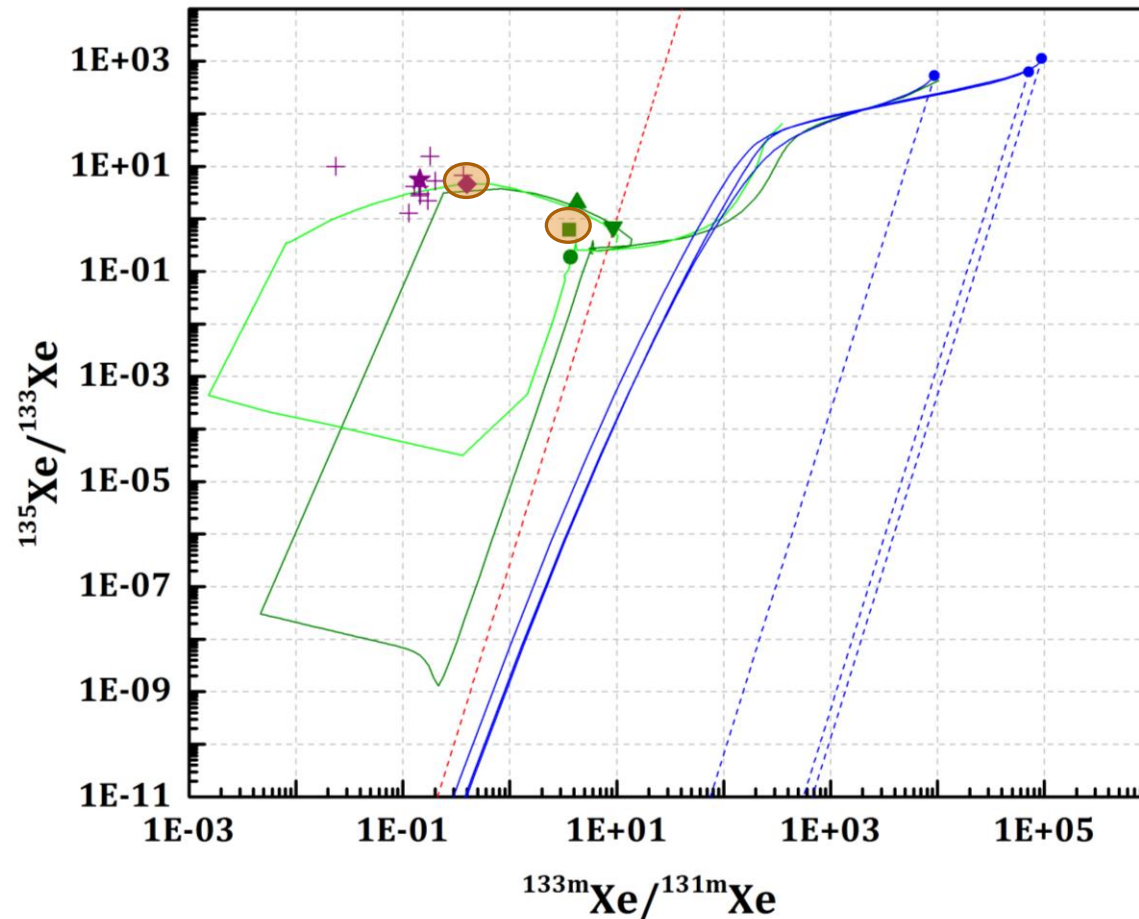
Annual release of NRRs (Bq/y) based on **fission** and **activation**



Comparison of releases based on **fission** with based on **activation**



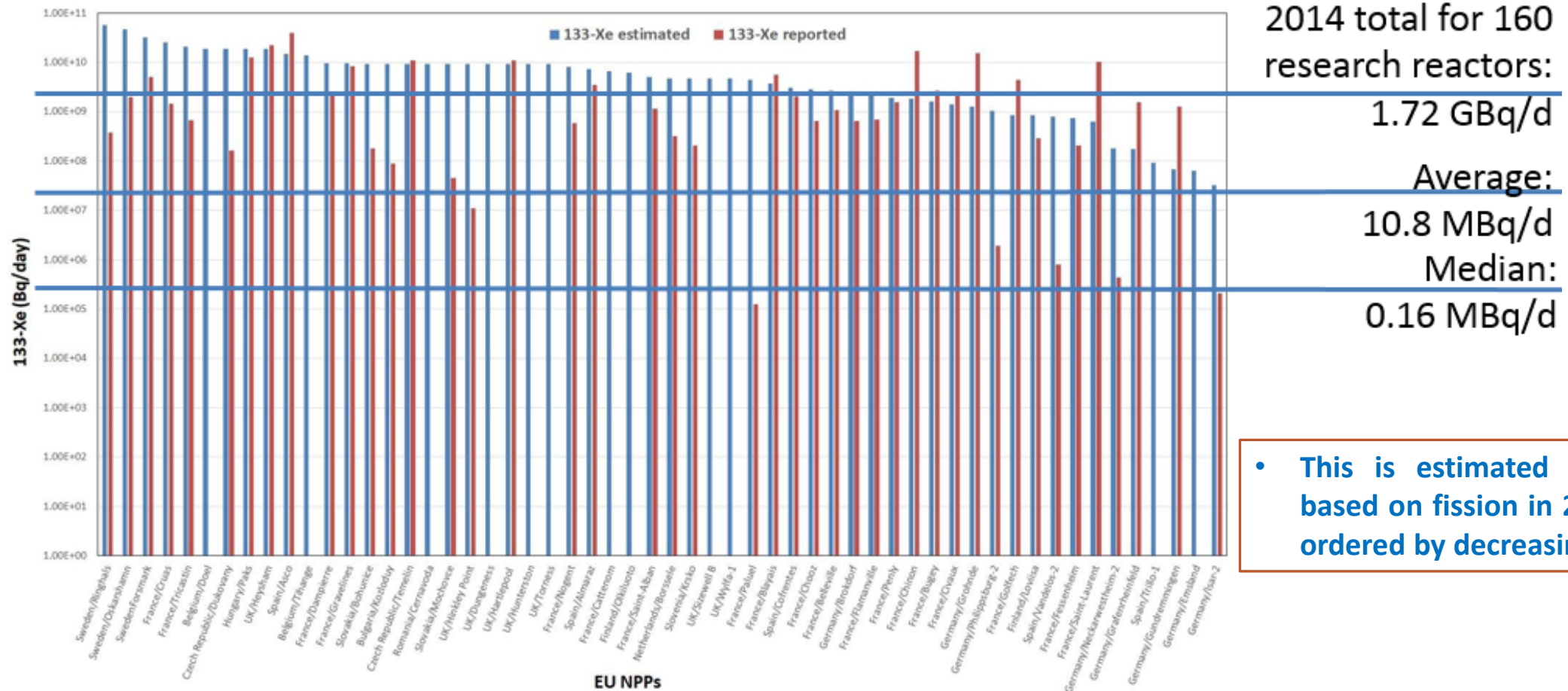
4-isotope plot



The 4-isotope plot shows a clear distinction between fission sources and activation sources.

- Tayyebi et al. (2017)
- Kalinowski et al. (2006)
- - - Separation line
- Booth
- ◆ Activation
- HANARO
- ▲ TRIGA II Vienna
- ▼ FRM II
- ★ HFIR (Median)
- + HFIR (10 years)
- Evolution of fission products in time with in-growth
- Fission of $^{235}\text{f}\text{U}$, $^{239}\text{f}\text{Pu}$ and $^{238\text{he}}\text{U}$ at $t=0$
- - - Evolution of fission products for xenon separated at $t=0$

^{133}Xe emission of NRRs in comparison with NPPs



Conclusions

Conclusions

- In this study, the contribution of all operational NRRs in global emission of radioxenon is estimated according to the few available experimental data and using the Booth plot based on fission. The activation contribution in radioxenon releases is also estimated.
- If activation is dominated in ^{133}Xe releases from the NRR, it seems to be an order of magnitude higher than the fission releases. The difference is even stronger for the other isotopes.
- According to this estimate, the world's entire research reactor fleet releases ^{133}Xe in the order of magnitude of $5\text{E}+09$ Bq/day (“reasonable radioxenon releases” according to Bowyer et al., 2013).
- In 2014 the total release of 160 NRRs together is still less than the release of many individual NPP.
- The distribution of source strengths for NRRs and NPPs are overlapping. There are 4 NRRs (in case of fission dominating) or 20 NRRs (if activation dominates) with a daily release >0.1 GBq/d. These are stronger sources than the 41 weakest emitters among NPPs. Therefore, NRRs cannot be ignored as a source.

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Thank you for your attention