



# Evolution of traceable Radon emanation sources from MBq to few Bq

by

Stefan Röttger, Annette Röttger, Florian Mertes  
on behalf of the traceRadon Consortium  
Coordinator: Annette Röttger (PTB)

Stefan Röttger

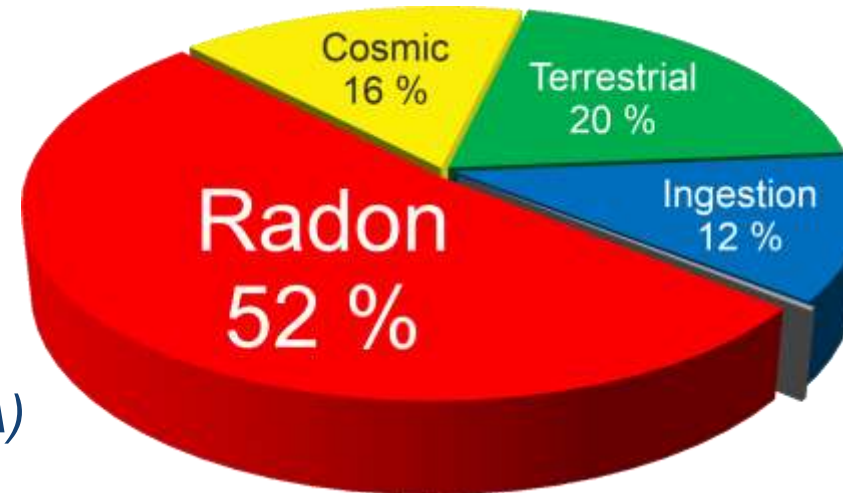
ICRM-LLRMT INFN-LNGS 2022-05-06



# $^{222}\text{Rn}$ Radon



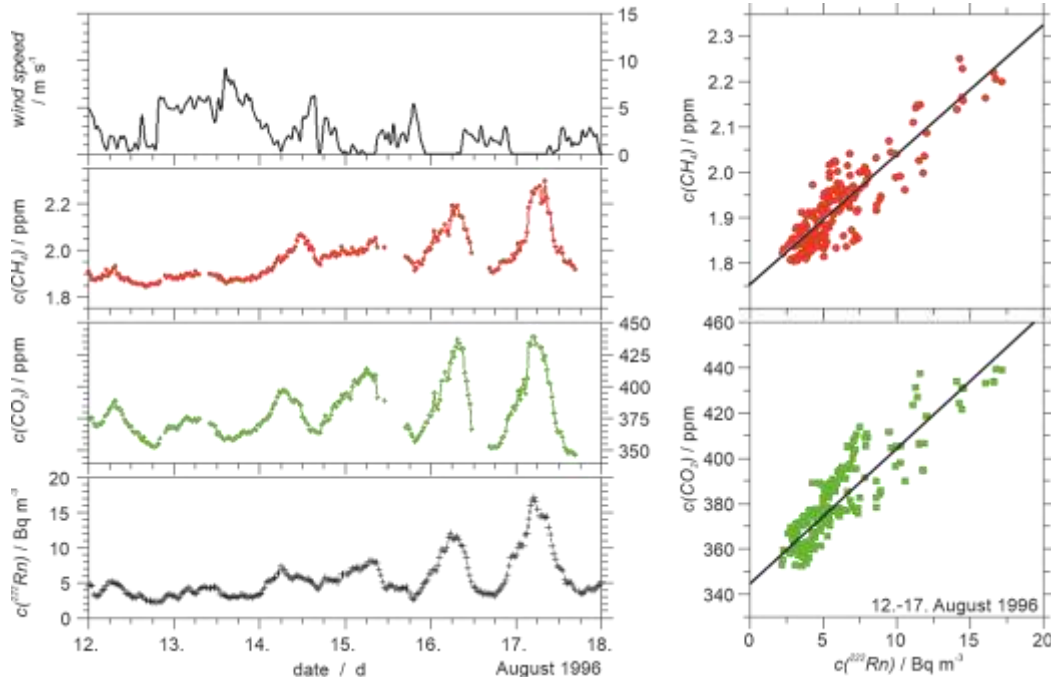
- $^{222}\text{Rn}$ : colourless and odourless radioactive noble gas ( $t_{1/2} = 3.8 \text{ d}$ ), naturally occurring in  $^{238}\text{U}$  decay chain
- Can be concentrated indoors
  - largest contribution to natural radiation exposure
  - important cause of lung cancer (e.g. Darby et. al. 2008)
- 2013/59 EURATOM *Basic safety standards*:
  - indoor activity concentration  $< 300 \text{ Bq}\cdot\text{m}^{-3}$  (1 atom out of  $10^{18}$  atoms)
  - identification of *Radon Priority Areas (RPA)*
- Outdoor air activity concentration  $(1 - 10) \text{ Bq}\cdot\text{m}^{-3}$



Exposure to natural radiation  
UNSCEAR 2008  
Total average individual dose:  
 $3 \text{ mSv}\cdot\text{a}^{-1}$



- $^{222}\text{Rn}$  is generated in the ground and takes part in atmospheric transport processes, but has only one well-defined sink (radioactive decay) because it is inert
  - Temporal / spatial distribution is determined by atmospheric transport
  - *Ideal proxy / tracer for modeling atmospheric processes*
  - Validation / improvement of transport models



## Radon Tracer Method (RTM):

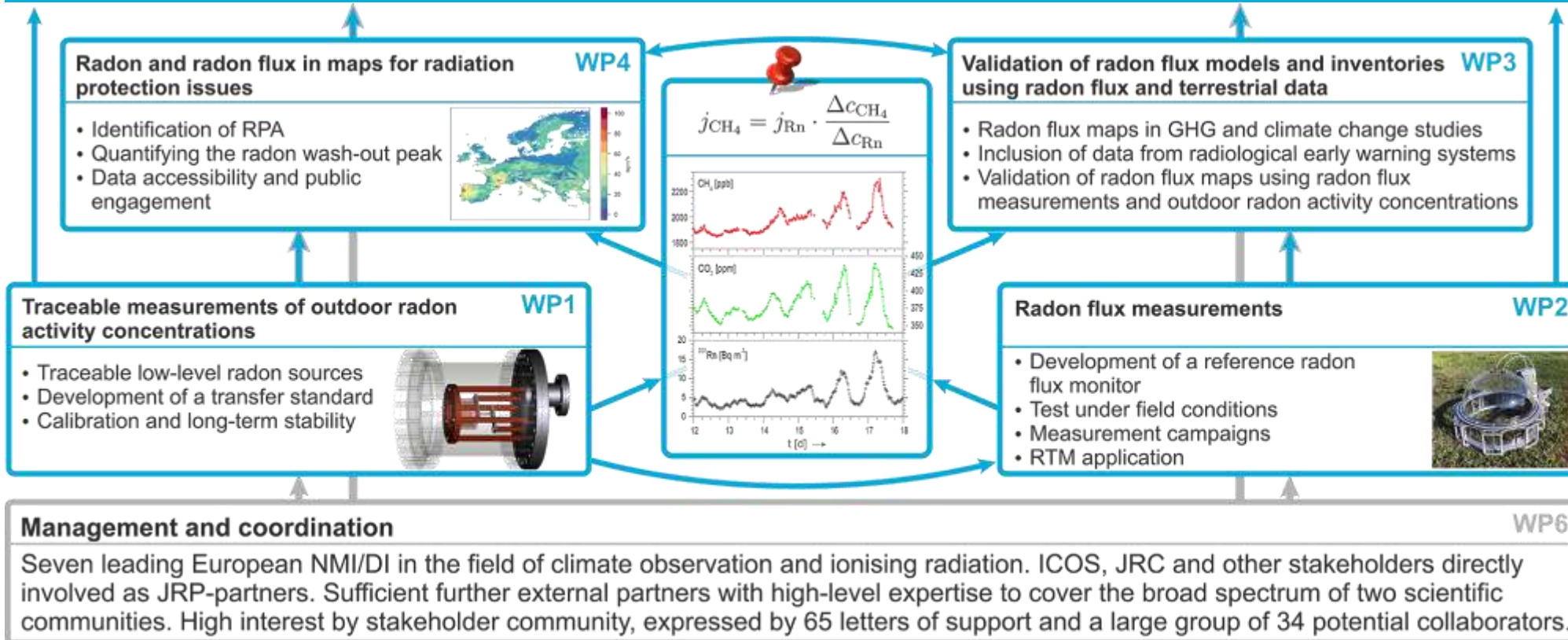
- The strength of the correlation allows the GHG fluxes to be estimated when the radon flux is known
- Comparability only with traceable calibration!

**EMPIR: traceRadon**

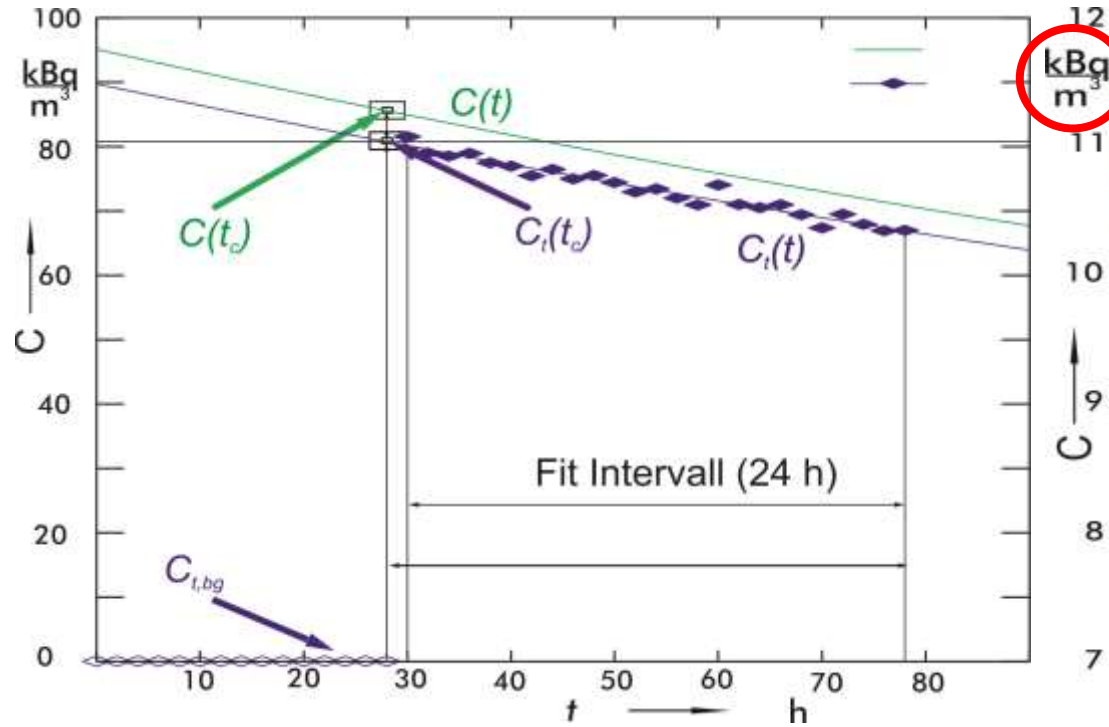


$^{222}\text{Rn}$  is the bridge between climate observation and radiation protection

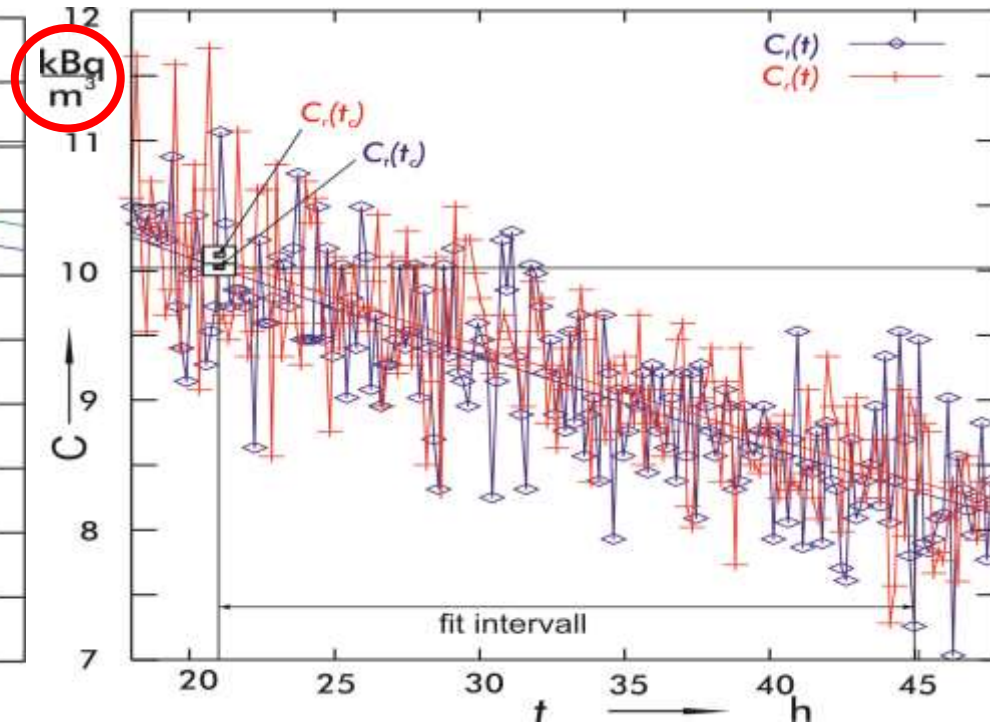
## Traceability to the SI system



# $^{222}\text{Rn}$ gas standard



$$C_r(t) = C_{r,ic} \cdot e^{(-\lambda t)} + C_{r,bg} \quad C_i(t) = C_{i,ic} \cdot e^{(-\lambda t)} + C_{i,bg} \quad k_i = \frac{k_r(C_r(t_c) - C_{r,bg})}{C_i(t_c) - C_{i,bg}}$$



$$C_r(t) = C_{r,ic} \cdot e^{(-\lambda t)} + C_{r,bg} \quad C_i(t) = C_{i,ic} \cdot e^{(-\lambda t)} + C_{i,bg} \quad k_i = \frac{k_r(C_r(t_c) - C_{r,bg})}{C_i(t_c) - C_{i,bg}}$$

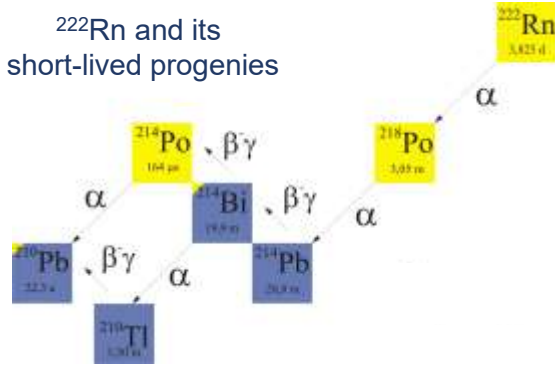
The representation of the activity concentration in the range of  $300 \text{ Bq} \cdot \text{m}^{-3}$  is a challenge to **metrology!**



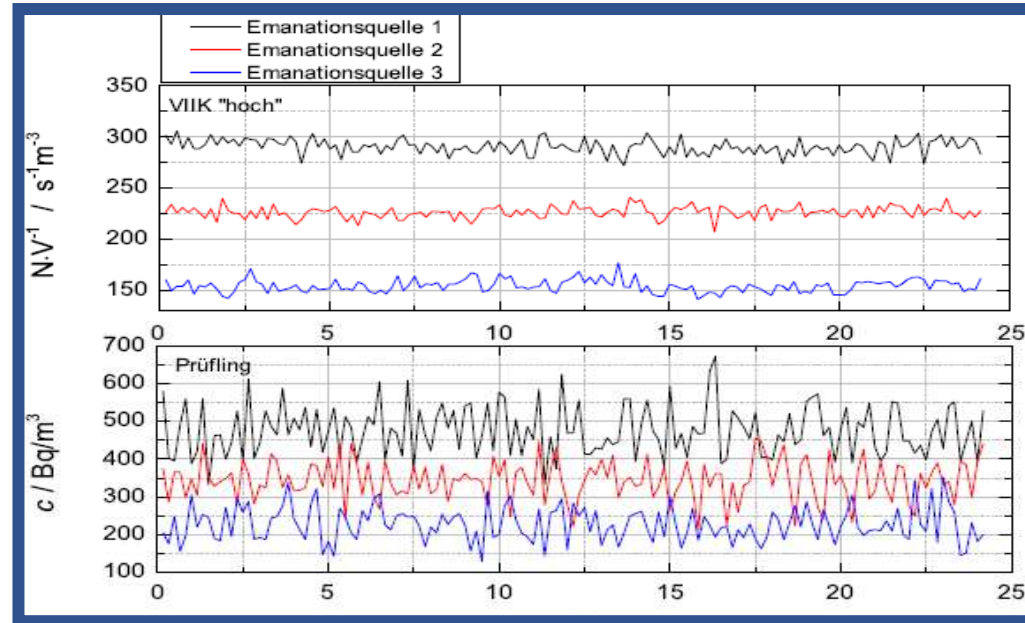
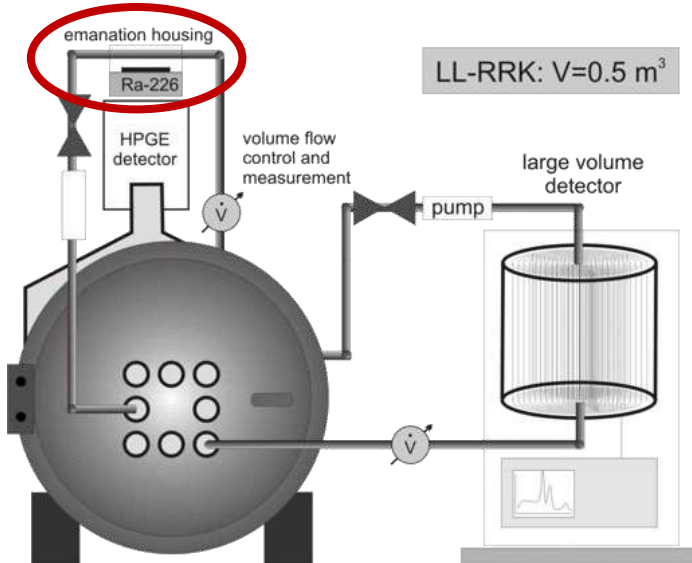
# Detector development



$^{222}\text{Rn}$  and its short-lived progenies

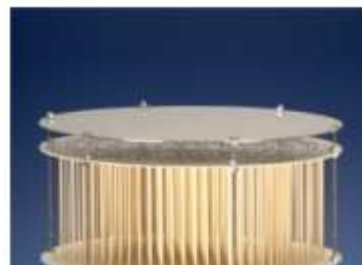


Preparation of EU-BSS by  
PhDs of Diana Linzmaier and Florian Mertes



## Low radon activity concentration measured precisely for the first time

Neues Messverfahren aus der PTB rechtzeitig zur Verschärfung der EU-Strahlenschutzrichtlinie



[PTB/es] Man sieht es nicht, man riecht es nicht, man schmeckt es nicht – aber es kann in hohen Dosen tödlich sein: Das natürliche radioaktive Edelgas Radon tritt vor allem dort aus dem Boden aus, wo der Untergrund aus Granit besteht. Es kann aber auch in Baumaterialien vorhanden sein. Dass Radon in hohen Dosen Lungenkrebs verursacht, ist längst bekannt – viele Arbeiter aus den Uran-Minen der Wismut-Werke der DDR sind daran gestorben. Inzwischen schätzen aber Wissenschaftler, dass Radon auch in niedrigen Konzentrationen eine Gefahr sein kann, und haben die Strahlenwirkung darum offiziell hochgestuft: Das Gas



# $^{222}\text{Rn}$ emanating sources (1)



## classical design

### Polyester-foil

Drop-cast  $^{226}\text{Ra}$   
wrapped in PE-foil

## Electrodeposited source

Deposition at  
 $30\text{ V} < U < 200\text{ V}$

## Implanted

Implantation of  $^{226}\text{Ra}$   
into W / Al after mass  
separation



# $^{222}\text{Rn}$ emanating sources (2)



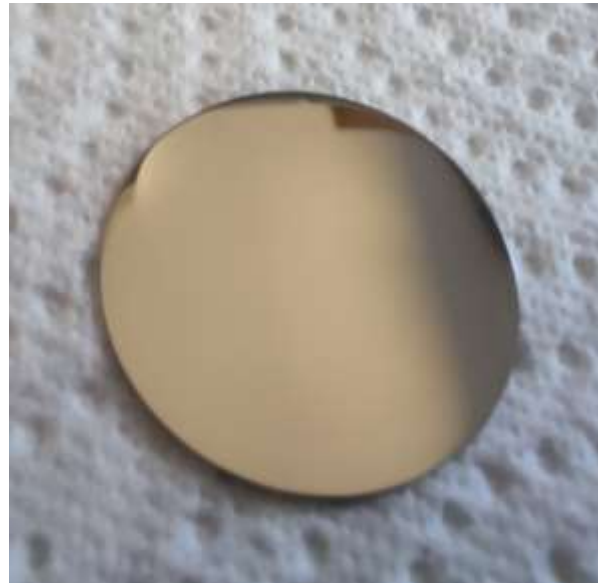
## stainless steel

deposition of  $^{226}\text{Ra}$  from  
 $^{226}\text{Ra}(\text{NO}_3)_2$  via tPVD



## p-type Si-wafer

deposition of  $^{226}\text{Ra}$  from  
 $^{226}\text{Ra}(\text{NO}_3)_2$  via tPVD



## PIPS 450 mm<sup>2</sup>

**300  $\mu\text{m}$  thickness**  
deposition of  $^{226}\text{Ra}$  from  
 $^{226}\text{Ra}(\text{NO}_3)_2$  via tPVD

**active source**



## Primary Characterisation of sources



### ${}^{226}\text{Ra}$ Activity:

- DSA  $\alpha$ -Spectrometry
- Autoradiography

Primary  ${}^{222}\text{Rn}$   
Source

### Emanation Power:

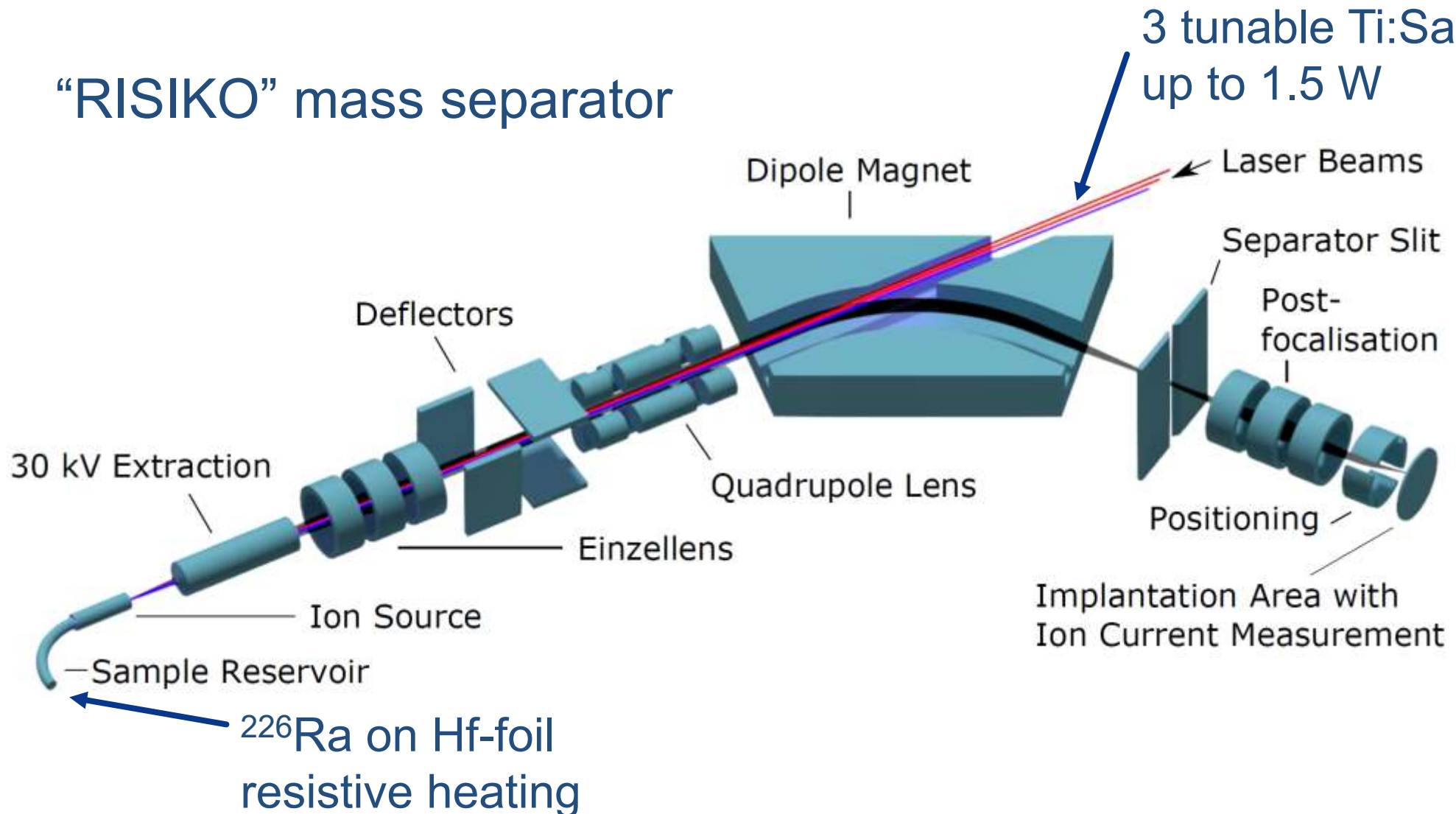
- $\gamma$ -Spectrometry (HPGe, LaBr<sub>3</sub>, CeBr<sub>3</sub>, Srl<sub>2</sub>)  
    ➔ Portable “on-line” measuring system
- Calibration of on-line system with enclosed / capsuled source of the same type and geometry



# Ion implanted sources



“RISIKO” mass separator

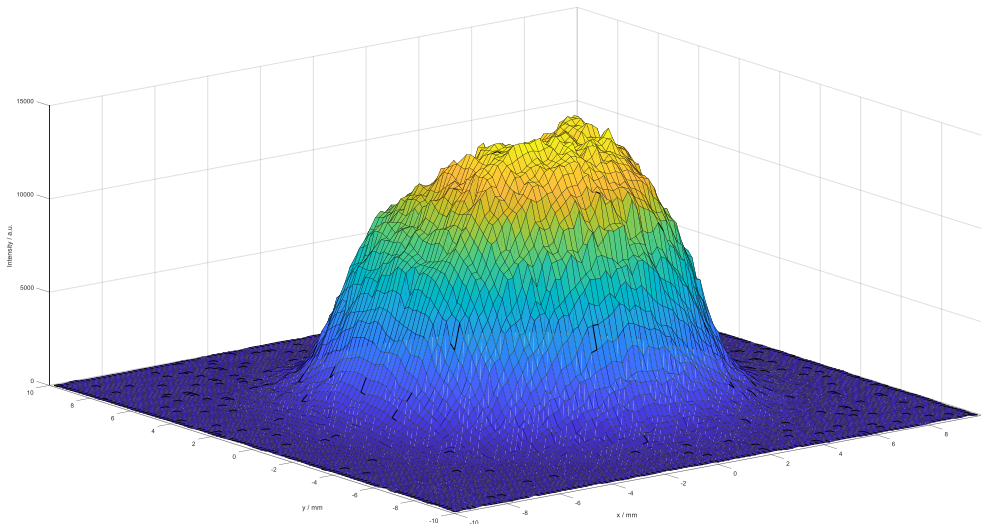


# Deposition vs implantation

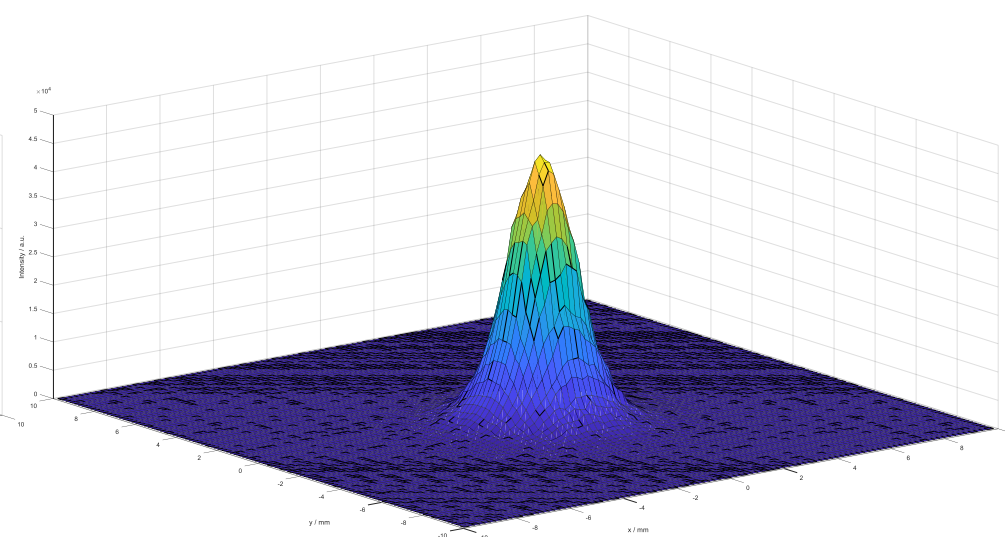
Implantation produces very defined distribution (3D-Gaussian)

➔ Beneficial for  $\alpha$ -spectrometry (FWHM, MC-calculations)

Electrodeposition



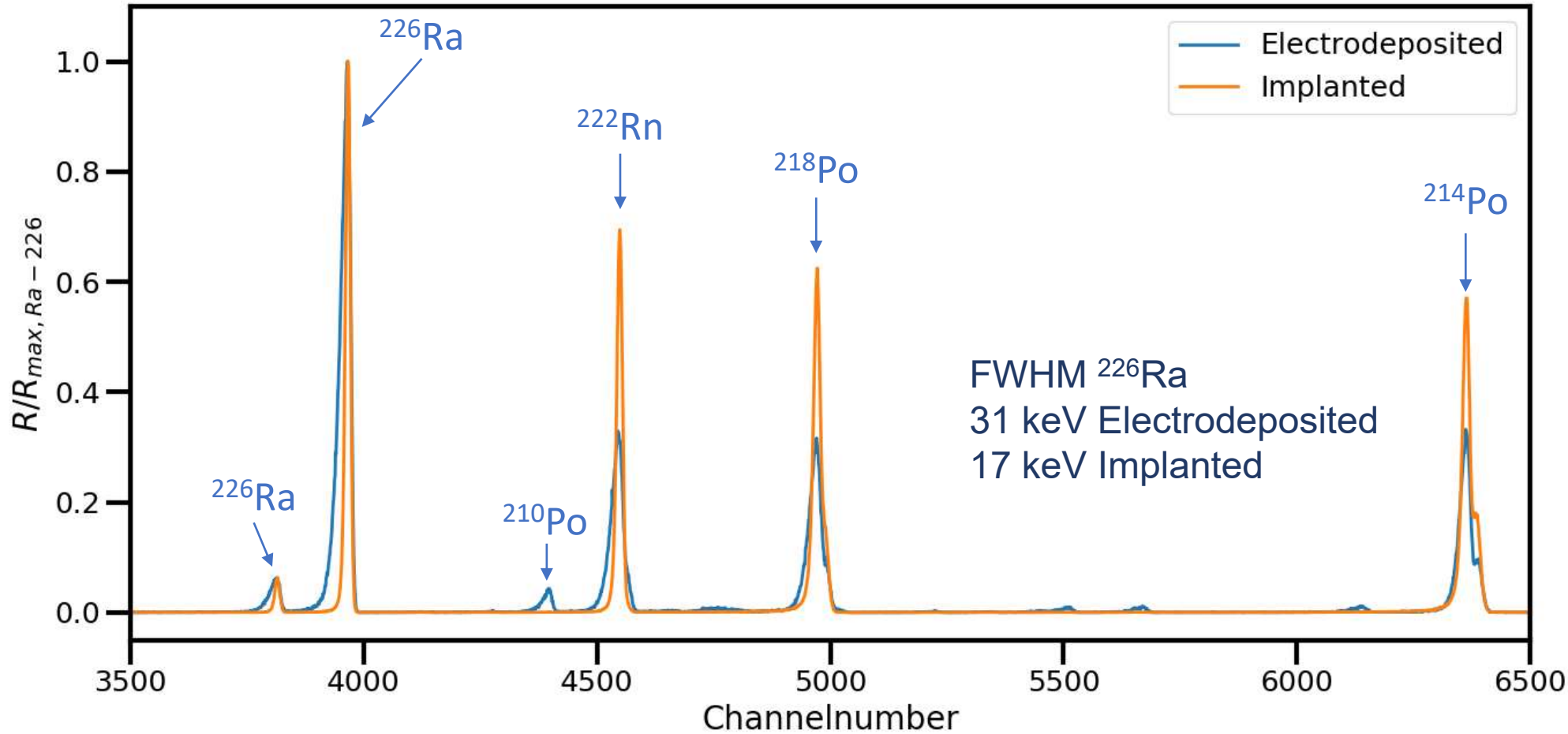
Implantation



# Deposition vs implantation



## $\alpha$ -spectra - comparison

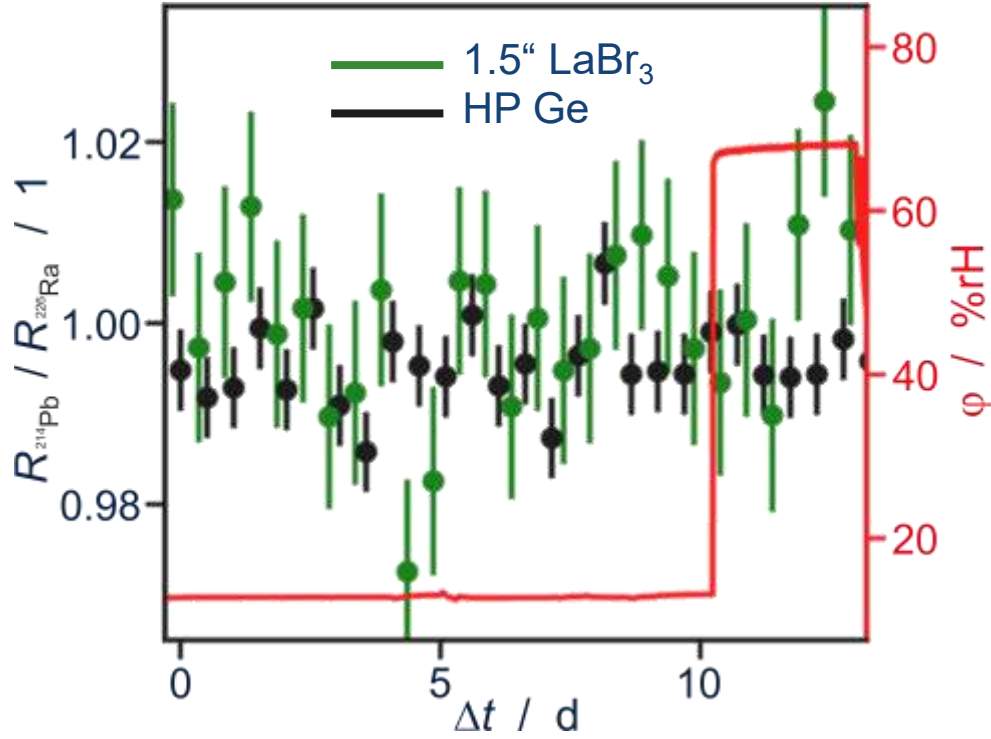


# environmental parameter

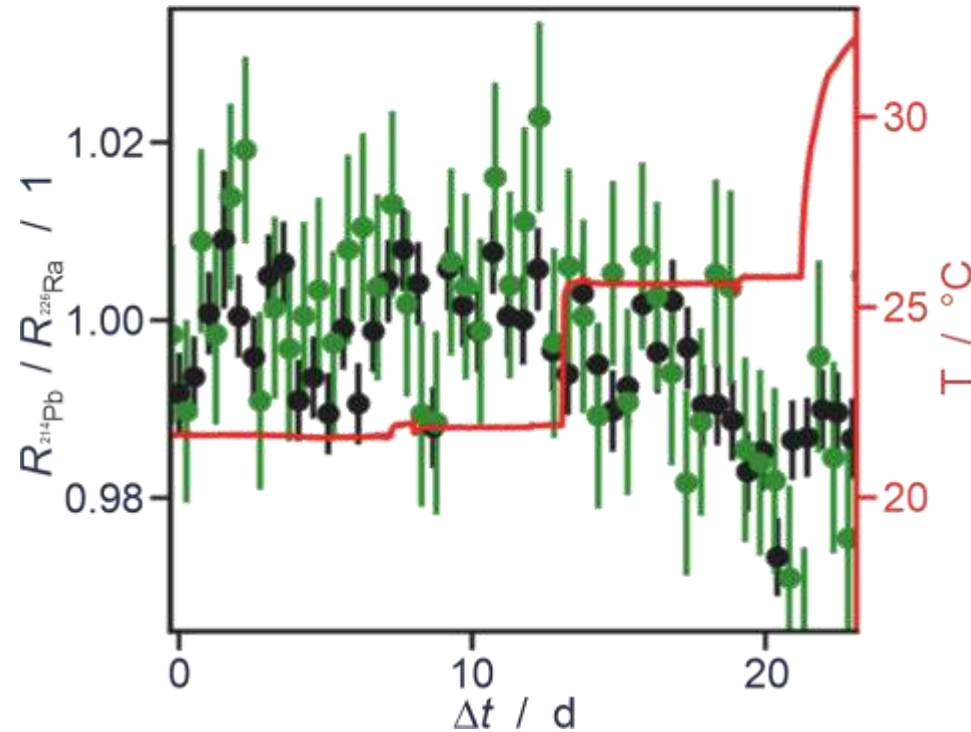


dependencies of emanation from

temperature



relative humidity



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## Ion implantation of $^{226}\text{Ra}$ for a primary $^{222}\text{Rn}$ emanation standard

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### ARTICLE INFO

**Keywords:**  
 Ion implantation  
 $^{226}\text{Ra}$  emanation  
 Laser ionization  
 Defined solid-angle alpha-particle spectrometry

### ABSTRACT

Laser resonance ionization at the RISKO 30 kV mass separator has been used to produce isotopically and isotopically pure and well quantified  $^{226}\text{Ra}$  emanation standards. Based upon laser spectroscopic preparation studies, ion implantation into aluminum and tungsten targets has been carried out, providing overall implantation efficiencies of 40% up to 60%. The absolute implanted activity of  $^{226}\text{Ra}$  was determined by the technique of defined solid angle alpha-particle spectrometry, where excellent energy resolution was observed. The  $^{226}\text{Ra}$  emanation coefficient of the produced targets was studied using alpha-particle and gamma-ray spectrometry, and yielded results between 0.23 and 0.34, with relative uncertainty on the order of 1%. No dependence exceeding a 1% change of the emanation on humidity could be identified in the range of 13% RH to 75% RH, whereas there were hints of a slight correlation between the emanation and temperature. Additionally, and as expected, the emanation coefficient was found to be dependent on the target material as well as the implanted dose.

### 1. Introduction

National and international guidelines and regulations drive the need for SI-traceable measurements of  $^{226}\text{Ra}$  at very low activity concentrations ( $<300\text{ Bq}\cdot\text{m}^{-3}$ ) in air. For such low concentrations, conventional

technique is commonly applied at on-line radioactive ion beam facilities such as CERN-ISOLDE (Fedoseev et al., 2017) or TRIUMF-ISAC (Lassen et al., 2017) for efficient production of exotic nuclides as well as off-line for rare isotope purification and enrichment. A suitable facility for the latter is the RISKO mass separator at Mainz University used for  $^{163}\text{Ho}$

## Implanted Implantation of $^{226}\text{Ra}$ into W / Al after mass separation



**Open Access:**  
<https://doi.org/10.1016/j.apradiso.2021.110093>

Article

## Development of $^{222}\text{Rn}$ Emanation Sources with Integrated Quasi $2\pi$ Active Monitoring

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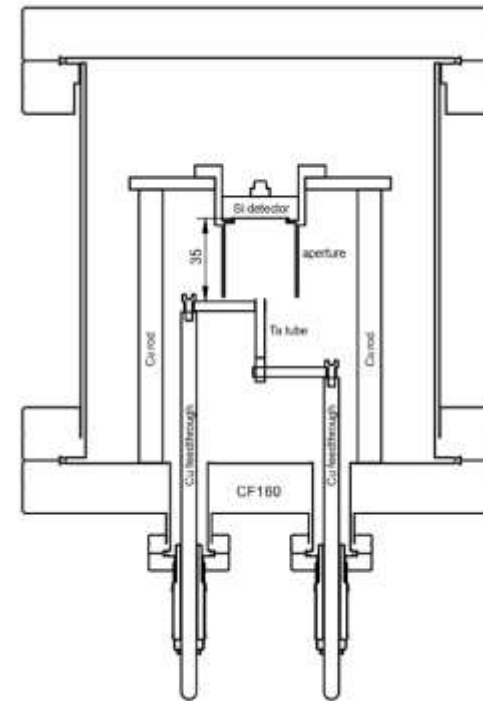
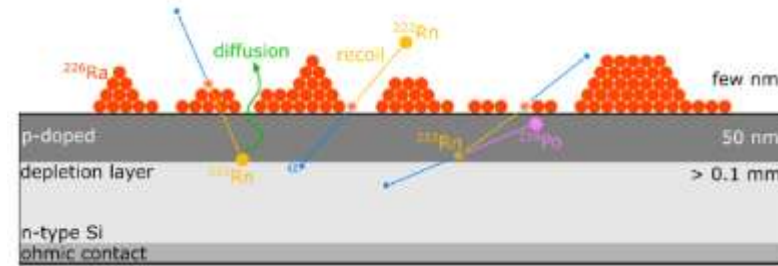
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**Abstract:** In this work, a novel approach for the standardization of low-level  $^{222}\text{Rn}$  emanation is presented. The technique is based on the integration of a  $^{222}\text{Rn}$  source, directly, with an  $\alpha$ -particle detector, which allows the residual  $^{222}\text{Rn}$  to be continuously monitored. Preparation of the device entails thermal physical vapor deposition of  $^{226}\text{RaCl}_2$  directly onto the surface of a commercially available ion implanted Si-diode detector, resulting in a thin-layer geometry. This enables continuous collection of well resolved  $\alpha$ -particle spectra of the nuclei, decaying within the deposited layer, with a detection efficiency of approximately 0.5 in a quasi  $2\pi$  geometry. The continuously sampled  $\alpha$ -particle spectra are used to derive the emanation by statistical inversion. It is possible to achieve this with high temporal resolution due to the small background and the high counting efficiency of the presented technique. The emanation derived in this way exhibits a dependence on the relative humidity of up to 15% in the range from 20% rH to 90% rH. Traceability to the SI is provided by employing defined solid-angle  $\alpha$ -particle spectrometry to characterize the counting efficiency of the modified detectors. The presented technique is demonstrated to apply to a range covering the release of at least 1 to 210  $^{222}\text{Rn}$  atoms per second, and it results in SI-traceable emanation values with a combined standard uncertainty not exceeding 2%. This provides a pathway for the realization of reference atmospheres covering typical environmental  $^{222}\text{Rn}$  levels and thus drastically improves the realization and the dissemination of the derived unit of the activity concentration concerning  $^{222}\text{Rn}$  in air.

**Keywords:**  $^{222}\text{Rn}$  emanation; physical vapor deposition; silicon detectors

**Open Access:**

<https://www.mdpi.com/1660-4601/19/2/840>



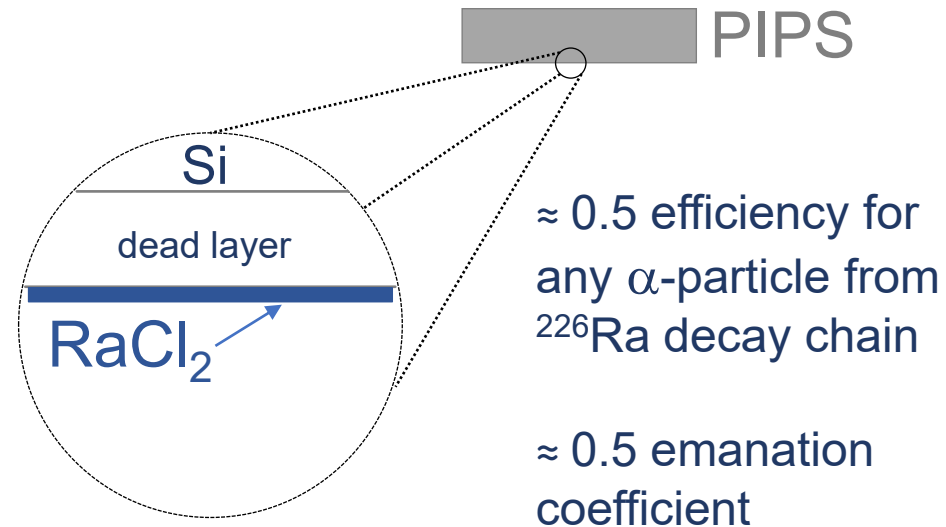
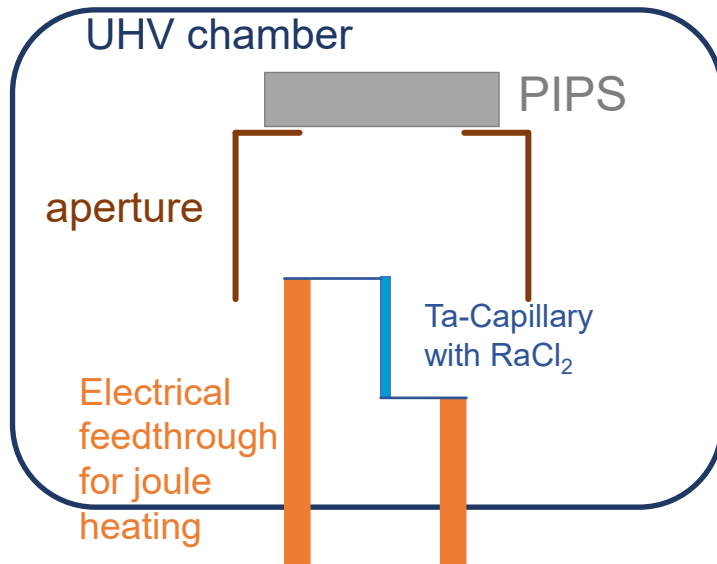
**Citation:** Mertes, F.; Röttger, S.; Röttger, A. Development of  $^{222}\text{Rn}$  Emanation Sources with Integrated Quasi  $2\pi$  Active Monitoring. *Int. J. Environ. Res. Public Health* **2022**, *19*, 840. <https://doi.org/10.3390/ijerph19020840>



# IRSD – idea

Can  $\alpha$ -spectrometry be used to make primary, extremely sensitive, on-line emanation source?

➔ *Modify PIPS with layer of  $\text{RaCl}_2$  by **thermal-PVD***



# IRSD – production model

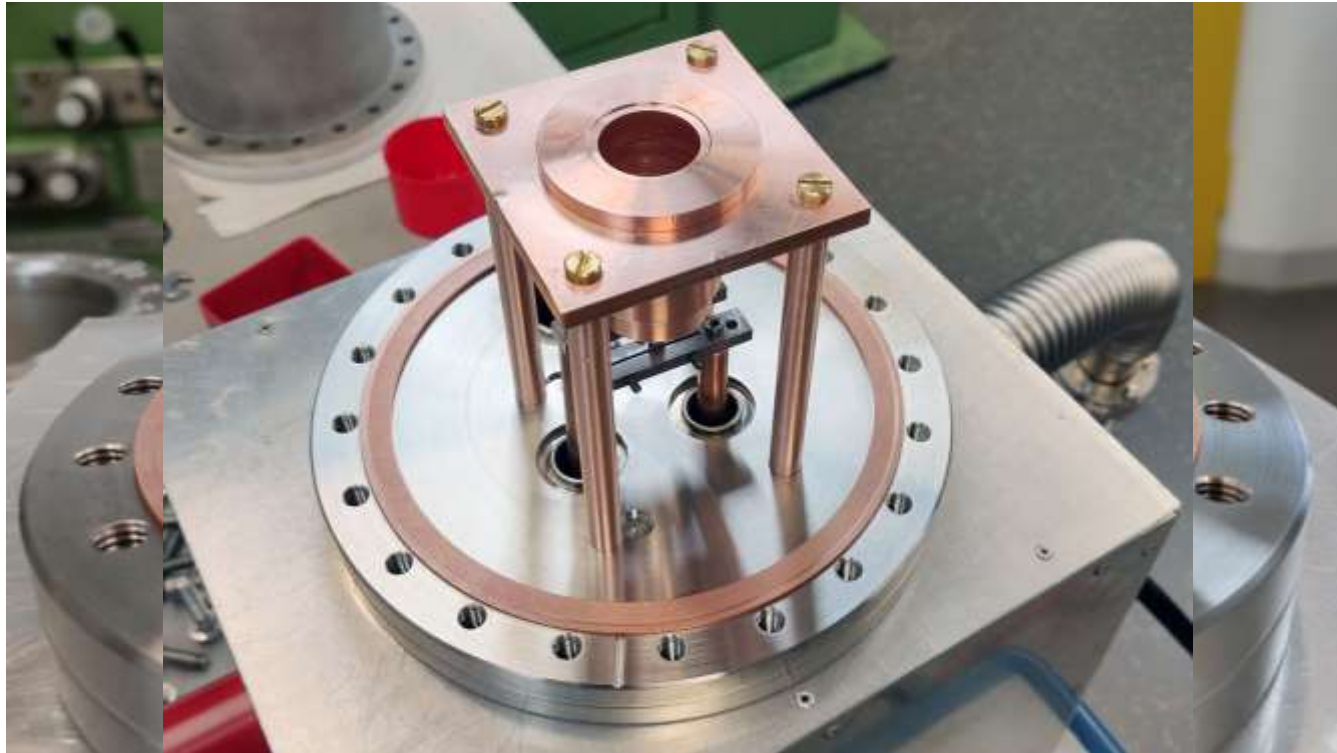


**Primary, on-line  
emanation sources  
model and simulation**



# IRSD – thermal PVD setup

**Primary, on-line emanation sources first realisation of suitable thermal PVD setup**



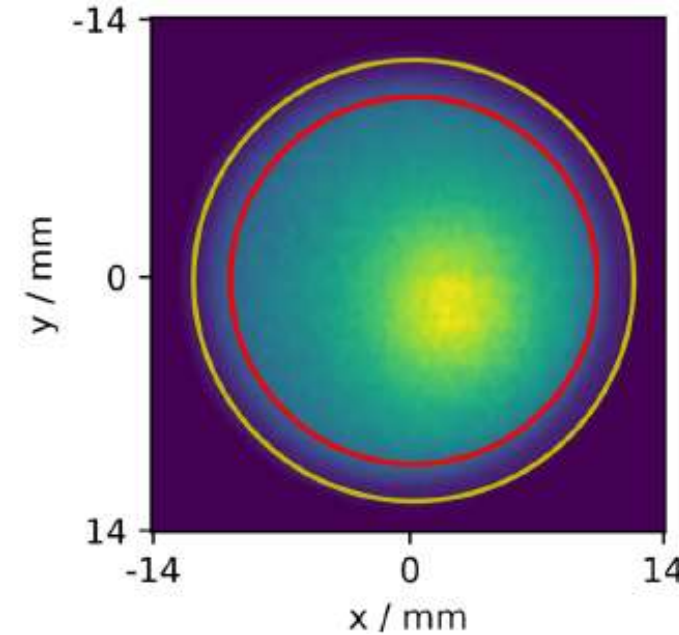
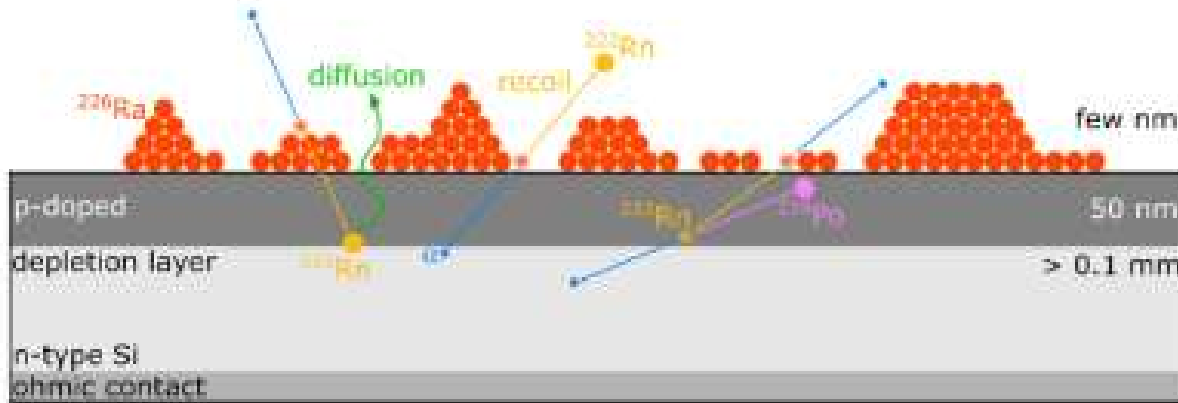
- $< 10^{-6}$  hPa
- Low vapor-pressure materials (Ta)
- est. up to 2000 °C



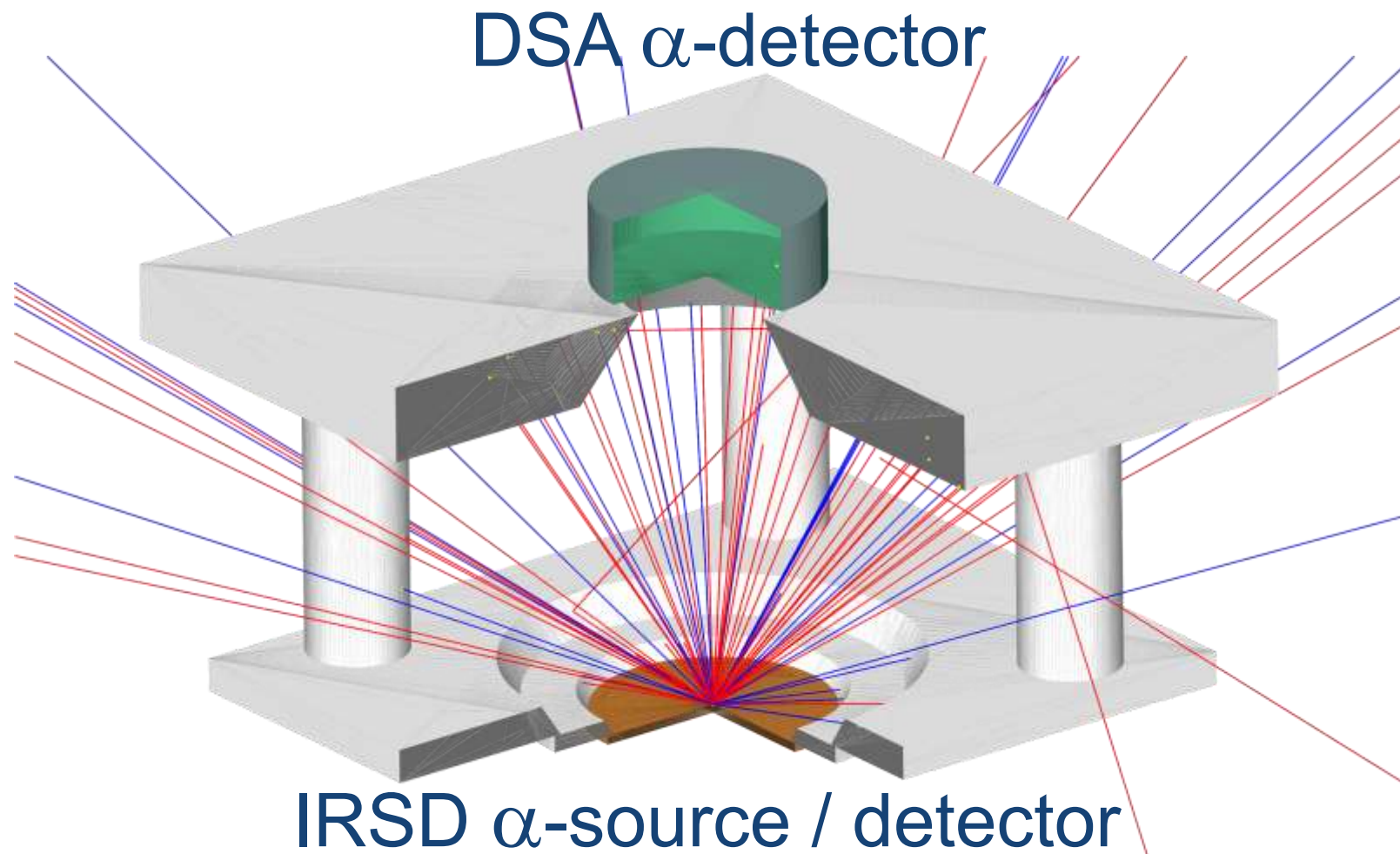
# Integrated Rn Source Detector (IRSD)



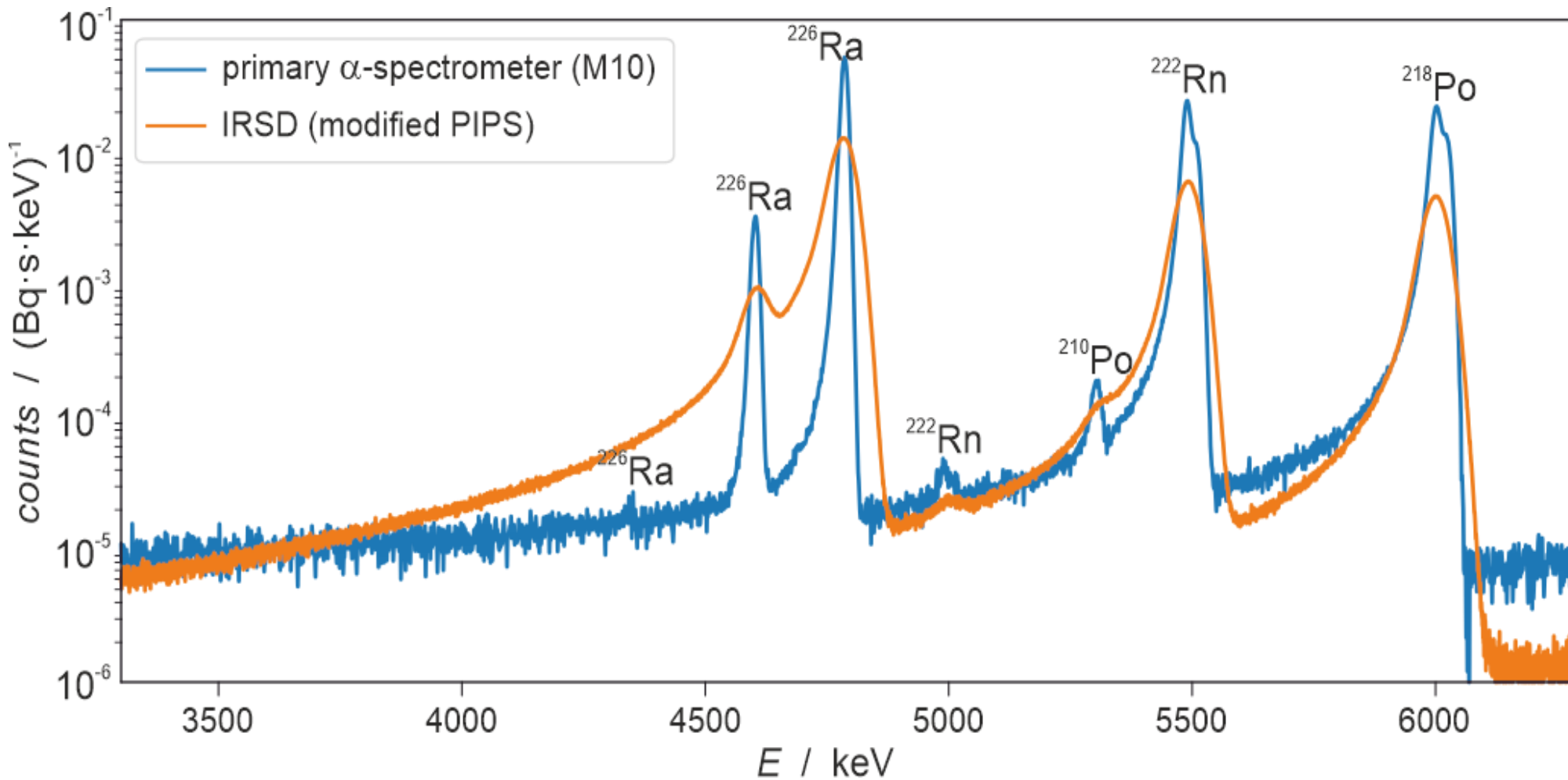
Photograph of an IRSD based on a 450 mm<sup>2</sup> Canberra PIPS® detector, modified with a layer containing **440 Bq <sup>226</sup>RaCl<sub>2</sub>** (left). A digital autoradiograph obtained from such a deposit where the inner diameter of the recessed Si-surface is given in yellow and the diameter of the shadowing aperture is given in red (below).



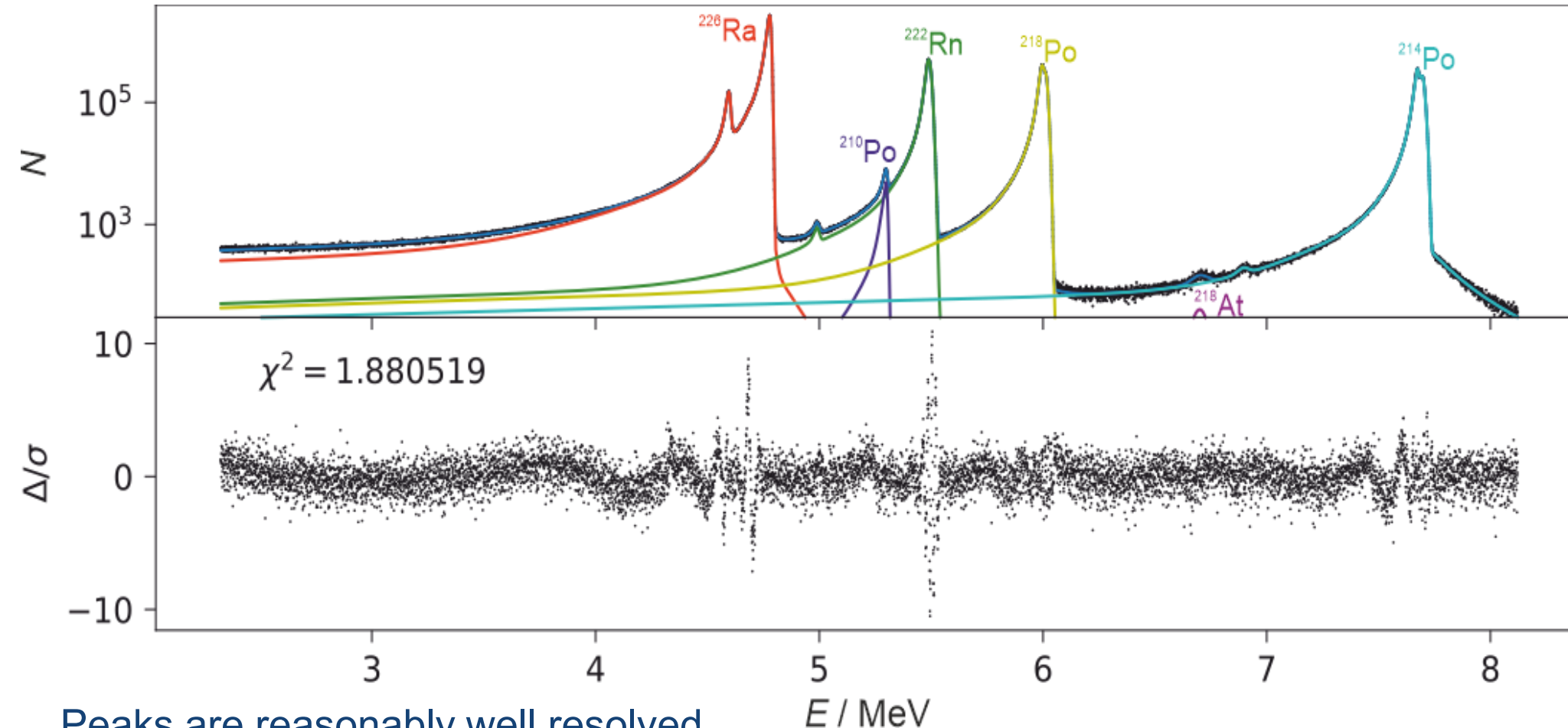
# Primary $^{226}\text{Ra}$ characterisation



## $\alpha$ -spectra - comparison



# $\alpha$ -spectrum of the IRSD



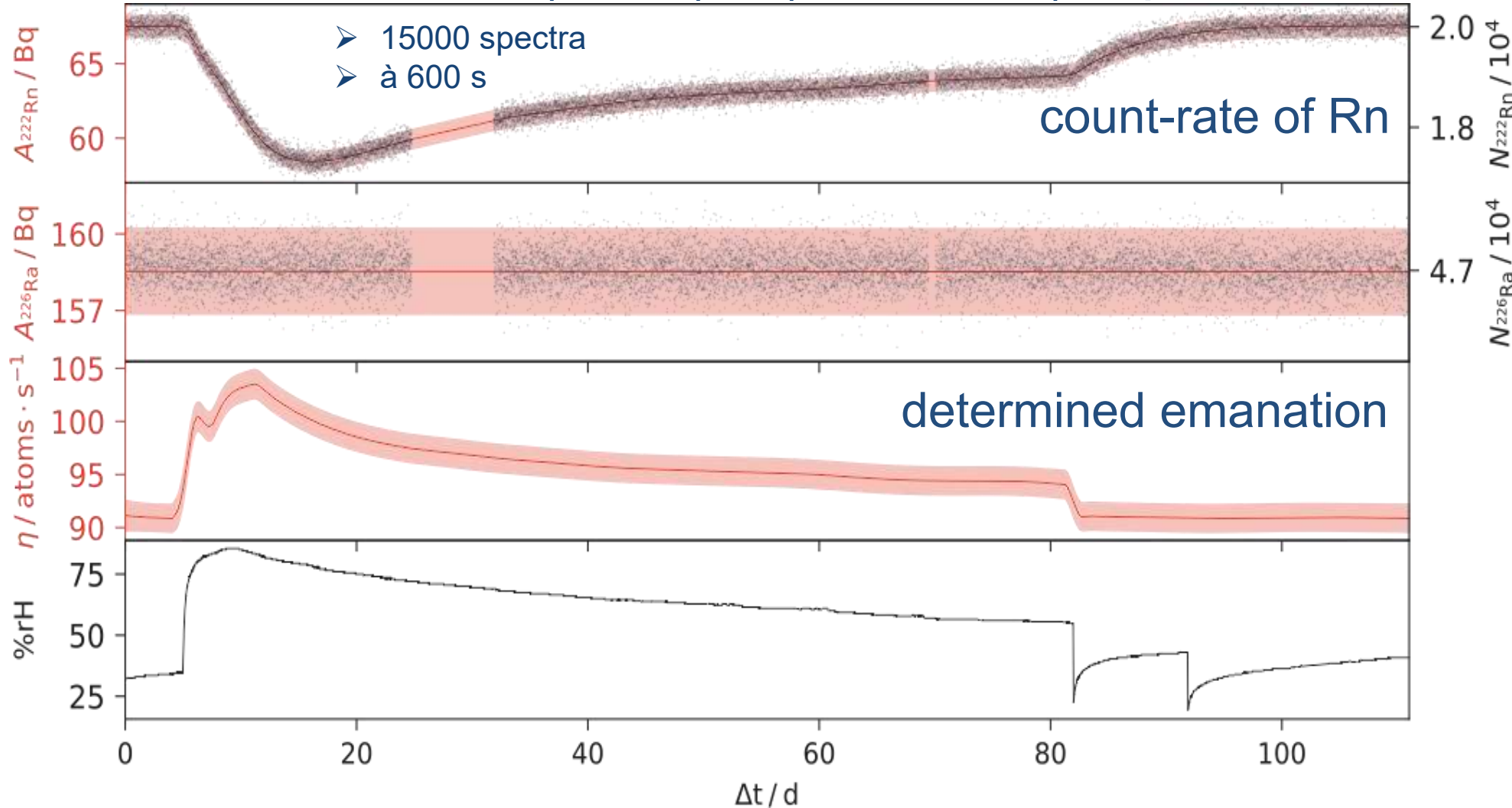
Peaks are reasonably well resolved

- ➡ Model with **different** mixtures of Exponentially Modified Gaussians
- ➡ Deviations in tailings with **penalty** term (for low and high energy tailing)



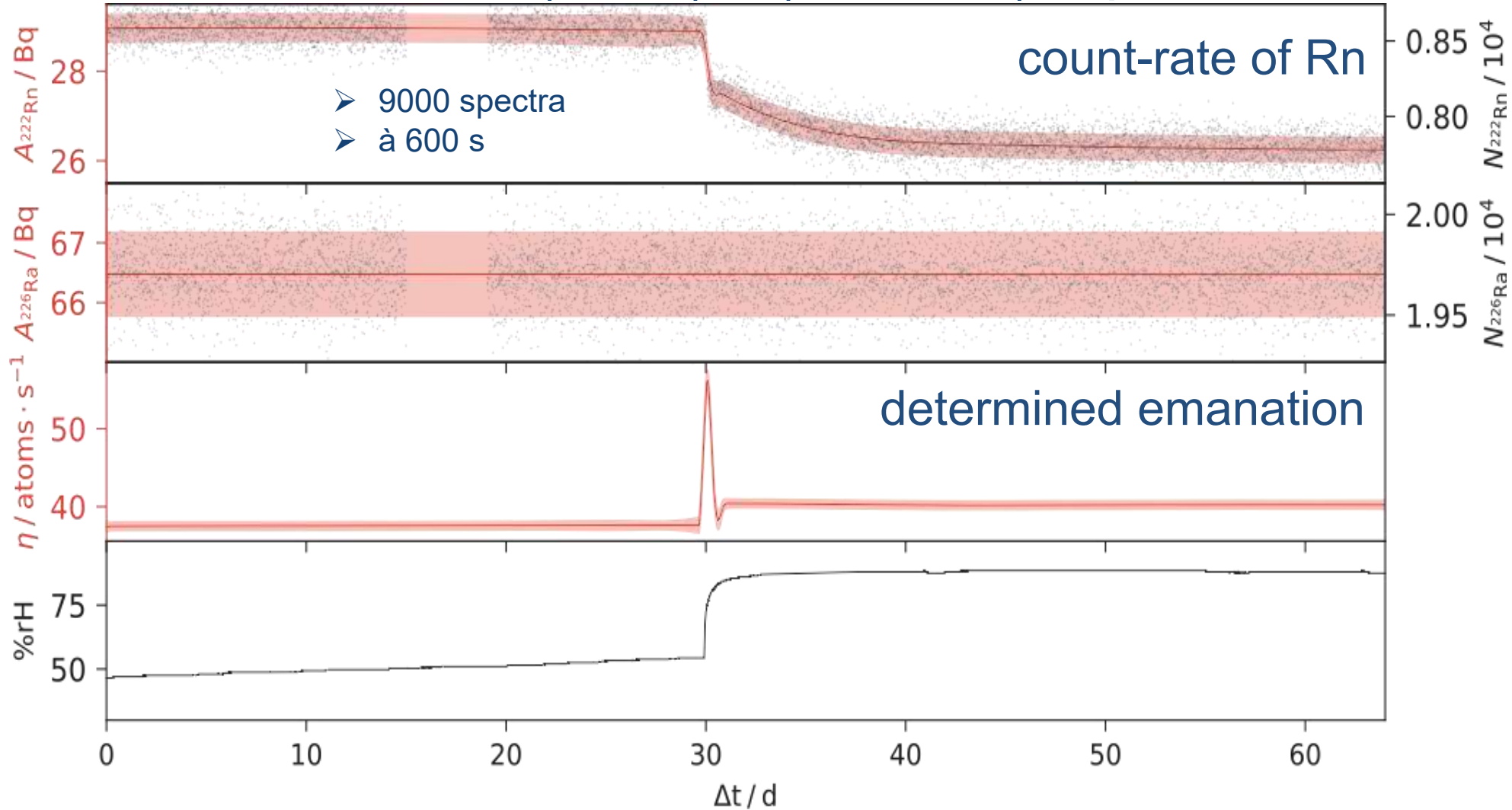
# Gaussian process SLDS-decon.

IRSD:  $A(^{226}\text{Ra}) = (158.6 \pm 1.7) \text{ Bq}$



# Gaussian process SLDS-decon.

IRSD:  $A(^{226}\text{Ra}) = (66.4 \pm 0.5) \text{ Bq}$



## Produced IRSD systems:

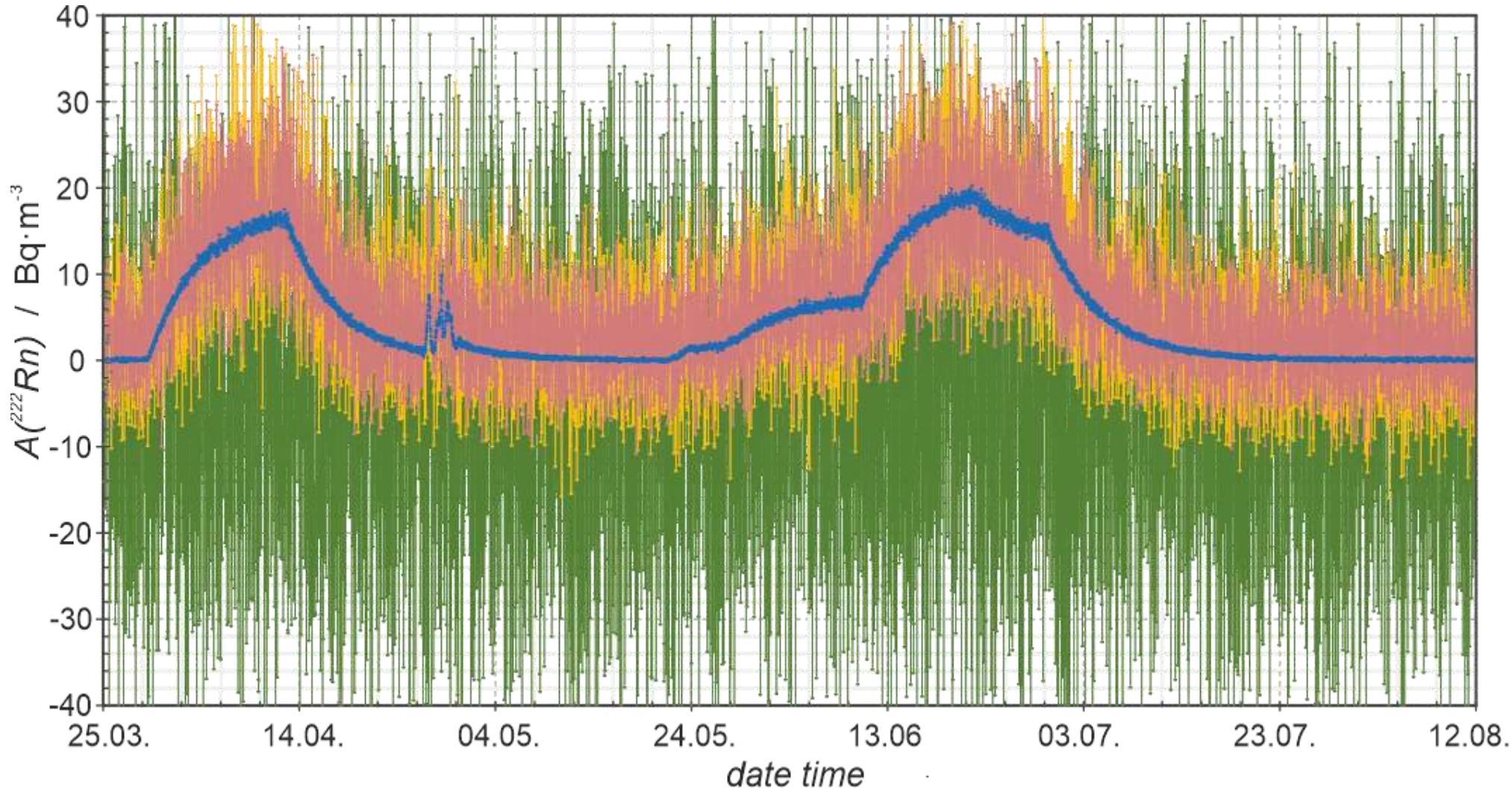
Detector Type	Active Area / Depletion Depth	$A(^{226}\text{Ra}) / \text{Bq}$	$\epsilon_{\text{Ra-226}} / \text{cps Bq}^{-1}$	observed mean $\eta / \text{atoms s}^{-1}$
Mirion PIPS®	450 mm <sup>2</sup> / 300 mm	1.91 ± 0.02	0.502 ± 0.006	0.999 ± 0.017
Ametek Ortec ULTRA®	450 mm <sup>2</sup> / 300 mm	66.4 ± 0.5	0.494 ± 0.004	see figures above
Mirion PIPS®	450 mm <sup>2</sup> / 300 mm	158.6 ± 1.7	0.494 ± 0.005	see figures above
Mirion PIPS®	450 mm <sup>2</sup> / 100 mm	442 ± 4	0.492 ± 0.005	209 ± 4

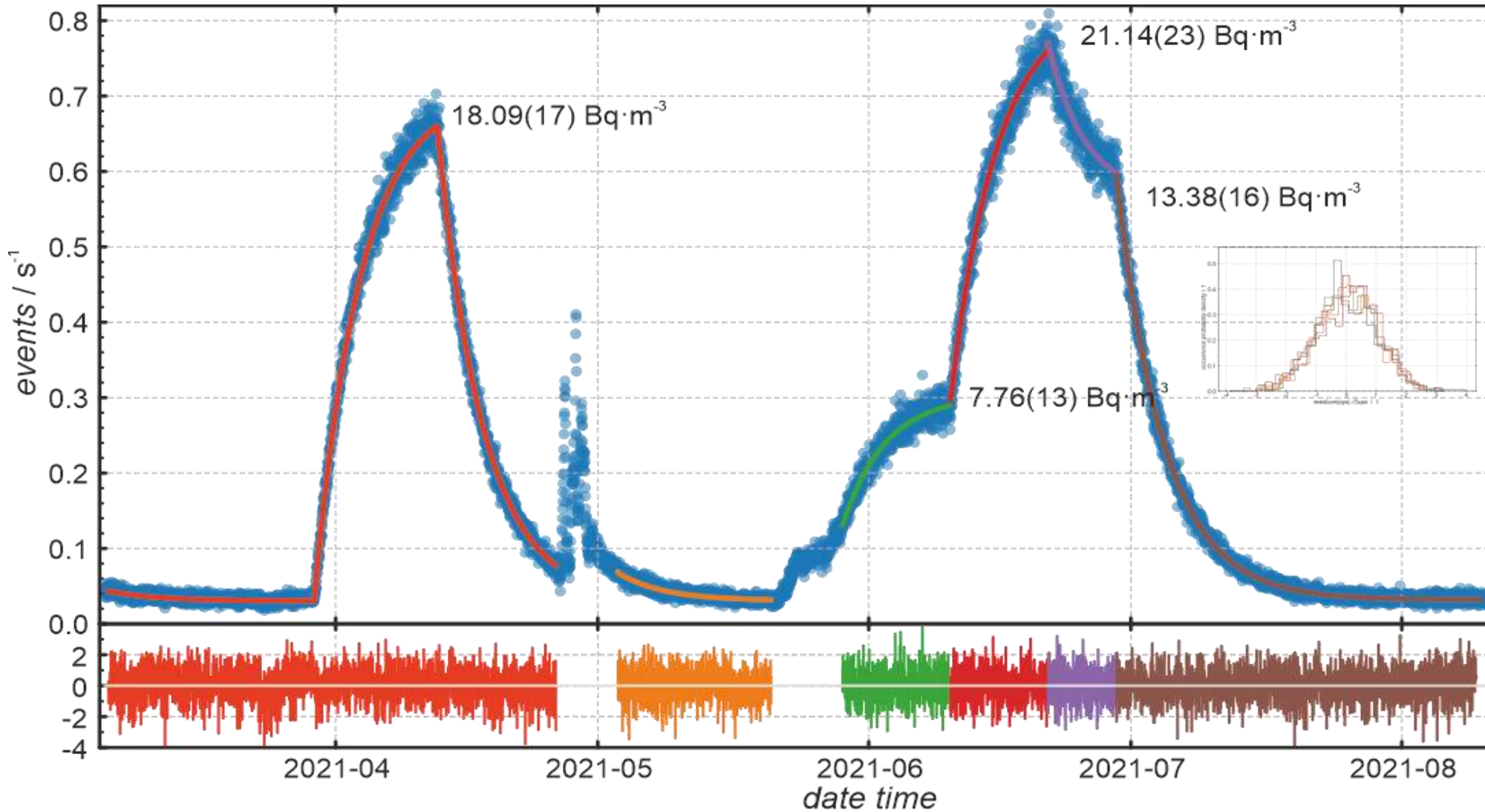
## IRSD uncertainty budget for 2 Bq <sup>226</sup>Ra:

Description and type	Value and uncertainty	Rel. uncertainty	Rel. contribution
Solid angle (systematic)	(0.00940 ± 0.00006) 4π sr	0.6 %	28.4 %
Backscattering <sub>DSA</sub> (systematic)	1 ± 0.002	0.2 %	3%
Tailing <sub>DSA</sub> (systematic)	1 ± 0.003	0.3 %	6.7 %
Tailing <sub>Si</sub> (systematic)	1 ± 0.003	0.3 %	6.7 %
<sup>226</sup> Ra rate <sub>DSA</sub> (stochastic)	(0.01796 ± 0.00015) s <sup>-1</sup>	0.8 %	55.1 %
<sup>226</sup> Ra rate <sub>Si</sub> (stochastic)	(0.9595 ± 0.0004) s <sup>-1</sup>	0.04 %	0.1 %
$\epsilon_{\text{Ra-226}}$	0.502 ± 0.006	1.2 %	

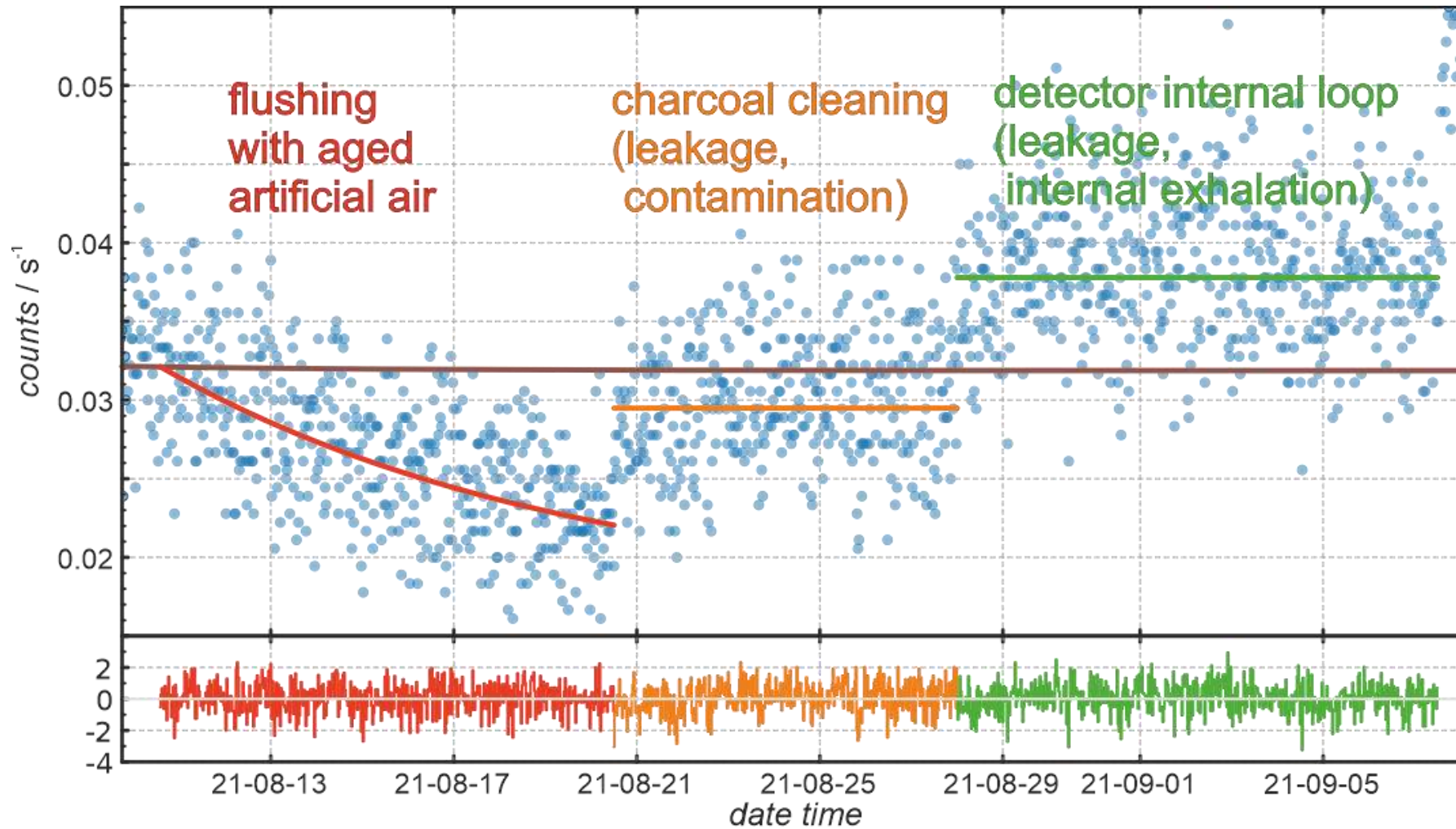


# Radon detector calibration





# traceRadon



# Conclusion



- $^{222}\text{Rn}$  emanation sources from MBq to few Bq are feasible
- Variety of production techniques with different features:
  - drop-cast:
    - + cheap, easy, high emanation
    - not primary, strong dependence
  - electrodeposition:
    - + cheap, easy, high emanation
    - dependence
  - ion implantation:
    - + highest quality, least dependency
    - difficult to produce, expensive, up-to-now not active
  - integrated Radon Source/Detector:
    - + high quality, active, easy to produce, not too expensive, high sensitivity
    - slight dependence, more difficult to use
- Wishlist: mass separated ion implanted IRSD with soft landing



## ... to the traceRadon-project partners:



## ... to the traceRadon-project collaborators:



... to the traceRadon-project Stakeholder Committee, Stakeholders, MSU, EURAMET,

... and for your attention!

