The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

Calibration of a reference instrument – an example

by Stefan and Annette Röttger

Training Course #1:
Precise and traceable Radon activity concentration measurements

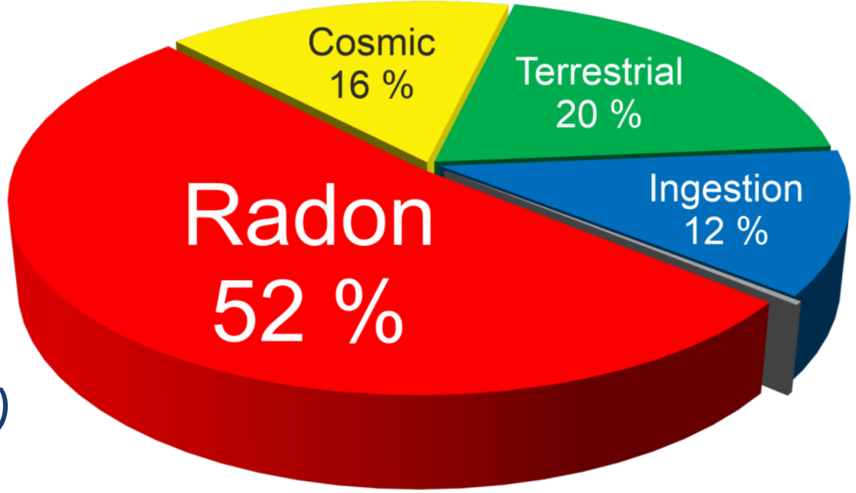
^{222}Radon

- ^{222}Rn: colourless and odourless radioactive noble gas ($t_{1/2}$ = 3.8 d),
  naturally occurring in ^{238}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 Bq·m^{-3}
    (1 atom out of 10^{18} atoms)
  - identification of *Radon Priority Areas (RPA)*

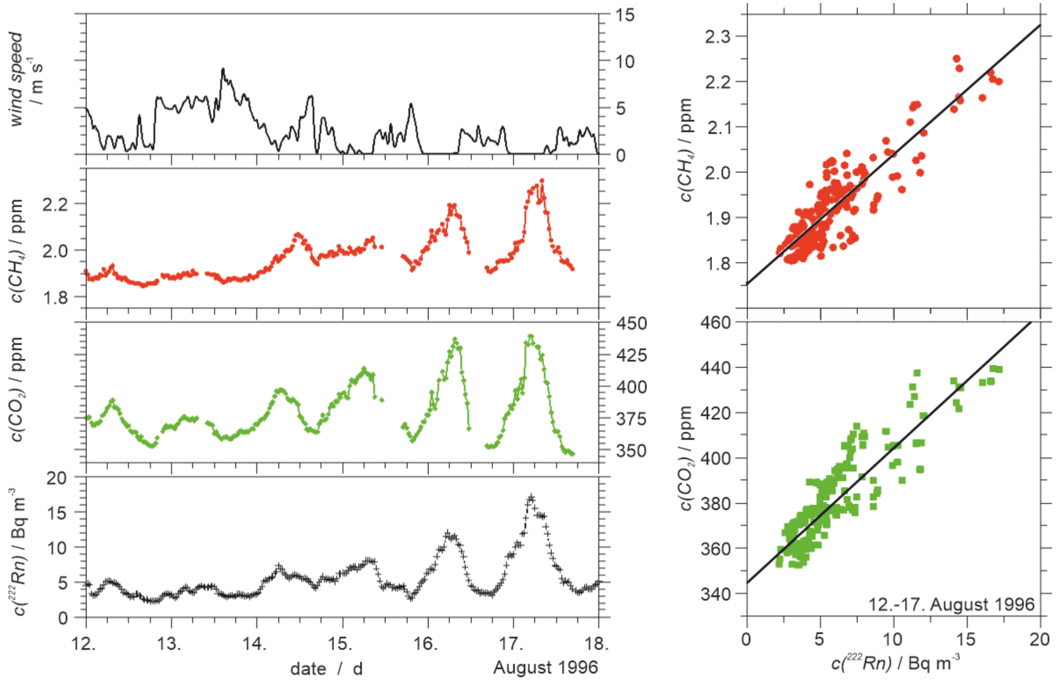
- Outdoor air activity concentration (1 – 10) Bq·m^{-3}



Exposure to natural radiation
UNSCEAR 2008
Total average individual dose:
3 mSv·a^{-1}

^{222}Radon and climate

- ^{222}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



Röttger et.al. Strahlenschutzpraxis 2021

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: 19ENV01 traceRadon

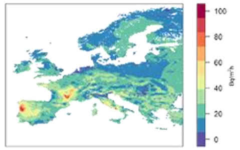
traceRadon

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

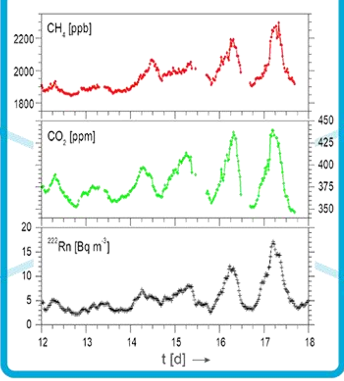
Traceability to the SI system

Radon and radon flux in maps for radiation protection issues **WP4**

- Identification of RPA
- Quantifying the radon wash-out peak
- Data accessibility and public engagement



$$j_{CH_4} = j_{Rn} \cdot \frac{\Delta c_{CH_4}}{\Delta c_{Rn}}$$

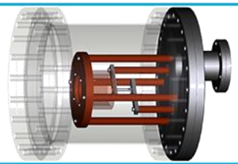


Validation of radon flux models and inventories using radon flux and terrestrial data **WP3**

- Radon flux maps in GHG and climate change studies
- Inclusion of data from radiological early warning systems
- Validation of radon flux maps using radon flux measurements and outdoor radon activity concentrations

Traceable measurements of outdoor radon activity concentrations **WP1**

- Traceable low-level radon sources
- Development of a transfer standard
- Calibration and long-term stability



Radon flux measurements **WP2**

- Development of a reference radon flux monitor
- Test under field conditions
- Measurement campaigns
- RTM application

Management and coordination **WP6**

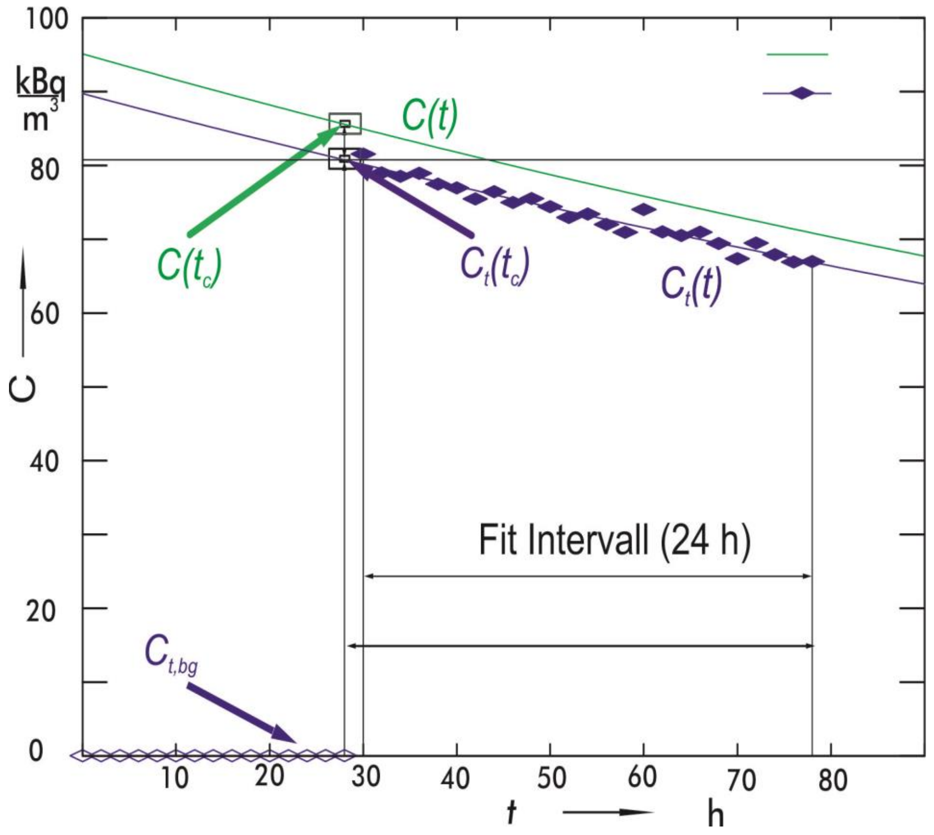
Seven leading European NMI/DI in the field of climate observation and ionising radiation. ICOS, JRC and other stakeholders directly involved as JRP-partners. Sufficient further external partners with high-level expertise to cover the broad spectrum of two scientific communities. High interest by stakeholder community, expressed by 65 letters of support and a large group of 34 potential collaborators.

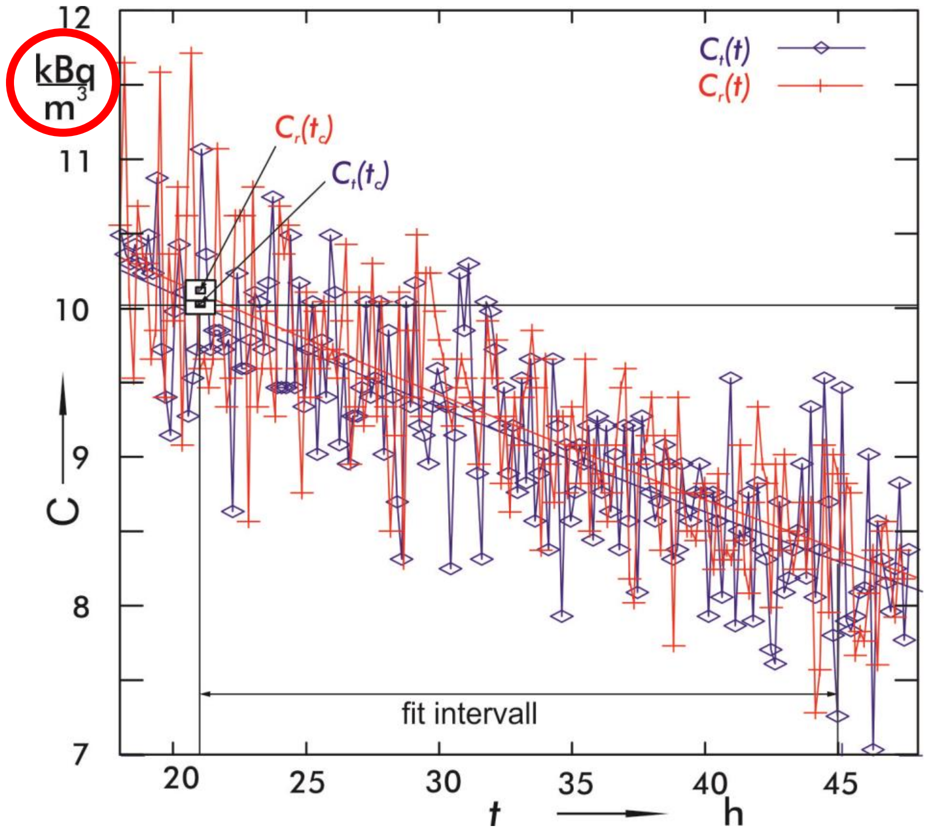
Physikalisch-Technische Bundesanstalt
Braunschweig und Berlin

^{222}Rn gas standard

trace Radon

EURAMET





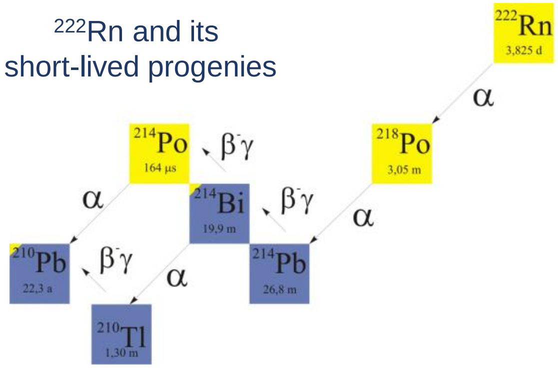
$C_r(t) = C_{r,tc} \cdot e^{(-\lambda \cdot t)} + C_{r,bg}$ $\quad C_t(t) = C_{t,tc} \cdot e^{(-\lambda \cdot t)} + C_{t,bg}$ $\quad k_t = \frac{k_r (C_r(t_c) - C_{r,bg})}{C_t(t_c) - C_{t,bg}}$

$C_r(t) = C_{r,tc} \cdot e^{(-\lambda \cdot t)} + C_{r,bg}$ $\quad C_t(t) = C_{t,tc} \cdot e^{(-\lambda \cdot t)} + C_{t,bg}$ $\quad k_t = \frac{k_r (C_r(t_c) - C_{r,bg})}{C_t(t_c) - C_{t,bg}}$

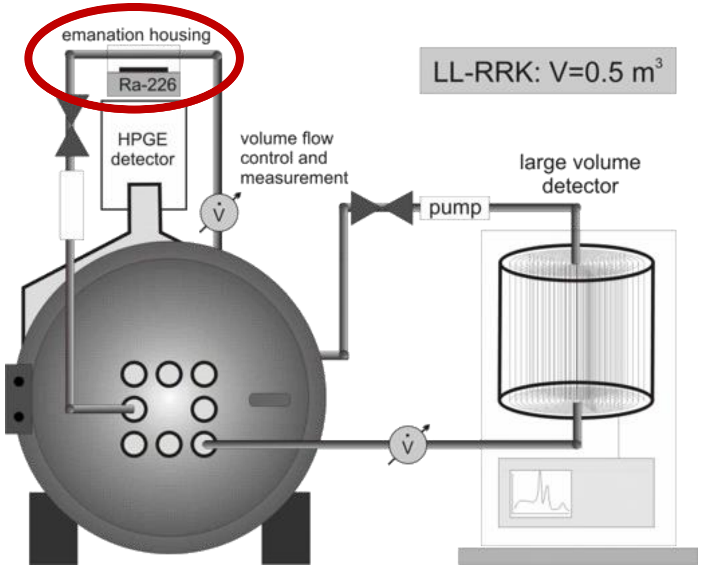
The representation of the activity concentration in the range of 300 Bq·m^{-3} is a challenge to metrology!

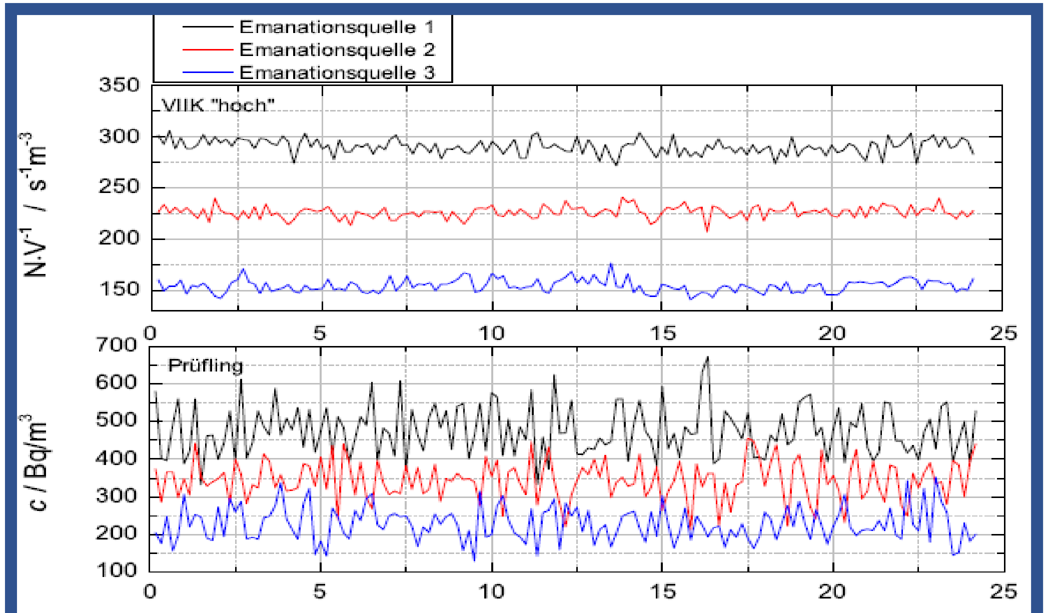
Detector development

^{222}Rn and its short-lived progenies



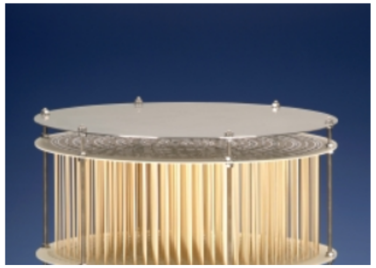
Preparation of EU-BSS by
PhDs of Diana Linzmaier and Florian Mertes





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}Rn emanating sources (1)

classical design

Polyester-foil

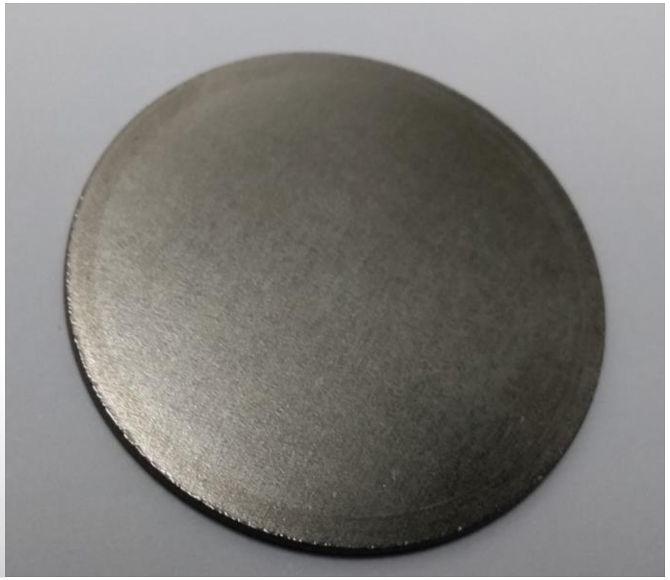
Drop-cast ^{226}Ra wrapped in PE-foil

Electrodeposited source

Deposition at $30\ V < U < 200\ V$

Implanted

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



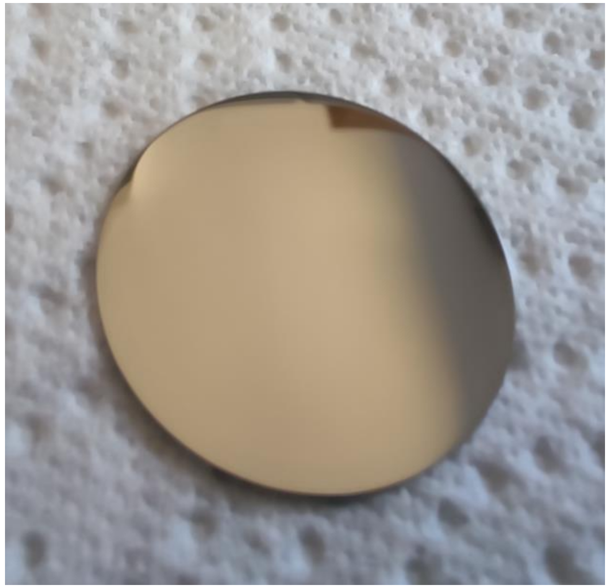
^{222}Rn emanating sources (2)

stainless steel

deposition of ^{226}Ra from $^{226}Ra(NO_3)_2$ via tPVD

p-type Si-wafer

deposition of ^{226}Ra from $^{226}Ra(NO_3)_2$ via tPVD



PIPS 450 mm²

300 µm thickness

deposition of ^{226}Ra from $^{226}Ra(NO_3)_2$ via tPVD

active source

Characterisation

Primary Characterisation of sources

$$^{226}_{88}Ra \rightarrow {}^{4}_{2}\alpha\,(4.87\ MeV) + {}^{222}_{86}Rn\,(86\ keV)$$

^{226}Ra Activity:

- DSA α-Spectrometry
- Autoradiography

Primary ^{222}Rn Source

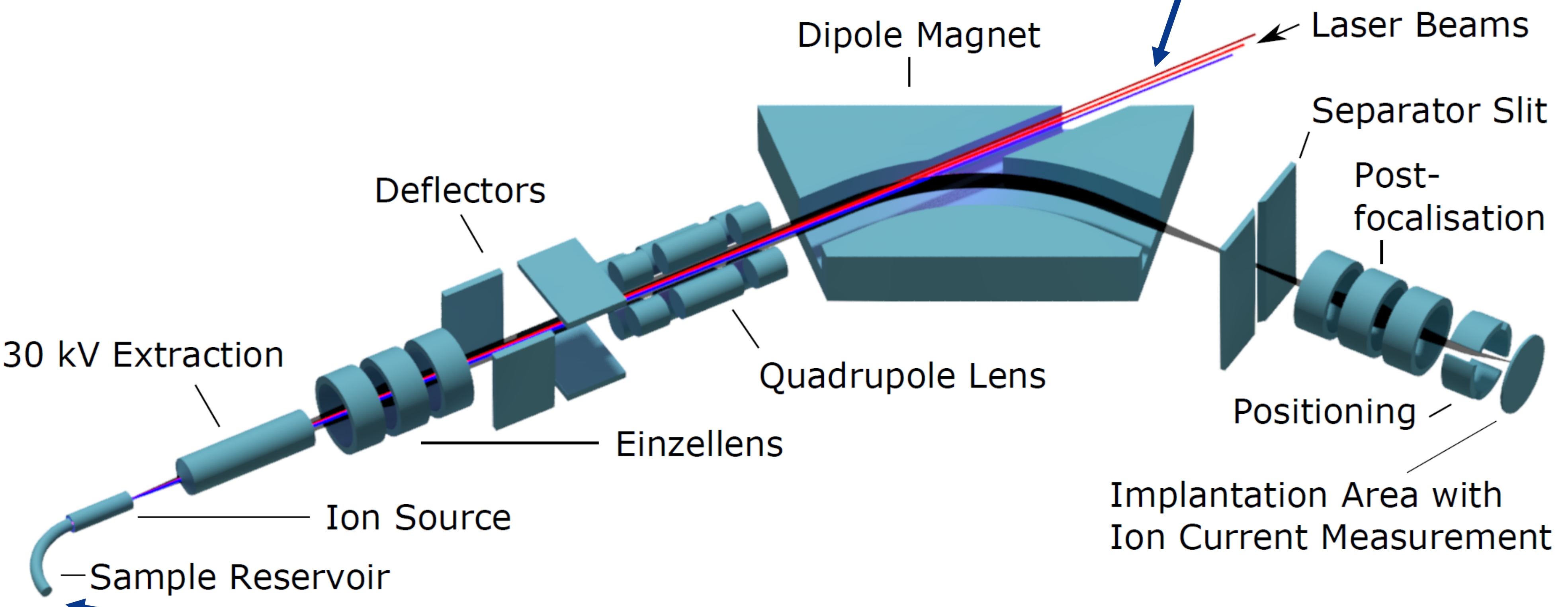
Emanation Power:

- γ-Spectrometry (HPGe, $LaBr_3$, $CeBr_3$, SrI_2)
  ➡ 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

3 tunable Ti:Sa
up to 1.5 W



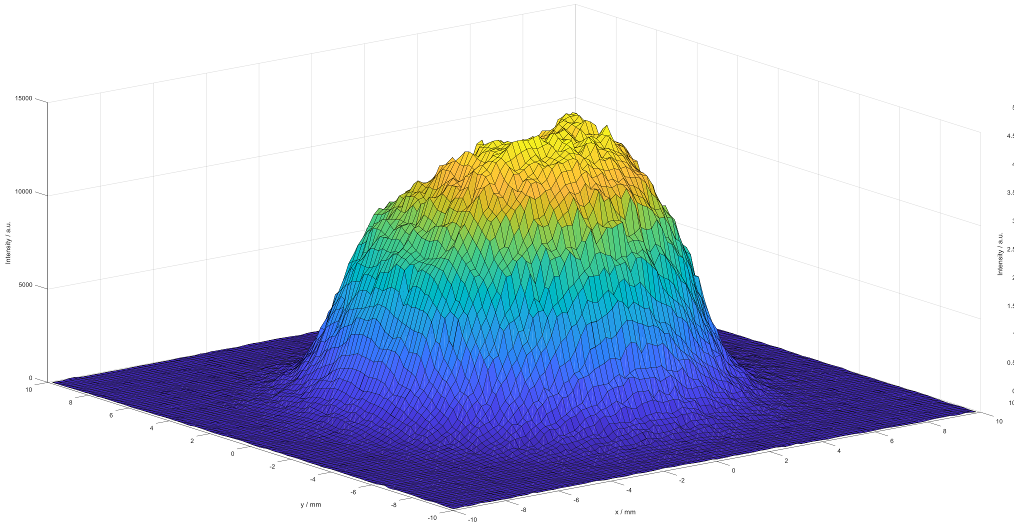
^{226}Ra on Hf-foil
resistive heating

Deposition vs implantation

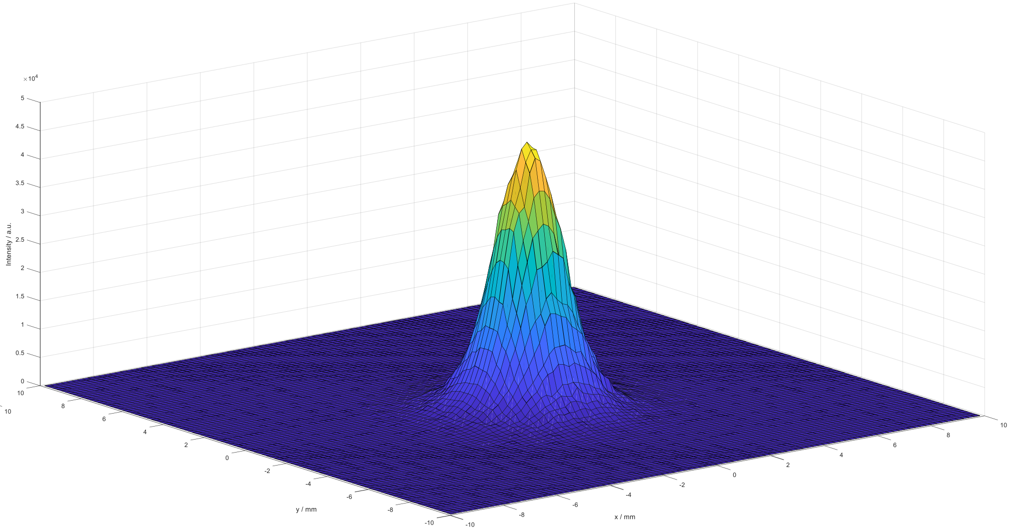
Implantation produces very defined distribution (3D-Gaussian)

➞ Beneficial for α-spectrometry (FWHM, MC-calculations)

Electrodeposition

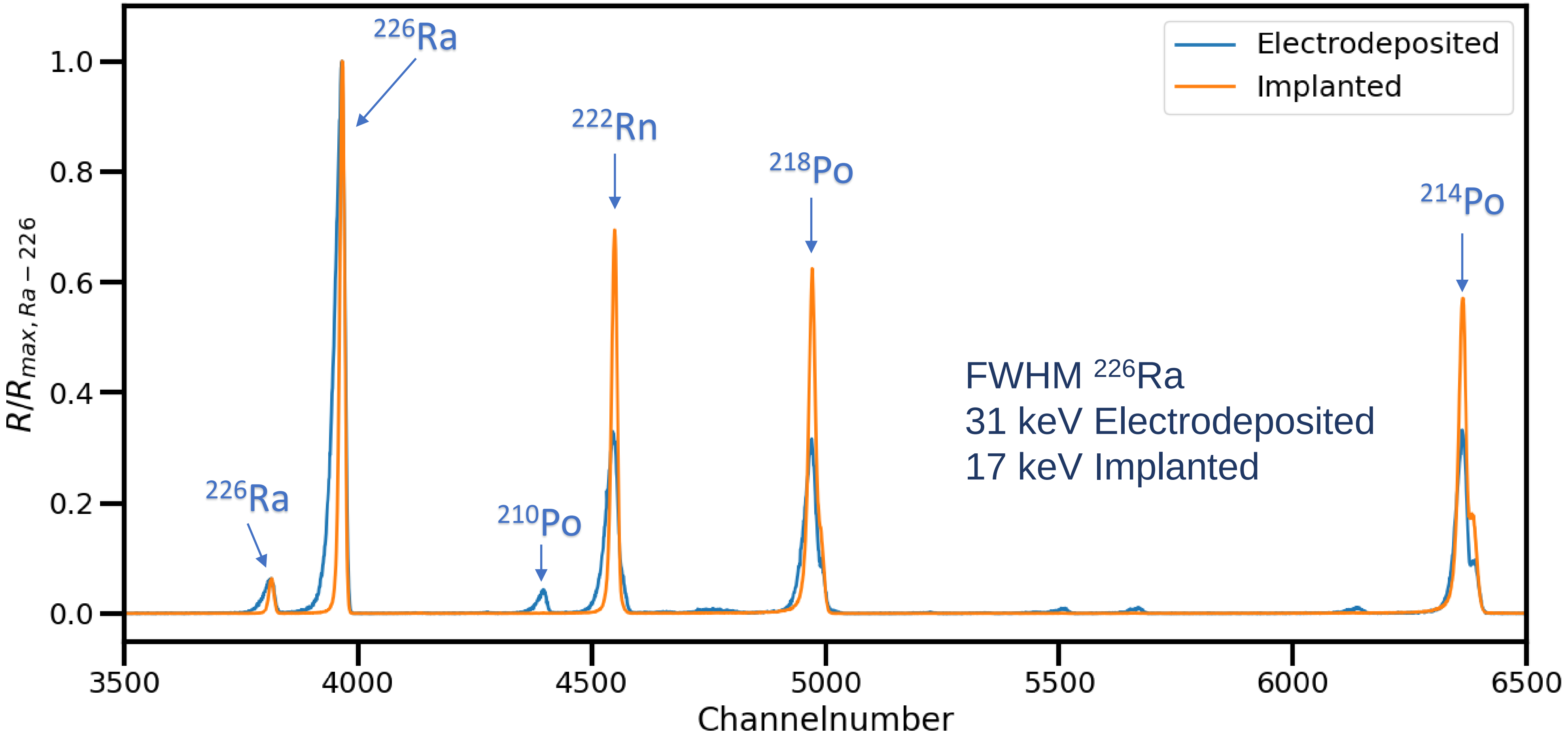


Implantation



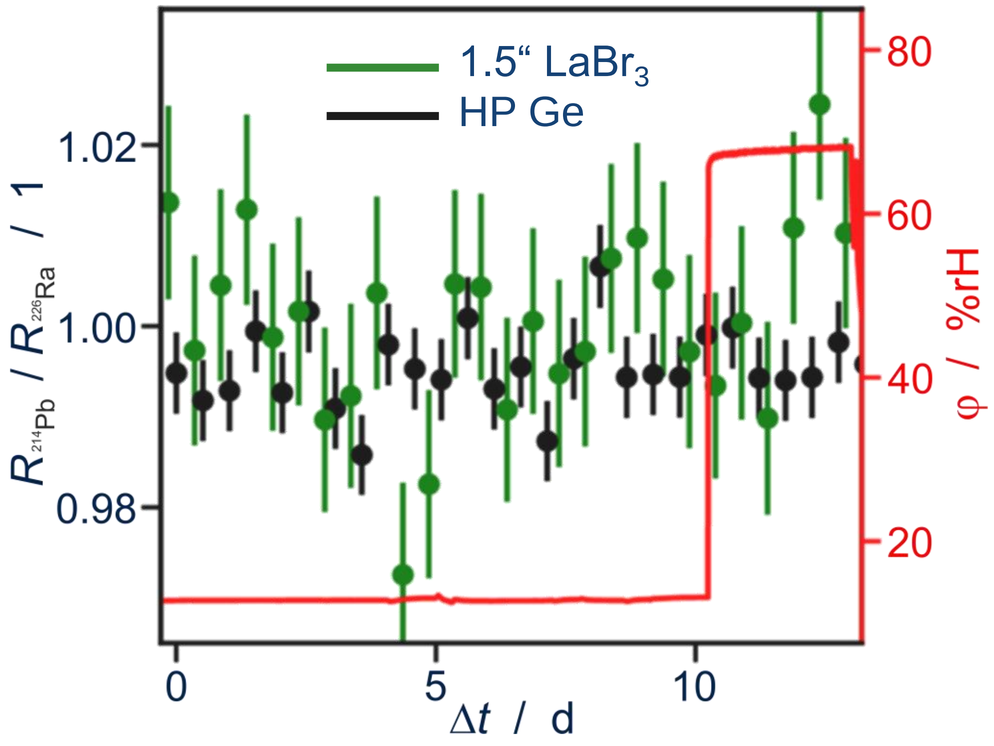
Deposition vs implantation

α-spectra - comparison



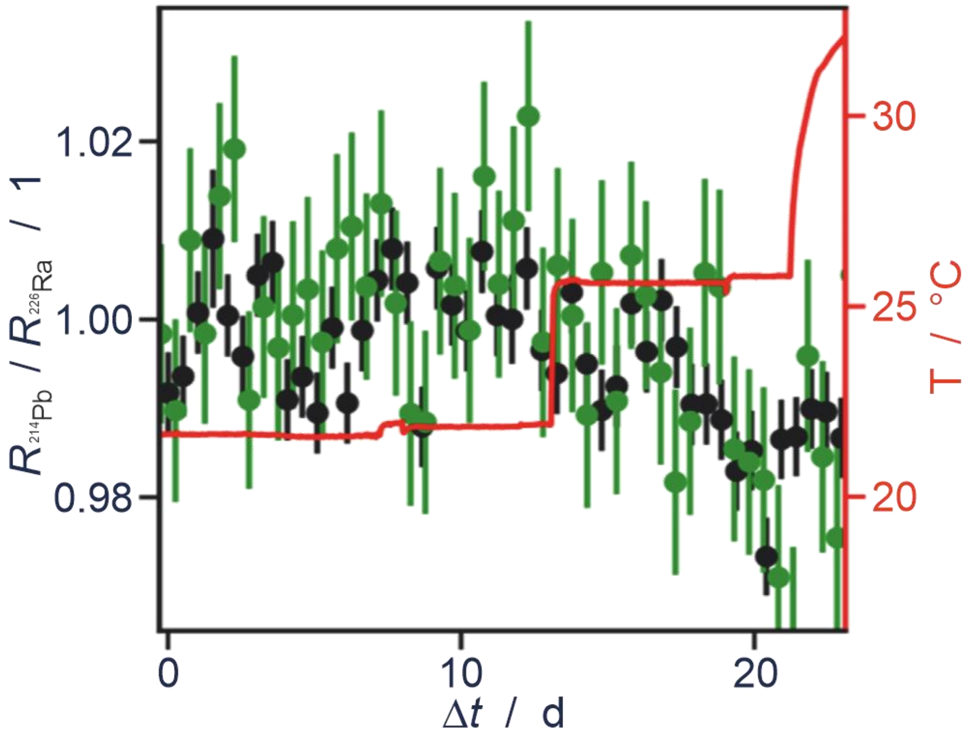
environmental parameter

dependencies of emanation from



relative humidity

temperature



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Mass separated ion implanted ^{226}Ra

Applied Radiation and Isotopes 181 (2022) 110093

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Ion implantation of ^{226}Ra for a primary ^{222}Rn emanation standard

Florian Mertes [a,*], Nina Kneip [b], Reinhard Heinke [b], Tom Kieck [b], Dominik Studer [b], Felix Weber [b], Stefan Röttger [a], Annette Röttger [a], Klaus Wendt [b], Clemens Walther [c]

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ARTICLE INFO

Keywords:
Ion implantation
^{222}Rn emanation
Laser ionization
Defined solid-angle alpha-particle spectrometry

ABSTRACT

Laser resonance ionization at the RISIKO 30 kV mass separator has been used to produce isotopically and isobarically pure and well quantified ^{222}Rn 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}Ra was determined by the technique of defined solid-angle α-particle spectrometry, where excellent energy resolution was observed. The ^{222}Rn emanation coefficient of the produced targets was studied using α-particle and γ-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 15 %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 ^{222}Rn at very low activity concentrations (<300 Bq·m^{-3}) in air. For such low concentrations, conventional technique is commonly applied at on-line radioactive ion beam facilities such as CERN-ISOLDE (Fedosseev 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 RISIKO mass separator at Mainz University used for ^{163}Ho

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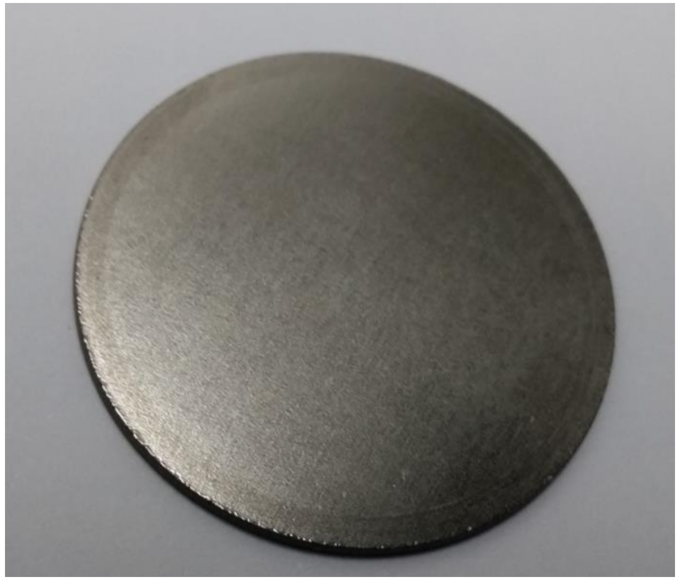
https://doi.org/10.1016/j.apradiso.2021.110093

Implanted

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



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Integrated Rn Source Detector (IRSD)

International Journal of *Environmental Research and Public Health*

MDPI

Article

Development of ^{222}Rn Emanation Sources with Integrated Quasi 2π Active Monitoring

Florian Mertes *, Stefan Röttger and Annette Röttger

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Abstract: In this work, a novel approach for the standardization of low-level ^{222}Rn emanation is presented. The technique is based on the integration of a ^{222}Rn source, directly, with an α-particle detector, which allows the residual ^{222}Rn to be continuously monitored. Preparation of the device entails thermal physical vapor deposition of $^{226}RaCl_2$ 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 α-particle spectra of the nuclei, decaying within the deposited layer, with a detection efficiency of approximately 0.5 in a quasi 2π geometry. The continuously sampled α-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 α-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}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}Rn levels and thus drastically improves the realization and the dissemination of the derived unit of the activity concentration concerning ^{222}Rn in air.

Keywords: ^{222}Rn emanation; physical vapor deposition; silicon detectors

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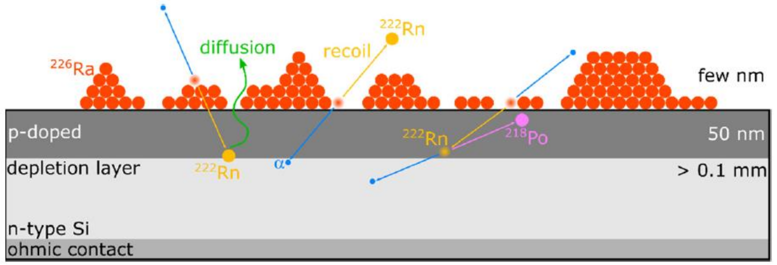
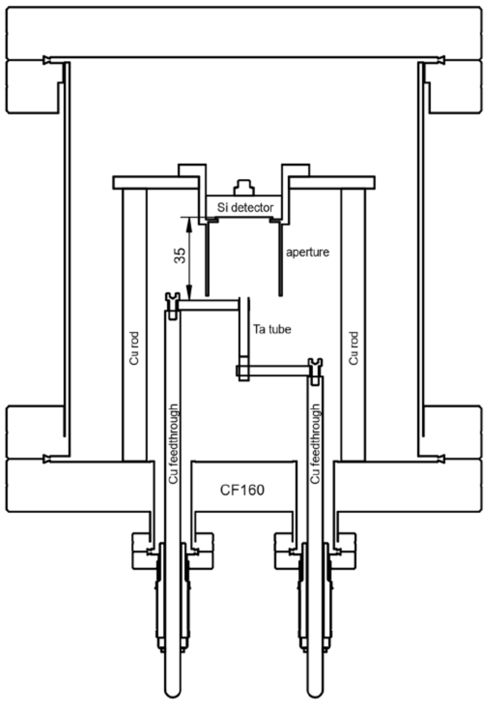
check for updates

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

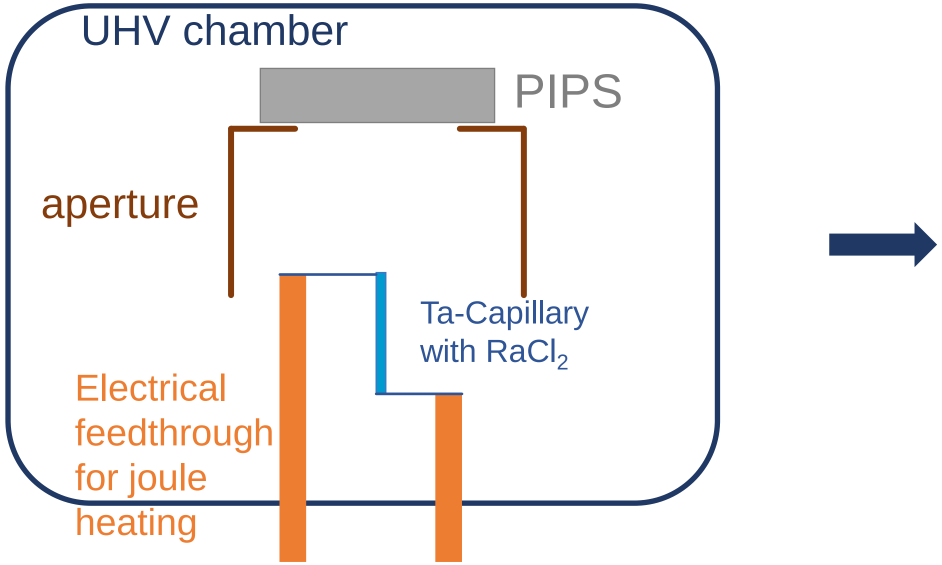


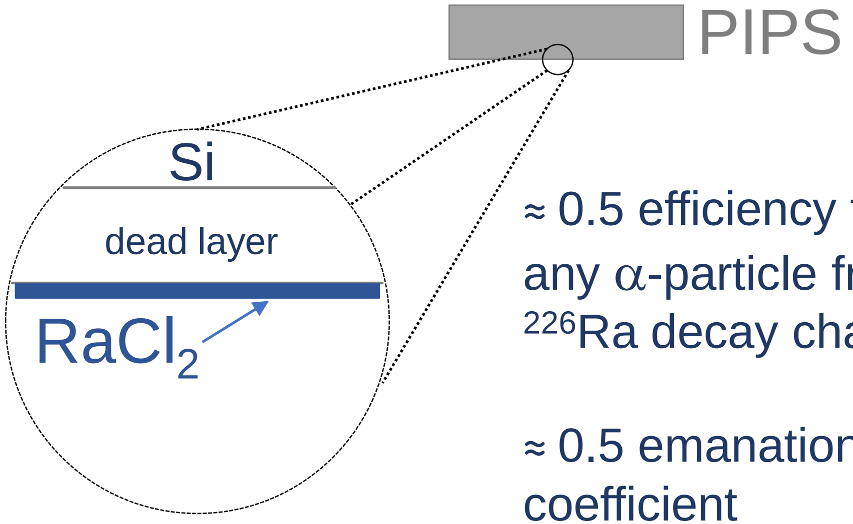


IRSD – idea

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

➡ *Modify PIPS with layer of $RaCl_2$ by **thermal-PVD***



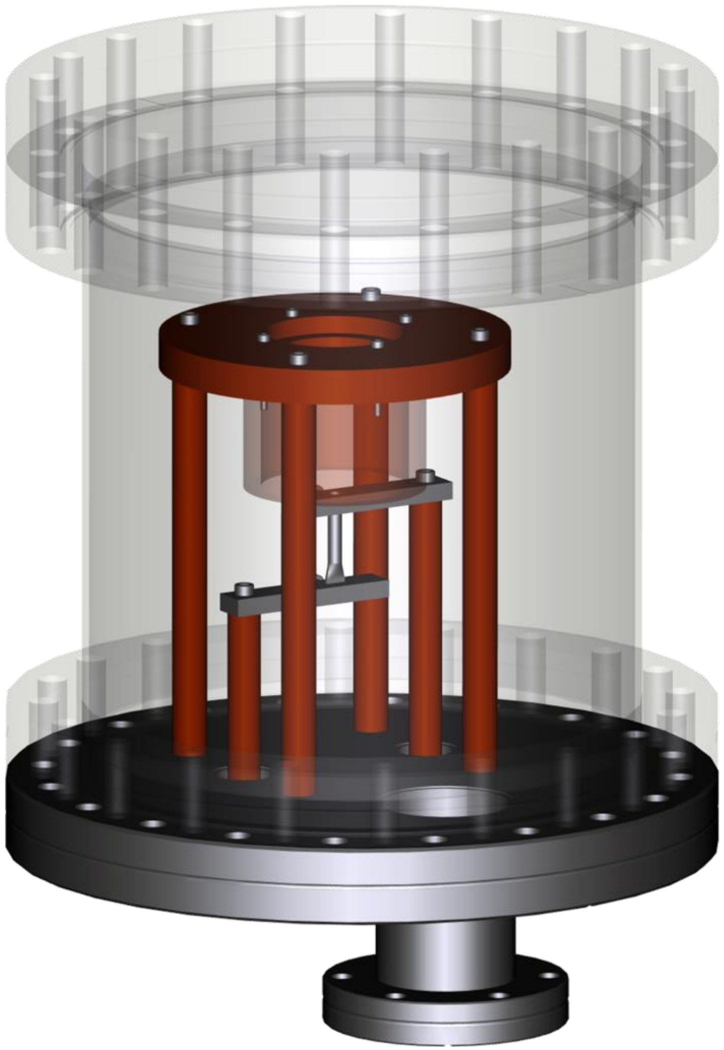


≈ 0.5 efficiency for any α-particle from ^{226}Ra decay chain

≈ 0.5 emanation coefficient

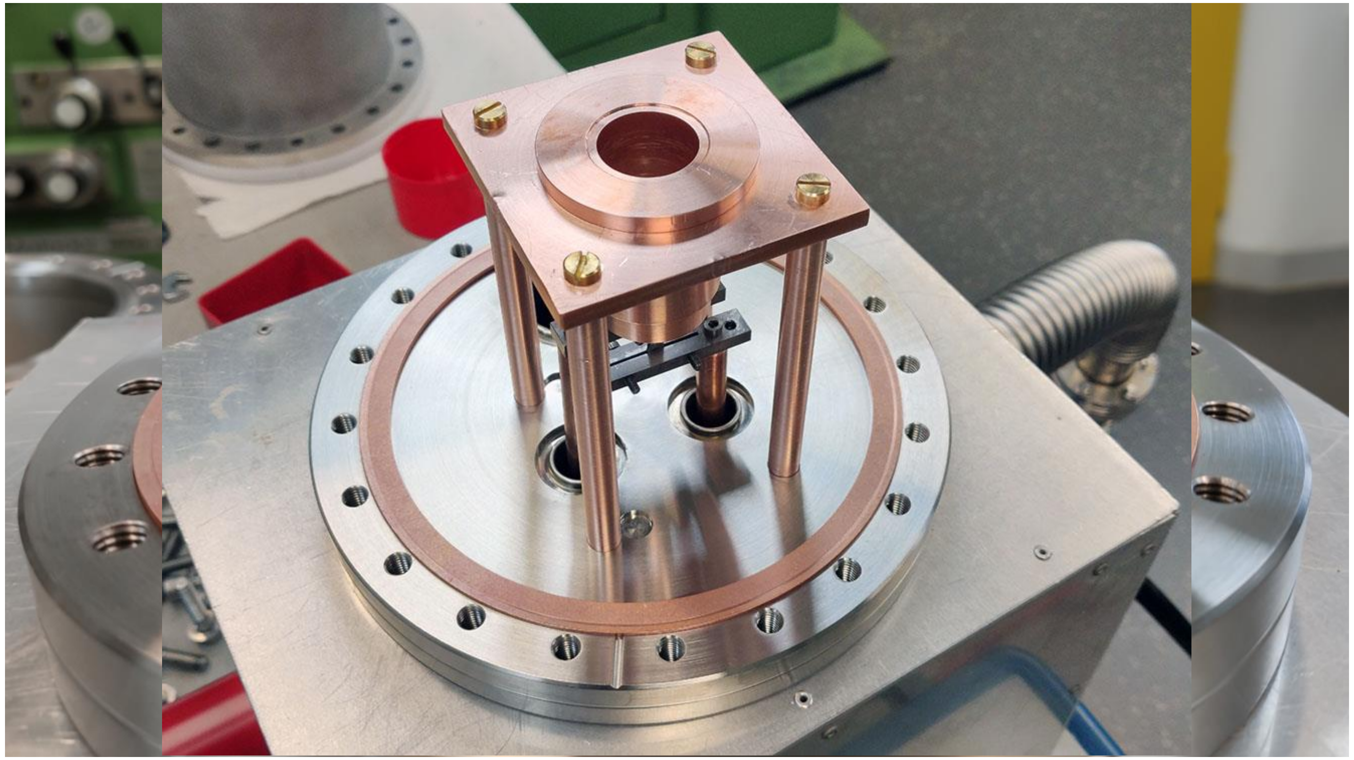
IRSD – production model

Primary, on-line emanation sources model and simulation



IRSD – thermal PVD setup

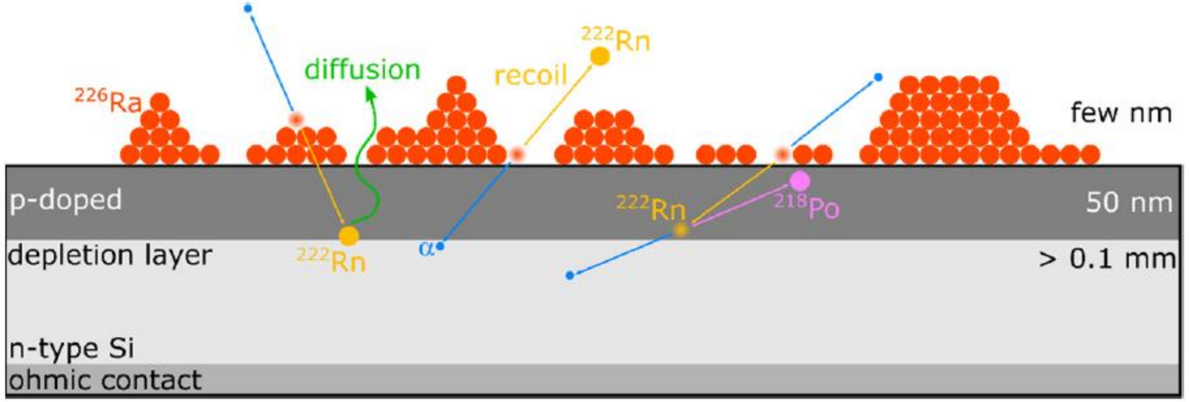
Primary, on-line emanation sources first realisation of suitable thermal PVD setup

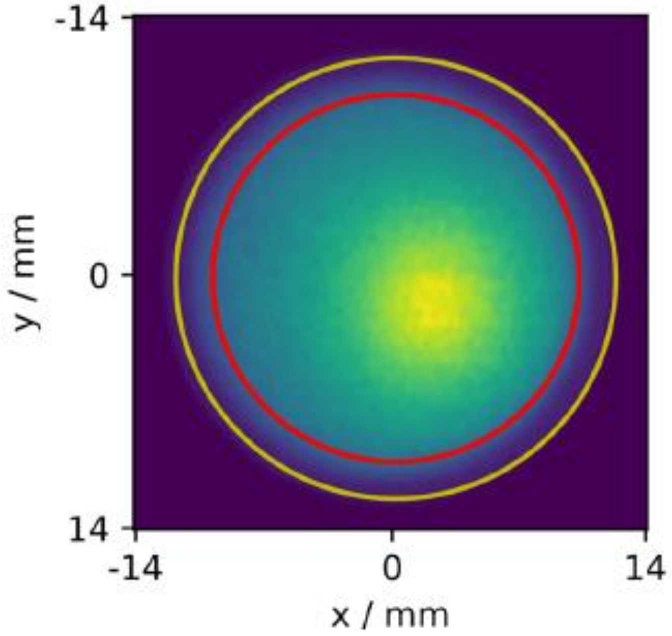


- $< 10^{-6}$ hPa
- Low vapor-pressure materials (Ta)
- est. up to 2000 °C

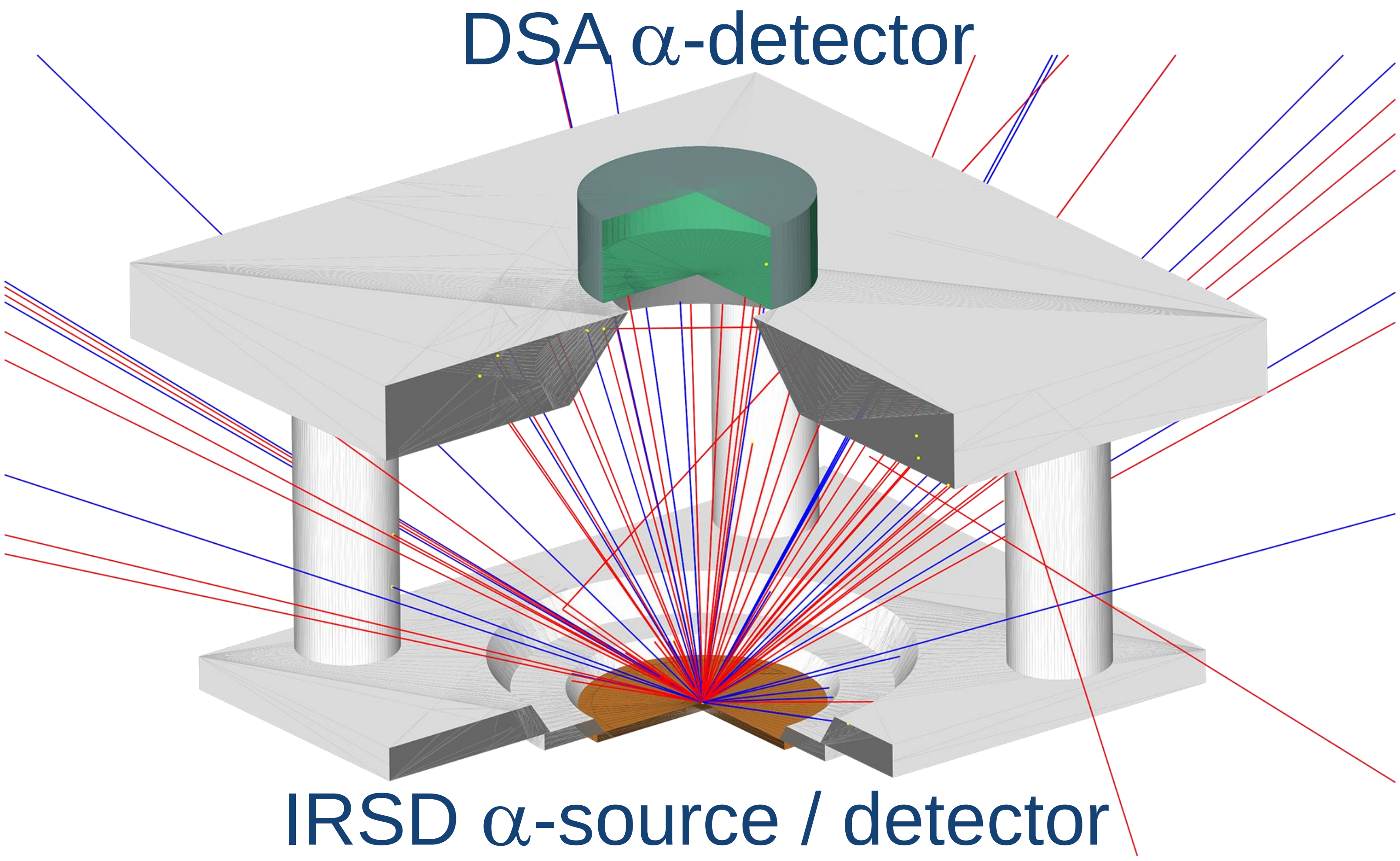
Integrated Rn Source Detector (IRSD)

Photograph of an IRSD based on a 450 mm^2 Canberra PIPS® detector, modified with a layer containing **440 Bq ^{226}RaCl$_2$** (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).



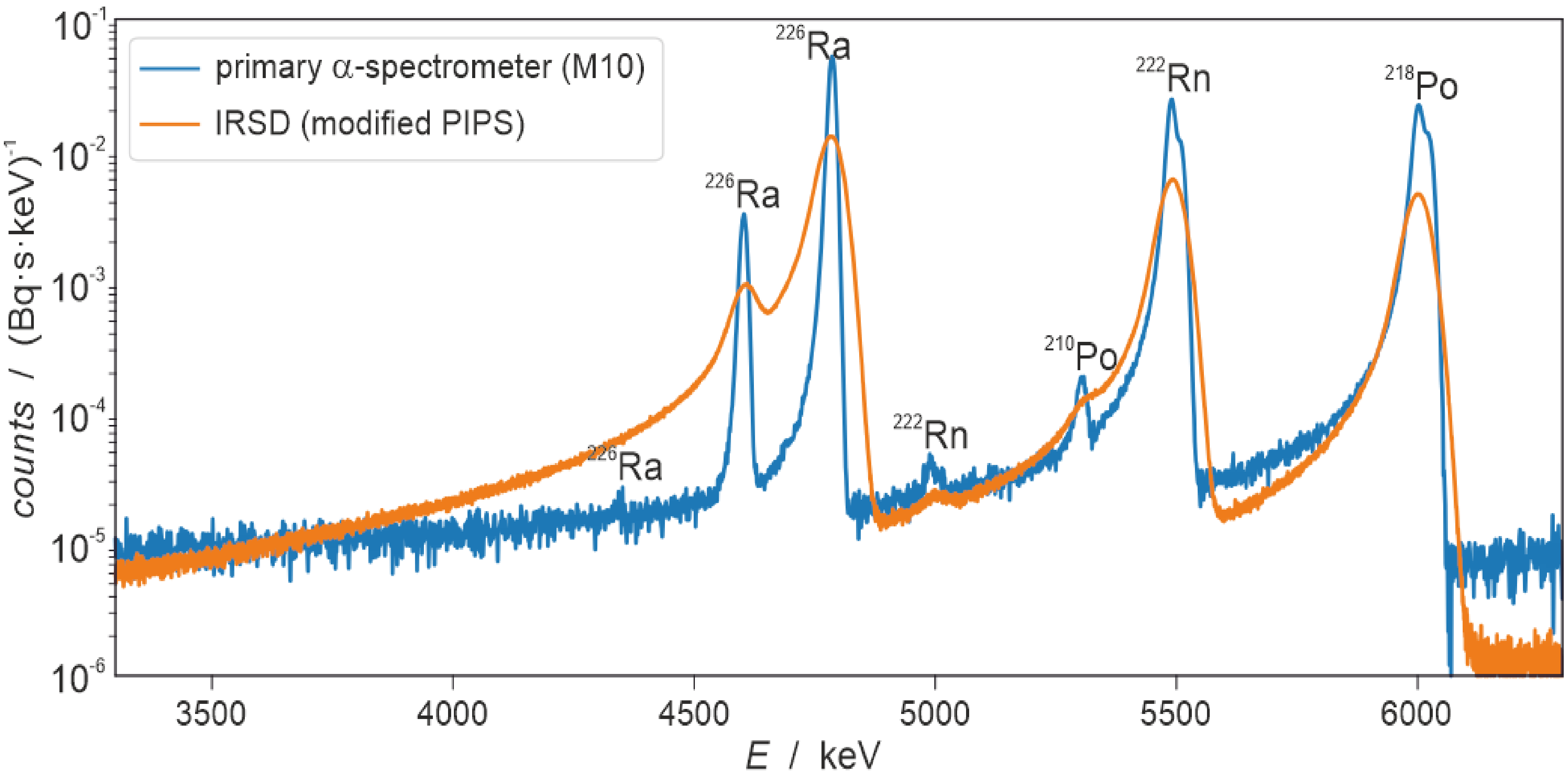


Primary ^{226}Ra characterisation

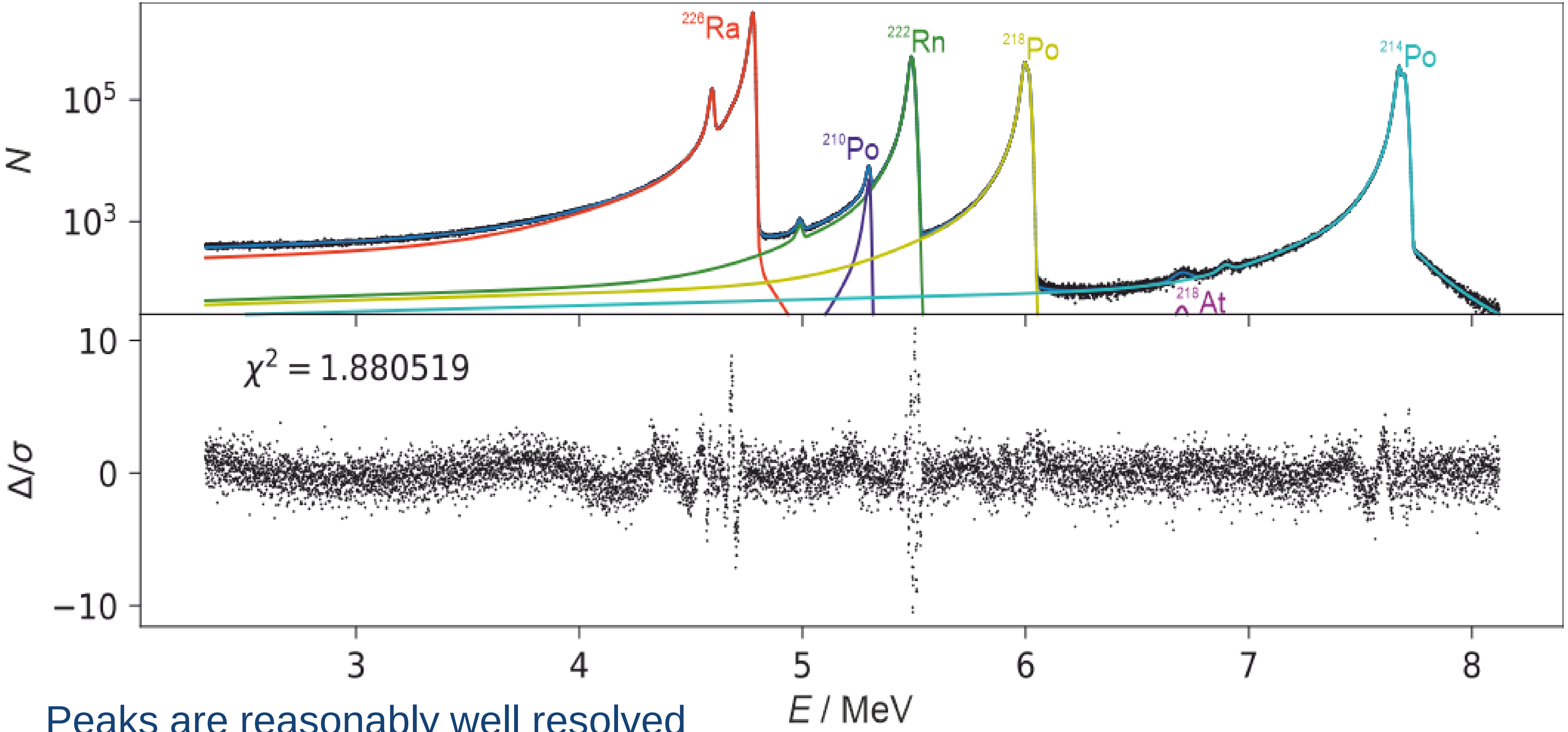


Primary ^{226}Ra characterisation

α-spectra - comparison



α-spectrum of the IRSD



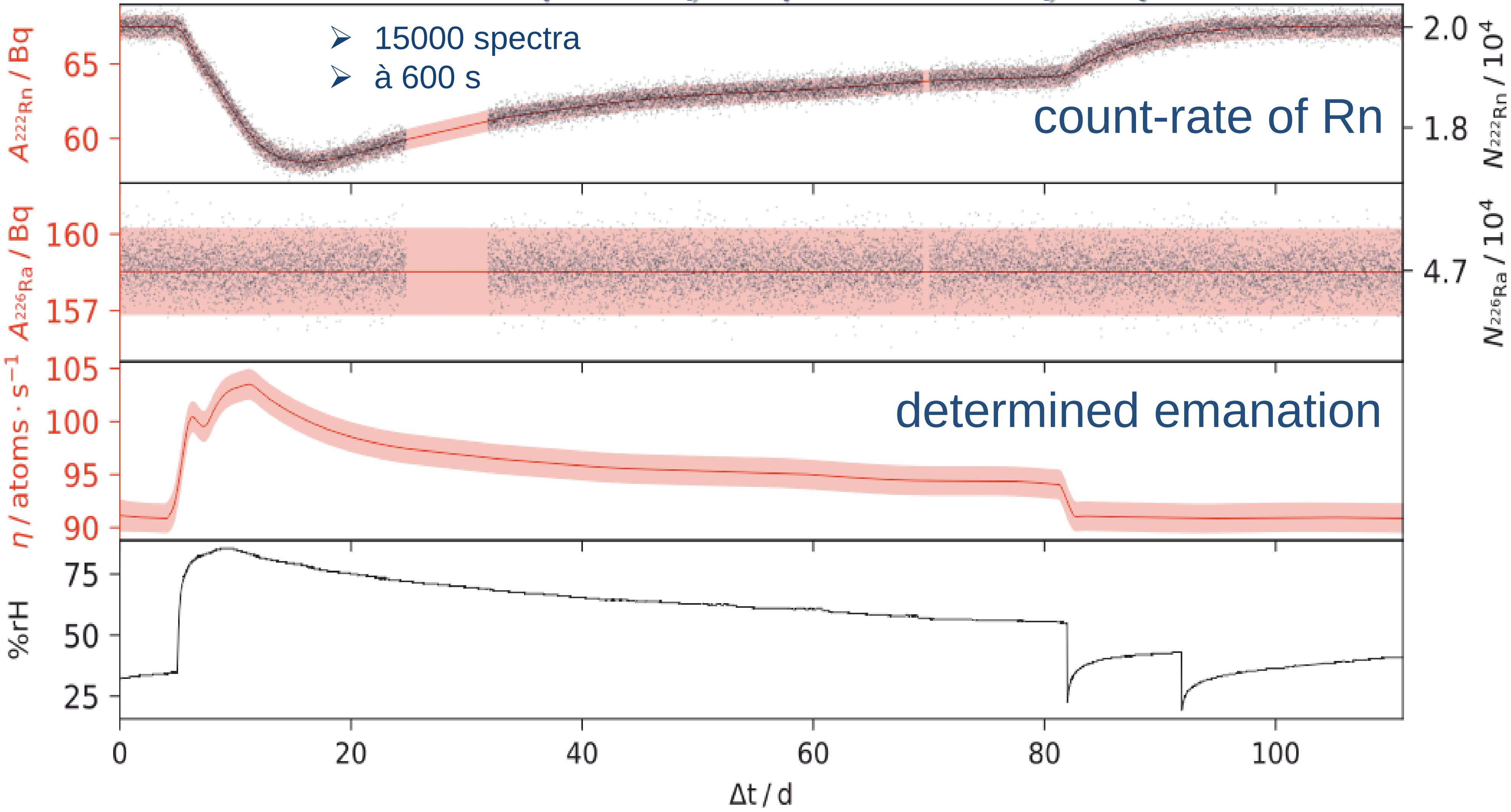
Peaks are reasonably well resolved

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

Gaussian process SLDS-decon.

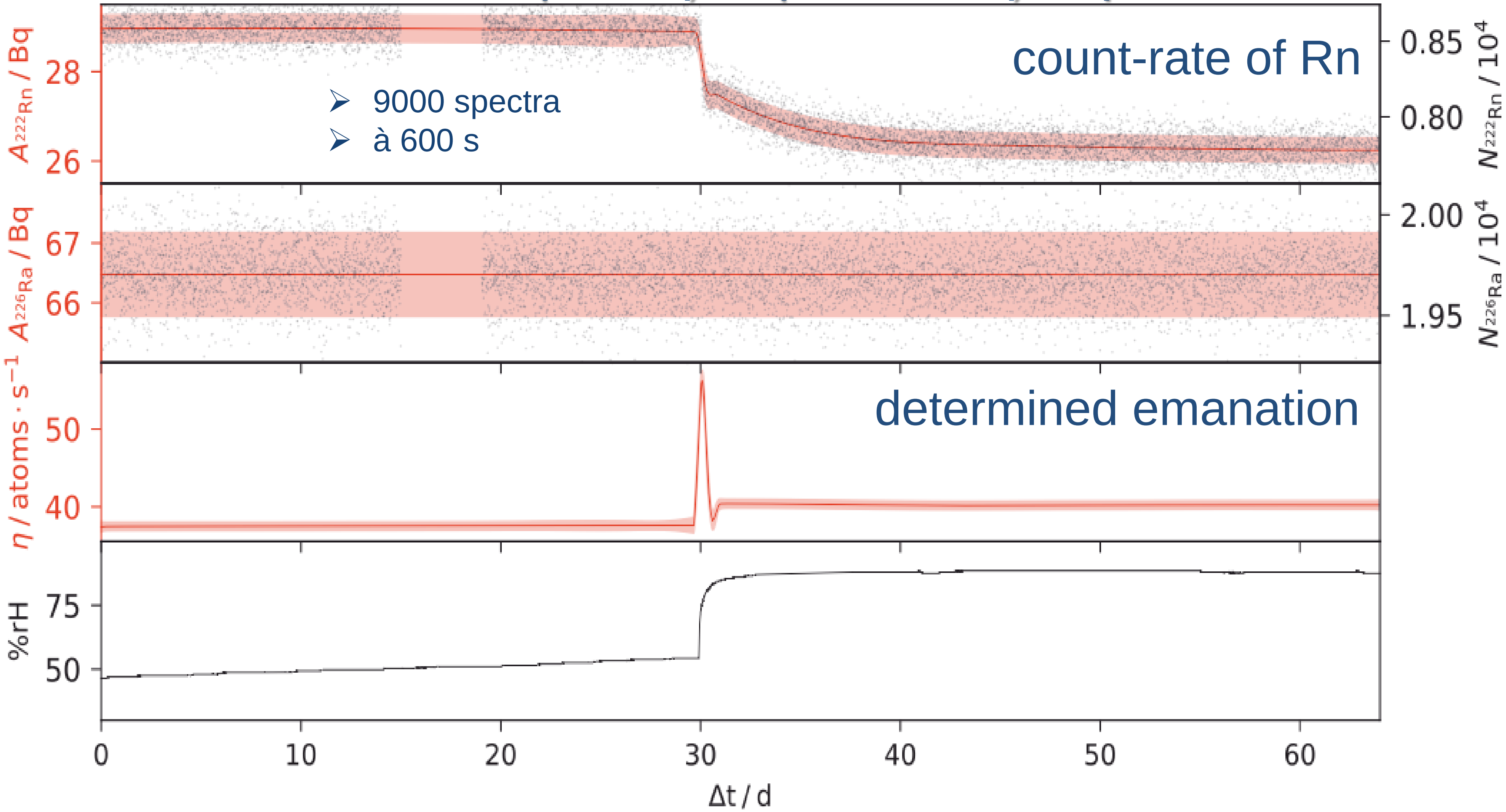
IRSD: A(^{226}Ra) = (158.6±1.7) Bq

- 15000 spectra
- à 600 s



Gaussian process SLDS-decon.

IRSD: A(^{226}Ra) = (66.4±0.5) Bq



IRSD – characteristics

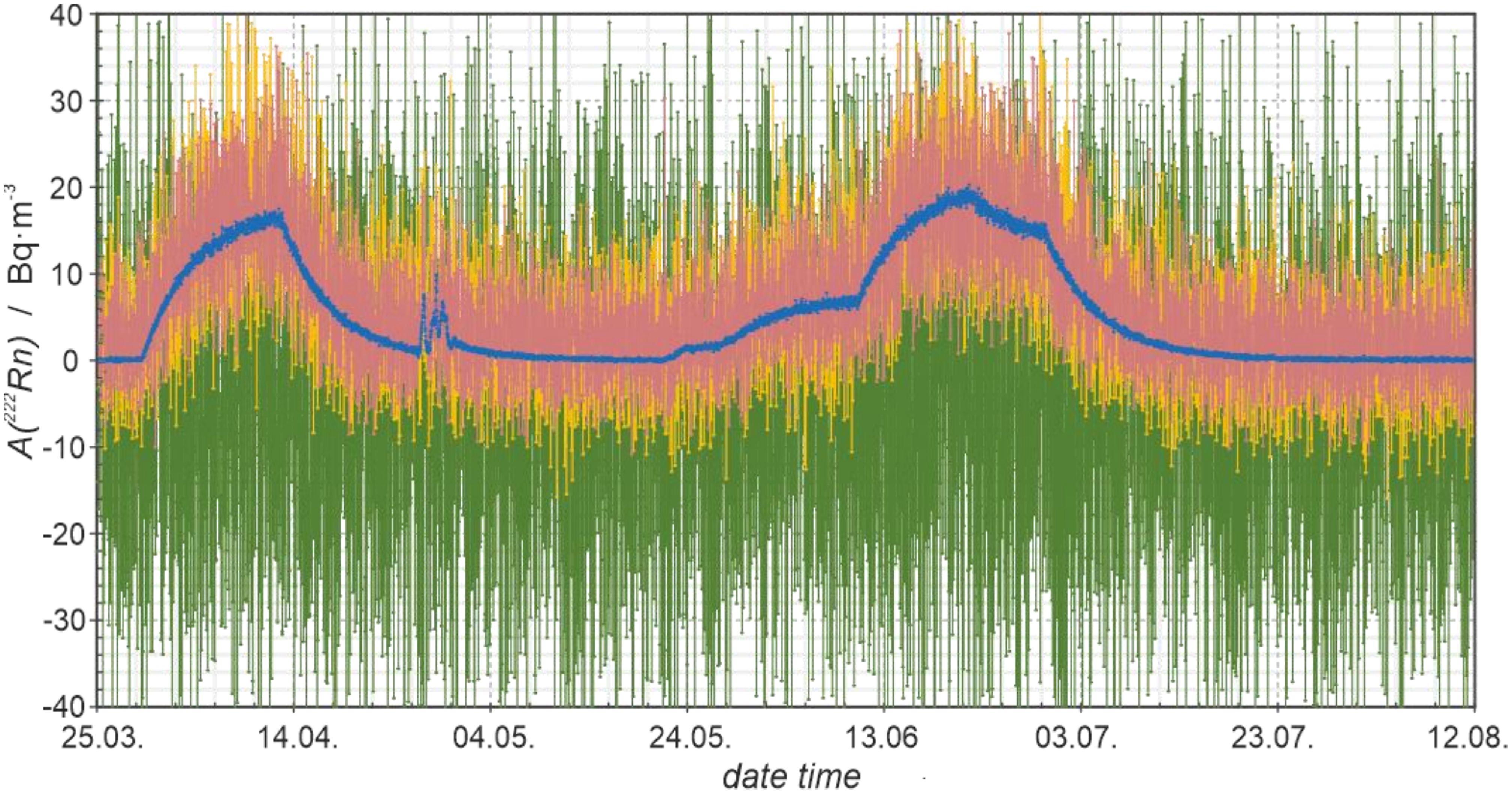
Produced IRSD systems:

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

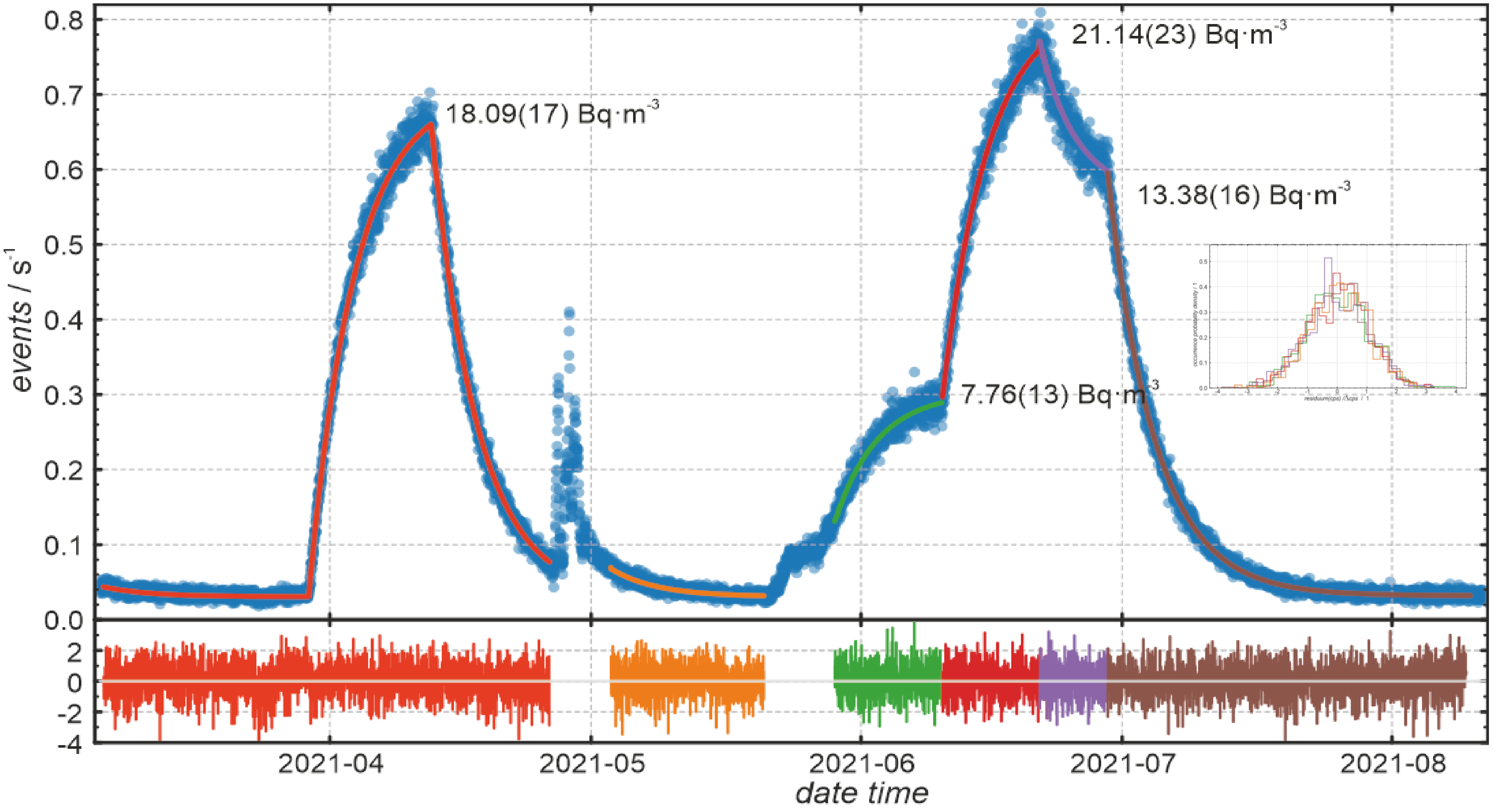
IRSD uncertainty budget for 2 Bq ^{226}Ra:

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

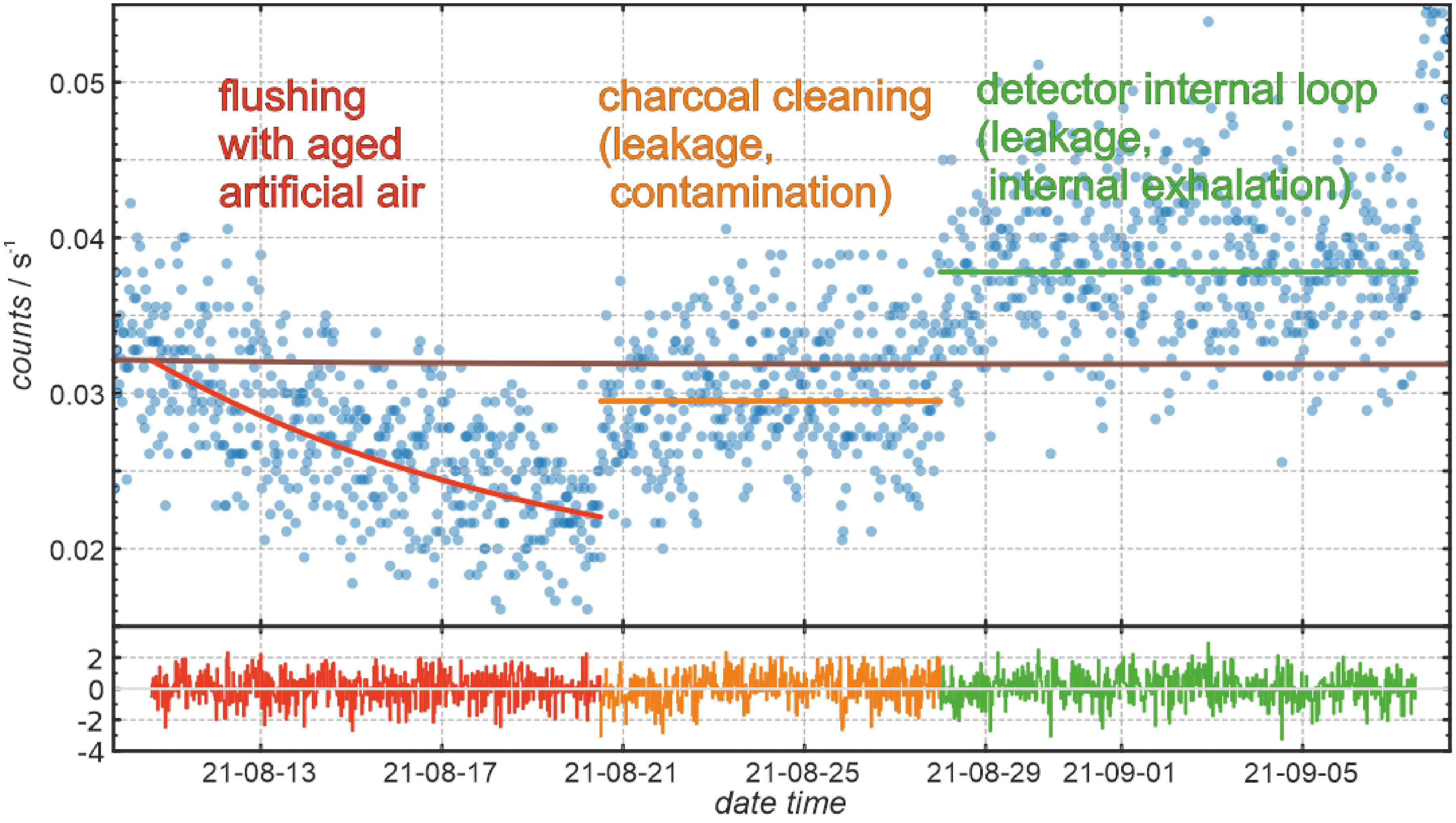
Radon detector calibration



calibration of ANSTO 200L



Background of ANSTO 200L



Publication of results

https://doi.org/10.1016/j.apradiso.2023.110726
Applied Radiation and Isotopes 196 (2023) 110726

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Evolution of traceable radon emanation sources from MBq to few Bq

Stefan Röttger[a,*], Annette Röttger[a], Florian Mertes[a], Viacheslav Morosch[a], Tanita Ballé[a], Scott Chambers[b]

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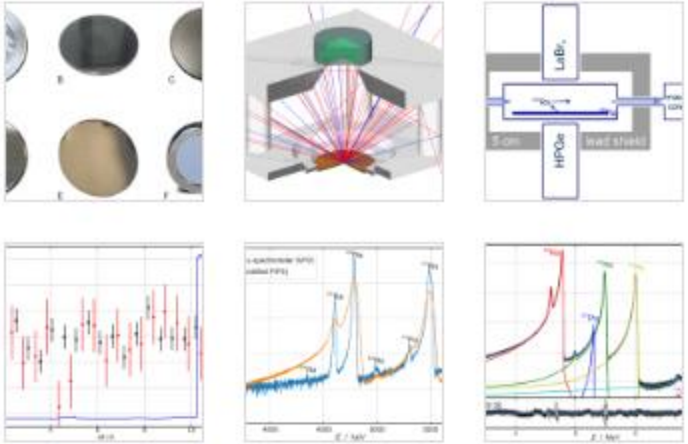
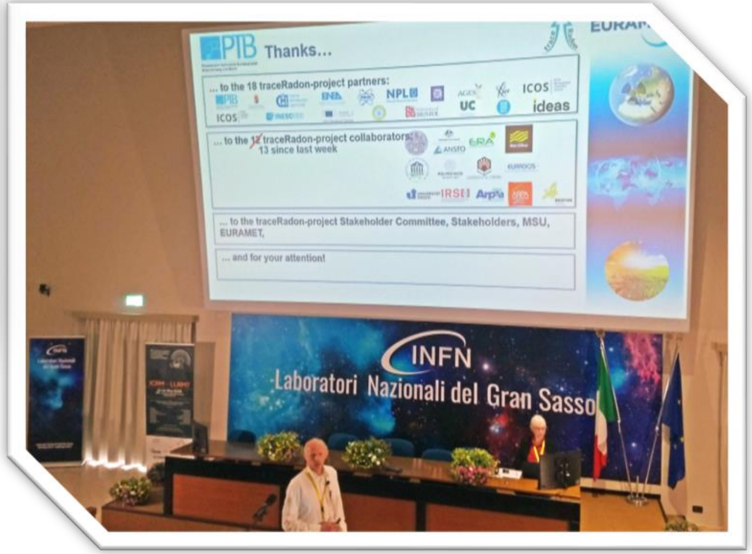
[b] Australian Nuclear Science and Technology Organisation (ANSTO), New Illawarra Road, Lucas Heights, NSW, 2234, Australia



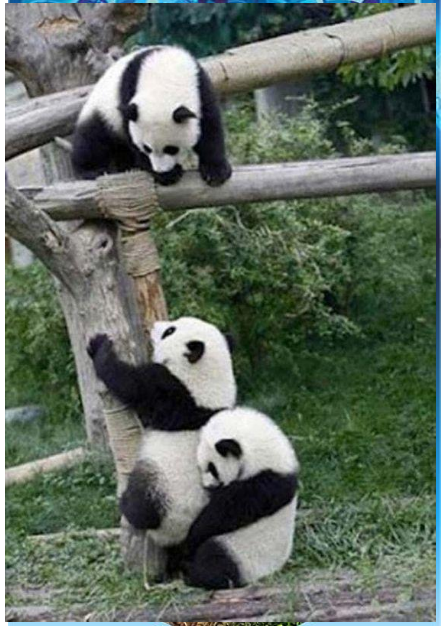
International Conference on Radionuclide Metrology – Low-Level Radioactivity Measurement Techniques

Plenary session,
S. Röttger



Conclusion

- ^{222}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



Thanks…

… to the traceRadon-project partners:

… to the traceRadon-project collaborators:

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

… and for your attention!