

New traceability chains for Radon: Metrology for use in climate change observation and radiation protection

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2nd International Conference on Applications of Radiation Science and Technology

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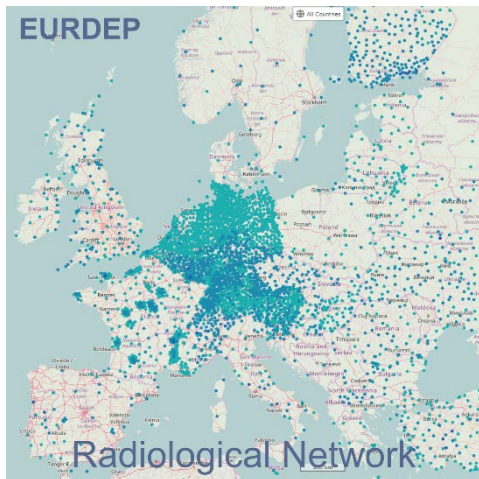
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Introduction – Why ?



Climate change is one of the greatest challenges of our time.

The temperature rise of the atmosphere of our planet, due to the greenhouse effect, is caused by the increase of GHG emissions.

- ICOS: Monitoring of GHG emissions, the dispersion of GHGs and the resulting GHG concentrations in air, is of utmost importance for appropriate climate change mitigation measures.
- EURDEP: Collection and exchange of radiological monitoring data between participating countries of the radiation in the environment.

Both networks could profit from radon measurements at the outdoor level. But **traceability to the SI system** is not established yet.



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



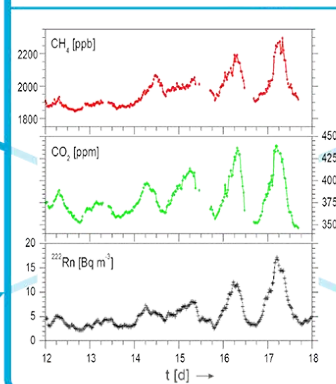
WP4

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

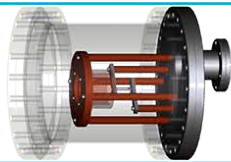
WP3

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



Traceable measurements of outdoor radon activity concentrations WP1

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



WP1

Radon flux measurements WP2

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



WP2

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.

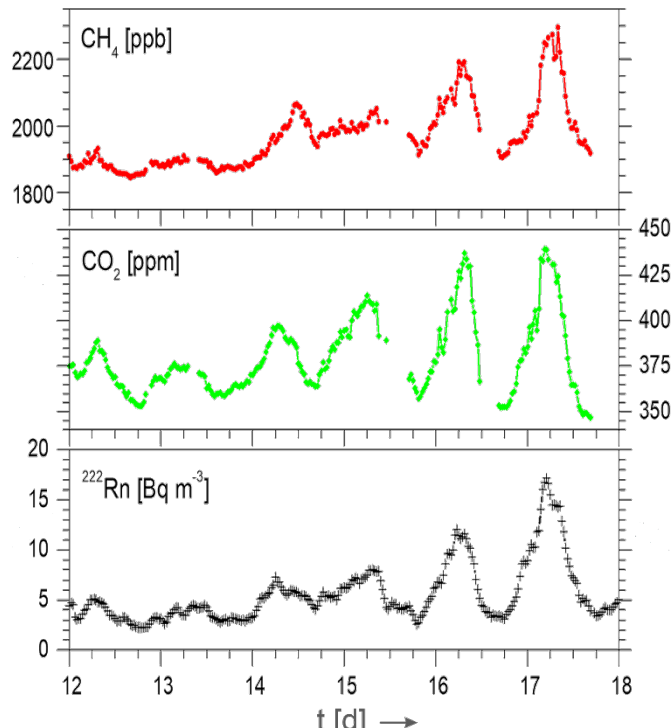
WP6





Why is Radon an issue in **climate observation**?

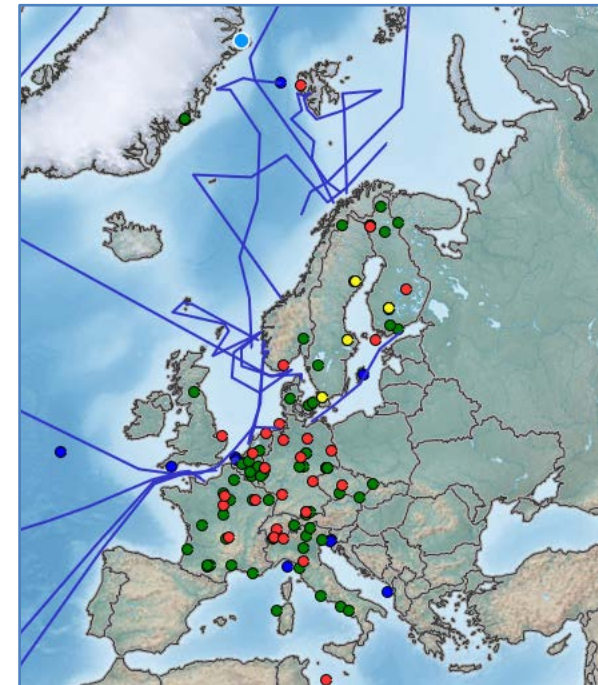
- **GHG flux measurements** are difficult though GHG concentration measurements are established.
- With radon activity concentration and radon flux measurements GHG fluxes can be **traced!**



ICOS Atmospheric Station Specifications:

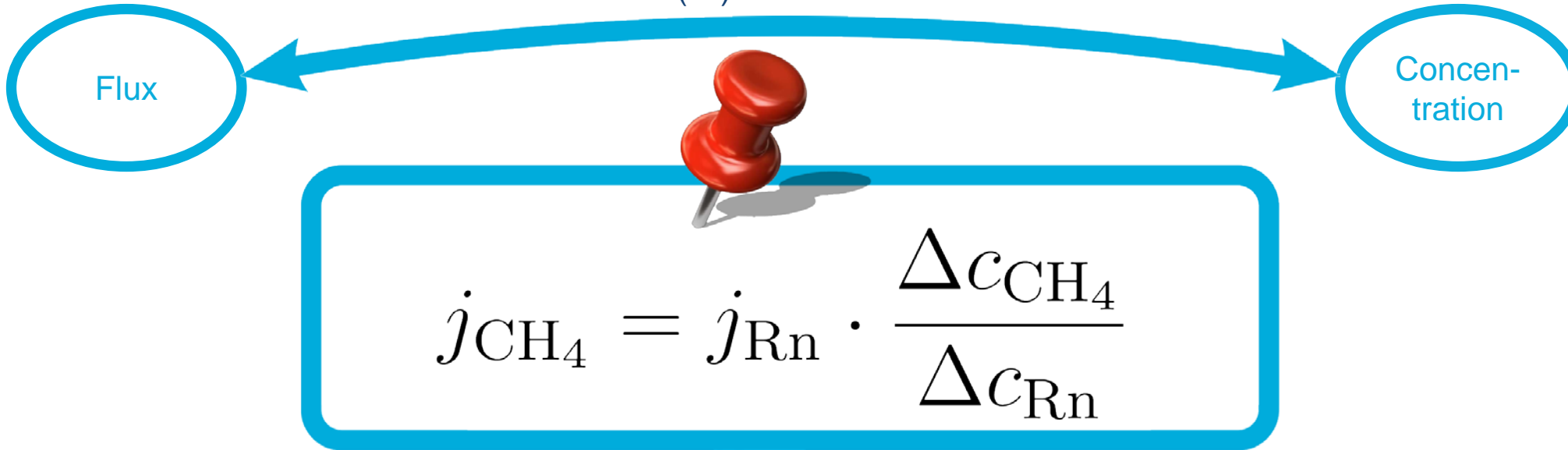
Radon monitor: “At the present stage, Radon-222 measurements are not mandatory in ICOS. However, Radon-222 is recognized as a very valuable measurement, in particular for trace gas flux estimates.”

- Determine source terms of GHG





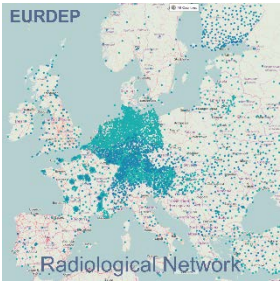
1. To develop traceable methods for the measurement of **outdoor low-level radon activity concentration** in the range of **1 Bq m⁻³ to 100 Bq m⁻³**, with uncertainties of **10 % for k = 1**, to be used in climate monitoring (...).
2. To develop the capability for traceable **radon flux measurements in the field**, based on the development of a radon exhalation reference system “exhalation bed” and a transfer standard (...).



3. To **validate current radon flux models and inventories** by the new traceable measurements of radon activity concentration and radon flux (...)



Objectives - Overview



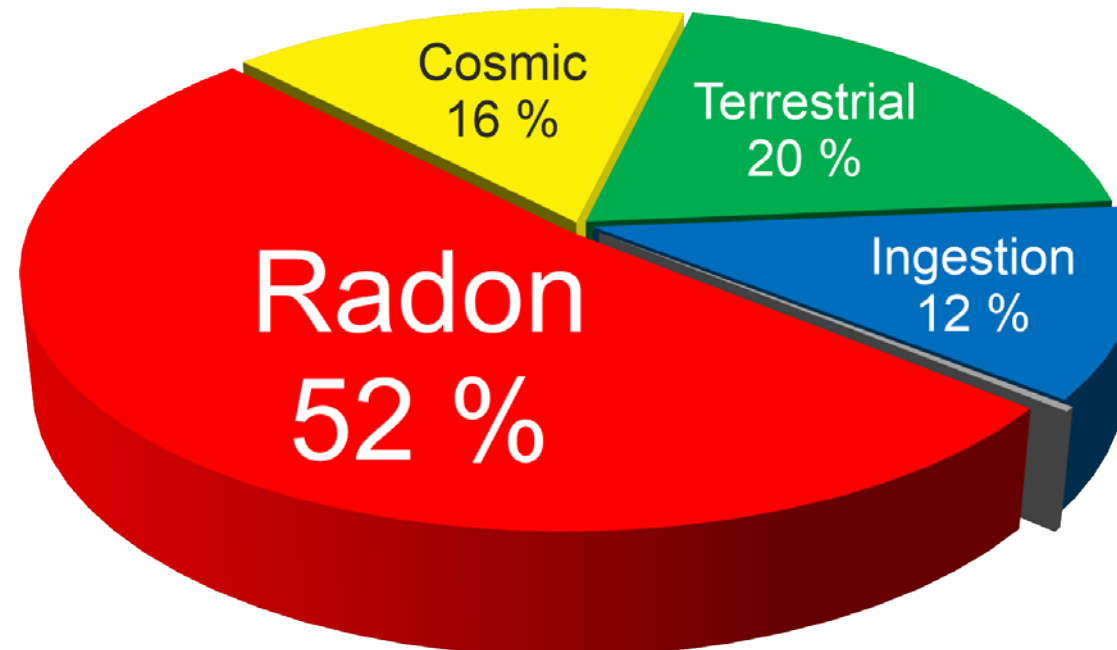
1. To develop traceable methods for the measurement of **outdoor low-level radon activity concentration** in the range of **1 Bq m⁻³ to 100 Bq m⁻³**, with uncertainties of **10 % for k = 1**, to be used in climate monitoring (...).
2. (...).
3. (...) To support the validation with dosimetric and spectrometric data from the **radiological early warning networks in Europe** (...).
4. To provide **easy to use dynamic radon and radon flux maps** for radiation protection in line with Council Directive 2013/59/EURATOM, including their use to identify **RPA and radon wash-out peaks** (...).

UNSCEAR, 2008:

Radon and its progeny contribute about half of the natural radiation dose to the public.

Public exposure to natural radiation:
Total average individual dose:

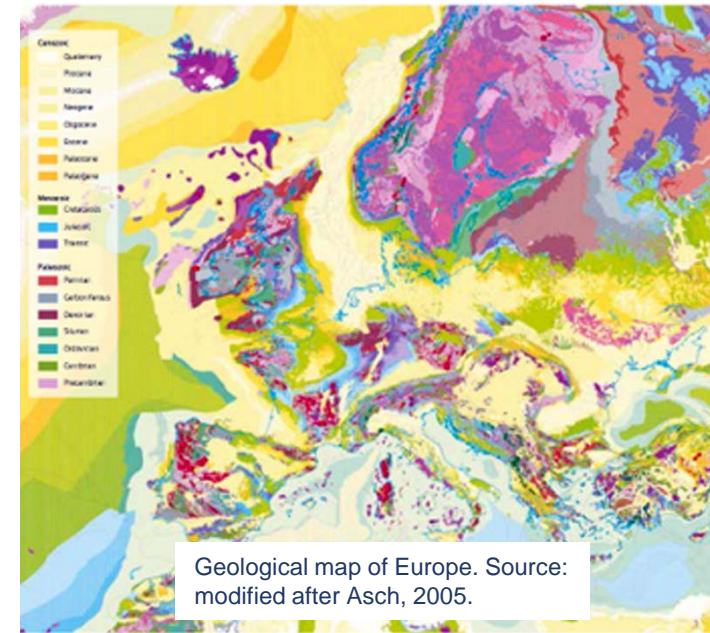
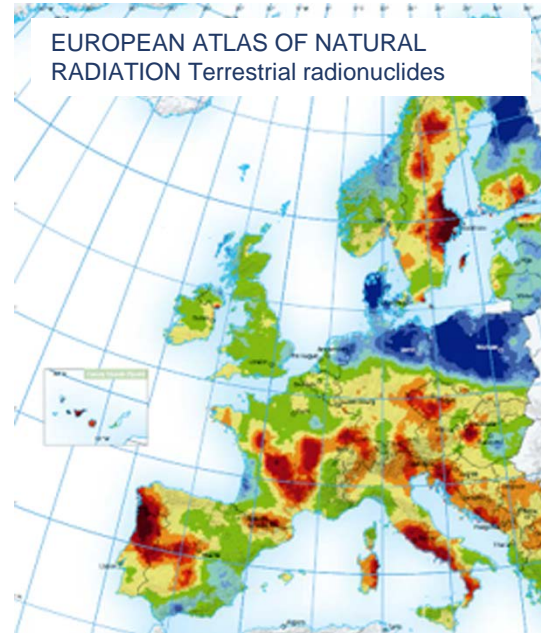
3 mSv a⁻¹



Objectives – static to dynamic

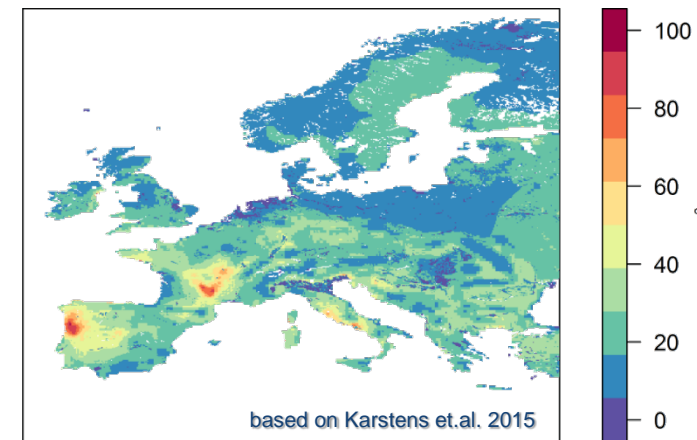
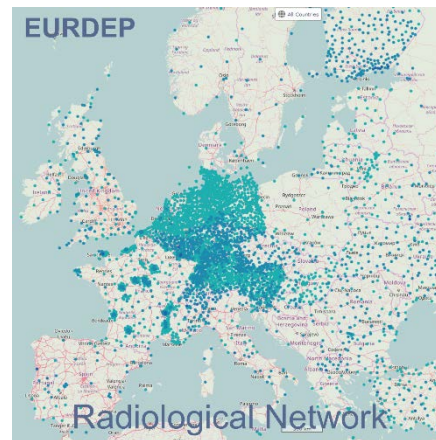


Static maps:



Dynamic maps:

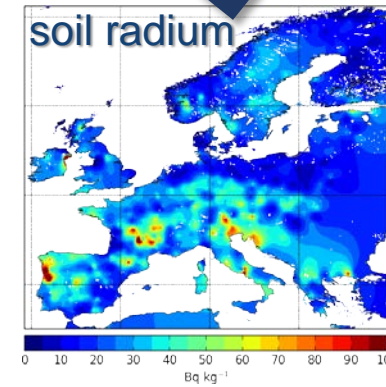
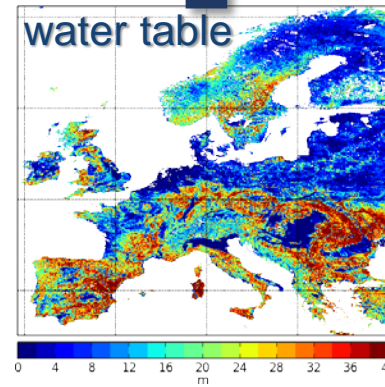
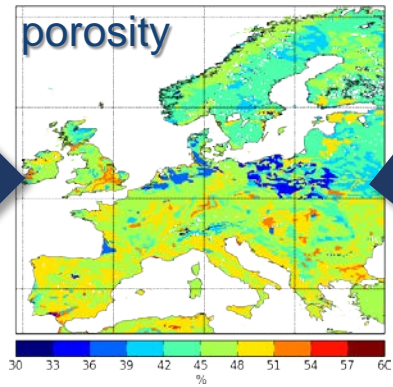
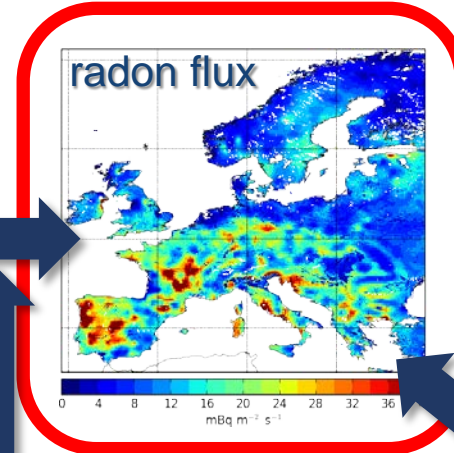
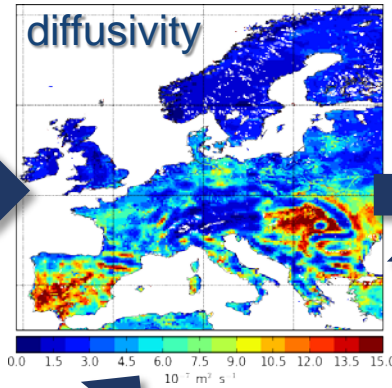
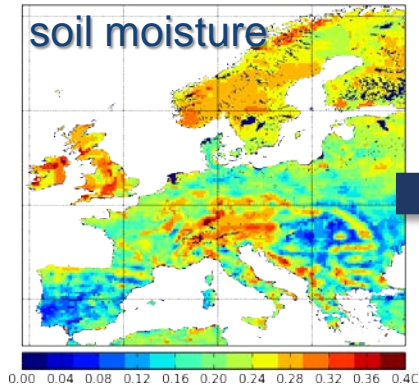
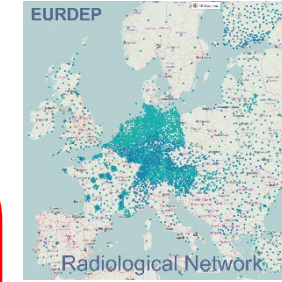
The early warning network shows online data for the dose rate. But **outdoor radon concentration** or even better online data on **radon flux** (emission) is missing!



Objectives – Joining forces



Our task: Joining forces in the field of radon



soil texture
% clay
% sand
% slit
bulk density





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New metrology for radon at the environmental level

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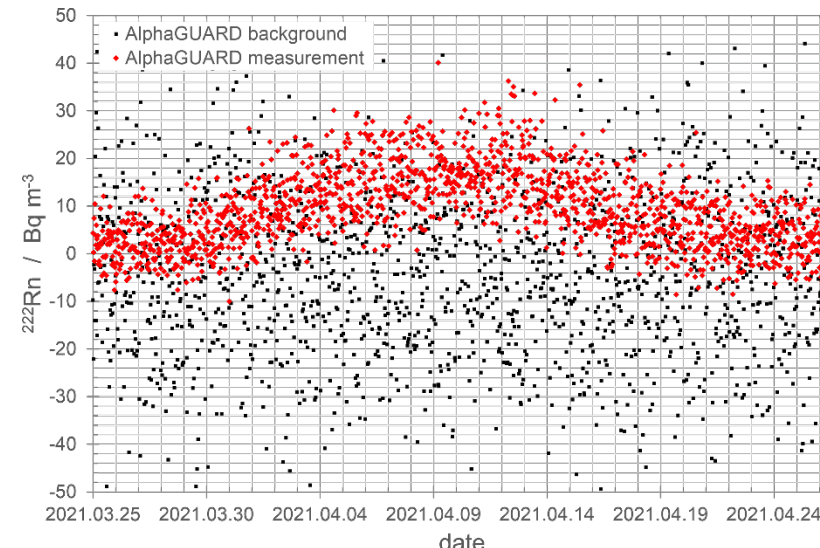
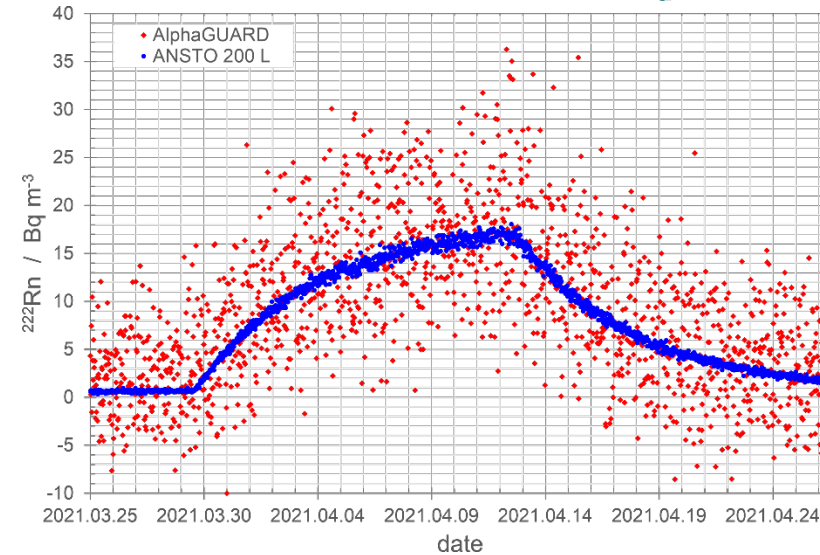
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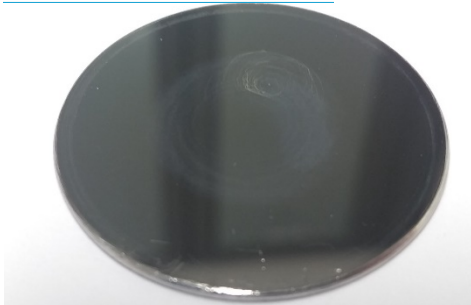
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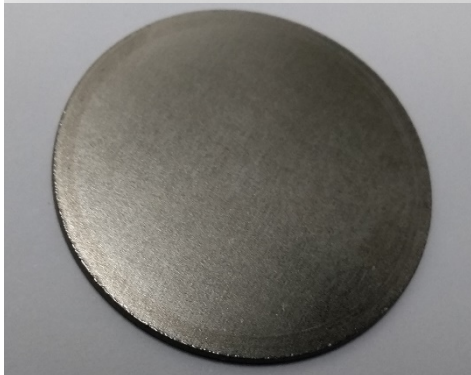
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Electro-deposited

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



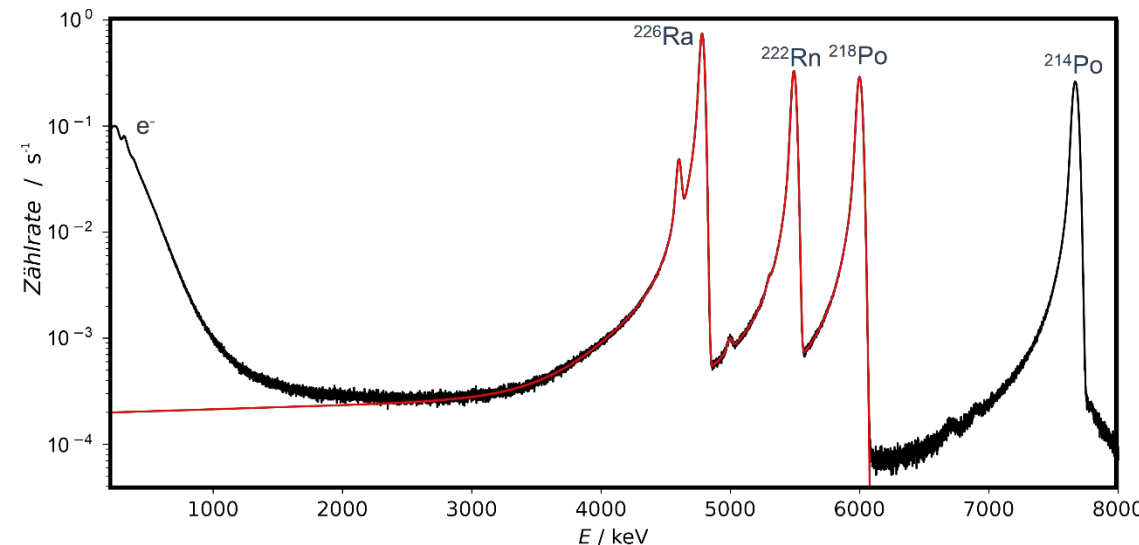
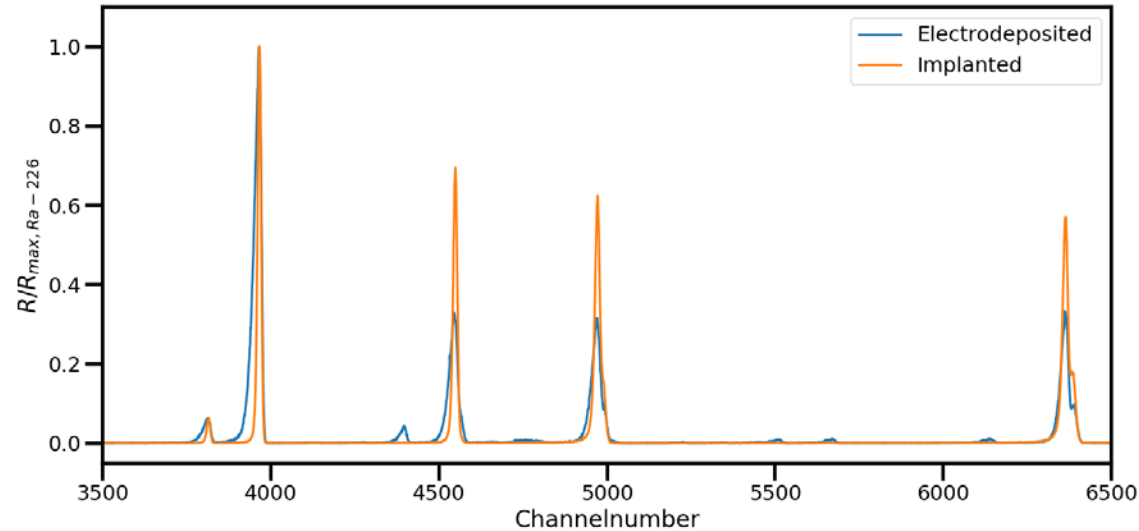
Implanted

Implantation of
Ra-226 into W / Al
after mass
separation



PIPS

450 mm², 300 μm
with 150 Bq ²²⁶Ra
layer





Article

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

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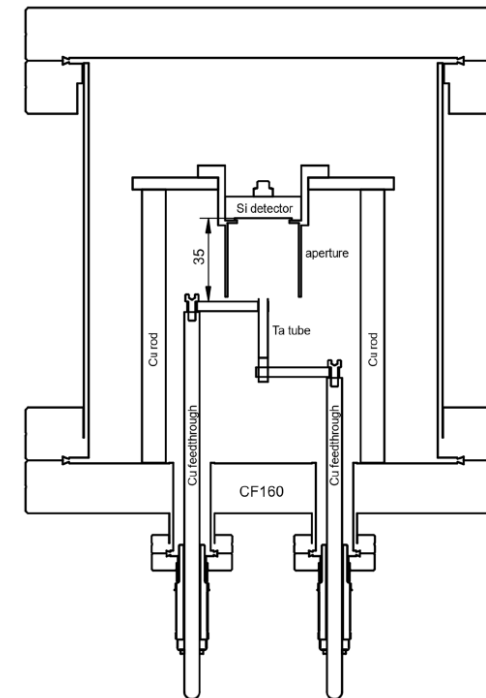
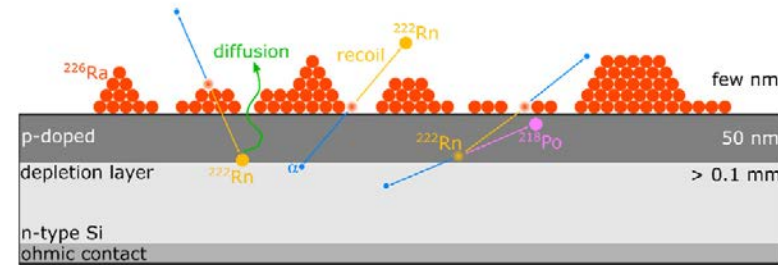
* Correspondence: florian.mertes@ptb.de

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 $^{226}\text{RaCl}_2$ 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 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

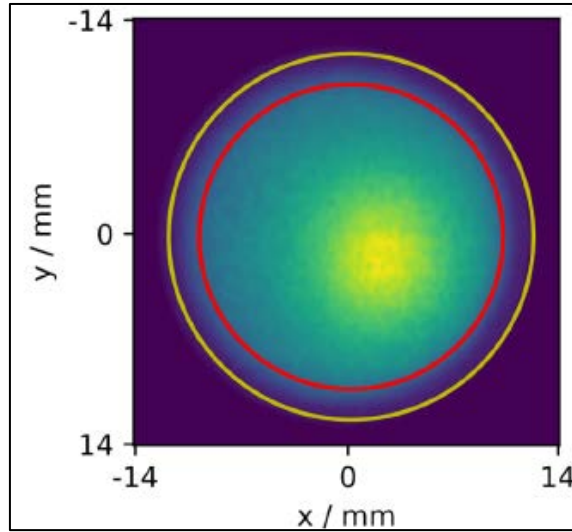
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<https://www.mdpi.com/1660-4601/19/2/840>



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>

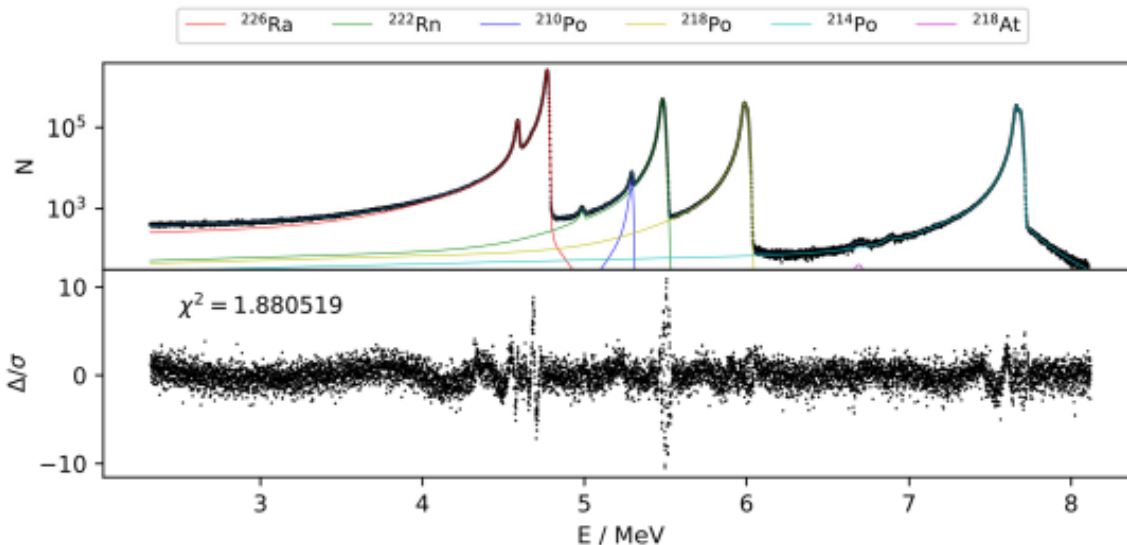




Photograph of an IRSD based on a 450 mm² Canberra PIPS® detector, modified with a layer containing 440 Bq ²²⁶RaCl₂ (left). A digital autoradiograph obtained from such a deposit where the inner diameter of the recessed Si-surface is given in yellow, and the diameter of the shadowing aperture is given in red (right).

What metrology can provide:

Description and Type	Value and Uncertainty	Rel. Uncertainty
Solid angle (systematic)	(0.00940 ± 0.00006) 4π sr	0.6%
Backscattering _{DSA} (systematic)	1 ± 0.002	0.2%
Tailing _{DSA} (systematic)	1 ± 0.003	0.3%
Tailings _{Si} (systematic)	1 ± 0.003	0.3%
²²⁶ Ra rate _{DSA} (stochastic)	(0.01796 ± 0.00015) s ⁻¹	0.8%
²²⁶ Ra rate _{Si} (stochastic)	(0.9595 ± 0.0004) s ⁻¹	0.04%
ϵ_{Ra-226}	0.502 ± 0.006	1.2%



Detector Type	Active Area/Depletion Depth	A(²²⁶ Ra)/Bq	ϵ_{Ra-226} /cps Bq ⁻¹
Mirion PIPS®	450 mm ² /300 μm	1.91 ± 0.02	0.502 ± 0.006
Ametek Ortec ULTRA®	450 mm ² /300 μm	66.4 ± 0.5	0.494 ± 0.004
Mirion PIPS®	450 mm ² /300 μm	158.6 ± 1.7	0.494 ± 0.005
Mirion PIPS®	450 mm ² /100 μm	442 ± 4	0.492 ± 0.005

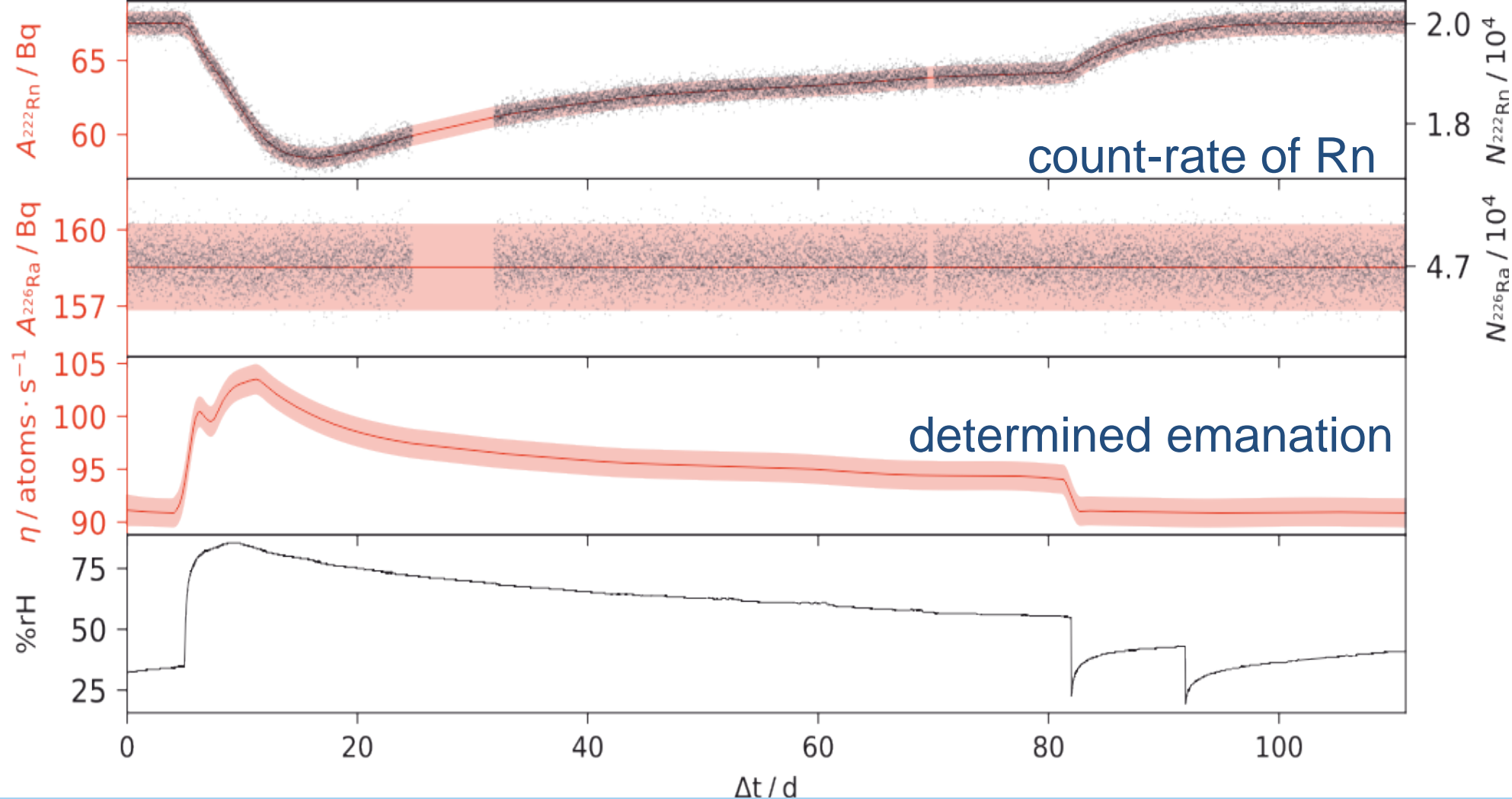


Achievements – 2d: Radon Activity



Switching Linear Dynamic System-deconvolution

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

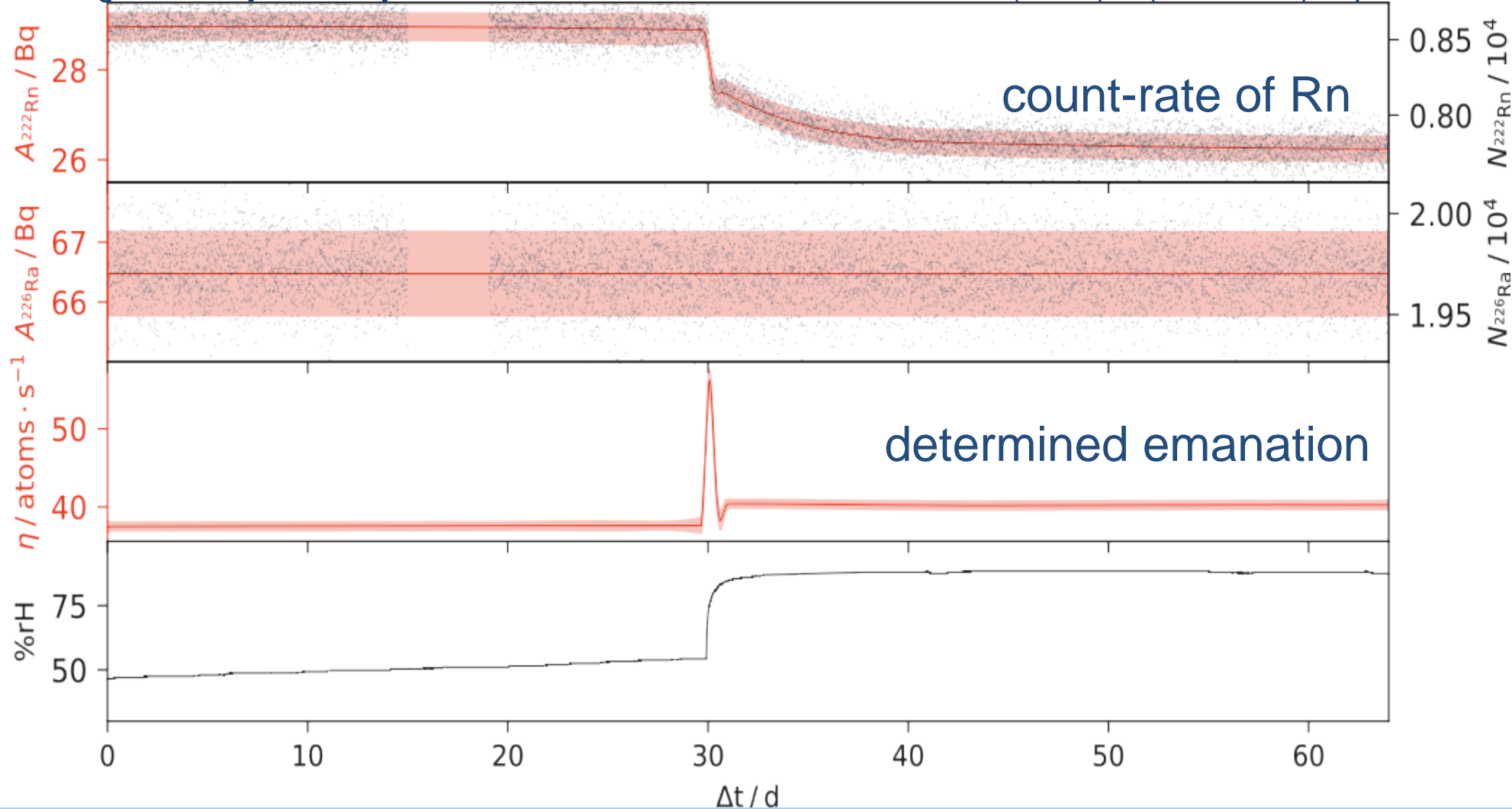


Achievements – 2d: Radon Activity



Switching Linear Dynamic System-deconvolution

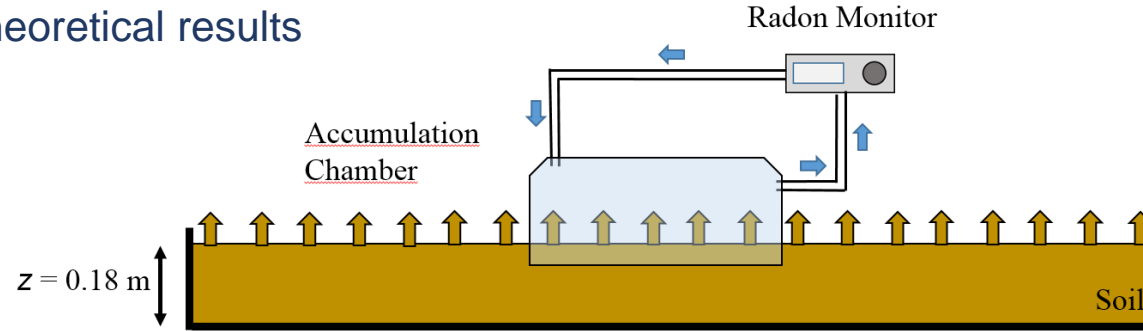
IRSD: $A(^{226}\text{Ra}) = (66.4 \pm 0.5) \text{ Bq}$





Calibration

Continuous radon flux systems according the experimental and theoretical results



Inter-comparison

- Good agreement between participants
- Static period is used to determine the leakages of the system and the applicability of linear assumption
- Integration time and device sensitivity are key to determine the radon flux

Next steps:

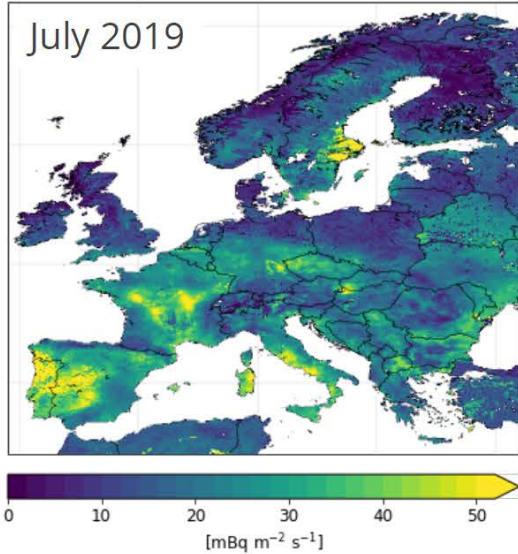
- Further data analysis (increasing period), optimize the methodology, check the time of linear assumption
- Produce the guidelines to installation and operation in field



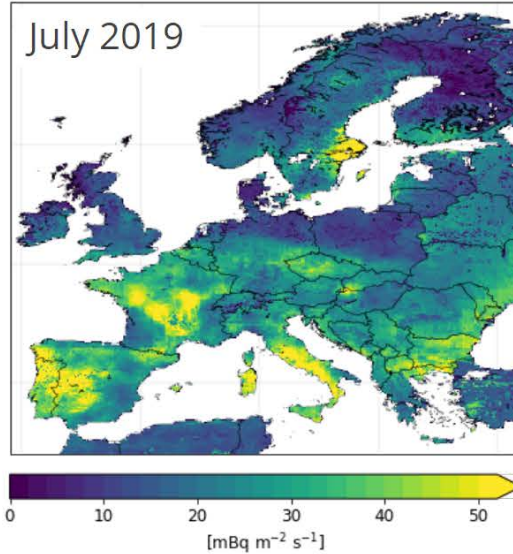
Achievements – 4: Data reanalysis



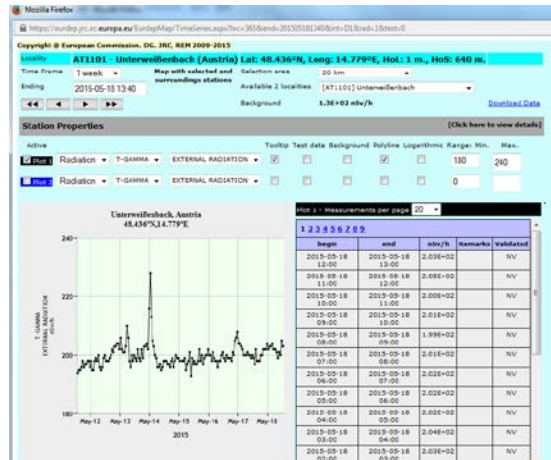
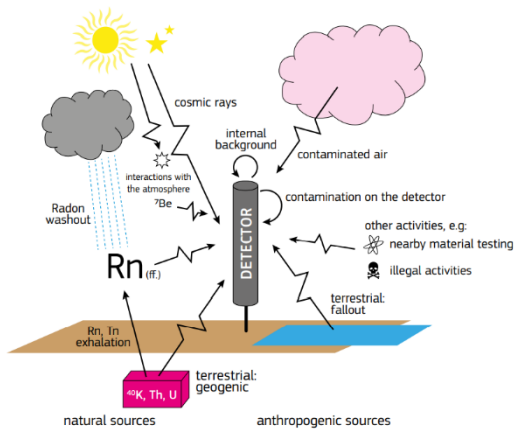
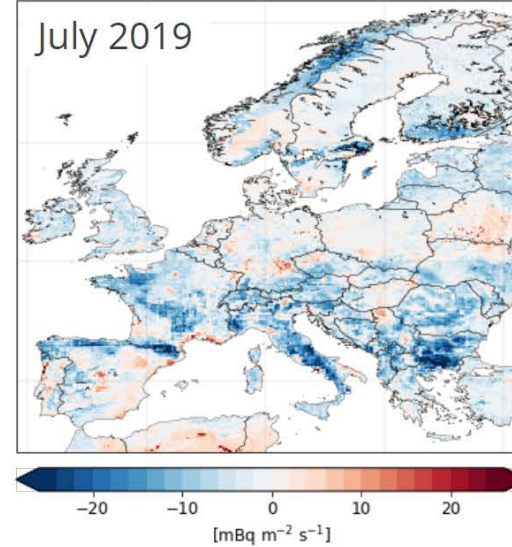
Radon flux based on
GLDAS-Noah v2.1 soil moisture



Radon flux based on
ERA5-Land soil moisture



Radon flux difference
ERA5-Land – GLDAS-Noah



The identification of the right origin of Ambient Dose Equivalent Rate (ADER) peaks is a crucial issue to prevent the impact of false alarm in the population.





The key targets to be reached by the end of this project (and to be exploited in the 5 years that follow the end of the project) are as follows:

- **New SI traceability** for measurement quantities used in climate observation and radiation protection;
- **New customer calibration services** for new types of measurement and new types of device. To develop a **first standard protocol for the application of the radon tracer method (RTM)** to enable retrieval of greenhouse gas fluxes at atmospheric climate gas monitoring stations and to use radon flux data for the identification of **Radon Priority Areas (RPA)**;
- To **validate current radon flux models and inventories** by the new traceable measurements of radon activity concentration and radon flux. To **support the validation with dosimetric and spectrometric data from the radiological early warning networks** in Europe;
- To provide easy to use **dynamic radon activity concentration and radon flux maps** for climate change research and radiation protection in line with Council Directive 2013/59/EURATOM, including their use to identify RPA and radon wash-out peaks;
- To facilitate the **take up of the technology and measurement infrastructure**.



1. Mertes, F et. al.: D3.3 Approximate sequential Bayesian filtering to estimate Rn-222 emanation from Ra-226 sources from spectra, <https://doi.org/10.5162/SMSI2021/D3.3>
2. Röttger, A. et al: *New metrology for radon at the environmental level 2021 Meas. Sci. Technol.* 32, 124008, <https://doi.org/10.1088/1361-6501/ac298d>
3. Radulescu, I et al.: Inter-comparison of commercial continuous radon monitors responses, Nuclear Instruments and Methods in Physics Research Section A, Volume 1021, 2022, 165927, <https://doi.org/10.1016/j.nima.2021.165927>
4. Mertes, F. et. al.: Ion implantation of ^{226}Ra for a primary ^{222}Rn emanation standard, Applied Radiation and Isotopes, Volume 181, March 2022, 110093, <https://doi.org/10.1016/j.apradiso.2021.110093>
5. Ćeliković, I. et. al.: Outdoor Radon as a Tool to Estimate Radon Priority Areas - A Literature Overview, Int. J. Environ. Res. Public Health 2022, 19, 662, <https://doi.org/10.3390/ijerph19020662>
6. Mertes, F et. al.: 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>
7. Rábago, D. et al.: Intercomparison of Radon Flux Monitors at Low and at High Radium Content Areas under Field Conditions, Int. J. Environ. Res. Public Health 2022, 19, 4213, <https://doi.org/10.3390/ijerph19074213>
8. Röttger, S. et al: Radon metrology for use in climate change observation and radiation protection at the environmental level, Adv. Geosci., 57, 37–47, 2022, <https://doi.org/10.5194/adgeo-57-37-2022>
9. Chambers, S. et al: Portable two-filter dual-flow-loop ^{222}Rn detector: stand-alone monitor and calibration transfer device, Adv. Geosci., 57, 63–80, 2022, <https://doi.org/10.5194/adgeo-57-63-2022>

This list is also available here: <https://www.euramet.org/repository/research-publications-repository-link/>



Last continent reached: Antarctica!



Thank you and thanks to

... to the 18 traceRadon-project partners:



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... to the 14 traceRadon-project collaborators:



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