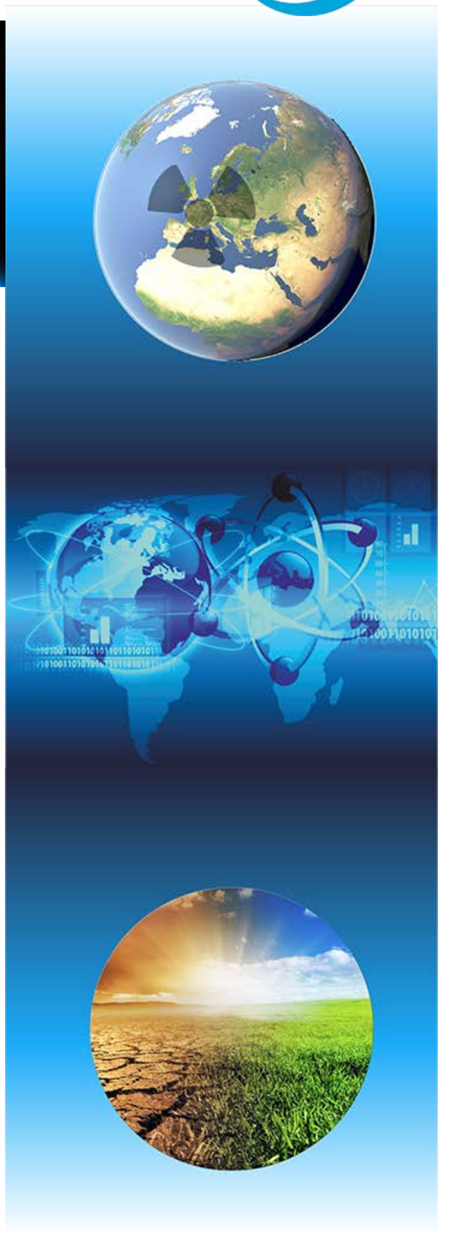
The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States



EXPLOTATION OF RESULTS: RADON METROLOGY FOR USE IN CLIMATE CHANGE OBSERVATION AND RADIATION PROTECTION

A. RÖTTGER[1*], S. RÖTTGER[1], F. MERTES[1], V. MOROSH[1], L. SZÜCS[2], C. YVER-KWOK[3], M. FUENTE[3], C. GROSSI[4], A. VARGAS[4], M. CAPOGNI[5], F. CARDELLINI[5], G. IURLARO[6], L. SILVI[5], L. SPERANDIO[6], A. RIZZO[6], I. VILARDI[6]

[1]Ionizing Radiation, Physikalisch-Technische Bundesanstalt, Braunschweig, Germany
[2]Section of Radiation Physical and Chemical Measurements, Budapest Főváros Kormányhivatala , Hungary
[3]Laboratoire des Sciences du Climat et de l'Environnement (LSCE/IPSL), CEA-CNRS-UVSQ, Université Paris-Saclay, 91191 Gif-sur-Yvette, France
[4]Institut de Tècniques Energètiques, Universitat Politècnica de Catalunya, Barcelona, 08028, Spain
[5]ENEA, National Institute of Ionizing Radiation Metrology (INMRI), C.R Casaccia, Via Anguillarese 301, 00123 Rome, Italy
[6]ENEA, Radioprotection Institute, C.R Casaccia, Via Anguillarese 301, 00123 Rome, Italy

A. Röttger, Coordinator of 19ENV01 traceRadon | 6th European Congress on Radiation Protection

Physikalisch-Technische Bundesanstalt
Braunschweig und Berlin

EMPIR: Funding 2019, Start 2020

EUROPEAN METROLOGY PROGRAMME FOR INNOVATION AND RESEARCH (EMPIR)

EMPIR is the main programme for European research on metrology. It coordinates research projects to address grand challenges, while supporting and developing the SI system of measurement units.

EMPIR follows on from the successful European Metrology Research Programme (EMRP), which issued its final call for projects in 2013. There is an increased focus within EMPIR on innovation activities to target the needs of industry and accelerate the uptake of research outputs.

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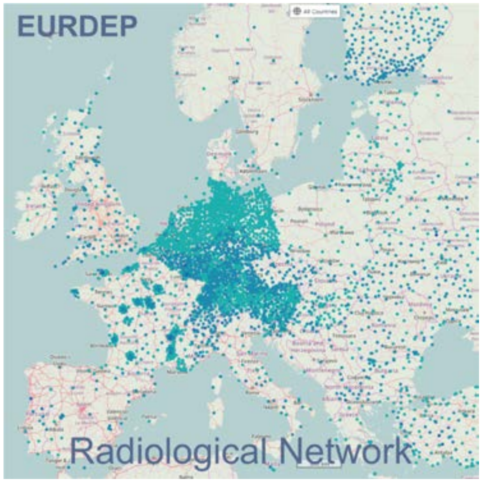
Introduction – Why ?



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.

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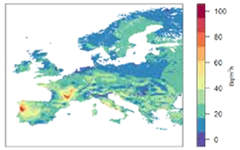
Introduction – What ?

Traceability to the SI system

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

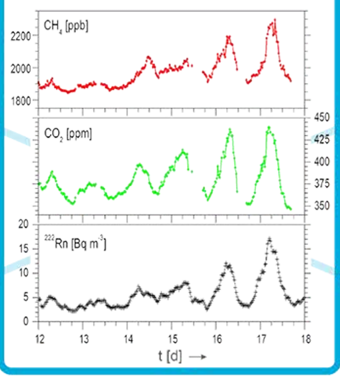
- Identification of RPA
- Quantifying the radon wash-out peak
- Data accessibility and public engagement



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

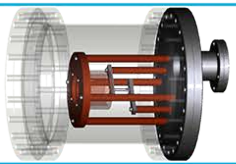
- Radon flux maps in GHG and climate change studies
- Inclusion of data from radiological early warning systems
- Validation of radon flux maps using radon flux measurements and outdoor radon activity concentrations

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



Traceable measurements of outdoor radon activity concentrations — WP1

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



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.

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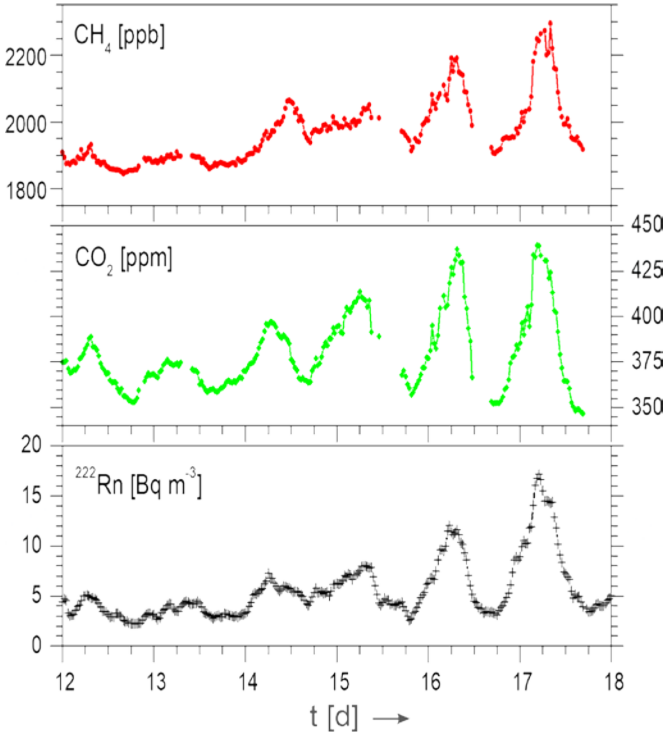
Introduction – For whom?

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

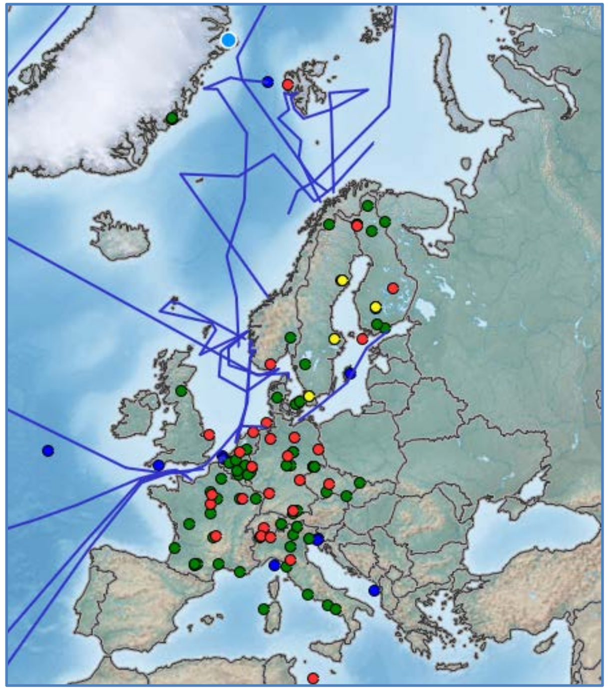
- ➢ GHG **flux measurements** are difficult though GHG concentration measurements are established.
- ➢ With radon activity concentration and radon flux measurements GHG fluxes can be **traced**!



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



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Introduction – Radon Tracer Method

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 (…) .

Flux

Concen-
tration

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

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

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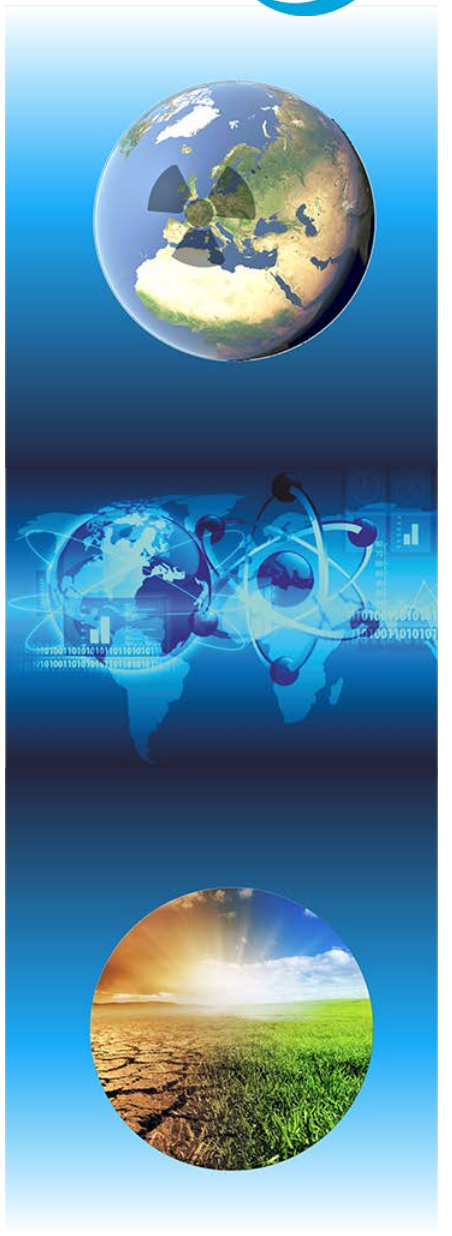
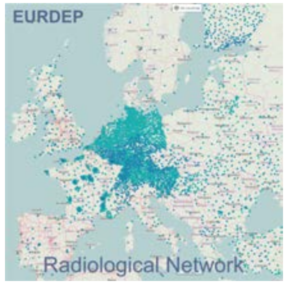


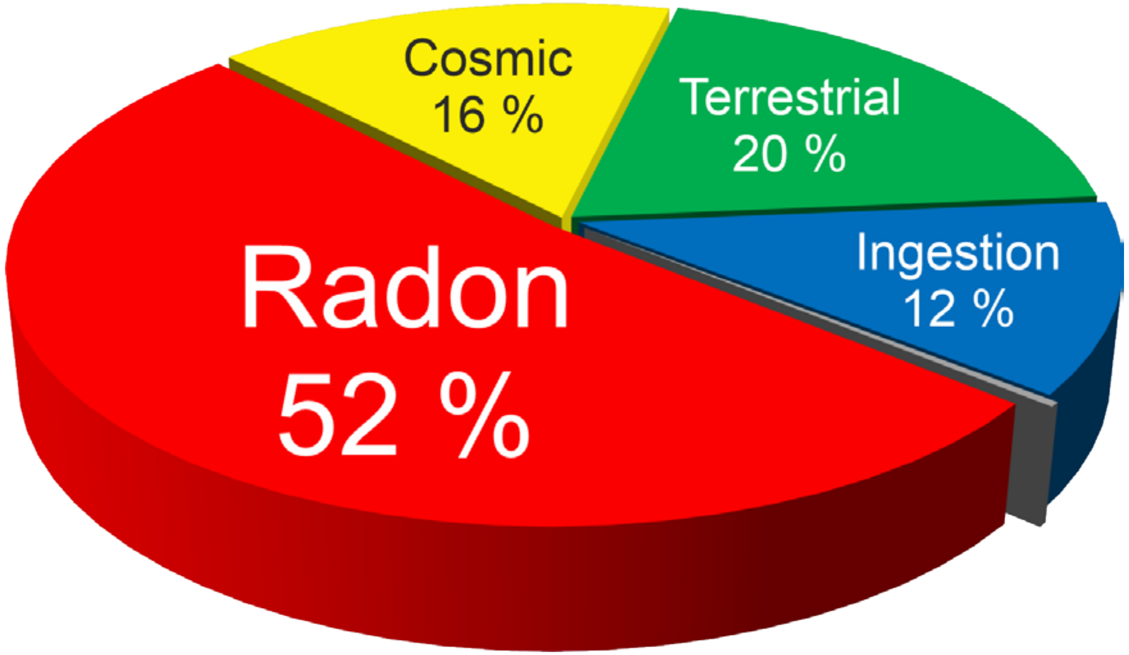
Objectives - Overview



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}



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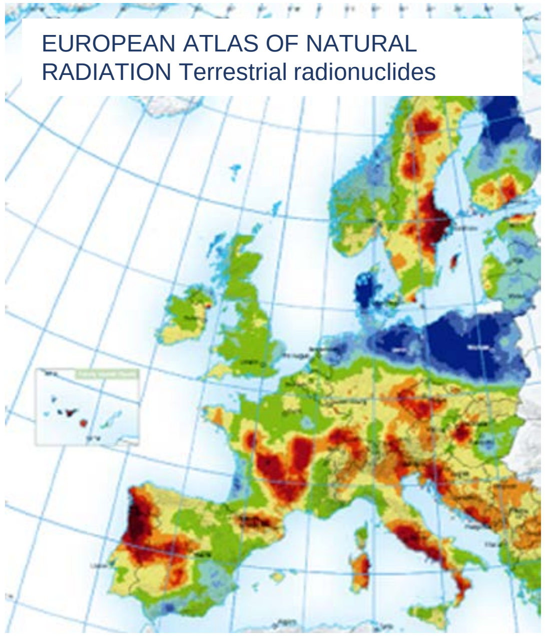
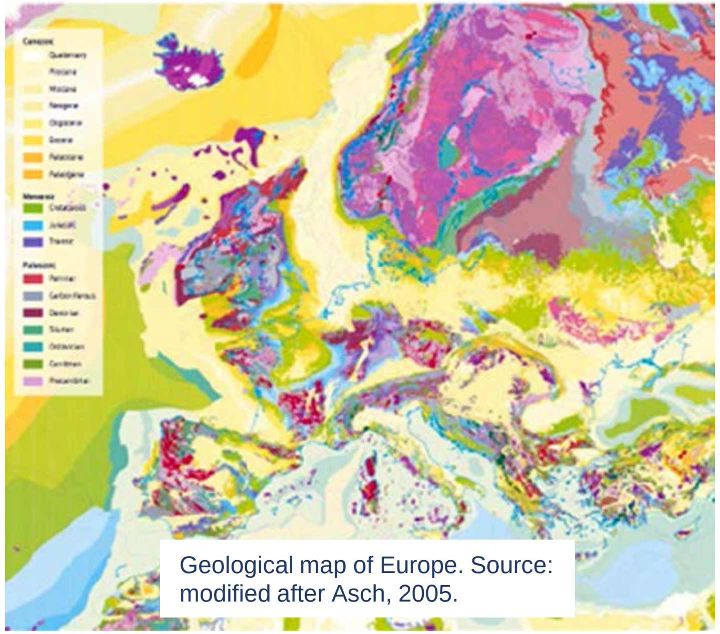
Objectives – static to dynamic

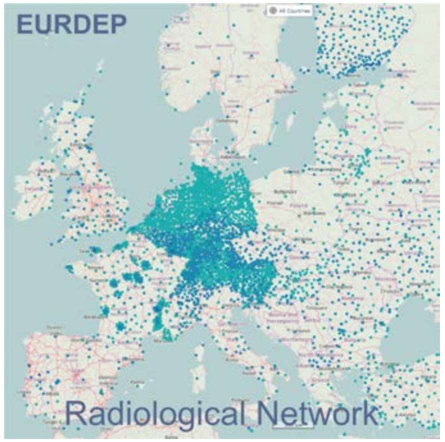
Static maps:

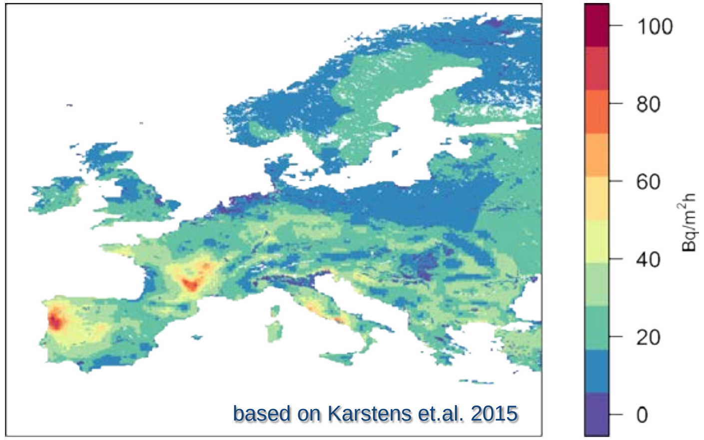




Dynamic maps:

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!





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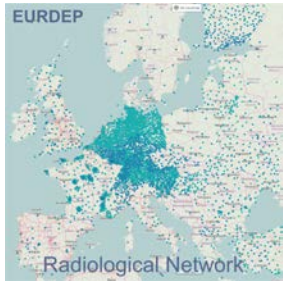
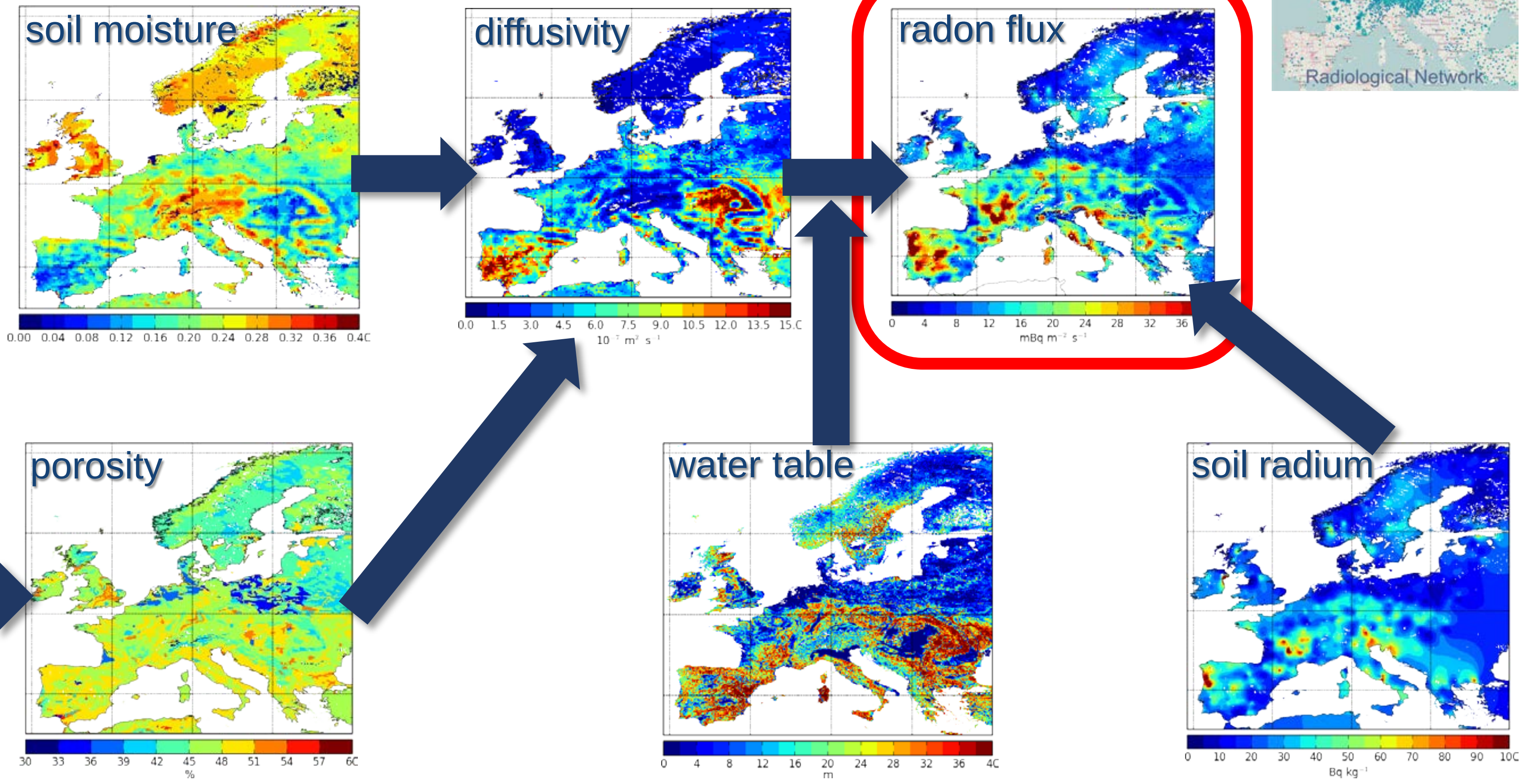
Objectives – Joining forces

Our task: Joining forces in the field of radon

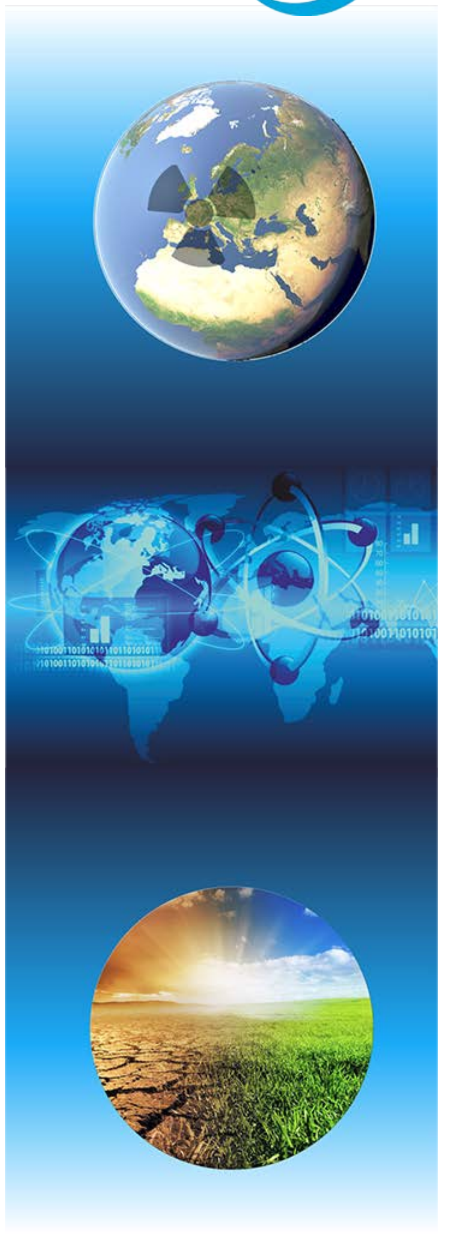


soil texture
% clay
% sand
% slit

bulk density



based on Karstens et.al. 2015



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Achievements – 1: New traceability

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New metrology for radon at the environmental level

Annette Röttger[1,*], Stefan Röttger[1], Claudia Grossi[2], Arturo Vargas[2], Roger Curcoll[2], Petr Otáhal[3], Miguel Ángel Hernández-Ceballos[4], Giorgia Cinelli[5], Scott Chambers[6], Susana Alexandra Barbosa[7], Mihail-Razvan Ioan[8], Ileana Radulescu[8], Dafina Kikaj[9], Edward Chung[9,10], Tim Arnold[9,10], Camille Yver-Kwok[11], Marta Fuente[11], Florian Mertes[1] and Viacheslav Morosh[1]

[1] Physikalisch-Technische Bundesanstalt, 38116 Braunschweig, Germany
[2] Institut de Tècniques Energètiques, Universitat Politècnica de Catalunya, 08028 Barcelona, Spain
[3] National Institute for NBC Protection, 26231 Milin, Czech Republic
[4] Department of Physics, University of Córdoba, 14071 Córdoba, Spain
[5] European Commission, Joint Research Centre, 21027 Ispra, Italy
[6] Australian Nuclear Science and Technology Organisation, 2234 Lucas Heights, Australia
[7] INESC TEC, 4200-465 Porto, Portugal
[8] Horia Hulubei National Institute for R&D in Physics and Nuclear Engineering, 077125 Magurele, Romania
[9] National Physical Laboratory, Teddington, Middlesex, United Kingdom
[10] School of GeoSciences, University of Edinburgh, Edinburgh, United Kingdom
[11] Laboratoire des Sciences du Climat et de l'Environnement, (LSCE-IPSL), CEA-CNRS-UVSQ, Université Paris-Saclay, 91191 Gif-sur-Yvette, France

E-mail: annette.roettger@ptb.de

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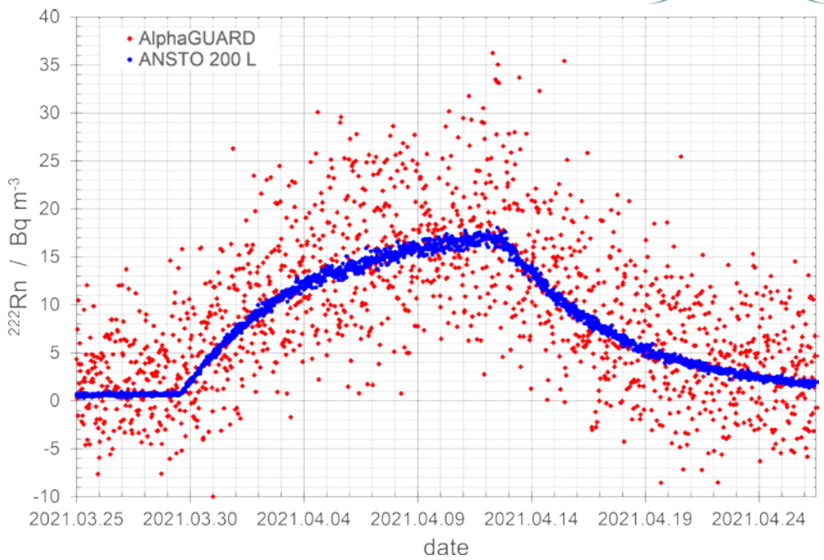

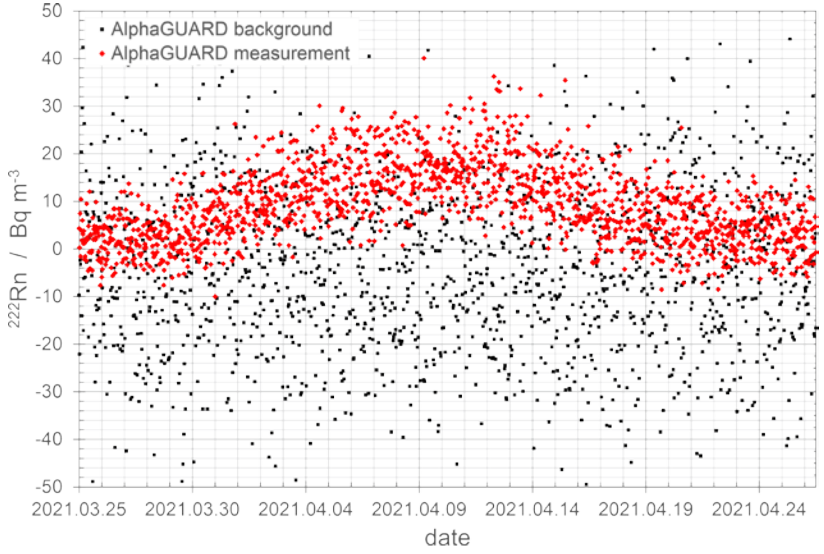

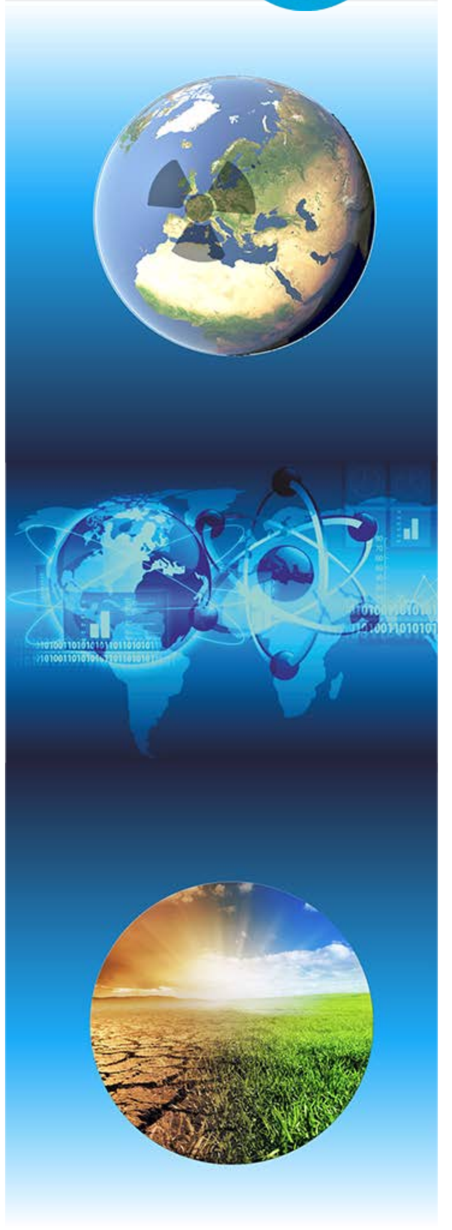

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Achievements – 2a: New sources

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Electro-deposited

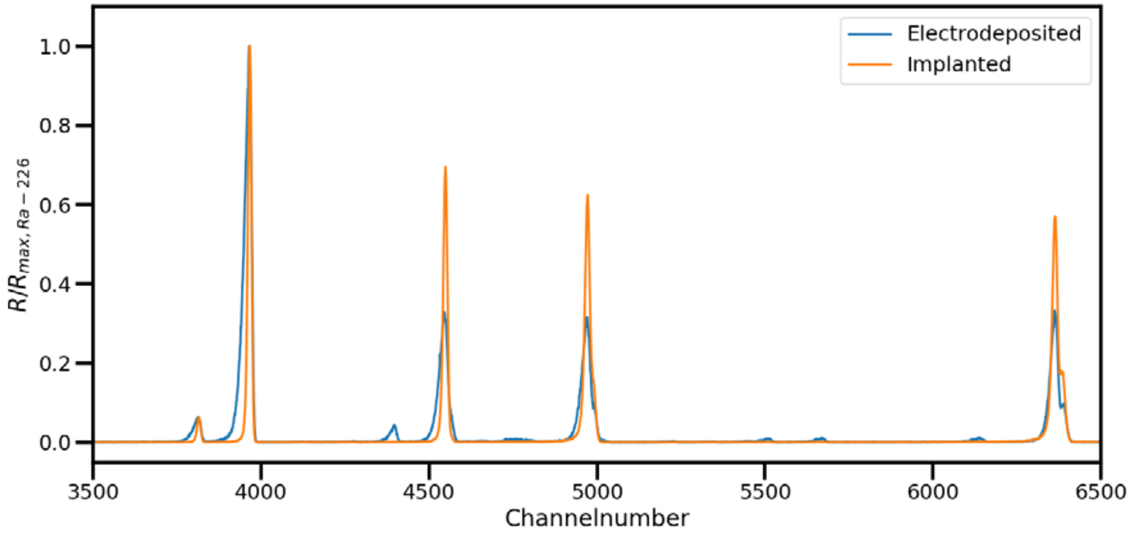
Deposition at
30 V < U < 200 V

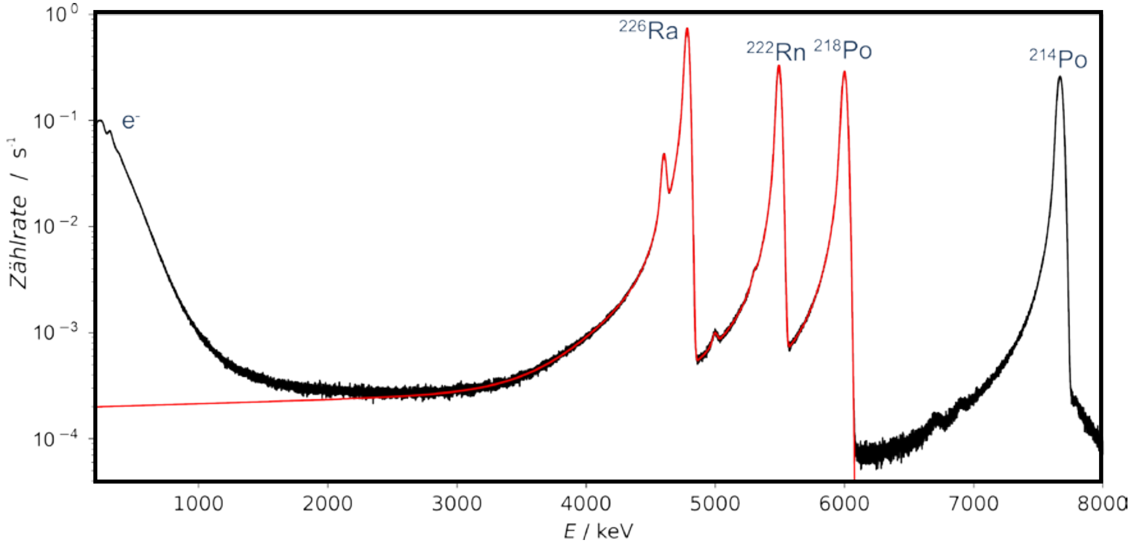
Implanted

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

PIPS

450 mm², 300 µm
with 150 Bq ^{226}Ra
layer





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Achievements – 2b: IRSD concept

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

Physikalisch-Technische Bundesanstalt, National Metrology Institute, 38116 Braunschweig, Germany; stefan.roettger@ptb.de (S.R.); annette.roettger@ptb.de (A.R.)
* 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 $^{226}RaCl_2$ 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

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

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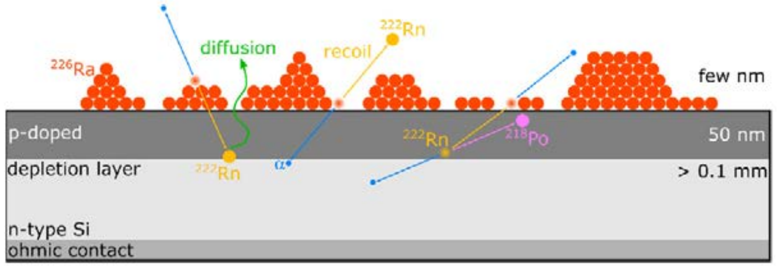

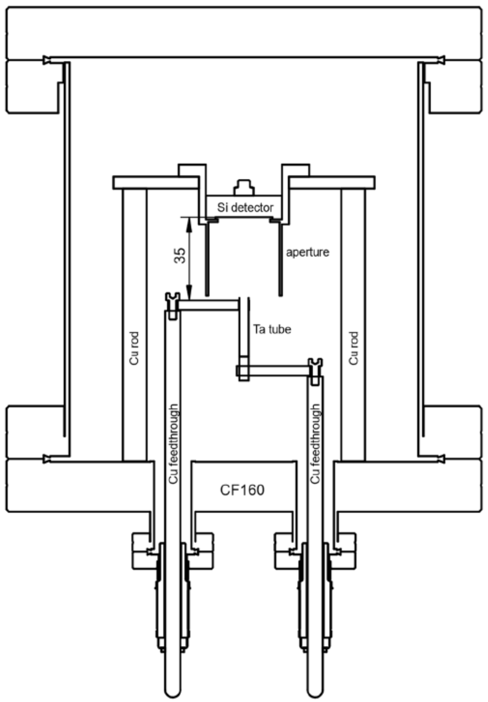

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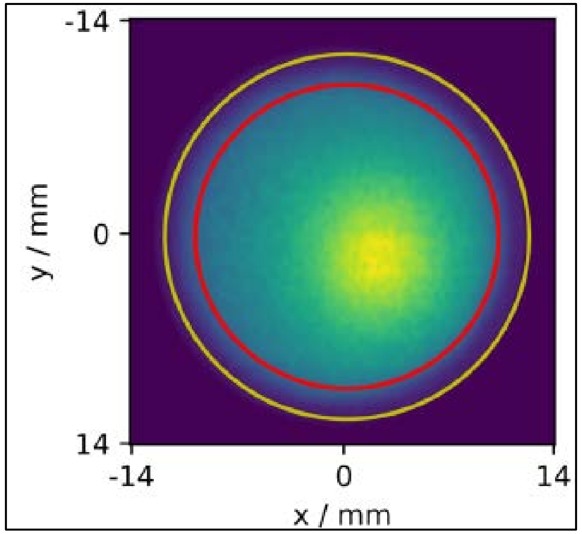
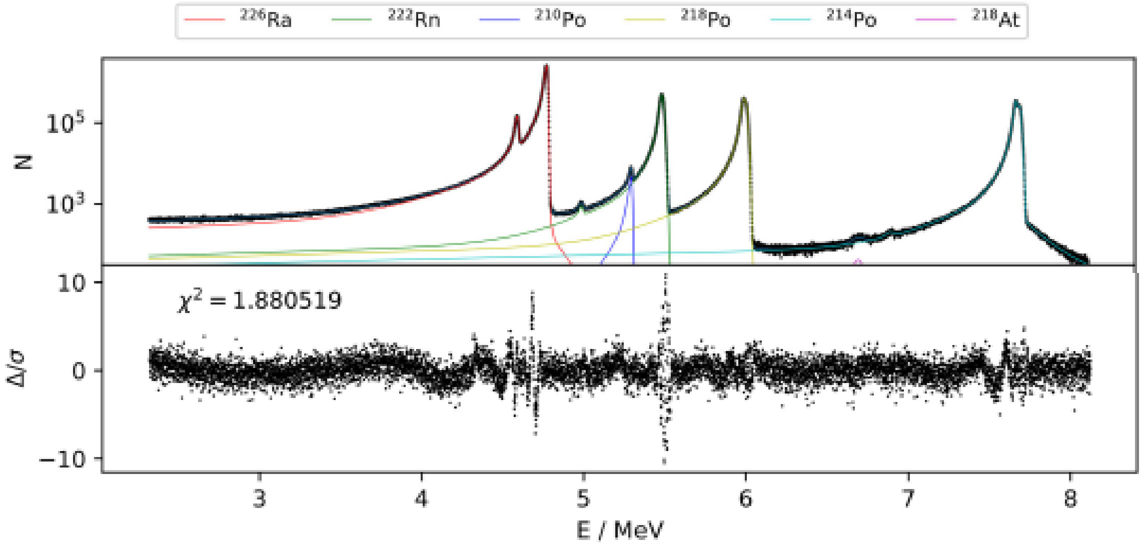
Achievements – 2c: IRSD application



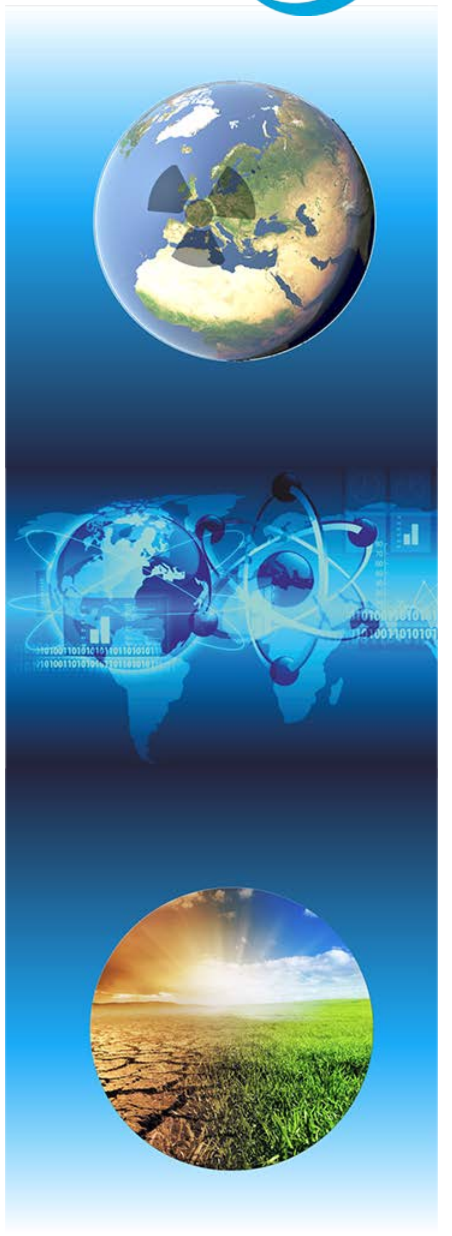
Photograph of an IRSD based on a 450 mm² Canberra PIPS® detector, modified with a layer containing 440 Bq $^{226}RaCl_2$ (left). A digital autoradiograph obtained from such a deposit where the inner diameter of the recessed Si-surface is given in yellow, and the diameter of the shadowing aperture is given in red (right).



What metrology can provide:

Description and Type	Value and Uncertainty	Rel. Uncertainty
Solid angle (systematic)	$(0.00940 \pm 0.00006)\ 4\pi$ sr	0.6%
Backscattering$_{DSA}$ (systematic)	1 ± 0.002	0.2%
Tailing$_{DSA}$ (systematic)	1 ± 0.003	0.3%
Tailing$_{Si}$ (systematic)	1 ± 0.003	0.3%
^{226}Ra rate$_{DSA}$ (stochastic)	$(0.01796 \pm 0.00015)\ s^{-1}$	0.8%
^{226}Ra rate$_{Si}$ (stochastic)	$(0.9595 \pm 0.0004)\ s^{-1}$	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 mm²/300 µm	1.91 ± 0.02	0.502 ± 0.006
Ametek Ortec ULTRA®	450 mm²/300 µm	66.4 ± 0.5	0.494 ± 0.004
Mirion PIPS®	450 mm²/300 µm	158.6 ± 1.7	0.494 ± 0.005
Mirion PIPS®	450 mm²/100 µm	442 ± 4	0.492 ± 0.005



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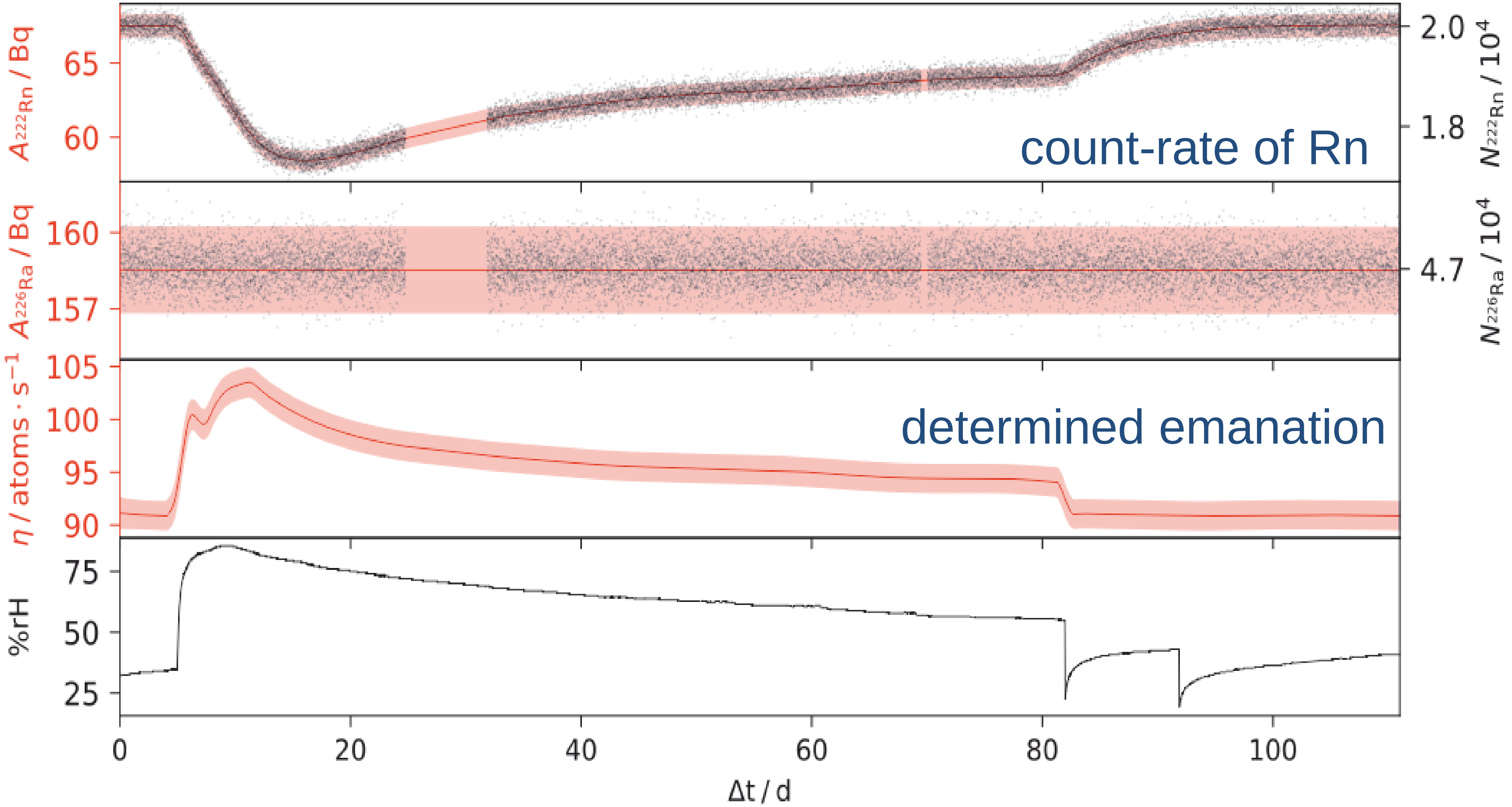
Achievements – 2d: Radon Activity

Switching Linear Dynamic System-deconvolution

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



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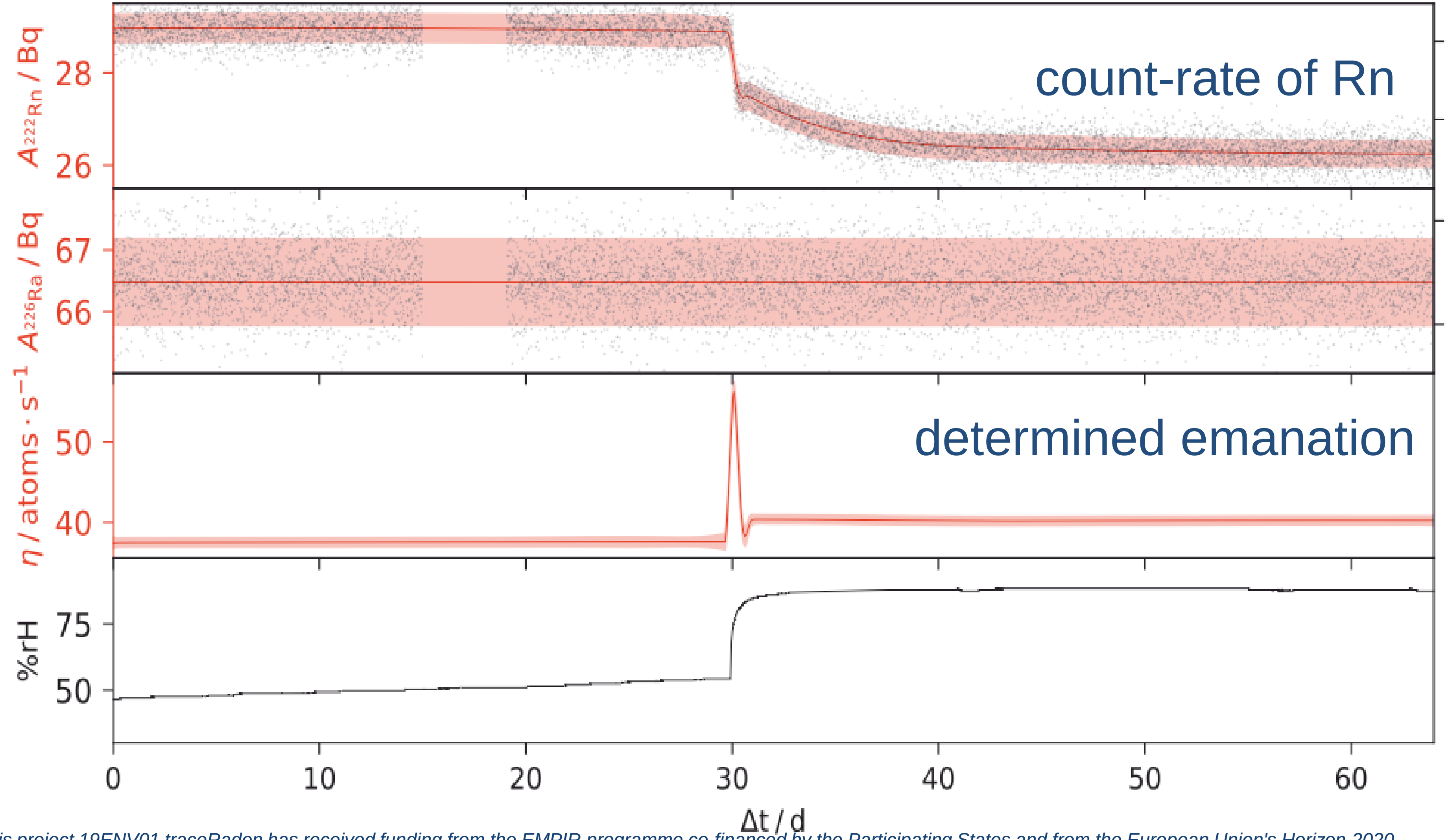
Achievements – 2d: Radon Activity

Switching Linear Dynamic System-deconvolution

IRSD: A(^{226}Ra) = (66.4 ± 0.5) Bq



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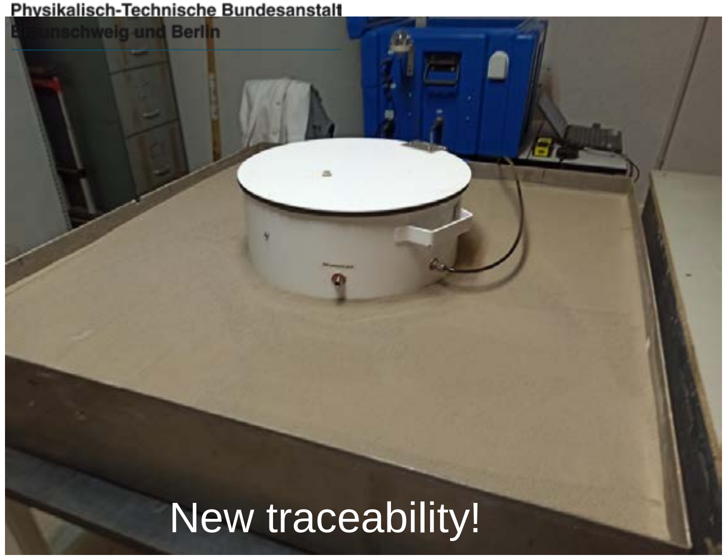
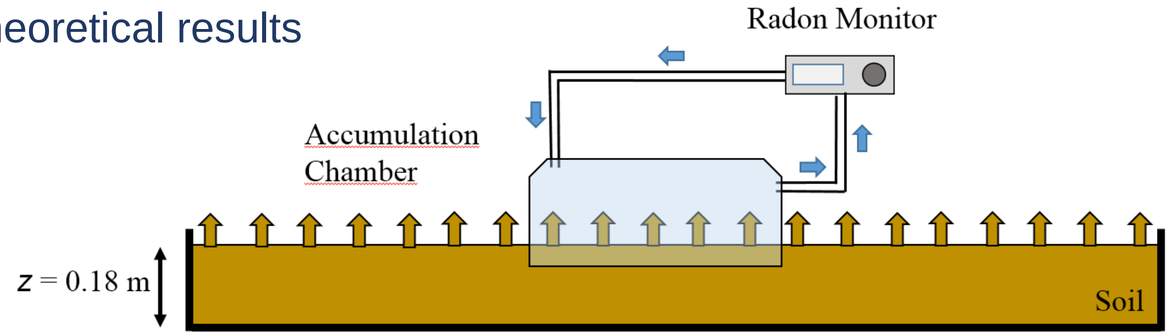
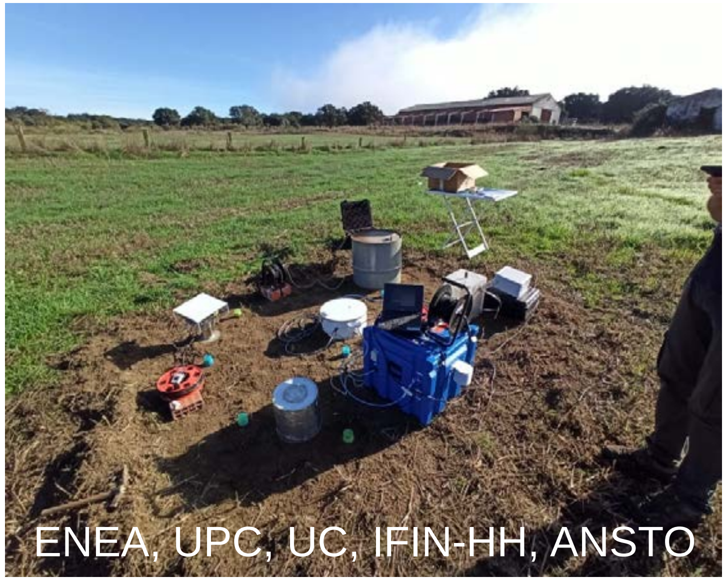
Achievements – 3: Radon flux



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

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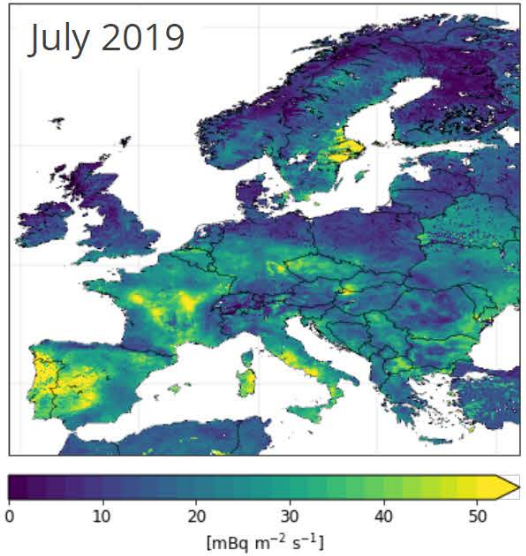
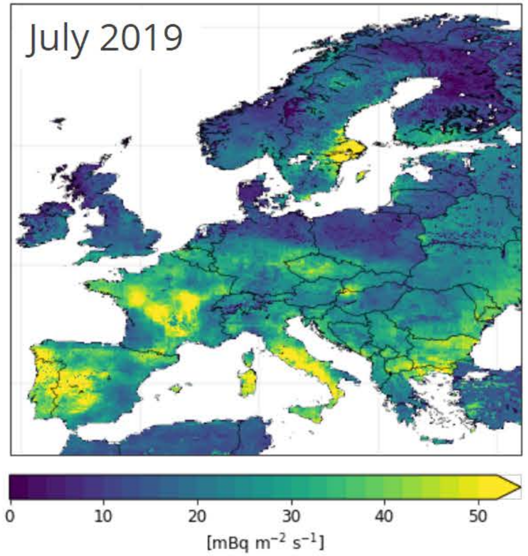
Achievements – 4: Data reanalysis

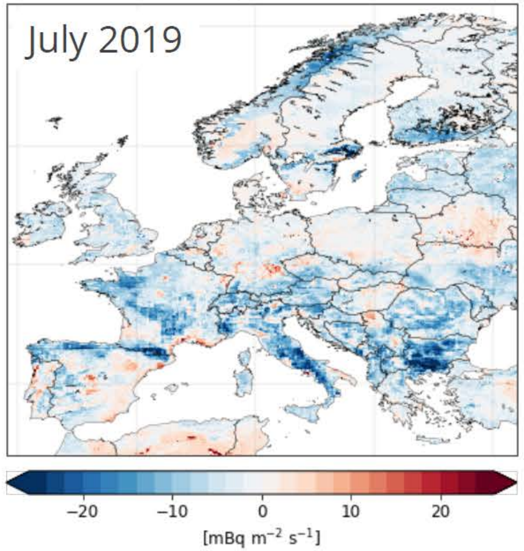
Radon flux based on GLDAS-Noah v2.1 soil moisture

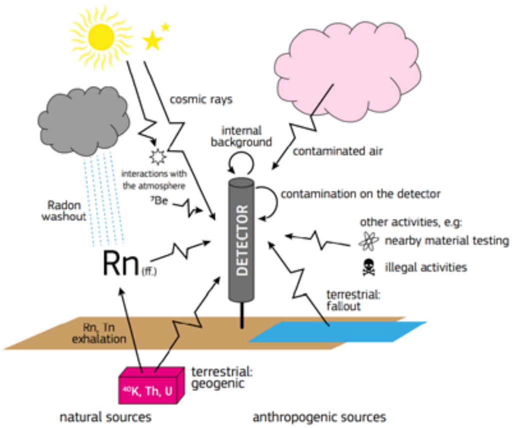


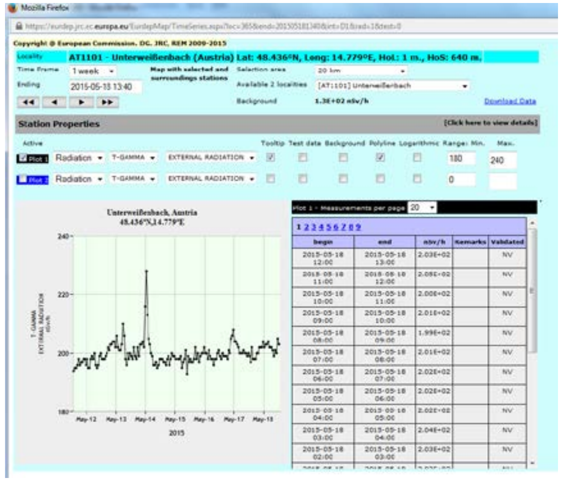
Radon flux based on ERA5-Land soil moisture



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.

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Physikalisch-Technische Bundesanstalt
Braunschweig und Berlin

Summary

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**.

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Worldwide collaboration

Last continent reached: Antarctica!

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Thanks…

… to the 18 traceRadon-project partners:

… to the 13 traceRadon-project collaborators:

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

… and for your attention!