Metrology chain for continuous radon flux observations: Strategy and Challenges

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in collaboration with
WP2 traceRadon project researchers

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

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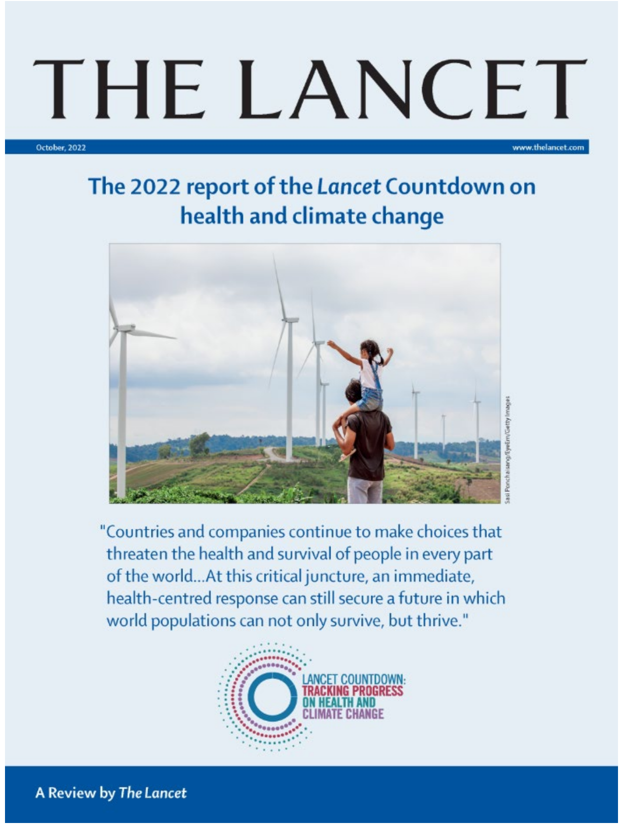
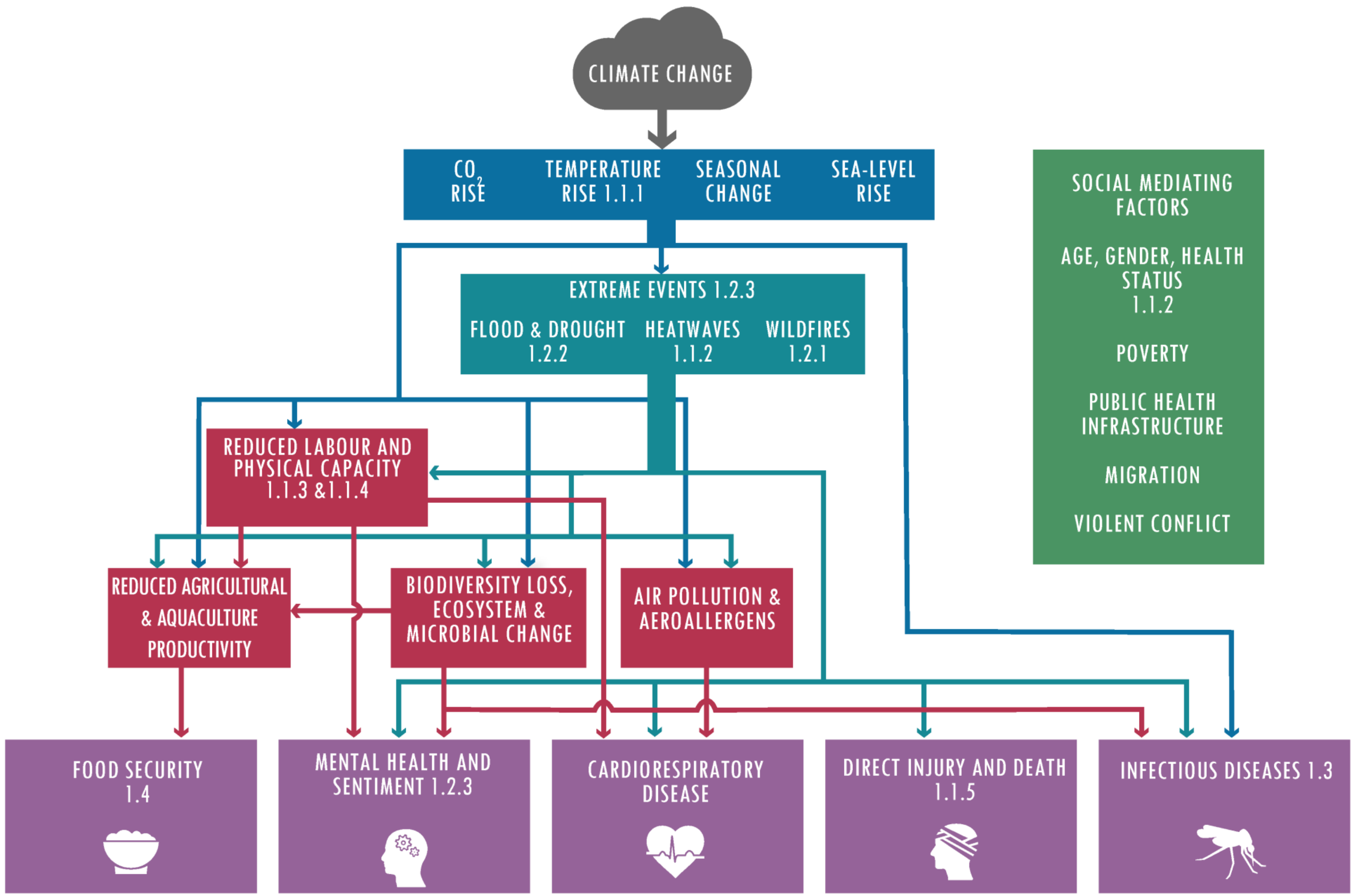
Contents

- Climate Change and Health
- ^{222}Rn from major natural radioactive exposure source to environmental tracer
- ^{222}Rn emissions from soil surfaces: European Radon flux maps
- High quality continuous radon flux observations: Metrology chain
- Intense radon flux campaigns in Europe
- Results and discussion: Is the linear method reliable?
- Further research steps: we know that we know something

A changing climate has profound implications for human health (The Lancet, 2022)



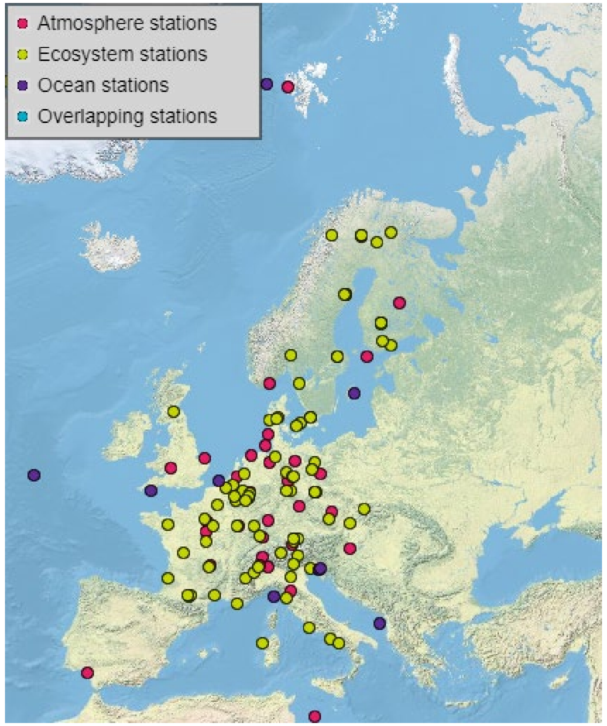
INDICATORS OF CLIMATE CHANGE IMPACTS, EXPOSURES, AND VULNERABILITY



Introduction: Climate change and Health: need of independent GHG emission inventories validation methods

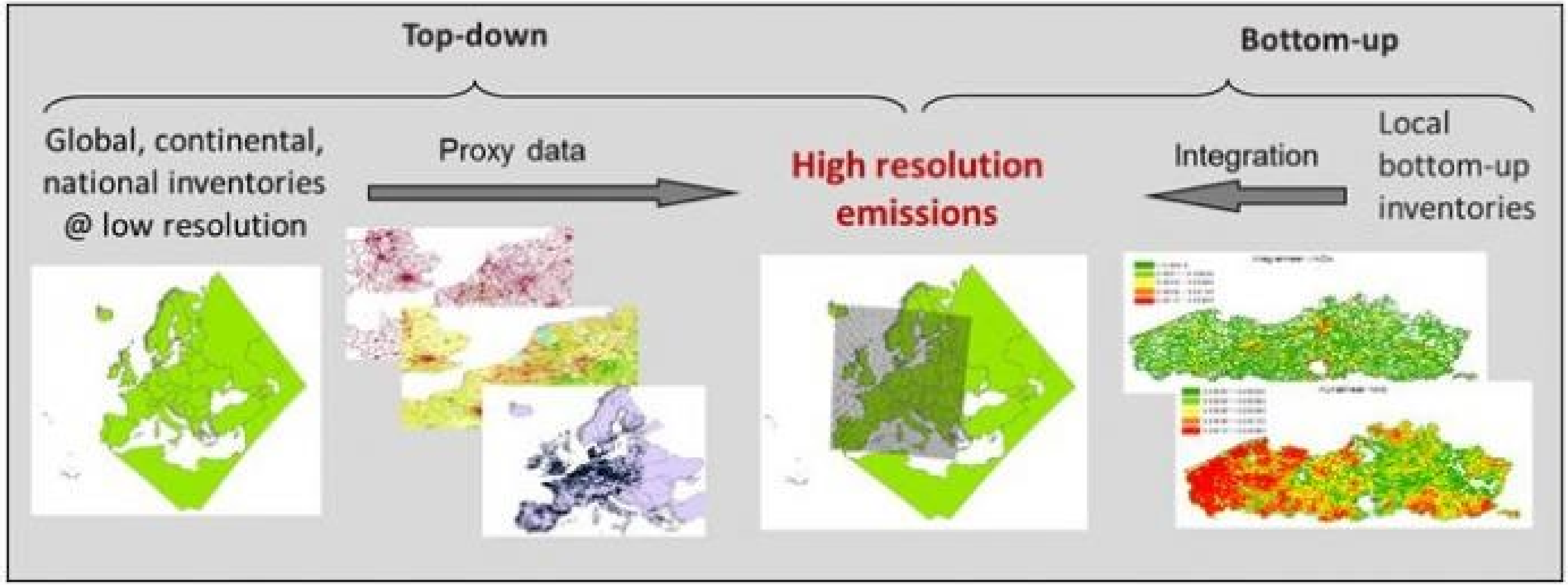
To improve and to harmonize outdoor GHGs and tracers concentration measurements and their fluxes over different ecosystems for the improvement of atmospheric transport models, GHG budget emissions and environmental radioactivity studies.

GHGs emissions, due to natural as well as anthropogenic sources, need to be estimated and reported by each national agency to the United Nations Framework Convention on Climate Change (UNFCCC).



Integrated Carbon Observation Netowrk (ICOS)

Introduction: Climate change and Health: need of independent GHG emission inventories validation methods



Indipendent methods and techniques are needed for reducing the uncertainties related with Top-Down and Bottom-Up methodologies and understanding their systematic inconsistencies

^{222}Rn from major natural radioactive exposure source to environmental tracer

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PROPRIETIES

- radioactive (alpha spectrometry)
- natural (^{226}Ra decay in grain soil/no need of monitored release)
- noble (no chemical reaction)
- $T_{1/2}$ = 3.8 days
- over the ocean its exhalation is taken as zero
- quite homogeneous fluxes IF compared with GHGs fluxes

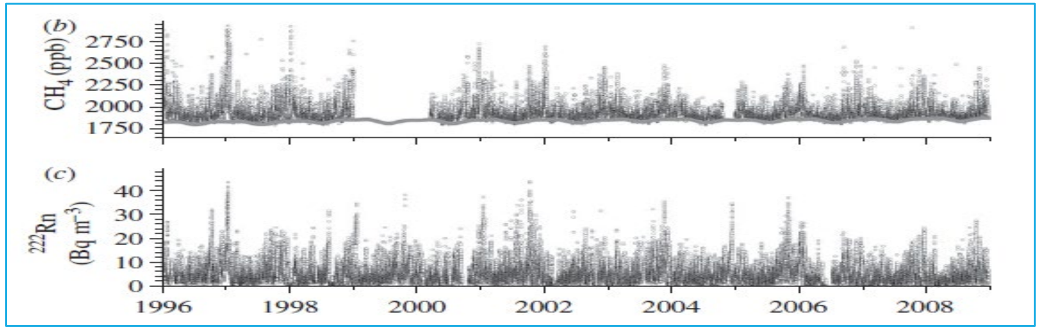
SOME APPLICATIONS

i) To improve inverse transport models
ii) To study atmospheric transport and mixing processes within the planetary boundary layer
iii) To experimentally estimate GHGs fluxes by using the Radon Tracer Method
iv) To refine baseline selection and characterisation techniques, and analyse air mass history and fetch at remote sites
v) To identify submarine groundwater discharges

^{222}Rn from major natural radioactive exposure source to environmental tracer

Horizon 2020 European Union Funding for Research & Innovation

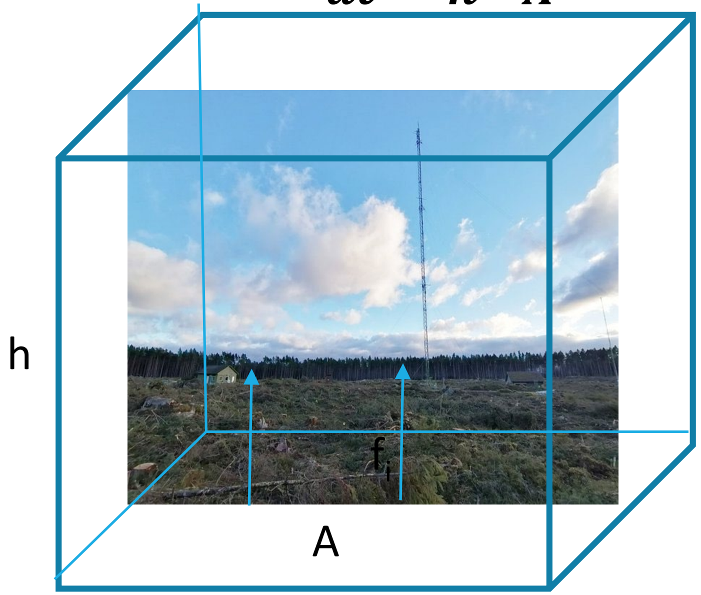
An independent method to estimate GHGs fluxes is the Radon Tracer Method (RTM) which allows experimentally estimating GHGs fluxes (Grossi et al., 2018) and thus improving Bottom-Up GHGs inventories.

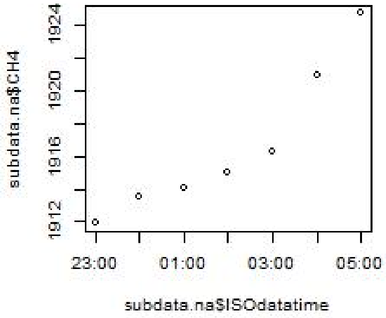


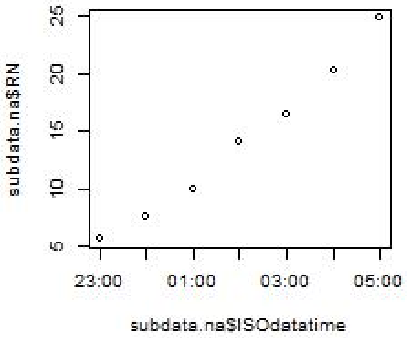
Levin et al., 2011

- **CO-LOCATED MEASUREMENTS OF RADON AND GHG CONCENTRATIONS IN AIR (Ci and Cj)**
- **WELL SOLVED RADON FLUX MAPS (fi)**
- **ATMOSPHERIC BOX MODEL with nocturnal layer height (h) and footprint A**

$$\frac{dc_i}{dt} = \frac{f_i}{h * A}$$







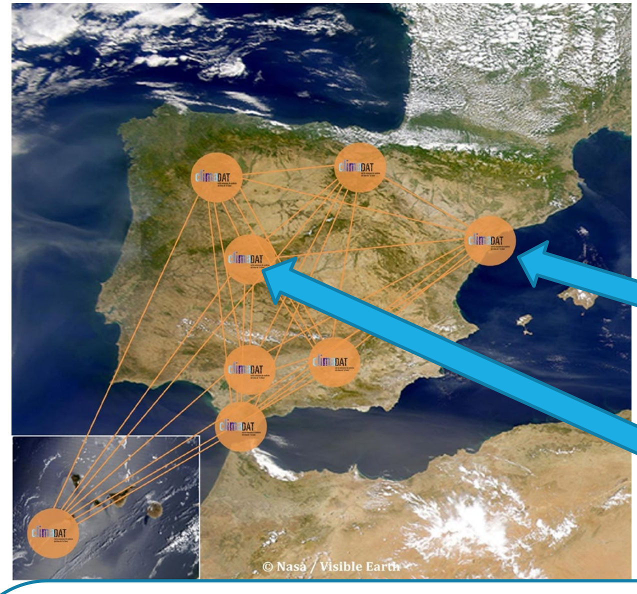
$$f_i = \frac{\frac{dc_i}{dt}}{\frac{dc_j}{dt}} \cdot f_j$$

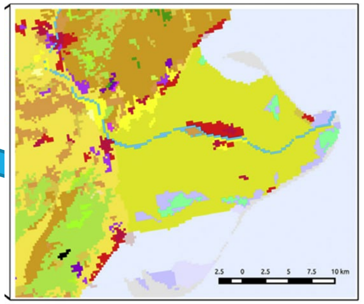
c_i GHG atmospheric concentration
f_i GHG flux
c_j atmospheric ^{222}Rn concentration
f_j ^{222}Rn flux

Knowing the flux of the tracer will allow us to calculate the flux of the GHG

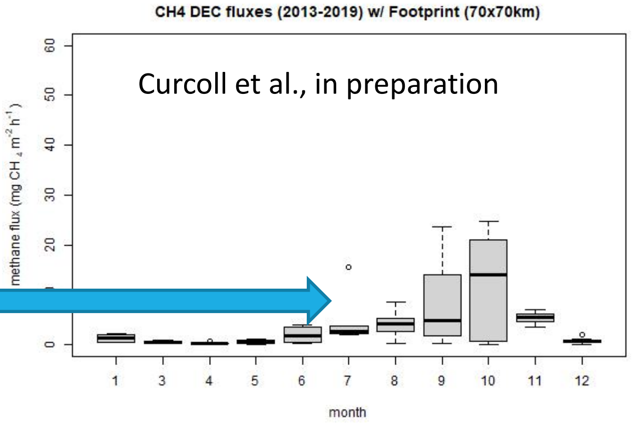
^{222}Rn from major natural radioactive exposure source to environmental tracer

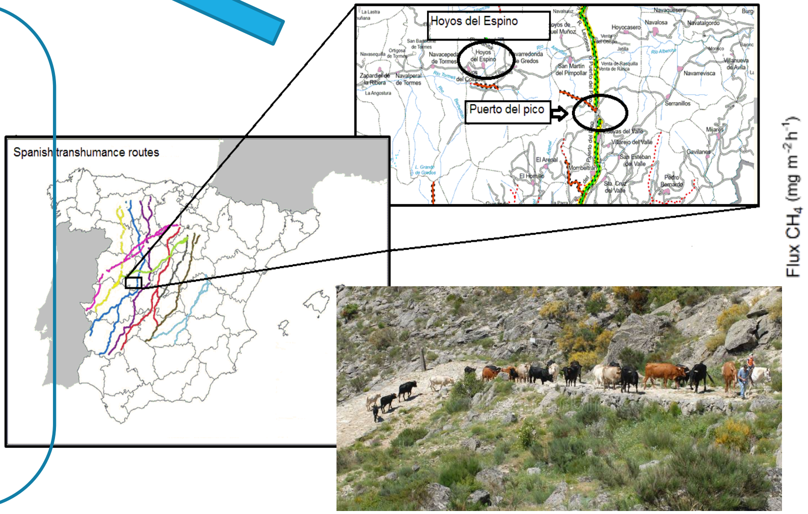
Application of **RTM to calculate CH_4 fluxes** at the Gredos and Iruelas (GIC3) and Delta D'Ebre station (DEC3) station within the IC3-UAB CLIMADAT Network during **cows transhumance periods and rice fields straw incorporation period**, respectively.

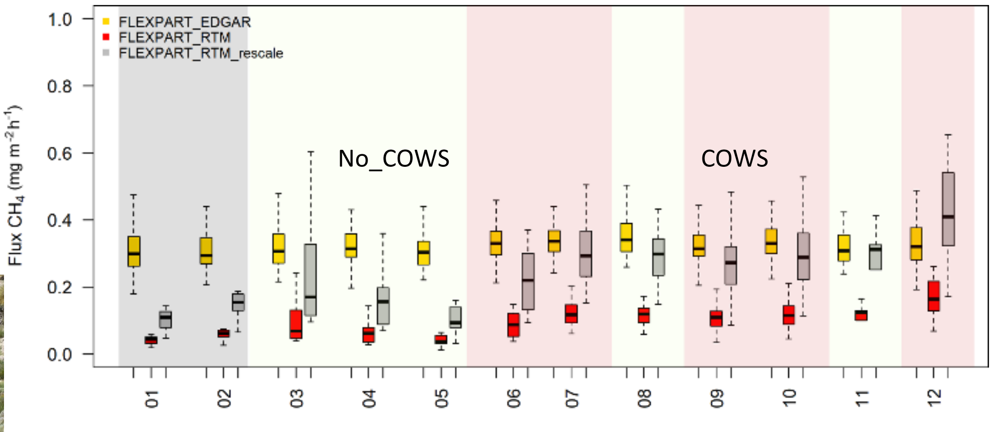




Àgueda et al., 2017





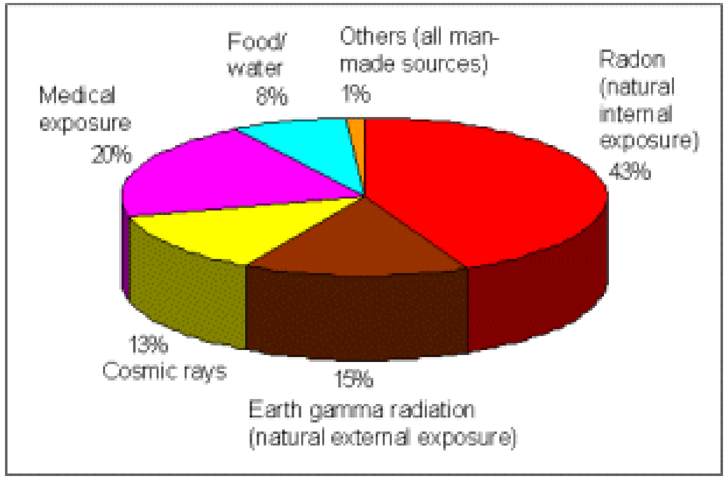


Grossi et al., 2018

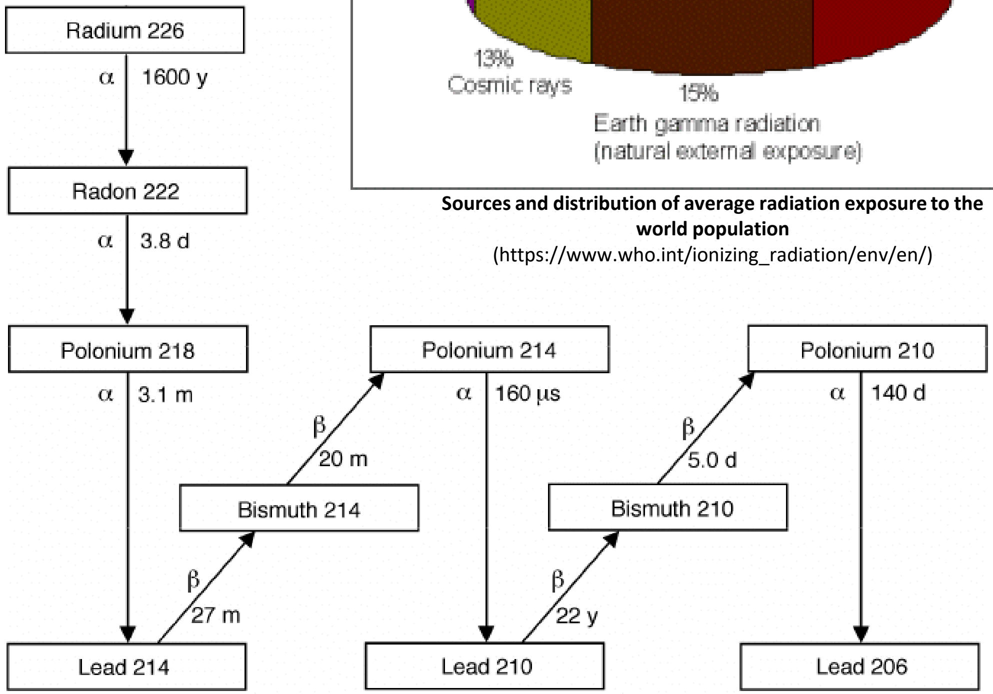
Radon emissions are easier to simulate that GHG ones but *again* models need to be validated

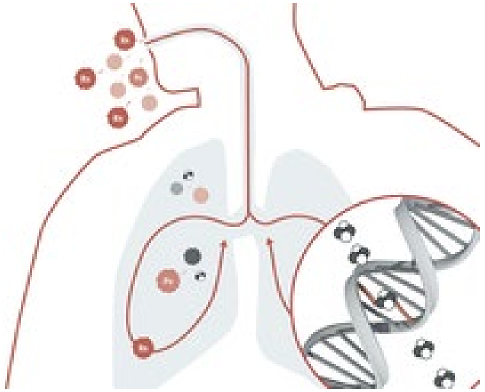
^{222}Rn from major natural radioactive exposure source to environmental tracer

The **natural radioactive noble gas radon** is an *Indoor Air Quality and a Radiation Protection Problem*



Sources and distribution of average radiation exposure to the world population
(https://www.who.int/ionizing_radiation/env/en/)





https://www.bfs.de/

2013/59/EURATOM – Article 103(3)

'**Member States shall identify areas** where the radon concentration (as an annual average) in a significant number of buildings is expected to exceed the relevant national reference level.'

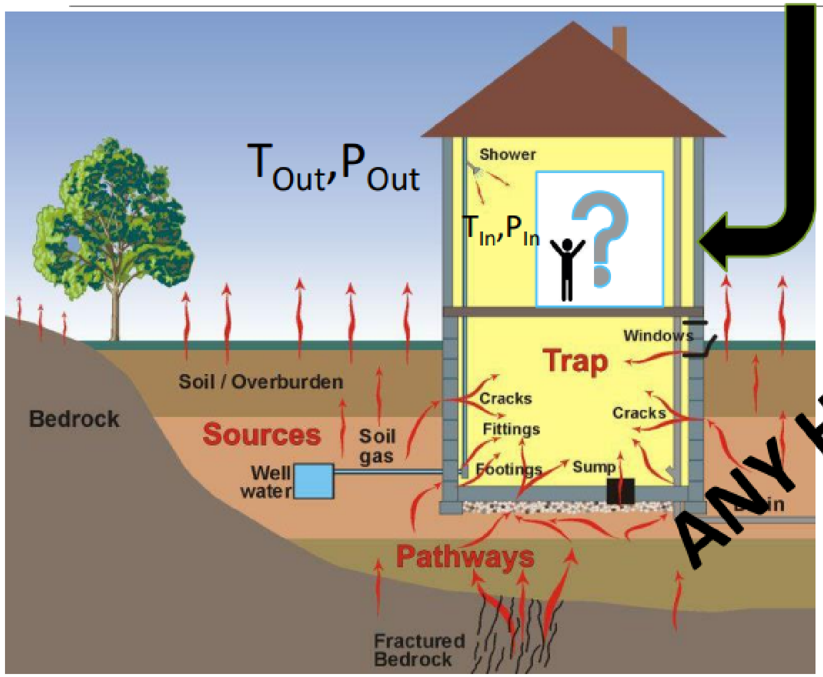
L 13/66 EN Official Journal of the European Union 17.1.2014

ANNEX XVIII

List of items to be considered in preparing the national action plan to address long-term risks from radon exposures as referred to in Articles 54, 74 and 103

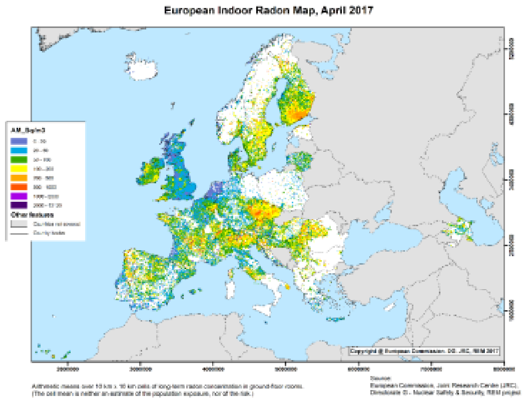
(1) Strategy for conducting surveys of indoor radon concentrations or soil gas concentrations for the purpose of estimating the distribution of indoor radon concentrations, for the management of measurement data and for the establishment of other relevant parameters (such as soil and rock types, permeability and radium-226 content of rock or soil).

The main aim is knowing the indoor ^{222}Rn concentrations at which the people are exposed in old building or may be exposed in new building

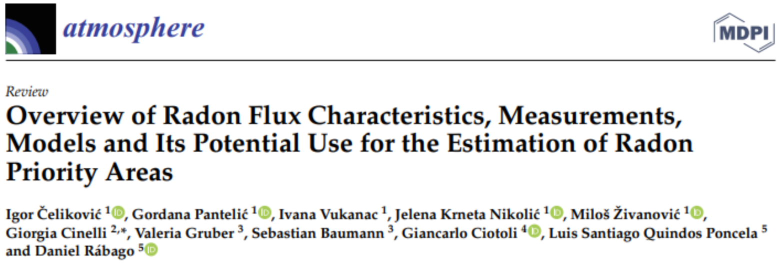


ANY HELP ?

EXPERIMENTAL LONG-TERM CONTINUOUS MEASUREMENTS ARE TOO DEMANDING FOR PRESENT BUILDINGS AND NOT FEASIBLE FOR FUTURE BUILDING



https://remon.jrc.ec.europa.eu/About/Atlas-of-Natural-Radiation/Indoor-radon-AM/Indoor-radon-concentration



atmosphere MDPI

Review

Overview of Radon Flux Characteristics, Measurements, Models and Its Potential Use for the Estimation of Radon Priority Areas

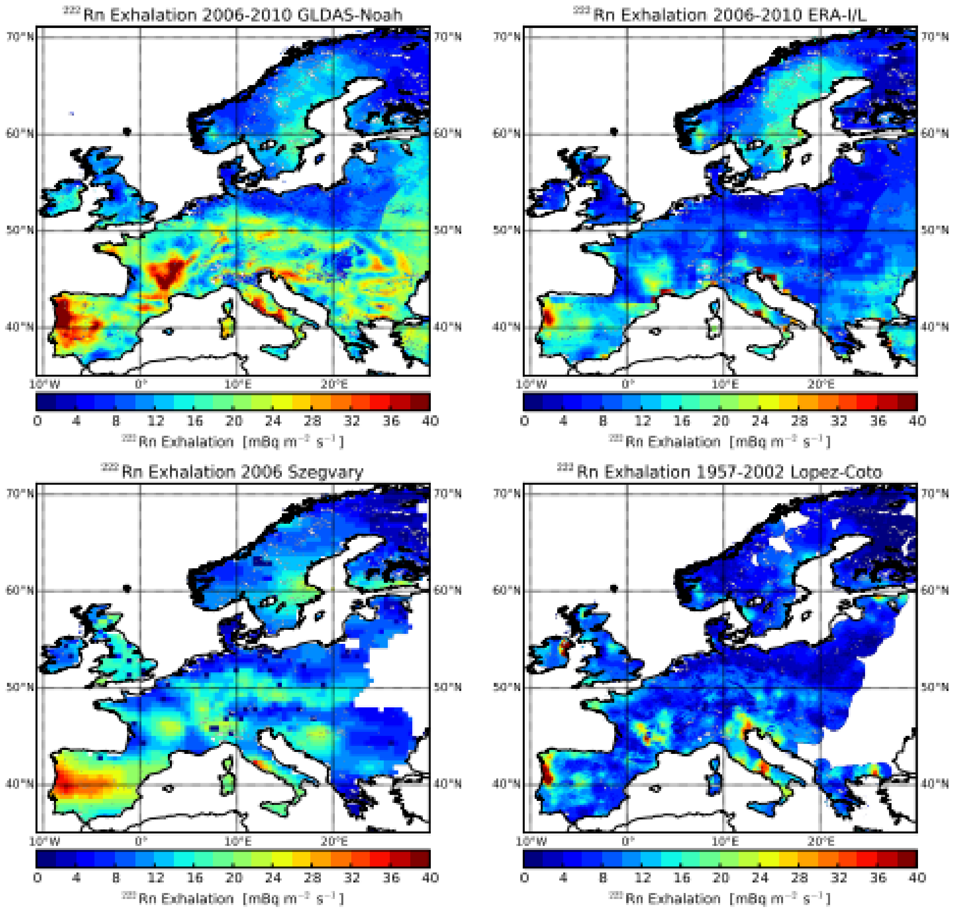
Igor Čeliković [1], Gordana Pantelić [1], Ivana Vukanac [1], Jelena Krneta Nikolić [1], Miloš Živanović [1], Giorgia Cinelli [2,*], Valeria Gruber [3], Sebastian Baumann [3], Giancarlo Ciotoli [4], Luis Santiago Quindos Poncela [5] and Daniel Rábago [5]

Due to the positive correlations between radon flux and radon quantities such as radon in soil gas and indoor radon, radon flux could be used as an input parameter for the estimation of RPA.

^{222}Rn emissions from soil surfaces: Theoretical approach and European radon flux maps

Available radon flux maps based on gas transport model in the soil could be a useful proxy to identify Radon Prone Area for population protection strategies and for applications of radon tracer.

High spatial and high temporal Radon flux models and/or inventories need to be validated using continuous observations in Europe



Karstens et al., 2015

^{222}Rn emissions from soil surfaces: Theoretical approach and European radon flux maps

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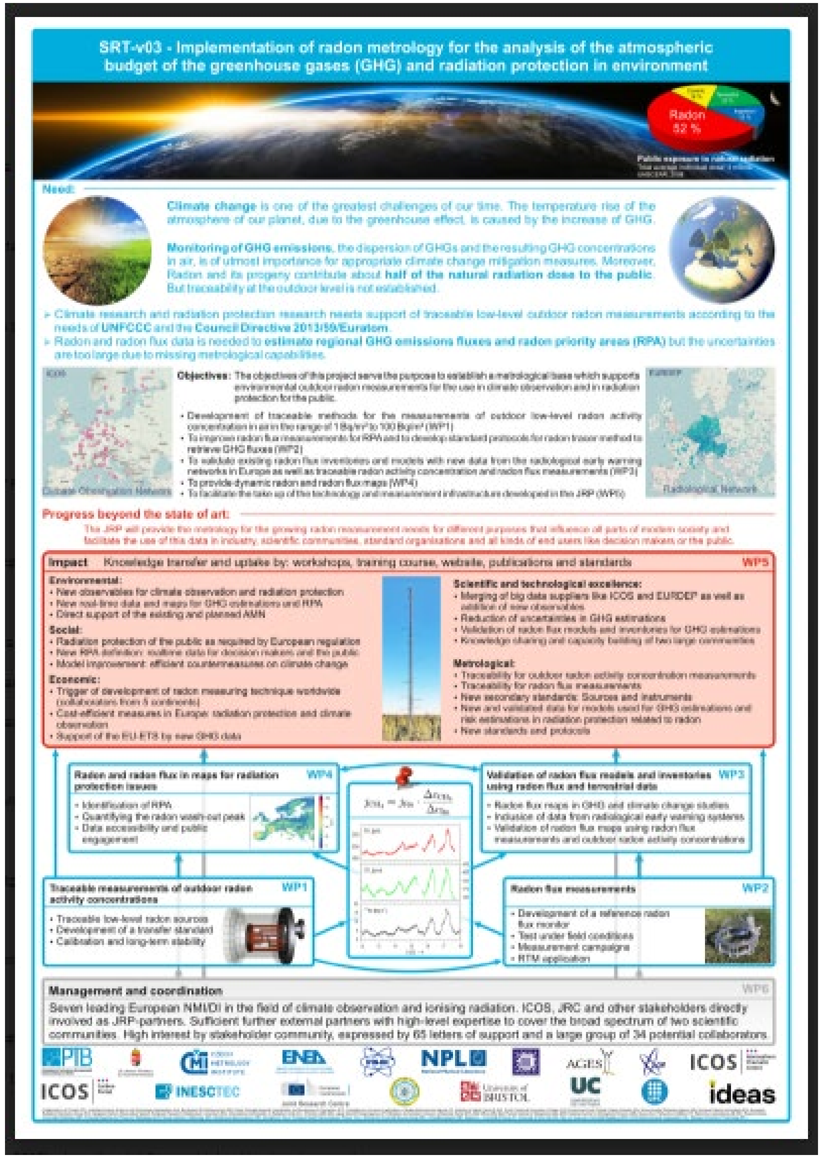

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EURAMET

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trace Radon

The WP2 and WP3 of the traceRadon Project aim to:

1. **Generate daily and monthly radon flux maps;**
2. **Offer a full metrology chain and infrastructure for continuous radon flux measurements;**
3. **Carry out four intense radon flux campaigns to generate reliable data for comparison with radon flux maps inputs and outputs**

^{222}Rn emissions from soil surfaces: Theoretical approach and European radon flux maps

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Modelling radon flux from soils (modified slide from Karstens et al., ICOS 2022)

The WP2 and WP3 of the traceRadon Project aim to:

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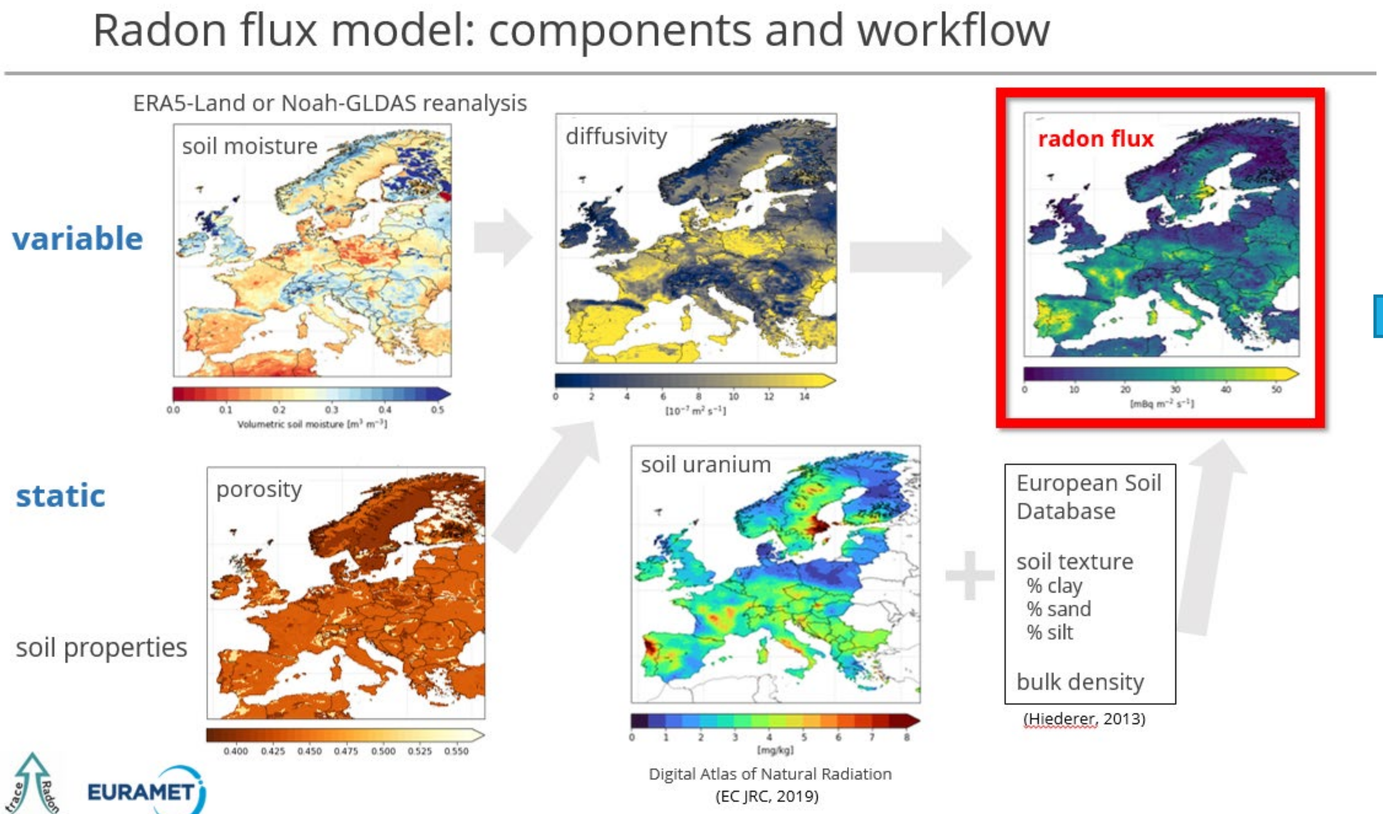

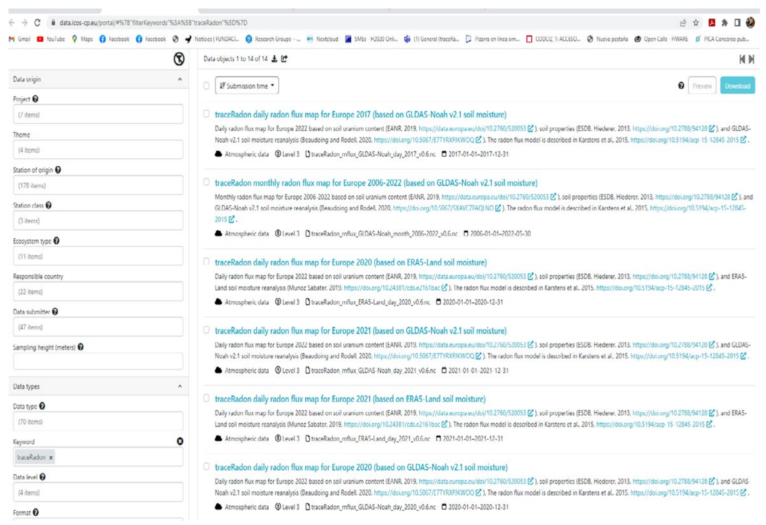

Daily and Monthly traceRadon European radon flux maps are now available at the ICOS Carbon Portal

High quality continuous radon flux observations: Metrology chain

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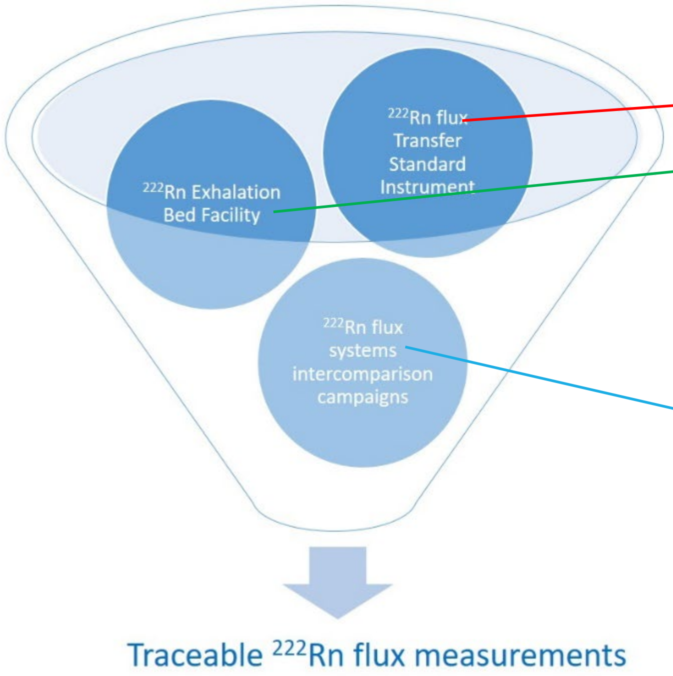
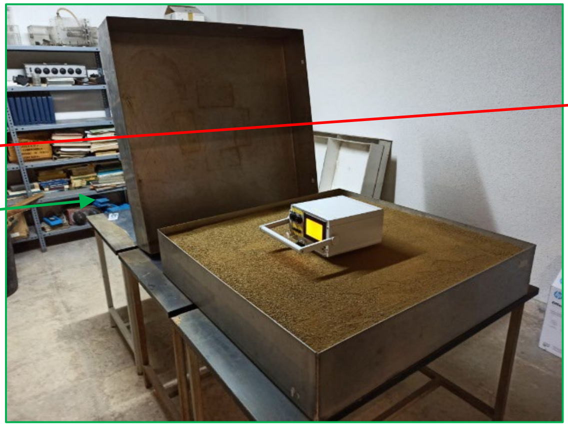




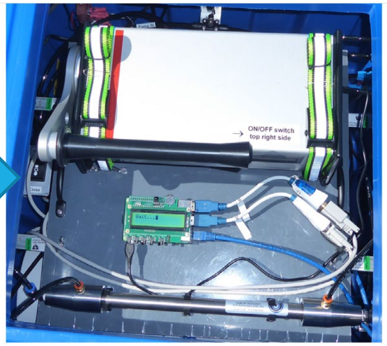
Selection of a ^{222}Rn flux Transfer Standard instrument

Diffusion chambers for radon flux measurements which may be coupled with continuous 'indoor' radon monitors

Table 4 list of chambers for radon flux measurements (sources are listed in Table 4)

Source	Device name	Creator	Volume	Emanation area	Automatic opening	Shape	Used with
1	AutoFlux	designed by Sylvester Werczynski and built by Ot Sisoutham in 2018/19	0.0188 m³	0.126 m²	yes	drum	AlphaGUARD
2	Emanometer diffusion chamber	ANSTO	0,018 m³	0,26 m²	no	Shallow conical	AlphaGUARD2 x 1L lucas cells (separated by 6 min flow path)
3	8100-401 Chamber Control Kit	LI-COR	4076.1 cm³	317.8 cm²	yes	hemisphere	-
4	radon flux measurement system	-	0.037 m³	0.21 m²	yes	drum	AlphaGuard
5	flux chambers	-	0.002 m³, 0.018 m³ and 0.352 m³	0.017 m², 0.071 m² and 2.32 m²	no	circular, circular, square	RAD7
6	The accumulation chamber	-	0.04398 m³	0.126 m²	yes	drum	-
7	UPC accumulation chamber	Universitat Politecnica de Catalunya	-	0,0069 m³	Only pump	box	DOSEman inside the chamber
8	UC accumulation chamber	Cantabria University	8000 cm³	400 cm²	Only pump	Drum	AlphE

AutoFlux system
Radon flux meas. every 3h
No maintenance is needed

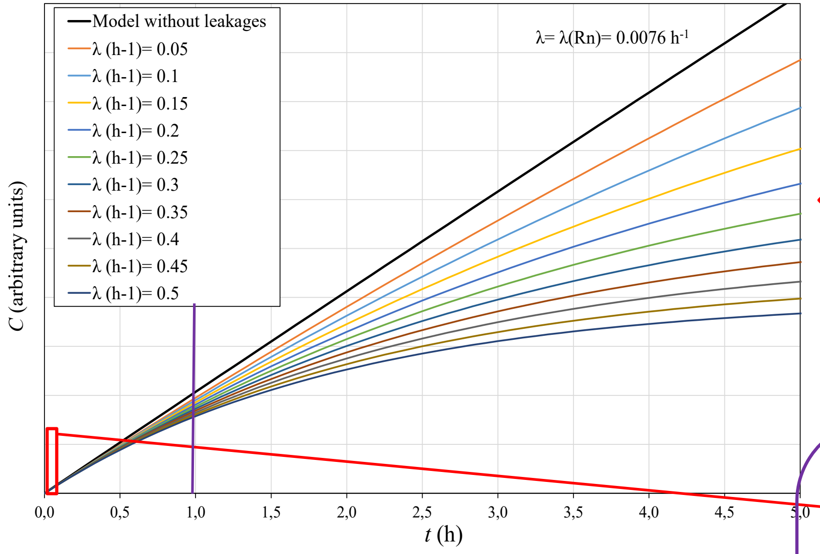


***AutoFlux* system running in the field. The radon activity concentration, internal air temperature, differential pressure and soil characteristics are measured within the white drum. Ambient temperature, humidity, pressure and rainfall are measured on the side of the transport case (~50 cm a.g.l.), and the main system components are located inside the waterproof transport case.**

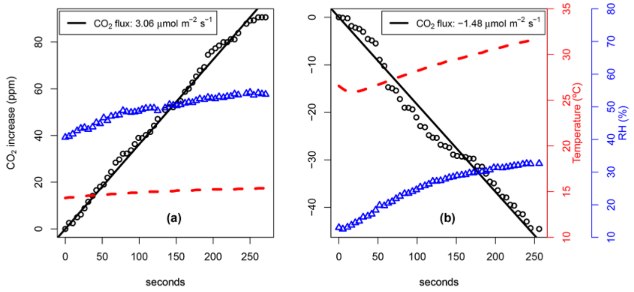
Variable (Label within the document)	Sensor	Location	Unit (S.I.)
Volumetric Water Content (VWC) in the soil	CSI CS655 Water Content Reflectometer	Inside Drum	m^3/m^3
Electrical soil conductivity (EC)	CSI CS655 Water Content Reflectometer	Inside Drum	dS/m
Water vapor pressure (VaporPress)	CSI CS655 Water Content Reflectometer	Inside Soil	kPa
Soil temperature (T)	CSI CS655 Water Content Reflectometer	Inside Soil	^0C
Drum air temperature (DrumTemp)	SDI-12 sensor Unidata 6508A	Inside Drum	^0C
Atmospheric air Pressure (AtmPress)	Integrated ATMOS-14 sensor	Outside attached to box	mbar
Ambient air Temperature (AirTemp)	Integrated ATMOS-14 sensor	Outside attached to box	^0C
Relative Humidity (RH)	Integrated ATMOS-14 sensor	Outside attached to box	%
Accumulated rain (Rain)	Hydreon RG-11 Optical Rain Gauge	Outside Drum	mm
Differential pressure between Drum and external atmosphere (DiffPress)	Novus NP785	Inside/Outside Drum	Pa

High quality continuous radon flux observations: Metrology chain

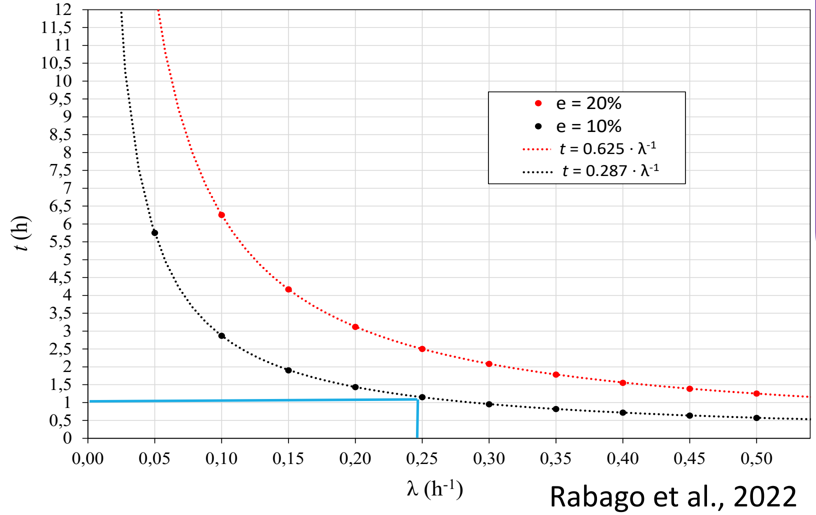
$$C_{Rn}(t) = \boxed{C_0 e^{-\lambda_{eff} t} + \frac{F \cdot A}{V_{eff} \cdot \lambda_{eff}} \left(1 - e^{-\lambda_{eff} t}\right)} \approx \frac{F \cdot A}{V_{eff} \cdot \lambda_{eff}} \cdot \lambda_{eff} t = \frac{F}{h_{eff}} \cdot t = b \cdot t$$



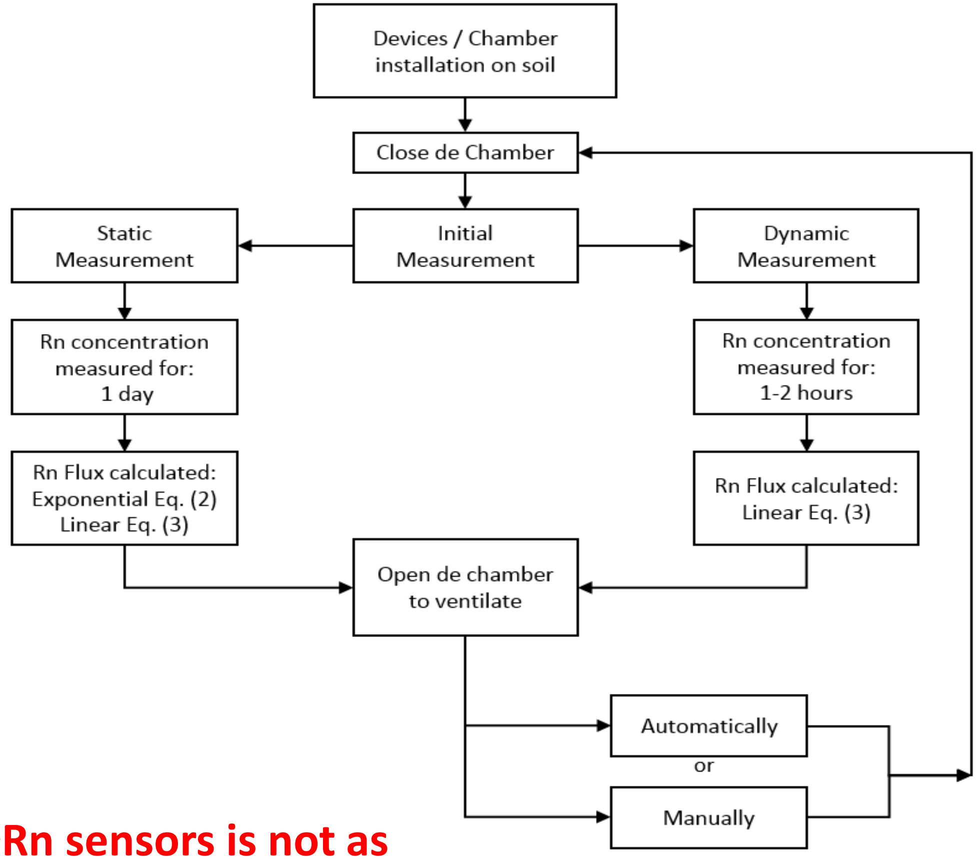
$$fco_2 = J_g = \frac{CO_{2\_slope} \cdot P \cdot V}{A \cdot T \cdot R},$$



Curcoll et al., 2022



Rabago et al., 2022



Rabago et al., 2022

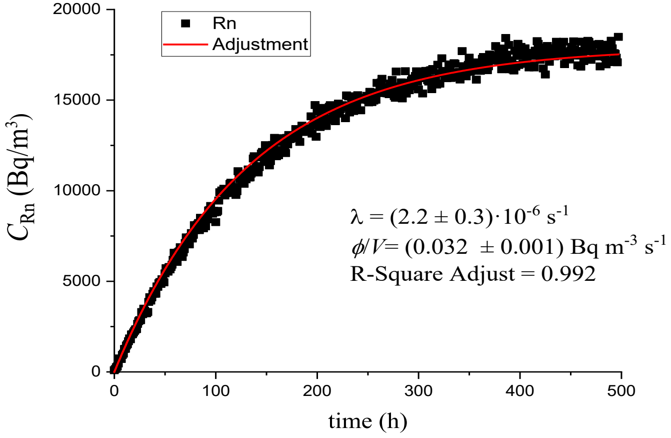
The response of the available ^{222}Rn sensors is not as fast as the one of CO_2 sensors

High ^{222}Rn exhalation bed facility

$$F = \varepsilon \cdot C_{\mathrm{Ra}} \cdot \rho \cdot L \cdot \lambda \cdot \tanh\left(\frac{z}{L}\right)$$

Soil parameters were directly measured by UC

EB was characterized both theoretically and experimentally at the Cantabria University under standard environmental conditions



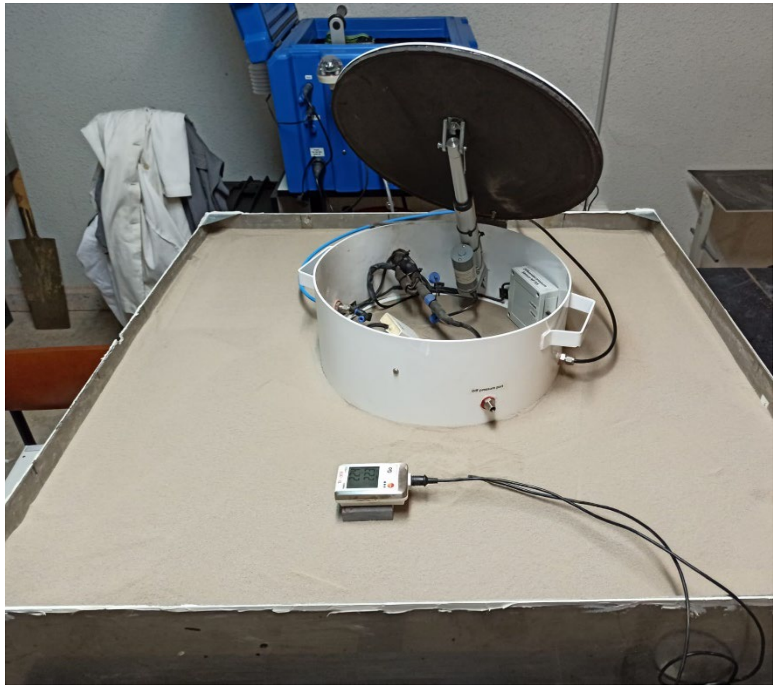
Grossi et al., submitted in AMT

Parameter	Symbol	Result
Emanation factor	ε	0.18 ± 0.03
Radium concentration	C_{Ra}	(19130 ± 350) Bq kg^{-1}
Bulk density	ρ	(1645 ± 2) kg m^{-3}
Grain density	ρ_g	(2570 ± 38) kg m^{-3}
Thickness	z	(0.165 ± 0.005) m
Mass Water content	w_c	(0.0132 ± 0.0004) kg/kg
Water saturation	w_s	(0.061 ± 0.008) m^3/m^3
Porosity	p	0.3599 ± 0.0001
Diffusion coefficient	D	$(3.47 \pm 0.08)\cdot 10^{-6}$ m^2/s
Diffusion length	L	(1.286 ± 0.015) m
Radon decay constant	λ	$2.0993(1)\cdot 10^{-6}$ s^{-1}
^{222}Rn Flux	$F_{Th\_EB} \pm u_{Th\_EB}$	**1918 ± 278 mBq m^{-2} s^{-1}**
Parameter/Variable	**Symbol**	**Result**
Radon emission rate	ϕ	(7.78 ± 0.29) Bq s^{-1}
Height of Chamber	h	(0.225 ± 0.005) m
Air temperature	T	(20.7 ± 0.3) °C
Mass water content in mass	w_c	(0.013 ± 0.001) kg/kg
Air moisture	RH	(47.0 ± 0.7)%
^{222}Rn Flux	$F_{Exp\_EB} \pm u_{Exp\_EB}$	**1757 ± 67 mBq m^{-2} s^{-1}**

Sensor	Manufacturer	Model	Range	Declared Accuracy
Temperature	Testo	175T2	(-35 to 55) ºC	±0.5 ºC
		Probe	(-40 to 120) ºC	±0.3 ºC
Soil moisture	ODYSSEY	Xtreem	(0 to 100) %	±1%
Pressure	ITEFI-CSIC	-	(-600 to 600) Pa	± 3 Pa

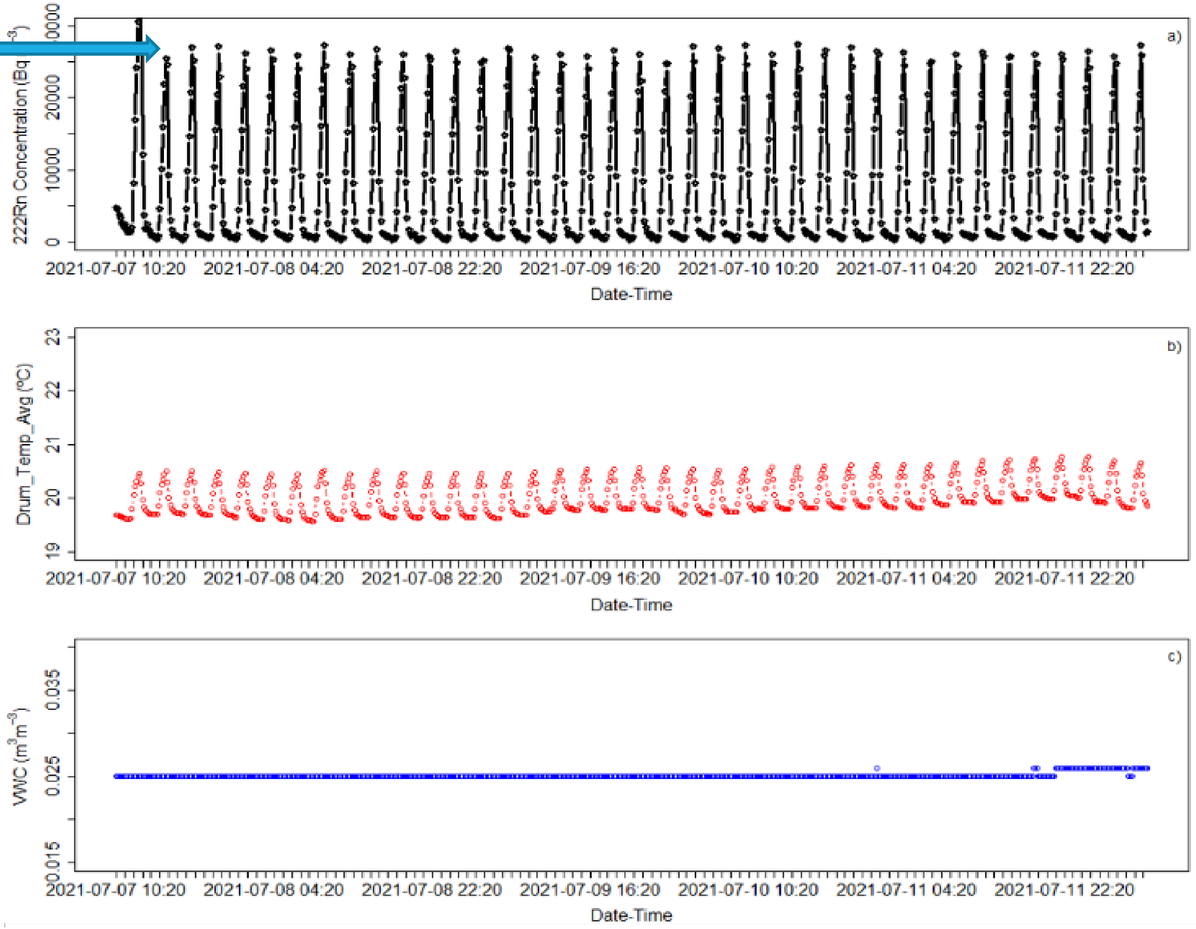
Calibration of the ANSTO Autoflux system

ANSTO *Autoflux* system was selected as TS: it was theoretically characterized and calibrated at the EB only for standard environmental conditions (due to project milestones and deliverable not experiments were done with extreme env. conditions and for low radon flux EB)



Constant response

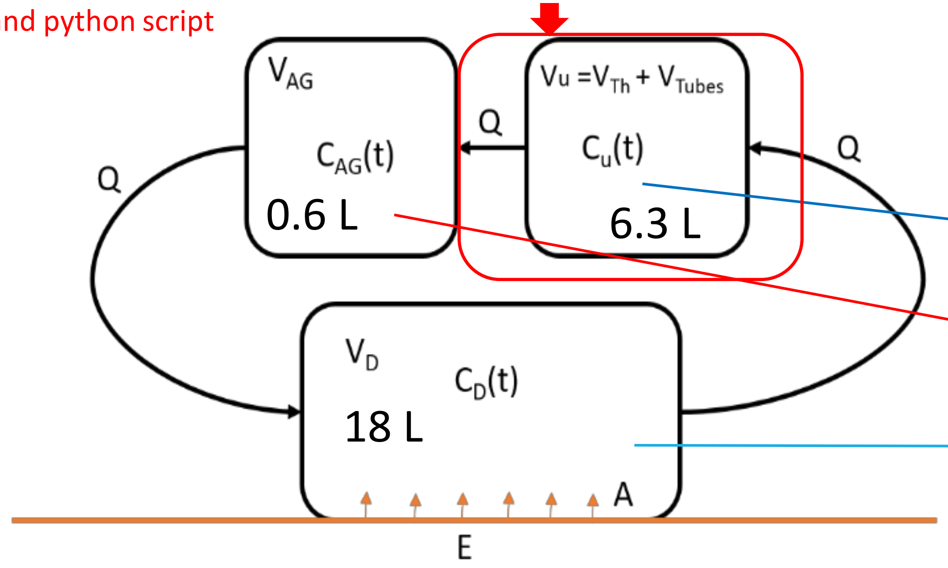
$$\mathbf{F} = \mathbf{b} \cdot h_{eff}$$



$$\left(\frac{u_{Cal\_Autoflux}}{F_{Cal\_Autoflux}}\right)^2 = \left(\frac{u_{Autoflux}}{F_{Autoflux}}\right)^2 + \left(\frac{u_{Exp\_EB}}{F_{Exp\_EB}}\right)^2 + \left(\frac{u_{F\_Corr}}{F_{Corr}}\right)^2$$

^{222}Rn flux Transfer Standard instrument

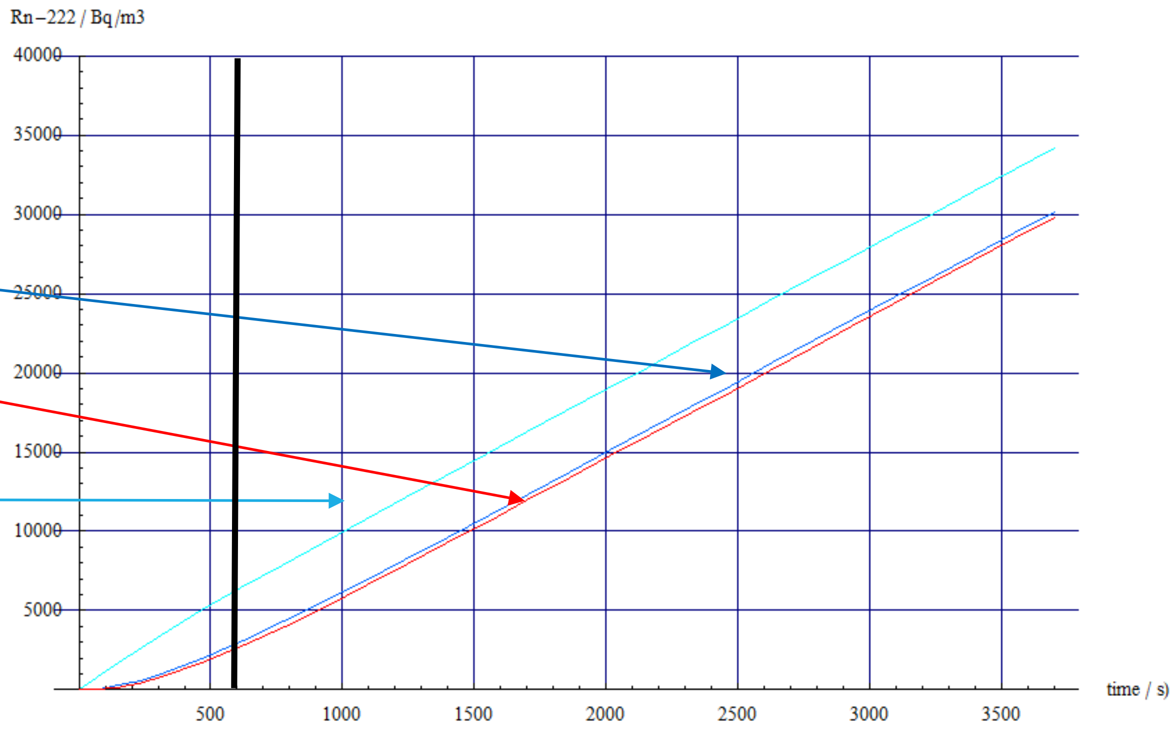
PLEASE NOTE: This volume was initially considered within the ANTO *Autoflux* user manual and python script



$$\frac{dC_D(t)}{dt} = \frac{F \cdot A}{V_D} - C_D(t) \cdot \frac{Q}{V_D} + C_{AG}(t) \cdot \frac{Q}{V_{AG}}$$

$$\frac{dC_u(t)}{dt} = C_D(t) \cdot \frac{Q}{V_D} + C_u(t) \cdot \frac{Q}{V_u}$$

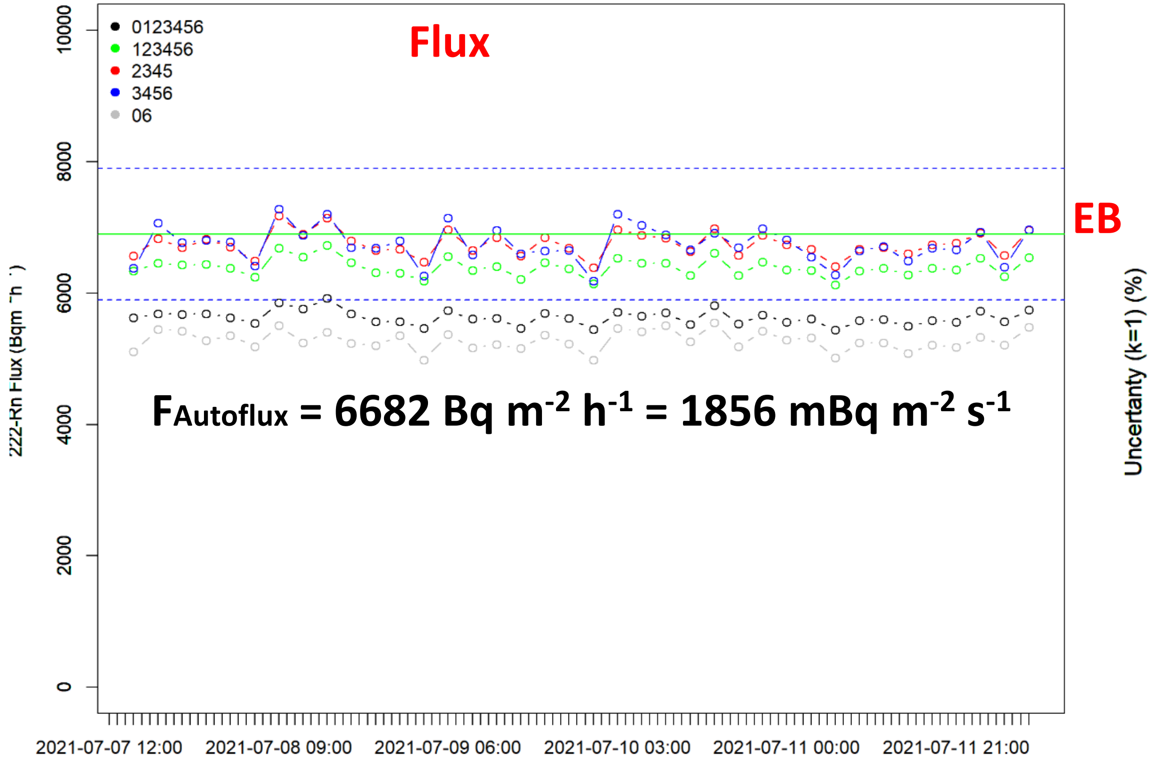
$$\frac{dC_{AG}(t)}{dt} = C_u(t) \cdot \frac{Q}{V_u} + C_{AG}(t) \cdot \frac{Q}{V_{AG}}$$

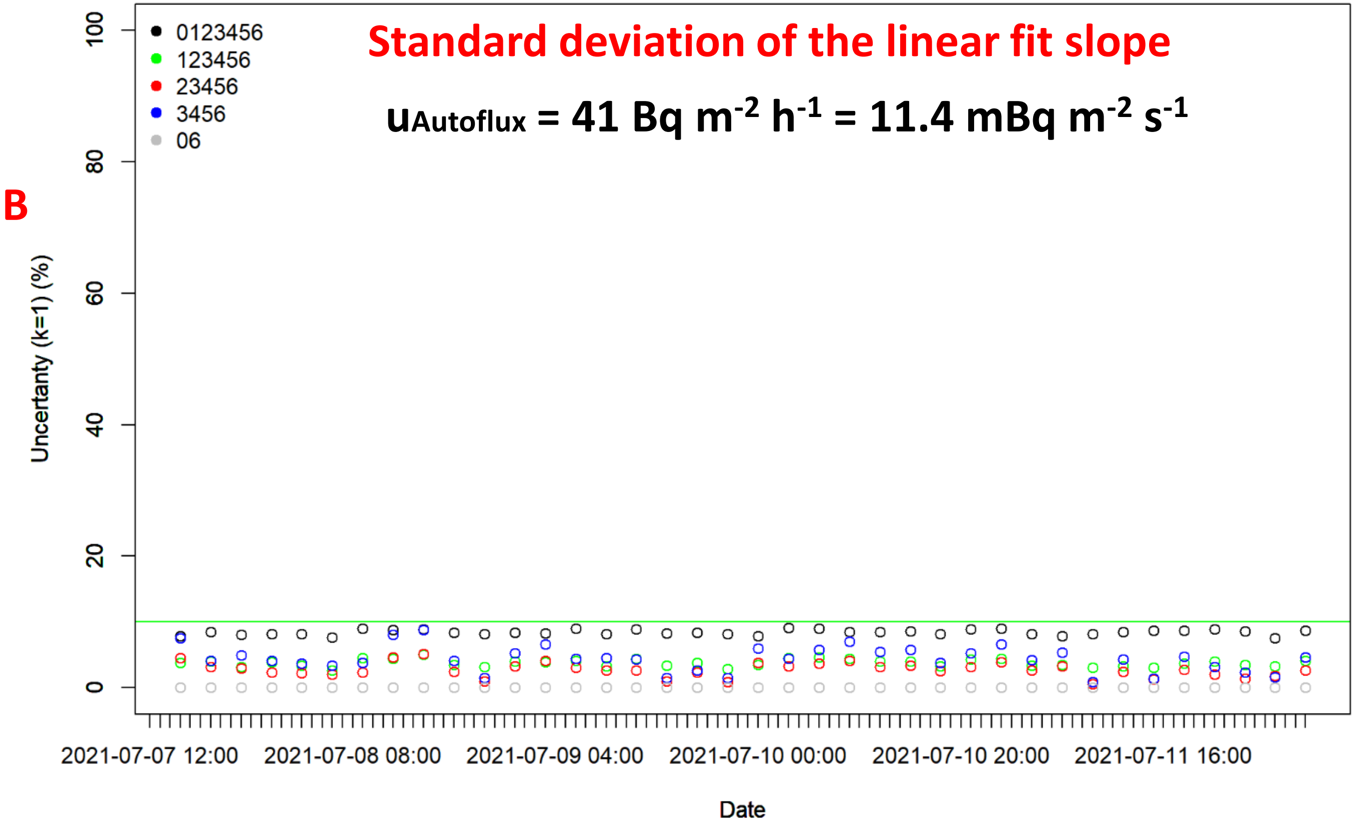


Simulated ^{222}Rn concentration behavior within each one of the volumes of the *AutoFlux* system during the hour for which the chamber was closed C_D (light blue line), C_u (blue line) and C_{AG} (red line).

High quality continuous radon flux observations: Metrology chain

^{222}Rn flux Transfer Standard instrument





$$\left(\frac{u_{Cal\_Autoflux}}{F_{Cal\_Autoflux}}\right)^2 = \left(\frac{u_{Autoflux}}{F_{Autoflux}}\right)^2 + \left(\frac{u_{Exp\_EB}}{F_{Exp\_EB}}\right)^2 + \left(\frac{u_{F\_Corr}}{F_{Corr}}\right)^2$$

$F_{Cal\_Autoflux} \pm u_{Cal\_Autoflux}$ = 0.95 ± 0.07

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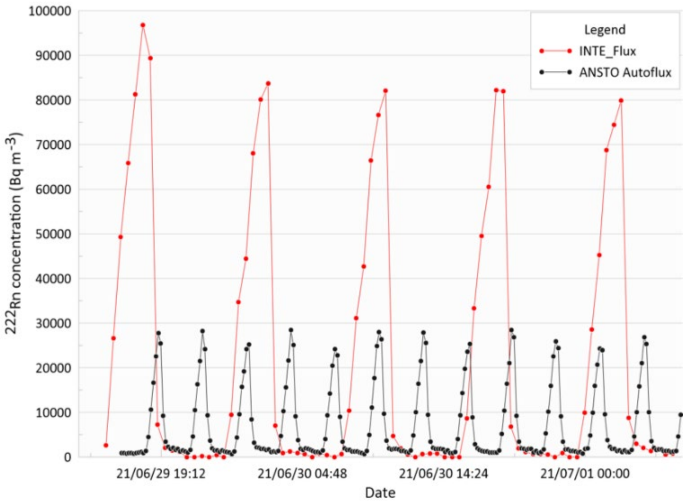
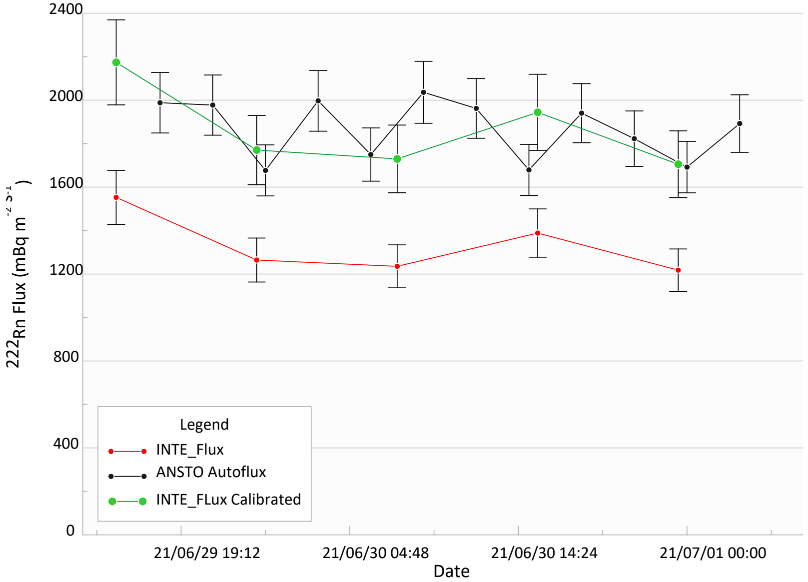
ANSTO *Autoflux* system was selected as TS: it was theoretically characterized and calibrated at the EB and used to calibrate secondary monitors

^{222}Rn flux client system



Grossi et al., submitted AMT

$F_{Cal} \pm u_{Cal}$ = 1.33 ± 0.12 (9%)



$$\left(\frac{u_{Cal}}{F_{Cal}}\right)^2 = \left(\frac{u_{Client}}{F_{Client}}\right)^2 + \left(\frac{u_{ref}}{F_{ref}}\right)^2 + \left(\frac{u_{Cal\_Autoflux}}{F_{Cal\_Autoflux}}\right)^2$$

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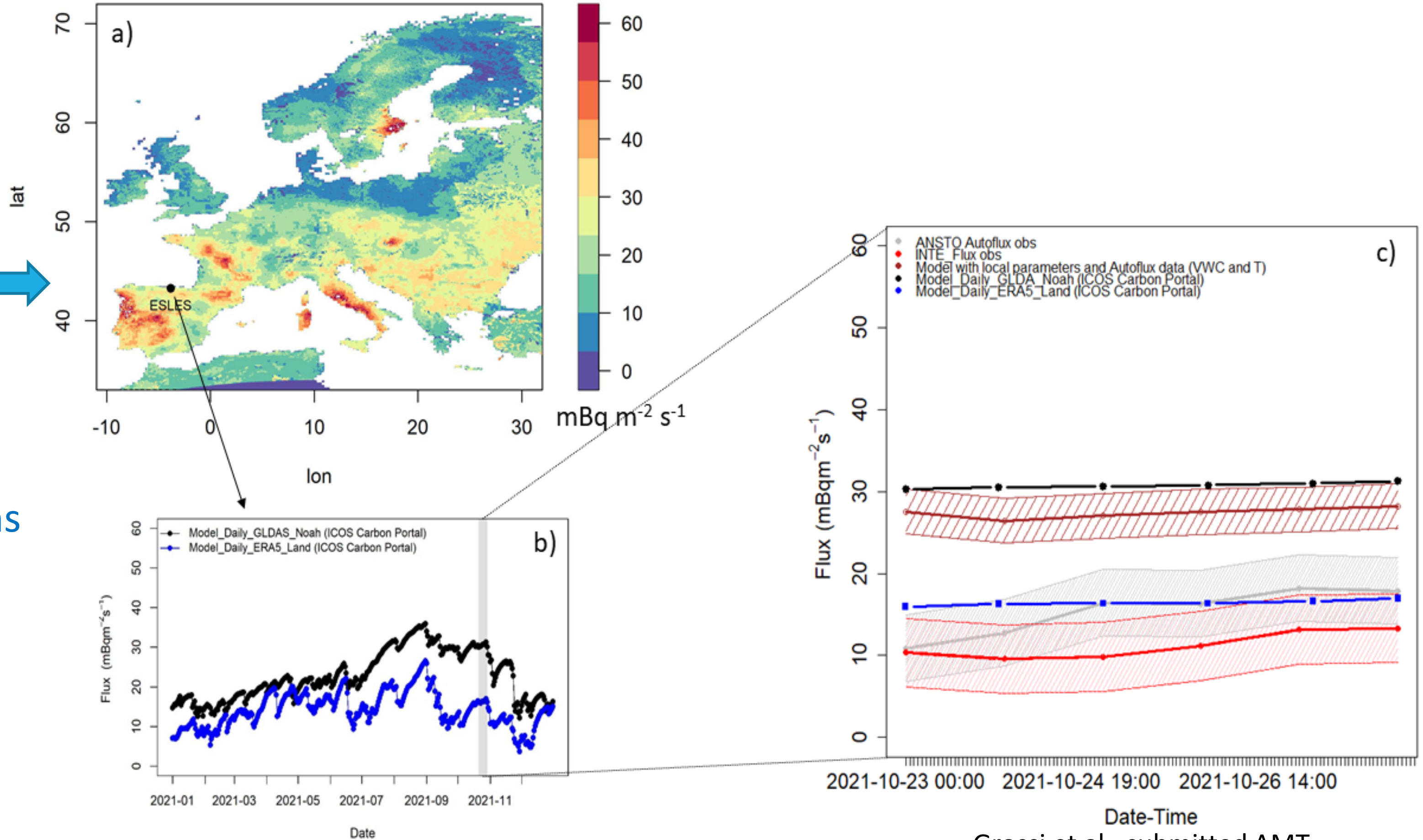
Two intercomparison campaigns of ^{222}Rn flux systems available from the literature were carried out in the northwester Spain in October 2021. Observations were compared with traceRadon radon flux daily outputs for the same period.

Rabago et al., 2022

^{222}Rn flux systems intercomparison campaigns



Grossi et al., submitted AMT

Intense radon flux campaigns in Europe

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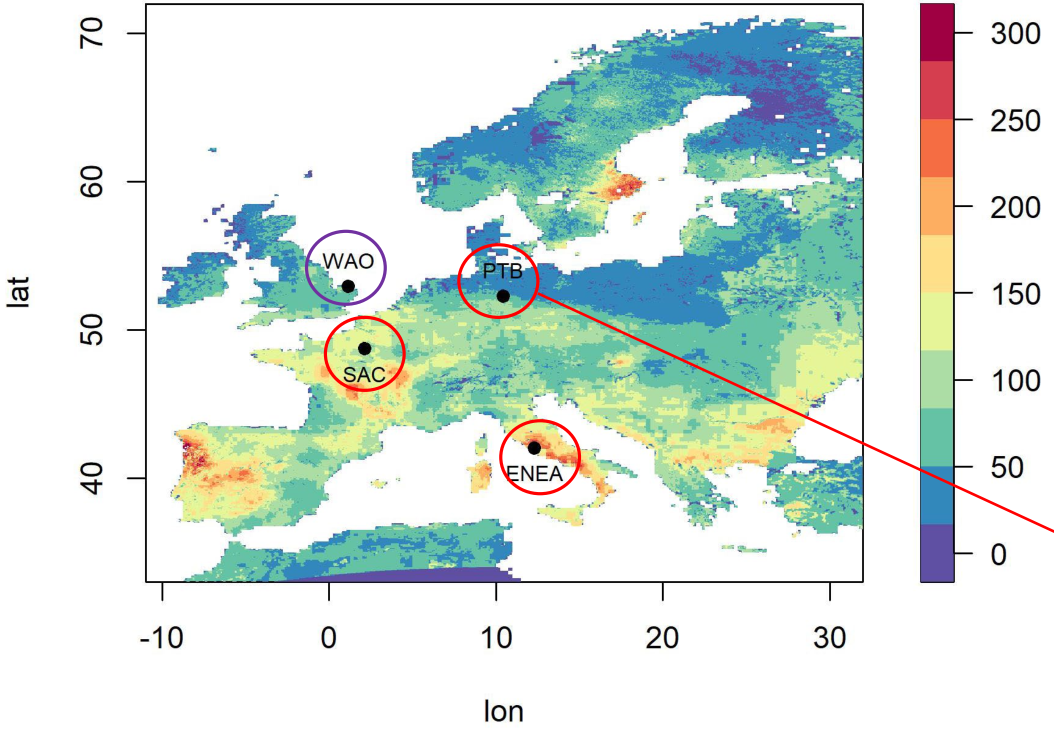
The WP2 and WP3 of the traceRadon Project aim to:

1. Generate daily and monthly radon flux maps;
2. Offer a full metrology chain and infrastructure for continuous radon flux measurements;
3. **Carry out four intense radon flux campaigns to validate radon flux maps and their input variables and parameters**

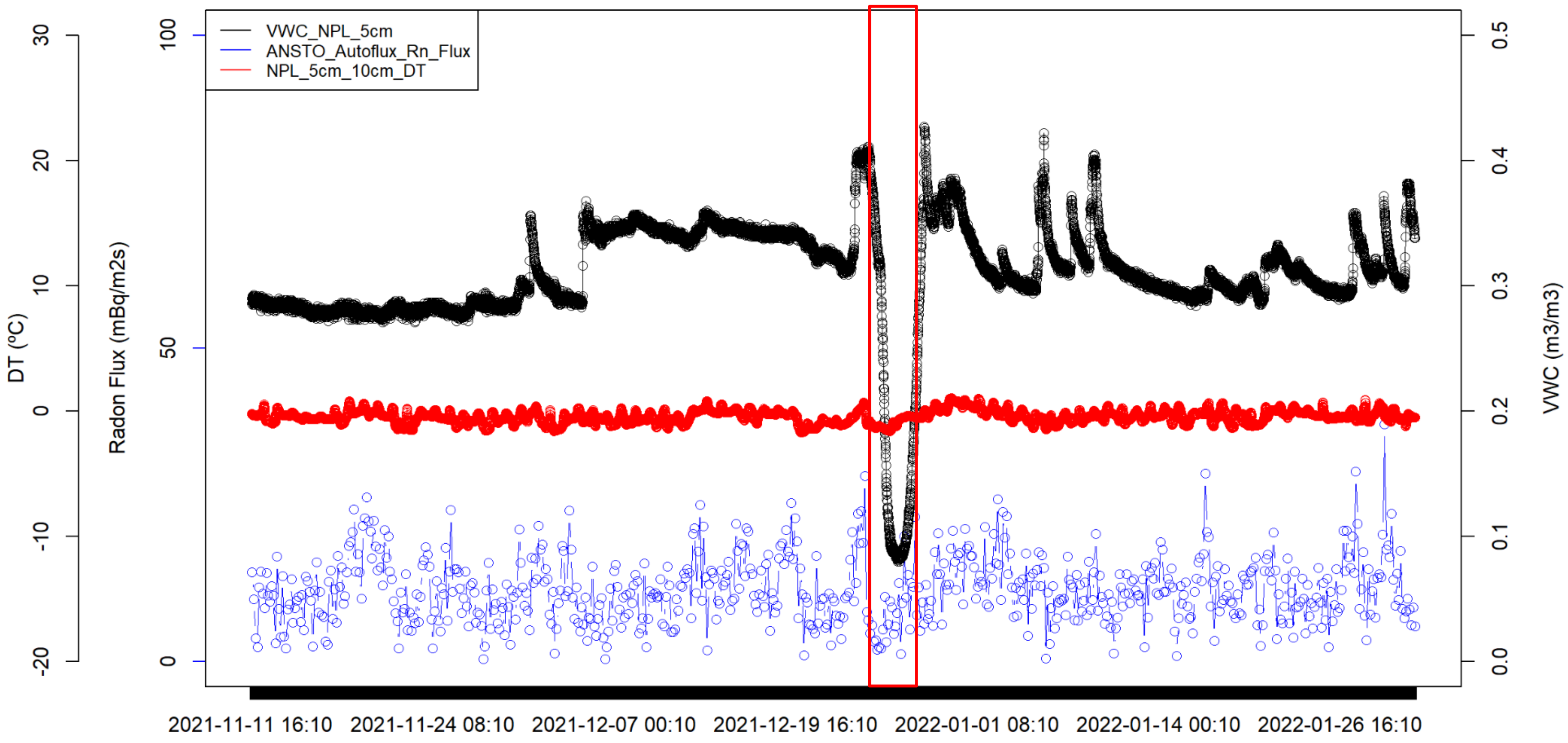


Each campaign lasted 3 months and several instruments and methods were applied to measure:

- ^{222}Rn Flux (UPC, UHEI);
- Environmental conditions in the soil (Volume Water Content, Temperature, etc.), within the accumulation chamber and air (NPL, UPC);
- Gamma spectrometry at 1 m from the ground (UPC);
- Environmental Dose Rate (PTB, ENEA);
- Radionuclides activity in the soil (ENEA);
- Physical characteristics of the soil (porosity, density, etc.) (home research group).

Intense radon flux campaigns in Europe

PTB



Soil Moisture

Soil temperature gradient between 5cm and 10 cm soil

3h resolution
^{222}Rn Flux

Intense radon flux campaigns in Europe

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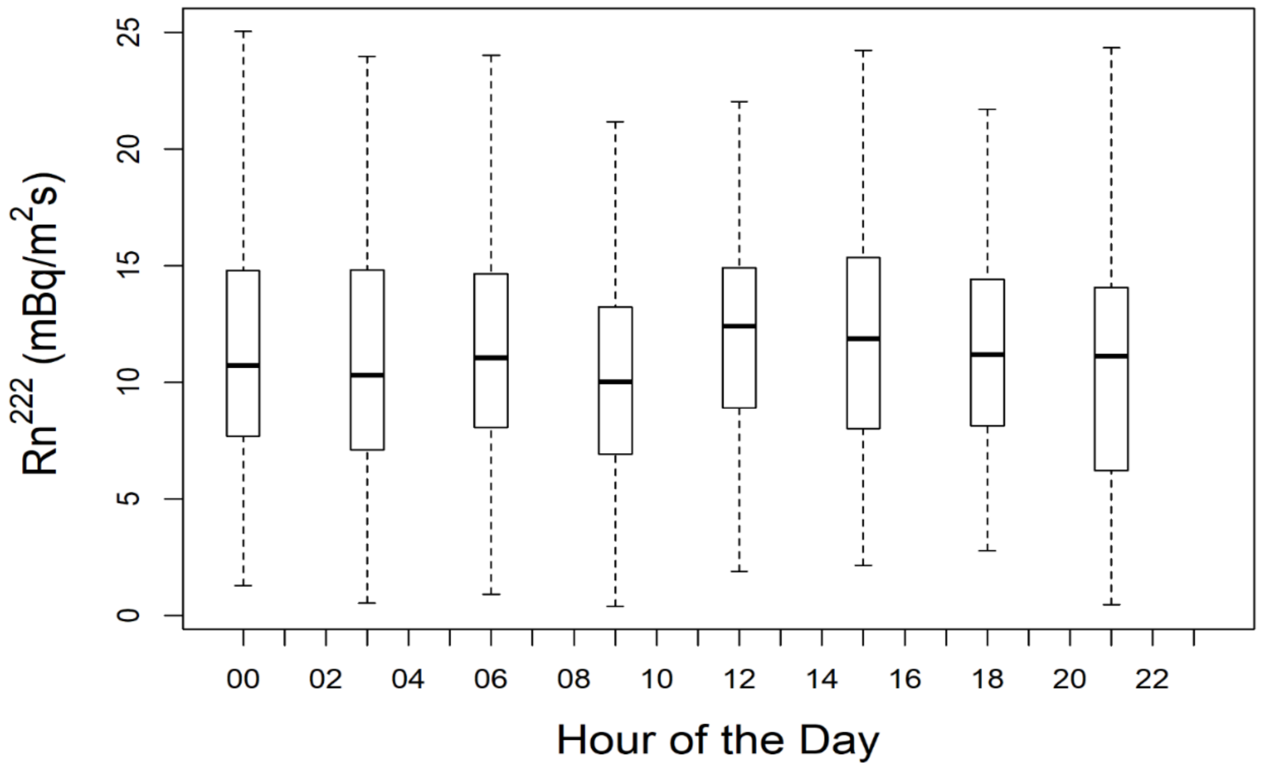
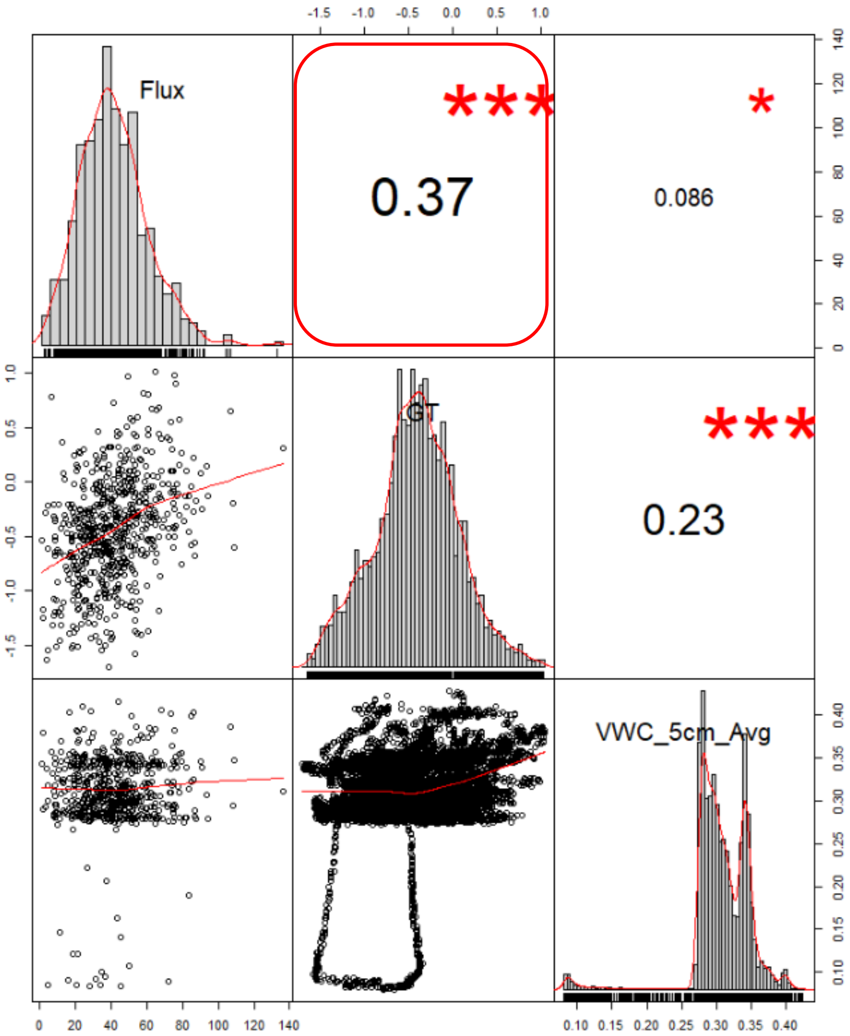
PTB

High sub diurnal variability is observed in the radon flux time series not justified as day/night variation





Intense radon flux campaigns in Europe

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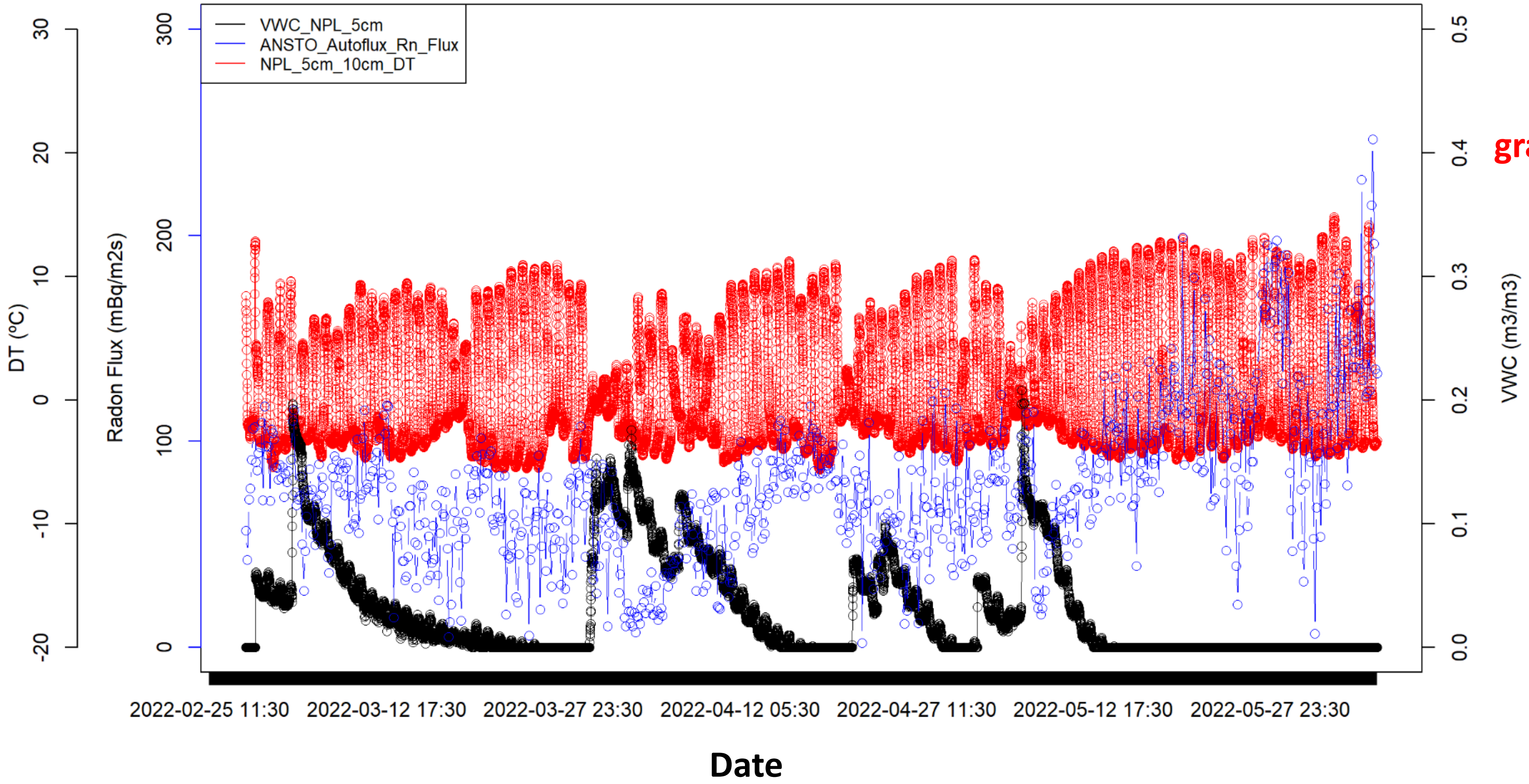
ENEA



Soil temperature gradient between 5cm and 10 cm soil

3h resolution ^{222}Rn Flux

Soil Moisture

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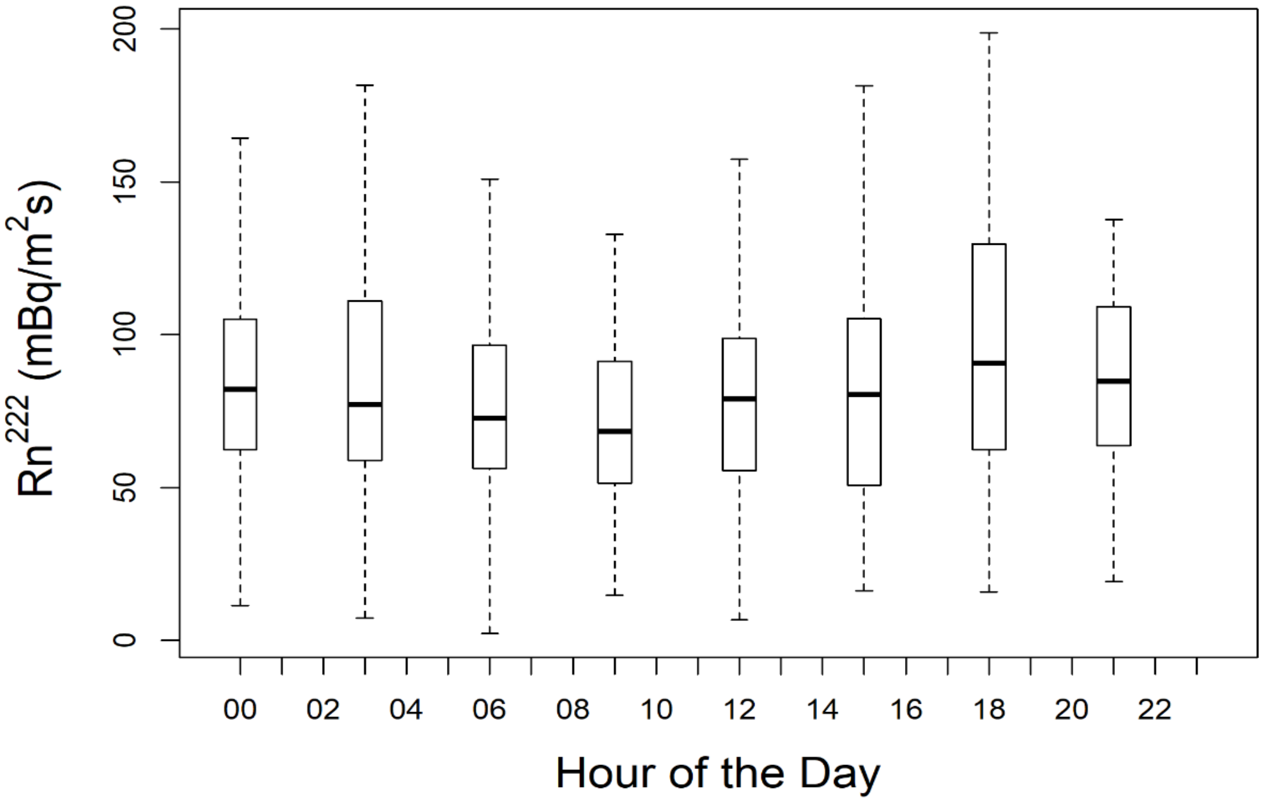
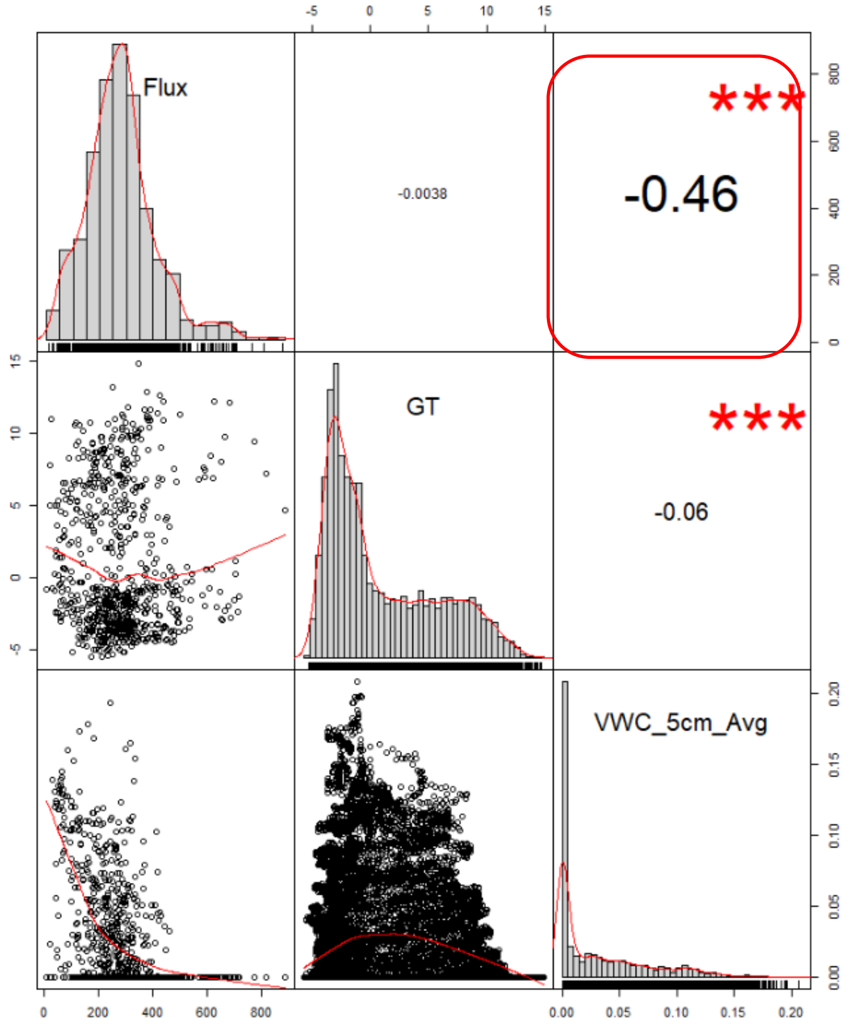
ENEA

High sub diurnal variability is observed in the radon flux time series not COMPLETELY justified as day/night variation





Intense radon flux campaigns in Europe

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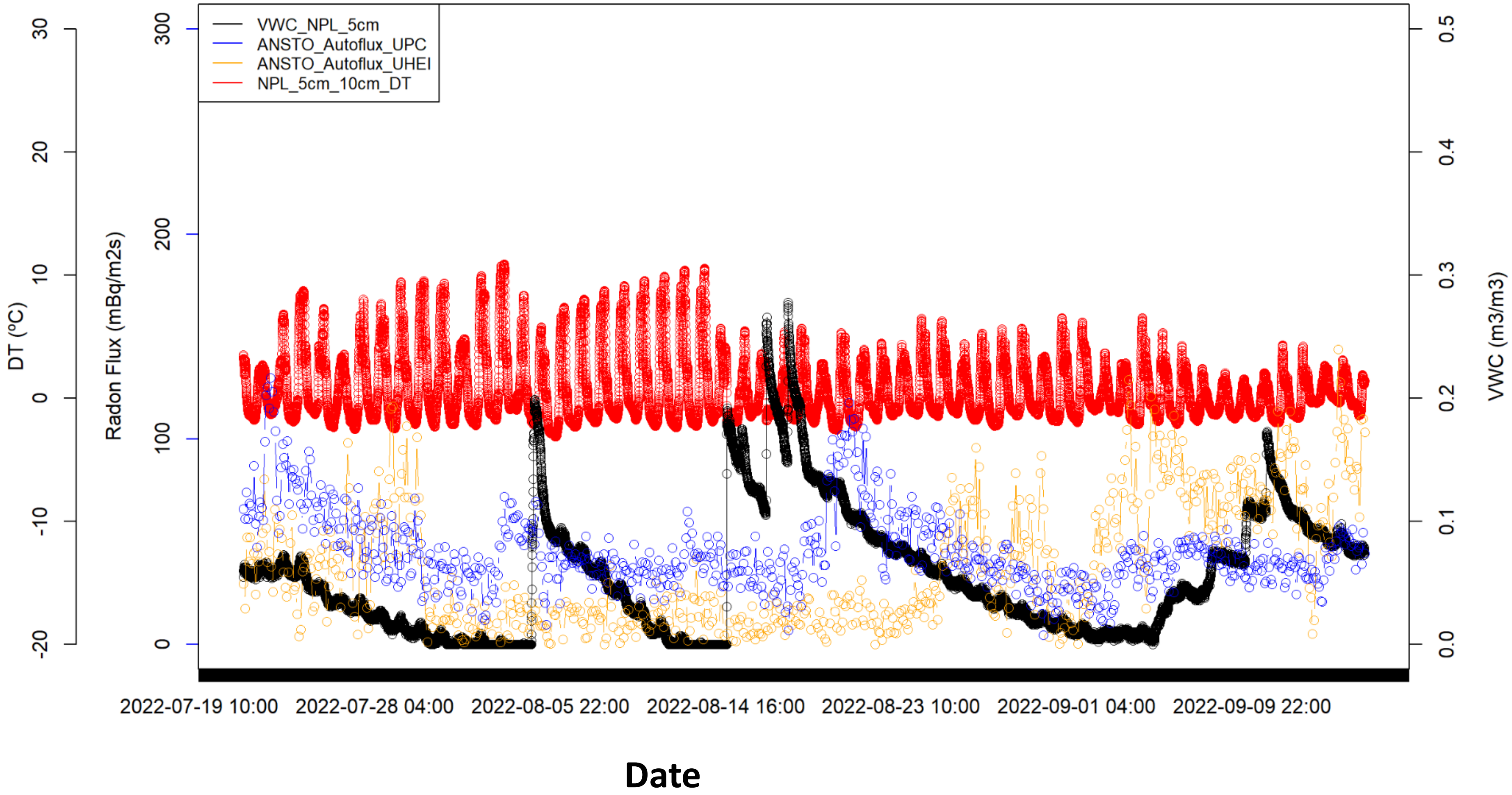
SACLAY



Soil temperature gradient between 5cm and 10 cm soil

3h resolution ^{222}Rn Flux UPC

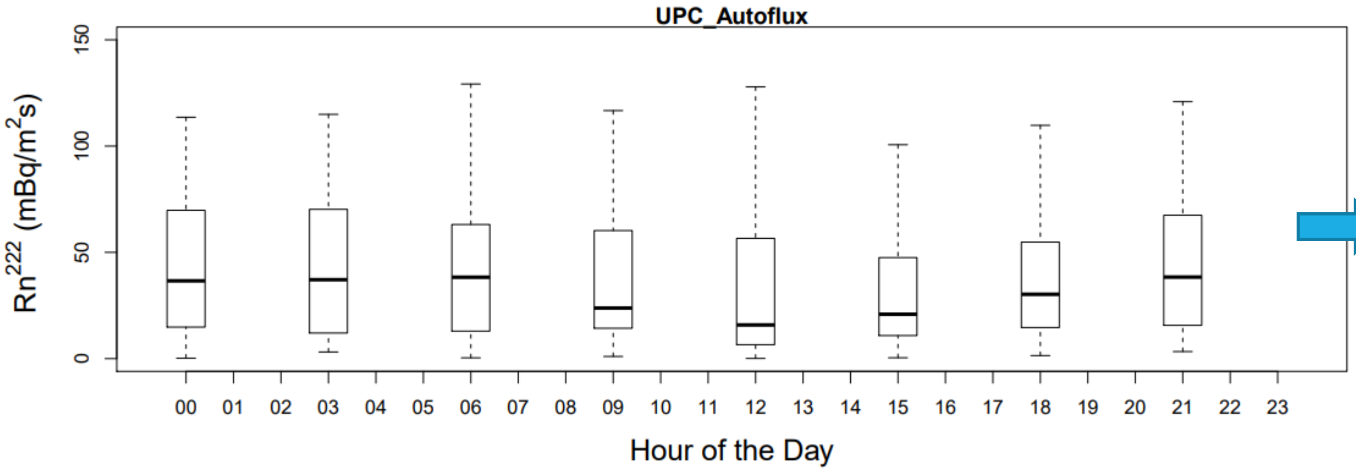
3h resolution ^{222}Rn Flux UHEI

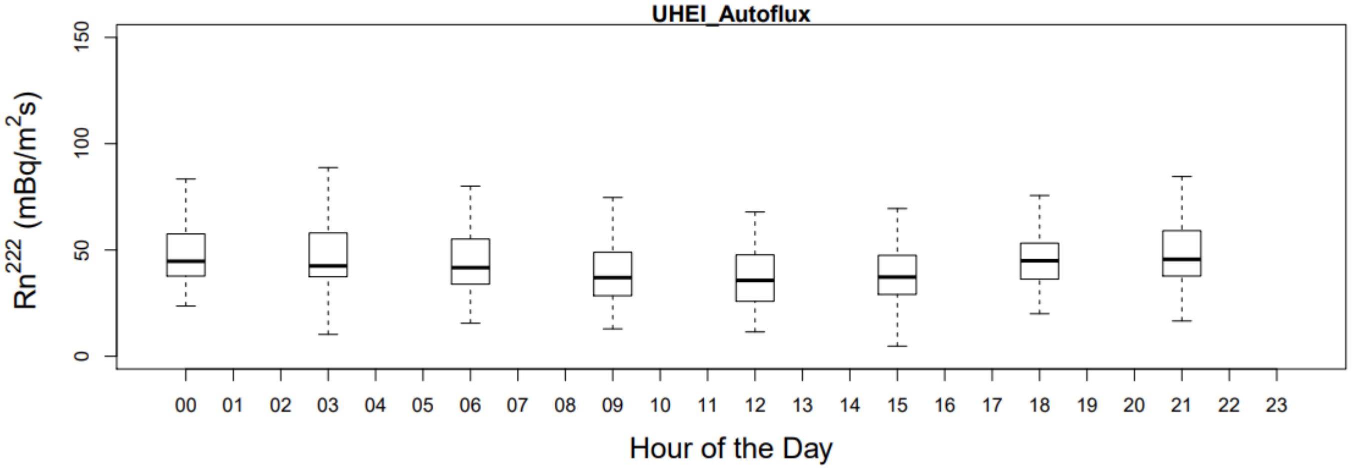
Soil Moisture

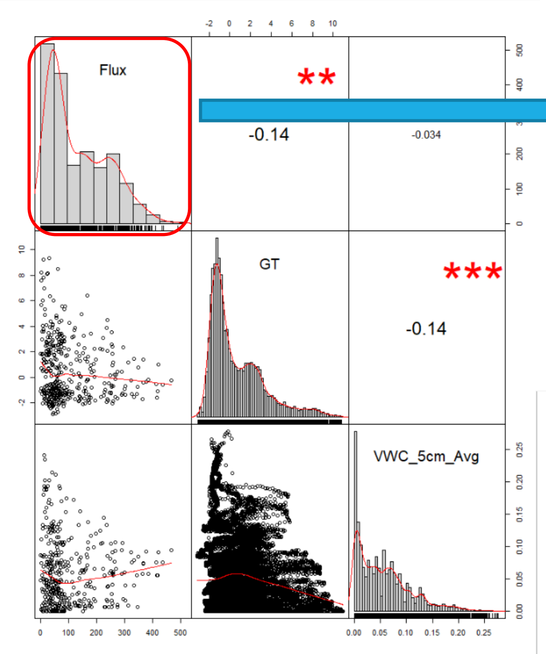
Intense radon flux campaigns in Europe

SACLAY

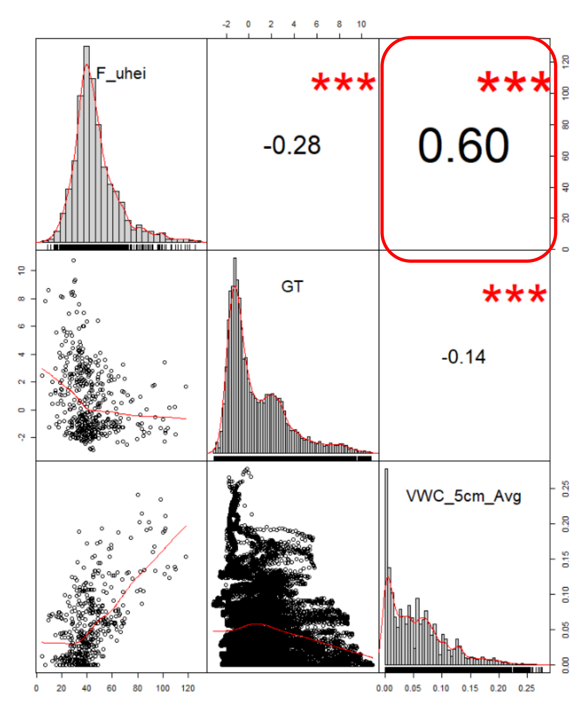
Higher sub diurnal variability is observed in the radon flux time series from ANSTO_UPC system







Different distribution



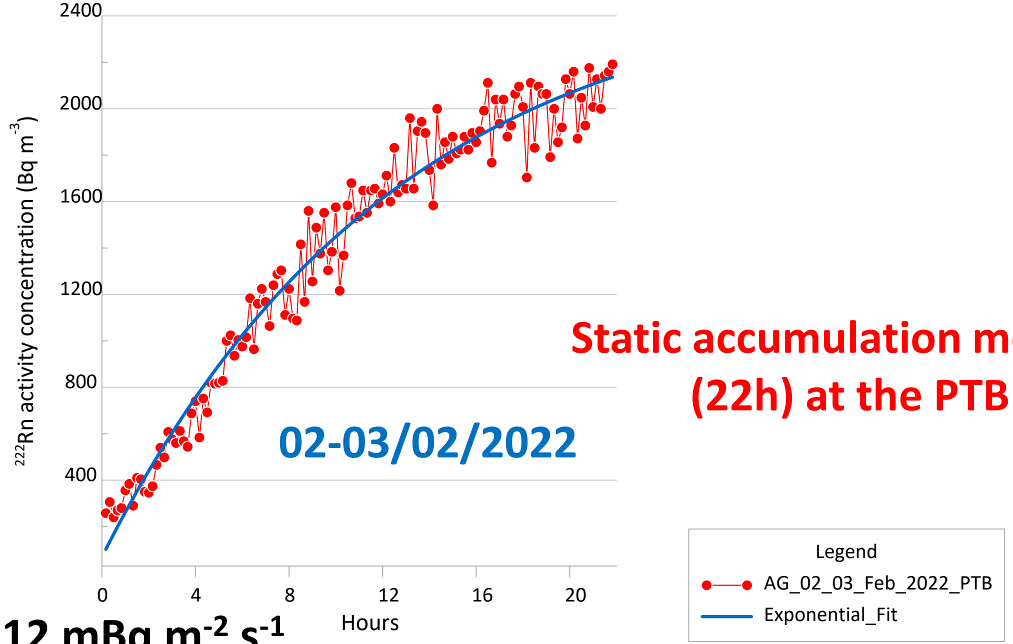
Linear Method with Flux systems: Is it reliable?

$\lambda^0 t$ = 0.08 for 1h measurement

Linear method can be applied

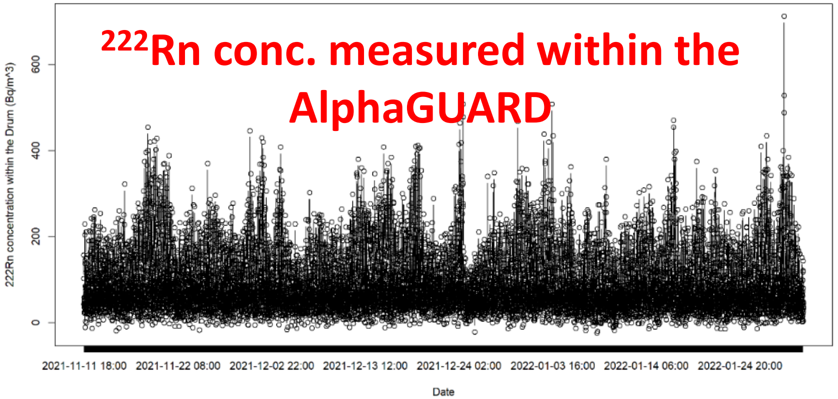
Where h is the effective height of the accumulation chamber equal to the Surface (A) divided by the volume (V_u)

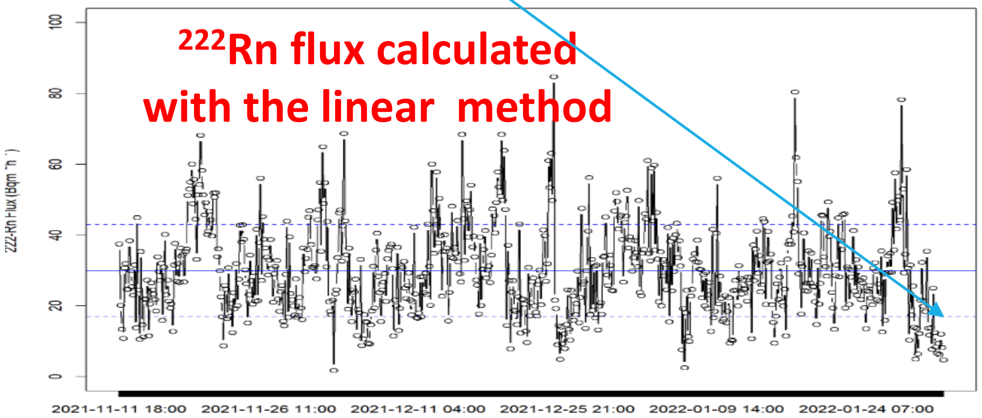
^{222}Rn activity concentration measured within the closed Autoflux drum at the PTB site



Static accumulation method (22h) at the PTB

Flux = $(C_0+a)*b*h=(2502+69.5)*0.08*0.204=$ **42 Bq m^{-2} h^{-1} = 12 mBq m^{-2} s^{-1}**





Mean = 11 Bq m^{-2} h^{-1}
St. Deviation = 3.6 Bq m^{-2} h^{-1}

Linear Method with Flux systems: Is it reliable?

$\lambda^0 t$ = 0.29/0.40 for 1h measurement

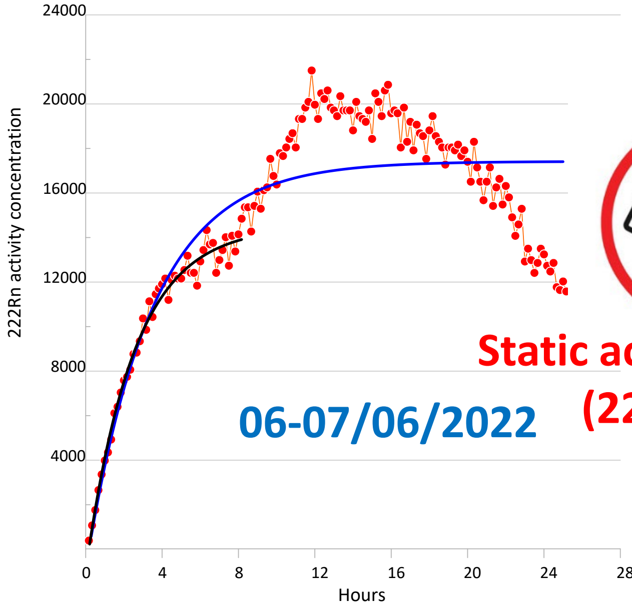
Where h is the effective height of the accumulation chamber equal to the Surface (A) divided by the volume (V_u)

Flux = $(C_0+a)*b*h=(18335-912)*0.29*0.204=$ **1031 Bq m^{-2} h^{-1} = 286 mBq m^{-2} s^{-1}**

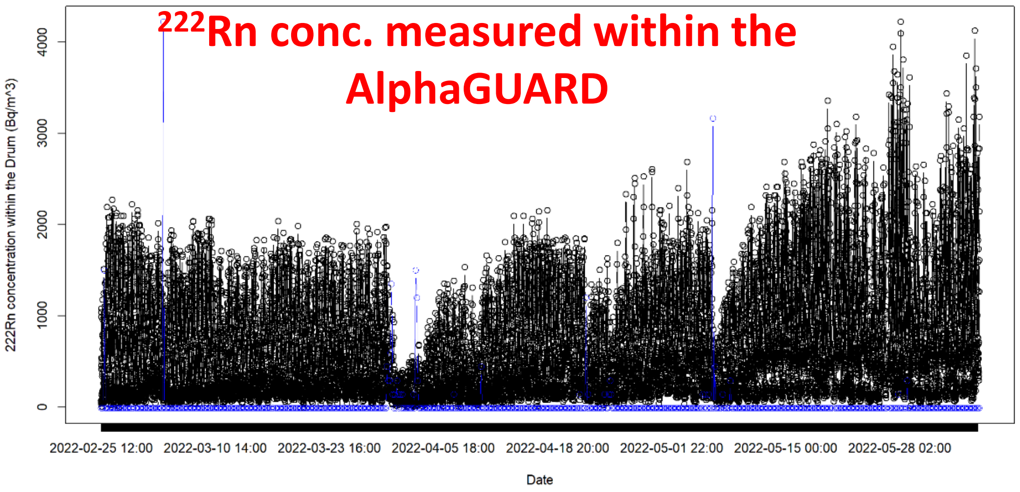
Flux = $(C_0+a)*b*h=(15508-993)*0.40*0.204=$ **1184 Bq m^{-2} h^{-1} = 329 mBq m^{-2} s^{-1}**

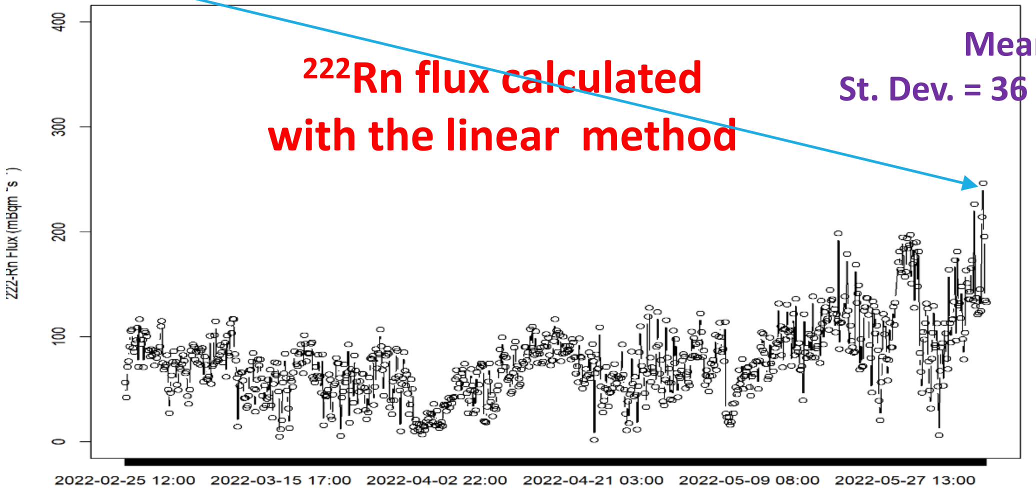
Flux = $b*h=3940*0.204=$ **803 Bq m^{-2} h^{-1} = 223 mBq m^{-2} s^{-1}**

^{22}Rn activity concentration measured within the closed Autoflux drum at ENEA site



Static accumulation method (22h) at the ENEA





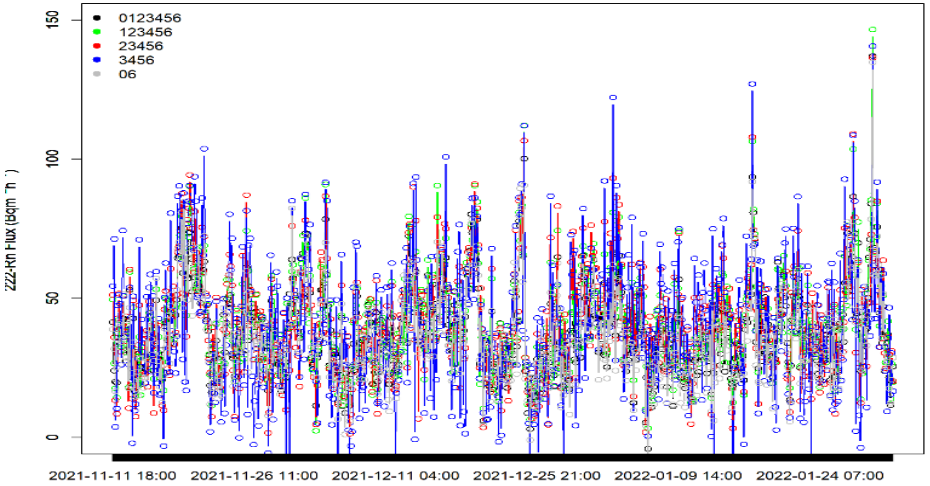
Mean = 78 mBq m^{-2} s^{-1}
St. Dev. = 36 mBq m^{-2} s^{-1}

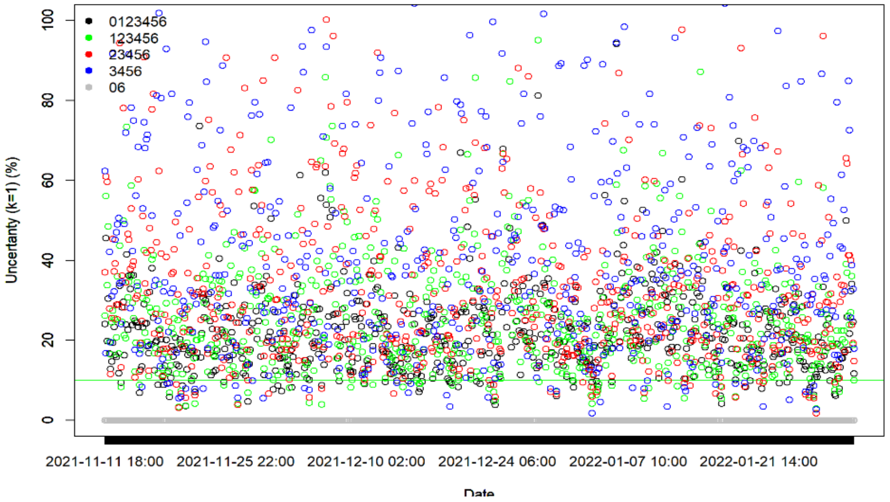
Linear Method with Flux systems: Is it reliable?

Flux values

Statistical Uncertanty

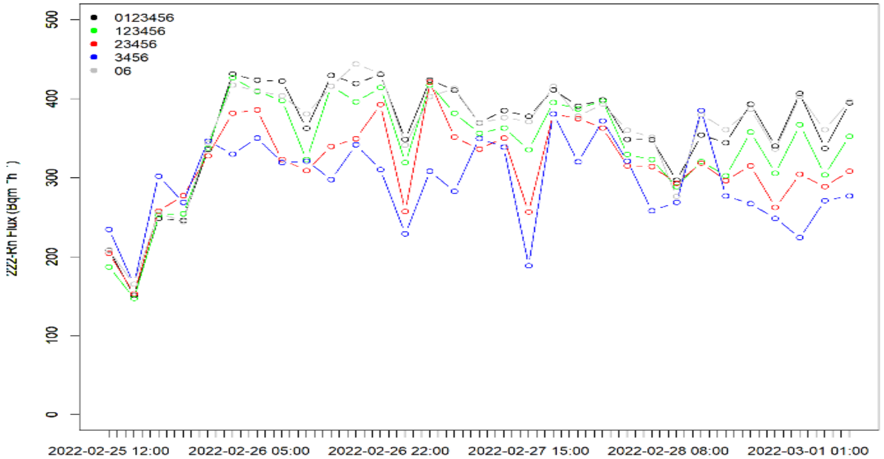
Low Flux (PTB)

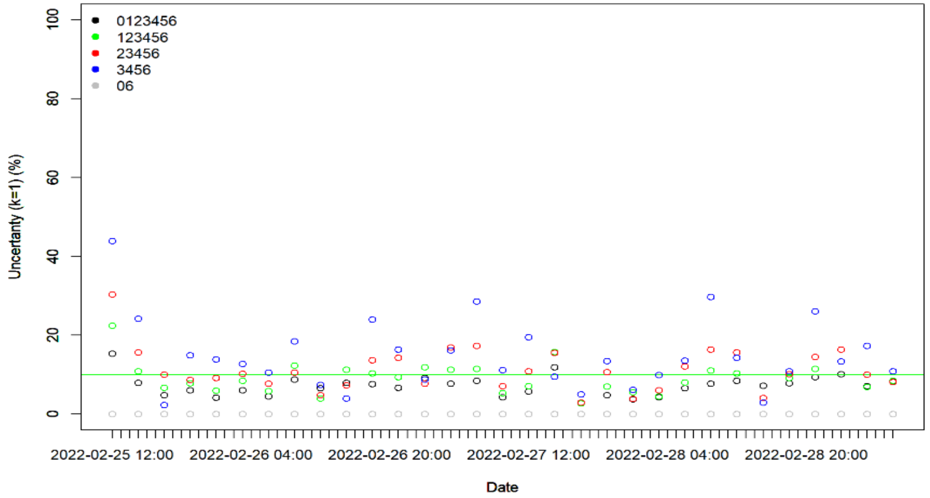




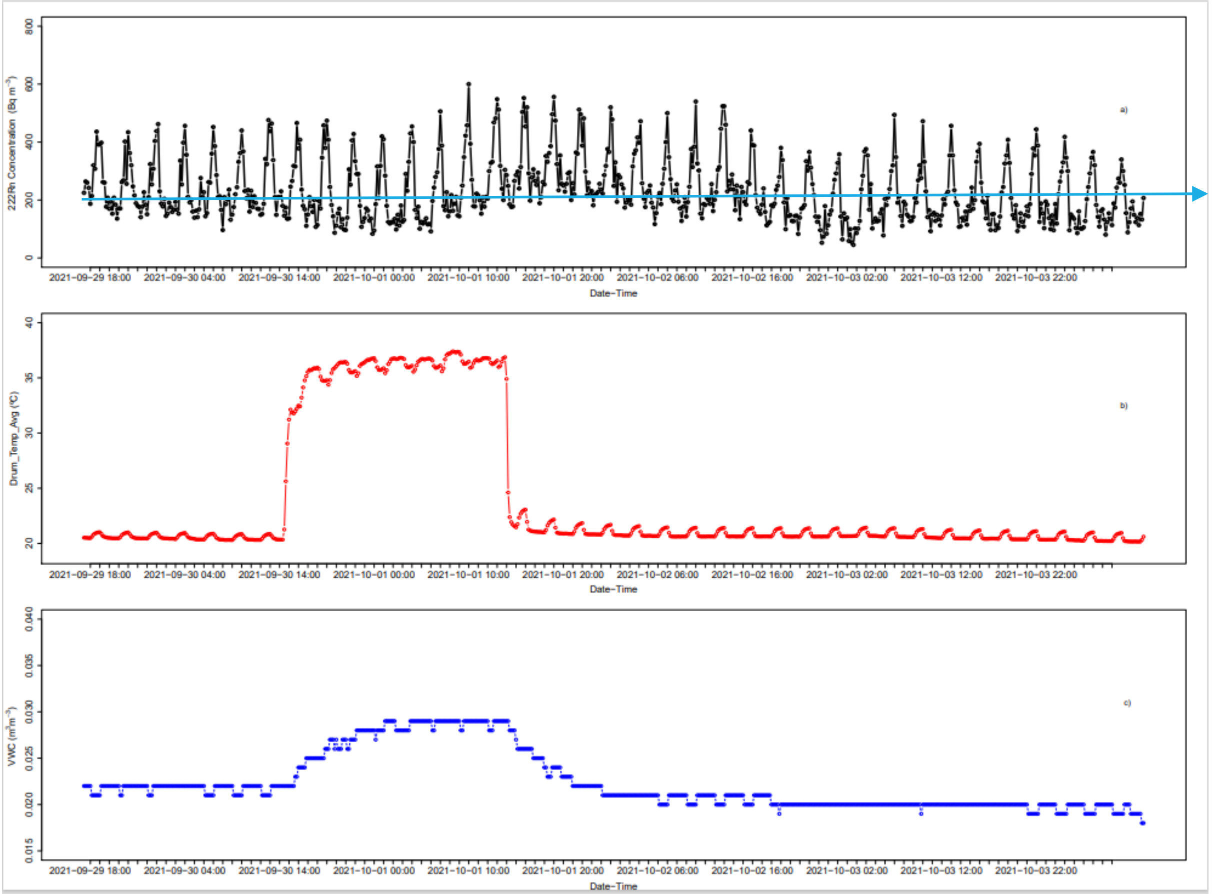
The AlphaGUARD monitor is not sensible enough to measure small radon increases within the accumulation chamber at low radon exhalation areas

High Flux (ENEA)



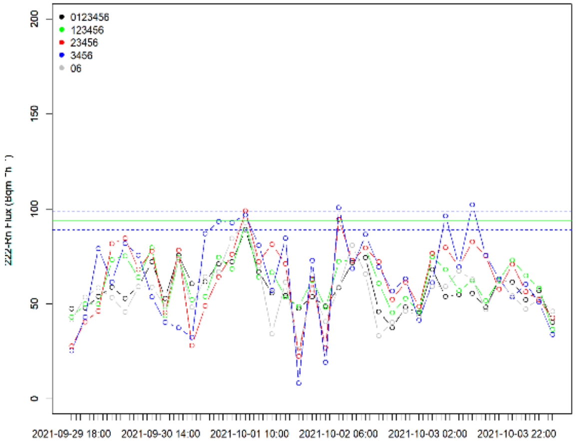


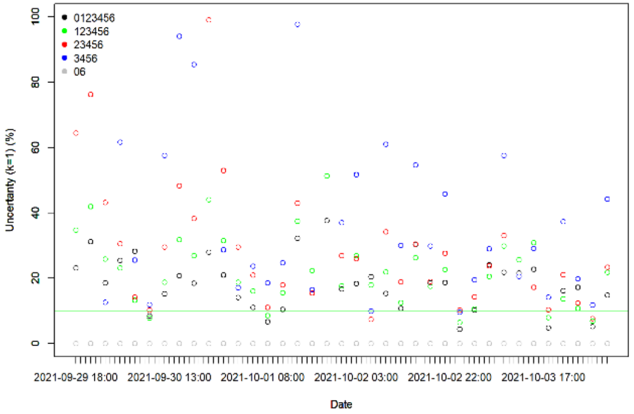
^{222}Rn flux Transfer Standard instrument with a low exhalation bed (25 mBq m^{-2} s^{-1})



The experiment was done during the weekend and the laboratory door was closed. The radon concentration increase in the ambient air is influencing the measurements initial conditions.

Due to the project deadlines the experiments were not repeated.





Conclusions and Further steps

- Rdon concentration measurements are not so fast as CO_2 measurements. Larger measurements (at least 30minuts) are needed to apply the linear method for radon flux calculations against the few minutes needed for CO2 meas. This could affect the first systems leakages.
- The ANSTO Autoflux system initially presented several incongruences in its defined volumes and in the description of the radon behavior within its different volumes. We were able to study and understand and correct them but this generated a huge delay within the traceRadon activities plan for WP2.
- Theoretical characterization and experimental calibration of the selected Autoflux ANSTO system as possible Transfer Standard was performed using only a constant and high reference flux and only under standard environmental conditions.
- No studies could be performed to observe the system response under extreme climate conditions in the soil or in the ambient air and for fluxes in the same orders of magnitude than from common soil.
- ^{222}Rn fluxes and environmental variables were measured at three sites during 2021-2022. Observed fluxes show high sub diurnal variability which should be better understood. So far we are not able to understand if the observed variability is due to real flux changes and to artefacts due to the chamber presence on the soil over long time (tens of minutes).
- Limitations of this transfer standard may be related to the fix position of the chamber in the soil. A Licor accumulation chamber could be a better option for further studies which is now possible to be bought separately.
- The EB was too simple. We need a facility with a deeper soil columns and sensors to observe the T, VWC, etc. gradients during the measurements.

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- Alphabetically
- Chronologically

2023

Outcomes of the traceRadon project: radon metrology for use in climate change observation and radiation protection at the environmental level

01 Mar 2023–01 Feb 2025 | Annette Röttger, Ute Karstens, Claudia Grossi, Helen Worden, Bin Yuan, Huilin Chen, and Hartwig Harder |

Information

An overlapping need exists between the climate research and the radiation protection communities to improve the metrology for atmospheric radon concentration and radon flux measurements. The EMPIR project 19ENV01 traceRadon works toward these goals for the benefit of both large scientific communities by providing the necessary infrastructure for measuring these aforementioned variables. In addition, it will generate data at four selected European sites for validation of radon flux models and inventories and will create the first standard protocol for applying the radon tracer method (RTM). The proposed special issue aims to collect the direct and indirect outcomes of the traceRadon project. It will include papers presenting results of the laboratory and field campaigns carried out within the project. In addition, papers directly related to the project goals will be welcome too.

Hide

traceRadon

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