EURADOS AM2019 winter school

Rn-222 as tracer for quantifying greenhouse gases fluxes: need of high quality and harmonized measurements of atmospheric concentrations and fluxes.

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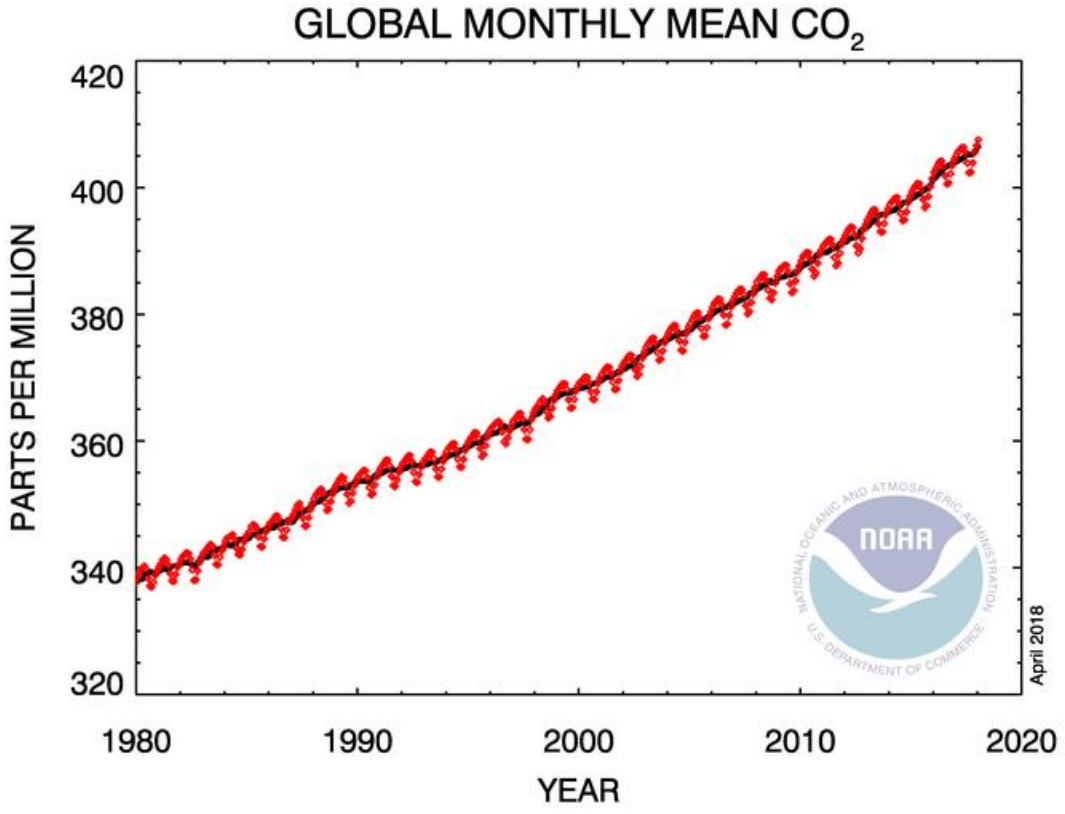
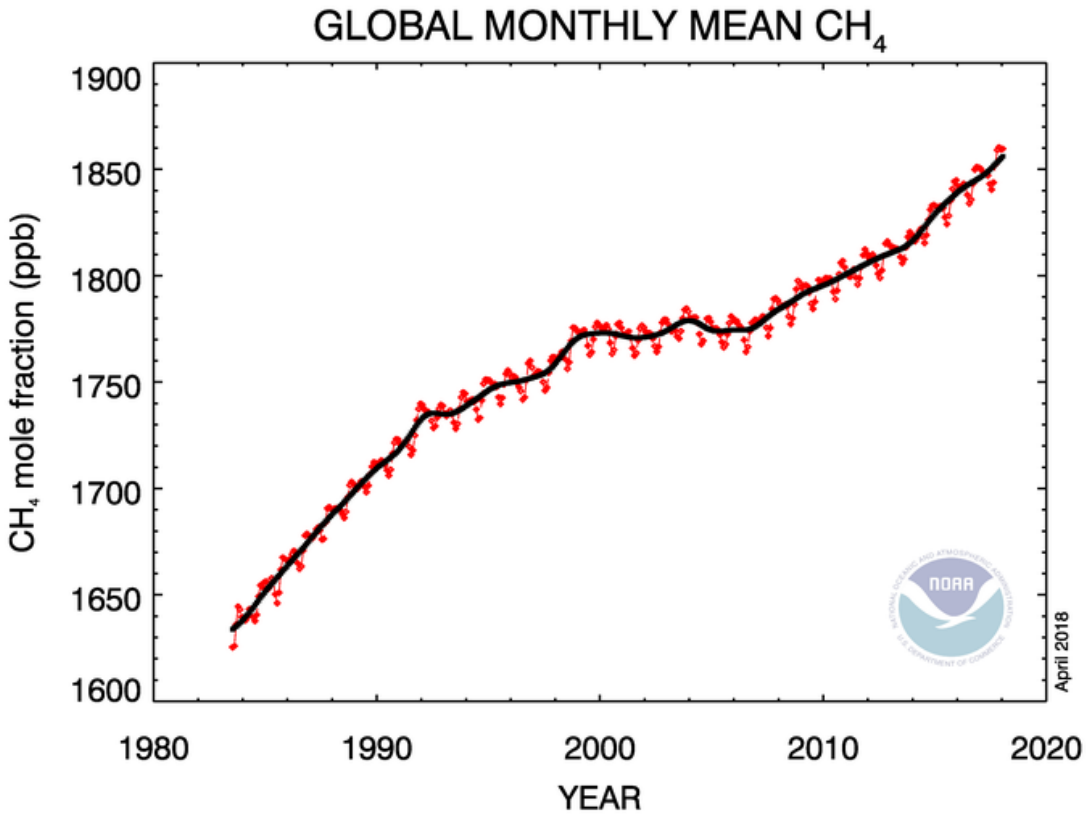
- Introduction
- Need: Improvement of Greenhouse Gases (GHGs) emission inventories
- Radon Tracer Method (RTM)
- State of the Art
- Conclusions and further steps

Introduction:
Climate Change is a matter of fact

Intergovernmental Panel on Climate Change (IPCC, 2013)

Introduction: Greenhouse gases increase is the cause





Introduction:
Reduction strategies have to be applied

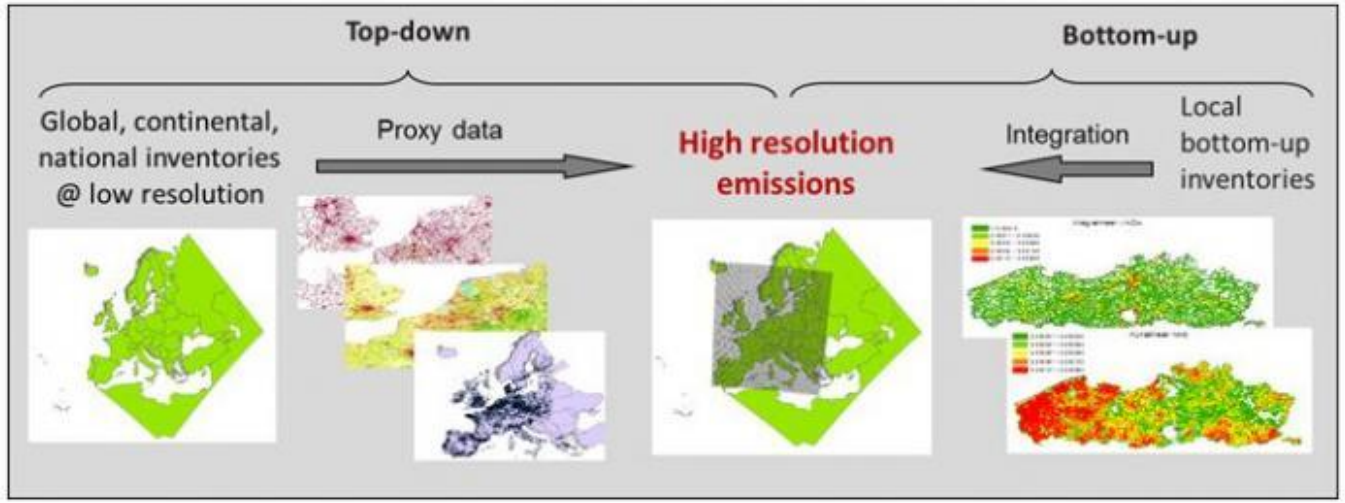
GHGs emissions, due to natural as well as anthropogenic sources, are currently estimated and reported by each national agency to the United Nations Framework Convention on Climate Change (UNFCCC).

Introduction:
Methods used to estimate GHGs emissions

Top-Down method
(based on atmospheric observations and inverse modelling)

Bottom-Up method
(based on fuel consumption and anthropogenic activity data)

- Low spatial resolution
- High uncertainties depending on atmospheric transport models setting
- No local information



- No direct measurements
- Only local information
- unknown uncertainties

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

^{222}Rn Tracer Method: Why may radon help us?

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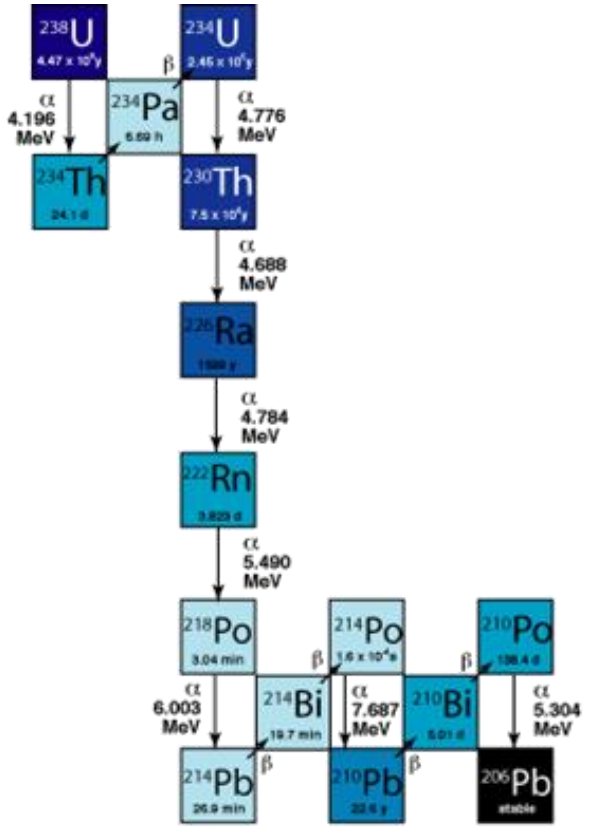
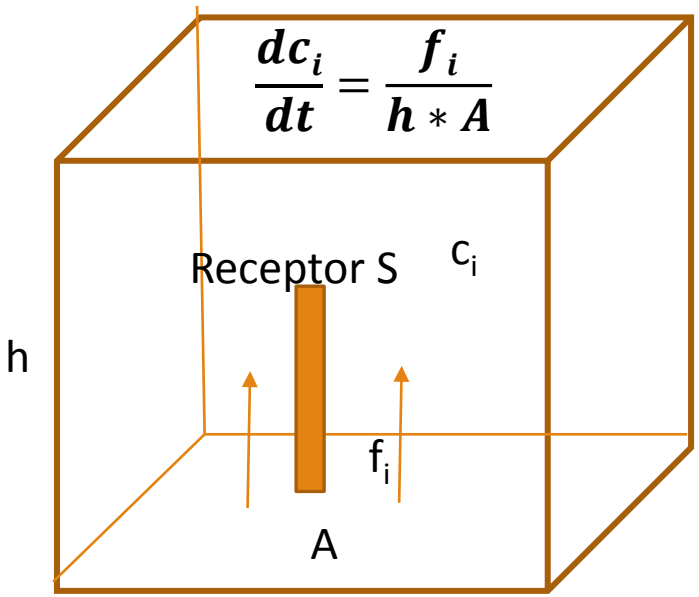
- radioactive measurement techniques are available to observe its atmospheric behavior
- its only sink into the atmosphere is due to its decay ($T_{1/2}$ = 3.8 d)
- Its source is only due to geophysics processes (^{226}Ra decay in grain soil/no need of monitored release)
- over the ocean its exhalation is taken as zero

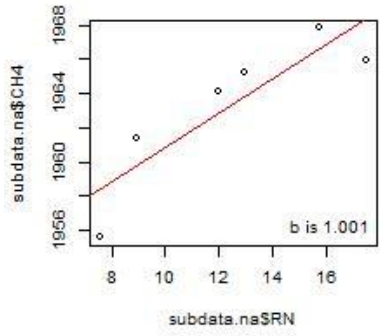


^{222}Rn Tracer Method

An independent method to estimate GHGs fluxes is the Radon Tracer Method (RTM) which allows experimentally estimating GHGs fluxes (Levin et al., 1999) and thus improving Bottom-Up GHGs inventories.

Atmospheric box model at night (D=0) for each gas





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

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

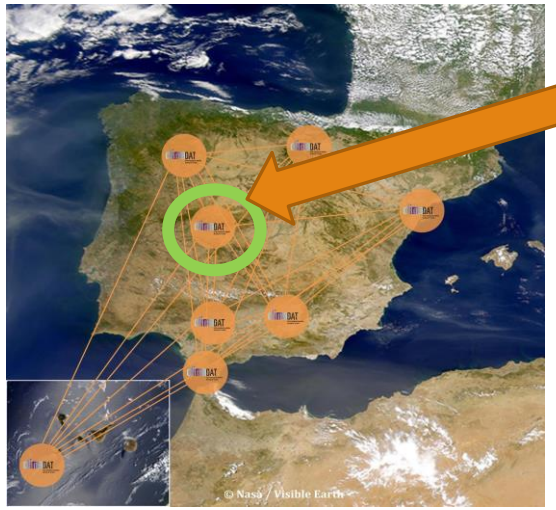
Knowing the flux of the tracer will allow us to calculate the flux of the GHG

^{222}Rn Tracer Method

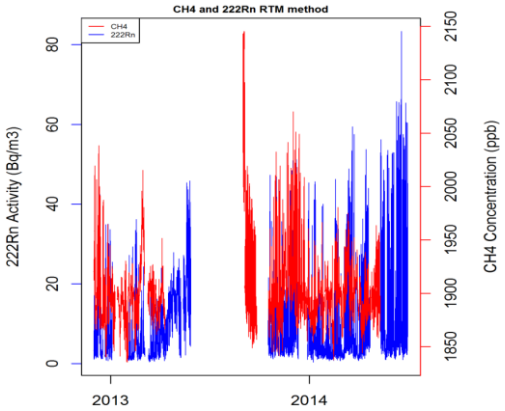
An **effective flux** is estimated for Rn-222 and CH4 considering mass origin

For each night over the time series (2013-2015)

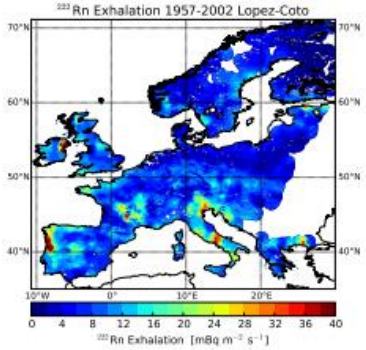
Gredos and Iruelas (GIC3) station within the Spanish CLIMADAT Network



Grossi et al., 2018

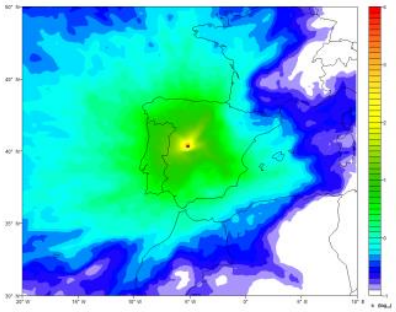


Rn-222 emission inventory
Lopez-Coto et al., 2013



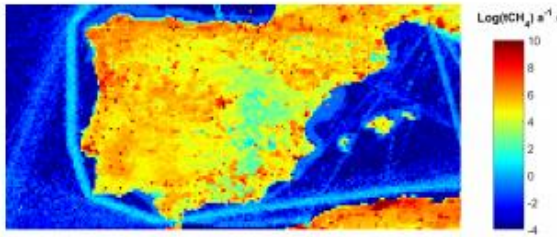
x

Foot-print of mass origin
ATM:Fluxpartv9.2



= ^{222}Rn effectiveFlux (used in the RTM to obtain **FR_CH4**)

CH4 emission inventory EDGAR, 2010



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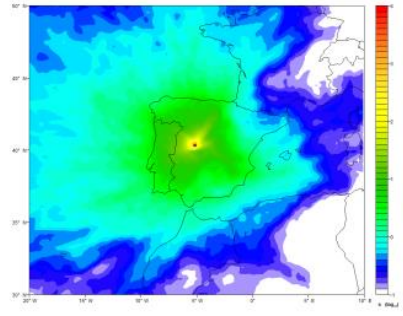
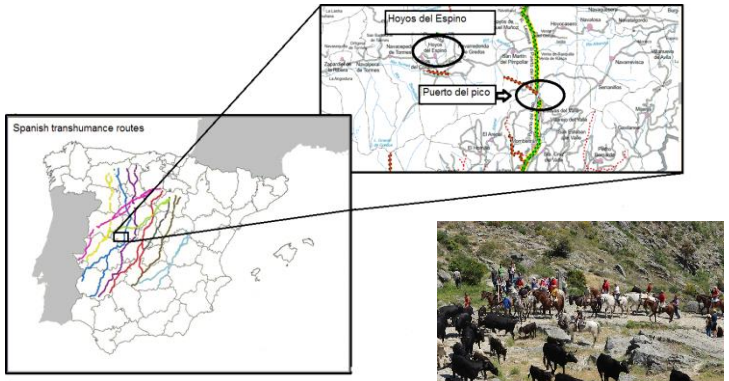
ATM:Fluxpartv9.2

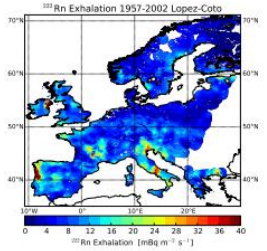


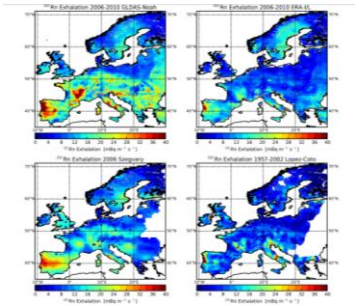
= CH_4 effectiveFlux (**FE_CH_4**)

^{222}Rn Tracer Method

Comparison between FR_CH_4 and FE_CH_4

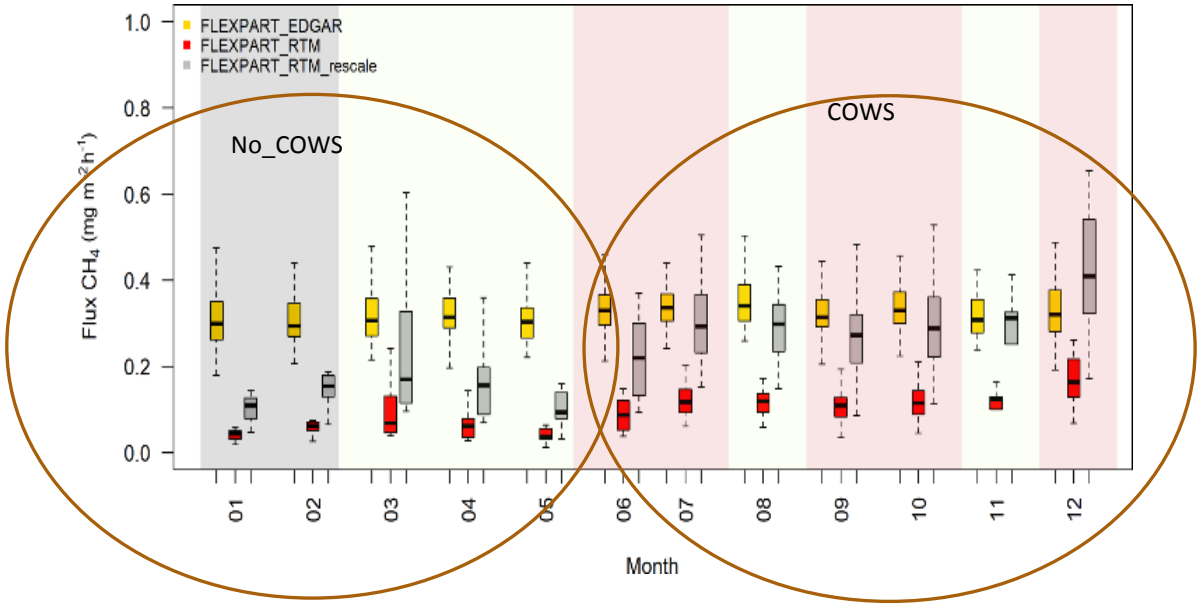






Karsten et al., 2015

Grossi et al., 2018



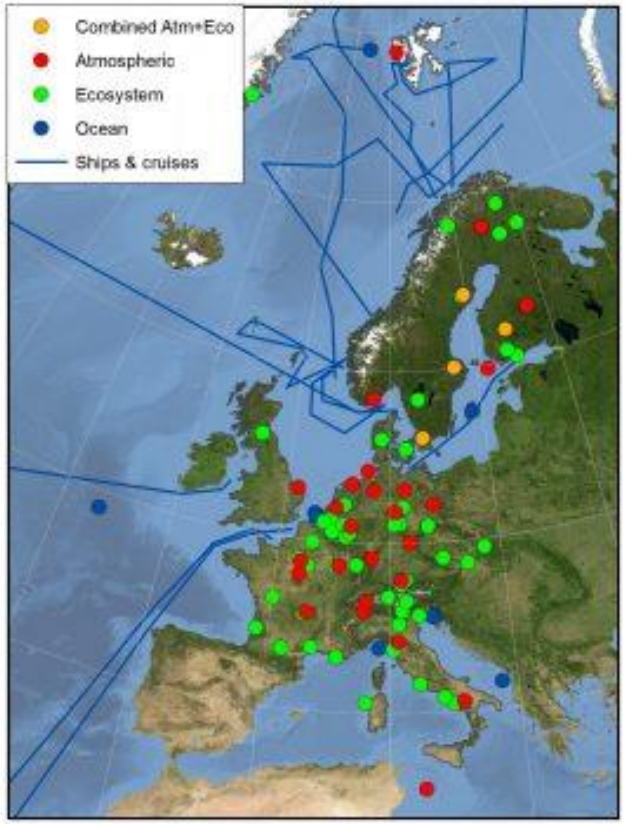
1) The **influence of the cows** can be detected by the RTM **but not by bottom-up** method using Edgar inventory.
2) The **importance of using validated ^{222}Rn flux models** and/or inventories is also shown in the plot.

^{222}Rn Tracer Method

In order to improve the RTM applications we will need:

1. High quality and harmonized ^{222}Rn concentrations observations with high spatial resolution
2. Reliable ^{222}Rn flux inventories with high spatial and temporal resolution
3. Standardization of the RTM method

State of the Art: ^{222}Rn concentrations observations



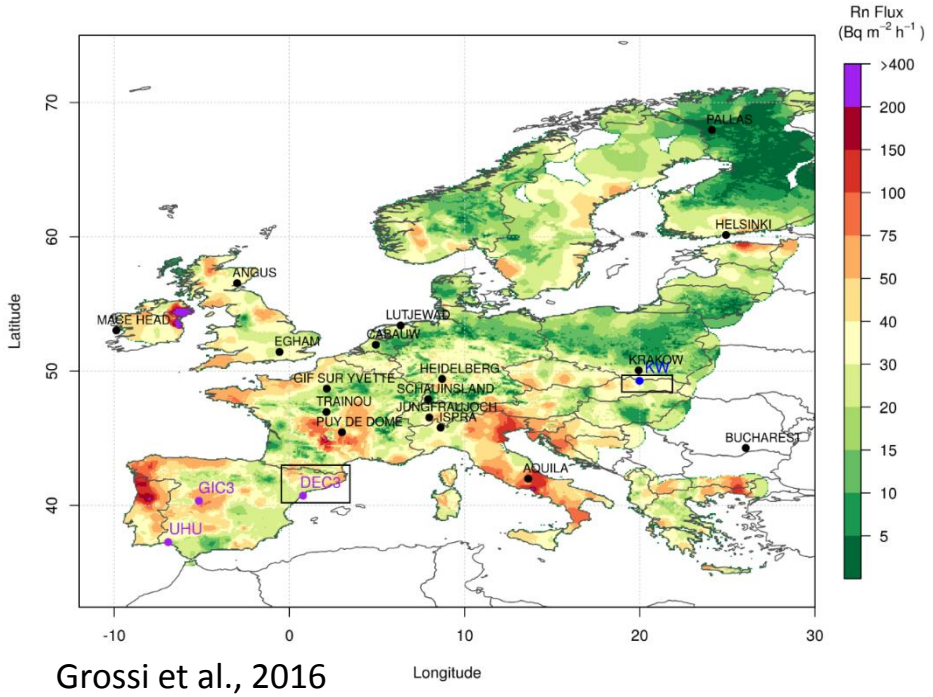
ICOS-RI

Is a pan-European Research Infrastructure

Integrated Carbon Observation System

ICOS-RI is an organization of twelve member countries and over 100 greenhouse gases measuring stations aimed at quantifying and understanding the greenhouse gas balance of the Europe and neighboring regions.

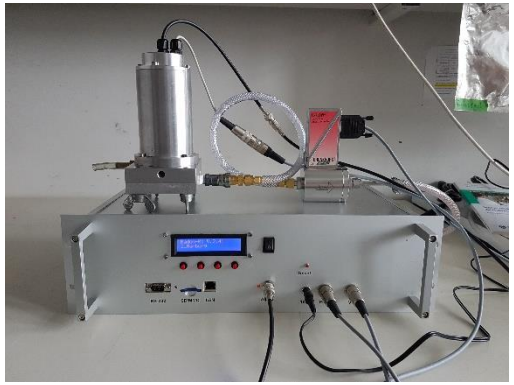
Atmospheric 222Rn stations in Europe



Grossi et al., 2016

State of the Art: ^{222}Rn concentrations observations

HEIDELBERG MONITOR
(Levin et al., 2002)



1-filter method
- portable
- ^{214}Po and need to assume an equlibrium factor between ^{214}Po/^{222}Rn

ANSTO MONITOR
(Zahorowski et al., 2004)

1500 L dual flow loop, two filter radon detector

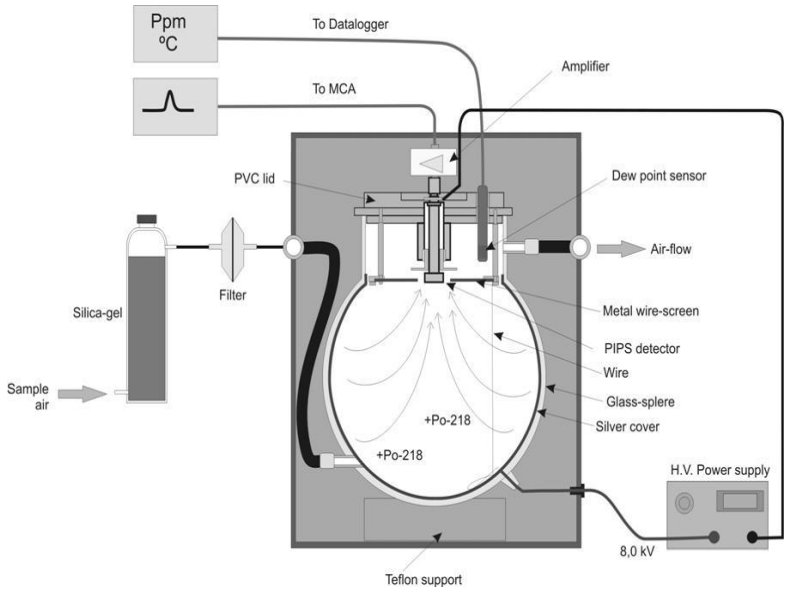
2-filters
-Large volume
-^{222}Rn decay products with large error associated for low concentrations

ARMON
(Grossi et al., 2012)

- Electrostatic collection
- Portable (20 L)
- ^{218}Po and ^{216}Po from ^{222}Rn decay only in the detection volume

State of the Art: ^{222}Rn concentrations observations

The Atmospheric Radon MONitor (ARMON) allows a spectrum resolution of ^{218}Po and ^{216}Po directly collected on PIPs detector after the ^{222}Rn and ^{220}Rn decay in the detection volume.



(ARMON scheme, Grossi et al., 2012)

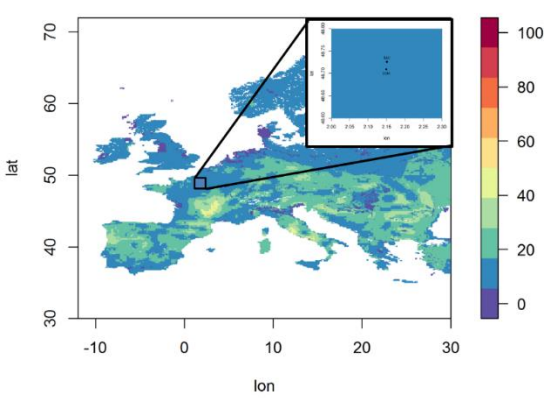
MDC – 100-150 mBq m^{-3}

Each ARMON device is calibrated at the INTE-UPC radon chamber.

Our laboratory was previously traceable to the German National Metrology Institute Physikalish-Technische Bundesanstalt (PTB) and now to the Swedish radiation safety authority.

We transfer the quantity radon activity concentration from an approved laboratory using a standard transfer instrument (Atmos 12 DPX).

State of the Art: ^{222}Rn concentrations observations



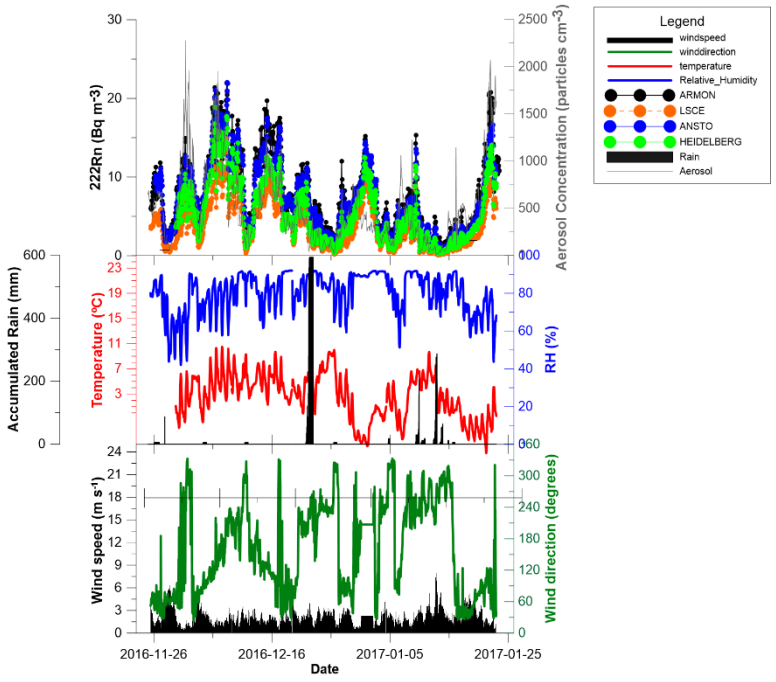
Grossi et al., in preparation

SAC
(Saclay)
100m

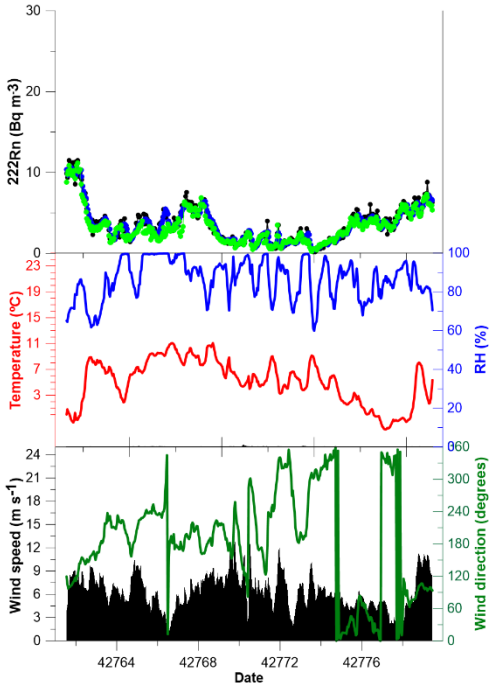
ODM (Orme des Merisiers)
5m

Monitor	Method	Alpha Spectrum	Remote Control	Dry air sample	Limits and/or corrections due to height of Inlet	Portability	References
Australian **N**uclear **S**cience and **T**echnology **O**rganisation (**ANSTO**)	**Dual-flow-loop two-filter ^{222}Rn**	**No**	**Yes**	**No**	**No**	**No**	**Chambers et al., 2011; Chambers et al., 2014; Schmithüsen et al., 2017**
Heidelberg **R**adon **M**onitor **(HRM)**	**Single-filter ^{222}Rn progeny**	**Yes**	**Yes**	**No**	**Yes**	**Yes**	**Levin et al., 2002; Levin et al., 2017; Schmithüsen et al., 2017**
LSCE Monitor **(LSCE)**	**Single-filter ^{222}Rn progeny**		**Yes**	**No**	**Yes**	**No**	**Schmithüsen et al., 2017**
Atmospheric **R**adon **MON**itor **(ARMON)**	**Eletrostatic deposition ^{222}Rn**	**Yes**	**Yes**	**Yes**	**No**	**Yes**	**Vargas et al., 2004; Grossi et al., 2012; Vargas et al., 2015; Grossi et al., 2016**

State of the Art: ^{222}Rn concentrations observations

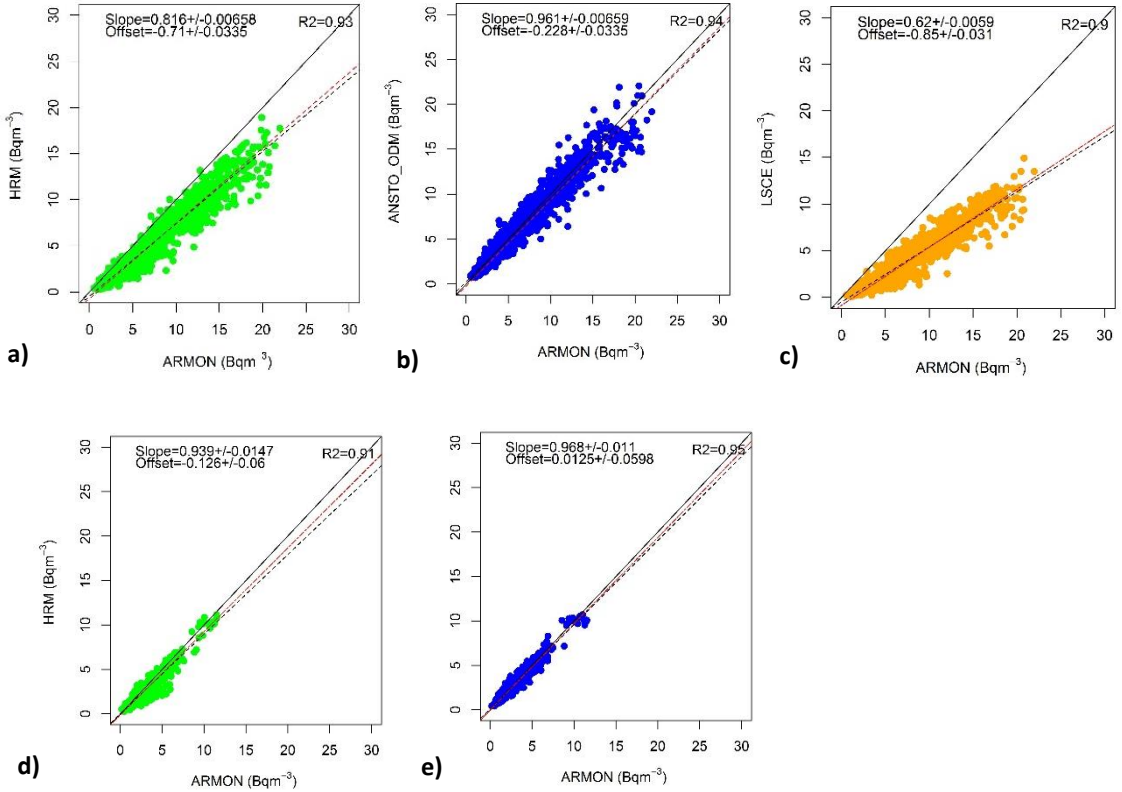


ODM 5 m agl



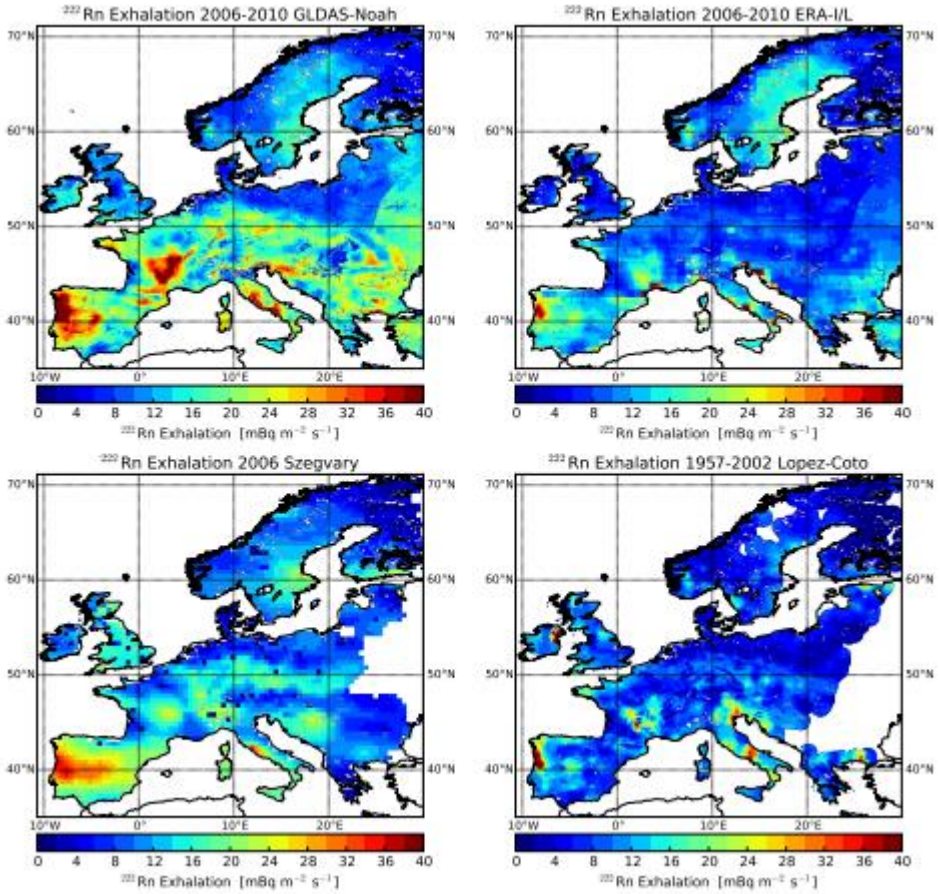
SAC 100 m agl

Grossi et al., in preparation



Arithmetic (dashed black line) and geometric (red line) linear regressions between each monitor and the ARMON at ODM (a,b,c) and at SAC (d,e).

State of the Art: ^{222}Rn flux



Available ^{222}Rn flux inventories need to be validated at European scale using experimental ^{222}Rn flux data

So far no standardised ^{222}Rn flux measurements techniques are used

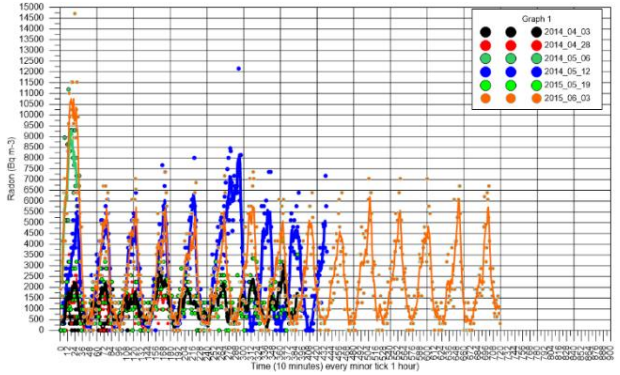
Karstens et al., 2015

State of the Art: ^{222}Rn flux

Design and characterization of continuous ^{222}Rn flux monitor

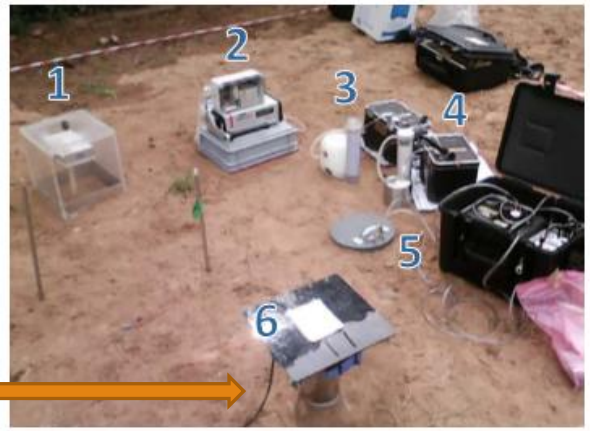
Continuous radon flux monitor

Measurements in Phosphogypsum with different humidity carried out in lab.



Intercomparison campaigns of radon flux monitors and/or techniques

Code	Research Group	Detector
1	Universidad de Cantabria (UC)	Radon Scout
2	Universitat Politecnica de Catalunya (UPC)	AplhaGUARD
3	Universitat Autonoma de Barcelona (UAB)	RAD 7
4	UAB	RAD 7
5	Geomnia	RAD 7
6	**UPC**	**Countinuous flux monitor**



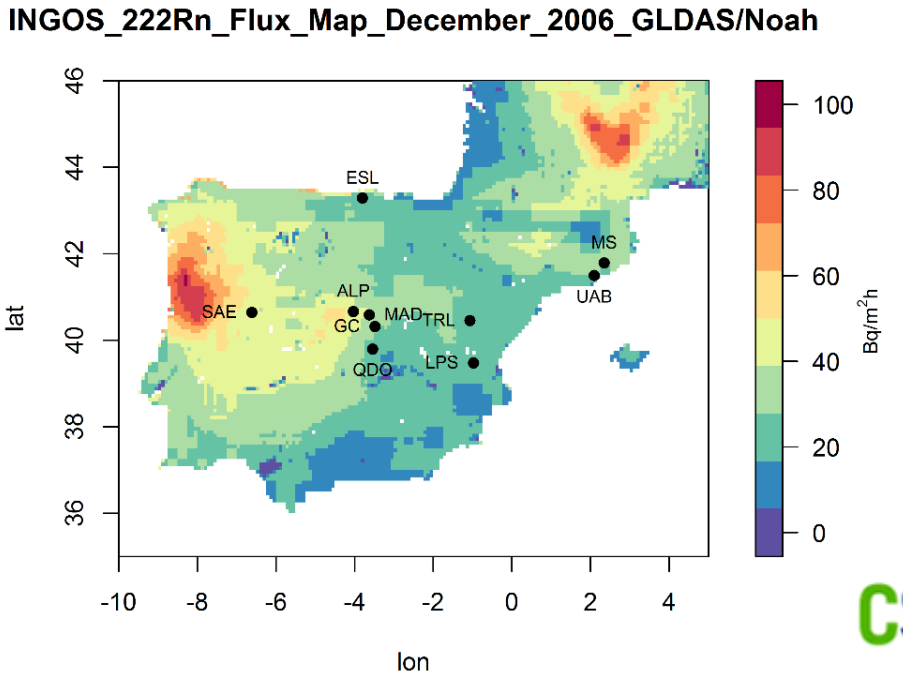
Moreno et al., in preparation

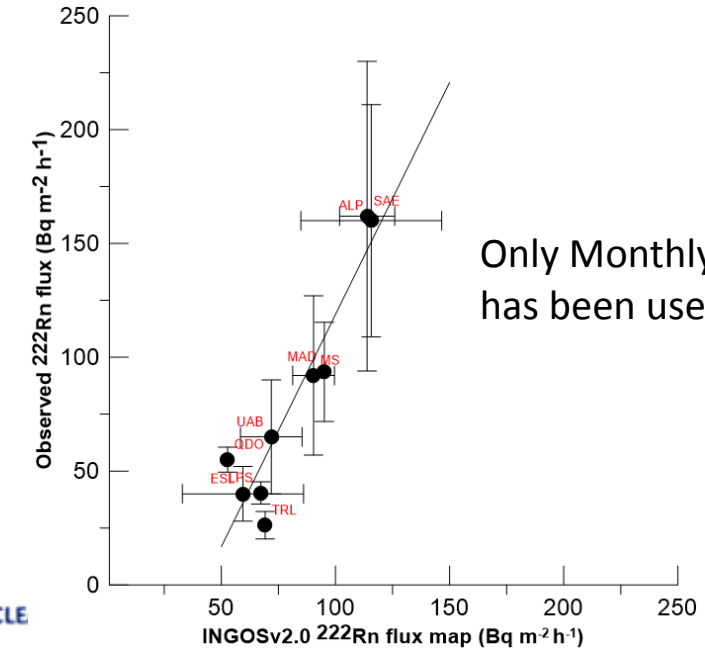
Universitat Autònoma de Barcelona

Universidad de Huelva

State of the Art: ^{222}Rn flux

Preliminary study on INGOSv2.0 map (Karstens et al., 2015) validation over Spain

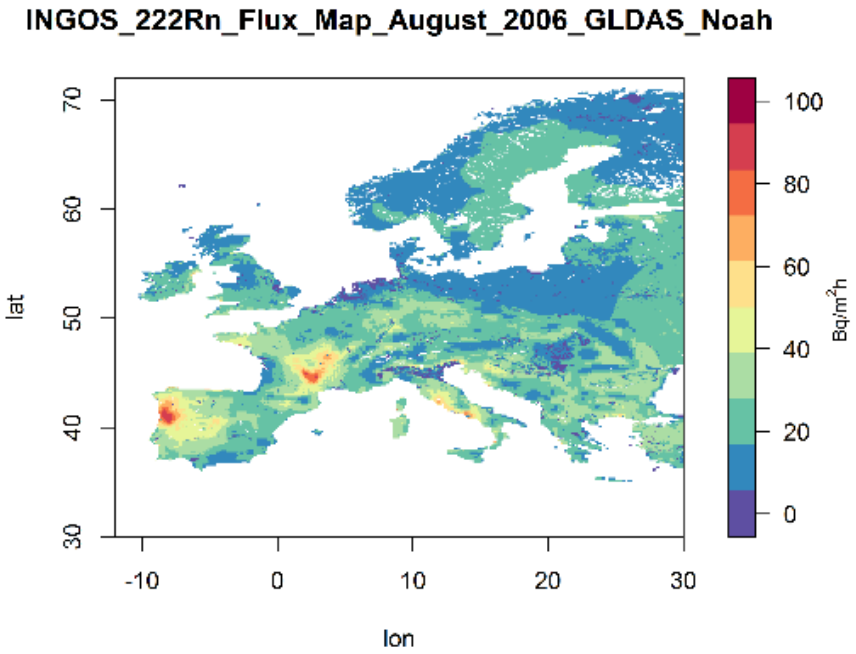


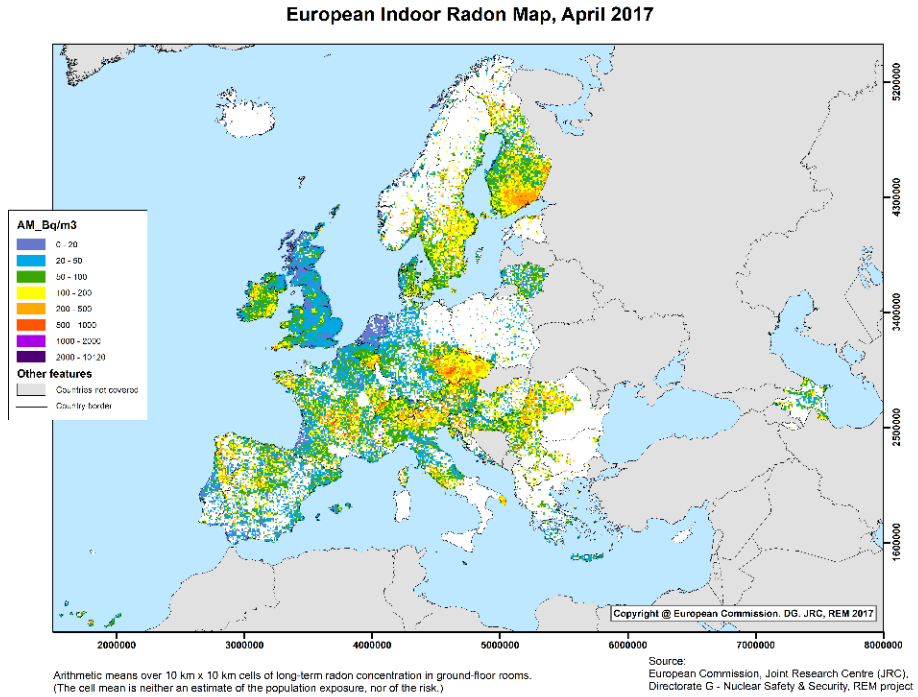


Only Monthly climatology has been used

State of the Art: ^{222}Rn flux

Validated radon flux model and also **radon atmospheric observations** could help in the assessment of **radon areas for radiation protection goals**





https://doi.pangaea.de/10.1594/PANGAEA.854715?format=html#mcol0_ds12803808

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

Conclusions and…

-The utility of the radon tracer method in helping to the improvement of greenhouse gases emission inventories implies the importance of standardize this method

- Atmospheric ^{222}Rn measurements are being carried out within European networks (ClimaDat, ICOS), because of the large radon tracer applications and it will be important to have as many stations as possible

- There is the need to harmonize all this data and compare the response of different atmospheric radon monitors

- ^{222}Rn flux inventories currently available have to be validated and this leads to the need of continuous ^{222}Rn flux data in Europe

….Further Steps

EMPIR

Potential Research Topic (Annette Röttger**)**

Implementation of radon metrology for the analysis for the atmospheric budget of greenhouse gases and radiation protection in the environment

MAIN AIM: to provide sound and novel metrological tools and relevant data for stakeholders, like ICOS, EC-JRC(EURDEP, EARN), ALMERA, IAEA, WHO , related with emission reduction strategies of greenhouse gases and radioprotection of general public.

Objectives:

- To establish metrological traceability of outdoor low-level radon activity concentration measurements as input for atmospheric networks and radiation protection networks.
- To support metrological infrastructure for radon flux measurements as input for identification of radon prone areas and for application of radon tracer method. To harmonise different radon flux measurement methods by intercomparison campaigns.
- To validate existing radon flux inventories and models using experimental radon activity concentration data and radon flux data. Including dosimetric and spectrometric data from the radiological early warning networks in Europe.
- To develop standard protocols for radon tracer method to retrieve GHGs fluxes at atmospheric climate gases monitoring stations.
- To provide dynamic atmospheric radon concentrations and radon flux maps for climate change research and radiation protection according COUNCIL DIRECTIVE 2013/59/EURATOM.

Thanks for your attention