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STATE OF THE ART OF RADON FLUX MEASUREMENTS

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Contents

- Radon flux from the soil (WHO)
- Utility of reliable radon flux measurements (WHY)
- Accumulation method (HOW)
- Challenge of high quality measurements (BUT)
- Current radon flux techniques (NOW)
- Contribution of traceRadon project (SO)

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WHO

Radon is released to the atmosphere after being transported by diffusion and advection from pore spaces

Radon flux/exhalation from the soil = Activity concentration of radon interchanged between the soil surface and the lower atmosphere for unit of area and of time (S.I. unit: Bq m^{-2} s^{-1})

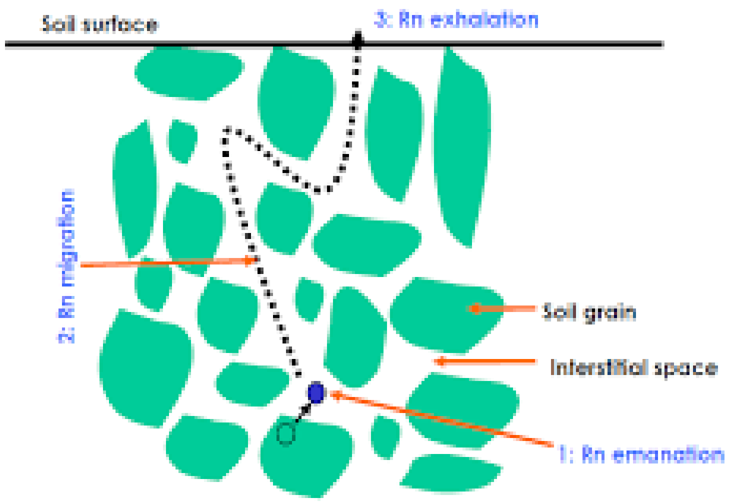

Figure 2.6: Radon emanation, migration and exhalation

Source: https://inis.iaea.org/collection/NCLCollectionStore/_Public/43/106/43106942.pdf

Fick's Law (Molecular Diffusion)

$$\vec{J}_d = -D \cdot \nabla C$$

Other physical conditions that may lead to a diffusive flux , in particular temperature gradients, pressure gradients and external forces are neglected in this expression. They are usually unimportant relative to the concentration gradient into the soil

Darcy's Law (Forced Convection)

$$J_a = vC \qquad \mathrm{v} = \frac{-K \cdot \nabla p}{\eta}.$$

$$\vec{J} = -D\vec{\nabla}C + \vec{v}C \quad \Rightarrow \quad \vec{\nabla} \cdot \vec{J} - \lambda C + A_0 = \frac{\partial C}{\partial t}$$

Nazaroff and Nero, 1988

WHY

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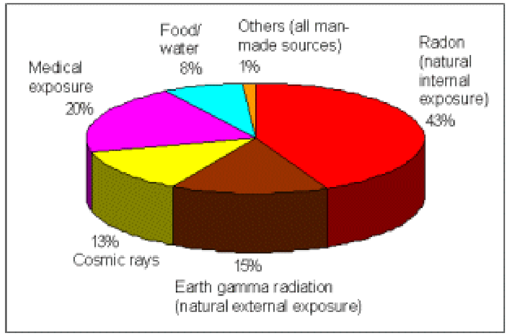
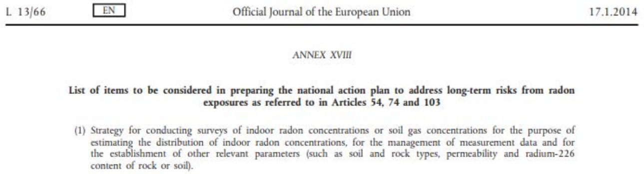
I.



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

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.’

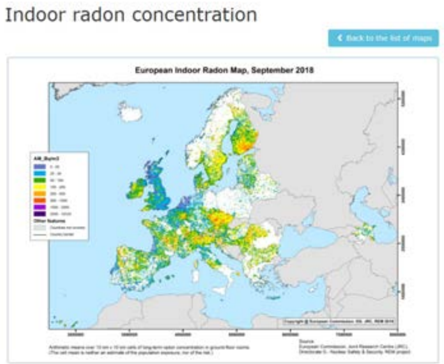


L 13/66 EN Official Journal of the European Union 17.1.2014

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

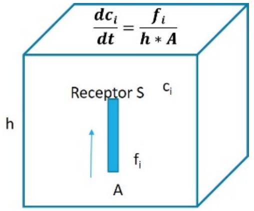
(1) Strategy for conducting surveys of indoor radon concentrations or soil gas concentrations for the purpose of estimating the distribution of indoor radon concentrations, for the management of measurement data and for the establishment of other relevant parameters (such as soil and rock types, permeability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



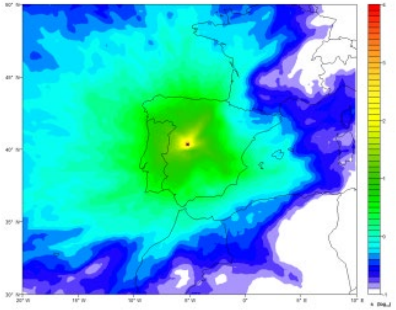
https://remon.jrc.ec.europa.eu/About/Atlas-of-Natural-Radiation/Digital-Atlas/Indoor-radon-AM/Indoor-radon-concentration

^{222}**Rn as tracer for, among others, improvement of atmospheric transport models and indirect evaluation of greenhouse gases fluxes**

II.

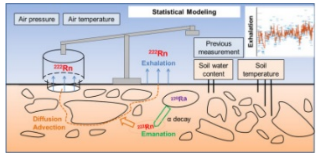


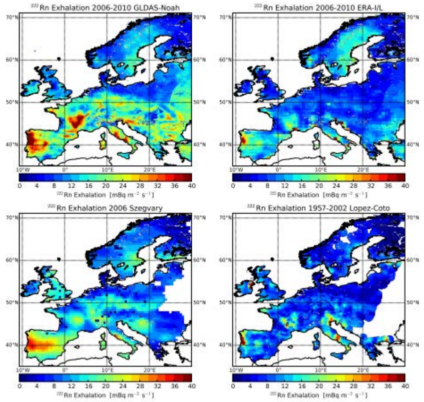
$$f_i = \frac{\frac{dc_i}{dt}}{\frac{dc_j}{dt}} \cdot f_j$$



Grossi et al., 2020

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





Karstens et al., 2015

HOW

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^{222}Rn fluxes can be measured using both integrated and continuous monitors coupled with chambers to use the Accumulation Method: ' The radon exhaled from the soil surface is accumulated during a time period (T) within a known volume (Moraswka, 1989; De Martino and Sabbarese, 1997; Grossi et al., 2011). The temporal variation of the radon activity concentration in the chamber is expressed by the equation:

$$\frac{dC(t)}{dt} = \frac{E_{Rn}}{V_u} - \lambda^0 C(t)$$

With the initial condition tan the radon activity concentration at the beginning of the measurements is C(t=0)=0.

E_{Rn} is the exhalation velocity of the radon escaping from the soil (Bq s^{-1}), V_u is the available chamber volume (m^3) and the constant $\lambda^0 = \lambda + \lambda_l$ is given by the sum of the radon decay constant and a factor taking into account possible leakages (is it constant? Which are the cause of its magnitude and changes?). The solution:

$$C(t) = \frac{E_{Rn}}{\lambda^0 V_u}(1 - e^{-\lambda^0 t})$$

$$E_{Rn} = J \cdot A$$

Under ideal 'hermetic' conditions $\lambda^0 = \lambda$

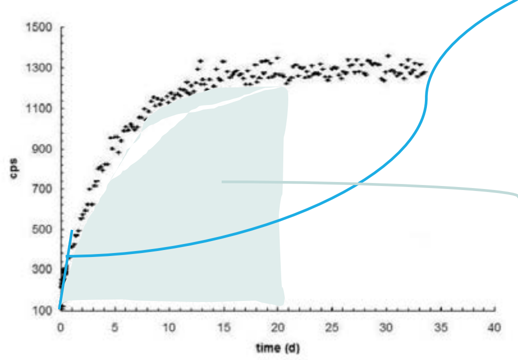


Figure 2.1: ^{222}Rn accumulation evolution in time inside a closed volume.

Grossi, 2012

Linear Approximation

$$C(t) = \frac{E_{Rn}}{V_u} t = \frac{FA}{V_u} t$$

Integrated measurement

$$C(Rn)Av = \frac{1}{T}\int_0^T C(t)dt = \frac{FxA}{\lambda^0 V_u}\left(1 - \left(\frac{1 - e^{-\lambda^0 t}}{\lambda^0 T}\right)\right)$$

A is the Surface area of the accumulation chamber

Grossi, 2011

BUT

Many Laboratory and in field studies on continuous measurements of radon fluxes have been carried out indicating **an influence of environmental parameters within the accumulation chamber and the exterior, setting conditions (insertion depth) on the accumulation method results both for total curve or linear approximations**.

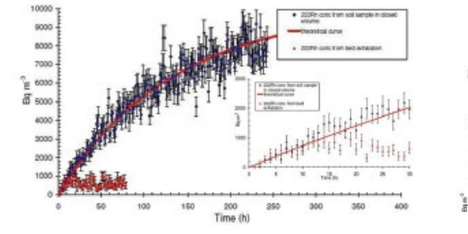


Figure 4.6: ^{222}Rn concentration exhaling from a phosphogypsum soil sample and from a phosphogypsum exhalation bed measured hourly by a DOSEman monitor.

Grossi, 2012

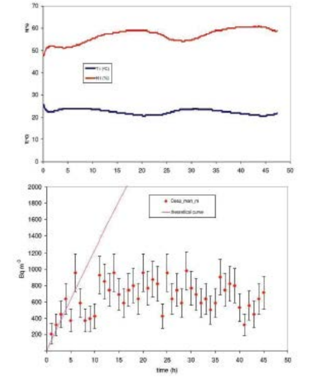
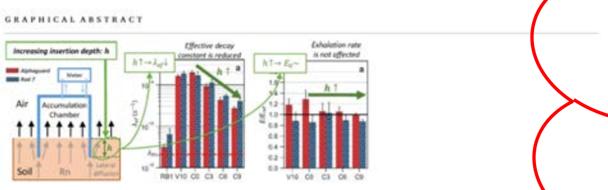


Figure 4.7: ^{222}Rn concentration exhaling from a phosphogypsum exhalation bed measured by a DOSEman monitor. Environmental parameters inside the accumulation volume were also measured by EasyLog USB sensor.

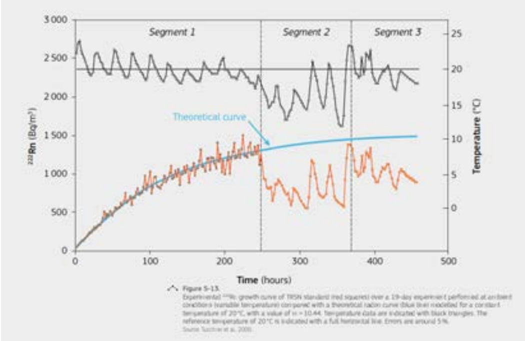


Influence of the accumulation chamber insertion depth to measure surface radon exhalation rates

I. Gutiérrez-Álvarez, J.L. Guerrero, J.E. Martín, J.A. Adame, J.P. Bolívar

GRAPHICAL ABSTRACT

There is a strong need of correctly quantifying all these influences to perform a protocol and guide to continuous radon flux measurements



European Atlas of Natural radiation Chapter 5

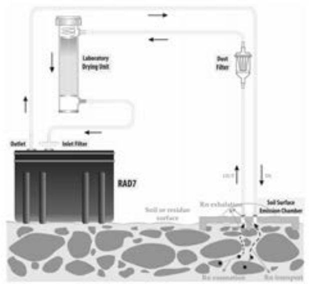


Fig. 2. A closed-loop radon exhalation rate measurement scheme using RAD7.

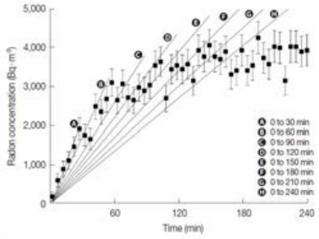


Fig. 5. Linear fitting of data 1-A.

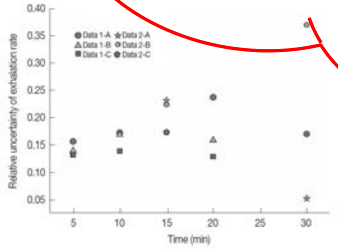


Fig. 7. Relative uncertainties compare with time interval at 2 hours measurement.

Seo et al., 2018

trace Radon

NOW

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EURAMET

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

Source	Device Name	Creator	Volume (m3)	Emanation area	Automatic opening	Shape	Used with
1	**AutoFlux**	**Designed by Sylvester Werczynski and built by Ot Sisoutham in 2018/19**	**0.019**	**0.13**	**Yes**	**Drum**	**AlphaGUARD**
2	Emanometer diffusion chamber	ANSTO	0,018	0,26	No	Shallow conical	AlphaGUARD2 x 1L Lucas cells (separated by 6 min flow path)
3	**8100-401 Chamber Control Kit**	**LI-COR**	**0.004**	**0.03**	**Yes**	**Hemisphere**	**-**
4	Radon flux measurement system	-	0.04	0.21	Yes	Drum	AlphaGUARD
5	Flux chambers	-	0.002, 0.018 and 0.35	0.02, 0.07 and 2.3	No	Circular, Circular, Square	RAD7
6	The accumulation chamber	Helmholtz Zentrum München (HMGU)	0.044	0.13	Yes	Drum	-
7	UPC accumulation chamber	UPC	0.002	0,01	Only pump	Cylinder	DOSEman
8	UC accumulation chamber	UC	0.008	0.04	Only pump	Drum	AlphE

Literature Review (Report of the activity A2.1.1 of the traceRadon Project located in the NPL share point)

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SO

Long-Term Chambers

Whether you choose a single chamber or multiplexed long-term configuration, the chambers can make all the difference between dependable, reproducible data and questionable results. LI-COR chambers are designed with a perforated base and patented pressure vent to preserve the integrity of your measurement.

Opaque Long-Term Chamber

The opaque long-term chamber (8100-104) is finished in durable, highly reflective white enamel. Rotating away from the soil collar, the motorized chamber allows for autonomous measurements in the field over long periods of time minimizing disturbance to the soil surface inside the collar.

Clear Long-Term Chamber

The clear chamber (8100-104C) is designed for long-term net carbon exchange measurements. It features a durable, clear chamber top so that plants are exposed to sunlight during measurements.

https://www.licor.com/env/products/soil_flux/system

LI-8150 Multiplexer

The LI-8150 Multiplexer provides the ability to measure the gas exchange from multiple long-term chambers, providing a large sample size that allows you to determine spatial and temporal flux rates with confidence. Capable of running autonomously for months, the LI-8150 system collects a continuous dataset from up to 16 chambers to characterize spatial and temporal variation in gas exchange over a research area. By integrating additional gas analyzers like the LI-7810 $CH_4/CO_2/H_2O$ Trace Gas Analyzer, long-term and multiplexed systems also support methane fluxes while third-party gas analyzer integration provides support for fluxes of isotopologues and other gases.

Why Choose the LI-8150?

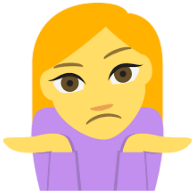
The LI-8150 Multiplexer enables you to collect a large sample size and characterize gas exchange for a study area or from treatment and control groups. A large sample size and long-term measurements help you characterize the flux much better than a single measurement. Continuous, long-term measurements help characterize diurnal and seasonal patterns.

- Supports any combination of clear and opaque chambers.
- Ideal for assessing both spatial and temporal variability of soil gas flux.

× Close

AlphaGUARD PQ2000

AlphaGUARD DF2000, D2000, D50



The UPC contacted with the Spain sale company to check the possibility of adapting this chamber to an AlphaGUARD sampling

Too expensive
The control system
May we skip its purchase?

SO

Overview of the ANSTO Autoflux system for long-term automatic radon flux monitoring

Scott Chambers, Sylvester Werczynski, Ot Sisoutham, Alan Griffiths, Alastair Williams
Isotope Tracing in Natural Systems
Contaminant Impacts (Atmosphere)

traceRadon **Workshop (A5.2.1)**

20th October 2020

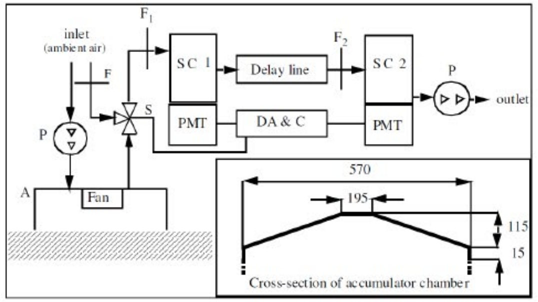
Work Package 2: Radon flux measurements

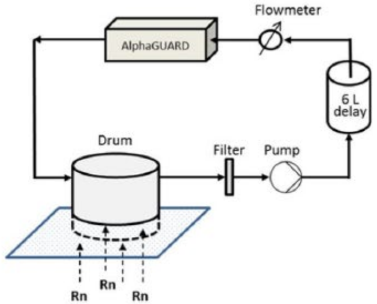
Candidate for testing and evaluation in A2.1.1

Science. Ingenuity. Sustainability.

Background

- The "autoflux" unit is a portable, automatic radon flux measuring device
- Based on a manual radon accumulator developed by Stewart Whittlestone, described in *Schery et al. (1989; JGR 94(D6), 8567-8576)*





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System characteristics

- The chamber pushes 5cm into the ground (internal lip) to reduce back diffusion
- Diameter 40 cm, active volume 0.0188 m^3, surface area 0.126 m^2
- Lid opens wide to expose surface to ambient conditions between flux measurements
- Automatic chamber operations controlled by a Campbell Scientific CR1000 logger; Radon concentrations measured using an AlphaGUARD (old or new model); Communication and data display controlled by Raspberry-Pi computer
- Minimum measurement cycle is 3-hourly (1-hour accumulation, 2-hour flushing) User can define longer measurement cycles if desired
- Chamber air is sampled in a closed loop at 0.9 L/min, and a 6 minute delay is incorporated before the AlphaGUARD to remove thoron
- The system additionally measures: soil water content and temperature (inside), external air temperature, relative humidity, absolute pressure sensor and rainfall
- The AlphaGUARD measures: air temperature, relative humidity and absolute pressure sensor of air inside the chamber as it measures radon
- The system can run unattended, continuously for many months

ANSTO

Additional information

AlphaGUARD PQ2000

AlphaGUARD DF2000, D2000, D50

- The Autoflux system can operate with either the AlphaGuard PQ2000 (old model) or the new DF2000 model
- The user needs to specify the required AG type prior, since they use different communication software (which needs to be pre-installed)
- So far the Autoflux system tests have been performed mainly in Australia, with predominantly dry conditions
- An Autoflux system operated at the Cabauw tower has experienced some problems with excessive moisture (blocking internal tubing)
- A drying system (capable of reducing ambient humidity by around 30%) has been included in the new model Autoflux systems
- While the system can run for long periods, it is prudent to make weekly maintenance visits (checking tubes for water, flow rates, removing overgrown grass in the lid, integrity of the system), so it is not suitable for very remote sites without a local support

ANSTO

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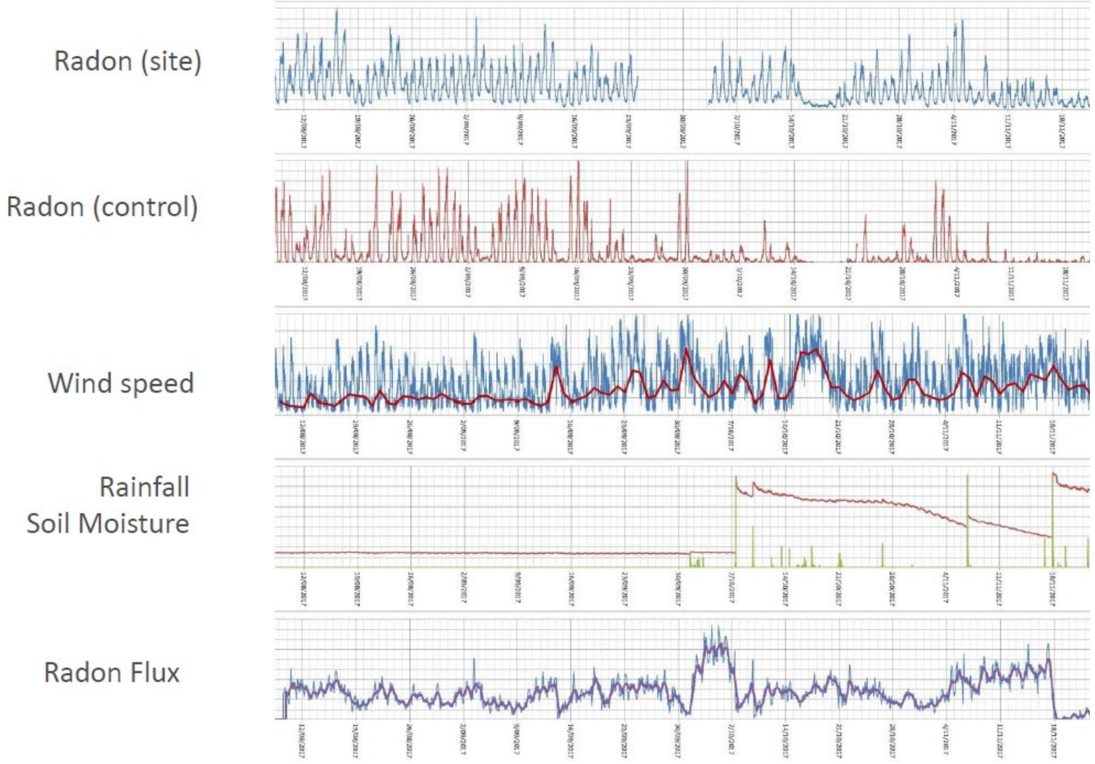
Example: Surat Basin, QLD (Australia)

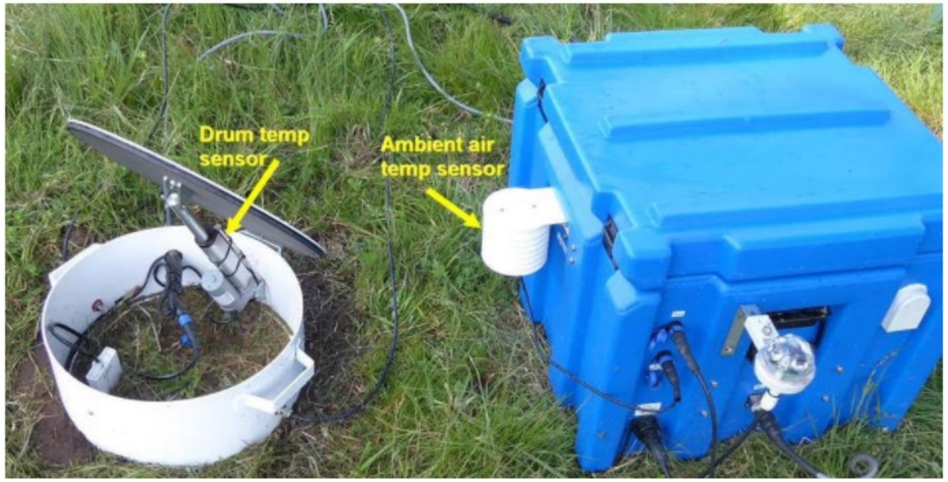


Possible reference flux monitor

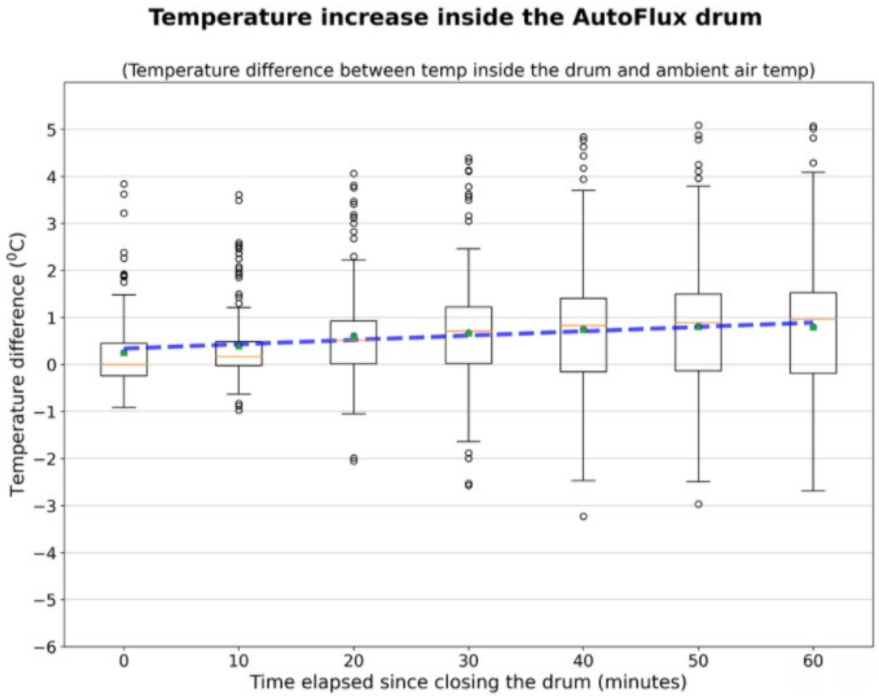
- As part of A2.1.5 UPC has offered to rent an Autoflux system for a year (for initial evaluation)
- An Autoflux system (with a white painted measurement chamber) is currently being prepared with an estimated dispatch date of November 2020
- Rental costs: AUD $7,200 per year (or 4,400 Euro per year)
- Purchase price: AUD $28,000 (17,000 Euro) (Excluding the AlphaGUARD)

White painted Autoflux drum to reduce self-heating

SO



https://chart-studio.plotly.com/~RadonFlux/788.embed



SO

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Task 2.1. to develop a continuous radon flux monitor for use as a transfer standard to calibrate existing radon flux monitors under laboratory conditions. Furthermore, to develop a radon exhalation reference system “exhalation bed”, (as the starting point of the traceability chain to the SI) and use this to calibrate the continuous radon flux monitor with an hourly uncertainty of 10 % for k=1.

Task 2.2. to use the radon flux monitoring capability developed in Task 2.1 and to harmonise existing radon flux methods and monitors under field conditions using intercomparison campaigns.

Task 2.3. to perform intense radon flux measurement campaigns in the field at AMNS or RMS using the calibrated continuous radon flux monitors from Tasks 2.1 and 2.2.

Task 2.4. to develop a protocol for the application of the RTM to enable future retrievals of GHG fluxes at atmospheric climate gas monitoring stations and to use radon flux data for the identification of RPA.

SO

	Month	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36
	Activity	Jun-20	Jul-20	Aug-20	Sep-20	Oct-20	Nov-20	Dec-20	Jan-21	Feb-21	Mar-21	Apr-21	May-21	Jun-21	Jul-21	Aug-21	Sep-21	Oct-21	Nov-21	Dec-21	Jan-22	Feb-22	Mar-22	Apr-22	May-22	Jun-22	Jul-22	Aug-22	Sep-22	Oct-22	Nov-22	Dec-22	Jan-23	Feb-23	Mar-23	Apr-23	May-23
		WP2																																			
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SUJCHBO	A2.1.1																																				
UPC	A2.1.2																																				
UC	A2.1.3																																				
UC	A2.1.4																																				
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ENEA	A2.3.4																																				
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UVSQ	A2.4.2																																				
UVSQ	A2.4.3																																				

Selection of a site to carry out the RTM method evaluation

From Activity 2.4.1 description, the ICOS AMNS will be chosen by UVSQ, in collaboration with LUND, INESC TEC, NPL, JRC and UPC according to their characteristics: i.e. the ability to make vertical radon gradient measurements and the capability for measuring Greenhouse Gases (GHGs) mixing ratio measurements and thus estimate GHG fluxes.

Within ICOS, a dozen sites are currently measuring atmospheric radon concentration activity using different monitors. However, data are transferred for only 6 sites to the ICOS database, and of these, only three are tall tower sites: Saclay, Trainou and Observatoire Pérenne de l'Environnement, all located in France. Tall tower sites allow to use gradient techniques, as well as the RTM, to evaluate fluxes (Yver et al., 2010).

The three sites measure greenhouse gases and meteorological parameters at different levels. For logistical reasons, we propose to perform the RTM evaluation first at Saclay, where radon flux monitors and other instruments can be installed more easily as it has been already done in the past.

Saclay (SAC) is located 30km south-west of Paris, 42.7217°N, 2.142°E, 160m asl. A 3-month intercomparison of radon monitors was previously carried out at this site in 2016 (Grossi et al., 2020).

Routine radon monitoring at SAC is conducted from 2m and 100m agl, while greenhouse gases are sampled at 15, 60 and 100m agl. Meteorological parameters are available at -0.1, 0.1, 1.5S, 10S*, 40S, 60S*, 80S*m agl (S denotes humidity, wind speed and direction measurements, temperature is measured at all indicated levels).

The above data are available in the ICOS database, since 2015 for GHG and meteorological data, since the end of 2016 for 100m radon, and since January 2019 for 2m radon.

The RTM has previously been applied at the nearby site of Gif-sur-Yvette, 2km west of SAC (Belviso et al., 2013, Belviso et al., 2020). Yver et al., 2009 summarized the radon flux estimates before this date that were ranging from 42 to 66 +/- 22 Bq m^{-2} h^{-1} with an average of 52 Bq m^{-2} h^{-1}. From 2006 to 2009, additional measurements were done and used to assess a new radon map (Karstens et al., 2015). The values found for SAC were varying between 18 to 54 Bq m^{-2} h^{-1} for observations and models.

Footprints are available on the ICOS Carbon Portal (https://stilt.icos-cp.eu/viewer/) since 2014.

REVIEW continual RADON MONITORS 4

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