19ENV01 traceRadon

Deliverable D4

Good Practice Guide including a standard protocol for the measurement of radon flux and atmospheric activity concentration for application in the radon tracer method (RTM) for greenhouse gas (GHG) flux estimates and for its application to derive data for Radon Priority Areas (RPA)

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Glossary

AMNS	Atmospheric Monitoring Network stations
ANSTO	Australian Nuclear Science and Technology Organisation
ATM	Atmospheric Transport Model
CP	Carbon Portal
EB	Exhalation Bed
EMPIR	European Metrology Programme for Innovation and Research
ECMWF	European Centre for Medium-Range Weather Forecast
GHG	Greenhouse Gas
ICOS	Integrated Carbon Observation System
IFS	Integrated Forecasting System
INESC TEC	Instituto de Engenharia de Sistemas e Computadores, Tecnologia e Ciência
JRC	Joint Research Center
LUND	Lunds Universitet
NPL	National Physical Laboratory
NRT	Near Real Time
RPA	Radon Priority Areas
RRI	Radon Reference Instrument
RTM	Radon Tracer Method
SI	Système international d'unités
STP	Standard temperature and pressure conditions
TS	Transfer Standard
UoB	University of Bristol
UPC	Universitat Politècnica de Catalunya
UVSQ	Université de Versailles Saint-Quentin-en-Yvelines
WP	Work package
WRF	Weather Research and Forecasting model

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1. Introduction

Here we present the deliverable D4, of the Work Package 2 (WP2) of the EMPIR joint research project 19ENV01 traceRadon. It is titled "Good Practice Guide including a standard protocol for the measurement of radon flux and atmospheric activity concentration for application in the radon tracer method (RTM) for greenhouse gas (GHG) flux estimates and for its application to derive data for Radon Priority Areas (RPA)".

It has been shown that accurate knowledge of environmental atmospheric radon activity concentrations and radon fluxes is key for improving GHG flux estimates for climate change research and radiological protection (Röttger et al., 2021, Levin etal., 2021). Climate related Atmospheric Monitoring Network Stations (AMNS) were established for measurements of GHGs in order to improve and to support the development of Atmospheric Transport Models (ATM) and to better understand GHG levels in the atmosphere by using long-term observations. Since atmospheric radon measurements are carried out at many such AMNS, this project will support European AMNS in performing atmospheric radon and radon flux measurements for a variety of radon tracer applications. The project will do this through its development of new low activity ^{222}Rn emanation sources, a reference instrument for atmospheric radon measurements and a traceability chain for low radon activity concentration measurements (from 1 Bq·m^{-3} to 100 Bq·m^{-3}). These efforts will support the comparability of real time atmospheric radon activity concentration data between different measurement sites and, over time, provide these radon measurements with the required traceability to the SI. In turn, this will lead to more reliable data for policy and decision makers to use in the combat against climate change.

In addition, a complete traceability chain, including a reference exhalation bed and a transfer standard system for continuous radon flux measurements, was also developed with the aim of helping in the validation of available radon flux models and inventories (Szegvary et al., 2009; Lopez-Coto et al., 2013; Karstens et al., 2015).

The overall aim of this project is the development of metrological capacity (reference monitors, transfer instruments and robust methodology) to measure low levels of radon in the environment, which can be used to determine emission reduction strategies of GHG and improve radiation protection of the general public.

The specific objectives are organized as work packages (WP) and described as follows:

- 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 and radiation protection networks. These methods include two new traceable ^{222}Rn emanation sources below 100 Bq·m^{-3}, a transfer instrument calibrated with these new sources to assure the traceability of the transfer instrument and a calibration procedure suitable to enable a traceable calibration of environmental atmospheric radon measurement systems in the field. (WP1)

- 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. To use this capability to harmonize existing radon flux instruments/methods by intercomparison campaigns. 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). (WP2)
- 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 improve process-based radon flux maps that will be available for use in the RTM, atmospheric dispersion modelling, and radiation protection. (WP3)
- To provide easy to use dynamic radon 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. (WP4)
- To facilitate the take up of the technology and measurement infrastructure developed in the project by the measurement supply chain (National Metrology Institutes, calibration laboratories), standards developing organizations (e.g. IEC, ISO) and end users in greenhouse gas monitoring and European radiological early warning networks. (WP5)

This deliverable in particular covers Task 2.4 of WP2: "RTM application at AMNS". The aim of this task is to develop a first general protocol for the application of the RTM to enable future retrievals of GHG fluxes at atmospheric climate gas monitoring stations and to use radon data for the identification of RPA. It builds on other traceRadon activities as well as the recent paper by Levin et al, 2021 as described below.

To reach this objective the following steps were taken:

- A review of ICOS (Integrated Carbon Observation System) AMNS sites was conducted in order to choose a station to evaluate the RTM. The characteristics of interest considered for the selection of the ICOS AMNS were: the ability to make vertical gradient measurements of radon and GHG mixing ratios and thus estimate GHG fluxes by at least two methods. The tall tower AMNS of SAC (Saclay, France) was proposed for the evaluation of the RTM.
- A sensitivity study of different parameters affecting the RTM output was conducted using the data from the chosen ICOS AMNS, SAC. In particular, the effect of deconvolution (correction of the dual-flow loop two filter radon detector response time (Griffiths et al., 2016) of the radon concentration on the correlation between radon and GHG as well as the uncertainties due to using different footprint models and radon flux maps was evaluated,

The deliverable is comprised of an overview section, followed by descriptions of the different steps taken to reach the objective of Task 2.4 as well as a short Summary combined with an Outlook. Finally, detailed reports of each activity and the publications produced in the framework of the deliverable can be found in the Annex.

1.1. Preliminary work

The deliverable D2 covered Task 1.2: Two transfer standard monitors were built and presented for low-level radon activity concentration and Task 1.3: Calibration and long-term stability of the TSs. A calibration procedure for the traceable measurement of atmospheric Rn-222 activity concentration in the field, with a specific focus on the needs of ICOS and other AMNS, was produced for D2. In this deliverable, the protocol for the measurement of atmospheric activity concentration for application in the RTM is based on the outcomes of deliverable D2.

The deliverable D3 covered Task 2.1: Designed, building and calibration of a new radon flux system which will be used as a transfer standard and Task 2.2: Intercomparison of available radon flux systems under field conditions. As a result of Task 2.1 and Task 2.2, deliverable D3 included guidelines for the installation, calibration and operation of a radon flux monitor. In this deliverable, the protocol for the measurement of radon flux for application in the RTM is based on the outcomes of deliverable D3.

The deliverable D5 *'Summary report on online available process-based radon flux maps with high temporal (daily) resolution'* covered Tasks 3.1. Its aim is to improve process-based radon flux maps for Europe that will be available for use in the RTM, atmospheric dispersion modelling, and radiation protection.

1.2. Radon Tracer Method

The Radon Tracer Method (RTM) has been used in many studies to evaluate the fluxes between atmosphere and ecosystems of trace gases such as CO_2, CH_4, N_2O or H_2 (e.g.: Levin et al., 1999, Schmidt et al., 2001, Biraud et al., 2000, Messager et al., 2008, Yver et al., 2009, Hammer et Levin, 2009, Lopez et al., 2012, Vogel et al., 2012, Belviso et al., 2013, Grossi et al., 2018, Belviso et al., 2020, Levin et al., 2021). Historically, the RTM has been applied in one of two ways: either to investigate regional-scale fluxes on an event basis (where an event may span hours or days), or to investigate local-scale fluxes on a nocturnal basis. Here, as we aim to propose an automated product, we are focusing on the nocturnal accumulation RTM.

The principle is based on the assumption of a constant flux J_{gas} in a well-mixed layer of height H during a nocturnal time window (8 to 10 hours window), thus we can write the temporal variation of its concentration as:

$$\frac{\overline{\Delta C_{gas}}}{\Delta t} = \frac{J_{gas}}{\overline{H}} \quad (1)$$

The same can be written for radon with an additional radioactive decay term.

$$\frac{\overline{\Delta C_{Rn}}}{\Delta t} = \frac{J_{Rn}}{\overline{H}} - \lambda_{Rn} C_{Rn} \quad (2)$$

If we combine equations 1 and 2 and we consider that for co-located measurement the height of the boundary layer is the same, we obtain:

$$J_{gas} = J_{Rn} \frac{\overline{\Delta C_{gas}}}{\overline{\Delta C_{Rn}}}\ decayterm \quad (3)$$

J_{Rn} is the ^{222}Rn flux over the region of influence, $\frac{\overline{\Delta C_{gas}}}{\Delta C_{Rn}}$ is the slope of the linear regression of observations between the gases. The overbar indicates that both mixing height and net surface flux of the catchment area are averaged for the observation period, and the ^{222}Rn 'decay term' is the factor used to correct for ^{222}Rn radioactive decay.

In this approach, the gas fluxes are considered similarly distributed in space and time, with no mixing of air from the free troposphere. The boundary layer height and the gas fluxes are assumed to remain constant during each event.

When we combine the RTM with air particle backtrajectories, we do not assume a regular region of influence to the radon concentration, but we consider that the influence of each grid cell around the station depends on the residence time of the incoming air over that grid cell (footprint). Hence, the radon flux J_{Rn} is calculated weighting the radon flux of each grid by a sensitivity value (source-receptor matrix) obtained with the backtrajectory model (Seibert and Frank, 2004). More details on this approach are described in Grossi et al., 2018.

In Levin et al. 2021, the limits of the method were thoroughly studied. The conclusions they reached are summarized here:

- The reliability of total nocturnal GHG emission estimates with the RTM critically depends on the accuracy and representativeness of the ^{222}Rn exhalation rates estimated from soils in the footprint of the site.
- Simply using ^{222}Rn fluxes as estimated by Karstens et al. (2015) could lead to biases in the estimated GHG fluxes as large as a factor of 2.
- RTM-based GHG flux estimates also depend on the parameters chosen for the nighttime correlations of GHG and ^{222}Rn, such as the nighttime period for regressions and the R^2 cut-off value for the goodness of the fit.

2. Development of a protocol for application of the RTM at AMNS

In this section, the course of action implemented for Task 2.4 is outlined. The subsections correspond to Activity Reports A2.4.1 and A2.4.2, which can be found in the Annex, and describe the steps in full detail.

2.1. Selection of sites to carry out the RTM evaluation (Activity Report A2.4.1)

The first part of Task 2.4 was performed by UVSQ with support from LUND, INESC TEC, NPL, JRC, UPC and ANSTO. It comprises a review of the ICOS AMNS where radon activity concentration is currently measured, with the intention to choose the suitable AMNS for RTM evaluation. The evaluation consisted of the analysis of a set of radon activity concentration, GHG mixing ratio and meteorological data from the selected AMNS station measured over a broad range of atmospheric conditions.

As per the description of activity A2.4.1, measurement of radon flux, as well as the radon activity and GHG concentrations at different heights of a 'tall tower' will be used to provide an understanding of how radon moves through different heights in air. The consistency and sensitivity of several approaches for the application of the RTM, will be evaluated and compared in order to produce a protocol for the application of the RTM, which will include installation guidance, measurement heights, equipment requirements and recommended instruments.

The site selected in the first place for this task is the ICOS Saclay tower. Saclay (SAC) is located 30 km south-west of Paris, 48.7217°N, 2.142°E, 160 m a.s.l. (above sea level). A 3-month intercomparison of radon and radon progeny monitors was previously carried out at this site in 2016 (Grossi et al., 2020).

The RTM has previously been applied at the nearby site of Gif-sur-Yvette, 2 km 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.

2.2. Evaluation of the RTM at Saclay ICOS station (Activity Report A2.4.2)

The next step was to evaluate the RTM at the selected AMNS, SAC ICOS station. The analysis included the early input from the improved radon flux maps in A3.1.3.

2.2.1. Description of the coding framework

The code is written in python and is hosted on the ICOS Carbon Portal (CP) JupyterLab. It thus takes advantage of the ICOS CP python package to access ICOS site data and already calculated footprints.

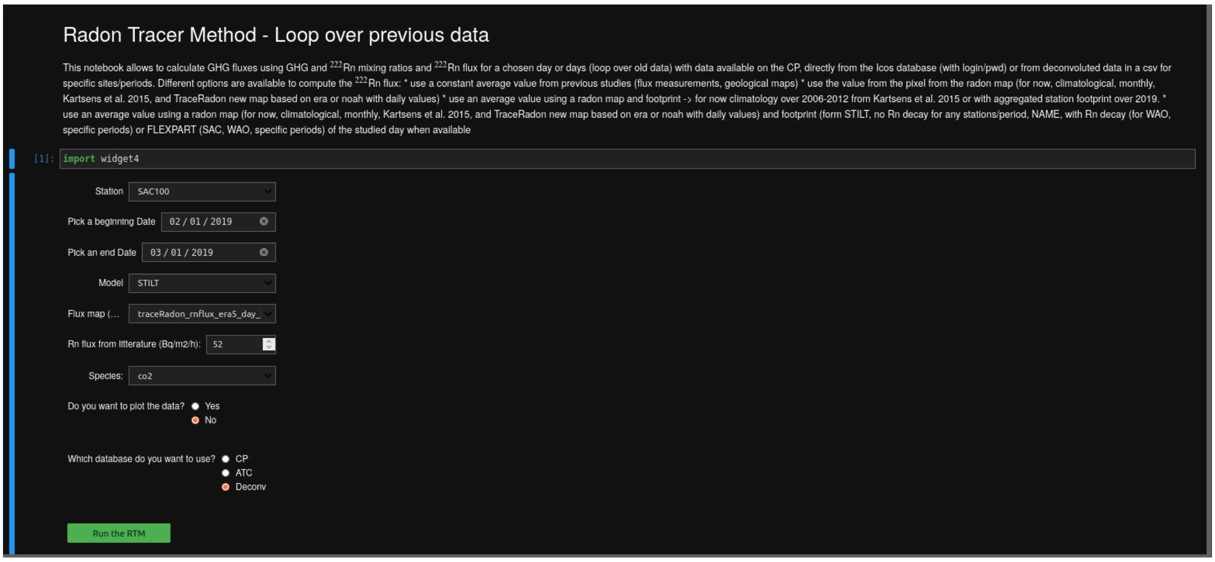

Figure 1: Screenshot of the RTM code developed during the project.

By default, it uses the footprints already calculated without radon decay by the Lagrangian model STILT as configured on the CP (available for all ICOS sites and more for at least 2018 to 2020). The STILT footprints are available every 3 hours and cover the region 33°S-73°N, 15°W-35°E with a resolution of

1/12° by 1/8°, approx. 10 km x 10 km. The STILT model is forced with European Centre for Medium-Range Weather Forecast (ECMWF) Integrated Forecasting System (IFS) operational analysis.

The radon exhalation maps used are either the InGOS one (Karstens et al., 2015), which is a climatology over 2006-2016 with one value per month, or the two new maps developed in WP3 (using either the reanalysed moisture data from ERA5-Land or GLDAS-Noah2.1) with a value per day and available from 2017 to May 2022. More information on the new maps can be found in the Deliverable Report D5. All maps can be downloaded at ICOS CP.

The maps and the footprints use a different grid, so when combined the radon exhalation map are regridded to the footprints.

The site to study can be chosen from the list available on the CP. The RTM can be applied to several species when data are available (CO_2, CH_4, N_2O and CO). Then, either it extracts the data from the CP NRT hourly data or if you have an access to the ICOS database with extraction rights, data with a smaller timestep can be extracted directly form the ICOS database.

By default, the code applies the RTM equation for the data between 21:00 to 06:00 UTC, which is a suitable window for most sites in Europe, but this window can be easily modified to fit with other latitudes or longitudes. The length of the window can be modified as well, for example to reproduce the tests from Levin et al. 2021.

No other criteria are applied but the correlation coefficient, the error on the linear regression, the number of data points and hours available for the calculation, the radon accumulation level and if the radon rise stopped before 08:00 UTC are recorded so the data can be filtered in a second step.

For the sensitivity study, we added the possibility of using radon and greenhouse gas data from csv files. Indeed, for the ANSTO detector, there is a measurement response time to consider, due to their design (a combined influence of their thoron delay volume, large measurement volume, and gross alpha counting approach). For optimal utilisation of radon measurements, a standardized protocol for data processing is required. This is not done yet in any ICOS radon data treatment chain. For this work, we have used a radon dataset derived from a preliminary standardized procedure, which is applicable to observations made by any similar (ANSTO made) radon monitoring system. The procedure to obtain the best estimate of atmospheric radon concentration (final product data) involves the traceability (WP1) and the post-processing of radon data, which includes the crucial deconvolution routine (step to correct for the instrument response time) as well as correction for standard temperature and pressure (STP).

We also added the possibility to use footprints from another model. For each model, it has to be tailored to it, depending on the grid size. The FLEXPART-WRF model version 3.3.2 (Brioude et al., 2013) run at UPC, is used here. This model uses WRF meteorological files as inputs for its backtrajectory calculations. This model was used with an output resolution of 0.05 degrees in order to fit with the new ERA-land and GLDAS-Noah2.1 radon maps. The backtrajectories were calculated for a 24h window time and assuming as footprint layer the 0-100 m height. For the Saclay site, the spatial window used was [42.9 - 54.5] LAT and [-6 - 16.2] LON.

2.2.2. Description of the different runs performed for this analysis

For the runs, we used the three different radon exhalation maps available (called hereafter InGOS, traceRadon_ERA5, traceRadon_Noah), two models (CP-STILT, WRF-FLEXPART) and two types of data (with and without the response time). Not all combinations are tested but all runs can go in pairs, with only one change from one to the other. Two months were chosen: February 2019 and August 2019 to observe the seasonal influence and as months with a good data coverage. The two models do not compute the radon decay term. It is applied as a fixed term in the equation (3) as in Schmidt et al., 2001.

Runs 1 and 2 (orange shaded cells in Table 1) were applied with the same input, except that radon data from the 1500L ANSTO monitor was used as calibrated detector output (not response time corrected) and as the best estimate (response time corrected) of the atmospheric radon concentration. This was done to study the influence of standardization on the efficiency of the RTM application. Runs 3 and 4 (yellow shaded cells in Table 1) were carried out using footprints calculated with the same CP-STILT model configuration and the same atmospheric concentration radon and GHG data. In this case the radon flux maps traceRadon-ERA5 and traceRadon_Noah were used to evaluate how radon fluxes calculated using different soil moisture reanalysis data could influence the final results. Finally, run 5 (blue shaded grid in Table 1) was executed with the same configuration of run 3, but using the FLEXPART-WRF based footprints which were calculated in the UPC cluster.

Table 1: Different runs for the sensitivity test

Run	Model	Radon map	Radon Data	Sites	Rn decay	Species
1	CP-STILT	InGOS	Not response time corrected ('raw')	SAC	No	CO_2
2	CP-STILT	InGOS	deconvoluted	SAC	No	CO_2
3	CP-STILT	traceRadon-Era5	deconvoluted	SAC	No	CO_2
4	CP-STILT	traceRadon-Noah	deconvoluted	SAC	No	CO_2
5	FLEXPART-WRF	TraceRadon-Era5	deconvoluted	SAC	No	CO_2

2.2.3. Results

Different ^{222}Rn fluxes for each night during the two months under study were used:

- constant radon flux value over the area of interest (52 Bq m^{-2} h^{-1});
- radon flux values obtained by available radon flux maps (InGOS, traceRadon_ERA5 and traceRadon_Noah) in the gridcell including the station. In the case of the InGOS map only a value for month was available where daily mean values are available for the two new traceRadon maps;
- radon fluxes values obtained coupling the previous radon flux maps with the ATM based footprints.

GHG fluxes within this study were calculated for every day during the months of February 2019 and August 2019 using, at least, two datapoints in the linear correlation between radon and CO_2.

The linear fits calculated between nocturnal radon and CO_2 data at the Saclay stations were then filtered to retain only the meaningful events using the following criteria: R^2 >0.6; error on the slope <50 %; radon increase over the night >1 Bq m^3.

Figure 2 shows the radon fluxes calculated at the Saclay station using the different methodologies as explained in details in the previous section.

Figure 2: Radon fluxes in February 2019 (top) and August 2019 (bottom)

Results show that winter fluxes are generally lower than summer ones, as it was expected from the literature because of the lower water content in the soil during dry period. Daily radon fluxes based on GLDA-Noah reanalysis offer, for this station and periods of time, higher values than the ones calculated using ERA5-Land data (red and green dots in the central panels of Figure 2). Unfortunately, from the preliminary work of A3.3.4 and A3.3.5, still on-going, it is not possible to state if one dataset is better than the other for this specific site. Specifically daily fluxes vary between 12 and 22 Bq h^{-1} m^{-2} for run 3 and in the range of 32 to 32 and 40 Bq h^{-1} m^{-2} for run 4, while run 2 is at 20 Bq h^{-1} m^{-2} in February 2019. In August 2019, they vary between 48 and 58 Bq h^{-1} m^{-2} for run 3, between 67 and 72 Bq h^{-1} m^{-2} for run 4 and run 2 is at 42 Bq h^{-1} m^{-2}.

Radon flux results calculated using radon flux maps and ATM footprints show as expected a different variability, but the range are in the same order of magnitude. In February, the fluxes vary between 19 and 38 Bq h^{-1} m^{-2} for run 2, 11 and 42 Bq h^{-1} m^{-2} for run 3, 36 and 71 Bq h^{-1} m^{-2} for run 4, and 15 and 38 Bq h^{-1} m^{-2} for run 5. In August, the fluxes vary between 26 and 64 Bq h^{-1} m^{-2} for run 2, 31 and 88 Bq h^{-1} m^{-2} for run 3, 44 and 119 Bq h^{-1} m^{-2} for run 4 and 30 and 115 Bq h^{-1} m^{-2} for run 5.

Figure 3 shows the results of the CO_2 fluxes obtained by RTM from Equation (3) using the different configurations presented in Table 1.

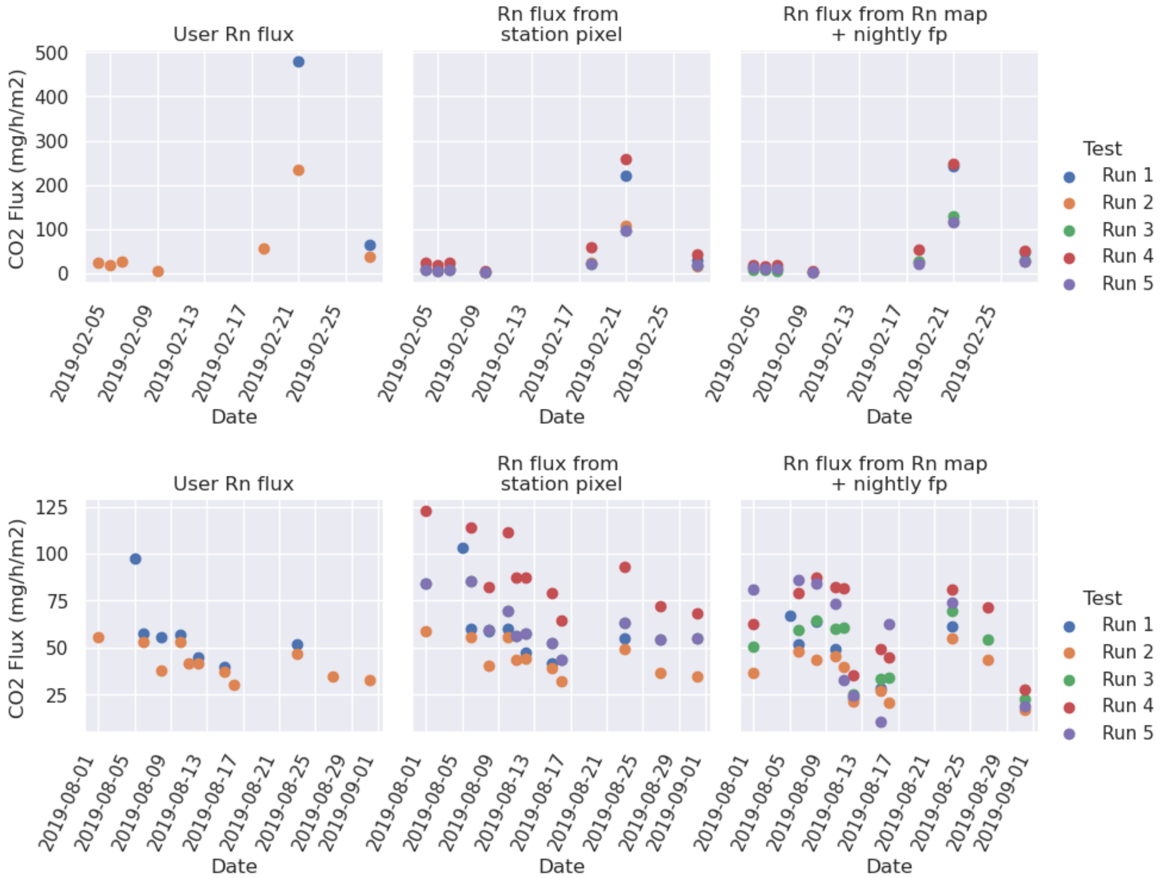

Figure 3: CO_2 fluxes calculated with the RTM for February 2019 (top) and August 2019 (bottom)

As can be expected, the variability on the radon fluxes is seen as well on the CO_2 fluxes. It is however interesting to notice that using the best estimate of atmospheric radon concentration, radon and GHG are more often seen as correlated and thus GHG flux can be calculated on more days. The standardized dataset allows to allocate the right sampling time for the radon measurement and thus when the two gases are influenced by the same air masses their correlation is better than when the data are not correcting and lagging behind.

In February, seven events are selected when optimized versus two without. In August, eleven events are selected when optimized versus six without.

For the year 2018, the CO_2 mean emission from the inventory EDGAR (Crippa et al., 2019) for the pixel around Saclay is 77 mg h^{-1} m^{-2}. The results from the RTM are of the same order of magnitude with fluxes computed using the exhalation map and the footprints, the average standard deviation between the runs is 6 mg h^{-1} m^{-2} (excluding Feb 21) in February and 14 mg $h^{-1}m^{-2}$ in August for a global average of 17 and 50 mg h^{-1} m^{-2} in February and August respectively.

From this sensitivity test, it appears important to estimate the radon fluxes with at least the two different radon exhalation maps developed in the project to be able to estimate the range of uncertainties of the calculated fluxes. It is also important to use the standardized data when needed in order to obtain a more realistic correlation between GHG and radon.

3. Protocol for the measurement of radon flux and atmospheric activity concentration for application in the RTM or to identify RPAs

To apply the RTM, we must first obtain properly calibrated GHG and radon data and apply STP correction on both gases. For the ANSTO radon detectors, it is also necessary to apply a response time correction (Griffiths et al., 2016).

Then, the radon flux for the time window of the calculation has to be estimated. Finally, the radon decay term can carry some additional uncertainty. Here, we used a constant value in equation (3) as in Schmidt et al., 2001. However, it could be directly calculated with the ATM models to obtain a value tailored to each situation.

For RPAs, radon concentration measurement could be used in model inversion to validate the flux maps and therefore needs to be optimized as well.

Within ICOS, the GHG data follow a standardized calibration procedure to ensure their quality. Uncertainty on the GHG measurement is very low compared to other terms.

Below, we compile the recommendations for the radon flux and concentration measurements.

3.1 Guidelines for the installation, calibration and operation of a radon flux monitor

A radon flux Transfer Standard (TS) system was developed under Task 2.1: Continuous radon flux monitor as a transfer standard, to support the creation of a complete metrology chain for the measurement of in situ radon fluxes from soils.

For this purpose, as reported in deliverable D3, the following steps were taken: i) an exhalation bed (EB) facility was designed and built based on the findings of a literature review of existing EB facilities and requirements; ii) a radon flux TS was selected based on the findings of a literature review of available radon flux systems; iii) the chosen TS was characterized, from both a theoretical and experimental point of view, using the EB under laboratory conditions and a box model; iv) the TS and the EB were used to calibrate other radon flux systems and a calibration protocol was produced; iv) the response of the TS and other available radon flux systems were compared at a reference site and guidelines were generated for in situ radon flux measurements (Rabago et al., 2022).

The TS and EB facility enabled the calibration of radon flux systems with a total calibration factor uncertainty of 6.4 % (k = 1) for radon fluxes in the order of a thousand $Bq \cdot m^{-2} \cdot h^{-1}$. From the conclusions of deliverable D3, radon flux systems used for in situ radon flux measurements should consider:

- A continuous radon monitor working in flow mode, with a low internal background, a temporal resolution not higher than 10 minutes, high precision, and, preferably, the ability to distinguish between radon and thoron contributions;
- An accumulation chamber that can be opened automatically at a set time interval, with a collar that can be correctly installed into the soil, with environmental sensors to monitor conditions inside and outside of the chamber as well as in the surface soil layer, painted in a reflective color to minimize solar heating of chamber air, with an effective height no bigger than 0.2 m to avoid low radon concentrations inside the chamber;
- A previous calibration under laboratory conditions using a TS and the EB, or being exposed directly in the field together with the TS for the transfer of the calibration/sensitivity factors;
- Application of the protocol of Figure 4 when the monitor is used in field measurements to determine the maximum accumulation time to be applied for the linear fit method to be reliable;

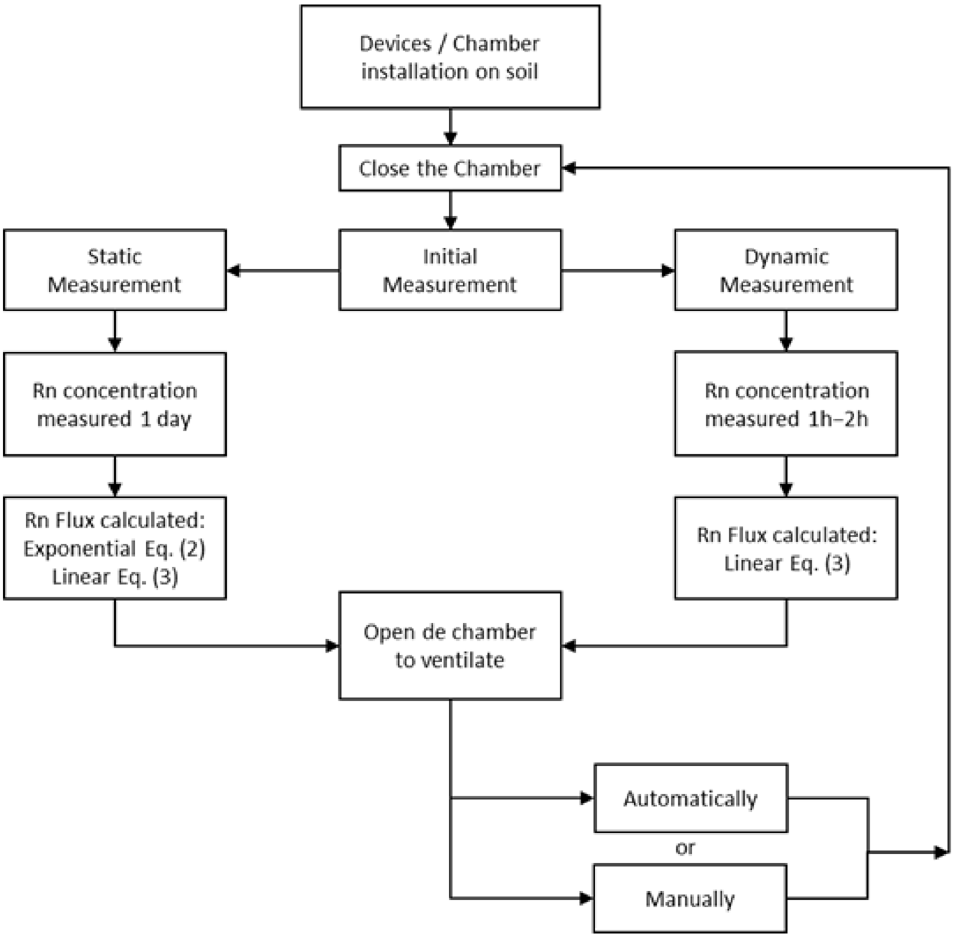

Figure 4. Flowchart of the methodology applied to radon flux measurements in field. (Extracted from D3)

- The use of a thoron delay volume in cases where the monitor is not capable of selective Rn-222 measurement when thoron is also present in the air. However, the user should be mindful of the fact that this delay volume also delays the temporal evolution of radon concentration in the monitor.
- A system with an automatic arm to carry out radon flux measurement at different points could be also recommendable.

In the case of the RTM, individual measurements close to the sites are not fully representative of the air masses reaching the station but can be a good indicator of the radon map accuracy for the station pixel. If radon flux measurement could be done within the average footprint of the site over different seasons, these would help radon flux map validation.

3.2 Guidelines for the choice, calibration and operation of radon atmospheric activity concentration monitors

From the conclusions of Activity 1.2.1, the following Table 2 of properties was produced for radon monitors to measure within atmospheric levels:

Table 2. Matrix of recommended properties for the in-field application of a transfer standard radon monitor for atmospheric measurements.

Property	Recommended range for in field applicability
Environmental Temperature (°C)	-25 - +50
Environmental relative Humidity (%)	10 - 100
Atmospheric Pressure (hPa)	620-1100
Measurable Atmospheric radon activity concentration (Bq m^{-3})	1 - 200
Sensitivity (cpm per Bq m^{-3})	> 0.3
Total uncertainty (%) for activity higher than 0 Bq m^{-3} and less of 100 Bq m^{-3} in 1 h (k = 2)	< 20
Volume and weight (kg)	< 1 m^3 < 50

Table 2 was designed for a transfer standard but any instrument at an AMNS should meet the same criteria in order to produce meaningful data outside of the volume and weight.

The uncertainties of two types of instruments used in AMNS that meet these criteria (ANSTO 200 L and ARMON) have been thoroughly evaluated in WP1 and summed up in D2. The uncertainty of the ARMON was described in detail in the combined Activity Report A1.2.2 and A1.2.3. It amounted to 0.42 Bq m^{-3} for k = 1 at a total ^{222}Rn activity concentration of 3.34 Bq m^{-3} (12.57 %). A new uncertainty budget was created for the ANSTO 200 L detector. The uncertainty of the ANSTO 200 L greatly depends on whether response-time-correction is regarded or not and on the variability of the internal flow used by the instrument. Without, it amounts to 0.213 Bq m^{-3} for k = 1 at a ^{222}Rn activity concentration of C_{Rn}= 3.476 Bq m^{-3} (6.13 %). With the correction, but using more realistic uncertainties, mostly on the internal flow uncertainty, it amounts to around 12 % for *k* = 1, comparable to the one by the 20L ARMON. The uncertainty of both the ARMON 20L as well as the ANSTO 200 L are well within the aspired goal of an uncertainty of 15 % for *k*= 1.

In combined activities A1.1.3 and A1.1.4, four calibration procedures have been developed. The first three are based on measurements in a controlled chamber and thus are only for laboratory measurements. They can be used as initial calibration before filed deployment. The fourth procedure could be used in the field as well as in the laboratory as it doesn't involve a controlled constant atmosphere. However, more development in the term of software, detailed instructions and outreach are needed before it can be widely implemented.

The activities A1.3.3 and A1.3.4 that involve short-term and long-term measurement campaigns will provide fine-tuned recommendations for the calibration strategy of each type of instrument.

However, what is needed to achieve good radon concentration measurement is:

- An instrument meeting the criteria from Table 2
- If possible, an initial calibration in a laboratory with a primary or secondary standard (source or transfer standard)
- Field calibration using protocols depending on the instruments
- Correction to standard temperature and pressure
- Depending on the instrument, a response-time correction

4. Summary and Outlook

The presented deliverable D4 is titled "Good Practice Guide including a standard protocol for the measurement of radon flux and atmospheric activity concentration for application in the radon tracer method (RTM) for greenhouse gas (GHG) flux estimates and for its application to derive data for Radon Priority Areas (RPA)" and covers Task 2.4 of WP2: "RTM application at AMNS". The application of the RTM was successfully carried out and a protocol developed. The quality needed for the RTM is also necessary for identifying RPAs and thus the guidelines for the flux and concentrations monitors are applicable for both subjects.

In summary:

Radon flux systems used for in situ radon flux measurements should consider:

- A continuous radon monitor working in flow mode, with a low internal background, a temporal resolution not higher than 10 minutes, high precision, and, preferably, the ability to distinguish between radon and thoron contributions;
- An accumulation chamber that can be opened automatically at a set time interval, with a collar that can be correctly installed into the soil, with environmental sensors to monitor conditions inside and outside of the chamber as well as in the surface soil layer, painted in a reflective color to minimize solar heating of chamber air, with an effective height no bigger than 0.2 m to avoid low radon concentrations inside the chamber;

- A previous calibration under laboratory conditions using a TS and the EB, or being exposed directly in the field together with the TS for the transfer of the calibration/sensitivity factors;
- Application of the protocol of Figure 4 when the monitor is used in field measurements to determine the maximum accumulation time to be applied for the linear fit method to be reliable;
- The use of a thoron delay volume in cases where the monitor is not capable of selective Rn-222 measurement when thoron is also present in the air. However, the user should be mindful of the fact that this delay volume also delays the temporal evolution of radon concentration in the monitor.
- A system with an automatic arm to carry out radon flux measurement at different points could be also recommendable.

Radon concentration systems used for in situ radon concentration measurements should consider:

- An instrument meeting the criteria from Table 2
- If possible, an initial calibration in a laboratory with a primary or secondary standard (source or transfer standard)
- Field calibration using protocols depending on the instruments
- Correction to standard temperature and pressure
- Depending on the instrument, a response-time correction

To apply the RTM and select data that are compliant with the underlying hypotheses of the method, one should consider:

- Using radon concentration measurements performed as recommended in 3.2.
- Using GHG concentration measurements that are quality controlled such as ICOS datasets.
- Using more than one radon flux exhalation map and if available more than one footprint model to estimate the radon flux uncertainty.
- If possible, comparing the radon flux with local measurements done as described in 3.1 within the mean footprint.
- Choose an adequate time window for the nocturnal accumulation gradient at your station: it can vary depending on the latitude/longitude of the station.
- Selecting data with a good correlation (i.e. R^2 >0.6), and a significant radon concentration rise (i.e. above 1Bq.m^{-3}) to select meaningful events.
- Perform a sensitivity study as in Levin et al., 2021 and here to evaluate the best criteria for an operational RTM calculation.
- Station sampling heights have to be taken into account; lower levels will allow to calculate local fluxes while higher levels may be decorrelated from the immediate surface during the night, being above the boundary layer height

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Annex: Activity Reports

- **Activity Report A2.4.1**
- **Activity Report A2.4.2**

JRP EMPIR 19ENV01: traceRadon

A2.4.1.M3 Selection of sites to carry out the RTM evaluation

Elaborated: UVSQ

Participants: LUND, INESC TEC, NPL, JRC, UPC, ANSTO

Version: 1

Date: 31. August 2020

Content

Introduction

Radon (^{222}Rn) is a naturally occurring radioactive gas that is ubiquitous in the environment, and as such contributes over half the total public exposure to radiation dose from natural sources (WHO, 2009). Due to its not so short (3.8 day) half-life and chemical inertness, radon can be used as a tracer for atmospheric transport and mixing studies and applied to geological studies (WMO, 2003; WMO, 2012; Yang et al., 2017; Chambers et al., 2018; Kikaj et al., 2019). One way to use radon as tracer is the Radon Tracer Method (RTM), which enables the estimation of local- to regional-scale fluxes of greenhouse gases for species with distributed sources (Levin et al., 1999).

Radon tracer method

The radon tracer method has been used in many studies to evaluate the fluxes between atmosphere and ecosystems of trace gases such as CO_2, CH_4, N_2O or H_2 (e.g.: Levin et al., 1999, Schmidt et al., 2001, Biraud et al., 2002, Messager et al., 2008, Yver et al., 2009, Hammer et Levin, 2009, Lopez et al., 2012, Vogel et al., 2012, Belviso et al., 2013, Grossi et al., 2018, Belviso et al., 2020).

The principle is based on the assumption of a constant flux J_{gas} in a well-mixed layer of height H, we can write the temporal variation of its concentration as

$$\frac{\Delta Cgas}{\Delta t} = \frac{Jgas}{H}$$

The same can be written for radon with an additional radioactive decay term.

$$\frac{\Delta Crn}{\Delta t} = \frac{Jrn}{H} - \lambda rnCr$$

If we combine the two, we eliminate the boundary layer height,

$$Jgas = Jrn\frac{\Delta Cgas}{\Delta Crn}\left(1 - \frac{\lambda rnCrn}{\frac{\Delta Crn}{t}}\right)$$

Jrn is the mean ^{222}Rn flux over the region of influence, ΔCgas/ΔCrn is the slope of the linear regression of observations between the gas and ^{222}Rn and λrn is the factor used to correct for ^{222}Rn radioactive decay.

In this approach, the gas fluxes are considered colocated spatially and temporally, with no mixing of air from the free troposphere. The boundary layer height and the gas fluxes are assumed to remain constant during each event.

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, 48.7217°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, 1.5$, 10$*, 40$, 60$*, 80$*m agl ($ 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 .

Conclusions

The site described above comply with requirements for application of the RTM and is also capable of characterising the vertical radon gradient. In addition to the existing radon monitoring at two levels the site could potentially accommodate an additional radon instrument either for intercomparison purposes, or for observations an intermediate level. It is also possible to install radon flux monitors and other instruments if necessary. Meteorological parameters and greenhouse gas observations are already available for several years, and are ongoing.

We therefore recommend SAC as appropriate site to perform the first evaluation of the RTM. In a second step, we plan to extend the evaluation to other sites such as Tacolneston or Weybourne in UK or Spanish sites, where Radon flux and meteorology may be different from those at SAC.

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JRP EMPIR 19ENV01: traceRadon

A2.4.2. Evaluation of the RTM at selected sites

Elaborated: UVSQ

Participants: LUND, NPL, UPC, ANSTO

Version: 1.1

Date: 26 September 2022

Content

Introduction

Radon (^{222}Rn) is a naturally occurring radioactive gas that is ubiquitous in the environment, and as such contributes over half the total public exposure to radiation dose from natural sources (WHO, 2009). Due to its not so short (3.8 day) half-life and chemical inertness, radon can be used as a tracer for atmospheric transport and mixing studies and applied to geological studies (WMO, 2003; WMO, 2012; Yang et al., 2017; Chambers et al., 2018; Kikaj et al., 2019). One way to use radon as tracer is the Radon Tracer Method (RTM), which enables the estimation of local- to regional-scale fluxes of greenhouse gases for species with distributed sources (Levin et al., 1999).

Radon tracer method

The nocturnal accumulation Radon Tracer Method (RTM) has been used in many studies to evaluate the fluxes between atmosphere and ecosystems of trace gases such as CO_2, CH_4, N_2O or H_2 (e.g.: Levin et al., 1999, Schmidt et al., 2001, Biraud et al., 2002, Messager et al., 2008, Yver et al., 2009, Hammer et Levin, 2009, Lopez et al., 2012, Vogel et al., 2012, Belviso et al., 2013, Grossi et al., 2018, Belviso et al., 2020, Levin et al., 2021).

The principle is based on the assumption of a constant flux J_{gas} in a well-mixed layer of height H during a nocturnal time window (8 to 10 hours window), thus we can write the temporal variation of its concentration as:

$$\Delta C^{-} gas \Delta t = Jgas H^{-} \quad [1]$$

The same can be written for radon with an additional radioactive decay term.

$$\Delta CRn^{-} \Delta t = JRn H^{-} - \lambda Rn CRn \quad [2]$$

If we combine the equations 1 and 2 and we considered that for lo-cate measurement the height of the boundary layer is the same, we obtain:

$$Jgas = JRn \Delta Cgas^{-} \Delta CRn^{-} decayterm \quad [3]$$

J_{Rn} is the ^{222}Rn flux over the region of influence, $\Delta Cgas^{-} \Delta CRn^{-}$ is the slope of the linear regression of observations between the gas. The overbar indicates that both mixing height and net surface flux of the catchment area are averaged for the observation period. ^{222}Rn and 'decay term' is the factor used to correct for ^{222}Rn radioactive decay.

In this approach, the gas fluxes are considered co-located spatially and temporally, with no mixing of air from the free troposphere. The boundary layer height and the gas fluxes are assumed to remain constant during each event.

When we combine the RTM with air particle backtrajectories, we do not assume a regular region of influence to the radon concentration, but we consider that the influence of each grid cell around the station depends on the residence time of the incoming air over that grid cell (footprint). Hence, the radon flux J_{Rn} is calculated weighting the radon flux of each grid by a sensitivity value (source-receptor matrix) obtained with the backtrajectory model (Seibert and Frank, 2004).

In Levin et al. 2021, the limits of the method were thoroughly studied. The conclusions they reached are summarized here:

- The reliability of total nocturnal GHG emission estimates with the RTM critically depends on the accuracy and representativeness of the ^{222}Rn exhalation rates estimated from soils in the footprint of the site.
- Simply using ^{222}Rn fluxes as estimated by Karstens et al. (2015) could lead to biases in the estimated GHG fluxes as large as a factor of 2.
- RTM-based GHG flux estimates also depend on the parameters chosen for the nighttime correlations of GHG and ^{222}Rn, such as the nighttime period for regressions and the R^2 cut-off value for the goodness of the fit.

Selection of a site to carry out the RTM method evaluation

From Activity 2.4.2, Saclay has been selected as the main site for the evaluation. Saclay (SAC) is located 30km south-west of Paris, 48.7217°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, 1.5$, 10$*, 40$, 60$*, 80$*m agl ($ 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.

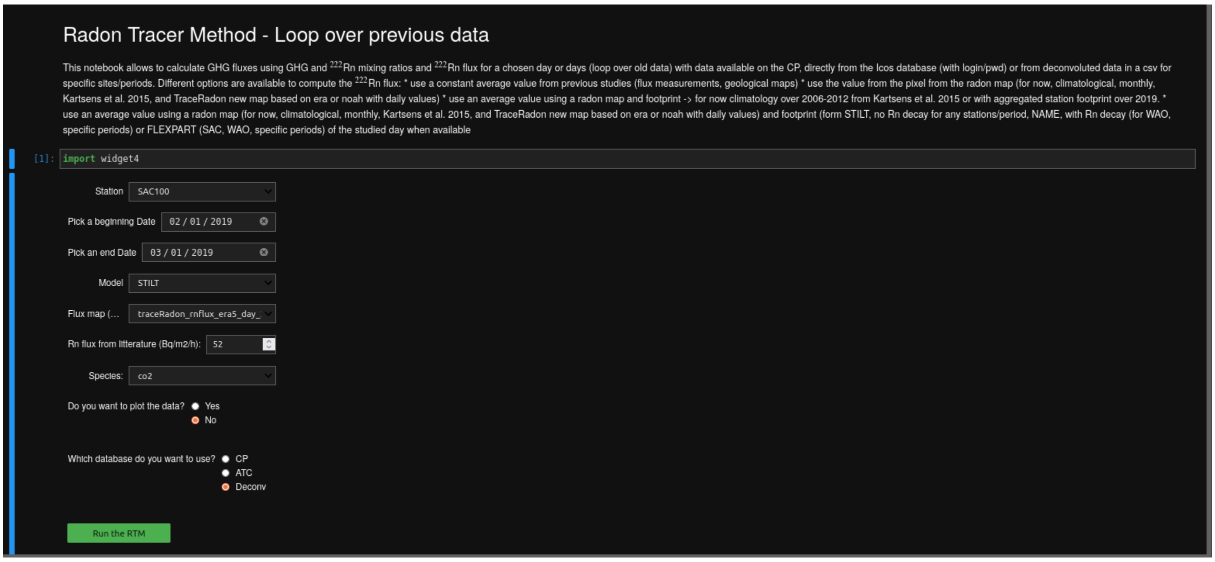
For comparison with the UK NAME model, we also use data from the Weybourne Atmospheric Observatory (WAO).

Routine radon monitoring at WAO is conducted at 10m with the greenhouse gas sampling and the meteorological parameters. The above data are available in the ICOS database, since March 2018 for radon and since August 2020 for greenhouse gases.

Description of the RTM framework

The code is written in python and is hosted on the ICOS Carbon Portal (CP) JupyterLab.

It thus takes advantage of the ICOS CP python package to access ICOS site data and already calculated footprints.



By default, it uses the footprints already calculated without radon decay by the Lagrangian model STILT as configured on the CP (available for all ICOS sites and more for at least 2018 to 2020). The STILT footprints are available every 3 hours and cover the region 33°S-73°N, 15°W-35°E with a resolution of 1/12° by 1/8°, approx. 10km x 10km. The STILT model is forced with European Centre for Medium-Range Weather Forecast (ECMWF) Integrated Forecasting System (IFS) operational analysis.

The radon exhalation maps used are either the InGOS one (Karstens et al., 2015) which is a climatology over 2006-2016 with one value per month or the two new maps developed in WP3 (using either the reanalysed moisture data from ERA5-Land or GLDAS-Noah2.1) with a value per day and available from 2017 to May 2022. More information on the new maps can be found in the Deliverable Report D5. All maps can be downloaded at ICOS CP.

The maps and the footprints use a different grid so when combined the radon exhalation map are regridded to the footprints.

The site to study can be chosen from the list available on the CP. The RTM can be applied to several species when data are available (CO_2, CH_4, N_2O and CO).

Then either it extracts the data from the CP NRT hourly data or if you have an access to the ICOS database with extraction rights, data with a smaller timestep can be extracted directly form the ICOS database.

The code applies the RTM equation for the data between 21:00 to 06:00 UTC which is a suitable window for most sites in Europe but this window can be easily modified to fit with other latitudes or longitudes. The length of the window can be modified as well for example to reproduce the tests from Levin et al. 2021.

No other criteria are applied but the correlation coefficient, the error on the linear regression, the number of data points and hours available for the calculation, the radon accumulation level and if the radon rise stopped before 08:00 UTC are recorded so the data can be filtered in a second step.

For the analysis here, we added the possibility of using radon and greenhouse gas data from csv files. . Indeed, for the ANSTO detector, there is a time-response delay due to their design (a thoron delay volume so only the ^{222}Rn is counted at the end). For these detectors, we thus need to correct for this delay using deconvolution routine. This is not done yet in ICOS data treatment chain and the influence of this correction on the RTM was tested here as the ANSTO detectors are used at SAC.

We also added the possibility to use footprints from another models. For each model, it has to be tailored to it, depending on the grid size. The FLEXPART-WRF model version 3.3.2 (Brioude et al.2013), run at UPC, is used here. This model uses WRF meteorological files as inputs for its backtrajectory calculations. This model was used with an output resolution of 0.05 degrees in order to fit with the new ERA-land and GLDAS-Noah2.1 radon maps. The backtrajectories were calculated for a 24h window time and assuming as footprint layer the 0-100m height. For the Saclay site, the spatial window used was [42.9 - 54.5] LAT and [-6 - 16.2] LON.

Descriptions of the different runs performed for this analysis

For the runs, we use the 3 different radon exhalation maps available (called hereafter InGOS, traceRadon_ERA5, traceRadon_Noah), two models (CP-STILT, WRF-FLEXPART), two types of data (not time delay corrected, deconvoluted (time delay corrected)). Not all combinations are tested but all runs can go in pairs, with only one change from one to the other. Two months were chosen: February 2019 and August 2019 to observe seasonal influence and as month with a good data coverage in both sites. The two models do not compute the radon decay term. It is applied as a fixed term in the equation 3 as in Schmidt et al., 2001.

Runs 1 and 2 (orange shaded cells in Table 1) were applied with the same input except radon data from ANSTO monitor was used with and without the deconvolution applied to correct the later response of the 1500L volume instrument. This was done to study if not deconvoluted data may affect the efficiency of the RTM application. Runs 3 and 4 (yellow shaded cells in table 1) were carried out using footprints calculated with the same CP-STILT model configuration and the same atmospheric concentration radon and GHG data. In this case the radon flux maps traceRadon-ERA5 and traceRadon_Noah was used to evaluate how radon fluxes calculated using different soil moisture reanalysis data could influence the final results. Finally, run 5 (blue shaded grid in table 1) was executed with the same configuration of run 3 but using the FLEXPART-WRF based footprints which were calculated in the UPC cluster.

Run	Model	Radon map	Radon Data	Sites	Rn decay	Species
1	CP-STILT	InGOS	Not time delay corrected ('raw')	SAC	No	CO_2
2	CP-STILT	InGOS	deconvoluted	SAC	No	CO_2
3	CP-STILT	traceRadon-Era5	deconvoluted	SAC	No	CO_2
4	CP-STILT	traceRadon-Noah	deconvoluted	SAC	No	CO_2
5	FLEXPART-WRF	TraceRadon-Era5	deconvoluted	SAC	No	CO_2

Table 1: Different runs for the sensitivity test

Results

Different ^{222}Rn fluxes for each night during the months under study were used:

- constant radon flux value over the area of interest (52 Bq m-2 h-1);
- radon flux values obtained by available radon flux maps (InGOS, traceRadon_ERA5 and traceRadon_Noah) in the gridcell including the station. In the case of the InGOS map only a value for month was available where daily mean values are available for the two new traceRadon maps;

- radon fluxes values obtained coupling the previous radon flux maps with the ATM based footprints.

GHG fluxes within this study were calculated for every day during the months of February 2019 and August 2019 using, at least, two datapoints in the linear correlation between Radon and CO_2.

The linear fits calculated between nocturnal radon and CO_2 data at the Saclay stations were then filtered to retain only the meaningful events using the following criteria: R^2>0.6; error on the slope <50 %; 222-Rn increase over the night >1Bq.m^3.

Figure 1 shows the radon fluxes calculated at the Saclay station using the different methodologies as it was explained in details in the previous section.

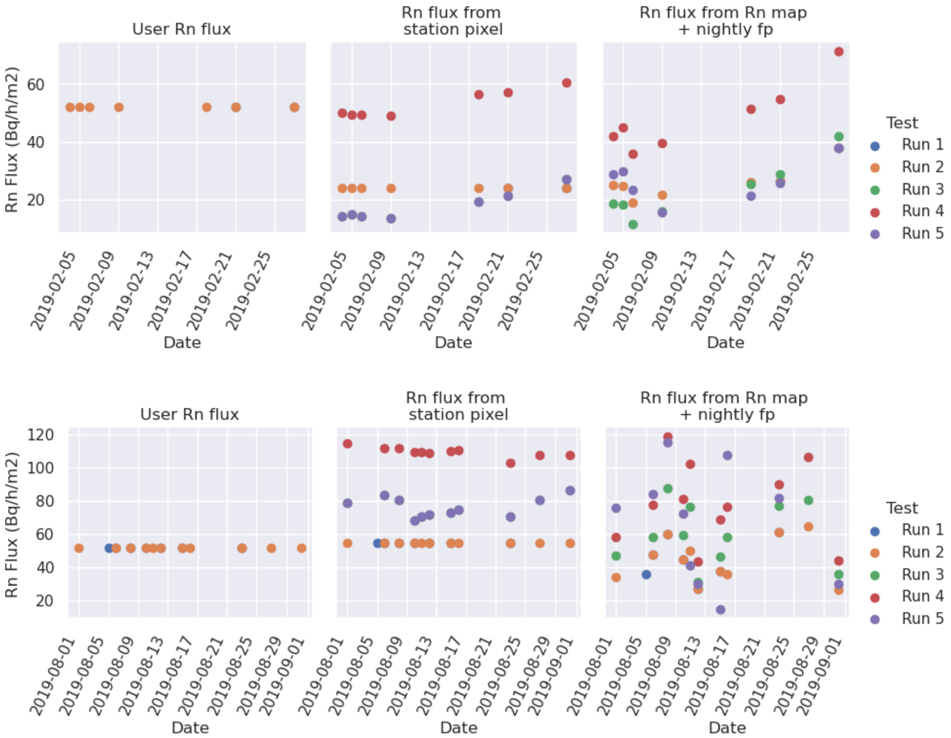


Figure 2: Radon fluxes in February 2019 (top) and August 2019 (bottom)

Results show winter fluxes are generally lower than summer ones as it was expected from the literature because of the lower water content in the soil during dry period. Daily radon fluxes based on GLDA-Noah reanalysis offer, for this station and periods of time, higher values than the ones calculated using ERA5-Land data (red and green dots in the central panels of Figure 2). Unfortunately, from the preliminary work of A3.3.4 and A3.3.5, still on-going, it is not possible to say if one dataset is better than the other one for this specific site. Specifically daily fluxes vary between 12 and 22 Bq.h^{-1}.m^{-2} for the run 3 and between 32 and 40 Bq.h^{-1}.m^{-2} for run 4 while run 2 is at 20 Bq.h^{-1}.m^{-2} in February and between 48 and 58 Bq.h^{-1}.m^{-2} for run

3 and between 67 and 72 Bq.h^{-1}.m^{-2} for run 4 while run 2 is at 42 Bq.h^{-1}.m^{-2} in August 2019. Radon flux results calculated using radon flux maps and ATM footprints show as expected a different variability but the range are in the same order of magnitude. In February, the fluxes vary between 19 and 38 Bq.h^{-1}.m^{-2} for run2, 11 and 42 Bq.h^{-1}.m^{-2} for run 3, 36 and 71 Bq.h^{-1}.m^{-2} for run 4 and 15 and 38 Bq.h^{-1}.m^{-2} for run 5. In August, the fluxes vary between 26 and 64 Bq.h^{-1}.m^{-2} for run2, 31 and 88 Bq.h^{-1}.m^{-2} for run 3, 44 and 119 Bq.h^{-1}.m^{-2} for run 4 and 30 and 115 Bq.h^{-1}.m^{-2} for run 5.

Figure 3 shows the results of the CO_2 fluxes obtained by RTM from Equation 3 using the different configurations presented in table 1.

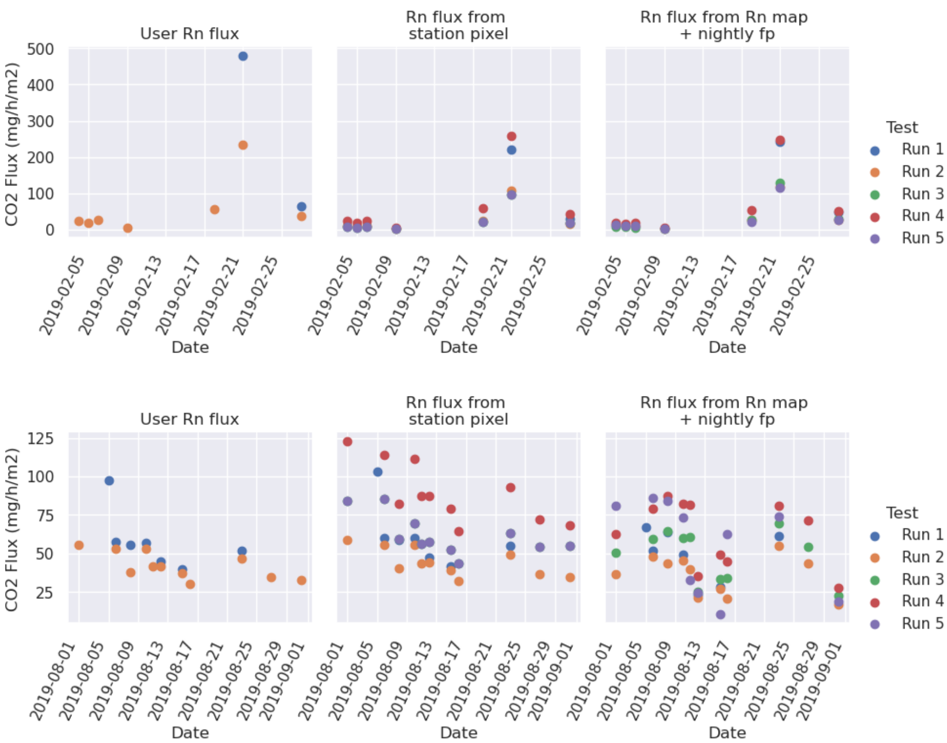


Figure 3: CO_2 fluxes calculated with the RTM for February 2019 (top) and August 2019 (bottom)

As can be expected, the variability on the radon fluxes is seen as well on the CO_2 fluxes. It is however interesting to notice that with deconvolution, radon and GHG are more often seen as correlated and thus GHG flux can be calculated on more days. The deconvolution allows to allocate the right sampling time for the radon measurement and thus when the two gases are influenced by the same air masses their correlation is better than when the data are not correcting and lagging behind.

In February, seven events are selected when deconvoluted versus two without. In August, eleven events are selected when devonvoluted versus six without.

For the year 2018, the CO_2 mean emission from the inventory EDGAR (Crippa et al., 2019) for the pixel around Saclay is 77 mg $h^{-1}m^{-2}$. The results from the RTM are of the same order of magnitude. Looking at the fluxes computed using the exhalation map and the footprints, the average standard deviation between the runs is 6 mg $h^{-1}m^{-2}$ (excluding Feb 21) in February and 14 mg $h^{-1}m^{-2}$ in August for a global average of 17 and 50 mg $h^{-1}m^{-2}$ in February and August respectively.

From this sensitivity test, it appears important to estimate the radon fluxes with at least the two different radon exhalation maps developed in the project to be able to estimate the range of uncertainties of the calculated fluxes. It is also important to deconvolute the data when needed in order to obtain a more realistic correlation between GHG and radon.

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