





19ENV01 traceRadon

The Radon Tracer Method – How to use it

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1. Introduction

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 ²²²Rn 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)

1.1. 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 CO₂, CH₄, N₂O or H₂ (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:

$$\frac{\overline{\Delta C}_{gas}}{\Delta t} = \frac{J_{gas}}{\overline{H}}$$
(1)

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

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

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

 J_{Rn} is the ²²²Rn flux over the region of influence, $\frac{\Delta C_{gas}}{\Delta C_{Rn}}$ 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 ²²²Rn 'decay term' is the factor used to correct

area are averaged for the observation period, and the $^{\rm 222}Rn$ 'decay term' is the factor used to correct for $^{\rm 222}Rn$ 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 ²²²Rn exhalation rates estimated from soils in the footprint of the site.
- Simply using ²²²Rn 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 ²²²Rn, such as the nighttime period for regressions and the R² cut-off value for the goodness of the fit.

2. The Radon Tracer Method software

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.

	Radon Tracer Method - Loop over previous data								
	This notebook allows to calculate GHG fluxes using GHG and ²²² Rn mixing ratios and ²²² Rn flux for a chosen day or days (loop over old data) with data available on the CP, directly from the loos database (with login/gwd) or from deconvoluted data in a cev for specific sites/periods. Different options are available to compute the ²²² Rn flux. 'use a constant average value from previous studies (flux measurements, geological maps) 'use the value from the pixel from the radon map (for now, climatological, monthly, Kartsens et al. 2015, and TraceRadon new map based on era or noah with daily values) 'use an average value using a radon map and footprint - for now climatology over 2006-2012 from Kartsens et al. 2015 or with aggregated station footprint over 2019. ' use an average value using a radon map (for now, climatological, monthly, Kartsens et al. 2015, and TraceRadon new map based on era or noah with daily values) and tootprint (form STILT, no Rn decay for any stations/period, NAME, with Rn decay (for WAO, specific period) or (EXPART (SAC, WAO, Specific periods) or the XPART (SAC, WAO, Specific period								
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	Run the RTM								

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 the 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. 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 05:00 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.

A log file is produced to inform about the success of the query.

Exercices

The version in exploredata is a lighter version than the full one that will be available later on.

Only the ERA5 flux maps for 2020 and 2021 have been regridded. For these exercises, please select only the 2020 or 2021 ERA5 flux map or the code won't work.

Also, the option to use ATC database data is not active here, only data from the CP will be used.

Go to <u>https://exploredata.icos-cp.eu</u>, enter a chosen login and the password 'francis'.

Go to "project_jupyter_notebooks/radon_tracer_method_for_ghg/"



If it's empty, you have to upload the zip file that was sent to you via a link. Then click on Python3 ipykernel. Unzip the zip file by typing in the open cell: !unzip radon_tracer_method_for_ghg.zip then run the cell with Shift+Enter

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Once the files installed, open RTM_widget.ipynb. Run the three cells. You should only have to run the second cell once during the session.

If you think the code is frozen, you can go to Kernel and choose "Restart kernel and clean all outputs" then rerun the first and third cells.

An interface will appear that allows to calculate greenhouse gas fluxes using the RTM.



In case of bugs, a log opens at the bottom of the page and you can check what is wrong.





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Checking the log file

For the first exercise, you will run the code for periods with missing data and the wrong year for the flux map to demonstrate the use of the log file.

- 1- Run 01-01-2021 to 03-01-2021 for BIR075 using the 2020 ERA5 flux map. Choose 'No' for the plotting option.
- 2- Run 20-03-2020 to 25-03-2020 for OPE120 and the CO species, with the ERA5 2020 flux map. Choose 'No' for the plotting option.

Open RTM.log file. To see updates in the log file, you need to close it and reopen it.

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Different messages are logged to help you follow the process, in particular in case you end up with an empty flux file. It is good practice to check the log at the end of a run.





Getting a flux file

Run data for a few days in 2021 for SAC100, TRN180 or OPE120 for CH4 using the 2021 ERA5 map. Click "yes" for plotting the data.

At the end of the run, new dialog open asking to select a file to plot. Click "select", then the file you want to plot, then "select" again and then "Plot". A pdf file is produced in the folder "Plots" with the footprints of each day of the period combined with the GHG flux.









Check in RTM.log file that you have lines with 'Flux calculated'.

Open the flux file in outputData. All the file names begin with RTMflux_then the species, the site, the name of the radon flux map and the dates.

In the file, you have 10 columns without header.

The columns are: 'Date', "Jghg", "R2", "Error", "Nb_pts", "Nb_hrs", "Rise_stop", "Rn_amplitude", "JRn", "Type".

- Jghg is the flux calculated for the species you have chosen
- R2 is the R2 of the correlation between RN and GHG data
- Error is the error on the fit
- Nb_pts is the number of data points used for the fit
- Nb_hrs is the number of hours used in the fit, for hourly data
- Rise_stop is the difference between the radon maximum and the last data point of the period. If it is positive, the radon stopped increasing before the end of the period showing it is a local/regional event, if it's negative, it may be a larger synoptic event and the flux will not be representative.
- Rn_amplitude is the difference between the Rn maximum and minimum over the window
- JRn is the radon flux used to do the calculation.
- Type indicates the way the Rn flux was calculated:
 - UserValue: the value entered in the dialog window
 - StationPixel: the value for the station pixel for the chosen flux map





- 2019Aggreg: the combination of the chosen flux map and the aggregated 2019 footprints
- DayFootprint: the combination of the chosen flux map and the footprint of the studied period

Plotting the data

To visualize the data, you can open Plot.ipnb. This is not an interface, just python code already ready. Read the comments to modify the code to plot what you want.

Use already compiled data, compare two Saclay files calculated with ERA5 or GLDAS flux maps.

Plots are saved in outputData.







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