

TTorch Report

*Title: **222Rn measurements: instrument comparison***

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Introduction

²²²Radon (²²²Rn), a radioactive noble gas, is used as a tracer in various applications, to test the reliability of transport models, able to reveal greenhouse gases' (GHG) sources, sinks and distribution, or to verify (e.g. national) GHG inventories (van der Laan et al., 2010). ²²²Rn is especially useful at the soil-air interface, where it undergoes the same transport processes as any other soil-derived GHG. For example, this makes ²²²Rn an ideal tracer to separate variations in soil carbon dioxide (CO₂) production from changes in the soil-atmosphere CO₂ transport, both being reflected in the total soil-atmosphere CO₂-flux. In this respect a wide knowledge on the ²²²Rn exhalations and their spatial and temporal variability is a prerequisite. Efforts have been made to predict this flux rate with widely available γ -dosimetry measurements (Szegevary et al., 2007), in order to produce European maps, but real ²²²Rn-flux measurements are still sparse, therefore needed. Since 2006, measurements of the soil ²²²Rn and CO₂ concentration (through soil probes) as well as the ²²²Rn and CO₂ soil fluxes (by means of an automatic soil chamber) are being carried out at Lutjewad station. In view of a broadening of the survey area, we are planning future campaigns, on different soil conditions in southern Italy. Therefore, the need of extensively testing and comparing the performance of the instrument to be brought back and used in Italy, RADIM-5, over the ²²²Rn monitor routinely used at Lutjewad. In particular, we need to check if RADIM-5 is sensitive at low concentrations of ²²²Rn and if the results of two systems agree well with each other. This present project is an essential step on the way to a unified measurement-based European ²²²Rn soil flux database. The two ²²²Rn detectors have to run continuously together. Once enough data have been collected, the instrument will be removed and we will convey again to analyze and to discuss the data acquired and, eventually, agree on further prospective of this collaboration.

Lutjewad sampling station and ²²²Rn monitors

The Lutjewad atmospheric measurement station is equipped with a 60 m tall metal frame tower with air intakes at 7 m, 40 m and 60 m above ground continuously monitoring most important anthropogenic long lived greenhouse gases CO₂, CH₄ and N₂O and also other tracers like CO, SF₆ and ²²²Rn. ²²²Rn is continuously being measured in the atmosphere using ANSTO ²²²Rn monitor which samples air from 60 m from the tower. ²²²Rn is also being measured in soil by deploying probes at depth of 27 cm into the soil and also ²²²Rn emanation rate from the soil into the atmosphere is carried out using the well known flow-through accumulator technique. Besides ²²²Rn, continuous measurement

of CO₂ is also performed at soil-atmosphere interface using Vaisala CO₂ detectors. Figure 1 shows the complete experimentation setup of the CO₂ and ²²²Rn in soil and also flux measurement.

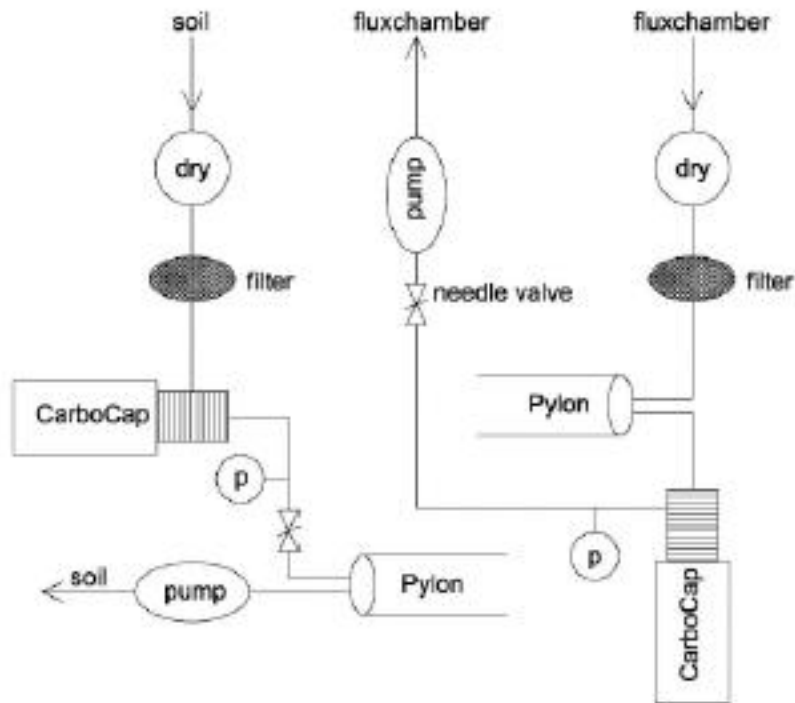


Figure 1 Lutjewad soil air sampling system.

An automated soil flux chamber is placed on the measurement site (mainly clay). It covers a surface area of ≈ 0.031 m² and its volume is ≈ 6.3 liter. If closed, air from the chamber is circulated through the ²²²Rn detector and a CO₂ detector (Vaisala CarboCap), after drying and filtering to remove ²²²Rn progeny.

The flow chamber is closed for 4 hours twice a day. During this time the increase of the CO₂ concentration in the chamber is measured. After 4 hours the pump stops and the flow chamber is opened for 8 hours to expose the soil inside to all meteorological influences. During these 8 hours the air in the detector is measured in 30 min. intervals. This considerably decreases the uncertainty of the ²²²Rn end-concentration measurement.

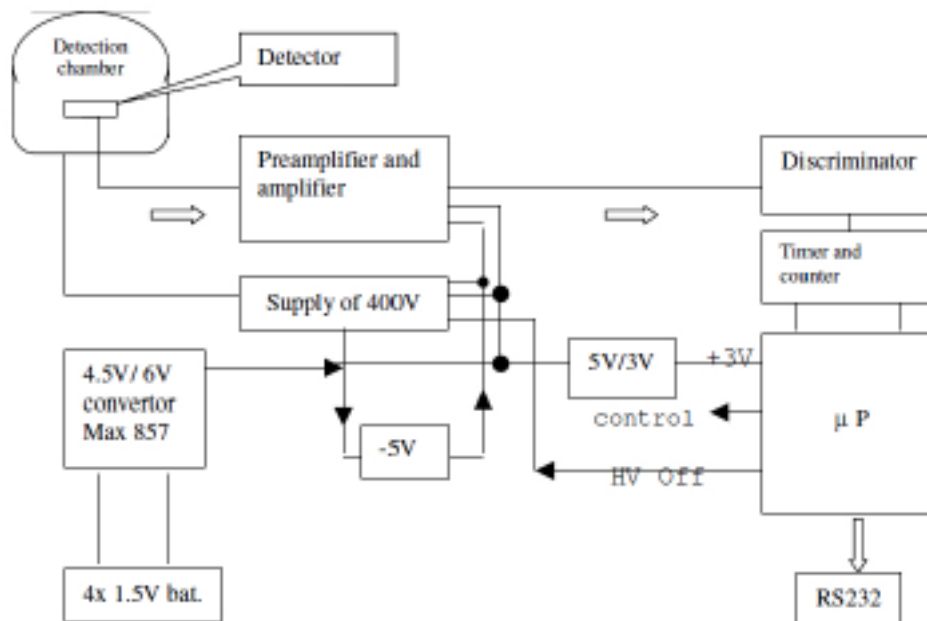
RADIM-5. Radon detector.

RADIM-5 is a commercial radon detector (www.jiriplch-smm.com). The concentration of radon is determined by measuring the α -activity of the decay products (daughters) of Radon, ²¹⁸Po (RaA) and ²¹⁴Po (RaC'), collected from the detection chamber on the surface of a semiconductor detector by an electric field. Half of the hemispherical chamber consists of a grid, covered with two layers of material. The material captures the radon products formed in the external space and protects the detector against light and dust. The dimensions of the RADIM-5 detection chamber is 140 ml. As the chamber dimensions are smaller, a source of 400 V is used in RADIM-5. Figure 2 shows the picture of RADIM-5 radon detector and also the block scheme of the electronics inside the detector box. According to the manufacturer RADIM-5

has the highest sensitivity and is least affected by the humidity among all other commercially available detectors, since the shape of the detection chamber, with a very effective electric lens, was designed by computer modelling and there is a special feature to reduce the effect of "electro-negative" ions.



(a)



(b)

Figure 2

Description of the work carried out during the visit

When first planned, this short visit aimed to install at Lutjewad a home-made ^{222}Rn detector on the active soil air sampling system. Unfortunately, some unsolved bugs of the acquisition system prevented us from using this detector. We opted, therefore, for testing RADIM-5, a commercial detector we had available, which was originally meant for indoor air quality monitoring.

During the visit we basically proceeded in two steps:

1. Laboratory tests. Testing the instrument itself and the data acquisition in laboratory conditions.
2. "In situ" installation and tests. Implementing a way to put RADIM-5 in place, minimising the interferences with the routine performance of the present system, and, finally, setting RADIM-5 monitor and letting it run besides the present working system, equipped with PYLON AB-5 ^{222}Rn detectors.

1. Laboratory tests

A radium source with a known activity (94.9 Bq) was placed inside an airtight container of volume 0.0039 m³, RADIM-5 was also placed inside the metal box and closed tightly, such that there are no leaks. Activity concentration or growth of radon inside the metal box can be calculated theoretically using the equation:

$$\left(\frac{A_t}{V}\right) = \frac{(1 - e^{-(1-\lambda_{Rn}t)})A_{Ra}}{V}$$

where:

A_t is the activity of radon at any instant (Bq);

V is the volume of the box (0.0039 m³);

A_{Ra} is the radium source activity (94.9 Bq);

λ_{Rn} is the decay constant of radon (h^{-1}).

Figure 3 shows the comparison of activity concentration calculated theoretically and also measured using RADIM-5. The values measured by RADIM-5 are slightly higher than the theoretical ones. This could be due to the uncertainty in the volume determination. The metal box was calculated to have a volume of 0.0039 m³ while the RADIM-5 detector itself occupies some volume, hence the total volume including the RADIM-5 detector should be less than 0.0039 m³. Considering this, a shaded area for the theoretical expectations has been drawn in Figure 4. Here the lower limit has been derived using the

actual volume of the metal box (0.0039 m^3), as the theoretical curve in figure 3, while the upper curve of the shaded area is obtained assuming that RADIM-5 detector a full body, occupying 0.0004 m^3 (total volume in this case would be 0.0035 m^3). From Figure 4, we see that the measured value fall in between the shaded area from which we can say that both the theoretical and measured values almost agree well with each other, i.e. the response from the RADIM-5 detector is reasonably accurate.

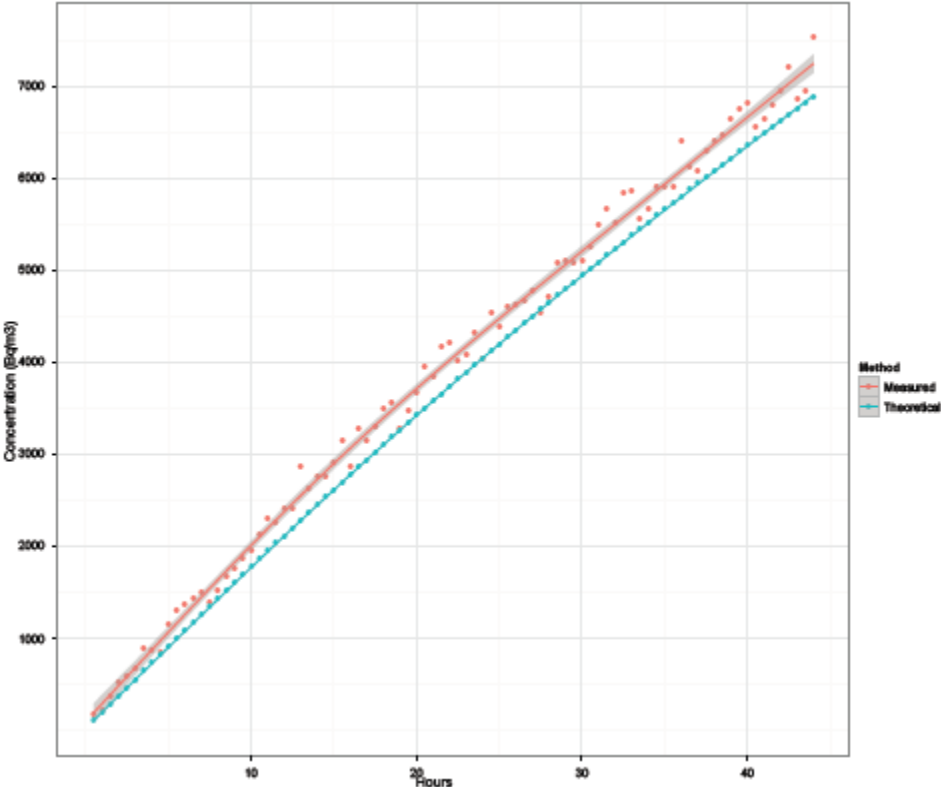


Figure 3

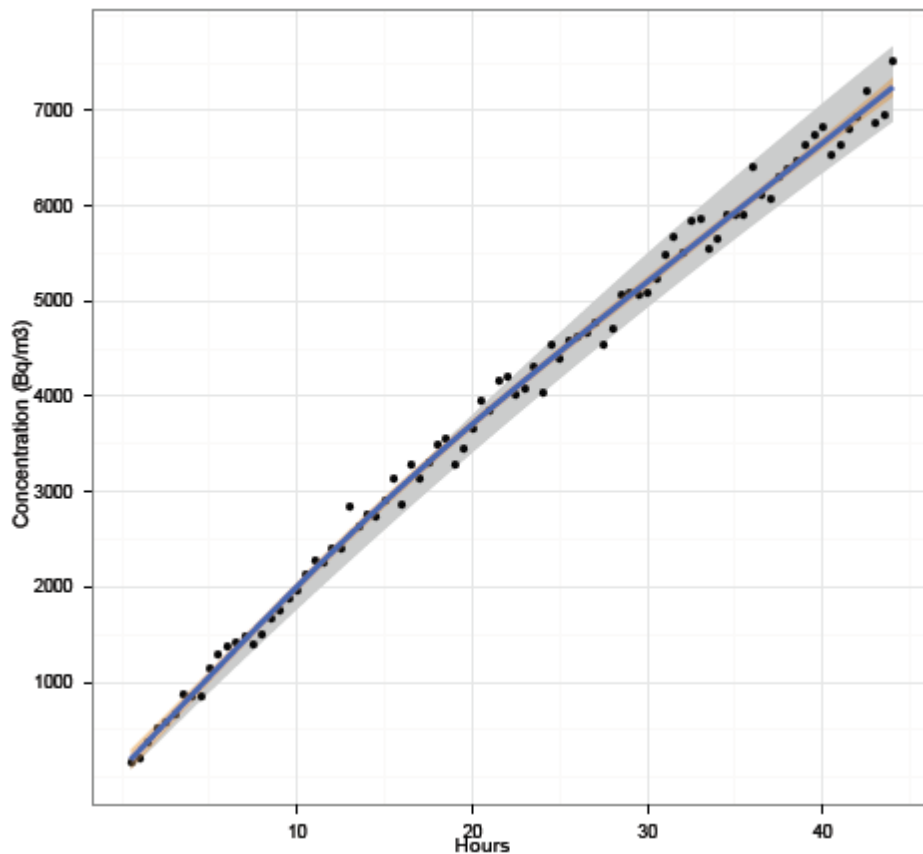
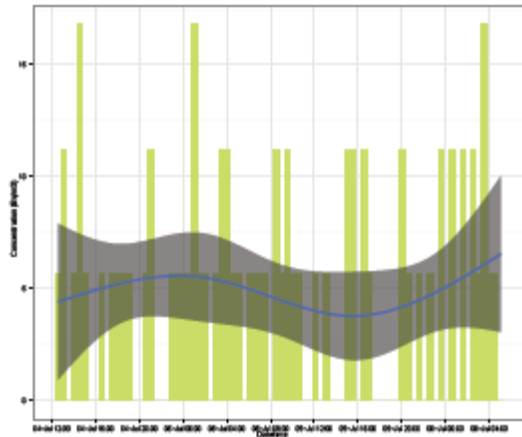
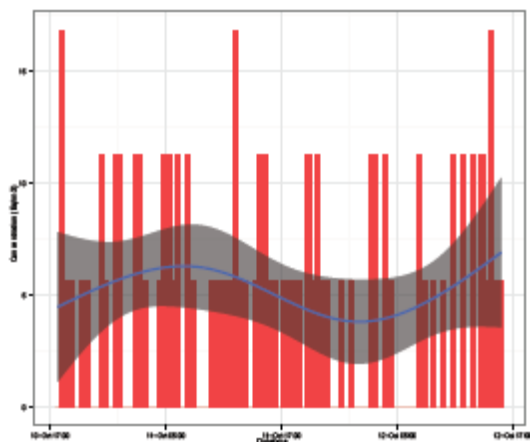


Figure 4

Before starting any experiments, it is important that we know the response of the detector itself in a radon free environment. This is useful for determining the response by the instrument itself. RADIM-5 detector was placed inside a metal box filled with N_2 and the background was measured twice. Figure 5 shows the background measurement done using the RADIM-5 detector and in both the cases the background was low and almost the same. These background measurements show that RADIM-5 detector can indeed be used as an alternative over conventional alpha detectors whose background counts increase upon time and, at some point in time, the detectors have to be replaced by new ones.



(a)



(b)

Figure 5

2. “In situ” installation

After background measurement and testing with known radium source, RADIM-5 was taken to Lutjewad and, as a preliminary measurement phase, RADIM-5 was placed inside a cylindrical container as shown in Figure 6 and the radon exhalation from the soil into the cylindrical head space was measured. Because of heavy rainfall during that time, the detector had to be taken out after seven days post deployment to prevent any damage to the electronics inside the system.

As final step RADIM-5 had to be on Lutjewad system besides the Pylon detector, for comparison measurements with the present radon system. RADIM-5 was put in a glass container and initially placed on the over-pressure side of the pump (Figure 7 a) and was allowed to measure simultaneously with the CO₂ and radon measurement system.

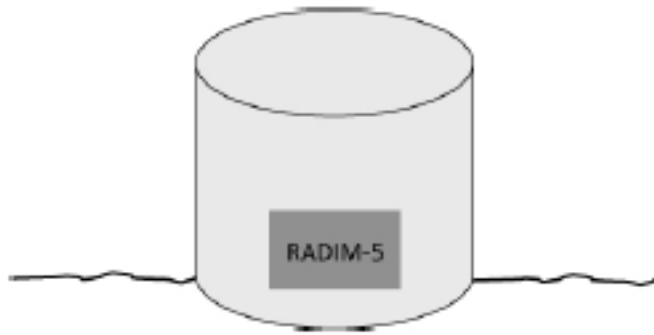


Figure 6

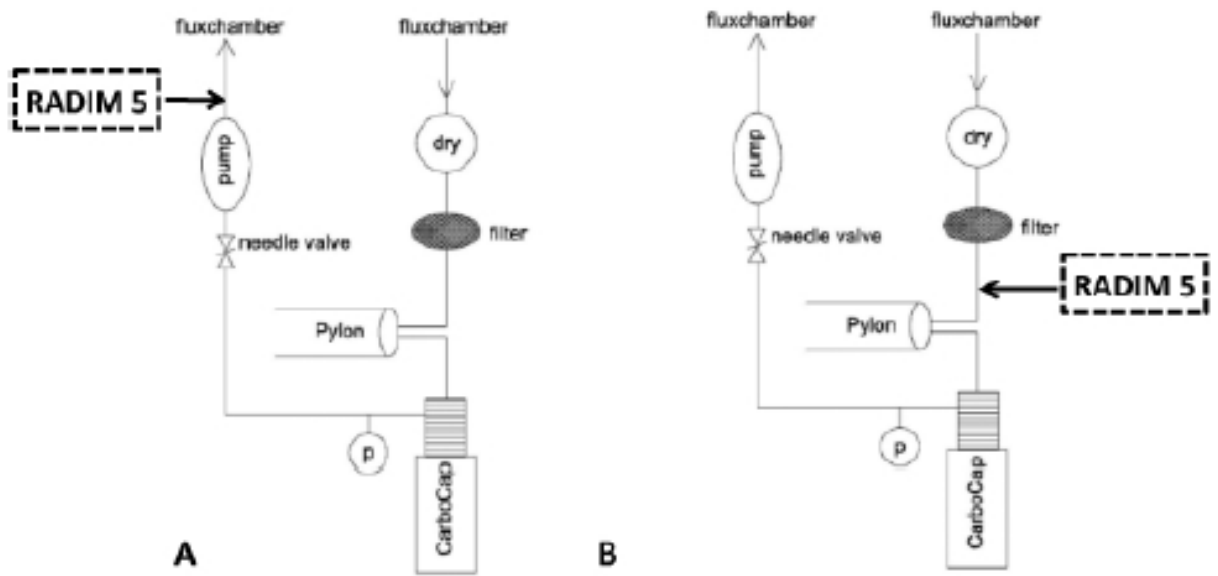


Figure 7

As we saw higher values than normal of radon and also CO₂ concentration, we speculated that this could be due to the big volume of the glass container we used to isolate and protect the RADIM-5 detector. Therefore, we concluded that the pump was not able to compensate the extra volume thereby, creating a low pressure in the system causing to suck air from the soil and hence higher values of CO₂ and radon. Hence, it was decided to reduce the volume of the container the RADIM-5 detector was put in and redo the measurement again. Now, the detector was placed in a smaller volume and again on the pushing end of the pump and measurement was performed again. As the situation improved a bit but not completely, after careful look at the results, it was decided to keep the RADIM-5 detector on the other end of the experiment setup, under pressure side or the sucking side (Figure 7 B). The system has been left in place to collect enough data for further analysis and comparison.

Conclusion and future work

In view of construction of a unified measurement-based European ²²²Rn soil flux database, two ²²²Rn detectors have been kept running together, one adapted to perform measurements on ²²²Rn soil flux, compared over another routinely run at Lutjewad for the same purpose. At this stage we have accomplished the laboratory testing stage, verifying that Radim-5 can indeed perform accurate ²²²Rn soil exhalation measurements. Furthermore, we have deployed the instrument in situ on a proper manner, in order to collect enough information on the comparison of our ²²²Rn monitor a the Pylon running at Lutjewad . Data analysis and drawing conclusion will be carried in a coming visit, when hopefully a positive response of the test can be the basis of future collaborations.

Szegvary T, Leuenberger MC, Conen F. Predicting terrestrial ²²²Rn flux using gamma dose rate as a proxy. Atmos. Chem. Phys. Discuss. 2007; 7: 1877-1892.

van der Laan S, Karstens U, Neubert REM, van Der Laan-Luijkx IT, Meijer HAJ. Observation-based estimates of fossil fuel-derived CO₂ emissions in the Netherlands using Δ¹⁴C, CO and ²²²Radon. Tellus B 2010; 62: 389-402.