Radon-222 flux measurements in Egham, Great Britain

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Introduction and motivation

The radioactive noble gas radon-222 can be used as tracer for various processes in the soil air and atmosphere. Radon-222 originates from the decay of radium-226 in the soil grains. It can be used for the parameterisation of diffusive fluxes of gases in the soil air. Moreover, atmospheric radon concentration measurements are used for the investigation of the change of the atmospheric boundary layer height and for estimating the residence time of air masses over continents.

For the use of radon as a tracer, the radon flux out of the soil must be known accurately. The large spatial heterogeneity of soils causes large spatial variations of radon fluxes. Moreover, changing soil moisture contents lead to temporal variations of the radon exhalation rate (the radon flux from the soil to the atmosphere). However, until now many models dealing with atmospheric radon concentration used a simple approach with a constant radon flux term [1, 2] or a northwards decreasing radon flux [2, 3]. In order to obtain better estimations of the radon exhalation rate, different radon flux maps for Europe were published in the last years. Szegvary et al. [4] published a radon flux map, which is based on an empirical relation between the radon flux and the γ-dose rate. Lopez-Coto et al. [5] presented a flux map that uses numerical modelling of radon transport in soils to estimate the radon exhalation rate.

Another approach for creating a radon flux map is currently developed by Karstens et al. [6]. This map uses radium concentration, grain size distribution, porosity and soil moisture content of the various soils to calculate the radon exhalation rate. In order to have validation data for this map, measurements of radon flux, soil properties and soil moisture were carried out at various locations. At our test site near Heidelberg, the temporal evolution of the radon flux, which is due to the changing soil moisture content, is investigated. In addition, measurements at different sites in Europe were carried out in order to have more information about the spatial radon flux variations. To have a direct benefit from these measurement campaigns, locations where the atmospheric radon concentration is continuously measured were chosen. In this way, the results of the measurements can be used for a better understanding of the measured atmospheric radon concentrations.

Scientific objectives

Direct radon flux measurements give important information for the interpretation of atmospheric radon concentration measurements. In Egham, a radon monitor for measuring the atmospheric radon concentration is running since almost one decade. However, no radon exhalation rates were measured in Egham up to now (pers. comm. with Rebecca Fisher). Thus, the radon flux measurements performed in Egham in September 2013 gave very important new information for a better interpretation of the atmospheric radon concentration measurements. Moreover, the measurements performed in Egham were used for the validation of the radon flux map of Karstens et al. [6].

Reason for choosing station

In Egham, continuous atmospheric radon concentration measurements are performed. Up to now, no radon exhalation rates were measured in Egham. The radon flux measurements carried out in September 2013 in Egham gave thus important new information for the interpretation of the measured atmospheric radon concentrations.

Method and experimental set-up

The radon exhalation rate can be measured directly by the so called accumulator method. Moreover, from measurements of soil properties and soil moisture, the radon exhalation rate can be calculated using a simple transport model of radon in the soil air [7]. In order to obtain all necessary parameters, different types of measurements were performed at three locations in Egham:

1. Radon flux measurements:
   For the radon flux measurements, two quadratic metallic frames (length: 50 cm, height: 25 cm) are used. These two frames are put ca. 5-10 cm deep in the soil. For sampling, the frames are closed with a cover. Radon diffuses from the soil into the "chamber" (frames + cover) and the radon concentration increases. After half an hour an air sample is taken from the chamber. It is analysed for its radon concentration in the laboratory in Heidelberg.
   From the radon concentration, the sampling time and the height of the "chamber" the radon flux density can be calculated.

2. Radium concentration measurements:
   A soil sample is taken with a core cutter and divided into pieces of 20 to 30 cm depth intervals. Afterwards it is dried in the laboratory and analysed for its radium content using gamma spectroscopy.

3. Soil moisture content, dry bulk density and porosity:
   A soil sample is taken with a core cutter with known volume. It is weighted and then dried in an oven. After drying, it gets weighted again. From the mass difference, the moisture content of the soil can be calculated. Furthermore, the porosity and the dry bulk density of the soil are obtained from the volume of the soil sampler and by measuring the grain density of the soil sample.

4. Grain size distribution:
   A representative soil piece is analysed for its grain size distribution. After destroying the organic carbon and removing the calcium carbonate, the soil sample is dried in an oven. With sieves with different sizes the single grain fractions are separated.

The measured soil properties and soil moisture content can be used for calculating the radon exhalation rate. Dörr [7] reports an equation for calculating the radon exhalation rate *j*:

$$\mathrm{j} = \mathrm{c_{Ra}}\, \rho_\mathrm{b}\, \varepsilon \sqrt{\mathrm{P}\, \lambda} \qquad (1)$$

with $\mathrm{c_{Ra}}$ the concentration of ^{226}Ra in the soil material, ρ_b the dry bulk density of the soil, ε the emanation factor, i.e. the fraction of produced radon that can leave the soil grains and reach the air in the pore space, P the permeability of the soil and $\lambda = 2.1 \cdot 10^{-6}\ \mathrm{s}^{-1}$ the decay constant of ^{222}Rn.

The permeability of the soil is calculated using the parameterisation of Millington and Quirk [8]:

$$P = \mathrm{D}_0 \frac{(\Phi - \theta_\mathrm{w})^2}{\Phi^{\frac{2}{3}}} \tag{2}$$

where Φ is the porosity of the soil, θ_w is the soil moisture content and $\mathrm{D}_0 = 1.1 \cdot 10^{-5}\ \mathrm{m}^2/s$ is the diffusion coefficient of radon in air. Hence, the radon exhalation rate can be calculated from the measured soil properties using Eq. (1) and Eq. (2).

Preliminary results and conclusions

Measurements were performed at three different locations on the campus of the Royal Holloway University in Egham. The results of the measured soil properties, soil moisture, the measured radon fluxes and the calculated radon fluxes are listed in Table 1. The measured and calculated radon fluxes are in good agreement with each other at the sites “grassland 1” and “forest”. At the site “grassland 2” the calculated radon flux is much higher than the measured one. It can be noticed that the measured soil moisture at site “grassland 2” is much lower than the soil moisture measured at the other two sites. The site “grassland 2” was located under a tree. Probably, the heterogeneous rainfall through the leaves of the tree caused varying soil moisture contents under the tree. Since the radon exhalation rate and the soil moisture content of the soil were not measured exactly on the same place, but in a distance of about 1 meter, this might cause different results for the measured and calculated radon exhalation rates.

Table 1: Overview of the measured soil properties, soil moisture and the measured and calculated radon fluxes at the three different sampling sites. The calculated radon fluxes are obtained from the measured soil properties and soil moisture using Eq. (1) and Eq. (2).

	Grassland 1	Grassland 2	Forest
Radium activity concentration / Bq/kg	13.7 ± 1.3	14.2 ± 1.0	28.8 ± 1.5
Dry bulk density / kg/m^3	1.23 ± 0.07	1.30 ± 0.02	1.11 ± 0.01
Emanation factor	0.323 ± 0.014	0.321 ± 0.013	0.375 ± 0.018
Porosity	0.51 ± 0.03	0.49 ± 0.02	0.56 ± 0.02
Soil moisture	0.20 ± 0.01	0.08 ± 0.01	0.20 ± 0.01
Measured Rn flux / mBq/(m^{2}s)	9.6 ± 1.6	5.9 ± 1.0	24.9 ± 7.0
Calculated Rn flux / mBq/(m^{2}s)	10.3 ± 1.3	14.7 ± 1.3	24.9 ± 2.0

The radon flux map of Karstens et al. [6] reports a radon flux of about 18 Bq/(m^{2}s) for September in Egham. The radon fluxes measured on the grassland sites are lower than the radon flux predicted by the radon flux map, whereas the radon flux measured at the forest site is higher.

Outcome and future studies

The measured and calculated radon exhalation rates are in agreement with the fluxes of the radon flux map of Karstens et al. [6]. Moreover, the measured and calculated radon exhalation rates are in good agreement at the sites “grassland 1” and “forest”. At the site “grassland 2”, the deviations between measured and calculated radon flux can possibly be explained by its location under a tree. This might have caused different soil moisture content at the place where the radon exhalation rate was measured with the accumulation chamber and the place, where the soil moisture was measured.

The measurement results can be used for a better analysis of the measured atmospheric radon concentrations in Egham. Since these measurements were carried out in the framework of a master thesis, no additional measurements are planned at the moment.

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