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Predicting terrestrial ^{222}Rn flux using gamma dose rate as a proxy*

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Abstract

^{222}Rn is commonly used as a natural tracer for validating climate models. To improve such models a better source term for ^{222}Rn than currently used is necessary. The aim of this work is to establish a method for mapping this source term by using a commonly measured proxy, the gamma dose rate (GDR). Automatic monitoring of GDR has been networked in 25 European countries by the Institute for Environment and Sustainability at the Joint Research Centre (JRC IES) in Ispra, Italy, using a common data format. We carried out simultaneous measurements of ^{222}Rn flux and GDR at 63 locations in Switzerland, Germany, Finland and Hungary in order to cover a wide range of GDR. Spatial variations in GDR resulted from different radionuclide concentrations in soil forming minerals. A relatively stable fraction (20%) of the total terrestrial GDR originates from the ^{238}U decay series, of which ^{222}Rn is a member. Accordingly, spatial variation in terrestrial GDR was found to describe almost 60% of the spatial variation in ^{222}Rn flux. Furthermore, temporal variation in GDR and ^{222}Rn was found to be correlated. Increasing soil moisture reduces gas diffusivity and the rate of ^{222}Rn flux but it also decreases GDR through increased shielding of photons. Prediction of ^{222}Rn flux through GDR for individual measurement points is imprecise but un-biased. Verification of larger scale prediction showed that estimates of mean ^{222}Rn fluxes were not significantly different from the measured mean values.

1 Introduction

^{222}Rn is commonly known as a hazardous radioactive (noble) gas in indoor air. Yet, ^{222}Rn is also often used as a natural tracer of air transport. Observations of atmospheric ^{222}Rn have been very useful in the evaluation of climate models simulating transport, transformation and removal processes of gases and aerosols (e.g. Rasch, 2000). Used in inverse mode, these models can provide information on location, extent and strength of sources and sinks of greenhouse gases based on the measurement of changes in their atmospheric concentrations (Chevillard, 2002; Gupta, 2004). Currently, the effective use of ^{222}Rn in this context is limited by the poor accuracy of the ^{222}Rn source function (WMO GAW report no. 155, 2004). Current practice is to assume a spatial and temporal uniform flux rate of 1 atom cm^{-2} s^{-1} from all ice-free land surfaces. Improvement of the source term was attempted by Schery and Wasiolek (1998), who created a global ^{222}Rn flux map based on porous media transport theory and calibrated with experimental radon flux data from Australia and Hawaii. It predicted regional variations of a factor of three not to be uncommon. However, current lack of detailed data on input parameters in large parts of the world results in the proposed map still being preliminary and depending on more data becoming available. Furthermore, additional flux measurements over a greater variety of conditions are needed for robust validation and eventual verification of the model. A different interpretation of the flux term was proposed by Conen and Robertson (2002), based on atmospheric profile measurements integrating over larger areas and indicating a decline in ^{222}Rn flux from ice-free land surface from 1 atom cm^{-2} s^{-1} at 30° N to 0.2 atom cm^{-2} s^{-1} at 70° N. This source term was found to improve predictions but it was speculated that ^{222}Rn flux might begin to decline well north of 30° N (Robertson et al., 2005). A more detailed source term is highly desirable to improve validation of atmospheric transport models since the quality of validation is directly proportional to the quality of the ^{222}Rn source term used.

Therefore, we are proposing a new method to describe the ^{222}Rn source term, initially focusing on the European continent. Our approach is to calibrate direct measurements of ^{222}Rn flux against terrestrial gamma dose rate (GDR). We made use of the high density of European GDR measurements, established after the nuclear reactor accident in Chernobyl in 1986, to produce a full description of the European ^{222}Rn source term.

2 Basic concept

The source of ^{222}Rn is ^{226}Ra, a member of the ^{238}U decay chain. Gamma spectroscopic analysis of soil surface samples (0-20 cm depth) in geologically diverse regions of Switzerland showed that ^{238}U contributes an almost constant proportion to the terrestrial GDR (Fig. 1a) and that ^{226}Ra activity is closely related to the ^{238}U activity (Fig. 1b). Large radioactive disequilibria of the uranium decay series have been found in the limestone Karst soils in the Jura mountains (Von Gunten et al., 1996). Selective migration of individual members of the ^{238}U decay chain could lead to an over- or underestimated GDR-based ^{222}Rn flux in such locations. However, such cases seem to be rare, as seen in the close correlations in Figs. 1a and b. Therefore, we assume that ^{222}Rn flux resulting from the decay of ^{226}Ra is directly related to terrestrial GDR. This assumption is probably a good first approximation but not entirely correct as indicated by the relatively large scatter in the ratio of ^{222}Rn flux to ^{226}Ra activity (Fig. 1c). Firstly, only part of the produced ^{222}Rn emanates into air filled pore space from where it might escape into the atmosphere and the fraction emanating may depend on grain size (Nazaroff, 1992). Secondly, differences in grain size and soil moisture modulate gas diffusivity and thus the fraction of emanated ^{222}Rn that may reach the atmosphere before decay. Thus, the proportion of ^{222}Rn produced that escapes into the atmosphere is variable and depends on factors other than ^{226}Ra content.

3 Methods

3.1 ^{222}Rn flux measurement techniques

A barely modified closed chamber method as described in Lehmann et al. (2000, 2003) was used to measure the ^{222}Rn flux. The main modification consisted in air from the chamber not being pumped through a series of two but only one alpha-decay detector (Alphaguard 2000 Pro, Genitron Instruments Frankfurt, Germany). The flow rate was

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0.5 l min^{-1}, a delay volume of 1.5 l was used to remove most of the ^{220}Rn with its half-life of 56 s (Lehmann et al., 2003 used the second detector, which was installed before the delay volume to evaluate also the ^{220}Rn flux). From there, the air passed to the detector where only ^{222}Rn was measured. The ^{222}Rn flux was estimated from the increase in ^{222}Rn activity measured in 10 min intervals over about 1.5 h. Remaining ^{220}Rn may have affected the absolute value of measured ^{222}Rn activity but not its increase over time, as ^{220}Rn concentrations reach a steady state between production and decay after about 7 min and we always rejected the first 10 min measurement interval. Due to radioactive decay of ^{222}Rn with a half-life time of 3.82 days the assumption of a linear increase of ^{222}Rn in the chamber must be corrected by a factor of +0.38 %. Two types of chambers were used: an automatically closing and opening chamber which measured autonomously the ^{222}Rn flux from soil over a longer time period. This flux chamber, a cylindrical box with a diameter of 20 cm and 25 cm height had a flap, which closed automatically 6 times a day for 1.5 hours to accumulate ^{222}Rn and was then opened for 2.5 h prior to the next measurement. A second analytical system was a manually closable chamber (a plastic box with the dimensions 35 cm×27 cm and 13 cm height) which was used for spot measurements.

Long-term measurements of ^{222}Rn fluxes were made at 7 different field sites of the Swiss Meteorological Service (MétéoSuisse). Normally, measurements took place for a duration of 3–4 weeks, except at the field site in Basel-Binningen, where continuous measurements were made over a year in order to estimate seasonal variations. Soil moisture at that location was measured with 4 TDR two-rod probes (rod length: 18 cm), connected with a multiplexer to a Tektronix 1502B (Tektronix, Inc., Wilsonville, USA). The signal was evaluated and logged with a data logger (CR10, Campbell Scientific, Inc., USA).

The manually closing chamber was used for in situ measurements of ^{222}Rn flux at 29 sites in Switzerland and South-West Germany, at 8 sites each in Southern (Helsinki region) and Northern Finland (Rovaniemi region) and at 12 sites in Hungary. Supplementary data from Scotland (Robertson, 2005) was included. These measurements (n=9) were done with the same analytical ^{222}Rn system. The difficulty of spot measurements of ^{222}Rn flux and GDR is to get representative values for the specific location. Especially precipitation has been found to have significant effects on GDR because of the deposition of Rn daughters associated with aerosols, but also on short-term variations in ^{222}Rn flux. Therefore, we avoided spot measurements during or immediately (4–8 h) after precipitation events. Additionally we studied on small scale spatial variability in a woodland in Basel (Lange Erlen) using a nested sampling design with lag distances of 0.5 m, 5 m and 50 m.

3.2 Gamma dose rate

An autonomous gamma probe (Gammatracer, Genitron Instruments Frankfurt, Germany) for continuous surveillance of the environmental gamma radiation was used for measuring GDR (10 H*). The gamma probe was placed 1 m above ground during the measurement. Since most of the measurements took place at locations of the national gamma monitoring networks, where GDR is continuously measured, the gamma probe was used as a reference probe. This allowed inter-comparison of different probes at the network sites. The terrestrial component of the gamma dose rate was obtained by subtracting the cosmic part (which depends on altitude above sea level and can be calculated) from the measured total GDR (Murith and Gurtner, 1994). A correction was made for the artificial radiation, which is mainly derived from ^{137}Cs from the Chernobyl powerplant accident in 1986, based on the "Atlas of Caesium deposition on Europe after the Chernobyl accident" (De Cort et al, 1998).

4 Results and discussion

4.1 Correlation of ^{222}Rn flux and terrestrial GDR at different locations

There is a linear relationship between ^{222}Rn flux and terrestrial GDR (Fig. 2), though the effect of heteroscedasticity is observed, i.e. the variability described by standard deviation depends on the mean value. This means high GDR values are associated with higher variability (an effect, which is often observed in nature). The measured data covers a range from almost 0 to 200 nSv h^{-1} respectively 0 to 250 Bq m^{-2} h^{-1}. Most soils in Europe have gamma dose rates between about 40 to 140 nSv h^{-1} well within this range. Very low GDR (~40 nSv/h and a ^{222}Rn flux less than 15 Bq m^{-2} h^{-1}) can be found at locations which have either a high water content and/or low or no mineral content like peat soils. Overall, almost 60% of the variation in ^{222}Rn flux can be described by the spatial variation of terrestrial GDR.

Still, there is a lot of variation, which may also be caused by the gamma probe and the ^{222}Rn measurement chamber integrating over different soil volumes. The measurement of GDR is mostly influenced by the variability of radionuclides and soil moisture near the soil surface (0 to 0.1 m) within a radius of about 10 m around its location. In contrast, measured ^{222}Rn flux is mostly influenced by ^{226}Ra content and soil moisture in 0 to 1 m soil depth but a three to four orders of magnitude smaller area. Thus, inhomogeneities in radionuclide and moisture distribution on this scale will affect both parameters to a different extent. The scatter in Fig. 2 is unlikely to be caused by short-term fluctuations in either parameter. Not only the short-term measurements (triangles) show the scattering effect, but also the long-term measurements (circles), which would smoothen out such short term effects. The nested sampling near Basel revealed that the coefficient of variation between measurements separated by a distance of 0.5 m was 19%, increasing to 21% and 36% for 5.0 and 50.0 m distances, respectively. The large coefficient of variation at the smallest distance may to a large part be caused by the error in our ^{222}Rn measurement, which we estimate to be around ±15% of the mean. For atmospheric tracer applications, regional information on the ^{222}Rn flux is

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required. The variability in the correlation between GDR and ^{222}Rn flux, which can be found on the local scale, seem to counter balance on the regional scale, as discussed later (see Sect. 4.4).

4.2 Correlation of ^{222}Rn flux and terrestrial GDR over time

Temporal variations in ^{222}Rn flux can be observed in GDR at the long-term measurement in Basel (Fig. 3) during the period from June to November 2006, where soil moisture and precipitation was also measured. At the beginning of July a prolonged dry period began without nearly any precipitation and soil moisture decreased almost constantly. During this period the ^{222}Rn flux was observed to increase by about 100% until the beginning of August. Simultaneously, GDR increased from 82 nSv/h to 98 nSv/h, which is nearly 20%. Decreasing soil moisture increases the air filled pore volume and with it the diffusivity of soil. Therefore, ^{222}Rn flux is larger when soils are dry and less ^{222}Rn decays before it may reach the soil surface (Grasty, 1997). At the same time, low soil moisture leads to reduced shielding of gamma-rays and a larger proportion of them can be detected in the atmosphere above the ground. Diurnal changes in the amplitude of GDR during periods without precipitation are supposed to be influenced by changes in Rn and Rn-progeny concentrations in the near surface air, where they accumulate during atmospherically stable conditions at night (Greenfield, 2002, 2003).

At end of September through the beginning of October three intense rain events were recorded (Fig. 3). These were days within a period of otherwise stable weather conditions, where during a short time period between 60 mm and 80 mm of rain fell, approximately the same amount for all three rain events. After each of the three events, the ^{222}Rn flux decreased immediately with the beginning of precipitation, probably because of the wet soil surface severely inhibiting ^{222}Rn diffusion into the atmosphere. The reaction of GDR was initially to the contrary. It suddenly increased after the first rain event from 85 nSv/h to 110 nSv/h, an increase of 29%. This effect is caused by outwash of particles from the lower atmosphere, carrying previously absorbed ^{222}Rn progeny back to the soil surface (Greenfield, 2002, 2003). The cummulative half-life of the short-lived ^{222}Rn progeny is about 50 min. Thus, the GDR decreased within a few hours once rain had stopped and was lower than it was before the rain event (~8–10%). The second and third rain event showed the same effect. The only difference between the three rain events was the amplitude of the peak at the start of each rain fall, which was smaller for the second and third compared to the first one because the atmosphere was getting increasingly cleaner of particles carrying ^{222}Rn progeny.

4.3 Factors affecting ^{222}Rn flux but not GDR

Our analysis of the correlation between ^{222}Rn and terrestrial GDR showed that both parameters are affected similarly by the radionuclide content of the soil and by soil moisture. However, there are also factors affecting ^{222}Rn flux without having a similar effect on GDR which we have not evaluated so far. Total pore space and tortuosity are important variables that affect ^{222}Rn flux (Nazaroff, 1992) but not GDR. A larger proportion of ^{222}Rn produced within the soil profile will escape to the atmosphere from coarse grained soils with a large total pore volume than from compacted fine grained soils, whereas the escape of gamma rays is unlikely to be affected by this. There already exist models for ^{222}Rn flux prediction based on geological and pedological factors, but such models require numerous parameters which are not well known due to the complicated interactions between different geological and pedological units influencing the ^{222}Rn flux (Ielsch et al., 2002). Temperature differences between air and soil have also been found to be a factor influencing ^{222}Rn flux (Nazaroff, 1992), which is driven by diffusion and possibly mass flow.

4.4 Verification on a regional scale

As mentioned in the introduction, our interest in describing the ^{222}Rn flux term is because of its application in the validation of atmospheric transport models. We therefore would like to be able to correctly predict regional averages of ^{222}Rn flux. To test our approach of using GDR as a proxy, we split our data in one part to produce the correlation function between ^{222}Rn flux and GDR (Switzerland, Germany, Scotland) and another part to verify the correlation (N- and S-Finland, Hungary). The correlation function derived was: $y = 0.995x–14.97$ (r^2=0.66), where y is the ^{222}Rn flux in Bq m^{-2} h^{-1} and x is the GDR in nSv h^{-1}. The measured regional means differed by a factor of up to 3, as considered not to be uncommon by Schery and Wasiolek (1998). Still, predicted means were within the error margin of the respective measured mean (Table 1).

5 Conclusions

Most of the spatial variation in ^{222}Rn flux may be explained by the variation in radionuclide activity in soils derived from different parent material. Soil moisture has been shown to have similar effects on ^{222}Rn flux as it has on GDR, except for short time periods during precipitation events. Considering additional parameters besides GDR, e.g. soil type, might further improve the prediction of ^{222}Rn fluxes on the small scale. However, it may also unnecessarily complicate prediction, especially if we are going to extend it to areas where required data may not be available. To predict average regional ^{222}Rn flux, the empirical correlation with GDR seems to suffice to produce regional means of ^{222}Rn flux within the error margin of measurements.

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References

Conen, F. and Robertson, L. B.: Latitudinal distribution of radon-222 flux from continents, Tellus, 54B, 127–133, 2002.

Chevillard, A., Ciais, P., Karstens, U., Heimann, M., Schmidt, M., Levin, I., Jacob. D., Podzun, R., Kazan, V., Sartorius, H., and Weingartner, E.: Transport of ^{222}Rn using the regional model REMO: a detailed comparison with measurements over Europe, Tellus, 54B, 850–871, 2002.
De Cort, M. and de Vries, G.: The European Union Radiological Data Exchange Platform: Reference Manual and European Automatic Monitoring Systems. EUR 16415 EN, EC.
Office for Official Publications of the European Communities, Luxembourg.
EUR (2005). Automatic mapping algorithms for routine and emergency monitoring data. EUR 21595 EN, EC, edited by: Dubois, G., Office for Official Publications of the European Communities, Luxembourg, 1996.
De Cort, M., Dubois, G., Fridman, Sh. D., Germenchuk, M. G., Izrael, Yu. A., Janssens, A., Jones, A. R.,Kelly, G. N., Kvasnikova, E. V., Matveenko, I. I., Nazarov, I. M., Pokumeiko, Yu. M., Sitak, V. A., Stukin, E. D., Tabachnyi, L. Ya., Tsaturov, Yu. S., and Avdyushin, S. I.: Atlas of caesium deposition on Europe after the Chernobyl accident, EUR Report 16733, EC, Office for Official Publications of the European Communities, Luxembourg, 1998.
Grasty, R. L.: Radon emanation and soil moisture effects on airborne gamma-ray Measurements, Soc. Geophys., 62(5), 1379–1385, 1997.
Greenfield, M. B., Domondon, A. T., Okamoto, N., and Watanabe, I.: Variation in γ-ray count rates as a monitor of precipitation rates, radon concentrations, and tectonic activity, J. Appl. Phys., 91(3), 1628–1633, 2002.
Greenfield, M. B., Domondon, A., Tsuchiya, S., and Tomiyama, G.: Monitoring precipitation and lightning via changes in atmospheric gamma radiation, application of accelerators in research and industry: 17th Int'l. conference, American Institute of Physics, 2003.
Gupta ML, Douglass AR, Kawa R, Pawson S.: Use of radon for evaluation of atmospheric transport models: sensitivity to emissions, Tellus Ser. B-Chem. Phys. Meteorol., 56(5), 404–412, 2004.
Ielsch, G., Ferry, C., Tymen, G., and Robe, M.-C.: Study of a predictive methodology for quantification and mapping of the radon-222 exhalation rate, J. Environ. Radioactivity, 63, 15–33, 2002.
Lebedyte, M., Butkus, D., and Morkunas, G.: Variations of the ambient dose equivalent rate in the ground level air, J. Environ. Radioactivity, 64, 45–57, 2002.
Lehmann, B. E., Lehmann, M., Neftel, A., and Tarakanov, S. V.: Radon-222 monitoring of soil diffusivity, Geophys. Res. Lett., 27, 3917–3920, 2000.

Lehmann, B. E., Ihly, B., Salzmann, S., Conen, F., and Simon, E.: An automatic static chamber for continuous ^{220}Rn and ^{222}Rn flux measurements from soil, Radiation Measurement, 38, 43–50, 2003.

Murith, C. and Gurtner, A.: Mesures in situ et irradiation externe, in: BAG, Environmental radioactivity and radiation exposure in Switzerland 1993, Swiss Federal Office of Public Health, Berne, 1994.

Nazaroff, W.: Radon transport from soil to air, Rev. Geophys., 30, 2/May, 137–160, 1992.

Rasch, P. J., Feichter, J., Law, K., Mahowald, N., Penner, J., Benkovitz, C., Genthon, C., Giannakopoulos, C., Kasibhatla, P., Koch, D., Levy, H., Maki, T., Prather, M., Roberts, D. L., Roelofs, G.-J., Stevenson, D., Stockwell, Z., Taguchi, S., Kritz M., Chipperfield, M., Baldocchi, D., McMurry, P., Barrie, L., Balkanski, Y., Chatfield, R., Kjellström, E., Lawrence, M., Lee, H. N., Lelieveld, J., Noone, K. J., Seinfeld, J., Stenchikov, G., Schwartz, S., Walcek, C., and Williamson, D.: A comparison of scavenging and deposition processes in global models: results from the WCRP Cambridge Workshop of 1995, Tellus, 52B, 1025–1056, 2000.

Robertson, B. R.: Radon Emissions to the Atmosphere and their use as Atmospheric Tracers, University of Edinburgh, PhD thesis, 2005.

Robertson, L. B., Stevenson, D. S., and Conen F.: Test of a northwards-decreasing Rn-222 source term by comparison of modelled and observed atmospheric Rn-222 concentrations, Tellus, 57B, 116–123, 2005.

Schery, S. D. and Wasiolek, M.: Modeling radon flux from the earth's surface. in: Radon and Thoron in the Human Environment, edited by: Katase, A. and Shimo, M., World Scientific, Singapore, 207–217, 1998.

Schery, S. D., Whittlestone, S., Hart, K. P., and Hill, S. E.: The flux of radon and thoron from australian soils, J. Geophys. Res., 94(D6), 8567–8576, 1989.

Von Gunten, H. R., Surbeck, H., and Rössler, E.: Uranium series disequilibrium and high thorium and radium enrichments in Karst formations, Environ. Sci. Technol., 30(4), 1268–1274, 1996.

World Meteorological Organization Global Atmosphere Watch, 1st International expert meeting on sources and measurements of natural radionuclides applied to climate and air quality studies, No 155, 2004.

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Table 1. Verification of the model in Finland and Hungary for regional mean values of measured and predicted ^{222}Rn flux.

	^{222}Rn flux measured	^{222}Rn flux predicted	n
S-Finland	100±17 Bq m^{-2} h^{-1}	102±13 Bq m^{-2} h^{-1}	8
N-Finland	32±11 Bq m^{-2} h^{-1}	41±6 Bq m^{-2} h^{-1}	8
Hungary	60±9 Bq m^{-2} h^{-1}	68±3 Bq m^{-2} h^{-1}	12

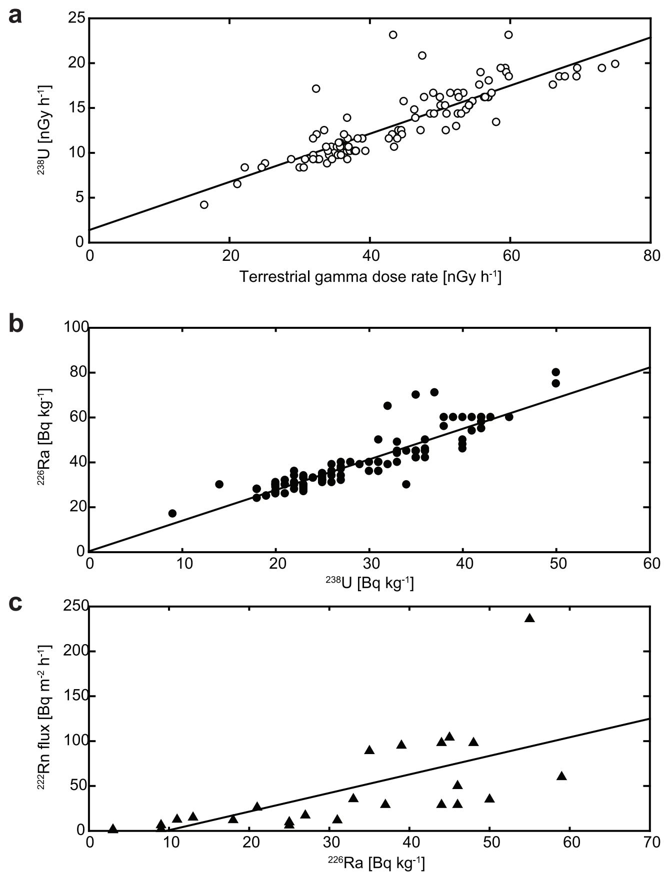

Fig. 1. Correlations between **(a)** the contribution of GDR originating from the ^{238}U decay series and total terrestrial gamma dose rate; **(b)** ^{226}Ra activity and ^{238}U activity and **(c)** ^{222}Rn flux at the soil surface and soil ^{226}Ra activity. Data for (a) and (b) was kindly provided by SUER (Section of Surveillance of Radioactivity, Switzerland).

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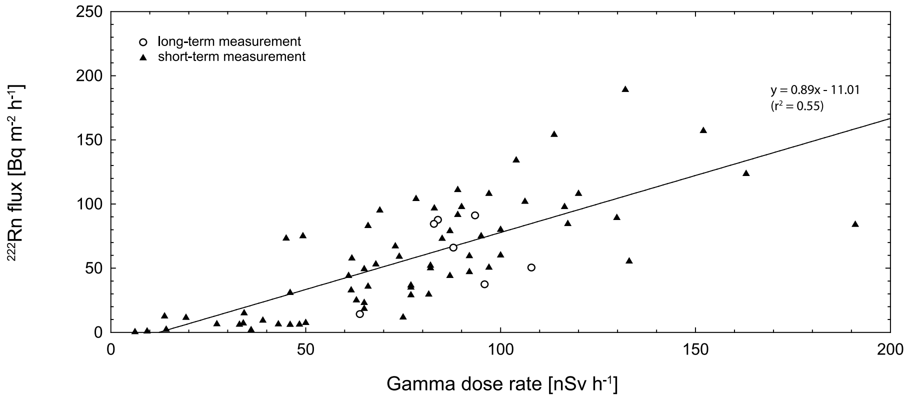

Fig. 2. Correlation of ^{222}Rn flux and terrestrial gamma dose rate measured at field sites in Switzerland, Germany, Scotland, Finland and Hungary.

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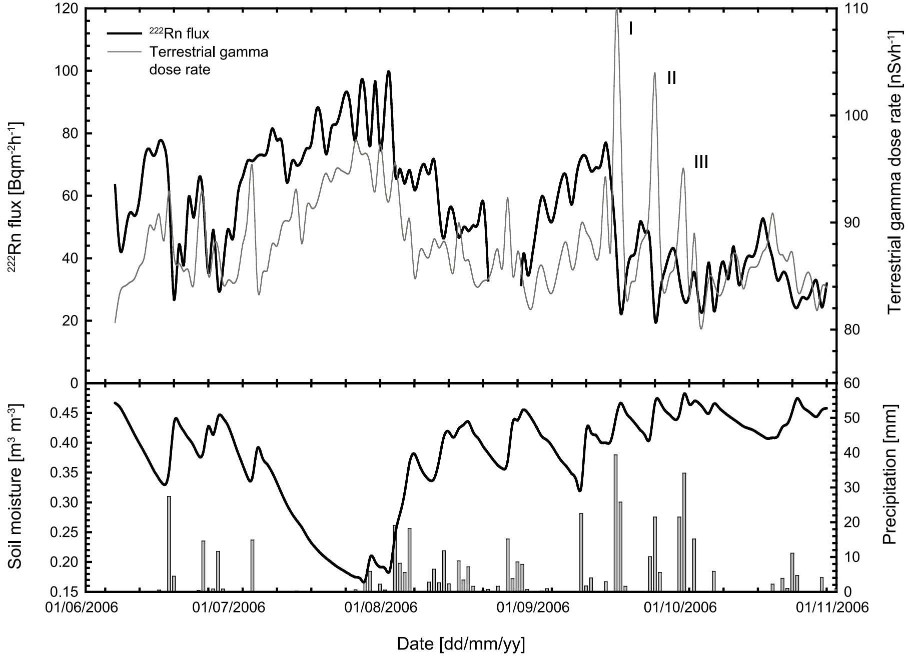

Fig. 3. ^{222}Rn flux, terrestrial gamma dose rate, precipitation and soil moisture time series from June to November 2006 in Basel (Switzerland). Heavy rain events are marked with I, II and III.