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Comparison of one- and two-filter detectors for atmospheric ^{222}Rn measurements

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Abstract

Parallel monitoring of ^{222}Rn and its short-lived progeny (^{218}Po and ^{214}Pb) were carried out from November 2007 to April 2008 close to the top of the Schauinsland mountain, partly covered with forest, in South-West Germany. Samples were aspirated from the same location at 2.5 m above ground level. We measured ^{222}Rn with a dual flow loop, two-filter detector and its short-lived progeny with a one-filter detector. A reference sector for events, facing a steep valley and dominated by pasture, was used to minimize differences between ^{222}Rn and progeny-derived ^{222}Rn activity concentrations. In the two major wind sectors covered by forest to a distance between 60 m and 80 m towards the station progeny-derived ^{222}Rn activity concentration was on average equal to 87% (without precipitation) and 74% (with precipitation) of ^{222}Rn activity concentration.

1 Introduction

^{222}Rn in the lower atmosphere originates from the decay of ^{226}Ra , a member in the decay series of ^{238}U , which is present in trace amounts in all soils. Emission rates of ^{222}Rn vary in space and time (Szegvary et al., 2009). Its only sink in the atmosphere is radioactive decay with a half-life of 3.8 days. This time scale is comparable to the lifetimes of short-lived atmospheric pollutants and the atmospheric residence time of water and aerosols. It is also comparable to important aspects of atmospheric dynamics, making it a useful tracer at local, regional or global scales for testing and validating atmospheric transport models (Israel, 1951; Jacob et al., 1997; Dentener et al., 1999; Taguchi et al., 2002) and for estimating the emission of greenhouse gases by mass balance approach (Dörr et al., 1983; Gaudry et al., 1992; Schmidt et al., 1996, 2001, 2003; Wilson et al., 1997; Biraud et al., 2000; Conen et al., 2002; Hirsch et al., 2006). Decay products of ^{222}Rn , such as ^{218}Po and ^{214}Pb cluster within less than one second forming small particles with diameters from 0.5 to 5 nm. Besides the cluster formation, these radionuclides attach to the existing aerosol particles in the atmosphere within

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1–100 s, forming the radioactive aerosol (Porstendörfer, 1994). Either way, they are subject to dry or wet surface deposition (Wyers and Veltkamp, 1997; Yamamoto et al., 1998; Akata et al., 2008; Petroff et al., 2008). ²²²Rn activity concentration in air is measured using either two-filter or one-filter detectors. Two-filter detectors involve a first filter removing all air-borne progeny from the air sample, a delay volume where air has a constant mean residence time and where new progeny is produced under controlled conditions, and a second filter to collect the newly produced progeny to be counted (e.g. Whittlestone&Zahorowski, 1998). Measuring ²²²Rn with a one-filter detector involves accumulation of its short-lived aerosol-bound progeny directly from the atmosphere onto one filter, its counting, and an assumption about the disequilibrium factor (activity of short-lived progeny/activity of ²²²Rn) between counted progeny and its precursor ²²²Rn (Haxel, 1953; Levin et al., 2002). Worldwide, a total of 23 stations forming part of the Global Atmosphere Watch program of the World Meteorological Organisation (GAW/WMO) are measuring atmospheric ²²²Rn activity concentrations (WMO, 2004). Nine of these stations are equipped with two-filter detectors and 14 use one-filter detectors. The principle difference between one- and two-filter detectors is that two-filter detectors sample from the atmosphere ²²²Rn gas while one-filter detectors sample aerosol-bound ²²²Rn progeny, which is subject to deposition depending on meteorological conditions. Our objective was to investigate what difference changing meteorological conditions may cause between ²²²Rn measurements with one- and two-filter detectors. After the inter-comparison of four different detectors, Collé et al. (1996) draw the following conclusion that stimulated our study: “Without question, continuous inter-comparison measurements over longer time intervals, two or more uninterrupted weeks or even months, would have been much better. Equally, it would have been more useful to conduct correlations with meteorological data and with ²²²Rn progeny measurements and equilibrium ratios.”

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2 Material and methods

2.1 Sampling site

The sampling site (Fig. 1) is located in the Black Forest in South-West Germany (47° 54' 15" N, 7° 54' 33" E, 1200 m a.s.l.) about 750 m North-East of the Schauinsland mountain top (1284 m a.s.l.). Air inlets of both measurement systems were next to each other at 2.5 m above ground. The Schauinsland is a westerly advanced mountain top of the Black Forest mountain range with steep slopes to neighbouring valleys to the North, South and West (Rhine Valley). The orography and local meteorological transport conditions were described in detail by Volz-Thomas et al. (1999) and Seibert et al. (2008). It is an intensive monitoring station equipped with a number of different sensors and belongs to the Federal Office for Radiation Protection of Germany (Bundesamt für Strahlenschutz, BfS). It is situated approximately 1000 m above the Rhine valley and is surrounded by meadows and woods. Dominating tree species around the station are *Picea abies* and *Fagus sylvatica*, with tree heights between 10 m and 20 m. In winter, the area around the station is usually covered with snow. During night, the Schauinsland is usually above the boundary layer inversion of the Rhine Valley. During day time, and particularly in summer, it mostly lies within the boundary layer (Schmidt et al., 1996). Meteorological parameters are continuously measured about 120 m South-South-East (SSE) of the station by the Federal Environment Agency (Umweltbundesamt), which is at the same time a regional Global Atmosphere Watch (GAW) station. During the measurement period from 12 October 2007 to 28 April 2008, the dominant wind sector was West-North-West (WNW) (Fig. 1), passing along the forested ridge and traversing only about 60 m grassland before reaching the air inlet at the station. Another frequent wind sector was North-North-East (NNE), along the rather flat, forested mountain top with grassland covering around 80 m between forest edge and station. A third wind sector is to the South-South-East (SSE). Flat grassland extends from the station in this direction for 160 m before the terrain falls off into a steep valley, the upper edge of which is in this direction covered by a narrow strip of mixed forest.

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We use the last sector as a reference sector while comparing effects of forest cover and precipitation on differences between one- and two-filter detectors in adjacent sectors.

2.2 Measurement techniques

2.2.1 Two-filter detector

The two-filter detector we used in this study has been described in detail by Whittlestone and Zahorowski (1998) and Brunke et al. (2002). Air is continuously drawn at a rate of $0.70 \times 10^{-3} \text{ m}^3 \text{ s}^{-1}$ through an inlet tube (diameter 5 cm diameter; length 10 m) and a first delay volume (two 0.200 m^3 barrels in series) to remove the short-lived ^{220}Rn ($t_{1/2}=56 \text{ s}$), then through a first particle filter to remove all ambient progenies of ^{222}Rn and ^{220}Rn . The cleaned air, containing ^{222}Rn but no progeny, then enters a second delay volume (0.75 m^3), where ^{222}Rn decay produces new progenies under controlled conditions. Air inside the second delay volume circulates at a rate of $0.013 \text{ m}^3 \text{ s}^{-1}$ in an internal loop, where it passes through a second filter retaining the newly formed progenies. Light pulses on a nearby ZnS surface are counted by a photomultiplier. Internal background during the measurement period was 1 count s^{-1} and sensitivity $0.30 \text{ count s}^{-1} \text{ per Bq m}^{-3}$. Three background measurements were carried out during the observation period. The instrument was calibrated monthly with a passive ^{222}Rn source (21.887 kBq ; calibrated against NIST standards; Pylon Electronics Inc., Ottawa, Canada).

2.2.2 One-filter detector

The one-filter detector used in this study is the BfS system (α/β Monitor P3), which is described in more detail in Stockburger and Sittkus (1966). Beside the continuous measurement of natural atmospheric radioactivity the detector system was mainly developed to monitor the artificial atmospheric β -activity from nuclear weapons fall-out and from releases of nuclear power plants, like during the incident in Chernobyl in

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spring 1986. The electronics for counting and data recording as well as the pumping system was modernized several times since 1966 but the detector system is still unchanged. Ambient air is continuously drawn through an aerosol filter (membrane filters (mixed cellulose ester) 1.2 μm , 150-250 mm ME 28 Schleicher&Schuell), where the progenies of ^{222}Rn and ^{220}Rn are quantitatively collected and the activity is measured with a (self-made) sandwich counter consisting of three independent proportional counters. In the first counter above the filter the lower energy α 's are measured, in the middle counter the high energy α 's are measured and in the third counter the β -activity is measured. The α -activity of the short-lived ^{222}Rn progenies ^{218}Po ($\alpha_E=6.0\text{ MeV}$, $t_{1/2}=3.05\text{ min}$) and ^{214}Po ($\alpha_E=7.69\text{ MeV}$, $t_{1/2}=164\text{ }\mu\text{s}$) collected on the filter is measured in situ mainly in the counter positioned directly above the filter. Corrections are made for count rate contributions coming from the progenies of ^{220}Rn which are mainly measured in the middle counter. Air is continuously pumped at $50\text{ m}^3\text{ h}^{-1}$ through an air duct (cross section $35\text{ cm}\times 45\text{ cm}$; length 5 m) over the filter for one week. After one week the pump is switched off, the filter is replaced, a 1 h check calibration using a $^{241}\text{Am}/^{90}\text{Sr}$ source is performed, followed by a background check with a new filter for an additional hour and then the air flow is started again. The sensitivity for short-lived ^{222}Rn progeny, expressed in ^{222}Rn equivalent, is 3.367 Bq cps^{-1} or $0.0673\text{ Bq m}^{-3}\text{ cps}^{-1}$ for an air flow rate of about $50\text{ m}^3\text{ h}^{-1}$. The background count rate used for data evaluation is 0.043 cps and was determined during a period of several days without an air flow. The background is controlled for 1 h after the weekly filter exchange. The ^{222}Rn equivalent activity concentration is calculated based on the assumption of equilibrium between ^{222}Rn activity and ^{218}Po and ^{214}Po activity in the atmosphere. The activity of ^{218}Po and ^{214}Po measured on the filter is only in equilibrium with the atmospheric ^{222}Rn , if the atmospheric activity is constant. If the latter changes, it is taken into account during the calculations by a correction factor which is a function of the half-life.

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3 Results and discussion

The time series of hourly values of atmospheric activity concentration of ^{222}Rn (measured with the two-filter detector), short-lived ^{222}Rn progeny, expressed in ^{222}Rn equivalent (measured with the one-filter detector), and meteorological parameters observed at Schauinsland station from October 2007 to April 2008 shows structures on the synoptical time scale (Fig. 2). Precipitation occurred from time to time with intensities ranging from 0.1 to 10.5 mm h⁻¹ in form of snow, rain or drizzle. Air temperature fluctuated between -10 °C to 10 °C with a mean of 1 °C. The relative humidity (RH) remained most of the time above 90% with some short periods of substantially smaller values, usually associated with southerly winds. Wind directions were already described above. Mean hourly wind speed ranged from 0.2 to 22.5 m s⁻¹. We note that atmospheric activity concentration of ^{222}Rn and short-lived ^{222}Rn progeny obtained by the different detector types follow a very similar pattern, even before harmonization of instrumental background and calibration. Activity concentrations of ^{222}Rn and short-lived ^{222}Rn progeny ranged from 0.5 to 10.8 Bq m⁻³ with a mean value of 2.8 (s.d.=1.5) Bq m⁻³ for activity concentration of ^{222}Rn , and from 0.1 to 10.7 Bq m⁻³ with a mean value of 1.8 (s.d.=1.3) Bq m⁻³ for short-lived ^{222}Rn progeny expressed in ^{222}Rn equivalent, respectively. Of all hourly values, 84% were below 4 Bq m⁻³. Close to the mountain top, changes in the origin of advected air, be it from the boundary layer or from the free troposphere, drive fluctuations in ^{222}Rn activity concentrations. This assumption is supported by the analysis of back-trajectories calculated using version 4.6 of NOAA Air Resources Laboratory's (ARL) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model for all hourly ^{222}Rn values (Draxler and Rolph, 2003). The upper quartile of observed ^{222}Rn activity concentrations was clearly associated with air masses that have reached the station from a lowest altitude, suggesting advection of boundary layer air masses (Fig. 3). In contrast, the lowest ^{222}Rn activity concentrations were found in air that has reached the station from a greater height and has most likely not been in contact with land surfaces for some time before arrival.

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3.1 Harmonization of instrumental background and calibration

Differences between measured activity concentration of ^{222}Rn and short-lived ^{222}Rn progeny are caused by differences in instrumental background and calibration in addition to changes of the progeny/ ^{222}Rn disequilibrium in air with meteorological conditions. As we are interested in the effect of meteorological conditions on ^{222}Rn estimates made by one- and two-filter detectors, we have to minimize differences caused by instrumental background and calibration, including the selection of an appropriate disequilibrium factor to transform short-lived ^{222}Rn progeny activity to ^{222}Rn activity concentration. To this end we selected conditions when progeny removal was considered minimal. Since forest canopies and precipitation increase the deposition rate (Petroff et al., 2008), we choose those data, when there was no precipitation and air arrived from the reference wind sector (120° – 180°). This air has travelled above a steep valley where only the upper slope is covered by a narrow strip of forest that does not extend onto the grassland plateau forming the last 160 m to the station. The correlation between measured activity concentrations for this selection (Fig. 4) is strong (Spearman rank correlation coefficient=0.946). There is an off-set of 0.382 Bq m^{-3} between detectors and values of short-lived ^{222}Rn progeny tend to be smaller than those of ^{222}Rn by a factor of 0.898. This is very close to the disequilibrium factor (0.85) estimated for this station by Schmidt (1999, as cited in Schmidt et al., 2003). Much larger differences between detectors have been reported (Collé et al., 1996). Because of physical plausibility we assume in our further analysis that the observed off-set is entirely due to internal instrumental effects and not explained by environmental factors. For the purpose of this study it is irrelevant to know which instrument is more accurate. We are interested in relative differences between ^{222}Rn and progeny-derived ^{222}Rn caused by meteorological conditions. For further analysis, we add 0.382 Bq m^{-3} to the short-lived ^{222}Rn progeny activity concentration measured with the one-filter detector and divide it by 0.898, thereby transforming short-lived ^{222}Rn progeny activity concentration into progeny-derived ^{222}Rn activity concentration. However, this way to

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harmonize background and calibration should not suggest that we think the two-filter detector is better background corrected or calibrated than the one-filter detector.

3.2 Effect of precipitation intensity

To investigate the effect of precipitation intensity, we selected all hourly values with precipitation larger than zero from the harmonized data set and sorted them into ranges with a similar number of observations in each range (Fig. 5). Within each range, there is a large variation in the ratio of progeny-derived ^{222}Rn to ^{222}Rn . We only can give plausible arguments for the reason of this behavior. Uncertainty in the measurements are certainly one cause. If this would be negligible, the ratio should always be ≤ 1 . Another reason may be associated with the process of wet deposition itself. A precipitation event, for example of 1 mm h^{-1} , may be caused by a short spell of large rain drops with small specific surface areas for interaction with aerosol. If so, its effect on wash-out of progeny is short and small. Alternatively, the same amount of rain may fall in a drizzle where the same amount of precipitation has an orders of magnitudes larger specific surface area and where interaction with short-lived progeny lasts the entire integration interval of the measurement. Despite the scatter of values within each range, our data suggests a weak tendency towards larger disequilibria with increasing precipitation intensity. Yet, it is impossible to provide precipitation-dependent factors to reliably convert progeny signal to ^{222}Rn concentration.

3.3 Effect of forest canopies

Aerosols, such as short-lived progeny of ^{222}Rn , can be collected by vegetation due to the interaction of aerosols with every vegetation surface (leaves, trunks, twigs, heads and fruits). Different mechanical processes generate the deposition. From smaller to larger particle sizes these are mainly Brownian diffusion, interception, inertial impaction and sedimentation. Compared to other types of land surfaces, research in the field of acid deposition to forest has shown largely increased deposition velocities above forest

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(Petroff et al., 2008). Smaller activity concentration of ^{214}Pb below canopy compared to above canopy have been reported (Wyers and Veltkamp, 1997). As indicated in Fig. 1, the Schauinsland station is partly surrounded by forest. To estimate the effect of forest canopy on differences between progeny-derived ^{222}Rn and ^{222}Rn , we plotted values from the three major wind directions for conditions when there was no precipitation. By default (point 3.3.1), the slope of the regression in the reference sector (120° – 180°) is 1 (Fig. 6a). Deviations from 1 in the two other sectors can be ascribed to the effect of forest canopy on progeny removal. On average, values of progeny-derived ^{222}Rn were 0.86 and 0.87 times those of ^{222}Rn in the forest covered sectors 240° – 300° and 0° – 60° , respectively (Fig. 6c, e).

3.4 Effects of precipitation and forest canopy

Ideally, we would have liked to compare progeny-derived ^{222}Rn and ^{222}Rn for the open wind sector, with and without precipitation, to get an estimate for the mean effect of precipitation only. Unfortunately, there were only 10 one-hourly intervals with precipitation from the open sector during the observation period. This is obviously not enough. For completeness, we nevertheless added the data to Fig. 6b. Consequently, the effect of precipitation, irrespective of intensity, can only be investigated in combination with the effect of forest canopy. Compared to forest canopies under dry conditions, precipitation reduced by 9% and 21% progeny-derived ^{222}Rn in the analyzed air for the wind sector 240° – 300° and 0° – 60° , respectively (Fig. 6d, f). Thus, the effect of precipitation seems to be of similar magnitude as the effect of forest canopy. Yet both influences can not be clearly separated because of a possible interaction between precipitation and forest canopy. It may well be that a forest canopy is more efficient in progeny removal when wet than when dry. During precipitation, average wind speed and air temperatures were similar, while relative humidity was larger, compared to conditions without precipitation (Table 1).

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4 Conclusions

The observations show that one- and two-filter systems are suitable to continuously monitor ²²²Rn in ground level air. Most of the time both systems follow the same pattern and produce very similar results, except under special meteorological conditions, when precipitation or forest canopy remove short-lived progeny from the air mass to be measured. Such effects are generally much smaller than the large fluctuations in activity concentrations of ²²²Rn and progeny-derived ²²²Rn on diurnal and synoptical time scales. The average altitude of air masses a few hours prior to arrival at a mountain station is expected to largely influence activity concentrations.

There is no clear relationship between precipitation intensity and the magnitude of the difference between progeny-derived ²²²Rn and ²²²Rn activity concentration. Thus, there is no precipitation-dependent factor to reliably convert progeny signal to ²²²Rn concentration. Disequilibrium between ²²²Rn and its short-lived progeny near the surface of a mountain top may be affected to a similar magnitude by the interaction between air and forest canopy and by wet deposition. Each factor may, cumulatively, reduce progeny-derived ²²²Rn activity concentration between about 10% and 15% compared to ²²²Rn activity concentration. These two effects and their influence on the ²²²Rn data were studied in this work and should be known for the interpretation and intercomparison of ²²²Rn data measured with different systems and at different sites.

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Table 1. Means and standard deviation (s.d.) of meteorological parameters for the three main wind sectors during dry (no precipitation) and wet (precipitation>0) conditions.

Wind sector		Wind speed (m s ⁻¹)		Temperature (°C)		Relative humidity (%)	
		mean	s.d.	mean	s.d.	mean	s.d.
120°–180°	dry	4.0	1.6	0.0	5.0	70.0	27.0
	wet	3.0	1.2	0.7	4.4	95.2	2.4
240°–300°	dry	6.1	3.4	2.5	4.1	75.4	24.4
	wet	8.2	3.8	0.9	3.0	96.5	4.7
0°–60°	dry	2.6	1.2	0.2	4.0	81.2	25.9
	wet	3.2	1.5	−0.7	3.2	97.7	2.1

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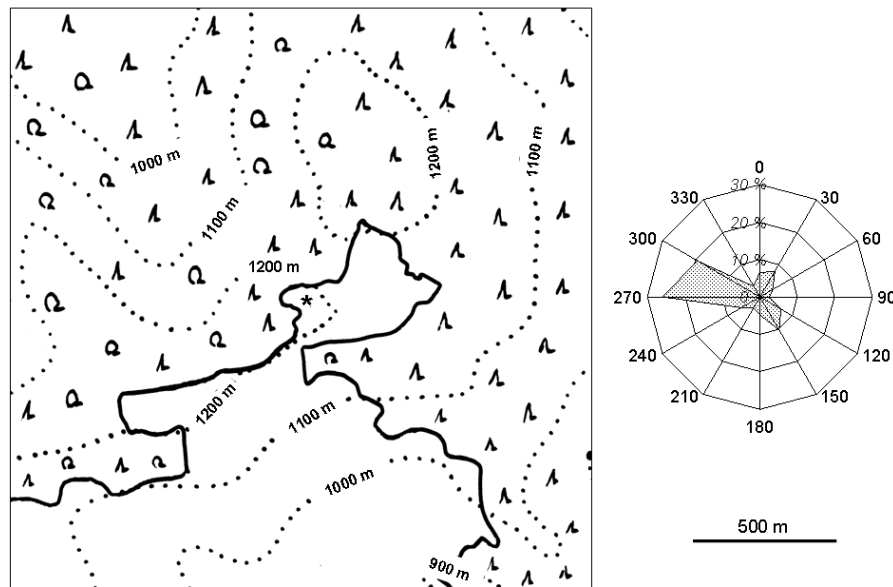


Fig. 1. (left) Sketch of topography and forest cover (solid line indicates forest edge) around the measurement station (asterisk in the centre) in the Black Forest. (right) Frequency distribution of wind directions for 30° sectors during the observation period. Wind from the sector 120°–180° is considered to have been least influenced by vegetation.

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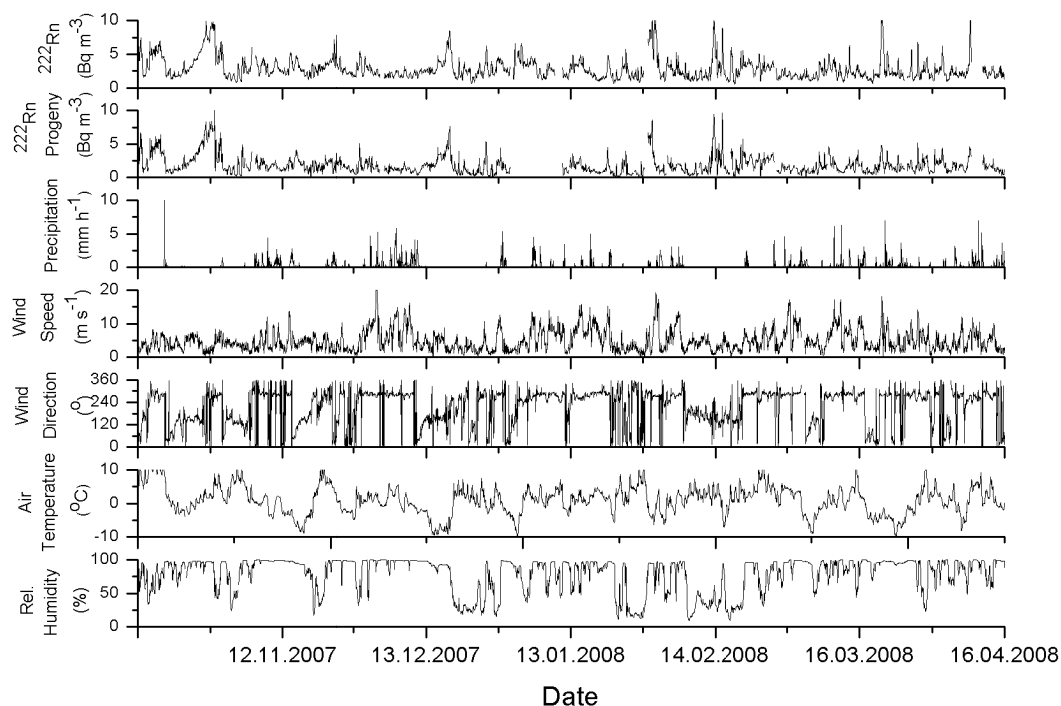


Fig. 2. Time series of hourly means of ^{222}Rn activity concentration (measured with a two-filter detector) and short-lived ^{222}Rn progeny, expressed in ^{222}Rn equivalent (measured with a one-filter detector) before harmonizing background and calibration between instruments, hourly precipitation, mean wind speed, wind direction, air temperature and relative humidity at Schauinsland station from October 2007 to April 2008.

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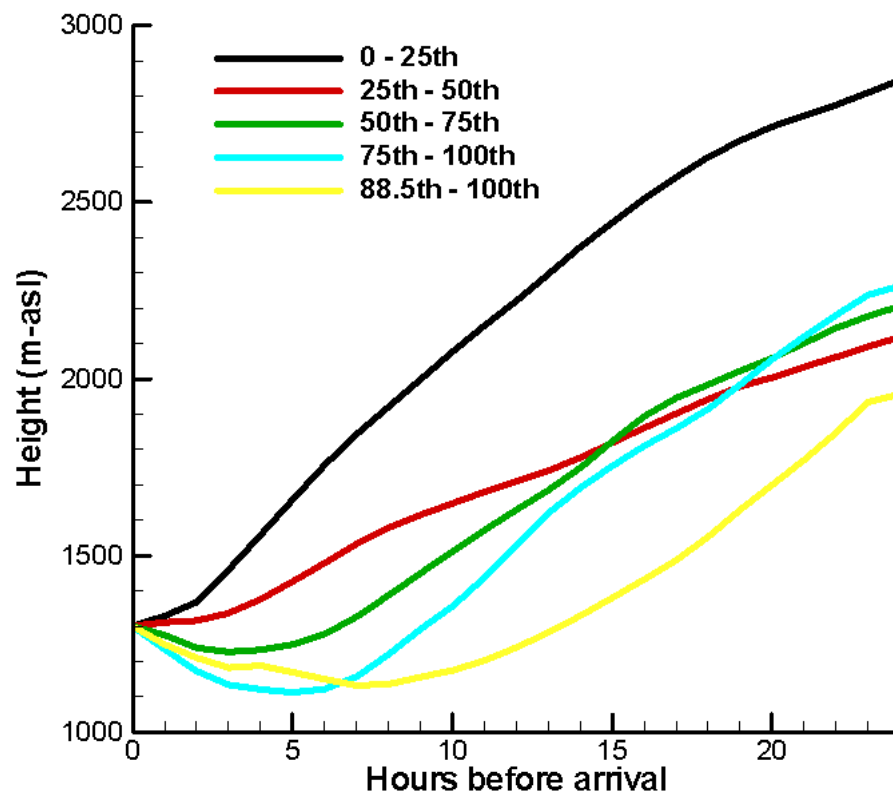


Fig. 3. Average altitude of air masses (ensemble means of single particle trajectories) during the 24 h before arrival at the station for the lowest (0–25th) to the highest (75th–100th) quartile of observed ^{222}Rn activity concentrations.

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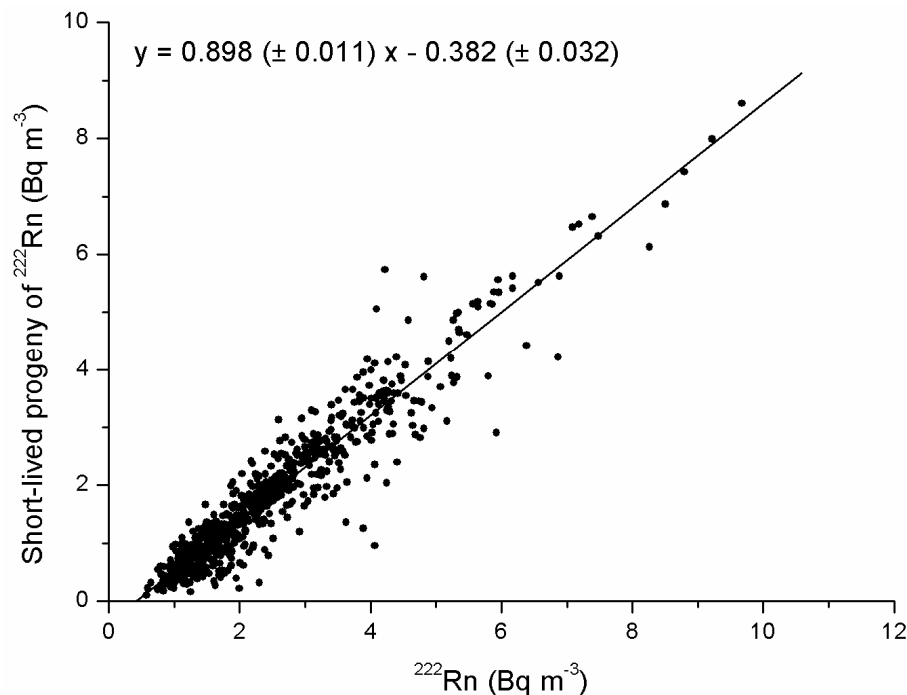


Fig. 4. Correlation between activity concentrations of ^{222}Rn and short-lived ^{222}Rn progeny (expressed in ^{222}Rn equivalent) as determined by two independently calibrated instruments for events with no surface wet deposition and wind from the reference sector (values in brackets are standard errors of regression parameters). The Spearman rank correlation coefficient r equals 0.946.

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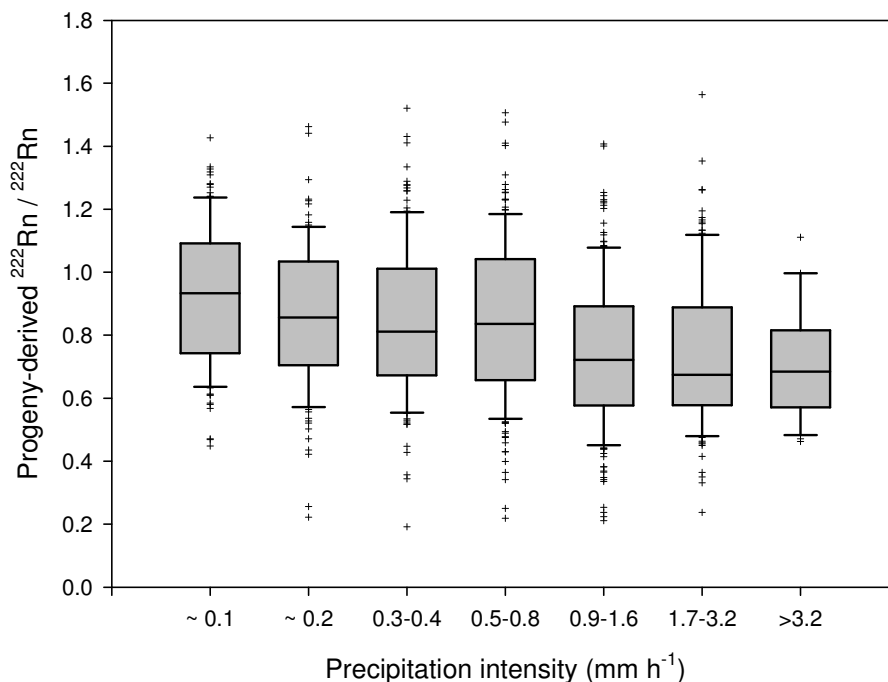


Fig. 5. Ratio of the activity concentrations of progeny-derived ^{222}Rn and ^{222}Rn summarized for different ranges of precipitation intensity (instrumental background and calibration have been harmonized between detectors). Boxes indicate median, upper and lower quartile, whiskers 10th and 90th percentile, crosses are outliers. Each range includes between about 120 and 180 hourly values, except for precipitation intensities $>3.2 \text{ mm h}^{-1}$ ($n=29$). The lowest precipitation intensities are near the detection limit of the instrument and therefore only approximate.

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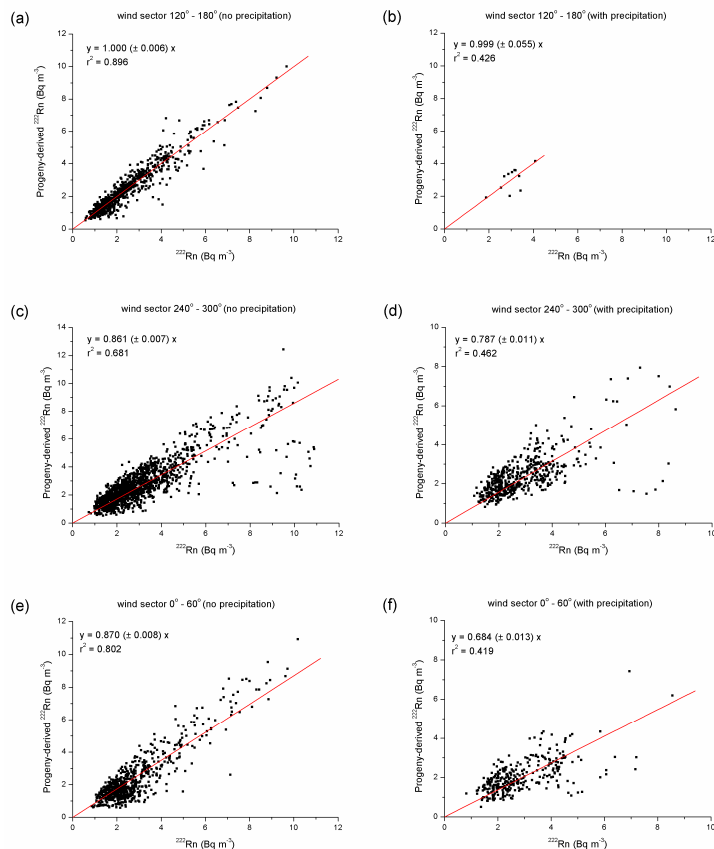


Fig. 6. Correlation between activity concentration of progeny-derived ^{222}Rn and ^{222}Rn for the reference sector (**a, b**) and the two sectors influenced by forest cover (**c, d, e, f**), for without precipitation (**a, c, e**) and with precipitation (**b, d, f**) (values in brackets are standard errors of regression parameters). Instrumental background and calibration have been harmonised between detectors.

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