

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

Y. Xia¹, H. Sartorius², C. Schlosser², U. Stöhlker², F. Conen¹, and W. Zahorowski³

¹Institute of Environmental Geosciences, University of Basel, Bernoullistrasse 30, 4056 Basel, Switzerland

²Federal Office for Radiation Protection (BfS), Willy-Brandt-Str. 5, 38226 Salzgitter, Germany

³Australian Nuclear Science and Technology Organisation, PMB 1, Menai, NSW 2234, Australia

Received: 11 January 2010 – Published in Atmos. Meas. Tech. Discuss.: 16 February 2010

Revised: 25 May 2010 – Accepted: 14 June 2010 – Published: 28 June 2010

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. The observations show that most of the time both detector types follow the same pattern. Still, there is no single disequilibrium factor that could be used to exactly transform short-lived progeny to ^{222}Rn activity concentration under all meteorological conditions.

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 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).

^{222}Rn 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 and Zahorowski, 1998). Measuring ^{222}Rn 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 ^{222}Rn) between counted progeny and its precursor ^{222}Rn (Haxel 1953, Levin et al. 2002). Worldwide, a total of 23 stations forming part of

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.



Correspondence to: Y. Xia
(yu.xia@unibas.ch)

the Global Atmosphere Watch program of the World Meteorological Organisation (GAW/WMO) are measuring atmospheric ^{222}Rn 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 ^{222}Rn gas while one-filter detectors sample aerosol-bound ^{222}Rn progeny, which is subject to deposition depending on meteorological conditions. Our objective was to investigate what difference changing meteorological conditions may cause between ^{222}Rn 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 ^{222}Rn progeny measurements and equilibrium ratios.*”

2 Material and methods

2.1 Sampling site

The sampling site (Fig. 1) is located in the Black Forest in South-West Germany ($47^{\circ}54'15''\text{N}$, $7^{\circ}54'33''\text{E}$, 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). The station 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. 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 the two other 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 membrane 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 (mesh wire, $20 \mu\text{m}$). Here, newly formed progenies deposit by Brownian diffusion. Light pulses on a nearby ZnS surface are counted by a photomultiplier. Internal background during the measurement period was around 1 cps and sensitivity $3.3 \text{ Bq m}^{-3} \text{ cps}^{-1}$. 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 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) $1.2 \mu\text{m}$, $150 \times 250 \text{ mm}$ ME 28 Schleicher & Schuell). The effective filter area is

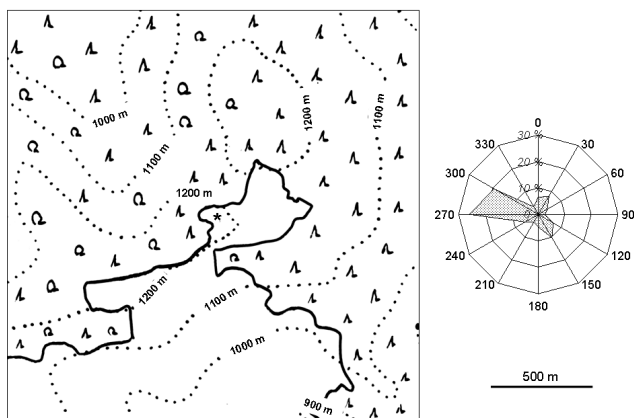


Fig. 1. Left: sketch of topography and forest cover (solid line indicates forest edge) around the measurement station (asterisk in the exact centre) in the Black Forest. Right: Frequency distribution of wind directions for 30° sectors during the observation period. Wind from the sector 120° – 180° is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

300 cm^2 . At a distance of 14 mm above the filter is a stack of three independent, methane-filled, proportional counters having the same length and width as the active filter area (Fig. 2). The proportional counters operate in the proportional range such that the lower counter measures α -activity from progeny of ^{222}Rn and ^{220}Rn . The middle counter detects the high energy α -activity of ^{212}Po (^{220}Rn progeny). The half life of ^{212}Po (10.6 h) is relevant for the time required to reach an equilibrium between activity in air and activity on the filter. Therefore, we can not always assume an equilibrium between activity in air and activity on the filter. Changes in atmospheric concentrations can occur before an equilibrium is reached on the filter. However, a determination of actual ^{212}Po activity in air is possible, if not only the activity on the filter but also its change over time is taken into account. By difference, the ^{222}Rn progeny activity is derived from the lower counter. The upper counter counts β particles only. Air is continuously pumped at $0.014\text{ m}^3\text{ s}^{-1}$ 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, an one hour 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 $0.014\text{ m}^3\text{ s}^{-1}$. 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 ^{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 con-

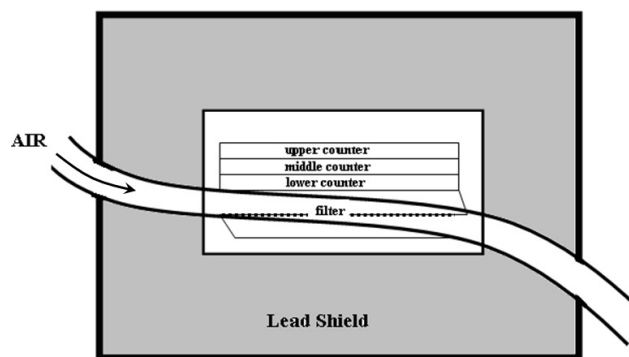


Fig. 2. The One-filter detector system contains a membrane filter and a stack of three independent, methane-filled, proportional counters having the same length and width as the active filter area. The middle counter detects the high energy α -activity of ^{212}Po (^{220}Rn progeny). Activity of ^{212}Po , together with the change in ^{212}Po activity over time, are used to determine total ^{220}Rn progeny contribution to total counts in the lower counter. By difference, the ^{222}Rn progeny activity is derived from the lower counter. The upper counter counts β particles only (redrawn from Stockburger and Sittkus, 1966).

stant. If the latter changes, it is taken into account during the calculations by a correction factor which is a function of the half-life.

The one-filter detector on Schauinsland represents one commonly applied principle to estimate atmospheric ^{222}Rn concentrations based on the collection and α -counting of both short-lived ^{222}Rn progeny (^{218}Po and ^{214}Po) from atmospheric air. For example, all one-filter detectors mentioned as operating at GAW stations in the WMO/GAW report No. 155 (2004) derive estimates of atmospheric ^{222}Rn from the combined detection of ^{218}Po and ^{214}Po . We are aware of other one-filter detectors that derive ^{222}Rn estimates exclusively from atmospheric ^{218}Po concentration such as the ‘Radgrabber’ (e.g. Lee and Larsen, 1997) or some commercial instruments. Also the two-filter detector we used, is not the only instrument measuring atmospheric ^{222}Rn instead of atmospheric ^{222}Rn progeny. Other instruments include those based on the design by Iida et al. (1996) and widely used in East Asia (e.g. Moriizumi et al., 2008), and the two filter detector developed by the Environmental Measurements Laboratory (EML) as described in Collé et al. (1996). Hence, the instruments in our study represent the two measurement principles of a majority of detectors currently in use.

3 Results and discussion

3.1 General description of data

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

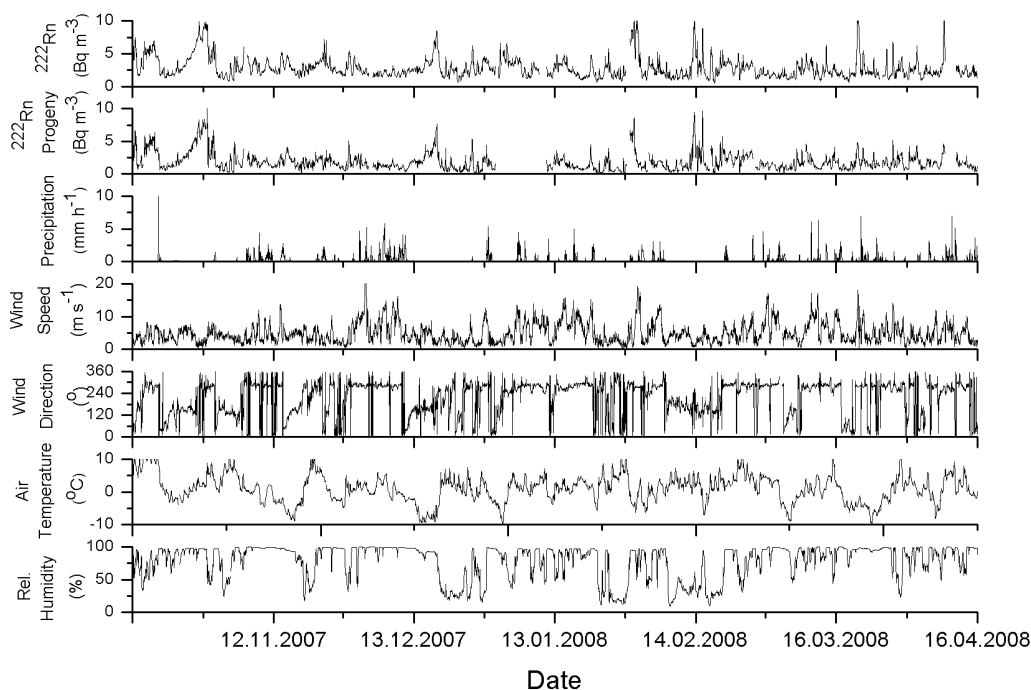


Fig. 3. Time series of hourly means of ^{222}Rn activity concentration (measured with a two-filter detector) and short-lived ^{222}Rn progeny, expressed in ^{222}Rn equivalent (measured with a one-filter detector) before harmonizing background and calibration between instruments, hourly precipitation, mean wind speed, wind direction, air temperature and relative humidity at Schauinsland station from October 2007 to April 2008.

(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. 3). 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 (HYSPPLIT) 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. 4). 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.

3.2 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

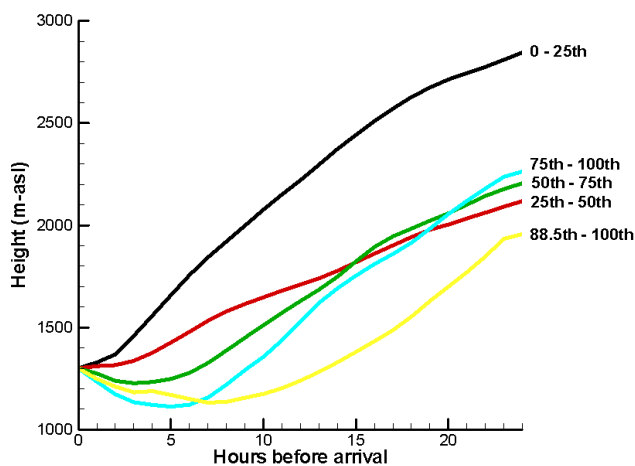


Fig. 4. Average altitude of air masses (ensemble means of single particle trajectories) during the 24 h before arrival at the station for the lowest (0–25th) to the highest (75th–100th) quartile of observed ^{222}Rn activity concentrations.

(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. 5) 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. An instrumental effect leading to this off-set, for example, could be an over-estimate of the ^{220}Rn progeny activity (^{212}Po) by the one-filter detector. This would lead to a lower estimate of short-lived ^{222}Rn progeny activity. 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 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.

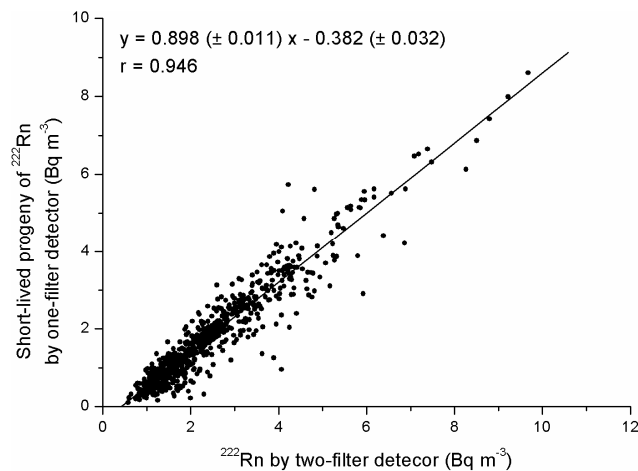


Fig. 5. Correlation between activity concentrations of ^{222}Rn (measured by two-filter detector) and short-lived ^{222}Rn progeny (expressed in ^{222}Rn equivalent; measured by one-filter detector) as determined by the two independently calibrated instruments for events with no surface wet deposition and wind from the reference sector (values in brackets are standard errors of regression parameters). The Spearman rank correlation coefficient r equals 0.946.

3.3 Effect of precipitation intensity

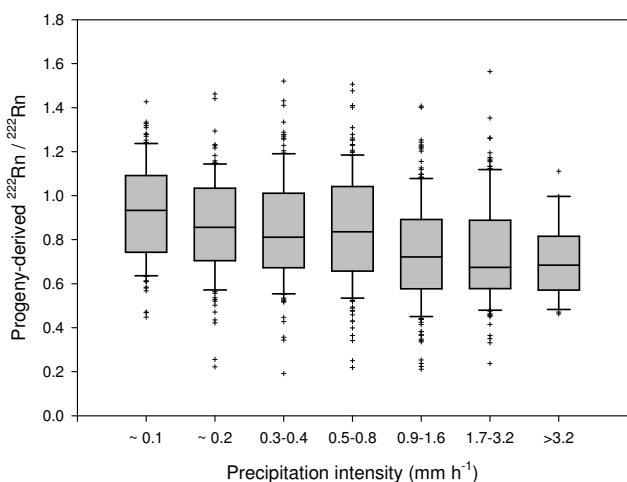
To investigate the effect of precipitation intensity, we selected all hourly values with precipitation larger than zero from the harmonized data set and sorted them into ranges with a similar number of observations in each range (Fig. 6). Within each range, there is a large variation in the ratio of 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.

3.4 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

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 ($^{\circ}\text{C}$)		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

**Fig. 6.** 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.

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 (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 (Sect. 3.2), the slope of the regression in the reference sector (120°–180°) is 1 (Fig. 7a). 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. 7c, e).

3.5 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. 7b. 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 progeny-derived ^{222}Rn in the analyzed air by 9% and 21% for the wind sector 240°–300° and 0°–60°, respectively (Fig. 7d, 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). The degree to which deposition of ^{222}Rn progeny is affected by forest canopies in various wind sectors would be different at other stations, which may be closer or further away from a forest edge, or where forest canopies are not similar to those on Schauinsland. The effect of precipitation is probably less site-specific. However, more generally, our results show that changing meteorological conditions affect the relative difference between

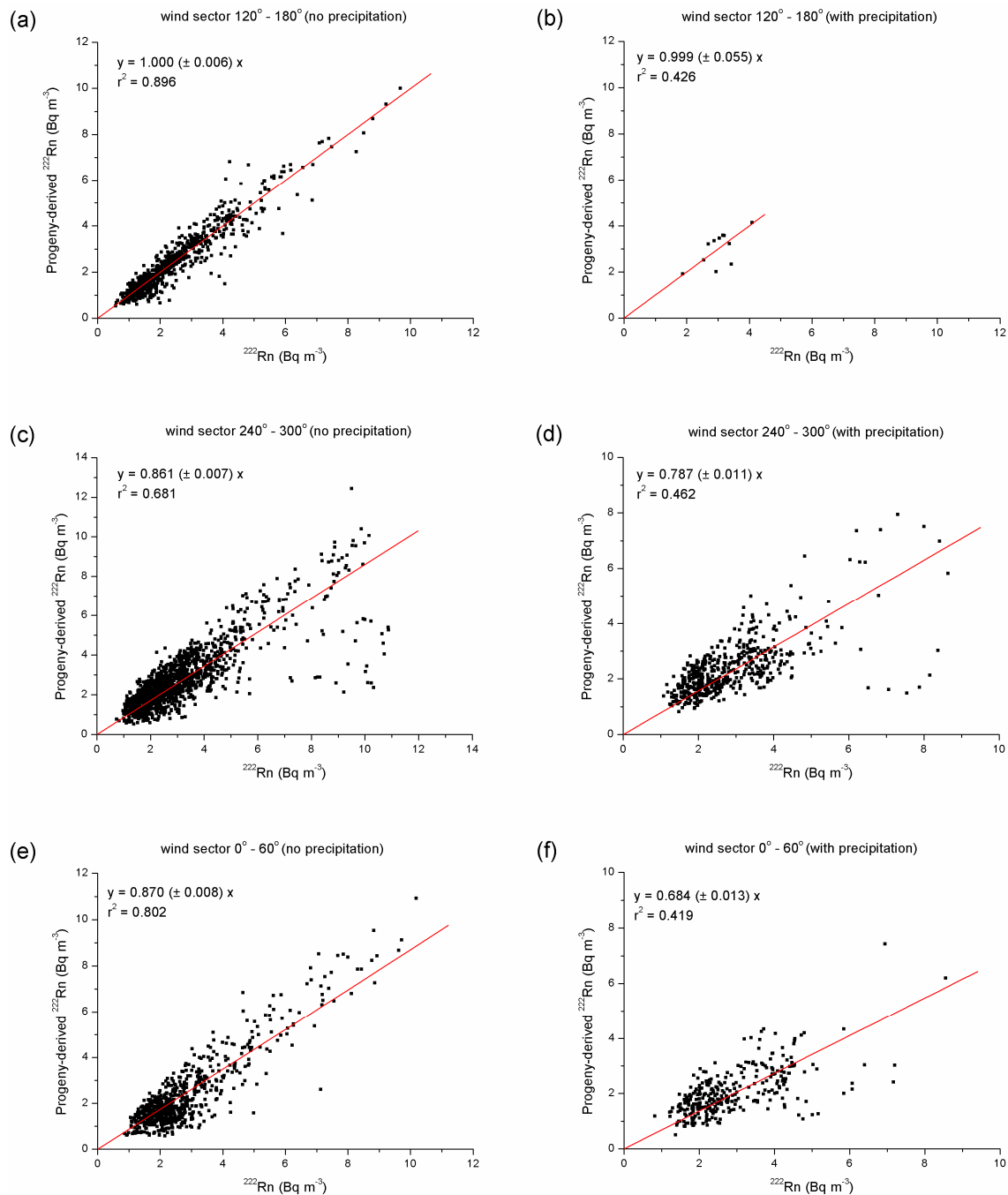


Fig. 7. Correlation between activity concentration of progeny-derived ^{222}Rn and ^{222}Rn for the reference sector (**a**, **b**) and the two sectors influenced by forest cover (**c**, **d**, **e**, **f**), for without precipitation (**a**, **c**, **e**) and with precipitation (**b**, **d**, **f**) (values in brackets are standard errors of regression parameters). Instrumental background and calibration have been harmonised between detectors.

one- and two-filter detectors. Consequently, there is not one single disequilibrium factor for a specific site that could be used to directly transform short-lived progeny to ^{222}Rn activity concentration. Site-specific disequilibrium factors cover a range of values depending on meteorological conditions. This more general outcome of our study applies to probably most other stations.

4 Conclusions

The observations show that one- and two-filter systems are suitable to continuously monitor ^{222}Rn 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 ^{222}Rn and progeny-derived ^{222}Rn 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 ^{222}Rn and ^{222}Rn activity concentration. Thus, there is no precipitation-dependent factor to reliably convert progeny signal to ^{222}Rn concentration. Disequilibrium between ^{222}Rn 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 ^{222}Rn activity concentration between about 10% and 15% compared to ^{222}Rn activity concentration. These two effects and their influence on the ^{222}Rn data were studied in this work and should be known for the interpretation and intercomparison of ^{222}Rn data measured with different systems and at different sites. Deviation of progeny-derived ^{222}Rn from directly measured ^{222}Rn activity concentration will be smaller where one-filter detectors specifically count ^{218}Po only, instead of the combined activity concentration of ^{218}Po and ^{214}Po .

Acknowledgements. This project was funded by the Swiss National Science Foundation (project no. 200020-117622/1). We would like to thank Frank Meinhardt from the German Environment Agency for providing the meteorological data for the site. The presented data would not be available without the conscientiousness of the local station operators at the BfS station Schauinsland in operating and maintain the systems. One of us (WZ) would like to acknowledge help of Jagoda Crawford and Sylvester Werczynski of ANSTO in back trajectory analysis. We thank two anonymous reviewers for their constructive and helpful comments.

Edited by: M. Weber

References

- Akata, N., Kawabata, H., Hasegawa, H., Sato, T., Chikuchi, Y., Kondo, K., Hisamatsu, S., and Inaba, J.: Total deposition velocities and scavenging ratios of ^7Be and ^{210}Pb at Rokkasho, Japan, *J. Radioanal. Nucl. Ch.*, 277(2) 347–355, 2008.
- Biraud, S., Ciais, P., Ramonet, M., Simmonds, P., Kazan, V., Monfray, P., O'Doherty, S., Spain, T. G., and Jennings, S. G.: European greenhouse gas emissions estimated from continuous atmospheric measurement and radon-222 at Mace Head, Ireland, *J. Geophys. Res.-Atmos.*, 105, 1351–1366, 2000.
- Brunke, E. G., Labuschagne, C., Parker, B., van der Spuy, D., and Whittlestone, S.: Cape point GAW station ^{222}Rn detector: factors affecting sensitivity and accuracy, *Atmos. Environ.*, 36, 2257–2262, 2002.
- Collé, R., Unterweger, M. P., Hutchinson, J. M. R., Whittlestone, S., Polian, G., Ardouin, B., Kay, J. G., Friend, J. P., Blomquist, B. W., Nadler, W., Dang, T. T., Larsen, R. J., and Hutter, A. R.: An international marine-atmospheric ^{222}Rn measurement inter-comparison in Bermuda Part II: Results for the participating laboratories, *J. Res. Natl. Inst. Stan.*, 101, 21–45, 1996.
- Conen, F., Neftel, A., Schmid, M., and Lehmann, B. E.: $\text{N}_2\text{O}/\text{Rn}$ -222-soil flux calibration in the stable nocturnal surface layer, *Geophys. Res. Lett.*, 29(2), 1025, doi:10.1029/2001GL013429, 2002.
- Dentner, F., Feichter, J., and Jeuken, A.: Simulation of the transport of Rn-222 using on-line and off-line global models at different horizontal resolutions: a detailed comparison with measurements, *Tellus*, 51(B), 573–602, 1999.
- Dörr, H., Kromer, B., Levin, I., Münnich, K. O., and Volpp, H. J.: CO_2 and Radon-222 as tracers for atmospheric transport, *J. Geophys. Res.*, 88, 1309–1313, 1983.
- Draxler, R. R. and Rolph, G. D.: Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) Model, <http://www.arl.noaa.gov/ready/hysplit4.html>, 2003.
- Gaudry, A., Kanakidou, M., Mihalopoulos, N., Bonsang, B., Bonsang, G., Monfray, P., Tymen, G., and Nguyen, B. C.: Atmospheric trace compounds at a European coastal site- Application to CO_2 , CH_4 and COS flux determinations, *Atmos. Environ.*, 26(A), 145–157, 1992.
- Haxel, O.: Eine einfache Methode zur Messung des Gehalts der Luft an radioaktiven Substanzen, *Zeitschrift für angewandte Physik*, 5, 241–242, 1953 (in German).
- Hirsch, A. I., Michalak, A. M., Bruhwiler, L. M., Peters, W., Dlugokencky, E. J., and Tans, P. P.: Inverse modelling estimates of the global nitrous oxide surface flux from 1998–2001, *Global Biogeochem.*, 20, GB1008, doi:10.1029/2004GB002443, 2006.
- Iida, T., Ikebe, Y., Suzuki, K., et al.: Continuous measurements of outdoor radon concentrations at various locations in East Asia, *Environment International*, 22, Supplement 1, S139–S147, 1996.
- Israel, H.: Radioactivity of the atmosphere, *Am. Meteorol. Soc.*, Boston, 155–161, 1951.
- Jacob, D. J., Prather, M. J., Rasch, P. J., Shia R. L., Balkanski, Y. J., Beagley, S. R., Bergmann, D. J., Blackshear, W. T., Brown, M., Chiba, M., Chipperfield, M. P., Degrandpre, J., Dignon, J. E., Feichter, J., Genthon, C., Grose, W. L., Kasibhatla, P. S., Kohler, I., Kritz, M. A., Law, K., Penner, J. E., Ramonet, M., Reeves, C. E., Rotman, D. A., Stockwell, D. Z., Vanvelthoven, P. F. J., Verver, G., Wild, O., Yang, H., and Zimmermann, P.: Evaluation and intercomparison of global atmospheric transport models using Rn-222 and other short-lived tracers, *J. Geophys. Res.*, 102, 5953–5970, 1997.
- Lee, H. N. and Larsen, R. J.: Vertical diffusion in the lower atmosphere using aircraft measurements of ^{222}Rn , *J. Appl. Meteorol.*, 36, 1262–1270, 1997.
- Levin, I., Born, M., Cuntz, M., Langendoerfer, U., Mantsch, S., Naegler, T., Schmidt, M., Varlagin, A., Verclas, S., and Wagenbach, D.: Observations of atmospheric variability and soil exhalation rate of radon-222 at a russian forest site, *Tellus*, 54(B), 462–475, 2002.
- Moriizumi, J., Ohkura, T., Hirao, S., et al.: Continuous Observation of Atmospheric Rn-222 Concentrations for Analytic Basis of Atmospheric Transport in East Asia, *J. Nucl. Sci. Technol.*, Supplement 6, 173–179, 2008.
- Petroff, A., Mailliat, A., Amielh, M., and Anselmet, F.: Aerosol dry deposition on vegetative canopies. Part 1: Review of present

- knowledge, *Atmos. Environ.*, 42, 3625–3653, 2008.
- Porstendörfer, J.: Properties and Behaviour of radon and thoron and their decay products in the air, *J. Aerosol Sci.*, 25, 219–263, 1994.
- Schmidt, M., Graul, R., Sartorius, H., and Levin, I.: Carbon dioxide and methane in continental Europe: a climatology, and ^{222}Rn -based emission estimates, *Tellus*, 48(B), 457–473, 1996.
- Schmidt, M.: Messung und Bilanzierung anthropogener Treibhausgase in Deutschland, PhD Thesis, Univ. of Heidelberg, Heidelberg, Germany, 1999 (in German).
- Schmidt, M., Glatzel-Mattheier, H., Sartorius, H., Worthy, D. E., and Levin, I.: Western European N_2O emissions: A top-down approach based on atmospheric observations, *J. Geophys. Res.*, 106, 5507–5516, 2001.
- Schmidt, M., Graul, R., Sartorius, H., and Levin, I.: The Schauinsland CO_2 record: 30 years of continental observations and their implications for the variability of the European CO_2 budget, *J. Geophys. Res.*, 108, 4619, doi:10.1029/2002JD003085, 2003.
- Seibert, P. and Skomorowski, P.: Untersuchung der orographischen Besonderheiten der Probennahmestellen Schauinsland und Freiburg und deren Auswirkungen auf die Genauigkeit von ad-jungierten atmosphärischen Ausbreitungsrechnung („Einzugsgebiete“), Schriftenreihe Reaktorsicherheit und Strahlenschutz, BMU-2008-713, 2008.
- Stockburger, H. und Sittkus, A.: Unmittelbare Messung der natürlichen und künstlichen Radioaktivität der atmosphärischen Luft, *Zeitschrift für Naturforschung*, 21a, 1128–1132, 1966 (in German).
- Szegvary, T., Conen, F., and Ciais, P.: European ^{222}Rn inventory for applied atmospheric studies, *Atmos. Environ.*, 43, 1536–1539, 2009.
- Taguchi, S., Lida, T., and Moriizumi, J.: Evaluation of the atmospheric transport model NIRE-CTM-96 by using measured radon-222 concentrations, *Tellus*, 54(B), 250–268, 2002.
- Whittlestone, S. and Zahorowski, W.: Baseline radon detectors for shipboard use: Development and deployment in the First Aerosol Characterization Experiment (ACE 1), *J. Geophys. Res.*, 103(D13), 16743–16751, 1998.
- Wilson, S. R., Dick, A. L., Fraser, P. J., and Whittlestone, S.: Nitrous oxide flux estimates from South-East Australia, *J. Atmos. Chem.*, 26, 169–188, 1997.
- WMO/GAW 1st International Expert Meeting on Sources and Measurements of Natural Radionuclides Applied to Climate and Air Quality Studies, WMO TD, No. 1201, 2004.
- Wyers, G. P. and Veltkamp, A. C.: Dry deposition of ^{214}Pb to conifers, *Atmos. Environ.*, 31(3), 345–350, 1997.
- Volz-Thomas, A., Geiß, H., and Kalthoff, N.: The Schauinsland 1999. Ozone Precursor Experiment (SLOPE96): Scientific background and summary of main results, *J. Geophys. Res.*, 105(D1), 1553–1561, 1999.
- Yamada, K., Iida, T., Ikebe, Y., Miyachi, H., Nagao, I., and Komura, K.: Time series analysis of atmospheric ^{222}Rn concentrations and meteorological factors in JAPAN, Proceeding of the 7th Tohwa University International Symposium, 1998.
- Yamamoto, M., Kofugi, H., Shiraishi, K., and Igarashi, Y.: An attempt to evaluate dry deposition velocity of airborne ^{210}Pb in a forest ecosystem, *J. Radioanal. Nucl. Ch.*, 227, 81–87, 1998.