Atmos. Chem. Phys. Discuss., 14, 3667–3706, 2014
www.atmos-chem-phys-discuss.net/14/3667/2014/
doi:10.5194/acpd-14-3667-2014

Atmospheric Chemistry and Physics
Open Access
Discussions

Factors controlling temporal variability of near-ground atmospheric ^{222}Rn concentration over Central Europe

M. Zimnoch[1], P. Wach[1], L. Chmura[1,2], Z. Gorczyca[1], K. Rozanski[1], J. Godlowska[2], J. Mazur[3], K. Kozak[3], and A. Jeričević[4]

[1]AGH University of Science and Technology, Faculty of Physics and Applied Computer Science, Krakow, Poland
[2]Institute of Meteorology and Water Management, National Research Institute IMGW-PIB Branch of Krakow, Poland
[3]The Henryk Niewodniczanski Institute of Nuclear Physics, Polish Academy of Sciences, Krakow, Poland
[4]Croatian Civil Aviation Agency, Zagreb, Croatia

Received: 4 December 2013 – Accepted: 20 January 2014 – Published: 10 February 2014

Correspondence to: M. Zimnoch (zimnoch@agh.edu.pl)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Specific activity of ^{222}Rn in near-ground atmosphere has been measured quasi-continuously from January 2005 to December 2009 at two continental sites in Europe: Heidelberg (south-west Germany) and Krakow (southern Poland). Atmosphere was sampled at ca. 30 m and 20 m, respectively, above the local ground. Both stations were equipped with identical instrumentation. Regular observations of ^{222}Rn were supplemented by measurements of surface fluxes of this gas in Krakow urban area, using two entirely different approaches. Atmospheric ^{222}Rn concentrations varied at both sites in a wide range, from less than 2 Bq m^{-3} to approximately 40 Bq m^{-3} in Krakow and ca. 35 Bq m^{-3} in Heidelberg. Averaged over entire observation period, the ^{222}Rn content in Krakow was approximately 30 % higher when compared to Heidelberg (5.86 ± 0.09 Bq m^{-3} and 4.50 ± 0.07 Bq m^{-3}, respectively). Distinct seasonality of ^{222}Rn signal was visible in both presented time series, with higher values recorded generally during late summer and autumn. The surface ^{222}Rn fluxes in Krakow also revealed a distinct seasonality, with broad maximum observed during summer and early autumn and minimum during the winter. Averaged over 5 yr observation period, the night-time surface ^{222}Rn flux was equal 46.8 ± 2.4 Bq m^{-2} h^{-1}. Although the atmospheric ^{222}Rn levels at Heidelberg and Krakow appeared to be controlled primarily by local factors, it was possible to evaluate the "continental effect" in atmospheric ^{222}Rn content between both sites, related to the gradual build-up of ^{222}Rn concentration in the air masses travelling between Heidelberg and Krakow. The mean value of this load was equal 0.78 ± 0.12 Bq m^{-3}. The measured minimum ^{222}Rn concentrations at both sites and the difference between them was interpreted in the framework of a simple box model coupled with HYSPLIT analysis of air mass trajectories. Best fit of experimental and model data was obtained for the average ^{222}Rn flux over the European continent equal 52 Bq m^{-2} h^{-1}, the mean transport velocity of the air masses within convective mixed layer of PBL on their route from the Atlantic coast to Heidelberg and Krakow equal 3.5 m s^{-1}, the mean rate constant of ^{222}Rn removal across the top of PBL equal to the

ACPD
14, 3667–3706, 2014

^{222}Rn decay constant and the mean height of the convective mixed layer height equal 1600 m.

1 Introduction

Radon-222 is an alpha-emitting radioactive inert gas with the half-life of 3.8 days. It is a product of the decay of ^{226}Ra which belongs to ^{238}U-decay series. Uranium-238 and its decay product, ^{226}Ra, are ubiquitous in the Earth's crust and in the soils. Radon-222 is being released into the pore space of the soils and diffuses into the atmosphere where it decays to lead ^{210}Pb via a chain of intermediate decay products. The flux of ^{222}Rn into the atmosphere is controlled by the source term (^{226}Ra content in the soil and its vertical distribution), by physical properties of the upper soil layer (mineral structure, porosity, water content) and to some extent by short-term variations of physical parameters characterizing the soil–atmosphere interface (atmospheric temperature and pressure) (e.g. Levin et al., 2003; Taguchi et al., 2011). Due to lack of important sinks apart of radioactive decay, ^{222}Rn is an excellent tracer for evaluation of atmospheric chemistry and transport models (Dörr et al., 1983; Jacob et al., 1997; Chevillard et al., 2002; Gupta et al., 2004; Bergamaschi et al., 2006; Zhang et al., 2008). Radon is also an important factor contributing to the ionization rate of Earth's atmosphere (Zhang et al., 2011). In the past two decades ^{222}Rn has been frequently used to assess surface emissions of greenhouse trace gases such as CO_2, N_2O and CH_4 (Schmidt et al., 1996, 2003; Levin et al., 1999, 2003; Biraud et al., 2000; Conen et al., 2002; Wilson et al., 1997).

Concentration of ^{222}Rn in near-ground atmosphere has been measured quasi-continuously over a five-year period at two continental locations in Europe: Heidelberg, south-west Germany and Krakow, southern Poland. The measurement sites are located at the same latitudinal band (ca. 50° N) in a similar urban settling, with the distance to the Atlantic Ocean equal approximately 600 km for Heidelberg and 1600 km for Krakow. Both measurement sites were equipped with identical instrumentation (Levin

ACPD
14, 3667–3706, 2014

et al., 2002). The atmosphere was sampled at a comparable level, ca. 20 and 30 m above the local ground at Krakow and Heidelberg, respectively. Regular observations of ^{222}Rn in near-ground atmosphere were supplemented by measurements of local surface fluxes of this gas in Krakow urban area using two different approaches. The observed temporal variability of radon concentration on diurnal, synoptic and seasonal time scales is presented and thoroughly discussed, with emphasis on seasonal changes.

2 Measurement sites

Krakow (approx. 800 000 inhabitants) is located in Southern Poland. Characteristic features of the local climate are generally weak winds and frequent inversions, sometimes extending over several days. The average wind speed for the period 2005–2007 was around 1.9 m s^{-1}.West and south-west direction of surface winds prevails. Westerly circulation is generally connected with stronger winds (wind speeds above 4 m s^{-1}). Periods characterized by low wind speeds (< 1 m s^{-1}), favouring accumulation of ^{222}Rn in near-ground atmosphere, constituted ca. 34 % of the total time considered. Monthly air temperature at the site reveals a distinct seasonal cycle, with summer maximum (July–August) reaching 19–24 °C and winter minimum (January–February) between −5 and +2 °C. Monthly precipitation rates are more irregular, with a broad maximum during summer and minimum during winter months. Radon measurement site was situated on the University campus located in the western part of the city (50°4′ N, 19°55′ E, 220 m a.s.l.), bordering recreation and sports grounds. Air intake for ^{222}Rn measurements was located on the roof of the Faculty of Physics and Applied Computer Science building, ca. 20 m above the local ground.

Heidelberg (approximately 130 000 inhabitants) is located in the upper Rhine valley, in South-West Germany. Monthly surface air temperatures vary between 1–3 °C during winter months and 18–22 °C during the summer. The local atmospheric circulation patterns in Heidelberg are dominated by alternate north/south flow along the Rhine valley, but also by frequent easterly winds from the Neckar valley (Levin et al., 1999). The average wind speed for the period 2005–2007 was around 3 m s^{-1} while low wind speed periods (< 1 m s^{-1}), constituted only ca. 9.8 % of the total time considered. In contrast to local, near-surface wind direction, backward air trajectories calculated for Heidelberg site clearly demonstrate predominance of westerly air masses. Air inlet for ^{222}Rn measurements was installed on the roof of the Institute of Environmental Physics building (49°24′ N, 8°42′ E, 116 m a.s.l.), ca. 30 m above the local ground.

3 Methods

3.1 Measurements of ^{222}Rn content in near-ground atmosphere

Regular measurements of ^{222}Rn content in near-ground atmosphere were performed with the aid of Heidelberg radon monitor. The instrument was developed at the Institute of Environmental Physics, University of Heidelberg, Germany (Levin et al., 2002), and made available for this study. The instrument measures specific activity of ^{222}Rn in air through its daughter products. The air is pumped with the constant flow rate through a glass filter (Whatman QM-A, 2.2 µm) which is placed directly over surface barrier detector measuring alpha particles emitted by ^{222}Rn and ^{220}Rn daughter products deposited on the filter. The instrument records alpha spectra accumulating in 30 min counting intervals. The spectra contain peaks representing decay products of ^{222}Rn (^{214}Po, $E_\alpha = 7.7$ MeV and ^{218}Po, $E_\alpha = 6$ MeV) as well as ^{220}Rn (^{212}Po, $E_\alpha = 8.8$ MeV and ^{212}Bi, $E_\alpha = 6.1$ MeV). By energy discrimination and dedicated data evaluation protocol taking into account disequilibrium between daughter products of ^{222}Rn at the end of each counting interval as well as empirically determined disequilibrium between ^{222}Rn gas and its daughter products in the atmosphere, the specific activity of the ^{222}Rn gas in air can be calculated. The mean disequilibrium has been tentatively determined by parallel measurement of the ^{222}Rn gas activity with an absolutely calibrated

ACPD
14, 3667–3706, 2014

slow-pulse ionization chamber and the Radon monitor to 1/1.367 for 20 m above ground in Heidelberg (Levin, 2002).

In the framework of this study, readings of Heidelberg radon monitor were compared with measurements of ^{222}Rn activity performed with the aid of a commercial radon instrument (AlphaGUARD PQ2000 PRO® – Genitron Instruments GmbH, Germany) which is functioning as an ionization chamber and measures directly alpha particles of ^{222}Rn. The instrument recorded hourly mean ^{222}Rn concentrations. Both detectors were operated in Krakow over one-week period, measuring air collected ca. 20 m above the ground. The results are shown in Fig. 1. Although overall good agreement between readings of both instruments was obtained (correlation coefficient $R^2 = 0.78$), it is apparent that the radon monitor does not fully capture fast changes of ^{222}Rn content in the atmosphere. This is expected for this indirect detection method employed by the instrument based on ^{222}Rn decay products and a constant disequilibrium factor between ^{222}Rn and the daughter products in air employed in the data evaluation protocol.

3.2 Measurements of surface fluxes of soil ^{222}Rn

Two different approaches were used to quantify the magnitude and temporal variability of surface fluxes of ^{222}Rn into the local atmosphere: (i) night-time ^{222}Rn fluxes were derived from measurements of atmospheric ^{222}Rn content near the ground, combined with quasi-continuous measurements of the mixing layer height within the Planetary Boundary Layer (PBL) and modelling of vertical ^{222}Rn profiles in the atmosphere using regional transport model, and (ii) point measurements of soil ^{222}Rn fluxes were performed using specially designed exhalation chamber system connected to AlphaGUARD detector. Both methods were used in parallel to derive night-time ^{222}Rn fluxes during the period from September 2005 to September 2006.

3.2.1 Sodar-assisted estimates of night-time ^{222}Rn fluxes

During the day, with active vertical convection of air in the lower atmosphere, radon emitted from the soil is dissolved in a large volume of mixing layer within PBL, leading to relatively low ^{222}Rn concentrations observed close to the ground. During late afternoon, when the vertical gradient of air temperature changes the sign, drastic reduction of vertical mixing occurs. This process leads to accumulation of ^{222}Rn in near-ground atmosphere during the night. The rate of nocturnal increase of ^{222}Rn concentration is controlled by the mixing layer height according to the mass balance equation:

$$H\frac{dC_m}{dt} = F_{in} - F_{out} \quad (1)$$

where:
H – height of the mixing layer
C_m – mean concentration of ^{222}Rn within the mixing layer
F_{in} – surface flux ^{222}Rn
F_{out} – flux of ^{222}Rn associated with removal processes (horizontal and vertical transport, radioactive decay). For nights with low wind speed ($< 1\,m\,s^{-1}$) and the adopted frequency of measurements, this term can be neglected.

During stable atmospheric conditions with low wind speeds, a distinct vertical gradient of ^{222}Rn concentration is established within the PBL. As the measurements of ^{222}Rn content are performed close to the surface, at the height of ca. 20 m, a correction factor k relating the increase of the mean ^{222}Rn concentration within the mixing layer (dC_m/dt) to the increase of the concentration observed close to the ground (dC_{surf}/dt) should be introduced to Eq. (1):

$$F_{in} = \frac{H}{k}\frac{dC_{surf}}{dt} \quad (2)$$

where:
H – mixing layer height

ACPD
14, 3667–3706, 2014

k– correction factor
C_{surf} – concentration of ^{222}Rn at the adopted measurement height.

The correction factor k was determined using vertical profiles of ^{222}Rn simulated by EMEP model. The Unified EMEP model (http://www.emep.int/) was developed at the Norwegian Meteorological Institute under the EMEP programme. In this work, the Unified EMEP model version rv2_6_1 was used. The model is fully documented in Simpson et al. (2012). It simulates the atmospheric transport and deposition of various trace compounds, as well as photo-oxidants and particulate matter over Europe. The Unified EMEP model uses 3 hourly meteorological data from PARallel Limited Area Model with the Polar Stereographic map projection (PARLAM-PS), which is a dedicated version of the HIgh Resolution Limited Area Model (HIRLAM) model for the use within EMEP. The model has been extensively validated against measurements (e.g. Jeričević et al., 2010). In the framework of the presented study the model was adopted to calculate regional transport of ^{222}Rn. The model domain (Fig. 2) covers Europe and the Atlantic Ocean with the grid size of 50 × 50 km and 20 layers in vertical, reaching up to 100 hPa.

The Unified EMEP model was used to simulate vertical profiles of ^{222}Rn concentrations in the atmosphere for the location of Krakow with hourly time resolution, assuming spatially constant exhalation rate of 1 atom $cm^{-2}\,s^{-1}$ over the continent for one full year (2005). Model results corresponding to the periods starting at sunset and ending 6 h later for each night were used. ^{222}Rn concentration at the measurement height (20 m above the ground.) was evaluated from the simulated profiles using exponential fit to the model data. The mean radon concentration in the mixing layer was calculated as the mean ^{222}Rn content in the lowest five layers of the model representing approximately first 600 m of the troposphere, weighted by the thickness of the corresponding layers:

$$C_m = \frac{\sum_{i=1}^{5} C_i h_i}{\sum_{i=1}^{5} h_i} \qquad (3)$$

Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper |
ACPD
14, 3667–3706, 2014

where:

C_m – modelled mean concentration of ^{222}Rn within the mixing layer,

C_i – mean ^{222}Rn concentration in the layer i (model data),

h_i – thickness of the layer i.

In the next step, the growth rates of the mean ^{222}Rn concentration within the mixing layer (ΔC_m) and at 20 m above the ground (ΔC_{surf}) were calculated for each considered period using linear regression fit to the data. Finally, k factor was calculated for each night:

$$k = \frac{\Delta C_{surf}}{\Delta C_m} \tag{4}$$

The monthly means of k values were calculated using the values of this parameter assigned for each night, after a two-step selection procedure. In the first step, only the periods characterized by well-defined growth rates ΔC_m and ΔC_{surf} ($R^2 > 0.8$) were selected. In the second step, periods characterized by high wind speeds within the first layer of the model ($v > 3\,m\,s^{-1}$) were removed. The mean monthly k values calculated using the above-outlined procedure are presented in Fig. 3 together with their uncertainties. The k value for January 2005 is missing because no single night in this month fulfilled the adopted selection criteria.

Since model results were available only for 2005, the monthly means of k values presented in Fig. 3 were further smoothed using CCGvu 4.40 routine (Thoning et al., 1989). The smoothed curve (heavy line in Fig. 3) was then used in calculations of surface fluxes of ^{222}Rn using Eq. (2), for the period June 2004–May 2009 (cf. Sect. 4.4).

The mixing layer height, H, was monitored using VDS sodar (Version 3) built by the Krakow Branch of the Institute of Meteorology and Water Management and operated in Krakow since 1994. Detailed description of the sodar system can be found in Netzel et al. (1995). Sodar records were analyzed manually. Stability of the surface layer was identified through unique features of sodar echoes and its range was defined by determining the height of the observed structures at the upper level where mixing processes

ACPD
14, 3667–3706, 2014

still exist (Piringer and Joffre, 2005). The sodar system was located inside a park complex, between the city centre and the industrial district, at the distance of ca. 6 km east from the location where atmospheric ^{222}Rn measurements were performed. To examine possible influence of the distance between the sodar and ^{222}Rn measurement sites on the calculated ^{222}Rn fluxes, two measurement campaigns were performed. During the first campaign lasting one month, the ^{222}Rn measurements were performed directly at the sodar site. Then, the sodar system was moved to the permanent location of the ^{222}Rn monitoring system. During both campaigns neither significant change in the mixing layer height variability nor the range of calculated fluxes were observed (Zimnoch et al., 2010).

3.2.2 Measurements of soil ^{222}Rn fluxes using chamber method

Sodar-assisted assessment of surface nigh-time ^{222}Rn fluxes in Krakow agglomeration during the period 2005–2009 was supplemented by point measurements of soil radon fluxes using the in-growth chamber method. The measurement site was located on the premises of the Institute of Nuclear Physics, Polish Academy of Sciences, situated in the western outskirts of the city, approximately 3 km north-west from the location of atmospheric ^{222}Rn measurements. The soil type at the chamber location was Endogleyic Cambisol (IUSS, 2007), dominated by silty clay loam. The mean concentration of ^{226}Ra in the soil profile, the precursor of ^{222}Rn, was equal $22 \pm 3\,\mathrm{Bq\,kg^{-1}}$ (Mazur, 2008).

Radon fluxes from the soil were measured using specially designed, automatic exhalation chamber, connected with AlphaGUARD monitor. The air trapped inside the chamber was circulated in a closed circuit for about 90 min and the concentration of radon accumulated in the chamber was recorded every 10 min. The radon flux was determined from the slope of the straight line fitted to individual measurement data (Mazur, 2008; Vaupotič et al., 2010). Special device was constructed which enabled automatic movement of the exhalation chamber, down to the soil for radon flux measurement and up for ventilation of the system (Mazur, 2008). The soil ^{222}Rn fluxes were measured up to eight times per day.

ACPD
14, 3667–3706, 2014

4 Results and discussion

Time series of hourly mean specific activity of atmospheric ^{222}Rn obtained at Heidelberg and Krakow between January 2005 and December 2009 are shown in Fig. 4. The gaps in time series of ^{222}Rn content from June till December 2007 in Krakow and in June–July 2006 in Heidelberg were caused by technical problems with radon monitors. It is apparent from Fig. 4 that ^{222}Rn content varies at both sites in a relatively wide range, from less than 2 Bq m^{-3} to approximately 40 Bq m^{-3} in Krakow and ca. 35 Bq m^{-3} in Heidelberg. The ^{222}Rn concentration in Krakow averaged over entire observation period (5.86 ± 0.09 Bq m^{-3}), is approximately 30 % higher when compared to Heidelberg (4.50 ± 0.07 Bq m^{-3} – cf. Table 1). Distinct seasonality of ^{222}Rn signal is visible in both presented time series, with higher values recorded during late summer and autumn.

4.1 Diurnal changes of ^{222}Rn content in near-ground atmosphere

Diurnal changes of specific activity of ^{222}Rn in near-ground atmosphere over Krakow and Heidelberg, averaged over entire observation period (January 2005–December 2009), separately for each season (spring, summer, autumn, winter), are summarized in Fig. 5a and b. Irrespectively of season, the measured ^{222}Rn content at both sites reveals characteristic behaviour, with elevated concentrations during night hours and reduced concentrations during mid-day. However, the shape and amplitude of daily changes of ^{222}Rn content varies significantly with the season and the observation site.

During winter months (December–February), daily variations of ^{222}Rn are remarkably similar at both locations. The average ^{222}Rn content for that period is equal 5.65 ± 0.17 Bq m^{-3} at Krakow and 5.19 ± 0.19 Bq m^{-3} at Heidelberg, with peak-to-peak amplitude of diurnal changes reaching ca. 1.5 Bq m^{-3} at both sites (cf. Fig. 5 and Table 1). Daily minima are shallow and reduced in duration. During spring and summer months (March–May and June–August, respectively) the peak-to-peak amplitude of daily ^{222}Rn variations increases significantly. This increase is particularly well

ACPD
14, 3667–3706, 2014

pronounced in Krakow (amplitudes equal 4.7 and 7.0 $Bq\,m^{-3}$, respectively) when compared to Heidelberg (ca. 2.5 and 3.5 $Bq\,m^{-3}$, respectively). At the same time, broader daily minima are observed at both sites, reflecting growing role of vertical mixing within the PBL, driven by longer exposure of the surface to the sunlight. Most pronounced differences between both sites are observed during autumn months (September–November). In Krakow, the maximum values of atmospheric ^{222}Rn content occur usually in early-morning hours (ca. 4–6 a.m.) and reach approximately 12 $Bq\,m^{-3}$, followed by a distinct minimum of ca. 5 $Bq\,m^{-3}$ recorded usually between 1 and 2 p.m. In Heidelberg, the early morning maximum reaches only ca. 8 $Bq\,m^{-3}$, while the minimum is maintained at approximately the same level as in Krakow (ca. 5 $Bq\,m^{-3}$).

Diurnal variations of ^{222}Rn content at both measurement sites, averaged over entire observation period, are shown in Fig. 5c. It is apparent that higher peak-to-peak amplitude of daily ^{222}Rn variations in Krakow is primarily due to higher build-up of ^{222}Rn during night hours at this site when compared to Heidelberg. The nocturnal build-up of radon in Krakow starts and ends up ca. 2 h earlier when compared to Heidelberg (approximately at 3 p.m. and 4 a.m., respectively). This difference may partly stem from the fact that while both stations are located in the same time zone, the true solar time is shifted by ca. 1 h.

The source strength of ^{222}Rn in the soil and the upward transport of this gas into the atmosphere should not vary significantly on hourly time scale, except perhaps frontal situations with relatively fast changes of atmospheric pressure or during prolonged rainfall events. Therefore, diurnal variations of the concentration of this gas in near-ground atmosphere presented in Fig. 5 should primarily reflect changes in the stability of the lower troposphere and the resulting intensity of vertical mixing.

4.2 Synoptic changes of ^{222}Rn content in near-ground atmosphere

While the amplitude of diurnal changes of ^{222}Rn content seems to be controlled primarily by intensity of vertical mixing, its variations on synoptic time scale (days to weeks) should be also a function of the origin of air masses passing through the given measurement site. One example illustrating the influence of synoptic-scale weather patterns on atmospheric ^{222}Rn content in Krakow is discussed below.

Temporal evolution of atmospheric ^{222}Rn content in Krakow during the period from 1 to 13 March 2005, is shown in Fig. 6 in relation to wind speed, precipitation rates and 96 h backward trajectories of air masses passing the measurement location. Between 1 and 4 March, the measured ^{222}Rn content revealed strong diurnal variations (Fig. 6a), with maximum ^{222}Rn concentrations reaching ca. 16 Bq m^{-3}, superimposed on the growing trend of mean daily concentration of this gas (Fig. 6b). Air masses passing the sampling site during this period originated in North-Central Europe (Fig. 6c). Very low wind speed facilitated gradual build-up of night-time ^{222}Rn in the local atmosphere during these days as well as an increase of day-time background level caused by increase in "continentality" of the air masses. From 5 March onwards, the course of atmospheric ^{222}Rn content has changed radically: diurnal variations almost disappeared (Fig. 6a) while the average daily ^{222}Rn content dropped from ca. 9.7 Bq m^{-3} for 4 March to 5.2 Bq m^{-3} for 5 March and 3.3 Bq m^{-3} for 6 March. This substantial change was clearly linked to change of circulation and sharp increase of wind speed (Fig. 6b and c). Between 5 and 7 March the sampling location was under influence of air masses originating over Russia and Estonia, passing westward at relatively high elevation over the Baltic Sea and then turning south-east in the direction of Poland. Between 8 and 12 March the sampling location was under influence of maritime air masses originating over the Arctic Sea and travelling southward at very high elevation with high speed, bringing precipitation in Krakow (mean daily rainfall between 0.5 and 2.0 mm). During the second part of this period, small increase of ^{222}Rn concentration correlated with significant reduction of trajectory elevation is observed.

The data shown in Fig. 6 clearly demonstrate a strong link between the measured ^{222}Rn content in near-ground continental atmosphere and weather-related phenomena such as history of air masses and wind speed. Concentrations of atmospheric ^{222}Rn may change dramatically on daily time scale in response to these factors.

ACPD
14, 3667–3706, 2014

4.3 Seasonal variations of ^{222}Rn content in near-ground atmosphere

Figure 7 shows monthly means of ^{222}Rn content in near-ground atmosphere, as observed in Krakow and Heidelberg between January 2005 and December 2009. The medians reveal distinct seasonal trend, with a broad minimum of ^{222}Rn content in spring and summer and maximum in autumn months. Monthly maxima are significantly higher in Krakow, reaching ca. 25 Bq m^{-3}, compared to approximately 20 Bq m^{-3} in Heidelberg. Monthly minima are comparable at both locations.

4.4 Surface fluxes of ^{222}Rn in Krakow

Using the methodology outlined in Sect. 3.2.1, the night-time fluxes of ^{222}Rn in Krakow were assessed for the period from June 2004 to May 2009. Each period considered in the calculations started typically at sunset and ended 6 h later. Average height of the mixing layer was calculated from hourly sodar data for each analyzed night. Individual nigh-time fluxes of ^{222}Rn calculated using Eq. (2) were subject to two-step data selection algorithm. In the first step, only the nights for which the growth rate of the measured ^{222}Rn concentration was well-defined ($R^2 > 0.8$), were selected. In the second step, the nights with standard deviation of the mean mixing layer height calculated from hourly values of this parameter derived from sodar measurements, larger than 30 m, were removed from the remaining dataset. Finally, the monthly mean values of ^{222}Rn flux were calculated for the selected nights (Fig. 8). For some months, the adopted data selection procedure resulted in significant reduction of the number of available data, leading to relatively large uncertainty of the corresponding monthly mean values of the calculated ^{222}Rn fluxes. The number of data used for calculation of the mean for each month varied between 5 and 23 with the mean value of 13.

As seen in Fig. 8, high monthly means of night-time fluxes of ^{222}Rn were obtained for autumn months (September, October, November), with the maxima reaching 80–90 Bq m^{-2} h^{-1}). Averaged over entire observation period, the mean ^{222}Rn flux is equal 46.8 ± 2.4 Bq m^{-2} h^{-1}, close to the value of ca. 46 Bq m^{-2} h^{-1} derived for the location of

Krakow from the map of ^{222}Rn flux in Europe obtained from terrestrial γ-dose rate data (Szegvary et al., 2009).

Direct measurements ^{222}Rn exhalation rates from the soil using the methodology described in Sect. 3.2.2 were performed over one-year period, from September 2005 till September 2006. Between 4 and 8 measurements have been conducted daily at the experimental plot chosen for observations. To allow direct comparison with the ^{222}Rn fluxes derived from parallel measurements of atmospheric ^{222}Rn content and observations of the mixing layer height, only the measurements performed during the days for which sodar-assisted ^{222}Rn fluxes were available, were considered. The comparison of ^{222}Rn fluxes derived using both approaches is shown in Fig. 9. The mean ^{222}Rn flux derived from chamber measurements ($50.3 \pm 8.4\,\mathrm{Bq\,m^{-2}\,h^{-1}}$) agrees very well with sodar-assisted estimate of this flux ($50.3 \pm 3.4\,\mathrm{Bq\,m^{-2}\,h^{-1}}$). A large discrepancy is apparent for February and March 2006, for which the chamber measurements differ from sodar-assisted estimates of the ^{222}Rn flux by a factor of ten. This large discrepancy may stem from the fact that winter conditions (frost, snow cover) may partly block the ^{222}Rn flux from the soil, resulting in low readings of the chamber method, whereas the sodar-assisted estimates of the ^{222}Rn flux are spatial averages over the footprint area of atmospheric ^{222}Rn measurements which is in the order of several square kilometres. In addition, these estimates include also ^{222}Rn emitted by building infrastructure of the city which is not the case for the chamber method. Thus, direct comparison of both methods may not be fully justified.

4.5 Factors controlling seasonality of ^{222}Rn content in near-ground atmosphere over Central Europe

Distinct seasonality of atmospheric ^{222}Rn concentrations apparent in the data presented in Figs. 4 and 7 may have its roots in several processes: (i) it may reflect seasonal bias in the origin of air masses arriving at the given measurement location e.g. prevalence of maritime air masses with low ^{222}Rn during summer and of continental air

ACPD
14, 3667–3706, 2014

masses with higher average ^{222}Rn content during autumn and winter, (ii) it may reflect seasonal bias in the stability of the lower atmosphere, with more frequent, prolonged inversions during autumn and winter when compared to summer, and, finally (iii) it may reflect seasonality in the source term i.e. seasonally varying ^{222}Rn exhalation rates from the soil. All these three factors may of course act together.

Backward trajectory analysis has been performed for Krakow and Heidelberg for the period from January 2005 till December 2009 using HYSPLIT model (Draxler and Rolph, 2011). 96 h backward trajectories were calculated for every hour within this time period. Multiple trajectories were displayed by creating an arbitrary grid over the computational domain, counting number of trajectories over each grid point (without multiple intersections of the same trajectory) and dividing by the total number of trajectories. The plotting routine was used (Rolph, 2011) to display spatial distribution of trajectory densities over the computational domain. The grid resolution has been set to one by one degree. The results were averaged separately for each season and each site and presented in Fig. 10. Yellow colour represents grid points where more that 10 % of all trajectories passed through. Blue, green and cyan colours represent trajectory density range between 5 and 10 %, 2 and 5 % and 1 and 2 %, respectively.

It is apparent from Fig. 10 that only minor seasonal variations occur in the spatial extension and shape of the trajectory density distribution maps for 94 h backward trajectories arriving in Krakow and Heidelberg. The contours are generally skewed towards W–E axis, reflecting dominance of westerly circulation. There are two distinct differences between Heidelberg and Krakow: (i) Heidelberg receives significantly higher proportion of maritime air masses originating over North Atlantic and arriving at the site within 96 h, when compared to Krakow, and (ii) the surface area of the 1 % contour map is larger for Heidelberg that for Krakow, indicating generally higher transport velocities and larger spatial extensions of 96 h backward trajectories for this site.

In order to better understand the factors controlling the observed seasonality of atmospheric ^{222}Rn levels at Krakow and Heidelberg the available data, averaged separately for each month and for entire observation period, were compared with other on-site

ACPD
14, 3667–3706, 2014

parameters which may influence this apparent seasonality (Fig. 11). Figure 11a shows monthly distribution of ^{222}Rn content in Krakow near-ground atmosphere, presented in the form of box-and-whisker plot for the entire observation period from January 2005 till December 2009. The monthly mean ^{222}Rn content shows a broad maximum in September-October-November. The long-term monthly minima of ^{222}Rn, as observed in Krakow and Heidelberg, are shown in Fig. 11b. Within the quoted uncertainties of the mean values they are indistinguishable at both sites in March, April and May. While the monthly ^{222}Rn minima are practically constant from March till August in Heidelberg, in Krakow they start to increase already in June. The monthly minima peak in October and November, for Heidelberg and Krakow, respectively.

Distinct seasonality observed in atmospheric ^{222}Rn concentration could be linked to the seasonality of ^{222}Rn emissions from the ground. Monthly mean night-time ^{222}Rn fluxes, as estimated for Krakow and averaged over entire observation period (cf. Fig. 8), are shown in Fig. 11c. Figure 11d shows the monthly means of ^{222}Rn fluxes derived from chamber measurements performed up to eight times per day in Krakow and averaged over the period from September 2005 till September 2006. The monthly night-time ^{222}Rn fluxes reveal a broad maximum during summer and early autumn (June–October). Although the mean monthly ^{222}Rn fluxes derived from chamber measurements are more variable, they also reveal a broad maximum during summer and early autumn. Very low ^{222}Rn flux values observed in February and March could result from local conditions at the measurement site. Comparison of Fig. 11a and b with Fig. 11c and d shows that the apparent seasonality in ^{222}Rn emissions from the ground cannot fully account for the observed seasonality of the ^{222}Rn concentrations in near-ground atmosphere. Also other factors should be involved.

Monthly means of surface air temperature recorded in Krakow, averaged over the period from January 2005 till December 2009, are shown in Fig. 11e. While the air temperature reveals distinct seasonal changes (the difference between coldest and warmest month is larger than 20 °C), monthly precipitation data are more variable and the seasonality is less marked. Comparison of local air and soil temperatures (four

ACPD
14, 3667–3706, 2014

different depths) is shown in Fig. 11f. It is worth to note that during autumn and winter soils are generally warmer than air, thus making the soil air unstable. This may facilitate transport of ^{222}Rn from the soil to the atmosphere.

The last two graphs in Fig. 11 (Fig. 11g and h) show seasonal variations of two parameters characterizing stability of the lower atmosphere in Krakow: the wind speed and the duration of inversion episodes. In Fig. 11g monthly means of the percentage of still periods (wind speed below 1 m s^{-1}), averaged over entire observation period (January 2005–December 2009), is shown. It is apparent that percentage of periods with wind speed below 1 m s^{-1} is highest in Krakow during autumn and winter. Also, the duration of inversion periods is highest during that time (October–February), contributing to enhanced stability of the lower atmosphere. Although the data presented in Fig. 11h refer to another 5 yr period (January 1994–December 1999), it is believed that this specific feature of the local atmosphere in Krakow is valid also for the period considered in this study (January 2005–December 2009).

4.6 Assessment of build-up of ^{222}Rn content over the European continent based on mass balance in the convective mixed layer

Fluxes of ^{222}Rn over the ocean are two to three orders of magnitude smaller than those over the continents (Schery and Huang, 2004). Consequently, maritime air masses entering the continent will have very low ^{222}Rn content and will be gradually laden with ^{222}Rn until new equilibrium is reached. Since both ^{222}Rn observation sites discussed in this study are located in the same latitudinal band (ca. 50° N) and are exposed to westerly circulation, with Heidelberg being situated ca. 600 km from the Atlantic coast and Krakow approximately 1000 km further inland, it was of interest to quantify the extent of ^{222}Rn build-up in the air masses on their way from Heidelberg to Krakow. The evolution of ^{222}Rn content within the PBL can be described by simple mass balance

ACPD
14, 3667–3706, 2014

equation:

$$\frac{dC_{Rn}}{dt} = S_{Rn} - (\lambda_d + \lambda_e)C_{Rn} \quad (5)$$

where:

C_{Rn} – concentration of ^{222}Rn in the convective mixed layer of PBL;

S_{Rn} – source term linked to the surface flux of ^{222}Rn.

$S_{Rn} = F_{Rn}/h$ where F_{Rn} is the surface flux of ^{222}Rn and h is the height of the convective mixed layer;

λ_d – decay constant of ^{222}Rn;

λ_e – rate constant associated with removal of ^{222}Rn across the PBL boundary.

Equation (5) implies perfect mixing within the PBL and assumes that the net exchange of ^{222}Rn due to horizontal transport perpendicular to the direction of air mass movement is equal zero. It has to be noted that convective mixed layer referred to in Eq. (5) extends to PBL boundary while the nocturnal mixing layer discussed in Sect. 3.2.1 is generally less extensive ($H < h$) and occupies only part of PBL (Stull, 1988). Analytical solution of Eq. (5), with the initial condition $C_{Rn}(o) = 0$, constant surface flux of ^{222}Rn and constant height of the convective mixed layer reads as follows:

$$C_{Rn}(t) = \frac{S_{Rn}}{\lambda_d + \lambda_e}\left(1 - e^{-(\lambda_d+\lambda_e)t}\right) \quad (6)$$

Substituting $S_{Rn} = F_{Rn}/h$ and $T_{eff} = 0.693/(\lambda_d + \lambda_e)$ it follows:

$$C_{Rn}(t) = \frac{F_{Rn} \cdot T_{eff}}{h \cdot 0.693}\left(1 - e^{-\frac{0.693}{T_{eff}}t}\right) \quad (7)$$

Equation (7) can be used to calculate the expected build-up of ^{222}Rn content within PBL for maritime air masses travelling eastward from the Atlantic Ocean towards Heidelberg and Krakow. It is apparent from the preceding discussion that local effects are decisive

ACPD
14, 3667–3706, 2014

in shaping up atmospheric ^{222}Rn levels at both sites. Therefore, careful screening and selection of ^{222}Rn data had to be performed before an attempt is made to quantify the "continental effect" between both sites.

In the first step, only trajectories arriving in Krakow, which also passed Heidelberg at the distance of less then 100 km were selected for further analysis (Fig. 12). Out of this population of trajectories, only those which have been observed in Krakow and in the vicinity of Heidelberg for at least 24 h, were selected. Finally, only the data representing minimum ^{222}Rn concentrations measured during these periods at both sites were considered, taking into account time lag due to atmospheric transport from Heidelberg to Krakow. The selected minimum ^{222}Rn concentrations (94 pairs), averaged over entire observation period (January 2005–December 2009) separately for each station are equal 1.12 ± 0.09 and $1.90 \pm 0.11\,\mathrm{Bq\,m^{-3}}$, for Heidelberg and Krakow, respectively. The difference ($0.78 \pm 0.12\,\mathrm{Bq\,m^{-3}}$) represents mean build-up of ^{222}Rn content between Heidelberg and Krakow.

Sensitivity analysis of Eq. (7) was made with an attempt to fit simultaneously three values: the measured mean minimum ^{222}Rn concentrations at both sites and the difference between them. Three parameters in Eq. (7) were treated as adjustable parameters: (i) the mean transport velocity (V) of the air masses within convective mixed layer of PBL, on their route from the Atlantic coast to Heidelberg and Krakow, (ii) the mean surface flux of ^{222}Rn (F_{Rn}) on the continent, and (iii) the rate constant of ^{222}Rn removal across the top of PBL (λ_e). Since HYSPLIT routine returns also the mixing height of the calculated backward trajectories, it was possible to derive the value of this parameter directly from the model output. The mixing heights were calculated for all trajectories which satisfied the selection criteria outlined above and the maximum mixing height for each trajectory was selected. Averaged over all trajectories considered in the calculation, this procedure yield the mean value of the convective mixed layer height equal 1083 ± 31 m. In calculations 1100 m was adopted. The selection of 94 h trajectories impose a lower limit on the velocity of air masses travelling from the Atlantic coast to Heidelberg and Krakow, equal ca. $2.8\,\mathrm{m\,s^{-1}}$. The values of transport

ACPD
14, 3667–3706, 2014

velocities used in sensitivity analysis varied between 3.0 and 4.0 $m s^{-1}$. It appeared that while the calculated ^{222}Rn concentrations at Heidelberg and Krakow depend on the assumed transport velocity, the difference between them is relatively insensitive to the actual value of this parameter. The rate constant of radon removal across the top of PBL (λ_e) and the mean surface ^{222}Rn flux varied in the calculations from $\lambda_e = 0.5\lambda_d$ to $\lambda_e = 1.5\lambda_d$ and from 30 to 60 $Bq m^{-2} h^{-1}$, respectively.

Goodness of the fitting procedure was quantified by calculating the sum of squared differences ($\Sigma(C_{Rn(m)} - C_{Rn(c)})^2$) between the measured ($C_{Rn(m)}$) and calculated ($C_{Rn(c)}$) mean minimum ^{222}Rn concentration at Heidelberg and Krakow, and the difference between them. The best fit ($\Sigma = 9.9 \times 10^{-4}$) was obtained for the following combination of the adjusted parameters: $V = 3.5\, m s^{-1}$, $F_{Rn} = 36\, Bq m^{-2} h^{-1}$ and $\lambda_e = \lambda_d$. Similarity between the rate constant of radon removal across the top of PBL and the decay constant of ^{222}Rn was suggested also by Lui et al. (1984). The mean ^{222}Rn surface flux obtained through the fitting procedure (36 $Bq m^{-2} h^{-1}$) is lower than the mean annual ^{222}Rn flux obtained for Krakow urban area in the framework of this study (ca. 47 $Bq m^{-2} h^{-1}$) and is significantly lower than the annual mean ^{222}Rn flux (ca. 57 $Bq m^{-2} h^{-1}$) estimated for Heidelberg area from long-term flux measurements at five locations with different soil texture (Schmidt et al., 2003). If the mean surface ^{222}Rn flux of 52 $Bq m^{-2} h^{-1}$ is assumed (arithmetic average of Heidelberg and Krakow best estimates of this parameter), an equally good fit of the measured minimum ^{222}Rn concentrations at both stations and the difference between them is also possible, although with significantly larger height of the convective mixed layer ($h = 1600$ m).

5 Conclusions

Systematic observations of ^{222}Rn content in near-ground atmosphere at two continental sites in Europe with similar characteristics, supplemented by measurements of surface ^{222}Rn fluxes, allowed a deeper insight into factors controlling spatial and temporal

ACPD
14, 3667–3706, 2014

variability of ^{222}Rn in near-ground atmosphere over Central Europe. The available data allowed to address the role of local and regional factors in controlling the observed atmospheric ^{222}Rn levels and their variability.

Atmospheric concentrations of ^{222}Rn at both observation sites vary on daily, synoptic and monthly time scales. Generally higher and more variable ^{222}Rn levels recorded in Krakow are mainly due to specific characteristics of the local atmosphere, such as lower wind speed and more frequent inversion periods of prolonged duration when compared to Heidelberg, leading to enhanced stability of the lower atmosphere at this observation site.

The surface ^{222}Rn fluxes estimated in the framework of this study reveal distinct seasonality, with a broad maximum during summer and early autumn and minimum during winter. The nigh-time ^{222}Rn flux in Krakow averaged over 5 yr observation period was equal approximately 48 Bq m^{-2} h^{-1}. Theoretical considerations (e.g. Nazaroff, 1992; Sasaki et al., 2004) as well as large body of experimental data (e.g. Rogers and Nielson, 1991; Greeman and Rose, 1996; Levin et al., 2002; Papachristodoulou et al., 2007; Sakoda et al., 2010) suggest that free pore space in the soil available for diffusion-controlled transport of gaseous ^{222}Rn exerts primary control over ^{222}Rn exhalation rates. This parameter can be approximated by volumetric water content in the soil profile. The maps of soil moisture available for the European continent (http://edo.jrc.ec.europa.eu) reveal generally lower values of this parameter during summer and autumn when compared to winter and spring, for large parts of continental Europe. This suggest that distinct seasonality of ^{222}Rn exhalation rates, as observed on the European continent, may have its origin in seasonal changes of moisture load in the upper soil level.

The presented data reveal a distinct asymmetry in the shape of seasonal variations of surface ^{222}Rn fluxes and ^{222}Rn levels in the local atmosphere of Krakow. While atmospheric ^{222}Rn content peaks in November, the ^{222}Rn exhalation rates reach their maximum in September–October. This distinct phase shift stems most probably from increased stability of the lower atmosphere during autumn months (higher percentage

ACPD
14, 3667–3706, 2014

of still periods, longer duration of ground-based inversion episodes). These factors may collectively lead to the observed ^{222}Rn maximum in the local atmosphere in November, despite of already weakening soil ^{222}Rn flux at that time of the year.

Although the atmospheric ^{222}Rn levels at Heidelberg and Krakow appeared to be controlled primarily by local factors, it was nevertheless possible to evaluate the "continental effect" in atmospheric ^{222}Rn content between both sites, related to gradual build-up of ^{222}Rn load of maritime air masses travelling eastward over the European continent. Satisfactory agreement obtained between the measured minimum ^{222}Rn concentrations at both sites and the difference between them, as compared to the modelled values derived from a simple box model coupled with HYSPLIT analysis of air mass trajectories, allowed to put some constraints on the parameters of atmospheric ^{222}Rn transport over the European continent and its surface fluxes.

Acknowledgements. The authors would like to thank I. Levin from the Institute of Environmental Physics, University of Heidelberg, for providing the radon monitor and the experimental data for Heidelberg, as well as for thoughtful comments on the early version of the manuscript. The work has been partially supported by EU projects EUROHYDROS and CARBOUEROPE, grants of the Ministry of Science and Higher Education (projects no. 2256/B/P01/2007/33 and 4132/B/T02/2008/43), and the statutory funds of the AGH University of Science and Technology (project no. 11.11.220.01). The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and READY website (http://www.arl.noaa.gov/ready.php) used in this publication.

References

Bergamaschi, P., Meirink, J. F., Muller, J. F., Körner, S., Heimann, M., Dlugokencky, E. J., Kaminski, U., Marcazzan, G., Vecchi, R., Meinhardt, F., Ramonet, M., Sartorius, H., and Zahorowski, W.: Model Inter-comparison on Transport and Chemistry – Report on Model Inter-comparison Performed Within European Commission FP5 Project EVERGREEN ("Global Satellite Observation of Greenhouse Gas Emissions"), European Commission, DG Joint Research Centre, Institute for Environment and Sustainability, 53 pp., 2006.

ACPD
14, 3667–3706, 2014

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 measurements and radon 222 at Mace Head, Ireland, J. Geophys. Res., 105, 1351–1366, doi:10.1029/1999JD900821, 2000.

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 B, 54, 850–871, 2002.

Conen, F., Neftel, A., Schmid, M., and Lehmann, B. E.: $N_2O/^{222}$Rn-soil flux calibration in the stable nocturnal surface layer, Geophys. Res. Lett., 29, 1025, doi:10.1029/2001GL013429, 2002.

Dörr, H., Kromer, B., Levin, I., Münnich, K. O., and Volpp, J.-J.: CO_2 and ^{222}Rn as tracers for atmospheric transport, J. Geophys. Res., 88, 1309–1313, 1983.

Draxler, R. R. and Rolph, G. D.: HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model access via NOAA ARL READY Website, available at: http://ready.arl.noaa.gov/HYSPLIT.php (last access: 3 February 2014), NOAA Air Resources Laboratory, Silver Spring, MD, 2011.

Greeman, D. J. and Rose, A. W.: Factors controlling the emanation of radon and thoron in soils of the Eastern USA, Chem. Geol., 129, 1–14, doi:10.1016/0009-2541(95)00128-X, 1996.

Gupta, M., Douglass, A. R., Kawa, S., and Pawson, S.: Use of radon for evaluation of atmospheric transport models: sensitivity to emissions, Tellus B, 56, 404–412, doi:10.1111/j.1600-0889.2004.00124.x, 2004.

IUSS Working Group WRB: World Reference Base for Soil Resources 2006, First Update 2007, World Soil Resources Reports 103, Food and Agriculture Organization, Rome, 118 pp., 2007.

Jacob, D., Prather, M., Rasch, P., Shia, R., Balkanski, Y., Beagley, S., Bergmann, D., Blackshear, W., Brown, M., Chiba, M.,
Chipperfield, M. P., de Grandpré, J., Dignon, J. E., Feichter, J., Genthon, C., Grose, W. L., Kasibhatla, P. S., Köhler, I., Kritz, M. A., Law, K., Penner, J. E., Ramonet, M., Reeves, C. E., Rotman, D. A., Stockwell, D. Z., Van Velthoven, P. F. J., Verver, G., Wild, O., Yang, H., and Zimmermann, P.: Evaluation and intercomparison of global atmospheric transport models using ^{222}Rn and other short-lived tracers, J. Geophys. Res., 102, 5953–5970, doi:10.1029/96JD02955, 1997.

ACPD
14, 3667–3706, 2014

Factors controlling variability of atmospheric ^{222}Rn over central Europe

M. Zimnoch et al.

Jeričević, A., Kraljević, L., Grisogono, B., Fagerli, H., and Večenaj, Ž.: Parameterization of vertical diffusion and the atmospheric boundary layer height determination in the EMEP model, Atmos. Chem. Phys., 10, 341–364, doi:10.5194/acp-10-341-2010, 2010.

Levin, I., Glatzel-Mattheier, H., Marik, T., Cuntz, M., and Schmidt, M.: Verification of German methane emission inventories and their recent changes based on atmospheric observations, J. Geophys. Res., 104, 3447–3456, 1999.

Levin, I., Born, M., Cuntz, M., Langendörfer, U., Mantsch, S., Naegler, T., Schmidt, M., Varlagin, A., Verclas, S., and Vagenbach, D.: Observations of atmospheric variability and soil exhalation rate of radon-222 at a Russian forest site, Tellus B, 54, 462–475, 2002.

Levin, I., Kromer, B., Schmidt, M., and Sartorius, H.: A novel approach for independent budgeting of fossil fuel CO_2 over Europe by $14CO_2$ observations, Geophys. Res. Lett., 30, 2194, doi:10.1029/2003GL018477, 2003.

Lui, S. C., McAfee, J. R., and Cicerone, R. J.: Radon 222 and tropospheric vertical transport, J. Geophys. Res., 89, 7291–7297, 1984.

Mazur, J.: Dynamics of radon exhalation rates in relation to meteorological parameters and properties of soil, available at: http://www.ifj.edu.pl/publ/reports/2008 (last access: 3 February 2014), Ph.D. Thesis, Henryk Niewodniczanski Institute of Nuclear Physics, Polish Academy of Science, Krakow, 140 pp., 2008 (in Polish).

Nazaroff, W.: Radon transport from soil to air, Rev. Geophys., 30, 137–160, doi:10.1029/92RG00055, 1992.

Netzel, P., Stano, S., and Zarębski, M.: Vertical doppler sodar VDS, Wiad. IMGW, 18, 119–125, 1995.

Papachristodoulou, C., Ioannides, K., and Spathis, S.: The effect of moisture content on radon diffusion through soil: assessment in laboratory and field experiments, Health Phys., 92, 257–264, doi:10.1097/01.HP.0000248147.46038.bc, 2007.

Piringer, M. and Joffre, S. (ed.): The Urban Surface Energy Budget and Mixing Height in European Cities: Data, Models and Challenges for Urban Meteorology and Air Quality. Final Raport of Working Group 2 of COST-715 Action, Demetra Ltd Publishers, Bulgaria, 2005.

Rogers, V. C. and Nielson, K. K.: Correlations for predicting air permeabilities and ^{222}Rn diffusion coefficients of soils, Health Phys., 61, 225–230, doi:10.1097/00004032-199108000-00006, 1991.

Rolph, G. D.: Real-time Environmental Application and Display sYstem (READY) website, available at: http://ready.arl.noaa.gov (last access: 3 February 2014), NOAA Air Resources Laboratory, Silver Spring, MD, 2011.

Sakoda, A., Ishimori, Y., Hanamoto, K., Kataoka, T., Kawabe, A., and Yamaoka, K.: Experimental and modeling studies of grain size and moisture content effects on radon emanation, Radiat. Meas., 45, 204–210, doi:10.1016/j.radmeas.2010.01.010, 2010.

Sasaki, T., Gunji, Y., and Okuda, T.: Mathematical modeling of Radon emanation, J. Nucl. Sci. Technol., 41, 142–151, doi:10.3327/jnst.41.142, 2004.

Schery, S. D. and Huang, S.: An estimate of the global distribution of radon emissions from the ocean, Geophys. Res. Lett., 31, L19104, doi:10.1029/2004GL021051, 2004.

Schmidt, M., Graul, R., Sartorius, H., and Levin, I.:. Carbon dioxide and methane in continental Europe: a climatology, and ^{222}Radon-based emission estimates, Tellus B, 48, 457–473, 1996.

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.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J.-P., Valdebenito, Á., and Wind, P.: The EMEP MSC-W chemical transport model – technical description, Atmos. Chem. Phys., 12, 7825–7865, doi:10.5194/acp-12-7825-2012, 2012.

Stull, R. B.: An Introduction to Boundary Layer Meteorology, Reidel Publishing Co., Dordrecht, 666 pp., 1988.

Szegvary, T., Conen, F., and Ciais, P.: European ^{222}Rn inventory for applied atmospheric studies, Atmos. Environ., 43, 1536–1539, doi:10.1016/j.atmosenv.2008.11.025, 2009.

Taguchi, S., Law, R. M., Rödenbeck, C., Patra, P. K., Maksyutov, S., Zahorowski, W., Sartorius, H., and Levin, I.: TransCom continuous experiment: comparison of ^{222}Rn transport at hourly time scales at three stations in Germany, Atmos. Chem. Phys., 11, 10071–10084, doi:10.5194/acp-11-10071-2011, 2011.

Thoning, K. W., Tans, P. P., and Komhyr, W. D.: Atmospheric carbon dioxide at Mauna Loa Observatory 2. Analysis of the NOAA GMCC data, 1974–1985, J. Geophys. Res., 94, 8549–8565, 1989.

ACPD

14, 3667–3706, 2014

Vaupotič, J., Gregorič, A., Kobal, I., Žvab, P., Kozak, K., Mazur, J., Kochowska, E., and Grządziel, D.: Radon concentration in soil gas and radon exhalation rate at the Ravne Fault in NW Slovenia, Nat. Hazards Earth Syst. Sci., 10, 895–899, doi:10.5194/nhess-10-895-2010, 2010.

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.

Zhang, K., Wan, H., Zhang, M., and Wang, B.: Evaluation of the atmospheric transport in a GCM using radon measurements: sensitivity to cumulus convection parameterization, Atmos. Chem. Phys., 8, 2811–2832, doi:10.5194/acp-8-2811-2008, 2008.

Zhang, K., Feichter, J., Kazil, J., Wan, H., Zhuo, W., Griffiths, A. D., Sartorius, H., Zahorowski, W., Ramonet, M., Schmidt, M., Yver, C., Neubert, R. E. M., and Brunke, E.-G.: Radon activity in the lower troposphere and its impact on ionization rate: a global estimate using different radon emissions, Atmos. Chem. Phys., 11, 7817–7838, doi:10.5194/acp-11-7817-2011, 2011.

Zimnoch, M. Godlowska, J., Necki, J. M., and Rozanski, K.: Assessing surface fluxes of CO_2 and CH_4 in urban environment: a reconnaissance study in Krakow, Southern Poland, Tellus B, 62, 573–580, doi:10.1111/j.1600-0889.2010.00489.x, 2010.

ACPD
14, 3667–3706, 2014

Table 1. Statistics of daily variations of ^{222}Rn activity in Krakow (KR) and Heidelberg (HD) for the period January 2005–December 2009.

Period		Average daily ^{222}Rn activity ($Bq\,m^{-3}$)	Average daily maximum ^{222}Rn activity ($Bq\,m^{-3}$)	Average daily minimum ^{222}Rn activity ($Bq\,m^{-3}$)	Peak-to-peak amplitude ($Bq\,m^{-3}$)
Winter	KR	5.65 ± 0.17	9.36 ± 0.27	2.99 ± 0.11	6.37 ± 0.23
(Dec–Feb)	HD	5.19 ± 0.19	7.93 ± 0.26	3.10 ± 0.14	4.83 ± 0.17
Spring	KR	4.00 ± 0.09	8.81 ± 0.23	1.52 ± 0.05	7.29 ± 0.22
(Mar–May)	HD	3.14 ± 0.08	5.74 ± 0.16	1.54 ± 0.05	4.21 ± 0.15
Summer	KR	5.50 ± 0.13	12.11 ± 0.30	2.02 ± 0.05	10.09 ± 0.27
(Jun–Aug)	HD	3.55 ± 0.07	6.88 ± 0.17	1.61 ± 0.04	5.27 ± 0.15
Autumn	KR	8.70 ± 0.23	15.59 ± 0.39	3.83 ± 0.13	11.76 ± 0.35
(Sep–Nov)	HD	6.08 ± 0.17	10.13 ± 0.27	3.38 ± 0.13	6.75 ± 0.21
Mean	KR	5.86 ± 0.09	11.28 ± 0.16	2.55 ± 0.05	8.72 ± 0.14
2005–2009	HD	4.50 ± 0.07	7.67 ± 0.12	2.42 ± 0.05	5.26 ± 0.09

ACPD
14, 3667–3706, 2014

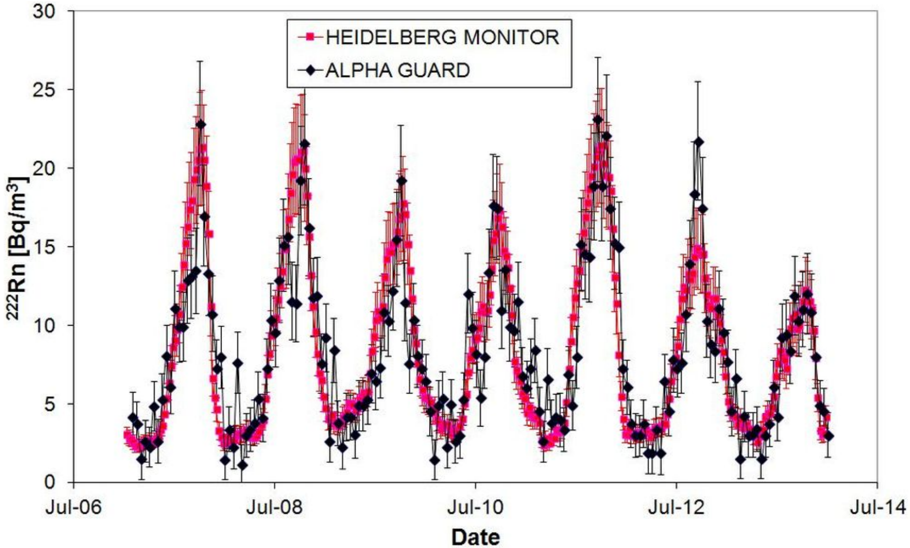

Fig. 1. Parallel measurements of ^{222}Rn concentration in near-ground atmosphere in Krakow between 6 and 14 July 2006, performed using Heidelberg radon monitor based on detection of ^{222}Rn daughter products and AlphaGUARD detector measuring directly alpha particles of ^{222}Rn. The data points correspond to 30 min and one-hour counting intervals for Heidelberg monitor and AlphaGUARD detector, respectively. Error bars represent uncertainties reported directly by AlphaGUARD (black) and 17 % constant relative uncertainty for Heidelberg monitor (red – Levin et al., 2002).

ACPD
14, 3667–3706, 2014

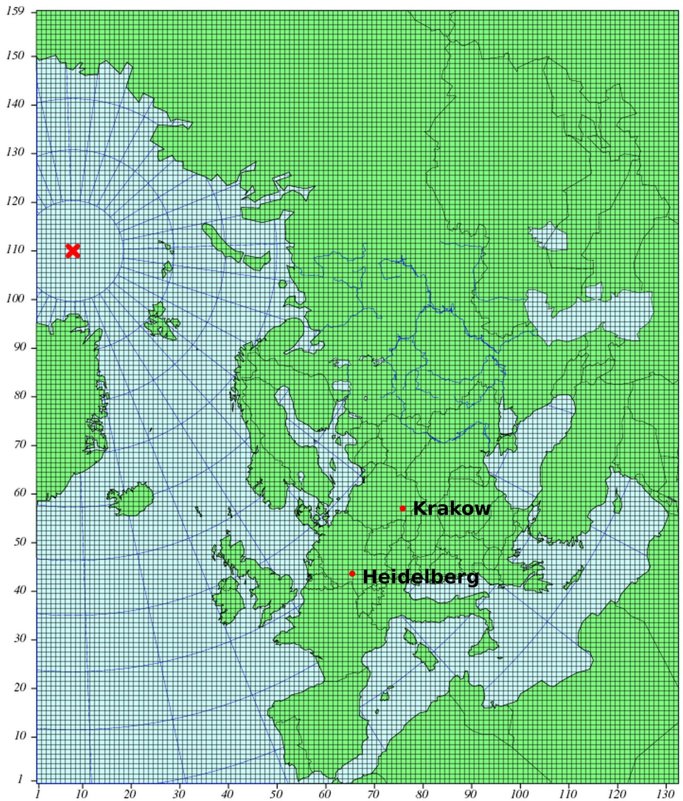

Fig. 2. The domain of Unified EMEP model used to simulate vertical distribution of ^{222}Rn in the lower atmosphere (see text for details). Size of the grid: 50 km × 50 km.

ACPD
14, 3667–3706, 2014

Factors controlling variability of atmospheric ^{222}Rn over central Europe

M. Zimnoch et al.

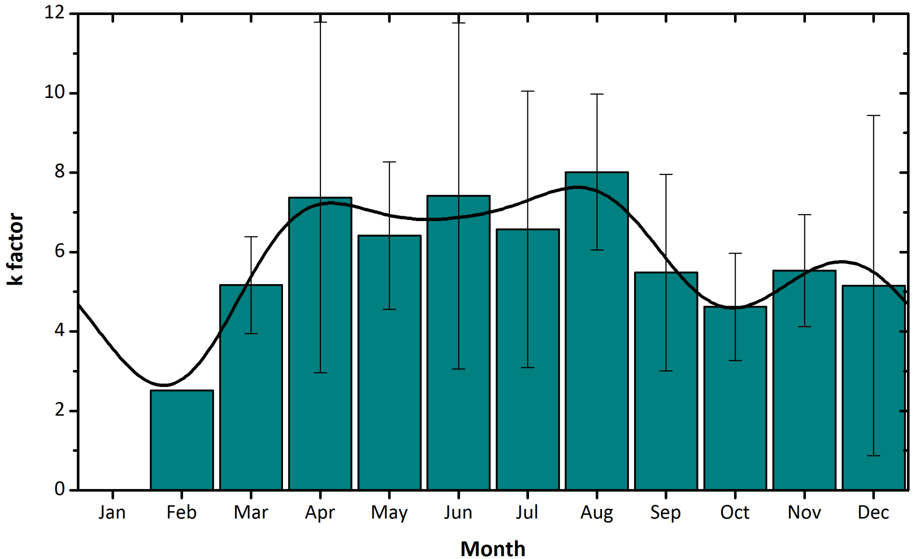

Fig. 3. Monthly mean values of the correction factor *k* calculated using vertical profiles ^{222}Rn simulated for Krakow by the Unified EMEP model (see text for details). Thin vertical bars indicate standard uncertainties of the mean *k* values. Heavy line represents best fit of monthly *k* values.

ACPD
14, 3667–3706, 2014

Factors controlling variability of atmospheric ^{222}Rn over central Europe

M. Zimnoch et al.

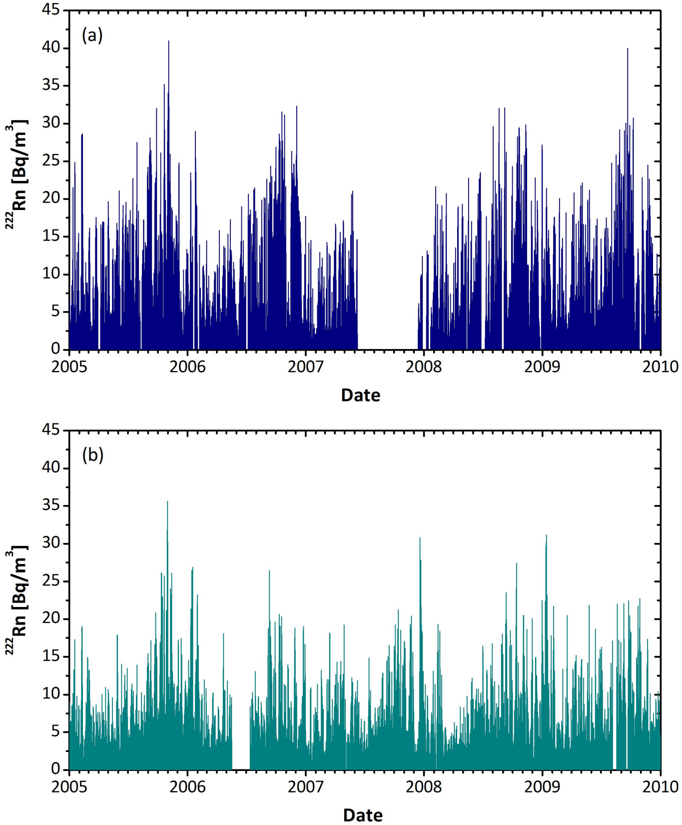

Fig. 4. Time series of hourly mean ^{222}Rn content in near-ground atmosphere, recorded between January 2005 and December 2009 in Krakow, southern Poland **(a)** and Heidelberg, south-west Germany **(b)**.

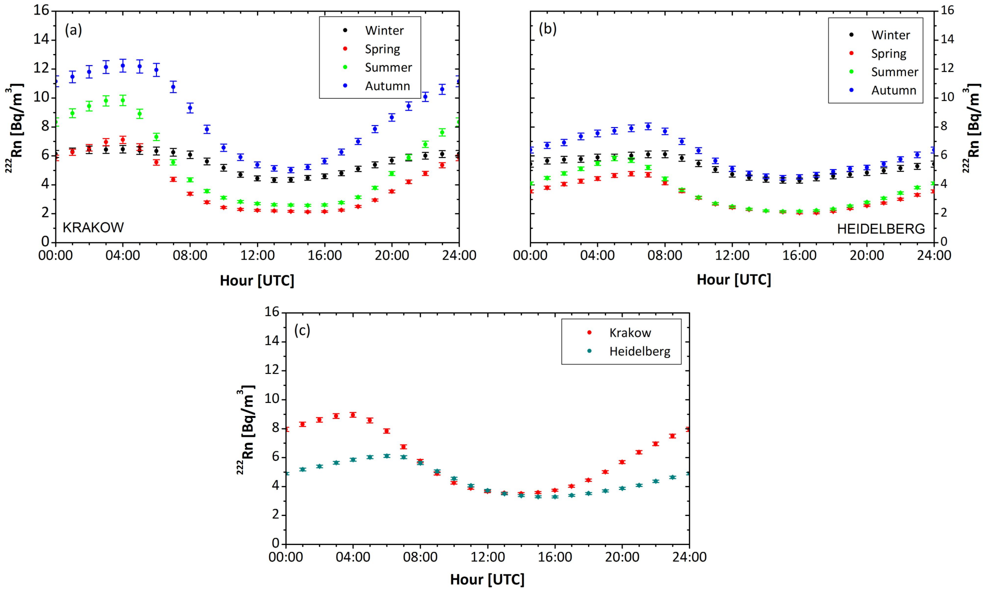

Fig. 5. Diurnal variations of ^{222}Rn specific activity in near-ground atmosphere over Krakow **(a)** and Heidelberg **(b)** during the period January 2005–December 2009, averaged separately for each hour and for four seasons: winter (DJF), spring (MAM), summer (JJA), autumn (SON). Diurnal variation of ^{222}Rn content at both sites averaged for entire observation period is also shown **(c)**. Vertical marks accompanying the data points indicate standard deviations of the calculated average values.

ACPD
14, 3667–3706, 2014

Factors controlling variability of atmospheric ^{222}Rn over central Europe

M. Zimnoch et al.

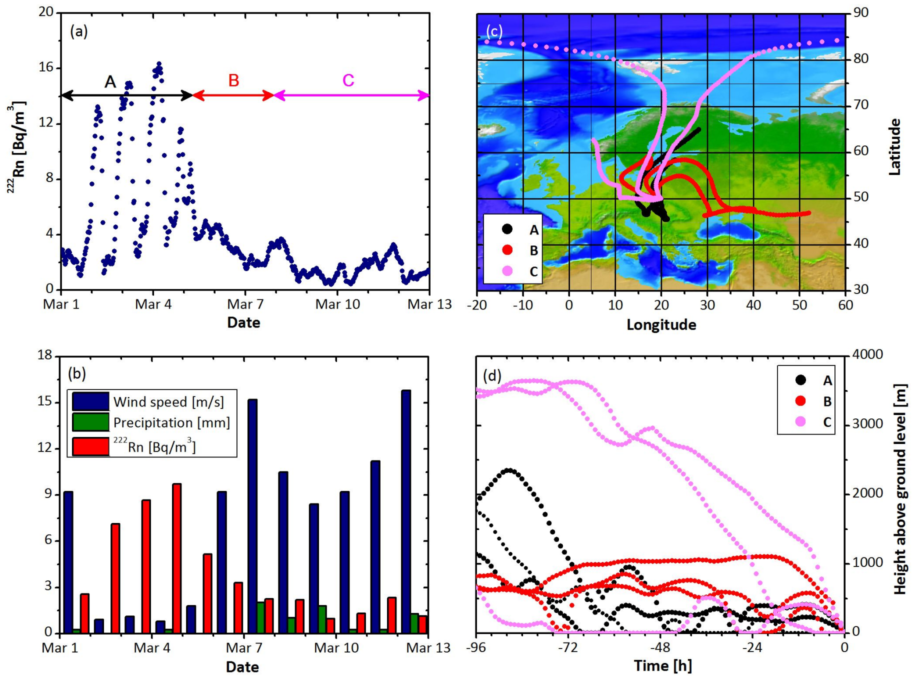

Fig. 6. (a) Changes of atmospheric ^{222}Rn in Krakow between 1 and 13 March 2005. Horizontal lines with arrows and letter symbols mark different air masses identified in **(c)**. **(b)** Daily means of atmospheric ^{222}Rn content, wind speed and rainfall amount in Krakow. **(c)** 96 h backward trajectories of the air masses arriving at Krakow (three trajectories representing beginning, centre and end-point of each period A, B and C marked in **(a)** are shown) and changes of their elevation above the ground along the route **(d)**.

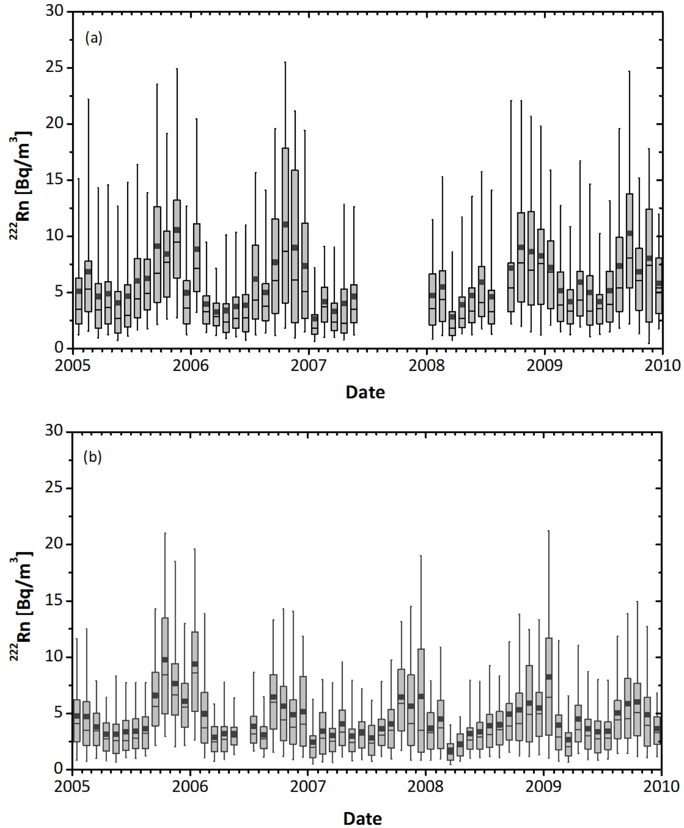

Fig. 7. Box-and-whisker plots of monthly ^{222}Rn activity recorded in near-ground atmosphere of Krakow **(a)** and Heidelberg **(b)** between January 2005 and December 2009.

ACPD
14, 3667–3706, 2014

Factors controlling variability of atmospheric ^{222}Rn over central Europe

M. Zimnoch et al.

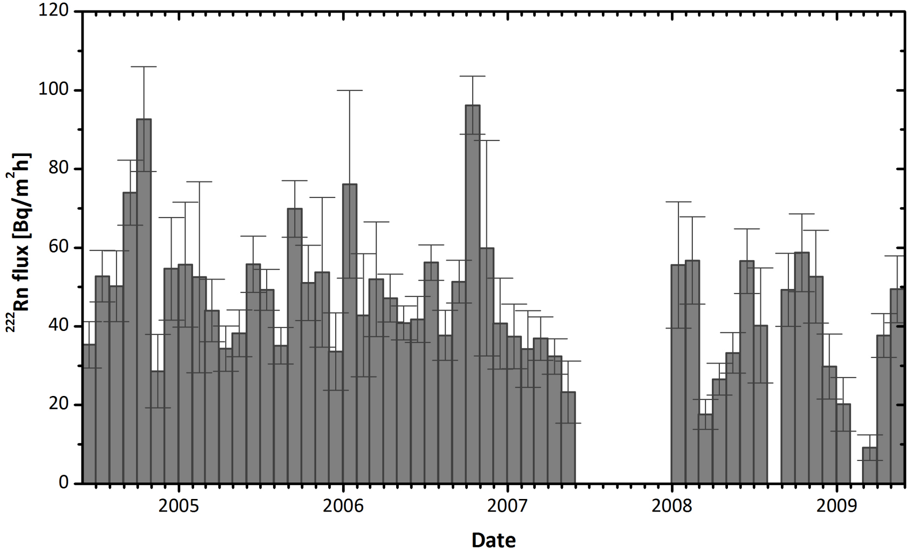

Fig. 8. Monthly mean night-time fluxes of soil ^{222}Rn in Krakow urban area for the period June 2004–May 2009, derived from atmospheric ^{222}Rn observations and sodar measurements of the mixing layer height. Error bars indicate standard deviations of the mean values.

ACPD
14, 3667–3706, 2014

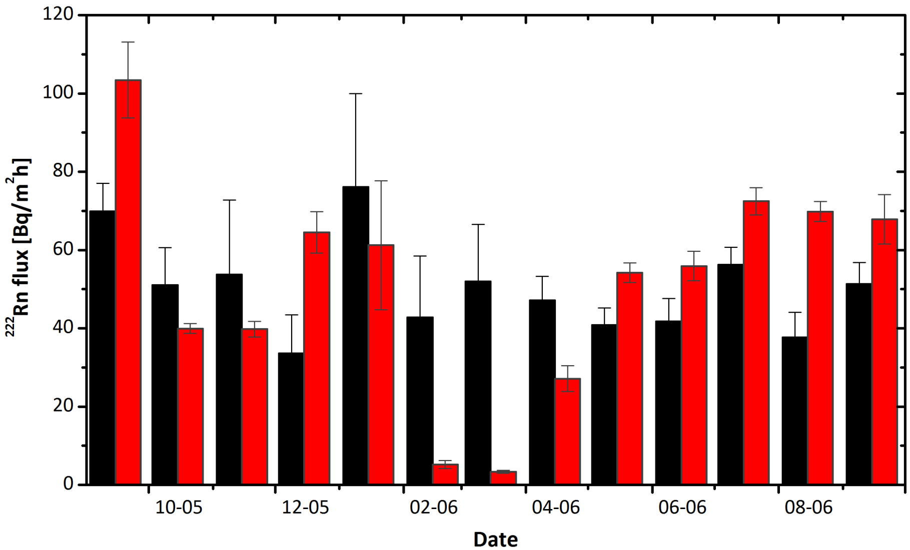

Fig. 9. Comparison of mean monthly night-time surface fluxes of ^{222}Rn in Krakow for the period from September 2005 till September 2006, derived from direct measurements of soil ^{222}Rn flux using chamber method (red bars) and from atmospheric ^{222}Rn observations combined with sodar measurements of the mixing layer height (black bars). Error bars indicate standard deviation of the mean values.

ACPD
14, 3667–3706, 2014

Factors controlling variability of atmospheric ^{222}Rn over central Europe

M. Zimnoch et al.

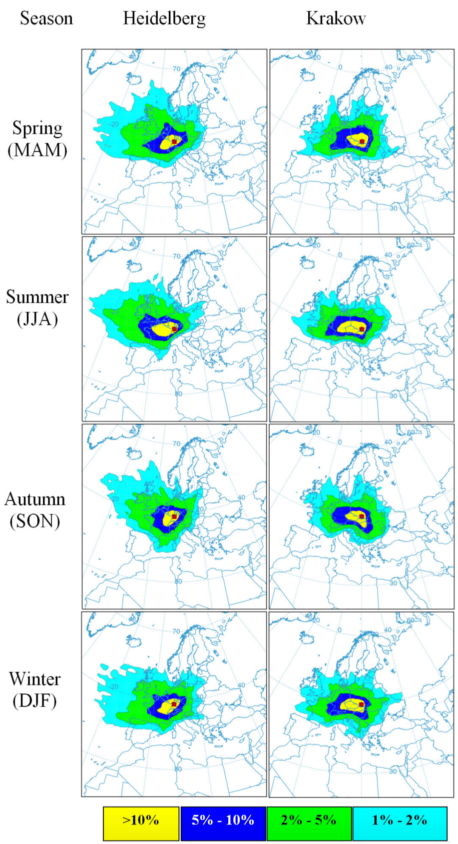

Fig. 10. Spatial distribution of 96 h backward trajectories of the air masses arriving in Heidelberg, southern Germany, and Krakow, southern Poland, during the period January 2005–December 2009, depictured for four seasons, calculated using HYSPLIT model (Draxler and Rolph, 2011; Rolph, 2011) – see text for details.

ACPD
14, 3667–3706, 2014

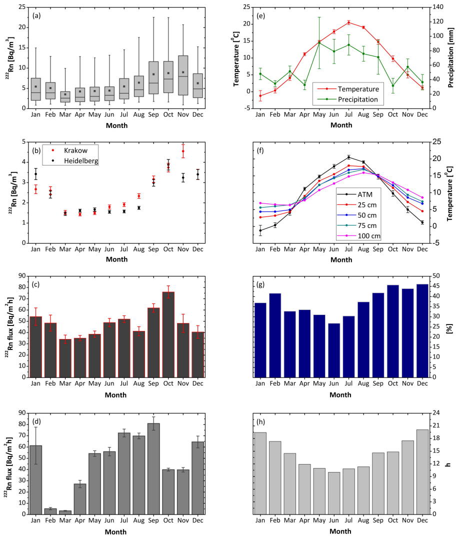

Fig. 11. (a) Monthly box-and-whisker plot of ^{222}Rn content in near-ground atmosphere in Krakow. Median is represented by horizontal line while arithmetic average is shown by full dot. **(b)** Monthly minima of atmospheric ^{222}Rn content, as observed in Krakow and Heidelberg, calculated as a mean value of daily ^{222}Rn minima for the given month, averaged over entire observation period from January 2005 till December 2009. **(c)** Monthly means of night-time ^{222}Rn fluxes into the local atmosphere in Krakow, averaged over the period from June 2004 till May 2009. **(d)** Monthly means of soil ^{222}Rn fluxes in Krakow derived from chamber measurements, averaged over the period from September 2005 till September 2006. **(e)** Monthly means of surface air temperature and precipitation in Krakow, averaged over the period from January 2005 till December 2009. **(f)** Monthly means of surface air and soil temperatures in Krakow. **(g)** Percentage of periods with wind speed below 1 m s^{-1} in Krakow, averaged over the period from January 2005 till December 2009. **(h)** Monthly means of number of hours with ground-based inversion per day, averaged over the period from January 1994 till December 1999.

ACPD
14, 3667–3706, 2014

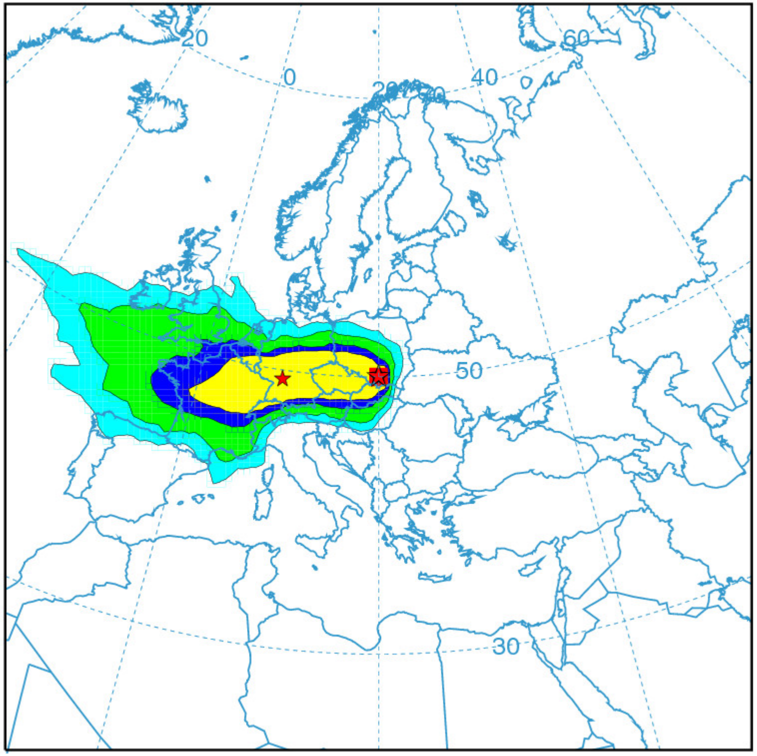

Fig. 12. Spatial distribution of 96 h backward trajectories of the air masses arriving in Krakow, Southern Poland, which passed Heidelberg, Southern Germany, at the distance of less than 100 km, calculated using HYSPLIT model (Draxler and Rolph, 2011; Rolph, 2011 – see text for details). The analysis comprised the period from January 2005 till December 2009.

ACPD
14, 3667–3706, 2014

Factors controlling variability of atmospheric ^{222}Rn over central Europe

M. Zimnoch et al.