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Determination of 222-Rn exhalation rates in an urban area - comparison of top-down and bottom-up approach

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Radon-222 is a radioactive noble gas, widely used as a tracer of atmospheric transport. The radon precursor, ^{226}Ra, belongs to ^{238}U series and is ubiquitous in the Earth's crust and in the soils. Due to its relatively long half-life ($T_{1/2}$ = 3.8 days), ^{222}Rn mostly diffuses out of the soil into the atmosphere, where it decays to ^{218}Po. The exhalation rate of ^{222}Rn into the atmosphere is controlled by the source term i.e. ^{226}Ra content in the soil and its vertical distribution, by physical properties of the upper soil layer such as mineral structure, porosity and water content and to some extent by short-term variations of physical parameters characterizing the soil-atmosphere interface (atmospheric temperature and pressure).

Radon-222 has been repeatedly used in the past decade as a tool to assess surface emissions of greenhouse gases such as CO_2 and CH_4 into the atmosphere, originating from distributed sources. The method requires a priori knowledge of ^{222}Rn exhalation rate in the given area and its temporal variability.

In this study we present a comparison of two independent methods of deriving exhalation rates of ^{222}Rn in an urban environment: (i) direct measurements utilizing the static chamber method, and (ii) determination of ^{222}Rn exhalation rates through measurements of mixing layer height in the lower atmosphere, combined with measurements of ^{222}Rn specific activity in near-ground atmosphere. The comparison was performed in Krakow, southern Poland, with parallel measurements of ^{222}Rn exhalation rates running from September 2005 to September 2006. Only night-time ^{222}Rn fluxes were compared.

Direct measurements of ^{222}Rn flux (static chamber method) were performed using the AlphaGUARD PQ2000 PRO® monitor together with an accumulation chamber working in an automatic system which made it possible to perform 4 measurements during 24 hours. The system was developed at the Institute of Nuclear Physics, Polish Academy of Sciences. The mixing layer height was measured with vertical doppler sodar system developed at the Department of Monitoring and Modelling Air Pollution, Institute of Meteorology and Water Management. Atmospheric concentrations of ^{222}Rn were measured using radon monitor based on alpha spectrometry of ^{222}Rn daughters captured from the air stream on a glass filter placed directly over the surface barrier detector measuring alpha particles emitted by ^{222}Rn daughter products. The radon monitor used for atmospheric ^{222}Rn measurements was developed at the Institute of Environmental Physics, University of Heidelberg, Germany, and made available for this study.

The night-time monthly mean ^{222}Rn exhalation rates were calculated from the data obtained using the methods briefly described above. Generally, high values of ^{222}Rn exhalation rates were observed during summer months (July, August, September) and reduced values during winter months (February, March). The maximum monthly mean value of ^{222}Rn exhalation rate (ca. 105 Bq m^{-2} h^{-1}) measured with the aid of chamber method was observed in September 2005, to be compared with ca. 65 Bq m^{-2} h^{-1} obtained from the indirect method. The minimum ^{222}Rn exhalation rate was recorded in February 2006 (ca. 20 Bq m^{-2} $h^{-1}$and 10 Bq m^{-2} h^{-1}, for the sodar-assisted and for the chamber measurements, respectively). Although the amplitude of seasonal changes of ^{222}Rn exhalation rate derived from chamber measurements was significantly higher than that obtained using the indirect method based on sodar measurements of mixing layer height and atmospheric measurements of ^{222}Rn concentration, a reasonably good agreement between both methods was obtained (r^2 = 0.67). The principal reason for the observed differences in the measured ^{222}Rn fluxes can be attributed to different footprint of both methods. While the chamber method yields ^{222}Rh exhalation rates averaged over an area of ca. 0.04 m^2 covered by the chamber, the footprint of the indirect method is in the order of several square kilometers, comparable to the size of the city.

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