

Seasonal and diurnal variation of outdoor radon (^{222}Rn) concentrations in urban and rural area with reference to meteorological conditions

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Abstract. The objective of the study was to investigate temporal variability of outdoor radon (^{222}Rn) concentration registered in the center of Łódź (urban station), at Ciosny (rural station) and Kraków (suburban station) in relation to meteorological parameters (i.e. air temperature, temperature vertical gradient, wind speed, soil heat flux, volumetric water content in soil) with special consideration of urban-rural differences. Continuous measurements of ^{222}Rn concentration (at 60 min intervals) were performed at a height of 2 m above the ground using AlphaGUARD[®] PQ2000PRO (ionization chamber) from January 2008 to May 2009. ^{222}Rn levels were characterized by a diurnal cycle with an early morning maximum and a minimum in the afternoon. The well-marked 24 h pattern of radon concentration occurred in summer at anticyclonic weather with cloudless sky, light wind and large diurnal temperature ranges. The urban measurement site was characterized by the lowest atmospheric ^{222}Rn concentration and an urban-rural differences of radon levels increased from winter to summer and during the nighttime periods. The maximum contrasts of ^{222}Rn levels between Łódź and Ciosny, reaching $-30\text{ Bq}\cdot\text{m}^{-3}$, were registered in June and July during the urban heat island (UHI) phenomenon (a positive thermal anomaly of a city if compared to rural area) and strong thermal inversion near the ground in the rural area.

Key words: outdoor radon (^{222}Rn) concentration • urban climate • seasonal variation • diurnal variation • meteorological parameters

Introduction

Radon gas (^{222}Rn) is a significant source of the natural radioactivity in the atmospheric boundary layer. It is produced in the soil by the radioactive decay of ^{226}Ra and then is released from the ground into the atmosphere (exhalation), where it is transported mainly by turbulent diffusion or convection and can migrate to the inside of a building. In Poland, radon inhalation is responsible for over 40% of the annual effective dose of ionizing radiation [3]. For that reason, the knowledge of fluctuations of natural atmospheric radioactivity in the lower atmosphere, the basic environment of the human living, given by radon is so important. Temporal variability of ^{222}Rn in the air near the ground in relation to weather elements (air pressure, wind velocity, precipitations, snow cover) and vertical mixing processes in the atmosphere were documented by various authors [1, 2, 4, 13, 15]. With a ground-level source, ^{222}Rn is a useful natural indicator of atmospheric mixing conditions that can be used to inter the behavior of other atmospheric pollutants within the atmospheric boundary layer [6, 8, 12, 14].

It is known that urbanization with the input of anthropogenic heat, moisture and pollutants into the atmosphere modify the local climate and the atmospheric boundary layer on urban areas differs from that of rural area. The most distinct modification of the

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local climate by towns is the UHI phenomenon – urban air temperature is higher than their corresponding rural values. Towns also modify wind velocity field, the air mixing-layer height, losses of solar radiation and causes an increase of cloudiness and rainfalls [9, 11]. Łódź, for the reason of its geographical position, size, building structure and great artificial heat emission, is distinguished by strong urban-rural contrasts of meteorological parameter. Over 50 years studies on climate of Łódź have revealed the highest UHI intensity in Poland, i.e. thermal urban-rural contrast reached 12°C (February 1996) and anthropogenic heat emission exceeded 100 W·m⁻² in winter [5, 7]. The aim of the present study was the analysis of temporal variability of ²²²Rn in the urban and rural areas in relation to local and mesoscale weather conditions with special consideration of urban-rural contrast.

Data and methods

Area and its subsoil radioactivity

Radon measurement sites are located in Central Poland (Łódź and Ciosny) and in the South (Kraków). In Łódź, ²²²Rn is measured at urban automatic meteorological station of the Department of Meteorology and Climatology of University of Łódź. The station is situated close to geometrical center of Łódź ($\phi = 51^{\circ}46'N$, $\lambda = 19^{\circ}28'E$), in a big downtown square. The Ciosny village is located in agrarian area, 25 km to the north from Łódź. ²²²Rn concentration at Ciosny is measured at a rural automatic hydro-meteorological station ($\phi = 51^{\circ}55'N$, $\lambda = 19^{\circ}24'E$) of the Department of Hydrology and Water Management of University of Łódź. In Kraków, ²²²Rn concentration is registered in the north-west part of the city (suburban area), on Radon Study Field ($\phi = 50^{\circ}05'N$, $\lambda = 19^{\circ}53'E$) of Laboratory of Radiometric Expertise, Institute of Nuclear Physics, Polish Academy of Sciences, Kraków. The subsoil of investigated area consists of sand and clay (Łódź),

glacial sand (Ciosny) and loessial soil on loess stratum to a depth of 8 m (Kraków).

Radium (²²⁶Ra) activity concentration, the precursor of ²²²Rn, in surface-layer soil at the measurement sites amounted to 13 Bq·kg⁻¹ (Łódź), 4 Bq·kg⁻¹ (Ciosny) and 22 Bq·kg⁻¹ (Kraków) whereas the mean value of ²²⁶Ra concentration in Poland equals 25 Bq·kg⁻¹ [3].

Instrumentation and data processing

The continuous measurements of ²²²Rn concentration (at 60 min intervals) in Łódź, (urban st.) Ciosny (rural st.) and Kraków (suburban st.) were made using one AlphaGUARD® PQ2000PRO) per one measurement point (ionization chamber, Genitron Instruments GmbH. The device was set up in a meteorological box at a height of 2 m above the ground from January 2008 to May 2009. In Central Poland, the following meteorological parameters were recorded simultaneously: air temperature, relative humidity, air pressure, wind speed, soil heat flux using HFP01 Heat Flux Plate (Campbell Scientific Ltd.) and volumetric water content in the soil using water content reflectometer (Campbell Scientific Ltd.). The daily courses of the differences in urban, rural and suburban radon levels were compared. The annual and seasonal means of 24 h patterns of ²²²Rn concentration were analyzed in relation to the courses of wind velocity, air temperature, and soil heat flux. The relationships between ²²²Rn concentration and temperature vertical gradient, volumetric water content and soil heat flux were studied. Additionally, the mesoscale weather conditions using synoptic charts were investigated. Detailed analysis of local and mesoscale weather pattern was performed for the period with the maximum of negative urban-rural differences of ²²²Rn concentration in Central Poland in June 2008. Seasonal measurements of ²²²Rn concentration in soil using the passive method (CR-39 track-etched detectors) were carried out in Łódź and Ciosny in 2008 as a background of radon measurements in the low troposphere.

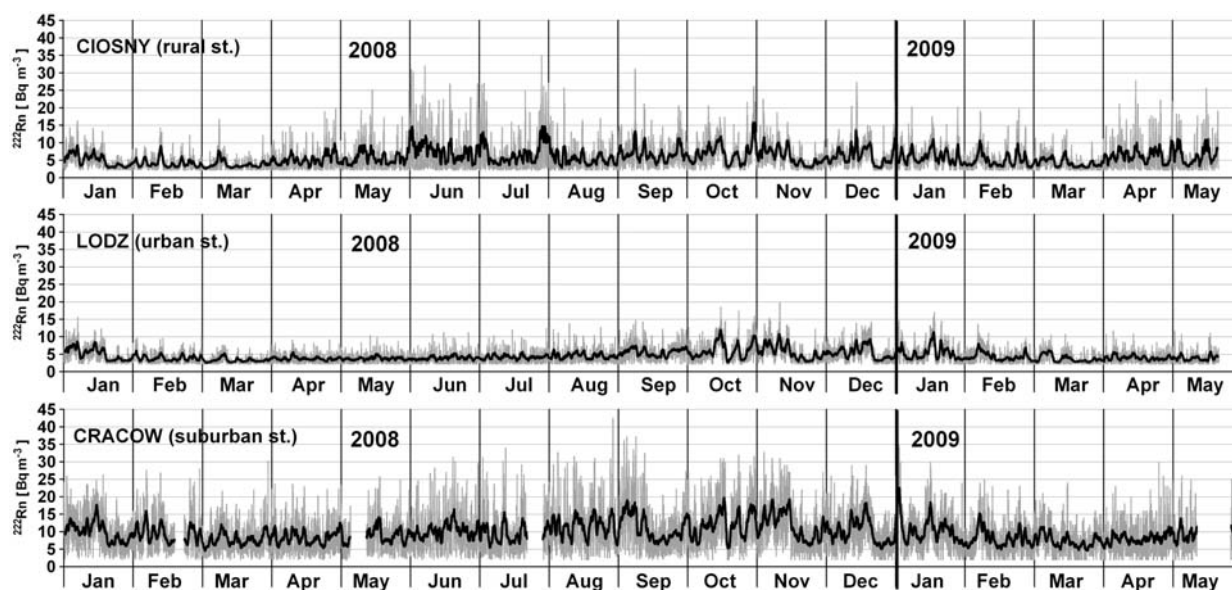


Fig. 1. Hourly ²²²Rn concentration in the air at 2 m above the ground at the rural (Ciosny), the urban (Łódź) and the suburban (Kraków) stations from January 2008 to May 2009. Black line – running average of 24 h.

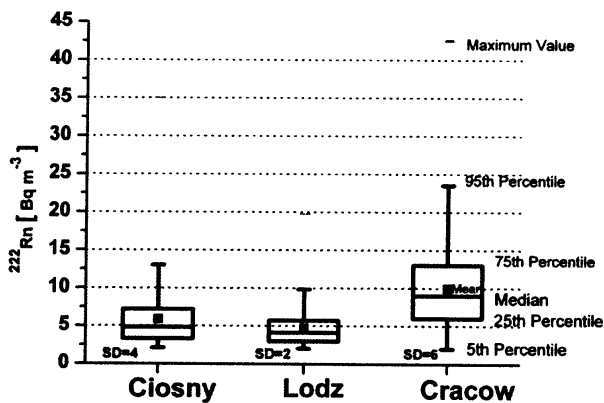


Fig. 2. Distribution of hourly ^{222}Rn concentration in the air at 2 m above the ground at the rural (Ciosny), the urban (Łódź) and the suburban (Kraków) stations from January 2008 to May 2009. SD (standard deviation) – in $\text{Bq}\cdot\text{m}^{-3}$.

Results and discussion

Measurements of ^{222}Rn in soil gas using track-etched detectors CR-39 at Łódź and Ciosny stations in particular seasons, i.e. February–March, July and November 2008 resulted in the average values of $2.9 \text{ kBq}\cdot\text{m}^{-3}$ and $2.3 \text{ kBq}\cdot\text{m}^{-3}$ (spring), $4.1 \text{ kBq}\cdot\text{m}^{-3}$ and $7.9 \text{ kBq}\cdot\text{m}^{-3}$ (summer) and $16.2 \text{ kBq}\cdot\text{m}^{-3}$ and $5.1 \text{ kBq}\cdot\text{m}^{-3}$ (autumn), respectively. In Kraków, radon concentration in soil did not exceed $60 \text{ kBq}\cdot\text{m}^{-3}$ (instantaneous measurements using an ionization chamber AlphaGUARD PQ2000PRO) [10].

Figure 1 shows the variation of hourly outdoor radon concentration in the atmosphere for the period from January 2008 to May 2009. An annual pattern with low radon levels in spring months (a minimum occurred in March every year at 3 stations) and the increase in autumn (Łódź) and in summer and autumn (Ciosny, Kraków) was identified. Outdoor ^{222}Rn concentrations

were distinguished very high variability and the maximum reached $20 \text{ Bq}\cdot\text{m}^{-3}$ in Łódź (November 9, 2008), $35 \text{ Bq}\cdot\text{m}^{-3}$ at Ciosny (July 28, 2008) and $43 \text{ Bq}\cdot\text{m}^{-3}$ in Kraków (August 28, 2008), see Fig. 1. The value of arithmetic mean (AM) amounted to $5 \text{ Bq}\cdot\text{m}^{-3}$ (Łódź), $6 \text{ Bq}\cdot\text{m}^{-3}$ (Ciosny) and $10 \text{ Bq}\cdot\text{m}^{-3}$ (Kraków) and standard deviation (SD) was $2 \text{ Bq}\cdot\text{m}^{-3}$, $4 \text{ Bq}\cdot\text{m}^{-3}$ and $5 \text{ Bq}\cdot\text{m}^{-3}$, respectively. As shows Fig. 2, 95% of the recorded ^{222}Rn levels were lower than $10 \text{ Bq}\cdot\text{m}^{-3}$ (Łódź), $13 \text{ Bq}\cdot\text{m}^{-3}$ (Ciosny) and $23 \text{ Bq}\cdot\text{m}^{-3}$ (Kraków). The annual pattern of ^{222}Rn concentration near the ground corresponded with variability of mesoscale weather conditions through the year. In Poland, February and March 2008 characterize high frequency of atmospheric instability resulting in cyclonic weather with active cold fronts (exchange of air masses), strong wind, precipitation and radon build-ups were rare in this period. Summer and autumn 2008, when the increases of ^{222}Rn concentration were recorded, are distinguished by greater frequency of anticyclonic, sunny weather with light wind and a surface-based thermal inversion on the study sites.

The diurnal pattern of ^{222}Rn concentration was revealed with a maximum in the early morning and a minimum in the afternoon at 3 stations (Figs. 3 and 4). This diurnal variation of ^{222}Rn concentration resulted from the formation of a surface thermal inversion at night and high turbulence in the daytime mixing layer was documented by various authors [2, 4, 13, 15]. In Kraków, the clear diurnal variations of radon levels occurred in all seasons with maximum amplitudes (about $10 \text{ Bq}\cdot\text{m}^{-3}$) in summer. In Central Poland, at rural site (Ciosny) the clear diurnal cycle was present during all seasons except winter, whereas at urban site (Łódź) it was found only in summer and autumn. The maximum diurnal amplitudes of ^{222}Rn concentration in summer did not exceed $10 \text{ Bq}\cdot\text{m}^{-3}$ and $4 \text{ Bq}\cdot\text{m}^{-3}$ for Ciosny and Łódź, respectively (Fig. 3). Łódź is distinguished by 1 h delay of extremely daily values of ^{222}Rn concentration in

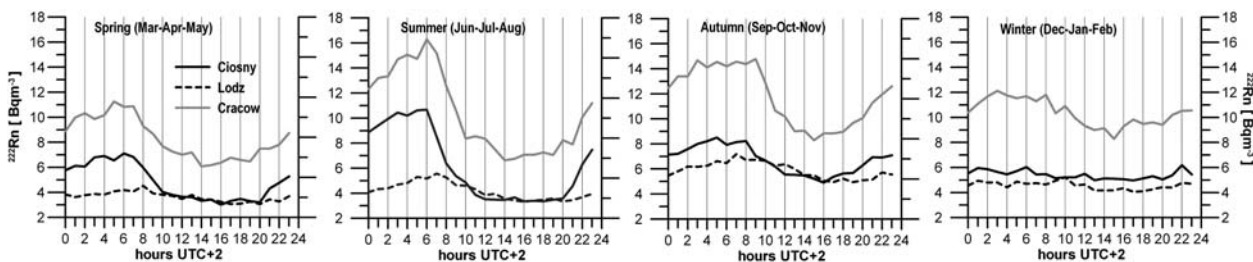


Fig. 3. Seasonal mean of 24 h pattern of ^{222}Rn concentration in the air at 2 m above the ground at the rural (Ciosny), the urban (Łódź) and the suburban (Kraków) stations in 2008.

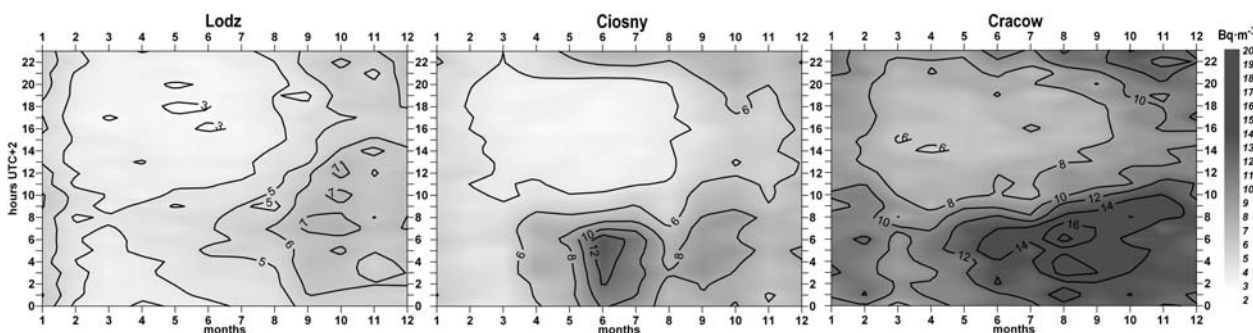


Fig. 4. Annual variability of 24 h pattern of ^{222}Rn concentration in the air at 2 m above the ground at the rural (Ciosny), urban (Łódź) and suburban (Kraków) stations in 2008.

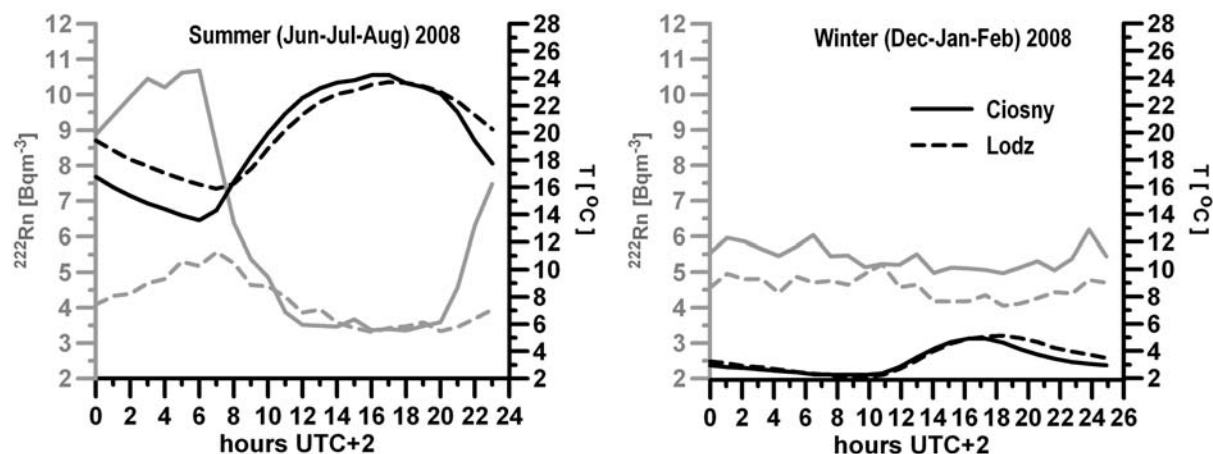


Fig. 5. The mean of 24 h pattern of ^{222}Rn concentration in the air and air temperature at 2 m above the ground at the rural (Ciosny) and urban (Łódź) stations in summer and winter 2008.

relation to rural and suburban site. The differences in the diurnal cycle between urban and rural sites resulted from a strong and fast radiative cooling of the rural site during night, resulting in a surface thermal inversion layer that trapped radon near the surface. Differences between the rural and urban site disappeared in daytime when mixing processes in the lower atmosphere developed (Figs. 3 and 4).

Figures 1 and 4 shows that the maximum contrasts of ^{222}Rn between Łódź and Ciosny were registered in May, June and July 2008. In these months, the UHI effect (UHI, a positive thermal anomaly of urban area if compared to rural area) was very frequent. The absolute maximum of urban-rural differences, $-30 \text{ Bq}\cdot\text{m}^{-3}$, occurred in June 2, 2008 during UHI with a maximum of intensity amounted to 8°C . The heat surplus at urban site caused a strong vertical mixing of air and a decrease of radon levels near the ground, whereas at rural site a strong surface thermal inversion occurred (5°C differences between 2 and 0.2 m) and it trapped radon in the air surface layer. The highest urban-rural contrasts of ^{222}Rn levels and the clear 24 h pattern of radon concentrations were registered during anticyclonic type weather conditions with a weak increasing pressure trend, wind speed $< 2 \text{ m}\cdot\text{s}^{-1}$, a high positive thermal gradient between 2 and 0.2 m height (temperature inversion) indicating a strong stable atmosphere. The disturbance of the daily pattern and a decrease of radon

levels near the ground was observed when a frontal cyclone associated with a strong wind and precipitation occurred. Figures 5, 6 and 7 show the mean daily variability of ^{222}Rn levels with reference to the daily courses of air temperature, wind velocity and soil heat flux through summer and winter 2008 at rural (Ciosny) and urban (Łódź) site. In general, the diurnal variation of near-surface ^{222}Rn concentration varied out of phase with air temperature and wind velocity (mechanical mixing in the boundary layer) and approximately in phase with the soil heat flux. Summer is characterized by stronger urban-rural contrasts of local climate characteristics and radon levels than in winter. In the summer months the increase of radon levels at nighttime and the vanish of these contrasts during the daytime were observed. In winter, a little 24 h variation of ^{222}Rn concentration were detected at both stations and generally, Ciosny is distinguished by higher values of radon levels. As can be seen in Fig. 5, in summer there were higher the daily amplitudes of air temperature and radon levels at rural station than in Łódź. In Łódź there was a time delay of the occurrence of air temperature and radon levels extremely values (Fig. 5). In general, in summer the rural station are characterized by lower wind velocity than urban station, especially in nocturnal hours when radon build-ups were observed (Fig. 6). Figure 7 shows the quite similar average courses of soil heat flux and radon levels in summer at both stations.

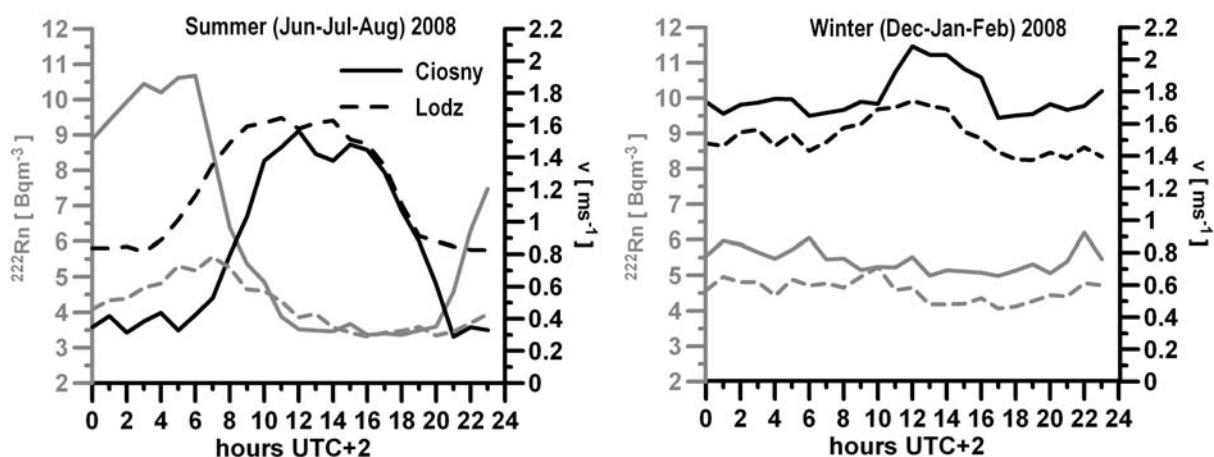


Fig. 6. The mean of 24 h pattern of ^{222}Rn concentration in the air and wind velocity at 2 m above the ground at the rural (Ciosny) and urban (Łódź) stations in summer and winter 2008.

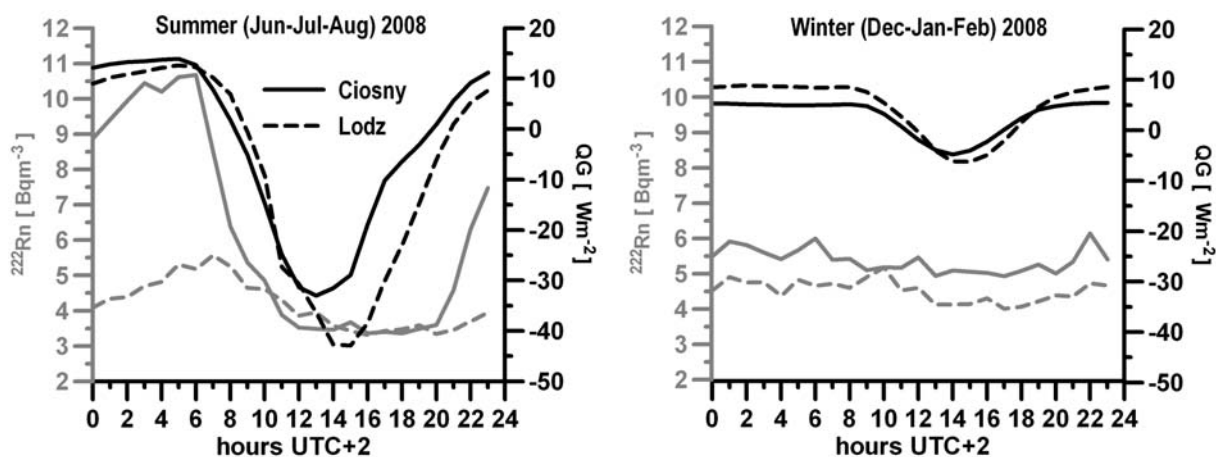


Fig. 7. The mean of 24 h pattern of ^{222}Rn concentration in the air at 2 m above the ground and soil heat flux (QG) at the rural (Ciosny) and urban (Łódź) stations in summer and winter 2008.

There is “downward” soil heat flux (negative values) in the daytime when radon levels decreased and “upward” soil heat flux (positive values) when radon increased. In general, soil heat flux and ^{222}Rn concentration varied in phase in the morning hours but a time delay of radon levels was observed in the evening. Further investigation is needed to determine the influence of heat flux in the soil on radon concentration in the surface air layer.

Conclusion

The urban-rural contrast of meteorological parameters (i.e. air temperature, thermal vertical gradient, wind velocity) can be used to determine near-surface radon build-ups. ^{222}Rn build-ups occurred at night and increased with stable atmospheric conditions from spring to autumn with a maximum during UHI. Radon build-ups were rare in winter as a result of atmospheric instability resulting in strong winds and precipitation. The present analysis indicated a positive correlation between ^{222}Rn levels and soil heat flux and a negative correlation between wind speed, air temperature, and volumetric water content in the soil. Further investigation is needed to confirm seasonal variation of ^{222}Rn concentrations in urban and rural area and to determine the long-term influence of individual meteorological parameters on radon levels in the air.

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