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# Soil heat flux and air temperature as factors of radon (Rn-222) concentration in the near-ground air layer

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**Abstract.** A unique, highly time-resolved, and synchronous three-year dataset of near-surface atmospheric radon-222 as well as soil heat flux and air temperature measurements at two sites (rural and urban) in Central Poland are investigated. The recognition of temporal variability of Rn-222 and selected meteorological variables in the urban and rural areas served to create two statistical models for estimation of this radionuclide concentration at 2 m a.g.l. The description of the relationships between the variables for individual months was established on the basis of an exponential function and an exponential function with time derivative of predictor to account for the hysteresis issue. The model with time derivative provided better results. The weakest fitting of modelled data to empirical ones is observed for the winter months. During subsequent seasons, air temperature as well as QG-driven (soil heat flux) models exhibited very high agreement with the empirical data (MBE, RMSE, MAE, and 'index of agreement' by Willmott were used to evaluate the models). A restriction in the use of QG for Rn-222 concentration was observed only in winter in the case of snow cover occurrence, which reduces the daily QG variability.

**Key words:** air temperature • Central Poland • exponential function • Rn-222 • soil heat flux

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## Introduction

Radon (Rn-222), a radioactive gas with a half-life of 3.82 days, is emitted naturally (0.72–1.2 atoms·cm$^{-2}$·s$^{-1}$) from ice-free, unsaturated terrestrial surfaces [1, 2]. The Rn-222 concentration in the near-surface air layer is undoubtedly strongly influenced by local conditions, which are connected with the environment of the gas emissions to the atmosphere (*inter alia*: geological formation, soil characteristics – radium content, permeability and porosity, temperature vertical profile, soil heat flux, and humidity) and microclimatic conditions occurring in the near-ground air layer [3]. Research on Rn-222 in the context of meteorology dates back to the 1920s [4] and has developed intensively since the 1960s. In most publications, Rn-222 has been used to study the atmosphere as a natural tracer of the processes of transport and dispersion of gases [5–11]. Rn-222 has also been used as an indicator of vertical mixing processes resulting in the atmospheric stability regime [12–17]. The temporal variability of the Rn-222 concentration with reference to the meteorological variables (such as wind velocity and direction, atmospheric pressure, air temperature, precipitation, and snow cover occurrence) was presented in the studies by [17–24].

Although Rn-222 research in the meteorological context has a long tradition, highly time-resolved, long-term, and synchronous measurements of near-ground atmospheric Rn-222 concentrations in different environments (*inter alia*: urban and rural areas with microclimate diversity) are not very common. In addition, statistical models for the estimation of Rn-222 concentrations in the near-surface air layer based on the meteorological variables are not well-documented in literature. Notably, the relationship between the atmospheric Rn-222 concentration and soil heat flux is poorly understood.

Soil heat flux as a component of the heat balance of the active surface (ground surface) indirectly characterizes the thermal properties of the substrate, which are important for the intensity of soil gas flow into the atmosphere. The sign and value of QG reflect the temperature differences between the surface and the deeper layers of the soil [25, 26]. Therefore, this element may give an overview of the microclimate in the near-ground air layer, indicating the degree of heating or cooling of the earth's surface; it may also be an indicator of the Rn-222 exhalation rate. The positive values of QG are a factor in soil gas migration to the surface of the earth, theoretically intensifying the Rn-222 exhalation into the atmosphere. The negative values of QG occur as a consequence of the inverse temperature profile of the substrate soil, which inhibit the transport of gases to the surface of the earth [27, 28].

The main goal of this study was to evaluate two meteorological variables, soil heat flux and air temperature, as predictors of the air Rn-222 concentration variability in the near-surface air layer. The partial objectives were: (i) recognition of the temporal variability of the Rn-222 concentration with reference to selected meteorological elements and assessment of the statistical relationships between the variables, (ii) creation of statistical models for estimation of the Rn-222 air concentrations based on the soil heat flux and air temperature, (iii) validation of the models for particular months, and (iv) comparison of modelled Rn-222 data based on the soil heat flux and air temperature values.

## Data and methods

The study area comprised two sites with different microclimatic conditions – the centre of Łódź (typical urban station, 51°46'10"N, 19°27'55"E, 214 m a.s.l.) and a rural, agrarian area in Ciosny village (25 km north of Łódź, 51°55'24"N, 19°24'38"E, 150 m a.s.l.). At the Łódź and Ciosny stations, continuous synchronous measurements of the radon (Rn-222) concentrations were conducted in the years 2008–2010, using an ionization chamber AlphaGUARD®PQ2000PRO (Saphymo GmbH) in the diffusion mode (averaging time: hour) placed in a meteorological box 2 m above the ground. The air temperature ($t$) was measured using AlphaGUARD®PQ2000PRO and the soil heat flux (QG) was measured by means of HFP01 Heat Flux Plate, Campbell Scientific Ltd. The plates for the measurements of heat flux in the soil were placed at a depth of approximately 10 cm. The surrounding land area was covered with grass at both the measuring stations. The heat flow was expressed in [W·m$^{-2}$]. Positive QG means heat flowing to the surface and negative QG is the propagation of heat from the surface of the active agent into the soil. The soil heat flux observations are logged by CR10X Campbell Scientific, Inc. as 10-min averages of 10-second readings and subsequently integrated into hourly averages for analysis.

The exponential function (model I) and the exponential function with time derivative of predictor to account for the hysteresis issue (model II) were selected, including but not limited to polynomial and power functions, to describe the relationship

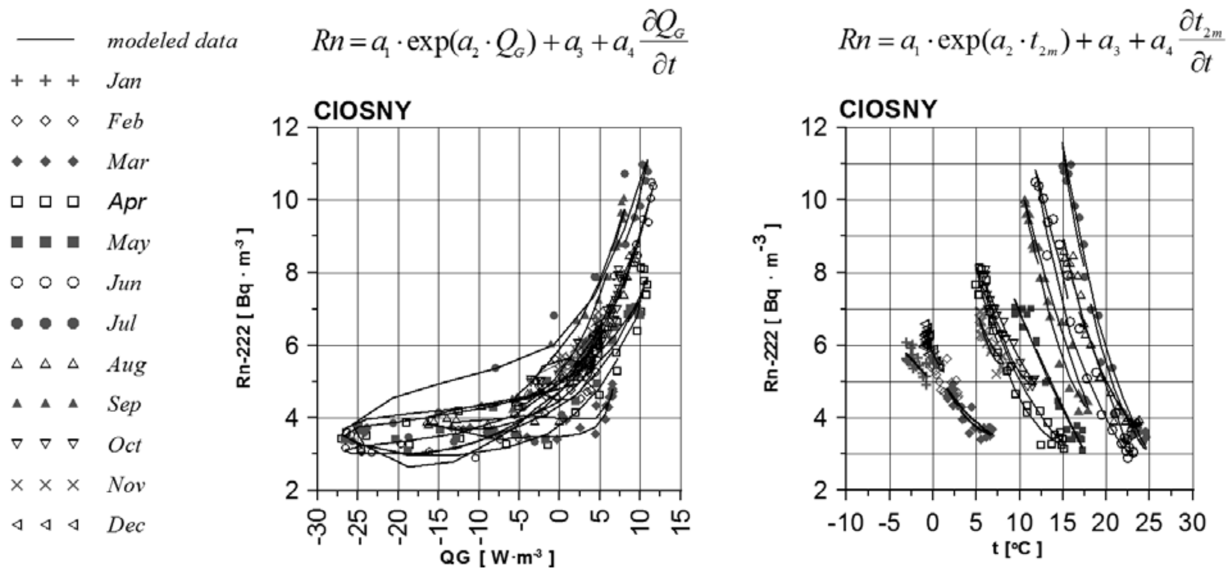


**Fig. 1.** Average hourly Rn-222 concentration as a function of soil heat flux (QG) and air temperature ($t$) for each month in Ciosny in 2008–2010 (dependence of average daily profiles). The lines mark the regression curves for consecutive months (model II with time derivative).

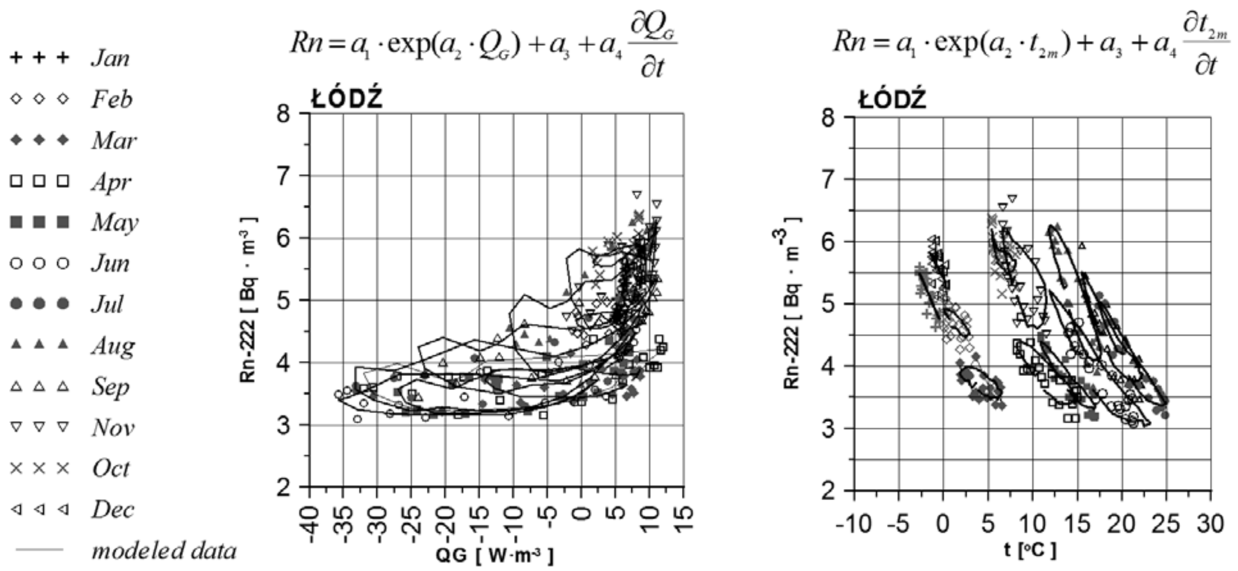


**Fig. 2.** Average hourly Rn-222 concentration as a function of soil heat flux (QG) and air temperature ($t$) for each month in Łódź in 2008–2010 (dependence of average daily profiles). The lines mark the regression curves for consecutive months (model II with time derivative).

the between monthly average hourly values of the Rn-222 concentrations and the meteorological variables. Estimates of the Rn-222 concentrations were performed for individual months due to the fact that the diurnal variability of the Rn-222 concentrations throughout the year was very high. The hysteresis effect was demonstrated in the form of a loop in the correlation plots of the Rn-222 concentration and QG and $t$, particularly evident in the months from April to October (Figs. 1 and 2).

The following are the statistical models of dependence of the Rn-222 concentration on heat flux or temperature, used in the study:

Model I ($t_{2m}$): $\mathrm{Rn} = a_1 \cdot \exp(a_2 \cdot t_{2m}) + a_3$

Model I (QG): $\mathrm{Rn} = a_1 \cdot \exp(a_2 \cdot Q_G) + a_3$

Model II ($t_{2m}$): $\mathrm{Rn} = a_1 \cdot \exp(a_2 \cdot t_{2m}) + a_3 + a_4(\partial t_{2m}/\partial t)$

Model II (QG): $\mathrm{Rn} = a_1 \cdot \exp(a_2 \cdot Q_G) + a_3 + a_4(\partial Q_G/\partial t)$

where, $t_{2m}$ – air temperature at 2 m a.g.l.

The function parameters were chosen using the method of least squares so that they describe the experimental results of the Rn-222 concentration most accurately. The fit of the data obtained using the presented models to the empirical values was evaluated based on the following fit statistics: systematic error (MBE), mean square error of measurement (RMSE), mean absolute error (MAE), and the index of agreement by Willmott ($d$) [29, 30]. The index of agreement ($d$) varies between 0.0 and 1.0, where a value of 1.0 expresses perfect agreement between $O$ (observed values) and $P$ (modelled values), and 0.0 describes complete disagreement.

Following are the fit statistics used in the study:
1) Mean bias error,

$$\mathrm{MBE} = N^{-1}\sum_{i=1}^{N}(P_i - O_i) = \frac{\sum P_i}{N_i} - \frac{\sum O_i}{N_i} = \overline{P}_i - \overline{O}_i$$

2) Root mean square error of measurement

$$\mathrm{RMSE} = \left[N^{-1}\sum_{i=1}^{N}(P_i - O_i)^2\right]^{0.5}$$

3) Mean absolute error

$$\mathrm{MAE} = N^{-1}\sum_{i-1}^{N}|P_i - O_i|$$

4) The index of agreement by Willmott

$$d = 1 - \left[\frac{\sum_{i=1}^{N}(P_i - O_i)^2}{\sum_{i=1}^{N}(|P_i'| + |O_i'|)^2}\right], 0 \le d \le 1$$

where: $N$ – number of cases, $O$ – observed values, $P$ – modelled values. $P' = P_i - \overline{O}$, $O' = O_i - \overline{O}$, where $\overline{O}$ – average value.

The expression in the denominator is referred to as the error potential (PE), and the expression in the numerator is the mean square error of measurement (MSE) [30]. Willmott's index of agreement $d$ is a more accurate fit statistic than those commonly used, that is, the correlation coefficient $R$ and the coefficient of determination $R^2$, which are applied for linear models.

For more details concerning the area of investigation, instrumentation and data processing, see the following papers: [17, 24].

## Results

The soil heat flux (QG) and air temperature ($t$), as compared with the other meteorological variables (e.g., soil heat flux, soil humidity, air temperature in the layer 0.2–2.0 m a.g.l., wind speed, and atmospheric pressure) are characterized by a clear

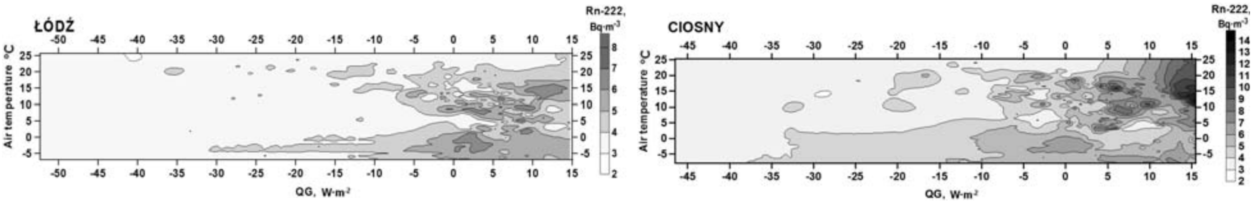


**Fig. 3.** Average Rn-222 concentrations depending on air temperature and soil heat flux (QG) in Łódź and Ciosny in 2008–2010 (based on the mean monthly hourly values).

diurnal cycle and the strongest statistical relationships with the atmospheric Rn-222 concentrations [24]. For this reason, QG and *t* were used to create a statistical model for the estimation of the Rn-222 concentrations at 2 m a.g.l. The daily profile of the Rn-222 concentration represents an inversion of the air temperature profile, and it varies approximately in phase with the soil heat flux. In Ciosny, the highest monthly average Rn-222 concentration was observed at a temperature range of 10–15°C, whereas for the urban areas, this range was 5–10°C. The highest monthly average concentrations of Rn-222 were recorded at QG > 5 $W \cdot m^{-2}$ and the lowest at QG < –10 $W \cdot m^{-2}$ at both sites (Figs. 1, 2, and 3).

Still, extreme concentrations of the radionuclide near the ground occur with some delay as compared to the minima and maxima of the meteorological variables. This hysteresis effect, whose physical reason is the inertia of the physical system, was taken into account in the prediction model II of Rn-222 levels at both stations.

An analysis of the temporal variability of the Rn-222 concentrations indicated a lack of clear diurnal cycles from January to March at both the urban and rural stations (Figs. 4 and 5). In this season of the year, there are two important factors determining the atmospheric Rn-222 levels: (i) the intense dilution processes of atmospheric mixing due to an increased frequency of cyclonic weather with strong winds and (ii) the snow cover weakening the exhalation process. The average daily amplitude of the Rn-222 concentrations at the stations begins to increase in April with a maximum in June (Ciosny – 11 $Bq \cdot m^{-3}$) and September in Łódź (3 $Bq \cdot m^{-3}$) (Figs. 4 and 5). The morning increase of the Rn-222 concentrations outside the city is distinctly higher

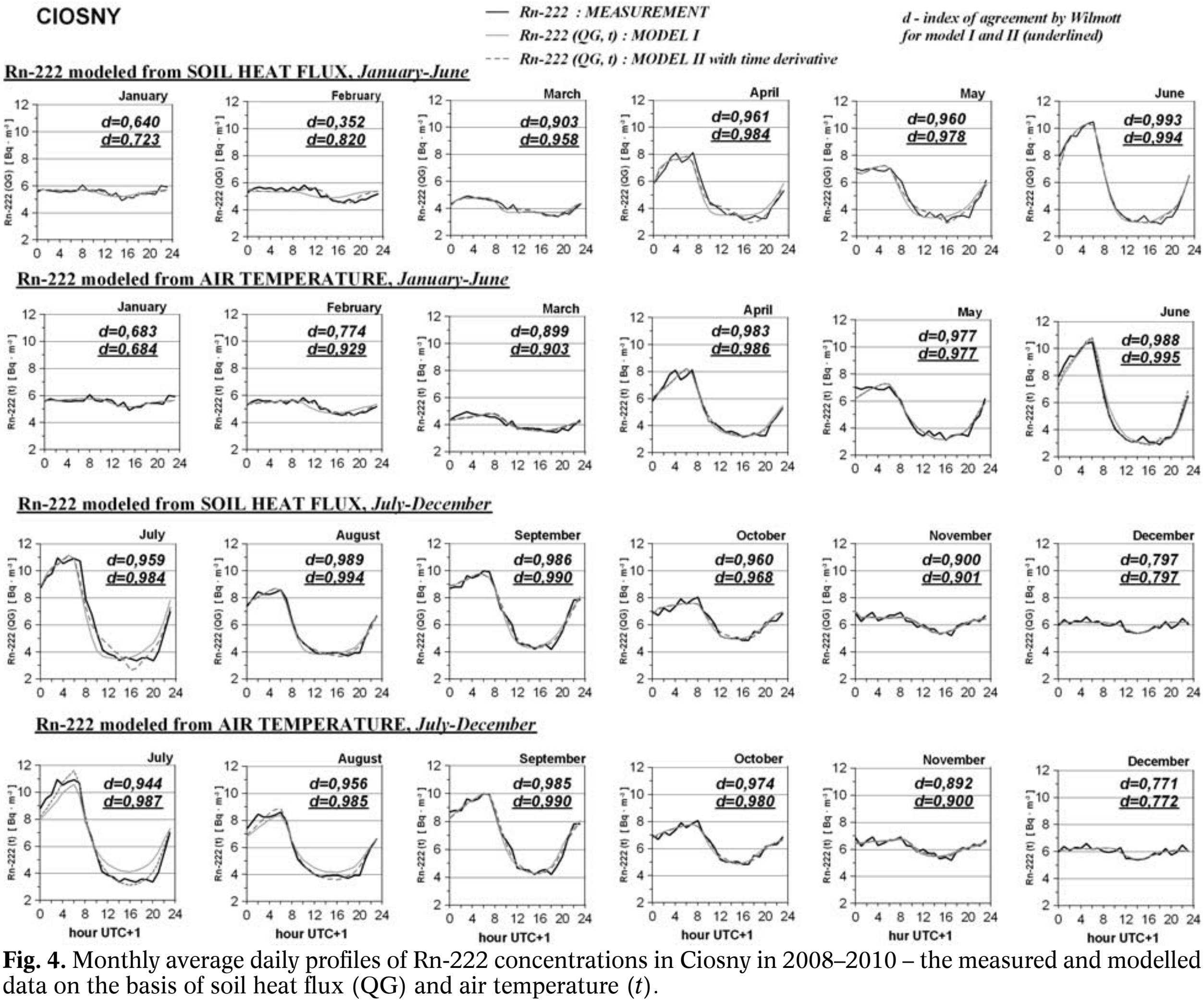


**Fig. 4.** Monthly average daily profiles of Rn-222 concentrations in Ciosny in 2008–2010 – the measured and modelled data on the basis of soil heat flux (QG) and air temperature (*t*).

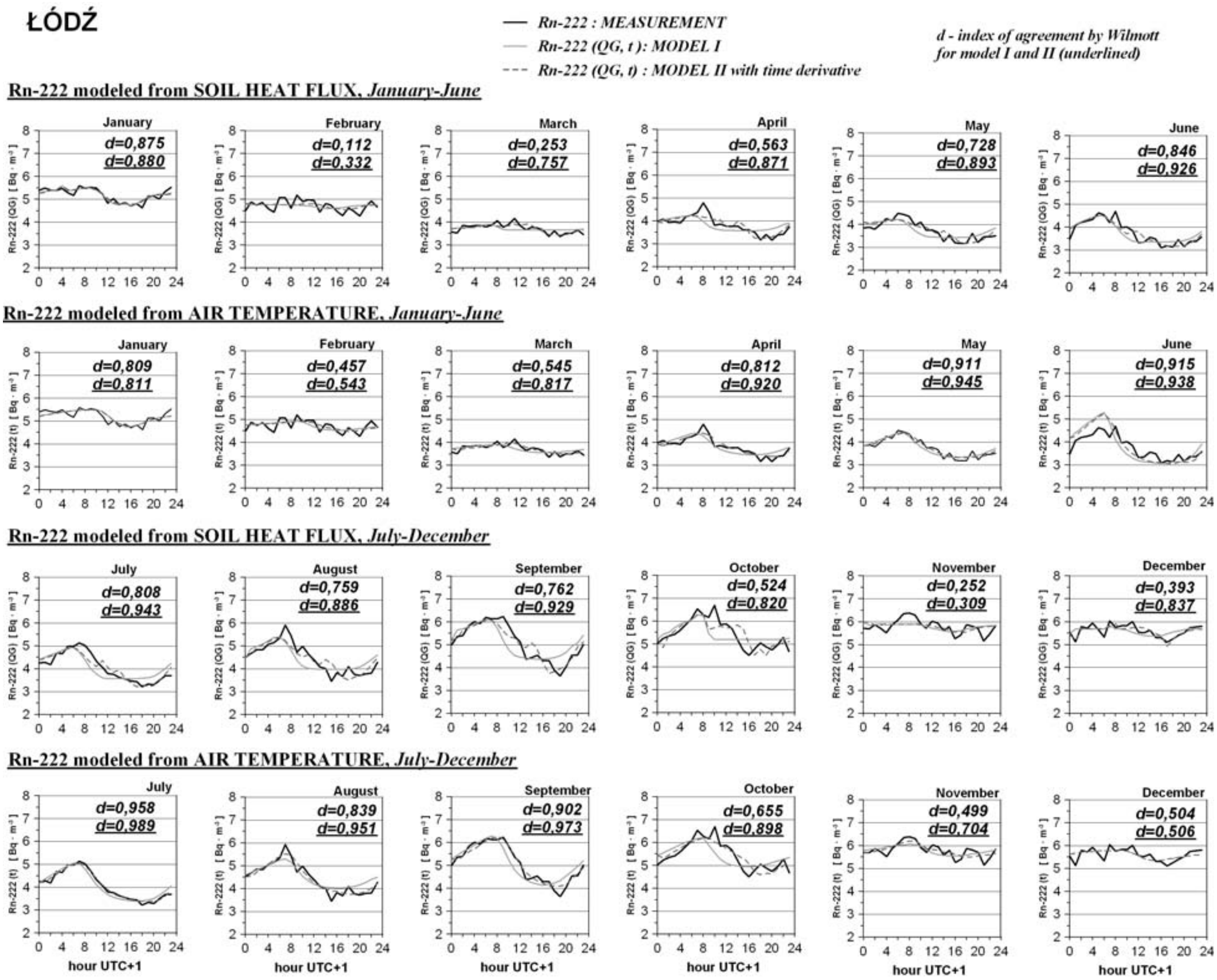


**Fig. 5.** Monthly average daily profiles of Rn-222 concentrations in Łódź in 2008–2010 – the measured and modelled data on the basis of soil heat flux (QG) and air temperature ($t$).

than in Łódź in all seasons except winter, which originates from the urban-rural difference in the diurnal evolution of the atmospheric boundary layer.

The air temperature as well as the soil heat flux--driven models exhibited very high agreement with the empirical data, especially from April to October. The index of agreement by Wilmott varied from 0.524 to 0.989 in Łódź and from 0.944 to 0.990 in Ciosny during this period. The models Rn(QG) and Rn($t$) with time derivative (model II) provided slightly better results in all months of the year than model I (Figs. 4 and 5). A restricted use of QG for the prediction of the Rn-222 concentrations was observed only in winter in the case of snow cover occurrence, which reduces the daily QG variability (e.g., February, November in Łódź, Fig. 5).

An analysis of the values of fit statistics ($d$, MBE, RMSE, and MAE) confirmed that the choice of the regression models was good. *Inter alia*, the systematic error in all cases reached a value close to 0.

The values of the coefficients of determination calculated for the experimental and modelled values of the Rn-222 concentrations indicated that the measured Rn-222 levels at the rural station Ciosny reflected slightly better the model data than those from the urban station in the city centre of Łódź (Figs. 6 and 7). In Łodź, models I and II based on the air temperature have given better results ($R^2$:

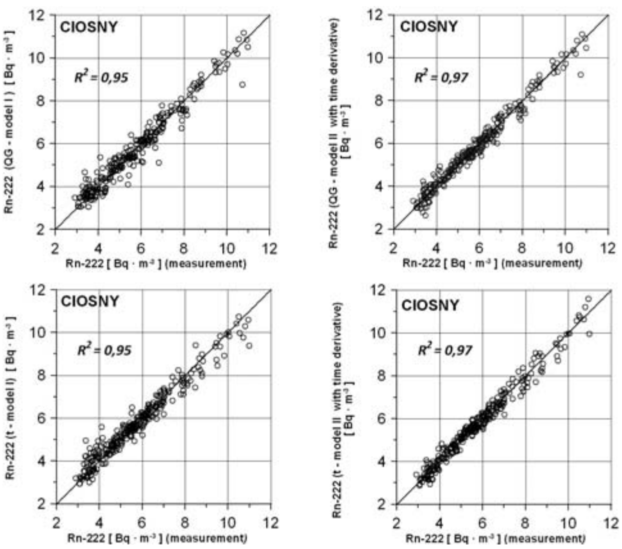


**Fig. 6.** Experimental and modelled values of Rn-222 concentrations from soil heat flux (QG) and air temperature ($t$) in Ciosny in 2008–2010 (calculations were based on the average daily values).

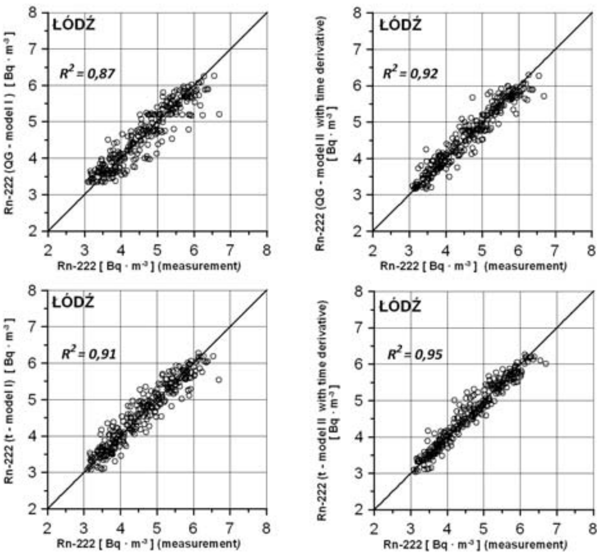


**Fig. 7.** Experimental and modelled values of Rn-222 concentrations from soil heat flux (QG) and air temperature ($t$) in Łódź in 2008–2010 (calculations were based on the average daily values).

0.91, 0.95) than the models based on the soil heat flux ($R^2$: 0.87, 0.92), but in Ciosny, the accuracy of both models was the same (Figs. 6 and 7).

## Conclusion

An analysis of the three-year data series of the soil heat flux and air temperature as predictors of the Rn-222 concentrations at the near-the-ground air layer at urban and rural areas showed the following results:

1. The soil heat flux (QG) and air temperature ($t$), as compared with other meteorological variables, have the strongest statistical relationships with the Rn-222 concentrations in the near-surface air layer. The hysteresis effect occurred in the diurnal profiles of the values of Rn-222 and the selected meteorological variables.
2. An increase in the Rn-222 levels was observed during positive values of QG in all months of the year (heat flowing to the surface – usually started at ~8.00 p.m. and lasted until morning hours), which could prove the important role of QG as a factor in soil gas migration to the surface of the earth, intensifying the Rn-222 exhalation.
3. The exponential function (model I) and exponential function with time derivative of predictor to account for the hysteresis issue (model II) were selected to describe the relationship between the monthly average hourly values of concentrations of Rn-222 and meteorological variables – QG and $t$.
4. The period from April to October was characterized by a good agreement between the observed and model-predicted values of the Rn-222 concentration.
5. Model II, taking into account the hysteresis effect, provided slightly better results.
6. In winter months, the snow cover occurrence reduced the daily QG variability and excluded this variable as a predictor of the atmospheric Rn-222 levels.
7. The soil heat flux and air temperature could be used as complementary predictors of the Rn-222 concentration in the near-surface air layer. The index of agreement by Willmott indicated both the meteorological variables as good predictors of the Rn-222 concentration in the near-surface air layer.

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