NUKLEONIKA 2020;65(2):115–119
doi: 10.2478/nuka-2020-0018

ORIGINAL PAPER



# $^{214}$Bi/$^{214}$Pb radioactivity ratio three-year monitoring in rainwater in Prague

**Fabrizio Ambrosino**,
**Lenka Thinová**,
**Miroslav Hýža**,
**Carlo Sabbarese**

**Abstract.** Continuous monitoring of natural gamma radiation in air has been carried out, during December 2014 – January 2018, with 1-min cyclic measurement in Prague, Czech Republic using a NaI(Tl) probe. The $^{214}$Bi/$^{214}$Pb ratio as a tracer in rainwater has been investigated to study its variations related to both the ambient dose equivalent rate per hour and the amount of rainfall. A hybrid methodology for time series analysis, composed of the aggregation of two signal decomposition methods (multiple linear regression and empirical mode decomposition) and one forecasting method (support vector regression), has been applied to identify the anomalies in the studied signals in order to better find correlations among them. The results show a strong correlation between the ambient dose equivalent rate and the $^{214}$Bi/$^{214}$Pb ratio values and between both these signals and rainfall amount ≥5 mm/h. Furthermore, the considered descendants of radon are mainly responsible for the overall ambient dose equivalent rate.

**Keywords:** $^{214}$Bi/$^{214}$Pb gamma-ray ratio • Ambient dose equivalent rate • Hybrid method • Rainfall • Time-series analysis

F. Ambrosino✉
Department of Mathematics and Physics
University of Campania "Luigi Vanvitelli"
Viale Lincoln 5, 81100 Caserta, Italy
and Faculty of Nuclear Sciences and Physical Engineering, Czech Technical University in Prague
Břehová 7, 11519 Prague, Czech Republic
E-mail: fabrizio.ambrosino@unicampania.it

L. Thinová
Faculty of Nuclear Sciences and Physical Engineering
Czech Technical University in Prague
Břehová 7, 11519 Prague, Czech Republic

M. Hýža
National Radiation Protection Institute (SÚRO)
Bartoškova 28, 14000 Prague, Czech Republic

C. Sabbarese
Department of Mathematics and Physics
University of Campania "Luigi Vanvitelli"
Viale Lincoln 5, 81100 Caserta, Italy

Received: 2 December 2019
Accepted: 17 January 2020

## Introduction

Long-term monitoring of environmental radioactivity is an integral part of any systematic approach of the radiation protection in a given area [1–6]. Such monitoring can be carried out in different ways and the most common ways are gamma-ray spectrometry and gamma dose rate measurements [1, 6–9]. In this framework, the continuous monitoring of radioactivity ratio of $^{214}$Bi/$^{214}$Pb as tracers in rainwater was carried out by several works [9–12]. During atmospheric thermal inversions, there is an increase in the concentration of natural gamma-emitting radionuclides of the short-lived $^{222}$Rn progeny ($^{214}$Pb and $^{214}$Bi with half-lives of 26.9 min and 19.9 min, respectively) that causes an increase in the total natural gamma background. These descendants of radon are incorporated in the raindrops and dragged to the ground [13]. In the atmospheric mixing of the overall variability in ambient gamma dose, radon variability attributable to diurnal changes contributes less than radon variability during precipitation periods [9, 10]. Therefore, any information about radon progeny concentrations could be useful for modelling the variation of the natural gamma background depending on the $^{222}$Rn flux and the variable atmospheric conditions [11, 12]. Moreover, the variations of radon and its short-lived progeny in air have been studied for a long time as indicators of height fluctuations of the mixed layer and potential air pollution conditions [3, 6, 12].

The present work addresses the study of $^{214}$Bi/$^{214}$Pb ratio monitored during three years (December 2014 – January 2018) in Prague, using a NaI(Tl) gamma probe, aiming to discover correlation of that signal with dose rate [μSv/h] and rainfall [mm/h]. The obtained time series of monitoring results have been analysed by a mathematical hybrid method on the basis of assembling more known signal processing methods that are able to identify anomalies.

## Materials and methods

### Gamma-ray monitoring and dose rate measurements

The 3-year monitoring of environmental radioactivity, from December 2014 to January 2018, was performed by gamma-ray spectrometry and continuous measurements of hourly ambient dose equivalent rate in air, using a 3" × 3" NaI(Tl) gamma probe of the NUVIA Czech company. The probe, covered by a canopy to avoid accumulations of raindrops and/or other materials, was placed in the open air on the large balcony (at third floor) of the building belonging to the Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University in Prague (N 50°05'28.1" – E 14°24'57.7"). The balcony is surrounded on three sides by the walls of the building; therefore measurements were also influenced by the gamma radiation of the ground, coming from the surrounding building materials.

The probe provides for the gamma-ray spectrum and the dose rate was calculated on the basis of the entire spectrum. The counting rates of the gamma-ray line at 352 keV and the sum of the 609 keV and 1120 keV gamma-ray lines were considered for measuring $^{214}$Pb and $^{214}$Bi activity concentrations, respectively. Moreover, the outside meteorological parameters, namely temperature [°C], relative humidity [%], pressure [hPa], wind speed [m/s], rainfall [mm/h], were recorded during the study period. All data were recorded using a 1-min interval and the daily average values were considered for statistical analysis.

### Time-series analysis method

The analysis of the recorded time series is performed to estimate various components present in the signals, to differentiate those due to known and/or unknown parameters (seasonal and meteorological effects) and to recognize the possible anomalies [14]. In the present work, the time-series analysis was performed by the application of a hybrid forecasting method which is able to combine multiple known methods, to combine the strengths of single ones with optimized algorithms and to achieve greater accuracy [15]. The used hybrid method was sequentially composed of two methods for the signal decomposition, i.e. multiple linear regression and empirical mode decomposition methods, and the other one for the forecasting of the time series, i.e. support vector regression method. This hybrid method was well-proven in several studies based on the statistical evaluation criteria of the uncertainty [15–19].

First, the multiple linear regression method estimates the contribution of the monitored meteorological parameters in the time series through a multiple linear regression model, based on the least-squares fit, to differentiate these known modulations within the time series. A new time series is obtained as the difference between the unprocessed time series and that derived from the model [14]. Then, at that time series, the empirical mode decomposition method is applied to further differentiate the unknown periodic modulations within the signal. The decomposition process is performed by iterative differences between the signal and the spline interpolations average from the local maxima and minima of the same signal [15, 16]. This process stops when the residual signal is constant or monotone [15]. At this point, the support vector regression method is applied, first on each component and then on their sum, to forecast the signal, via regression model, with a function having the lowest deviation from the original data set. The last stage of the application of the hybrid method to the studied time series is the recognition of anomalies which are obtained by the difference between the raw starting time series and the forecasted signal [17, 18]. The entire procedure described for the time-series analysis is performed under normalized estimate of the data as *Z*-score (datum minus average divided by standard deviation), developed and tuned in Matlab® environment.

## Results and discussion

### Statistical analysis of the data

A statistical analysis has been performed on the recorded data during the monitoring period of three years. The average value of hourly ambient dose equivalent rate was 0.101 ± 0.002 μSv/h (Fig. 1), which corresponds to an average value of 0.885 ± 0.002 mSv/year. This value is consistent, within the uncertainty, with the worldwide average of the dose rate due to the natural background gamma radiation of ~0.88 mSv/year from external/cosmic (~0.4 mSv/y) and terrestrial (~0.48 mSv/y) sources, estimated by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [20]. The terrestrial gamma-radiation source was also considered since the used probe is influenced by the radiation coming from the ground andthe building materials surrounding the balcony where the instrument was placed.

In the present study, to estimate the degree of similarity (or dissimilarity) between two discrete time series, as a function of the displacement of one relative to the other, the cross-correlation parameter was adopted [16]. The average cross-correlation value of 70 ± 3% was found between the raw dose rate and the rainfall signals, and the other meteorological signals are almost uncorrelated with the dose rate.

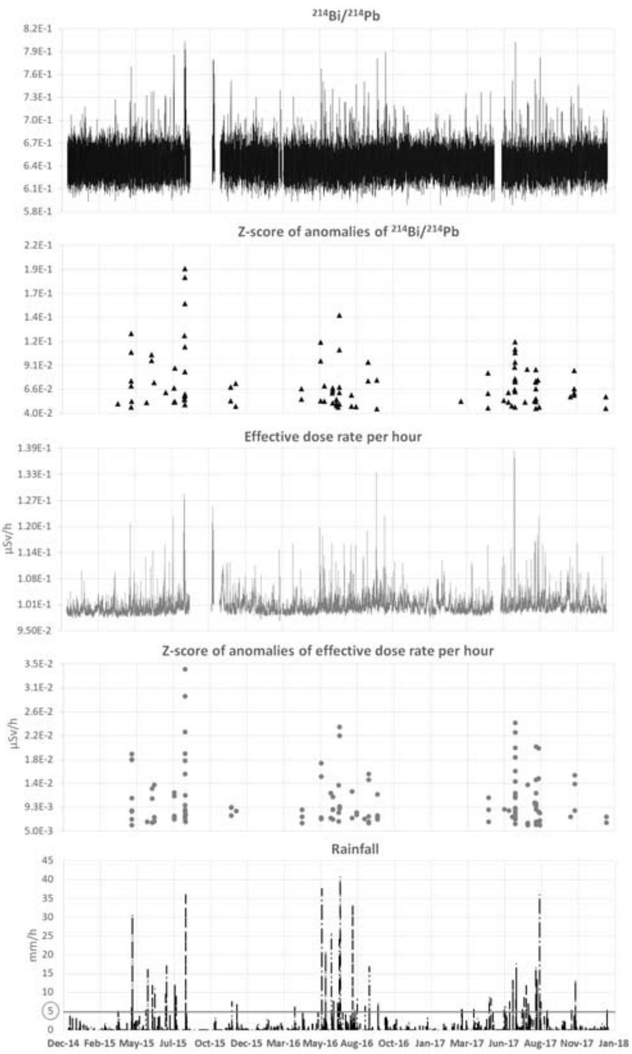


**Fig. 1.** Comparison between the daily time series of $^{214}Bi/^{214}Pb$ ratio (in black) and hourly ambient dose equivalent rate (in grey) with their anomalies results as *Z*-score (in black triangles and grey circles, respectively) obtained from the application of the proposed hybrid method for time-series analysis (multiple linear regression + empirical mode decomposition + support vector regression) during the period December 2014–January 2018. On the bottom of the graph, a further comparison between the above signals and the days when rainfall occurs (with dashed lines) is shown, where strong correlation is found with rainfall ≥5 mm/h (above the horizontal grey line).

## Analysis of the radon progeny

The developed hybrid method was applied to the daily time series of $^{214}Bi/^{214}Pb$ ratio and the time series of dose rate, during the period December 2014 – January 2018, to identify the possible anomalies in both signals. In Fig. 1, the results of the time-series analysis compared with the raw starting signals are reported.

To evaluate the degree of similarity (or dissimilarity) between two-point process series (i.e. anomalies results), the Ripley's spatial descriptive estimator was adopted [19, 21]. This parameter estimate shows point patterns occur over a given time interval. This was performed fixing each point in the first process and changing the distance in days (ranging ±30 days) between it and all points in the second process, and then an average value was taken. This analysis proves that anomalies of $^{214}Bi/^{214}Pb$ ratio and dose rate time series were clustered within ±2 days on average. The cross-correlation between the two studied raw time series has also been calculated: the average value of 84 ± 2% was obtained. From this statistical analysis, the progeny of radon is the main reason for the overall ambient dose equivalent rate [11, 12, 22].

In Fig. 1, a further comparison is also shown where the previous signals are compared with the amount of rainfall. The cross-correlation parameter between the raw $^{214}Bi/^{214}Pb$ ratio and the rainfall time series showed the average value of 89 ± 2%, while the raw $^{214}Bi/^{214}Pb$ ratio signal was not correlated with the other climatic parameters. The Ripley's spatial descriptive estimation demonstrated that both anomalies of $^{214}Bi/^{214}Pb$ ratio and dose rate time series were clustered within ±3 days on average with respect to the days with rainfall ≥5 mm/h.

It is worth pointing out that the high values of $^{214}Bi/^{214}Pb$ ratio in the raw signal, greater than or equal to 0.7, seem to be associated with the simultaneous occurrence of low wind speed and high relative humidity (Fig. 2), most likely due to atmospheric water vapour condensation [11, 12].

The present study confirms the results obtained by other authors in similar studies through different measurements and methodologies of analysis [1, 9–13]. Anomalies of $^{214}Bi/^{214}Pb$ ratio, during rainfall events, reflect anomalies of the $^{222}Rn$ (their progenitor) that has a half-life of about 3.82 days and is

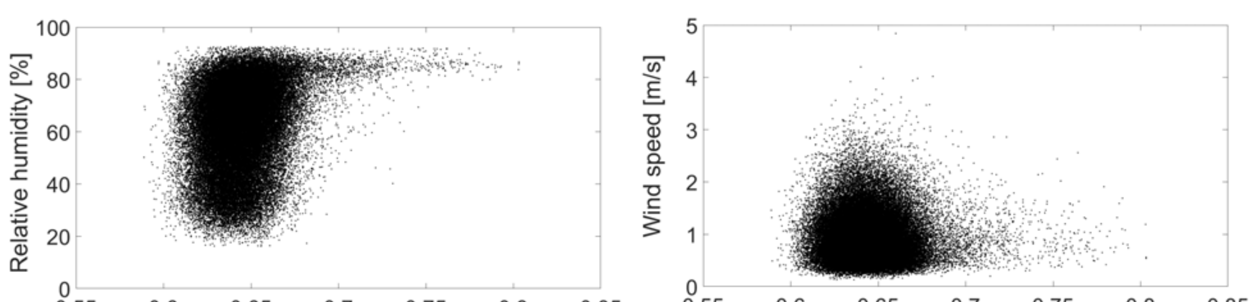


**Fig. 2.** Two graphs show the recorded meteorological signals of relative humidity (in %), and wind speed (in m/s) vs. the raw time series of $^{214}Bi/^{214}Pb$ ratio.

incorporated in the raindrops due to its solubility in water [11–13, 23].

## Conclusion

The $^{214}Bi/^{214}Pb$ radioactivity ratio has been measured continuously for three years (December 2014 – January 2018) in Prague, Czech Republic. The ambient dose equivalent rate and meteorological parameters were also monitored. The relationship among the investigated signals has also been studied through the application of a hybrid forecasting method to identify anomalies in the time series. The used hybrid method first decomposes the raw time series and then forecasts it; finally, the anomalies are obtained by the difference between the raw time series and the forecasted signal. The following results are obtained:

i) The time series of dose rate showed an annual average value of 0.885 ± 0.002 mSv/year, consistent, within the uncertainty, to the worldwide average of the dose rate due to the natural background gamma radiation of Σ0.88 mSv/year from external/cosmic and terrestrial sources.
ii) Anomalies of $^{214}Bi/^{214}Pb$ ratio and of dose rate time series were clustered within ±2 days on average; the average value of the cross-correlation parameter between the two time series was 84 ± 2%.
iii) The cross-correlation parameter between the dose rate and the rainfall signals was equal to 70 ± 3% on average, and the other meteorological parameters were uncorrelated.
iv) The cross-correlation parameter between the $^{214}Bi/^{214}Pb$ ratio and the rainfall signals was equal to 89 ± 2% on average, and the other meteorological parameters were uncorrelated.
v) Both anomalies of $^{214}Bi/^{214}Pb$ ratio and dose rate time series were clustered within ±3 days on average with respect to the days when rainfall occurs ≥5 mm/h.
vi) The high values of $^{214}Bi/^{214}Pb$ ratio signal (≥0.7) were associated with the simultaneous occurrence of low wind speed and high relative humidity.

The overall results obtained in the present work outline two important points: the progeny of radon is the main reason for the overall ambient dose equivalent rate and seasonal variation in the frequency of rainfall has strong effects on the environmental radioactivity of short-lived radon progeny.

## ORCID

*F. Ambrosino* http://orcid.org/0000-0003-2122-9130

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