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APPLICATION OF POLONIUM CONCENTRATION FOR ESTIMATION THE MIXING HEIGHT IN AIR POLLUTION PROBLEMS

Krajny E, Osrodka L.[1], Skowronek J.[2], Skubacz K.[3] and Wojtylak M.[4]

[1]Institute of Meteorology and Water Management (IMGW), Department of Monitoring and Environmental Research, Warsaw, Branch of Katowice, Bratkow 10, 40 045 Katowice, Poland, leszek.osrodka@imgw.pl
[2]Institute for Ecology of Industrial Areas (IETU), Kossutha 6, 40 844 Katowice, Poland, j.skowronek@ietu.katowice.pl
[3]Central Mining Institute (GIG), Laboratory of Radiometry, Pl. Gwarkow 1, 40 166 Katowice, Poland, k.skubacz@gig.katowice.pl
[4]Silesian University (US), Institute of Mathematics, Bankowa 14, 40 007 Katowice, Poland, monitoring.katowice@imgw.pl

ABSTRACT

The paper presents a new method of evaluation of the mixing layer height (MLH). The data analysis bases on study of variation of chosen meteorological parameters and its comparison with change of the radon Rn-222 concentration in soil gas and polonium Po-218 concentration in the atmospheric air. The Fourier-, Wavelet-, Range Rescaled- and Regression analysis were used for this aim. The obtained results show that the variation of the Po-218 concentration in response to the diurnal changes of mixing state of the lower troposphere may become a good tracer to estimate the atmosphere dynamics and transport of pollution by the turbulent mixing of atmospheric air. The statistically significant relation between the MLH and Po-218 concentration in the air was found and, as a result, the statistical model was developed to define the MLH during the hot season of the year (May-October).

Key Words: Mixing Layer Height (MLH), Air Quality, Natural Radioactivity, Alpha Spectroscopy

1. INTRODUCTION

The air pollution strongly depends on thermodynamic state of the atmosphere. Among meteorological factors, the mixing layer height (MLH) is of the great importance. This parameter influences the form and intensity of diffusion in the atmosphere and, as a consequence, the concentration of primary and secondary pollutants in the lower atmosphere (Garratt, 1992). It shows characteristic diurnal variation but can not be measured immediately. Therefore it is evaluated indirectly by measuring of the others quantities.There is no commonly accepted method of the MLH evaluation due to different genesis of its generation in the boundary layer of the atmosphere taking into account different time of day. The Sodar was used for this aim since 80's of the last century (Reitebuch and Emeis, 1998). Although the obtained outcomes are not satisfactory for every meteorological conditions, this

method nevertheless makes it possible to achieve the unmistakable results of so-called equivalent MLH. On the other hand however, the remote sensing monitoring is relatively expensive which limits their application. This work is aimed at presentation of the cheaper method of the MLH evaluation that relies on measurement of natural radioactivity in the atmosphere.
The description of the MLH on base of measurements of the natural radioactivity is one of the methods used in evaluation of the mixing process in the atmosphere (Allegrini et al., 1994, Fujinami and Esaka, 1998, Kataoka et al., 2001, Porstendörfer et al., 1991). According to this, the concentration of the chosen natural occurring radioactive pollutants in air was observed. Up to now, the radon concentration in atmospheric air was taken into account. Radon (strictly, Rn-222), is a member of the uranium series and, under normal conditions, occurs as gas. As a result of turbulence and convection, radon can be transported from soil to considerable heights above the ground and contributes to the ionization of the atmosphere. Such measurements can be however difficult because the natural level of radon concentration in the atmospheric air is rather low. Radon itself decays by alpha emission and its half-life is equal to 3.82 day. The isotopes of polonium, bismuth and lead called the short-lived radon daughters are generated as a result of the radioactive decay of radon (Nazaroff, 1992). The new approach relies on measurements of the polonium concentration (Po-218) in the atmosphere close to the ground. The polonium has considerable shorter half-life (3.05 minutes) than radon, so between these two isotope the equilibrium state will be reached within very short time. On the other hand, the polonium-218 as a free atom, claster or aerosol can be simply collected on a filter and its activity precisely measured by an alpha spectroscopy system. Such method makes it possible to considerably decrease a lower limit of detection in relation to measurement of radon concentration.
The particular emphasis was laid to confirm the assumption about quasi-stable radon concentration in the soil gas. The presented method of evaluation of the MLH, after automatization of the measurement process, can be used for operational modeling of the imission of air pollution.

2. INSTRUMENATION AND MEASUREMENTS

The measurement sites were located in city of Katowice and Cracow (southern part of Poland) and in Ostrava (north part of Czech Republic), close to the meteorological stations of the National Weather Services (NWS). The stations collect meteorological data according to the WMO standards. Additionally, there were conducted gradient measurements of the following meteorological elements: temperature and velocity at 2 and 10 m above ground and the gradient of Richardson's Number (Ri) was calculated basing on these results.

2.1 Barasol

The measurements of the Rn-222 concentration in soil gas were performed on the depth of 1m by means of the Barasol probe. The probe is adjusted to long-term measurements all year round. This device is equipped with a circular silicon detector of the PIPS type with effective area of 450 mm^2 (see Table 1).

The measurements were performed in 1999-2003 in Katowice and Cracow and the results were averaged over a period of 15 minutes.

Table 1. BARASOL probe - basic specification.

Probe length	Diameter	Temperature range	Pressure limit	Background	Measuring range
800 mm	60 mm	-20 to +60 ^{0}C	100 kPa	1 impulse/h	0.1 - 3000 kBq/m^3

2.2 Alpha spectroscopy

Measurements of the Po-218 concentration in air were performed by means of the alpha spectroscopy (Krajny et al., 2005, Osrodka et al., 2003). Immediately after decaying of radon, about 80-82 % of polonium-218 occurs as positive ions that are attached to air gas molecules and water particles within 10^{-7} second (Reineking and Porstendörfer,1986). Free atoms and clasters are called "unattached fraction". Then the clasters, after being bound to the other air-suspended particles, can generate the bigger particles called "attached fraction". During measurements, the air together with all particles is drawn through a filter with adjusted flow rate. A special semiconductor CAM PIPS detector with effective area of 1700 mm^2 was placed above this filter to detect alpha radiation emitted by polonium, that was separated out of the air stream and deposited on the filter (Hindus, 1982). Our detector was a part of an alpha spectroscopy system. Therefore there was possibility to distinguish alpha particle energy and identify isotopes collected on the filter (see Table 2).
Activity of the polonium was measured continuously during air sampling. In years 2001-2004 from May to October anywhere from ten and twenty one-day measurements of Po-218 concentration were performed above the ground under different meteorological conditions. These experiments however were conducted during the well-marked variation of the MLH. The polonium concentrations were then compared with the reference results obtained by the sodar instrumentation.

Table 2. Alpha spectroscopy instrumentation - basic specification.

Nucleopore polycarbonate filter		Flow rate	Sampled time	Lower limit of detection (at 5% significance level)
pore size	effective diameter			
0,8 μm	40 mm	60 dm^3/min	1 hour	0.15 Bq/m^3

2.3 Remote sensing

The remote sensing method was used to collect reference data related to the MLH. The monostatic sodar SAMOS-4C (Katowice) and REMTECH PA2 Doppler sodar (Cracow and Ostrava) were included to perform such measurements. The Doppler sodar was able to evaluate the characteristic of atmospheric boundary layer (vertical wind component, mixing height and inversion level) by means of the Doppler analysis. Physical parameter data of the boundary layer were collected in real time

every six seconds by SAMOS and every half-hour by REMTECH. The frequency ranged from 1.6 to 2 kHz The instruments are optimized for long-range detection up to 1200 m above ground. The data have been analyzed automatically.
Data related to the physical parameters of the atmospheric boundary layer were collected in real time as well. However these result were averaged over a period of 1 hour to harmonize it with measurements of the polonium concentration in air.

3. METHODOLOGY

3.1 Radon concentration in the soil gas

The method of evaluation of the MLH relies on assumption that the radon (Rn-222) concentration in the soil gas is quasi-stable. Such statistical methods as Fourier-, Wavelet-, Range Rescaled analysis were applied to analyze the time series of radon concentration and verify this hypothesis.
This analysis was performed after removal of results that had been affected by geological- and geophysical events. On the other hand, the Linear Regression analysis was used to reveal relations between radon concentration in the soil gas and polonium concentration in air and meteorological conditions, especially the MLH

Frequency analysis

The Fourier method was applied to define the global frequencies (annual and seasonal components) in relation to variation of radon concentration. The FFT algorithm (Fast Fourier Transformation) was include to fulfill this task (Cormen et al., 1990).
The Wavelet analysis helped to find the diurnal variation. This analysis is a generalization of the Fourier analysis and makes it possible, for obtained results related to any natural phenomena, where periodicity seems to occur, to reveal the followings:

- for any frequency, further called the scale, local short-term oscillations,
- oscillations with the same scale and time-changeable amplitude,
- oscillations with time-changeable scale.

The wavelet transform relies on conversion of the data series $x_1,........,x_n$ (real or complex numbers) into the Form Table (from values into frequency values to obtain a relation: frequency-measured value):

$$W_k(s) = \sqrt{\frac{1}{s}} \sum_{j=1}^{n} x_j \overline{\Psi(\frac{j-k}{s})} \qquad \text{k=1,...,n, s=s}_1\text{,...,s}_m \qquad (1)$$

where sign „—" means conjugation of the complex number, function Ψ (real or complex function) is called a basic wavelet, $s_1,...,s_m$ correspond to scales (periods) which have been accepted for wavelet analysis. The Morlet wavelet was applied in the analysis (Bialasiewicz, 2000, Farge, 1992, Torrence et al., 1998).

Range Rescaled Analysis - Hurst exponent

The Hurst exponent (Hurst, 1951, Mandelbrot, 1983) is a statistical tool, so all-purpose, that it can be applied for the fractal analysis of the time series. Hurst did a non-dimensional indicator by dividing of the oscillation range related to specific data series by standard deviation related to obtained observations:

$$\underset{c,H\in R}{\vee}\ \underset{n_0\le n\le N/2}{\wedge}\left(\frac{R}{S}\right)_n = c\cdot n^H \qquad (2)$$

where $(R/S)_n$ is a rescaled range, $M=d\cdot n$ is a series length, d number of subseries, n number of elements that belong to the subseries, n_0 – minimal length of the subseries, $n\in[10; M/2]$, N – number of observations, c – constant, H – Hurst exponent.
By using a double-logarithmic scale, the curve $n\rightarrow(R/S)_n$ can be plotted where its slope is an estimate of the H value. The main point of the analysis is to rescale the range and relies on analyzing of the range R of the time series taking into account different time periods n.The conclusions can be drawn in relation to independence and memory length of the process (even when there are not the Gaussian series) when comparing the series to the similar range in case of the independent random variables. There are three classes of the time series depending on the value of the Hurst exponent: antipersistent or ergodic series ($0 \le H < 0.5$), "white noise" ($H = 0.5$) and persistent series ($0.5 < H \le 1$).

Results of the Analysis

Fourier analysis
According to the Fourier analysis, there was not observed the cyclic variation of the Rn-222 concentration in the soil gas up to depth of 1m (Fig. 1, 2).

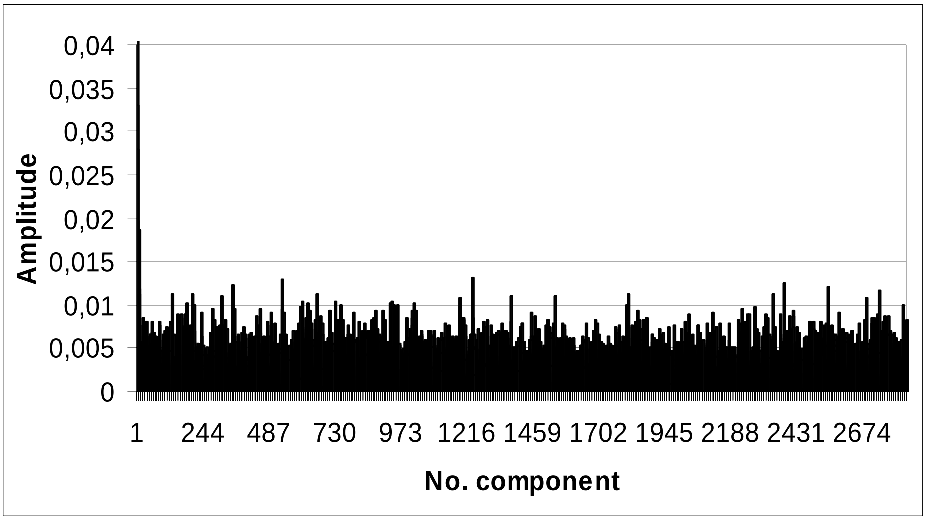

Figure 1. The Fourier spectrum of the radon (Rn-222) concentration in the soil gas during the period 11 July – 7 September 1999 (Katowice).

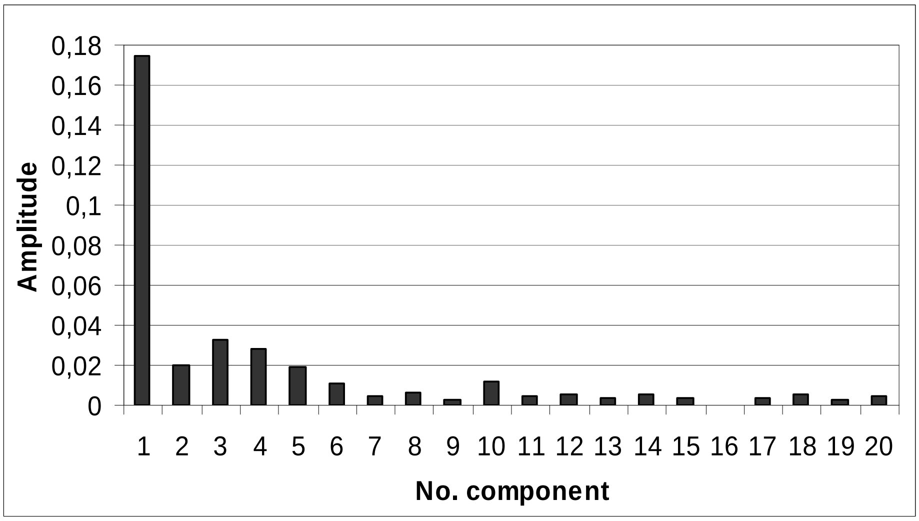

Figure 2. The Fourier spectrum of the radon (Rn-222) concentration in the soil gas taking into account the first 20 components of the FFT analysis, during the period 11 July – 7 September 1999 (Katowice).

Wavelet analysis

The analysis of the amplitude of the wavelet transform gave no reason for separation of any diurnal variation of Rn-222 concentration in the soil gas taking into account diurnal variation of the meteorological conditions. (Fig. 3). So, the short-term changes of the meteorological conditions do not influence the changes of the radon 222 concentrations. On the other hand, the long-term changes of these conditions can affect indirectly the radon concentration in the soil gas.

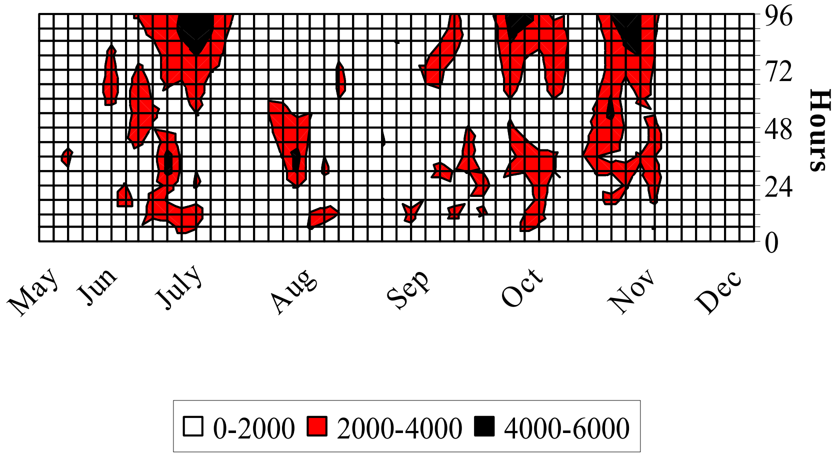

Figure 3. Local amplitude of the Morlet wavelet for the respective one-hour-scales of the radon (Rn-222) concentration in the soil gas in 2000 year (Katowice).

Hurst exponent

According to the Range Rescaled Analysis, the Hurst exponent for the radon concentration in the soil gas ranged from 0.811 up to 0.996 and was greater than 0.9 for the most time. So the changes of Rn-222 concentration in the soil gas are persistent and the phenomena of the radon occurrence in the soil up to 1m have a long-term memory (big positive correlation) or using terms of the chaotic dynamics: there is a subtle sensitivity to the initial conditions (Fig. 4).

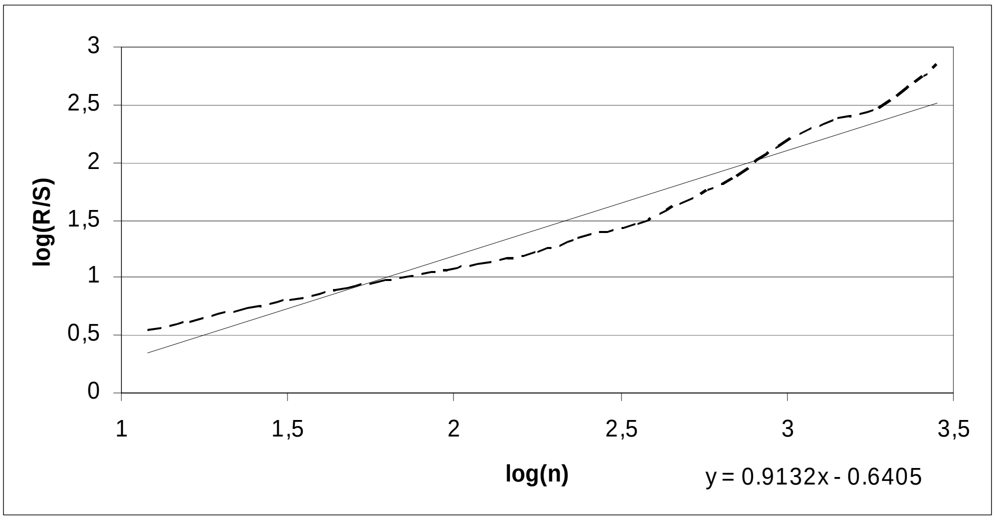

Figure 4. The Hurst exponent (H) for of the radon (Rn-222) concentration in the soil gas during the period 11 July – 7 September 1999 (Katowice).

3.2 Polonium concentration in the atmospheric air

Results of the analysis

The concentration of polonium in atmospheric air shows characteristic diurnal variation with the maximal value in the early morning (3-5 UTC) and minimum value around midday (12-14 UTC) when the shaky thermic dynamical stratification of the atmosphere occurs and as a result the range of the mixing layer is maximal (Fig. 5). The strongest, statistically significant correlation (α=0.05) has been observed between polonium-218 concentration and air temperature (r=-0.759) or wind velocity (r=-0.829). Basing on the comparative analysis for Po-218 concentration and MLH, the following correlation coefficients have been achieved: r = -0.761, -0,575 i -0.580, in Katowice, Cracow and Ostrava respectively, at significance level of α=0.05. The differences were caused by different meteorological conditions that occurred during field experiments. One of the basic parameters influencing directly the Po-218 concentration in air was wind velocity. When the average diurnal air velocity is greater than 4 m/s, the correlation was considerably lower.

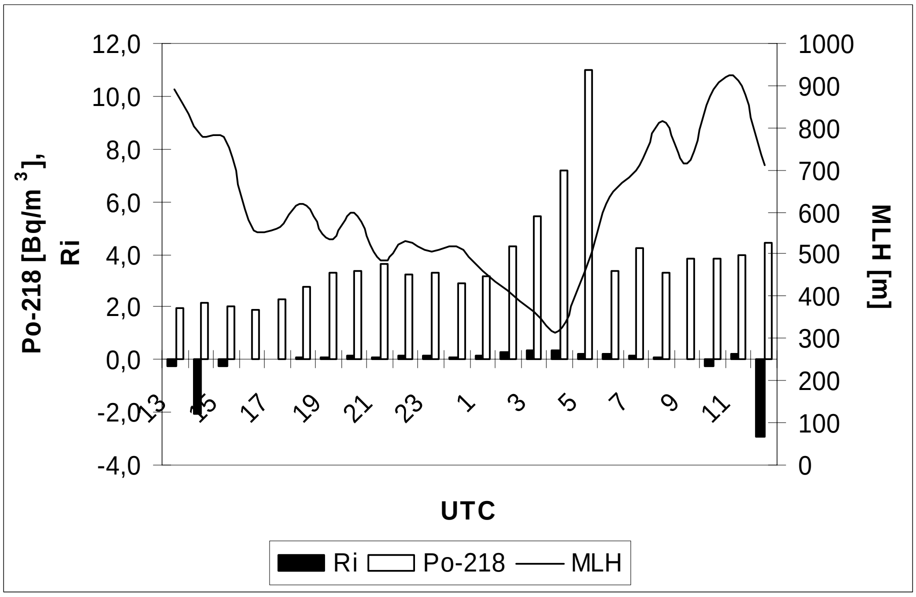

Figure 5. Diurnal changes of the polonium (Po-218) concentration in air and the mixing layer height (MLH) value with the Richardson number (Ri), 6-7 October 2004 year (Ostrava).

4. MODEL FOR ESTIMATION OF THE MLH

The mathematical model was developed taking into account the results of measurements performed in years 2000-2003 in Katowice and Cracow. These experiments made it possible to draw conclusion that between Po-218 concentration in air near to ground (C_{Po}) and equivalent MLH value (h_{eqmix}) occurs a statistically significant relation at significant level of 0.05. This relation can be described by the following mathematical formula:

$$h_{eqmix} = \left(\frac{C_{Po}}{const} \right)^{\beta} \tag{3}$$

where *const* is a local constant that depends on geographical conditions, β=-1.55 - exponent, that does not depend on measurement site. This model was tested during hot season in 2004 in Katowice. The obtained data showed that the model gives satisfactory results when the meteorological conditions are well defined.

5. CONCLUSIONS

1. The frequency and fractal analysis showed that the radon (Rn-222) concentration in the soil gas has a quasi-stable character.
2. The mathematical analysis showed that polonium (Po-218) concentration near to ground has a characteristic diurnal variation and is well correlated with meteorological elements. As a result, this concentration can be a satisfactory indicator of the MLH in the atmosphere.

3. A statistical model for evaluation of the equivalent MLH basing on measurement of the Po-218 concentration in air was developed. It can be presented as a power function. This mathematical model was satisfactory verified for the hot season (months May-October). The mixing conditions seem to be underestimated when the averaged diurnal air velocity is greater than 4 m/s. The examined model can be successfully applied under different geographical conditions.
4. The measuring process should be automatized and present work concentrates on this aim.

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