Aerosol removal coefficients based on ^7Be , ^{210}Pb , and ^{210}Po radionuclides in the urban atmosphere

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Abstract

In this study, the aerosol removal coefficients based on ^7Be , ^{210}Pb and ^{210}Po radionuclides in the urban air, in Lodz, Poland, were investigated over 3 years, between May 2014 and December 2017. Results representing the summer/warm and winter/cold seasons were applied to quantity and quality estimates of aerosol removal processes. The values for the removal processes were closely dependent on the meteorological conditions; therefore, a set of nine meteorological parameters was employed in the analysis. The multiple regression method was applied to explain the relationship between the removal coefficients of aerosols and independent factors identified using Principal Component Analysis.

Keywords Aerosols residence time · Aerosol removal · PCA · Multiple Regression · Radionuclides in the air · Aerosols

1 Introduction

Most natural and artificial radionuclides occurring in the atmosphere are aerosol-borne tracers to study atmospheric transport processes. Radiotracers can be introduced to the atmosphere as emissions from the earth's surface (solid state products of gaseous ^{222}Rn decay, including ^{210}Po and ^{210}Pb); natural radioactivity produced by cosmic radiation; artificial radioactivity introduced by nuclear weapon tests, nuclear incidents or human activity. The presence of ^7Be ($T_{1/2} = 53.3$ d) nuclei in the atmosphere is a result of processes occurring in the upper troposphere and lower stratosphere by spallation reactions, and disintegration of nuclei of nitrogen and oxygen atoms that have been hit by cosmic ray neutrons. ^7Be concentration varies with geomagnetic latitude and atmospheric depth (Chham et al. 2018; Papastefanou and Ioannidou 1995; Ioannidou and Paatero 2014). After nucleus formation, ^7Be nuclei attach to atmospheric aerosol particles (Grundel and Porstendorfer 2004; Rodas Ceballos et al. 2016) and then play a role in the formation and growth of the aerosols.

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^{210}Pb ($T_{1/2} = 22.3\text{y}$) is mainly produced in the atmosphere near ground level by the decay of the noble radioactive gas ^{222}Rn emanating from soil and further decays of short-lived progenies. ^{210}Pb activity concentration at the ground level of the atmosphere varies depending on meteorological conditions, season, efficiency of radon emanation from the ground, height of atmospheric boundary layer and the soil moisture. In the city of Lodz ^{210}Pb activity in fractionated aerosol profiles changes from 0.2 to 1.2 mBq m^{-3} (Długosz-Lisiecka 2019, 2015a, b). ^{210}Po ($T_{1/2} = 138\text{ d}$) is a ^{210}Pb decay product, however, its presence in urban air, especially in winter, is also linked with fossil fuel combustion or other human activities.

The sources of ^7Be , ^{210}Pb , and ^{210}Po radionuclide distributions are relatively well-known and can therefore be considered as radiotracers of all processes occurring in the atmosphere. Radioactive tracers quickly attach to the surface of aerosols and nuclei to become carrier aerosols (Baskaran 2011; Winkler et al. 1998; Rodas Ceballos et al. 2016; Papastefanou and Ioannidou 1996). The movement of aerosols in the urban air depends on the physicochemical processes in the vertical air profile and the motion of air masses. The effectiveness of removing aerosols depends on size distribution and the level under sea and meteorological conditions (Mohan et al. 2018; Budhavant et al. 2017).

The size distribution of the radionuclides in the aerosol particles is the result of a combination of atmospheric processes such as coagulation of ultrafine particles, fog and cloud droplet formation, evaporation and condensation, washout, rainout and sedimentation, and of contributions of, for example, dust storms and combustion products to the tropospheric aerosol mixture. The aerosol size distribution is the result of all interdependent processes present in the atmosphere. The composition of the fractionated dust profile can vary significantly depending on the origins of the particles (Cao et al. 2019; Huang et al. 2019).

Connecting cosmogenic radionuclide ^7Be with other ^{222}Rn radionuclide progeny can assist understanding of the transport phenomena in the atmosphere (Persson and Holm 2014; Długosz-Lisiecka and Bem 2010, 2012). Discrepancies between the aerosol residence times calculated on ^7Be , ^{210}Pb , and ^{210}Po radionuclide concentrations are caused by the following main atmospheric processes: wet and dry deposition, mass exchange between stratosphere and troposphere, vertical transport in the troposphere, and horizontal transport (Daish et al. 2005; Camacho et al. 2009). Dynamic processes in the atmosphere, weather conditions and seasons cause significant changes in the efficiency of aerosol removal and aerosol residence times. Therefore, the dynamics of radionuclide decay has negligible contribution to the removal process of aerosols. The atmospheric residence time is determined by the total removal rate, resulting from all removal processes from the atmospheric boundary layer. The aerosol residence times in the atmosphere vary widely from minutes to days, and even months (van Pul et al. 1998). It depends on the particle size, altitude in the atmosphere and atmospheric processes such as wet and dry deposition (Paatero et al. 2017; Heinrich and Pilon 2013; Talpos 1997; Krajny et al. 2014).

2 Aim of the study

The aim of this study was to identify of a correlation between aerosol removal coefficients λ [day^{-1}] and meteorological parameters like temperature T [$^{\circ}\text{C}$], relative humidity H [%], air pollution P [$\mu\text{g m}^{-3}$], cloud cover C [octa], rainfall R [mm], air pressure PR [hPa], wind velocity V [ms^{-1}], insolation S [W m^{-2}], and rainfall rate D [mm h^{-1}] in the urban air. For interpretation of this correlation, multidimensional exploration techniques were used.

Chemometric tools enable the building of a linear regression model and check the accurate prediction of a dependent variable. Therefore, to predict the value of aerosol residence times based on the value of several variables, multiple regression methods were applied. Aerosol removal coefficients were applied as fingerprints of natural atmospheric processes.

3 Materials and methods

In this study, equipment for fractionated aerosol collection and radiometric analysis were used. Dust samples were collected using a portable high volume sampler, which fractionated particulate matter into separate fractions. The aerosol sampling station was located one meter above the ground. The high volume (TISCH) station had a regulated air flow rate and the efficient cutoff diameters (ECDs) ranged from 0.4 to 7 microns.

Due to the need for quantity analysis of the chosen radionuclides and the relatively low concentrations, various measuring techniques were applied. The method for measuring radionuclide activity consist of two stages. In the first, an instrumental variant, the dust filters (Millipore, nitrocellulose, 0.45 μm) were weighed, pressed into a disc form, and then analyzed using gamma spectrometry (with an HPGe detector). All gamma emitters were analyzed using a low background spectrometric system based on HPGe (with maximum relative 30% efficiency, Canberra) with a 9 inch NaI guard detector (Scionix) for Compton suppression (Długosz-Lisiecka 2016, 2019). Radionuclides ^7Be and ^{210}Pb were analyzed using a modern low-level gamma spectrometric technique. Detection efficiency was estimated using ISOCS/LabSOCS and reference materials based on IAEA soil 327 RMs.

In the framework of this study, the radiochemical method of ^{210}Po separation was used as has been described elsewhere (Długosz-Lisiecka and Bem 2010; Długosz-Lisiecka 2015a, b).

In this study, together with the radionuclide concentrations, several other data (Temperature T, Cloud cover C, Humidity H, Rain R, Wind velocity V) were applied. The source of this data was the Institute of Meteorology and Water Management, National Research Institute, Poland.

The results obtained were statistically analyzed using the Statistica v.10 software package for principal component analysis (PCA) and cluster analysis, and regression methods widely used for source apportionment. Using these methods, the components accounting for the variance of the elements were extracted and subsequently identified as atmosphere dynamic parameters. PCA was applied for variables using varimax orthogonal rotation with Kaiser Normalization and an eigenvalue greater than 1. All methods have been described elsewhere (Długosz-Lisiecka 2016, 2019). Radiometric analysis was conducted in an ISO 17025-certified laboratory system.

Residence time RT was calculated as the difference between the measured AMAD of ^7Be (or ^{210}Pb , or ^{210}Po) aerosols and the diameter of Aitken nuclei (AMDA) equal to 0.015 μm by the MGR(0.0045 $\mu\text{m/day}$) according to equation 1.

$$RT = \frac{AMAD - AMDA}{MRT} \quad [\text{day}] \quad (1)$$

$$\frac{1}{RT} = \lambda_{diff} + \lambda_{sed} + \lambda_{wetdep} \quad [\text{day}^{-1}] \quad (2)$$

The residence time RT of aerosols is correlated with the dynamics of all processes presented in the atmosphere, e.g. dynamic diffusion λ_{diff} , sedimentation λ_{sed} , and wet and dry deposition λ_{wetdep} (2) (Koch 1996). In this study $\frac{1}{RT} = \lambda$ has been defined as the removal coefficient.

4 Results and discussion

^7Be , ^{210}Pb , and ^{210}Po radionuclides are suitable for the study of atmospheric transport processes (Baskaran 2011; Hirose et al. 1993; Grabowska et al. 2003). All three radionuclides have different origins, but after adsorption to the surface of aerosols, their behavior is strongly linked with particles suspended in the atmosphere.

The most important characteristic of aerosols affecting their transport and residence time is their size. The activity median aerodynamic diameter (AMAD) is the diameter above and below which 50% of radionuclide activity takes place.

Aerosol residence time for all three radionuclides was calculated based on AMAD using the method described by Papastefanou and Ioannidou (1995) (1).

McMurry and Wilson (1984) defined the Mean Growth Rate (MGR from 0.004 to 0.005 $\mu\text{m}/\text{day}$) parameter for describing aerosol particle increase according to the theory of secondary ambient aerosol growth by condensation and coagulation.

Fig. 1 Radionuclide residence times RT results from May 2014 to December 2017

Aerosol residence times result from stratosphere–troposphere transport, leaching dynamics, or the spread of radionuclides of various origins and therefore strongly depend on the aerodynamic diameter of particles, and play the role of a fingerprint in this process.

The average aerosol residence time based on the ^7Be , ^{210}Pb , and ^{210}Po radionuclides were equal to 6.06, 6.57, and 6.36 days (see Fig. 1), respectively, for whole fractionated dust profiles. The average value from all three radionuclides was equal to 6.33 days. Similar values were reported in other locations e.g. 7–9 days for ^7Be activity size distribution in ground-level air in the Thessaloniki Region (Greece) at sea level. Aerosol residence times reported by (Ioannidou and Paatero 2014) calculated based on the $^{10}\text{Bi}/^{210}\text{Pb}$ activity ratio was 8 days. From $^7\text{Be}/^{210}\text{Pb}$ activity ratios, Koch (1996) calculated mean tropospheric residence times of 5–6 days.

Aerosol residence time based on the $^{210}\text{Po}/^{210}\text{Pb}$ activity ratio showed overestimated values (Długosz-Lisiecka 2016; Semertzidou et al. 2016), higher than the results of the present work. A previous study confirmed that artificial injection of ^{210}Po activity into the atmosphere dramatically increased calculated aerosol residence times. In this work, residence times calculated using AMADs applied to the ^{210}Po radionuclide showed similar values to those for ^{210}Pb and ^7Be (Table 1.).

PCA was used for statistical identification patterns in residence time results based on ^7Be , ^{210}Pb , and ^{210}Po radionuclides, and meteorological parameters corresponded to their differences and similarities (Table 2). PCA was used as a tool for data analysis on a total of 32 aerosol size distribution profiles collected between May 2014 and December 2017. An analysis of 12 species(variables) was conducted according to the Kaiser criterion, therefore only eigenvalues greater than 1 for the extracted factors were included (Table 2). Variables with factor loadings >0.5 were employed to extract the eigenvalues and eigenvectors from the correlation matrix. For winter and summer, five significant factors were calculated which accounted for 88.1% and 85.5% of the total variance.

In winter, five principal components (PC1, PC2, PC3, PC4, PC5) accounted for, respectively, 24.7%, 17.3%, 15.2%, 19.7%, and 11.2% of the variance. The values listed in Table 2 for PC1 illustrate a positive correlation with cloud cover C(0.80), relative humidity H(0.93) and a negative correlation with the insolation parameter S(-0.92). The second component

Table 1 Average ^{210}Pb , ^{210}Po , and ^7Be radionuclide residence times values [day] and mean air pollution P [$\mu\text{g}/\text{m}^3$] and other meteorological parameters in summer and winter

	summer	winter
Residence time [days] RT_{Be} (AMAD(Be) $\pm$ SD)	5.60 $\pm$ 0.67 (0.612 $\pm$ 0.070)	6.66 $\pm$ 0.85 (0.734 $\pm$ 0.088)
Residence time [days] RT_{Pb} (AMAD(Pb) $\pm$ SD)	6.20 $\pm$ 0.70 (0.687 $\pm$ 0.079)	7.06 $\pm$ 0.99 (0.768 $\pm$ 0.110)
Residence time [days] RT_{Po} (AMAD(Po) $\pm$ SD)	6.58 $\pm$ 0.60 (0.721 $\pm$ 0.066)	6.09 $\pm$ 0.91 (0.680 $\pm$ 0.099)
precipitation [mm] R	49.67 $\pm$ 36.74	44.46 $\pm$ 23.67
Pressure [hPa] PR	994.3 $\pm$ 1.77	995.4 $\pm$ 4.8
Cloud cover [octa] C	4.92 $\pm$ 0.68	6.18 $\pm$ 0.71
Wind velocity [ms^{-1}] V	3.03 $\pm$ 0.38	3.78 $\pm$ 0.54
Temperature [$^{\circ}\text{C}$] T	16.07 $\pm$ 3.05	3.14 $\pm$ 3.84
Relative humidity [%] H	71.11 $\pm$ 5.07	83.6 $\pm$ 5.33
Air pollution [$\mu\text{g}/\text{m}^3$] P	49.7 $\pm$ 16.3	72.2 $\pm$ 74.0
Rainfall rate [mmh^{-1}] D	1.29 $\pm$ 0.68	0.371 $\pm$ 0.216
Insolation [Wm^{-2}] S	224.7 $\pm$ 46.9	74.25 $\pm$ 40.9

Table 2 Principal component Analysis PCA results, variable and its loading factors

	winter	Principal Components : variable (loading factors)	
	0.247	PC1	C(0.80); H(0.93); S(-0.92)
	0.173	PC2	PR(0.84);
	0.152	PC3	T(0.81); D(0.91)
	0.197	PC4	R(0.77); P(0.94);
	0.112	PC5	V(0.87)
	0.881	Sum	
	summer		
	0.255	PC1	R(-0.76); C(-0.74); H(-0.90); S(0.88)
	0.175	PC2	D(0.89)
	0.144	PC3	PR(-0.92); V(0.72)
	0.143	PC4	P(0.87); T(-0.55)
	0.717	Sum	

(PC2) accounted for 17.3% of the variance and was determined by atmospheric pressure – PR(0.84). PC3 was correlated with air temperature T(0.81) and the rainfall rate D(0.91). Total precipitation R(0.77) and air pollution P(0.94) were attributed to PC4. The final principal component (PC5) was directly linked with wind velocity V(0.87).

In summer, the principal components (PC1,PC2,PC3,PC4) changed their contribution and, respectively, accounted for 25.5%, 17.5%, 14.4% and 14.3% of the variance. In summer, the variables were linked with the principal components in a different configuration (Table 2).

There was significant correlation between variables due to principal component configurations. Similar significant ($p < 0.05$) correlations between variables exhibited Spearman correlation coefficients, which enabled the prediction of groups of linked parameters, e.g. in summer, PC1 can be defined as a multiplication of the four variables: precipitation - R, cloud cover-C, relative humidity - H and insolation - S. All these variables are highly correlated (positively or negatively). In this case, PC1 can be defined as wet deposition.

In the results from nine variables (C,H,S,PR, T,D,R,P,V) in five different parameters (PC1,PC2, PC3, PC4, PC5) based on PCA, it was possible to apply multiple regression.

This statistical method was useful for predicting the value of a variable based on new independent variables (PC1,PC2,PC3,PC4 and PC5), where each represented the impact of a different physical parameter or a set of parameters.

In this approach, $\frac{1}{RT_{Be}}; \frac{1}{RT_{Pb}}; \frac{1}{RT_{Po}}$, corresponding to λ_{Be} , λ_{Pb} , λ_{Po} , aerosol removal coefficients were investigated based on the ^7Be , ^{210}Pb and ^{210}Po radionuclides.

According to the above equation (2), removal coefficients were a simple sum of sedimentation, wet and dry deposition, and dependent on the meteorological conditions. Multiple regression enabled expression of a linear function of different parameters X_i , where “i” represented the number of significant ($p < 0.05$) parameters, described as PC coefficients (3).

$$\lambda_{Be} = \sum X(i) * PC(i)_{w,s} \quad (3)$$

All three radionuclides (^7Be , ^{210}Pb , ^{210}Po) had different origins but residence times results calculated on the base of activity size distribution looked similar. This suggested similar processes influenced the aerosol residence times and affected their removal.

Removal coefficients based on the various radionuclides fluctuated corresponding to dynamic processes occurring in the atmosphere. The nature of the variables that made up significant PC coefficients exhibited different factors that linked the removal processes for all three radionuclides. In winter, multiple regression equations described aerosol deposition processes.

$$\lambda_{w,Be}(R = 0.76) = 0.987*PC1_w(p = 0.008) - 0.759*PC2_w(p = 0.011) \quad (4)$$

$$\lambda_{w,Pb}(R = 0.89) = 0.562*PC1_w(p = 0.047) - 0.614*PC2_w(p = 0.009) \quad (5)$$

$$\lambda_{w,Po}(R = 0.81) = 0.626*PC2_w(p = 0.012) - 0.531*PC4_w(p = 0.032) + 0.558*PC5_w(p = 0.030) \quad (6)$$

In the case of λ_{Be} and λ_{Pb} removal coefficients, meteorological conditions C,H,S, and PR transformed to $PC1_w$ and $PC2_w$ parameters and resulted in 76% and 89% variation. The remaining 24% and 11% of aerosol particles were removed by radioactive decay of 7Be and ^{210}Pb . The removal coefficient based on ^{210}Po activity size distribution can be calculated based on the $PC2_w$, $PC4_w$, and $PC5_w$ parameters, which accounted for 81% of the total variance. The remaining 19% of aerosol removal was linked with ^{210}Po radioactive decay.

In summer, multiple regression equations were applied to describe aerosol removal:

$$\lambda_{s,Be}(R = 0.89) = -0.451*PC1_s(p = 0.018) + 0.681*PC4_s(p = 0.002) \quad (7)$$

$$\lambda_{s,Pb}(R = 0.64) = 0.478*PC3_s(p = 0.048) + 0.525*PC4_s(p = 0.039) \quad (8)$$

$$\lambda_{s,Po}(R = 0.61) = -0.538*PC3_s(p = 0.022) + 0.1935(p = 0.000001) \quad (9)$$

where:

in winter:
 $PC1_w = C*H*S$
 $PC2_w = PR$
 $PC3_w = T*D$
 $PC4_w = R*P$
 $PC5_w = V$

in summer:
 $PC1_s = R*C*H*S$
 $PC2_s = D$
 $PC3_s = PR*V$
 $PC4_s = P*T$

In summer, the removal coefficient $\lambda_{s,Be}$ based on 7Be activity size distribution can be described by $PC1_s$ and $PC4_s$, which accounted for 89% of its variation (7). The remaining 11% of the removal variation was 7Be radioactive decay. $\lambda_{s,Pb}$ removal coefficients can be described by $PC3_s$ and $PC4_s$ in 64% of the variation contribution. For $\lambda_{s,Po}$ removal coefficient estimation, $PC3_s$ and a free parameter equal to 0.1935 can be applied, which accounted for 61% of total variation. In summer, for $\lambda_{s,Pb}$, $\lambda_{s,Po}$ removal coefficients described by a significant variable, pressure, wind velocity, air pollution, and temperature are strongly connected with horizontal and vertical transport processes (effectively, air mixing processes). The 36% and 39% of aerosol removal not explained by meteorological parameters were probably linked to radioactive decay, long-range transport of fine particles, and washout from urban regions.

The quality of the surface layer of the atmosphere has a significant impact on the biosphere and human health. Most of the atmosphere's mass is accumulated in the layers closest to Earth. About 75% of the atmosphere's mass is concentrated in a 10 km layer

adjacent to Earth's surface, therefore dynamic processes in this region have significant impact on the removal of suspended particles from the atmosphere. The atmospheric residence time of aerosols is highly reduced because of the efficient removal of particles by several atmospheric processes, highly correlated with meteorological conditions (van Pul et al. 1998). ^7Be , ^{210}Pb , and ^{210}Po activity size distribution fluctuates due to radionuclide origin and local dynamic processes. For aerosol residence time the majority of the aerosols can be transported on a local to regional scale. Assuming average wind speed equal $\bar{V} = 3.5 \text{ ms}^{-1}$ aerosols, each day, can reach the distance even 300 kilometers.

In order to investigate aerosol removal coefficients calculated on the basis of ^7Be , ^{210}Pb , and ^{210}Po activity size distribution in the ground air layer, PCA analysis and multiple regression methods were applied.

Aerosol removal coefficients defined on the basis of different radionuclide systems obtain various results. Koch (1996) estimates ^{210}Pb and ^7Be is removed almost entirely by deposition: in 81% and 63% by convective precipitation, 7% and 5% by large scale precipitation and in 12% and 3% in dry deposition, respectively. Balkanski et al. (1993) on the basis of ^{210}Pb suppose that wet deposition accounts for 86% of the global deposition flux, the remaining 14% being due to dry deposition. Sykora et al. (2017) on the basis of ^{222}Rn and ^{210}Pb calculate the removal constant equal $2.6 \cdot 10^{-6} \text{ sec}^{-1}$. In this study for summer season $\lambda_{s,Be}$, $\lambda_{s,Pb}$, $\lambda_{s,Po}$ average removal coefficients are equal $2.1 \pm 0.3 \cdot 10^{-6} \text{ sec}^{-1}$, $1.9 \pm 0.2 \cdot 10^{-6} \text{ sec}^{-1}$, and $1.8 \pm 0.2 \cdot 10^{-6} \text{ sec}^{-1}$ respectively for ^7Be , ^{210}Pb and ^{210}Po . In winter season all three coefficients significantly change and are equal: $1.7 \pm 0.2 \cdot 10^{-6} \text{ sec}^{-1}$, $1.7 \pm 0.3 \cdot 10^{-6} \text{ sec}^{-1}$, and $1.9 \pm 0.3 \cdot 10^{-6} \text{ sec}^{-1}$, respectively.

5 Conclusions

The residence time of atmospheric aerosol particles can be estimated by means of radioactive nuclides as tracers, which become attached to aerosol particles and are removed with them as they are scavenged by precipitation or undergo dry fallout. Negatively correlated temperature (T) and relative humidity (H) in the wet season caused increased stability in the air, which led to higher values of residence times in the atmosphere, calculated on the basis of natural radionuclides ^7Be and ^{210}Pb . The lower aerosol residence times in winter/autumn seemed to be due to the removal and may be attributed to the condensation processes as well as coagulation between the attached aerosol particles. This caused a continuous increase of aerosol particle size and therefore the deposition probability increased. This process was observed for ^{210}Po radionuclides deposited on a large particle diameter.

In summer/dry season, high temperatures and low humidity increased air mixing processes and contributed to significantly lower aerosol residence times and more effective horizontal aerosol transport.

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