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The deficit of ^{210}Po activity confirms an efficient escape of polonium from dedusting installations. Eleven gigabecquerels of ^{210}Po per year are released into the urban air. However, industrial polonium emission cannot pose a radiological hazard as it is only slightly higher than the natural dose. On the other hand, in some agglomerations like Belchatow city in central Poland or Silesia region, inhabitants are likely to be more vulnerable to elevated ^{210}Po concentrations in the urban air. Therefore the activity concentration of ^{210}Po should be more carefully controlled.

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ARTICLE TYPE

Excess of Polonium-210 activity in the surface urban atmosphere. Part 1. Fluctuation of the ^{210}Po excess in the air

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The concentration of ^{210}Pb, ^{210}Bi, and ^{210}Po in the urban atmosphere of Lodz city has been measured from February 2010 to May 2010 and from May 2011 to April 2012. The seasonal change in the activity ratio for ^{210}Po/^{210}Pb and ^{210}Bi/^{210}Pb indicates that the observed fluctuations are independent of the concentration of tropospheric ^{210}Pb and its decay products, particularly ^{210}Po. A simple calculation method has been proposed for the estimation of the excess of ^{210}Po in the urban aerosols in relation to the fraction of its activity formed from ^{210}Pb. On the basis of results obtained, one can conclude that a substantial part of the ^{210}Po in urban air is coming not from the decay of atmospheric ^{222}Rn but from input from artificial sources. The highest levels of measured total ^{210}Po activity were observed during the winter period. This observation suggests that the main source of ^{210}Po in the investigated region could be related to anthropogenic emission from domestic heating systems and local coal power plants, rather than other sources such as soil resuspension or stratospheric air intrusion as usually suggested in the literature.

Introduction:

^{210}Pb (half-life = 22.3 years), ^{210}Bi (half-life = 5 days), and ^{210}Po (half-life = 138 days) isotopes (Equation (1)) and their mutual relations have been widely used as chronometers and markers of a variety of natural processes occurring in the atmosphere, for example, air transport of aerosols or removal of suspended matter (Baskaran and Shaw, 2001).

$$^{210}_{82}Pb \xrightarrow{\beta^-} {}^{210}_{83}Bi \xrightarrow{\beta^-} {}^{210}_{84}Po \xrightarrow{\alpha} {}^{206}_{82}Pb \text{ (stable)} \qquad (1)$$

Most of the above mentioned isotopes are highly particle-reactive and become attached as high mobility clusters to aerosols soon after their formation. This generally takes from 40 s to 3 min. Finally they are scavenged from the atmosphere by precipitation. The removal of radionuclides from the atmosphere occurs by dry or wet deposition with raindrops[1].

Nucleation of gas molecules produces ultrafine aerosols in the range of 1 to 10 nm. As a result of condensation of gases and further coagulation caused by collisions of the particles, these ultrafine aerosols grow rapidly to a size of 0.01–1 µm, which is classified as fine aerosol. Further growth of particles above 1 µm size is however much slower. Aerosol particles formed by condensation of the gas molecules finally accumulate in the 0.01–1 mm size range, the so-called accumulation mode. These small particles are removed from the atmosphere mainly by scavenging by cloud droplets and subsequent washout.

^{210}Pb radionuclide is bound mainly to aerosol particles with submicron diameters. The adsorption of the radionuclide ^{210}Pb on the surface of solid aerosol particles followed by production of its two daughters is the basis for the aerosol resident time estimations. Disequilibrium of the activity of ^{210}Pb and its daughters ^{210}Bi and ^{210}Po in fresh air particles can be used to calculate the aerosol residence time, based on the ^{210}Bi/^{210}Pb and ^{210}Po/^{210}Pb nuclide activity ratios. Both methods are widely used for such calculations[1,2,3,4,5,6].Unfortunately, the two methods give different values of the residence times for the same investigated sample

Usually the one-box compartment model has been applied for the description of the activity ratios of ^{210}Pb/^{210}Bi and ^{210}Pb/^{210}Po in aerosol samples. The measured ratios are then used to estimate the mean residence time of solid particles in the air. Calculation of the residence times for each pair of radioisotopes has often led to inconsistent values. Therefore, Poet[7] suggested that part of the ^{210}Po activity, ΔA_{Po}, in the aerosol samples comes from additional sources such as resuspension of soils or volcanic eruption of ^{210}Po. This radionuclide is usually in radioactive equilibrium with its parent ^{210}Pb.

Poet[7] estimated an aerosol residence time ranging from 11 to 77 days. The lowest mean aerosol residence time of 0–38.9 days was found for samples collected in Alaska[8]. The longest mean residence time of 320 days was estimated from ^{210}Po/^{210}Pb activity ratio data in Arkansas[9]. The same authors pointed out that residence time values, T_R, based on the ^{210}Po/^{210}Pb activity ratios are longer than those calculated based on the ^{210}Bi/^{210}Pb activity ratios. They concluded that the observed difference could be a result of the presence in the aerosol mixture of particles other than these three radionuclides remaining in secular radioactive equilibrium. This means that the ^{222}Rn decay products present in the troposphere might arise from additional sources other than natural emanation of ^{222}Rn from the surface soil.

The most probable additional sources of ^{210}Po suggested in the literature are resuspension of the soil[7] , fly ashes emitted from energetic coal[10], and biomass combustion[11]. Other likely sources that can be mentioned are stratospheric input of older air masses (stratospheric air intrusion), sea sprays, volcanic[12] and industrial emissions[13] and finally car exhaust fumes[14]. In most of them, ^{210}Po and ^{210}Bi are in radioactive equilibrium with their parent ^{210}Pb.

Poet[7] estimated that up to 85% of ^{210}Po in the air has a terrestrial origin. Lambert[12] linked high concentrations of ^{210}Po in relation to ^{210}Pb with erupted volcanic gases, coal burning, and forest fires. Moore[15] indicated that anthropogenic sources of ^{210}Po from phosphate fertilizer dispersion and fossil fuel burning could constitute up to 7% of the total ^{210}Po influx to the atmosphere. Carvalho[13] suggested that increased ^{210}Po/^{210}Pb ratios in the precipitates in Lisbon (Portugal) were caused by the industrial emission of ^{210}Po. Wallner[16] measured an unexpectedly high

$^{210}Po/^{210}Pb$ airborne activity ratio equal to 7.5 in the Alps mountain region. The highest $^{210}Po/^{210}Pb$ activity ratio, which was equal to 33.7, was observed at the site of a Syrian mine[17]. A similar disequilibrium phenomenon ($^{210}Po/^{210}Pb$ higher than 9) in particulate matter of marine origin was reported by Radakovitch[18]. In urban air, $^{210}Po/^{210}Pb$ does not exceeds 0.1, while in fresh aerosols the typical value[5] is only 0.02.

The average annual ^{210}Pb activity concentrations in the urban air in Lodz city are about 0.5 mBq/m^3, whereas corresponding activities of ^{210}Po are ten times lower[5]. Therefore, an additional input of ^{210}Pb and ^{210}Po radionuclides in radioactive secular equilibrium makes an important contribution to the total activity of ^{210}Po only.

Combustion of fossil fuels with a predominant contribution of coal in urban areas of central Poland leads to the dispersion of natural radionuclides into the environment[19,20]. The mineral components, including radioactive elements, are present in all kinds of crude coals although at low concentration. After combustion, the concentrations of ^{210}Pb, ^{210}Bi, and ^{210}Po radionuclides in fly ash are much higher (up to 30 times) than in crude coals[21]. The activity concentrations of ^{210}Pb and ^{210}Po radionuclides in freshly formed fly ash particles are strongly dependent on their diameter. For particles with an aerodynamic diameter of less than 1.1 µm, the enrichment factor for these radionuclides may be as high as 26 and 31 for ^{210}Pb and ^{210}Po respectively[22]. The enrichment factor values for solid particles are lower for lignite or domestic coal combustion.

The high enrichment factor of the element Po is a result of its efficient volatility. The volatility of ^{210}Po was recognized previously as a reason for its loss during the sample preparation process prior to its concentration analysis in different types of samples. Measured activity loss[23] begins at temperatures above 100 ^{o}C, with 90% loss at 300 ^{o}C.

Combustion of fossil fuels at high temperature may also lead to the high activity concentration of polonium emission. In the open air, pure elemental Po sublimes at 700 ^{o}C, but usually before reaching this temperature it is mainly oxidized to a PoO_2 species. For the coal combustion temperature of ~1200 ^{o}K, the corresponding vapour pressures of PoO_2 and elemental Bi and Pb are equal to 4.76, 0.3, and 0.1 mm Hg, respectively[24] . At temperatures above 1600 ^{o}C, complete evaporation of polonium can be expected[21].

High-temperature processes like coal combustion and other anthropogenic activities of the foundry or phosphate industry can be considered as possible additional sources of the excess of ^{210}Po unsupported by ^{210}Pb in the urban air[21,25].

For the reason presented above, in the vinicity of coal combustion facilities (domestic or central heating and also electric power production stations) only the $^{210}Bi/^{210}Pb$ method should be used for determination of the aerosol residence time.

The main aim of the present work was to verify the relation between seasonal fluctuations of the total ^{210}Po activity, A_{Po}, and corresponding values of the excess of this radionuclide concentration, ΔA_{Po}'. The latter is based on real aerosol residence time determinations in the urban area.

Materials and methods

Sampling and pre-treatment:

Total Suspended Particle (TSP) samples were collected by the ASS-500 station situated in the centre of Lodz city, Poland. This aerosol-collecting unit operates as part of the national air monitoring system. In a typical experiment, approximately 50 000 m^3 of air was filtered. During the seven days of collection, 2.5 to 4 g of the dust was collected by a 40 cm × 40 cm square filter. Immediately after the collection of aerosols, the filters were divided into two equal parts.

Radionuclides determinations:

The first piece of filter was used for fast ^{210}Bi determination. ^{210}Bi was washing out from filter by 100 mL of 2 M HCl solution. The resulting filtrate solution has been diluted to 1 M HCl and carried on DOWEX 1x8 resin. For ^{210}Bi elution 100 mL 1.8 M H_2SO_4 was passed through the chromatography column and then was extracted by 5% trioctylphiosphine oxide (TOPO) in toluene. Radiochemical separation and activity measurement of ^{210}Bi have been described elsewhere[5].

From the second part of the filter, a circle 3 cm in diameter was cut out and placed in a beaker filled with 50 ml of 1M HCl. After 24 hours of leaching, the solution was filtered, and prior to the spectrometry measurement, ^{210}Po present in the solution was separated by spontaneous deposition on silver discs. The activity of ^{210}Po was determined using an alpha spectrometry system. Details of the method used were described by us elsewhere[26,27].

The remaining second part of the filter was pressed into the form of a disc 5.2 cm in diameter and 0.4 cm thick, which represents standard geometry for all instrumental γ-spectrometry determinations[25] of ^{210}Pb. The detection limits for these radionuclides calculated according to Currie's formula[28] were 5.2 µBq/m^3 for ^{210}Bi, 5 µBq/m^3 for ^{210}Pb, and 1.19 µBq/m^3 for ^{210}Po.

The quality assurance of the used analytical procedures and instrumental γ-spectrometry measurements was checked using various type standard reference material: IAEA 327 (soil), IAEA 300 (sediment). Good compliance of the determined activity value of ^{210}Po with a certified reference sample was achieved, as previously reported[26].

In the additional set of experiments, the fractionated samples of aerosols with diameters in the range from 0.1 to 10 µm were collected by an eight-stage cascade Andersen Impactor. Radiochemical separation and activity measurements of the samples collected were carried out as described previously[6].

Both methods based on the $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ activity ratios should give the same real aerosol residence time, T_{RK}, calculated from Equation (2). If this assumption is correct, possible influx of the additional ΔA_{Po} activity can be estimated using calculated T_{RK} values in Equation (3)[4,7,26]:

$$T_{RK} = \frac{(A_{Bi} - A_{Po})}{\lambda_{Bi}(A_{Pb} - A_{Bi}) - \lambda_{Po}(A_{Bi} - A_{Po})} \tag{2}$$

$$\Delta A_{Po} = A_{Bi} - \frac{\lambda_{Bi}(A_{Bi} - A_{Po})}{\lambda_{Bi}(A_{Pb} - A_{Bi}) - \lambda_{Po}(A_{Bi} - A_{Po})}(A_{Pb} - A_{Bi}) \tag{3}$$

where:

$A_{Pb,\ Bi,\ Po}$ – measured activities of ^{210}Pb, ^{210}Bi, and ^{210}Po radionuclides in the surface urban air,

$\lambda_{Po,\ Bi}$ – the ^{210}Po and ^{210}Bi- decay constants, respectively.

ΔA_{Po} – the excess of the unsupported ^{210}Po activity.

Additional input of unsupported ^{210}Po is considered, the following equations can be written (Equation (4)):

$$A_{Pb} = A_{Bi}(1 + \frac{1}{\lambda_{Bi}T_R}) \tag{4}$$

$$A_{Bi} = (A_{Po} - \Delta A_{Po}')(1 + \frac{1}{\lambda_{Bi} T_R}) \qquad (5)$$

where:

ΔA_{Po}’ is an excess of ^{210}Po activity that does not originate from ^{210}Bi radionuclide previously adsorbed on the aerosol particles. A simple transformation of Equation (5) leads to Formula (6):

$$\Delta A_{Po}' = \frac{A_{Po} - [A_{Bi} - A_{Po}] T_R \lambda_{Po}}{1 + T_R \lambda_{Po}} \qquad (6)$$

After calculating the T_R values from Equation (2) or (4) and inserting these data in Equation (5), one can simply obtain from Equation (6) the values for ΔA_{Po}’, the unsupported excess activity of ^{210}Po radionuclide in air particulate matter.

On the basis of real residence times, the corresponding theoretical values of A_{tPo} for ^{210}Po activity resulting from the decay of ^{210}Pb adsorbed on the aerosol particles at a given concentration A_{Pb} were calculated according to Equation (7)

$$A_{tPo} = A_{Pb}(1 - \exp(-\lambda_{Bi} t) + \frac{\lambda_{Bi}}{(\lambda_{Bi} - \lambda_{Po})})[\exp(-\lambda_{Bi} t) - \exp(-\lambda_{Po} t)] \qquad (7)$$

The effective doses D related to the excess of ^{210}Po due to aerosol inhalation for a child up to one year old and for adult inhabitants of Lodz city were estimated on the base of Equation (8).

$$D = DCF * A_{Po}' * BR * t \qquad (8)$$

where:

DCF – dose conversion factor for medium breathing rate [Sv/Bq]
BR – breathing rate assumed as 1 m^3/h
A_{Po}’ – excess of ^{210}Po in the air [μBq/m^3]
t – time of radiological hazard [h]

Results:

Determining the excess of Po-210 activity

In this study ^{210}Pb, ^{210}Bi and ^{210}Po activity concentrations in fresh air-filters were analyzed for excess of ^{210}Po activity estimation.

The final yield of bismuth separation by method was calculated from the ^{210}Bi activity determination in three old air filters with known activities of ^{210}Pb. The results are presented in Table 1. Filters older than two years should have ^{210}Pb, ^{210}Bi, and ^{210}Po activity equilibrium.

Table 1. The yield of ^{210}Bi separation and activity determination

Code of the filters	^{210}Pb activity, A_{Po} [μBq/m^3]	^{210}Bi activity, A_{Bi} [μBq/m^3]	Chemical yield %
851	479.3	471.4	98.3
838	367.4	306.5	83.4
920	187.7	138.1	73.6

The average total detection and ^{210}Bi separation efficiency value of 85% has been taken for ^{210}Bi concentration calculation by this method. For all collected aerosol samples, the real residence times were calculated according to Equation (2). The excess of ^{210}Po activity has been calculate on the base of Equation 3.

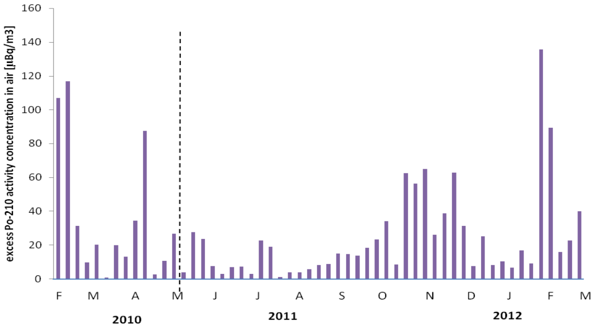

Fig. 1. Excess ^{210}Po (ΔA_{Po}’) activity concentration in the air (the vertical line represents a pause in the sampling period).

The fluctuation of the calculated values of the ΔA_{Po}’ activities in the examined periods is shown in Fig. 1. Weekly data, after averaging for each season of the year, are also listed in Table 2.

The activity fluctuation during the period of the whole year, ranging from 3548.0 to 133.5 µBq/m^3, from 1025.8 to 54.6 µBq/m^3, and from 136.9 to 9.44 µBq/m^3 for ^{210}Pb, ^{210}Bi, and ^{210}Po, respectively. The results of activity measurements were grouped for each of four seasons: spring (March–May), summer (June–August), autumn (September–November), and winter (December–February).

The comparison of the experimentally determined values of ^{210}Po activities, A_{Po}, with those calculated from Equation (7) is shown in Fig 2. One can see that in all cases the experimentally determined concentrations of ^{210}Po considerably exceed these coming from ^{210}Pb decay products during the residence time of aerosol particles.

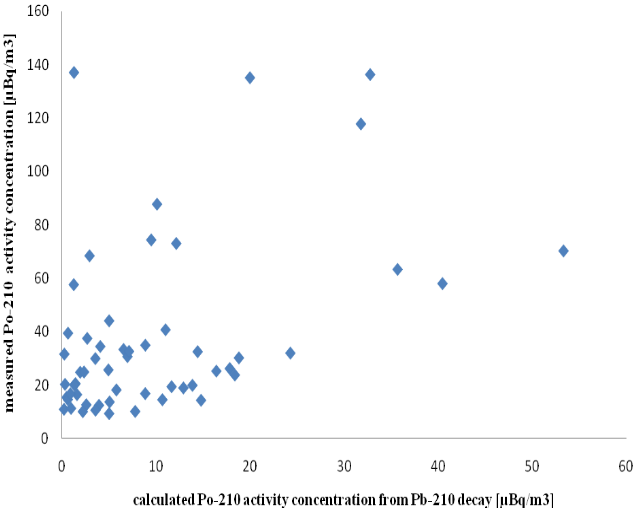

Fig. 2. ^{210}Po measured activity concentration, A_{Po}, and ^{210}Po activity concentration in-growths from ^{210}Pb decay (A_{tPo})

The measured ^{210}Po/^{210}Pb activity ratio ranged from 0.020 to 0.156 (averaging 0.069). It is interesting that the ratio is much lower in the summer period than in the winter period. The average value of the activity ratio as well as the calculated values of ΔA_{Po}’ in the air and in the suspended dust (TSP) for each season is shown in Table 2.

Table. 2 Seasonal change of excess Po-210 activity and residence time based on Af_{Po}/A_{Pb} activity ratio.

Season	Measured A_{Po}/A_{Pb} activity ratios	Calculated A_{tPo}/A_{Pb} activity ratios	$\Delta A_{Po}{}'$ µBq/m^3	$\Delta A_{Po}{}'$ Bq/g	T_R [days]
Spring	0.072	0.023	22.84 ± 12.04	0.406	8.27
Summer	0.055	0.028	8.80 ± 4.76	0.257	10.75
Autumn	0.055	0.021	35.53 ± 16.71	0.545	8.21
Winter	0.092	0.026	42.46 ± 36.13	0.608	10.17

The average values of ^{210}Po excess in air show a clear seasonal trend but low correlation with ^{210}Pb activity levels. The estimated seasonal change in ΔA_{Po}' is statistically significant. This may suggest that the observed excess of ^{210}Po in the winter period is coming from a source characteristic for this season.

The activity concentration of ^{210}Po in becquerels per gram of the total suspended matter varies from 0.608 Bq/g in the winter to a value of 0.257 Bq/g in the summer. These values considerably exceed the measured activity concentration of this radionuclide in the resuspended soil particulates or in the captured fly ashes (0.03 and 0.026 Bq/g, respectively). The dependence of the activity concentrations of ^{210}Po and ^{210}Pb in the urban dust and ^{210}Po in the fly ashes on the particulate aerodynamic diameters is presented in Fig. 3

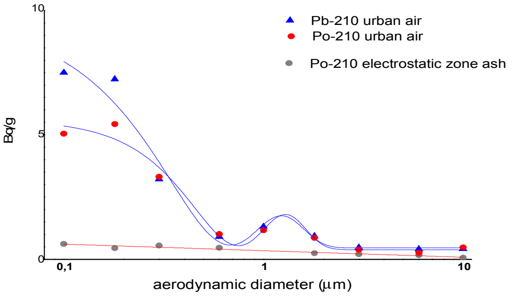

Fig. 3. ^{210}Po activity concentration [Bq/g] in fractionated urban aerosols and fractionated fly ash cached in electrostatic precipitation system in coal power plant.

The fraction of ^{210}Po with the highest activity is adsorbed on aerosol particles below 1 µm in size. The fate of the airborne radionuclides is determined by the behaviour of the aerosol particles in the atmosphere. The very low activities of the fly ashes indicate that even escaping fly ashes with diameters below 0.1 µm did not have sufficiently high activities < 1 Bq/g to be considered as a polonium source. This means that the high activity concentration of ^{210}Po, reaching average values of up to of 4.71 Bq/g for aerosols in the size range of 0.1–0.3 µm, are the result of high particle-reactivity.

Identification of source Po-210 excess

In order to clarify the nature of the additional activity of ^{210}Po, its concentration in dust samples of various origin has been analysed.

Table 3. ^{210}Po activity concentrations in the environmental samples.

Sample origin	^{210}Po Activity concentration [Bq/g]
Fractioned dust with diameter < 0.3 µm	4.71 ± 1.6
Fly ash from electrostatic precipitation of coal power station	0.264 ± 0.016
Dust from foundry processes	0.522 ± 0.023
Surface soil	0.029 ± 0.003 (on the basis of 210Pb activity)
Car flue gases	0.129 ± 0.010

Additional emission of ^{210}Po during the combustion of coal can be confirmed by analysis of the ^{210}Po to ^{210}Pb ratio in the fresh fly ash samples. The results of such a comparison are presented in Table 4.

Table 4. Activity concentration of ^{210}Poand ^{210}Pb [Bq/g] and their activity ratio in fly ash samples.

Sample type	^{210}Po [Bq/g]	^{210}Pb [Bq/g]	^{210}Po/^{210}Pb activity ratio
Electrofilter sample 1 – Lodz	0.106	0.239	0.44
Electrofilter sample 2 – Lodz	0.090	0.209	0.43
Electrofilter sample 3 – Lodz	0.059	0.137	0.44
Electrofilter sample 1 – Chorzów	0.086	0.159	0.54
Electrofilter sample 2 – Chorzów	0.117	0.211	0.55

The average ^{210}Po activities in the captured fresh fly ashes are about half of the activity concentrations measured for the parent radionuclide ^{210}Pb. $^{210}PoO_2$ molecules which are released during combustion are then gradually adsorbed on the solid aerosol particles.

Activity concentration is strongly dependent on the aerodynamic diameter of the solid particles of aerosol[21]. Generally, fine particulate matter carries more ^{210}Po than the coarser fractions. The activity concentration increases with the decrease of aerosol diameter, causing an increased specific surface area of the particles. Similar behavior of lead and polonium on the surface of fine aerosols was presented in other works[21,29,30]. The activity concentration of ^{210}Po in particles collected on the filters with aerodynamic diameter in the range of 0.1–10 µm (attached fraction) and the activity concentration of fly ash collected in the electrostatic area is presented in Fig. 3.

The particles with diameters lower than 0.1 µm suspended in the atmosphere are not retained in conventional filters. Urban air can play the role of a reservoir for ^{210}Po in unattached form, which can move freely with air masses during the whole year and with the highest activity concentration in cold seasons. Fine particles not captured in the electric field, and thus freely leaving the chimneys of power plants, can be one of the main sources of increased activity of air and soil in the region of the emission source.

Different fate of ^{210}Po from its parents ^{210}Pb and ^{210}Bi in the fresh country air[16] is evident. Because of the higher volatility of ^{210}Po, the distribution coefficient between air and aerosols is higher than for ^{210}Pb and ^{210}Bi radionuclides. The behavior of ^{210}Po radionuclide is similar to the behavior of artificial ^{131}I isotope[31]. Masson[32] confirmed that even 90% of ^{131}I occurs in particulate matter in the air. ^{210}Po and ^{131}I isotopes show similar natures in air and can stay in gaseous form longer than in particle form.

Part of the polonium emitted into the atmosphere can be deposited on the surface of particles, thus increasing its activity concentration. For the 0.1–0.18 µm fraction, the measured activity concentration was equal to 5 Bq/g. The escaping unsupported polonium fraction and its coagulation and condensation are the most significant phenomena responsible for the rise of the ^{210}Po concentration in the urban air.

Polonium in the gaseous form that escapes from domestic central heating systems cannot pose a hazard for residents or power plant workers. Health risks posed by ^{210}Po from coal combustion can however be a serious problem, especially in the vicinity of major point sources of dust emission located in Lodz city centre.

Inhaled urban dust particles containing these isotopes released from coal burning may contribute to enhanced inhalation doses in the human lung. Therefore, the exact determination of the ^{210}Po released unsupported by ^{210}Pb and its compounds are significant for correct dose rate assessment (Equation (8)).

The effective doses D related to the excess of ^{210}Po due to aerosol inhalation for a child up to one year old (dose conversion factor: 15 µSv/Bq) and for adult inhabitants of Lodz city (3.3 µSv/Bq) were estimated to be 5.2 and 1.1 µSv/year, respectively. When estimating total radiological exposure, the gaseous form of ^{210}Po should also be taken into account. Emitted unsupported ^{210}Po in the volatile form can play an important role for the inhabitants of the central part of Lodz. The total radiological exposure of children and adults may be about 20 and 4 µSv/year, respectively. An increase in radiological hazard around local power plants has been confirmed previously[18] in surface soil up to 30 cm depth.

Conclusions

The excess of ^{210}Po activity concentration in the air was studied. The main source of polonium was connected with the activity of power plants and domestic coal typical for the winter season[33]. On average, about 50% of the total activity of ^{210}Po is discharged in the vicinity of the power plant.

The deficit of ^{210}Po activity confirms an efficient escape of polonium from dedusting installations. Eleven gigabecquerels of ^{210}Po per year are released into the urban air. However, industrial polonium emission cannot pose a radiological hazard as it is only slightly higher than the natural dose. On the other hand, in some agglomerations like Belchatow city in central Poland or Silesia region, inhabitants are likely to be more vulnerable to elevated ^{210}Po concentrations in the urban air. Therefore the activity concentration of ^{210}Po should be more carefully controlled. In such a region, the aerosol residence time method based on the ^{210}Po/^{210}Pb activity ratio is useless and should be replaced by a more appropriate ^{210}Bi/^{210}Pb method.

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Notes and references

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